Below Threshold Harmonics Dependence with Phase-Matching Parameters in Argon

Armando V. F. Zuffi
IPEN – CNEN/SP
São Paulo, Brazil
armandozuffi@gmail.com

Andreia A. Almeida
IPEN – CNEN/SP
São Paulo, Brazil
aalmeida.fis@gmail.com

Nilson D. Viera Jr.
IPEN – CNEN/SP
São Paulo, Brazil
nilsondiasvieirajr@gmail.com

Ricardo E. Samad
IPEN – CNEN/SP
São Paulo, Brazil
resamad@gmail.com

Abstract—This work reports the generation of Below Threshold Harmonics generated in Argon by ultrashort laser pulses. The 3rd, 5th, 7th and 9th harmonics of the laser were generated in the UV and VUV regions, and the frequency conversion occurred in flowing gas in a glass nozzle into a vacuum chamber. The harmonics generated show a blue-shift and an efficiency that depend on the laser pulses intensity and also on the gas pressure. These parameters determine the phase-matching condition, which governs the energy conversion from the laser into the harmonics, and this study shows that is possible to tune the harmonics intensity and wavelength, providing a versatile light source.

Keywords—nonlinear optics, harmonic generation, ultrafast optics, plasma physics, ultrashort pulses.

I. Introduction

Below Threshold Harmonics generation techniques (BTH) have been studied for having enormous potential for coherent light generation in the VUV region [1-3] which is of fundamental interest for time-resolved spectroscopy [4, 5], materials processing, precision manufacturing and surface physics [6-8]. Moreover, the harmonic generation below the ionization threshold of gases has been shown to be an important technique for understanding the electronic dynamics close to the gas ionization energy [9, 10].

The BTH generation in gases demands laser intensities higher than in solids due to the density of generating centers that is two orders of magnitude smaller [11]. However, coherent light generation at wavelengths below 200 nm in solids become impractical because the solids become opaque in this spectral region [12]. The harmonic generation in solids below 200 nm is only possible using sophisticated techniques [13-15]. On the other hand, in noble gases, the absorption edge is located at its 1st ionization energy which allows the BTH generation up to 50.4 nm in helium, as an example [16].

In the BTH generation an abrupt decrease of the generation efficiency occurs with the increase of the harmonic order. Therefore, intensities over tens of GW/cm² are needed to generate the 5th and higher order harmonics, which are easily achieved by ultrashort laser pulses. Commonly available CPA laser system [17] reach intensities of TW/cm² with modest focusing. At these intensities, the laser pulses ionize a portion of the gas during its propagation and begin to interact with the generated plasma, which can reduce harmonic generation efficiency. Therefore, the laser pulse intensity has to be balanced to efficiently generate the harmonics without wasting energy in plasma generation. An alternative is to work inside a vacuum chamber, restricting the presence of the gas only to the region of the harmonics generation. Thus, ultrashort laser pulses interact with the gas in a nozzle a few millimeters long [18].

The maximum efficiency in the harmonic generation occurs in phase-matching condition [19] that is when the wavefronts of the laser and harmonic fields remain in phase in the region of harmonic generation. Consequently, the harmonic fields suffer a constructive interference in the nonlinear media, making the conversion of energy into efficient generation. The phase difference between laser and the q^{th} harmonic field is given by the phase-mismatch term, Δk_q, which, for BTH generated in gases by focused ultrashort laser pulses is [20-22]:

$$\Delta k_q = -2\pi q\, p\Delta\delta\, (\eta - 1) / \lambda_0 - \eta\, p\, N_{atm}\, r_e\, \lambda_0\, (q - 1/q) + q / (z_R + z^2 / z_R) \qquad (1)$$

where q is the harmonic order, p is the gas pressure in atm, $\Delta\delta = \delta_0 - \delta_q$ is the difference between the refractive index of the laser and the q^{th} harmonic per unit of pressure (in atm), η is the ionized gas fraction, λ_0 is the laser wavelength, N_{atm} is the density of atoms at 1 atm, r_e is electron radius, z_R is the laser Rayleigh length and z is the laser beamwaist position with respect to the nonlinear medium center. The first term on the right side in equation (1) is related to the neutral gas dispersion, the second is caused by the dispersion of the plasma, and the third is the Gouy phase [22]. BTH phase-matching can be achieved by balancing these terms until the condition $\Delta k_q = 0$ is reached.

In practice, attaining phase-matching in a thick nonlinear medium is difficult because the mismatch increases with the medium length. In the condition $\Delta k_q \neq 0$, the distance that the q^{th} harmonic field must propagate in the nonlinear medium to have its highest efficiency is [11, 22]:

$$L_{C,q} = \pi/\Delta k_q \qquad (2)$$

where $L_{C,q}$ is the coherence length for the q^{th} harmonic. Therefore, for efficient harmonic generation a nonlinear medium length of $L_{C,q}$ is desirable. Those properties show that the harmonic generation efficiency depends not only on the magnitude of the nonlinearity (microscopic properties) [11], but also on the macroscopic properties associated with the propagation of the laser beam and the harmonic fields.

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In this work we report the generation of BTH in flowing Argon by ultrashort laser pulses. The harmonics were identified and characterized. The BTH dependence with laser intensities and gas pressure was studied in order to balance the conditions for phase-matching from the ionized gas fraction, η, in (1). Given that, it was possible to observe effects that interfere and modify the harmonic generation, such as plasma induced blueshift [23]. That mapping of the BTH generation parameters was fundamental to improve the understanding of the phenomena involved, as well as providing a light source in the VUV region for future applications.

II. Experimental Setup

The BTH were generated by ultrashort laser pulses from a Ti:Sapphire CPA laser system. The laser system is composed by a main oscillator (Femtolasers Rainbow pumped by a Coherent Verdi V6 laser) that generates pulses with approximately 1.5 nJ and 6 fs FWHM (Full Width at Half Maximum), at 78 MHz repetition rate, with 370 nm of bandwidth at -10 dB. The pulses generated by this oscillator are injected into the CPA amplifier (Femtolasers Femtopower Compact Pro HR/HP). The amplification process produces pulses centered at 781 nm with about 35 nm bandwidth (FWHM), up to 650 µJ, 25 fs (FWHM), at 4 kHz repetition rate in a laser beam with a M^2 factor of 1.5. The CPA laser system is pumped by a Q-Switched Nd:YLF laser (Photonics Industries DM30-527) that emits up to 10 mJ, 300 ns laser pulses at 4 kHz repetition rate.

The harmonics are generated inside a cylindrical vacuum chamber with an internal diameter of 50 cm, a volume of approximately 5×10^4 cm^3, and several accesses for beam injection and instrumentation, as shown in the Fig. 1 scheme. The vacuum is made by a TMH 521/TMU 521 turbomolecular pump, backed by a mechanical pump, producing final pressures under 10^{-6} mbar inside the chamber. To generate the harmonics, the laser pulses are focused by a 50 cm focal length lens (plano-convex singlet, 50.8 mm in diameter) in a gas nozzle that is inside the vacuum chamber. With this and considering that the transverse profiles of the laser beam are Gaussian, the beamwaist size at the focus is about 35 µm [24]. Thus, the laser system can reach intensities higher than 10^{14} W/cm². The nozzle consists of a 2.3 mm inner diameter glass capillary that is etched with ultrashort pulses in a homemade machining system to reduce its wall thickness, so it can be drilled by the same beam that generates the harmonics. This determines in the nozzle an interaction length of 2.7 mm (assuming that the gas pressure drops abruptly outside the nozzle), as shown in Fig. 2, bored by the laser on diametrically opposite spots previously machined. Argon flows through the nozzle at a 100 mbar pressure and the laser enters and exits the interaction region [22, 25]. The harmonics wavelengths can be filtered out by a glass plate that can be placed after the nozzle by vacuum compatible displacement actuators controlled by a computer (Newport NSA12V6). The glass plate works like a contrast technique to observe harmonics with higher signal-to-noise ratio, such as 9th harmonic [22, 24].

The harmonics are separated by a VUV monochromator (McPherson 234/302), which covers the spectral range from 30 to 300 nm and has a concave holographic diffraction grating with 200 mm focal length, aperture f/4.5 and resolution of 1 Å at 584 Å. The detection is made by a photomultiplier tube (PMT) with a scintillator coated with sodium salicylate. The harmonics signal is sent to a Lock-In amplifier and stored on a computer.

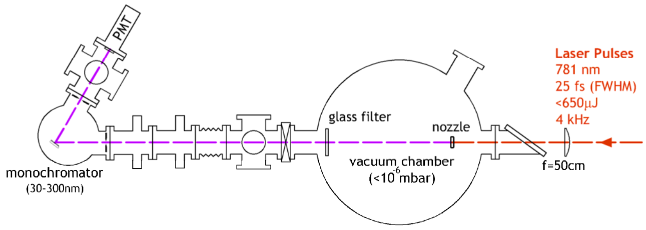

Fig. 1. Scheme of the experimental setup in which the harmonics were generated and measured.

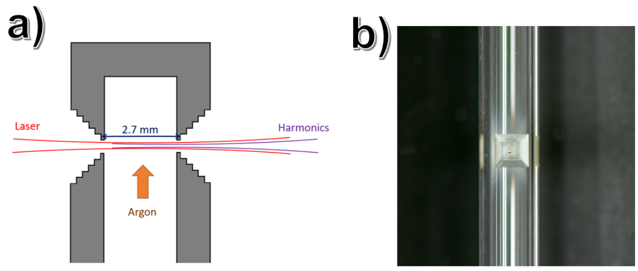

Fig. 2. a) Scheme of the glass nozzle with lateral view showing the laser-gas interaction region. b) Longitudinal view of the glass nozzle after machining and drilling by ultrashort laser pulses.

III. Results

A. Spectral characterization of the BTH in Argon

As previously discussed in Zuffi *et al* [24], all BTH supported in Argon (3rd, 5th, 7th and 9th) were generated and spectrally characterized. These harmonics were obtained for a laser intensity of 3.4×10^{14} W/cm² and a pressure of 100 mbar in the nozzle. From these parameters, the positions of the spectral centers of mass of the harmonics were 258.98, 154.52, 109.88, 86.84 nm, for 3rd, 5th, 7th and 9th, respectively. Those experimental values present a blueshift of about 1% with respect to theoretical values from the laser spectral center of mass given by $\lambda_{CM,q} = \lambda_{CM,0}/q$. This displacement is a consequence of the plasma induced blueshift [22, 23], as verified by the expression of the maximum percentual displacement in [22]:

$$\delta\omega_{max} = \sqrt{[m\, ln(2) / \pi]\, L\eta p N_{atm} / (c\tau_p N_{crit})} \qquad (3)$$

where L is the interaction length of the nozzle, η was assumed to be 1% [26], c is the speed of light in vacuum, τ_p = 25 fs is the pulse duration (FWHM), N_{crit} is the critical density of free electrons in the medium [27], which is 1.8×10^{21} cm^{-3} for Argon, and m is the multiphotonic absorption process order; it is assumed that ionization occurs only by a multiphotonic absorption of order 10. Such consideration is plausible since the Keldysh parameter [28] under our experimental conditions was found to be 0.8, indicating that the ionization occurs preferentially by this process instead of tunneling. The expression (3) is directly proportional to the ionization fraction, η. Therefore, the blueshift increases along with the laser intensity at the nozzle position. This can be seen in Fig. 3 where BTH spectra from 80 to 280 nm measured with different pulses intensities are shifted relative to each other. The spectra are normalized by the 3rd harmonic intensity, and the purple spectrum, obtained with 3.4×10^{14} W/cm² is blue shifted relative to the one obtained at 1.8×10^{14} W/cm², both generated at 100 mbar of pressure in the nozzle. Besides that, the Table I shows the comparison of the spectral centers of mass between the 3rd, 5th and 7th harmonic for these two laser intensities.

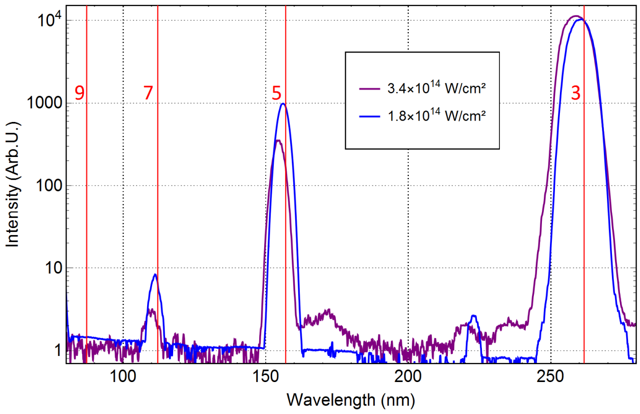

Fig. 3. Spectra of the BTH generated in Argon with laser intensity of 1.8×10^{14} (blue line) and 3.4×10^{14} W/cm² (purple).

TABLE I. BTH SPECTRAL CENTERS OF MASS COMPARISON

order	*λ_{CM} (nm)* *3.4×10^{14} W/cm²*	*λ_{CM} (nm)* *1.8×10^{14} W/cm²*
3rd	258.98 ± 0.01	260.30 ± 0.01
5th	154.52 ± 0.01	156.06 ± 0.01
7th	109.88 ± 0.01	111.11 ± 0.01

The 3rd, 5th and 7th harmonics can be clearly seen in Fig. 3, and their intensities are observed to rapidly decline with the harmonic order increase due to microscopically properties [8]. The 9th harmonic signal could not be seen and must have been masked by scatterings of the laser wavelength on the detection system [22, 24]. Therefore, the 9th harmonic behavior was not studied.

B. BTH dependence on parameters related to phase-matching in Argon.

Fig. 3 also reveals that the harmonics relative intensities change with different laser intensities. These differences must be a consequence of the macroscopic properties associated with the propagation of the laser and harmonic fields. According to the macroscopic properties, the coherence length and its relation to the interaction length of the nozzle are essential for the BTH efficient generation. Thus, the coherence lengths were estimated for the 3rd, 5th and 7th harmonic by expression (1). For this, the same parameters assumed in the expression (3) in the maximum percentage displacement of the blueshift were used, along with z = 0 and z_R = 3.5 mm. The refractive indexes per unit of pressure for the 3rd and 5th harmonics wavelengths in Argon were obtained in the literature [29], and the 7th harmonic value was estimated by an extrapolation of the Sellmeier formula [22, 30, 31]. Table II shows the spectral centers of mass, refractive indexes per unit of pressure and coherence lengths for the 3rd, 5th and 7th harmonics. Fig. 4 illustrates each harmonic generation efficiency by coherence length in the nozzle interaction length, with solid lines indicating the intensity effectively achieved during propagation within the nozzle and dashed lines, its projection for longer interaction lengths (longer nozzles).

TABLE II. BTH COHERENCE LENGTHS ANALYSIS

order	*λ_{CM} (nm)*	*δ_q (atm^{-1})*	*$L_{c,q}$ (mm)*
laser	781.00	1.0000276	
3rd	258.98	1.0000298	3.7
5th	154.52	1.0000360	2.3
7th	109.88	1.0000540	1.7

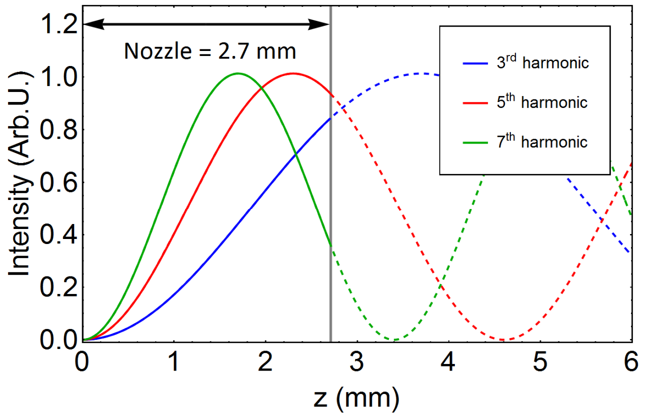

Fig. 4. Harmonic generation efficiency by coherence length in the nozzle interaction length.

For the parameters assumed, an efficient generation of the 3rd and 5th harmonic occurs because the coherent length is close to the interaction (nozzle) length. For the 7th harmonic the generation occurs with about 40% of the maximum generation efficiency. Note that several conditions have been assumed, especially that the ionization fraction is 1%, which is a sensitive parameter for control through the balance of the gas pressure in the nozzle (density of generating centers) and the laser intensity. These experimental parameters control both macroscopic and microscopic properties of the harmonic generation. Thus, the next step of this work is an investigation of the harmonic generation dependence with laser intensity and pressure in the nozzle.

C. BTH dependence with laser intensity

The spectra of each harmonic for different laser intensities were measure, as exemplified in Fig. 5 for the 3rd harmonic. The gas pressure in the nozzle was kept constant in 100 mbar for all harmonics.

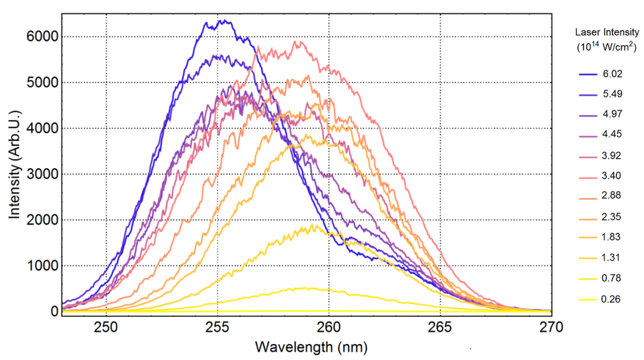

Fig. 5. 3rd harmonic spectra dependence with laser intensities (legend on the right of the graph).

The position of the spectral center of mass, λ_{CM}, and area of the harmonic spectrum (proportional to the harmonic energy) were determined using the measured spectra. Fig. 6 presents these analyses for the 3rd, 5th and 7th harmonics.

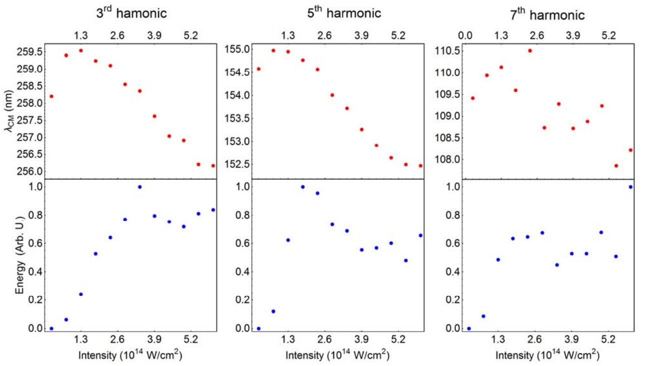

Fig. 6. Dependence of the spectral center of mass (red) and energy (blue) with laser intensities for 3rd, 5th and 7th harmonic generated in Argon.

The plasma induced blueshift is observed in each harmonic by the displacement of the spectral center of mass (in red). The total displacements observed were about 4.0, 2.5 and 2.0 nm for the 3rd, 5th and 7th harmonics, respectively. Although a redshift was observed for the lower intensities, we attribute this discrepancy to large noise/signal ratios that prevent the center of mass to be accurately determined, and to inhomogeneous self-phase modulation [11, 32] that at low intensities should be a more predominant mechanism than the plasma blueshift. The self-phase modulation should also be responsible for the bandwidth decrease with laser intensity grow, as shown in Fig. 5 for the 3rd harmonic. In addition, the 7th harmonic results have a greater dispersion of data that is due to the large noise/signal ratio.

The harmonic energy analysis (in blue) clearly shows a saturation for all harmonics, indicating a competition between the various nonlinear processes that occur in the gas. The saturation was observed at different intensities for different harmonics. This is related to phase-matching conditions and shows harmonic generation efficiency dependence with the macroscopic properties.

D. BTH dependence with gas pressure in the nozzle

Fig. 7 shows the position of the spectral center of mass and area of the harmonic dependence with gas pressure in the nozzle for the 3rd, 5th and 7th harmonics. In these measurements the laser intensity was kept constant in 3.4×10^{14} W/cm² in the nozzle position for all harmonics.

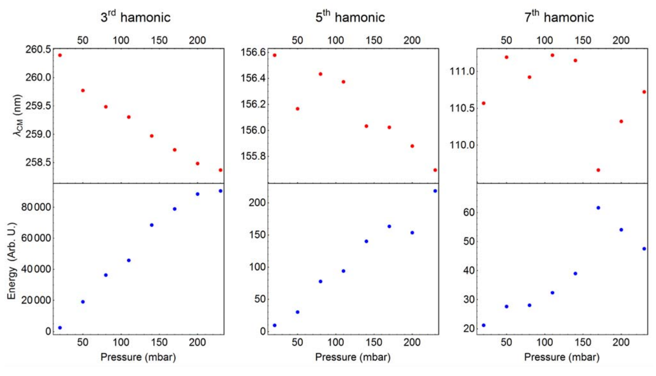

Fig. 7. Dependence of the spectral center of mass (red) and energy (blue) with gas pressure for the 3rd, 5th and 7th harmonics generated in Argon.

Increasing the gas pressure in the nozzle raises the generating centers density, which has a similar effect as increasing the laser intensity. Therefore, the results shown in Fig. 7 are similar to the ones on Fig. 6. The plasma blueshift induced was observed in the spectral center of mass analysis (in red). However, the displacements, of about 2.0, 1.0 and 1.0 nm for the 3rd, 5th and 7th harmonics, respectively, are smaller than the ones observed when changing the laser intensity. Moreover, the 7th harmonic results in Fig. 7 have greater dispersion than the ones shown in Fig. 6, with a less clear trend. With respect to the harmonic energy analysis (in blue), the saturation tendency is only observed close to 230 mbar, the maximum pressure used in the experiments. Although this saturation is poorly evidenced in our results, we believe that at pressures above 230 mbar the generation efficiency will drop, like what happens with HHG [33].These results confirm that the macroscopic properties are essential for generating the BTH efficiently.

IV. Conclusions

In this work we explored a significant part of the theoretical and practical knowledge for the BTH generation in Argon. This approach is poorly explored in the literature relating the plasma effects in the BTH generation. Moreover, we performed a mapping of the BTH dependence with the laser intensity and gas pressure, both essential parameters for controlling the phase-matching. Finally, we have shown that is possible to tune both the BTH wavelength, through the induced plasma blueshift, and the generation efficiency by varying these parameters. Unfortunately, higher intensities and pressures could not be studied due to equipment limitations.

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