Long-range third-harmonic generation in air using ultrashort intense laser pulses

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The filamentation process taking place during the propagation of the ultrashort and intense laser pulse in atmosphere has stimulated a lot of research interest because of many promising applications such as remote sensing1–3 and lightning discharge control.4–6 The mechanism for the femtosecond laser pulses propagating over long distance in optical medium is governed by the dynamic interplay between the self-focusing due to the Kerr effect and the defocusing from low-density plasma induced by multiphoton/tunnel ionization. The defocusing effect of the plasma balances the self-focusing effect and leads to a limited beam diameter at the self-focal region and hence a limited peak intensity. This is known as intensity clamping.7–9

Recently, theoretical and experimental results using a 1 m focal length lens have demonstrated that during femtosecond laser-induced filamentation in air an ultrashort third-harmonic pulse is generated forming a two-colored filament.15 This two-colored filamentation phenomenon is due to a nonlinear intensity-dependent phase locking between the fundamental and the third-harmonic pulses which could extend the phase-matching condition over longer propagation distance.

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The filament formation from intense femtosecond Ti:sapphire laser pulse (centered around 800 nm) has been observed as far as 2 km in the atmosphere.16 From the application’s point of view, the generation of high intensity third-harmonic pulse over long range in air opens exciting perspectives such as remote sensing using the Lidar technique.2,3 In particular, the strong interaction between the harmonic pulse over long range in air opens exciting applications for atmospheric remote sensing of pollutants and bioaerosols. © 2005 American Institute of Physics. [DOI: 10.1063/1.2033148]
FIG. 1. Wave forms of the backscattered nitrogen fluorescence (dashed line) and third-harmonic (solid line) detected as a function of the propagation distance. The effective focal length \( F \) of the sending telescope was (a) \( F = 40 \) m, (b) \( F \rightarrow \infty \) and the effective focal length \( F \) for the initially divergent laser beam was (c) \( F = -150 \) m and (d) \( F = -75 \) m. The nitrogen fluorescence signal and the backscattered third-harmonic signal were averaged over 300 and 100 laser shots, respectively.

The receiver for the backscattered third-harmonic signal consisted of a second photomultiplier tube with 1 ns response time and a collecting lens made of fused silica (diameter of 50 mm). Two dielectric mirrors with high reflectivity centered at 800 nm with a 100 nm bandwidth and a 4-mm-thick UG11 absorption filter were put in front of the photomultiplier tube to filter out the scattered laser light and transmit the nitrogen fluorescence bands. The spatial resolution of the backscattered nitrogen fluorescence signal is limited by the lifetime of the nitrogen fluorescence which is around 1–2 ns, i.e., 30–60 cm along the propagation axis.

In Figs. 1(a)–1(d), the experimental results of the backscattered nitrogen fluorescence (dash line) and third-harmonic (solid line) are shown using the 45 mJ laser pulses with different divergences. The nitrogen fluorescence and the backscattered third-harmonic signals were averaged over 300 and 100 laser shots, respectively. The double peaks wave forms from the nitrogen fluorescence in Fig. 1 are due to multiple filaments and were confirmed by the observation of two hot spots on a burn paper. Multiple filaments occurred in the experiment due to inhomogeneous intensity distribution in the transverse cross section of the laser pulse. The backscattered third-harmonic wave forms show strong peaks accompanied by weaker ones. The less disturbed wave forms of the nitrogen fluorescence result from the higher signal-to-noise ratio of the backscattered signal due to a larger collecting area of the detector. As shown in Figs. 1(c) and 1(d), the self-induced plasma filaments are observed at further distances up to more than 35 m using initially a negatively chirped pulse and larger diverging laser beam. For all initial convergence or divergence of the laser beam, the backscattered third-harmonic signals in Fig. 1 are detected at the same position as the backscattered nitrogen fluorescence and both signals have similar lengths. The relative amplitudes of the backscattered third-harmonic signals as a function of the distance are also comparable to the nitrogen fluorescence wave forms.

Along the propagation path in air, the duration of negatively chirped laser pulse shortens due to the compensation through the positive group velocity dispersion in air and its peak power increases until self-focusing and filamentation start. This precompensation by the atmospheric group velocity dispersion and the initial convergence or divergence of the laser beam allows us to control the position of the filament in air. In our current case, the distance is more than 35 m. This distance, in principle, can be extended significantly by changing the laser parameters. During the filamentation process, the self-phase locking mechanism between the fundamental pulse and the third-harmonic pulse is dependent on the plasma density and the intensities of both the fundamental and third-harmonic pulses. This implies and is confirmed in this work (see later) that the third-harmonic pulse is generated in the filament’s volume. In the filament, the density of free electrons grows during filamentation and scatters the trailing edge of both the third-harmonic and the fundamental pulses. The rise of the nitrogen fluorescence signal in air is directly related to the increase of the probability of ionization. Thus, the increase of plasma density implies an increase of the backscattered third-harmonic signal as shown in Fig. 1. The results show a perfect overlap between the backscattered third-harmonic signal and the nitrogen fluorescence wave forms. These experimental results convince us that the backscattered third-harmonic signal is generated mainly inside the volume of the self-induced weak plasma filaments and are direct confirmation of the cofilamentation of the fundamental and third-harmonic pulses.

Recently, it had been demonstrated that the third-harmonic pulse generated in air is strongly deformed in space and time similar to the deformation of the fundamental pulse through self-phase modulation, self-steepening, and cross-phase modulation. The resulting spectrum of the third harmonic is significantly broadened and merged into the blue side of the supercontinuum of the fundamental pulse resulting in a continuous broad spectral band extending into the ultraviolet down to 230 nm. However, the broad backscattered supercontinuum becomes fully developed only at the end of the plasma column; this is the result of the distance-cumulative effects of self-phase modulation and self-steepening of the fundamental laser pulse and the cross-
phase modulation with the third-harmonic pulse. Therefore, throughout the whole filament, the backward-emitted nitrogen fluorescence is clearly resolved from the backscattered continuum of both the third-harmonic and the fundamental pulses. During the cofilamentation in air, the peak intensity of the third-harmonic pulse reaches about $10^{12}$ W/cm$^2$. Based upon our current results, it is evident that intense third-harmonic pulses could be generated wherever the filament of the fundamental reaches; recent reports indicates that the filament can reach a few kilometers in the atmosphere. This finding, coupled to the effective broad spectral content of the third harmonic, would allow potential applications in remote sensing of atmospheric pollutants and bioaerosols through ultraviolet (third-harmonic) induced-fluorescence.

Experimental results confirm the possibility to control third-harmonic generation in air over long propagation distance using divergent and negatively chirped fundamental pulse. The backscattered third-harmonic signal originated from the self-induced plasma filaments and was detected remotely using a Lidar technique. The cofilamentation of the high intensity fundamental and third-harmonic pulses over long distance show promising applications for atmospheric remote sensing.

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