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Third-order harmonic generation by self-guided femtosecond pulses in air

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Strong third-order harmonic (TH) emission is observed with a conversion efficiency higher than 10^{-3} from a plasma channel formed by self-guided femtosecond laser pulses propagating in air. The main characteristics of TH emission in various conditions and the phase-matching condition between the fundamental and the TH wave are investigated. An optimized condition is found, under which the TH conversion efficiency is maximized. Our experimental results show that radiation of the emission in ultraviolet wavelength range makes a major attribution to TH emission, whereas the effects of self-phase modulation are not important when intense laser pulses interact with gaseous media.

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I. INTRODUCTION

The propagation of intense laser pulses in transparent matter causes strong nonlinear effects such as self-focusing [1,2], self-phase modulation (SPM) [3–5], four-wave mixing [6–9], and stimulated Raman processes. Propagation of an intense laser pulse in air will lead to self-focusing due to the Kerr effect. The mechanism for femtosecond laser pulses propagating over long distance in air is the balance between the Kerr self-focusing due to the nonlinear effects in air and defocusing due to the tunneling ionization and the diffraction of the laser beam [10–12]. Recently, much attention has been paid to harmonic generation by the self-guided femtosecond pulses in gaseous media. The dynamic equilibrium between the self-focusing due to nonlinear intensity-dependent refractive index and defocusing due to plasmas in the channel can support a long plasma channel in the interaction of the intense laser pulses and gaseous media. This is beneficial to get high harmonic conversion efficiency in the interaction, because the conversion efficiency between the fundamental wave and harmonic wave does not only depend on laser intensity, but also on interaction length. Usually, tight focusing is used to gain higher laser intensity in the interaction region, but it also leads to very short interaction length. Moreover, the ionization of gas always prevents high laser intensity due to defocusing. Therefore extending the interaction length is a promising way to get high conversion efficiency. We can get very long interaction length because of self-guiding when intense laser pulses interact with gaseous media. Theoretical and experimental results [16–21] have demonstrated that a femtosecond laser pulse can propagate many Rayleigh lengths due to self-guiding. The third-order harmonic (TH) generation in gaseous media has been studied for many years. Fedotov *et al.* [22] have studied the effects of the temporal and spatial self-action of light in atmospheric air. Marcus *et al.* [23] and Zhu *et al.* [24] tightly focused a femtosecond laser beam in atmospheric pressure methane and studied the TH generation process. In the process of ultrashort laser pulse propagation in air, the radiation of UV waves is considered as a result of the self-phase modulation, which leads to supercontinuum spectra expanding to UV waves when the ultrashort laser pulse propagates very long distance in air. However, the physics mechanism of TH generation in air is still not well understood.

In this paper, we will exploit the relationship between the TH generation and self-guiding propagation of intense laser pulses in air. It is demonstrated that the energy conversion efficiency of the fundamental wave to TH wave is very high, with a maximum efficiency up to 1.2×10^{-3}, because the interaction length is largely prolonged due to the self-guiding propagation of an ultrashort laser pulse in air. Our experimental results show that the emission of UV waves should be mainly responsible for the TH emission generation rather than self-phase modulation, when ultraintense laser pulses interact with air.

In this paper, we will exploit the relationship between the TH generation and the self-guiding propagation of ultraintense laser pulses in air. It is demonstrated that the energy conversion efficiency of the fundamental wave to TH wave is very high, with maximum efficiency reaching 1.2×10^{-3}, because the interaction length is largely prolonged due to the self-guiding propagation of ultrashort laser pulse in air. In earlier publications, in the process of ultrashort laser pulse propagation in air, the radiation of UV wave is considered as a result of the self-phase modulation, which leads to supercontinuum spectra expanding to UV waves when the ultrashort laser pulse propagates very long distance in air. Our experimental results show that the emission of UV waves should be attributed to TH rather than self-phase modulation, when the ultraintense laser pulse interacts with air.

II. EXPERIMENTAL SETUP

Figure 1 is a schematic diagram of the experimental setup. The laser system is a Ti:sapphire chirped-pulse amplification

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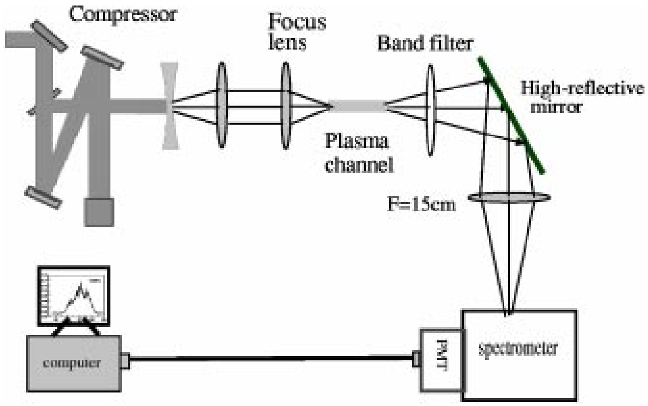

FIG. 1. A schematic representation of the experimental setup.

system (JG-II), which can provide up to 640 mJ energy, in 30 fs pulses, at a central wavelength of 800 nm. To prevent defocusing effects due to high intensity, in our experiment the maximum pulse energy used is about 28 mJ. In order to reduce the necessary distance before the formation of laser plasma channel, femtosecond laser pulses are slightly focused with different positive lens ($f=25,40,60,80,100$ cm) in air. The optical breakdown can be clearly observed in air with the appearance of the spark at the focus of the lens. In experiment, we observe that ultrashort laser pulses propagate many Rayleigh lengths, forming a long plasma channel. Generally, the length of the channel varies with the focal length of the focusing lens. A band filter is used to suppress the fundamental signal, which propagates in the same direction. The TH wave is collected into a photomultiplier tube with a $f=25$ cm quartz lens, and the TH spectra are detected with a spectrometer and processed by a computer. In the experiment, the characteristics of the spectra of the TH wave are studied in various conditions. We find that there exists an optimum condition, under which maximum conversion efficiency from the fundamental wave into the TH emission can be obtained.

III. RESULTS AND DISCUSSION

In order to study the characteristics of the TH emission in various conditions, we measure the spectrum of the fundamental wave emission in air from the position where filaments are formed by focusing the laser beam. The broadened spectrum of the fundamental wave due to SPM is observed as shown in Fig. 2. The solid and dotted curves represent the spectral profile with and without an $f=25$ cm lens, respectively. It can be seen that the fundamental spectra are greatly braodened because of the formation of the plasma filament channel in the air. This phenomenon can be attributed to self-phase modulation, which can greatly broaden the spectral component. The profile of the broadened spectrum is asymmetric and exhibits a blue shift. The broadening at short wavelength is more obvious than that at long wavelength. This is very consistent with theory and experiments [5].

After the filaments, very strong TH emission can be observed by applying a band filter, which can filter out fundamental waves. The TH spectrum is shown in Fig. 3. The peak

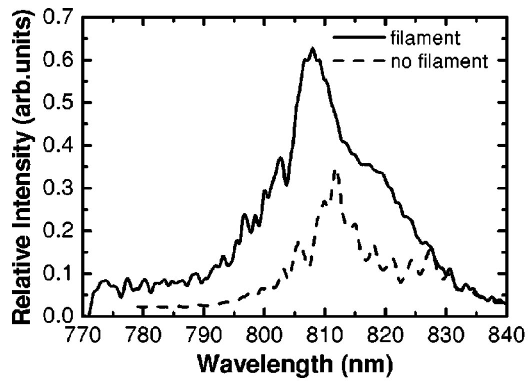

FIG. 2. The spectrum of the fundamental wave with and without lens.

of the TH spectrum occurs at 274 nm, which is red-shifted relative to the three fold (270 nm) of the fundamental wavelength (810 nm). Moreover, the spectral profile of the TH is clearly oscillatory. This spectral modulation may simultaneously be caused by two mechanisms. One is that the TH emission is generated with the modulated fundamental light as in Fig. 2. The other one is that the TH itself may experience self-phase modulation [14].

As is well known, the phase matching is necessary for an efficient conversion of the fundamental wave to the TH emission. The phase matching condition requires [13]

$$n(\omega)=n(3\omega). \quad (1)$$

Usually, air is a homogeneous medium and has normal dispersion. So it is very difficult to satisfy this condition. But when intense laser pulses form plasma channel in the air, this condition is easily satisfied. Since the fundamental wave intensity is much stronger than that of the TH emission, the intensity-dependent refractive index due to the Kerr effect in fundamental waves is much larger than that in the TH emission. Hence, Eq. (1) can be written as

$$n_0(\omega)+n_2I(\omega)-n_e/2n_c(\omega)=n_0(3\omega)-n_e/2n_c(3\omega), \quad (2)$$

where n_0 is the refractive index of the air, and n_2 is the Kerr nonlinear coefficient, n_e is the electron density. Here we

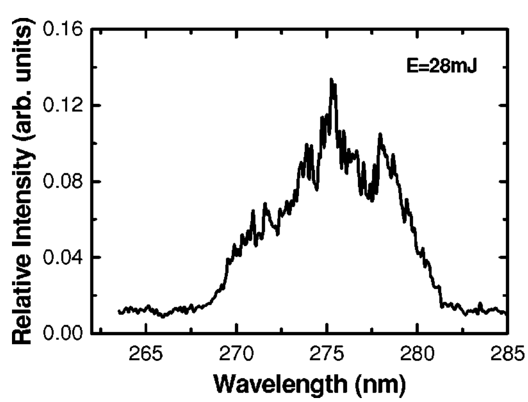

FIG. 3. The spectra of the TH with a fundamental wave laser energy of $E=28$ mJ and a lens with a focal length of $f=40$ cm.

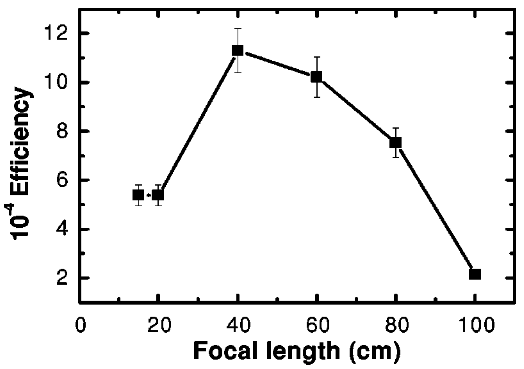

FIG. 4. The conversion efficiency between the fundamental wave and the TH versus focal length of the lens.

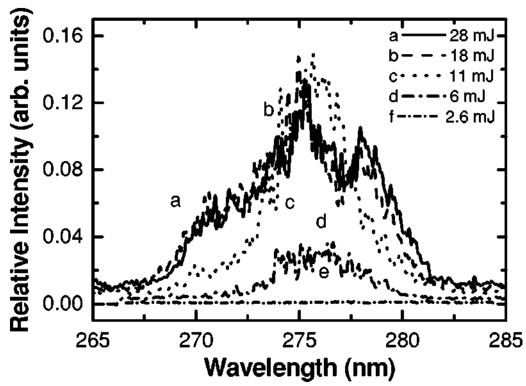

FIG. 5. The spectrum of the TH versus focal length of the lens.

have neglected the intensity-dependent refractive index arising from the TH emission, because the intensity of the TH emission is very small. Considering the electron density in plasma channel is about 10^{16}–10^{17}/cm^{-3} as measured in Ref. [11], we can get phase-matching laser intensity of about 10^{13}–10^{14} W/cm^2, which is consistent with the laser intensity required for self-guiding in air [11,21]. Generally, the output intensity of the TH emission is about [15]

$$I_{3\omega}(Z)=\frac{(3\omega)^2}{n_{3\omega}n_{\omega}^3\varepsilon_0 c^4}I_{\omega}^3 l_{eff}^2, \tag{3}$$

where $l_{eff}=\chi_{eff}Z(\chi_{eff}=\hat{e}_{3\omega}\chi^{(3)}:\hat{e}_{\omega}\hat{e}_{\omega}\hat{e}_{\omega})$ is the effective interaction length, which represents the interaction length in which the phase-matching condition is maintained. From Eq. (3), the conversion efficiency is proportional to the product of the fundamental wave intensity and the effective length.

In our experiment, the output energy of the TH emission versus focal length of the lens is shown in Fig. 4. The highest conversion efficiency is larger than 1.2×10^{-3}. It is interesting to note that the conversion efficiency to the TH emission is maximized when the focal length of the lens is about 40–60 cm. From Eq. (3), we can explain the existence of this optimum focal length corresponding to the maximum conversion efficiency, because the output power of the TH emission is proportional to the laser intensity and the effective interaction length l_{eff}. So for a shorter (long) focal length, the laser intensity is higher (lower) in the channel, but l_{eff} will be shorter (longer). Thus, there exists an optimum focal length, with which the product of the laser intensity with effective interaction length will be maximized. On the other hand, for a shorter focal length, the conversion efficiency will be reduced due to ionization, which consumes a lot of fundamental wave energy.

With a fixed focal length of the lens ($f=40$ cm), the TH spectral profile is plotted versus various pulse energies in Fig. 5. For higher energy, the spectral width of the TH is almost constant and the spectral profile of the TH is similar to that of the fundamental wave as shown in Fig. 5 (curves and a and b). For lower energy, the TH spectrum becomes narrower as shown in Fig. 5 (curves c and d). With increasing energy, the oscillation of the TH spectra becomes more serious. This can be explained as follows. For higher energy of the fundamental wave, the self-phase modulation covers more spectral components and causes more serious oscillations in the fundamental wave. Thus, the higher fundamental wave energy results in stronger modulation and wider spectra in the TH emission. When the laser energy at the fundamental wave is less than 2.6 mJ in our experiment, the TH emission is not observable. At this energy, the corresponding laser intensity can be estimated to be 2.5×10^{13} W/cm^2 from the focus spot size. This fundamental laser intensity may be considered as threshold for generating TH emission in air. We can evaluate the approximate threshold of the laser intensity required for generation of the TH in theory. The ionization term in Eq. (2) can be omitted at threshold laser intensity, so the laser threshold intensity is

$$I(\omega)=[n_0(3\omega)-n_0(\omega)/n_2]\approx 2\times10^{13}\ \text{W/cm}^2. \tag{4}$$

This is almost the same as the direct estimate of the experimental value.

For a fixed energy ($E=28$ mJ) of laser pulse, the measured TH spectral profile versus various focal lengths of the lens is shown in Fig. 6. With increasing focal length, the TH spectra are red shifted, which is largest at $f=60$ cm as shown by curve C in Fig. 6. When the focal length is longer than this focal length, the TH spectra become blue shifted. The reason for this phenomenon may be self-phase modulation. That is to say, relative red shift or blue shift may be decided by product of the laser intensity and effective interaction length, because the product of laser pulse intensity

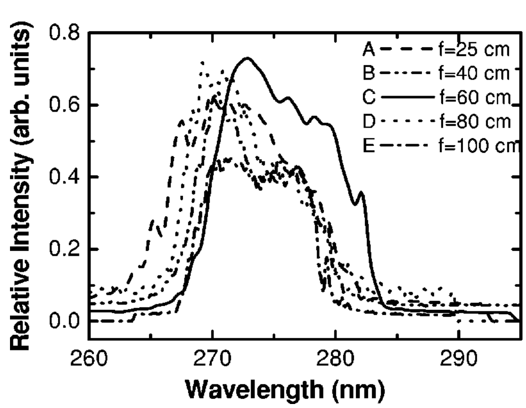

FIG. 6. The spectrum of the TH versus laser energy.

with effective interaction length is maximum at around $f = 60$ cm, which corresponds to the maximum conversion efficiency 1.2×10^{-3} [25–27].

IV. CONCLUSION

Because of the self-guiding of the ultrashort laser pulses in air, the interaction length between the ultrashort laser pulse and air is greatly elongated. During the interaction of the ultrashort laser with air, very strong TH emission is observed, with a maximum conversion efficiency higher than 10^{-3}. At the same time, the characteristics of the TH spectra are investigated, for different focal lengths and different energies of the laser pulse. We find that there exists an optimum focal length of the lens for the conversion efficiency to the TH emission for a constant input fundamental energy. This means that there is an optimum product of the laser intensity with interaction length. Experimental results show that the radiation of UV waves in supercontinuum should attribute to harmonic generation instead of self-phase modulation when the intense laser pulses interact with gaseous media. Moreover, our experimental results not only show that the supercontinuum, which is observed during the propagation of the ultrashort laser pulse in air, originated from SPM or stimulated Raman processes, but also demonstrate that the third-order harmonic contributes to the supercontinuum generation.

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