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Intense broadband THz generation from femtosecond laser filamentation

Tie-Jun Wang*, Shuai Yuan, Yanping Chen, and See Leang Chin

Centre d’Optique, Photonique et Laser (COPL) and Département de Physique, de Génie Physique et D’Optique,
Université Laval, Québec, Québec G1V 0A6, Canada

*Corresponding author: tie-jun.wang.1@ulaval.ca; tjw502007@yahoo.com

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Broadband and energetic terahertz (THz) pulses can be remotely generated in air through filamentation. We review such THz generation and detection in femtosecond Ti:sapphire laser induced remote filaments. New results are presented on the direct relationship between THz generation in a two color filament and induced N₂ fluorescence through population trapping during molecular alignment and revival in air. This further supports the new technique of remote THz detection in air through the sensitive measurement of N₂ fluorescence.

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1. Introduction

Terahertz (THz) wave is an electromagnetic radiation in the frequency interval between 0.1 and 10 THz. Compared to other frequency windows, THz has its special advantages for spectroscopy and imaging. For instance, from gigahertz (GHz) to THz frequencies, numerous organic molecules exhibit strong absorption and dispersion due to rotational and vibrational transitions. These transitions are specific to the targets and enable THz fingerprinting in spectroscopy. THz waves can also penetrate many common materials such as paper, plastic, and ceramic, glass, wood, and clothes; THz has a low photon energy (4 meV @ 1 THz, one million times weaker than an X-ray photon) and will not cause harmful photoinization in biological tissues, etc. THz science and technology based on table-top laser system is finding applications. With the rapid development of THz source technology, numerous or- ganic molecules can be imaged by THz waves due to novel advantages for spectroscopy and imaging. For instance, THz radiation is a promising alternative for imaging due to rotational and vibrational transitions. These transitions are specific to the targets and enable THz fingerprinting in spectroscopy. THz waves can also penetrate many common materials such as paper, plastic, and ceramic, glass, wood, and clothes; THz has a low photon energy (4 meV @ 1 THz, one million times weaker than an X-ray photon) and will not cause harmful photoinization in biological tissues, etc. THz science and technology based on table-top laser system is finding use in an increasingly wide variety of important applications. With the rapid development of THz source technique, MV/cm level of THz field is available based on table-top laser system[1]. High-field physics in the THz range becomes a very exciting frontier[2–40].

However, the delivery of energetic THz pulses for spectroscopy at a remote position in the atmosphere is limited by linear defraction and the strong attenuation due to water vapor absorption. THz generation from femtosecond laser remote filamentation in gases, especially in air, is a promising alternative because the technique allows the generation of intense near single-cycle THz pulses at a long distance by controlling the remote onset of the filament via controlling the initial laser parameters. Generation of intense THz pulses with large bandwidth from inside the filament in air is an active area of current research on THz science. Such broadband and rather powerful THz pulses would provide a new prospective tool for remote THz nonlinear optics and spectroscopy.

In this review, we briefly summarize the development of THz pulse generation from femtosecond laser-induced plasma filament, mainly from the experimental points of view, with emphasis on remote generation and detection of high energy, broadband THz pulses. In particular, we shall present new results on the direct measurement of THz generation and N₂ fluorescence in a two color filament. Because of population trapping, the two signals followed each other during molecular alignment and revival in air. The results support the new technique of remote detection of THz pulses through the sensitive measurement of N₂ fluorescence as reported in Ref.[39,40].

2. THz generation from femtosecond laser filamentation

The first single-cycle THz generation from a photo-induced plasma was reported by Hamster et al. in 1993 by focusing laser pulses with a power of 1 TW and a duration of 100 fs into a He gas[3]. The THz emission mechanism[4] in their experiments was based upon space charge separation by the ponderomotive force generated by the optical beam, leading to a conical THz emission. Since then, other techniques have been used to generate intense THz emission. For example, by applying a transverse DC bias to the plasma region, an order-of-magnitude increase in the THz field was reported by Löffler et al.[5,6]. Recently, Houard et al.[7] and Chen et al.[8] have applied external electric fields transversely to single-color filaments and Liu et al.[9] have applied an external electric field longitudinally to single-color filaments. 3 orders enhancement of THz amplitude with up to 10 kV/cm external electric field has been reported[7]. By applying a helical electrical field along a plasma region, Lu et al.[10] could generate an elliptically polarized THz wave. An intense THz generation can also be generated from air plasma driven by an intense few-cycle laser pulse[11,12]. The effect of the carrier envelope phase of few-cycle laser pulses on THz emission was predicted by Wu et al.[11] and later on experimentally demonstrated by Bai et al.[12].

A four-wave rectification method by focusing the fundamental wave (FW) and second-harmonic wave
using a converging lens followed by an axicon resulting in efficiency from two-color plasma ported more than 2 times increase in THz generation.

The FW of a Ti:sapphire based laser at a distance of remote THz generation from the filaments induced by as mentioned above. D'Amico et al. demonstrated that THz emission from a two-color plasma filament could be amplified by another two-color plasma. The THz wave amplitude from a two-color filament can also be enhanced by more than eight times with an optimal aperture-limited pump laser beam according to Ref. [18] by Peng et al. Replacing the focusing lens with a gold off-axis parabolic mirror, Blanchard et al. reported more than 2 times increase in THz generation efficiency from two-color plasma[19]. More recently, by using a converging lens followed by an axicon resulting in a tailored two-color laser filament in air, Manceau et al. reported significant THz temporal pulse shortening from a uniform plasma strings[20]. The polarization of THz emission from a two-color filament in air can be coherently controlled by changing the optical phase between the FW and its SHW and (c) THz radiation energy versus time sequence of the two color beams. At a distance of 16 m, forward conical emission was observed whose origin they attributed to a Cerenkov-type mechanism from the laser-pulse induced plasma wake. Their conclusion was based on the measurement obtained with a heterodyne detector (sensitive only to 0.1 THz with a bandwidth of 4 GHz). Later, by using an electro-optic sampling (EOS) technique (sensitive to frequencies below 4 THz), it was demonstrated by Chen et al.[33] and Zhang et al.[34] that the THz pulses in the forward direction of a filament were elliptically polarized. The physical mechanism is that filament induced birefringence in gases provides a phase delay between the two orthogonal components of the THz field, leading to an elliptically polarized THz emission. Liu et al.[35] demonstrated that by sending a sequence of two femtosecond IR laser pulses at 800 nm separated by less than 3 ns, forming two overlapping filaments in air (so called bifilamentation), they can generate 1 order of magnitude more intense THz emission than the transient-Cherenkov THz emission[32] from the plasma filament. The origin is attributed to the emission from a bimodal transmission line created by a pair of neighboring plasma filaments. Even so, the remotely generated THz pulse by femtosecond laser filamentation of the FW of a Ti:sapphire based laser system is still very weak (less than nJ/pulse).

For remote high energy, broadband THz generation, so far, two-color filamentation is strongly desired and is also challenging. Phase compensation between the two color beams is necessary so that the fundamental and second harmonic pulses overlap in the remote filament zone with parallel linear polarizations; this needs to be precisely controlled. One could set up a delay stage and a pair of ultra-thin wedges in one arm of the two-arm scheme (FW and SHW in each arm, respectively). However, fluctuation and mechanical instability would be serious in the two-arm scheme especially for long distance experiments according to our experience. Recently, an in-line phase compensation scheme was reported by Dai et al.[21] to improve the stability. We adopted the setup in Ref. [21]

3. Remote THz generation

An important reason why THz generation from femtosecond laser induced plasma filaments in air recently attracts much attention is that THz emission can be generated close to a remote target. Remote THz generation minimizes the importance of propagation issues as mentioned above. D’Amico et al. reported the first remote THz generation from the filaments induced by the FW of a Ti:sapphire based laser at a distance of 30 m from the laser in 2007[32]. A radially polarized
together with a reflective-type telescope (Fig. 1(a)) to remotely generate high energy THz pulses in air\([36]\) using high energy large beam diameter (~30 mm) Ti:sapphire laser pulses. Here is a brief explanation of our design.

In order to decrease the nonlinear effects inside the optics of the in-line setup and also considering the large pump beam size, optics with large aperture had to be used. For frequency doubling, a 0.5-mm-thick KDP crystal with aperture >30 mm was chosen. At the output, for maximum conversion efficiency, the polarizations of the FW and SHW were orthogonal. When they passed through a birefringent crystal—X-cut BBO with a clear aperture >30 mm, the FW was delayed compared to the SHW (so-called negative delay). All the following optics and propagation media (air) provided positive delays, which would result in the temporal overlap of the two pulses in a predetermined filament zone. A pair of ultra-thin wedges (BK7, rectangular: 50×30 (mm), 4° wedge, angle, and 0.5 mm minimum thickness) could precisely control the relative phase between the two beams. A dual-wavelength wave plate (DWP) with a clear aperture of 30 mm rotated one of the polarizations by 90° and the other by 180°. The latter means that the polarization is un-affected. Thus, at the output of the DWP, the polarizations of the two pulses are parallel. A reflective-type telescope could project the two-color filament at a long distance. Its aberrations were minimized by using a concave off-axis parabolic reflector as a secondary mirror for the telescope. The THz radiation was collected and focused on a pyroelectric detector (Coherent P4-45CC) by a 90° off-axis, 4-inch in diameter Au-coated parabolic mirror. A 0.38-mm-thick 4-inch-diameter Si filter with another 0.5-mm-thick, 4×4 (mm) square Si-filter, stuck at the center of the former, were used to filter out the FW and SHW beams. The Si filter could transmit frequency <300 THz. Another 1.6-mm-thick Teflon was used as a long-pass wavelength filters to verify the spectral components (<5.5 THz) of the THz pulse. By minimizing the aberration of a telescope and optimizing the pump pulse duration\([37]\), the beam profile, the delay time and the polarization scheme between the FW and SHW pulses (Fig. 1(b)), a record pulse energy of 0.57 μJ with frequency less than 5.5 THz and 2.8 μJ for the entire super-broadband pulse (Fig. 1(c)) were measured at a `remote’ distance of 16 m\([36,38]\).

One can also remotely control this strong THz radiation by pre-aligned air molecules through rotational Raman excitation. During the filamentation of femtosecond laser pulses in air, most of the molecules survive in the plasma filament due to weak molecular ionization\([41]\). However, the clamped intensity of \(\sim 5 \times 10^{13} \text{ W/cm}^2\) in the filaments of the laser pulses is strong enough to align the neutral molecules through rotational Raman excitation, resulting in field-free periodic revivals. The typical revival periods of \(\text{N}_2\) and \(\text{O}_2\) molecules in air are 8.4 and 11.6 ps, respectively. Recently, Durand et al. reported a control of THz emission from bifilamentation of two FW pulses based on molecular lensing effect\([42]\). The emission was characterized by a heterodyne THz detector, which was sensitive at 0.1 THz with a bandwidth of 4 GHz. Wu et al.\([43]\) and Wang et al.\([44]\) reported the molecular alignment effect on THz emission from two-color filaments in air. Air molecules were pre-aligned by the FW pulses\([43]\) or a pair of two-color laser pulses\([44]\). THz emission was generated by another pair of two-color laser pulses. By tuning the delay time between the alignment laser pulses and THz generating laser pulses around the air molecular revival time, significant modulations of the amplitude of the THz electric fields were observed. The peak-to-peak amplitude of THz electric field as a function of delay time is shown in Fig. 2 at each revival delay position of \(\text{N}_2\) and \(\text{O}_2\) including quarter, half, three-quarter, and full revival periods. \(T_{\text{N}_2}\) and \(T_{\text{O}_2}\) are typical revival periods of \(\text{N}_2\) and \(\text{O}_2\).

More recently, a long distance dependence of remote THz generation from two-color filamentation in air was investigated by Daigle et al.\([45]\). THz signals were still observed when the filaments were positioned 55 m from the source. But the results obtained revealed that as the filament bunch moved towards longer distances, the produced THz signal decreased monotonically. Through a simple numerical simulation the decaying mechanisms were identified as an eventually unavoidable group velocity mismatch between the two-color pulses and the diffraction/divergence of the non-filamenting second harmonic pulse\([45]\). For example, assuming that both the pulses of FW and SHW had a pulse duration of 75 fs, the maximum interaction length of the two pulses was limited to 1.8 m according to the Eq. (7) in Ref. [45]. Any section of a filament whose length exceeded this value would not produce significant THz signal. Moreover, when the propagation distance increased, the intensity of the SHW pulse decreased due to its divergence. The production of strong THz pulses beyond 100 m from the current laser source is very challenging. A promising method to have strong THz generation beyond 100 m consists of increasing the clamped intensity by using a laser pulse with a longer wavelength since the generated THz field is proportional to the intensity of the FW pulse in FWM\([31]\). Good beam profile, small F-number of focusing telescope beyond 100 m, and stable spatial and temporal control of two-color beam would also contribute to this remote THz generation.

### 4. Remote THz detection

Towards THz remote sensing, Prof. Zhang’s group recently introduced a technique for standoff THz wave...
sensing. Adapted from Liu et al.\cite{39,40}.

The mechanism of THz-REEF from a plasma could be attributed to population trapping\cite{41,42,43} in the high-lying Rydberg states in molecules caused by intense two-color femtosecond laser pulses. Those trapped states could be ionized through collision with energetic electrons so as to form more ions. Also, the THz pulse could either further field ionize directly the trapped high-lying Rydberg states or heat the free electrons first which then impact-ionized these trapped states. The consequence was that the ion population was increased, which resulted in the enhancement of the fluorescence from the neutral N$_2$ molecules or molecular ions\cite{44}. The higher the THz field was, the stronger the fluorescence would be. Hence, the variation of the N$_2$ fluorescence follows the THz field variation. Since N$_2$ fluorescence in an air filament is in the UV and near UV (such as 337 and 391 nm)\cite{44}, sensitive detection using PMT or ICCD would enhance the THz radiation significantly and could be applied to remote sensing.

We recently performed an experiment to confirm the above mechanism of enhancement of THz detection by looking simultaneously at the THz emission and the fluorescence, for example, at 337 nm of N$_2$ during molecular alignment and revival inside a filament in air. The schematic setup is shown in Fig. 4. Collinear laser pulses at a repetition rate of 1 kHz from beam I and beam II with precisely controlled delay time were focused by a 50-cm focal length lens and then passed through 0.1-mm-thick type-I-barium borate (BBO) crystal to produce second harmonic pulses. Pulse energies from beam I and beam II measured after the lens were 0.38 and 0.5 mJ, respectively. Through two-color filamentation in air, THz pulse was characterized by the electro-optical sampling (EOS) technique\cite{31}. The THz signal from beam II was optimized by varying the BBO angle which was then fixed. The optimum angle $\alpha$ between the optic axis and the FW’s polarization direction was $\alpha \approx 55^\circ$\cite{44}. The linear polarizations and temporal schemes of the two pairs of two-color pulses after the BBO crystal are depicted as the inset of Fig. 4. The angle between the two polarizations of the FW and SHW was $\sim 35^\circ$. N$_2$ fluorescence at 337 nm was detected by a gated PMT covered by a 337-nm interference filter. In our experiments, the first pair of two-color laser pulses from beam I shown in the inset of Fig. 4 created filaments in air, resulting in the alignment and revival of air molecules. Since the SHW was around 2 orders of magnitude weaker than the FW, the FW played the dominant role in this molecular alignment event. The second pair of two-color laser pulses from beam II was used to generate THz pulses to probe/interact with the aligned and revived molecules. By scanning the relative delay time between the two pairs of pulses from beam I and beam II, THz electric field from the second pair of two-color pulses in the forward direction and the filament-induced 337-nm N$_2$ fluorescence from the side were measured (Fig. 4).

Figures 5(a) and (b) show the calculated response functions ($R(t)$) of the molecular rotational Raman effect (alignment and revival) in air created by the pulses from beam I as a function of delay time at half-revival of N$_2$ (Fig. 5(a)) and full revival of O$_2$ with three quarter revival of O$_2$ (Fig. 5(b)) given by the expression of $R(t) = n_{2,rot} \sum_{J=0}^{\infty} F_J(\sin(\omega_Jt)$, where $n_{2,rot} = 32\beta^2(\omega)^2 \pi^4N/hcn_0$ is the overall magnitude of the rotational Raman effect, $\omega_J = 4\pi Bc (2J + 3)$ is the angular frequency difference between the coupled rotational levels, $B$, $N$, $h$, $c$, $n_0$, and $\beta(\omega)$ are the rotational constant of the molecule, the density of the considered gas, Planck’s constant, the speed of light, linear refractive index at the laser wavelength, and the anisotropy of the molecular polarizability, respectively\cite{44}. $F_J = (\rho_J + 2 - \rho_J)Z_J(2J + 1)/(2J + 3)$ is a function of the transition moment between two coupled rotational levels ($J+2$ and $J$), of
the population difference ($\rho_{J+2} - \rho_J$) between these levels and of a weighting factor $Z_J$ describing the multiplicity of each level. When the rotational Raman response function, $R(t)$, is $>0$, the average molecular axis is parallel to the polarization of the exciting laser field and the refractive index change felt by another incoming laser pulse is positive. In our experiment, the exciting field is the 800-nm strong pulse in the first pair (I) of pulses in Fig. 4 and the so-called incoming pulse is the second pair (II) of pulses in Fig. 4. This would result in more focusing (guiding) of the second pair of pulses into the filament zone formed by the first pair of pulses. When $R(t)$ is $<0$, the average molecular axis is perpendicular to the polarization of the exciting laser field and the refractive index change of the incoming laser pulse is negative.

The 337-nm fluorescence is proportional to the total number of $N_2$ ions which includes the ions in the ground state, $N_2^+$, and in the excited states, $N_2^{++}$[49]. The experimental observations of 337-nm fluorescence are shown at the half-revival time of $N_2$ in Fig. 5(c) and the full revival time of $N_2$ with three quarter revival of $O_2$ in Fig. 5(d). When the $N_2$ molecules are aligned parallel to the polarizations of the FW of beam I, both the FW and the SHW of beam II would experience an induced refractive index change which is positive. This corresponds to guiding (focusing), resulting in stronger filamentation, hence more ionization. This would give rise to more 337-nm fluorescence. At the same time, stronger THz pulses would be generated through FWM[31]. These stronger THz pulses would ionize some of the trapped Rydberg states[46–48] converging to $N_2^+$ and $N_2^{++}$ resulting in even more ionization, hence, more 337-nm fluorescence. When the aligned molecules are perpendicular to the polarizations of the FW and SHW, the induced refractive index change is negative. This corresponds to de-focusing, resulting in weaker filamentation. Weaker THz pulses will be generated through FWM. Thus, there would be less ionization of the trapped Rydberg states converging to $N_2^+$, $N_2^{++}$ under weaker THz field, resulting in less 337-nm fluorescence. The consequence is that THz and fluorescence follow the alignment and revival; hence, follow each other. That is to say, when the THz field is increased in a filament zone, the induced extra $N_2$ fluorescence also increases proportionately. This allows sensitive remote measurement using know sensitive optical detectors such as photomultipliers or ICCD.

5. Conclusion

In summary, we briefly review THz generation from femtosecond laser induced plasma filament. Since the THz source from filaments can avoid the propagation issues of diffraction and absorption, emphasis is put on the remote generation and detection of this high energy, broadband THz pulses. Towards remote high energy THz generation, THz pulses from two-color laser filamentation at a long distance are discussed; towards THz remote sensing, the technique of THz-REEF is reviewed. We also present new results on the direct relationship between THz generation in a two color filament and induce $N_2$ fluorescence through population trapping during molecular alignment and revival in air. This further supports the remote sensing technique of THz-REEF. We believe that by combining the techniques of the remote high energy broadband THz generation and the remote THz detection, broadband standoff THz spectroscopy is feasible.

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