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ABSTRACT

Metals have been studied as terahertz sources for decades. Recent research has shown the potential of metals in generating extremely high THz pulse energy excited by intense laser pulses. To avoid the metal surface debris caused by laser pulses, here, we report the results of the broadband terahertz wave emission from a flowing liquid metal line excited by subpicosecond laser pulses. The THz signal emitted from the liquid gallium line shows the stronger field with the broader bandwidth compared to the signal from water under the identical optical excitation conditions. Our preliminary study suggests that the liquid metals have the potential to serve as efficient and powerful THz sources for the intense lasers with a high repetition rate.

Over the last decade, several terahertz (THz) frontiers such as condensed-matter dynamics,[1] strong THz field-matter interaction,[2–4] imaging applications,[5] and nonlinear THz spectroscopy,[6–8] have attracted considerable interest. Taking these opportunities requires the development of powerful and efficient THz sources. One way is to use laser-induced plasma excited by ultra-intense laser pulse on solid or gas targets.[2,9,10] Gopal *et al.* reported the 0.7 mJ THz radiation generation from the rear surface of a 5 μm thick stretched titanium metal foil with an intense femtosecond laser pulse, wherein the sheath acceleration dominates the ion acceleration process.[9] Also, an electric field of more than 8 MV/cm of THz waves excited by two-color air-plasma was obtained with the help of combining a thin dual-wavelength half-waveplate and a Brewster-angled silicon window.[2]

Metallic targets have been considered as great THz radiation sources because of their relative lower ionization thresholds and higher molecule densities compared with gas, which allows THz wave generation using lower pump energy.[11] In addition, there is no need to consider the phonon absorption compared to solid crystals.[12] Early studies on THz wave generation from metallic targets have been widely reported.[4,9,13–17] Specifically, an intense and coherent THz wave emission with a pulse energy as high as 55 mJ from a copper foil target irradiated by a high-intensity picosecond laser pulse has been released recently.[4] Liquid metal is taken into consideration because it can provide a pristine, unperturbed smooth surface repeatable for continuous laser shots.

In this Letter, we report the use of a flowing liquid gallium (LG) line as a THz radiation emitter. The THz signal generated from LG by single-color optical excitation is collected in a transmission geometry. THz wave emission from air, liquid water, and liquid metal with identical excitation conditions is measured. THz signals from liquids (LG and water) are stronger than those from air. Also, a 1.7 times enhancement of THz peak field is obtained from the LG line compared with that from water. Similar polarity flips of THz electric field according to the relative position between the incident laser and target liquid line are observed for both cases of LG and water. In addition, it is measured that a longer optical pulse duration works better for LG, indicating that the ionization process is likely attributed to the cascade ionization. The ionized electrons are accelerated by the laser ponderomotive force, and the THz wave is radiated when they pass through the metal–air interface.

For the targets, we consider the metals with relative low melting points and convenience to obtain. Table I lists the melting point, surface tension, density, viscosity, and ionization energy of the candidates at certain temperatures. The most commonly used liquid metal is the mercury because of the lowest melting point among all metals; however, it is not considered in our experiments due to the toxicity concern. Among all metals in this Table I, liquid gallium is a promising candidate for a THz source since the melting point of gallium (Ga) is just above the room temperature (30 °C). Also, compared with other candidates listed in Table I, Ga owns better chemical stability and

TABLE I. Physical properties for selected metals.[20–22] The surface tension, density, and viscosity of each candidates are at the temperature listed in the first column.

Metal targets	Melting point (°C)	Surface tension (N/m)	Density (g/cm^3)	Viscosity (Pa·s)	Ionization energy (eV)
Mercury (Hg) (at 20 °C)	−38.8	0.487	13.534	0.0015	10.44
Cesium (Cs) (at 60 °C)	28.5	0.675	1.843	0.0058	3.90
Gallium (Ga) (at 35 °C)	29.8	0.708	6.109	0.0020	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

physical safety, which make it an ideal liquid metal target and widely used in x-ray generation applications as well.[18,19]

The surface tension of LG at 35 °C is 0.708 N/m, which is more than 10 times higher than that of water (0.0690 N/m) at the same temperature.[23] The high surface tension helps to form a smoother surface of a flowing liquid line. Furthermore, the density of LG (6.109 g/cm^3 at 35 °C) is larger than that of solid gallium (5.91 g/cm^3).[21] A solid Ga ingot with 99.998% purity was melted into and maintained at the liquid phase at 35 °C during all measurements.

The optical setup of THz wave generation from liquid metal employs a Ti:sapphire amplified laser with the center wavelength of 800 nm and the repetition rate of 1 kHz. The p-polarized laser pulse with the energy of 0.4 mJ and the pulse duration of 370 fs is focused by a 50 mm focal length lens at the liquid metal line (LML). The laser focal size on the LML and the Rayleigh length of the laser beam are 5 μm and 51 μm, respectively. The schematic diagram of the setup is shown in Fig. 1(a). The movement of the liquid line near the focus along the x and z direction is finely controlled by a two-dimensional translation stage. The forward propagating THz signal is detected by a standard electro-optical sampling (EOS) using a 3 mm thick ⟨110⟩ cut ZnTe crystal. More details of the experimental setup can be found in an earlier work.[24] A commercial peristaltic pump is used to create a LG line flowing at 3.8 m/s steadily with a diameter of 210 μm as shown in Fig. 1(b). As the flow rate of the liquid metal is greater than 1 m/s, the liquid target at the focal point is replenished and restored after each laser pulse under the laser repetition rate of 1 kHz.

The temporal THz waveforms from air, water, and LG are recorded under the same optical excitation conditions, as shown in Fig. 2(a). It is worth noticing that the signal from air plasma is enlarged by 10 times in the plot. When optical pulse duration is varied from 65 fs to 420 fs by controlling the negative chirp, we find that the optimal pulse durations for LG and water are both 370 fs. Positive and negative chirp give the similar results. All the THz signals as shown in Fig. 2(a) are measured with the identical optical pulse duration of 370 fs and pulse energy. The diameter of the liquid water line and the LG line is produced using a same needle with an inner diameter of 210 μm, and the laser focal positions on the water and LG lines are

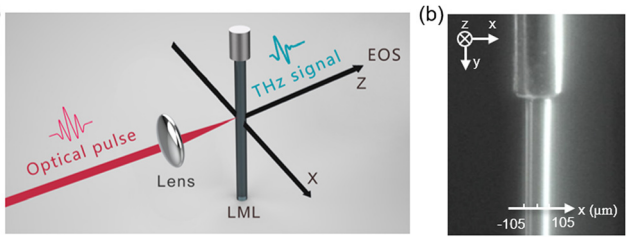

FIG. 1. (a) Schematic of the experimental setup. A two-dimensional translation stage is used to control the position of the x-axis and the z-axis of the LML. The optical beam propagates along the z-axis, and THz emission is measured by a standard EOS in the forward direction. (b) Photo of a flowing LG line in ambient air. The diameter of the LG line is 210 μm. The high surface tension of LG makes an excellent surface quality.

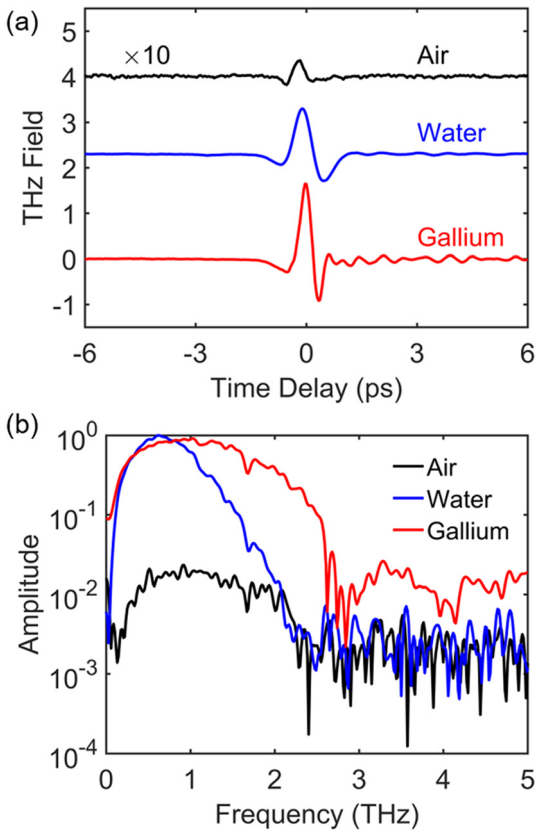

FIG. 2. Comparison of THz waves generated from air and a 210 μm diameter lines of water and gallium with a single-color excitation scheme. (a) Comparison of THz field strengths in air plasma, water, and liquid gallium. The THz signal from single color air plasma is enlarged by 10 times. (b) Corresponding comparison in the frequency domain.

optimized, respectively, to obtaining the maximum THz field in the forward direction.

Remarkably, the THz signal from the LG is much stronger than the signal from the air. Considering air-plasma which prefers to a shorter pulse duration, the THz signal from air excited by the shorter pulse is also measured, and the result is still one-order weaker than the LG signal. The THz electric field generated from ionized LG is 1.7 times stronger than that from water at 35 °C. This might be attributed to the higher density of ionized electrons from the LG line. The corresponding comparison in the frequency domain is plotted in Fig. 2(b). It is noteworthy that the THz signal from LG has a broader bandwidth than that from water and a stronger spectral amplitude in a high frequency. The spectrum of LG contains more high frequency components than the water spectrum because of the high absorption of THz radiation by liquid water at a higher frequency. The frequency bandwidth of the THz signal emitted from the LG l obtained in our setup is limited by the detector phonon bandwidth of crystal ZnTe.

To further study the THz field strength dependence on the focal location on the LG line, the LG line is scanned along the x-axis. Note the z position of the two-dimensional stage is optimized for the THz field previously. We scan the liquid lines from the negative position to the positive position along the x-axis for each measurement, and the normalized THz strength of both water and LG lines as a function of the x position are plotted in Fig. 3. When x equals to 0, the optical pump pulse is focused at the center of the LG line.

The curves show that both THz signals generated from water and LG possess two peaks but with reversed polarity. This is caused by the flipped dipole for the opposite incident angle.[25] The dashed black and red lines in the figure illustrate the position of the peak THz fields of water and LG, and the separation between the two maxima of the LG signal is larger than the case of water. The x positions of maximum peak THz field from water and LG lines are 70 μm and 102 μm, respectively. This means that the positions where the peak THz fields are emitted from the LG line are closer to the edge of the liquid line. Note that the diameter of the liquid line and the laser focused spot size are 210 μm and 5 μm, respectively. Thus, in the case of LG, only part of the optical pulse is focused at the edge of the liquid line when the peak THz field is generated, which might be explained by the low penetration depth of LG for both optical and THz wavelengths. In addition, both p and s-polarized THz fields is measured with the p-polarized pump beam. We find that the p-polarized THz field is one order of magnitude higher than the s-polarized signal.

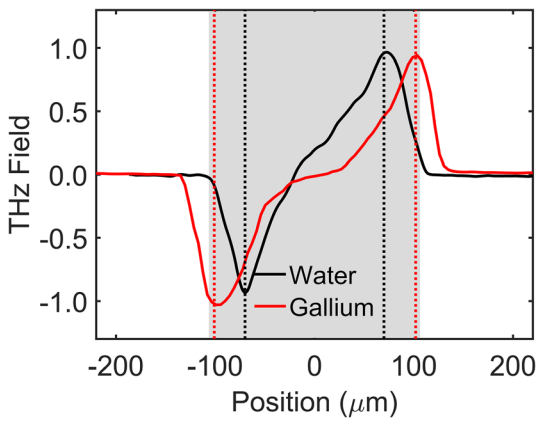

FIG. 3. Normalized THz field strength as a function of the x-axis position from liquid water and LG lines. The dashed black and red lines represent the x-axis position of the maximum THz fields for water and LG, respectively. The width of the gray area stands for the diameter of the LG line. The positions for obtaining the maximum THz field strengths for water and LG are 70 μm and 101 μm, respectively.

The mechanism of the THz wave from the LG line might be attributed to the coherent transition radiation when the laser-induced electrons pass through the metal-air interface.[14,17] The skin depth of LG at both optical (800 nm) and THz (~300 μm) wavelengths are estimated to be around 7.7 nm and 60 nm, respectively, which is much less than the diameter of LG line (210 μm). Thus, ionization starts from the surface at LG. Since a longer pulse duration works better for THz wave generation from LG, like water, enough seed electrons are ionized through multiphoton ionization at the front part of the optical pulse. Then cascade ionization occurs.[25,26] The mean-free-time of electrons in ionized LG is estimated to be less than 1 fs, which is much shorter than the laser pulse duration. This indicates that the collisions of electrons continuously occur for a long period of time and leads to an exponential increase in the number of electrons.[26] Furthermore, another possible reason for the THz radiation emission enhancement from LG than water is the large number of electrons ionized from LG due to the higher molecular density of gallium. After the ionization process, the electrons are accelerated by the ponderomotive force as a result of the non-uniform density gradient distribution of plasma, and THz wave is emitted when the electrons cross the boundary of LG and air.

In summary, we report the preliminary experimental data to reveal the possibility of strong THz emission from the LG line excited by subpicosecond laser pulses. Although the flip of THz waveforms from LG observed by scanning along the x-axis shows similar characteristics with water signal, the mechanism of THz waves generation from liquid metals likely differs from the generation process of liquid water. The strongest THz field we measure from LG with the forward detection is 0.6 kV/cm, but we believe higher THz field can be obtained by increasing pump pulse energy and using sideway detection in later experiments. Further experiments still need to be conducted. The THz wave emission pattern, especially the sideway and the backward direction, is an important experiment to understand the THz radiation mechanism. In addition, the THz wave emission from different liquid metal targets will be investigated to acquire a strong THz source. One other good liquid metal candidates is Galinstan, which is a eutectic alloy with its composition of gallium, indium, and tin. This eutectic alloy, with its melting between −19 °C and +11 °C dependent on the ratio of compositions, is at the liquid state at room temperature. Our results suggest that liquid metal targets have the potential to be strong THz sources for intense lasers with a high repetition rate and may provide a perspective to study the physics of laser-metal interactions.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

[1]T. Kampfrath, K. Tanaka, and K. A. Nelson, "Resonant and nonresonant control over matter and light by intense terahertz transients," Nat. Photonics **7**, 680 (2013).

[2]T. I. Oh, Y. J. Yoo, Y. S. You, and K. Y. Kim, "Generation of strong terahertz fields exceeding 8 MV/cm at 1 kHz and real-time beam profiling," Appl. Phys. Lett. **105**(4), 041103 (2014).

[3]X. Li, T. Qiu, J. Zhang, E. Baldini, J. Lu, A. M. Rappe, and K. A. Nelson, "Terahertz field-induced ferroelectricity in quantum paraelectric $SrTiO_3$," Science **364**(6445), 1079 (2019).

[4]G. Liao, Y. Li, H. Liu, G. G. Scott, D. Neely, Y. Zhang, B. Zhu, Z. Zhang, C. Armstrong, E. Zemaityte, P. Bradford, P. G. Huggard, D. R. Rusby, P. McKenna, C. M. Brenner, N. C. Woolsey, W. Wang, Z. Sheng, and J. Zhang, "Multimillijoule coherent terahertz bursts from picosecond laser-irradiated metal foils," Proc. Natl. Acad. Sci. U. S. A. **116**, 3994 (2019).

[5]M. A. Brun, F. Formanek, A. Yasuda, M. Sekine, N. Ando, and Y. Eishii, "Terahertz imaging applied to cancer diagnosis," Phys. Med. Biol. **55**(16), 4615 (2010).

[6]X.-C. Zhang, A. Shkurinov, and Y. Zhang, "Extreme terahertz science," Nat. Photonics **11**(1), 16 (2017).

[7]M. Tanaka, H. Hirori, and M. Nagai, "THz Nonlinear Spectroscopy of Solids," IEEE Trans. Terahertz Sci. Technol. **1**(1), 312 (2011).

[8]S. Baierl, M. Hohenleutner, T. Kampfrath, A. Zvezdin, A. Kimel, R. Huber, and R. Mikhaylovskiy, "Nonlinear spin control by terahertz-driven anisotropy fields," Nature Photonics **10**(11), 715–8 (2016).

[9]A. Gopal, P. Singh, S. Herzer, A. Reinhard, A. Schmidt, U. Dillner, T. May, H.-G. Meyer, W. Ziegler, and G. G. Paulus, "Characterization of 700 μJ T rays generated during high-power laser solid interaction," Opt. Lett. **38**(22), 4705 (2013).

[10]F. Buccheri and X.-C. Zhang, "Terahertz emission from laser-induced microplasma in ambient air," Optica **2**(4), 366 (2015).

[11]E. G. Gamaly, A. V. Rode, and B. Luther-Davies, "Ablation of solids by femtosecond lasers: Ablation mechanism and ablation thresholds for metals and dielectrics," Phys. Plasmas **9**(3), 949 (2002).

[12]B. Ferguson and X.-C. Zhang, "Materials for terahertz science and technology," Nat. Mater. **1**(1), 26 (2002).

[13]H. Hamster, A. Sullivan, S. Gordon, and R. W. Falcone, "Short-pulse terahertz radiation from high-intensity-laser-produced plasmas," Phys. Rev. E **49**(1), 671 (1994).

[14]W. P. Leemans, C. G. R. Geddes, J. Faure, C. Tóth, J. Van Tilborg, C. B. Schroeder, E. Esarey, G. Fubiani, D. Auerbach, and B. Marcelis, "Observation of terahertz emission from a laser-plasma accelerated electron bunch crossing a plasma-vacuum boundary," Phys. Rev. Lett. **91**(7), 074802 (2003).

[15]K. Filip, K. Petr, and C. Jean-Louis, "Optical rectification at metal surfaces," Opt. Lett. **29**(22), 2674 (2004).

[16]E. V. Suvorov, R. A. Akhmedzhanov, D. A. Fadeev, I. E. Ilyakov, V. A. Mironov, and B. V. Shishkin, "Terahertz emission from a metallic surface induced by a femtosecond optic pulse," Opt. Lett. **37**, 2520 (2012).

[17]G. Liao, Y. Li, Y. Zhang, H. Liu, X. Ge, S. Yang, W. Wei, X. Yuan, Y. Deng, B. Zhu, Z. Zhang, W. Wang, Z. Sheng, L. Chen, X. Lu, J. Ma, X. Wang, and J. Zhang, "Demonstration of coherent terahertz transition radiation from relativistic laser-solid interactions," Phys. Rev. Lett. **116**, 205003 (2016).

[18]D. Von Der Linde, K. Sokolowski-Tinten, C. H. Blome, C. Dietrich, P. Zhou, A. Tarasevitch, A. Cavalleri, C. W. Siders, C. P. J. Barty, J. Squier, K. R. Wilson, I. Uschmann, and E. Forster, "Generation and application of ultrashort x-ray pulses," Laser Part. Beams **19**(1), 15 (2001).

[19]G. Korn, A. Thoss, H. Stiel, U. Vogt, M. Richardson, T. Elsaesser, and M. Faubel, "Ultrashort 1-kHz laser plasma hard x-ray source," Opt. Lett. **27**(10), 866 (2002).

[20]A. V. Grosse, "Surface tension of the alkali metals from the melting point to the critical region," J. Inorg. Nucl. Chem. **30**(5), 1169 (1968).

[21]V. K. Prokhorenko, V. V. Roshchupkin, M. A. Pokrasin, S. V. Prokhorenko, and V. V. Kotov, "Liquid gallium: Potential uses as a heat-transfer agent," High Temp. **38**(6), 954 (2000).

[22]D. W. G. White, "The surface tensions of indium and cadmium," Metall. Mater. Trans. B **3**(7), 1933 (1972).

[23]A. Karbalaei, R. Kumar, and H. J. Cho, "Thermocapillarity in microfluidics—A review," Micromachines **7**, 13 (2016).

[24]Q. Jin, E. Yiwen, K. Williams, J. Dai, and X.-C. Zhang, "Observation of broadband terahertz wave generation from liquid water," Appl. Phys. Lett. **111**(7), 071103 (2017).

[25]Q. Jin, E. Yiwen, S. Gao, and X.-C. Zhang, "Preference of subpicosecond laser pulses for terahertz wave generation from liquids," Adv. Photonics **2**(1), 1 (2020).

[26]P. K. Kennedy, "A first-order model for computation of laser-induced breakdown thresholds in ocular and aqueous media. I. Theory," IEEE J. Quantum Electron **31**(12), 2241 (1995).