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

M. Sayrac[a,b,*]

[a] Department of Physics, Faculty of Science, Cankırı Karatekin University, Cankırı, Turkey
[b] Department of Physics & Astronomy, Texas A&M University, College Station, TX, 77843-4242, USA

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ABSTRACT

Dependence of the variation of the gas pressure on the high harmonic generation (HHG) from nitrogen molecule (N_2) and atmospheric air using ultrashort intense laser pulses is measured. The optimum pressure point for generation of maximum harmonic signal is found for both sample. Enhancement and extension of the HH orders are observed at around optimum pressure value. A theoretical calculation based on one-dimensional model is used to explain this effect.

1. Introduction

To generate efficient extreme ultraviolet (XUV) radiation is much needed tool for imaging of matter by short wavelength and observation of coherence properties of matter. One of the prominent source to generate for this radiation is high harmonic generation (HHG). HHG is a unique nonlinear process in which atoms/molecules ionized by an intense laser field at frequency ω_0 produces radiations of higher frequencies $q\omega_0$ where q is an odd integer. HHG driven by IR lasers can span a frequency range from UV to soft X-ray regions [1,2], and it has various applications in physics, chemistry, and biology [2,3]. HHG is usually realized by using an amplified femtosecond laser pulses that can be produced from a table top laser systems. For this reason HHG is considered as a major approach of obtaining a coherent table top source of coherent XUV radiation [4].

HHG phenomena can be understood in a semi-classical picture [5,6], i.e an electron is tunnel ionized by the 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 its parent ion, it subsequently releases 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 great advantages of HHG source is that the emission is perfectly synchronized to the driving laser field. One can obtain a train of sub-femtosecond pulses of coherent light separated by half of the driving laser cycle when a number of high harmonic fields are phase-locked [8]. The maximum HH orders is determined by the cutoff energy which depends on the ionization potential (I_p) of the used atom, and the ponderomotive energy ($U_p(\mathrm{eV}) = 9.33 \times 10^{-20} \times I \times \lambda^2$ where $I(\mathrm{W/cm^2})$ being the laser intensity and λ (nm) being the fundamental wavelength. U_p refers to free electrons averaged kinetic energy gained in the laser electric field [6], and the cutoff energy is given as $Emax = I_p + 3.17U_p$ [9,10].

* Correspondence address: Department of Physics, Faculty of Science, Cankırı Karatekin University, Cankırı, Turkey.
E-mail address: muhammedsayrac@karatekin.edu.tr

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In addition to advantage of HHG, such as its compactness and availability of wide range of researchers, conversion efficiency of HHG is remarkable low, i.e. conversion of the driving IR laser radiation into its harmonics can be achieved in the range of 10^{-7}–10^{-5} per harmonic [11,12]. Thus, the possibility to generate a high density photon flux is limited by this low conversion efficiency, and it is further exacerbated by the mismatch of phase velocities of the fundamental and harmonic fields and a large absorption/dispersion in the medium [13]. In addition, geometrical phase shift (Gouy phase) arises when the laser beam is focused into gas medium [14]. The above mentioned de-phasing effects 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 pressure are measured, and both sample present similar pressure dependence since N_2 is 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 model of Ref. [13].

2. Experimental setup

HHG experiment is performed by using a Ti:Sapphire laser system which produces infrared pulses with a 1 kHz repetition rate, 50 fs pulse in duration, 800 nm spectrally central wavelength and pulse energy of upto 1 mJ. The focal length of 40 cm lens is used to reach peak intensity at the focus (1.5×10^{14}W/cm^2). Harmonics are generated in a gas jet (GJ) produced by burning holes with the laser beam, and the GJ is made of a squeezed nickel (Ni) tube (0.8 mm outer and 0.6 mm inner size). The GJ is enclosed, as is shown in Fig. 1, in a cell, and the main portion of the ejected gas is removed by a separate roughing pump (Oerlikon, Scroll 15), which helps to reach at relatively high pressures in the interaction region (R2). The differential pumping is created between the region of high pressure R2 and the main chamber, region R1. Region R3 is connected to an additional roughing pump, removing the main portion of the gas directly from the gas jet cell before it reaches region R1. The input hole (IH) and the output hole (OH) of the cell for passing the radiation are of 1.5 mm diameter (see Fig. 1b). The cell has also a 1.5 mm tube hole (TH) on the top for the Ni tube, sealed at the end. A bottom hole (BH) with a similar diameter is used for centering the Ni tube. The laser beam is focused on the gas jet to produce high harmonics, which are detected using an XUVspectrometer (McPherson, 248/310G). In this spectrometer, XUV radiation is diffracted by a grating (133.6 groves/mm) to a micro-channel plate (MCP), which detected the different HHs. The image from the phosphorous screen mounted at the back side of the MCP is projected onto a charge-coupled device (CCD) camera; this latter image of spectrally resolved HHs is acquired with a LabVIEW program for subsequent processing.

The propagation path of the XUV radiation to the spectrometer is pumped to reduce the re-absorption of the HH due to the residual gas and to maintain operation pressure ($\lesssim 5 \times 10^{-6}$ millibars) of the MCP of the XUV spectrometer.

The pressure requirements of the MCP and pressure loads on the turbo molecular pump (Pfeiffer, TMU-521-YP), used for evacuating the main chamber, set the upper limit of the achievable pressure in the gas jet. Maximum pressure limit ($\sim 7 \times 10^{-3}$ millibars) in the main chamber is directly related to the load on the turbo molecular pump that is determined mainly by the flow to the main chamber through the holes IH and OH. The large front and back openings in the gas jet cell, directly connected through the tubing to the roughing pump, as is shown in Fig. 1b, assure that only a small portion of the total gas flow leaks to the main chamber.

The maximum pressure reached in the interaction region (R2) is estimated to be ~2 bars for N_2 and air. The gas leakage due to the openings in the gas cell (IH and OH) to the main chamber (R1) (see Fig. 1) determines the maximal pressure ratio in the gas jet, when the valve to the additional roughing pump is opened or closed. This ratio is experimentally determined by comparing the outputs of HHs in two different cases with opened or closed valve by adjusting the flow through the gas jet and measuring the pressure in the main chamber. This pressure ratio, or more precisely the pressure gain, is experimentally found by comparing the HH spectrum with closed and open pump, and it is around ~15 for both sample. The gas jet pressure

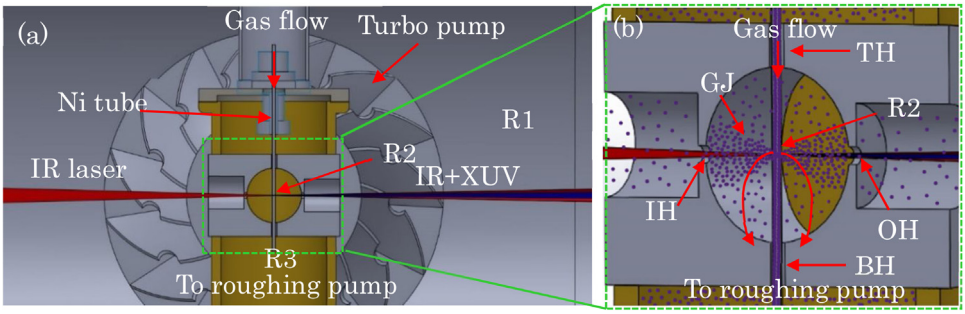

Fig. 1. Schematic of the experimental arrangement of the gas jet assembly and differential pumping (a) Section view of the differential gas cell setup, (b) Enlarged view of the central part of the gas cell. IR: infrared radiation, XUV: extreme ultraviolet radiation, IH: input hole, OH: output hole, TH: tube hole, BH: bottom hole, and GJ: gas jet.