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UNITED STATES OF AMERICA

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TECHNICAL REPORT

# Science of Broadband THz Wave Photonics: Generation and Detection with Gases

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**14. ABSTRACT**

Our scientific objective is to understand the science of broadband THz wave generation and detection with gases. Specifically, we are studying on using ambient air and selected gases as THz wave emitters and sensors with excitation by pulsed laser for remote detection. In the proposed two (2) optional years, we will continue to systematically study inert gases and molecular gases which are used to emit and detect THz waves under different conditions (photo-ionization potential, pressure, third-order optical nonlinearity, and static breakdown field threshold). We will continue the investigation of the ability to utilize THz sources to remotely detect and spectrally identify weapons of mass destruction-linked aerosols or other airborne particles. Our targeted distance for the remote THz wave generation and detection is one kilometer.

**15. SUBJECT TERMS**

Terahertz wave, laser-induced air plasma, remote detection, laser

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# UNIT CONVERSION TABLE

## U.S. customary units to and from international units of measurement[*]

| U.S. Customary Units | Multiply by →<br>← Divide by[†] | International Units |
|---|---|---|
| **Length/Area/Volume** | | |
| inch (in) | 2.54 × $10^{-2}$ | meter (m) |
| foot (ft) | 3.048 × $10^{-1}$ | meter (m) |
| yard (yd) | 9.144 × $10^{-1}$ | meter (m) |
| mile (mi, international) | 1.609 344 × $10^{3}$ | meter (m) |
| mile (nmi, nautical, U.S.) | 1.852 × $10^{3}$ | meter (m) |
| barn (b) | 1 × $10^{-28}$ | square meter ($m^2$) |
| gallon (gal, U.S. liquid) | 3.785 412 × $10^{-3}$ | cubic meter ($m^3$) |
| cubic foot ($ft^3$) | 2.831 685 × $10^{-2}$ | cubic meter ($m^3$) |
| **Mass/Density** | | |
| pound (lb) | 4.535 924 × $10^{-1}$ | kilogram (kg) |
| unified atomic mass unit (amu) | 1.660 539 × $10^{-27}$ | kilogram (kg) |
| pound-mass per cubic foot (lb $ft^{-3}$) | 1.601 846 × $10^{1}$ | kilogram per cubic meter (kg $m^{-3}$) |
| pound-force (lbf avoirdupois) | 4.448 222 | newton (N) |
| **Energy/Work/Power** | | |
| electron volt (eV) | 1.602 177 × $10^{-19}$ | joule (J) |
| erg | 1 × $10^{-7}$ | joule (J) |
| kiloton (kt) (TNT equivalent) | 4.184 × $10^{12}$ | joule (J) |
| British thermal unit (Btu) (thermochemical) | 1.054 350 × $10^{3}$ | joule (J) |
| foot-pound-force (ft lbf) | 1.355 818 | joule (J) |
| calorie (cal) (thermochemical) | 4.184 | joule (J) |
| **Pressure** | | |
| atmosphere (atm) | 1.013 250 × $10^{5}$ | pascal (Pa) |
| pound force per square inch (psi) | 6.984 757 × $10^{3}$ | pascal (Pa) |
| **Temperature** | | |
| degree Fahrenheit (°F) | [T(°F) − 32]/1.8 | degree Celsius (°C) |
| degree Fahrenheit (°F) | [T(°F) + 459.67]/1.8 | kelvin (K) |
| **Radiation** | | |
| curie (Ci) [activity of radionuclides] | 3.7 × $10^{10}$ | per second ($s^{-1}$) [becquerel (Bq)] |
| roentgen (R) [air exposure] | 2.579 760 × $10^{-4}$ | coulomb per kilogram (C $kg^{-1}$) |
| rad [absorbed dose] | 1 × $10^{-2}$ | joule per kilogram (J $kg^{-1}$) [gray (Gy)] |
| rem [equivalent and effective dose] | 1 × $10^{-2}$ | joule per kilogram (J $kg^{-1}$) [sievert (Sv)] |

[*] Specific details regarding the implementation of SI units may be viewed at http://www.bipm.org/en/si/.

[†] Multiply the U.S. customary unit by the factor to get the international unit. Divide the international unit by the factor to get the U.S. customary unit.

2015-11-16

## Project Description

Our scientific objective is to understand the science of broadband THz wave generation and detection with gases. Specifically, we are studying on using ambient air and selected gases as THz wave emitters and sensors with excitation by pulsed laser for remote detection. In the proposed two (2) optional years, we will continue to systematically study inert gases and molecular gases which are used to emit and detect THz waves under different conditions (photo-ionization potential, pressure, third-order optical nonlinearity, and static breakdown field threshold). We will continue the investigation of the ability to utilize THz sources to remotely detect and spectrally identify weapons of mass destruction-linked aerosols or other airborne particles. Our targeted distance for the remote THz wave generation and detection is one kilometer.

Our tasks are (I) Basic Science, (II) Experimental Verification, (III) Exploring Limitations, and (VI) Remote Operation. In the two (2) optional years, our additional projects under above tasks are: direct measurement of THz field distribution inside of a laser induced filament; THz wave with arbitrary polarization; demonstration of intense THz wave generation (target for 5 to 10 MV/cm); and systematic study of THz wave generation with the excitation of sub-10 fs laser pulses.

## Summary on the achievements in the two optional years

(1) In order to further understand the basic science, we performed numerical simulation based on our full quantum mechanical model to reproduce our experimental results on phase-dependent THz wave emission spectrum from two-color laser-induced gas plasma (our simulation results along with the experimental ones will be submitted to Physical Review Letters in a few months);

(2) In an effort to increase the THz wave emission efficiency and get high-field THz wave generation from laser-induced gas plasma we use different laser polarization combinations and different gases species with higher nonlinearity to generate THz waves. We have shown that a circularly polarized fundamental beam (800 nm) combined with a linearly polarized second harmonic (400 nm) can generate higher THz emission in comparison to the case when linearly polarized 800-nm beam is combined with linearly polarized 400-nm beam. We verified that some metallic vapors, due to its higher optical nonlinearity, can generate much higher THz wave emission at low pressure compared to those regular gases (i.e., nitrogen and those inertia gases) at the similar pressure. Very importantly, we have recently demonstrated that, using airy laser beam, the THz emission efficiency can be further increased. Besides, we have demonstrated that very thin metal films excited by two-color laser fields can generate broadband THz waves, which provides an alternative intense THz source;

(3) For the purpose of remote sensing, we have successfully extended THz wave remote sensing distance in ambient air from the previous 10 meters to 20 meters, and finally to 30 meters, using THz radiation enhanced emission of fluorescence (THz-REEF) by designing a new optical fluorescence collection system. We also indicated that, laser-induced gas plasma not only can emit forward THz radiation but also backward radiation, which gives more options for THz wave remote sensing (using either THz radiations from the two opposite emission directions as the remote sensing source). More importantly, THz wave generation from micro-sized plasma has been demonstrated very recently. This new achievement greatly reduces the requirement on the laser pulse energy (intensity) for THz wave generation from gas plasma and pushes THz remote sensing closer to the reality.

## Detailed achievements we have got in the research of two optional years

### *(1) Improvement on remote THz wave sensing distance using THz Radiation-Enhanced Emission of Fluorescence (THz-REEF) from previous 10 m to 20 m, and finally 30 m.*

We have performed remote THz wave sensing and extended the maximal sensing distance from previously reported about 10 m to more than 30 m using THz-REEF. **Fig.1** shows the experimental setup and THz waveforms sensed at different distances (0.1 m, 7 m, 14 m, 20 m, and 30 m, respectively). Currently the maximal sensing distance is limited by either the lab space or signal-to-noise ratio (SNR). In order to increase the SNR we will improve both the THz electric field and electronics used to get the signal from fluorescence emission of the plasma. Another factor that affects the SNR is the air turbulence in the atmosphere. We proposed to use adaptive optics to mitigate the THz signal and/or fluorescence fluctuation.

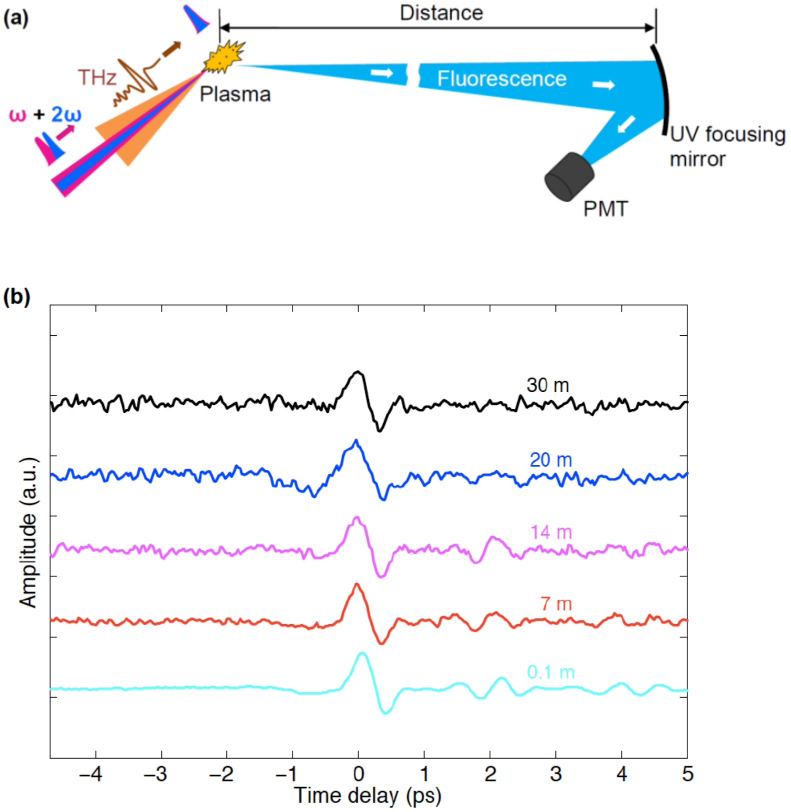
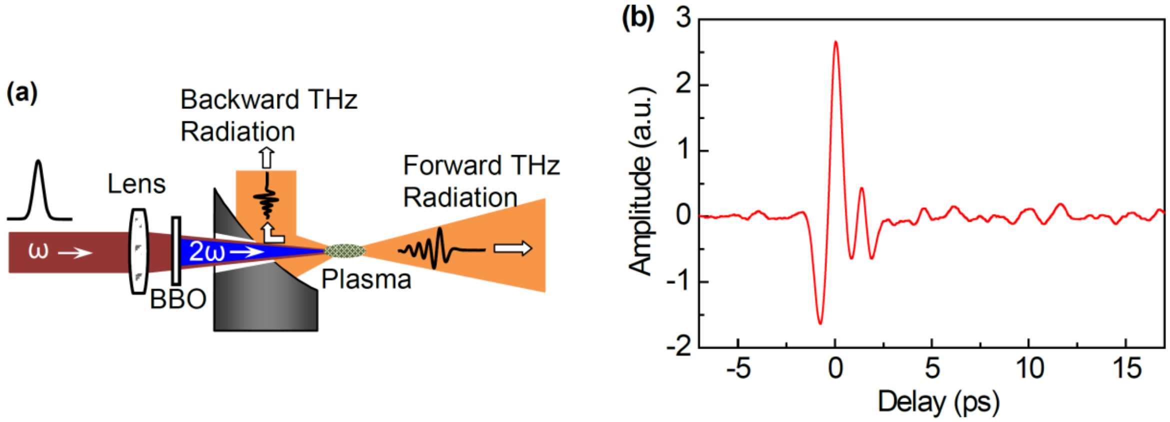


**Fig. 1**. **(a)** Schematic of the experimental setup used to remotely sense THz waves using two-color (ω and 2ω) THz radiation enhanced emission of fluorescence (THz REEF) at different distances; **(b)** THz waveforms sensed/detected at distances of 0.1 m, 7 m, 14 m, 20 m, and 30 m respectively. PMT, photomultiplier tube.

### *(2) Backward THz radiation from two-color laser-induced gas plasma*



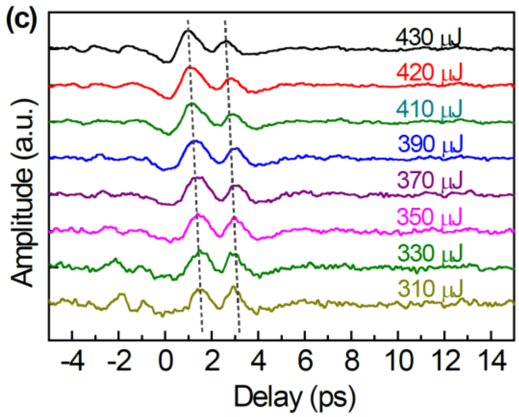


**Fig.2.** **(a)** Schematic illustration of the generation and collection of backward pulsed THz radiation from two-color laser induced gas plasma; **(b)** a typical THz waveform generated from the plasma pumped by a total pulse energy of about 500 $\mu$J using a lens with an effective focal length of 100 mm; **(c)** pump pulse energy dependence of THz waveforms: the results show that the waveform shifts to the left in delay as the pulse energy increases. The dashed lines are used to indicate the peak positions of the detected THz waveforms using electric-optic sampling with 1-mm <110> cut ZnTe crystal.

As shown in **Fig.2**, we found that there is very good THz wave emission in the backward direction with respective to laser pulse (ω and 2ω pulses) propagation direction. which gives more options for THz wave remote sensing (using either THz radiations from the two opposite emission directions as the remote sensing source)

### *(3) Phase-dependent THz wave emission spectrum in two-color laser-induced gas plasma*

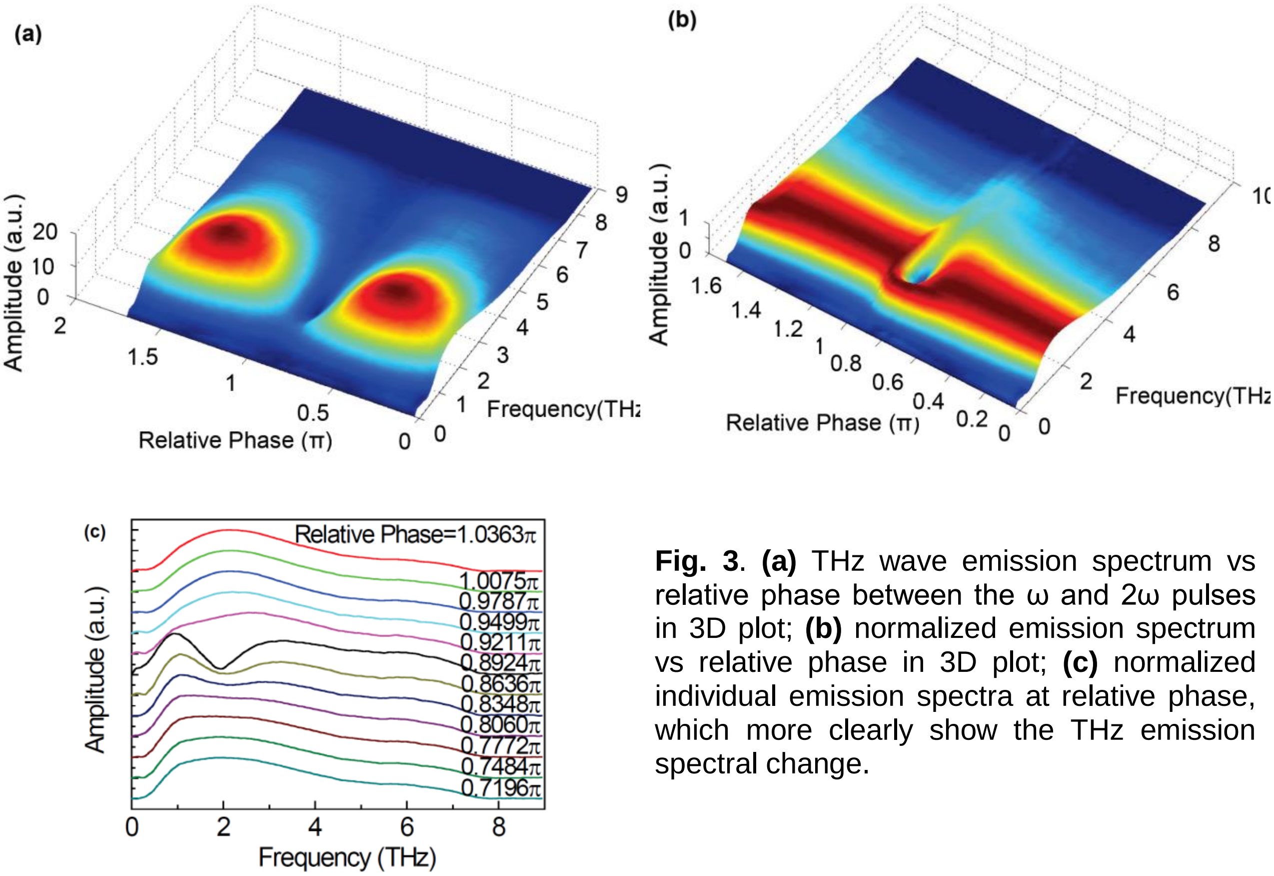


**Fig. 3**. **(a)** THz wave emission spectrum vs relative phase between the ω and 2ω pulses in 3D plot; **(b)** normalized emission spectrum vs relative phase in 3D plot; **(c)** normalized individual emission spectra at relative phase, which more clearly show the THz emission spectral change.

We have found that the change in the relative phase between the fundamental laser pulse and its second harmonic not only causes the THz wave emission efficiency change but also induces the emission spectral change, as shown in **Fig. 3**. The emission spectrum changes slightly (almost keeps unchanged in the normalized case in **Fig. 3(b)**) as the phase changes. However, the spectral change becomes dramatic when the relative phase approaches the value at which the THz emission efficiency becomes the lowest. The physical mechanism of such spectral change is investigated by numerical simulation based on our quantum mechanical model.

### *(4) Improvement on the efficiency of THz wave generation from laser-induced gas plasma using circularly polarized fundamental pulse (ω) in combination with linearly polarized second harmonic pulse (2ω)*

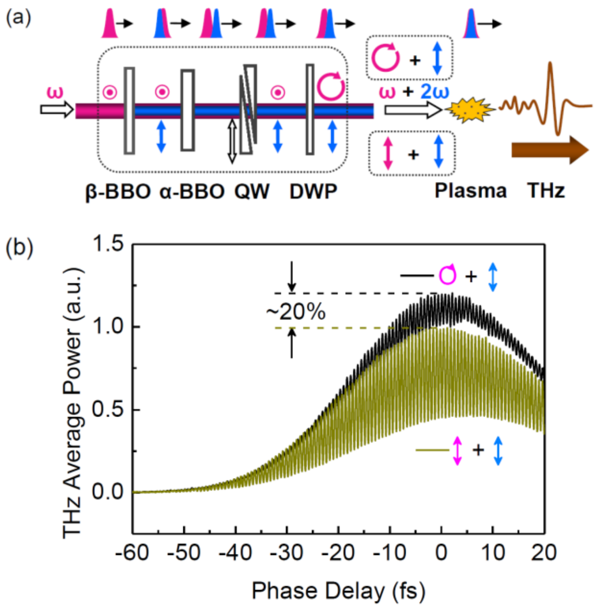
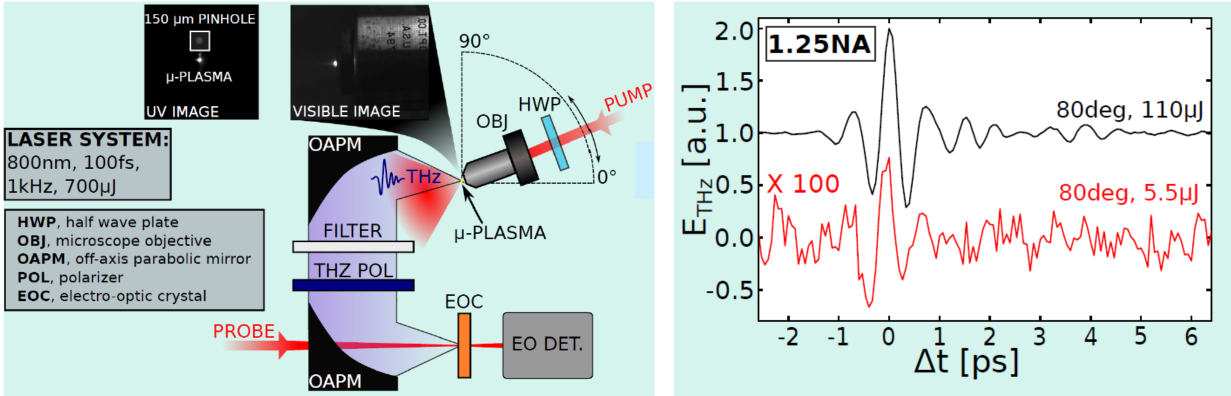


**Fig. 4**. **(a)** An inline phase compensator used to generate circularly or linearly polarized fundamental pulse (ω) in combination with a linearly polarized second harmonic pulse (2ω) pulse is also used to control the relative phase between ω and 2ω pulses; **(b)** two phase curves obtained with different polarization combinations (circular ω + linear 2ω and linear ω + linear 2ω). The maximum of each phase curve indicates the maximal emission efficiency of each case. QW, quartz wedge; DWP, dual-wavelength waveplate.

**Fig. 4(a)** shows an inline phase compensator used to generate circularly or linearly polarized fundamental pulse (ω) in combination with a linearly polarized second harmonic pulse (2ω) pulse. The same phase compensator is also used to control the relative phase between ω and 2ω pulses. **Fig.4(b)** indicates the difference in THz wave emission efficiency using different polarization combinations of the ω and 2ω pulses. An increase of the THz wave emission efficiency of ~20% is currently observed using circularly polarized fundamental pulse in combination with linearly polarized second harmonic pulse. Other polarization combinations will be tested in search for the maximal THz emission efficiency in the next reporting period. The THz average power is measured with a pyroelectric detector. The theoretical investigation will be performed to explain the experimental observation.

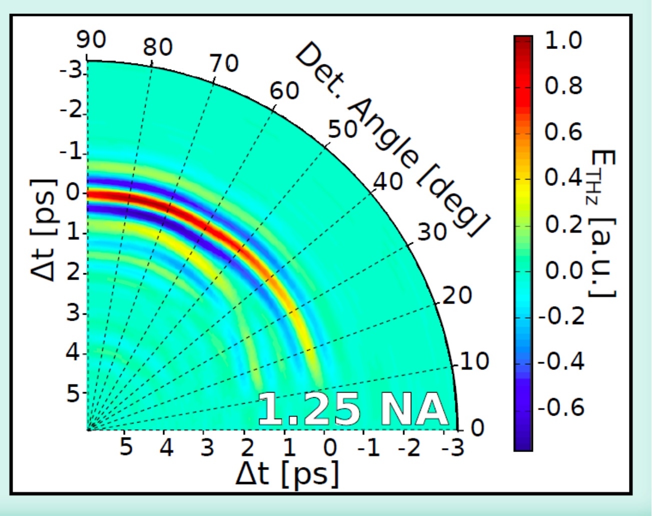
### *(5) Generation of THz radiation from a laser-induced micro-plasma*

We demonstrated THz wave generation using the lowest pulse energy down to μJ level using laser-induced micro-plasma created by focusing laser pulses into air or selected gases with a microscope objective. The pulse energy required for THz wave generation from micro-plasma has been reduced by an order of magnitude.



a)

(b)

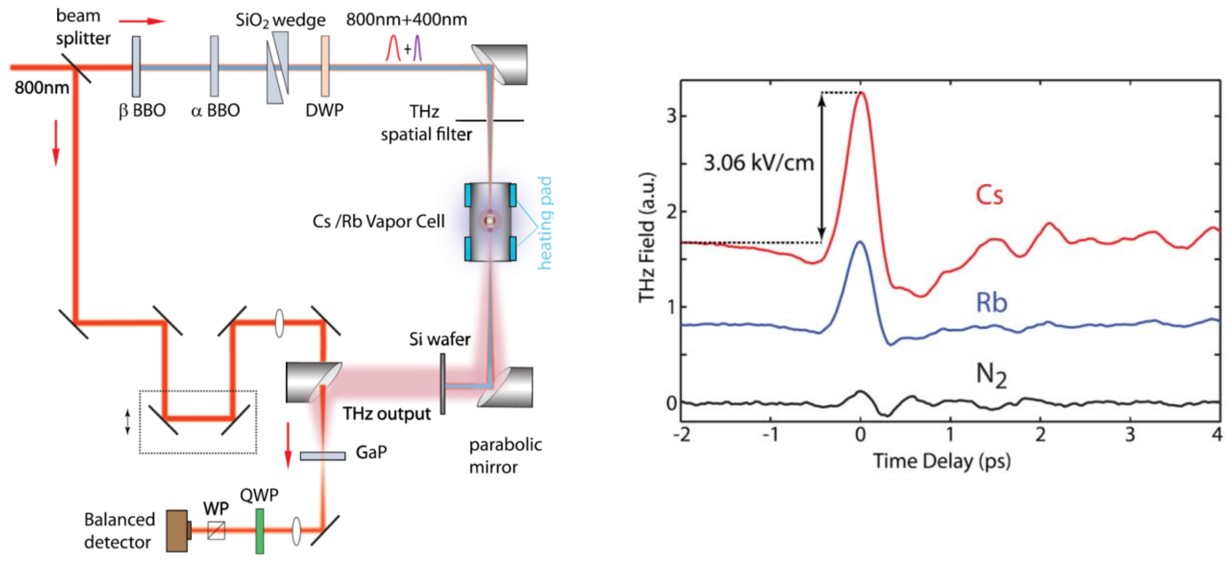


(c)

**Fig.5.** **(a)** Schematic illustration of generation and detection of pulsed THz radiation from laser-induced micro-plasma created by focusing the laser pulses with a microscope objective; **(b)** typical THz waveforms generated from micro-plasmas created with pulse energies of 110 and 5.5 $\mu$J, respectively, using a microscope objective with a numerical aperture (NA) of 1.25; **(c)** angular distribution of THz radiation from a laser-induced micro-plasma in air.

### *(6) Generation of THz waves from alkali metallic vapors, rubidium and cesium gases*

**Fig. 6 (a)** shows the experimental setup for THz wave generation from $N_2$, Rb, and Cs gases. **Fig. 6(b)** shows THz time waveforms recorded at a pressure of 0.45 Torr with an input pulse energy of 0.6 mJ, produced in Cs vapor (red), Rb vapor (blue), and nitrogen gas (black), respectively. From **Fig. 6(b)** one can see THz field strength generated from Cs gas is about one order of magnitude higher than that from nitrogen gas, indicating the potential of alkali gases for higher-field THz wave generation.



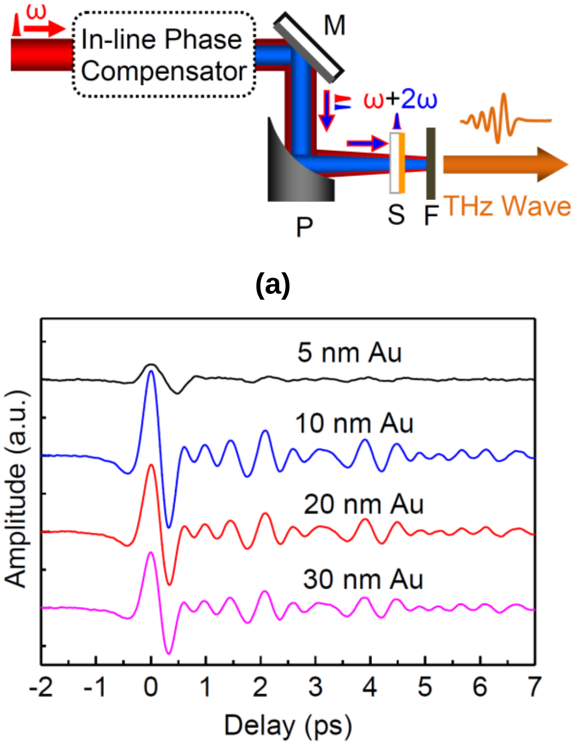
**(a)** **(b)**

**Fig. 6**. **(a)** Schematic of the experimental setup. DWP: dual-wavelength wave plate; QWP: quarter-wave plate; WP: Wollaston prism. The α-BBO is used to compensate the dispersion between the two-color waves. The SiO2 wedge pair functions as an inline phase compensator to modulate the relative phase between the two-color waves. The two parabolic mirrors on the right have a focal length of 4 in. The THz spatial filter (an iris) is to block the residual THz wave emitted from β-BBO. The silicon wafer is used to block the residual two-color waves output from the cell. The vapor cell has two end windows made with 1.59-mm-thick fused silica. pulses in 3D plot; **(b)** THz time waveforms recorded at a pressure of 0.45 Torr with an input pulse energy of 0.6 mJ, produced in Cs vapor (red), Rb vapor (blue), and nitrogen gas (black), respectively.

### ***(7) THz wave generation from thin metal films excited by asymmetrical optical field in transmission geometry***

We experimentally demonstrated terahertz (THz) wave emission from thin metal (gold) films excited by asymmetrical optical fields synthesized using an in-line phase compensator. By driving the electrons in thin metal films asymmetrically, THz wave emission is observed at normal incidence of two-color pump beams. Coherent control of THz wave emission from metal films suggests that a similar mechanism to that of the air-plasma THz source excited by two-color laser fields can be used to describe the generation processes.

**Fig. 7(a)** Schematic diagram of the experimental setup. M, high-reflection mirror at both 800 nm (ω) and 400 nm (2ω); P, parabolic mirror; S, thin gold film coated on z-cut sapphire crystal substrate; F, high-resistivity silicon filter used to block the residual 800 nm and 400 nm beams while passing through the THz beam. **Fig.7(b)** shows THz waveforms generated at normal incidence from Au films with different thicknesses, 5 nm, 10 nm, 20 nm, and 30 nm, respectively. For each waveform in the figure, the THz signal is optimized by changing the relative phase between 800 and 400 nm excitation pulses. Experimental results indicate when the thickness of gold film approaches 10 nm maximal THz wave emission can be obtained.



**(a)**

**(b)**

**Fig. 7.** **(a)** Schematic diagram of the experimental setup. M, high-reflection mirror at both 800 nm (ω) and 400 nm (2ω); P, parabolic mirror; S, thin gold film coated on z-cut sapphire crystal substrate; F, high-resistivity silicon filter used to block the residual 800 nm and 400 nm beams while passing through the THz beam. **(b)** THz waveforms generated at normal incidence from Au films with different thicknesses, 5 nm, 10 nm, 20 nm, and 30 nm, respectively.

**Publication List (published)**

1. Anna Mankova and X.-C. Zhang, "Terahertz time-domain and FTIR spectroscopy of tris-crown interaction", Chemical Physical Letters, Vol. 554, 201 (2012).
2. Xiaofei Lu and X.-C. Zhang, "Generation of Elliptically Polarized Terahertz Waves from Laser-Induced Plasma with Double Helix Electrodes", Physical Review Letters, Vol. 108, No 12, 123903 (2012).
3. K. Iwaszczuk and X.-C. Zhang et al, "Terahertz field enhancement to the MV/cm regime in a tapered parallel plate waveguide", Optics Express, Vol. 20, No. 8, 8344 (2012).
4. D. Brigada and X.-C. Zhang, "Chemical identification with information-weighted terahertz spectrometry", IEEE Transactions on Terahertz Science and Technology, Vol. 2, No. 1, 107 (2012).
5. Ikurou Umezu, X.-C. Zhang et al, "Emergence of very broad infrared absorption band by hyperdoping of silicon with chalcogens", Journal of Applied Physics, Vol. 113, No. 21, 213501 (2013).
6. A.V. Borodin, and X.-C. Zhang et al, "Transformation of terahertz spectra emitted from dual-frequency femtosecond pulse interaction in gases", Optics Letters, Vol. 38, No. 11, 1906 (2013);
7. Anna Mankova and X.-C. Zhang, "Terahertz time-domain and FTIR spectroscopy of tris-crown interaction", Chemical Physical Letters, Vol. 560, 55 (2013).
8. X.-C. Zhang, "Bright THz Source", Nature Photonics, Vol. 7, 670 (2013).
9. Xuan Sun and X.-C. Zhang, "Terahertz Radiation in Alkali Vapor Plasmas", Applied Physics Letters, Vol. 109, No. 19, 191106 (2014).
10. Jianming Dai and X.-C. Zhang, "Terahertz wave generation from thin metal films excited by asymmetrical optical fields", Optics Letters, Vol. 39, No. 4, 777 (2014).
11. Xiaofei Lu and X.-C. Zhang, "Investigation of ultra-broadband terahertz time-domain spectroscopy with terahertz wave gas photonics", Frontiers of Optoelectronics, Vol. 7, No. 2, 121 (2014).
12. Jingle Liu and X.-C. Zhang, "Terahertz radiation-enhanced-emission-of-fluorescence", Frontiers of Optoelectronics, Vol. 7, No. 2, 156 (2014).
13. I. Chen Ho and X.-C. Zhang, "Application of broadband terahertz spectroscopy in semiconductor nonlinear dynamics", Frontiers of Optoelectronics, Vol. 7, No. 2, 220 (2014).
14. Benjamin Clough and X.-C. Zhang, "Toward remote sensing with broadband terahertz waves", Frontiers of Optoelectronics, Vol. 7, No. 2, 199 (2014).
15. YutingW Chen and X.-C. Zhan, "Anti-reflection implementations for terahertz waves", Frontiers of Optoelectronics, Vol. 7, No. 2, 243 (2014).
16. Jing Zhang, "Polarization-dependent study of THz air-biased coherent detection", Optics Letters, Vol. 39, No. 14, 4096 (2014).
17. X.-C. Zhang, "Message from incoming editor-in-chief", Optical Letters, Vol. 39, No. 1, ED1 (2014).
18. Fabrizio Buccheri and X.-C. Zhang, "Terahertz emission from laser-induced microplasma in ambient air", Optica, Vol. 2, No. 4, 366 (2015).

19. Fabrizio Buccheri and X.-C. Zhang, "Terahertz wave generation based on laser-induced microplasma", IEEE Transactions on Terahertz Science and Technology, Vol. 8, No. 2, 58 (2015).
20. Liangliang Zhang, Kaijun Mu, Yunsong Zhou, Hai Wang, Cunlin Zhang, and X.-C. Zhang, "High-power THz to IR emission by femtosecond laser irradiation of random 2D metallic nanostructures", Nature: Scientific Reports, Vol. 5, 12536 (2015).

## Papers in preparation

1. Liangliang Zhang, Fabrizio Buccheri, Cunlin Zhang, and Xi-Cheng Zhang, "Terahertz emission from thin metal films with porous nanostructures", to be published on Applied Physics Letters, 2015.
2. Kang Liu , D. G. Papazoglou, A. D. Koulouklidis, S. Tzortzakis, and X.-C. Zhang, "Terahertz wave emission from abruptly autofocusing beam induced air plasma", in preparation, to be submitted to Applied Physics Letters, 2015.

## Presentations (oral)

1. Kham Lim and X.-C. Zhang et al, "Broadband THz detection in the counter-propagating configuration using THz-enhanced plasma fluorescence", CLEO 2013, San Jose, CA, June 13, 2013
2. X.-C. Zhang, "THz wave air photonics, covering the gap and beyond", Seminar at National Chiao Tung University, Tsingchu, Taiwan, March 25, 2013
3. X.-C. Zhang, "Next rays, T-ray!", Tin Ka Pin Lecture at National Chiao Tung Univerity , Tsingchu, Taiwan, March 27, 2013
4. X.-C. Zhang, "THz wave air photonics, covering the gap and beyond", Short Pulse Strong Field Laser Physics Symposium Honoring See Leang China, Quebec City, Quebec, Canada, May 22, 2013
5. X.-C. Zhang, "Terahertz Technology and Application", SPIE Defense, Security, and Sensing 2013, Baltimore, MD, May 2, 2013
6. X.-C. Zhang, "Passion for THz wave photonics", Lecture at Shanghai University of Science and Technology, Shanghai, China, June 5, 2015
7. X.-C. Zhang, "Handheld THz spectroscopy (micro-Z)", International 4rd THz-Bio Workshop, Seoul, Korea, February 7, 2013
8. X.-C. Zhang, "Introduction of THz", Seminar at National Central University, Taiwan, March 26, 2013
9. X.-C. Zhang, "THz wave photonics", The sixth International Symposium on Ultrafast Photonics Technologies, Rochester, New York, October 22, 2013
10. Kang Liu, Fabrizio Buccheri, Jianming Dai, and X.-C. Zhang, "Light Filamentation Science", ARO-MURI program review at Washington DC, November 18, 2013

11. Jianming Dai and X.-C. Zhang, "THz wave generation from nm thick metal films excited by asymmetrical optical fields", International 5th THz-Bio workshop, Seoul, Korea, April 3, 2014
12. Jianming Dai and X.-C. Zhang, "THz wave generation from nm thick metal films excited by asymmetrical optical fields", lecture at Wuhan National Laboratory for Optoelectronics, Wuhan, Hubei, China, June 10, 2014
13. Jianming Dai and X.-C. Zhang, “Broadband Terahertz Wave Emission from Thin Films Excited by Two-Color Laser Fields”, IRMMW-THz 2014, Tucson, AZ, USA, Sept. 14~19, 2014.
14. Jing Zhang and X.-C. Zhang, “Measurement of birefringence insides an air plasma by THz-ABCD”, IRMMW-THz 2014, Tucson, AZ, USA, Sept. 14~19, 2014.
15.
16. Xuan Sun and X.-C. Zhang, “THz wave generation from Cesium vapor”, IRMMW-THz 2014, Tucson, AZ, USA, Sept. 14~19, 2014.
17. Fabrizio Buccheri and X.-C. Zhang, "THz wave generation from micro-plasma", SPIE/COS Photonics Asia 2014, Beijing, China, November 2014.
18. Fabrizio Buccheri, Kang Liu, and X.-C. Zhang, "Emission of THz radiation from micro-plasma", 5th International Symposium on Filamentation (COFIL2014), Shanghai, China, September 20, 2014.
19. Fabrizio Buccheri and X.-C. Zhang, "Terahertz wave emission from laser-induced micro-plasma", 39th International Conference on Infrared Millimeter and Terahertz Waves (IRMMW-THz 2014), , Tucson, Az, United States, September 2014. (Keynote talk)

**Patent(s)**

1. Jingle Liu and X.-C. Zhang, "A plasma diagnostic method using terahertz-wave-enhanced fluorescence", Issued on February 12, 2014, US Patent# 8653462.

**Awards**

1. X.-C. Zhang, "William F. Meggers Award" from Optical Society of America, October 12, 2012
2. Leva McIntire, "Newport Award" from Newport Corporation, March 6, 2013
3. Leva McIntire, "Horton Fellowship" from Horton Fellowship for supporting student research acitivities, April 15, 2013
4. Leva McIntire, "NASA Honor Award" from NASA Honor Award for Team Achievement, June 21, 2013

5. X.-C. Zhang, "Kenneth J Button Prize recipient for 2014 for Outstanding Contributions to the Science of the Electromagnetic Spectrum" from IRMMW-THz 2014, September 16, 2014
6. X.-C. Zhang, "Editor-In-Chief" from Optics Letters, January 1, 2014
7. Kang Liu, "Research Mobility Travel Grant", February, 2014

## Opportunities for training and professional development has the project provided

Graduate students, Fabrizio Buccheri, Kang Liu, Xuan Sun, and Leva McIntire have been well trained by being involved in the project supported by DTRA. Kang Liu is one of the two students who are the main players to recently extending the THz remote sensing distance from the previous 10 m to 20 m and finally to 30 m.

Moreover, two undergraduate students (currently in their 4rd year), Natalie Pastuszka and Christoper Marsh in the Institute of Optics at University of Rochester will join the THz wave air photonics team. They are currently supported by NSF REU for a year and will potentially become graduate students to continue to work on THz wave air photonics for further training and career development.

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