Effect of two-color laser pulse duration on intense terahertz generation at different laser intensities

Chenhui Lu,[1] Tao He,[1] Liqiang Zhang,[1] Hui Zhang,[2] Yunhua Yao,[3] Shufen Li,[4] and Shian Zhang[3,*]

[1]*College of Mechanical Engineering, Shanghai University of Engineering Science, Shanghai 201620, People's Republic of China*

[2]*Institute of Science, Information Engineering University, Zhengzhou 450001, People's Republic of China*

[3]*State Key Laboratory of Precision Spectroscopy and Department of Physics, East China Normal University, Shanghai 200062, People's Republic of China*

[4]*College of Physics and Information Engineering, Quanzhou Normal University, Quanzhou 362000, People's Republic of China*

(Received 4 May 2015; published 30 December 2015)

We study theoretically terahertz generation in two-color laser-gas interactions based on a transient photocurrent model. We show that terahertz generation depends on the laser pulse duration and intensity of the two-color laser field. Furthermore, the terahertz amplitude increases with an increase of the laser pulse duration at low laser intensity, but decreases with an increase of the laser pulse duration at high laser intensity. Our analysis shows that the ionization events play an important role in terahertz generation and the terahertz amplitude is determined by the superposition of contributions from individual ionization events.

DOI: 10.1103/PhysRevA.92.063850 PACS number(s): 42.65.Re, 32.80.Fb, 52.50.Jm

I. INTRODUCTION

Recently, the generation of an intense terahertz (THz) pulse via two-color laser focusing in air has attracted considerable attention [1–18] not only for related applications [19–21], such as THz imaging [22,23], but also for the understanding of physical mechanisms [3,4,8,12,15,18]. In this two-color scheme, a fundamental femtosecond laser field combining with its second-harmonic field is focused in air to generate gaseous plasma and the ionized electron accelerated by this two-color pulse will form a quasi-dc photoinduced current, which could emit an intense and broadband THz pulse. This intense THz generation depends on all the parameters of two-color pulses, which have been extensively studied in theory and experiment, such as the effects of intensity or phase [1,2,5,8–12,16–18], the two-color laser intensity ratio [11,24], and the laser polarization on THz generation [25,26].

As an important parameter, the pulse duration can effectively control the times of gas ionization and subsequent electron motion and therefore should have a significant effect on THz generation. Many previous studies have demonstrated that the amplitude of THz pulse increases with an increase of pulse duration at low laser intensity [5,27,28]. By contrast, some works show that the THz pulse amplitude decreases with an increase of pulse duration at high laser intensity or a few-cycle laser pulse [17,29,30]. This issue needs further discussion. Generally speaking, the THz yield is determined by the amplitude of low-frequency electron current, which can be obtained by the free-electron residual current density (RCD) calculation [16–18]. This RCD calculation is useful for estimating the THz pulse yield as it considers only the ultimate current produced by the whole electron motion; but it cannot illustrate the physical mechanism completely and further analysis in the THz spectral domain is needed [18]. Babushkin *et al.* [27] proposed an alternative approach based on the THz spectral analysis. In this model THz generation is associated with a stepwise increase of the plasma formation due to tunneling ionization. The THz spectral components can be attributed to an interference of the discrete and ultrabroadband radiation bursts that are produced around these ionization events. This method considers the electron generation and motion on the discrete attosecond scale, which is very useful for a further understanding of THz generation and can be applied in spectral shaping of THz pulses. So in this paper we comprehensively study the pulse duration effects on the THz generation at different laser intensities and analyze our results in the THz frequency domain.

We simulate the THz generation in the two-color laser field based on the transient photocurrent model. Our results show that the THz amplitude increases and then saturates as the laser intensity increases while the laser pulse duration remains constant. However, the dependence of the THz amplitude on pulse duration is determined by the laser intensity. It is shown that the THz amplitude increases at low laser intensity and decreases at high laser intensity with an increase of the pulse duration. Finally, we analyze the physical mechanism of THz generation in the frequency domain. It is shown that the amplitude of the THz spectrum is determined by the superposition of contributions from individual ionization events, which intrinsically depends on the electron density and velocity of each ionization event.

II. THEORETICAL MODEL

Our theoretical simulation is based on the transient photocurrent model [8–11]. Here the two-color laser field employed is a superposition of the fundamental laser field and its second-harmonic field, which can be expressed as

$$E(t) = \exp(-2\ln 2t^2/\tau^2)[E_1 \cos\omega_0 t + E_2 \cos(2\omega_0 t + \phi)], \tag{1}$$

where E_1 and E_2 are, respectively, the field amplitude of the fundamental laser field and its second-harmonic field, τ is the pulse duration, ω_0 is the central frequency of the fundamental laser field, and ϕ is the relative phase between the fundamental laser field and its second-harmonic field. For the intensity regime of our simulation (10^{14}–10^{15} W/cm^2), the Keldysh parameter $\gamma \leqslant 1$ and tunneling ionization is the dominant

*sazhang@phy.ecnu.edu.cn

ionization route. The ionization ratio can be calculated by the static tunneling model [31,32] or the Ammosov-Delone-Krainov model [33,34]. Here the well-known static tunneling model is employed [18,27] and the ionization rate can be expressed by

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

where $\varepsilon(t) = E(t)/\varepsilon_a$, ε_a is the electric field in atomic units, $\alpha_{\rm ST} = 4\omega_a r_{\rm H}^{5/2}$, and $\beta_{\rm ST} = (2/3) r_{\rm H}^{3/2}$, with ω_a the atomic frequency unit $\omega_a = \kappa^2 m e^4/\hbar^3 \approx 4.13 \times 10^{16}\,{\rm s}^{-1}$ and r_H the ionization potential of the gas molecule relative to hydrogen atom $r_{\rm H} = U_{\rm ion}/U_{\rm ion}^{\rm H}$. In our simulation we use $U_{\rm ion} = 15.6\,{\rm eV}$ (for N_2 gas) and $U_{\rm ion}^{\rm H} = 13.6\,{\rm eV}$. Given the ionization rate $W_{\rm st}$, the increasing rate of the electron density can be expressed by

$$\frac{dN_e(t)}{dt} = W_{\rm st}[N_g - N_e(t)], \quad (3)$$

where $N_e(t)$ is the time-dependent electron density and N_g is the initial neutral gas density. Here the electron density calculation considers the so-called local current (LC) limit. That is to say, only a small volume of gas is irradiated with the pump field. For a high laser intensity ($\geqslant 10^{15}\,{\rm W/cm^2}$), the gas molecules will be completely ionized after the passage of the laser pulse. We use the final ionization degree $W_{\rm fi}$ as a measurement of the electron density, which is given by

$$W_{\rm fi} = N_e(t = \infty)/N_g. \quad (4)$$

For complete ionization, $W_{\rm fi} = 1$. Once freed from the parent molecular, the electron will oscillate with the laser field and the electron velocity at a subsequent time can be written as

$$v(t,t') = -\frac{e}{m}\int_{t'}^{t} dt'' E(t''), \quad (5)$$

where t' is the time when the electron is born. The initial velocity of the electron is assumed to be zero. Considering the contribution of all ionized electrons, the generated transverse electron current can be expressed by

$$J(t) = \int_{t_0}^{t} ev(t,t') \exp[-\gamma(t - t')] dN_e(t'), \quad (6)$$

where $dN_e(t')$ represents the change of electron density in the interval between t and t', $v(t,t')$ is the velocity of these electrons at time t, and γ is the phenomenological electron-ion collision rate ($\gamma \cong 5\,{\rm ps}^{-1}$ at atmospheric pressure) [27]. The time-evolving electron current $J(t)$ can generate an electromagnetic pulse at a THz frequency in the far field and the amplitude of the THz field is proportional to the derivate of the electron current $J(t)$ and is written as

$$E_{\rm THz} \propto \frac{d}{dt}[J(t)]. \quad (7)$$

Finally, the THz radiation spectrum is obtained from the Fourier transform of $E_{\rm THz}(t)$, i.e., $E_{\rm THz}(\omega) = F[E_{\rm THz}(t)]$. In our simulation, the central frequency of the fundamental laser field is set to be $\omega_0 = 12\,500\,{\rm cm}^{-1}$, the relative intensity ratio and phase of the two laser fields are $r = \frac{I_{2\omega}}{I_\omega} = 0.2$ and $\phi = 0.5\pi$, respectively, and $I_{\rm total}$ represents the total laser intensity of the two-color laser field.

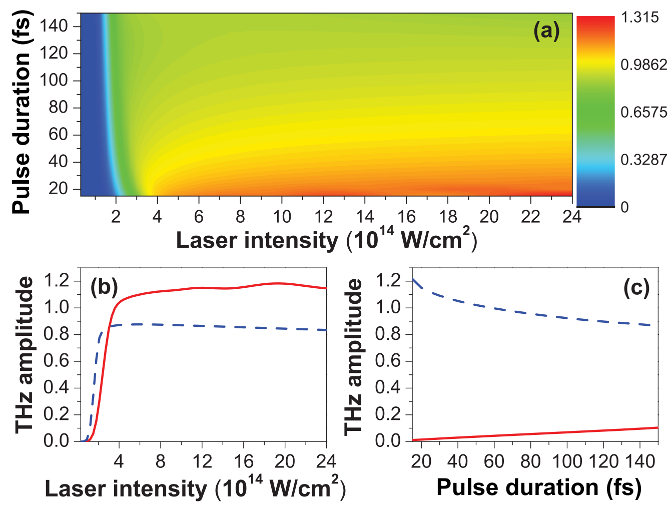

FIG. 1. (Color online) (a) Contour plot of the THz amplitude as a function of the laser pulse duration τ and the laser intensity $I_{\rm total}$. (b) The THz amplitude as a function of the laser intensity $I_{\rm total}$ with a pulse duration of $\tau = 20$ (red solid line) and 140 fs (blue dashed line). (c) The THz amplitude as a function of the pulse duration τ with a laser intensity of $I_{\rm total} = 120$ (red solid line) and 2000 TW/cm^2 (blue dashed line).

III. RESULTS AND DISCUSSION

In this work we comprehensively study the effects of laser pulse duration and intensity on THz generation. Figure 1(a) shows the contour plot of the THz spectral amplitude as a function of the laser pulse duration τ and pulse intensity $I_{\rm total}$. As can be seen, the THz amplitude increases with an increase of the total laser intensity and reaches saturation at a certain laser intensity. It can also be seen in Fig. 1(b) that the THz amplitude with a pulse duration of $\tau = 20$ (red solid line) and 140 (blue dashed line) fs possesses a similar functional dependence on the laser intensity. Moreover, when the pulse duration is short, the THz spectral amplitude reaches a minimal value at low laser intensity and a maximal value at high laser intensity. That is to say, the effects of pulse duration on the THz spectral amplitude are different for the low and high laser intensities. To illustrate this point, Fig. 1(c) further shows the THz spectral amplitude as a function of pulse duration τ with a laser intensity of $I_{\rm total} = 120$ (red solid line) and 2000 (blue dashed line) TW/cm^2. As can be seen, the THz spectral amplitude increases and decreases with an increase of pulse duration for low and high laser pulses, respectively.

Generally, the amplitude of the THz pulse relies on the ionization of gas molecules. Therefore, we first demonstrate the effects of the laser pulse duration τ and intensity $I_{\rm total}$ on the electron density. Figure 2 shows the final ionization degree $W_{\rm fi}$ as a function of the laser pulse duration τ and intensity $I_{\rm total}$. As can be seen, the final ionization degree $W_{\rm fi}$ increases with an increase of the laser intensity $I_{\rm total}$ and will reach saturation for a certain intensity. Compared with Fig. 1(a), it can be found that the THz generation is correlated with the electron density. When the laser intensity is relatively low, only some of the gas molecules are ionized, the electron density increases with an increase of the laser pulse intensity $I_{\rm total}$ and duration τ, and the corresponding THz amplitude also increases. When

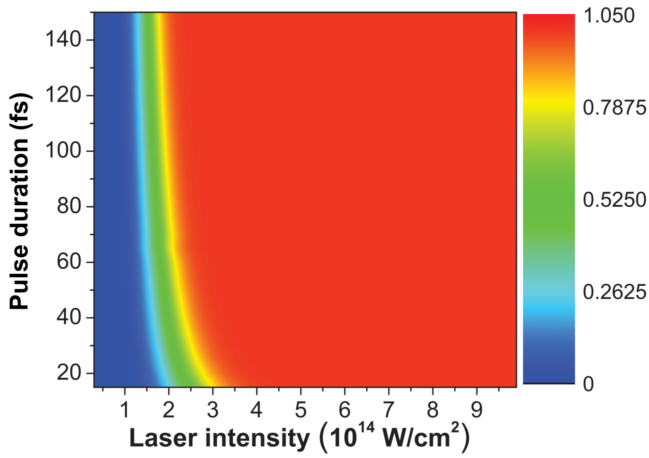

FIG. 2. (Color online) Contour plot of the final ionization degree $W_{\rm fi}$ as a function of the laser pulse duration τ and laser intensity $I_{\rm total}$.

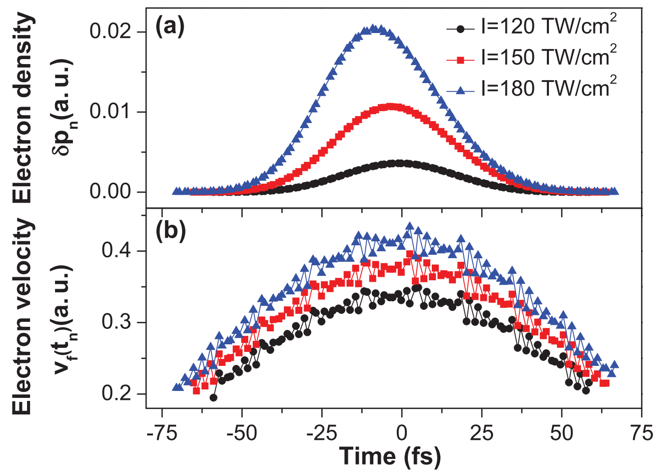

FIG. 3. (Color online) (a) Electron density $\delta\rho_n$ and (b) velocity $v_f(t_n)$ with a laser intensity of $I_{\rm total} = 120$ (black circles), 150 (red squares), and 180 (blue triangles) TW/cm^2 for a laser pulse duration of $\tau = 100$ fs.

the laser intensity is high enough, the gas molecules can be completely ionized, the electron density reaches saturation, and the corresponding THz amplitude will not increase with the increase of laser intensity $I_{\rm total}$. It is noteworthy that the THz amplitude can be further increased with a decrease of the laser pulse τ for the high laser intensity. Therefore, the results should be discussed further and a more accurate analysis is needed.

In this paper the analysis approach proposed by Babushkin *et al.* [27] is used to explain the THz spectral amplitude in our simulation. In this model, the free-electron density $\rho(t)$ increases in short attosecond-scale ionization events that only occur near the extreme of the two-color laser field at times t_n. This ionization event is well separated from others and has a well-defined amplitude $\delta\rho_n$ and shape $H_n(t)$. Therefore, the electron density can be written as a discrete version

$$\rho(t) = \sum_n \delta\rho_n H(t - t_n), \tag{8}$$

where H is a smooth step function. Taking into account the electron velocity, the electron current can be written as a sum of these separated attosecond currents

$$J(t) = \sum\nolimits_n J_n(t) = \sum_n q\delta\rho_n H(t - t_n)[v_f(t) - v_f(t_n)], \tag{9}$$

where $v_f(t) = \frac{q}{m}\int_{-\infty}^{t} E(\tau)d\tau$ and q are the free-electron velocity and electron charge, respectively. In the LC limit, the generated THz pulse in the frequency domain can be obtained by the Fourier transform of Eq. (9) (neglecting the slowly varying and pump items):

$$E_{\rm THz}(\omega) \propto \sum_n C_n e^{i\omega t_n}, \tag{10}$$

with

$$C_n = q\delta\rho_n v_f(t_n), \tag{11}$$

where C_n is the amplitude of the nth attosecond current burst that is produced around the nth ionization event and $\delta\rho_n$ and $v_f(t_n)$ represent, respectively, the electron density and velocity for the nth ionization event. As can be seen in Eq. (10), the THz spectrum is a superposition of contributions from these individual ionization events. Due to the spectral phase $e^{i\omega t_n}$, these contributions cancel each other at the high-frequency part and interfere instructively at the low-frequency part. Finally, the THz spectral can be obtained and its amplitude is determined by the whole amplitude of the current bursts $C_{\rm total} = \sum_n C_n$. This approach provides an easy prediction of the THz spectrum for arbitrary input field shapes, which is also useful for analyzing our results.

Since the amplitude of the nth current burst C_n is determined by the electron density $\delta\rho_n$ and velocity $v_f(t_n)$, we discuss the THz generation by analyzing the two quantities at different laser pulse intensities and durations and also illustrate their roles in limiting the THz output. We first study the dependence of the two quantities on the laser pulse intensity $I_{\rm total}$. Figure 3 shows the electron density $\delta\rho_n$ [Fig. 3(a)] and velocity $v_f(t_n)$ [Fig. 3(b)] with a laser intensity of $I_{\rm total} = 120$ (black circles), 150 (red squares), and 180 (blue triangles) TW/cm^2 for a laser duration of $\tau = 100$ fs. As can be seen, the ionization events evenly distribute on both sides of the laser pulse center, the electron density $\delta\rho_n$ rapidly increases, and the electron velocity $v_f(t_n)$ also increases with an increase of the laser intensity. This is because the ionization rate rapidly increases at low laser intensity and the electron velocity is determined by the amplitude of the laser field at the ionization events, thus the electron density massive increases with a small change of laser intensity and the electron velocity also increases with an increase of the laser intensity. Finally, the whole amplitude of the current bursts $C_{\rm total}$ will increase with the laser intensity, which can result in a more intense THz pulse generation.

However, when the laser intensity is high enough, the ionization rate approaches saturation and the gas molecules will be completely ionized in a few optical periods. As can be seen in Fig. 4(a), when the laser intensity is significantly increased, almost all of the ionization events will occur before the central position of the laser pulse and the electron density $\delta\rho_n$ can hardly increase. Furthermore, the corresponding

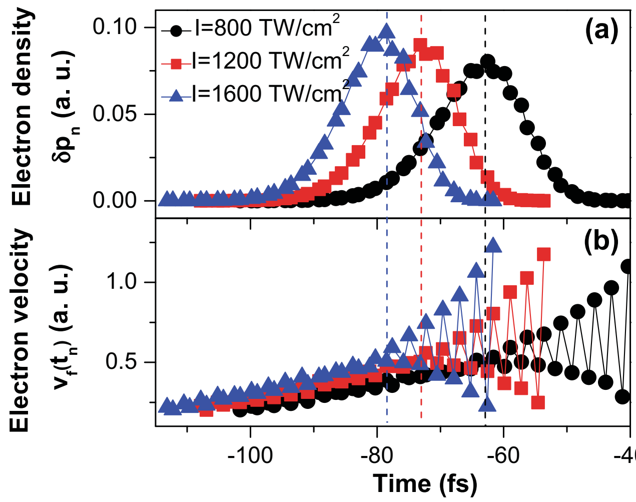

FIG. 4. (Color online) (a) Electron density $\delta\rho_n$ and (b) velocity $v_f(t_n)$ with a laser intensity of $I_{\text{total}} = 800$ (black circles), 1200 (red squares), and 1600 (blue triangles) TW/cm^2 for a laser pulse duration of $\tau = 100$ fs. The dashed lines indicate the velocity of the central ionization event.

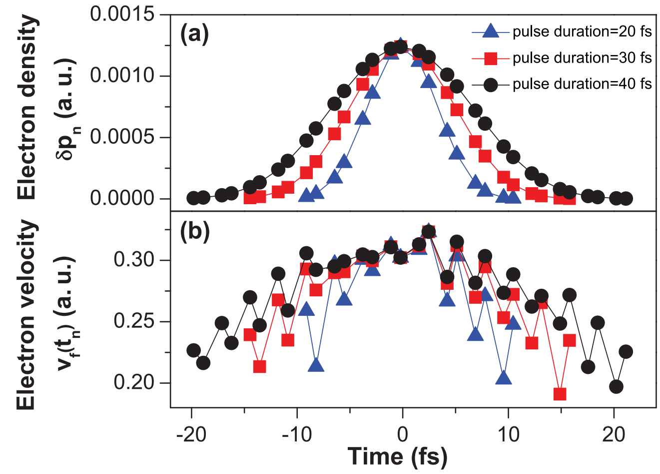

FIG. 6. (Color online) (a) Electron density $\delta\rho_n$ and (b) velocity $v_f(t_n)$ with a laser pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs for a laser pulse intensity of $I_{\text{total}} = 100\,\text{TW/cm}^2$.

electron velocity $v_f(t_n)$ is almost the same for different laser intensities. As shown in Fig. 4(b), the ionization times are different, but the electron velocities at the central ionization events and both sides are almost the same. Therefore, in this case the whole amplitude of the current bursts C_{total} is determined by the sum of the electron density $\delta\rho_n$. Since the electron density approaches saturation for a high laser intensity, the amplitude of the THz spectra also reaches the maximum value. In order to further illustrate the point, we compare the whole amplitude of the current bursts C_{total} with the final ionization degree W_{fi} at different laser intensities. Figure 5 shows the final ionization degree W_{fi} [Fig. 5(a)] and the whole amplitude of the current bursts C_{total} [Fig. 5(b)] as a function of the laser intensity I_{total} for a pulse duration of $\tau = 20$ (black solid line) and 100 (red dashed line) fs. As can be seen, when the gas molecules are completely ionized for a certain laser intensity, the whole amplitude of the current bursts C_{total} also reaches the maximum at this laser intensity and can hardly increase with the increase of laser intensity, which results in the maximal THz radiation. This result can be regarded as corresponding to Fig. 1(b).

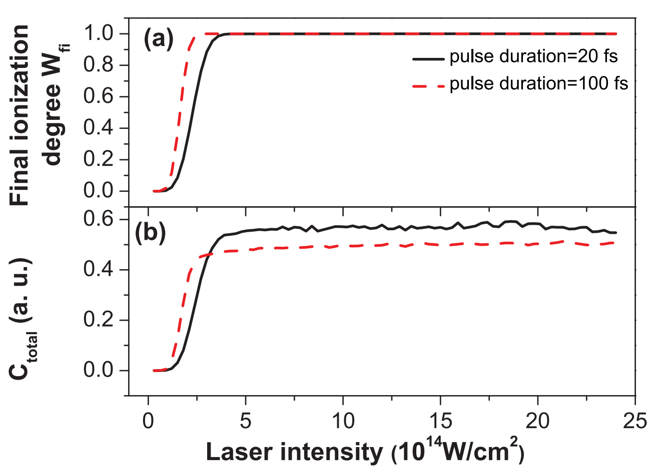

FIG. 5. (Color online) (a) Final ionization degree W_{fi} and (b) whole amplitude of current bursts C_{total} as a function of laser intensity I_{total} for a laser pulse duration of $\tau = 20$ (black solid line) and 100 (red dashed line) fs.

Next we study the dependence of the two quantities on the laser pulse duration τ. Figure 6 shows the electron density $\delta\rho_n$ [Fig. 6(a)] and electron velocity $v_f(t_n)$ [Fig. 6(b)] with a pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs for a total laser intensity of $I_{\text{total}} = 100\,\text{TW/cm}^2$. As can be seen, both the electron density $\delta\rho_n$ and the electron velocity $v_f(t_n)$ increase with the increase of pulse duration for the same ionization events distributed on both sides of the pulse center and more ionization events will occur for long pulse duration. To further determine the THz amplitude, we also present the amplitude of the nth current burst C_n for the three laser pulse durations. As can be seen in Fig. 7, the number of ionization events increases with an increase of the pulse duration τ and the amplitudes of the ionization events are larger for the longer pulse duration at the same ionization times. Moreover, the whole amplitude of the current bursts C_{total} obtains the maximal value at the pulse duration $\tau = 40$ fs, as shown in the inset of Fig. 7. Since the THz amplitude is determined by the whole amplitude of the current bursts C_{total}, therefore the intense THz radiation can be obtained at long pulse duration for low laser intensity. In general, the envelope of the laser field slowly varies for long pulse duration and the laser field amplitude is large at the same ionization times. Thus this laser can produce more free electrons with high velocity and also increases the number of ionization events, which ultimately results in more intense THz radiation. In this case, the increase of THz amplitude can be roughly attributed to the increase of the electron density. However, the volume of neutral gas is constant in the LC limit, so the gas molecules can be completely ionized at high laser intensity ($I_{\text{total}} \geqslant 10^{15}\,\text{W/cm}^2$) and the case will be different.

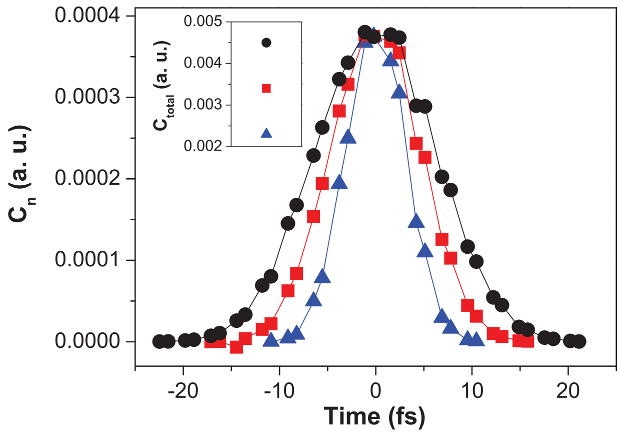

FIG. 7. (Color online) Amplitude of the nth current burst C_n with a pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs for a total laser intensity of $I_{\mathrm{total}} = 100\,\mathrm{TW/cm^2}$. The inset shows the whole amplitude C_{total} for the three pulse durations.

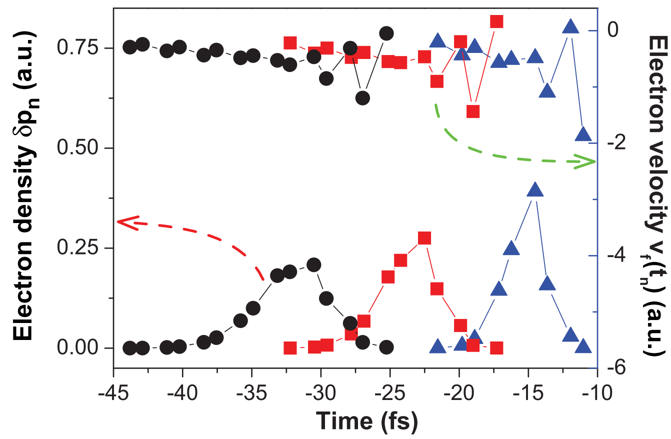

FIG. 9. (Color online) Electron density $\delta\rho_n$ and velocity $v_f(t_n)$ of the nth ionization event with a total laser intensity of $I_{\mathrm{total}} = 2000\,\mathrm{TW/cm^2}$ for a pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs.

For comparison, Fig. 8 shows the amplitude of the nth current burst C_n with a pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs for a total laser intensity of $I_{\mathrm{total}} = 2000\,\mathrm{TW/cm^2}$. As can be seen, the number of ionization events decreases, while the amplitudes of the main ionization events increase as the pulse duration τ decreases. It is noteworthy that the whole amplitude of these current bursts C_{total} is mainly determined by the amplitudes of the main burst currents since the amplitudes of the ionization events at the both sides are small and negligible. As shown in the inset of Fig. 8, the whole amplitude of these current bursts C_{total} obtains the maximal value at a pulse duration of $\tau = 20$ fs, which results in the more intense THz radiation.

To illustrate this issue, we further discuss the THz generation by analyzing the electron density $\delta\rho_n$ and velocity $v_f(t_n)$ under the same conditions. Figure 9 shows the electron density $\delta\rho_n$ and velocity $v_f(t_n)$ with a pulse duration equal to 20 (blue triangles), 30 (red squares), and 40 (black circles) fs

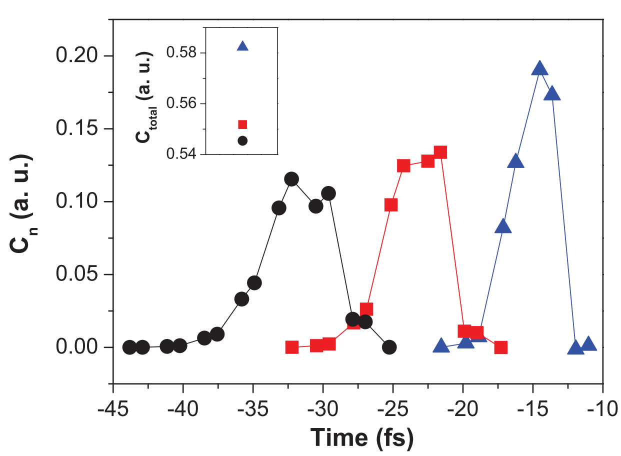

FIG. 8. (Color online) Amplitude of the nth current burst C_n with a pulse duration of $\tau = 20$ (blue triangles), 30 (red squares), and 40 (black circles) fs for a total laser intensity of $I_{\mathrm{total}} = 2000\ \mathrm{TW/cm^2}$. The inset shows the whole amplitude C_{total} for the three pulse durations.

for a total laser intensity of $I_{\mathrm{total}} = 2000\,\mathrm{TW/cm^2}$. As can be seen, the electrons are all generated in a few ionization events and more electrons can be produced in the central ionization events with a decrease of pulse duration τ. This result can be explained by the ionization process. Since the envelope of the laser field rapidly changes for a short pulse duration and the gas density is limited, the gas molecules will be completely ionized in a few ionization events and the ionization events that are close to the center position of laser pulse can produce more electrons in a single ionization event. In other words, when the laser intensity is high enough, the short pulse duration can reduce the ionization event numbers and concentrate the electron production on a few ionization events. Figure 9 also shows the electron velocity $v_f(t_n)$ of each ionization event for the three pulse durations. As can be seen, the electron velocity of the main ionization events will slightly increase with a decrease of the pulse duration. Therefore, the amplitude of the main ionization events obtains the maximal value and the whole amplitude of them is also maximal for the short laser pulse duration, which results in the maximal THz radiation.

Finally, it is important to further investigate the effect of pulse duration on the THz generation at the same laser flux since the input flux is usually fixed and the pulse duration can be flexibly controlled in real experimental conditions. For consistency, we define the laser flux $F = I_{\mathrm{total}}\tau$ for simplicity. Figure 10 presents the contour plot of the THz amplitude as a function of the laser pulse duration τ and laser flux F. As can be seen, the THz amplitude increases with an increase of the laser flux F and can further increase with a decrease of the laser pulse duration τ. The previous analysis will be still suitable for these results. When the laser flux F remains constant, the laser intensity I_{total} will increase if the pulse duration τ decreases. The electron density $\delta\rho_n$ and velocity $v_f(t_n)$ can significantly increase as the laser intensity I_{total} increases, which results in an increase of THz generation under the incomplete ionization condition. When the gas molecules are completely ionized, the pulse duration plays an important role in controlling the number of ionization events. The number of ionization events decreases as the pulse duration τ decreases. This means that

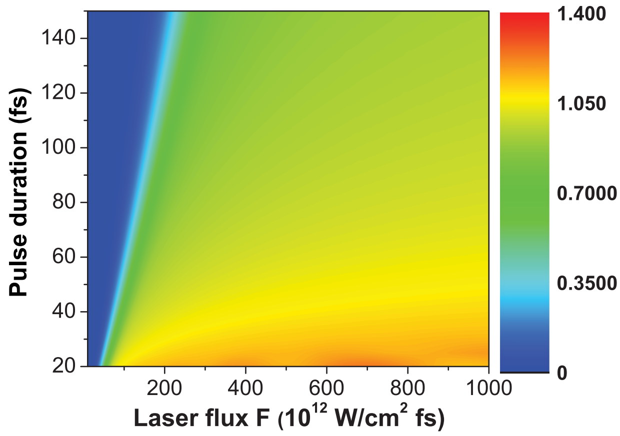

FIG. 10. (Color online) Contour plot of the THz amplitude as a function of the laser pulse duration τ and laser flux F.

the short pulse duration can further decrease the ionization event occurrence and concentrate the electron production in a few ionization events, which will produce the maximal electron current and result in the maximal THz output. In other words, the gas molecules can rapidly absorb a photon in a few laser optical periods and produce the maximal net electron current for short pulse duration although the laser flux is the same. It also can be seen in Fig. 1(a) that the THz output for the pulse duration $\tau = 20$ fs and laser intensity $I_{\text{total}} = 20\,\text{TW/cm}^2$ is stronger than that for the laser pulse duration $\tau = 80$ fs and intensity $I_{\text{total}} = 5\,\text{TW/cm}^2$ despite the equal laser flux. Consequently, these results further indicate that the tunneling ionization process plays an important role in THz generation.

IV. CONCLUSION

We have studied theoretically the dependence of THz generation on the laser pulse duration at different laser intensities. Our results show that the THz amplitude increases with an increase of laser pulse duration at low laser intensity and decreases with an increase of laser pulse duration at high laser intensity. Furthermore, the physical mechanisms of THz generation at low and high laser intensities were discussed via THz spectrum analysis. It was shown that the THz spectral amplitude is the superposition of contributions from individual ionization events and the electron density and velocity of each ionization event play an important role in limiting the THz output. We believe that these theoretical results will be useful for further understanding the THz generation and THz pulse shape control.

ACKNOWLEDGMENTS

This work was partly supported by the National Natural Science Foundation of China (Grants No. 11304396 and No. 51305254) and the Open Foundation of Zhejiang Provincial Key Laboratory of Part Rolling Forming Technology, China (Grant No. ZKL-PR-200305).

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