

Enhancement of Terahertz Radiation from a Transversely Asymmetric Femtosecond Laser Filament

Jiayu Zhao,* Jiajun Yang, Xiaofeng Li, Feifan Zhu, Li Lao, Yan Peng,* and Yiming Zhu*

Cite This: *ACS Photonics* 2025, 12, 2034–2043

Read Online

ACCESS |



Metrics & More



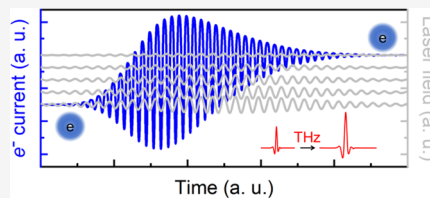
Article Recommendations



Supporting Information

ABSTRACT: Plasma filaments via femtosecond laser ionization in air have been extensively studied as a significant terahertz (THz) source, based on which one particular objective is to enhance the efficiency and intensity of THz radiation. To this end, various strategies have been explored, including modulating the pump laser through temporal asymmetry of the carrier or envelope, or by tailoring the spectral amplitude profile asymmetrically. Apart from the above “asymmetric” operations in time and frequency domains, here we proposed a straightforward and practical method based on the spatial asymmetry. Specifically, an opaque blade was employed to partially obstruct the cross-section of the pump laser beam, resulting in a notable THz power enhancement of up to 60%. To interpret this improvement, we introduced a spatially asymmetric photocurrent mechanism: the created steep gradient of the inhomogeneous laser field enhances the electron drift motion, which in turn generates a stronger transverse current, leading to the observed increase in THz signal strength. To summarize, the proposed experimental method is highly accessible without requiring additional modulation devices, significantly lowering the application threshold. And its mechanism is also versatile, not only enhancing THz transverse wave radiation in both single- and dual-color field configurations, but also potentially applying to other setups, such as tilting focusing lenses or frequency-doubling crystals, warranting a reevaluation of previous studies. Additionally, our approach optimizes energy usage, enabling stronger THz radiation under low-power laser pump conditions and further allowing the blocked laser energy to be redirected for enhanced functionalities.

KEYWORDS: terahertz enhancement, transverse asymmetry, laser field, plasma filament, electron drift and current



INTRODUCTION

Terahertz (THz) waves are gaining increasing attention across a wide range of fields, including spectroscopy, imaging, materials science, biomedicine, and chemistry.¹ The generation of strong and broadband THz radiation, capable of spanning the THz gap from 0.1 to several tens of THz, remains a focal area of research within the THz science and technology community.^{2,3} Enabled by ultrashort laser pulses, several intense and broadband THz sources have been developed, such as photoconductive antennas,⁴ optical rectification in nonlinear crystals,⁵ and laser-induced plasma.^{6–8} Among these, the focus of this work is the THz source of laser plasma filament, which is created at the focus of femtosecond laser pulses in air and is capable of emitting broadband THz waves.^{9,10} Since the pioneering work by Hamster et al.,¹¹ significant efforts have been made in enhancing the efficiency and output of THz radiation.

For example, the THz radiation generated by the interaction of two-color femtosecond laser pulses with gas plasma has attracted wide attention.^{12–15} Compared to the single-color scheme,¹⁶ the combination of fundamental laser (ω) and its second harmonic (2ω) built an asymmetric electric field across individual optical cycles, which can generate a residual current and stronger THz output. One can also use multiple carrier harmonics to shape an asymmetric sawtooth profile of the laser

field for THz enhancement.¹⁷ Additionally, THz radiation can be enhanced by utilizing a pumping laser pulse with a temporally asymmetric envelope.¹⁸ In this case, the laser pulse is specially shaped with slow turn-on but rapid turn-off in the temporal domain, which can accelerate electrons at higher velocities, resulting in stronger THz radiation than the normal two-color laser pulses with the same power. Moreover, the frequency-domain asymmetry can also be employed to achieve modulation of THz emissions.¹⁹ By filtering out the noncentral frequency components from the pumping laser spectrum, tunable THz power yield with broader bandwidth is generated.

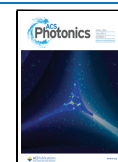
The aforementioned techniques have significantly optimized THz radiation through asymmetrically temporal field carriers, pulse envelopes and spectral manipulation, which raises the question of whether asymmetrically spatial modulation is also feasible. In this regard, actually, previous studies have reported that symmetrically spatial modulation can enhance the THz radiation. For example, Peng et al.²⁰ utilized an aperture to

Received: December 10, 2024

Revised: March 25, 2025

Accepted: March 27, 2025

Published: April 1, 2025



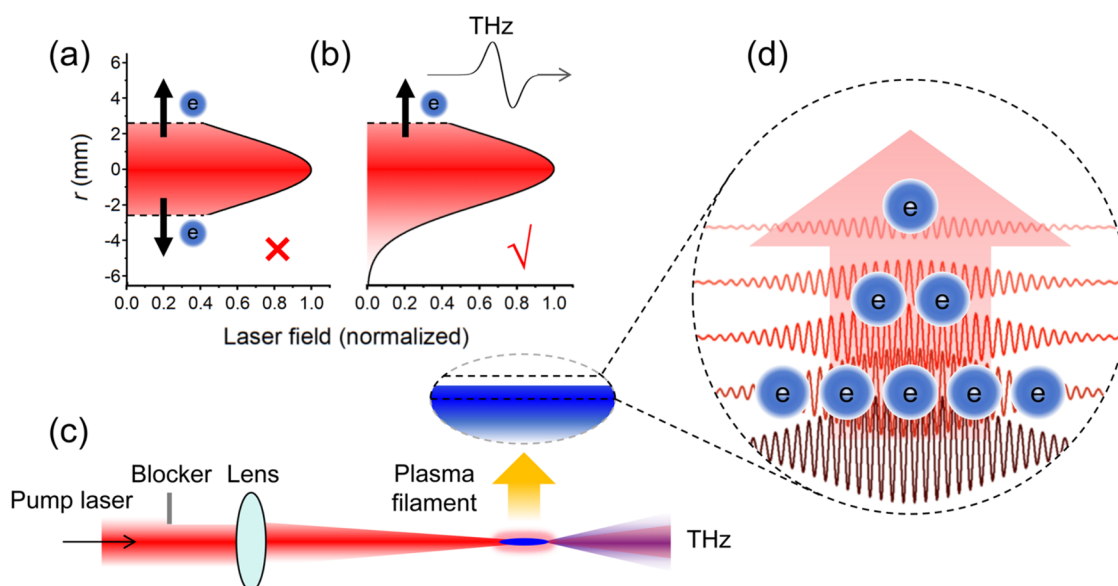


Figure 1. Transverse distribution of the laser field under (a) symmetric and (b) asymmetric blocking conditions. The former has become ineffective for THz enhancement under low pump power conditions, whereas the latter may offer a viable solution. (c) The proposed experimental scheme for enhanced THz wave generation via asymmetric blocking of the laser spot, followed by its filamentation and THz generation. (d) The underlying mechanism for THz enhancement: under the influence of an asymmetric laser electric field, electrons experience an intensified transverse drift motion, which facilitates the THz generation.

reduce the diameter of a pump laser beam with an energy of 14 mJ/pulse, and observed a trend of initial increase followed by a decrease in the THz signal. This phenomenon can be roughly understood by the following fact. When the pumping laser intensity is high,^{20,21} a dense plasma is formed and strongly defocuses the laser pulse from coupling into the plasma, which leads to intensity clamping inside the filament²² and saturation of THz output. While the symmetrically spatial modulation on the laser beam could control the laser intensity and keep the plasma density within the range suitable for THz radiation, thereby enhancing the THz yield.²⁰ However, this approach was found to be less effective under low-power pumping laser conditions (below 8 mJ/pulse) as reported by the similar research group.²³ This time, reducing the laser beam diameter could only result in a decrease of the THz output. This can be interpreted as follows: under low-power laser pump conditions, there is no THz saturation effect, and thus the original method fails to further enhance the THz generation (as shown in Figure 1a).

In addition to the above method of breaking the THz output saturation to enhance its radiation, another approach involves increasing the transverse displacement of plasma electrons within the laser filament, such as by applying a DC bias.^{24,25} Recently, transversely asymmetric modulation methods have been proposed, gaining increasing attention.^{26,27} These techniques utilize shockwaves generated by a prepulse to drive the transverse diffusion of electrons during the main pulse filamentation. Furthermore, a recent theoretical work by Jia et al.²⁸ suggests that creating near-field localization of the laser field along a nanoscale conical structure can produce a highly asymmetric transverse electric field, which in turn can also enhance THz generation. However, all of these modulation methods—whether based on an applied DC field, an additional filament's shockwave, or near-field nanostructures—are external modifications to the existing filament in the experimental setup. As a result, these

approaches may not offer significant advantages for remote enhancement of THz radiation at a distance from the filament.

Inspired by the above works, we realized that a simple method of spatially asymmetric modulation is undoubtedly partial beam blocking (Figure 1b). Hence, in this work, we focused on exploring whether THz enhancement could be achieved using this method (Figure 1c), particularly with a low-power pumping laser beam (~ 1.8 mJ/pulse), where there is no THz output saturation. Specifically, we proposed that the partial beam blocking could introduce a stepwise decay of the laser electric field, and along this sharp gradient, the transverse drift of electrons will be intensified, leading to an enhanced THz radiation (Figure 1d). To investigate this, we theoretically suggested a spatially asymmetric photocurrent (SAPC) model considering the transverse gradient effect of the asymmetric laser field distribution in order to account for the possible THz enhancement. Then, regarding the experimental verifications, we initially followed similar procedures described in the literature,²³ and blocked the laser beam spot from symmetric two/four sides simultaneously. As expected, there was no considerable enhancement of the THz signal, which confirms that there was indeed no output saturation. Subsequently, we asymmetrically blocked the beam spot and observed a first increase and then decrease of the THz power. Based on the above experimental phenomena under asymmetrically spatial modulation, a simple method of THz enhancement has been demonstrated.

The advantages of our experimental method and its SAPC mechanistic explanation lie in their broad applicability, which can be summarized as follows. (i) Simplicity of experimental technique: Our method for generating a laterally asymmetric laser electric field by blocking part of the laser spot is remarkably straightforward. Unlike previously reported approaches, this “self-simplification” experimental design eliminates the need for additional modulation devices on the laser filament, thus significantly lowering the application threshold and enhancing accessibility. (ii) Versatility of the mechanism:

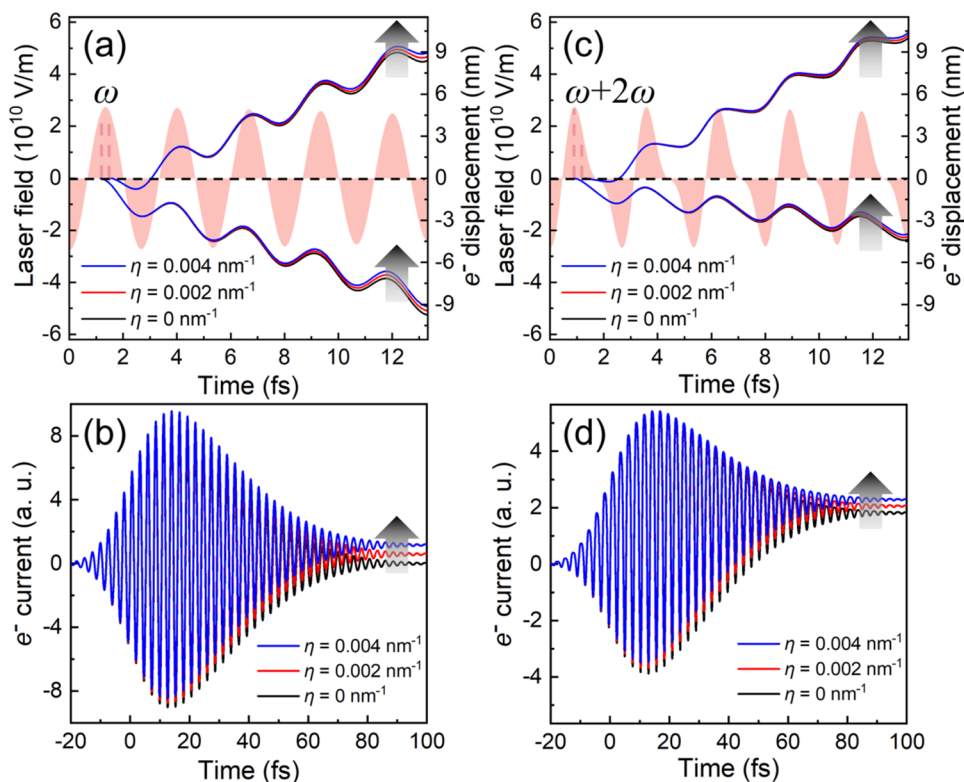


Figure 2. (a) The trajectories of electrons born at two different phases with respect to the single-color laser field (light red) are calculated with our model for $\eta = 0$ (black line) and $\eta \neq 0$ (red and blue lines). (b) The corresponding drift currents. (c, d) Similar calculations for the two-color case.

The mechanism we propose for transversely asymmetric laser electric fields is highly adaptable. It not only enhances THz transverse wave radiation in both single- and dual-color laser pumping schemes, as demonstrated in our experiments, but can also be extended to other experimental configurations. For instance, the THz enhancement can also be achieved by tilting the focusing lens or frequency-doubling crystals, as reported in refs.^{29–32} Our findings suggest that the physical mechanisms underlying these studies, which do not account for asymmetric laser fields, may need to be revisited in light of our theoretical insights. (iii) Improved energy utilization: Our approach effectively harnesses laser energy to generate stronger THz radiation under low-power laser pump conditions—a notably more challenging scenario compared to high-power setups. Moreover, the blocked or reflected laser energy can be redirected into the experimental setup, enabling energy conservation and additional functionalities beyond the THz output enhancement (see Discussion).

THEORY

To account for both the on-axis temporal laser electric field and the off-axis transverse field inhomogeneity (gradient variations), we established the PC model¹² in two dimensions, encompassing both longitudinal and transverse components. This allowed us to compute the evolution of electron trajectories (r) and velocities (v) over time (t) within a nonuniform field ($E(r, t)$). The calculation formulas are as follows:

$$e \cdot E(r, t) = m_e \cdot a \quad (1)$$

where e , m_e and a represent the electric charge, the effective mass and the accelerated velocity of an electron, respectively. Here

$$E(r, t) = (1 - \eta)E_0(t) \quad (2)$$

where r is the radial distance and η indicates the inhomogeneous coefficient of the laser field ($\eta = 0$ if the laser spot is not blocked). Then, the evolution of r and v with different η as a function of t can be obtained by solving the following two equations simultaneously:

$$v(t) = \int_0^t a(t') dt' = \int_0^t (e/m_e)(1 - \eta)E_0(t') dt' \quad (3)$$

$$r(t) = \int_0^t v(t') dt' \quad (4)$$

Initially, v and r are set as 0. Other calculations, including those for ionization-induced electron generation and the formation of drift currents, utilize formulas similar to those in refs 10,12 and can also be found in Section A of the Supporting Information. Briefly, the constructed asymmetric laser field above could strip off bound electrons and produce a nonvanishing transverse plasma current, which is computed with the plasma density and electron velocity under the laser field. This photocurrent surge, occurring on the time scale of the photoionization, could further produce an electromagnetic pulse at THz frequencies. And the final THz radiation is determined by the time derivative of the drift current. Additionally, the laser electric field $E_0(t)$ is considered in two cases: $E_0(t) = E_1 \cos(\omega t)$ for the single-color laser pump, and $E_0(t) = E_1 \cos(\omega t) + E_2 \cos(2\omega t + \theta)$ for the two-color condition, where E_1 and E_2 are the amplitudes of the fundamental laser centered at 800 nm (ω) and its second

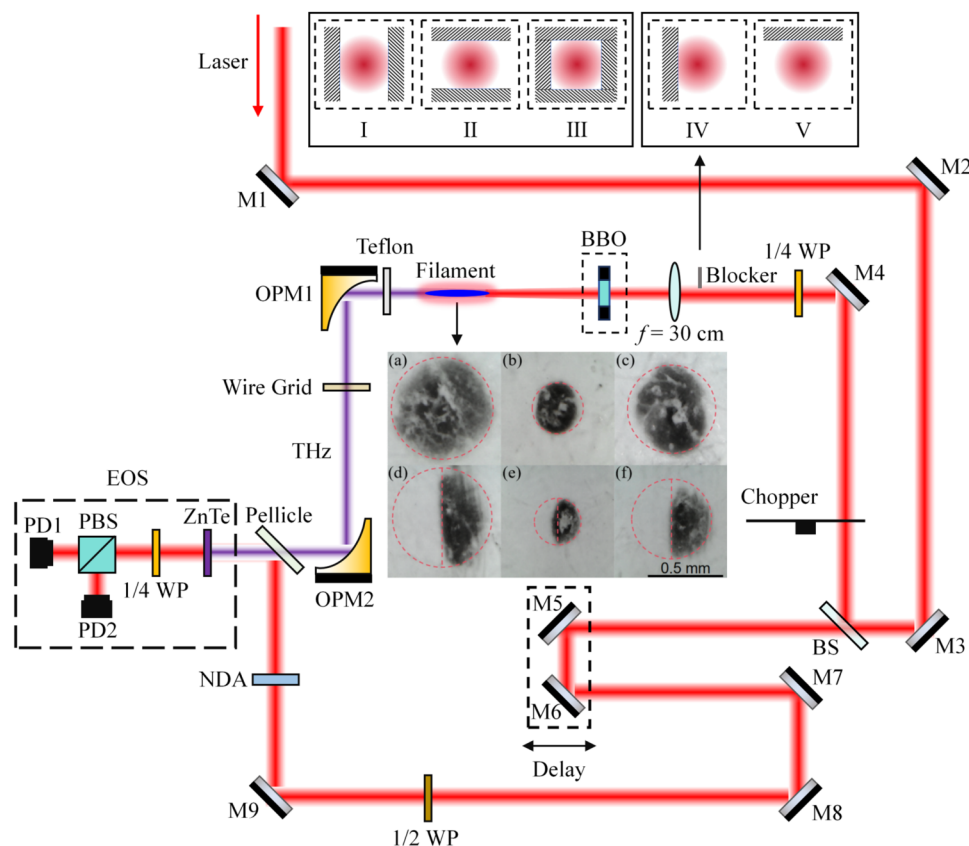


Figure 3. Experimental setup. BS: beam splitter. WP: wave plate. BBO (optional): frequency-doubling crystal. OPM: off-axis parabolic mirror. NDA: neutral density attenuator. EOS: electro-optic sampling system. PBS: polarization beam splitter. PD: photodetector. Insets: the blocking methods on the laser spot (I–V), and the laser ablation patterns on the thermosensitive paper along the filament without laser beam blocking (a–c) or half of the beam was blocked (d–f). (Note: due to thermal effects, the patterns on the thermosensitive paper are larger than the actual cross-section of the filament.).

harmonic (2ω) with the ratio of about 1:0.45, and θ is the ω – 2ω relative phase difference set as $\pi/2$.

For the former case, we calculated the drift trajectories of two electrons born at the middle stage (after $t = 0$ fs) of the laser pulse (-100 fs $< t < 100$ fs), which has a duration of about 50 fs as shown in Figure 2a for five optical cycles (in light red color). Specifically, the two electrons are liberated on opposite slopes of the laser field with phases of $-\pi/10$ and $\pi/10$, respectively. And the calculation results are shown as black lines in Figure 2a for initial conditions without the laser beam blocking ($\eta = 0$), which can be seen that, the drift effects of the two electrons nearly cancel each other. Moreover, the computed electron current as shown as the black line in Figure 2b also exhibits a zero residual value after the laser pulse at $t = 100$ fs. These observed electron drift and current results align with our understanding of the little transverse THz wave radiation under monochromatic field excitation. However, when $\eta \neq 0$, which corresponds to the transversely asymmetric experimental conditions in our study, the lateral drift symmetry of the electrons can be broken, as shown by the red and blue lines along the arrows in Figure 2a. As a result, the corresponding electron currents have a nonzero residual value as shown in Figure 2b, which could induce the THz radiation and its further amplification.

For the latter case, similar calculations were carried out, and the results are shown in Figure 2c–d. Specifically in Figure 2c, even if $\eta = 0$, the drift trajectories of the two electrons (black lines) exhibit lateral asymmetry. This can be easily understood

that the two-color field can essentially produce stronger THz radiation compared to the single-color case (black lines in Figure 2a), due to the temporal asymmetry of the two-color laser field. However, when $\eta \neq 0$, which corresponds to the case proposed by us with the presence of a transversely asymmetric laser field, the asymmetry of the electrons' trajectories is further enhanced as indicated by the red and blue lines. Thus, our asymmetric blocking method could further enhance the THz radiation beyond what is achieved with the two-color laser field alone (Figure 2d).

In summary, as illustrated in Figure 2, our model has been built upon a framework similar to the classic PC model, but with the addition of a key factor η , which accounts for the transversely asymmetric laser field. When $\eta = 0$, the calculation results (black lines in Figure 2) are fully consistent with previous reports. However, when $\eta \neq 0$, meeting our experimental conditions, the calculated results (red and blue lines) can preliminarily infer that electrons are driven to drift along the field gradient direction, and this enhances their transverse displacements, which in turn increases the residual current and THz radiation. Based on the theoretical analysis above, particularly the residual current (drift tail current) shown in Figure 2d, which is proportional to the electric field amplitude of the THz radiation, we anticipate that the THz power output can be enhanced by approximately 57% when using a bichromatic field with $\eta = 0.004$ nm $^{-1}$, corresponding to a lateral electric field gradient reduction range of $1/\eta = 250$ nm. Here, $1/\eta$ is set as 250 nm in order to ensure $0 < 1 - \eta <$

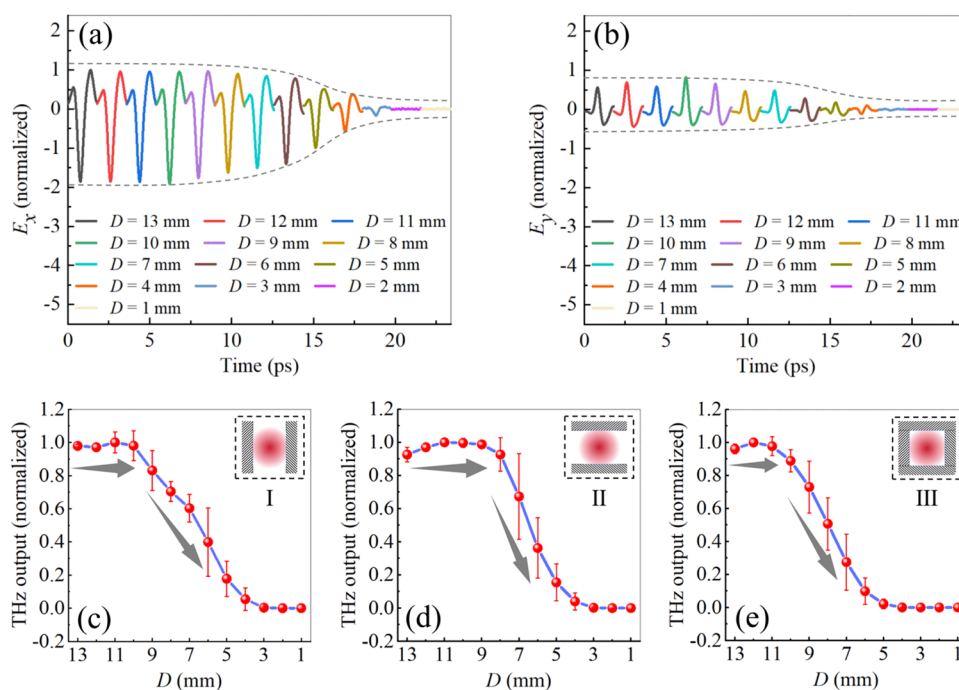


Figure 4. (a–c) represent the variations of x - and y -polarized THz temporal signals and the corresponding THz output with the decreasing laser spot size, in case of blocking the laser beam from both left and right sides simultaneously. (d) and (e) represent the THz yield change in the other two cases, i.e., blocking from top and bottom simultaneously, and in orthogonal directions, respectively.

1, where the maximum lateral electron displacement is estimated to be around $r = 200$ nm under the conventional two-color field pumping, thus guaranteeing a decreasing lateral gradient of the laser electric field $E(r,t)$ in eq 2. In practical experiments, when the laser beam is partially blocked, the range of $1/\eta$ depends on several factors, including the material and edge steepness of the blocking element and focusing conditions, among others. And as the gradient range increases, η would decrease and result in a lower THz enhancement as shown in Figure 2b,d.

EXPERIMENTAL RESULTS

Design of the Setup. Our experimental setup is illustrated in Figure 3. The pump laser (~ 1.8 mJ/pulse, 800 nm, 50 fs, 1 kHz) was focused by a lens, and then passed through a BBO crystal (optional) to generate the single- or dual-color laser field (for the latter case, the ω - 2ω relative phase was set as $\sim \pi/2$ by adjusting the BBO position). At the focal point, an air plasma filament (~ 10 mm in length) was created to radiate THz waves, which were then collected by an electro-optic sampling (EOS) system in x - and y -polarized directions, respectively. Meanwhile, in order to adapt to the different blocking directions, the pumping laser was converted from linear into circular polarization via a $\lambda/4$ plate. Here are some other considerations on experimental design details, including the spatial modulation (blocking) methods and the selection of the focal length of the lens.

- (1) Regarding the blocking methods: We could symmetrically or asymmetrically block the laser beam spot with metal plates to study the THz amplitude variations. First, we intended to verify the reported results in the literature that symmetric beam blocking with low laser power input cannot enhance the THz radiation. Here, we updated the aperture method²³ by using blockers to

symmetrically block the laser beam in three different ways: (I) from left and right, (II) from top and bottom, and (III) from orthogonal directions, as displayed in the inset of Figure 3. On the other hand, to contrast with the above experiments, we adopted an asymmetric blocking method by covering the laser beam with only one blocker, including from the left (IV) or from the top (V) in Figure 3.

- (2) Focal length of the lens and the filament mode cleaning effect: During laser filamentation, under extreme focusing conditions, the spatial mode cleaning effect becomes significant. For example, (i) when the focal length is long as 5.66 m, the sufficiently filament propagation distance could result in a filament length of up to 5 m, and theoretically smooth the laser field distribution into an ideal Gaussian mode;³³ (ii) And if the strongly focused laser with a focal length of 75 mm produces a filament length of only 0.2 mm, the sharp transverse gradients of the laser field pumped by a half-covered laser spot could be even totally eliminated.³⁴ In the former case, the spatial mode cleaning is dominated by the self-focusing effect of the long filament, while in the latter case, the dominant effect is the strong ionization via the tight geometrical focusing. Here, in our blocking experiments, in order to suppress this effect to form a transversely asymmetric plasma column, a proper focal length of 30 cm was chosen for the lens to create an intermediate filament length of approximately 10 mm (differing by about 2 orders of magnitude from 0.2 mm and 5 m, respectively). Thus, our spatial cleaning effect could be nonpronounced, which is also evident from the following test. We performed single-laser-pulse ablation on a thermosensitive paper placed perpendicular to the plasma filament. The insets in Figure 3 show the ablation patterns at the beginning,

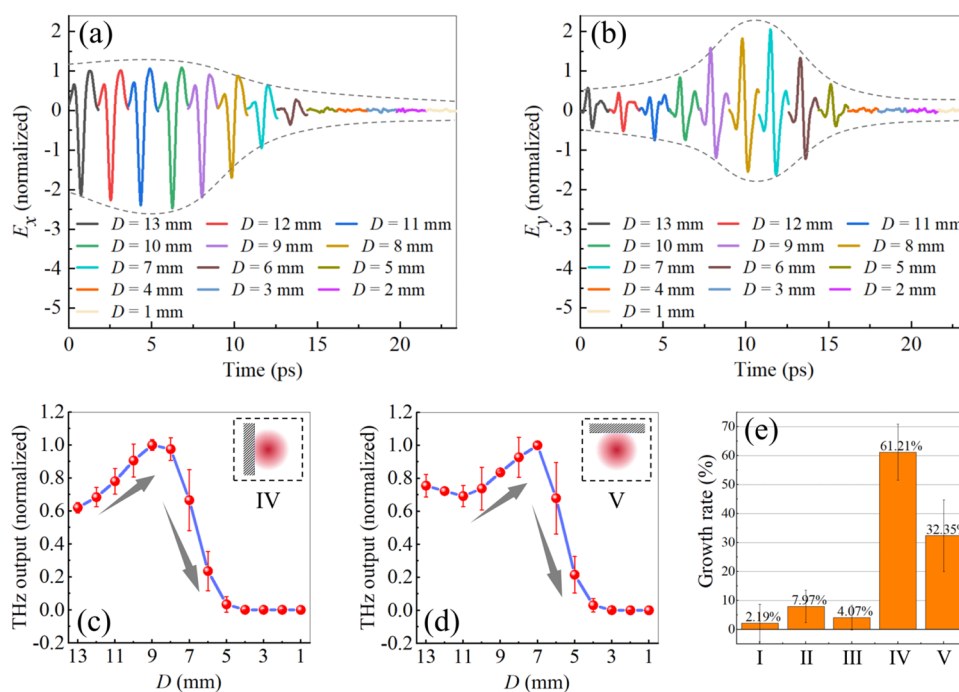


Figure 5. (a–c) represent the variations of x - and y -polarized THz temporal signals and the corresponding THz yield with the decreasing laser spot size, in case of blocking the laser beam from the left side. (d) represents the THz output change in another case, i.e., blocking from the top. (e) summarizes the growth rate of THz yield with the five blocking methods in Figures 4–5.

middle and end of our filament, which were analyzed to determine the degree of energy distribution asymmetry within the filament cross-section. It can be seen that the patterns are approximately circular with ablation areas in (a) and (c) larger than (b), consistent with the evolution of the laser beam along the filament region. (d–f) of the inset illustrate the ablation marks after blocking the left half of the laser spot, showing that the transverse intensity profile of the filament is essentially semi-circular, hinting an acceptable transverse gradient of the laser field distribution.

Next, we plan to validate our theory under both single- and two-color laser field experimental conditions. The latter will be conducted first because its THz signal is much stronger, making us easier to detect it and optimize the entire optical system. After completing the two-color field experiments, the single-color case can be fulfilled by simply removing the BBO crystal.

Symmetrically Spatial Modulation in the Two-Color Case. Taking Inset (I) of Figure 3 as an example, initially, two blockers were tangent to edges of the laser spot, and then they were moved in both directions toward the beam center simultaneously. The original beam diameter was about 13 mm, and the step size of each blocker movement was 0.5 mm. During this process, we recorded the x - and y -polarized THz signals as a function of the distance between blockers (D), as shown as E_x and E_y in Figure 4a,b, respectively. One can see that, when D just started to be decreased, it did not cause significant changes of the THz amplitude. This is because the pump laser is a Gaussian beam characterized by the strong intensity in the center but much weaker at the periphery. Afterward, as D continued to be reduced, the THz pulse amplitude gradually decreased as expected. At the same time, the THz yield (estimated by $A_x^2 + A_y^2$, where A_i is the peak-to-

peak amplitude of E_i) also showed a similar trend of a plateau followed by a continuous declination (Figure 4c).

Next, we repeated the above experimental processes by symmetrically blocking the beam from top and bottom simultaneously as (II) in the inset of Figure 3, and found that the THz yield also exhibited a combination of a plateau and a falling stage as D decreased in Figure 4d. So did the results of the orthogonal blocking method as described in Figure 4e, which was carried out with four blockers (III in the inset of Figure 3). The above studies verified that the method of symmetrically spatial modulation cannot enhance the THz radiation from the laser filament induced by a low input laser energy as previously demonstrated by ref²³

Asymmetrically Spatial Modulation in the Two-Color Case. For the asymmetrical case, the recorded E_x and E_y of the method (IV) are displayed in Figure 5a,b (see also Section B of the Supporting Information for complete THz temporal waveforms, spectra and polarization). Unlike the symmetric method as in Figure 4a,b with continuous reduction, this time, the THz signal showed enhancement as the beam size D decreased. And the change of THz output showed similar trend in Figure 5c, which increased first and then rapidly decreased. So did the method (V) with results in Figure 5d. To summarize the above experimental results, we analyzed the THz growth rate as $G = (P_{\max} - P_0)/P_0$ in Figures 4c–e and 5c–d, where P_0 and $P_{\max}$ represent the first and maximum values of the THz output for a given blocking method, respectively. The calculated results are shown in Figure 5e for the used five different beam blocking methods. And the error for G was calculated by $G^{\text{error}} = (P_{\max}/P_0) \sqrt{(P_{\max}^{\text{error}}/P_{\max})^2 + (P_0^{\text{error}}/P_0)^2}$, where P_0^{error} and $P_{\max}^{\text{error}}$ are corresponding errors of P_0 and $P_{\max}$ (see also Section C of the Supporting Information). It can be found that the growth rate of the THz signal under the first three

symmetric blocking methods was approximately 2–8%, which is negligible considering experimental errors like weak scattering effects when the blocker interacted with the laser spot. In contrast, the asymmetric blocking method has much larger growth values as 30–60%, which is proved to be a valid technique for achieving THz enhancement, also in good agreement with the theoretical prediction of 57%.

It is worth mentioning that, in view of Figure 4c,d, it can be observed that the plateau range width of THz output varies by 1–2 mm along the D axis. Similarly, this phenomenon can also be seen in Figure 5c,d, where the rise period widths of THz output are also slightly different. This can be attributed to the reason that the laser beam spot itself was not perfectly circular, leading to discrepancies when being blocked from orthogonal directions, as well as the fluctuations of growth rates between the first three (or the last two) in Figure 5e. Moreover, to complement the above experiments, we have also performed experiments using bidirectional blockers with different step sizes (Section D of the Supporting Information), or with different positions of the blocker (Section E of the Supporting Information), further demonstrating the feasibility of our THz enhancement scheme by the transverse asymmetry.

As for the far-field spatial profile of the THz radiation with the blocker, one might expect a slight distortion on the original THz donut without the blocker, as the plasma filament is no longer symmetrical. About this point, we experimentally measured the overall divergence angle of the THz radiation profile at about 2 cm after the filament with an adjustable diaphragm, as shown in the inset of Figure 6. By decreasing the

blocking did not significantly alter the original divergence angle of the THz radiation without the blocker.

Asymmetrically Spatial Modulation in the Single-Color Case. At last, we repeated the experiments under the single-color field condition (with BBO removed), and were also able to observe the enhanced THz output, as shown by red waveforms in Figure 7a–b. The other gray signals with disordered fluctuations are noises due to the low generation efficiency of transverse THz waves in the single-color case. The overall THz yield estimated by $A_x^2 + A_y^2$ is displayed in Figure 7c as a function of the remaining laser spot diameter D . It can be seen that the THz enhancement mainly appears when the laser beam's cross-section is half-blocked ($D = 6$ – 7 mm), which can be attributed to the maximum laser field amplitude and transverse current generated at this location. This can also be seen in Figure 7d, in which the calculated residual current increases with the growth of the laser field amplitude E_0 , and thus the THz radiation also becomes stronger.

DISCUSSION

The above experimental results corroborated our theory that the enhanced THz radiation arises from the additional transverse drift current of electrons driven by an asymmetric laser electric field. An accompanying theoretical extension of this model would suggest that if the additional current has an opposite sign to the existing one, it would instead result in an attenuation of THz radiation. To verify this prediction, we adjusted the BBO-to-focus distance (BFD) by a dephasing length, altering the sign of the residual current while maintaining the same laser spot blocking condition (see Section F of the Supporting Information). The experimental findings aligned well with the theoretical prediction, further validating the proposed model. In addition to the aforementioned theory of transverse asymmetry in the laser electric field, other mechanisms might also assist in enhancing the THz radiation, such as (i) the created large gradient of the plasma density or (ii) the laser ponderomotive force along the asymmetric laser electric field.

For (i), when one single edge of the laser beam spot is partially blocked and then focused to create a laser filament in air, the radial distribution of the plasma density (N_e) is also asymmetrical. Clearly, there is a sharp N_e difference across the blocking position. And due to this steep gradient (dN_e/dr), electrons are strongly driven to move across the boundary, which can be described as an enhanced electron diffusion effect. By contrast, the positive ions are much heavier and nearly immobile. Hence, the diffusion motion of free electrons leads to further ($\pm$) charge separation inside the plasma filament, which could possibly contribute to the enhancement of THz radiation. For (ii), the transverse laser ponderomotive force is also a candidate for the underlying mechanism of driving the possible THz enhancement. The presence of a transverse electric field gradient could indeed enhance transverse ponderomotive effects, which in turn can accelerate the electron motion. Moreover, the direction of electron motion driven by the ponderomotive force aligns with the diffusion direction due to the N_e gradient. While these two mechanisms do not contradict our SAPC model, their contributions to the enhanced THz amplitude are likely smaller, as estimated by the electron velocity in Section G of the Supporting Information.

For further improvements of the experiments and applications, the used blocker can be upgraded into a D-

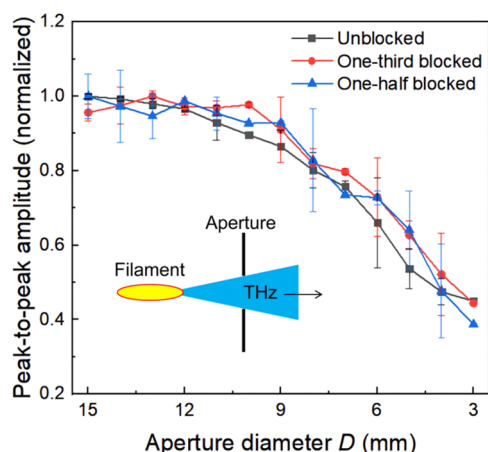


Figure 6. Detected THz peak-to-peak amplitude as a function of the aperture diameter set after the filament induced by a unblocked or blocked laser beam.

aperture size of the diaphragm, we observed that, whether the laser beam was unblocked, one-third or one-half blocked, the reductions of the THz peak-to-peak amplitude are mostly consistent. This indicates that with our current experimental configurations, the blocking of the laser beam does not have a significant impact on the THz radiation angle. We attributed these observations to the following possible reasons: (i) The blocking of the laser beam did not induce strong scattering of the plasma filament as can be seen from (a–f) in Figure 3; (ii) Although the transverse asymmetry of the plasma filament intensified the electron diffusion and drift, the THz enhancement is only about 30–60%. In view of (i) and (ii), the

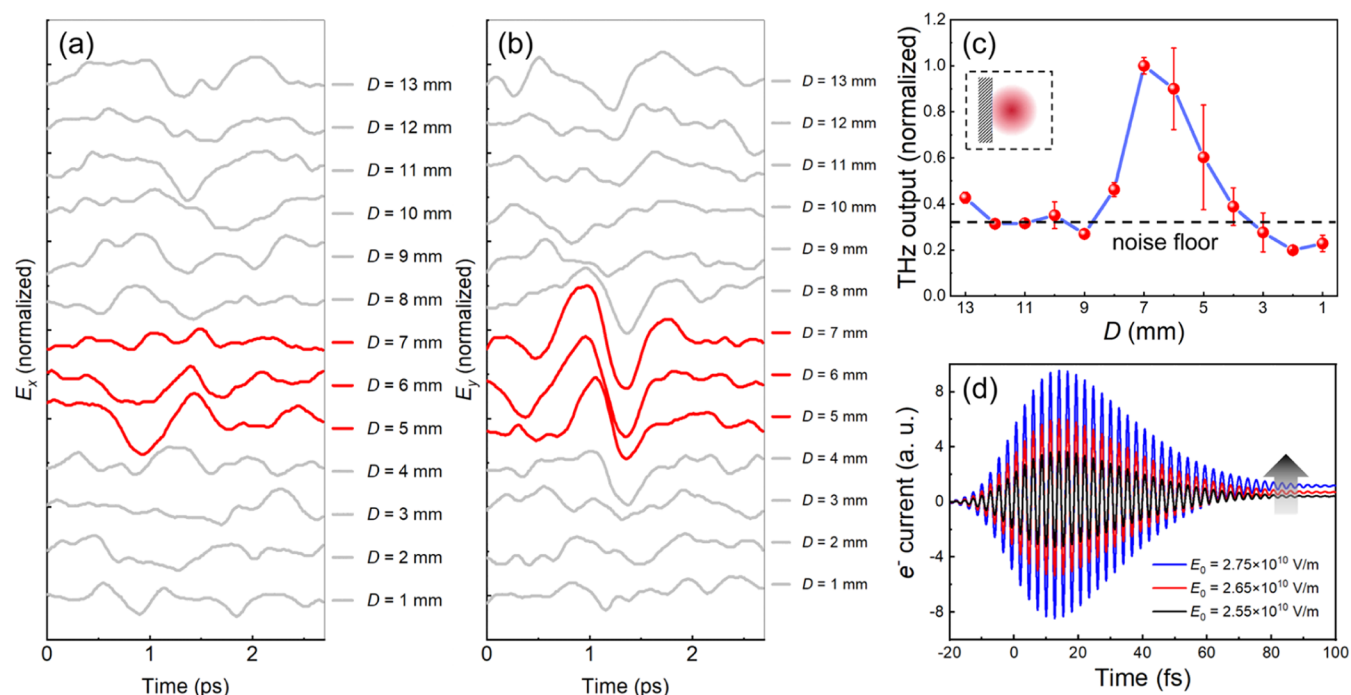


Figure 7. (a) and (b) show the x - and y -polarized THz temporal signals (with vertical offsets) pumped by the single-color laser field with different beam spot apertures D . (c) The THz output as a function of D . (d) Calculation results of the drift current as the laser field amplitude E_0 is varied.

shaped mirror, which could not only enhance the THz radiation, but also allow the laser to be divided into a reflected beam and a transmitted one for saving the laser energy and more purposes. This can be seen in Section H of the [Supporting Information](#). Following our approach, one can also develop different experimental techniques, such as based on the light field control methods to construct a non-Gaussian laser spot with a unidirectional gradient electric field via a spatial light modulator, for the purpose of enhanced THz burst during laser filamentation.

CONCLUSIONS

In summary, this work investigated the transverse modulation of a laser beam spot by symmetrical or asymmetrical blocking with blockers positioned from the left, right, top, bottom, or orthogonal directions. The modulated laser beam was then focused to generate plasma filaments and THz radiation. Impressively, the asymmetrical manners resulted in significant THz enhancements, ranging from 30 to 60%. To interpret these experimental results, we built a SAPC model in two dimensions by considering both the longitudinal temporal laser electric field and its transverse asymmetry. Specifically, the transversely blocked laser beam created a filament column with a pronounced gradient of the inhomogeneous electric field, which further increased the asymmetry of electron displacement. Then, this asymmetry effect led to a strengthened residual current following the laser pulse, which ultimately gave birth to the observed increase in THz power. In contrast, with symmetrical blocking, the opposing electron motions on symmetric sides canceled each other out, preventing any amplification of the overall THz radiation. The proposed experimental method is accessible, requiring no additional modulation devices and lowering the application threshold. It enhances THz transverse wave radiation in both single- and dual-color field setups and may apply to other configurations,

such as tilting lenses or frequency-doubling crystals, prompting a reevaluation of prior studies. Additionally, it optimizes energy usage, enabling stronger THz radiation under low-power lasers and repurposing blocked energy for enhanced functionalities.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsphotonics.4c02464>.

Additional experimental details, methods, and theoretical models, including new designs of the experimental setup (PDF)

AUTHOR INFORMATION

Corresponding Authors

Jiayu Zhao – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China; Shanghai Institute of Intelligent Science and Technology, Tongji University, Shanghai 200092, China; Email: zhaojiayu@usst.edu.cn

Yan Peng – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China; Shanghai Institute of Intelligent Science and Technology, Tongji University, Shanghai 200092, China; orcid.org/0000-0002-8273-5566; Email: py@usst.edu.cn

Yiming Zhu – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China; Shanghai Institute of

Intelligent Science and Technology, Tongji University, Shanghai 200092, China; Email: ymzhu@usst.edu.cn

Authors

Jiajun Yang – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China

Xiaofeng Li – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China; orcid.org/0009-0004-9121-2494

Feifan Zhu – Terahertz Technology Innovation Research Institute, Terahertz Spectrum and Imaging Technology Cooperative Innovation Center, Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology, Shanghai 200093, China

Li Lao – Tera Aurora Electro-optics Technology Co., Ltd, Shanghai 200093, China

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsphotonics.4c02464>

Funding

National Natural Science Foundation of China (61988102, 62335012, 62435010), National key research and development program (2023YFF0719200, 2022YFA1404004), the Youth Sci-Tech “Qimingxing” Program of Shanghai (22QC1400300), Science and Technology Commission of Shanghai Municipality (21S31907400). The authors thank Prof. Xuebin Bian for fruitful discussion.

Notes

The authors declare no competing financial interest.

REFERENCES

- (1) Leitenstorfer, A.; Moskalenko, A. S.; Kampfrath, T.; Kono, J.; Castro-Camus, E.; Peng, K.; Qureshi, N.; Turchinovich, D.; Tanaka, K.; Markelz, A. G.; Havenith, M.; Hough, C.; Joyce, H. J.; Padilla, W. J.; Zhou, B. B.; Kim, K.; Zhang, X. C.; Jepsen, P. U.; Dhillon, S.; Vitiello, M.; Linfield, E.; Davies, A. G.; Hoffmann, M. C.; Lewis, R.; Tonouchi, M.; Klarskov, P.; Seifert, T. S.; Gerasimenko, Y. A.; Mihailovic, D.; Huber, R.; Boland, J. L.; Mitrofanov, O.; Dean, P.; Ellison, B. N.; Huggard, P. G.; Rea, S. P.; Walker, C.; Leisawitz, D. T.; Gao, J. R.; Li, C.; Chen, Q.; Valusis, G.; Wallace, V. P.; Pickwell-MacPherson, E.; Shang, X. B.; Hesler, J.; Ridler, N.; Renaud, C. C.; Kallfass, I.; Nagatsuma, T.; Zeitler, J. A.; Arnone, D.; Johnston, M. B.; Cunningham, J. The 2023 Terahertz Science and Technology Roadmap. *J. Phys. D: Appl. Phys.* **2023**, *56* (22), No. 223001.
- (2) Zhang, X. C.; Shkurinov, A.; Zhang, Y. Extreme Terahertz Science. *Nat. Photonics* **2017**, *11* (1), 16–18.
- (3) Bergé, L.; Kaltenecker, K.; Engelbrecht, S.; Nguyen, A.; Skupin, S.; Merlat, L.; Fischer, B.; Zhou, B.; Thiele, J.; Jepsen, P. U. Terahertz Spectroscopy from Air Plasmas Created by Two-Color Femtosecond Laser Pulses: The ALTESSE Project. *Europhys. Lett.* **2019**, *126* (2), No. 24001.
- (4) Nilforoushan, N.; Kidd, C.; Fournier, A.; Palomo, J.; Tignon, J.; Dhillon, S.; Lhuillier, E.; Li, L.; Davies, A. G.; Linfield, E. H.; Freeman, J. R.; Mangeney, J. Efficient THz Generation from Low-Temperature-Grown GaAs Photoconductive Antennas Driven by Yb-Doped Fiber Amplifier at 200 kHz Repetition Rate. *Appl. Phys. Lett.* **2023**, *123* (24), No. 241107.
- (5) Wu, X. J.; Kong, D. Y.; Hao, S. B.; Zeng, Y. S.; Yu, X. Q.; Zhang, B. L.; Dai, M. C.; Liu, S. J.; Wang, J. Q.; Ren, Z. J.; Chen, S.; Sang, J. H.; Wang, K.; Zhang, D. D.; Liu, Z. K.; Gui, J. Y.; Yang, X. J.; Xu, Y.; Leng, Y. X.; Li, Y. T.; Song, L. W.; Tian, Y.; Li, R. X. Generation of 13.9-mJ Terahertz Radiation from Lithium Niobate Materials. *Adv. Mater.* **2023**, *35* (23), No. 2208947.
- (6) Liao, G. Q.; Liu, H.; Scott, G. G.; Zhang, Y. H.; Zhu, B. J.; Zhang, Z.; Li, Y. T.; Armstrong, C.; Zemaityte, E.; Bradford, P.; Rusby, D. R.; Neely, D.; Huggard, P. G.; McKenna, P.; Brenner, C. M.; Woolsey, N. C.; Wang, W. M.; Sheng, Z. M.; Zhang, J. Towards Terawatt-Scale Spectrally Tunable Terahertz Pulses via Relativistic Laser-Foil Interactions. *Phys. Rev. X* **2020**, *10* (3), No. 031062.
- (7) Koulouklidis, A. D.; Gollner, C.; Shumakova, V.; Fedorov, V. Y.; Pugžlys, A.; Baltuška, A.; Tzortzakis, S. Observation of Extremely Efficient Terahertz Generation from Mid-Infrared Two-Color Laser Filaments. *Nat. Commun.* **2020**, *11* (1), No. 292.
- (8) Zhao, J. Y.; Chu, W.; Wang, Z.; Peng, Y.; Gong, C.; Lin, L.; Zhu, Y. M.; Liu, W. W.; Cheng, Y.; Zhuang, S. L.; Xu, Z. Z. Strong Spatial Confinement of Terahertz Wave Inside Femtosecond Laser Filament. *ACS Photonics* **2016**, *3* (12), 2338–2343.
- (9) Chen, Y. X.; He, Y. H.; Liu, L. Y.; Tian, Z.; Zhang, X. C.; Dai, J. M. Plasma-Based Terahertz Wave Photonics in Gas and Liquid Phases. *Photonics Insights* **2023**, *2*, R06.
- (10) Zhao, J. Y.; Liu, W. W.; Li, S. C.; Lu, D.; Zhang, Y. Z.; Peng, Y.; Zhu, Y. M.; Zhuang, S. L. Clue to a Thorough Understanding of Terahertz Pulse Generation by Femtosecond Laser Filamentation. *Photonics Res.* **2018**, *6* (4), 296–306.
- (11) Hamster, H.; Sullivan, A.; Gordon, S.; White, W.; Falcone, R. W. Subpicosecond, Electromagnetic Pulses from Intense Laser-Plasma Interaction. *Phys. Rev. Lett.* **1993**, *71* (17), 2725–2728.
- (12) Kim, K. Y.; Glowina, J. H.; Taylor, A. J.; Rodriguez, G. Terahertz Emission from Ultrafast Ionizing Air in Symmetry-Broken Laser Fields. *Opt. Express* **2007**, *15* (8), 4577–4584.
- (13) Kim, K. Y.; Taylor, A. J.; Glowina, J. H.; Rodriguez, G. Coherent Control of Terahertz Supercontinuum Generation in Ultrafast Laser-Gas Interactions. *Nat. Photonics* **2008**, *2* (10), 605–609.
- (14) Babushkin, I.; Kuehn, W.; Köhler, C.; Skupin, S.; Bergé, L.; Reimann, K.; Woerner, M.; Herrmann, J.; Elsaesser, T. Ultrafast Spatiotemporal Dynamics of Terahertz Generation by Ionizing Two-Color Femtosecond Pulses in Gases. *Phys. Rev. Lett.* **2010**, *105* (5), No. 053903.
- (15) Babushkin, I.; Skupin, S.; Husakou, A.; Köhler, C.; Cabrera-Granado, E.; Bergé, L.; Herrmann, J. Tailoring Terahertz Radiation by Controlling Tunnel Photoionization Events in Gases. *New J. Phys.* **2011**, *13* (No), No. 123029.
- (16) D’Amico, C.; Houard, A.; Franco, M.; Prade, B.; Mysyrowicz, A.; Couairon, A.; Tikhonchuk, V. T. Conical Forward THz Emission from Femtosecond-Laser-Beam Filamentation in Air. *Phys. Rev. Lett.* **2007**, *98* (23), No. 235002.
- (17) Martínez, P. G. D.; Babushkin, I.; Bergé, L.; Skupin, S.; Cabrera-Granado, E.; Köhler, C.; Morgner, U.; Husakou, A.; Herrmann, J. Boosting Terahertz Generation in Laser-Field Ionized Gases Using a Sawtooth Wave Shape. *Phys. Rev. Lett.* **2015**, *114* (18), No. 183901.
- (18) Du, H. W.; Xu, C. THz Generation from Slow Turn-On, Rapid Turn-Off Femtosecond Laser Pulses Interaction with Gas Plasma. *Infrared Laser Eng.* **2022**, *51* (5), 174–181.
- (19) Yang, N.; Du, H. W. THz Pulse Generation from Shaped Femtosecond Laser Pulses Interaction with Gas-Plasma. *Opt. Express* **2014**, *22* (21), 25494–25499.
- (20) Peng, X. Y.; Li, C.; Chen, M.; Toncian, T.; Jung, R.; Willi, O.; Li, Y. T.; Wang, W. M.; Wang, S. J.; Liu, F.; Pukhov, A.; Sheng, Z. M.; Zhang, J. Strong Terahertz Radiation from Air Plasmas Generated by an Aperture-Limited Gaussian Pump Laser Beam. *Appl. Phys. Lett.* **2009**, *94* (10), No. 101502.
- (21) Oh, T. I.; You, Y. S.; Jhaji, N.; Rosenthal, E. W.; Milchberg, H. M.; Kim, K. Y. Scaling and Saturation of High-Power Terahertz Radiation Generation in Two-Color Laser Filamentation. *Appl. Phys. Lett.* **2013**, *102* (20), No. 201113.

- (22) Chin, S. L. *Femtosecond Laser Filamentation*; Springer: New York, 2010.
- (23) Sheng, W.; Tang, F.; Zhang, Z. L.; Chen, Y. P.; Peng, X. Y.; Sheng, Z. M. Spectral Control of Terahertz Radiation from Inhomogeneous Plasma Filaments by Tailoring Two-Color Laser Beams. *Opt. Express* **2021**, 29 (6), 8676–8684.
- (24) Houard, A.; Liu, Y.; Prade, B.; Tikhonchuk, V. T.; Mysyrowicz, A. Strong Enhancement of Terahertz Radiation from Laser Filaments in Air by a Static Electric Field. *Phys. Rev. Lett.* **2008**, 100 (25), No. 255006.
- (25) Löffler, T.; Jacob, F.; Roskos, H. G. Generation of Terahertz Pulses by Photoionization of Electrically Biased Air. *Appl. Phys. Lett.* **2000**, 77, 453–455.
- (26) Huang, H. H.; Nagashima, T.; Hatanaka, K. Shockwave-Based THz Emission in Air. *Opt. Express* **2023**, 31 (4), S650–S661.
- (27) Huang, H. H.; Nagashima, T.; Kumagai, K.; Hayasaki, Y.; Juodkazis, S.; Tikhonchuk, V. T.; Hatanaka, K. Fine 3D Control of THz Emission in Air with Dual Femtosecond Laser Pre-Pulses at Tunnelling Ionisation Regime. *Light: Adv. Manuf.* **2024**, 5 (3), 30.
- (28) Jia, G. R.; Zhao, D. X.; Zhang, S. S.; Yue, Z. W.; Qin, C. C.; Jiao, Z. Y.; Bian, X. B. Enhanced THz Radiation from Spatially Inhomogeneous Fields. *Chin. Phys. Lett.* **2023**, 40 (10), No. 103202.
- (29) Zhang, Z. L.; Chen, Y. P.; Yang, L.; Yuan, X. H.; Liu, F.; Chen, M.; Xu, J. Q.; Sheng, Z. M.; Zhang, J. Dual-Frequency Terahertz Emission from Splitting Filaments Induced by Lens Tilting in Air. *Appl. Phys. Lett.* **2014**, 105 (10), No. 101110.
- (30) Manceau, J.-M.; Averchi, A.; Bonaretti, F.; Faccio, D.; Trapani, P. D.; Couairon, A.; Tzortzakis, S. Terahertz Pulse Emission Optimization from Tailored Femtosecond Laser Pulse Filamentation in Air. *Opt. Lett.* **2009**, 34 (14), 2165–2167.
- (31) Yu, Z. Q.; Zhang, N.; Wang, J. X.; Dai, Z. J.; Gong, C.; Lin, L.; Guo, L. J.; Liu, W. W. 0.35% THz Pulse Conversion Efficiency Achieved by Ti:Sapphire Femtosecond Laser Filamentation in Argon at 1 kHz Repetition Rate. *Opto-Electron. Adv.* **2022**, 5 (9), No. 210065.
- (32) Su, Q.; Liu, W. W.; Lu, D.; Qi, P. F. Influence of the Tilting Angle of a BBO Crystal on the Terahertz Radiation Produced by a Dual-Color Femtosecond Laser. *IEEE Trans. Terahertz Sci. Technol.* **2019**, 9 (6), 669–674.
- (33) Prade, B.; Franco, M.; Mysyrowicz, A.; Couairon, A.; Buerling, H.; Eberle, B.; Krenz, M.; Seiffer, D.; Vasseur, O. Spatial Mode Cleaning by Femtosecond Filamentation in Air. *Opt. Lett.* **2006**, 31 (17), 2601–2603.
- (34) Heins, A. M.; Guo, C. Spatial Mode Cleaning in Radically Asymmetric Strongly Focused Laser Beams. *Appl. Phys. B: Laser Opt.* **2013**, 113 (3), 317–325.