Noncollinear interaction of femtosecond filaments with enhanced third harmonic generation in air

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We demonstrate that the interaction between two intense femtosecond filaments noncollinearly crossed in air results in third harmonic generation of more than two orders enhancement in energy conversion as compared with that from a single filament. The dependences of the third harmonic generation on the noncollinear crossing angles, input intensity ratios, and input pulse polarizations are investigated. © *2009 American Institute of Physics*. [doi:10.1063/1.3227658]

The propagation of ultrashort intense laser pulse in air was demonstrated to induce a self-guided filamentation channel[1] and facilitates many research interests, such as pulse self-compression,[2] spatial self-filtering and intensity clamping with self-stabilized high intensity in the core,[3] supercontinuum generation,[4] remote sensing,[5] white-light lidar,[6] and terahertz emission.[7] Recently, there has been an ever growing interest in the study of interaction between two or more light filaments since it exhibits distinctive features, such as beam fusion,[8] repulsion, fission,[9] and spiraling.[10] In addition, the spectrum of the probe pulse was demonstrated to be tunable by using the interaction of two beams,[11] which was also used to controllably trap and destruct the intense optical filament as well as its sequential propagation dynamics.[12] These new characteristics give rise to a lot of possibilities for potential applications by using such nonlinear interactions instead of a single filament. It was demonstrated that the nonlinear frequency conversion, such as third harmonic (TH) generation,[13] could be ensured efficiently by intense filaments in air. For efficient TH generation, it is important to have the phases of the generated TH and the fundamental wave (FW) to be locked each other over a long propagation distance so as to defeat the energy backconversion, which was demonstrated achievable by intense filaments.

In this paper, we use the strong interaction of two noncollinear filaments to induce an efficient TH generation in air with more than two orders of pulse energy enhancement as compared with that from a single filament, whose dependences on the noncollinear crossing angles, intensity ratios, and relative polarizations between the input pulses are studied.

Our experiments were carried out with the output from an amplified Ti:sapphire laser system (50 fs, 800 nm, 1 kHz), which was split into pump and probe pulses with pulse energy of ~0.85 mJ for each one. By using two concave mirrors ($f = 100$ cm), both the pump and probe pulses were focused to produce two noncollinearly crossed filaments in air. The length of the individual filament was measured to be ~5 cm. The delay between these two pulses was tuned with a motorized translation stage in the pump arm, whose field polarization could be rotated by using a half-wave plate. The positive or negative delay accounts for the probe or pump pulse was ahead with respect to the other one, respectively. At the end of the probe filament, two fused silica prisms were used to separate the generated TH pulse from the FW pulse and then measured by a photomultiplier tube (PMT), vice versa for the pump, and we focused on the probe here. The spatial overlapping between these two noncollinear pulses around the crossing point was checked by imaging the optical fields to a charge-coupled device (CCD), and the zero delay was determined by detecting the maximum TH generation with a PMT.

Interaction between these two noncollinear filaments was observed as the pump and probe pulses were temporally synchronized. Constructive and destructive interference between the incident pulses of the same polarizations could produce periodic distribution of local intensity in the overlapped region, leading to the corresponding changes of the local refractive index owing to the Kerr effect.[14] We monitored periodic structures induced by filament interaction with a thin plate inserted nearby the crossing point at a grazing angle to reflect part of optical fields, and directly recorded the intensity distributions with a CCD after an optical imaging system. The observed intensity spatial distribution was dependent upon the crossing angles and incident intensity ratios as shown in Fig. 1. The period of the interference

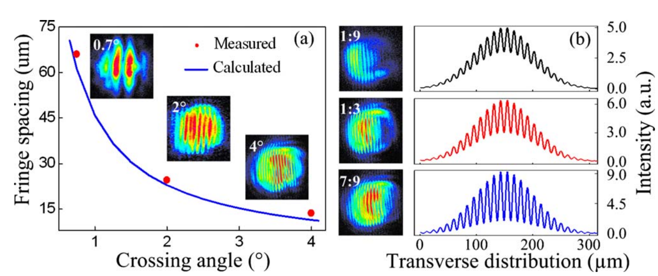

FIG. 1. (Color online) (a) The transverse fringe periods and the spatial distributions (insets) of the noncollinearly interacted filaments at various crossing angles. (b) The transverse distributions of the noncollinearly interacted filaments at different intensity ratios of the pump and probe pulses and the simulated intensity modulations.

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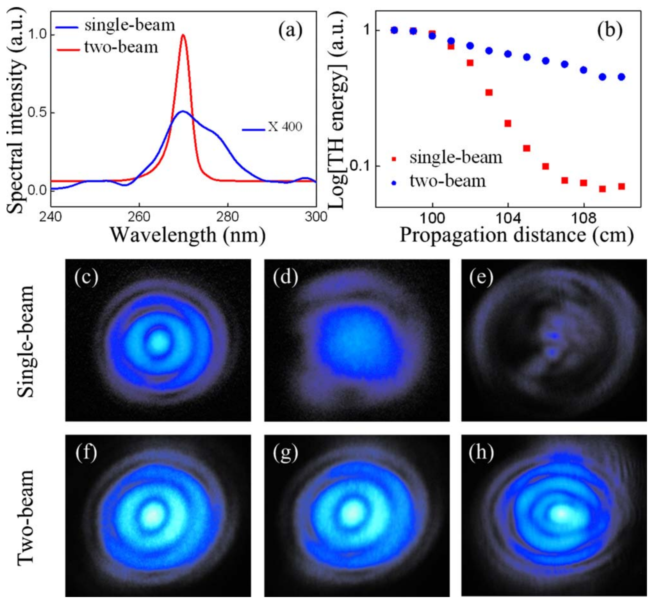

FIG. 2. (Color online) (a) The measured spectral profile of the generated TH field. (b) The TH energy vs the propagation distance. [(c)–(h)] The spatial distributions of the TH field at propagation distances of 100, 102, and 120 cm. The pump and probe filaments were crossed with an angle of 3°.

pattern varied with the crossing angles as $\lambda/2\sin(\alpha/2)$ [shown in Fig. 1(a)], where λ is the central wavelength and α is the crossing angle of the noncollinear input beams. As the relative intensities of the pump and probe pulses (defined respectively as I_{pump} and I_{probe}) varied, the modulation depth of the fields inside the overlapped region changed, as shown in Fig. 1(b) for $I_{pump}/I_{probe}=1:9$, 1:3, and 7:9.

Accompanied with the filament interaction, a significant enhancement of the TH generation was observed along the FW propagation direction. The enhancement of the TH generation through filament interaction referred to the observed increase of the TH energy generated from the probe filament by interacting with the pump filament as compared to the case of a single probe filament (without pump). As shown in Fig. 2(a), the spectral intensity of the generated TH field was enhanced by a factor of 770 for the peak frequency component as compared with the case of a single filament. A detailed evolution of the generated TH at various propagation distances after the concave focusing mirror were shown in Fig. 2(b) and Figs. 2(c)–2(h), respectively, for the pulse energy measured with a PMT and the spatial distributions recorded with a CCD camera.

For a single probe filament, as shown in Fig. 2(b), the TH energy was kept almost unchanged inside the filament (from 98 to 100 cm) due to its nonlinear phase locking with the FW filament.[13] Multiring structures of TH were observed during the filamentation as shown in Fig. 2(c), where the divergent angles of the first, second, and third rings were measured to be 2.25, 4.09, and 5.86 mrad, respectively. The TH generation in the filament could be divided into on- and off-axis parts, which were respectively generated from the FW filament core with nonlinearly modified refractive index and off-axis noncollinear phase matching between the FW and TH pulses.[15] The divergent angle of the off-axis TH was approximately 6 mrad,[16] which agreed with that of the third ring observed in our experiment. The first and second TH rings were possibly originated from Fraunhofer diffraction of the on-axis TH in the filament core. The Fraunhofer diffraction induced by a hole of the diameter D could be expressed as $I(\theta)\sim J_1(\pi D\theta/\lambda)^2/(\pi D\theta/\lambda)^2$, where J_1 is the first-order Bessel function, θ and λ are the divergent angle and wavelength of laser beam, respectively. By assuming a TH filament diameter of $D=170$ μm, the divergent angles of the first and second diffraction orders were estimated to be 2.26 and 4.20 mrad, respectively, which accounted for the first and second TH rings observed in our experiments. The filament ceased as the FW peak intensity decreased in the filamentation channel, which broke the counterbalance between the Kerr self-focusing and plasma defocusing. In this region, the TH decreased rapidly with the propagation distance, as shown in Fig. 2(b) (red square). The on-axis TH underwent free diffraction and the multiringed TH was eventually disappeared, as shown in Figs. 2(d) and 2(e).

In the presence of the temporally synchronized pump filament, as shown in Figs. 2(f)–2(h), the multiring profiles of the generated TH from the probe sustained for a much longer propagation distance as compared to that of a single filament, indicating significant elongation of the filamentation dynamics of the generated TH pulse with self-confined beam diameter. It was consistent with the observed TH enhancement shown in Fig. 2(b) (blue circle) with filament interaction. For the propagation distance from 98 to 100 cm, the TH generation was almost the same for the cases with and without pump pulse, indicating that the observed enhancement of the TH generation was originated from the arrested energy backconversion from the generated TH to the FW pulses with filament interaction, which could be ascribed to the nonlinear phase locking of the generated TH with the FW pulse inside the elongated filament. Such filament elongation could be caused by the experienced spatiotemporal modulation of the pulses during the filament interaction. It has been demonstrated that the filamentation could be promoted by tuning the spatiotemporal profiles (chirp or beam diameter) of the incident laser pulses.[17,18]

In order to systematically investigate the noncollinear filament interaction that enhanced TH generation from the probe filament, we then tuned the crossing angles, relative polarizations, and intensity ratios of the pump and probe pulses. The TH generation based on the interaction between the pump and probe filaments were closely dependent on the changes of the pump pulse, which led to a control of the probe filament and therefore the enhanced TH generation through the filament interaction. Figure 3(a) shows the measured TH generation from the probe filament when the field polarization of the pump pulse was rotated with respect to the probe one. Obviously, the maximum and minimum TH energy were observed as their polarizations were, respectively, parallel and orthogonal to each other, corresponding to the cases with and without interference in the overlapped region, as typically shown in the insets of Fig. 3(a) for the spatial intensity distribution for different polarizations. The parallel polarized pump and probe filaments with local field intensity modulation inside the overlapping region were demonstrated to induce the enhanced TH in the far-field region. However, as shown in Fig. 2(b), the TH generation inside the overlapping region was almost the same when the pump pulse was tuned on or off. The observed TH enhancement was mainly caused by the decreased energy backconversion of the generated TH to the FW pulses along their propagations. It was the filament interaction with

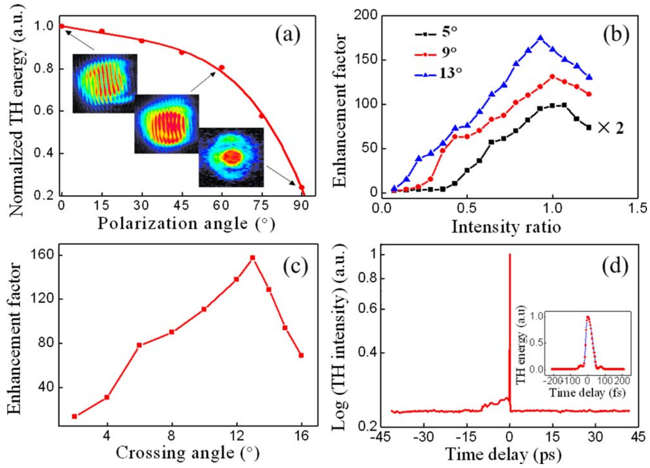

FIG. 3. (Color online) The dependences of the TH generation on (a) the relative polarization, (b) intensity ratio, (c) crossing angle, and (d) time delay between the pump and probe pulses. The pump-to-probe intensity ratio was set as 1:1 for (a), (c), and (d), and the crossing angle was fixed at 13° for (a) and (d). The inset is the detailed evolution of TH energy as a function of time delay around zero delay.

interference-induced periodic structures inside the interaction zone that dominated the succeeding filamentation dynamics of the FW and TH pulses, which consequently resulted in the observed TH enhancement in the far-field region.

Such kind of filament interaction was expected to be closely dependent on the relative intensity ratios and crossing angles of the pump filament, which tuned the modulation depths and periods of the field fringes inside the interaction zone. As shown in Fig. 3(b) for various crossing angles, the enhancement of the TH generation from the probe filament increased gradually with the increase of the pump intensity, and reached its maximum as the intensity ratio between the pump and probe was close to one with a maximum modulation depth, which then decreased as the pump intensity was further increased. Here, we define the enhancement factor as $S_{\mathrm{TH\_two}}/S_{\mathrm{TH\_single}}$, where $S_{\mathrm{TH\_two}}$ and $S_{\mathrm{TH\_single}}$ stand for the measured energy of the generated TH pulse from the probe filament when the pump filament was turned on and off, respectively. Similarly, as shown in Fig. 3(c) for the intensity ratio of 1:1, the enhancement of the TH generation increased first and then decreased as the crossing angle increased from 2.0° to 16.0°, which accordingly tuned the periods of the field fringes inside the interaction zone. The TH generation reached the maximum enhancement factor of 174 at the crossing angle ~13°, which corresponded to significantly promoted filamentation dynamics of the FW and TH pulses as discussed above. Figure 3(d) shows the enhanced TH as a function of time delay between the pump and probe pulses. Except for the case where the pump and probe pulses were temporally synchronized within the pulse duration as shown in the inset of Fig. 3(d), the enhancement of the TH generation was still observed for about 10 ps after the extinction of the pump pulse, which indicated the influence of the ionization-induced plasma on the TH generation with a lifetime of several picoseconds.[19] It was consistent with the fact that the third-order nonlinearity $\chi^{(3)}$ of the gas could be enhanced as the existence of the laser-driven plasma generated by the pump pulse,[20] leading to the observed TH enhancement in our experiment as shown in Fig. 3(d) when the pump pulse was several ps ahead of the probe pulse.

In summary, we have demonstrated that the TH generation form the probe filament could be dramatically enhanced through the noncollinear filament interaction, which closely depended on the relative polarizations, crossing angles, and intensity ratios of the pump and probe pulses. This may serve as a promising approach to generate and control bright ultrashort ultraviolet radiations to monitor chemical pollutants and biological species in air.

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