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Enhanced harmonic conversion efficiency in the self-guided propagation of femtosecond ultraviolet laser pulses in argon

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ABSTRACT Harmonic generation during the self-guided propagation of femtosecond ultraviolet (UV) laser pulses (248-nm, 450-fs) in argon is investigated. The third (82.7-nm) and fifth (49.6-nm) harmonics are generated in the UV filament. The energy-conversion efficiencies for the harmonics are found to be at least two orders of magnitude higher than those reported in the literature for similar gas pressures. The enhancement is attributed to the quasi-phase matching of the harmonics due to the self-guiding of the driving pulse.

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1 Introduction

Nonlinear propagation of femtosecond laser pulses in gases and associated effects have been intensively investigated during the last decade. Phenomena such as self-focusing, self-phase modulation, filamentation and conical emission have been studied under various conditions [1–5]. One of the most interesting nonlinear phenomena is the generation of secondary coherent radiation sources in the VUV (vacuum ultraviolet) and XUV spectral regions. Such secondary sources find interesting applications, e.g. in XUV interferometry [6] or in the study of molecular dynamics and XUV spectroscopy [7]. The efficient generation of such sources is directly connected to the phase-matching control between the involved light waves. For a recent review on phase-matching control in gases see e.g. [8] and references therein. Lately, approaches using self-guided femtosecond pulses for phase-matching control of harmonic generation have been explored [9–11].

More recently, Aközbek et al. [12] demonstrated, both experimentally and theoretically, self-phase locking between the fundamental and the third (266.7 nm) harmonic in an IR femtosecond filament propagating in air at atmospheric pressure. The third-harmonic pulse maintained both its peak intensity and energy over distances much longer than the characteristic phase-mismatch length. They showed that this is due to a nonlinear phase-locking mechanism between the two pulses in the filament and is independent of the initial material wave-vector mismatch. We will come back to this point with more details later on. For clarity reasons, we notice that the term phase locking (used in [12]) stands for a constant phase difference while, in the case of absolute phase matching, this difference is equal to zero.

In this paper, we report the first demonstration of (quasi-) self-phase matching for the third and the fifth harmonics produced by a UV femtosecond laser filament in argon. Experiments were performed for different gas pressures, between 50 and 1000 mbar. Important conversion efficiencies of the produced harmonics are shown. Numerical simulations reveal the crucial role of the nonlinear propagation in the process.

2 Experimental setup

A schematic diagram of the experiment is shown in Fig. 1. A femtosecond excimer (KrF) oscillator–amplifier system in conjunction with a dye laser [13] is used. It produces linearly polarized pulses at 248 nm with a duration of 450 fs and an energy up to 10 mJ at a repetition rate of 5 Hz. The laser system consists of a double-chamber Lambda-Physik excimer laser and an ultra-short dye-laser system. The XeCl excimer laser oscillator beam pumps a series of dye lasers to produce a sub-picosecond green (496-nm) laser pulse. These pulses are frequency doubled in a nonlinear BBO crystal and amplified twice in the KrF cavity of the excimer laser. Laser diagnostics were performed on-line with a 1-m Littrow-type spectrograph and a multiple-shot autocorrelator at 248 nm. The amplified spontaneous emission (ASE) in the laser output was 10^{-6}:1. The final laser beam had a rectangular profile of 26 mm × 8 mm and a full divergence angle of 0.15 mrad. The energy distribution was flat along the largest dimension and roughly Gaussian along the smallest one.

The laser beam was slightly focused into a 2.5-m-long tube containing the argon gas with a flat–convex lens ($f = 9.4$ m) located 6.3 m from the entrance window of the tube (Fig. 1). Argon was chosen as the best-suited nonlin-

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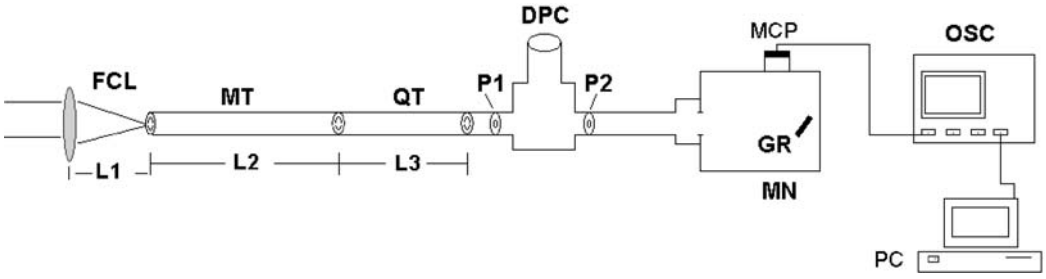

FIGURE 1 Experimental setup. FCL – flat–convex lens ($f = 9.4$ m); MT – metallic tube; QT – quartz tube; P1, P2 – pinholes (200 μm and 1 mm respectively); DPC – differential pumping cell; MN – monochromator (Acton VM502-X); GR – holographic grating coated with Al and MgF_2 (1200 grooves/mm); MCP – multi-channel-plate detector; OSC – oscilloscope; PC – data-storage computer; L1: 6.3 m; L2: 1.5 m; L3: 1.0 m

ear medium for efficient generation of harmonics, especially for the third one [14]. The tube consisted of a 1.5-m-long metallic part and a 1-m-long quartz tube. The argon pressure in the tube could be varied from 50 to 1000 mbar. The produced filament was arrested by a 200-μm pinhole placed at the end of the quartz tube. The pinhole formed the junction to a differential pumping system coupled to a monochromator (Acton VM502-X). The monochromator was equipped with a 1200-g/mm Al and MgF_2-coated holographic grating functioning from 30-nm to 550-nm wavelength range. The signal at the exit of the monochromator was recorded with a single-stage micro-channel plate (MCP) detector connected to a LeCroy 450-MHz digital oscilloscope. The grating and MCP detector were appropriately calibrated for the whole studied spectrum range. A resolution of 2 nm was achieved with a slit of 0.5 mm at the exit of the monochromator.

3 Results

Filamentation of the UV laser pulse in air was demonstrated and studied in previous works [1–3]. For experimental conditions similar to those of the present work, a filament of more than 3-m long was found, starting well in front of the geometrical focus at about 6 m [1, 2]. The fluorescence emission technique [15] was employed to observe filamentation at different gas pressures of interest here. Measurements of the emission spectrum of argon gas in the spectral range from 300 to 450 nm along the 1-m quartz tube, performed perpendicular to the laser axis, showed a plateau profile indicating that filamentation of the UV short laser pulse occurs in argon at various pressures in the range from 50 to 1000 mbar.

A three-dimensional code was used to simulate the propagation of ultra-short UV-laser pulses in argon for different pressures. The code resolves an extended nonlinear Schrödinger equation, coupled with the density of electrons produced by optical field ionization and avalanche. More details about the code can be found in [1, 16]. The main results of the simulations show UV filamentation in argon for pressures in the range 50 to 1000 mbar. Filaments of various lengths are obtained for the different gas pressures. The maximum intensity in the filament is about 10^{13} W/cm^2 and the maximum electron density about 10^{16} cm^{-3}. The laser beam profile at 250 mbar is shown in Fig. 2. The filament is formed at 8 m from the lens and remains robust over more than 2 m.

A typical power spectrum of the fundamental as well as the third and fifth harmonics in argon at 250-mbar is shown in Fig. 3. The relative amplitudes of the three peaks are not representative, as the MCP detector used in our experiment had a quantum efficiency of 2.4×10^{-1} electrons/photon at the third and fifth harmonics but only 9.16×10^{-7} electrons/photon at the laser wavelength. Nevertheless, even after the appropriate corrections were made for the detection system, important conversion efficiencies were revealed for the observed harmonics, 0.02% for the third harmonic and 0.01% for the fifth harmonic. These values are higher by a factor of 10^2 to 10^4 than those reported in the literature under similar experimental conditions, i.e. in a low-pressure gas cell but without filamentation. In the following we will explain the origin of these high conversion efficiencies.

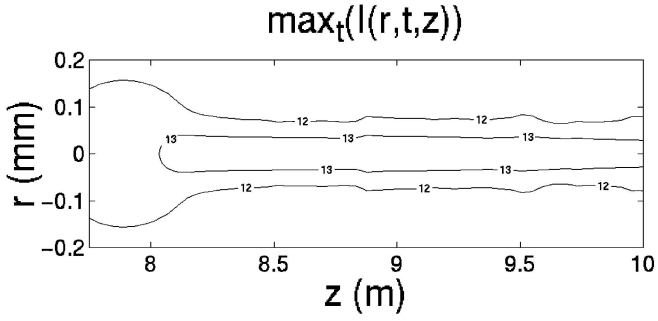

FIGURE 2 Filament spatial profile along the propagation at 250 mbar. The labels indicate the domains where the intensity exceeds 10^{12} and 10^{13} W cm^2

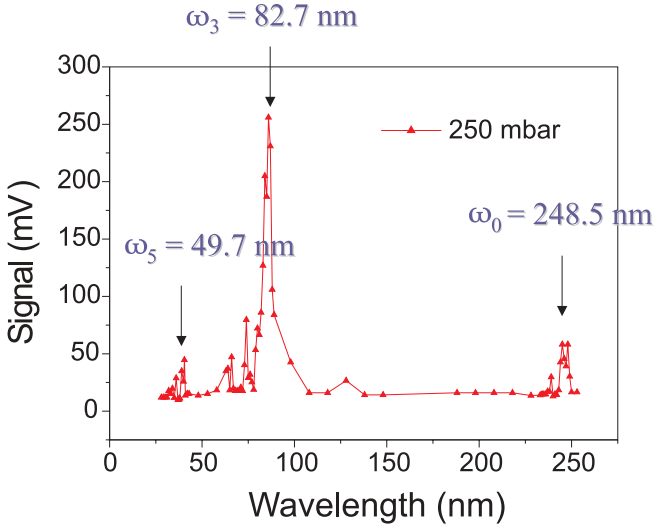

FIGURE 3 Power spectrum with the harmonics after propagation in argon at 250 mbar

A series of harmonic energy measurements have been performed as a function of argon pressure. Results for the third and the fifth harmonics are shown in Fig. 4a and b, respectively. The error bar on the experimental data is estimated to be around 15%, which is mainly due to fluctuations of the laser energy. A sinusoidal-like response is found with half-period of $\Delta P_3 = 0.085$ atm and $\Delta P_5 = 0.095$ atm for the third and the fifth harmonics, respectively. This sinusoidal-form response is well known under the name of Maker fringes [17] and is due to the group-velocity dispersion that limits the constructive superposition of the produced harmonics. In the following it is shown that the observed dispersion is much smaller than expected and actually results from the self-guiding of the driving laser pulse.

As is well known, the intensity of the harmonic depends on the phase matching between the pump and the harmonic waves. Three major effects contribute to the dephasing between the fundamental and the harmonics [18], (a) ionization, (b) the Gouy phase shift on focusing and (c) the phase change due to atomic group velocity dispersion. For our experiment the dephasing due to ionization is negligible, because of the relatively low electron density, which has been measured previously [1, 2] and calculated by numerical simulations in our present experimental conditions to be less than $10^{17}\,\mathrm{cm}^{-3}$. Therefore, the total phase mismatch Δk for the qth-harmonic generation mainly depends on the atomic dispersion and on the Gouy phase shift and can be expressed as [19]

$$\Delta k = \Delta k_{\mathrm{disp}} + \Delta k_{\mathrm{geom}}\,,$$

where (a) $\Delta k_{\mathrm{disp}} = k_q - qk_1$ is the dispersive part, with $k_1 = n_1\omega_1/c$ (n_1 the refractive index of the fundamental, ω_1 the frequency of the fundamental and c the speed of light in vacuum) and $k_q = n_q\omega_q/c$ (n_q the refractive index of the qth harmonic and ω_q the frequency of the qth harmonic) and (b) $\Delta k_{\mathrm{geom}} = (1-q)\tan^{-1}(z/b)$, where z denotes the direction of propagation and b is the confocal parameter, which is given by $b = 2\pi r_{\mathrm{o}}^2 n/\lambda_{\mathrm{o}}$ (where r_{o} is the radius of the beam at focus, n is the refractive index of the fundamental and λ_{o} is the fundamental wavelength). In our calculations, the values of Δk_{geom} as a function of pressure were found to be three orders of magnitude smaller than Δk_{disp}; thus Δk_{geom} can also be neglected in the calculations and thus one can simply admit $\Delta k \approx \Delta k_{\mathrm{disp}}$. To calculate this dispersion one needs to know the index of refraction that is calculated using a three-pole Sellmeier equation:

$$\frac{3}{2}\frac{n_i^2-1}{n_i^2+2} = \frac{C_1}{\lambda_1^{-2}-\lambda_i^{-2}} + \frac{C_2}{\lambda_2^{-2}-\lambda_i^{-2}} + \frac{C_3}{\lambda_3^{-2}-\lambda_i^{-2}}\,, \tag{1}$$

where n_i is the index of refraction at the wavelength λ_i at one atmosphere. In the calculation, for argon, three resonances are included at $\lambda_1 = 106\,\mathrm{nm}$, $\lambda_2 = 104.8\,\mathrm{nm}$ and $\lambda_3 = 60.9\,\mathrm{nm}$. The values of the constants are $C_1 = C_2 = 25.2528\times10^4\,\mathrm{cm}^{-2}$ and $C_3 = 595.379\times10^4\,\mathrm{cm}^{-2}$.

Now, let us retrieve the relation of the third-harmonic intensity as a function of gas pressure as in the experimental procedure. The third-harmonic intensity is given by

$$I_3(z) = \frac{\omega_3^2}{n_3 n_1^3 c^4 \varepsilon_0^2}\left|\chi^{(3)}\right|^2 I_1^3 \frac{\sin^2(\Delta k z/2)}{(\Delta k/2)^2}\,, \tag{2}$$

where $I_1(r,t) = I_{10}\,\mathrm{e}^{-r^2/r_{\mathrm{o}}^2}\,\mathrm{e}^{-t^2/\tau_{\mathrm{o}}^2}$ is the input intensity for a Gaussian laser pulse, $\chi^{(3)}$ is the third-order susceptibility, Δk is the phase mismatch and z is the interaction length.

Relation (2) can be expressed as a function of pressure by replacing $\Delta k = \Delta k_0 P$ and $\chi^{(3)} = \chi_0^{(3)} P$, where $P = P_i/P_0$ with P_i the given pressure (in mbar) and $P_0 = 1000\,\mathrm{mbar}$. Then one has

$$I_3(z) \propto \left|\chi_0^{(3)}\right|^2 P^2 \frac{\sin^2(\Delta k_0 P z/2)}{(\Delta k_0 P/2)^2}\,. \tag{3}$$

Relation (3) goes through minima and maxima whose amplitude does not depend on the gas pressure. The pressure difference between a minimum and a maximum is then given by

$$\Delta P = \frac{\pi}{\Delta k_0 z} = \frac{L_{\mathrm{d3}}}{z}\,,$$

where $L_{\mathrm{d3}} = \pi/\Delta k_0$ is the phase-mismatch length. Using the values for the dispersion of argon calculated by the Sellmeier equation (1) one has $L_{\mathrm{d3}} = 0.5$ mm and $L_{\mathrm{d5}} = 0.032$ mm. The last parameter left in the above expression is the interaction length. In our experiment the interaction length coincides with the filament length, which was at least 1 m. Nevertheless, in the case of a pulsed laser source (like fs pulses) an effective interaction length has to be defined, which is limited by the coherence length, L_{coh}, of the source ($L_{\mathrm{coh}} > z > L_{\mathrm{d}}$). If $\Delta\lambda$ is the spectral width of the harmonic, the coherence length L_{coh}

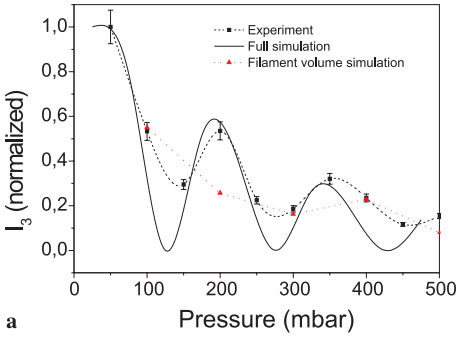

FIGURE 4 The third (**a**) and fifth (**b**) harmonic intensities as a function of pressure (see text)

is defined as

$$\frac{2\pi}{\lambda}\Delta n L_{\mathrm{coh}} = \frac{2\pi}{\lambda+\Delta\lambda}\Delta n L_{\mathrm{coh}} + \frac{\pi}{2}\,,$$

$$\frac{\Delta\lambda}{\lambda} = \frac{L_{\mathrm{d}}}{2L_{\mathrm{coh}}}\,.$$

In our case $\Delta\lambda = 1$ nm and $L_{\mathrm{coh3}} = 62$ mm; $L_{\mathrm{coh5}} = 2$ mm. If we assume that these values limit the interaction length, we obtain a system of Maker fringes with half-period $\Delta P_3 = 0.008$ atm and $\Delta P_5 = 0.015$ atm, while experimentally we have $\Delta P_3 = 0.085$ atm and $\Delta P_5 = 0.095$ atm. There is a factor 10 in the estimated and observed periods, in the sense that the measured mismatch is smaller.

These results suggest (partial) phase-matching conditions in our experiment. Indeed, if the phase mismatch is taken 10-times smaller the experimental results are reproduced with high accuracy, as represented by the solid lines in Fig. 4a and b. This is the first experimental demonstration of (partial) phase matching in harmonic generation using an ultraviolet self-guided driving pulse (filament).

As mentioned in the introduction the same phenomenon has been previously observed in infrared laser pulse filamentation by Aközbek et al. [12]. In order to illustrate the key physical elements that lead to this effect, let us examine the amplitude variation of the laser electric field in the presence of the third harmonic. This can be expressed in a simplified form as

$$\frac{1}{2}\frac{\partial A_\omega^2}{\partial z} = -\frac{1}{4}\nabla_\perp\left(A_\omega^2\nabla_\perp\phi_\omega\right) + \frac{1}{2}I_0 n_2 w_0^2 k_\omega^2 A_\omega^3 A_{3\omega}\sin\left(3\phi_\omega - \phi_{3\omega}\right)\,,$$

where A_ω, $A_{3\omega}(\phi_\omega, \phi_{3\omega})$ are the amplitudes (slowly varying phases) of the fundamental and the third harmonic, respectively, n_2 is the nonlinear index of refraction and w_0 is the beam waist. Now, it has been proven both experimentally and numerically that the intensity in the filamented pulse is clamped due to the dynamic-equilibrium between Kerr self-focusing and plasma defocusing [10, 20], that is: $\partial A_\omega^2/\partial z \approx 0$. Also, the wavefront in the filament is nearly flat [10], that is: $\nabla_\perp\phi_\omega \approx 0$. Thus, in order to preserve the intensity clamping in the presence of the third harmonic one must have $\sin(3\phi_\omega - \phi_{3\omega}) \approx 0$. This means that the phase difference between the fundamental and the harmonic is maintained. The phase difference is also independent of the medium, as experiments ([12] and the present work) in both air and argon prove. For a more detailed analysis the reader is invited to refer to [12].

A last point that deserves our attention is the observed attenuation in the amplitude of the oscillations in Fig. 4a and b as a function of the gas pressure. This is an original result which can easily be explained by the variation of the filament diameter, and in consequence the filament volume, with gas pressure. This point is crucial as for a given interaction length, the effective volume in which harmonics are produced changes. Our propagation code shows a reduction of the filament diameter as the pressure increases and reproduces well the observed harmonic amplitude attenuation as shown by the triangle-dotted line in Fig. 4a.

4 Conclusion

In conclusion, we have provided the first experimental demonstration of (partial) phase matching in the generation of the third (82.7-nm) and fifth (49.6-nm) harmonics produced by an ultraviolet self-guided femtosecond laser pulse (248 nm, 450 fs) in argon. Numerical simulations of the filamentation itself and the resulting harmonic generation reproduce well the experiments. Intensity clamping in the filamented laser pulse is the key element for the observed (partial) phase matching and the consequent increased conversion efficiency in the harmonic generation. The efficient generation of XUV radiation, shown here, can find various applications, for example, in XUV spectroscopy and interferometry.

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