Fourth-order harmonic generation during parametric four-wave mixing in the filaments in ambient air

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(Received 4 June 2010; published 11 October 2010)

We propose a simplified scheme for efficient 200-nm pulse generation in isotropic medium using 800-nm radiation. The observation and optimization of the fourth-order harmonic generation during two-color filamentation in air are reported. This parametric process is based on four-wave mixing of the fundamental and the second-order harmonic radiation of the Ti:sapphire laser. A systematic study of the influence of the laser intensity, the polarization, the chirp, and the pulse duration on the fourth-order harmonic output from the filaments has been carried out. The fourth-order harmonic conversion efficiency was estimated to be of the order of 10^{-4}.

DOI: 10.1103/PhysRevA.82.043812 PACS number(s): 42.65.Jx, 52.38.Hb, 42.65.Ky

I. INTRODUCTION

Third-order harmonic generation (THG) during self-channeling (filamentation) in air using high-power 800-nm femtosecond laser pulses has become a versatile method for coherent UV (~266 nm) radiation generation [1–6]. The filamentation causes pulse self-compression, spatial self-filtering, intensity clamping with self-stabilized high intensity in the core, supercontinuum generation, white-light generation, and terahertz emission. These studies were performed under conditions of self-guided propagation of a laser beam through a gas medium when Kerr-induced self-focusing was compensated for by self-defocusing caused by the free electrons generated in the ionized channel. These THG experiments were carried out in a loose-focusing geometry that is required for phase matching in a positively dispersed medium. An increase of the interaction length in this geometry leads to an increase of third-order harmonic conversion efficiency.

It is well known that intensity inside the air filament can be maintained at intensity as high as 5×10^{13} W cm^{-2}. At these conditions, one can expect the creation of optimal phase-matching conditions for various parametric processes, in particular fourth-order harmonic generation (FHG) in the case of two-color laser (i.e., beam containing fundamental as well as second-order harmonic radiation). One can assume that low-order nonlinearity of air allows for efficient generation of the fourth-order harmonic through the parametric processes of the wave mixing in filaments.

Parametric four-wave-mixing (FWM) processes in atomic media have been studied for a long time starting from early laser-atom experiments. In particular, the $\omega + \omega + 2\omega \rightarrow 4\omega$ process was analyzed in strontium vapors, and relatively high efficiencies (3×10^{-5}) were reported in the case of resonance-induced enhancement of wavelength conversion [7]. FWM in gases was studied using different gases as well, and the vacuum UV radiation ($\lambda \approx 100$–200 nm) has been generated using different laser sources (see Ref. [8] and references therein). The interest in FWM has been growing as an alternative to the direct harmonic generation. Tunable, visible, ultrashort pulses can be created using FWM by filamentation in a gas cell [5]. Few-cycle pulses in the deep UV and mid-IR have also been generated by this method [9,10]. In particular, in Ref. [9], generation of UV femtosecond pulses has been reported by FWM through filamentation in neon gas. The fundamental (ω) and second-order harmonic (2ω) pulses of a 25-fs Ti:sapphire laser were focused into neon gas, and 20-μJ pulses with the center wavelength of 260 nm were produced by a FWM process ($2\omega + 2\omega - \omega \rightarrow 3\omega$) through a 15-cm-long filament. They also observed the generation of pulses at 200 nm (i.e., FHG) with an energy of 2 μJ, which was assigned to the cascaded processes ($\omega + \omega + 2\omega \rightarrow 4\omega$ or $3\omega + 2\omega - \omega \rightarrow 4\omega$).

In this article, we systematically analyze the generation of fourth-order harmonic through the parametric mixing of the fundamental wave and second-order harmonic of the Ti:sapphire laser ($\omega + \omega + 2\omega \rightarrow 4\omega$) in air filaments. We analyze the influence of various experimental parameters (laser intensity, polarization, chirp, and pulse duration) on the FHG in air. Nonlinear frequency conversion demonstrated in this study, such as fourth-order harmonic ($\lambda \approx 200$ nm) generation of IR femtosecond pulse, is an efficient approach to obtaining ultrashort laser pulses in the UV spectral region at the very edge of the transmittance of air.

II. EXPERIMENTAL DETAILS

In the experiments, a commercial Ti:sapphire femtosecond laser system (Alpha 10/US: Thales Laser S.A., France) was used, which delivered 790-nm, 45-mJ pulses of 48 fs at a repetition rate of 10 Hz. The pulses were focused by a convex lens of focal length 2 m in ambient air. The value of the Rayleigh length (before filamentation) with the lens used was about 20 mm. In the focal area, 100-to-130-mm-long filaments and white-light generation were observed. At these conditions, broadening of the third-order harmonic emission was also observed. A type-I phase-matched β-barium borate (BBO) crystal of 1 mm thickness was inserted in the beam between the lens and its geometrical focus. The conversion efficiency to the second-order harmonic ($\lambda = 395$ nm) in BBO was measured to be 8%. The intensity of fundamental radiation inside the second-order harmonic crystal was maintained at such a level that no self-phase modulation (SPM), appreciable chirp, or white-light generation were introduced on the residual

radiation by the BBO crystal. At this conversion efficiency, filamentation induced by the second-order harmonic wave itself was observed when the BG39 filter was inserted after the crystal to remove the unconverted fundamental radiation. The characteristics of the light filaments formed by second-order harmonic ($\lambda \approx 395$ nm) femtosecond laser pulses have previously been experimentally investigated [11]. The 395-nm femtosecond laser pulses have several characteristics, which are beneficial for the generation of plasma filaments in air. In particular, the critical power required for self-focusing is in the range of 0.3–0.6 GW, which is almost ten times less than the critical power in the case of a 790-nm laser. We analyzed the influence of pulse characteristics on the FHG in air by changing the duration of the output pulse between 48 and 2300 fs. Note that we analyzed the on-axis fourth-order harmonic radiation propagating through the 2-mm-wide slit placed in front of the dispersive prism. The harmonic radiation propagating through the filaments was dispersed by a fused silica prism and analyzed by a fiber spectrometer (USB2000: Ocean Optics). The predispersion through the prism was necessary to avoid saturation of the spectrometer by the unconverted fundamental radiation.

III. RESULTS AND DISCUSSION

A. Finding the conditions for fourth-order harmonic generation

Generation of the off-axis third-order harmonic component due to the phase-matching between the fundamental and third-order harmonic waves started at pump powers around the critical power related with filament formation. In the far field, the off-axis component appeared as a ring whose diameter corresponds to a phase-matching cone. At pump powers above the critical power for self-focusing, most of the third-order harmonic energy was concentrated in the ring pattern. In our case, it was a cone of diameter 80 mm at the distance of 1600 mm from the filaments. The same can be said about the fourth-order harmonic radiation, which repeated the same properties. It may be noted that the maximum intensity of the observed fourth-order harmonic appearing as a result of the $\omega + \omega + 2\omega \rightarrow 4\omega$ sum mixing did not correlate with conditions of maximum third-order harmonic yield. Also note that the maximum intensity of fourth-order harmonic was observed for a longer wavelength ($\lambda \approx 201.5$ nm) with regard to the exact position of the fourth-order harmonic ($\lambda \approx 790/4$ nm $= 197.5$ nm). Efficient FWM was observed in the case of artificially chirped 790-nm pulses, which is discussed in what follows (see Sec. III B).

The combined action of self-focusing and plasma generation in air plays an important role in on-axis and off-axis phase matching along the filament, resulting in high conversion efficiency of the THG [12]. In our case, THG conversion efficiency in air up to 10^{-3} was achieved, which is in the range of previously reported data [13]. This high conversion efficiency is induced by phase locking between the generated third-order harmonic wave and the fundamental wave inside the filament, which maintains the phase-matching condition for THG over a long distance (see also [14,15], where another interpretation is given).

In articles of Kolesik *et al.* [14,15], it is demonstrated numerically that the supercontinuum generation and the THG that accompany the optical filamentation in a nonlinear dispersive bulk medium can be described as first-order scattering processes. In particular, for an incident ultrashort pulse, the angle-resolved spectrum of the transmitted pulse is shown to be accurately determined using first-order scattering of the incident field from the refractive index nonlinearly modified due to the optical filament. Thus, although an optical filament is a highly nonlinear object, the accompanying supercontinuum generation and THG are driven parametrically by the filament. Overall, one may say that there is no general consensus regarding the THG mechanism. In fact, the phase-locking mechanism first proposed has actually been demonstrated not to occur in Ref. [14,15].

The two-color pump, using the fundamental and second-order harmonic fields, has become a practical way of high-order harmonic enhancement in gas and plasma media [16–20]. The use of a two-color field breaks the inversion symmetry and allows for the generation of the odd harmonics, which are more intense than those generated with the fundamental field alone, and even harmonics as intense as the odd ones. It may be noted that, in some cases, the intensity of low-order even harmonics was higher than that of the odd harmonics in the same spectral range.

In the case of two-color pump, at small fundamental beam energy, only third-order harmonic radiation on the coaxial direction was observed, while the increase of fundamental beam energy led to the appearance of a ringlike structure of the third-order harmonic and fourth-order harmonic radiation. The transformation of the third-order harmonic beam shape following the increasing pump energy implies that one or more nonlinear optical effects are playing important roles in the THG process during the filamentation.

The phase modulation of the laser radiation can easily change the spectrum and intensity of the harmonics. It may be noted that the effective spectral broadening and tuning of the harmonics in the range of few nanometers using the chirp variation can be achieved only in the case of broadband radiation. SPM of the laser pulse is widely used to generate additional frequencies. In particular, insertion of optical medium (like a glass slab) in the path of the laser pulse leads to moderate spectral broadening of the laser pulse by SPM. One can expect that propagation of a focused intense laser pulse through the air can also cause significant SPM-induced broadening of the driving laser pulse and remarkably change the harmonic spectrum during the formation of filaments.

It was observed that, during filamentation, the propagating laser pulse experiences a modification of its spectral and temporal structures. The SPM due to the refractive index change by plasma formation and by the Kerr effect induces spectral broadening [Fig. 1(a)]. The SPM in air induces spectral broadening toward the shorter wavelength in the leading edge of the pulse, while the Kerr effect causes the spectral broadening toward the longer wavelength in the leading edge and shorter wavelength in the trailing edge [see the fundamental radiation spectra propagating through the focal area at low and high intensity of initial laser pulse, Fig. 1(a)]. The spatial and spectral characteristics of the 790-nm radiation creating air filaments were analyzed. The initial spatial shape of the compressed driving beam was

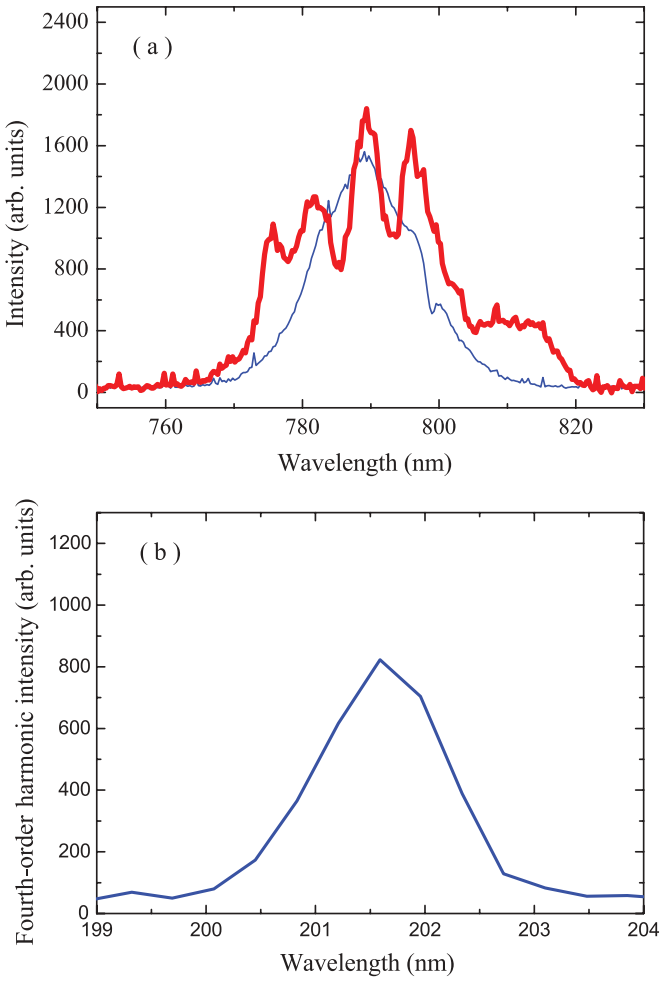

FIG. 1. (Color online) (a) Spectral shapes of the fundamental radiation after propagation through the focal area at low (thin curve) and high (thick curve) energies of laser pulses. (b) Spectral distribution of the fourth-order harmonic.

relatively smooth, without any significant variation of laser intensity along the beam. The spatial shape of the laser beam creating the filaments and propagating through the air, even after some smoothing during the long pass, showed the hot spots created due to the filamentation of the separated parts of beam. Strong SPM and white-light generation in the area of filament formation also caused a considerable change of the spectral shape of the driving laser beam. The spectra of the driving pulse before and after propagation through the air filaments considerably differ from each other. While the spectral width of the laser pulse before focusing always remained in the range of 20 nm, the observation of a dramatic increase of spectral width of the focused laser radiation propagating through the air filaments can be attributed to aforementioned nonlinear optical processes. At the same time, the bandwidth and spectral shape of the fourth-order harmonic remained mostly unchanged in a broad range of variable parameters of experiments, which indicates the nonsaturated conditions of FHG (Fig. 1). The FHG was observed under the conditions when long filaments were produced in air.

B. Optimization of the fourth-order harmonic yield

The influence of chirp variation on the fourth-order harmonic efficiency was studied. The chirp of fundamental radiation was varied by changing the separation between the gratings in the compressor chamber. Reducing the separation from the chirp-free conditions resulted in positively chirped pulses, and an increase of the distance between the gratings provided negatively chirped pulses. At negative chirp, the pulse contains short wavelength components of the laser spectrum in the leading part of the pulse and vice versa. The chirp contained in the laser pulse also affects the temporal structure of the laser pulse.

A comparison of the fourth- and third-order harmonic spectra and fundamental spectra was made at different intensities. While the third-order harmonic radiation, in most of cases, showed a blue shift, the fourth-order harmonic radiation shifted toward red. The spectral width of fourth-order harmonic emission remained constant (~1.4 nm) compared to the ~30-nm bandwidth of the propagated fundamental radiation. A significant variation in the fundamental spectrum was observed at intensities close to the ionization threshold, whereas the fourth-order harmonic spectrum showed a smooth envelope even at considerably higher intensity. This feature can be explained by the dynamics of the sum frequency generation during the initial stage of laser-air interaction. The intense ultrashort pulse produces a temporal variation of the electron density gradients, which makes the spatial and spectral shapes of the self-modulated pulse propagating through the focal spot complicated. The intensity-induced phase modulation could also lead to variations in the spatial shape of fundamental and harmonic radiation [21]. However, in the case of fourth-order harmonic emission, no significant spectral modulation was observed.

Most previous studies of the THG in loose-focusing configuration were carried out at the conditions of plasma formation, when self-channeling and long-pass filamentation considerably influenced the THG in air [2]. The ionization of the air consumed a part of the fundamental energy. The growth of the third-order harmonic yield in this case was determined by the interplay of (a) the growth of the product of the fundamental wave's intensity and the effective length of interaction, (b) the depletion of the laser intensity owing to the ionization of air, and (c) the phase modulation leading to the phase mismatching of THG. The growth of conversion efficiency under these conditions was accompanied by considerable third-order harmonic spectral broadening and beam shape distortion.

Previous studies on THG in air and other gases with positive dispersion demonstrated different dependencies on $I_{3\omega}(I_{1\omega})$, depending on experimental conditions (pulse duration, focusing conditions, gas pressure, etc.). In our case, the influence of pulse duration and chirp variation on the fourth-order harmonic emission was studied at fixed pulse energy. The pulse duration of the fundamental radiation was gradually increased from 48 to 2300 fs by changing the distance between the compressor gratings. Figure 2(a) shows the $I_{4\omega}(t_p)$ dependence on pulse duration at a fundamental pulse energy of $E = 44$ mJ. The optimal pulse duration (300–400 fs) was defined at a range of pulse intensities, which allows generation of the maximum fourth-order harmonic intensity. No fourth-order harmonic was observed at zero chirp.

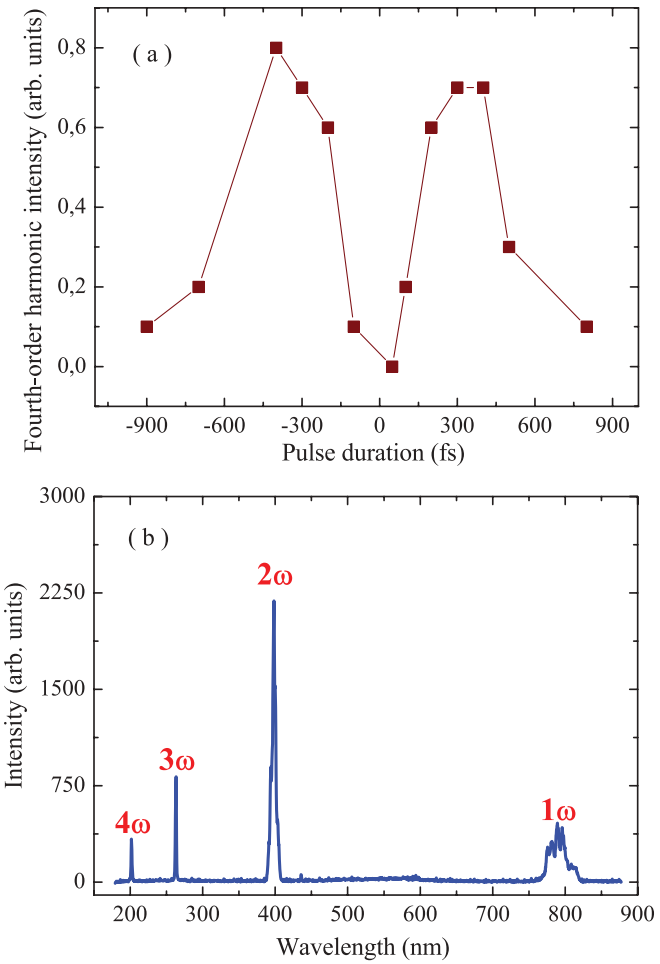

FIG. 2. (Color online) (a) Fourth-order harmonic output as a function of pulse duration for positively and negatively chirped laser radiation. Positive and negative values of pulse duration correspond to positively and negatively chirped pulses. (b) Harmonic spectra at optimal conditions for the FHG.

Nonanalogous behavior was observed when the intensity of fundamental radiation was varied by changing the pulse energy. The reason for this could be some change in the divergence of the fundamental beam, as well as different roles of the radiation in the generation of free electrons at different pulse durations. The partial separation between fundamental and second-order harmonic pulses in the filaments cannot fully exclude the FWM. At the same time, with increasing the chirp, the pulse length increases such that the overlap of the two pulses is increased. This may perhaps lead to conversion over a narrow bandwidth (as not all wavelengths in the two chirped pulses overlap), as was observed in experiment, and it also explains the symmetric efficiency for negative and positive chirps [Fig. 2(a)].

Now we analyze the observed $I_{4\omega}(t_p)$ dependence from the point of view of the delay between the two chirp-free pump pulses (ω and 2ω) inside the filaments. Due to the group velocity mismatch between the ω and 2ω waves in the type-I BBO crystal, the 790-nm beam is delayed ($\Delta_{\text{cryst}} = l_{\text{cryst}}[(n^o_\omega)_{\text{group}}/c - (n^e_{2\omega})_{\text{group}}/c] \approx 188$ fs for 1-mm-thick BBO) with respect to the 395-nm beam due to $n^o_\omega > n^e_{2\omega}$ in this negative uniaxial crystal ($n^o_\omega \approx 1.6605, n^e_{2\omega} \approx 1.5681$ [22], $(c/n^o_\omega)_{\text{group}}$ and $(c/n^e_{2\omega})_{\text{group}}$ are the group velocities for the o and e waves in BBO at 790 and 395 nm, respectively; $n_{\text{group}} = n - \lambda(dn/d\lambda)$ is the group refractive index calculated from the Sellmeier relations). The duration of the 395-nm pulse is given by $\Delta t_{2\omega} \approx [(\Delta_{\text{cryst}})^2 + 0.5(\Delta t_\omega)^2]^{1/2}$ [23]. Hence, the 395-nm beam has a longer pulse duration, corresponding to the induced delay and a certain percentage (~50%) of the 790-nm pulse duration. The latter is because the energy of the fundamental radiation is in general not high enough in the pulse wings to effectively generate the second-order harmonic. Thus, one can estimate $\Delta t_{2\omega}$ at the output of the BBO crystal to be about 191 fs. The second-order harmonic crystal was inserted in the experimental scheme at an appropriate position after the focusing lens so that no impeding processes were observed after propagation of the laser radiation through the crystal. After leaving the crystal, the delay between the two pulses gets reduced due to propagation in air [$n^{\text{air}}_\omega \approx 1.000\,274$, $n^{\text{air}}_{2\omega} \approx 1.000\,283$, $(n^{\text{air}}_\omega)_{\text{group}} \approx 1.000\,280$, $(n^{\text{air}}_{2\omega})_{\text{group}} \approx 1.000\,304$]. In the area of filament formation, which was approximately $L = 700$ mm away from the BBO crystal, the delay between two pulses decreases by $\Delta_{\text{air}} = L\ [(n^{\text{air}}_{2\omega})_{\text{group}}/c - (n^{\text{air}}_\omega)_{\text{group}}/c] \approx 55$ fs. Thus, the final delay between two pumps inside the filaments became ≈133 fs.

Under these conditions (i.e., for chirp-free pulses), no fourth-order harmonic was observed. Although the 48-fs, 790-nm pulse partially overlaps with the trailing part of the 191-fs, 395-nm pulse, the conditions for parametric FWM remained far from optimum. The increase of fundamental pulse duration by introducing the positive or negative chirp led to considerable improvement in the overlapping of two pumps inside the filaments. From our experiment [Fig. 2(a)], one can define a range of fundamental pulse duration (300–400 fs) at which the most efficient FWM occurs. This range corresponds to the most effective overlap of the chirped fundamental pulse and broadened second-order harmonic pulse, which led to the strongest FHG. Longer fundamental pulses (>400 fs) caused a decrease of fourth-order harmonic efficiency due to considerable decrease of the laser intensity. Thus, the optimization of the pulse duration is due to interplay between the overlapping of the pump pulses in the filaments and their relative intensities. Our studies show that the FHG takes place despite the significant difference between the two pump intensities [$I(\omega)$:$I(2\omega) \cong 12$:1], and without any precise temporal and spatial matching of the two pumps. The results show that, by appropriate chirping of the fundamental radiation, one can achieve a considerable improvement of the fourth-order harmonic yield.

Figure 2(b) shows the case when all three (second, third, and fourth order) harmonics, together with fundamental radiation, were recorded. The relative intensities of these harmonics cannot be taken into consideration since the conditions for recording of these spectral components were optimized only for the fourth-order harmonic radiation. The harmonics, other than fourth-order harmonic, were seen in this spectrum since their scattered radiation was quite strong.

The influence of fundamental and second-order harmonic intensities on the fourth-order harmonic output was also studied. The yield of the fourth-order harmonic increased

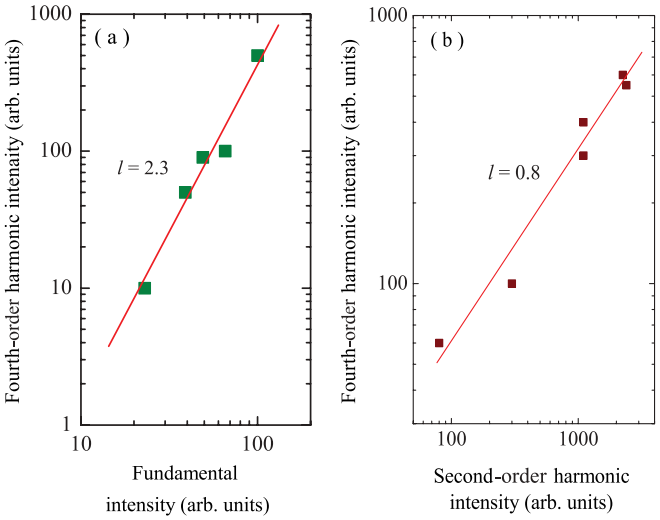

FIG. 3. (Color online) Dependences of the fourth-order harmonic output on the (a) fundamental and (b) second-order harmonic intensity.

almost quadratically with the fundamental wave intensity [Fig. 3(a), scaling of 2.3], while the $I_{4\omega}(I_{2\omega})$ plot showed approximately linear dependence [Fig. 3(b), scaling of 0.8]. Such a behavior is expected in a perturbative regime of the sum mixing [8]. The FWM intensity in that case should follow the dependence $I_{4\omega} \sim (I_{1\omega})^2 \times I_{2\omega}$, which was close to the observations in our experiment. The slopes of the observed dependences $I_{4\omega}(I_{1\omega})$ and $I_{4\omega}(I_{2\omega})$ (2.3 and 0.8, respectively) are close to the expected values (2 and 1).

Finally, we discuss the results of FWM studies using the variation of polarizations of the pump pulses. Initially, the effect of circularly polarized fundamental radiation on the generation of the fourth-order harmonic was studied. A quarter-wave plate was inserted in front of the focusing lens to change the polarization of the laser beam. In this case also, the FHG was observed, although its intensity was two to three times less compared to that with linearly polarized fundamental beam. The observation of the FHG in the case of circularly polarized incident laser light can be explained as follows. Circularly polarized light is composed of two orthogonal linear polarizations differing in phase by a quarter wave. Of these, the component which is an ordinary ray for the BBO crystal (type I phase matched) gets converted to second-order harmonic, and the other orthogonal component, which becomes extraordinary for the BBO crystal goes through the crystal unconverted (no phase matching). As the refractive indices and group velocities for the ordinary and extraordinary components (of the fundamental) in the BBO crystal are quite different, the two components get separated in time and do not give rise to circularly polarized fundamental, but act as independent radiation pulses. So we have a case of second-order harmonic (extraordinary) and fundamental (ordinary) orthogonal to each other (two-color field) and the remaining fundamental field (unconverted extraordinary, single color field) producing the fourth-order harmonic radiation independently.

Subsequently, the polarizations of two beams (ω and 2ω) were made parallel by inserting a $\lambda/2$ plate after the BBO crystal. In this case, at the optimally chirped fundamental radiation, the FHG was decreased, probably due to the group velocity dispersion (GVD) between these two pulses (ω and 2ω), which get partially separated in time. To confirm the influence of GVD on the FHG efficiency, optical glasses of different thickness were inserted after the nonlinear crystal. With 10-mm-thick glass slides inserted after the BBO crystal, no fourth-order harmonic was observed. Fourth-order harmonic started appearing at the thickness of glass slides of 1 mm. With a microslide of thickness of 0.3 mm, the intensity of fourth-order harmonic became stronger (about an order of magnitude higher than that for 1-mm glass slide), while it further increased (by a factor of about 3) when no optical materials were inserted in the path of fundamental and second-order harmonic waves. The optimization of the conditions for strongest fourth-order harmonic output was the same as earlier, that is, when the negatively chirped orthogonally polarized 300-fs pulses were used.

It has been shown that in the case of high-order harmonic generation, an orthogonally polarized two-color field can generate harmonics much more strongly than the fundamental field alone [18]. The strong harmonic generation is due to the formation of a quasilinear field, the selection of a short quantum-path component, which has denser electron wave packet, and high ionization rate. With suitable control of the relative phase between the fundamental and second-order harmonic field, this particular field significantly enhances the short-path contribution while diminishing other electron paths, resulting in a clean high harmonic spectrum [18]. These assumptions are based on a nonperturbative approach for the description of the high-order harmonic generation. In the case of lower-order nonlinearities, the perturbative theory [8] describes the behavior of harmonics and parametric processes.

Apart from the phase modulation, another parameter of a femtosecond laser pulse, which also changes during filamentation of the laser beam, is the polarization of radiation. Initial linear polarization of the laser radiation changes after creation of filaments [24]. Various random directions of polarization appear in this radiation during propagation through the focus of lens, together with the prevailing linear polarization component. The presence of harmonics even after rotation of polarization by quarter- and half-wave plates could be attributed to the simultaneous rotation of randomly rotated polarizations of the parts of the laser beam, which can cause the appearance of linearly polarized components in an overall pattern of a polarization state of the laser beam. These weak components are still strong enough and can cause the generation of low-order harmonics and/or low-order parametric processes. The harmonic intensity in that case is defined by the intensity of these weak linearly polarized components. The existence of harmonics during introduction of considerable ellipticity and depolarization is in a stark contrast with conventionally accepted case of a strong influence of the deviation from linear polarization on the harmonic output (both for low- and high-order harmonics).

Measurement of the FHG conversion efficiency was carried out relative to the THG conversion efficiency. The relative intensities of these two waves were measured to be $I_{3\omega}$:$I_{4\omega} \approx$ 10:1, which gives the FHG conversion efficiency to be $\sim 10^{-4}$.

The conversion efficiency of a fourth-order harmonic was also estimated by direct comparison of the intensities of ω and 2ω waves. Their relative intensities were measured using a calibrated photodiode after dispersing the radiation in the fused-silica prism. In this case, the FHG conversion efficiency was estimated to be 2×10^{-4}, which agrees with aforementioned value (10^{-4}) within a factor of two.

We now analyze the difference between our approach and results of Ref. [9] regarding the FHG in neon and air. The differences between our scheme and the one in Ref. [9] are as follows. In the previously mentioned work, the second-order harmonic was obtained using a separate channel where efficient (35%) conversion to the 2ω wave was achieved. The second-order harmonic and the fundamental beams (with equal energies of 0.5 mJ) were then focused by concave mirrors ($r = 2$ m) independently and were spatially and temporally overlapped inside a stainless-steel gas cell filled in with neon. It is clear that there are obvious difficulties in overlapping these two beams (in space) and pulses (in time), especially for short pulse durations. Our case consisted of a simple scheme consisting of the focused fs beam with a BBO crystal inserted in its path between the focusing lens and air filaments. The ratio between fundamental and second-order harmonic waves was 12:1, due to the low conversion efficiency of the fundamental wave to the second-order harmonic wave. Surprisingly, even this ratio was enough for efficient FHG using optimally chirped laser pulses. From the reported FHG efficiency, one can calculate the energy of 200-nm radiation obtained in our experiments to be 4.5 μJ.

As for complicatedness of the setup in Ref. [9], it has both positive and negative sides. In particular, it has freedom to vary the delay between two pumps, which allows optimization of the parametric processes. One cannot say that our setup is problem-free and is ideal for FHG. Delaying the two pulses for optimum FHG is not possible in our scheme, but the absence of the delay line is compensated for by introducing chirp. However, this reduces the intensity, the conversion efficiency, and the bandwidth (compared to Ref. [9]).

It is obvious that the difference in the conversion efficiencies reported in our study (10^{-4}) and in Ref. [9] (10^{-3}) is determined by the ratio between the interacting ω and 2ω waves (12:1 in our case and 1:1 in their case). The conversion efficiency in this case is just a technical parameter, which can be considerably improved, particularly by corresponding changes of the waves' ratio, perhaps by putting the second-order harmonic crystal in the unfocused beam, followed by reflective focusing optics. The point to be noted is that, even at this relatively small conversion efficiency, we were able to systematically define the main physical processes, which allow the optimization of FWM in air.

IV. CONCLUSIONS

In conclusion, we have proposed a simplified scheme for efficient generation of 200-nm radiation in isotropic medium using 800-nm radiation. The observation and optimization of the FHG during two-color filamentation in atmospheric air are reported. From the observed scaling exponents, it is shown that this parametric process is based on FWM of the appropriately chirped fundamental and second-order harmonic radiation of Ti:sapphire laser. The fourth-order harmonic conversion efficiency was estimated to be $\sim 10^{-4}$. A systematic study of the influence of laser intensity, polarization, chirp, and pulse duration on the fourth-order harmonic output from the filaments has been carried out.

ACKNOWLEDGMENTS

R. A. Ganeev gratefully acknowledges the support from the TWAS-UNESCO Associateship Scheme. He also thanks Raja Ramanna Centre for Advanced Technology for financial support and the invitation to carry out this work. The authors gratefully acknowledge the help from S. R. Kumbhare, S. Sebastin, R. P. Kushwaha, and the laser team in carrying out this work.

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