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Abstract: Three orders of magnitude in the enhancement of the third-order harmonic (TH) generation induced by the interaction of two femtosecond filaments crossing with small angles in the air is achieved. The dependences of the TH generation on the time delay, the relative polarization, the input laser intensity ratios between the probe and pump beam are measured with the crossing angle of 3.5°, and the results with quasi-vertical crossing angle are also shown for comparison.

The measured spectra of TH generation without pump beam (black solid line), with pump beam with cross angle θ set to be 3.5° (red dash line), and quasi-vertical (blue dot line)

Enhancement of third-order harmonic generation by interaction of two IR femtosecond filaments

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1. Introduction

The field of femtosecond pulse filamentation in transparent media [1–29] was intensively investigated after the invention of chirped pulse amplification. One of the most interesting nonlinear phenomena occurring in the filament is harmonic generation, and the third-order harmonic (TH) generation has drawn considerable interest due to the most popular medium, i.e. air. Especially, third-order harmonic generation microscopy has been applied to probe laser plasma plumes from solid targets and recognized as a useful nonlinear imaging method to observe spatial inhomogeneities in almost all kind of materials. Femtosecond laser pulses propagating in neutral media undergo filamentation [8] with self-stabilized intensity clamping in self-guided channels owing to the dynamic balance between Kerr self-focusing and plasma defocusing, which induce plenty of nonlinear optical phenomena. In particular, it was reported that an efficient conversion (≈0.2%) from a femtosecond laser pulse to its TH has been achieved in air and noble gases [9,10].

Recently, the interaction between two or more filaments has attracted more attentions since it shows distinctive features [11–19], and the TH generation induced in femtosecond filaments can be increased by two orders of magnitude by adding a second pump filament to intercept the probe filament [13–19].

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The enhancement of TH generation was explained to be originated from an increase of the nonlinear coefficient correlated with the induction of the plasma named bulk effect or a neutral air-plasma interface effect [14,20–22]. Most recently, it was reported that the TH enhancement has been observed by the breaking of the large cancellation of TH in a filament perturbed by a pump pulse [17,18]. The nonlinear phase shift in the filament partially compensates for the destructive interference between the TH generated before and after geometrical focus caused by Gouy phase shift effect, leading to a weak TH generation. By truncating the probe filament with a pump filament, the influence of symmetrical Gouy phase shift could be weaken or eliminated, and the TH generation could be significantly enhanced. However, the experimental results about the enhancement of TH generation reported by various groups are conflicting in some aspects. Suntsov et al. [14,15] found a large TH enhancement caused by the local plasma formed by the interaction between two femtosecond filaments subsisting for more than forty picoseconds, which has no relation with the relative polarization. But Yang et al. [16] found that the TH enhancement only occurs when the two co-polarized laser pulses with small crossing angles overlap in time, and it is originated from the formation of plasma grating. Kosareva et al. [23] found that the interaction among multiple filaments enhances the clamped fundamental laser intensity, which may induce the increase of the TH yield of the probe filaments, due to the pump-probe interference effect. The understanding of the physical mechanism responsible for the TH enhancement is still not complete.

In this work, by using intense interaction between two non-collinear femtosecond filaments, named the probe filament and the pump filament, the TH generation induced in the probe filament is investigated. An efficient enhancement of TH generation about three orders of magnitude is achieved with small crossing angle between the two filaments, as compared with that from a single filament. With the crossing angle between the probe beam and the pump beam set to be $3.5^\circ$ or quasi-vertical, the dependences of TH enhancement factor on the time delay, the relative polarization and the input laser intensity ratios between the probe and the pump pulses are investigated.

2. Experimental setup

The experiments were conducted by using a Ti:Sapphire chirped-pulse amplification laser system operating at 1 kHz, delivering a 3.0 mJ pulse centered around 810 nm with pulse duration about 33 fs. The schematic of experimental setup is shown in Fig. 1. The fundamental laser beam was split into two arms, the pump and the probe, by a beamsplitter BS1. The pump beam with pulse energy up to 0.9 mJ was focused using a 60 cm-focal-length lens L1, and formed a femtosecond filament in the air with a length about 1.5 cm. The probe beam with pulse energy up to 0.6 mJ was focused using a 117 cm-focal-length lens L2 in order to obtain a longer filament with a length about 3 cm. M4 and L1 mounted on an automatic translation stage can be adjusted to let the probe filament cross the pump filament with a small angle. The time delay between the probe beam and the pump beam can be adjusted by a delay line denoted by Delay.

The plasma fluorescence of interaction area of the two femtosecond filaments was collected and projected onto a sensitive charge-coupled device (CCD) camera, so the interaction area can be investigated in time. For the measurements of TH spectrum, the TH generation was separated from the fundamental beam and reflected to a fused silica lens by two 266 nm high reflective mirrors, and then focused into a spectrometer (Ocean Optics, USB4000). The TH spectra obtained from the probe filament was recorded by the spectrometer placed at a distance about 55 cm away from the center of the interaction area. The relative TH intensity was normalized to the peak frequency component of TH spectra.

3. Experimental results and discussion

Accompanied with the interaction between the probe filament and the pump filament, a significant enhancement of TH generation can be observed along the probe beam propagation direction. First, the dependences of the TH generation on the time delay $\tau_d$, the relative polarization and the input laser intensity ratios between the probe and the pump beams, and the relative position that probe filament with $\theta$ set to be $3.5^\circ$ and quasi-vertical respectively. The enhancement factor in different conditions was defined as TH intensity divide by TH intensity generated.
from a single probe filament. The spectral intensity of TH generation was enhanced by a factor about 1040 with \( \theta \) set to be 3.5\(^\circ\) and only about 85 with \( \theta \) set to be quasi-vertical. Obviously, the TH generation has been significantly enhanced through truncating the probe filament by a pump filament. Note that there is a second-order peak appearing in the blue side originated from the self-phase modulation and the cross-phase modulation effect, which dominate the spectral broadening of TH generation to the UV regime.

The dependences of the TH enhancement factor on the time delay \( \tau_d \) are shown in Fig. 3a, while the dependences of the normalized TH intensity on the relative polarization between the probe and the pump are illustrated in Fig. 3b, with the cross angle \( \theta \) set to be 3.5\(^\circ\) and quasi-vertical respectively. The TH enhancement factor sharply grows with the increase of time delay \( \tau_d \), reaching a maximum at \( \tau_d = 250 \) fs, and then stabilizes at \( \tau_d = 500 \) fs, which is in good agreement with results in [14]. With longer time delay, the TH enhancement factor exhibits a slow monotonic decay well described by the function \( a(1 + b\tau_d)^{-1} \) with decay constant \( \beta \approx 24 \) fs\(^{-1}\), which agree with that measured in [24] for free electron density decay in a femtosecond filament. This result indicates that the enhancement of TH generation is correlated with the free-electron density in the interaction area. Plasma grating [25–28] can tolerate ultra-high-intensity laser field beyond the clamping intensity limit for free electron generation [23], and the local peak plasma density was enhanced up to about 2 orders of magnitude within a UV plasma grating [29]. The presence of the charged particles can effectively enhance the third-order nonlinear optical susceptibility, which is named bulk effect, which has no relation with the relative polarization. For the enhancement of the TH generation from the probe filament interacting with the pump filament, the bulk effect may play an important role.

For more information, the dependences of the normalized TH intensity on the relative polarization are shown in Fig. 3b, and the tendencies with two different cross angles are different with the previous results reported in [14] and [16]. It can be seen that the intensity of TH generation exhibits an exponential decay with the increasing relative polarization, and it decays more quickly with \( \theta \) set to be quasi-vertical. The interaction between the probe filament and the pump filament along the propagation direction, which overcome intensively the effect of Gouy phase shift, stops the energy of the TH signal flowing back to the fundamental pulse, and this effect has a relation with...
the relative polarization. Besides the bulk effect, this effect may also play an important role on the enhancement of TH generation by comparing the results obtained from the co-polarized and cross-polarized beams interaction. As is known, the intense interaction between two noncollinear filaments can induce the enhancement of the laser peak intensity of the interaction area due to the light filed interference [23], and the higher fundamental laser peak intensity can result in more intense nonlinear effects, such as self-modulation, self-steepening effect, cross-phase modulation effect and so on. In other words, the interaction between the two filaments results in the spatiotemporal modulation of the laser pulses in the interaction area [16], which can increase the length of the interaction area for the change of the relation between the self-focusing and self-defocusing. The length of interaction area, i.e., the length of phase locking between the fundamental pulse and generated TH, is effectively elongated when the crossing angle is very small, which induces the enhancement of the TH generation. Meanwhile, the bulk effect also has a relation with the interaction length [15], which induces the different TH enhancement factors obtained from co-polarized beams interacting with the two crossing angles.

Fig. 4a and Fig. 4b show the distributions of fluorescence of the interaction areas when the relative positions is 9.95 and 0.40 mm corresponding to the Fig. 4a and Fig. 4b,
respectively. Fig. 4c and Fig. 4d shows the dependences of normalized TH intensity on the relative position that the pump filament crossing the probe filament with $\theta$ set to be 3.5° and quasi-vertical. Fig. 4a shares the same abscissa as Fig. 4c, meanwhile Fig. 4b shares the same abscissa as Fig. 4d. From the comparison between the results obtained from the two crossing angles, the tendencies of TH generation changing with relative position are similar, but the effective lengths are different. The electron densities at the head and tail of pump filament are so low that when probe crossing with pump at this area, TH generation can only be enhanced in a short range with $\theta$ set to be quasi-vertical. However, plasma grating could bound free electron together in a much longer range with $\theta$ set to be 3.5°, so the enhancement of TH generation can be found in a region more than 10 mm. The pulse energy of pump beam is changed from 0.2 to 0.9 mJ. The dependence of the normalized TH intensity on the intensity ratios is shown Fig. 4e with $\theta$ set to be 3.5°. For intensity ratio in the range from 0.35 to 1.50, TH generation from the probe filament increases gradually with the increase of pulse energy of pump beam.

### 4. Conclusion

In summary, three orders of magnitude enhancement of TH generation from a probe filament truncated by a pump filament with 3.5° crossing angle is demonstrated. The dependence of TH generation on the relative polarization between the probe beam and the pump beam exhibits an exponential decay, which is firstly observed. Although the effect of overcoming the Gouy phase shift and the bulk effect may be contributed to the enhancement of the TH generation, the enhancement of the clamped fundamental laser intensity in the interaction area plays an important effect on the enhancement of the TH generation from the probe filament. The intensity of TH generation closely depends on the relative polarization, the relative position that the probe filament crossing the pump filament, and the intensity ratio.

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