Pressure dependence of high order harmonic generation in nitrogen molecular gas and atmospheric air

M. Sayrac, A.A. Kolomenskii, H.A. Schuessler

Department of Physics, Faculty of Science, Cankiri Karatekin University, Cankiri, Turkey
Department of Physics & Astronomy, Texas A&M University, College Station TX 77843-4242, USA
Science Program, Texas A&M University at Qatar, Doha 23874, Qatar

Abstract

The effect of the variation of the gas pressure on the high harmonic generation (HHG) from nitrogen molecular gas (N₂) and atmospheric air with ultrashort intense laser pulses is measured. The optimum pressure in the interaction region of a gas jet for maximizing the HHG yield is determined for both gases. Enhancement of the HHG output and its extension to higher harmonic orders are observed around the optimum pressure value of 0.33 bar. Theoretical calculations based on one-dimensional models explain this effect and provide reasonable agreement with experimental results.

1. Introduction

An efficient source of extreme ultraviolet (XUV) radiation is a much needed tool for imaging of matter and spectroscopic studies with short wavelength radiation. A promising approach to generate such radiation is by high harmonic generation (HHG). HHG is a unique nonlinear process, in which atoms/molecules are ionized by an intense laser field with a frequency ω₀, and as a result, radiation at higher frequencies qω₀ is produced, where q is an odd integer. HHG driven by IR lasers can span a frequency range from UV to soft X-rays [1,2], and it has various applications in physics, chemistry, and biology [2,3]. HHG is usually realized by using amplified femtosecond laser pulses that are generated with a table top laser system. For this reason HHG is considered as a major approach of obtaining a table top source of coherent XUV radiation [4].

The HHG phenomenon can be understood in a semi-classical picture [5,6]. An electron is initially tunnel-ionized by an intense electromagnetic field of a laser pulse, and the freed electron is accelerated in the oscillating laser field and gains kinetic energy. If the electron recombines with the parent ion, it can release its kinetic energy, emitting harmonics of the fundamental field. Bright harmonic radiation is emitted, if the emission from many atoms in the medium adds constructively, i.e. phase-matched [7]. One of the important advantages of the HHG source is that the XUV emission is perfectly synchronized to the driving laser field. Consequently, one can obtain a train of sub-femtosecond pulses of coherent light separated by half of the driving laser cycle, since high harmonic (HH) fields are phase-locked [8]. The maximum HH order is determined by the cutoff energy, which depends on the ionization potential (Iₚ) of the used atomic system, and the ponderomotive energy Uₚ(eV) = 9.33 × 10⁻²⁰ × I × λ², where I(W/cm²) is the laser intensity, and λ (nm) is the fundamental wavelength. Uₚ refers to the average kinetic energy of a free electron, gained in the oscillating laser electric field [6], and the cutoff energy is given as Eₘₐₓ = Iₚ + 3.17Uₚ [9–11].

The HHG sources, possessing such advantages as compactness and availability to researchers, have also a disadvantage, i.e. the...
conversion efficiency of the fundamental radiation into HHs is remarkably low, in the range of $10^{-7} - 10^{-5}$ per harmonic [12,13]. Thus, the possibility to generate a high density photon flux is limited by this low conversion efficiency. It can be further exacerbated by the mismatch of phase velocities of the fundamental and harmonic fields (frequency dispersion) and a large absorption in the medium [14]. In addition, a geometrical phase shift (Gouy phase) arises when the laser beam is focused into gas medium [15]. The above mentioned de-phasing effects and absorption set limitations for the efficient harmonic generation.

In this study, enhancement and extension of the HHs yield by using a differentially pumped gas cell are presented. Depending on the phase matching conditions, the HHs output can be suppressed or enhanced. The HHG in N$_2$ and atmospheric air for various interaction pressures are measured, and both gases present similar pressure dependences, since N$_2$ is a major component of the atmospheric air (about 78% N$_2$). In this experiment, the gas jet is enclosed in a differentially pumped cell to enable measurements over a wide range of pressure values. The mechanism of pressure dependence of HHG is elucidated by using one dimensional models.

2. Experimental setup

We use a Ti:Sapphire laser system that produces 800 nm, 50 fs pulses of up to 1 mJ pulse energy with a repetition rate of 1 kHz. The laser beam is focused by a 40 cm focal length lens reaching the peak intensity at the focus of $1.5 \times 10^{14}$W/cm$^2$. Harmonics are generated in a gas jet (GJ) produced by burning holes in a squeezed 1 mm diameter nickel (Ni) tube. The GJ is enclosed in a cell, and the main portion of the ejected gas is removed by an additional roughing pump (Oerlikon, SC 30D), which helps to reach relatively high pressures in the interaction region (R2), as shown in Fig. 1. The differential pumping is created between the region of high pressure R2 and the main chamber, region R1. Thus, the additional roughing pump is removing the main portion of the gas directly from the gas jet cell (region R3) before it reaches region R1. The focused laser beam passes through a 1.5 mm input hole (IH) and an output hole (OH1) of the cell. The cell has also a 1.5 mm tube hole (TH) on the top for the Ni tube, sealed at the lower end. Produced high harmonics are detected using an XUV spectrometer (McPherson, 248/310 G). In this spectrometer, XUV radiation is diffracted by a grating (133.6 groves/mm) and then imaged onto a micro-channel plate (MCP) backed by a phosphor screen. The image from the phosphor screen, mounted at the back side of the MCP, is projected onto a charge-coupled device (CCD) camera.

3. Experimental results

The HHG yield is measured with enabled differential pumping (the valve from the space R3 to the roughing pump is opened, Fig. 1; the valve itself is not shown in the figure). The HHs spectra for N$_2$ are shown in Fig. 2. The maximum pressure in the interaction region (R2) is reaching up to 2 bar, and HHs from 15th to 25th orders are observed, as presented in Fig. 2. The maximum HH output is achieved at the optimum pressure of 0.33 bar for all harmonics, as is shown in Fig. 3. Around the optimum pressure value (~0.33 bar), the generation of the 25th harmonic is observed, as can be seen in Fig. 2, which was not observed outside of the optimum pressure region.

The maximum pressure of up to 2 bar is reached with the atmospheric air filled gas jet, and the observed HHs from 15th to 25th order are shown in Fig. 4. The maximum HH output is achieved at the optimum pressure of 0.33 bar for all harmonics, as shown in Fig. 5. Around the optimum pressure value, again the generated 25th harmonic appears, as is seen in Fig. 4(a, b).

Since the air mostly consists of nitrogen (I$_p$(N$_2$) = 15.6 eV [16]), the experimentally observed cutoff for N$_2$ and atmospheric air both take place at 25$^{th}$ order (~38.7 eV). Also, the abovementioned formula for the cutoff energy gives the cutoff for N$_2$ ~ 40.5 eV (~31 nm), which is in close agreement with the experimental results. Some discrepancy between theoretical and experimental results can be due to imperfections of the optics and the divergence of the laser beam, which cause the lower laser intensity and lower HH yield than is expected in the medium [10,17].
Fig. 2. Results for N$_2$: (a) raw image of the HH spectrum (from 15th to 23rd) taken by the CCD camera. (b) HHG spectra at three different pressures. The extension of the cutoff to 32 nm (25 H) is noticeable in (b) at pressure value of 0.33 bar.

Fig. 3. Yields of HHs vs. gas jet pressure in the interaction region for N$_2$ gas. HHs from 15H to 25H are observed. The optimum pressure value is found around 0.33 bar.

Fig. 4. Results for air: (a) raw image of the HH spectrum (from 15th to 25th) taken by the CCD camera. (b) HHG at three different pressures in the gas jet. The extension of the cutoff to 32 nm (25 H) is noticeable in both the raw data (a) and in the plotted spectrum (b) at pressure value of 0.33 bar.
Theoretical calculations and comparison to experiment

The HHG output depends on the gas pressure, absorption and phase matching conditions, and it can be modelled by the one dimensional description of Ref. [14]

\[
S_{HH} \propto \frac{4L_{coh}^2 P^2 A_q^2}{1 + 4\pi^2 \left( \frac{L_{med}}{L_{coh}} \right)^2} \times \left( 1 + e^{-L_{med}/L_{abs}} - 2\cos \left( \frac{L_{med}}{L_{coh}} \right) e^{-L_{med}/L_{abs}} \right)
\]

(1)

Fig. 5. Yield of HHs from 15H to 25H as a function of the gas jet pressure in the interaction region observed for atmospheric air. The optimum pressure value is around 0.33 bar. The appearance of the 25th harmonic is observed around the optimum pressure value.

4. Theoretical calculations and comparison to experiment

The HHG output depends on the gas pressure, absorption and phase matching conditions, and it can be modelled by the one dimensional description of Ref. [14]

\[
S_{HH} \propto \frac{4L_{coh}^2 P^2 A_q^2}{1 + 4\pi^2 \left( \frac{L_{med}}{L_{coh}} \right)^2} \times \left( 1 + e^{-L_{med}/L_{abs}} - 2\cos \left( \frac{L_{med}}{L_{coh}} \right) e^{-L_{med}/L_{abs}} \right)
\]

(1)

It uses the assumption that the gas density as well as the induced dipole are constant along the segment \(L_{med}\) occupied by the uniform medium (for calculations with this model we used \(L_{med} = 0.4\) mm), where the HHG takes place. Consequently, we refer to this model as a "segment source model". Here, \(S_{HH}\) is the number of HH photons emitted per unit time, \(P\) is the gas pressure in the region 2 (R2), \(A_q\) is the dipole amplitude of the \(q^{th}\)-order harmonic; \(L_{abs}\) is the absorption length, and \(L_{coh}\) is the coherence length \(L_{coh} = \pi/|\Delta k|\), where the wave vector mismatch \(\Delta k\) between infrared and XUV light is given as

\[
\Delta k = \Delta k_{at} + \Delta k_{elec} + \Delta k_{Gouy} + \Delta k_{fat}
\]

(2)

Here the atomic dispersion is described by \(\Delta k_{at} \sim 2\pi q P(1 - \eta_f)(n_d - n_q)/\lambda\), where \(q\) is the harmonic order, \((n_d - n_q)\) is the difference of the refractive indices of the gas per 1 bar for the driving and the HH wavelength, respectively; \(\eta_f\) is the ionization fraction (typically, for our range of intensities \(\eta_f < 1\)), \(P\) is the gas pressure. The electronic dispersion provides \(\Delta k_{elec} \sim PqN_e\lambda\) [18,19], and is determined by \(P\), \(q\) and also depends on the free electron density per 1 bar \(N_e\), classical electron radius \(r_e\), and the wavelength \(\lambda\). The third term is due to the geometrical phase shift (Gouy phase) and is determined by \(\Delta k_{Gouy} \sim (q - 1)^2\tan^{-1}(z/z_R)/\partial\tau, \) where \(z_R\) is the Rayleigh range, and \(z\) is the displacement from the focal point along the laser beam [20–22]. The last term in Eq. (2) gives \(\Delta k_{fat} = -\alpha\nabla I\), where \(\nabla I\) is the intensity gradient, and \(\alpha\) is a coefficient depending on the quantum path of short or long electron trajectories. It was estimated for the short trajectory \(\alpha \sim 2 \times 10^{14}\text{cm}^2/W\) and for the long trajectory \(\alpha \sim 22 \times 10^{14}\text{cm}^2/W\) [23–25]. The first and the second terms in Eq. (2) having opposite signs can be adjusted such that the mismatch is minimized, i.e. \(\Delta k \to 0\).

Evaluating all the contributions to the wave vector mismatch of Eq. (2) allows to determine the coherence length \(L_{coh}\), and by using it and the absorption length, which was calculated from the available data [26], to find the pressure dependence of the HHs output from Eq. (1). It is assumed that the long and short trajectories equally contribute to the total harmonic signals [23,27].

Another 1D-model that we used for comparison assumes that the gas density and the dipole density are gradually distributed in the medium [28], consequently we will refer to this model as the "distributed source model". The output of a HH of order \(q\) is described by

\[
I_q = \int_0^{L_{med}} A_q(x) \exp\left[-\int_z^{L_{med}} \left[ x'/(L_{abs}(x')) + ix'/L_{coh}(x') \right] dx' \right] dx
\]

(3)

where \(A_q(x)\) is the amplitude of the partial contribution to the HH output from the element of the medium \(dx\) and the absorption length, \(L_{abs}\) and the coherence length, \(L_{coh}\) are functions of the position \(x\); \(L_{med}\) is the total length of the gaseous medium, which affects the HHG process. The induced dipole density we assume to be proportional to the molecular density \(n\) and power \(\gamma\) of the field amplitude \(E\)

\[
A_q(x) \propto n E^\gamma
\]

(4)
Here we assume that $n$ has the following distribution

$$n = n_0 l^2 / (x^2 + l^2)$$

with a characteristic length $l = 0.17 \text{mm}$ and that the laser field amplitude $E$ has the spatial distribution

$$E = E \left[ l_r^2 / (x^2 + l_r^2) \right]^{0.5}$$

where $l_r = 0.45 \text{cm}$ is the Rayleigh range in the focal region. The power law for the HH output intensity dependence was measured and approximated to be $2 \gamma \approx 3$. The coherence and absorption lengths for different gas densities were calculated from the data of Ref. [26].

Figs. 6 and 7 show the comparison of calculations with the two 1D-models and the experimental results for HHs from 21st to 25th order for N$_2$ and atmospheric air. The results of calculations (red dashed lines for the segment source model and dashed black lines for the distributed source model) are in reasonable agreement with the experimental data (blue dotted lines) in Figs. 6 and 7, where the yields are normalized to 1 for better comparison. The comparison shows that the distributed source model overall better describes the pressure dependences for different HHs.

As presented in Figs. 3 and 5, the HH yields for N$_2$ and air initially increase with increasing pressure reaching the maximum, and then they decrease with the further increase of gas pressure. The simulations show that the phase mismatch is strongly affecting the HHs emission. The latter depends on the pressure in the interaction region (R2). The coherence and absorption lengths were calculated for different harmonics and various pressures. It was found that the coherence length reaches the maximum (small phase mismatch) at approximately the same optimal pressure values, where the maximum of the HH yield was observed. As an example we present the results of calculations of $L_{\text{coh}}$ and $L_{\text{abs}}$ for 21$^{\text{st}}$ and 25$^{\text{th}}$ HHs in Fig. 8.

With the increasing gas jet pressure, the phase mismatch proportional to $|\Delta k|$ goes through the minimum $|\Delta k| \rightarrow 0$, which corresponds to $L_{\text{coh}} \rightarrow \infty$, achieved at the pressure value $\sim 0.33 \text{ bar}$, where the HH emission coherently builds up. For pressures higher than the optimal one, $|\Delta k|$ starts increasing, which leads to the decrease of the HH yield.

As Fig. 8 shows, the maximum coherence length is realized for 21$^{\text{st}}$ and 25$^{\text{th}}$ harmonics at about the same pressure. Only slight dependence of the coherence length on the order of HHs explains why the optimal pressure only to small degree varies for different HHs (Figs. 3 and 5). At the optimum pressure value, the higher concentration of the gas atoms provides strong HH output. We note that the extension of the gas jet to a larger distance is suppressed by the additional pumping of the gas cell. Thus, the reabsorption of the generated XUV radiation is decreased with the help of the differentially pumped cell, increasing the HH output as a result.

---

Fig. 6. Comparison of calculated and experimental results at varying pressure values for N$_2$ gas and HHs of different orders: (a) 21$^{\text{st}}$ order (b) 23$^{\text{rd}}$ order (c) 25$^{\text{th}}$ order. Blue dotted lines are the experimental results, black dashed lines show the result for the distributed source model of Eq. (3), and red dashed lines are the calculations with the segment source model of Eq. (1) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).
5. Conclusions

The pressure influence on the HHG in N₂ and atmospheric air with near-IR ultrafast laser fields is experimentally investigated. The experimental results are compared with two 1D theoretical models with constant on a segment and distributed HH sources that give a reasonable agreement (somewhat better for the distributed source model) with the experimentally measured pressure dependences of HH outputs. The optimum pressure is required for phase matching to generate HHs efficiently, and the optimum values are found to be about the same ∼0.33 bar for N₂ and atmospheric air at intensities ∼1.5 × 10¹⁴W/cm² and at given experimental conditions. Near this pressure value a significant increase in the HHs output was observed.

This study shows the importance of the phase matching for generating coherent extreme ultraviolet radiation using HHG process, which can be realized with a table top system at a relatively high repetition rate. The increased XUV flux is useful for applications in photoelectron spectroscopy, imaging of small size objects (nano or biological samples), and XUV pump-probe experiments.

Fig. 7. Comparison of calculated and experimental results for HHs of different orders at varying air pressure values: (a) 21st order (b) 23rd order (c) 25th order. Blue dotted lines show the experimental results, black dashed lines are the calculations with the distributed source model (Eq. (3)), and red dashed lines present the results of the segment source model (Eq. (1)) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

Fig. 8. Coherence and absorption lengths: (a) for 21st harmonic for N₂ gas, (b) for 25th harmonic for atmospheric air for different pressures. Blue line is the coherence length, black line is the absorption length. The dashed vertical line shows the pressure value, where the mismatch $\Delta k \to 0$ and correspondingly, $L_{coh} \to \infty$ (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).
Acknowledgements

This work was supported by the Robert A. Welch Foundation Grant No. A1546 and the Qatar Foundation under the grant NPRP 8-735-1-154.

References


