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13. SUPPLEMENTARY NOTES

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14. ABSTRACT

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RPPR Final Report
as of 14-Jul-2021

Agency Code: 21XD

Proposal Number: 70858EL **Agreement Number: W911NF-17-1-0428**

INVESTIGATOR(S):

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DUNS Number: 041294109 EIN: 160743209
Report Date: 29-Sep-2021 Date Received: 08-Jul-2021
Final Report for Period Beginning 01-Sep-2017 and Ending 29-Jun-2021
Title: Extreme THz Science
Begin Performance Period: 01-Sep-2017 **End Performance Period:** 29-Jun-2021
Report Term: 0-Other
Submitted By: Xi-Cheng Zhang Email: xi-cheng.zhang@rochester.edu
Phone: (585) 275-0333
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 3 **STEM Participants:** 10

Major Goals: Matters are generally classified within four phase 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.
Our group is the first group in the world, who demonstrated broadband THz wave generation from a flowing liquid target through laser induced micro-plasma. Liquid target as THz source presents unique properties. Specifically, liquids have a comparable material density to that of solids, meaning that laser pulses over a certain area will interact with three orders more molecules than an equivalent cross-section of gases. In contrast with solid targets, the fluidity of liquid allows each laser pulse to interact with a fresh area on the target. Thus, the material damage or degradation is not an issue with the high repetition rate intense laser pulses. These properties make 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. By employing the liquids target, we create a new topic in the THz community, which is “THz liquid photonics” to explore extreme THz science from a new perspective.

Accomplishments: This final report covers research project under ARO support at The Institute of Optics, University of Rochester. Our goal is to explore extreme THz science with short laser pulses. We are proudly reporting that we have achieved all the major tasks we proposed during this very challenging period (pandemic Covid-19). Significant progress was made by our group in the topic of THz liquid photonics under the support of this grant. We published over 34 peer-reviewed papers and gave 48 colloquium and invited presentations in international conferences. Some of our results are summarized in the following: (1) Demonstrate broadband THz generation from flowing liquid targets under single/two-color optical excitation. (2) The preference of longer pulse during for optical excitation in THz generation from liquids. (3) THz generation from different liquids, such as cryogenic liquid (liquid nitrogen), liquid metal, liquids with different polarities. (4) Enhanced generation efficiency under double-pump optical excitation. (5) Strong sideway THz generation from liquid targets. (6) Development of the liquid circulating system for creating free-standing liquid targets with the high-repetition rate laser excitation.

Training Opportunities: Involvement: Two PhD students Kang Liu and Qi Jin were graduated under the support of his grant. Outstandingly, Qi Jin published 4 first-author papers during his PhD program. Kaia Williams, Greg Lier, and Justin Murante who were undergraduate students of UR, were supported by 2018 UPAP, 2019 URAP and 2021 URAP (Undergraduate Research Apprenticeship Program) for their summer internship. Two PhD students Kareem Garriga and Steven Fu are working on the project of extreme THz emission from liquids.

RPPR Final Report

as of 14-Jul-2021

Results Dissemination:

Colloquium Presentations

(1) "From Speculation to Demonstration: THz Wave Emission from Water," Physics Department, Capital Normal University of Beijing, Beijing, China, June 19, 2017.
(2) "Observation of Broadband Terahertz Wave Generation from Liquid Water," OSA Nonlinear Optics conference, Hawaii Island, Hawaii, July 19, 2017.
(3) "Observation of Broadband Terahertz Wave Generation from Liquid Water," Keynote, IRMMW-THz 2017, Cancun Mexico, August 29, 2017.
(4) "THz Wave Emission from Water," Plenary, Russia-Japan-USA-Europe Symposium on Fundamental & Applied Problems of Terahertz Devices & Technologies (RJUSE TeraTech-2017), Troy, NY, Oct. 2, 2017.
(5) "Terahertz Wave Generation from Liquid Water," 8th International THz-Bio Workshop, ENEA-Frascati, Italy, Oct. 5, 2017.
(6) "Frontiers of THz Photonics," Plenary, the 10th International Photonics and OptoElectronics Meetings (POEM 2017), Wuhan, China, Nov. 4, 2017.
(7) "Puzzle of THz wave emission from liquid water with ultrafast laser pulses," Plenary, MTSA2017&TeraNano-8, Okayama, Japan, Nov. 20, 2017.
(8) "Let THz light shine out of darkness," OSA Traveling Lecture Howard University, Washington DC, April 10, 2018.
(9) "Recent Development on THz Aqueous Photonics," 9th International symposium on ultrafast phenomena and THz wave, Changsha, China, April 23, 2018.
(10)"Let THz light shine out of darkness," GIPO seminar, National Taiwan University, Taipei, Taiwan, May 4, 2018.
(11)"THz Aqueous Photonics and Beyond," Spring Taiwan THz Workshop (2018S T-TW) & Forum, National Tsing Hua University, Hsinchu, May 7, 2018.
(12)"Past, present, and expectation of THz photonics," Seminar, National Chiao Tung University, Hsinchu, May 8, 2018.
(13)"Past, present, and expectation of THz photonics," Seminar, Research Center for Applied Sciences, Academia Sinica, Taipei, May 14, 2018.
(14)"Next Rays? T-ray!" (In Mandarin), International day of light symposium, National Taiwan University, Taipei, May 16, 2018
(15)"Next Rays? T-ray!" Seminar, National Cheng Kung University, Tainan, May 18, 2018.
(16)"Past, present, and expectation of THz photonics," Seminar, National Sun Yat-Sen University, Kaohsiung, May 21, 2018.
(17)"Past, present, and expectation of THz photonics," Seminar, National Chiao Tung University, Tainan, May 22, 2018.
(18)"Next Rays? T-ray!" Seminar, National Central University, Taoyuan, Taiwan, May 23, 2018.
(19)"Mission Impossible: THz Aqueous Photonics," Department of Physics, Applied Physics and Astronomy, Rensselaer Polytechnic Institute, Troy, NY, Sept. 22, 2018.
(20)"Mission Impossible: THz Aqueous Photonics," National Research Center for Optoelectronics, HUST, Wuhan, Oct, 18. 2018.
(21)"Mission Impossible: THz Aqueous Photonics," The Institute of Optics, University of Rochester, NY, USA. Nov. 12, 2018.
(22)"Let THz Light Up the Darkness," 2nd Shaanxi Postgraduate Academic Conference on Optics & Photonics, Xian, China, Nov. 16, 2018.
(23)"Mission Impossible: THz Loves Water," Xian Institute of Optics and Mechanics, Xian, China, Nov. 17, 2018.
(24)"Recent Advances of THz Science and Technology," Physical Society of Taiwan Annual Meeting, National Pingdong University, Pingdong, Tiawan, Feb. 5, 2020.

Invited Conference Presentation

(1) "Observation of Broadband Terahertz Wave Generation from Liquid Water," Qi Jin, Kaia Williams, Yiwen E, Jianming Dai and X. -C. Zhang, Nonlinear Optics Conference, Hawaii, USA, July 19, 2017
(2) "Terahertz Wave Generation from Liquid Water," Qi Jin, Kaia Williams, Yiwen E, Jianming Dai and X. -C. Zhang, THz Bio Conference, Rome, Italy, Oct. 4, 2017.
(3) "THz Wave Emission from Water," Yiwen E, Qi Jin, Kaia Williams, Jianming Dai and X. -C. Zhang, RJUSE TeraTech, Troy, Rensselaser Polytechnic Institute, New York, USA, Oct. 1, 2017.
(4) "THz science, technology, and applications," X.-C. Zhang, Beihang University, Beijing, China, Oct. 19, 2017.
(5) "Last piece of puzzle: THz wave emission from liquid water," X.-C. Zhang, Institute of Physics, Chinese Academy of Science, China, Oct. 27, 2017.
(6) "Last piece of puzzle: THz wave emission from liquid water," X.-C. Zhang, POEM 2017, Wuhan, China, Nov. 4,

2017.
(7) "Automatic spectrum recognition by real-time terahertz spectrometer," Ying-hao Yuan, Chang-Geng Li, Zheng Zhou, Tom Tongue, Xi-Cheng Zhang, POEM 2017, Wuhan, China, Nov. 5, 2017.
(8) "Last Piece of Puzzle." X.-C. Zhang, Beijing Advanced Innovation Center for Imaging Technology, Beijing, China, Nov. 11, 2017.
(9) "THz aqueous photonics," ASILS10, American University of Shajah, Shahja, March 11, 2018.
(10)"THz aqueous photonics," X.-C. Zhang, Plenary talk, 18th National Optical Fiber Communication and 19th Integrated Optics Conference, Changchun, China, July 14, 2018.
(11)"Let's THz light shine out of darkness," X.-C. Zhang, Plenary talk, Light Conference 2018, Changchun, China, July 18, 2018.
(12)"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.
(13)"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.
(14)"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.
(15)"Mission impossible: generation of THz wave from liquid water," X.-C. Zhang, Westlake Forum, Hangzhou, China, Oct. 26, 2018.
(16)"THz Aqueous Photonics and Beyond," French-German THz conference, Kaiserslautern, Germany. April 2-5, 2019.
(17)"Mission Impossible: Generation of THz wave from liquid water," Optics and Photonics 2019, Dubai, AUE. April 15, 2019.
(18)"Next Rays? T-ray!, part I" Siegman International School on Lasers, Rochester, July 27, 2019.
(19)"Next Rays? T-ray!, part II" Siegman International School on Lasers, Rochester, July 31, 2019.
(20)"Challenges and opportunities for THz wave liquid photonics," Tutorial, International Conference on Information Optics and Photonics, Xian, China, Aug. 7, 2019
(21)"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]
(22)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.
(23)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.
(24)Yiwen, X.-C. Zhang, “Recent progress on THz liquid photonics”, the 10th International Symposium on Ultrafast Phenomena and THz Waves (ISUPTW 2021), Chengdu, China, June 16-19, 2021.

Honors and Awards: 2020Dr. Zhang is appointed as Executive Editor in Chief of Light, Science & Application (Nature Springer) 2020-2023.

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
2017	Australian Academy of Science Selby Fellow, Australia

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: Graduate Student (research assistant)
Participant: Kareem Garriga
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Steven Fu
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Kang Liu
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Qi Jin
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Kaia Williams
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Justin Murante
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Greg Lier
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Yiwen E.
Person Months Worked: 7.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: PD/PI
Participant: X.-C. Zhang

Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

ARTICLES:

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Applied Physics Letters
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Volume: 113 Issue: 18 First Page #: 181103
Date Submitted: 8/12/19 12:00AM Date Published: 10/1/18 4:00AM
Publication Location:
Article Title: Terahertz wave generation from liquid water films via laser-induced breakdown
Authors: Yiwen E, Qi Jin, Anton Tcypkin, X.-C. Zhang
Keywords: terahertz generation, terahertz spectroscopy, plasma
Abstract: Understanding the physics of terahertz (THz) wave generation from water is essential for developing liquid THz sources. This letter reports detailed experimental measurements of THz wave emission by focusing intense laser pulses onto water films. The simulation based on a ponderomotive force-induced dipole is supported by the observation of the THz intensity dependence on the laser incidence angle. This work provides fundamental insights into the THz wave generation process in water and an alternative perspective for studying laser-induced breakdown in liquids.
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Journal: Applied Physics Letters
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Publication Location:
Article Title: Terahertz wave emission from a liquid water film under the excitation of asymmetric optical fields
Authors: Qi Jin, Jianming Dai, Yiwen E, Xi-Cheng Zhang
Keywords: terahertz generation, terahertz spectroscopy, plasma
Abstract: Liquid water excited by intense two-color laser pulses radiates electromagnetic waves at terahertz frequencies. Compared with one-color excitation, two-orders of magnitude enhanced terahertz energy are observed by using asymmetric optical excitation with the same total excitation pulse energy and focusing geometry. Modulation of the terahertz field is achieved via the coherent con- trol approach. We find that modulated and unmodulated terahertz energies have, respectively, qua- dratic and linear dependence on the laser pulse energy. This work, as part of terahertz aqueous photonics, paves an alternative way of studying laser-liquid interactions and developing intense ter- ahertz sources.
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Journal: Optics Express
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Date Submitted: 8/12/19 12:00AM Date Published: 4/1/19 4:00AM
Publication Location:
Article Title: High Kerr nonlinearity of water in THz spectral range
Authors: Anton N. Tcypkin, Maksim V. Melnik, Maria O. Zhukova, Irina O. Vorontsova, Sergey E. Putilin, Sergei A
Keywords: Kerr coefficient, THz nonlinearity, Water
Abstract: The values of the nonlinear refractive index coecient for various materials in the terahertz frequency range exceed the ones in both visible and NIR ranges by several orders of magnitude. This allows to create nonlinear switches, modulators, systems requiring lower control energies in the terahertz frequency range. We report the direct measurement of the nonlinear refractive index coecient of liquid water by using the Z-scan method with broadband pulsed THz beam. Our experimental result shows that nonlinear refractive index coecient in water is positive and can be as large as 7?1010 cm2/W in the THz frequency range, which exceeds the values for the visible and NIR ranges by 6 orders of magnitude. To estimate n2, we use the theoretical model that takes into account ionic vibrational contribution to the third-order susceptibility. We show that the origins of the nonlinearity observed are the anharmonicity of molecular vibrations.
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Publication Location:
Article Title: Terahertz wave generation from ring-Airy beam induced plasmas and remote detection by terahertz-radiationenhanced- emission-of-fluorescence: a review
Authors: Kang Liu, Pingjie Huang, Xi-Cheng Zhang
Keywords: terahertz generation, terahertz spectroscopy, plasma
Abstract: With the increasing demands for remote spectroscopy in many fields ranging from homeland security to environmental monitoring, terahertz (THz) spectroscopy has drawn a significant amount of attention because of its capability to acquire chemical spectral signatures non-invasively. However, advanced THz remote sensing techniques are obstructed by quite a few factors, such as THz waves being strongly absorbed by water vapor in the ambient air, difficulty to generate intense broadband coherent THz source remotely, and hard to transmit THz waveform information remotely without losing the signal to noise ratio, etc. In this review, after introducing different THz air-photonics techniques to overcome the difficulties of THz remote sensing, we focus mainly on theoretical and experimental methods to improve THz generation and detection performance for the purpose of remote sensing through tailoring the generation and detection media, air-plasma.
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Volume: 12 Issue: 1 First Page #:
Date Submitted: 8/12/19 12:00AM Date Published: 7/1/19 4:00AM
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Article Title: Strong Terahertz Radiation from a Liquid-Water Line
Authors: Liang-Liang Zhang, Wei-Min Wang, Tong Wu, Shi-Jia Feng, Kai Kang, Cun-Lin Zhang, Yan Zhang, Yu-
Keywords: terahertz generation, terahertz spectroscopy, plasma
Abstract: Terahertz radiation generation from liquid water has long been considered impossible due to strong absorption. A few very recent works reported terahertz generation from water, but the mechanism is not clear and the efficiency demands to be enhanced. We show experimentally that strong single-cycle terahertz radiation with field strength of 0.2 MV cm?1 is generated from a water line (or column) of approximately 200 ?m in diameter irradiated by a mJ femtosecond laser beam. This strength is 100-fold higher than that produced from air using single-color pumping. We attribute the mechanism to the laser- ponderomotive-force-induced current with the symmetry broken around the water-column interface. This mechanism can explain our following observations: the radiation can be generated only when the laser propagation axis deviates from the column center; the deviation determines its field strength and polarity; it is always p polarized no matter whether the laser is p or s polarized. This study
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Date Submitted: 8/12/19 12:00AM Date Published: 5/1/19 4:00AM
Publication Location:
Article Title: Flat liquid jet as a highly efficient source of terahertz radiation
Authors: Anton N. Tcypkin, Evgenia A. Ponomareva, Sergey E. Putilin, Semen V. Smirnov, Sviatoslav A. Shtump
Keywords: terahertz generation, terahertz spectroscopy, plasma
Abstract: In conclusion, we would like to note that we have highlighted the main experimental conditions that have significantly contributed to the effective generation of terahertz radiation in flat liquid jets. We applied a mathematical model of a subpicosecond pulse strong field interaction with a transparent isotropic medium for the generation of terahertz radiation in liquid jets which predicts the results of experiments. We claim that it is necessary to take into account not only the ionization energy of the medium, but also its molecular density and absorption coefficient to obtain the maximum efficiency of terahertz generation. Comparing all selected liquids, acetone shows the highest efficiency of terahertz generation due to the optimal ratio of all parameters of the medium. We show that it is necessary to consider the correlation between the thickness of the jet and the duration of the pump pulse to obtain the most efficient terahertz generation. Quasi-quadratic dependence of the terahertz ener
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Volume: 8 Issue: 1 First Page #:
Date Submitted: 8/12/19 12:00AM Date Published: 6/1/19 4:00AM
Publication Location:
Article Title: Spatial sampling of terahertz fields with sub-wavelength accuracy via probe-beam encoding
Authors: Jiapeng Zhao, Yiwen E, Kaia Williams, Xi-Cheng Zhang, Robert W. Boyd
Keywords: Computational imaging, Terahertz imaging,
Abstract: Recently, computational sampling methods have been implemented to spatially characterize terahertz (THz) fields. Previous methods usually rely on either specialized THz devices such as THz spatial light modulators or complicated systems requiring assistance from photon-excited free carriers with high-speed synchronization among multiple optical beams. Here, by spatially encoding an 800-nm near-infrared (NIR) probe beam through the use of an optical SLM, we demonstrate a simple sampling approach that can probe THz fields with a single-pixel camera. This design does not require any dedicated THz devices, semiconductors or nanofilms to modulate THz fields. Using computational algorithms, we successfully measure 128 × 128 field distributions with a 62-?m transverse spatial resolution, which is 15 times smaller than the central wavelength of the THz signal (940 ?m). Benefitting from the non- invasive nature of THz radiation and sub-wavelength resolution of our system, this simple approach c
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CONFERENCE PAPERS:

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Conference Name: Nonlinear Optics
Date Received: 12-Aug-2019 Conference Date: 16-Jul-2019 Date Published: 22-Jul-2019
Conference Location: Waikoloa Beach, Hawaii
Paper Title: Terahertz Photonics in Liquids
Authors: Q. Jin, Y.W. E, J.M. Dai, L.L. Zhang, C.L. Zhang, A. Tcypkin, S. Kozlov, and X.-C. Zhang
Acknowledged Federal Support: **Y**

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Conference Name: 2018 43rd International Conference on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz 2018)
Date Received: 12-Aug-2019 Conference Date: 09-Sep-2018 Date Published:
Conference Location: Nagoya
Paper Title: THz Aqueous Photonics and Beyond
Authors: Q. Jin, Y.W. E, L.L. Zhang, C.L. Zhang, A. Tcypkin, S. Kozlov, X.-C. Zhang
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Conference Location: Hangzhou, China
Paper Title: Investigation of liquid properties on emitting terahertz wave under ultrashort optical excitation
Authors: Shenghan Gao, Qi Jin, Yiwen E., X.-C. Zhang
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Conference Location: Buffalo, NY, USA
Paper Title: Spatial Measurement of Terahertz Fields by Encoding Probe Beam
Authors: Jiapeng Zhao; Yiwen E; Kaia Williams; Xi-Cheng Zhang; Robert W. Boyd
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Paper Title: Broadband THz Wave Generation from Flowing Liquid Nitrogen
Authors: Yiwen E; Yuqi Cao; Fang Ling; Alexander P. Shkurinov; Yiming Zhu; X.-C. Zhang
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Authors: Yuqi Cao; Yiwen E; Anton Tcypkin; Pingjie Huang; X.-C. Zhang
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Paper Title: Terahertz Wave Emission from Liquid Metal
Authors: Yuqi Cao; Yiwen E; Pingjie Huang; X.-C. Zhang
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Conference Location: Online Only, China
Paper Title: THz liquid photonics and beyond
Authors: Yiwen E., Qi Jin, Jianming Dai, Yuqi Cao, Fang Ling, Kaia Williams, Mervin Lin Pac Chong, Gregoire Le
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Authors: Jianming Dai, Qi Jin, Yiwen E., Kaia Williams, X.-C. Zhang
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DISSERTATIONS:

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Authors: Qi Jin
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Partners

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We had the collaboration with the Capital Normal University, Beijing, China and ITMO University, St. Petersburg, Ru

I certify that the information in the report is complete and accurate:
Signature: X.-C. Zhang
Signature Date: 7/8/21 4:36PM

Extreme THz Science

Final Progress Report

Report Period: 09/01/2017--07/31/2021 (including one-year no-cost-extension)

Grant Contract #: W911NF-17-1-0428
RDRL-ROS-I Proposal Number: 70858-EL

Principal Investigators: **X.-C. Zhang**
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Authors: X.-C. Zhang and Yiwen E

Submitted: Dr. Joe Qiu, ARO

Abstract:

This final report covers research project under ARO support at The Institute of Optics, University of Rochester. Our goal is to explore extreme THz science with short laser pulses. We are proudly reporting that we have achieved all the major tasks we proposed during this very challenging period (pandemic Covid-19). Significant progress was made by our group in the topic of THz liquid photonics under the support of this grant. We published over 34 peer-reviewed papers and gave 48 colloquium and invited presentations in international conferences. Some of our results are summarized in the following: (1) Demonstrate broadband THz generation from flowing liquid targets under single/two-color optical excitation. (2) The preference of longer pulse during for optical excitation in THz generation from liquids. (3) THz generation from different liquids, such as cryogenic liquid (liquid nitrogen), liquid metal, liquids with different polarities. (4) Enhanced generation efficiency under double-pump optical excitation. (5) Strong sideway THz generation from liquid targets. (6) Development of the liquid circulating system for creating free-standing liquid targets with the high-repetition rate laser excitation.

Matters are generally classified within four phase 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.

Our group is the first group in the world, who demonstrated broadband THz wave generation from a flowing liquid target through laser induced micro-plasma. Liquid target as THz source presents unique properties. Specifically, liquids have a comparable material density to that of solids, meaning that laser pulses over a certain area will interact with three orders more molecules than an equivalent cross-section of gases. In contrast with solid targets, the fluidity of liquid allows each laser pulse to interact with a fresh area on the target. Thus, the material damage or degradation is not an issue with the high repetition rate intense laser pulses. These properties make 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. By employing the liquids target, we create a new topic in the THz community, which is “THz liquid photonics” to explore extreme THz science from a new perspective.

1. Accomplished

Papers Published and Submitted

(1) Fabrizio Buccheri, Kang Liu, X.-C. Zhang, "Terahertz Radiation Enhanced Emission of Fluorescence from Elongated Plasmas and Microplasmas in the Counter-propagating Geometry," Appl. Phys. Lett. 111, 091103 (2017); doi: http://dx.doi.org/10.1063/1.4990143.

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(10) 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). http://dx.doi.org/10.1063/1.5054599

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Emission- of- Fluorescence: A Review," Invited paper, Front. Optoelectron., 2019, 12(2): 117–147 https://doi.org/10.1007/s12200-018-0860-7

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1. Training

Involvement: Two PhD students Kang Liu and Qi Jin were graduated under the support of his grant. Outstandingly, Qi Jin published 4 first-author papers during his PhD program. Kaia Williams, Greg Lier, and Justin Murante who were undergraduate students of UR, were supported by 2018 UPAP, 2019 URAP and 2021 URAP (Undergraduate Research Apprenticeship Program) for their summer internship. Two PhD students Kareem Garriga and Steven Fu are working on the project of extreme THz emission from liquids.

2. Dissemination

Colloquium Presentations

(1) "From Speculation to Demonstration: THz Wave Emission from Water," Physics Department, Capital Normal University of Beijing, Beijing, China, June 19, 2017.

(2) "Observation of Broadband Terahertz Wave Generation from Liquid Water," OSA Nonlinear Optics conference, Hawaii Island, Hawaii, July 19, 2017.

(3) "Observation of Broadband Terahertz Wave Generation from Liquid Water," Keynote, IRMMW-THz 2017, Cancun Mexico, August 29, 2017.

(4) "THz Wave Emission from Water," Plenary, Russia-Japan-USA-Europe Symposium on Fundamental & Applied Problems of Terahertz Devices & Technologies (RJUSE TeraTech-2017), Troy, NY, Oct. 2, 2017.

(5) "Terahertz Wave Generation from Liquid Water," 8th International THz-Bio Workshop, ENEA-Frascati, Italy, Oct. 5, 2017.

(6) "Frontiers of THz Photonics," Plenary, the 10th International Photonics and OptoElectronics Meetings (POEM 2017), Wuhan, China, Nov. 4, 2017.

(7) "Puzzle of THz wave emission from liquid water with ultrafast laser pulses," Plenary, MTSA2017&TeraNano-8, Okayama, Japan, Nov. 20, 2017.

(8) "Let THz light shine out of darkness," OSA Traveling Lecture Howard University, Washington DC, April 10, 2018.

(9) "Recent Development on THz Aqueous Photonics," 9th International symposium on ultrafast phenomena and THz wave, Changsha, China, April 23, 2018.

(10) "Let THz light shine out of darkness," GIPO seminar, National Taiwan University, Taipei, Taiwan, May 4, 2018.

(11) "THz Aqueous Photonics and Beyond," Spring Taiwan THz Workshop (2018S T-TW) & Forum, National Tsing Hua University, Hsinchu, May 7, 2018.

(12) "Past, present, and expectation of THz photonics," Seminar, National Chiao Tung University, Hsinchu, May 8, 2018.

(13) "Past, present, and expectation of THz photonics," Seminar, Research Center for Applied Sciences, Academia Sinica, Taipei, May 14, 2018.

(14) "Next Rays? T-ray!" (In Mandarin), International day of light symposium, National Taiwan University, Taipei, May 16, 2018

(15) "Next Rays? T-ray!" Seminar, National Cheng Kung University, Tainan, May 18, 2018.

(16) "Past, present, and expectation of THz photonics," Seminar, National Sun Yat-Sen University, Kaohsiung, May 21, 2018.

(17) "Past, present, and expectation of THz photonics," Seminar, National Chiao Tung University, Tainan, May 22, 2018.

(18) "Next Rays? T-ray!" Seminar, National Central University, Taoyuan, Taiwan, May 23, 2018.

(19) "Mission Impossible: THz Aqueous Photonics," Department of Physics, Applied Physics and Astronomy, Rensselaer Polytechnic Institute, Troy, NY, Sept. 22, 2018.

(20) "Mission Impossible: THz Aqueous Photonics," National Research Center for Optoelectronics, HUST, Wuhan, Oct, 18. 2018.

(21) "Mission Impossible: THz Aqueous Photonics," The Institute of Optics, University of Rochester, NY, USA. Nov. 12, 2018.

(22) "Let THz Light Up the Darkness," 2nd Shaanxi Postgraduate Academic Conference on Optics & Photonics, Xian, China, Nov. 16, 2018.

(23) "Mission Impossible: THz Loves Water," Xian Institute of Optics and Mechanics, Xian, China, Nov. 17, 2018.

(24) "Recent Advances of THz Science and Technology," Physical Society of Taiwan Annual Meeting, National Pingdong University, Pingdong, Tiawan, Feb. 5, 2020.

Invited Conference Presentation

(1) "Observation of Broadband Terahertz Wave Generation from Liquid Water," Qi Jin, Kaia Williams, Yiwen E, Jianming Dai and X. -C. Zhang, Nonlinear Optics Conference, Hawaii, USA, July 19, 2017

(2) "Terahertz Wave Generation from Liquid Water," Qi Jin, Kaia Williams, Yiwen E, Jianming Dai and X. -C. Zhang, THz Bio Conference, Rome, Italy, Oct. 4, 2017.

(3) "THz Wave Emission from Water," Yiwen E, Qi Jin, Kaia Williams, Jianming Dai and X. -C. Zhang, RJUSE TeraTech, Troy, Rensselaser Polytechnic Institute, New York, USA, Oct. 1, 2017.

(4) "THz science, technology, and applications," X.-C. Zhang, Beihang University, Beijing, China, Oct. 19, 2017.

(5) "Last piece of puzzle: THz wave emission from liquid water," X.-C. Zhang, Institute of Physics, Chinese Academy of Science, China, Oct. 27, 2017.

(6) "Last piece of puzzle: THz wave emission from liquid water," X.-C. Zhang, POEM 2017, Wuhan, China, Nov. 4, 2017.

(7) "Automatic spectrum recognition by real-time terahertz spectrometer," Ying-hao Yuan, Chang-Geng Li, Zheng Zhou, Tom Tongue, Xi-Cheng Zhang, POEM 2017, Wuhan, China, Nov. 5, 2017.

(8) "Last Piece of Puzzle." X.-C. Zhang, Beijing Advanced Innovation Center for Imaging Technology, Beijing, China, Nov. 11, 2017.

(9) "THz aqueous photonics," ASILS10, American University of Shajah, Shahja, March 11, 2018.

(10) "THz aqueous photonics," X.-C. Zhang, Plenary talk, 18th National Optical Fiber Communication and 19th Integrated Optics Conference, Changchun, China, July 14, 2018.

(11) "Let's THz light shine out of darkness," X.-C. Zhang, Plenary talk, Light Conference 2018, Changchun, China, July 18, 2018.

(12) "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.

(13) "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.

(14) "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.

(15) "Mission impossible: generation of THz wave from liquid water," X.-C. Zhang, Westlake Forum, Hangzhou, China, Oct. 26, 2018.

(16) "THz Aqueous Photonics and Beyond," French-German THz conference, Kaiserslautern, Germany. April 2-5, 2019.

(17) "Mission Impossible: Generation of THz wave from liquid water," Optics and Photonics 2019, Dubai, AUE. April 15, 2019.

(18) "Next Rays? T-ray!, part I" Siegman International School on Lasers, Rochester, July 27, 2019.

(19) "Next Rays? T-ray!, part II" Siegman International School on Lasers, Rochester, July 31, 2019.

(20) "Challenges and opportunities for THz wave liquid photonics," Tutorial, International Conference on Information Optics and Photonics, Xian, China, Aug. 7, 2019

(21) "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]

(22) 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.

(23) 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.

(24) Yiwen, X.-C. Zhang, “Recent progress on THz liquid photonics”, the 10th International Symposium on Ultrafast Phenomena and THz Waves (ISUPTW 2021), Chengdu, China, June 16-19, 2021.

3. Plans

THz liquid photonics provides a new platform for 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 and solid targets, which should be further studied to draw a thorough picture for the laser-induced ionization process in liquid. Currently, the THz field generated from liquid under single-color excitation is 2 orders higher than that from single-color air plasma. The optimal conditions for the maximum generation efficiency are underexplored. Further experiments include target geometry design, laser pulse shaping, target material testing, external field modulating and so on.

Furthermore, one of the most interesting projects is to study THz wave generation from superfluid such as $_4$He, which flows with no viscosity, and its thermal conductivity increases by a factor of ~10^6 for the temperature at the λ-point. This causes heat in 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 serves as a perfect system for the optical study on the laser-matter interaction by focusing laser beam on the liquid.

4. Honors and Awards

2020 Dr. Zhang is appointed as Executive Editor in Chief of Light, Science & Application (Nature Springer) 2020-2023.

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

2017 Australian Academy of Science Selby Fellow, Australia Nothing to Report

5. Tech Transfer

Nothing to Report

6. Personnel Metrics

Please complete the below tables, providing the information for this reporting period only. Add rows as needed.

(1) Faculty

Name	Percent Supported
Xi-Cheng Zhang	10%

(2) Research Associate

Name	Percent Supported
Yiwen E	70%

(3) Graduate Students

Name	Discipline	Percent Supported
Kareem Garriga	Optics	50%
Steven Fu	Optics	100%
Jiacheng Zhao	Optics	0%
Kang Liu	Optics	50%

Qi Jin	Optics	100%

(4) Undergraduate Students

Name	Discipline	Percent Supported
Mervin Lim Pac Chong	Optics	0%
Justin Murante	Optics	100% URAP 2021
Kelsey Lee	Optics	0%
Greg Lier	Optics	100% URAP 2019
Kaia Williams	Optics	100% URAP 2018

7. Ph.D. Degree Awarded

Kang Liu, who was partially worked on the THz project under this grant, was awarded her Ph.D. degree in Optics in 2018.

Qi Jin, who was directly working on the THz project under this grant, was awarded his Ph.D. degree in Optics in 2020.

8. International Collaborations

We had the collaboration with the Capital Normal University, Beijing, China and ITMO University, St. Petersburg, Russia on the THz emission from liquid targets. Our collaboration only limited to the joint publications. There is no ARO financial support to Russian and Chinese students/faculty. we also collaborate with the students and faculty at the Laboratory for Laser Energetics (LLE), University of Rochester, to explore the THz emission from solid targets under the excitation of the multi-terawatt laser. The preliminary results prove the strong THz emission with low frequency components (< 1.5 THz).

9. Scientific Progress and Accomplishments

Developing efficient and robust terahertz (THz) sources is of incessant interest in the THz community for their wide applications. With successive effort in past decades, numerous groups have achieved THz wave generation from solids, gases, and plasmas. However, liquid, especially liquid water has never been demonstrated as a THz source. One main reason leading the impediment is that water has strong absorption characteristics in the THz frequency regime.

A thin water film under intense laser excitation was introduced as the THz source to mitigate the considerable loss of THz waves from the absorption. Laser-induced plasma formation associated with a ponderomotive force-induced dipole model was proposed to explain the generation process. For the one-color excitation scheme, the water film generates a higher THz electric field than the air does under the identical experimental condition. Unlike the case of air, THz wave generation from liquid water prefers a sub-picosecond (200-800 fs) laser pulse rather than a femtosecond pulse (~50 fs). This observation results from the plasma generation process in water.

For the two-color excitation scheme, the THz electric field is enhanced by one-order of magnitude in comparison with the one-color case. Meanwhile, coherent control of the THz field is achieved by adjusting the relative phase between the fundamental pulse and the second-harmonic pulse.

To eliminate the total internal reflection of THz waves at the water-air interface of a water film, a water line produced by a syringe needle was used to emit THz waves. As expected, more THz radiation can be coupled out and detected. THz wave generation from other liquids were also tested.

1 Terahertz Wave Generation from a Water Film

1.1 Free-Flowing Thin Water Film

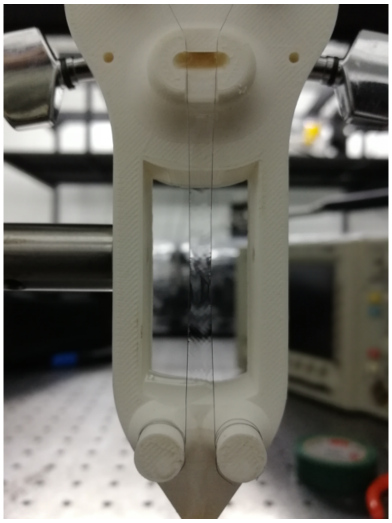

Fig. 1 Photo of the water film. Two aluminum wires are separated by 4 mm. The thickness of the water film is controlled by the water flow rate.

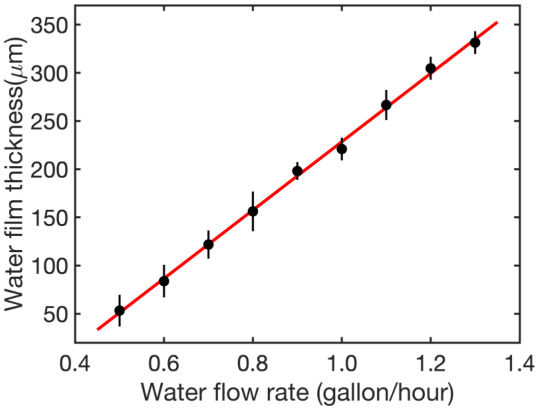

Fig. 2 Thickness of the water film versus the water flow rate.

Bulk liquid water is a strong absorber in the THz frequency range, making liquid water has historically been sworn off as a THz source. To mitigate the considerable loss of THz waves, the water with much less than 1 mm thickness is an intuitive choice to study THz wave generation. Gravity-driven, free-flowing water films have been efficaciously used owing to their simple design and almost unmatched ability to generate a thin, continuous, and stable film of liquid water in free space. The gravity-assisted flow along two metal wires forms the water film. The thickness of the water film is adjusted by the height difference between an upper reservoir and the top of the wires mounted in an aluminum frame. The system is circulated by a peristaltic pump.

By using the concept of water flowing along wires, a similar water film system was built. These wires have a diameter of 170 μm, and are separated by 4 mm. Under the effect of water's surface tension, the water goes along the wires and forms a water film as shown in **Fig. 1**. The thickness of this water film was adjusted by throttling the water flow rate. An optical second-harmonic intensity autocorrelator was used to measure and calibrate the thickness of the water film. The corresponding result is plotted in **Fig. 2**, which shows a linear relationship between the thickness of the water film and the flow rate. In this system, the thickness of the water film can be varied from 50 μm to 330 μm.

1.2 Experimental Set-up

Fig. 3 schematically shows the experimental set-up for the generation of THz waves. An amplifier laser (Ti: sapphire) with 800 nm central wavelength and 1 kHz repetition rate was used. An optical polarizer was placed to assure the p-polarized optical beam. The following half-wave plate (HWP) further adjusted the polarization of the optical beam. The laser pulse duration is adjusted by moving the compressor stage integrated within the laser. The original pulse duration is 58 fs. The incident angle of the laser beam on the water film was tilted to 25° from the normal to reduce the water sputtering onto the surface of optics. The thickness of the water film was set to be 177 μm. The laser beam was focused into the water film by a 1-inch effective focal length parabolic mirror, forming a plasma inside the water film. Filters were placed to block the remaining laser beam as well as any white light simultaneously generated from the water film in addition to the THz radiation. A tungsten wire-grid polarizer was applied as the THz polarizer. Standard electro-optic sampling (EOS) with a 3 mm thick <110>-cut ZnTe was used to detect the THz electric field with a cutoff frequency around 2.5 THz. The flow velocity of the water is about 1.3 m/s, meaning that the water film flows about 1.3 mm between two laser pulses. This distance is much greater than the diameter of the focal spot of the laser beam, which indicates that each THz pulse will not be affected by previous interaction between the water and laser pulses.

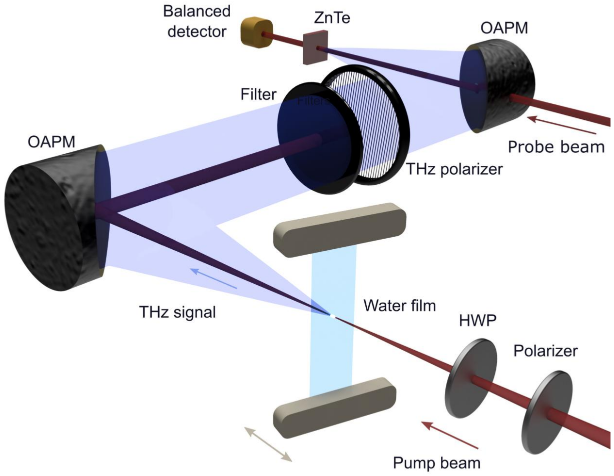

Fig. 3 Experimental set-up for THz wave generation from a water film. Broadband THz wave is generated by tightly focusing the laser beam into a gravity-driven wire-guided free-flowing water film. The water film can be moved in the laser propagation direction by a mechanical translation stage. OAPM, off-axis parabolic mirror. HWP, half-wave plate.

1.3 Terahertz Radiation from a Water Film

By using the above experimental set-up, THz wave generation from a water film is achieved. The THz waveform is shown as curve B in **Fig. 4(a).** Since EOS provides coherent signals, the obtained THz radiation is obviously not just a tail of black-body radiation. To confirm that the THz radiation is mainly emitted from the water film rather than the air plasma, the water film is translated along the direction of laser propagation. The schema of relative positions between the water film and the plasma is shown on the left-hand side of **Fig. 4(a).** The corresponding THz waveforms are plotted on the converse side of **Fig. 4(a).** For curve A, the focal point of the laser is behind the film: the laser beam passes through the water and is focused to generate THz waves from an air plasma. For curve B, the focal point is near the center of the water film: a plasma is formed inside the water film, and the THz field emitted from liquid water is measured. For curve C, the laser beam is focused and forms an air plasma before the water film. Little THz radiation is detected due to the strong absorption of the water film. It is noticeable that the THz signals from air plasma will be clearly observed if the thickness of the water film is reduced to 100 µm or less. Curve D is shown as a reference: no water film is present and only the THz wave generated from air plasma is detected.

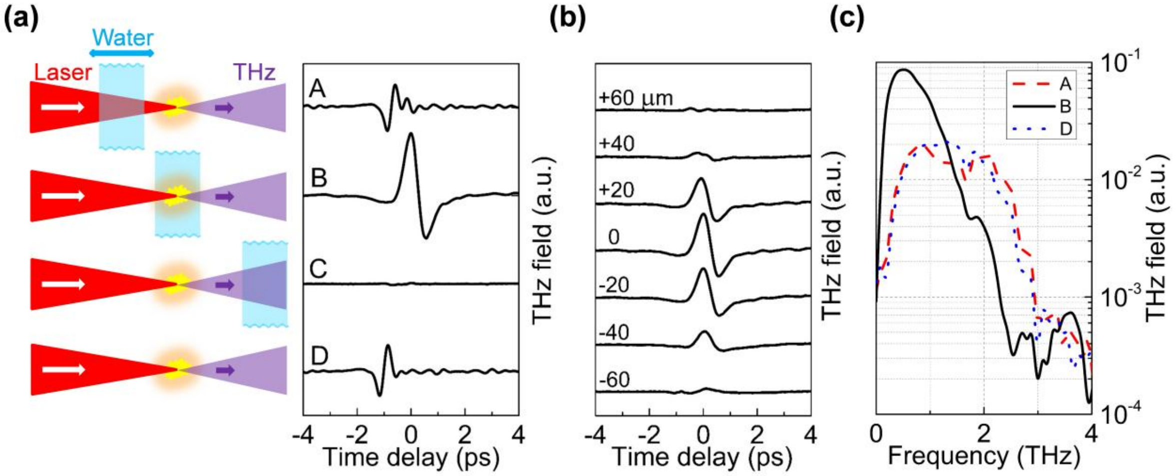

Fig. 4 Measurements of the THz fields when the water film is translated along the direction of laser propagation. (a) THz waveforms are plotted from curve A to curve C when the water film is before, near, and after the focus, respectively; Curve B shows the THz waveform generated from liquid water; Curve D is the reference with no water film. Yellow spark and bluish pane represent the plasma and the water film respectively. THz emission angle shown in the figure. is not meant to be indicative of actual THz emission pattern. (b) THz waveforms when the water film is moved near the focal point. The 0 position is set to the place with the strongest THz field. Relative positions are listed with the corresponding waveforms. The negative sign means the water film is located after the focal point. The positive sign indicates the opposite case. (c) Comparison between the THz field from water and that from air plasma in the frequency domain. The dashed, solid, and dotted spectra correspond to curve A, curve B, and curve D in (a), respectively. The laser pulse is temporally stretched to 550 fs for these measurements.

By scanning the water film along the optical axis, THz radiation from different sources can be clearly differentiated. The timing distinctions of the waveforms in **Fig. 4(a)** are indicative of different generation sources. A time delay is observed from the THz waveform from liquid water compared with other generations. **Fig. 4(b)** shows the measurements of THz waveforms as the water film is tracked along the direction of laser propagation marking a relative position across -60 µm to +60 µm. The measurement shows that the emitted THz waves are significantly sensitive to the relative position between the water film and the focus. The THz radiation can be detected only within a roughly 60 µm scanning range of the water film. It should be mentioned that no THz radiation is detectable when only part of the plasma is located outside the range of the water film. The plasma located on the interface does not give a spurious THz signal.

1.4 Comparison between Terahertz Radiation from Water and Air

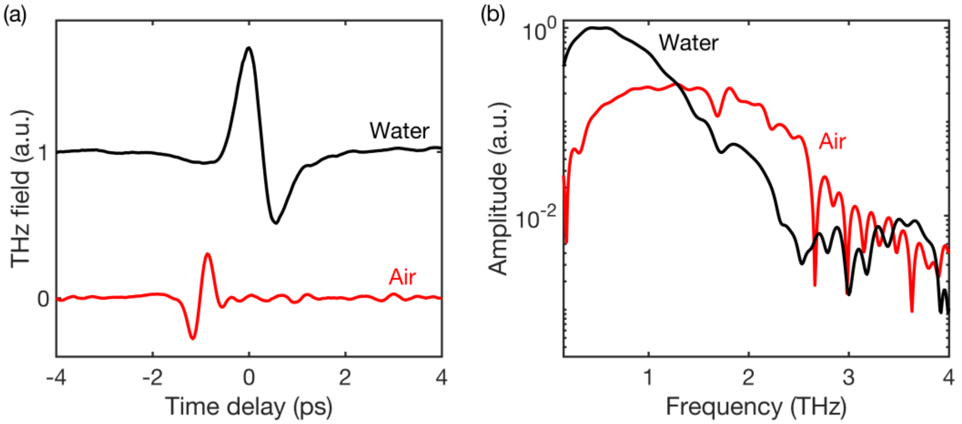

Fig. 5 Comparison of THz waves generated from water and air in the (a) time domain and (b) frequency domain.

Ambient air is one of the most extensively used THz sources. It is necessary to compare the THz radiation generated from water and air in the same experimental condition. Actually, curve B and curve D in **Fig. 4(a)** offer a reasonable comparison. To make it clear, only these two signals are plotted in **Fig. 5**. **Fig. 5(a)** depicts that the THz field from the water film is 1.8-times stronger than that from the air. The corresponding comparison in the frequency domain is shown in **Fig. 5 (b).** Note that the feature around 1.7 THz is caused by the water vapor absorption. The measured THz radiation from the water has more low-frequency components and less high-frequency components with a peak at 0.5 THz. In addition, the bandwidth is narrower than the signal from air plasma. Their difference in the frequency domain may result from the fact that liquid water absorbs more high-frequency components than low-frequency components in the THz frequency region. It should be mentioned that the result in **Fig. 5** only shows the comparison in this experimental condition. The signal from liquid water will be rather stronger if other optimal conditions (the incident angle of the laser beam on the water film, the thickness of the water film, the detection angle, etc.) are satisfied, which will be discussed in later sections.

1.5 Effect of Optical Polarization and Pulse Energy

Polarization and pulse energy of the optical beam are two important parameters that may affect THz wave generation from liquid water. The relationship between the THz radiation and the optical polarization is exposed by measuring the THz fields generated from the optical excitation beams with different polarizations. Since the water-air interface reflects more s-polarized THz waves according to the Fresnel equations, the p-polarized component of the THz field is studied here. A wire-grid THz polarizer was applied for the measurement. The polarization of the optical beam was controlled by the HWP in the optical path (see **Fig. 3**). The corresponding result is shown in **Fig. 6**. 0° refers to the p-polarized optical beam and 90° refers to the optical beam with s-polarization. It is shown that strong THz radiation is achieved with a p-polarized optical beam, while an s-polarized optical beam offers a sparse contribution. One possible explanation is that the polarization of the THz field is dependent on the optical polarization, as the measured p-polarized component of the THz field has a cosine squared relationship with the angle of the optical polarization. Also, the different Fresnel loss of the optical beam caused by the air-water interface with different polarization will change the laser intensity inside the water film, thus, lead to the variation of THz radiation. A similar phenomenon was reported wherein a particle-in-cell model was applied to simulate the observation. This result goes against the case of THz wave generation from one-color laser-induced air plasma. It is well known that the THz radiation from air plasma with one-color optical excitation does not depend upon the polarization of the optical beam, which means that the THz energy keeps constant with various optical polarizations.

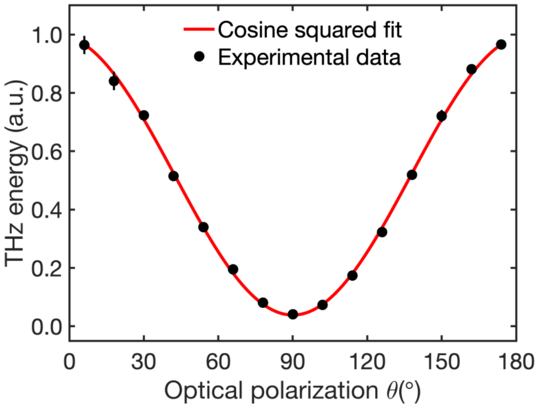

Fig. 6 The energy of p-polarized THz field from liquid water with different linearly optical polarization. 0° and 90° refer to p-polarized and s-polarized optical beam respectively.

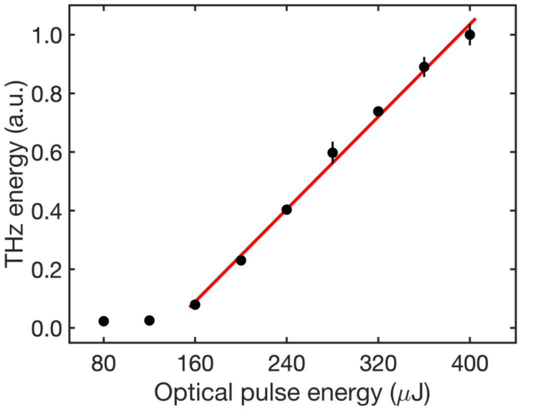

Fig. 7 Normalized THz energy from liquid water as a function of incident optical pulse energy. The water film will be broken if the energy of the excitation pulse is over 420 μJ.

Furthermore, the linear energy dependence observed in **Fig. 7** is different from the quadratic relation in the case of THz wave generation from the one-color laser-induced air plasma. **Fig. 7** also shows a laser excitation threshold at about 160 μJ for the detectable THz field from liquid water under this experimental condition. The 177 μm thick water film will be broken if the energy of the excitation pulse is over 420 μJ. This rupture may be caused by the occurrence of shock waves, plasma expansion, and water ejection when high-intensity laser pulses are focused into liquid water. These effects weaken the stability of the water film as laser energy is increased.

1.6 Effect of Water Film’s Thickness

The wire-guided free-flowing water film's thickness can be easily controlled by throttling the flow rate, which facilitates the study of how the water film's thickness impacts on the THz radiation. THz waveforms generated from water films with different thicknesses are plotted in **Fig. 8**. To make sure that the water film was not broken under the excitation of intense laser pulses, the thickness of the water film was set to be greater than 130 µm in this measurement. **Fig. 8** shows that the peak of the THz waveform linearly shifts in time with various thicknesses. This time shift comes from the path difference of THz waves in water with difference thickness. The central frequency of the THz radiation is around 0.5 THz in this case (see **Fig. 4(c)** and **Fig. 5**). From the linear fitting (red dash line), the refractive index of water at 0.5 THz can be calculated as n_{THz} = 2.29 if increasing the thickness of the water film is assumed to go symmetrically with respect to the center of the film. Furthermore, the attenuation of the THz field amplitude with the increased thickness is observed in **Fig. 8**. The attenuation of amplitude results from the increasing absorption of a thicker water film. From the exponential fitting (red dash curve), the absorption coefficient of water at 0.5 THz is calculated to be 146.2 cm^{-1}. The calculated refractive index and absorption coefficient are very close to the values found in previous work: n_{THz} = 2.27 and α = 150 cm^{-1}. It is noteworthy that only the thickness of the water film was changed, and no further optimization was applied in this measurement.

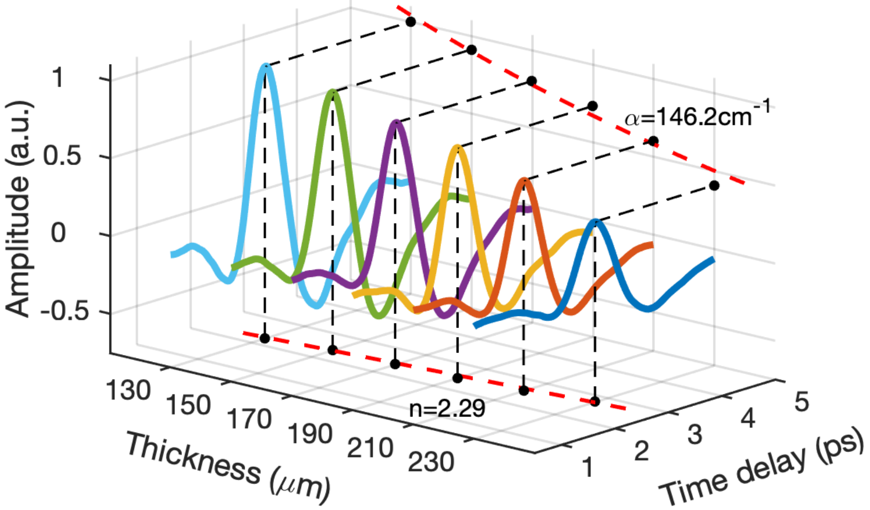

Fig. 8 THz wave generation from water films with different thicknesses. The refractive index of water at 0.5 THz is calculated to be 2.29 from the time shift of the THz field. The absorption coefficient of water at 0.5 THz is calculated to be 146.2 cm-1 from the attenuation of the THz field's amplitude.

1.7 Forced-Flowing Thin Water Film

THz wave generation from a gravity-driven free-flowing water film has already been demonstrated. However, it is observed that the film will be broken if the laser intensity is too high, which may impede the development of intense liquid sources for THz waves. To solve this problem, a nozzle jet was utilized to produce a forced-flowing water film to bear intense laser pulse. A liquid jet with a pressure of 30 psi was used to create a 5 mm wide, 120 µm thick water film, as shown in **Fig. 9**. By using this forced-flowing water film, the laser pulse energy can be greatly increased without worrying about the occurrence of rupture, which assures that a plot of energy dependence with a large range can be seen. Similar to **Fig. 7**, **Fig. 10** also shows that the THz energy linearly increases with the optical pulse energy (up to 2 mJ). No saturation is observed within this range as well. This result is indicative of a way to realize intense THz radiation from water: using intense laser pulses for excitation.

Fig. 9 Photo of a 120 μm thick water film formed by a water jet with a flat nozzle. The laser beam is focused into the center of the film where is flat and stable.

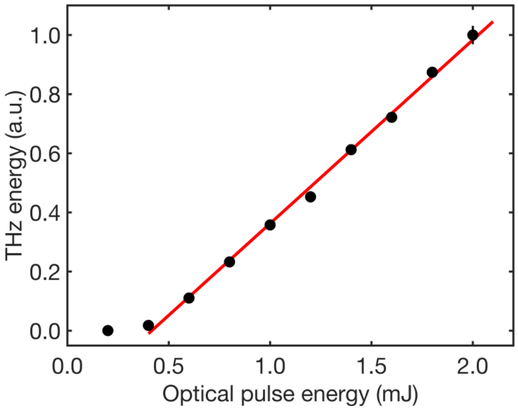

Fig. 10 Normalized THz energy as a function of the optical pulse energy. The red line shows a linear fit.

1.8 Mechanism of the Generation Process

With these experimental observations, the physical mechanism of THz wave generation from water can be analyzed. Laser-induced plasma formation associated with a dipole radiation model is proposed to explain the generation process. **Fig. 11** is a cross-section diagram of the generation process in a water film. Intense laser pulses ionize water molecules through multi-photon ionization (MPI), tunneling ionization (TI), and cascade ionization (avalanche ionization) in the focal volume. Plasmas (electrons and ions) are formed in the ionized area. The quasi-free electrons (will be called as free electrons in later sections for simplicity) in water experience the ponderomotive force and move towards the areas of lower electron density due to the density-gradient distribution. Simultaneously, other ionized particles are relatively stationary due to their large masses. Since the electrons move slower than the envelope of the laser pulse, the density of the ionized carriers always keeps identical in the forward direction. As a result, electrons are accelerated backward and create a dipole oriented along the laser propagation direction, which emits electromagnetic waves including THz frequencies. Reflection of the optical beam at the air-water interface and refraction of the THz beam at the water-air interface are considered. THz beams that experience multiple reflections between the interfaces are ignored due to the strong absorption of water.

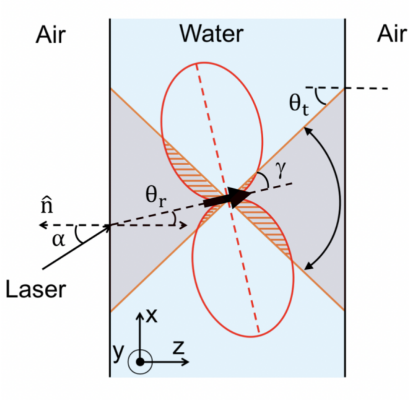

Fig. 11 2D cross-section of the THz wave generation process in a water film. Intense pulses ionize water at the focal point in the direction of the refracted laser beam. The angle of incidence on the air-water interface is α. The black arrow shows the dipole orientation direction. Due to the total internal reflection at the water-air interface, THz emission at 0.5 THz can be coupled out only when -24.6° < θt < +24.6°.

1.8.1 Laser-Induced Plasma Formation

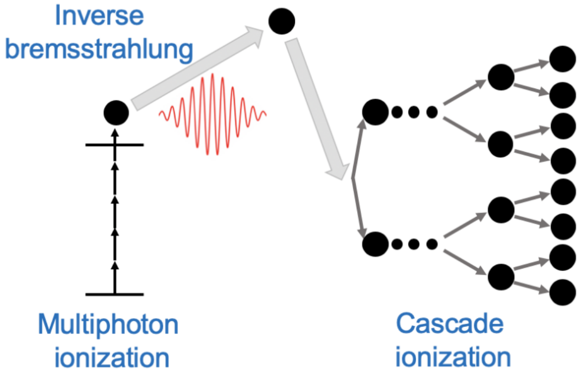

Fig. 12 Visualization diagram of laser-induced plasma formation.

Electrons are produced through MPI/TI and cascade ionization at the focus. MPI/TI directly ionizes water molecules while cascade ionization desires the presence of free electrons for initiation. These free electrons come from the background electrons and MPI/TI. Free electrons absorb photons to gain energy from laser pulses through collision with surrounding atoms or molecules, which is known as inverse bremsstrahlung absorption. Water can be treated as an amorphous semiconductor with a bandgap of E_g = 6.5 eV. Once achieving the energy greater than the ionization energy E_g, a free electron will be produced. This results in two free electrons of lower energy, which in tum absorb energy from the laser and ionize two more electrons, and so on. Thus, the process leads to a cascade of electrons. **Fig. 12** shows the process of plasma formation. It needs to be mentioned that TI occurs or dominates over MPI when the laser intensity is sufficiently high to decrease the potential barrier allowing electrons to escape. The Keldysh parameter γ is widely used to indicate which process is dominant.

$$\gamma = \sqrt{E_g/U_p} \tag{1}$$

where U_p is the laser ponderomotive potential energy. $\gamma > 1$ implies MPI while $\gamma < 1$ indicates TI.

Assuming the laser pulse propagates in the z-direction, the electric field envelope E(z, r, t) follows the equation:

$$\frac{\partial E}{\partial z} = \frac{i}{2nk_0}\nabla_T^2 E + ik_0 n_2 |E|^2 E - \frac{i\beta_2}{2}\frac{\partial^2 E}{\partial t^2} - \frac{\sigma}{2}(1+i\omega\tau)\rho E - \frac{\beta^{(k)}}{2}|E|^{2K-2}E. \tag{2}$$

The terms on the right-hand side represent transverse beam diffraction, nonlinear self-focusing, group velocity dispersion (GVD), plasma absorption and defocusing, and multiphoton absorption, respectively. Here, n is the refractive index, k0 is the wave vector, n_2 is the nonlinear coefficient, β_2 represents GVD, $\sigma = (ke^2\tau/m\omega\varepsilon_0)/(1+\omega^2\tau^2)$ is the cross-section for inverse bremsstrahlung absorption, ω is the optical frequency, τ is the electron collision time, and $\beta^{(K)}$ is the nonlinear coefficient for K-photon absorption. The last term need be modified to $w(\rho_{\text{water}}\text{-}\rho)E_g E/2I$ when TI dominates. w is the ionization rate, ρ_{water} is the water molecular density, and I is the laser intensity. The electron density ρ(z, r, t) satisfies the rate equation:

$$\frac{\partial\rho}{\partial t} = \left(\frac{\partial\rho}{\partial t}\right)_{mp} + \eta_{cas}\rho - \eta_{diff}\rho - \eta_{rec}\rho^2. \tag{3}$$

The first two terms on the right-hand side correspond to the generation of electrons from MPI and cascade ionization. The first term becomes $w(\rho_{\text{water}}\text{-}\rho)$ in the case of TI. The other two terms in Equation 3 describe the loss of electrons from diffusion and recombination. The laser pulse is considered to have a Gaussian profile with its intensity as follows:

$$I(t) = 0.94\frac{\epsilon_P}{\tau_P}\exp\left[-4ln2\left(\frac{t}{\tau_P}\right)^2\right]/(0.5\pi\omega_0^2) \tag{4}$$

where ε_p is the laser pulse energy, τ_p is the laser pulse duration, and w_0 is the beam waist at the focus. In condensed media, Keldysh derived an approximate expression for the multiphoton ionization rate as:

$$\left(\frac{\partial\rho}{\partial t}\right)_{mp} \approx \frac{2\omega}{9\pi}\left(\frac{m\omega^2}{2\hbar\omega}\right)^{1.5}\left(\frac{e^2}{8mE_g\omega c\epsilon_0 n}I\right)^K + \exp(2K)\Phi\left(\sqrt{2K - \frac{2E_g}{\hbar\omega}}\right) \tag{5}$$

with $\Phi(x) = \exp(-x^2)\int_0^x y^2 dy$.

In the case of TI, the ionization rate can be calculated by Ammosov-Delone-Krainov (ADK) model:

$$\Phi(x) = \exp(-x^2)\int_0^x y^2 dy \tag{6}$$

The cascade ionization rate is given by:

$$\eta_{cas} = \frac{\sigma}{n^2 E_g} I - \frac{m\omega^2\tau}{M(1+\omega^2\tau^2)} \tag{7}$$

where M is the mass of a water molecule. The first term on the right-hand side results from the inverse bremsstrahlung absorption. The second term relates to the energy transfer from the electrons to heavy molecules.

1.8.2 Dipole Radiation Model

The ionized electrons are accelerated to form a dipole in the direction of the refracted laser beam that emits THz radiation as shown in **Fig. 11**. To confirm the applicability of the dipole radiation model, the following experiments were conducted. The laser pulses with horizontal polarization, 800 nm central wavelength, and 1 kHz repetition rate were delivered from a Ti: sapphire amplified laser. It was focused into a 120 μm thick water film by a 2-inch effective focal length lens (F/4) ionizing water molecules at the focus. A 2 mm thick <110>-cut ZnTe crystal configured for EOS and a commercially available Golay cell were both used for the detection of THz waves. An illustration of the incident angle α for the laser beam and the angle of detection β for the detector is shown in **Fig. 13**. These two angles are both defined with respect to the laser propagation direction (z-axis) and can be changed by rotating the film and detector, respectively. $\hat{n}$ is the surface normal of the water film. The sign of the angle is negative/positive when it is measured clockwise/counter-clockwise from the z-axis. The optical pulse duration is tuned to be 300 fs for maximizing the THz signal with the 120 μm thick water film.

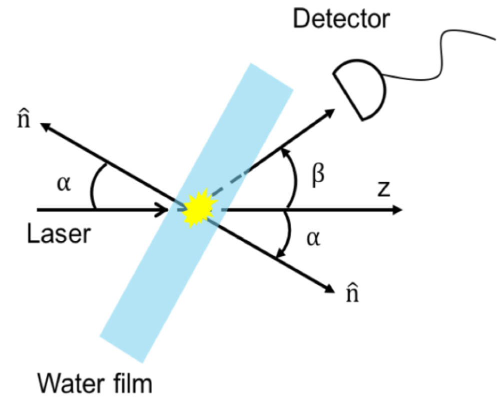

Fig. 13 Illustration of incident angle α and angle of detection β. All angles are defined with respect to the z-axis.

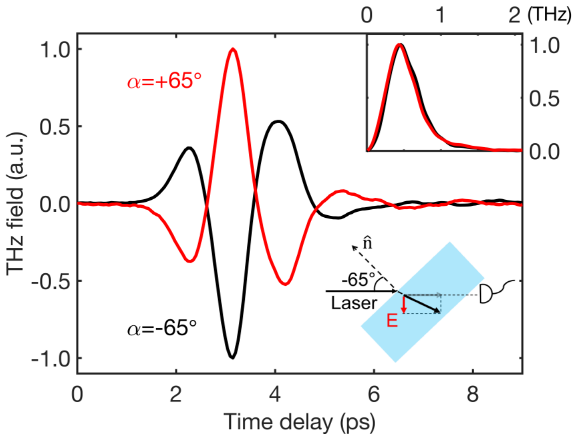

Fig. 14 THz waveforms generated from a water film with opposite angles of incidence (α = ±65°). The corresponding spectra and dipole approximation illustration are shown in the insets.

Fig. 14 plots the THz waveforms generated from the water film with two opposite angles of incidence (α = ±65°) and detected in the laser propagation direction. The corresponding spectra are shown in the inset. The central wavelength and full width at the half maximum (FWHM) are both about 0.5 THz. Therefore, the parameters (index of refraction and absorption coefficient) at 0.5 THz are used in the following calculation and simulation. As show in **Fig. 14**, the waveform keeps its amplitude but completely flips over when the angle of incidence changes its sign. This observation applies to all opposite angles of incidence. It can be inferred that the dipole orientation is along the propagation direction of the refracted laser beam, illustrated as a black arrow in **Fig.**

14. The projected signal from the dipole reverses in direction when the angle of incidence changes the sign, which accounts for the flipped waveforms.

1.9 Radiation Pattern

In the last section, a dipole model was proposed to explain the mechanism of THz wave generation from liquid water. Similar to the case in air, a spatial net charge distribution created by the ponderomotive force acts like a Hertzian dipole and radiates THz waves. However, the existence of the interfaces makes the scenario more complicated. Specifically, the refraction of the laser beam induced by the first interface (air-water) changes the orientation of the dipole while the total internal reflection and refraction of the THz beam caused by the second interface (water-air) significantly affect the radiation angle of THz waves. The model also predicts that backward THz radiation should be present even though only the forward radiation has been paid attention in the previous study. Thus, simulations and measurements of the THz radiation pattern can be very useful to further verify the dipole radiation model.

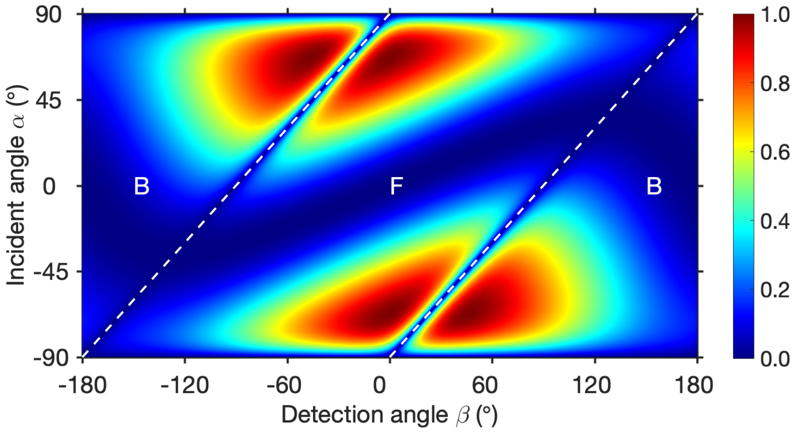

Fig. 15 Simulation result of normalized THz energy $I_{THz}(\alpha, \beta)$ using the dipole radiation model. The dashed lines indicate the cases of $|\alpha - \beta| = 90°$, which means the detector is located in the plane of the water film. These dash lines separate the plots into three parts, labeled as "B", "F", and "B". "B" and "F" indicate backward and forward propagating THz signal, respectively.

At the air-water interface, refractive angle $\theta_r(\alpha)$ and transmittance $T_1(\alpha)$ for a given α is obtained according to Snell's law and the Fresnel equations, which are determined by the refractive indices of the 800 nm optical beam in air and water. The maximum $\theta_r(\alpha)$ is 48.8° when $\alpha = 90°$. THz waves radiated by the dipole propagating in the water film are attenuated due to the absorption of water. If the thickness of the water film is d, the absorption in the different direction $\theta_t(\beta)$ from the source can be described as $\exp[-\alpha_{THz}d/2\cos\theta_t(\beta)]$, where α_{THz} is the power absorption coefficient of water. Multiple reflections of THz waves are neglected in the calculation due to the strong absorption of water. Additionally, the dipole radiation energy is proportional to $\sin^2(\gamma)$, where $\gamma(\alpha, \beta) = \theta_t(\beta) - \theta_r(\alpha)$ is the angle measured with respect to the dipole direction. Finally, the THz waves are detected after passing through the water-air interface with a transmittance $T_2(\beta)$. To sum up, the angular dependence of THz energy on α and β is described as:

$$I_{THz}(\alpha,\beta) \propto T_1(\alpha)T_2(\beta)\sin^2[\gamma(\alpha,\beta)]\exp\left(-\frac{\alpha d}{2cos\theta_t(\beta)}\right) \quad (8)$$

Fig. 15 plots the simulation result of the normalized THz energy $I_{THz}(\alpha, \beta)$. A micro-plasma is created in the water film with the tightly focused geometry. This plasma can be considered as a point source emitting THz waves in all directions. Thus, besides the forward (F) propagating signal, the signal propagating in the backward (B) direction is also expected. These two parts are separated by the dashed line in the plot and labeled separately. Due to the symmetric geometry of the model, the energy distribution pattern for forward and backward propagating THz signals are the same when the plasma is located at the center of the water film. The dashed lines also indicate the case of |α - β| = 90°, which means that the detector is put in the plane of the water film. When |α - β| > 90°, the THz waves propagate in the backward direction.

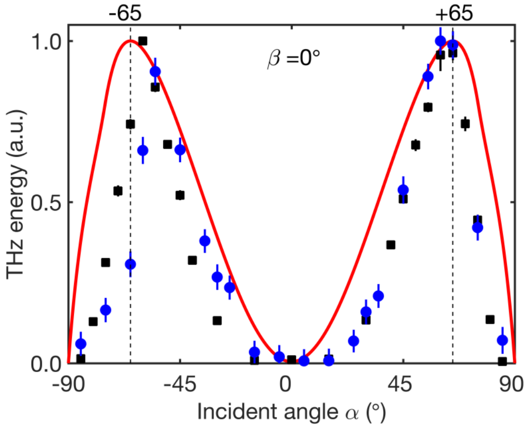

Fig. 16 THz energy versus the angle of incidence α with β = 0°. The black squares are the data measured by EOS and the blue dots are measured by a Golay cell. Only forward propagating signals can be detected for β = 0°.

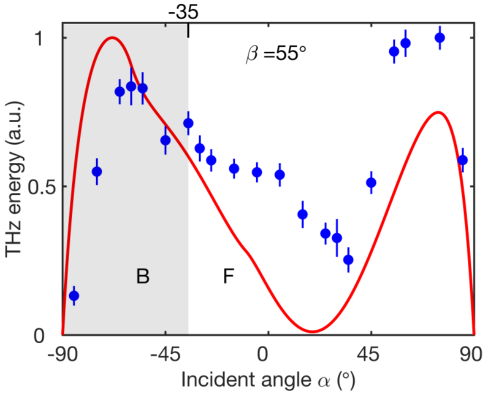

Fig. 17 THz energy versus the angle of incidence β = 55°. The blue dots are measured by a Golay cell. Backward propagating signals are detected for -90° < α < -35°.

To verify the simulation result, the THz signal versus α was measured when β was fixed at 0° or 55°, both EOS and a Golay cell were used for the detection in the measurement. The corresponding results are plotted in **Fig. 16**. The red solid line shows the simulation result. The EOS result (black squares) is obtained from the temporal integration of the whole THz waveform. The result from the Golay cell is plotted as the blue dots. As shown in the plot, the optimal incident angle of the laser beam is 65°, which results from the dipole's orientation. The coincidence of the experimental data and the simulation exhibits the validity of the dipole radiation model. Note that only the forward THz waves can be measured when β = 0°. In addition, the calculation with α = 65° indicates that 80% of the THz energy dissipates due to the total internal reflection at the water-air interface and the strong absorption of water.

Even though EOS generally offers a better signal-to-noise ratio, its optical alignment for the detection of radiation pattern is complicated. By contrast, a Golay cell is capable of measuring radiation pattern including both forward and backward directions easily. Thus, the Golay cell is used for the case of β = 55° (**Fig. 17**). In this case, the signal comes from the backward radiation THz waves is observed when -90° < α < -35°. The detectable backward propagating THz signal supports the dipole radiation model. Compared to the simulation result (solid line), stronger signals are measured in the forward direction. This may be a consequence of plasma deviation from the center of the film.

1.10 Effect of Optical Pulse Duration

It has reported that long pulse helps enhance x-ray generation from a liquid plasma. To study the dependence of the THz radiation on the optical pulse duration, the laser pulse was stretched in time to achieve different pulse duration. **Fig. 18** shows the normalized THz energy from water and air plasma versus the optical pulse duration. The THz energy from water or air plasma is normalized to the corresponding maximum respectively.

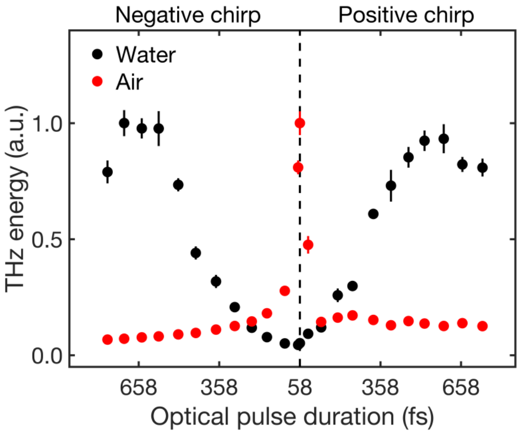

Fig. 18 Normalized THz energy from liquid water and air plasma with different pulse duration of the laser beam. Black squares represent the THz energy from liquid water and red dots represent the case of air plasma. The optical pulse duration is at its minimum of 58 fs when no frequency chirp is applied. On the left-hand side of the figure, negative chirps are applied to increase the optical pulse duration while the case of positive chirps is shown on the right-hand side of the figure. The energy of the laser pulse is 0.4 mJ for these measurements.

The optical pulse duration is at its minimum of 58 fs when no chirp is applied. The left-hand side of **Fig. 18** shows the case of negative chirps, where the low-frequency component of the pulse lags the high-frequency component. Positive chirps indicate the opposite, and the corresponding measurements are shown on the right-hand side of **Fig. 18**.

Unlike the THz radiation from air plasma, where the signal is maximized at a minimum pulse duration with no additional chirp, liquid water generates a maximum field at a longer pulse duration. Furthermore, by comparing the left part and the right part of **Fig. 18**, it is shown that the frequency chirp of the optical beam is not a dominant factor compared with the contribution from the pulse duration. This can also be supported by **Fig. 19**, which plots the similar spectra of THz radiation by using the same optical pulse duration but opposite chirps. For simplicity, only positive chirps will be applied in future investigation. The above observations may result from the dependence of plasma formation in water upon the optical pulse duration. To confirm the assumption, further experiments and simulations are indispensable. More in-depth discussions will be shown later.

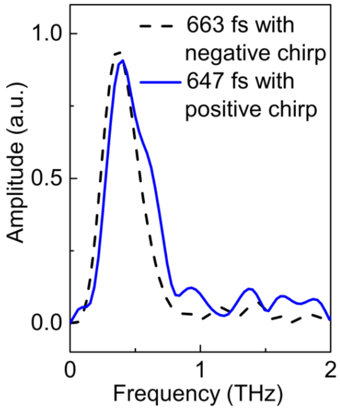

Fig. 19 Spectra of THz radiation generated from the water film with a negative chirped 663 fs pulse and that with a positive chirped 647 fs pulse.

2 Terahertz Radiation from Liquid Water under Two-Color Excitation Scheme

2.1 Glory of Two-Color Excitation Scheme in Gases

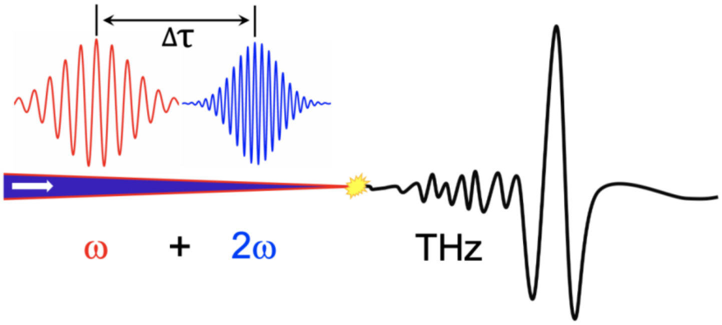

Fig. 20 Schematic diagram of THz wave generation from a two-color laser pulses induced air plasma. An intense femtosecond laser beam ω and its second harmonic 2ω are focused to generate plasma in the air. In the most common way, a β-BBO crystal is applied for the generation of 2ω pulse. The output THz waves are determined by the phase delay between ω pulse and 2ω pulse.

THz wave generation through the tunneling ionization process in gases induced by two-color (fundamental frequency ω and its second-harmonic 2ω) femtosecond laser pulses is a significant milestone in the THz community due to its impressive intensity, remarkably broad bandwidth, and applications in nonlinear interactions and THz spectroscopy. There is no exaggeration to say that THz wave generation under the two-color excitation scheme has been always one of the most popular topics since it was reported by Cook and Hochstrasser in 2000. A typical schematic diagram is shown in **Fig. 20**. An intense femtosecond laser pulse ω and its second harmonic 2ω are focused into the air to generate air plasma for the emission of THz waves. Commonly, the 2ω laser pulse is generated by applying a type-I ω-barium borate (BBO) crystal to the ω laser pulse. The output THz radiation is controlled by the phase delay between ω pulse and 2ω pulse. Compared with using only ω pulses for the excitation, the mixing of ω and 2ω pulses has shown to provide enhancement of THz wave generation efficiency by several orders of magnitude.

The generation mechanism of the two-color excitation scheme was phenomenologically assigned to a four-wave-mixing model as:

$$E_{THz}(t) \propto \chi^{(3)} E_{2\omega}(t) E_{\omega}^{*}(t) E_{\omega}^{*}(t) cos\phi \tag{9}$$

where E is the field amplitude of the frequency components specified by the subscript, $\chi^{(3)}$ is the effective component of the third-order nonlinear susceptibility tensor of the gas, and is the relative phase between ω pulse and 2ω pulse. The relationship in Equation (9) was experimentally demonstrated from an energy dependence measurement by Xie et al. in 2006. The THz amplitude is linearly increased with the intensity of ω pulse and is proportional to the square root of the intensity of 2ω pulse. Even though the four-wave-mixing model successfully offers an intuitive and straightforward description, it fails to explain some observations in experiments. For example, the occurrence of a clear threshold cannot be predicted by the model. Instead, the threshold should be associated with the photoionization process happened in the air. Additionally, the four-wave-mixing model is not indicative of the enhancement of THz radiation that caused by an applied asymmetric laser field.

In 2007, Kim et al. proposed a transient photocurrent model that could solve the problems mentioned above. In this model, a net photocurrent produced by asymmetric optical fields through the photoionization radiates THz waves. Compared with THz wave generation from gas plasmas induced by one-color laser pulses, the asymmetric electron motion introduced by the two-color laser fields leads to a net dipole moment and hence, much stronger THz emission. In 2009, Karpowicz and Zhang developed a full quantum model to depict a complete physical picture of the generation process by numerically solving the time-dependent Schrödinger equation. The model accurately describes the formation and acceleration of the relevant wave packets. Compared with other THz wave generation techniques, the two-color excitation scheme in gases provides the THz radiation with intense electrical fields (> 8 MV/cm) as well as broad spectral information (> 100 THz).

In addition, the two-color laser pulses enable the coherent control of THz waves. A femtosecond ω pulse generates a 2ω pulse while passing through a β-BBO crystal. The perpendicular polarized ω and 2ω pulses pass through an α-BBO with its slow axis aligned with the polarization of the ω beam and the fast axis aligned with the polarization of the 2ω beam. Thus, the 2ω pulse leads the ω pulse after passing through the ω-BBO crystal. A fused silica wedge pair is used to finely control the phase delay between the ω and 2ω pulses. Finally, a tunable dual-band wave plate is used to control the polarization of the ω beam and the 2ω beam. Modulation of THz waves generated from air plasmas has been achieved by changing the relative phase between ω and 2ω pulses via the phase compensator. It is worth underlining that Kumar et al. also reported THz wave generation from liquids by focusing femtosecond laser pulses into a cuvette filled with target liquids. Their observations also indicate that the laser-induced plasma formation in liquid materials plays a critical role in the THz wave generation process. The electrons ionized from water molecules are regarded as quasi-free electrons, and therefore the THz wave generation process in water resembles the process in air. Thus, the two-color excitation scheme should also work in liquid water. Stronger THz waves and corresponding modulation are expected by using the asymmetric excitation scheme in water as well. Manipulation of strong THz emission would have widespread applications in different research fields, such as THz nonlinear optics and electron acceleration. In the following sections, the THz wave generation from a thin water film under two-color laser excitation will be discussed.

2.2 Experimental Setup of the Two-Color Excitation Scheme

The schematic diagram of the experimental setup is shown in **Fig. 21**. A femtosecond amplified Ti: sapphire laser with a central wavelength of 800 nm and a repetition rate of 1 kHz was used.

Unless otherwise stated, the laser pulse duration used in this experiment was 58 fs. A β-BBO crystal was used for the generation of 2ω pulses through frequency-doubling, and an in-line phase compensator was applied to accurately control the relative phase between ω and 2ω pulses by changing the mechanical insertion of one of the fused silica wedges. A phase compensator was composed of an α-BBO crystal, a pair of wedges, and a dual-wavelength wave plate (DWP). The energy of 2ω pulses was about 10% of the entire excitation laser energy. Both ω and 2ω pulses were vertically polarized after they pass through the phase compensator. Subsequently, ω and 2ω laser pulses were co-focused into a 120 µm thick water film by a 1-inch effective focal length parabolic mirror to generate THz waves. The focal point of the laser beam was set to be close to the center of the water film. A liquid jet was employed to obtain the water film. A high-resistivity silicon wafer was used as a filter to block the residual laser beams while allowing the THz waves to pass through. The THz electric field was detected by a 3 mm thick <110>-cut ZnTe crystal through EOS. Also, the corresponding THz energy was measured by a commercially available Golay cell with a combination of different filters that eventually blocked all the high-frequency components. The angle of incidence on the water film was optimized to be 61°.

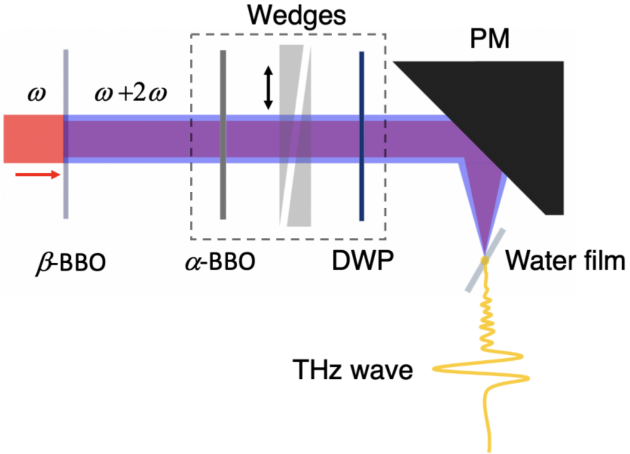

Fig. 21 Schematic diagram of the experimental setup. A phase compensator composed of an α-BBO crystal, a pair of wedges, and a dual-wavelength wave plate (DWP) is applied to control the relative phase between ω and 2ω pulses. PM, parabolic mirror with an effective focal length of 1-inch.

2.3 Comparison between Terahertz Radiation from One-Color and Two-Color Excitation Scheme

By using the above experimental setup, THz emission from liquid water under the two-color excitation scheme is achieved. Remarkably, the THz electric field generated from the two-color excitation scheme is about 10-times stronger than that from the one-color excitation scheme at the laser pulse duration of 58 fs, as shown in **Fig. 22(a).** The one-order of magnitude increased THz electric field is indicative of the two-orders of magnitude enhanced THz energy. The corresponding spectra are shown in **Fig. 22(b).** It is noteworthy that the enhancement of the THz electric field with the asymmetric excitation scheme in water may not be as high as that in air. This could arise from the fact that a short laser pulse duration works well for the case in air, but a longer pulse duration is favored in the ionization process in liquid water, where cascade ionization dominates.

Comparatively, experimental results in the case of a longer laser pulse duration (300 fs), which is obtained by chirping the original 58 fs pulse, are shown in **Fig. 22(c)** and **Fig. 22(d).** The scales of the vertical axis in **Fig. 22(a)** and **Fig. 22(c)** are the same. Compared to the one-color case, the two-color excitation scheme provides 11% enhanced THz electric field when the pulse duration is 300 fs. Such a reduced enhancement rate may be caused by multiple effects. For example, the ω and 2ω pulses may have uneven chirps, which reduces the asymmetry of the ionized electron motion and finally decreases the overall generation efficiency of THz waves. The lower enhancement with a longer pulse duration may also result from the significant drop in second-harmonic generation efficiency as the pulse duration increases. In the experiment, the energy of 2ω pulse decreases by more than 60% when the pulse duration increases from 58 fs to 300 fs.

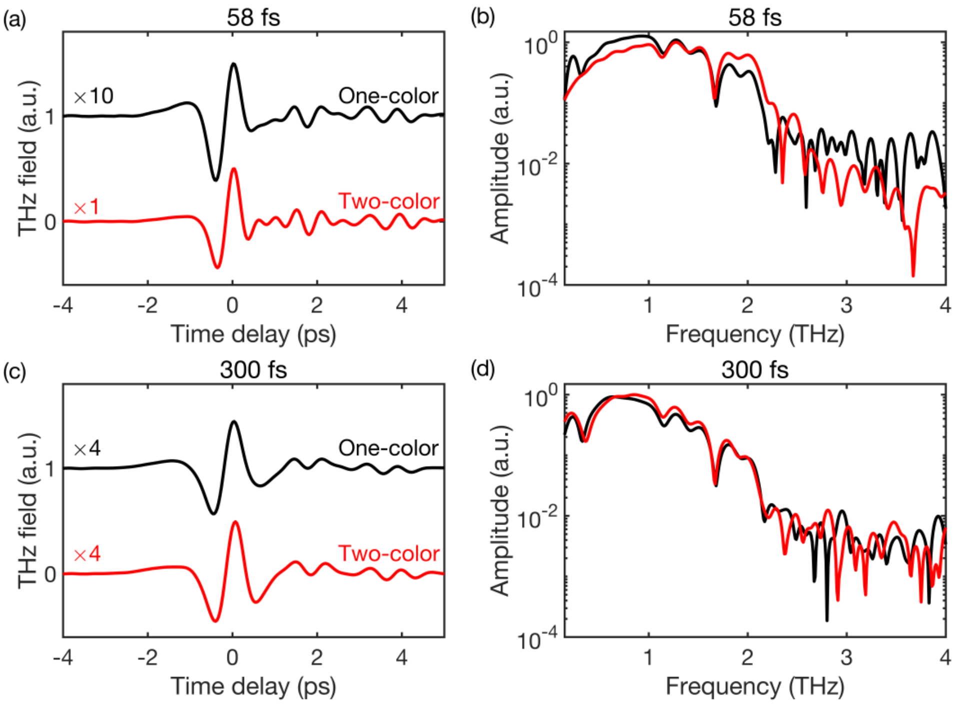

Fig. 22 Comparison of THz waves generated from a 120 μm thick water film with one-color and two-color excitation schemes. (a), (b) Comparison in the case of a short optical pulse duration (58 fs) in the time domain and frequency domain, respectively. (c), (d) Comparison in the case of a long optical pulse duration (300 fs) in the time domain and frequency domain, respectively. Unified normalization ratios are labeled.

2.4 Modulation of THz Fields

Phenomenologically, the transient photocurrent model can be used to explain the generation process in water and would predict the modulation of THz fields generated from a water film as well, which is experimentally confirmed, as shown in **Fig. 23**. Specifically, **Fig. 23(a)** shows that the polarity of the THz electric field is completely flipped over by changing the relative phase φ by π. The inset of **Fig. 23(a)** plots the THz field as a function of optical phase delay between π and 2π pulses, which indicates that the polarity of the THz electric field is gradually changed with the optical phase delay.

Moreover, an overall phase scan for THz wave emission from the water film is obtained by gradually adjusting the phase between π and 2π pulses at an attosecond-level accuracy while monitoring the THz energy with a Golay cell, as shown in **Fig. 23(b).** The noise floor is also shown in the figure. The modulated portion shows the phase modulation while the unmodulated portion remains blank at the bottom of the figure. By comparing the energy levels in the figure, the modulated and unmodulated components are estimated to be 70% and 30%, respectively. It needs to be mentioned that similar modulation can be achieved with a longer optical pulse duration (300 fs) as well.

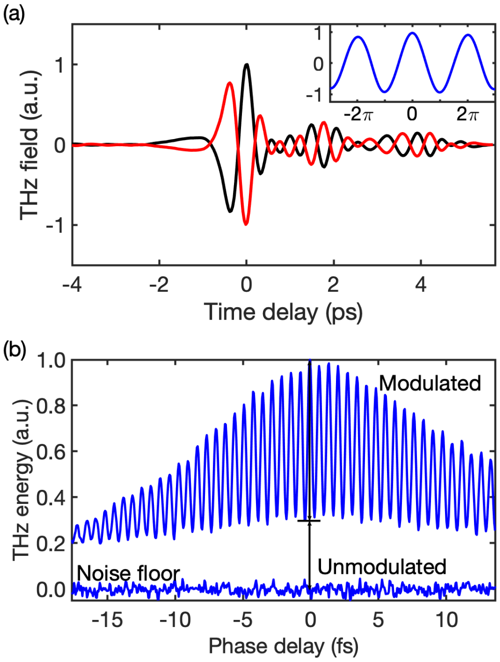

Fig. 23 Modulation of THz wave generation from a water film. (a) Comparison of THz waveforms obtained when the relative phase between π and 2π pulses is changed by π through the change of the insertion of one of the wedges in the phase compensator. Inset, THz electric field as a function of the phase delay between π and 2π pulses. (b) An overall phase scan for THz wave radiation from the water film obtained by gradually changing the phase between π and 2π pulses while monitoring the THz energy by using a Golay cell. The range of the phase delay is limited by the full length of the wedge.

The modulated and unmodulated THz waves are related to different generation processes in the plasma. For the further study, the corresponding THz energy varies with the excitation laser pulse energy was measured. **Fig. 24** plots the THz energy as a function of the total excitation pulse (ω and 2ω) energy. The unmodulated THz energy (red circles) shows a linear dependence on the laser pulse energy. For the modulated THz energy (blue dots), the modulation does not appear until the excitation pulse energy is beyond 200 μJ. Subsequently, the measurement matches a quadratic fitting above the threshold. In addition, the energy dependence measured from EOS (blue squares) is coincident with the modulated result from the Golay cell.

Similar to the case in air, the modulated THz energy mainly comes from electron acceleration in the transient photocurrent model, while in the full quantum model, the modulated THz radiation may also result from the buildup of bremsstrahlung from electron-atom collisions. In contrast, the unmodulated THz energy may arise 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 be attributed to part of the broadband radiation from the combination of thermal bremsstrahlung from electrons and electronion recombination. Moreover, the energy dependence in **Fig. 24** indicates that the ratio of the modulated THz energy to unmodulated THz energy increases with the laser pulse energy. The unmodulated component is stronger with weak excitation pulses while the modulated component will dominate if intense laser pulses are used.

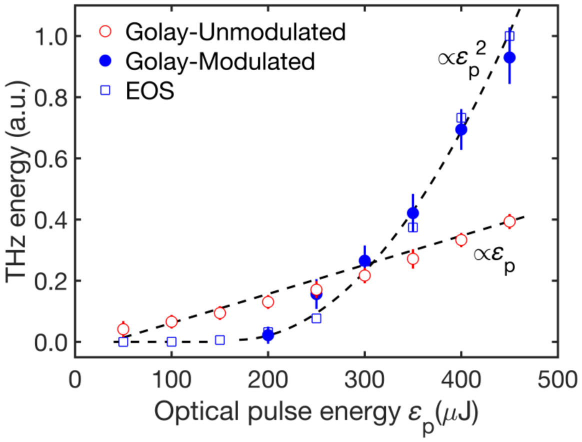

Fig. 24 Normalized THz energy from liquid water as a function of the total excitation optical pulse (ω and 2ω) energy. Blue squares, THz energy calculated from the temporal integral of the THz waveform measured by EOS. Blue dots, modulated THz energy measured by the Golay cell. Red circles, unmodulated THz energy measured by the Golay cell. The maximum pulse energy is limited by the available laser pulse energy in the experiment.

In consideration of the fact that the liquid source can quickly replenish itself due to its fluidity, the THz wave emission can be dramatically scaled up by increasing the excitation laser energy, which reveals liquid water's potentiality to emit intense THz waves. Although no saturation occurs in **Fig. 24** when the laser pulse energy is up to 450 μJ, it is worth mentioning that the saturation should be observed when the excitation energy is sufficiently high. The measured THz electric field strength is estimated as 1.1 kV/cm when the excitation laser pulse energy is 450 μJ. Realistically, the measured value is much weaker than the generated THz radiation due to the absorption of the water film itself and total internal reflection on the water-air boundary, etc.

Consequently, the THz wave radiation from the air at the interface is negligible, in comparison to the contribution from the water film. In fact, the measurable THz field from the air plasmas at the interfaces should be even smaller. Specifically, the water film will either absorb 83% of the THz energy from the plasma located close to the first interface (air-water) or decrease the laser intensity by affecting the focusing geometry for the plasma near the second interface (water-air). Based on the above discussion, THz waves generated in the experiment are mainly attributed to the plasma within the water film rather than the air plasma.

3 Broadband THz wave generation from liquid metal

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 wave generation process. Besides, the surface tension of liquid metal is much higher than that of water, meaning that it forms 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. 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 can be obtained.

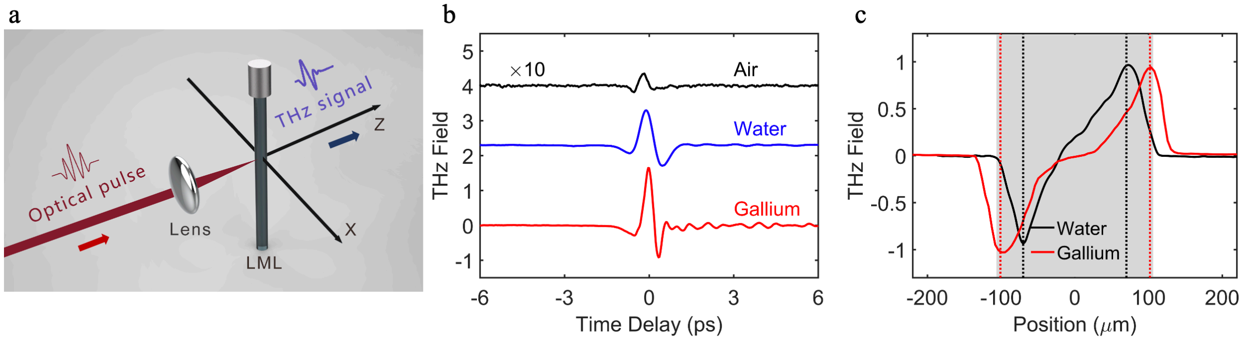

Fig. 25 THz generation from liquid metal. a Schematic of the experimental setup. A two-dimensional translation stage is used to control the position of the x-axis and the z-axis of the liquid metal line. The optical beam propagates along the z-axis. THz emission is measured by a standard electro-optical sampling. **b** Comparison of THz waveforms generated from air, water and liquid gallium under single-color excitation. The target diameter is 210 μm. The THz signal from air plasma is enlarged by 10 times. **c** Normalized THz field strength plotted 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.

Fig. 25a 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 **Fig. 25b**. THz signals from both gallium and water lines are much stronger than that from air plasma. The THz signal generated from liquid gallium is 1.7 times stronger in field than that from water at 35 °C. The THz signal from liquid gallium shows a narrower pulse width, indicating that the THz signal from liquid gallium shows a broader bandwidth than that from water. More components at high frequencies are generated, which are also confirmed by their spectra.

Fig. 25c plots the dependence of THz field strength on the x-position of the source. The position of liquid gallium 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 penetration 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 via 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 THz signal from water, the mechanism of THz waves generation from liquid metals likely differs from the generation process of liquid water. Further exploration still needs 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.

4 Broadband THz wave generation from liquid nitrogen

Compared to liquid water, liquid nitrogen (LN) is a good candidate for tests based on the following properties: (1) LN 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 has a much lower viscosity (164 mPa·s at -196 °C) than water (889 mPa·s at 25 °C). (3) LN is a nonpolar liquid, which has a much lower absorption in THz regime. (4) LN 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 has been demonstrated recently under double pump geometry with either single- or two-color excitation. The shockwave in LN created by the laser pulse may affect the interaction of the next pulse. To eliminate the influence between laser pulses, a flowing LN line is required. However, the room temperature is much higher than the boiling point of LN, which is a challenge to create a flowing line at room temperature.

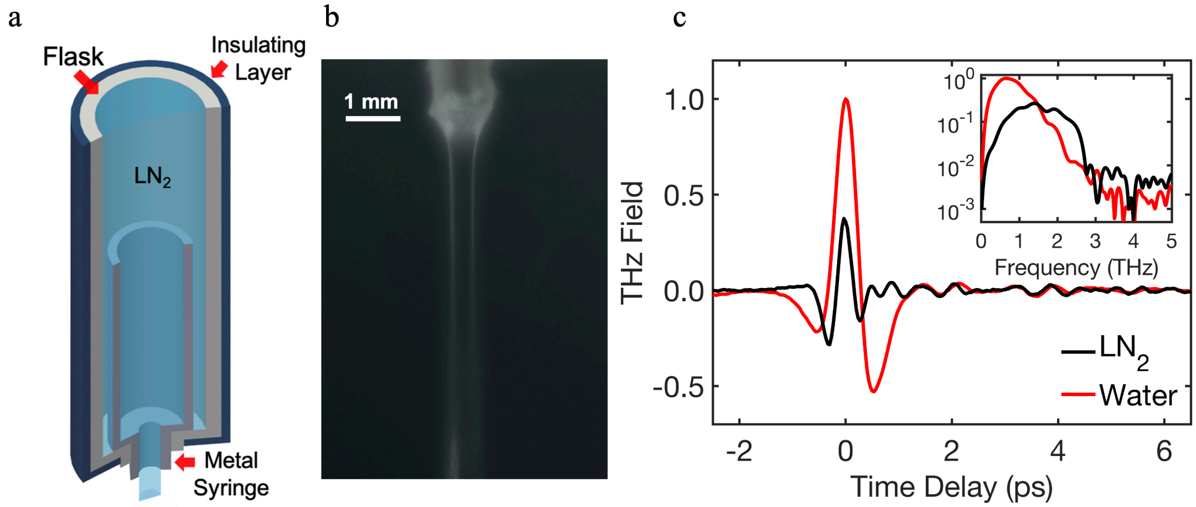

Fig. 26. THz generation from liquid nitrogen (LN). a Diagram of the apparatus for guiding an LN line. By filling it with LN, the liquid in the flask prevents the liquid in the syringe transferring heat. **b** A photo of a flowing LN line in an ambient environment. The transparency of the line indicates the liquid phase clearly with a smooth surface. **c** Comparison of THz waveforms from water and LN under the same optical excitation. Inset: Corresponding spectra.

When liquid contacts with a hot surface whose temperature is much higher than the boiling point, vaporization creates an insulating layer preventing the liquid from boiling rapidly. This is known as the Leidenfrost effect. Benefiting from this effect, it is possible to create a flowing LN line at ambient environment. Additionally, another factor concerned is the transient vaporization of LN caused by the pressure difference. Usually, LN is stored in a Dewar with a higher pressure inside than the outside. The boiling point highly depends on the pressure and 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 LN will be ejected out from the jet. To overcome this problem, a phase separator and a custom-designed LN reservoir are needed. As shown in **Fig. 26a**, the LN reservoir consists of a syringe, a flask, and an insulating layer. The metal syringe is immersed in a flask filled with LN 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, 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 in the ambient environment. **Fig. 26b** is a photo of the flowing LN 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.

Fig. 26c shows the THz waveforms from the LN line and a water line under the same experimental conditions. The THz peak field from LN is 0.4 times weaker than that from water. However, the THz signal from LN shows a shorter pulse duration. By fitting the envelope of the THz signal in field, the THz signal from LN has a pulse duration of 0.6 ps. For the THz signal from water, the THz pulse duration is about 1 ps. The corresponding spectra are shown in the inset. The LN shows a broader bandwidth with more high-frequency components. There are two possible reasons. First, LN has a low absorption in THz regime because it's a nonpolar liquid. Additionally, the vaporized N2 keeps purging the system to preserve the high-frequency components.

5 Strong THz sideway emission from a liquid line

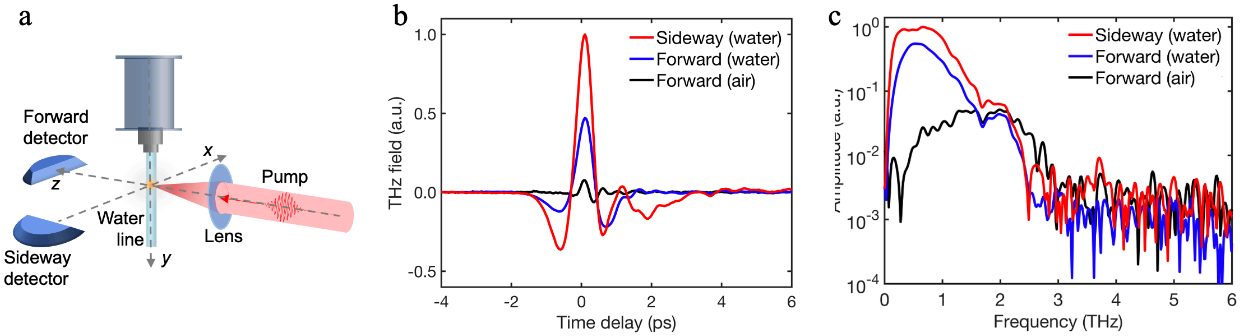

Fig. 27 Comparison of forward and sideway detection. a Setup schematic for forward and sideway detection. The pump beam is along *z*-axis. The water line flows in *y* direction. Two EO detectors are in z and -x directions, respectively. **b** Comparison of THz waveforms. The laser pulse energy is 0.4 mJ. Corresponding spectra are shown in **c**.

Similar to THz wave generation from a micro-plasma in air, a thin liquid target can also emit THz wave in the sideway directions. **Fig. 27a** schematically illustrates the system for the sideway measurement, in which the THz radiation propagating perpendicular to z direction is measured via EOS. 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. **Fig. 27b** shows the comparison of THz waveforms generated from water and air, respectively. Here, a 210 μm-thick-water line is used as a THz 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. In this case, the optimal pulse duration is about 534 fs. And the shortest pulse duration (~ 140 fs) is used for the air plasma generation.

The waveforms in **Fig. 27b** 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 direction. There is no detectable THz signal from air in sideway in this case. 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 **Fig. 27c**. Both signals from water in sideway and forward direction shows a narrower bandwidth than that from air plasma. The strong sideway THz signal might result from 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.

6 Preliminary results of THz wave generation from solid targets

We are collaborating with the Laboratory for Laser Energetics (LLE) researchers on exploring intense THz wave generation from relativistic electrons, shown in **Fig. 28**. This preliminary test on the solid targets. The laser used is the Multi-Terawatt (MTW) laser (a high intensity laser pulse (I > 10^{18} W/cm^2), 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 peak power at focus is 10^{19} W/cm^2.

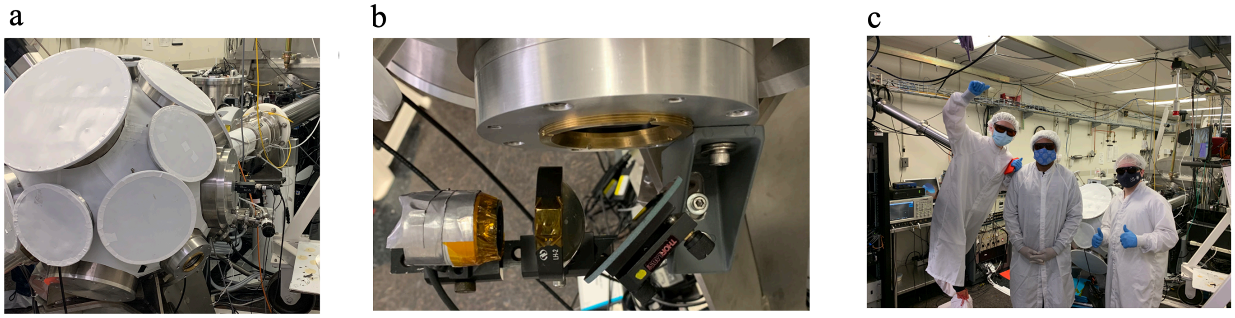

Fig. 28 a The target chamber for THz wave generation using a multi-terawatt laser at LLE, Rochester. **b** THz detection from one of the chamber windows. **c** PhD students, Gerrit Bruhaug (right) and Kareem Garriga (middle) and Research Associate Dr. Yiwen E (left) in the testing room.

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. We also tried the wire target and Cu target. All the preliminary results are in line with our expectations. We are working on the single shot measurement of coherent THz pulses to get more valuable information.