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Spectral shifts of the fundamental and third harmonic radiation in air induced by self-focused femtosecond laser pulses

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ABSTRACT

The large spectral shifts of the fundamental and third harmonic waves (up to 20 nm and 50 nm, respectively) during femtosecond laser pulse filamentation in air have been demonstrated by using focusing lenses of long focal length. It was shown that in a wide range of laser intensities this process does not saturate and linearly depend on the pump pulse energy.

1. Introduction

Third harmonic generation (THG) in gases is a radiation source capable to produce the coherent and powerful ultraviolet (UV) and vacuum ultraviolet (VUV) radiation [1–4]. THG can find applications not only for the efficient frequency tripling over a wide range of wavelengths, but also in a variety of scientific and technological areas including the atmospheric pollutants detection through third harmonic (TH) induced-fluorescence [2], laser-induced plasma diagnostic [5], ultrafast spectroscopy of atoms and molecules [6], time-resolved and high-resolution biological imaging [7], etc. [2,6].

Unfortunately, usually the efficiency of THG is limited due to the fact that its phase matching conditions cannot be satisfied in a medium of normal dispersion. Moreover, in the tight-focusing limit the TH wave generated prior the focus interferes destructively with TH wave generated beyond focus [8]. Also, when femtosecond laser pulses are focused to achieve more efficient frequency conversion, the other nonlinear effects compete with THG. Thus, the self-focusing and plasma electron defocusing lead to the femtosecond laser pulse filamentation in air and the intensity clamping [9]. Therefore the light peak intensity usually is limited to about 10^{13} W/cm^2 during pulse propagation in air [10]. Nevertheless, even the clamped laser intensity is sufficient for the efficient THG [9,10]. Thus, the achieved efficiency of TH generated by infrared (IR) femtosecond laser pulses in air is about 0.2 percent [3,4]. Moreover, during femtosecond laser-induced filamentation in air a two-color filament can be formed and the plasma-related processes can lessen the tight phase matching constraint between the fundamental harmonic (FH) and the TH pulses. As a result the phase-matching conditions of THG can be satisfied over the longer propagation distances [4,11]. However, during the femtosecond laser filamentation in air the spectra of TH and FH pulses are broadened due to electron plasma generation [12,13], self-phase modulation (SPM) [14,15], cross-phase modulation (XPM) [6,16] and the temporal self-steepening [17,18]. The spatial structure of TH is also complicated. Thus, when FH power is less than the critical power needed for self-focusing, the far-field pattern of the TH beam consists of only the central part. When the pump power exceeds the critical power, the conical emission of TH appears and the far-field pattern of the TH beam consists of two parts: central one and the ring around it, i.e. the conical TH beam [1,2]. Note that TH central part experiences spectral blue-shift of peak wavelength while spectrum of the conical part is red-shifted for the large FH pulse energies. However, in previous papers reported spectral shifts of TH were of the order of 3–4 nm and saturated quickly with the pump energy [1,2]. Moreover, recently it was demonstrated that at least a part of TH generated by the femtosecond laser pulses in gases can be a result of a noncollinear six-wave mixing (SWM) [19,20]. Thus unfortunately, the physical mechanisms of femtosecond filamentation in gases are not fully understood and the complicated spatial and spectral structure of TH strongly limit the number of potential applications of such a UV light source [2,4].

This motivated us to study the spectral and spatial properties of the FH and TH generated in air by focused femtosecond laser pulses of variable energy. In contrast to other papers [1,2] for the

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experiments we have used lenses of longer focal length (1–2 m) which allowed us to increase the spectral shifts of TH to about 20 nm. Furthermore, the registered dependencies were not saturated and spectral shifts of FH and TH increased linearly with the pump pulse energy. Also note that TH spectral shifts were proportional to the measured plasma filament length which was also linearly dependent on the FH pulse energy.

2. Experimental setup

In our experiments TH was generated in atmospheric air by focused femtosecond laser pulses. All experiments were performed using a Ti:sapphire laser system, which generated the ultrashort light pulses of 120 fs (FWHM) duration with a central wavelength of 800 nm at a repetition rate of 1 kHz, single pulse energy of up to 3.6 mJ (Fig. 1). The diameter of the Gaussian beam at the $1/e^2$ level was about 8 mm. Laser pulse energy could be varied by a half-wave plate and two polarizers. The pump beam was focused using lenses of various focal length. The TH was generated in a plasma filament created in air by the FH laser pulses. The TH signal from the fundamental radiation was separated using the dichroic mirrors and interference filters. Spectral characteristics of TH were registered using a spectrometer "Ocean Optics". During these measurements the conical and central parts of TH signal were focused into the fiber delivering light to the spectrometer. In addition to these measurements, the fluorescence images produced on the paper screen by the far-field TH radiation were registered by digital camera. In all experiments the pulse energy of pump beam was controlled by using the energy detector "Ophir 30A-SH".

3. Results and discussion

At the high laser pulse energies (more than 1 mJ per pulse) a bright blue TH central spot surrounded by the ring has been observed on the screen. Typical experimentally registered far-field patterns of the TH emission are shown in Fig. 2. Note that the divergence of TH conical part have not depended on the FH pulse energy E but decreased with the focusing lens focal length f. When the lenses of short focal length (f=300 mm) were used, the conical and central parts of TH overlapped in space and the spatial structure of the far-field radiation was not distinct (Fig. 2a).

In order to investigate the interaction length of the FW and TH pulses, we have measured the dependencies of the plasma filament length L on the FH pulse energy E for different focal lengths of focusing lenses. Thus, when the visible by naked eye plasma column appeared (E=1 mJ, see Fig. 3a), the registered by a CCD camera filament length was found to be about 5 cm (note that in this case the Rayleigh length $z_0 = \xi^2\pi/\lambda$ was about 6.3 cm, where $\lambda = 800$ nm is the wavelength, $\xi = 127$ μm is the estimated beam waist for f=1000 mm). For higher pump pulse energies the plasma column length linearly increased with E (see Fig. 3b). The slope values of linear fit γ were the following: 15.9 cm/mJ (f=2000 mm), 8.2 cm/mJ (f=1400 mm) and 5.2 cm/mJ (f=1000 mm). Note that the length of the plasma column increased faster for the lenses of longer focal length f. Note also that for E=1 mJ, the peak power P=7.8 GW (for the Gaussian pulse of 120 fs duration at FWHM) is larger than the critical power $P_{cr} = 2.9$ GW for self-focusing ($P_{cr} = (0.61\lambda)^2\pi/(8n_0n_2)$ where n_0 and $n_2 = 3.2 \times 10^{-19}$ cm^2/W are the linear and nonlinear refractive indexes of air, respectively) and plasma filaments appeared for all the used focal length lenses. Thus, the plasma formation onset was strongly dependent on P but weakly depended on f under our experimental conditions. Moreover, when the filament appeared (E=1 mJ), the variations of the plasma filament length L were less than 5 percent for all the used focal length lenses. At the maximum pulse energy (E=3 mJ) the variations were about 7 (for f=1000 mm), 10 (for f=1400 mm) and 15 (for f=2000 mm) percent. So the longer plasma column was formed, the bigger were variations of L. Note that the THG conversion efficiency was of the order of $10^{-6} - 10^{-5}$ for E=2 mJ and focal lengths of $f = 1000$–2000 mm.

The FH spectral broadening registered beyond the filament is illustrated in Fig. 4(a). The spectra of FH were registered using long focal length (f=2000 mm) lens because it forms the long plasma filament (up to 50 cm) (see Fig. 3 (b)) and as a result the spectral broadening is larger than that obtained by using lenses of shorter focal length. As one can see (see Fig. 4(a)), the FH spectra consist of the blue-shifted and red-shifted parts with respect to the central pump wavelength of 800 nm for the $E > 1$ mJ. In addition, the peak wavelength of TH conical part (of about 274 nm) was linearly dependent on the FH pulse energy E and red-shifted with respect to the central pump wavelength divided by three (see Fig. 4(b)). However, when E was less than 0.55 mJ, the peak wavelength of TH central part was about 266 nm, which corresponds to the non-shifted TH wavelength. In contrast to the conical TH emission, the

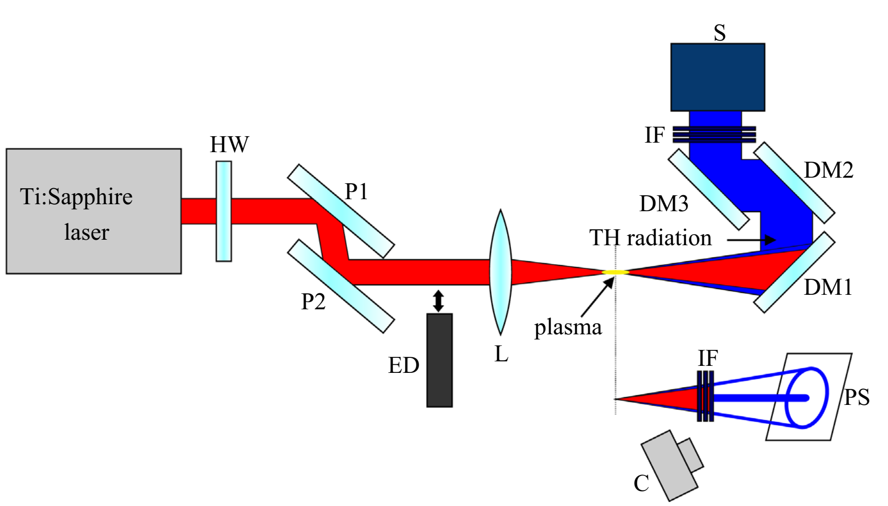

Fig. 1. Schematic of the experimental setup: HW, half-wave plate; P1–P2, polarizers; ED, energy detector; L, focusing lens; DM1–DM3, dichroic mirrors; IF, interference filters; S, spectrometer; C, digital camera; PS, paper screen.