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THz Photonics in Water and Beyond

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14. ABSTRACT
In the THz frequency range, laser-induced air plasma has become one of the most popular THz sources in the research laboratories. It can generate THz waves with a field strength of over MV/cm level by two-color optical excitation. The transient field is strong enough to induce nonlinearity. Recently, THz wave generation from ionized liquids, especially from liquid water, has also been successfully demonstrated. Liquids are preferable to general targets, as a flowing liquid line provides a fresh area for each excitation pulse, so the chaos caused by the previous pulse will not influence the next one. This makes it possible to use a high repetition rate laser for excitation. THz wave generation from ionized liquids presents photoionization processes that are different from those in gases.
The most important difference is the preference of liquid for excitation with a sub-picosecond pulse. In gas, the shortest pulse always generates strongest THz field, but in liquids, in a big surprise we have found that a laser pulse with longer pulse duration offers stronger THz emission!

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1 Introduction

The wavelength of terahertz (THz) radiation is between that of infrared light and microwaves, and thereby it has less energy than the band gap of many nonmetallic materials, enabling it to penetrate and image most solid objects. This could offer specialized uses in manufacturing, medical imaging, and security, such as in airport scanners. Moreover, since THz radiation has low photon energy, it is unlikely to lead to damage from photo-ionization, in contrast to the commonly used X-ray imaging, which also has a large penetration depth but with much higher photon energies.

Liquid water subjected to pulsed laser excitation emits unexpected THz radiation, offering possibility for inexpensive imaging devices. Electromagnetic radiation in the THz range has been generated from a variety of materials, including solids, plasmas and gases. However, investigators have doubted that liquid water, which strongly absorbs in the THz range, could be coaxed to emit THz radiation. Our "THz photonics in water and beyond" research project at University of Rochester has developed a new method to generate broadband THz radiation using a liquid water line with short laser pulse excitation, as reported in numerous publications from 2018 to 2021.

In our developed method, an 800-nm laser pulsed at a repetition rate of 1 kHz focused onto a flowing thin film of water is able to generate THz waves. While the interaction between laser light and water is still not fully understood, together with our collaborators from China and Russia, we believe that we have achieved a more detailed understanding of the physics involved. By calculating the electron density, we found that longer pulse durations generate more electrons in liquid. This is caused by the collision of electrons, which plays an important role in the ionization process. Cascade ionization – an electron avalanche in the ionized liquid, leads to an exponential increase in the number of electrons. For the gas case, however, the lifetime of electrons is always longer than the pulse duration, so the collision effect is always not considered. Our explanation illuminates the influence of optical pulse duration on laser-induced ionization in radiating THz waves, and contributes insight into the development of intense liquid THz sources to provide an alternative opportunity for THz nonlinear phenomenon.

THz wave generation from gas, cluster, solid and plasma has been substantially studied for decades. However, the demonstration of THz wave generation from liquid sources was conspicuously absent, especially from liquid water due to its infamously strong absorption characteristics in the THz regime. It is reasonable to expect that liquids might have unique properties if they could be harnessed as THz sources. Liquids have a high molecular density, close to that of solids, meaning that light over a certain area interacts with many more molecules than an equivalent cross-section of a gas material. The fluidity makes liquids very good candidates of high-repetition-rate targets for the study of high-energy-density plasma with ultra-intense laser pulses.

2. Key Results of the Project Accomplishments:

[a] My group has published and submitted 27 referred journal papers (including 4 pending) during the project period. Specifically, we had published 4 papers in 2018, 8 papers in 2019, and 9 papers in 2020-21. we gave 15 invited and 18 contributed talks during this period. [b] My last international trip is to a plenary talk at Taiwan Physical Society on Feb. 5, 2020. [c] Remote teaching is challenging. I taught two undergraduate/master level courses online. [d] Since July 2020, I am serving as the Executive Editor-in-Chief of Light, Science, and Applications after I stepped down from six years Editor-in-Chief of Optics Letters by the end of 2019. I am covering all the European submissions. Below list shows my service, honors, and scholarly accomplishments during the project period. [e] I have reached 3.3 million Traveling Miles in United Airline for academic events. This is more than six round trips from the earth to the moon (earth – moon, average 238,855 miles one way).

Matters are generally classified within four states: solid, liquid, gas, and plasma. Three of four states of matter (solid, gas, and plasma) have been used for THz wave generation with short laser pulse excitation for decades, including the recent vigorous development of THz photonics in gases (air plasma). However, the demonstration of THz generation from liquids was conspicuously absent. It is well known that water, the most common liquid, is a strong absorber in the far infrared range. Therefore, liquid water has historically been sworn off as a source for THz radiation. Recently, broadband THz wave generation from a flowing liquid target has been experimentally demonstrated through femtosecond laser induced micro-plasma. Liquid target as THz source presents unique properties, making liquids very promising candidates for the study of high-energy-density plasma, as well as the possibility of being a candidate for the next generation of THz sources.

2.1 Background

With tremendous advancements in laser technology, light-induced ionization in matter including gas, [1-6] clusters, [7-9] liquids, [10-16] and solids [17-20] have attracted considerable interest in generating coherent, intense, broadband THz waves through nonlinear processes. When a laser intensity is above the ionization threshold, abundant electrons and ions are created. [21-23] These charged particles can generate transient currents radiating electromagnetic (EM) waves, which cover the spectrum from microwaves to X-rays. [2, 24, 25] Recently, laser-produced air plasma becomes one of the most popular THz sources, generating THz waves with a field strength over MV/cm level under two-color (fundamental and its second harmonic beams) optical excitation. [26, 27] This opens up a new avenue of extreme THz science. [28] Concurrently, detecting fluorescence and acoustic waves from a plasma is crucial in characterizing the process of laser-matter interaction. Moreover, the fluorescence and acoustic wave can be enhanced by external THz fields through the collision process of accelerated particles. THz-field enhanced radiation emission of fluorescence (THz-REEF), [29-31] and THz-field enhanced acoustics (TEA) [32, 33] have been demonstrated for detecting broadband and coherent THz waves remotely.

THz wave generation from air plasma was first observed by Hamster et al. [1] in 1993 by focusing optical beams into a small volume of ambient air. It is awe-inspiring due to its simplicity. A THz source is achieved by using only one singlet lens. The photoexcited electrons in air plasma experience a ponderomotive force due to the density-gradient distribution, moving towards the areas with lower electron density. Since electrons cannot move faster than the laser beam, the plasma density keeps identical in the laser propagating direction resulting in that electrons are accelerated in the backward direction to create a dipole along the direction of laser propagation. [1, 2, 34] The THz wave emitted from the single-color air plasma show a conical energy distribution. [35]

In 2000, it was reported by Cook et al. [3] that the generation efficiency of THz waves from laser-produced gas-plasma is significantly enhanced under two-color excitation. This technology is a milestone in the THz community due to its capability of generating an intense THz pulse with a peak field over MV/cm as well as an ultrabroad bandwidth. For the two-color air plasma, the property of the THz wave can be coherently controlled by individually tuning the polarization and phase of each optical beam. [5, 36, 37] The generation process and the capability of coherent control are interpreted by a four-wave mixing model, [3, 5] a transient current model, [38-40] and a full quantum mechanical model numerically solving the time-dependent Schrödinger equation. [41] In the reciprocal process, ionized gas matter with a high third-order susceptibility can be correspondingly used to detect ultra-broadband THz pulses through the process of THz field induced second harmonic generation. [42, 43] The THz air-biased-coherent-detection (THz-ABCD) is capable of detecting the entire THz spectrum without the limitation of the phonon absorption existing in crystal detectors. [44]

To boost THz generation efficiency, some efforts have been done in ionized noble gases [45] and atomic clusters. [8, 9] However, further studies reveal electron density saturation and laser intensity clamping effect. [46, 47] Meanwhile, plasma produced in solid material shows advantages in generating intense THz waves. In 2008, Sagisaka et al. firstly reported THz wave generation from moving electrons on a solid target surface. [48] From then on, the record of THz pulse energy has been constantly created in solid targets. [17] Recently, it has been observed by Liao et al. that THz pulse energy exceeding millijoule (mJ) is generated from picosecond laser-irradiated metal foils. [18] For a solid target, it is difficult to realize a continuously replenishing for each pulse. Usually, the target is replaced for each pulse. Accordingly, the single-shot measurement is needed for detecting the signal. Instead, liquid targets can be circulated easily by adding a pump system. The recent demonstration of THz wave generation from liquid targets has attracted considerable attention due to its superiorities.

The demonstration of using liquids as a THz source was first reported in 2017 by two groups in different geometries. My group observed THz wave generation from a thin, free-standing water film. [10] Dey et al. reported

THz wave generation from ionized liquids by focusing intense laser into a cuvette. [11] With successive studies, [12-15, 49-51] THz wave generation from liquids show different properties comparing to that from other targets. In this paper, we review recent studies of THz wave generation from flowing liquid targets as well as the ultrafast dynamics of liquids under intense THz excitation. A comparison of THz wave generation from air plasma and liquid plasma is provided. Different liquids as THz sources are discussed in detail. Terahertz liquid photonics shows a potential to developing new type THz sources, and also offers a new perspective to investigate the process of laser-liquid interaction.

2.2 THz wave generation from air

THz generation process in air plasma is intimately tied with gas ionization, it requires that the intensity of the optical pulse is greater than 10^{12} W/cm^2. This can be achieved through geometric focusing intense laser pulses with lenses or curved mirrors. When the intensity exceeds the ionization threshold, a plasma channel with mm to cm lengths is formed in air. This plasma channel stabilizes the beam at very small diameters (30 to 100 μm) and maintains high intensities over ranges much longer than the Rayleigh length of a traditional, geometrically focused beam. THz wave is emitted by the transient current formed in the air plasma.

Figure 1 shows the schematic of a typical two-color air plasma system. The femtosecond laser beam is split into the pump beam and the probe beam. The pump beam is focused into air to create an air plasma, generating THz waves. After THz filters, the THz signal is coherently measured by the probe beam through either electro-optical sampling (EOS) or air-biased-coherent-detection (ABCD). The excitation of using one fundamental beam is known as single-color excitation. Two-color excitation is realized by inserting a frequency doubling crystal, usually BBO, into the focused pump beam to generate the second harmonic pulse. Then, both fundamental and its second harmonic are focused into air to create a plasma. Comparing to the THz signal under single-color excitation, the THz field from two-color air plasma is 3 orders higher with a much broader bandwidth.

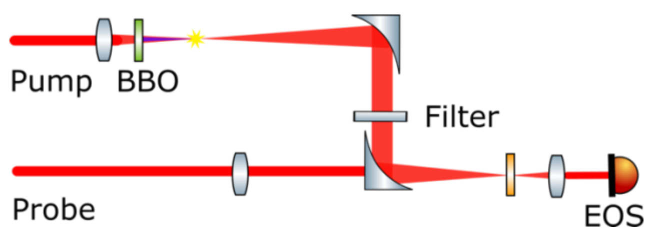


Figure 1. A typical THz two-color air plasma system. The laser beam is split into pump beam and probe beam. The air plasma is produced by focusing the pump beam. One BBO crystal is inserted to create the second harmonic pulse. The THz signal is detected trough electro-optic sampling (EOS).

2.3 THz wave generation from water

Compared with gas material, liquids are easier to be ionized due to the relatively low critical potential. [52-54] As the most common liquid on Earth, water has strong cohesion, adhesion, and surface tension, which are beneficial in forming a free-standing liquid targets with smooth surfaces. It's also colorless, transparent, chemically stable, non-toxic, and inodorous. These properties make water one of the ideal objects for the research community. Besides, water is an excellent solvent for a wide variety of substances. To study the THz wave generation from water, the same setup as shown in **Figure 1** is employed, and a thin liquid target is inserted at the focus. Instead of an air plasma, ionization in liquid is created.

2.4 Flowing liquid targets

Solid and liquid material has a similar molecular density, which is about 3 orders higher than that of gas at the standard ambient temperature and pressure. However, solid targets under an excitation of intense laser pulses are hard to make the measurements reproducible when the target surface is degraded by laser radiation through heart or direct ionization. Instead, the fluidity of liquid material makes the target is able to provide a fresh area for each pulse. Benefiting from this, the influence between optical excitation pulses can be eliminated. Using liquid targets as sources has been studied for decades in the generation of extreme ultraviolet radiation and x-ray for the requirement of the laser induced high-density plasma. [55-57] For THz wave, however, most liquids show a strong absorption. Such as the most common liquid - water, the absorption coefficient at 1 THz is about 220 cm^{-1}. [58, 59] In order to reduce absorption of THz signal in water, the target with a thickness less than 500 μm is required for THz wave generation.

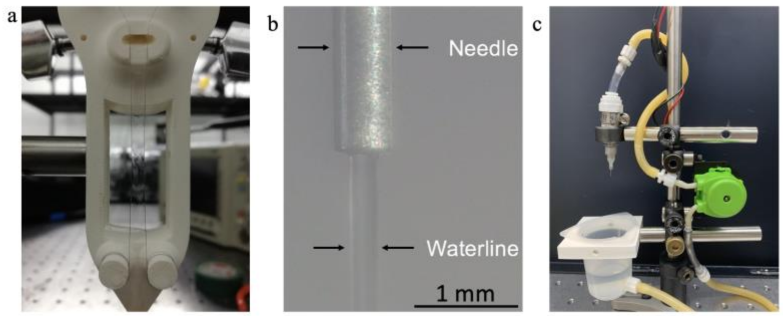


Figure 2. Liquid targets and the liquid system. a A gravity-driven water film. The thickness is 210 μm. **b** A pump-driven water line with a diameter of 260 μm, which is guided by a syringe needle. **c** A home-built liquid system, which needs only 50 ml water for several hours running. A syringe is used as a jet for producing a liquid line.

The flowing target can be driven either by the gravity or a pump. **Figure 2a** shows a gravity-driven water film, in which the film is guided by two parallel metal wires. The transparency in the visible range of the film indicates a thin thickness as well as the smoothness of the film surface. The surface tension of liquid water is relatively high, which is able to support a film with thickness in the level of several micrometers. The thickness of the film is tunable by changing the flow rate or the gap between the parallel wires. Higher flow rate produces thicker films. A liquid jet is another way of making a flowing target. A circulating system can be easily made by pumping liquids from a collecting reservoir to nozzles. The liquid jet can be shaped by the nozzle shape. **Figure 2b** shows a pump-driven water line. In this case, a syringe needle with an inner diameter of 260 μm works as a nozzle. **Figure 2c** is a home-build liquid line system, which consists of a peristaltic pump, a jet (syringe needle), a liquid reservoir and some pipes. This system is compact and easy to build, which is able to produce the liquid lines with diameters from 90 μm to 510 μm by using different syringe needles. Moreover, a test of hours operation shows that it only needs 50 ml liquid material in the circulating system. Also, the liquid line from this system is very stable and has an excellent liquid-air interface. It should be noted that the smoothness of the liquid target surface is important to get a better focus of the laser beam without too much scattering.

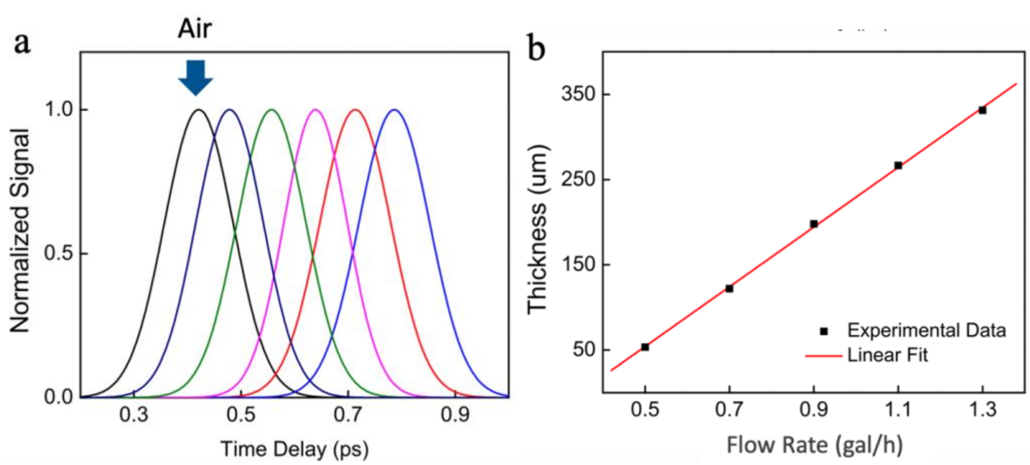


Figure 3. Calibration of the water film thickness. a The original (air) signal from the autocorrelation system is plotted in the black line. With the water film in one of the two beams of the autocorrelation system, the signal peak will move linearly with the increase of thickness. **b** The linear dependence of the thickness on the flow rate. The thickness of the water film can be continuously tuned from 50 μm to 350 μm.

To calibrate the thickness of a liquid film precisely, we can use an optical autocorrelation system by inserting the film into one of the two beams. [60] **Figure 3a** shows the measurement results. The black line is the original signal from the autocorrelation system without a water film. After inserting the water film, the peak delays to a certain position because of the different optical path caused by the liquid film. The peak position moves linearly with the increase of the film thickness. **Figure 3b** shows the linear dependence of the thickness on the flow rate. In our case, the thickness of the wire-guided water film can be tuned continuously from 50 μm to 350 μm. The accuracy of the controlled film thickness is within 20 μm.

2.5 Single-color optical excitation

THz wave generation form liquids was firstly demonstrated under single-color excitation. Because the thickness of a liquid target is much thinner than 1 mm, to confine most pulse energy in the target, the length of ionization region should be shorter than the thickness. Therefore, creating a micro-plasma in the liquid is needed by focusing a laser beam through a high numerical-aperture (NA) objective. [34]

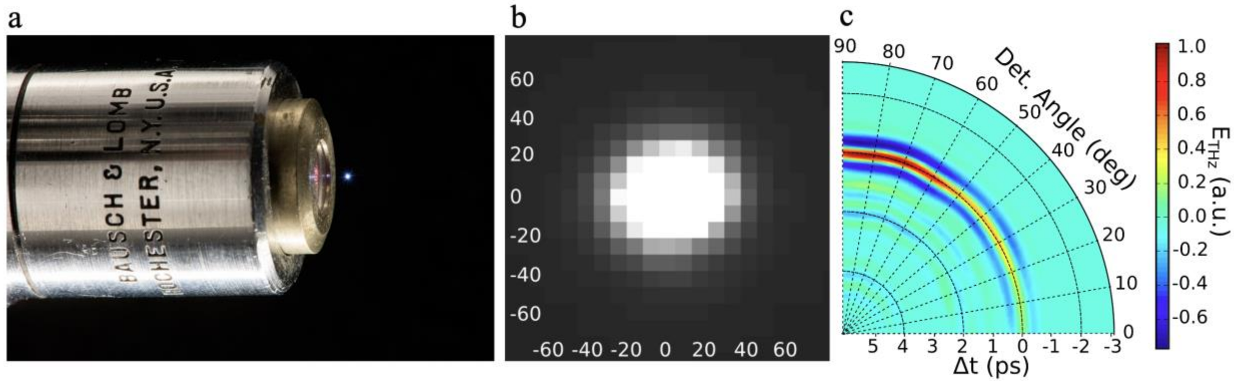


Figure 4. Micro-plasma and the signal distribution. [34] **a** A photo of the micro-plasma fluorescence obtained by focusing the laser beam with a high NA microscope objective. **b** A magnified image of the micro-plasma (unit is μm). However, the plasma spot's contribution to the THz wave is about one μm or less. **c** Measured THz field as a function of the THz emission angle. The optimal angle is 80 degree from laser propagation direction.

Table 1. Comparison between micro-plasma and elongated plasma.

Micro-plasma	Filament (Elongated Plasma)
Length: < 500 μm Width: < 10 μm	Length: few mm up to meters Width: ~ 100 μm
Tight focusing of the laser (NA > 0.1) Higher peak laser intensity (> 5×10^{14} W/cm^2) Higher peak electron densities (~10^{18}-10^{19} cm^{-3})	Loose focusing of the laser (NA<< 0.1) Lower peak laser intensity (~1×10^{14} W/cm^{20}) Lower peak electron densities (~ 10^{15}-10^{16} cm^{-3})
Position does not change with laser energy	Position changes with laser energy
Lower laser energy threshold (< 1 μJ)	Higher laser energy threshold (~ 50 μJ)

Figure 4a is a photo of a micro-plasma induced by a 0.85 NA air immersion microscope objective in ambient air, where the laser pulse energy is 65 μJ. The fluorescence spot at the focal point is shown in **Figure 4b**. This longitudinal and transverse sizes of this micro-plasma is smaller than 30 μm, which are measured by an iCCD camera. **Figure 4c** is the measured THz field as a function of the detection angle. The direction for the maximum THz emission is nearly perpendicular with respect to the optical axis. We attribute this sideway THz radiation to the steep Ponderomotive potential at the focal plane, which accelerates the free electrons created by photoionization. The comparison of micro-plasmas with the elongated plasmas is summarized in **Table 1**. The

concept of a micro-plasma is employed for exploring THz wave generation from thin liquid targets, meaning a lens with a short-focal length is used in the setup.

Because of the high molecular density, scattering and nonlinear effects in water are much stronger than those in air. By focusing a laser beam with same pulse energy and pulse duration, the fluorescence is much brighter in water. **Figure 5a** is a photo of a flowing water line, which is ionized by a tightly focused laser beam. By using a traditional THz time-domain spectroscopy system, a THz waveform is measured in the forward direction (laser propagation direction). **Figure 5b** is a typical THz pulse emitted from water. This signal is detected through EOS, which has a dynamic range over 1100 and a signal-noise ratio (SNR) better than 300 when the pulse energy is 0.4 mJ. Specifically, the dynamic range of a system is defined by the ratio of the signal peak level and the noise level of the background, which indicates the sensitivity range of a system. And the SNR is defined by the ratio of the maximum signal and the fluctuation of the maximum signal itself, which describes the stability of a system. Therefore, the SNR can never be greater than the dynamic range. Here, the relatively low SNR mainly results from the vibration of the target surface. The inset shows the corresponding spectrum. The center wavelength is near 0.5 THz, which is due to the strong absorption at higher frequency in water.

a

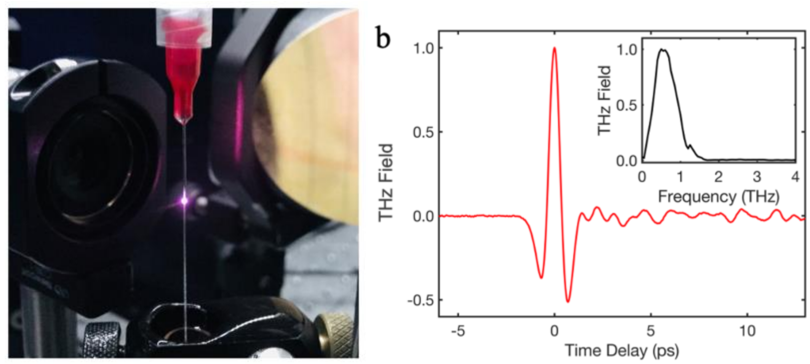


Figure 5. THz generation form liquid water under single-color ionization. a A photo of a fluorescence from a laser induced micro-plasma of a water line. **b** A typical THz waveform generated from a water line and its corresponding spectrum (inset).

In contrast to gas target without interfaces, the liquid and solid targets bear the target/air interfaces in the generation process. Because many solid metal targets are not transparent to the optical laser beam and THz wave, the detection is generally conducted in a reflection geometry. For a free-standing transparent liquid target, the transmitted geometry is in favor. The system can be optimized by air plasma first, then it can be switched for studying the liquid generation process by inserting a liquid target at the focus without changing other optical elements. In this case, the optical beam refracts at the air/liquid interface first. Because water is easier to be ionized than air, the ionization sometimes starts at the surface when the optical intensity exceeds the ionization threshold of water but lower than that of air. The refraction angle of the optical beam is a key parameter for maximizing the THz signal. **Figure 6a** shows normalized THz energy with respect to the incidence angle (α). The positive and negative signs of α indicate that the film is rotated in a clockwise or counterclockwise direction, respectively. The squares show the THz energy obtained from the integral of the THz waveforms. The error is the standard deviation of multiple measurements. The solid line is a simulation result based on a dipole approximation model, in which the electrons in the ionized area experience the ponderomotive force and create a dipole along the direction of laser propagation. Both, the experiment and simulation results show that the THz field is maximized when the incidence angle is about 65°, which results from the transmittance of the p-polarized excitation laser at the air/water interface and the dipole orientation direction caused by the refraction of the surface. **Figure 6b** plots THz waveforms generated from the water film when the incidence is ±65°. The THz field has the same peak value but opposite polarity. The insets show the experimental geometry and the flipped direction in the projection of the dipole (red arrow) for two cases. The opposite direction of the dipole projection results in the flipped waveforms accordingly.

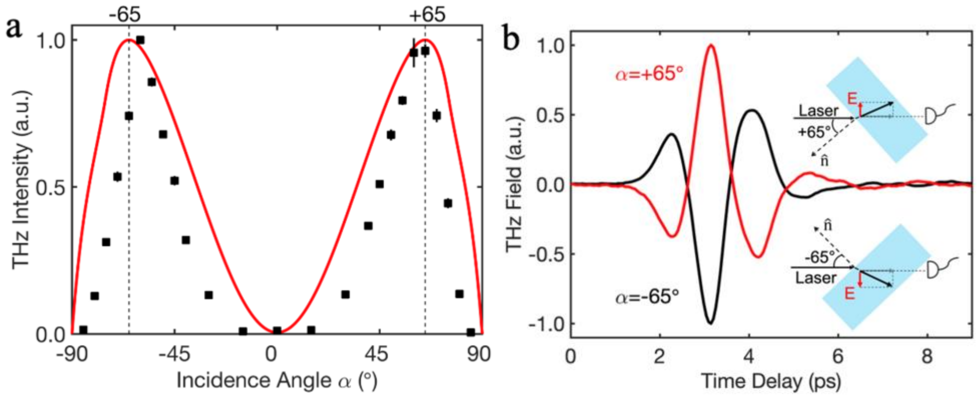


Figure 6. The dependence of THz signal on the incidence angle α. [10, 12] a THz intensity with different incidence angles is studied by rotating the water film. The signal is maximized when α is 65°. The squares are the experimental results, and the red line is a simulated result from a dipole model. **b** The waveforms show optics polarities with opposite incidence angle. The insets explain the observation with a contribution of the flipped dipole projection (E) of opposite incidence angles.

Another influence of the target/air interface is the total internal reflection for the emitted THz wave, which greatly decreases the transmission of THz wave at the flat surface. In other word, a liquid line or droplet target could offer stronger THz signals than the flat film. L. Zhang et al. demonstrated intense THz radiation from a water line. [15] Instead of rotating the water film to get an optimized incidence angle, THz signal from a liquid line is optimized by scanning the position of the liquid line across the laser focus. During the movement, the effective thickness in the waterline varies continuously, as well as he optical incidence angle. A similar incidence angle dependence but a much stronger signal is obtained by using a liquid water line than a flat film with the comparable thickness.

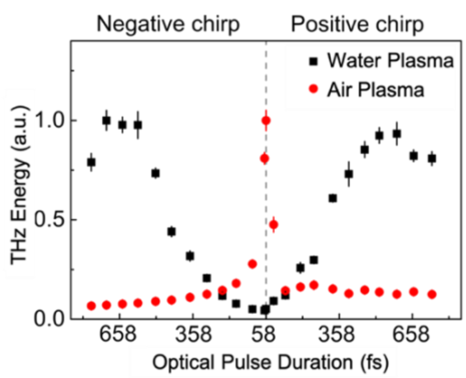


Figure 7. Normalized THz energy with different optical pulse durations. [10] THz signal generated by water plasma prefers an optical excitation with a longer pulse duration. However, the generation from air plasma is always in favor of the shortest pulse.

One remarkable property of THz wave generation form liquid is the preference of the longer optical pulse duration [10, 50]. **Figure 7** plots the dependence of THz energy on the optical pulse duration. For air plasma, the maximum THz signal is always obtained when the pulse is close to the Fourier-transform-limited pulse. Higher peak power always leads to a stronger ionization in the air case. However, THz wave generation from liquids surprisingly prefers a longer pulse. As shown in **Figure 7**, THz signal increases with an increased pulse duration by adding a frequency chirp in the pulse. At a certain pulse duration, the THz signal reaches its maximum. By comparing the THz spectra respectively generated by negatively and positively chirped pulse, we didn't find apparent difference in both the bandwidth and amplitude, which suggests that the chirp of the pulse doesn't play an important role here. Instead, pulse duration is the key parameter. This phenomenon can be explained by the different ionization processes in liquid and air, respectively. A high molecular density of liquid causes a more frequent collision. The mean-free time of ionized electrons is about 1 fs in a liquid, leading to hundreds of cycles of collisions happen within the ionization process when the pulse duration is sub-picosecond. Therefore, the electron density is mainly decided by the cascade ionization process in liquid. However, multiphoton and tunneling ionization are more important in the case of gas. For more information on the theoretical analysis about single-color excitation, please check refs. [15, 50, 61, 62]

2.6 THz emission in sideway

Similar to THz wave generation from a micro-plasma in air, a thin liquid target also emits THz wave in the sideway directions. **Figure 8a** schematically illustrates the system for the sideway measurement, in which the THz radiation propagating perpendicular to z direction is measured by a sideway detector. For a fair comparison, the forward detector measures the signal in z direction by using the same EOS crystal to assure the same detection efficiency. The pump beam is focused by a 2-inch focal length lens onto a water line. **Figure 8b** shows the comparison of THz waveforms generated from water and air, respectively. Here, a 210 μm water line is used as a source. The pump pulse energy is 0.4 mJ for all cases. All the signals are optimized for getting the maximum signal by tuning the pulse duration. For water, the optimal pulse duration is 534 fs. And the shortest pulse duration (~ 140 fs) is used for the air plasma generation. The results show that the THz signal generated from water is much stronger than that from ambient air. And the THz signal generated from air plasma is mainly in the forward detection. There is no detectable THz signal from air in sideway. To get the sideway signal from air plasma, a lens with a higher NA is needed. However, the signal from liquid water in sideway is about twice as much as that of its forward signal. The corresponding spectra are shown in **Figure 8c**. Both signals from water shows a narrower bandwidth than that from air plasma. The strong sideway THz signal is stemmed from either the strong scattering from the liquid or the shorter longitudinal dimension of the "micro water plasma" caused by the much higher molecular density in liquid. This observation also confirms that the THz generation from liquid water has a much higher efficiency than that from air.

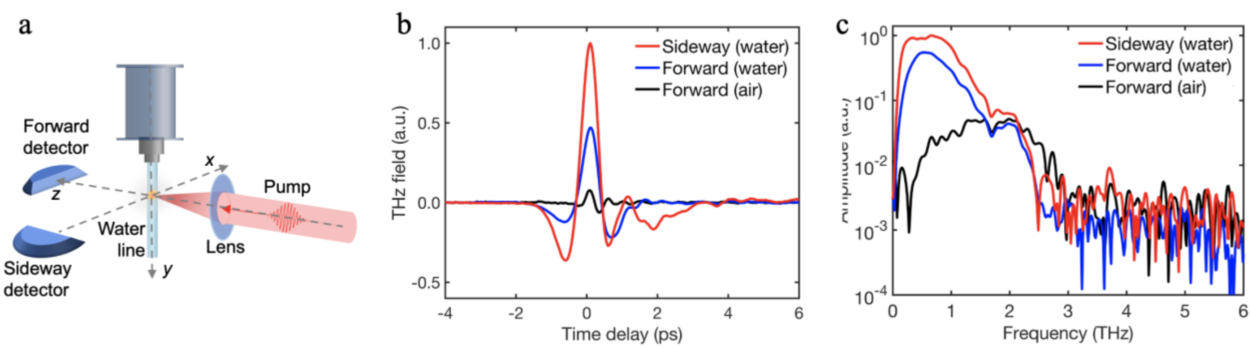


Figure 8. Comparison of forward and sideway detection. a Setup schematic for forward and sideway detection. The pump beam is along *z*-axis. The water line flows in *y* direction. Two EO detectors are in z and -x directions, respectively. **b** Comparison of THz waveforms. The pulse energy is 0.4 mJ. Corresponding spectra are shown in **c**.

2.7 Two-color optical excitation

By mixing the fundamental beam and its second harmonic, THz wave generation efficiency from a gas-plasma is greatly improved, which is usually known as two-color excitation. A similar enhancement is expected to boost the THz signal from liquid. However, instead of several orders, THz field generated under a two-color excitation in liquid water is about 10-times stronger than that from the one-color excitation scheme. [13] This may be caused by the short mean free path length of the electrons in water, which results in a decrease of transient photocurrent. [39, 63] Alternatively, ionized electrons in liquid are quasi free and less sensitively to the symmetric field.

Similar to a gas case, by precisely tuning the phase between the fundamental and its second harmonic beams, the THz field can be coherently modulated [13]. **Figure 9a** shows that the THz waveform from liquid water under two-color excitation is completely flipped over by changing the relative phase of two pump beams by π. The inset plots the THz peak field as a function of the relative phase. **Figure 9b** shows the THz wave energy measured by a Golay cell as a function of the relative optical phase. The noise floor is measured when the THz signal is blocked. It is interesting that some of the signals cannot be modulated through the coherent phase control. By calculating from the noise level, the modulated components are about 70% of the total energy. The modulated and unmodulated THz waves reflect different generation processes. For the further exploration, the excitation power dependence for these two components is studied, which is shown in **Figure 9c**.

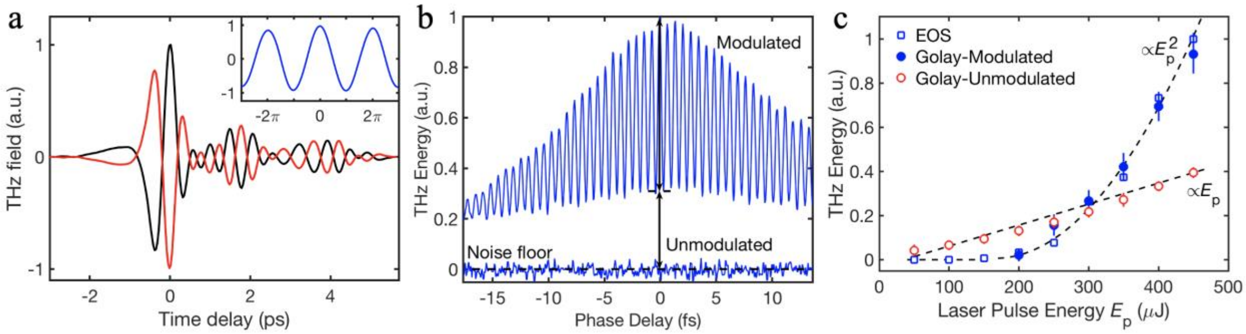


Figure 9. THz generation from liquid water under two-color excitation. [13] **a** Two THz waveforms obtained when the relative phase is π and 2π. Inset, THz electric field as a function of the phase delay. **b** THz wave energy with a tuned phase between 800 nm and 400 nm pulses. The THz energy was monitored by a Golay cell. **c** Normalized THz energy from liquid water as a function of the total excitation laser pulse energy. Blue squares show the THz energy calculated from the temporal integral of the THz waveform measured by EOS. Blue dots indicate modulated THz energy measured by a Golay cell. Red circles present unmodulated THz energy measured by the Golay cell.

The unmodulated part (red circles) shows a linear dependence on the laser pulse energy. For the modulated part (blue dots), there is a threshold (0.2 mJ), beyond which the signal appears. Its tendency matches a quadratic function above the threshold. This result is also coincident with the result measured from EOS (blue squares). Unlike EOS, the Golay cell can measure both coherent and incoherent signal. Thus, the modulated portion is mainly from the coherent signal resulting from the buildup of bremsstrahlung from electron-atom collisions in the electron acceleration process. [41] In contrast, the unmodulated part may include both coherent and incoherent signal arising from multiple physical processes. For instance, a spatial net charge distribution created by the ponderomotive force radiates THz waves. Since no threshold is observed for the unmodulated portion, the THz wave emission can also be attributed to part of the broadband radiation from the combination of thermal bremsstrahlung from electrons and electron-ion recombination. Besides providing an approach to reveal more information about plasma behaviors in liquids, two-color excitation offers an insight into developing a liquid THz source that may have applications in THz nonlinear optics and THz-driven electron acceleration.

2.8 THz wave generation from other liquids

The demonstration of THz wave generation from liquid water opens one new avenue for studying THz liquid sources. In contrast to solid material, two distinguish properties are usually used to characterize liquid material. One is viscosity, which depends on the friction among molecules. The other one is surface tension, which relates to the surface that resists force and keeps the liquid together. Besides, ionization threshold, electron density buildup and THz dispersion are important parameters in the THz generation process. Liquids with different properties emit THz waves with different properties. Measuring THz wave from different liquids reflects the ultrafast dynamics of the laser-liquid interaction process. In this chapter, we discuss THz wave generation from different liquids.

2.8.1 Liquid nitrogen

Compared to liquid water, liquid nitrogen (LN_2) is a good candidate for tests based on the following properties: (1) LN_2 has a surface tension of 8.85 mN/m at -196 °C, which is the lowest surface tension to our knowledge except liquid helium. For water, it is 71.97 mN/m at 25 °C. (2) LN_2 has a much lower viscosity (164 mPa·s at -196 °C) than water (889 mPa·s at 25 °C). (3) LN_2 is a nonpolar liquid, which has a much lower absorption in THz regime. [64] (4) LN_2 liquid phase is at a cryogenic temperature (-196 °C). It's interesting to study different dynamics of liquids with a huge temperature difference under the same optical excitation. THz wave generation from a bulk LN_2 has been demonstrated recently under double pump geometry with either single- or two-color excitation. [16] The shockwave in liquid created by the laser pulse may affect the interaction of the next pulse. To eliminate the influence between pulses, a flowing LN_2 line is required. However, the room temperature is much higher than the boiling point of LN_2, which is a challenge to create a flowing line at room temperature.

Actually, when liquid contacts with a hot surface whose temperature is higher than the boiling point, vaporization creates an insulating layer preventing the liquid from boiling rapidly. This is known as the Leidenfrost effect [65, 66]. Benefiting from this effect, it is possible to create a flowing LN_2 line at ambient environment. Additionally, another factor should be concerned is the transient vaporization of LN_2 caused by the pressure difference. Usually, LN_2 is stored in a Dewar with a higher pressure inside than the outside. The boiling point highly depends on the pressure, which drops with the decrease of the pressure. It means that the boiling point inside the Dewar is higher than outside. If a Dewar is connected to a jet directly, a mixture of gas and liquid phases ejects out from the jet. To get rid of this problem, a phase separator and a custom-designed LN_2 reservoir are needed. As shown in **Figure 10a**, the LN_2 reservoir consists of a syringe, a flask, and an insulating layer. The metal syringe is immersed in a flask filled with LN_2 for maintaining the cryogenic temperature. Outside the flask, a thick insulating layer is employed to resist the heat transition between the flask and the ambient environment. The volume of the flask is 600 ml. While filling the flask with LN_2, the syringe is blocked at the beginning until the setup is cooled down. Benefiting from the high thermal conductivity of the metal syringe, the setup reaches the thermal balance quickly. After removing the block. A steady liquid line is formed at the ambient environment. **Figure 10b** is a photo of the flowing LN_2 line. The diameter of the flow is estimated to be 400 ± 5 μm, which is determined by the inner diameter of the syringe needle. The high transparency indicates a smooth surface as well as a stable flow.

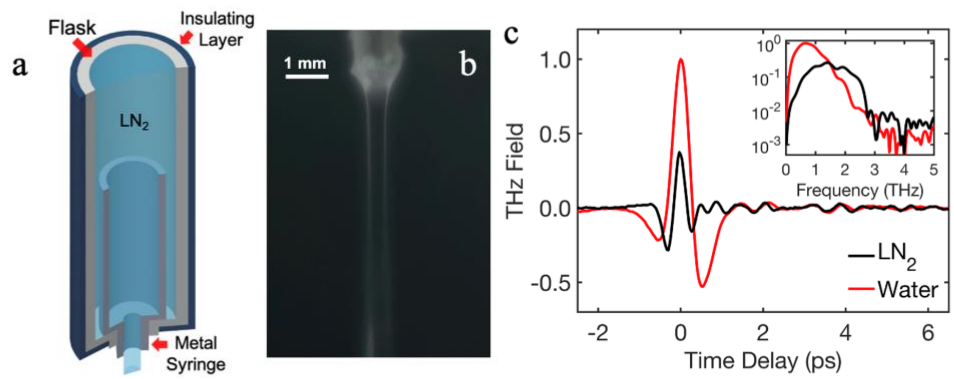


Figure 10. THz generation from liquid nitrogen. [67] **a** Diagram of the apparatus for guiding an LN_2 line. By filling it with LN_2, the liquid in the flask prevents the liquid in the syringe transferring heat. **b** A photo of a flowing LN_2 line in an ambient environment. The transparency of the line indicates the liquid phase clearly with a smooth surface. **c** Comparison of THz waveforms from water and LN_2 under the same optical excitation. Inset: Corresponding spectra.

Figure 10c plots the THz waveforms from the LN_2 line and a water line under the same experimental conditions. The peak field from LN_2 is 0.4 times weaker than that from water. However, the THz signal from LN_2 shows a shorter pulse duration. By fitting the envelope of the signal in the field, the signal from LN_2 has a pulse duration of 0.6 ps. For the water signal, the pulse duration is about 1 ps. The corresponding spectra are shown in the inset. The LN_2 shows a broader bandwidth with more high-frequency components. There are two possible reasons. First, LN_2 has a low absorption in THz frequency because it's a nonpolar liquid. Additionally, the vaporized N_2 keeps purging the system to preserve the high frequency components. More details of THz wave generation from a flowing LN_2 can be found in our published paper. [67]

2.8.2 Liquid gallium

Metal targets have been considered as great THz radiation sources because of their relatively lower ionization thresholds, which allow THz wave generation by using a lower pump energy compared with water. It would be interesting to discuss the different contributions of electrons excited by laser pulses and free electrons originally existing in the liquid metal in the THz generation process. Besides, the surface tension of liquid metal is much higher than that of water, meaning that it is able to form a smoother surface of a flowing liquid line. Because of the chemical stability and physical safety, liquid metal, such as gallium, has been widely used in x-ray generation applications as well. [68-70]

Table 2 lists the melting point, surface tension, density, viscosity and ionization energy of several selected liquid metals. Among those, mercury has the lowest melting point. For the toxicity concern and chemical stability, gallium is more in favor for lab users. The melting point of gallium is 30 °C. Using a heater attached liquid circulating system, a high-quality liquid gallium line is obtained.

Table 2. Properties of some liquid metals comparing with water.[92-94]

Metal Targets	Melting point (°C)	Surface tension (N/m)	Density (g/cm3)	Viscosity (Pa·s)	Ionization energy (eV)
Water (at 20°C)	**0**	**0.073**	**0.998**	**0.0010**	**6.5**
Galinstan (at 20°C)	-19.0	0.718	6.440	0.0024	-
Mercury (Hg) (at 20°C)	−38.8	0.487	13.534	0.0015	10.44
Francium (Fr) (at 30°C)	27.0	0.051	1.870	-	4.07
Cesium (Cs) (at 60°C)	28.5	0.675	1.843	0.0058	3.90
Gallium (Ga) (at 53°C)	**29.8**	**0.723**	**6.080**	**0.0019**	**5.98**
Rubidium (Rb) (at 100°C)	39.3	0.854	1.460	0.0048	4.18
Phosphorus (P) (at 50°C)	44.0	0.698	1.740	0.0016	10.64
Indium (In) (at 170°C)	156.6	0.558	7.020	0.0019	6.08

Figure 11a shows the schematic of using a liquid gallium line as a THz source. The laser beam is focused by a 2-inch focal length lens on the liquid metal target. The position of the liquid line near the focus is finely controlled by a two-dimensional translation stage. The diameter of the gallium line is 210 μm. A flowing rate is about 3.8 m/s, which can be controlled by the pump. The forward propagating THz signal is detected, as shown in **Figure 11b**. THz signals from both gallium and water lines are much stronger than that from air plasma. The signal generated from liquid gallium is 1.7 times stronger in field than that from water at 35 °C. Also, it is noteworthy that the THz signal from liquid gallium shows a broader bandwidth than that from water. More components at high frequencies are generated.

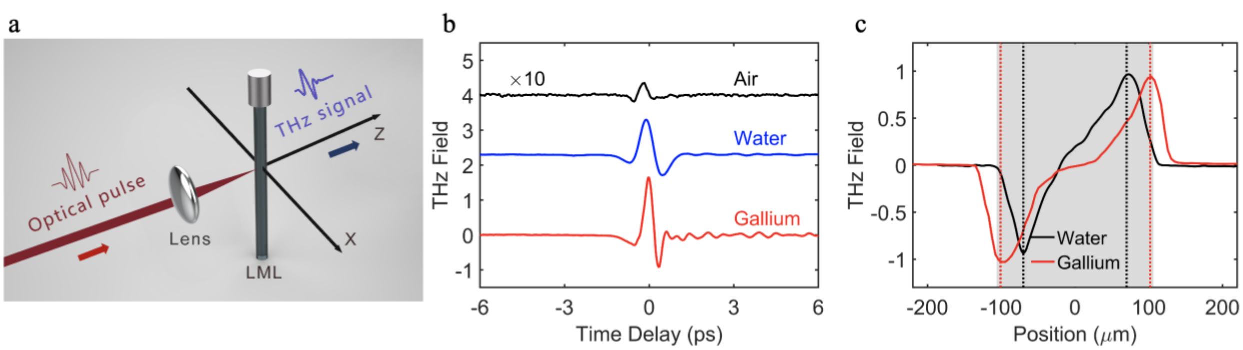


Figure 11. THz generation from liquid Gallium. [71] **a** Schematic of the experimental setup. A two-dimensional translation stage is used to control the position of the x-axis and the z-axis of the liquid metal line. The optical beam propagates along the z-axis. THz emission is measured by a standard electro-optical sampling. **b** Comparison of THz waveforms generated from air, water and liquid gallium under single-color excitation. The target diameter is 210 μm. The signal from air plasma is enlarged by 10 times. **c** Normalized THz field strength as a function of the x-axis position. The dashed lines represent the x-axis position of the maximum THz fields. The gray area shows the diameter of the liquid line.

Figure 11c plots the dependence of THz field strength on the x-position of the source. The position of liquid line is scanned across the focus. When x is 0, the optical pump pulse is focused on the center of the gallium line. For a comparison, a similar measurement of water is also plotted. The dashed black and red lines illustrate the x position when the THz fields reach maximum for two liquids, respectively. The measurement clearly shows that the separation between the two maxima of the gallium signal is larger than that of water. The water line has a same diameter as that of the gallium line, which is shown by the gray area. The different sign of the electric field indicates waveforms with opposite polarities. This result shows that the THz signal is generated at the surface of gallium.

The skin depths of liquid gallium for optical and THz waves are estimated to be 7.7 nm and 60 nm, respectively. Therefore, ionization can only happen at the surface of liquid gallium. Since a longer pulse duration works better for THz wave generation from liquids, enough seed electrons are ionized through multiphoton ionization or tunneling ionization at the front part of the optical pulse. Then cascade ionization occurs, in which collisions of electrons lead to an exponential increase in the number of electrons. Then, electrons are accelerated by the ponderomotive force as a result of the non-uniform density gradient distribution of plasma. Eventually, THz wave emits from liquid metal.

Although the flip of THz waveforms from liquid gallium observed by scanning along the x-axis shows similar characteristics with a water signal, the mechanism of THz waves generation from liquid metals likely differs from the generation process of liquid water. Further exploration still need to be conducted. The THz wave emission pattern, especially the sideway and the backward detection, is an important experiment to understand the THz radiation mechanism. More details of THz generation from a flowing liquid gallium line can be found in Ref. [71].

2.8.3 Liquids with different polarity indices

Molecular polarity describes the separation of positive and negative charged particles of a molecule. A polar molecule possesses a dipole contributing to a strong absorption in THz range. [72] Generally, liquids made of nonpolar molecules show a low absorption. Polarity index is a quantitative parameter that defines how polar a liquid is. [73] To investigate the influence of liquid absorption, liquids with different polarity indices are tested. **Table 3** lists liquids with different polarity indices. Stronger THz signals are expected from liquids with smaller polarity indices.

Table 3. List of selected liquids with different polarity.

Liquid	Polarity	Boiling T (°C)
Alpha-pinene	~ 0	156
Isooctane	0.1	99
Butyl chloride	1.0	78
P-xylene	2.5	138
Methylene chloride	3.4	240
Ethyl acetate	4.3	77
Pyridine	5.3	115
Ethanol	6.3	78
Water	9.8	100

Figure 12a shows the peak-valley values of THz electric fields obtained from four different liquids, which are α-pinene, p-xylene, ethanol, and water. Their polarity indices are 0 (nonpolar), 2.4, 5.2, and 9, respectively. Among these liquids, α-pinene offers the strongest THz signal, which is about 1.8-times stronger in field than that from water under the identical experimental condition (liquid line diameter, laser pulse energy, laser pulse duration,

F/#, etc.). **Figure 12b** plots the comparison in the corresponding spectra. Water generates the THz signal with much less high-frequency components. This results from the stronger absorption of water in the high THz frequency region. Although ethanol exhibits a similar peak-valley value of the THz field to water, it generates more high-frequency THz waves. The widest bandwidth is achieved by p-xylene while the highest amplitude is obtained by α-pinene. Even though there are some other factors that may also influence the THz radiation from different liquids such as refractive index, surface tension, and viscosity, the result shows that molecular polarity or absorption coefficient of liquid plays an important role in the generation process of THz waves both in peak field and pulse bandwidth.

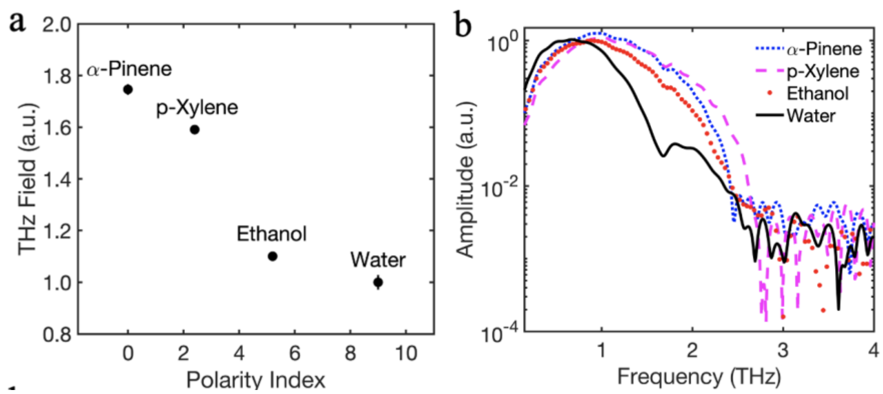


Figure 12. THz generation from liquids with different polarity indices. a THz fields generated from four liquids, which are α-pinene, p-xylene, ethanol, and water. Their polarity indices are 0, 2.4, 5.2, and 9, respectively. **b** Corresponding spectra.

2.9 THz wave enhancement under double-pump optical excitation

Double-pump optical excitation is a technology to boost the generation efficiency via the nonstationary nonlinearity enhancement, in which two pump beams with a certain time delay are used for excitation. Usually, the first pump ionizing target first is relatively weaker in power than the second pulse. The time delay is optimized to get the highest signal. To boost the generation efficiency for different wavelengths, the time delay may vary greatly. The double-pump excitation was originally a practical strategy to enhance the x-ray generation efficiency from liquids. Berglund et al. reported an eight-times increase in the intensity of extreme-ultraviolet emission by using ethanol droplets as targets for a double-pump excitation. [74] Additionally, Anand et al. reported a 68-times increase in the intensity of hard x-ray emission from methanol droplets when irradiated by double-pulsed femtosecond laser pulses with a delay time of 10 ns. [75] Double-pump excitation is also studied in THz wave generation [16, 76-79]. Recently, THz generation enhancement by several orders of magnitude in power is observed from gallium droplets under double pump excitation. [80]

Figure 13a shows a schematic of double-pump excitation on a liquid line for THz wave generation in top view. Two pump beams propagating collinearly with a time delay $\Delta\tau$ are focused on the liquid line. The position of the liquid line is precisely controlled by a two-dimensional stage in the x-y plain. When the line moves across the focus, the incidence angle of two pumps changes accordingly because of the curved surface of the liquid line. THz signal generated from liquid water is shown in **Figure 13b**. The top two waveforms show the results from two individual pumps, respectively. The time delay $\Delta\tau$ between two pumps is reflected on the time delay of THz waveforms, which is 3.9 ps. This time delay is longer than the pulse duration, and smaller than the lifetime of the photoexcited electrons (nanosecond level). The greater pulse energy of Pump 2 produces a stronger THz signal. Two waveforms keep same shape indicating a good overlap of the two beams. The bottom waveform shows the result of the double-pump excitation. The THz signal generated by pump 2 is enhanced because of the existing of the ionization by pump 1. The waveform keeps the same, which indicates the spectral components are enhanced equally.

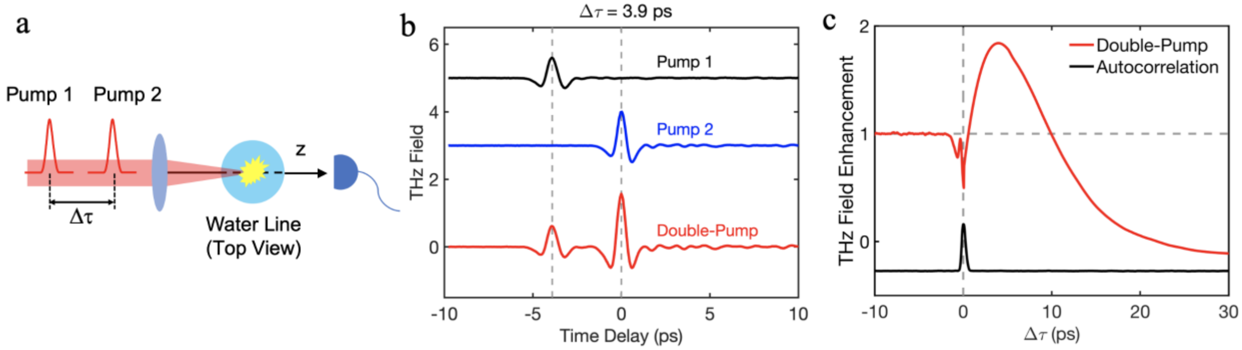


Figure 13. THz generation under double-pump excitation. [79] **a** Schematic of double pump excitation. Two optical pumps propagating collinearly with a certain time delay Δτ are focused on a water line (top view). Δτ is controlled by a delay stage. The THz signal is detected through EOS. **b** THz waveforms generated by pump 1 (black line) and pump 2 (blue line), respectively. The bottom red line shows the waveform generated by two pumps when Δτ is 3.9 ps. **c** The red line shows the dynamic of the THz peak field versus Δτ. The black line plots the autocorrelation signal of the two pumps, from which the 0 timing is determined. Δτ > 0 means that the pump 1 ionizes the target first.

The temporal dependence of the THz field enhancement on the time delay is shown in **Figure 13c**. By tuning Δτ, the peak field of the THz waveform generated by the pump 2 is recorded. The zero timing of Δτ is determined by the peak of the autocorrelation signal (black line) of two pumps. The enhancement dynamic (red line) results from either the interaction of two plasmas produced by two pumps, or the influence of the first-produced plasma on the pump 2. It could not be the direct interaction of two beams due to the enhancement happens in a picosecond time scale, which is a much longer than that of two pulses interaction (~ 100 fs). As shown in the plot, when Δτ < 0, the pump 2 arrives at the water line first and generates a THz signal. The second pump comes later and has no influence on it. Therefore, the signal keeps the same peak value (normalized to 1) until pump 1 catches up pump 2. The signal starts to change when two pumps temporally overlap with each other. The enhancement gradually increases until reaches the maximum. A maximum enhancement of 1.8 times in field is observed in this case. Additionally, the enhancement lasts a long time. It is noteworthy that the time delay supporting the enhancement is over 10 ps. Eventually, the THz field decreases because of the absorption of the first plasma. More details can be found in Ref. [79]. For the double-pump excitation, one possible reason of the enhancement is that the pre-existing plasma provides more initial electrons for the cascade ionization. Under single-color excitation, the multiphoton or tunneling ionization processes give rise to the initial ionization of target at the front of the pulse. Instead, under double-pump excitation, the pre-plasma generated by the first pulse provides a higher electron density.

2.10 Ultrafast dynamics in liquids excited by intense THz waves

When a strong enough electric field is applied to an isotropic media, birefringence is created, which is well-known as the Kerr effect. [81, 82] As the development of THz sources in recent decades, it was realized that the intense THz pulse induces transient Kerr effect, as known as THz Kerr effect (TKE). [83] This finding offers a new method of investigating the ultrafast evolution of Kerr effect in picosecond timescale. The hydrogen bond vibrations as well as other molecular motions in aqueous solutions could be characterized in the THz range, which play an important role in understanding the thermodynamic properties of liquids. [84] By employing the technique of THz Kerr effect, the low-frequency molecular dynamics is able to be studied experimentally.

Figure 14a shows the diagram of a THz Kerr effect setup. An intense THz pulse with the maximum peak electric field strength of 14.9 MV/cm is focused on a gravity-driven, free-sanding liquid film. The liquid samples could be pure water or aqueous salt solutions with different concentrations. One optical beam colinearly propagates with THz beam is measured in a balanced photodetector. The THz induced birefringence is recorded by measuring the polarization change of the optical beam. **Figure 14b** shows a bipolar THz Kerr effect response with significant oscillation characteristics in liquid water. The response linearly scales with the peak-intensity of THz pulse, indicating that the Kerr effect is dominant here. The broadband THz pulse covers two modes of intermolecular motions, which are hydrogen bond bending and stretching vibrations, respectively. The positive signal caused by hydrogen bond stretching vibration and the negative signal caused by hydrogen bond bending vibration, indicating that the polarizability perturbation of water presents competing contributions under bending and stretching conditions. More details can be found in the Ref. [85].

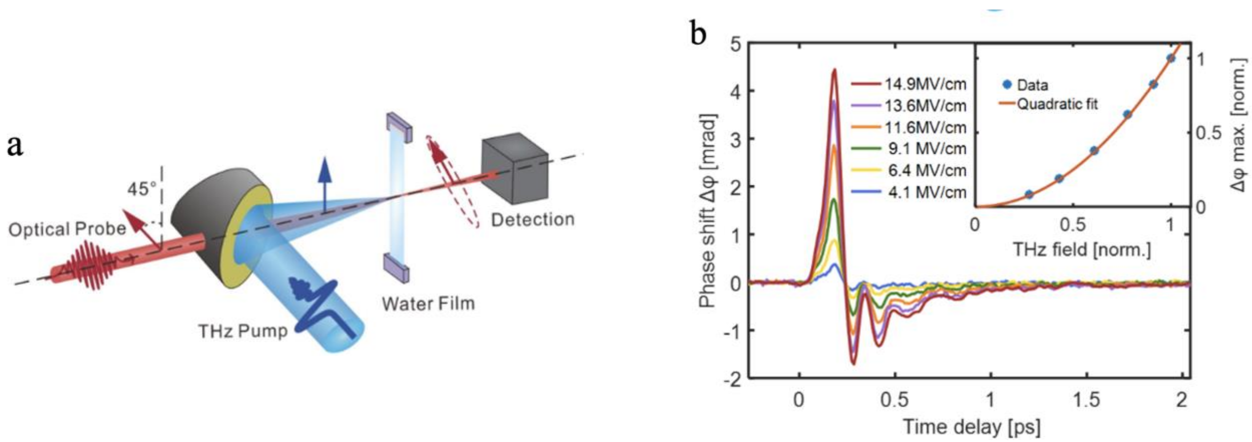


Figure 14. Terahertz Kerr effect (TKE) in a water film. [85] **a.** Diagram of the experimental setup. An intense THz pulse is focused onto a free-standing water film. The THz field induced Kerr effect is detected though an optical probe. **b.** TKE with different THz peak field strengths. Inset: THz field strength dependence.

To simulate the molecular motions, the Lorentz dynamic equation is adopted in the model, which is based on the perturbation in the dielectric tensor caused by the intermolecular vibration modes. The resulting polarizability anisotropy can be expressed by the refractive index associated with the dielectric susceptibility: [85, 86]

$$\sum_{i=1,2}\Delta n_i = \frac{1}{2\varepsilon_0 n_0}\left[\varepsilon_y - \frac{1}{2}(\varepsilon_x + \varepsilon_z)\right] = \frac{1}{2n_0}\left(\frac{\partial \chi_{\parallel}}{\partial Q_s}q_2 - \frac{\partial \chi_{\perp}}{\partial Q_b}q_1\right) \tag{1}$$

where ε_0 is the vacuum dielectric constant and n_0 is the refractive index of liquid water. q_1 and q_2 represent the anisotropic perturbations caused by hydrogen bond bending and stretching vibrations, respectively. Q_b and Q_s represent the bending and stretching vibration amplitudes of a single hydrogen bond unit, respectively. $\chi_{\parallel}$ and $\chi_{\perp}$ represent the perturbations of the dielectric susceptibilities parallel and perpendicular to the hydrogen bond direction caused by the intermolecular modes. These two intermolecular motions result in opposite birefringence contributions to the total refractive index, i.e., $\Delta n_1 < 0$, $\Delta n_2 > 0$.

In addition, q_i satisfies the Lorentz dynamic model, which describes the motion of the damped harmonic oscillator: [87, 88]

$$\frac{\partial^2 q_i(t)}{\partial t^2} + \gamma_i \frac{\partial q_i(t)}{\partial t} + \omega_i^2 q_i(t) = a_i E^2(t) \quad \text{(i=1, 2)} \tag{2}$$

where γ_i, ω_i represent the damping coefficient and inherent frequency, respectively, with values of $\gamma_1 = 115\ cm^{-1}$, $\gamma_2 = 165\ cm^{-1}$, $\omega_1 = 60\ cm^{-1}$, and $\omega_2 = 190\ cm^{-1}$. Here, $a_i = \beta_i \frac{\mu_0^2}{3\sqrt{2}k_B Tm}$ represents the coupling factor between the square of the THz pump electric field and the driving term in **Equation (2).** We use the above model to decompose and simulate the experimental data in the time domain, as shown in **Figure 15**. The data can be decomposed into (i) positive responses with electronic (Electr.) and hydrogen bond stretching (Stretch.) contributions and (ii) negative responses with Debye relaxation (Debye) and hydrogen bond bending (Bend) contributions.

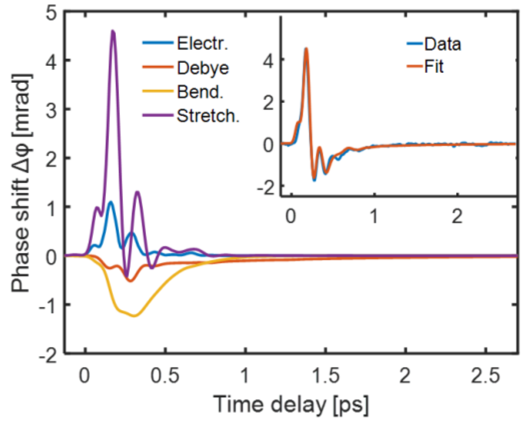


Figure 15. Simulation of the TKE response. [85] The simulation results of the TKE response in the time domain for the electronic (Electr.), Debye relaxation (Debye), hydrogen bond bending vibration (Bend) and hydrogen bond stretching vibration (Stretch.) contributions. Inset: a comparison between the sum of all the contributions and the measured data.

For ionic aqueous solutions, the TKE signals of NaCl, NaBr and NaI solutions with different concentrations are measured, as shown in **Figure 16a**. Accordingly, the amplitude and shape of the THz Kerr effect responses varies with exchanging anions when the counter-ion is Na^+. These signals are also theoretically decomposed into the weak electronic responses and different molecular motion modes, [89] as show in **Figure 16b**.

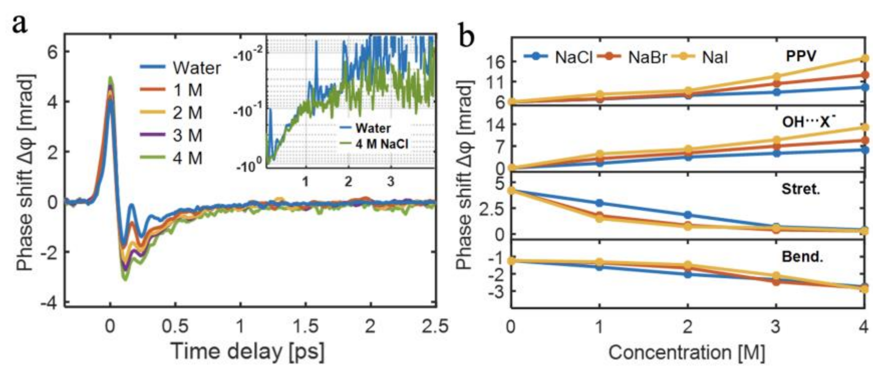


Figure 16. Terahertz Kerr effect response on different concentrations. [89] **a** TKE responses of water and NaCl solutions with different concentrations. Inset: an enlarged logarithmic view of the negative polarity responses. **b** The upper panel shows the measured peak-to-peak values (PPV) of TKE responses for NaCl, NaBr, NaI solutions with different concentrations. The other panels show the extracted maximum values of TKE responses induced by $OH\cdots X^-$ hydrogen bond vibration ($OH\cdots X^-$), the hydrogen bond bending (Bend.), and hydrogen bond stretching modes (Stret.).

For the bending mode, the amplitude of negative polarity response gradually increases as the concentration increases. However, the damping coefficient and inherent frequency do not change significantly in this over-damped oscillation system. For the stretching mode, the positive polarity response gradually decreases along with the weakening of oscillation characteristics as the concentration increases, because the addition of ions dilutes the hydrogen bond density and destroys the water network structure. However, as the concentration increases, the positive polarity response increases, indicating that the interaction of ions with water provides a larger positive polarity response than that from water-water hydrogen bond stretching mode. The enhanced positive response origins from the special hydrogen bond vibration formed between the anions and the water molecules ($OH…X^-$, X^- : Cl^-, Br^-, I^-), [90, 91] and the addition of anions will greatly reduce the water-water hydrogen bond vibration. As the ion concentration increases, the low-frequency molecular motion in water is gradually dominated by ion-water hydrogen bond motion.

For the $OH…X^-$ hydrogen bond vibration mode, the THz Kerr effect response increases significantly with the increase of concentration for all the three kinds of solutions. In particular, the NaI solution exhibits higher anisotropy response at the same concentration. This is because for the low surface charge density of anion (such as I^-), the formed OH…X^- hydrogen bond has relatively long bond length and low binding energy. [91] In addition, the center frequency of 3.9 THz of the THz pulse is closer to the inherent frequency of the OH…I^- hydrogen bond vibration. Therefore, an enhanced dipole moment and a greater polarizability anisotropy under OH…I^- hydrogen bond vibration mode could be achieved with the resonant THz electric field excitation.

The THz Kerr effect measurement directly observes the ultrafast temporal evolution of intermolecular hydrogen bond dynamics in aqueous solutions at the sub-picosecond scale. The THz Kerr effect technique helps us to explore the low-frequency dynamics and successfully obtain the THz Kerr effect responses in various aqueous solutions, such as pure water and halide anion aqueous solutions. The results provide an experimental basis for further insight into the ultrafast evolution of transient structures in complex aqueous solutions. We believe that THz Kerr effect method can also be applied to detect the physical mechanisms of the complex interaction of reagents with solvent water molecules, which can provide the further insight into the effects of hydrogen bond network on the biochemical reaction and the related structural dynamics in water environment.

2.11 Outlook

Currently, the THz field generated form liquid under single color excitation is 2 orders higher than that from single-color air plasma, which is comparable to the signal from the two-color air plasma. A detailed comparison between THz generation from air and water is shown in **Table 4**. Please note, this comparison is only valid under certain experimental conditions: 2-inch focal length lens, 0.4 mJ pulse energy, 60 fs Fourier transform limited pulse. Broadband THz wave generation from liquid is a very recent research topic, detailed systematic theoretical and experimental studies are needed. The optimal conditions for the maximum generation efficiency are underexplored. Some work can be done in target geometry design, laser pulse shaping, target material testing, external field modulating and so on.

Table 4. Comparison between THz generation from air and water.

	Air	Water
Optimal optical pulse duration	Shortest	Sub picosecond
Central THz frequency	1~1.5 THz	0.5~1 THz
THz signal bandwidth	1~1.5 THz	0.5~1 THz
Two-color enhancement in power	$\sim10^6$	$\sim10^2$
Fluorescence and white light	Relatively week	Very brighter
Sideway signal (2 in lens is used)	No measurable signal	Strong sideway signal

Furthermore, it would be interesting to study THz generation from superfluid such as ^{4}He, which flows with no viscosity, and its thermal conductivity increases by a factor of $\sim10^6$ for the temperature at the λ-point. This causes heat in the body of the liquid to be transferred to its surface so quickly that vaporization takes place only at the free surface of the liquid. Thus, there are no gas bubbles in the body of the superfluid ^{4}He. Therefore, the superfluid ^{4}He is a perfect system for the optical study on the laser-matter interaction by focusing laser beam in the liquid. THz liquid photonics provides a new platform in the THz community, which offers an opportunity for developing liquid THz sources and exploring the light-liquid interaction. THz wave generation from liquids presents its specialties and superiorities compared to the gas or solid targets, which should be further studied to help plot a thorough picture for the laser-induced ionization process in liquid. All efforts in this topic will help pave the way toward developing intense liquid THz sources and offer a better insight into the laser-liquid interaction process.

3. Services during the Project Period

Community Service

2018 - 2020	Member, Optical Society Nominating Council.
2019	General Co-Chair, POEM Conference, Wuhan, China, Nov. 2019
2-19	Co-Chair, SPIE/COS Asia Photonics, Hangzhou, China, Oct. 2019
2019	Co-Chair, CIOP, Xi'an, China, Aug. 2019
2020 – 2022	Executive Editor-in-Chief, Light, Science and Applications, A journal with Nature Springer.
2020 – 2022	Advisor, Ultrafast Science Journal. A new journal with SPIE.
2021	Chair, Technical Program Committee, IRMMW-THz annual conference.
2021	Consultant, Materia Medica Holding, Russia. Approved by University of Rochester. The consulting fees have been scheduled directly to Universities to support graduate students.
2021	General Chair, 10th International Symposium on Ultrafast Phenomena and Terahertz Waves (ISUPTW 2021)
2021	Technical Program Chair, The 12th International Conference on Information Optics and Photonics (CIOP2021)

4. Honors and Awards during the Project Period

2018	Humboldt Prize, Alexander von Humboldt Foundation, Germany
2018	Visiting Chair Professor, National Taiwan University, Taiwan
2018	Visiting Chair Professor, National Chiao-Tung University, Taiwan

During my time working as the Editor-in-Chief of Optics Letters, I did not publish my own research papers in Optics Letters to avoid potential conflict of interest. This decision was my own choice since the first day I served as Editor-in-Chief of Optics Letters. However, I have published 8 editorials in Optics Letters during the period of 2014 - 2019. **Figure 17a** is a photo of completion of PI's final term as the Editor-in-Chief.

a b c

Figure 17. a Receiving appreciation for the service as Editor-in-Chief of Optics Letters (2019). **b** Receiving the Humboldt Prize, AvH Foundation (2018). **c** Meeting the President of Germany Frank-Walter Steinmeier during the Humboldt Prize reception (2018).

The PI received Alexander von Humboldt Prize in 2018. **Figure 17b** and **17c** show the honorable events of receiving the Humboldt Prize certificate and reception in Berlin. During the pandemic, the PI gave a popular talk: "5 minutes in optics" online in summer 2020. Over 33,000 audiences worldwide attended my talk. The PI currently serves as Executive Editor-in-Chief of Light, Science and Application, Nature Springer.

5. Dissemination during the Project Period

Papers Published and Submitted During This Period

1. X.-C. Zhang and Fabrizio Buccheri, “THz Photonics of Micro-Plasma and Beyond,” Lithuanian Journal of Physics, Vol58, pp1-14, (2018). DOI: https://doi.org/10.3952/physics.v58i1.3647
2. Fabrizio Buccheri, Pingjie Huang, and X.-C. Zhang, “Generation and Detection of Pulsed Terahertz Waves in Gas: from Elongated Plasmas to Microplasmas,” Invited paper, Frontiers of Optoelectronics, (2018). doi.org/10.1007/s12200-018-0819-8.
3. Y. E, Q. Jin, A. Tcypkin, and X.-C. Zhang, “Terahertz Wave Generation from a Liquid Water Film via Laser-Induced Breakdown,” Appl. Phys. Lett. 113, 181103 (2018); doi: http://dx.doi.org/10.1063/1.5054599.
4. Q. Jin, J.M. Dai, Y. E, and X.-C. Zhang, “Terahertz wave emission from a liquid water film under the excitation of asymmetric optical fields,” Appl. Phys. Lett., (2018). SciLight feature article.
5. Kang Liu, Pingjie Huang, and X.-C. Zhang, “Terahertz Wave Generation from Ring-Airy Beam Induced Plasmas and Remote Detection by Terahertz- Radiation- Enhanced- Emission- of- Fluorescence: A Review,” Invited paper, Frontiers of Optoelectronics, (2019). 10.1007/s12200-018-0860-7.
6. A.N. Tcypkin, M.V. Melmik, M.O. Zhukova, I.O. Vorontsova, S.E. Putilin, S.A. Kozlov, and X.-C. Zhang, “High Kerr nonlinearity of water in THz spectral range,” Optical Express. Vol. 27, Issue 8, pp. 10419-10425 (2019). https://doi.org/10.1364/OE.27.010419
7. L.-L. Zhang, W.-M. Wang, T. Wu, S.-J. Feng, K. Kang, C.-L. Zhang, Y. Zhang, Y.-T. Li, X.-Q. Yan, Z.-M. Sheng, and X.-C. Zhang, “Strong Terahertz Radiation from a Liquid-Water Line,” Phys. Rev. Applied, Phys. Rev. Applied vol. 12, 014005, (2019). DOI: 10.1103/PhysRevApplied.12.014005
8. A.N. Tcypkin, E.A. Ponomareva, S.E. Putilin, S.V. Smirnov, S.A. Shtumpf, M.V. Melnik, Y. E, S.A. Kozlov and X.-C. Zhang, “Flat liquid jet as a highly efficient source of terahertz radiation,” Optics Express, vol. 27, 15485-15494, (2019). https://doi.org/10.1364/OE.27.015485
9. Jiapeng Zhao, Yiwen E, Kaia Williams, Xi-Cheng Zhang, and Robert Boyd, “Spatial Sampling of Terahertz Fields with Sub-wavelength Accuracy via Probe-Beam Encoding,” Light: Science & Applications 2047-7538, (2019). https://doi.org/10.1038/s41377-019-0166-6
10. Y.W. E, Q. Jin, and X.-C. Zhang, “Enhancement of terahertz emission by a preformed plasma in liquid water,” Applied Physics Letters, 115, 101101 (2019). https://doi.org/10.1063/1.5119812. DOI: 10.1063/1.5119812.
11. Evgenia A. Ponomareva, Anton N. Tcypkin, Semen V. Smirnov, Sergey E. Putilin, E Yiwen, Sergei A. Kozlov, and Xi-Cheng Zhang, “Double-pump technique − one step closer towards efficient liquid-based THz sources” Opt. Express 27(22), 32855-32862 (2019). https://doi.org/10.1364/OE.27.032855
12. Si-Chao Chen, Zheng Feng, Jiang Li, Wei Tan, Liang-Hui Du, Jianwang Cai, Yuncan Ma, Kang He, Haifeng Ding, Zhao-Hui Zhai, Ze-Ren Li, Cheng-Wei Qiu, Xi-Cheng Zhang & Li-Guo Zhu, "Ghost spintronic THz-emitter-array microscope," Light: Science & Applications, 9, 99 (2020). https://doi.org/10.1038/s41377-020-0338-4
13. Hang Zhao, Yong Tan, Liangliang Zhang, Rui Zhang, Mostafa Shalaby, Cunlin Zhang, Yuejin Zhao & Xi-Cheng Zhang, "Ultrafast Hydrogen-Bond Dynamics of Liquid Water Revealed by Terahertz-induced Transient Birefringence" by Light: Science & Applications 9:136 (2020). https://doi.org/10.1038/s41377-020-00370-z
14. Q. Jin, Y.W. E, S.H. Gao, and X.-C. Zhang, “Preference of subpicosecond laser pulses for terahertz wave generation from liquids,” Advanced Photonics, 2(1), 015001-1, (2020). https://doi.org/10.1117/1.AP.2.1.015001
15. Yan Peng, Bowei Xu, Shiwei Zhou, Zhaozhao Sun, Haicheng Xiao, Jiayu Zhao, Yiming Zhu, Xi-Cheng Zhang, Daniel Mittleman, Songlin Zhuang, "Experimental measurement of the wake field in a plasma filament created by a single-color ultrafast laser pulse", Physical Review E, 102, 063211 (2020). https://doi.org/10.1103/PhysRevE.102.063211
16. Yuqi Cao, Yiwen E, Pingjie Huang, and X.-C. Zhang, “Broadband Terahertz Wave Emission from Liquid Metal,” Applied Physics Letters, (2020). Appl. Phys. Lett. 117, 041107 (2020). https://doi.org/10.1063/5.0015507

17. Kareem J. Garriga Francis, Mervin Lim Pac Chong, Yiwen E, and X.-C. Zhang, "Terahertz Nonlinear Index Extraction via Full-Phase Analysis," Optics Letters, Vol. 45, 5628-5631 (2020). https://doi.org/10.1364/OL.399999
18. Yiwen E, Yuqi Cao, Fang Ling, and X.-C. Zhang, "Flowing cryogenic liquid target for terahertz wave generation," AIP Advances, 09 October (2020). https://doi.org/10.1063/5.0023106
19. Xinrui Liu, Maksim Kulya, Nikolay Petrov, Yaroslav Grachev, Mingzhao Song, Anton Tcypkin, Sergey Kozlov, and Xi-Cheng Zhang, "Spectral Fresnel filter for pulsed broadband terahertz radiation," AIP Advances 10, 125104 (2020); https://doi.org/10.1063/5.0024456
20. Chen Xie, Yehao Ma, Yiwen E, Pingjie Huang, Xi-Cheng Zhang, "THz spectroscopic decomposition and analysis in mixture inspection using soft modeling methods," Journal of Infrared, Millimeter, and Terahertz Waves. (2020). https://doi.org/10.1007/s10762-020-00755-6
21. Q. Jin, Y. E, and X.-C. Zhang, "THz aqueous photonics," Frontiers of Optoelectronics, 14(1): 37–63 (2021). https://doi.org/10.1007/s12200-020-1070-7
22. Evgenia Ponomareva, Azat Ismagilov, Sergey Putilin, Anton Tsypkin, Sergei Kozlov, and Xi-Cheng Zhang, "Varying pre-plasma properties to boost terahertz wave generation in liquids," Communications Physics. (2020). COMMSPHYS-20-0530B. Commun Phys 4, 4 (2021). https://doi.org/10.1038/s42005-020-00511-1.
23. Na Zhang, Jia-Sheng Ye, Sheng-Fei Feng, Xin-Ke Wang, Peng Han, Wen-Feng Sun, Yan, Zhang, X.-C. Zhang, "Generation of long-distance stably propagating Bessel beams," OSA Continuum, (2021). https://doi.org/10.1364/OSAC.420125
24. Yan Peng, Jieli Huang, Jie Luo, Zhangfan Yang, Liping Wang, Xu Wu, Xiaofei Zang, Chen Yu, Min Gu, Qing Hu, Xicheng Zhang, Yiming Zhu, Songlin Zhuang, "Three-step one-way model in terahertz biomedical detection, " PhotoniX, (2021). Pending. PHOX-D-21-00016R1.
25. Jiapeng Zhao, Jianming Dai, Boris Braverman, Xi-Cheng Zhang, and Robert W. Boyd, "Compressive ultrafast sensing enabled by programmable temporal fan-out," Optica. (2021). Submitted.
26. Kareem Garriga Francis, Yuqi Cao, Yiwen E, Fang Ling, Mervin Lim Pac Chong, Xi-Cheng Zhang, "Forward THz Wave Generation from Liquid Gallium in the Non-relativistic Regime", Optics Letters, (2020). Under review.
27. Anil Malik, Bhati, Ruchi and X.-C. Zhang, "Broadband, Ultrasensitive and Ultrahigh Q- Factor Metamaterial Terahertz sensor," APC Photonics. (2021). Pending.
28. Y. E, L. Zhang, A. Tcypkin, S. Kozlov, C. Zhang, X.-C. Zhang, "Broadband THz sources from gases to liquids'', Ultrafast Science, 2021. https://downloads.spj.sciencemag.org/ultrafastscience/aip/9892763.pdf

6. Invited Conference Presentations and Seminars During This Period

1. "THz aqueous photonics," ASILS10, American University of Shajah, Shahja, March 11, 2018.
2. "THz aqueous photonics," X.-C. Zhang, Plenary talk, 18th National Optical Fiber Communication and 19th Integrated Optics Conference, Changchun, China, July 14, 2018.
3. "Let's THz light shine out of darkness," X.-C. Zhang, Plenary talk, Light Conference 2018, Changchun, China, July 18, 2018.
4. "THz Aqueous Photonics and Beyond," Q. Jin, Y.W. E, L.L. Zhang, C.L. Zhang, A. Tcypkin, S. Kozlov, X.-C. Zhang, Plenary talk, IRMMW-THz Conference, Nagoya, Japan, Sept 10, 2018.
5. "Generation of intnse and coherent THz wave from liquid water," X.-C. Zhang, Q. Jin, Y.W. E, L.L. Zhang, C.L. Zhang, A. Tcypkin, S. Kozlov, SPIE Asia Photonics, Beijing, Oct. 12, 2018.
6. "Pulse duration dependence of THz generation in water and ethanol," S.E. Putilin, S.A. Stumpf, S.V. Smirnov, Y. E, M.V. Melnik, E.A. Ponomareva, V.G. Bespalov, S. A. Lozlov, X.-C. Zhang, SPIE Asia Photonics, Beijing, Oct. 12, 2018.
7. "Mission impossible: generation of THz wave from liquid water," X.-C. Zhang, Westlake Forum, Hangzhou, China, Oct. 26, 2018.
8. "THz Aqueous Photonics and Beyond," French-German THz conference, Kaiserslautern, Germany. April 2-5, 2019.

9. "Mission Impossible: Generation of THz wave from liquid water," Optics and Photonics 2019, Dubai, AUE. April 15, 2019.
10. "Next Rays? T-ray!, part I" Siegman International School on Lasers, Rochester, July 27, 2019.
11. "Next Rays? T-ray!, part II" Siegman International School on Lasers, Rochester, July 31, 2019.
12. "Challenges and opportunities for THz wave liquid photonics," Tutorial, International Conference on Information Optics and Photonics, Xian, China, Aug. 7, 2019
13. "Enhanced emission of terahertz wave from liquid water," Yiwen E, Qi Jin, X.-C. Zhang, SPIE/COS Photonics Asia, Hangzhou, China, Oct. 21, 2019. [11198-6]
14. Yuqi Cao, Yiwen E, Pingjie Huang, and X.-C. Zhang, "Liquid Metal for Terahertz Wave Emission," PA20-PA116-21, SPIE/COS Asia Photonics, Beijing, China. Oct. 11-13, 2020.
15. Yiwen E, Qi Jin, Jianming Dai, Yuqi Cao, Fang Ling, Kaia Williams, Mervin Lim Pac Chong, Gregoire Leir, Kareem Garriga Francis, Anton Tcypkin, LiangLiang Zhang, Cunlin Zhang, and X.-C. Zhang, "THz Liquid Photonics and Beyond", PA20-PA116-32, SPIE/COS Asia Photonics, Beijing, China. Oct. 11-13, 2020.

7. Contributed Conference Presentations during This Period

1. J.M. Dai, Q. Jin, Y.W. E, K. Williams, X.-C. Zhang, "Using liquid water as broadband terahertz wave emitter," SPIE Commercial + Scientific Sensing and Imaging, SI18C-SI103-22 (2018).
2. Artur Gleim, Vladimir Egorov, Vladimir Chistiakov, Artur Vasiliev, Andrei Gaidash, Anton Kozubov, Semen Smirnov, Sergei Kynev, Nikita Buldakov, Oleg Sadov, Andrey Shevel, Sergei Khoruzhnikov, Oleg Bannik, Lenar Gilyazov, Konstantin Melnik, Narkis Arslanov, Ilnur Latypov, Sergei Moiseev, Xi-Cheng Zhang and Sergei Kozlov, "Scalable QKD networks based on subcarrier wave architecture and software-defined paradigm: ITMO University initiative in Russian quantum networking." QCrypt2018, Shanghai, China (2018)
3. "Features of few-cycle THz pulses nonlinear refraction, A.N. Tspkin, S.E. Putilin, M.S. Kylya, M.V. Melnik, M.A. Kniazev, A.A. Drozdov, I.O. Vorotsova, V.G. Bespalov, X,-C. Zhang, S.A. Kozlov, SPIE Photonics Asia, Beijing, Oct. 12, 2018.
4. "THz Photonics using Liquids," Q. Jin, Y.W. E, J.M. Dai, L.L. Zhang, C.L. Zhang, A. Tcypkin, S. Kozlov, X.-C. Zhang, OSA Nonlinear Optics. Hawaii, July 2019.
5. "Double pulse excitation for enhancing THz generation in liquid jets," IRMMW-THz 2019, Paris, Fence, Sept. 2019.
6. "THz Aqueous Photonics and Beyond," IRMMW-THz 2019, Paris, Fence, Sept. 2019.
7. "Spatial Sampling of Terahertz Fields with Sub-wavelength Accuracy via Probe Beam Encoding", J.P. Zhao, B. Boyd, X.-C. Zhang, SPIE OPOT, Hangzhou, China, Nov. 24, 2019.
8. Anton Tsypkin, Evgenia Ponomarava, Sergei Putilin, Yiwen E, Sergei Kozlov, X.-C. Zhang, "Comparison of various liquids as sources of terahertz radiation from one-color laser filament" SPIE/COS Photonics Asia, Beijing, Oct. 21, 2019. [11198-7]
9. Shenghan Gao, Qi Jin, Yiwen E, X.-C. Zhang, "investigation of different liquid properties on emitting THz wave under ultrashort optical excitation," SPIE/COS Photonics Asia, Beijing, Oct. 21, 2019. [11198-8]
10. Shenghan Gao, Zeqing Jin, Yiwen E, X.-C. Zhang, "Fabrication and testing of the smallest flute on syringe needles," SPIE/COS Photonics Asia, Beijing, Oct. 21, 2019.
11. Jiapeng Zhao, Yiwen E, Kaia Williams, X.-C. Zhang, and Robert W. Boyd, "Spatial Sampling of Terahertz Fields with Sub-wavelength Accuracy via Probe Beam Encoding," SPIE Photonic West, San Francisco, Jan. 28, 2020.
12. Jiapeng Zhao, Yiwen E, Kaia Williams, Xi-Cheng Zhang, Robert W. Boyd, "Spatial Measurement Of Terahertz Fields By Encoding Probe Beam", 45th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz) 2020.
13. Kareem Garriga, M. Chong, Yiwen E, and X.-C. Zhang, "Nonlinear spectroscopy at terahertz frequencies", PA20-PA116-47 SPIE/COS Asia Photonics, Beijing, China.

14. Fang Ling, Yiwen E, Kareem J. Garriga Francis, Bin Zhang, X.-C. Zhang, "Influences of gold nanoparticles on terahertz generation from water," PA20-PA116-45, SPIE/COS Asia Photonics, Beijing, China. Oct. 11-13, 2020.
15. Yiwen E, Yuqi Cao, Fang Ling, Alexander P. Shkurinov, Yiming Zhu, and X.-C. Zhang, "High-repetition-rate, cryogenic liquid target for terahertz wave generation," PA20-PA116-61 SPIE/COS Asia Photonics, Beijing, China. Oct. 11-13, 2020.
16. Yuqi Cao, Yiwen E, Anton Tcypkin, Pingjie Huang, and X.-C. Zhang, "Terahertz Wave Generation at Different Water Temperatures," 45th IRMMW-THz 2020, Buffalo, NY, USA. Nov. 8-13, 2020.
17. Yiwen E, Yuqi Cao, Fang Ling, Alexander P. Shkurinov, Yiming Zhu, and X.-C. Zhang, "Broadband THz Wave Generation From Flowing Liquid Nitrogen," 45th IRMMW-THz 2020, Buffalo, NY, USA. Nov. 8-13, 2020.
18. Kareem Garriga, Mervin Chong, Yiwen E, and X.-C. Zhang, "THz Nonlinear Dielectrics," 45th IRMMW-THz 2020, Buffalo, NY, USA. Nov. 8-13, 2020.
19. Yiwen E., Qi Jin, Jianming Dai, Yuqi Cao, Fang Ling, Kaia Williams, Mervin Lin Pac Chong, Gregoire Leir, Kareem J. Garriga Francis, Anton N. Tcypkin, Liangliang Zhang, Cunlin Zhang, and Xicheng Zhang "THz liquid photonics and beyond", 1155902, SPIE/COS Asia Photonics, Beijing, China, 2020.

8. PhD students graduated during this period

Ms. Kang Liu, Ph.D. 2018; Thesis topic: Tailoring air-plasma toward advanced THz pulses generation and detection. Current affiliation: Apple (San Jose).

Mr. Qi Jin, Ph.D. 2019; Thesis topic: Terahertz Aqueous Photonics. Xerox (Webster, NY)

9. Research Personnel supported by this program at University of Rochester

Prof. X.-C. Zhang (Principal investigator)

Dr. Yiwen E (postdoc/research associate)

Dr. Jin Qi (now works at Xerox)

Mr. Kareem Garriga, Ph.D. candidate.* (2021 summer under Apple internship)

Mr. Steven Fu, Ph.D. candidate. * (Hong Kong student)

* passed Ph.D. qualify test and candidacy test*.

10. International Collaboration through Visiting Students and Scholars

1. Dr. Yiwen E, Institute of Physics, Chinese Academy of Science, China, 1/12/17 – present
2. Dr. Pingjie Huang, Zhejiang University, China, 9/30/17 – 9/29/19
3. Dr. Takeshi Moriyasu, University of Fukui, Japan, 8/15/18 – 3/31/19
4. Ms. Yuqi Cao, Zhejiang University, 7/1/18 – 11/1/18; and 10/27/19 – 10/26/20
5. Ms. Yuelin Chen, Sun Yat-sen University, 7/15/2019 - 9/6/2019
6. Ms. Fang Ling, Sichuang University, 11/13/2019 - 7/4/2021

11. Personnel Supported

Professor X.-C. Zhang received summer salary support under this grant. Dr. Yiwen E and two PhD students received financial support. Kareem Garriga and Steven Fu were supported for their doctoral degrees. We have joined international collaborations with Japan, Russian and China on joint conference presentations and journal papers, but there is NO financial support to international collaborators using our government grants. **Figure 18** is the screen shot of Zhang's group meeting on June 11, 2021.

Figure 18 Screen shot of Zhang's group meeting. Mervin Lim (MS student), Fang Ling (visiting scholar), Gerrit Bruhaug (PhD student), Yiwen E (Research Associate), X.-C. Zhang (PI), Justin Murante (undergraduate student), Kelsely Lee (undergraduate student), Steven Fu (PhD student), and Kareem Garriga (PhD student).

Name: Total Number of Months: Project role: Contribution to Project: State, U.S. territory, and/or country of residence: Collaborated with individual in foreign country: Country(ies) of foreign collaborator: Travelled to foreign country: If traveled to foreign country(ies), duration of stay:	X.-C. Zhang 2.74 months PI Lead the group to conduct the research project. NY, USA Yes Australia, Japan, Germany, Russia, China Travel to many countries, but not funded by this AFOSR grant.

Name: Total Number of Months: Project Role: Contribution to Project: State, U.S. territory, and/or country of residence: Collaborated with individual in foreign country: Country(ies) of foreign collaborator: Travelled to foreign country: If traveled to foreign country(ies), duration of stay:	Dr. Yiwen E 18.00 months Postdoc/Res Associate Dr. E has performed research on THz photonics in water. She also participated in an experiment at the LLE NY, USA

Name: Total Number of Months: Project role: Contribution to Project: State, U.S. territory, and/or country of residence: Collaborated with individual in foreign country:	Qi Jin 11.00 months PhD Graduate Student Dr. Jin has performed research on THz photonics in water. NY, USA

Country(ies) of foreign collaborator: Travelled to foreign country: If traveled to foreign country(ies), duration of stay:	

Name: Total Number of Months: Project role: Contribution to Project: State, U.S. territory, and/or country of residence: Collaborated with individual in foreign country: Country(ies) of foreign collaborator: Travelled to foreign country: If traveled to foreign country(ies), duration of stay:	Kareem Garriga 17.00 months PhD Graduate Student Mr. Garriga has performed research on THz photonics in water. He also participated in an experiment at the LLE NY, USA

Name: Total Number of Months: Project role: Contribution to Project: State, U.S. territory, and/or country of residence: Collaborated with individual in foreign country: Country(ies) of foreign collaborator: Travelled to foreign country: If traveled to foreign country(ies), duration of stay:	Steven Fu 4.00 months PhD Graduate Student Mr. Fu has performed research on THz photonics in liquid. NY, USA

Has there been a change in the active other support of the PD/PI(s) or senior/key personnel since the last reporting period? Nothing to report

What other organizations have been involved as partners? During my one-year sabbatical, I have studied in and visited ten countries: Australia (one month), China (one month), Japan, Italy, New Zealand, United Arab Emirates, Russia, Taiwan (one month), Mexico, and Germany (one month). I met a milestone of 3 million traveling miles and maintained Global Service status in the MileagePlus Program with United Airlines. In the midst of all this, two PhD students graduated. I have been named as the Australian Academy of Science Selby Fellow and received the Humboldt Prize and National Taiwan University Chair Professorship I had the honor to meet the President of Germany and the President of the Humboldt Foundation during the reception. There was not any financial support from this AFOSR grant involved in this collaboration. I have disclosed and contacted the Program Manager (Gernot Pomrenke) for the joint publication with Russian and Chinese team (email).

Have other collaborators or contacts been involved? "Nothing to Report."

12. What was the impact of the project?

Matter has four states, solid, liquid, gas, and plasma. THz wave generation from solids, gases, and plasmas has been well studied for decades. However, there was no demonstration of THz emission from a liquid source previously, especially from liquid water due to its infamously strong absorption characteristics in the THz regime. Recently we have observed the generation of broadband THz waves from liquid water by tightly focusing laser pulses into a thin water film (about 100 - 200 um).

Laser-induced ionization in matter occurs when a laser pulse with sufficient intensity is focused into a target material, creating electrons and ions through nonlinear processes of laser-matter interaction. Photoionization has attracted considerable interest for generating intense, broadband waves in the terahertz (THz) portion of the electromagnetic spectrum. THz pulse energy up to millijoules has been observed by using an intense laser pulse to irradiate a metal foil.

In the THz frequency range, laser-induced air plasma has become one of the most popular THz sources in the research laboratories. It can generate THz waves with a field strength of over MV/cm level by two-color optical excitation. The transient field is strong enough to induce nonlinearity. Recently, THz wave generation from ionized liquids, especially from liquid water, has also been successfully demonstrated. Liquids are preferable to general targets, as a flowing liquid line provides a fresh area for each excitation pulse, so the chaos caused by the previous pulse will not influence the next one. This makes it possible to use a high repetition rate laser for excitation. THz wave generation from ionized liquids presents photoionization processes that are different from those in gases. The most important difference is the preference of liquid for excitation with a sub-picosecond pulse. In gas, the shortest pulse always generates strongest THz field, but in liquids, in a big surprise we have found that a laser pulse with longer pulse duration offers stronger THz emission!

Discussion

Since the use of short-pulse laser excitation on photoconductors and electro-optic crystals in late 1980s', the development of THz science and technology has been limited to the marginal power level of available lasers with nano- to milli-Joule laser pulse energy. All our previous projects on liquid water with short laser pulses are low peak laser power. As we proposed in our project title, "THz photonics in water and beyond", after we demonstrated THz wave generation from water lines, we also used liquid nitrogen and liquid metals as the THz sources, and published the results by the end of the project.

LLE and renewal proposal

With the completion of the previous AFOSR project, we have submitted a renewal proposal. We propose to investigate THz wave emitters under ultra-intense laser pulse excitation. We propose to explore the upper limit of THz wave emitters with the application of the most intense lasers (kJ pulse energy and sub-ps pulse duration) in the world at the Laboratory for Laser Energetics at Rochester. We propose to investigate THz photonics by using unique lasers (from **J** to **kJ** pulse energy) originally constructed for laser fusion at the Laboratory for Laser Energetics. Proposed tasks including different THz emitter targets, excitation laser parameters, and spectral/temporally resolved detection. The development of a single-shot THz characterization technology is imperative for low repetition rate lasers, which will also be demonstrated in our lab.

Preliminary Results with Laser Pulse Energy > 10 J (Unpublished)

We are collaborating with LLE researchers on terahertz (THz) wave generation from relativistic electrons, shown in **Figure 19**. This preliminary test on the solid targets is a collaboration with the Laboratory for Laser Energetics (LLE), University of Rochester. The laser used is the Multi-Terawatt (MTW) laser, which can shoot a 13 J pulse every 30 min with a central wavelength at 1053 nm. Under the best compression, the pulse duration is about 700 fs. The MTW laser parameters are:

- Wavelength: 1053 nm
- Pulse duration: ~ 1 ps
- Pulse energy: 13 -18 J
- ~30 min between shots
- ~10^{19} W/cm^2 at peak

By focusing the laser pulse onto a 20-μm thick CH target, a THz signal is detected by using a pyroelectric detector. The frequency of the signal is mainly located below 3 THz. This observation agrees with the theoretical prediction of the coherent transition radiation model.

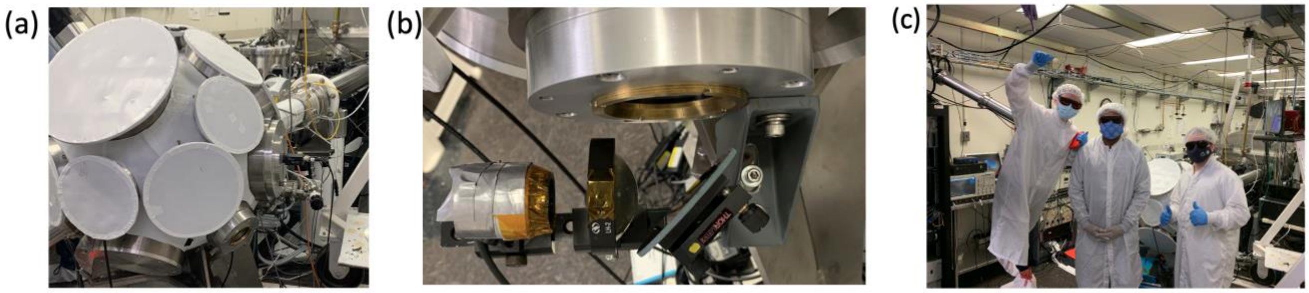


Figure 19. (a) The target chamber for THz wave generation using a multi-terawatt laser at LLE, Rochester. (b) THz detection from one of the chamber windows. (c) PhD students, Gerrit Bauhuag (right) and Kareem Garriga (middle) and Research Associate Dr. Yiwen E (left) in the testing room.

Chair evaluation for 2020 (The Institute of Optics)

I am pleased to share my 2020 annual valuation for teaching, research, and service at The Institute of Optics, Univ. of Rochester. I received all "Exceptional" scores during the most challenging period (Covid-19 pandemic) in my faculty career.

Chair Evaluation
2020 Calendar Year

Evaluee: XI-CHENG ZHANG
Department: OPTICS INSTITUTE
Last Update: May 18, 2021 2:25 PM

Contributors: P. Scott Carney

Teaching Evaluation

Rating: **5** (Exceptional)

Comments: XC was due a semester off of teaching last Fall. As we brainstormed for ideas to retain our MS students (whom we were afraid were especially vulnerable to melt, especial out international MS students), he stepped up and taught a special class on THz sources on-line. It was instrumental in our high retention rates. Moreover, this demonstrated X-C's dedication to the department and the university.

Research Evaluation

Rating: **5** (Exceptional)

Comments: As always, X-C maintains a high level of research output. His core group is a little smaller than in the past, but he is collaborating more. He has several proposals in and I expect this will be a very good year for him.

Service Evaluation

Rating: **5** (Exceptional)

Comments: X-C performs on three difficult service assignment. First, he is our representative on the Faculty Council, a body so dysfunctional that the one time i attended in his place, a member called for the abolitions of the council itself. Second, he serves on the ABET committee along with Andrew Berger and Wayne Knox (chair). Thirdly, X-C is cochair of the admissions committee and handles all MS applications. At my request, he implemented a rapid-response rolling admissions policy. His careful work with the committee has ensured a high-quality of students while also producing much needed revenue for ASE.

Overall Evaluation

Rating: **5** (Exceptional)

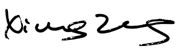
Comments: X-C is an exceptional member of the faculty and that has become very clear this year. While his teaching in prior years has occasionally been his weakest of the three categories, this year it is his strongest, a testament to his ability to rise to the needs of the department.

Summary

Ultra-Intense THz Sources: To explore THz science under extreme conditions, intense THz sources are essential. We have started a preliminary test using ps lasers with >10 J pulse energy at the LLE. Preliminary experimental results measured that THz wave emitted from thin solid targets in forward and sideway directions. X-ray and THz pulse are generated simultaneously. With the use of strong lasers (> kJ pulse energy), new ultra-intense THz sources will provide unique platform and opportunity for new sciences under intense THz field. 100 GV/m THz field using the lasers with kJ pulse energy and sub-ps pulse duration is possible if the saturation of THz emission could be managed. This AFOSR project offered us the opportunity to explore new science and technology on light-matter interaction. We have tried our very best to meet the proposed goal and performed well during this most challenging period.

Meliora

Submitted respectfully



Xi-Cheng Zhang

References cited in this final report

[1] H. Hamster, A. Sullivan, S. Gordon, et al., "Subpicosecond, electromagnetic pulses from intense laser-plasma interaction," Physical Review Letters, Vol. 71, no. 17, pp. 2725-2728, 1993.

[2] H. Hamster, A. Sullivan, S. Gordon, et al., "Short-pulse terahertz radiation from high-intensity-laser-produced plasmas," Physical Review E, Vol. 49, no. 1, pp. 671-677, 1994.

[3] D. J. Cook, and R. M. Hochstrasser, "Intense terahertz pulses by four-wave rectification in air," Optics Letters, Vol. 25, no. 16, pp. 1210-1212, 2000. https://doi.org/10.1364/OL.25.001210.

[4] M. Clerici, M. Peccianti, B. E. Schmidt, et al., "Wavelength Scaling of Terahertz Generation by Gas Ionization," Physical Review Letters, Vol. 110, no. 25, pp. 253901, 2013.

[5] X. Xie, J. Dai, and X. C. Zhang, "Coherent Control of THz Wave Generation in Ambient Air," Physical Review Letters, Vol. 96, no. 7, pp. 075005, 2006.

[6] J. Dai, J. Liu, and X. C. Zhang, "Terahertz Wave Air Photonics: Terahertz Wave Generation and Detection With Laser-Induced Gas Plasma," IEEE Journal of Selected Topics in Quantum Electronics, Vol. 17, no. 1, pp. 183-190, 2011. https://doi.org/10.1109/JSTQE.2010.2047007.

[7] F. Jahangiri, M. Hashida, T. Nagashima, et al., "Intense terahertz emission from atomic cluster plasma produced by intense femtosecond laser pulses," Applied Physics Letters, Vol. 99, no. 26, pp. 261503, 2011. https://doi.org/10.1063/1.3672814.

[8] T. Nagashima, H. Hirayama, K. Shibuya, et al., "Terahertz pulse radiation from argon clusters irradiated with intense femtosecond laser pulses," Optics Express, Vol. 17, no. 11, pp. 8907-8912, 2009. https://doi.org/10.1364/OE.17.008907.

[9] K. Mori, M. Hashida, T. Nagashima, et al., "Directional linearly polarized terahertz emission from argon clusters irradiated by noncollinear double-pulse beams," Applied Physics Letters, Vol. 111, no. 24, pp. 241107, 2017. https://doi.org/10.1063/1.4991736.

[10] Q. Jin, Y. E, K. Williams, et al., "Observation of broadband terahertz wave generation from liquid water," Applied Physics Letters, Vol. 111, no. 7, pp. 071103, 2017. https://doi.org/10.1063/1.4990824.

[11] I. Dey, K. Jana, V. Y. Fedorov, et al., "Highly efficient broadband terahertz generation from ultrashort laser filamentation in liquids," Nature communications, Vol. 8, no. 1, pp. 1184, 2017. https://doi.org/10.1038/s41467-017-01382-x.

[12] Y. E, Q. Jin, A. Tcypkin, et al., "Terahertz wave generation from liquid water films via laser-induced breakdown," Applied Physics Letters, Vol. 113, no. 18, pp. 181103, 2018. https://doi.org/10.1063/1.5054599.

[13] Q. Jin, J. M. Dai, Y. W. E, et al., "Terahertz wave emission from a liquid water film under the excitation of asymmetric optical fields," Applied Physics Letters, Vol. 113, no. 26, pp. 261101, 2018. https://doi.org/10.1063/1.5064644.
[14] A. N. Tcypkin, E. A. Ponomareva, S. E. Putilin, et al., "Flat liquid jet as a highly efficient source of terahertz radiation," Optics Express, Vol. 27, no. 11, pp. 15485-15494, 2019. https://doi.org/10.1364/OE.27.015485.
[15] L.-L. Zhang, W.-M. Wang, T. Wu, et al., "Strong Terahertz Radiation from a Liquid-Water Line," Physical Review Applied, Vol. 12, no. 1, pp. 014005, 2019. https://doi.org/10.1103/PhysRevApplied.12.014005.
[16] A. V. Balakin, J.-L. Coutaz, V. A. Makarov, et al., "Terahertz wave generation from liquid nitrogen," Photonics Research, Vol. 7, no. 6, pp. 678-686, 2019. https://doi.org/10.1364/PRJ.7.000678.
[17] A. Gopal, P. Singh, S. Herzer, et al., "Characterization of 700 μJ T rays generated during high-power laser solid interaction," Optics Letters, Vol. 38, no. 22, pp. 4705-4707, 2013. https://doi.org/10.1364/OL.38.004705.
[18] G. Liao, Y. Li, H. Liu, et al., "Multimillijoule coherent terahertz bursts from picosecond laser-irradiated metal foils," Proceedings of the National Academy of Sciences, Vol. 116, no. 10, pp. 3994, 2019. https://doi.org/10.1073/pnas.1815256116.
[19] A. Woldegeorgis, T. Kurihara, M. Almassarani, et al., "Multi-MV/cm longitudinally polarized terahertz pulses from laser–thin foil interaction," Optica, Vol. 5, no. 11, pp. 1474-1477, 2018. https://doi.org/10.1364/OPTICA.5.001474.
[20] G.-Q. Liao, Y.-T. Li, Y.-H. Zhang, et al., "Demonstration of Coherent Terahertz Transition Radiation from Relativistic Laser-Solid Interactions," Physical Review Letters, Vol. 116, no. 20, pp. 205003, 2016. https://doi.org/10.1103/PhysRevLett.116.205003.
[21] Laser Induced Breakdown Spectroscopy (Cambridge University Press, 2006).
[22] F. Anabitarte, A. Cobo, and J. M. Lopez-Higuera, "Laser-Induced Breakdown Spectroscopy: Fundamentals, Applications, and Challenges," ISRN Spectroscopy, Vol. 2012, no., pp. 12, 2012. https://doi.org/10.5402/2012/285240.
[23] B. Kearton, and Y. Mattley, "Sparking new applications," Nature Photonics, Vol. 2, no., pp. 537, 2008. https://doi.org/10.1038/nphoton.2008.173.
[24] S. Corde, K. Ta Phuoc, G. Lambert, et al., "Femtosecond x rays from laser-plasma accelerators," Reviews of Modern Physics, Vol. 85, no. 1, pp. 1-48, 2013.
[25] J. Yoshii, C. H. Lai, T. Katsouleas, et al., "Radiation from Cerenkov Wakes in a Magnetized Plasma," Physical Review Letters, Vol. 79, no. 21, pp. 4194-4197, 1997.
[26] D. Kuk, Y. J. Yoo, E. W. Rosenthal, et al., "Generation of scalable terahertz radiation from cylindrically focused two-color laser pulses in air," Applied Physics Letters, Vol. 108, no. 12, pp. 121106, 2016. https://doi.org/10.1063/1.4944843.
[27] T. I. Oh, Y. J. Yoo, Y. S. You, et al., "Generation of strong terahertz fields exceeding 8 MV/cm at 1 kHz and real-time beam profiling," Applied Physics Letters, Vol. 105, no. 4, pp. 041103, 2014. https://doi.org/10.1063/1.4891678.
[28] X. C. Zhang, A. Shkurinov, and Y. Zhang, "Extreme terahertz science," Nature Photonics, Vol. 11, no., pp. 16, 2017. https://doi.org/10.1038/nphoton.2016.249.
[29] J. Liu, and X. C. Zhang, "Terahertz-Radiation-Enhanced Emission of Fluorescence from Gas Plasma," Physical Review Letters, Vol. 103, no. 23, pp. 235002, 2009. https://doi.org/10.1103/PhysRevLett.103.235002.
[30] J. Liu, J. Dai, S. L. Chin, et al., "Broadband terahertz wave remote sensing using coherent manipulation of fluorescence from asymmetrically ionized gases," Nature Photonics, Vol. 4, no., pp. 627, 2010. https://doi.org/10.1038/nphoton.2010.165.
[31] B. Clough, J. Liu, and X. C. Zhang, ""All air-plasma" terahertz spectroscopy," Optics Letters, Vol. 36, no. 13, pp. 2399-2401, 2011. https://doi.org/10.1364/OL.36.002399.
[32] J. Liu, B. Clough, and X. C. Zhang, "Enhancement of photoacoustic emission through terahertz-field-driven electron motions," Physical Review E, Vol. 82, no. 6, pp. 066602, 2010. https://doi.org/10.1103/PhysRevE.82.066602.
[33] B. Clough, J. Liu, and X. C. Zhang, "Laser-induced photoacoustics influenced by single-cycle terahertz radiation," Optics Letters, Vol. 35, no. 21, pp. 3544-3546, 2010. https://doi.org/10.1364/OL.35.003544.

[34] F. Buccheri, and X.-C. Zhang, "Terahertz emission from laser-induced microplasma in ambient air," Optica, Vol. 2, no. 4, pp. 366-369, 2015. https://doi.org/10.1364/OPTICA.2.000366.
[35] C. D'Amico, A. Houard, M. Franco, et al., "Conical Forward THz Emission from Femtosecond-Laser-Beam Filamentation in Air," Physical Review Letters, Vol. 98, no. 23, pp. 235002, 2007. https://doi.org/10.1103/PhysRevLett.98.235002.
[36] J. Dai, N. Karpowicz, and X. C. Zhang, "Coherent polarization control of terahertz waves generated from two-color laser-induced gas plasma," Phys Rev Lett, Vol. 103, no. 2, pp. 023001, 2009. https://doi.org/10.1103/PhysRevLett.103.023001.
[37] H. Wen, and A. M. Lindenberg, "Coherent Terahertz Polarization Control through Manipulation of Electron Trajectories," Physical Review Letters, Vol. 103, no. 2, pp. 023902, 2009. https://doi.org/10.1103/PhysRevLett.103.023902.
[38] K.-Y. Kim, "Generation of coherent terahertz radiation in ultrafast laser-gas interactions," Physics of Plasmas, Vol. 16, no. 5, pp. 056706, 2009.
[39] K.-Y. Kim, J. H. Glownia, A. J. Taylor, et al., "Terahertz emission from ultrafast ionizing air in symmetry-broken laser fields," Optics Express, Vol. 15, no. 8, pp. 4577-4584, 2007. https://doi.org/10.1364/OE.15.004577.
[40] K.-Y. Kim, A. Taylor, J. Glownia, et al., "Coherent control of terahertz supercontinuum generation in ultrafast laser–gas interactions," Nature Photonics, Vol. 2, no. 10, pp. 605, 2008. https://doi.org/10.1038/nphoton.2008.153.
[41] N. Karpowicz, and X.-C. Zhang, "Coherent terahertz echo of tunnel ionization in gases," Physical Review Letters, Vol. 102, no. 9, pp. 093001, 2009.
[42] J. Dai, X. Xie, and X. C. Zhang, "Detection of Broadband Terahertz Waves with a Laser-Induced Plasma in Gases," Physical Review Letters, Vol. 97, no. 10, pp. 103903, 2006.
[43] N. Karpowicz, J. Dai, X. Lu, et al., "Coherent heterodyne time-domain spectrometry covering the entire "terahertz gap"," Applied Physics Letters, Vol. 92, no. 1, pp. 011131, 2008. https://doi.org/10.1063/1.2828709.
[44] Q. Wu, and X. C. Zhang, "7 terahertz broadband GaP electro-optic sensor," Applied Physics Letters, Vol. 70, no. 14, pp. 1784-1786, 1997. https://doi.org/10.1063/1.118691.
[45] Y. Chen, M. Yamaguchi, M. Wang, et al., "Terahertz pulse generation from noble gases," Applied Physics Letters, Vol. 91, no. 25, pp. 251116, 2007. https://doi.org/10.1063/1.2826544.
[46] G. Rodriguez, and G. L. Dakovski, "Scaling behavior of ultrafast two-color terahertz generation in plasma gas targets: energy and pressure dependence," Optics Express, Vol. 18, no. 14, pp. 15130-15143, 2010. https://doi.org/10.1364/OE.18.015130.
[47] S. L. Chin, T. J. Wang, C. Marceau, et al., "Advances in intense femtosecond laser filamentation in air," Laser Physics, Vol. 22, no. 1, pp. 1-53, 2012. https://doi.org/10.1134/S1054660X11190054.
[48] A. Sagisaka, H. Daido, S. Nashima, et al., "Simultaneous generation of a proton beam and terahertz radiation in high-intensity laser and thin-foil interaction," Applied Physics B, Vol. 90, no. 3, pp. 373-377, 2008. https://doi.org/10.1007/s00340-008-2931-8.
[49] S. Feng, L. Dong, T. Wu, et al., "Terahertz wave emission from water lines," Chinese Optics Letters, Vol. 18, no. 2, pp. 023202, 2020.
[50] Q. Jin, E. Yiwen, S. Gao, et al., "Preference of subpicosecond laser pulses for terahertz wave generation from liquids," Advanced Photonics, Vol. 2, no. 1, pp. 015001, 2020.
[51] A. Ismagilov, E. Ponomareva, M. Zhukova, et al., "Liquid jet-based broadband terahertz radiation source," Optical Engineering, Vol. 60, no. 8, pp. 082009, 2021.
[52] F. Williams, S. Varma, and S. Hillenius, "Liquid water as a lone-pair amorphous semiconductor," The Journal of Chemical Physics, Vol. 64, no. 4, pp. 1549-1554, 1976.
[53] D. N. Nikogosyan, A. A. Oraevsky, and V. I. Rupasov, "Two-photon ionization and dissociation of liquid water by powerful laser UV radiation," Chemical Physics, Vol. 77, no. 1, pp. 131-143, 1983.
[54] R. A. Crowell, and D. M. Bartels, "Multiphoton ionization of liquid water with 3.0− 5.0 eV photons," The Journal of Physical Chemistry, Vol. 100, no. 45, pp. 17940-17949, 1996.
[55] L. Malmqvist, L. Rymell, and H. M. Hertz, "Droplet-target laser-plasma source for proximity x-ray lithography," Applied Physics Letters, Vol. 68, no. 19, pp. 2627-2629, 1996. https://doi.org/10.1063/1.116203.

[56] M. Berglund, L. Rymell, H. M. Hertz, et al., "Cryogenic liquid-jet target for debris-free laser-plasma soft x-ray generation," Review of Scientific Instruments, Vol. 69, no. 6, pp. 2361-2364, 1998. https://doi.org/10.1063/1.1148944.

[57] K. M. George, J. T. Morrison, S. Feister, et al., "High-repetition-rate (≥ kHz) targets and optics from liquid microjets for high-intensity laser–plasma interactions," High Power Laser Science and Engineering, Vol. 7, no., pp. e50, 2019. https://doi.org/10.1017/hpl.2019.35.

[58] L. Thrane, R. H. Jacobsen, P. U. Jepsen, et al., "THz reflection spectroscopy of liquid water," Vol. 240, no. 4, pp. 330-333, 1995.

[59] C. Ro/nne, L. Thrane, P.-O. Åstrand, et al., "Investigation of the temperature dependence of dielectric relaxation in liquid water by THz reflection spectroscopy and molecular dynamics simulation," The Journal of Chemical Physics, Vol. 107, no. 14, pp. 5319-5331, 1997. https://doi.org/10.1063/1.474242.

[60] T. Wang, P. Klarskov, and P. U. Jepsen, "Ultrabroadband THz Time-Domain Spectroscopy of a Free-Flowing Water Film," IEEE Transactions on Terahertz Science and Technology, Vol. 4, no. 4, pp. 425-431, 2014. https://doi.org/10.1109/TTHZ.2014.2322757.

[61] S. Stumpf, E. Ponomareva, A. Tcypkin, et al., "Temporal field and frequency spectrum of intense femtosecond radiation dynamics in the process of plasma formation in a dielectric medium," Laser Physics, Vol. 29, no. 12, pp. 124014, 2019. https://doi.org/10.1088/1555-6611/ab4dac.

[62] E. A. Ponomareva, S. A. Stumpf, A. N. Tcypkin, et al., "Impact of laser-ionized liquid nonlinear characteristics on the efficiency of terahertz wave generation," Optics Letters, Vol. 44, no. 22, pp. 5485-5488, 2019. https://doi.org/10.1364/OL.44.005485.

[63] S. L. Chin, Femtosecond laser filamentation (Springer, 2010).

[64] J. Samios, U. Mittag, and T. Dorfmüller, "The far infrared absorption spectrum of liquid nitrogen," Molecular Physics, Vol. 56, no. 3, pp. 541-556, 1985. https://doi.org/10.1080/00268978500102511.

[65] J. G. Leidenfrost, De aquae communis nonnullis qualitatibus tractatus (Ovenius, 1756).

[66] B. S. Gottfried, C. J. Lee, and K. J. Bell, "The leidenfrost phenomenon: film boiling of liquid droplets on a flat plate," International Journal of Heat and Mass Transfer, Vol. 9, no. 11, pp. 1167-1188, 1966. https://doi.org/10.1016/0017-9310(66)90112-8.

[67] Y. E, Y. Cao, F. Ling, et al., "Flowing cryogenic liquid target for terahertz wave generation," AIP Advances, Vol. 10, no. 10, pp. 105119, 2020. https://doi.org/10.1063/5.0023106.

[68] O. Hemberg, M. Otendal, and H. Hertz, "Liquid-metal-jet anode electron-impact x-ray source," Applied Physics Letters, Vol. 83, no. 7, pp. 1483-1485, 2003.

[69] D. H. Larsson, P. A. Takman, U. Lundström, et al., "A 24 keV liquid-metal-jet x-ray source for biomedical applications," Review of Scientific Instruments, Vol. 82, no. 12, pp. 123701, 2011.

[70] E. Espes, T. Andersson, F. Bjornsson, et al., "Liquid-metal-jet x-ray tube technology and tomography applications," in *Digital Imaging XVIII*(2015), pp. 7-11

[71] Y. Cao, Y. E, P. Huang, et al., "Broadband terahertz wave emission from liquid metal," Applied Physics Letters, Vol. 117, no. 4, pp. 041107, 2020. https://doi.org/10.1063/5.0015507.

[72] Y.-S. Lee, Principles of terahertz science and technology (Springer Science & Business Media, 2009).

[73] L. R. Snyder, "Classification of the solvent properties of common liquids," Journal of Chromatography A, Vol. 92, no. 2, pp. 223-230, 1974.

[74] M. Berglund, L. Rymell, and H. M. Hertz, "Ultraviolet prepulse for enhanced x-ray emission and brightness from droplet-target laser plasmas," Applied Physics Letters, Vol. 69, no. 12, pp. 1683-1685, 1996. https://doi.org/10.1063/1.117027.

[75] M. Anand, S. Kahaly, G. R. Kumar, et al., "Enhanced hard x-ray emission from microdroplet preplasma," Applied Physics Letters, Vol. 88, no. 18, pp. 181111, 2006. https://doi.org/10.1063/1.2200761.

[76] E. A. Ponomareva, A. N. Tcypkin, S. V. Smirnov, et al., "Double-pump technique–one step closer towards efficient liquid-based THz sources," Optics Express, Vol. 27, no. 22, pp. 32855-32862, 2019.

[77] E. A. Ponomareva, A. O. Ismagilov, S. E. Putilin, et al., "Varying pre-plasma properties to boost terahertz wave generation in liquids," Communications Physics, Vol. 4, no. 1, pp. 1-7, 2021.

[78] H.-H. Huang, T. Nagashima, W.-H. Hsu, et al., "Dual THz Wave and X-ray Generation from a Water Film under Femtosecond Laser Excitation," Nanomaterials (Basel, Switzerland), Vol. 8, no. 7, pp. 523, 2018. https://doi.org/10.3390/nano8070523.

[79] Y. E, Q. Jin, and X.-C. Zhang, "Enhancement of terahertz emission by a preformed plasma in liquid water," Applied Physics Letters, Vol. 115, no. 10, pp. 101101, 2019. https://doi.org/10.1063/1.5119812.

[80] P. M. Solyankin, B. V. Lakatosh, M. S. Krivokorytov, et al., "Single Free-Falling Droplet of Liquid Metal as a Source of Directional Terahertz Radiation," Physical Review Applied, Vol. 14, no. 3, pp. 034033, 2020. https://doi.org/10.1103/PhysRevApplied.14.034033.

[81] J. Kerr, "XL. A new relation between electricity and light: Dielectrified media birefringent," The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science, Vol. 50, no. 332, pp. 337-348, 1875. https://doi.org/10.1080/14786447508641302.

[82] J. Kerr, "XLIII. On rotation of the plane of polarization by reflection from the pole of a magnet," The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science, Vol. 3, no. 19, pp. 321-343, 1877. https://doi.org/10.1080/14786447708639245.

[83] M. C. Hoffmann, N. C. Brandt, H. Y. Hwang, et al., "Terahertz Kerr effect," Applied Physics Letters, Vol. 95, no. 23, pp. 231105, 2009. https://doi.org/10.1063/1.3271520.

[84] F. Perakis, L. De Marco, A. Shalit, et al., "Vibrational spectroscopy and dynamics of water," Chemical reviews, Vol. 116, no. 13, pp. 7590-7607, 2016.

[85] H. Zhao, Y. Tan, L. Zhang, et al., "Ultrafast hydrogen bond dynamics of liquid water revealed by terahertz-induced transient birefringence," Light: Science & Applications, Vol. 9, no. 1, pp. 136, 2020. https://doi.org/10.1038/s41377-020-00370-z.

[86] R. W. Boyd, Nonlinear optics (Academic press, 2020).

[87] W. Demtröder, Laser spectroscopy: vol. 2: experimental techniques (Springer Science & Business Media, 2008).

[88] M. Levenson, Introduction to Nonlinear Laser Spectroscopy 2e (Elsevier, 2012).

[89] H. Zhao, Y. Tan, R. Zhang, et al., "Anion–water hydrogen bond vibration revealed by the terahertz Kerr effect," Optics Letters, Vol. 46, no. 2, pp. 230-233, 2021. https://doi.org/10.1364/OL.409849.

[90] I. A. Heisler, and S. R. Meech, "Low-Frequency Modes of Aqueous Alkali Halide Solutions: Glimpsing the Hydrogen Bonding Vibration," Science, Vol. 327, no. 5967, pp. 857-860, 2010. https://doi.org/10.1126/science.1183799.

[91] D.-Y. Wu, S. Duan, X.-M. Liu, et al., "Theoretical Study of Binding Interactions and Vibrational Raman Spectra of Water in Hydrogen-Bonded Anionic Complexes: $(H_2O)_n^-$ (n = 2 and 3), $H_2O\cdots X^-$(X = F, Cl, Br, and I), and $H_2O\cdots M^-$ (M = Cu, Ag, and Au)," The Journal of Physical Chemistry A, Vol. 112, no. 6, pp. 1313-1321, 2008.

[92] A. V. Grosse, "Surface tension of the alkali metals from the melting point to the critical region," Journal of Inorganic and Nuclear Chemistry, Vol. 30, no. 5, pp. 1169-1174, 1968.

[93] V. Y. Prokhorenko, V. V. Roshchupkin, M. A. Pokrasin, et al., "Liquid gallium: potential uses as a heat-transfer agent," High Temperature, Vol. 38, no. 6, pp. 954-968, 2000.

[94] D. White, "The surface tensions of indium and cadmium," Metallurgical and Materials Transactions B, Vol. 3, no. 7, pp. 1933-1936, 1972.