Spectral control of terahertz radiation from inhomogeneous plasma filaments by tailoring two-color laser beams

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Abstract: Terahertz (THz) radiation from an inhomogeneous plasma filament generated by focusing two-color femtosecond laser pulses into argon gas filled in a chamber is investigated experimentally by tailoring the Gaussian pump laser beams with an iris, where broadband THz emission over 10 THz is produced. It is found that the collected far-field THz radiation includes not only coherent but also partial-coherent components of the THz waves, which are emitted from the different parts of the inhomogeneous plasma filament with different plasma densities, contributing correspondingly to the different frequencies of the THz spectrum. Our results suggest that the THz spectrum can be manipulated by controlling the plasma density distribution of the filaments.

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1. Introduction

Terahertz (THz) radiation from the plasma filament generated by focusing a fundamental ($\omega_0$) and its second harmonic ($2\omega_0$) femtosecond laser pulses into the ambient air was demonstrated first by Cook et al. [1]. This technique has drawn significant attention [2–10] due to its high peak electric field and broadband spectrum, which can find wide applications such as THz nonlinear spectroscopy, THz high-field physics, and so on. Many models [1–7] were proposed successively to explain the underlying THz generation mechanism in this two-color laser mixing scheme.

In the meanwhile, some approaches to control or optimize the THz radiation from two-color laser induced plasma filaments have been proposed. For example, a few efforts have been made by changing the parameters of the pump lasers, including the wavelength [8], duration [9], polarization [10–12], and energy [6,13] and by changing the species and pressure [14,15] of the gas interacting with the laser. Other ideas have been explored by adjusting the ratio [16] or the relative phase [17] between the fundamental laser pulse and its second harmonic, including the frequency doubling beta-barium borate (BBO) crystal rotation angle and BBO-to-focus distance, and by using different pump laser beam focusing condition, including using focus lens with different focal length [13], by controlling the pump laser beam wavefront by tilting the focus lens [18]. It is found that THz radiation can be controlled by tailoring the Gaussian pump laser beams with iris [19], a hybrid approach that changes both the energy and the transversal intensity distribution of the Gaussian pump laser beams. Among above mentioned approaches, the plasma filament length or structure connected intrinsically with the THz radiation are significantly affected by changing the pump laser energy [6,13] or the beam focusing conditions [18]. In these studies, however, the contributions of different parts along the filament string to the entire THz...
spectrum are still not clear. In Ref. [19], it is noted that THz output can be manipulated and optimized by tailoring the Gaussian pump laser beams with iris, however, investigation on how the beam tailoring affects the plasma filament length or structure and subsequently the entire THz radiation spectrum (0.1-10 THz) have not been performed. Such investigation is helpful to understand fully the THz generation process and the underlying mechanism of the THz radiation.

In this study, we examine the entire THz spectrum by controlling the spatiotemporal structure of the plasma filament simply with an iris to tailor the two-color Gaussian pump femtosecond-laser beams. We investigate how different parts of the inhomogeneous plasma filament contribute to the entire THz spectrum.

2. Experiment and results

The schematic experimental setup used for THz generation and detection is shown in Fig. 1. Our Ti: sapphire laser system delivers laser pulses at the central wavelength of 800 nm with energy of ∼8.7 mJ in 50 fs duration at a repetition rate of 1 kHz. The laser beam is split into a pump beam and a probe beam. The pump beam is focused by a convex lens with a focal length of 40 cm placed before the incident quartz window for generation of elongated plasma filaments. A 100-µm thick BBO crystal (Type I) is placed after the incident quartz window in a stainless chamber to produce the second harmonic (SH) pulses. An iris is placed before the focal lens to tailor the Gaussian pump laser beams. A schematic Gaussian pump laser beam and its tailored beam by an iris with its aperture diameter of \( D \) are shown at the inset in the top left corner of Fig. 1. The phase delay \( \Phi \) between the fundamental (\( \omega_0 \)) and the SH (2\( \omega_0 \)) pulses is controlled by moving the BBO crystal along the laser direction. Both the fundamental and SH pulses are focused into the ambient argon (Ar) gas filled in the chamber. This chamber is capable of filling different gas under different gas pressure, especially designed to mount the focal lens, the BBO crystal, and the off-axis parabolic (OAP1) mirror for generation, collection, and collimation of the THz emission. Before the experiment, air is pumped out from the chamber then the argon is injected into it as the pressure reaches up to 1 atm. The output THz beam is collimated and refocused using other three OAP mirrors outside the chamber. A silicon window with high resistance is used as a filter to pass THz radiation and filter out unwanted remnant \( \omega_0 \), 2\( \omega_0 \) and supercontinuum light from the plasma filament. The probe beam is focused with a convex lens through a hole in the back of the OAP4 mirror.

Considering the THz spectrum generated from the plasma filament would cover wide THz range even over tens THz, a super broadband detection technique, air-biased-coherent-detection (ABCD) technique [20] is used to measure the THz waveform, as shown in the area of the green rectangle at the top right of Fig. 1. This ABCD system is filled with nitrogen (N\(_2\)) gas to avoid the vapor absorption in all measurements. By optimizing the rotation angle of the BBO crystal and its position relative to the focus (i.e. the phase delay \( \Phi \) between the \( \omega_0 \) beam and the 2\( \omega_0 \) beam), an intense, highly directional, broadband THz pulse is generated. We reduce continually the aperture diameter \( D \) of the iris with an interval of 1 mm and measure the corresponding THz radiation with the ABCD system at different \( D \) values. A camera close to a lateral optical window is used to record the fluorescence images of the plasma filaments at different \( D \) as shown in the right inset of Fig. 1.

In the experiment the Gaussian pump laser beam has a diameter of 16 mm (86.5% of the total laser energy) and energy of 8.7 mJ before the iris. The largest diameter of the iris \( D \) is 19 mm that can be considered as the iris-free case, i.e. no limitation to the pump laser beams. The THz waveforms are measured at different \( D \) adjustable from 19 mm to 9 mm to tailor the pump laser beams. Figures 2(a) and 2(b) show respectively the typical measurements of THz pulses and their corresponding Fourier transformed THz spectra as a function of the iris diameter \( D \). The focused laser energy through the iris decreases nonlinearly with the decrease of \( D \) as shown in the inset of Fig. 2(a). In the case of \( D = 19 \) mm (i.e., the iris-free case), it is found that the strongest THz
radiation is located at ~ 0.8 THz and the THz spectrum covers the range from 0.1 to over 12.5 THz. Due to the tailored intensity distribution of the laser beams, we observe that the nonlinear variation of the THz waveforms and its frequency spectrum. For the sake of clarity, we only show the typical THz waveforms and their spectra in $D = 19, 17, 14, 11$ mm in Fig. 2. Figure 2(a) shows that the THz waveform nearly does not change when $D$ decreases from 19 to 14 mm, only the second half of the waveform is weakly reduced in strength. The corresponding THz spectrum with frequencies over $\sim 2$ THz nearly does not change when $D$ decreases from 19 to 14 mm, as shown in Fig. 2(b). When $D$ is reduced to 11 mm, the pump laser energy decreases from 8.6 mJ ($D = 19$ mm) to 6.5 mJ. In this case, not only is the strength of THz signal weakened but also the second half of the waveform is shortened evidently. In the corresponding spectrum, the low frequency part below $\sim 6$ THz especially the part around $\sim 0.8$ THz decreases significantly, while the high frequency part over 6 THz remains nearly the same with the large $D$ values as shown in Fig. 2(b). This phenomenon is totally different from the iris-free case, where the strength of all frequencies of the whole THz spectrum decreases when the Gaussian pump laser energy is reduced (a separate experiment not shown here). These results indicate that THz radiation from two-color laser plasma filaments can be controlled by tailoring the pump laser beams with an iris.

Except for the four-wave mixing (FWM) model [1], nearly all current models such as the transient photocurrent (PC) model [3], transient ionization current (IC) model [4], and linear-dipole array (LDA) model [6] demonstrate that the THz radiation in the two-color laser scheme is associated with the plasma density of the filaments. To interpret above phenomena, we investigate the evolution of the spatial structure of the plasma filaments. Considering the plasma density is relative to the fluorescence and the higher the plasma density, the stronger fluorescence intensity [21], we take pictures of the lateral fluorescence from the plasma filaments in different $D$ in order to obtain the longitudinal plasma distribution. Typical fluorescence images of plasma filaments obtained with iris diameters of $D = 19, 17, 14,$ and $11$ mm in ambient argon are shown in Fig. 3. These images reflect the evolution of the argon plasma filaments controlled by the iris. The right insets in Fig. 3 show the corresponding lateral fluorescence intensity distributions along the
Fig. 2. The nonlinear variation of THz signal in argon at 1 atm with different iris aperture diameter $D$. Typical THz temporal waveforms in (a) and their Fourier transformed spectra in (b) as well as the energy of the tailored pump laser pulses in different $D$ in the inset of (a).

plasma filament under different $D$. The “0” positions shown in the right insets correspond to the blue dashed line.

Fig. 3. Typical fluorescence images of plasma filament distributions with iris diameters $D = 19$, 17, 14, and 11 mm in ambient argon. These images reflect the evolution of the argon plasma filament by tailoring the two color pump laser beams by the iris. The right panel shows the corresponding lateral fluorescence intensity distributions along the plasma filament in different $D$.

From Fig. 3, one can observe that in the case of $D = 19$ mm, i.e., the iris free case, an elongated two spindle-like filament string with a length of $\sim 28$ mm is generated, similar to the phenomena observed in previous studies [22–24], showing an inhomogeneous plasma distribution with two maximum fluorescence intensities along the plasma filament and weaker fluorescence in the leading edge and the tail of the filament. The exact two maximum positions can be observed more clearly in the right panel of Fig. 3. These two positions correspond to the two maximum
plasma densities. According to the LDA model [6], all parts of the filament string contribute to the collected far-field THz radiation shown in Fig. 2. However, one does not know which part of the filament contributes to which frequency components of the whole THz spectrum. Therefore, we reduce the aperture $D$ of the iris from 19 mm to 14 mm and one can see that the second spindle-like filament fragment become smaller and shorter, which is consistent with the decrease of the THz pulse length correspondingly as shown in Fig. 2(a). When examining the filament structure carefully, one may find that the first maximum fluorescence position (i.e. the maximum plasma density position) is moving a little when $D$ is changed. If we go back to Fig. 2, only the THz radiation with frequencies less than $\sim 2$ THz decrease, those radiations over 2 THz does not change. This means that the disappeared tail with a lower plasma density only contributes to the THz radiation with frequencies less than $\sim 2$ THz. If $D$ is reduced continually to 11 mm, not only the second spindle-like filament part disappears but also the first one become slimmer and shorter. The quantitative change of the fluorescence of the filament can be observed clearly from the right insets in Fig. 3. If we check the THz spectra of Fig. 2 once again, when $D = 11$ mm, only the THz radiation with its frequencies less than $\sim 6$ THz decrease, while the others over $\sim 6$ THz nearly does not change. This indicates that the tailored part, i.e., the second spindle-like filament fragment (disappeared part) does not contribute to the THz radiation with frequencies over 6 THz, but contribute to that of less than 6 THz, especially mainly to the strongest radiation at $\sim 0.8$ THz and its vicinities.

To understand the evolution of the spatial structure of the filament under different $D$, we have performed another experiment to check the transverse distribution of the plasma filament. For the sake of simplicity and considering that the maximum plasma density position moves slightly if $D$ is changed, we insert a quartz sheet at the first maximum fluorescence position of the filament in the typical cases of $D = 19$ mm (i.e., the iris-free case), 14 mm, 13 mm, and 12 mm as shown at the dashed red line (shown in the left panel of Fig. 3). We record the ablation patterns damaged by plasma filaments and residual laser beams when $D$ is reduced. Considering the laser jitter effect, we measured firstly one-shot ablation patterns and found that their boundaries are too vague to be observed clearly. As a compromise on clear boundaries and laser beam jitters, we measured laser ablation patterns with two laser shots. The typical ablation patterns (two shots accumulated for each figure) on the quartz sheet are shown in Figs. 4(a), 4(b), 4(c), and 4(d) for $D = 19$, 14, 13, and 12 mm, respectively. The corresponding real focusing laser energies after the iris are 5.4, 5.4, 5.1 and 4.8 mJ, respectively. One can see that the ablation area becomes smaller as $D$ is decreased, indicating that the transverse size of the plasma filament is decreased with the decrease of $D$, in accord with the phenomenon that the plasma filament becomes slimmer if $D$ is reduced as we observed in Fig. 3. To check the tailoring effect of the pump laser beams to the boundary of the plasma filament, we compare the iris limited cases in Figs. 4(b), 4(c), and 4(d) with the iris-free case in Fig. 4(a), and cannot observe evident difference of the ablation boundaries between in the iris limited cases and the iris-free case. This indicates that the tailored pump laser beams do not induce a much sharper plasma boundary compared with the full Gaussian pump laser beams in iris-free case.

In the cases of $D = 17$ mm and 14 mm, only the peripheral rings of the Gaussian pump laser beams are “tailored”. In these cases, due to that the “tailored energy” of the pump laser beams is too small to affect the transverse size and the length of the plasma filament evidently, two spindle-like filament string (i.e. the focusing-refocusing phenomenon [22–24]) can still be observed, indicating that the remnant main energy of the pump laser beams is still enough to support the refocusing process. If $D$ is decreased more, for example $D = 11$ mm, 12 mm, too much energy of the peripheral pump laser beams is “tailored”, resulting in insufficient energy of the remnant pump laser beams to support to refocusing. Therefore, the second sprinkle-like filament disappeared. However, with the change of $D$ from 19 mm (iris-free case) to 12 mm, as shown in Fig. 4, the transverse size of the main plasma filament does not change too much compared with
Fig. 4. Typical ablation patterns (two shots accumulated) on a quartz sheet damaged by plasma filaments and residual laser beams in the iris free-case ($D = 19$ mm) in (a) and in the cases of different typical iris aperture diameters $D = 14$, $13$, $12$ mm in (b), (c), and (d), respectively.

the longitudinal change of the plasma filament. In other words, the change of the THz radiation could be attributed mainly to the evolution of the longitudinal plasma distribution along the filament when the pump laser beams are tailored with the iris when $D$ is larger than $11$ mm.

To investigate how the different parts of the entire plasma filament contribute to the THz radiation in more detail, we calculate the THz fields contributed by different laser components due to the introduction of the iris, i.e., the laser energy passing through the iris aperture and blocked by the iris. These two laser components contribute to different part of plasma filaments, which are named as the remanent filament and removed filament in the following. According to the superposition principle of fields, the collected far-field THz signal $E_{\text{THz}}$ of the entire plasma filament is the superposition of THz waveforms from all parts along the filament with different plasma densities. If pump laser beams are tailored by the iris with different $D$, the total THz field can be described with $E_{\text{THz}} = \Sigma E_i = E_R + E_T$, in which $E_R$ is the THz field of the remanent filament and $E_T$ is that of the removed filament when the pump laser beams are tailored. Thus we obtain the THz fields $E_T$ of removed parts of the filament at different $D$ ($=17$ mm, $14$ mm, $11$ mm) and their corresponding Fourier transformed THz spectra as shown in Figs. 5(a)–5(f), in which the THz field and spectrum of the entire filament in the iris-free case ($D = 19$ mm) are also shown for the comparison. One can see clearly from Figs. 5(a), 5(c), and 5(e) that the THz waveforms from removed parts of the filaments at different $D$ have different pulse lengths and show different lagging phases compared with that from the remanent filaments: $\Delta t = t_T - t_R = 0.040$ ps, $0.053$ ps, $0.027$ ps when $D = 17$ mm, $14$ mm, $11$ mm, respectively. Figures 5(b), 5(d), and 5(f) show that removed parts of the filaments at different $D$ have distinct different frequency spectrum due to that the different parts of the filament hold distinct different plasma densities. In Fig. 5(f), it is clearer than in Fig. 2(b) and validates that the removed part, i.e., the second spindle-like filament...
fragment in the iris-free case only contribute to the THz radiation with frequencies less than 6 THz.

Fig. 5. Typical THz waveforms in (a), (c), (e) and their spectra in (b), (d), (f) from the remanent filaments and their corresponding removed parts of plasma filaments vs. that from the entire plasma filaments \( (D = 19 \text{ mm}) \) at three typical iris aperture diameters \( D = 17, 14, 11 \text{ mm} \), respectively.

3. Discussion

The plasma filament formation is a very complicated process, depending on the focusing conditions such as the focal length of focusing lens, the laser beam size, the laser pulse energy, as well as the density of gas and so on. The eventually formed filament structure depends on the dynamic balance between some nonlinear effects [25] such as diffraction, Kerr self-focusing, plasma nonlinearity. In our case, due to multiple beam refocusing and longitudinal filament instability, an elongated two spindle-like filament string is generated. By tailoring the Gaussian pump laser beams with an iris, a hybrid approach that changes both the energy and the transversal intensity distribution of the Gaussian pump laser beams, the second spindle-like filament fragment
become weaker and even disappear with the decrease of the iris aperture $D$. This suggests that the peripheral parts of the Gaussian laser beams play a main role in the generation of the second filament density peak along the laser propagation direction. This density peak has lower plasma densities and as a result contributes to the lower frequency components of the THz radiation. According to the LDA model [6], for an elongated inhomogeneous plasma filament, adjacent dipole arrays with the same plasma densities emit the same THz waves and then superpose coherently in the far-field. Furthermore, Fig. 5 shows different phases of the THz waveforms and pulse lengths as well as the different frequency ranges are mainly due to the different parts of the inhomogeneous plasma filament. These results not only validate the LDA model but also suggest that, the far-field collected THz wave include not only coherent but also partial coherent components from different parts with different plasma densities.

In our experiment, the strongest THz radiation locates at $\sim$0.8 THz, corresponding to the plasma density of $n_e = 7.95 \times 10^{15}$ cm$^{-3}$ according to the relationship between the plasma density $n_e$ and the frequency $f_{pe} (\approx f_{THz})$ that reads $\omega_{pe} = 2\pi f_{pe} = (e^2 n_e / m_e \epsilon_0)^{1/2}$. Here $e$ denotes the electric charge, $m_e$ the effective mass of the electron, and $\epsilon_0$ the permittivity of free space. The plasma densities correspond to the two positions with maximum luminescence along the filament shown in Fig. 3 can be estimated from Fig. 5(f). The highest frequency of the THz radiation is $\sim$12.5 THz, locating at the first maximum luminescence position of the first spindle-like filament fragment and corresponding to the plasma density of $n_e = 1.94 \times 10^{18}$ cm$^{-3}$. The second highest frequency of the THz radiation locating at the maximum luminescence position of the second spindle-like filament fragment is less than 6.0 THz, corresponding to the plasma density of $n_e = 4.47 \times 10^{17}$ cm$^{-3}$. The fact that strongest THz radiation locates at $\sim$0.8 THz indicates that the most plasma distribution along the entire filament especially the second spindle-like filament fragment fall in the plasma density $n_{em}$ of around $7.95 \times 10^{15}$ cm$^{-3}$ and its vicinity. Each local THz emitter (dipole) with this same plasma density and similar densities along the filament superpose coherently in the far field and contribute to the strongest THz radiation, whereas others THz emitters with dispersive plasma densities far from $n_{em}$ superpose coherently in part in the far field and contribute to the rest frequencies of the THz radiation. Therefore, THz radiation including the output and spectrum can be controlled by “designing” a plasma filament with suitable plasma density distribution.

4. Conclusions

In conclusion, we have demonstrated experimentally a simple method to control the plasma filament distribution and subsequently the THz radiation spectrum by tailoring the Gaussian pump laser beams with an iris. The THz radiation is produced from two-color femtosecond laser pulses induced elongated inhomogeneous argon plasma filament in broad spectrum over 10 THz. It is found that the collected far-field THz radiation includes not only the coherent superposition but also some partial coherent superposition of the THz waves emitted from the dipole array along the filament. Different parts of the filament with different plasma densities contribute correspondingly to different frequencies of the THz radiation. Our results not only validate the LDA model but also show that it is feasible to manipulate the frequency spectrum of THz radiation by our simple method. This provides an effective way to manipulate the THz spectrum for applications.

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