X-shaped third harmonic generated by ultrashort infrared pulse filamentation in air

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The authors report the measurement of the angularly resolved spectrum of the third harmonic generated in a femtosecond filament in air and its evolution with increasing pump power. Pumped by a focused infrared ultrashort pulse with a carrier wavelength of 1270 nm, a pulse duration of ~20 fs, and pulse energy up to 487 μJ, the generated third harmonic is composed of an on-axis emission and a conical ring emission. When the pump power is sufficiently high, angularly resolved spectra with significant X-like feature could be observed, indicating the formation of nonlinear X wave at third harmonic. © 2008 American Institute of Physics. [DOI: 10.1063/1.2830017]

In propagation of intense laser pulse in Kerr nonlinear medium, stable optical filament could be generated as a result of the balance between beam self-focusing due to Kerr nonlinearity, and self-defocusing due to linear diffraction and plasma formation. Owing to the very high intensity achieved in filamentation, several nonlinear phenomena could arise, such as supercontinuum generation, efficient third harmonic (TH) generation, and conical emission. In particular, due to the potential application in laser frequency conversion to shorter wavelength, TH generation through optical filaments in air has been intensively studied. Spatiotemporal coupling during nonlinear propagation would naturally result in complex spatiotemporal structures in both the fundamental pulse and other products of interaction such as TH generation that emerge from the output. The traditional spectrum detection technique, however, is not sufficient for the diagnosis of the complex wavepacket. To overcome this problem, a simple but powerful diagnostic method, namely, the measurement of angularly resolved spectra, has been developed recently. Angularly resolved spectra were proved to be fruitful in the area of ultrashort pulse propagation in various kind of nonlinear media, through which surprisingly detailed portrait of complex wavepacket could be obtained. For example, in Kerr media with normal or anomalous dispersion, an intense Gaussian wavepacket could spontaneously transform into nonlinear X or O wave after its nonlinear propagation. We note that most of the experimental studies were focused on the wavepacket transformation of the fundamental wave (FW), whereas in Kerr media, due to the self-group and phase locking between the fundamental wave (FW) and the TH in their nonlinear propagation, the wavepacket of TH should undergo similar spatiotemporal modulation as that of FW. X-waves at TH were observed by propagation of 800 nm pulses either in subatmospheric pressure noble gas or in air and X-waves were also obtained in the simulation of TH generation using 800 nm pulses as FW. If the TH pulse with a complex structure could be diagnosed by use of the angularly resolved spectrometer, it will offer us an opportunity for obtaining a deep understanding of the evolution of the TH pulse and the mechanism of the wave coupling in filamentation experimentally. So far, the popular carrier wavelength of an ultrashort pulse laser source is around 800 nm. The TH (carrier wavelength, 267 nm) spectral signal is rather weak and should usually be measured by a photomultiplier tube instead of a charge coupled device (CCD) due to the strong absorption by oxygen and the relatively poor detection sensitivity of CCD in the UV region. Recently, the rapid development of optical parametric amplification (OPA) technology makes it possible to generate laser pulse with energy of hundreds of microjoules in the infrared region. Since the TH of an infrared pulse could be easily detected by a CCD, the angularly resolved spectrum of TH was expected to be attainable with the infrared pump source.

In this letter, we report on the experimental measurement of angularly resolved spectrum of third harmonic generated by intense infrared pulse after its filamentation in air. The X featured angularly resolved spectrum of TH indicates that the TH wavepacket is transformed into nonlinear X wave after filamentation, and the intensity-dependent spectrum gives a clear evidence of the strong nonlinear phase locking between the FW and the TH during their copropagation.

The experimental setup is shown in Fig. 1(a). The pump source of the OPA system (TOPAS-C, Light Conversion) is an amplified Ti:sapphire laser system (Legend, Coherent), which produces ultrashort pulses with a pulse duration of 40 fs, a carrier wavelength of 795 nm, and a single pulse energy of 2.5 mJ at 1 KHz repetition rate. The carrier wavelength from the OPA is tuned to 1270 nm, with a maximum single pulse energy of up to 487 μJ. The temporal duration of the IR pulse from OPA is measured to be ~20 fs (assuming a Gaussian profile) by a home-built second-order single-shot autocorrelator (SSA). The autocorrelation trace is shown in the inset of Fig. 1(a). After being separated from the idler using a broadband reflection mirror (M1, high reflection at 1100–1300 nm), the infrared pump pulse is then tightly focused by a gold coated concave mirror with a focal length of...
250 mm to generate a single light filament in air near the geometrical focal point. After filamentation, an angularly resolved spectrometer was employed to portrait the angular resolved spectrum of the output TH pulse; the far-field spatial pattern as well as the filament is thus captured on a digital camera (DS5, Nikon, Japan). The angularly resolved spectrometer consists of a positive lens (L1, focal length: 300 mm) and a grating spectrometer (SpectraPro 300i, Acton), where L1 is acting as a spatial Fourier transformer to generate a two-dimensional k_x-k_y spectrum of the output TH at the entrance slit of the grating spectrometer. The central part of the k_x-k_y spectrum (k_y=0 in this case) is then selected out by the slit and is further Fourier transformed by the spectrometer. Finally, the angular-resolved (k_y-ω) spectrum is recorded by a CCD.

For a pump pulse energy of 487 µJ, a 3–4 cm long light filament [Fig. 1(b)], accompanied by bright blue (423 nm) conical ring of the TH pulse at the end of the filament [Fig. 1(c)] could be observed. The center of the filament is ~5 mm before the geometrical focal point of the mirror M3. The conversion efficiency of the TH generation is measured to be ~0.02%. Comparing with the high THG conversion efficiency of the order of 0.1% (Refs. 2 and 13) in air with shorter wavelength pump (800 nm), the THG efficiency with the IR pump laser obtained in this experiment is significantly lower. However, since the conversion efficiency of THG in femtosecond laser filamentation relies critically on the pump laser parameters (e.g., wavelength, pulse energy, beam size, spatial profile, and so on) as well as the focal length, one might be able to improve the THG efficiency by optimizing the IR pump laser parameters and the focusing conditions. The far-field pattern indicates that the output TH pulse splits up into an on-axis part and an off-axis conical ring emission due to the tight-focusing geometry. The measured divergence angle with respect to the propagation axis of the conical TH is centered approximately at 4.5 mrad. According to the linear phase-matching-condition of off-axis TH generation in air, the conversion efficiency of the TH generation is measured to be ~0.1%.

\[
\theta(\omega) = \tan^{-1}\left\{\frac{\sqrt{2}K_c(3\omega)K_c(3\omega) - 3K_c(\omega)}}{K_c(3\omega)}\right\},
\]  

where \(\theta(\omega)\) is the phase-matching angle of TH generation with FW frequency of \(\omega\), \(K_c(\omega) = 4.948 \times 10^6 \text{ m}^{-1}\) and \(K_c(3\omega) = 1.4846 \times 10^7 \text{ m}^{-1}\) are the propagation constants of FW and TH, respectively, the calculated phase-matching angle for TH generation at 423 nm is 4.02 mrad, which agrees reasonably well with the measured divergence angle.

Figure 2 shows the evolution of the angular-resolved spectrum (\(\theta-\lambda\)) of TH with the increase of the pumping pulse power from 30 to 487 µJ. The off-axis noise pattern [indicated by the dashed ring in Figs. 2(c) and 2(d)] locating around 395 nm could be found. This background noise is due to some unavoidable leakage/scattering of the second harmonic (SH) of the pump source of OPA, which is generated from a quadratic nonlinear crystal in the OPA. For input pulse energy below 100 µJ, only on-axis TH exists and no conical emission could be observed [Fig. 2(a)]. With pump energy above 100 µJ [see Figs. 2(b)–2(d)], off-axis TH conical emission appears with the divergence angle increasing with intensity, while the on-axis TH spectrum is gradually blue shifted to 370 nm. Interestingly, when the pump pulse energy is further increased to >430 µJ, two hyperbolic (or X-like) structured tails (a strong one is located around 410 nm and a weak one at 384 nm) emerge and grow rapidly, as shown in Fig. 2(d). In a normal dispersive medium, due to spatiotemporal modulation instability (MI), the spatiotemporal modes with high parametric gain could be found to have hyperbolic structures in the Fourier plane (e.g., \(K-\Omega\) plane) which shows up in far-field spectra. Thus, we conclude that the hyperbolic structured tails in the TH \(\theta-\lambda\) spectra originated from the MI during the THG process, where the hyperbolic tails could obtain high gain and grow exponentially with the increasing pump intensity.

In Fig. 3, the spectra of the on-axis TH and the ring TH are compared with a reference TH spectrum, which is the cubic intensity of input FW spectrum with a threefold decrease in wavelength. The output TH spectra are not only much broader than the input reference TH spectrum, but also significantly blue shifted. It has been shown that, due to a strong energy exchange or coupling between the FW and TH
of the output TH pulse. The intensity-dependent spectral broadening and frequency shift of the on-axis TH implies the existence of strong cross-phase modulation which originates from the nonlinear phase and group coupling between the FW and TH wavepackets.

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In conclusion, we have measured the far-field angularly resolved spectrum of the TH generated after the filamentation of the intense ultrashort IR pulse and investigated its dependence on the pump pulse energy. Similar to the case of THG using 800 nm, the conical TH contains significantly more energy than the on-axis TH since the off-axis component satisfies the phase matching condition of THG. The hyperbolic-structured tails on the far-field spectrum were observed, which indicate the formation of the nonlinear X wave of the output TH pulse.