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Enhancement and modulation of terahertz radiation by multi-color laser pulses*

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We study theoretically intense terahertz radiation from multi-color laser pulse with uncommon frequency ratios. Comparing the two-color laser scheme, of which the uncommon frequency ratio should be set to be a specific value, we show that by using multi-color harmonic laser pulses as the first pump component, the lasers as the second pump component can be adjusted in a continuous frequency range. Moreover, these multi-color laser pulses can effectively modulate and enhance the terahertz radiation, and the terahertz yield increases with the increase of the wavelength of the uncommon pump component and is stable to the laser relative phase. Finally, we utilize the electron densities and velocities of ionization events to illustrate the physical mechanism of the intense terahertz generation.

Keywords: THz generation, multi-color scheme, laser-plasma interaction, uncommon frequency ratio

PACS: 42.65.Re, 32.80.Fb, 52.50.Jm **DOI:** 10.1088/1674-1056/27/8/084209

1. Introduction

Terahertz (THz) radiation from laser-plasma interactions has attracted increasing interest not only for the capability of obtaining intense THz radiation, but also for various applications in ultrafast dynamic, molecular spectroscopy, and biological-medical imaging.[1–5] The conventional emission source in the scheme was based on the femtosecond laser pulse near 800 nm from a Ti:sapphire generator and its second-harmonic field, which jointly focus on air to generate gaseous plasma and then emit the intense THz radiation.[6–11] This scheme has been widely studied, which has illustrated that the residual directional current is responsible for the generation of intense THz radiation, and that its amplitude is determined by the ionization process and electron motion.[12–24] However, the interaction between the gas and femtosecond laser is complicated and the effect of wave mixing is still hard to ignore in the THz-generation mechanism, so many studies have dedicated many efforts to addressing this issue.

Recently, Vvedenskii *et al.* proposed an ionization-induced multiwave mixing theory to explain the THz-generation mechanism by two-color laser pulse with uncommon frequency ratios. In the calculation, they have utilized the conventional laser pulse of 800 nm as a fundamental pulse, and a laser pulse of 1600, 1200, or 533 nm as the second pulse to generate THz radiation, which is virtually as effective as the commonly used laser with a wavelength of 400 nm.[25] Then, the study by Wang *et al.* shows that more frequency ratios can be used to generate the stable THz radiation,[26] and they further presented the first experimental demonstration of THz generation with the uncommon frequency ratios.[27] In their experiment, the combination of 400-nm and 1600-nm pulses and the combination of 800- and 1200-nm pulses were used to generate the intense THz pulse. Currently, the ionization-induced multiwave mixing method is extended to the effective generation of the extremely short midinfrared, visible, and deep-ultraviolet pulses.[28] The above studies show that more wavelength laser pulses can be used to generate THz radiation, however, the available frequency ratios of the two-color laser pulse are limited since they should fix at specific values, and the two pulse energies should be pretty close. In our previous study, we utilized the non-harmonic three-color laser pulses to generate the intense THz pulse. Our results show that the THz radiation can be effectively enhanced and tuned by the negative detuning of the shortest wavelength and the positive detuning of the longest wavelength, but the available frequency range of the three-color laser is also limited and discontinuous since we can only obtain the tunable THz spectrum by detuning the laser pulse wavelength around their harmonic frequencies.[29] Maybe more-harmonic laser pulses can solve this issue but they are hard to further obtain in a practical experiment. We expect to introduce more color laser pulses which are usually utilized to construct the laser pulse,

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thus we propose a comprehensive solution based on the above two methods. Here in this work, a standard harmonic five-color laser pulse is utilized, and its first four harmonic pulses are used as the first pump component, and the fifth pulse is used as the second pump component but with continuously tunable frequency ratios. By using these uncommon five-color laser pulses, the available laser wavelength is widely expanded since most of the laser pulses usually used under laboratory conditions can be utilized to generate the THz radiation, and actually a more intense and tunable THz radiation can be obtained.

So in this paper, we simulate the THz production by a multi-color laser field with uncommon frequency ratios based on the transient photocurrent model. Our results show that the uncommon frequency ratios of the laser pulse can be adjusted continuously, and its relative phase does not affect the THz yield. We also show that the THz yield can further increase with the uncommon pulse wavelength increasing and the THz spectrum can be effectively modulated. Specifically, the low spectral frequency components remain unchanged, and the high spectral frequency components emerge in the THz spectrum and can be effectively detuned by varying the wavelength of the uncommon laser pulse. Finally, we analyze the physical mechanism of THz generation in the uncommon multi-color laser gas interactions.

2. Theoretical model

Our theoretical simulation is based on the transient photocurrent model.[11,15,16,18,19,30] The generally employed multi-color laser field can be expressed as

$$E(t)=\sum_{k=1}^{N}E_k(t)=\sum_{k=1}^{N}\varepsilon_k(t)a_k\cos(k\omega_0 t+\phi_k), \tag{1}$$

where $\varepsilon_k(t)$, a_k, and ϕ_k are the envelope, relative amplitude, and phase of the k-th harmonic, respectively. Here, we assume that Gaussian envelopes $\varepsilon_k(t)=E_0\,\mathrm{e}^{-t^2/\tau^2}$ with amplitude E_0 and duration τ being identical for all colors, with $a_k=1/k$ and $\phi_k=(-1)^k\pi/2$ for a single color. Thus, the standard harmonic multi-color lasers with the sawtooth shapes are obtained which can significantly improve THz convention efficiency.[24] In our simulation, the standard harmonic five-color laser pulse is exploited and the central frequency of the fundamental laser field is set to be $\omega_0=187.5$ THz ($\lambda_0=1600$ nm) for simplicity, since its first four harmonics laser pulses (1600, 800, 533, and 400 nm) contain the main spectral range of laser sources, which can be accessible in practice, and the four laser pulses can be seen as the first pump component. Furthermore, we focus on the laser pulse with uncommon frequency ratios on the THz generation, thus change the shortest wavelength λ_5 of the five-color laser pulse with arbitrary frequency ratios (only long wavelength laser pulses are utilized since the higher harmonics are becoming harder to obtain in experiment). For the intensity regime of our simulation (10^{14} W/cm^2–10^{15} W/cm^2), the Keldysh parameter is $\gamma\leq 1$ and the tunneling ionization is the dominant ionization route. The ionization ratio can be calculated with either the static tunneling (ST) model[31] or the Ammosov–Delone–Krainov (ADK) model.[32] Here, the well-known static tunneling model is employed, and the ionization rate can be expressed as

$$W_{\mathrm{st}}=\frac{\alpha_{\mathrm{ST}}}{|\varepsilon(t)|}\exp\left[-\left(\frac{\beta_{\mathrm{ST}}}{\varepsilon(t)}\right)\right], \tag{2}$$

where $\varepsilon(t)=E(t)/\varepsilon_{\mathrm{a}}$, ε_{a} is the electric field in atomic units, $\alpha_{\mathrm{ST}}=4\omega_{\mathrm{a}}r_{\mathrm{H}}^{5/2}$, $\beta_{\mathrm{ST}}=(2/3)r_{\mathrm{H}}^{3/2}$, ω_{a} is the atomic frequency unit with $\omega_{\mathrm{a}}=\kappa^2me^4/\hbar^3\approx 4.13\times10^{16}$ s^{-1}, and r_{H} is the ionization potential of the gas molecule relative to the hydrogen atom $r_{\mathrm{H}}=U_{\mathrm{ion}}/U_{\mathrm{ion}}^{\mathrm{H}}$. In our simulation, we use $U_{\mathrm{ion}}=15.6$ eV (for N_2 gas) and $U_{\mathrm{ion}}^{\mathrm{H}}=13.6$ eV. Given the ionization rate W_{st}, the increasing rate of the electron density can be expressed as

$$\frac{\mathrm{d}N_{\mathrm{e}}(t)}{\mathrm{d}t}=W_{\mathrm{st}}\left[N_{\mathrm{g}}-N_{\mathrm{e}}(t)\right], \tag{3}$$

where $N_{\mathrm{e}}(t)$ is the time-dependent electron density and N_{g} is the initial neutral gas density. We use the final ionization degree W_{fi} as a measurement of the electron density, which is given by

$$W_{\mathrm{fi}}=N_{\mathrm{e}}(t=\infty)/N_{\mathrm{g}}. \tag{4}$$

For complete ionization, $W_{\mathrm{fi}}=1$. In our simulation, the amplitude of the fundamental laser pulse E_0 is so set so that a moderate ionization degree $W_{\mathrm{fi}}=0.4$ is produced at the standard five-color pulse, since the effects of the laser parameters will be different at higher laser intensities.[33,34] Once freed from the parent molecule, the electron will oscillate with the laser field, and the electron velocity at a subsequent time can be written as

$$v(t,t')=-\frac{e}{m}\int_{t'}^{t}\mathrm{d}t''E(t''), \tag{5}$$

where t' is the ionization instant. Considering the contribution from all ionized electrons, the generated transverse electron current can be expressed as

$$J(t)=\int_{-\infty}^{t}ev(t,t')\exp\left[-\gamma(t-t')\right]\mathrm{d}N_{\mathrm{e}}(t'), \tag{6}$$

where $\mathrm{d}N_{\mathrm{e}}(t')$ represents the change of the electron density in the interval between t' and $t'+\mathrm{d}t'$, $v(t,t')$ can be seen as the velocity of an electron born at $-\infty$, which undergoes variations at the ionization instant t' of the pulse (denoting the electron drift velocity), and γ is the phenomenological electron-ion collision rate ($\gamma\cong 5$ ps^{-1} at atmospheric pressure).[30] The time–dependent electron current $J(t)$ can generate an electromagnetic pulse at THz frequency in the far field. The amplitude of

the generated THz field is proportional to the time-derivative of the electron current $J(t)$ and is written as

$$E_{\mathrm{THz}} \propto \frac{\mathrm{d}}{\mathrm{d}t}[J(t)]. \tag{7}$$

Finally, the THz radiation spectrum is obtained by the Fourier transform of $E_{\mathrm{THz}}(t)$, i.e., $E_{\mathrm{THz}}(\omega) = \mathrm{FFT}[E_{\mathrm{THz}}(t)]$. Throughout this paper, we consider a Gaussian pulse envelope with $\tau = 80$ fs, thus the THz spectrum with a narrow band in the low-frequency spectral component can be obtained,[30,34] and we also consider the THz yield

$$U_{\mathrm{THz}} \propto \int_0^{\omega_{\mathrm{co}}} E^2(\omega)\mathrm{d}\omega$$

below the frequency range $\omega_{\mathrm{co}} = 100$ THz.

3. Results and discussion

In this work, we first utilize an uncommon laser pulse with a wavelength of $\lambda = 1200$ nm to replace the shortest wavelength pulse ($\lambda_5 = 320$ nm) of the standard five-color laser pulse. Figure 1 shows the THz spectra, respectively, for the standard five-color laser pulse with the shortest wavelength of $\lambda_5 = 320$ nm and for the five-color uncommon laser pulse with the shortest wavelength of $\lambda_5 = 1200$ nm. As can be seen, the amplitude of the THz spectrum basically remains constant, but high spectral frequency components emerge in the THz spectral range and obtain a relatively high amplitude. Since the intensity of the low frequency components remains constant and the intensity of the high frequency components is significantly enhanced, the total THz yield can further increase. In our previous studies,[29] we have analyzed the physical mechanism of the high-frequency spectrum generation with the non-harmonic three-color laser pulses and illustrated that the spectrum emergence is attributed to the modulation of the ionization event contributions, it can also be used to explain the spectrum modulation in this case. Furthermore, the five-color laser pulse with uncommon frequency ratio can keep the amplitude of low-frequency spectral components approximately constant, which is significantly superior to the nonharmonic three-color laser pulses and the other incommensurate two-color laser schemes.[30,35]

In a conventional two-color or uncommon two-color and multi-color laser scheme, the THz generation is sensitive to the relative phase of the laser pulses. Thus, we also study the dependence of THz generation on the relative phase of the uncommon multi-color laser pulse. Figure 2 shows the THz spectra for the uncommon five-color laser pulse of $\lambda_5 = 1200$ nm with the relative phases of $\phi_5 = -\pi$, -0.5π, 0, 0.5π, and π. As seen, the amplitude of low spectral frequency components remains unchanged and the amplitude of high spectral frequency components slightly changed with relative phase ϕ_5 varying. It means that the THz yield is phase stable to the replaceable uncommon laser pulse. It can also be seen in the inset of Fig.2 that the THz yield U_{THz} is periodically modulated by the relative phase ϕ_5, but the fluctuation is very small. We believe that these results will bring convenience to the setup of experiments.

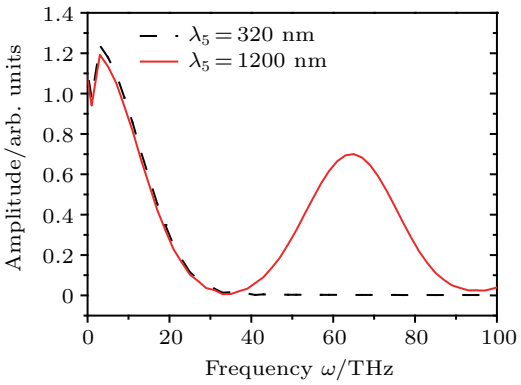


Fig. 1. (color online) THz spectra for the harmonic five-color laser pulse (black dashed line) and the uncommon five-color laser pulse with a replaceable wavelength of $\lambda_5 = 1200$ nm (red solid line).

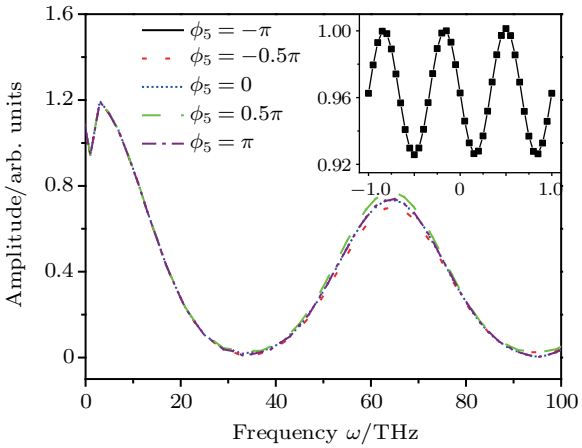


Fig. 2. (color online) THz spectra for the uncommon five-color laser pulse with the replaceable wavelength of $\lambda_5 = 1200$ nm and the relative phases of $\phi_5 = -\pi$ (black solid line), -0.5π (red dotted line), 0 (blue short dotted line), 0.5π (green dashed line), and π (purple dash–dotted line), with inset showing THz yield U_{THz} varying with relative phase $\phi_{5/\pi}$.

We further show that laser pulse with various frequencies can be used as an uncommon pump component to generate THz radiation. Here, we study the dependence of the wavelength of the uncommon laser pulse on THz generation. Figure 3 shows the THz yield U_{THz} as a function of wavelength λ_5 of the replaceable laser pulse. As seen, various long laser pulses can be used to replace the shortest wavelength laser of the standard five-color laser pulse and enhance the THz generation. Moreover, the THz yield U_{THz} basically increases with the replaceable wavelength λ_5 increasing. It is noted that the THz yield is relatively small at the wavelength $\lambda_5 = 1050$ nm, since we only consider the THz yield below the frequency $\omega_{\mathrm{co}} = 100$ THz and the high frequency spectral components

goes out of the scope in this case, but the THz radiation is also enhanced actually in a wider frequency range.

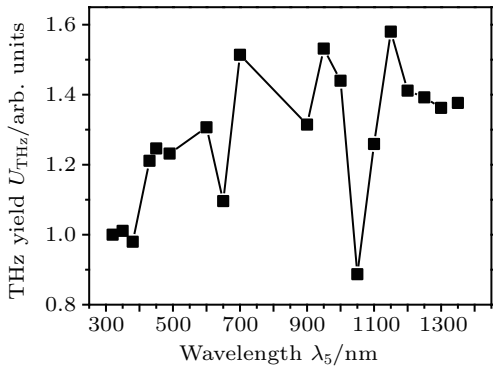


Fig. 3. THz yield $U_{\rm THz}$ versus laser wavelength λ_5 of the uncommon five-color laser pulse. It is noted that the laser wavelengths near the pump harmonic spectral range are excluded from the replaceable wavelengths to keep the whole laser energy constant.

The analysis approach proposed by Babushkin *et al.* has been widely used to estimate the THz yield in previous studies.[29,30,34] In this model, the THz spectrum is a superposition of contributions from individual ionization events, which can be written as $E_{\rm THz}(\omega) \propto \sum_n C_n \mathrm{e}^{\mathrm{i}\omega t_n}$, where $C_n = q\delta\rho_n v_{\rm f}(t_n)$, $\delta\rho_n$, and $v_{\rm f}(t_n)$ are the amplitude, electron density, and velocity for the nth ionization events. It is noted that the amplitude of nth current burst C_n depends on the electron density $\delta\rho_n$ and velocity $v_{\rm f}(t_n)$ of individual ionization events, which determine the THz radiation amplitude ultimately. So, we further discuss the THz generation by analyzing the two quantities for the multi-color laser pulses with uncommon frequency ratios to finally illustrate the enhancement mechanism of THz radiation. Figure 4 shows the increasing rates of electron density $\mathrm{d}N_{\rm e}(t)/\mathrm{d}t$, respectively, for the standard and uncommon five-color laser in an optical period and their corresponding field amplitudes. As can be seen, the standard five-color laser pulse has a well designed sawtooth shape, and its adjacent events show a similar ionization rate. By contrast, the waveform of the uncommon five-color laser pulse is changed and its asymmetry is further increased. Specifically, the positive amplitude increases and the negative amplitude decreases, and the durations of field extrema also increase for the uncommon laser pulse. Since the ionization rate depends on the amplitude of the laser pulse and the ionization time-length depends on the shape of the laser field peak, one of the adjacent events obtains the high amplitude and long duration, and the other suffers the low amplitude at the long duration. It means that the uncommon laser pulse can concentrate the electrons generated in a few ionization events. These results are also well consistent with our pervious study,[23] and the long wavelength laser pulse obtains a high optical period, and thus producing the electrons in fewer ionization events. Obviously, the replaceable long wavelength laser pulse changes the shape of optical cycle, thus affecting the THz generation.

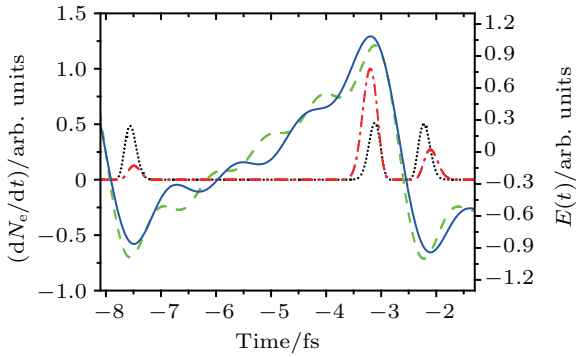


Fig. 4. (color online) Time-dependent field amplitudes of harmonic five-color laser pulse (green dashed line) and uncommon five-color laser pulse with the replaceable wavelength of $\lambda_5 = 1200$ nm (blue solid line) in an optical period, and the corresponding increasing rate of electron density $\mathrm{d}N_{\rm e}(t)/\mathrm{d}t$ for the harmonic (black dotted line) and uncommon (red dash–dotted line) laser pulse.

Figure 5 shows the main ionization events and their corresponding electron velocities. As seen, the electrons are generated in fewer ionization events for the uncommon five-color laser pulse, and the ionization rate in one of the adjacent ionization events is significantly increased and the others decrease. However, the electron velocities of the enhancement ionization events slightly decrease, while the electron velocities of the suppressive ionization events remain constant. Considering all contributions of these ionization events, the electron current significantly increases, which can generate the intense THz radiation.

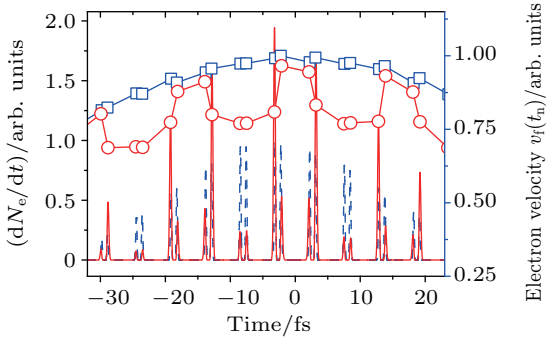


Fig. 5. (color online) Time-dependent increasing rate of electron density $\mathrm{d}N_{\rm e}(t)/\mathrm{d}t$ for the harmonic five-color laser pulse (blue dashed line) and the uncommon five-color laser pulse with the replaceable wavelength of $\lambda_5 = 1200$ nm (red solid line) in the pulse duration, and the corresponding electron velocity $v_{\rm f}(t_n)$ for the harmonic (blue squares) and uncommon (red circles) laser pulse.

It is also indicated that the THz spectrum can be effectively modulated by varying the wavelength of the replaceable laser pulse. Figure 6 shows the THz spectra, respectively, for the standard (black solid line) and uncommon five-color laser pulses with the replaceable wavelength of $\lambda_5 = 1280$ (red dashed line), 1220 (blue dotted line), and 1150 nm (green dashed line). As seen, the peaks of the high frequency spectral component shift with replaceable wavelength λ_5 changing and the tuning range contains the main THz spectral range.

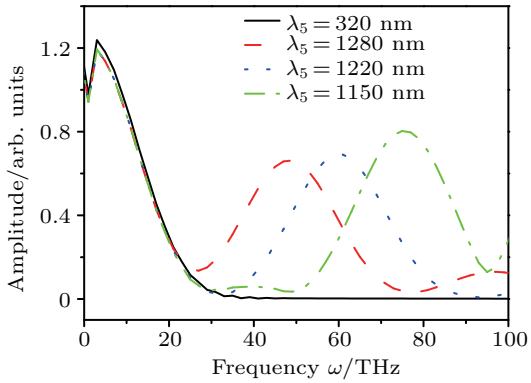


Fig. 6. (color online) THz spectra for the harmonic five-color laser pulse (black solid line), and the uncommon five-color laser pulse with a replaceable wavelengths of $\lambda_5 = 1280$ nm (red dashed line), 1220 nm (blue dotted line), and 1150 nm (green dash–dotted line).

In this paper, our theoretical simulations are based on the transient photocurrent model and we focus on the microscopic effect of the uncommon wavelength laser in the multi-color laser pulse on THz generation. In the current experimental study, He *et al.* introduces another laser pulse of 800 nm into the conventional two-color setup, and through changing the polarization and energy of the introducing laser pulse, the spatial plasma density can be modulated, and thus changing the THz spectral shape.[36] Therefore, we believe that our uncommon multi-color scheme can be used for further experimental studies and provide a way of studying the intense and shaped THz generation.

4. Conclusions

In this work, we have theoretically studied the THz radiation generated by multi-color laser pulses with uncommon frequency ratios. Our results show that by using the multi-color laser pulse, the uncommon frequency components can be extended into a broader and continuous scope. Furthermore, the THz yield can be further increased by increasing the uncommon laser wavelength of the multi-color laser pulse. In addition, it is also shown that the middle and high spectral frequency components of the THz spectrum can be effectively tuned by varying the uncommon wavelength. Finally, our analysis shows that the uncommon multi-color laser pulses can concentrate the electrons generated in a few ionization events and maintain the electron velocity, which results in the intense and shaped THz pulse generation. We believe that these theoretical results can serve as a basis for further experimental studies.

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