Spectral modulation of third-harmonic generation by molecular alignment and preformed plasma*

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We demonstrate spectral modulation of third-harmonic generation from molecular alignment effects. The third harmonic spectrum is broadened or narrowed under different influences of cross-phase modulations originating from various molecular alignment revivals. Furthermore, the spectrum and spatial distribution of the generated third harmonic pulse change dramatically in the presence of a preformed plasma. Under the influence of a preformed plasma, a narrower third harmonic spectrum is observed, and the conical third-harmonic pulse increases while the axial part decreases. The investigation provides an effective method to modulate the spectral characteristic and spatial distribution of third-harmonic generation from intense femtosecond filament.

Keywords: third-harmonic generation, laser filament, molecular alignment

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1. Introduction

Third harmonic (TH) generation from intense laser filament in gas attracts much attention due to its simple operation and relatively high conversion efficiency up to $\sim 0.2\%$.[1–7] It can provide high-intensity ultraviolet coherent light source by means of group-phase-locking between the fundamental-wave and TH pulses. Many investigations have been made in order to increase the conversion efficiency and modulate the intensity of the generated TH pulse. The TH intensity is enhanced by several orders of magnitude as the filament is intercepted by a second intense infrared pulse.[8,9] The plasma grating induced by noncollinear interaction between two filaments has been proved to enhance the TH conversion dramatically.[10] In addition, the TH conversions both from single-color filament and from dual-color filament are modulated by rotational wavepackets.[11,12] Furthermore, the spectral and angular distributions of the TH generation from filaments in air have been studied in detail.[7] Compared with extensive research on TH intensity enhancement, investigation on TH spectral modulation is relatively scarce. As the frequency shift of the generated TH is important for the ultraviolet source, this would be of great significance for being used to modulate the ultraviolet spectrum.

In this paper, molecular alignment induced by the impulsive rotational Raman excitation is used to modulate the spectrum of TH generation from fundamental-wave (FW) femtosecond filament. As is well known, the characteristics of followed pulse filament were altered at different revivals of molecular pre-alignment.[13–17] Here, the spectrum of TH generation is broadened or narrowed while the FW pulse is tuned to match different molecular alignment revivals. In the experiment, a preformed plasma is also important to influence the spectral characteristic of the TH pulse. The results show that the conical TH generation increases remarkably while the axial part decreases under the influence of the preformed plasma. Meanwhile, the TH spectrum is narrowed and shows slight red-shift.

2. Experimental setup

The experimental setup is shown in Fig. 1. In the experiment, a pulse with a central wavelength at 800 nm, pulse duration of 45 fs, repetition rate of 1 kHz delivered from a Ti:Sapphire laser system was split into two beams. One beam passed through the β-BBO crystal to produce 400-nm pulse as the pump pulse to align molecules in atmospheric air. Another 800-nm beam named probe pulse was focused to generate TH pulse during filamentation, which passed through the pre-aligned molecules during its propagation. The polarization of the pump pulse was adjusted by inserting a half-wave plate at 400-nm. The pump and probe pulses were combined by a dichroic mirror after being focused by convex lenses

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($f_{400\ \mathrm{nm}} = 500$ mm, $f_{800\ \mathrm{nm}} = 800$ mm), respectively. The focuses of the two beams were spatially overlapped with each other. The energies of the pump and probe pulses were 0.29 mJ and 1.04 mJ, respectively. A step motor was used to precisely adjust the time delay between the pump and probe pulses. The residual pump and probe pulses were filtered by several mirrors with high reflectivity at 267 nm at a 45° incident angle. The spectral characteristics of the TH pulse from probe pulse affected by molecular alignment and preformed plasma were measured with a spectrometer (Ocean Optics HR 4000).

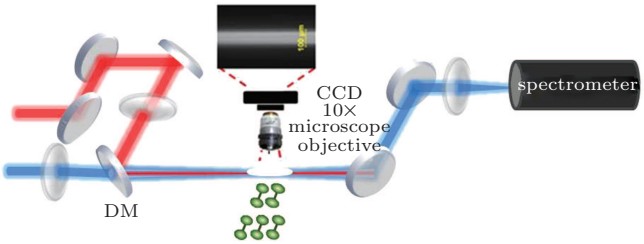


Fig. 1. (color online) Experimental setup. Blue path represents the pump pulse, which is focused to align molecules; red path denotes the probe pulse used to produce TH generation. DM: dichroic mirror, used to combine pump and probe pulse. Microscope objective and CCD are used to image and capture the laser filament. Spectrometer is used to measure the spectrum of TH generation.

3. Results

3.1. TH modulation by molecular alignment

Firstly, the influence on the spectral modulation of TH generation by molecular alignment is investigated. The pump pulse energy is reduced to a relatively low value so as to avoid any possible plasma influence. Accordingly, the TH generation from the probe pulse is affected only by the molecular alignment. The polarization of the pump pulse is adjusted to be parallel to that of the probe pulse. The time delay between pump and probe pulse is tuned by adjusting the step motor. The spectrum of the TH pulse is measured by the spectrometer. The modulations of spectral width and intensity of the TH pulse at different molecular alignment revivals are shown in Fig. 2(a). The results of TH intensity show the same trend variability as the molecular alignment revivals with parallel polarizations for the pump and probe pulses. According to the result, the intensity of the central wavelength is extracted to represent its change with molecular alignment revival. The experimental results in Fig. 2(b) show that the intensity of the central wavelength increases at the parallel molecular alignment revivals and decreases at the perpendicular alignment revivals. As reported in Ref. [18], the phase-matching condition is satisfied in a longer propagation distance in the filament due to the phase-locking between the FW and TH pulse, which leads to the high-efficiency generation of TH pulse from the FW filament. Also, the generated TH energy from the probe pulse filament is expressed as $I^{3\omega}(\tau) \propto |\chi^{(3)}(\tau)|^2$, where $I^{3\omega}$ is the TH intensity, and $\chi^{(3)}$ is the third-order nonlinear susceptibility of atmospheric air. In this experiment, the influence on the TH generation from the molecular alignment is mainly reflected in its effect on third-order nonlinear susceptibility. As reported in Ref. [19], $\chi^{(3)}$ is related to $\langle\langle\cos^2\theta(t)\rangle\rangle$ and $\langle\langle\cos^4\theta(t)\rangle\rangle$, where $\langle\langle\cos^2\theta(t)\rangle\rangle$ represents the alignment degree, $\langle\langle\cos^4\theta(t)\rangle\rangle$ is the high-order terms presenting alignment degree, and θ is the angle between prealigned molecular axis and the polarization of the followed pulse. The effect of $\langle\langle\cos^4\theta(t)\rangle\rangle$ can be ignored since it is one magnitude less than $\langle\langle\cos^2\theta(t)\rangle\rangle$. Therefore, third-order nonlinear susceptibility is mostly influenced by molecular alignment degree. The third-order nonlinear susceptibility increases when the prealigned molecular axis is parallel to the polarization of the followed pulse, resulting in the higher conversion of TH generation from followed pulse filament. In contrast, the TH generation decreases at the perpendicular alignment revivals since the third-order nonlinear susceptibility is reduced. The measured results of TH intensity in the experiment accord well with the alignment degree at different molecular alignment revivals shown in Fig. 2. Besides, as described in Refs. [20]–[22], remarkable enhancement of TH emission can be observed with the redistribution of laser energy during the reconstruction of the two-colored filament. Therefore, damage to the original phase-lock between fundamental and third-harmonic pulse partly leads to the enhancement of TH generation in our experiment.

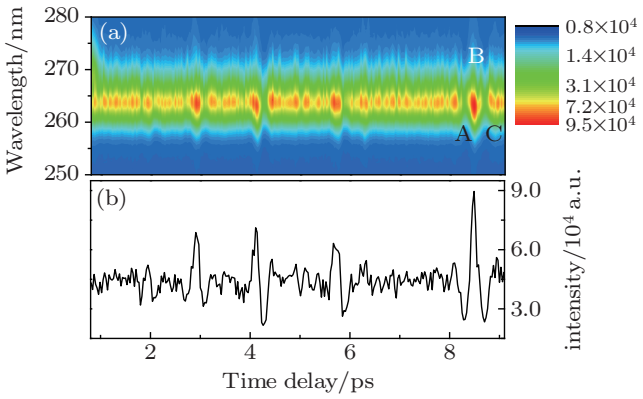


Fig. 2. (color online) (a) Modulation of TH intensity and spectrum at different molecular alignment revivals. (b) The intensity of TH central wavelength varying with the molecular alignment revival. The unit a.u. is short for arbitrary units.

After that, the spectrum of generated TH is measured for further analyzing its effect on the TH generation by molecular alignment. In the experiment, the output spectrum of TH generation is measured while the probe pulse is tuned to match various revival times. The spectral intensities here are normalized to analyze the spectral broadening or narrowing as shown in Fig. 3 at time delays A, B, and C marked in Fig. 2(a). The red line shows the spectrum of TH generation with the influence of molecular alignment revivals, while the blue one shows the spectrum at random alignment revivals. It is clearly shown in Fig. 3 that the spectrum of TH generation is broad-

ened at time delays A and C. According to the above analysis, the prealigned molecular axis is perpendicular to the polarization of the followed probe pulse at these revivals. The refractive index experienced by the probe pulse decreases. It works equivalently like inserting a concave lens in the propagation of probe pulse, giving rise to the defocusing effect. This elongates the filament length of the probe pulse. Therefore, the cross-phase modulation experienced by TH pulse is further enhanced, resulting in spectral line broadening of the TH pulse. In contrast, the spectrum exhibits slight blue-shift accompanied by narrowing the width while the time delay between the pump and probe pulses is tuned at delay B as shown in Fig. 2(a). On this occasion, the prealigned molecular axis is parallel to the polarization of probe pulse, and the increased refractive index makes it function as a convex lens. The filament length of the probe pulse is shortened due to the further focusing effect from the prealigned molecules. Therefore, the cross-phase modulation experienced by TH pulse decreases in comparison with the spectrum at random revivals. The spectral broadening is weakened, resulting in a narrower spectral width.

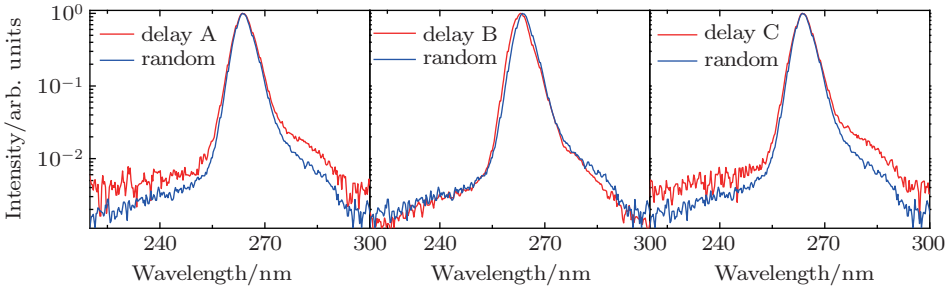


Fig. 3. (color online) Spectra of TH generation at different molecular alignment revivals. The red line shows the spectrum with the effect of molecular alignment, while the blue one represents the spectrum of TH generation at random revivals. Delays A, B, and C correspond to the molecular alignment revivals in Fig. 2.

3.2. TH modulation by preformed plasma

Then, the pump pulse energy is increased to 0.29 mJ to produce the preformed plasma. The profile distribution and spectrum of the generated TH are detected as the pump pulse is tuned substantially before the probe pulse filament temporally. As the refractive index induced by molecular alignment is about 10^{-5}, much less than that induced by plasma effect, which is about 10^{-4}, here, the modulation of TH intensity and spectrum from molecular alignment are ignored. As reported in Ref. [7], the conical emission appears simultaneously with the axial TH component since the probe pulse increases above the critical power. The third-harmonic generation from axial part of the probe pulse leads to the appearance of axial TH component. Compared with the axial one, the conical TH component is closely related to the off-axial phase matching of the third harmonic generation with ring-shaped probe pulse arising from the chromatic dispersion and spatial electron-density gradient of the plasma. By contrasting the spectra of the axial and conical TH components, the generated axial TH shows slight blue-shift due to the refocusing of the trailing part of probe pulse, and red-shifted conical TH component is generated because of the diffraction of the steep front part. Therefore, the axial part of TH generation has shorter central wavelength than the conical one. The output beam pattern of the TH generation is captured by a digital camera in our experiment. Two insets (b) and (a) in Fig. 4 exhibit the spatial profiles of TH generation in the far-field with and without preformed plasma, respectively. In the absence of a preformed plasma, the generated TH distributes not only axially, but also conically. In the probe pulse filament, the generated axial TH pulses before and after the focus interfere destructively, reducing the axial TH intensity. Meanwhile, the overlapping of the probe and axial TH pulse spatiotemporally promotes the back conversion from the axial TH pulse to the probe pulse during their propagations in the filament. But for the conical TH component, it is not overlapped with the ring-shaped probe pulse spatiotemporally, avoiding the back conversion to probe pulse. Therefore, the axial part of the TH intensity is weak compared with the conical one without the preformed plasma. In the presence of a preformed plasma, the conical component of TH generation increases dramatically, and its distribution profile here is only conical as shown in the inset b. As is well known, the refractive index induced by the plasma is proportional to $\delta n_{\rm plas} \sim -\rho/2\rho_{\rm cr}$, where ρ is the electron density of the plasma and $\rho_{\rm cr} \sim \lambda^{-2}$ is the critical density closely related to the laser wavelength. The plasma reduces the refractive index and functions as a concave lens in the propagation of the followed probe pulse. The defocusing effect by the plasma further elongates the length of the probe pulse filament. According to this, the destructive interference between the axial TH pulses before and after the focus is enhanced, and also, the elongated filament increases back conversion from axial TH pulse to the probe pulse, resulting in significant reduction of the axial TH component. The preformed plasma intensifies the chromatic dispersion and divergence of the ring-shipped probe pulse. Based on this, the conical TH is enhanced since the third-harmonic generation process from the ring-shaped probe pulse increases. Also, the phase matching process between ax-

ial part of the FW pulse and a preferential angle is improved with a preformed plasma. The conical part also increases with the improvement of the phase matching. Therefore, under the influence of preformed plasma, the TH generation from probe pulse filament can be distributed only conically. This is a simple method to adjust the TH profile distribution from the filament. A higher divergence and spectral red-shifted TH can be generated, while the density of the preformed plasma increases to further affect the probe pulse.

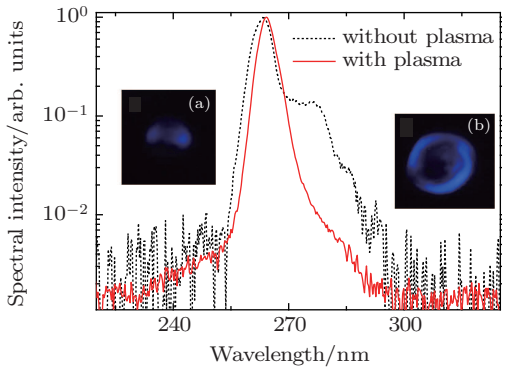


Fig. 4. (color online) Spectra of TH generation from the probe pulse (black dashed line) and under the influence of preformed plasma (red line). Inset (a) shows distribution profile of TH generation in the far-field, recorded by digital camera, and inset (b) shows the distribution profile of TH generation in the presence of preformed plasma.

The experimental results in Fig. 4 display the normalized spectra of TH generation with and without preformed plasma. Without a preformed plasma, the spectrum of TH generation is wide in the consideration of the self-phase modulation during its propagation in the filament. Also, both the axial and conical component of the TH generation appear in this condition. The spectrum is wide with slight red-shifted conical and blue-shifted axial components. Compared with the spectrum without a preformed plasma, a slight red-shifted spectrum appears because of the conical TH component diffracted by the preformed plasma, and accords well with the results reported in Ref. [7]. According to the Gaussian distribution of the laser beam, the central part of the filament has a higher intensity. The axial part of TH generation experiences obvious self-phase modulation, resulting in its wider spectrum. Since the intensity of the ring-shaped probe pulse is smaller than the intensity in the central part, the self-phase modulation experienced by the conical TH component decreases. Therefore, the spectral broadening effect of the conical TH is weaker than the spectrum without preformed plasma. The experimental results demonstrate that the far-field profile distribution of generated TH from femtosecond filament could be redistributed by a preformed plasma. Also, this preformed plasma could be used to modulate the TH spectrum from a laser filament.

4. Conclusions and perspectives

In this work, we study the spectral modulation of TH generation induced by the molecular alignment revivals and preformed plasma. The experimental results show that the spectral line broadening or narrowing could be achieved by tuning the time delay between the probe and pump pulses at different molecular alignment revivals. Also, the peak intensity of the central wavelength increases or decreases under the influence of molecular alignment. The redistribution of TH profile in the far-field and adjustment of TH spectrum could be achieved with the effect of preformed plasma. The results provide an effective method to generate controllable ultraviolet sources.

References

[1] Macklin J J, Kmetec J D and Goudon C L 1993 *Phys. Rev. Lett.* **70** 766
[2] Steingrube D S, Schulz E, Binhammer T, Vockerodt T, Morgner U and Kovaĉev M 2009 *Opt. Express* **17** 16177
[3] Fedotov A B, Koroteev N I, Loy M, Xiao X and Zheltikov A M 1997 *Opt. Commun.* **133** 587
[4] Kim H T, Kim I J, Tosa V, Lee Y S and Nam C H 2004 *Appl. Phys. B* **78** 863
[5] Sutherland J R, Christensen E L, Powers N D, Rhynard S E, Painter J C and Peatross J 2004 *Opt. Express* **12** 4430
[6] Huillier A L, Kmetec J D and Goudon C L 1993 *Phys. Rev. Lett.* **70** 774
[7] Théberge F, Aközbek N, Liu W, Filion J and Chin S L 2007 *Opt. Commun.* **276** 298
[8] Suntsov S, Abdollahpour D, Papazoglou D G and Tzortzakis S 2009 *Opt. Express* **17** 3190
[9] Suntsov S, Abdollahpour D, Papazoglou D G and Tzortzakis S 2010 *Phys. Rev. A* **81** 033817
[10] Yang X, Wu J, Pang Y, Tong Y, Yuan S, Ding L, Xu Z and Zeng H P 2009 *Appl. Phys. Lett.* **95** 111103
[11] Hartinger K and Bartels R A 2008 *Opt. Lett.* **33** 1162
[12] Li M, Li W and Zeng H P 2013 *Opt. Express* **21** 23
[13] Wu J, Cai H, Lu P, Bai X, Ding L and Zeng H P 2009 *Appl. Phys. Lett.* **95** 221502
[14] Cai H, Wu J, Bai X, Pan H and Zeng H P 2010 *Opt. Lett.* **35** 49
[15] Cai H, Wu J, Couairon A and Zeng H P 2009 *Opt. Lett.* **34** 827
[16] Cai H, Wu J, Li H, Bai X and Zeng H P 2009 *Opt. Express* **17** 21060
[17] Chrissanthopoulos A, Hohm U and Wachsmuth U 2000 *J. Mol. Struct.* **526** 323
[18] Aközbek N, Iwasaki A, Becker A, Scalora M, Chin S L and Bowden C M 2002 *Phys. Rev. Lett.* **89** 143901
[19] Marburger J H and Dawes E 1968 *Phys. Rev. Lett.* **21** 556
[20] Zhang Z, Lu X, Zhang Y, Zhou M, Xi T, Wang Z and Zhang J 2010 *Opt. Lett.* **35** 974
[21] Xi T, Lu X and Zhang J 2009 *Opt. Commun.* **282** 3140
[22] Feng L, Lu X, Xi T, Liu X, Li Y, Chen L, Ma J, Dong Q, Wang W, Sheng Z, He D and Zhang J 2012 *Phys. Plasmas* **19** 072305