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Generation of third harmonic emission in propagation of femtosecond laser pulses in air*

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The main characteristics of the third harmonic emission generated by femtosecond laser pulses propagating in air are investigated by numerically solving the coupled nonlinear Schrödinger equations. Strong third harmonic emission is observed with a maximum conversion efficiency as high as 0.43%. The on-axis phase difference between fundamental beam and third harmonic is investigated. The result is in good agreement with the phase-locking mechanism. Dependence of the conversion of third harmonic emission on focusing conditions is also studied. The results are also compared with those of experiments.

Keywords: third harmonic emission, plasma channel
PACC: 4265K, 4265J

1. Introduction

As the rapid development of studies on the propagation of femtosecond laser pulses in air, the process of third harmonic (TH) emission has attracted much interest.[1−8] The TH emission is a key process for understanding important features in the propagation of the self-channelled femtosecond laser pulses, such as conical emission and spectral supercontinuum.[3−7] Although the peak intensity in the plasma channel is clamped down to 10^{14}W/cm^2,[2,9,10] it is sufficient to generate strong TH emission. The combined effects of self-focusing and plasma generation in the channel contribute to the phase-matching condition, [7] resulting in a conversion efficiency of TH emission as high as 0.5% in theory [6] and 0.3% in experiment.[1−4] It was shown that the conversion efficiency remains roughly constant in the whole filament, much longer than the coherence length. An explanation of the phase-locking mechanism is that the envelope phase difference between the fundamental pulse and the TH emission almost maintains π in the filament range.[2,6] The TH pulse also has the conical emission[6,7] which implies that the TH emission experiences a more important spectral diffraction. In addition, the strong TH emission has a great influence on the spectral supercontinuum generation, which is responsible for the spectrum extending to UV wavelength.[3,5,6] This is important for the application of atmospheric sensing. However, the potential mechanisms in the process of TH generation have not been well understood so far.

In this paper, we present numerical simulation studies on the main characteristics of TH emission during the propagation of femtosecond pulses in air. We demonstrate the process of TH emission and the conversion efficiency as a function of the propagation distance. We also study the phase-locking mechanism by calculating the on-axis phase difference between the fundamental pulse and the TH emission. The different conversion of TH emission under different focusing conditions is investigated and the results are compared with those of experiments.

2. Numerical simulation

The theoretical model to describe TH emission in the propagation of femtosecond laser pulses in air is based on a set of coupled equations.[2,6] The equations include the extended nonlinear Schrödinger equation coupled with the evolution equation of the electron density produced by the multiphoton ionization. The models in Refs.[2] and [6] are slightly different, but the outcomes are similar and in good agreement with

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experiments. Thus we follow the full spatiotemporal model and numerically solve the equations to present simulations on the characteristics of TH emission. In the frame moving at the group velocity of fundamental beam, the coupled equations can be written as:[2,6]

$$\Big(\mathrm{i}\frac{\partial}{\partial z}+\frac{1}{2k_0}\nabla_\perp^2-\frac{k_\omega''}{2}\frac{\partial^2}{\partial t^2}\Big)E_\omega + k_0n_2(|E_\omega|^2E_\omega+2|E_{3\omega}|^2E_\omega+E_\omega^{*^2}E_{3\omega}) - \frac{k_0}{2}\frac{\rho}{\rho_\mathrm{c}}E_\omega+\frac{\mathrm{i}}{2}\beta_\omega^{(K_\omega)}|E_\omega|^{2K_\omega-2}E_\omega=0\,, \quad (1)$$

$$\Big(\mathrm{i}\frac{\partial}{\partial z}+\frac{1}{6k_0}\nabla_\perp^2+\mathrm{i}\Delta\upsilon^{-1}\frac{\partial}{\partial t}-\frac{k_{3\omega}''}{2}\frac{\partial^2}{\partial t^2}-\Delta k\Big)E_{3\omega} + 3k_0(n_2^{3\omega}|E_{3\omega}|^2E_{3\omega}+2n_2|E_\omega|^2E_{3\omega}+n_2E_\omega^3/3) - \frac{k_0}{6}\frac{\rho}{\rho_\mathrm{c}}E_{3\omega}+\frac{\mathrm{i}}{2}\beta_{3\omega}^{(K_{3\omega})}|E_{3\omega}|^{2K_{3\omega}-2}E_{3\omega}=0\,, \quad (2)$$

$$\frac{\partial\rho}{\partial t}=(\sigma_\omega|E_\omega|^{2K_\omega}+\sigma_{3\omega}|E_{3\omega}|^{2K_{3\omega}})(\rho_0-\rho)\,. \quad (3)$$

In the coupled equations, the beam is assumed to be linearly polarized and propagate along the z-axis. We take into account several linear and nonlinear effects in the propagation dynamics including diffraction, group-velocity dispersion, self-phase modulation and cross-phase modulation, electron defocusing and multi-photon ionization loss. The fundamental beam and the TH emission are coupled through cross-phase modulation, which causes the energy transfer between them. The parameters we used are as follows. The input fundamental beam has a central wave length $\lambda_0=800$ nm and a central wave number $k_0=2\pi/\lambda_0$. The group velocity mismatch $\Delta\upsilon=[\upsilon_\mathrm{g}(3\omega)^{-1}-\upsilon_\mathrm{g}(\omega)^{-1}]^{-1}=0.44$ cm^2/fs accounts for the temporal walk-off. The coefficients of group-velocity dispersion are $k_\omega''=0.2$ fs^2/cm and $k_{3\omega}''=1.0$ fs^2/cm, and $\Delta k=3k_0(n_\omega-n_{3\omega})=-5.0$ cm^{-1} refers to the linear wave vector mismatch. Here, $n_2=4\times10^{-19}$ cm^2/W and $n_2^{3\omega}=8\times10^{-19}$ cm^2/W represent nonlinear refractive indices of the fundamental pulse and the TH emission, respectively. In addition, the electrons of plasma are generated by multiphoton ionization of oxygen in our calculation. Therefore, in this case, the numbers of photons required for ionization are $K_\omega=8$ and $K_{3\omega}=3$, and the coefficients of multiphoton ionization are $\sigma_\omega=2.88\times10^{-99}$ s$^{-1}\cdot$cm^{16}/W^8 and $\sigma_{3\omega}=1.91\times10^{-28}$ s$^{-1}\cdot$cm^6/W^3 for the two optical fields. According to this, the coefficient of multiphoton ionization absorption can be calculated by the equation $\beta_{\omega(3\omega)}=\sigma_{\omega(3\omega)}\times K_{\omega(3\omega)}\rho_0\hbar\omega_{(3\omega)}$ for the fundamental beam and the TH emission, where $\rho_0=5.4\times10^{18}$ cm^{-3} is the density of neutral oxygen atoms. $\rho_c=1.8\times10^{21}$ cm^{-3}, ρ_c is the plasma critical density defined at 800 nm.

3. Results and discussion

We numerically solve the set of coupled equations to investigate the propagation dynamics of the fundamental beam and the TH emission. The initial fundamental beam is modelled as an unperturbed Gaussian function with a waist (FWHM) of 3 mm and a pulse duration (FWHM) of 30 fs. Thus, it can be written as

$$E_\omega(x,y,z=0,t)=E_0\exp\Big(-\frac{x^2+y^2}{w_0^2}-\frac{t^2}{\tau^2}-\frac{\mathrm{i}kr^2}{2f}\Big)\,.$$

Here, $E_0=\sqrt{2P_\mathrm{in}/\pi w_0^2}$ denotes the peak intensity of initial fundamental IR pulse. w_0 represents the beam waist and τ is the pulse duration. We take the initial condition to be one colour beam, thus, the input third harmonic is defined as $E_{3\omega}(x,y,z=0,t)=0$. In order to investigate the characteristics of THG by the self-channelled IR pulse in air, we first take into account the case of input power $P_\mathrm{in}=3P_\mathrm{cr}$, where P_cr is the self-focusing threshold, and the initial focal length $f=1$ m. After that we simulate the different conversions of the THG under different focusing conditions, using the condition similar to that in Ref.[3].

Our simulations demonstrate the characteristics of TH emission when fundamental IR pulse with an input power of $P_\mathrm{in}=3P_\mathrm{cr}$ propagates in air, as shown in Fig.1. Figure 1(a) shows that the on-axis intensities of the fundamental (solid line) and the TH (dashed line) pulses evolve as a function of propagation distances. The common feature of the on-axis intensities of the two optical fields is that they have the same evolution tendency as the propagation. The ultrashort laser pulses self-focus before the focal point due to nonlinear Kerr effect, ensuring that the on-axis intensity increases rapidly. Consequently, the electrons induced by ionization tend to defocus the beam. This results in the decrease of intensity. After defocusing, the most interesting feature is that the beam refocuses with propagation, thus creating the second peak in the profile with propagation distance. The results are in accord with the dynamic spatial replenishment.[11] From Fig.1(b), we can obtain strong TH emission with a maximum conversion efficiency as high as 0.43% and the roughly constant evolution curve of conversion efficiency in the filament range. The current explanation for this phenomenon is the phase-locking mecha-

nism, i.e., the phase difference $\Delta\phi = 3\phi_\omega - \phi_{3\omega}$ maintains the constant π over the whole filament length. To further understand the mechanism, we calculate the on-axis phase difference between the fundamental pulse and the TH emission, as shown in Fig.1(c). In Fig.1(c), we can observe that the on-axis phase difference almost remains a constant π in the propagation direction except for the following two cases. When the energy of the TH emission becomes zero or small, the phase difference deviates from the constant.

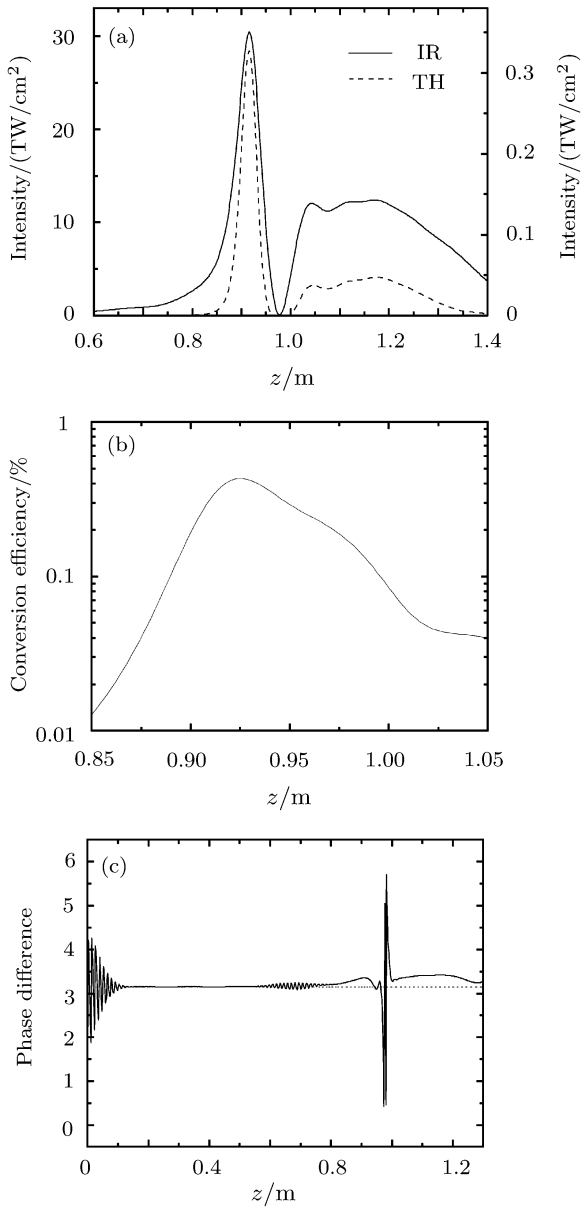


Fig.1. (a) On-axis intensity (r=0, t=0) of the fundamental (solid line) (left-hand scale) and third harmonic (dotted line) (right-hand scale) pulses. (b) The TH conversion efficiency as a function of propagation distance. (c) The on-axis phase difference as a function of propagation distance. The dashed line denotes the constant π.

The other case is that, when pulses cross the geometrical focal plane, the phase difference fluctuates from π. We suggest that this may be caused by the Guoy shift.[12] As the fundamental Gaussian beam passes through the focus, it experiences π phase shift. This will affect its coupling with the TH emission through the nonlinear term E_1^3. The combined effects including self-phase modulation, cross-phase modulation and Guoy shift cause the TH emission to be partially absorbed by the fundamental pulse. As a result, the energy conversion efficiency of TH emission decreases.

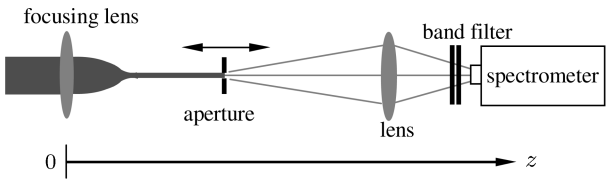


Fig.2. FIG1 Schematic of the experimental setup.

In early experiments,[3] the process of TH emission in the propagation of femtosecond pulse was investigated under different focusing conditions. The experimental setup is shown in Fig.2. The laser system is a Ti:sapphire chirp-pulse amplification system (JG-II). In the experiments, the maximum pulse energy was about 28mJ and the pulse duration 30fs. The pulses were focused by different lenses (f = 25, 40, 60, 80, 100 cm). It was found that the energy conversion efficiency of the TH emission was maximized when the focal length of the lens reached a proper value. In order to study the underlying mechanism in the process of TH emission in air, we carry out the simulation under similar conditions. In the simulation, the input IR pulse is taken to be a standard Gaussian shape and focused by lenses with different focal lengths. Because of the computational limitation, we do not simulate the beam with a centimetre waist. In our simulation, the femtosecond laser beam exhibits a waist of 3mm (FWHM); correspondingly, the input energy is $E_{\rm in} = 0.28$ mJ. Figure 3 shows the comparison between simulation and experiment[3] about the maximal energy conversions of the TH emission which are achieved by the nonlinear propagation of the fundamental pulse in air under different initial focusing conditions. The dotted line represents the simulation results and the solid line represents the experimental results.[3] The simulation results show that, under a certain focusing condition, the conversion efficiency of

the TH emission reaches its maximum after the start of filamentation during the nonlinear propagation.

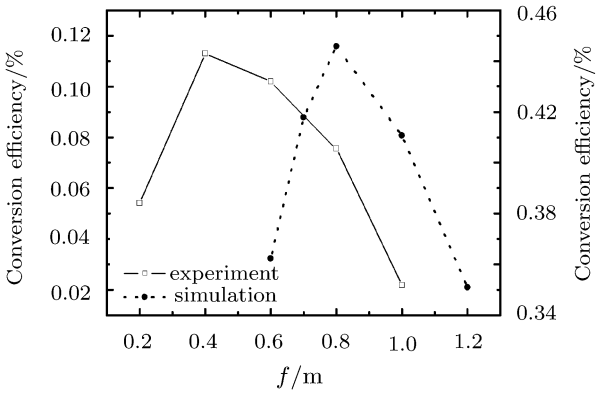


Fig.3. The conversion efficiency of the TH emission versus the focal length of the lens. The solid line denotes the experimental result (left-hand scale) and the dotted line denotes the simulation result (right-hand scale).

The interesting feature is that the maximum conversion efficiency of TH varies with the focal length of the lens, and the optimum focusing condition for the TH conversion does exist as shown in Ref.[3]. Comparing the simulation with the experiment, we find that the point of optimum condition is different with different initial condition, but the similarity is the tendency that it is beneficial to the TH emission by use of a moderate focal length of the lens in a range neither too short nor too long. In the simulation results, two colour filaments are formed on axis, and the energy of the TH emission is mainly concentrated into its filament resulting from the on-axis phase match.[7] If we do not take account of the intensity difference in the transverse plane of the filament, then the larger the on-axis fluence of the TH emission, the higher the energy of the TH emission. Figure 4 shows the on-axis intensity profile of the fundamental pulse (a) and the TH pulse (b) at the propagation distance where the maximum energy conversion efficiency is achieved in the channel, and the area under the curve gives the energy fluence on axis. Under different focusing conditions the reshape of the pulse is different in the nonlinear propagation.[13] We can see that when the initial focal length varies from 0.6 to 0.8 m, the longer the focal length, the higher the intensity of fundamental pulse in the channel, and the greater the conversion efficiency of TH emission correspondingly. As the focal length increases (f = 1.0 m, 1.2 m), the temporal splitting occurs at the profile of fundamental pulse. The interplay between the group-velocity dispersion and self-focusing leads to the pulse splitting which arrests the increase of intensity.[13] The decrease of the intensity at the centre of the pulse results in the decrease of electron density generated by ionization, as compared with the case of f=0.8m. Because the combined effects of self-focusing and electron generation play an important role in the phase matching condition,[7] the on-axis phase matching condition cannot be achieved, as in the case of f = 0.8 m. The asymmetric splitting profile appears in the TH pulse. One peak occurs at the leading pulse and a plateau is kept in the tail edge. From Fig.4(b), we can see that the in the case of f = 0.8 m, the energy fluence is maximum as compared with the other cases. Thus, the conversion efficiency of the TH emission is maximum.

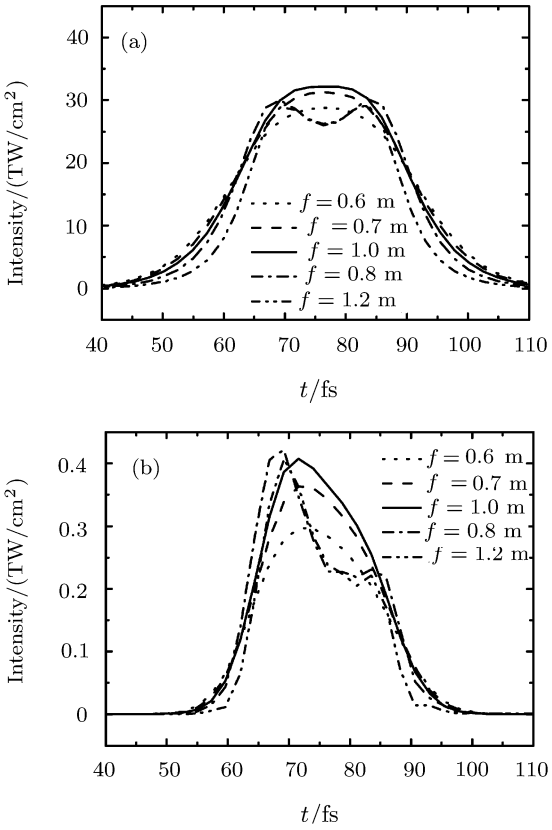


Fig.4. The on-axis temporal profile of the fundamental (a) and the TH (b) pulses.

4. Conclusion

In conclusion, we have presented simulations on the characteristics of TH emission in the propagation of femtosecond laser pulse in air by numerically solving the coupled nonlinear Schrödinger equations. In the simulation results, the strong third harmonic emis-

sion is observed with a conversion efficiency as high as 0.43%. The on-axis phase difference between the fundamental pulse and the TH pulse maintains a constant π except two cases, as is in good agreement with the phase-locking mechanism. It is argued that the phase shift of the fundamental pulse may have close correlation with the decrease of TH conversion efficiency, when the beam crosses the focal plane. In addition, the simulations performed under different focusing conditions confirm the existence of optimum focusing condition for the TH conversion as shown in experiment.

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