Review Vol. 39, No. 3 / March 2022 / Journal of the Optical Society of America B A43

Journal of the Optical Society of America B

OPTICAL PHYSICS

Progress, challenges, and opportunities of terahertz emission from liquids

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Received 18 October 2021; revised 5 December 2021; accepted 7 December 2021; posted 8 December 2021; published 7 January 2022

Broadband terahertz (THz) wave emission from flowing liquid targets has been demonstrated under short optical pulse excitation. Observations have been reported by using liquid THz sources, including optimal angle of incidence, preference of subpicosecond pulse excitation, and strong sideway emission. Compared with solid targets, the fluidity of liquid allows each laser pulse to interact with a fresh area, which makes it possible to use a table-top laser with a high repetition rate for excitation. Liquids with a comparable material density to solids make them promising candidates for the study of high-density plasma and bright THz sources. In this paper, we review recent progress, challenges, and opportunities of THz emission from liquids. This topic may offer new possibilities in the exploration of THz liquid photonics and may play an indispensable role in the study of laser-liquid interaction.

https://doi.org/10.1364/JOSAB.446095

1. INTRODUCTION

In the last few decades, the terahertz (THz) wave with spectrum from 0.3 to 10 THz has been studied in many aspects. By measuring the absorption resonances of molecules, THz spectroscopy is widely used as a technique to characterize matters [1–3]. As the high-frequency limit for most electronic devices, THz waves are currently applied to the sixth generation of communications with an ultrabroad bandwidth [4]. Certain ultrafast dynamics, such as chemical reactions [5], relaxation of phonons in crystal [6,7], collision of carriers in semiconductors [8], and elementary excitation in solids [9], present a time scale of a picosecond, making THz waves practical in monitoring these processes. Moreover, since an intense THz electric field can induce ultrafast switching in matters, the THz wave controlled nonlinear phenomenon has attracted much research attention [10–12]. Therefore, the development of THz technology directly impacts applications in advanced sciences.

Solids, gases, and plasmas have long been studied as THz sources [13–15]; however, the use of liquids as THz wave emitters is still underdeveloped, possibly because the most common liquid water shows a high absorption at THz frequencies. With the application of flowing liquid targets in the generation of extreme ultraviolet light and soft x rays under femtosecond laser excitation [16–18], it is reasonable to expect strong THz wave generation from ionized liquids. Liquids have a comparable material density to solids, making 3 orders of more molecules interact with laser field than an equivalent cross section of gases. The fluidity enables the liquid target to replenish the surface quickly. Accordingly, target surface damage or material degradation is not an issue with the excitation of kilohertz (kHz) repetition rate laser pulses. These motivations make liquids promising candidates for intense THz sources.

Since the first demonstrations of THz wave generation from a liquid film [19] and liquid in a cuvette [20] in 2017, varieties of experiments with different liquids have been studied for THz wave generation [21,22], including water, acetone, liquid nitrogen (LN) [23,24], and liquid metal [25–27] with different target geometries, such as liquid wedge [28], liquid line [29–32], and droplets [27]. Moreover, the use of flowing liquid target as a sample has also been applied in the study of ultrafast hydrogen bond dynamics [33], quasifree electrons in ionized liquid [34], and THz third-order nonlinear response [35].

2. FLOWING LIQUID TARGETS FOR OPTICAL EXCITATION

It is well known that some liquids strongly absorb far-infrared electromagnetic waves. For example, liquid water has a continuous absorption covering THz regime with the power absorption coefficient $\alpha = 220\ \mathrm{cm}^{-1}$ at 1 THz [36]. Such a high absorption makes it very difficult to measure THz emission from liquid water. Therefore, reflection geometry or thin targets are more practical.

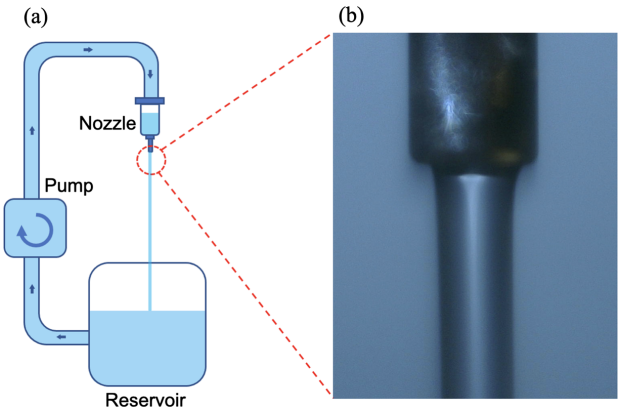


Fig. 1. (a) Schematic diagram of a liquid target system. Liquid is pumped from a reservoir and ejects through a nozzle. (b) Photo of a flowing water line with a diameter of 260 μm.

To prepare a thin flowing liquid target, a liquid pumping system is required to support certain pressure and flow rate. The shape, direction, and pressure of the liquid flow can be further controlled by the jet nozzle. Figure 1(a) shows the schematic diagram of a liquid flow system. A peristaltic pump is used to pump the fluid from the reservoir to the nozzle. The flow rate is controllable by varying the rotor speed inside the pump. With a proper design of the system, a small volume (<50 ml) of liquid is needed for hours of operation without interruption. The consumption of liquid is mainly from the evaporation in the ionization process.

The target geometry stability is important to get a good signal-noise-ratio in measurement. In fluid mechanics, a turbulent/laminar flow is defined by the molecule trajectory within the flow [37]. A turbulent flow indicates that the molecules move irregularly with an unstable speed in random directions. On the contrary, in a laminar flow, there is no lateral mixing. The liquid is regarded as several layers moving smoothly, producing a good quality (stable and smooth surface) of flow for optical excitation. For different liquids, the flow rate needs to be finely controlled to form a laminar flow. The maximum flow rate of a liquid target is limited by the condition of avoiding the turbulence in the flow. And the minimum flow rate is decided by the repetition rate of optical pulses for excitation and the diameter of focal spot on the liquid target. A certain flow rate is required to avoid the disturbance caused by the previous laser pulse. If the laser has a repetition rate of 1 kHz, the flow rate should be higher than 1 m/s when the beam waist is 1 mm at the focal point. For different liquids and excitation geometry, a proper flow rate should be carefully selected to get a qualified liquid target for optical excitation.

A. Liquid Line

Liquid line is a relatively feasible approach to produce a flowing liquid target in the lab. A syringe needle can be used as a nozzle to form a cylinder shape of flow. Figure 1(b) shows the photo of a 260 μm water line. The uniform reflection in the photo showcases the smoothness and the stability of the flow. The diameter of the flow is as same as the inner diameter of the needle when the flow is laminar. However, because of the acceleration of the gravity, the flow becomes turbulent after a certain distance from a tip and breaks into droplets eventually [38]. The laminar flow only survives near the needle tip. Usually, the focus of laser beam is set about 3 mm from the tip.

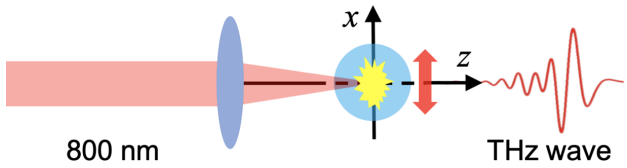


Fig. 2. Schematic diagram of a liquid line under the excitation of focused optical beam.

Figure 2 shows the schematic diagram of a liquid line under the excitation of the focused optical beam. To eliminate the potential contribution from air plasma, the technique of "microplasma" [39] is employed to confine the laser pulse energy within a small volume, in which a high numerical-aperture (NA) objective is used to focus laser pulse on the target. To detect THz emissions, either electro-optic sampling or THz power detectors can be used.

B. Liquid Film

The utilization of flowing liquid film is an appealing method in absorption or emission spectroscopy to guarantee a continuous refresh of the liquid, as well as to avoid liquid degradation. Unlike a liquid line with a curved surface, the liquid film with flat surfaces is more favorable in the quantitative discussion. For the study at THz frequencies, Wang *et al*. used the wire-guided film in the measurement of index and absorption coefficient of liquid water [40]. Recently, the flowing water jet has also been used for the investigation of THz Kerr effect to unveil ultrafast dynamics in liquid sample [41,42]. The broadband THz wave generation from a flowing water film under 800 nm optical excitation was demonstrated in 2017 [19].

There are different approaches to produce a thin, flowing liquid film. Figure 3(a) shows the wire-guided gravity-driven water film. Benefiting from the high surface tension of water, a flow guided by two wires forms a thin film. The thickness of the film depends on the diameter of the wire, surface tension of the liquid, and flow rate. Usually, the thickness of guided film is greater than 100 μm. For some liquids with relatively low viscosity, the film is very easy to break when an intense laser is focused onto it, which is usually hard to recover. Also, the thickness is not uniform across the film. At the center of the film, it is usually thinner. Figure 3(b) shows a water film formed by a slit nozzle. The similar design has been used in dye laser systems for decades [44,45]. This method provides a better control of pressure than that of the gravity-driven liquid film, keeping the shape of the film unchanged under intense laser excitation. However, these approaches hardly produce a liquid film with the thickness under 10 μm. By colliding two laminar jets (cylinder liquid lines) [46], a uniform thin film is formed as shown in Fig. 3(c). With this method (flatjet), the thickness of the film can possibly to be less than 5 μm. Table 1 lists pros and cons of different methods to produce a flowing liquid film.

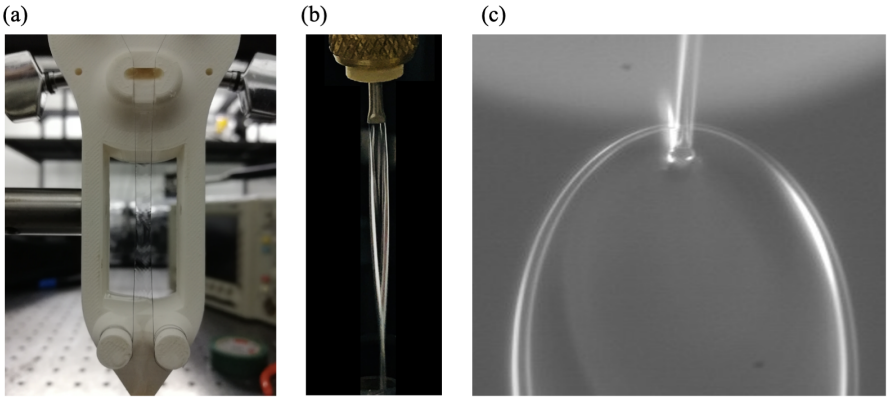


Fig. 3. Thin liquid films produced by three different methods. (a) Wire-guided free-flowing film; (b) thin film by a slit nozzle; (c) flatjet [43], a uniform thin film formed by the collision of two laminar jets.

Table 1. Comparison of Different Methods for Making a Liquid Film

	Pros	Cons
Gravity-driven film	Simple in operation and construction, low maintenance, adjustable thickness.	Delicate film structure, uneven surface profile, no possibility for use with high-power systems.
Jet nozzle	Easy in operation, high stability, ease of use with various liquids, relatively simple in construction, highly adjustable thickness.	System can be costly, uneven surface profile, the liquid target quality highly depends on the nozzle.
Flatjet	Use with various liquids, extremely flat surface profile.	Expensive, films cannot be made larger than 1 mm or thicker than 10 μm.

C. Droplets

The continuous flow in liquid line or film is sometimes restricted in the experiments due to the laser induced fragments (debris), which could damage the optics near the target. With mass-limited liquid droplets, the debris problem is negligible. Also, the amount of required liquid for test declines considerably, which is favorable in the test of costly materials. By synchronizing the timing between falling droplets and laser pulses, droplets have been widely studied in a high brightness extreme ultraviolet light and x ray generation, respectively [17,47,48]. In 2020, Solyankin *et al.* demonstrated directional THz radiation from liquid metal droplets under double-pump optical excitation, in which two pulses with a certain delay are used for excitation [27]. The THz generation efficiency is optimized by varying the delay between two pulses.

3. PROPERTIES OF THz EMISSION FROM LIQUIDS

Using ionized materials to generate electromagnetic waves for different wavelengths has been studied for decades with the fast development of laser technology. For the THz wave, air plasma under two-color excitation as one of the brightest THz emitters generates intense THz field with ultrabroad bandwidth, attracting considerable attention in the past few years. Liquid plasma as a new type of source in the THz community has shown different properties.

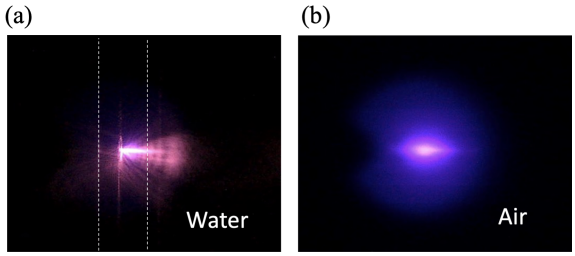


Fig. 4. Photos of the plasma fluorescence taken by a CCD camera from sideway. (a) Liquid plasma in a 210 μm water line. The dashed lines indicate the interface of the liquid line. (b) Air plasma.

Figure 4 shows the fluorescence of water and air plasmas taken sideway by a CCD camera. In this paper, "sideway" indicates the direction that is perpendicular to the laser propagation direction in the horizontal plane. A 2 in. focal length lens is used to focus the laser beam. Since the liquid target introduces a liquid/air interface (indicated by the dash line in the figure), the fluorescence compared to that from air plasma is more directional. The direction of the brightest fluorescence highly depends on the relative position between the laser focus and liquid target. In contrast, the fluorescence from the air plasma is more uniform as shown in Fig. 4(b). The theoretical model of liquid plasma is more complicated than that of air plasma. More physical processes are involved in the ionization process. Some theoretical models have been proposed to explain THz generation from liquids [49–52]. In this section, the properties of THz emission from liquid material are discussed.

A. Optimal Angle of Optical Incidence

Figure 5(a) shows a typical THz waveform from a 210 μm flowing water line detected through electro-optic sampling (EOS) with a 2-mm-thick ZnTe crystal. The laser pulse energy is 0.4 mJ with p polarization. The setup is initially optimized with a single-color air plasma. By inserting the flowing water line at the laser focus and increasing the pulse duration of the laser pulses, the THz signal generated from liquid water is measured. With a fast Fourier transform, the spectrum shows that the

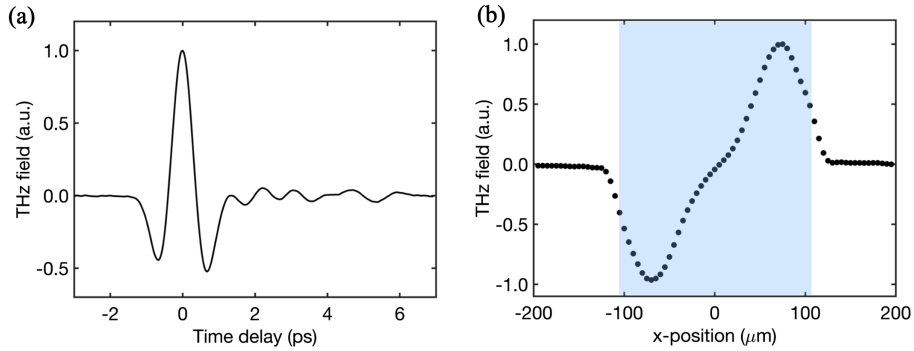


Fig. 5. (a) Typical THz waveform generated from a 210 μm water line. The optical pulse energy is 0.4 mJ. (b) Position dependence of the THz peak field in the x direction.

center frequency locates 0.5 THz with a bandwidth of 0.7 THz. Compared to the signal from air plasma, the bandwidth is relatively narrower, which could result from the high absorption of liquid water at higher THz frequency and the subpicosecond pulse excitation.

The THz yield highly depends on the relative position of the optical focus to the liquid. For a liquid line, the angle of incidence continuously varies when the line is moved across the focus in the x direction (shown in Fig. 2). The dependence of the THz peak field on the x position is plotted in Fig. 5(b). The rectangle shadow shows the diameter of the water line. By scanning the water line in the x direction, two peak values with an opposite polarity are obtained. We have checked that all waveforms at different x positions have a similar waveform without obvious time-delay difference. A similar result is also observed when a water film is used as a source when the angle of incidence is varied by rotating the liquid film [53]. For the case of a 210 μm water line, the maximum signal is measured in the forward direction when the optical focus is about 75 μm from the center of the line. The corresponding angle of incidence is ∼46°, which is determined by asymmetry optical excitation, projection of dipole component in x direction, and total internal reflection of THz wave at the liquid/air interface [31,53].

B. Optimal Pulse Duration for Optical Excitation

For either single-color or two-color laser excited air plasma, the THz signal is always maximized when the pulse is or close to Fourier transform limited pulse. In this case, the pulse duration is shortest for the system. Accordingly, the highest peak power produces a bright air plasma, in which the white light (WL) fluorescence from air plasma is also maximized. However, this behavior is different in a liquid plasma. Figure 6(a) shows the scattered WL spectrum sideway from a water plasma. The black line shows the spectrum without filtering. The saturated signal is from the scattered laser, which has a central wavelength of 800 nm. Besides, a broadband WL is recorded with a central wavelength at 500 nm and a side peak at 657 nm. Here, the WL is generated from the self-phase modulation and self-steepening in liquid water [54,55].

To obtain the dependence on pulse duration, a shortpass filter is used to filter out the laser, and the integrated WL intensity versus pulse duration is recorded and shown as the blue squares in Fig. 6(b). The WL intensity from a water line increases first with the increased pulse duration and saturates when the pulse duration is about 600 fs. The dependence of the THz peak field on the pulse duration is also measured in the same setup, which is plotted in the black dots. Specifically, the THz peak field is maximized when the pulse duration is about 370 fs and then drops fast with the increased pulse duration.

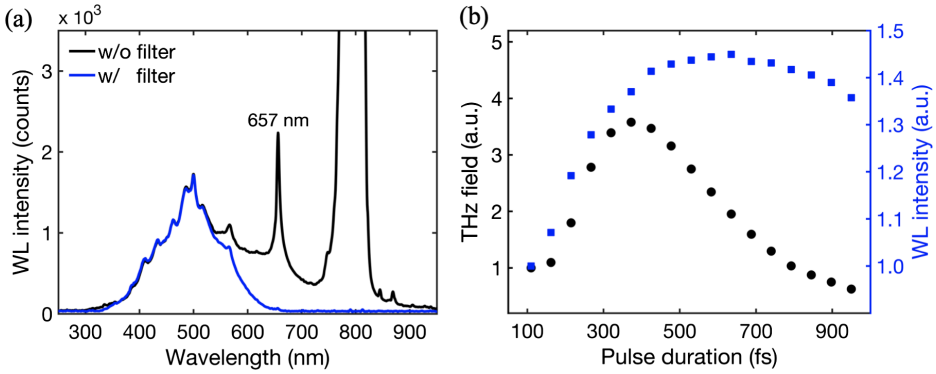


Fig. 6. (a) Scattered white light (WL) intensity detected sideway of a water plasma. The laser is centered at 800 nm. The blue line shows the spectrum after filtering out the pump beam by using a shortpass filter. For comparison, the result without filter is plotted in the black line. (b) Dependence of the THz peak field and WL intensity on the excitation pulse duration, respectively.

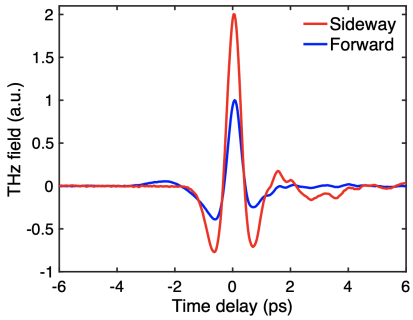


Fig. 7. Comparison of THz waveforms measured in the sideway and forward directions via EOS.

By controlling the pulse duration of excitation, both WL and the THz peak field are enhanced, which could be explained by the photoionization process in liquid. Near the focal point, initial ionization happens through the process of multiphoton absorption or tunneling ionization. The ionized electrons are accelerated by the remaining part of the pulse through the inverse Bremsstrahlung effect [55]. Subsequently, more molecules are ionized through collisions with the accelerated electrons possessing a kinetic energy greater than the ionization potential. This process is called avalanche ionization. In gas material, the mean free time of electrons is typically a few hundreds of femtoseconds, which is longer than or comparable to the pulse duration. Accordingly, the collision does not play an important role in the ionization process. However, in the photoionization of liquid, avalanche ionization is crucial since the much higher ($\sim 10^3$) molecular density comparing with that of gas leads to a much shorter mean free time ($\sim$1 fs) [56]. On the other hand, liquid has a lower ionization potential than gas material, which makes it easier to trigger the multiphoton ionization process. Therefore, the optimal pulse duration for THz generation in liquid results from maximizing the signal through both multiphoton and avalanche ionization. A detailed calculation of the electron density under different pulse durations for excitation can be found in [29].

C. Sideway Emission

In liquid plasma, the dimensions of the ionization region are about a few microns to a few tens of microns, which are much narrower than those in gas plasma under the same optical excitation conditions [57,58]. The energy losses of the laser are much higher in liquid because of the higher electron density in liquid plasma, which quickly exhausts the laser energy and reduces the peak power below the critical power for ionization. In an analogy to a microplasma in gas material, a strong sideway emission is expected for the steep ponderomotive potential at the focal plane for a length-limited liquid plasma.

By using a THz energy detector or EOS, a strong sideway emission is observed in experiments. Figure 7 shows a comparison of the THz waveforms measured sideway and forward from a 210 μm flowing water line. The peak field in the sideway direction is about twice that in the forward direction. Since the radiation pattern or angular distribution highly depends on the target position near the focus, this measurement is individually optimized for the two configurations (sideway and forward) by scanning the target across the focus both in x and z direction. Besides the short length of plasma, the curved surface of the target and the scattering of the plasma also affect the angular distribution of THz emission. A detailed study of radiation pattern will be very helpful to understand the generation and diffraction of THz emission in far field, which is still a challenge in both experimental and theoretical study. If the radiation pattern can be controlled by tailoring the target geometry, a directional THz emission with a much stronger electric field is expected.

4. THz EMISSION FROM VARIETIES OF LIQUIDS

The demonstration of THz wave generation from liquid water opens a new avenue for THz liquid sources. Different liquid materials can be characterized by parameters, such as viscosity, surface tension, and polarity. Viscosity reflects the friction among liquid molecules. And surface tension describes the resistance of liquid surface against an external force. Polarity of liquid originates from the separation of positive- and negative-charged particles of a molecule. To study THz emission from varieties of liquids is meaningful to help understand the electron dynamics in a liquid photoionization process.

A. Liquid Nitrogen

As the most common cryogenic liquid, LN shows great distinctions with respect to common liquids like water. Specifically, LN has an extremely low viscosity (164 mPa · s at −196°C) and surface tension (8.85 mN/m at −196°C). As a nonpolar liquid, LN also shows a low absorption at THz range. The absorption coefficient is 0.8 cm^{-1} at 1 THz [59]. THz generation from LN was first demonstrated by Balakin *et al.* [24], in which the reflected THz wave from the surface of LN is detected in both single- and double-pump excitation. A comparable THz power to the generation from air plasma was detected. In 2020, E *et al.* demonstrated broadband THz wave generation from a flowing LN line at ambient environment [23]. A broader bandwidth than that from water is observed, which indicates that absorption at THz frequency hinders the bandwidth of THz signal generated from liquid water.

B. Liquid Metal

In recent years, metal targets have been widely studied under relativistic laser excitation. When a high intensity laser pulse ($I > 10^{18}$ W/cm^2) is focused onto a target, relativistic electrons are produced with a velocity approaching the speed of light [60]. Relativistic laser-plasma interaction draws attention since it generates bright x rays and highly collimated electron beams with energies in the 10–100 MeV regime [61]. Meanwhile, being accelerated by the laser wakefield, self-trapped electron bunches are produced with a longitudinal dimension in the order of the laser pulse duration [62]. Such a short electron bunch can generate intense THz radiation through a variety of mechanisms, such as synchrotron radiation [63], diffraction radiation [64], and transition radiation [65]. Recently, it has been observed by Liao *et al.* that THz pulse energy exceeding millijoule (mJ) is generated from picosecond laser-irradiated metal foils [66]. For the experiments related to relativistic

Table 2. Comparison of Common THz Plasma Sources in Non-Relativistic and Relativistic Region

	Non-Relativistic Region		Relativistic Region
Laser intensity for excitation (W/cm^2)	$<10^{13-18}$		$>10^{18}$
Target preference	Gas, cluster	Liquid	Solid (Metal)
Pulse duration	Shortest	Subpicosecond (unresolved)	(unresolved)
THz generation mechanism	pondermotive force; four wave mixing; asymmetry field induced current.		coherent transition radiation; sheath field acceleration; transient electron currents.

region, single-shot measurement is used for the large laser facility with a low repetition rate.

In the non-relativistic region ($I < 10^{18}$ W/cm^2), gas and cluster are more favorable since the target degradation is not an issue for high repetition rate pulses. Table 2 shows a comparison of common THz plasma sources in non-relativistic and relativistic region with some unresolved challenges. The flowing metal target provides a possibility to study THz generation from metal material in the non-relativistic region with high repetition rate lasers.

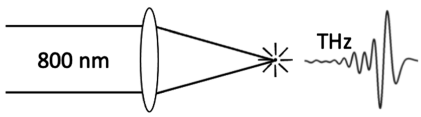
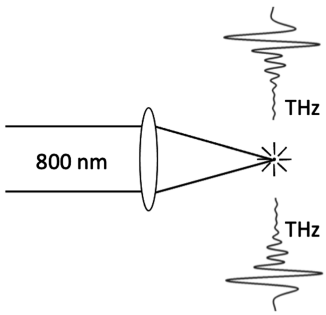
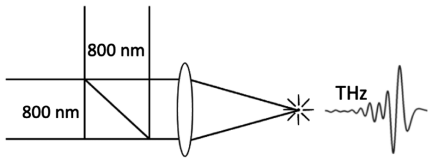
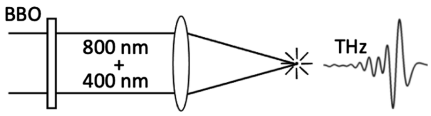
Liquid gallium (Ga) is first demonstrated as a liquid metal source for THz wave generation because of its chemical stability, physical safety, and accessibility. In 2020, Cao *et al.* investigated the THz emission from a flowing Ga line under single-color excitation [25]. The preliminary result shows that liquid Ga generates a stronger signal than that from liquid water. By scanning the Ga line across the laser focus, flipped waveforms are detected at opposite angle of incidence, which shows a similar trend to that of water. Meanwhile, the much thinner penetration depths in liquid metal for both the laser and THz wave make the generation process take place only at the surface of the liquid target. With two-color excitation, no obvious enhancement in the THz field is observed, which demonstrates that liquid metal presents a different mechanism from that of liquid water. More detailed THz generation from a flowing liquid gallium line can be found in [25,26]. Additionally, gallium droplets are studied for a directional THz radiation [27]. The result showcases that the THz polarization highly depends on the second-order susceptibility.

5. FUTURE PERSPECTIVES

Since the first demonstration of broadband THz wave generation from water in 2017 [19], there have been many groups showing great interests in the study of THz emission from liquids. Based on the recent published reports, some key observations of THz emission from liquid targets are summarized in the following:

(1) THz wave emission from water is stronger than that from single-color air plasma under the comparable laser condition.

Table 3. Comparison of THz Field Generated from Water under Different Excitation Geometries

	Excitation Geometry	Detection	THz Field (kV/cm)[a]
Single-color excitation		Forward	~10
		Sideway	~20
Double-pump excitation		Forward	~30
Two-color excitation		Forward	~100

[a]The estimated typical strength of THz field highly depends on the parameters of optical excitation and dimensions of the liquid target. Here, the laser pulse energy for excitation is about 1 mJ.

(2) Excitation with longer optical pulse duration (subpicosecond) in liquid is better than the femtosecond pulse excitation.
(3) Brightest WL fluorescence does not offer the strongest THz signal at the same time.
(4) Sideway THz emission is stronger than that from the forward direction.
(5) Liquid metal (Ga) emits a stronger and broader THz signal than that from liquid water.

A comparison is summarized in Table 3 of different optical excitation geometries and the corresponding THz fields.

However, obstacles still remain in the pursuit. Major renovations and replenishments need to be accomplished beyond the current state-of-the-art. They are as follows:

(1) Improvement of THz generation efficiency in liquids. Two-color excitation [67] and double-pump excitation [68–70] have been tested in a liquid target and showed a limited enhancement of THz signal. Alternative methods such as different wavelength excitation and external field control need to be put forth to either increase plasma density or manipulate electron dynamic.
(2) Control of THz radiation pattern and bandwidth. By tailoring the target geometry and optical excitation configuration, it is expected to acquire more directional THz radiation with a broad bandwidth.
(3) Ultrafast plasma dynamics in liquid plasma. A thorough simulation or theoretical model for liquid plasma is going to help understand the liquid plasma physics in depth.
(4) Currently, the mechanism of THz wave emission under laser excitation is different in the non-relativistic and relativistic region. With the development of laser technology, the output of the table-top laser system is approaching the relativistic region. A systematic study of power dependence in THz generation covering two regions will shed light to the rich physics beneath laser–matter interaction.
(5) THz wave generation from liquid without photoionization. Current liquid emitters are based on the ionization process to provide electrons, which limits the application in biological study.

Exploration of THz emission from liquids is an interdisciplinary, innovative, and important project, from which peculiar electron dynamics can be unveiled and advanced technology for THz sensing/spectroscopy would be developed.

Funding. Air Force Office of Scientific Research (FA9550-21-1-0300, FA9550-21-1-0389); National Science Foundation (ECCS-1916068); Army Research Office (W911NF-17-1-0428); Russian Science Foundation (19-12-00097); National Natural Science Foundation of China (12074272); Beijing Municipal Natural Science Foundation (JQ18015).

Acknowledgment. The authors would like to thank Qi Jin, Kareem Garriga Francis, Fang ling, Yuqi Cao, Jianming Dai, and Evgenia Ponomareva. A. Tsypkin and S. Kozlov are supported by the RSF. L. Zhang and C. Zhang are supported by the Beijing Natural Science Foundation and the NSFC.

Disclosures. The authors declare no conflicts of interest.

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