Süleyman Demirel Üniversitesi Fen Edebiyat Fakültesi Fen Dergisi
Süleyman Demirel University Faculty of Arts and Sciences Journal of Science
2022, 17(1): 36-43
DOI: 10.29233/sdufeffd.956371
Atıf için / For Citation: M. Sayrac, "High harmonic generation produced in molecular nitrogen using ultrashort optical pulses", *Süleyman Demirel Üniversitesi Fen Edebiyat Fakültesi Fen Dergisi*, 17(1), 36-43, 2022.

Araştırma Makalesi

High Harmonic Generation Produced in Molecular Nitrogen using Ultrashort Optical Pulses

Muhammed SAYRAC[*1]

[1]*Sivas Cumhuriyet University, Department of Nanotechnology Engineering, 58140 , Sivas, Turkey*

**corresponding author e-mail: muhammedsayrac@cumhuriyet.edu.tr*

(Alınış / Received: 23.06.2021, Kabul / Accepted: 28.12.2021, Yayımlanma / Published: 27.05.2022)

Abstract: Generation of coherent extreme ultraviolet (XUV) pulses is a nonlinear process of high harmonic generation (HHG). HHG produced in molecular nitrogen (N_2) medium has been obtained using ultrashort intense laser pulses. The strong laser field was focused onto a gas cell to produce short wavelengths having photon energy up to 54 eV. The highest photon energy is experimentally observed as the 35^{th} order. The absorption of harmonics restricts the harmonic yield efficiency. The harmonic yield is affected by the interaction length and medium pressure. The harmonic yield changes with the medium parameters. The simulation for the absorption length was performed using the Mathematica program. Variation of the harmonic signal has been attributed to the absorption of harmonics, and it is compared with the absorption length.

***Key words**:* High harmonic generation, XUV, Ultrafast optics, Molecular nitrogen

Ultrahızlı Optik Darbeler Kullanılarak Azot Molekülünden Yüksek Mertebeli Harmonik Üretimi

Öz: Aşırı ultraviyole (XUV) optik darbelerinin üretilmesi, doğrusal olmayan bir yüksek harmonik üretim (HHG) sürecidir. Ultra kısa yoğun lazer darbeleri moleküler nitrojen (N_2) üzerine odaklanarak yüksek mertebeli harmonikler elde edilmiştir. Güçlü lazer alan şiddeti bir gaz hücresine odaklanmış ve 54 eV foton enerjisine kadar optik dalgaboyları üretilmiştir. Deneysel olarak en yüksek foton enerjisi 35. harmonik mertebesi olarak gözlemlenmiştir. Harmoniklerin soğurulması (absorpsiyon) harmonik sinyal verimliliğini kısıtlamaktadır. Harmonik verim, gaz hücresi uzunluğu ve ortam basıncı gibi parametre değerlerinden etkilenmektedir. Ortam parametreleri nedeniyle harmonik verim azalmaktadır. Absorpsiyon uzunluğu için simülasyon Mathematica programı kullanılarak yapılmıştır. Harmonik sinyalin değişimi, harmoniklerin absorpsiyonuna atfedildi ve absorpsiyon uzunluğu ile karşılaştırdı.

***Anahtar kelimeler**:* Yüksek harmonik üretimi, XUV, Ultrahızlı optik, Nitrojen molekülü

Muhammed SAYRAC, muhammmedsayrac@cumhuriyet.edu.tr, ORCID: https://orcid.org/0000-0003-4373-6897

1. Introduction

Obtaining chemical reactions in real-time is an important success of laser technology, which produces short optical pulses on the order of femtosecond time scale (1fs=10^{-15}s) [1-3]. High harmonic generation (HHG) paths in a way that the optical pulses allow one to observe the electron dynamics in atomic and molecular systems, which dynamics occur shorter time scale on the order of femtosecond or attosecond (1as= 10^{-18}s) [1, 2].

HHG is a promising method for generating short pulses in the XUV spectral region [4, 5]. When an intense laser field interacts with nonlinear media (such as atoms, molecules, or solids), high order harmonics of the initial radiation are emitted [6-9]. This coherent XUV source is essential for many practical applications such as biological imaging [10-13], the study of electron dynamics [14], attosecond science [15, 16], and so on. The optimized HHG can find an application on coherent diffractive imaging (CDI) of nanostructures or biological samples [10-13]. Even though HHG is an ideal method to produce short pulses at high photon energies (up to ~keV), the total photon flux is not sufficient for many practical applications. HHG has been studied over the past two decades because it has proved to be a powerful coherent tabletop X-ray source [4, 5]. There are many experimental and theoretical studies to increase the efficiency of HHG in atomic and molecular gases. However, there are still many open questions for the efficiency of the high harmonic generation because HHG strongly depends on the macroscopic conditions such as phase matching, which can vary for different experimental approaches and design.

The three-step model [17, 18] semi-classically explains the HHG process. In this model, an electron is initially subjected to an intense laser field with a magnitude comparable to that of the Coulomb field that binds the electrons to the nucleus. The strong laser field suppresses the potential barrier, and so allows quantum tunneling to free the electron from the nucleus. In the second step, the freed electron is accelerated by the oscillating laser field. The electron is either completely ionized or it is accelerated back to its parent ion about one–half of the optical cycle. In the third step, if the electron re-collides with the parent ion, it may emit a very short burst of x-ray radiation, in the form of high-energy photons. This coherent process occurs every half cycle of the laser pulse. For this reason, the emitted radiation is a spectral comb of odd-order harmonics of the fundamental field. In the HHG process, the intense laser field of frequency ω_0 interacts with nonlinear medium, higher frequencies ($q\omega_0$, q is an odd integer) of the fundamental field are emitted [19].

After the discovery of this powerful process, there are several studies concentrated on improving the properties of harmonics, such as pulse duration, number of photons, and efficiency of harmonics. These studies have shown that the properties of the harmonic source depend on the parameters of driving laser intensity and wavelength, and the pressure of the interaction region [20-22].

For this reason, the harmonic source provides the study of strong laser field-matter interaction, and it provides the observation of the generation system due to its short pulse duration. The harmonics mostly depend on the generating medium since the molecular symmetry and rotational vibrational dynamics bring additional parameters to control harmonic efficiency [23].

The cutoff energy is the summation of the ionization potential (I_p) of the used medium and the ponderomotive energy ($U_p = I/4\omega_0^2$) [24]. U_p is the free electrons averaged kinetic energy gained in the oscillating laser electric field [18]. The highest harmonic energy (cutoff, $E_{cutoff} = h\nu_{\max}$) is given as [25]

$$E_{cutoff} = I_p + 3.17U_p \quad (1)$$

Ponderomotive energy shows that harmonics are proportional to the driving field intensity (I) and its wavelength (λ) [17, 18].

Due to the cutoff formula, there are several parameters to vary for increasing the efficiency of HHs and for extension of the cutoff harmonics, for example, the highest harmonic energy is linearly proportional to the laser intensity and the wavelength. By controlling these two variables, the harmonic signal can be optimized, and the harmonic spectrum can be extended to high-order harmonics.

In this paper, the harmonic spectrum produced in N_2 molecules is studied by using a strong laser field. High order harmonics are obtained up to 35H orders, corresponding photon energy of 54eV. The external field pulse energy is set to 6mJ and 3mJ, and the power change affects the harmonic yield. The N_2 medium is important since it is simple and its molecular revival time is 8.2 ps [26]. In addition, the air mostly consists of N_2. For these reasons, the investigation of N_2 medium using a high power laser system for the generation of high order harmonics is in the interest of this study.

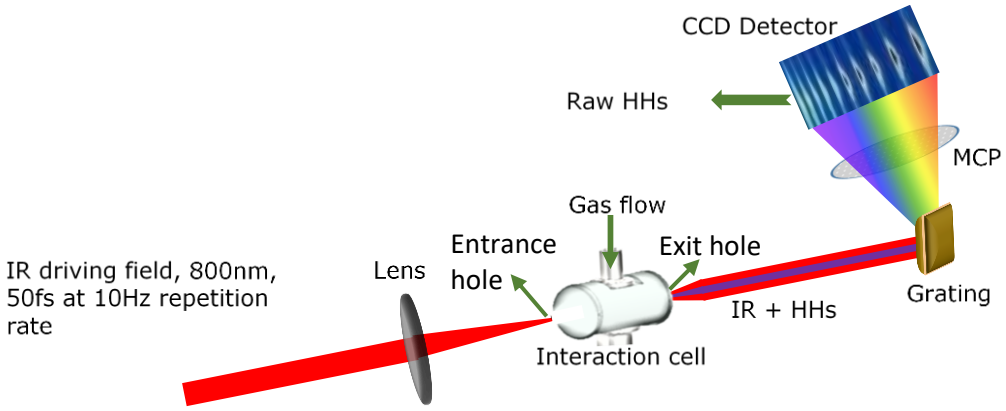

Figure 1. Demonstration of the experimental setup of high harmonic generation. CCD: Charged coupled device; HHs: High harmonics; IR: Infrared radiation; MCP: Microchannel plate

2. Material and Method

The state of the laser system producing 6mJ optical pulses with 50fs pulse duration at a 10Hz repetition rate is focused on the molecular medium. The central wavelength of the driving laser source is 800nm. The pressure in the interaction chamber is 4×10^{-3}mbar, and the pressure in the gas cell is 80mbar. The laser field is focused onto a gas cell by using a 40cm focal length lens. The beam size at the focus is about ~60-80μm. The intensity of the driving laser field at the focus is 1×10^{15}W/cm^2 for 6mJ pulse energy, and it is 5×10^{14}W/cm^2 for 3mJ pulse energy. High harmonics are generated in a gas cell, which has 7mm length and 5mm diameter. The “Entrance hole” and “Exit hole” are taped with aluminum foil tape to localize the gas flow inside the gas cell. After the focused laser beam passes through the holes, the generated XUV radiation propagates through to the McPherson XUV spectrometer, consisting of a grating and a microchannel plate (MCP) detector coupled to a phosphorous screen. The images of harmonics are captured by using a charged coupled device (CCD) camera. The integration time of the CCD camera is set to 20s, which is the optimum time duration for the used experimental parameters to prevent saturation of the CCD. The experimental arrangement of HHG is shown in Fig. 1. The generated short-wavelength source energy per optical pulse is expected in the nanojoule range, corresponding to the

power of ~3kW [27]. This high-order harmonic radiation can be used as a seeding source for particle acceleration systems [28-30].

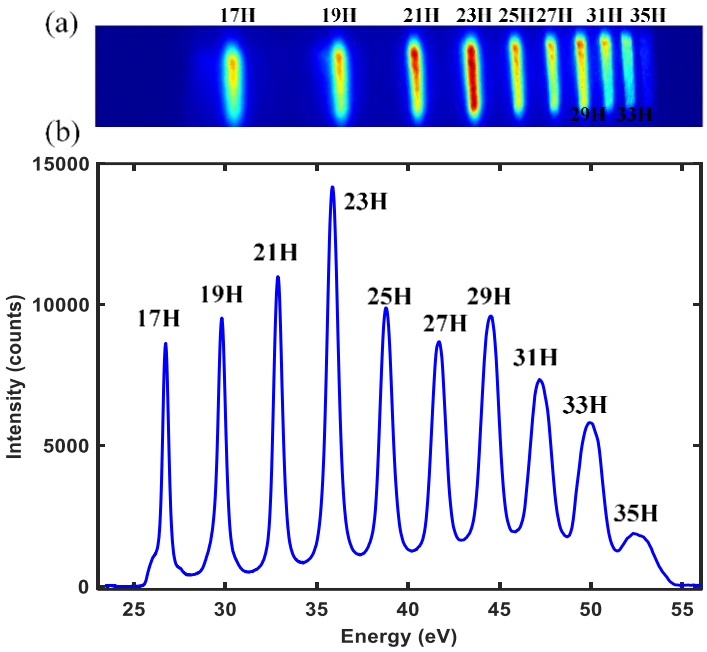

Figure 2. The pulse energy of the driving field is 6mJ. (a) Raw data of harmonic spectrum in molecular nitrogen is recorded using CCD camera, (b) Harmonic yield as a function of photon energy (eV)

3. Results

The generated harmonics spectrum contains a series of well-resolved harmonic peaks, which corresponds to the harmonic orders from 17H (~26eV) to 35H (54eV), (corresponding wavelength from ~47nm to 22nm). A high harmonic spectrum in pure N_2 is observed by using 6mJ pulse energy at a 10Hz repetition rate, Fig. 2. Figure 2(a) shows the harmonic spectrum recorded by the CCD camera. Harmonics yield is well observed up to 35H, Fig. 2 (b). The pulse energy of the driving laser field is 6mJ (1×10^{15}W/cm^2), and the integration time of the CCD camera is set to 20s, and the interaction region pressure is 4×10^{-3}mbar, which corresponds to 80mbar pressure inside the gas cell, Fig. 2.

When the pulse energy is decreased by 50% (3mJ), the harmonic signal is decreased by a factor of ~7 to 57 for different harmonic orders, Fig. 3 (b). The harmonic orders of 17H and 19H are less decreased (factor of ~7). The higher harmonic orders from 21H to 35H are decreased as a factor of ~22 to ~57, Table 1. Interaction chamber pressure is set to $4x10^{-3}$mbar (80mbar inside the gas cell) for generating the optimum harmonic spectrum.

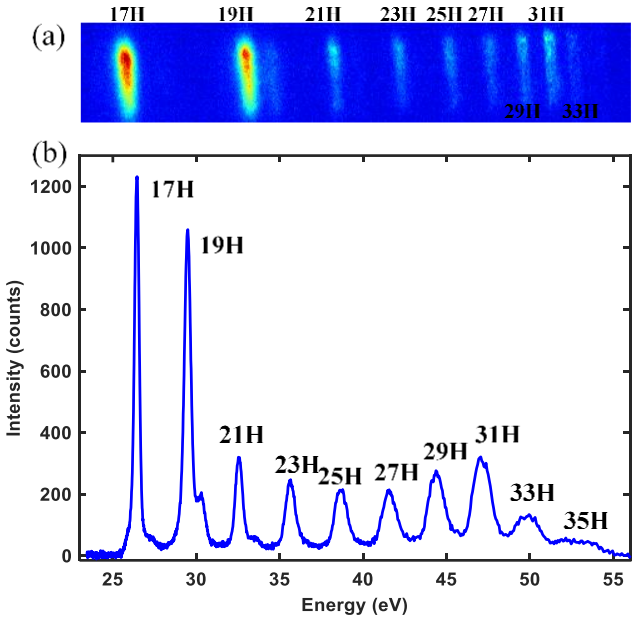

Figure 3. The pulse energy of the driving field is 3mJ. (a) Raw data of harmonic spectrum in molecular nitrogen is recorded using CCD camera, (b) Harmonic yield as a function of photon energy (eV)

Ionization potential (I_p) of N_2 molecules is 15.7eV [31]. The experimentally observed cutoff harmonic is 35H order. The cutoff harmonic calculation using Eq. 1 gives the cutoff harmonic as 33H order. The experimental value of cutoff harmonics is in close agreement with the calculation. The variation of the harmonic order comes from the intensity miscalculation due to imperfection of optics and variation of laser intensity at the focus. The harmonic orders present a saturation effect, in which harmonic yield has almost equal counts when the 6mJ pulse energy is used, Fig. 2 (b). When the pulse energy is decreased to 3mJ, the harmonic spectrum presents well-known features, namely a few strong harmonics, the plateau region, and eventually sharp cutoff, Fig. 3(b).

Table 1. Harmonic order signal ratio of 6mJ to 3mJ pulse energy

Harmonic order signal ratio $r = \frac{S_{\mathrm{HHG\_6mJ}}}{S_{\mathrm{HHG\_3mJ}}}$										
HH order	17	19	21	23	25	27	29	31	33	35
Wavelength (nm)	47.06	42.11	38.10	34.78	32	29.63	27.59	25.81	24.24	22.86
Photon energy (eV)	26.35	29.45	32.55	35.65	38.75	41.85	44.95	48.05	51.15	54.25
r	7	9	34.4	57.4	46	40.5	34.8	22.9	43.3	38

Table 1 presents a harmonic signal ratio of 6mJ to3mJ ($r = \frac{S_{\mathrm{HHG\_6mJ}}}{S_{\mathrm{HHG\_3mJ}}}$). The r-value is increased with increasing harmonic orders (shorter wavelength or higher photon energy). This can be explained by the fact that absorption rises at a shorter wavelength. The harmonic yield efficiency is restricted by absorption. The harmonic signal is affected by interaction length and medium pressure. The harmonic yield decreases because of the medium. The absorption length (L_{abs}) is determined from Refs. [27, 32],

$$L_{abs} \cong -\frac{1}{Log[T]} \tag{2}$$

where *T* is the transmission value at a specific wavelength. The transmission parameters are obtained from Ref. [33]. The pressure in the interaction chamber is 4×10^{-3}mbar, which corresponds to 80mbar pressure in the gas cell. Absorption increases after 30eV, Fig. 4. In addition, the signal ratio is given in Table 1 (highlighted column), which is in agreement with Fig. 4, i.e. absorption dominates the harmonic yield after ~32eV photon energy.

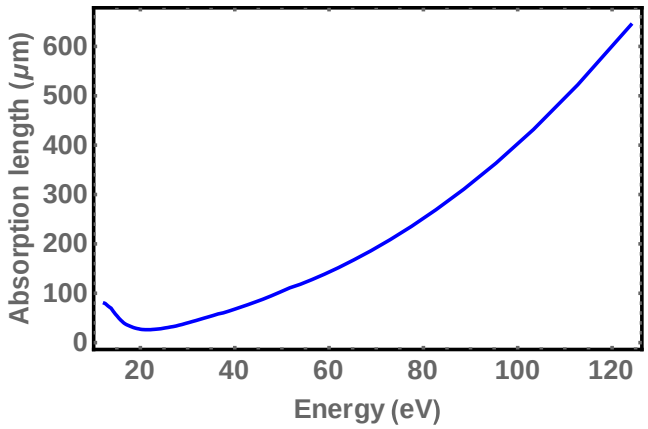

Figure 4. (a) Absorption length for N_2 molecule

4. Conclusion

High harmonic generation is produced in N_2 molecule by using the laser system producing sub-terawatt optical pulses having 50fs pulse duration at a 10 Hz repetition rate. Harmonic spectrum is produced in molecular nitrogen medium. The harmonic orders are produced up to 35H orders, which corresponds to ~54eV photon energy. The harmonic orders present well-resolved peaks, Fig. 2, 3. The decrease of the pulse energy results in a decrease in the harmonic yield because the ponderomotive energy linearly depends on the intensity of the optical field, and the electron is more dependent on Coulomb potential. The cutoff harmonic is calculated by using the above-mentioned formula, Eq. 1. The calculation gives the 33rd harmonic order, which is consistent with the experimental cutoff of 35H orders. Figure 4 presents that the absorption length increases with the increment of photon energy. Absorption plays a vital role on N_2 molecules for the used experimental parameters. The harmonic signal ratio is estimated in Table 1. The difference in cutoff harmonics between calculations and experiments comes from the imperfection of optic and intensity variation of the laser field during the experiment. The harmonic intensity (counts) decreases as the driving laser intensity is decreased.

The molecular N_2 is a promising source for the generation of coherent XUV sources having photon energy up to 54eV. This powerful source is useful for pump-probe spectroscopy, biological imaging studies, and so on.

Author Statement

Muhammed Sayrac: Experimental Data, Original Draft Writing, Visualization.

Acknowledgment

I am thankful to A. Kolomenski and H. Schuessler for providing the opportunity to perform the experiments described in this paper at Texas A&M University.

Conflict of Interest

As the authors of this study, we declare that we do not have any conflict of interest statement.

Ethics Committee Approval and Informed Consent

As the authors of this study, we declare that we do not have any ethics committee approval and/or informed consent statement.

References

[1] A. H. Zewail, "Femtochemistry: Atomic-scale dynamics of the chemical bond," *J. Phys. Chem. A*, 104, 5660-5694, 2000.

[2] X. F. Li, A. L'Huillier, M. Ferray, L. A. Lompré, and G. Mainfray, "Multiple-harmonic generation in rare gases at high laser intensity," *Phys. Rev. A*, 39, 5751-5761, 1989.

[3] A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. A. McIntyre, K. Boyer, and C. K. Rhodes, "Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases," *J. Opt. Soc. Am. B*, 4, 595-601, 1987.

[4] Z. Chang and P. Corkum, "Attosecond photon sources: the first decade and beyond," *JOSA B*, 27, B9-B17, 2010.

[5] H. C. Kapteyn, M. M. Murnane, and I. P. Christov, "Extreme nonlinear optics: Coherent X rays from lasers," *Phys. Today*, 58, 39-44, 2005.

[6] K. Ishikawa, "Photoemission and ionization of He+ under simultaneous irradiation of fundamental laser and high-order harmonic pulses," *Phys. Rev. Lett.*, 91, 043002, 2003.

[7] E. J. Takahashi, T. Kanai, K. L. Ishikawa, Y. Nabekawa, and K. Midorikawa, "Dramatic enhancement of high-order harmonic generation," *Phys. Rev. Lett.*, 99, 053904, 2007.

[8] F. Brizuela, C. M. Heyl, P. Rudawski, D. Kroon, L. Rading, J. M. Dahlström, J. Mauritsson, P. Johnsson, C.L. Arnold, and A. L'Huillier, "Efficient high-order harmonic generation boosted by below-threshold harmonics," *Sci. Rep.*, 3, 1410, 2013.

[9] T. Kanai, E. J. Takahashi, Y. Nabekawa, and K. Midorikawa, "Destructive interference during high harmonic generation in mixed gases," *Phys. Rev. Lett.*, 98, 153904, 2007.

[10] S. Eisebitt, J. Luning, W. F. Schlotter, M. Lorgen, O. Hellwig, W. Eberhardt, and J. Stohr, "Lensless imaging of magnetic nanostructures by X-ray spectro-holography," *Nature*, 432, 885-888, 2004.

[11] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pepin, J. C. Kieffer, P. B. Corkum, and D. M. Villeneuve, "Tomographic imaging of molecular orbitals," *Nature*, 432, 867-871, 2004.

[12] M. D. Seaberg, D. E. Adams, E. L. Townsend, D. A. Raymondson, W. F. Schlotter, Y. Liu, C. S. Menoni, L. Rong, C.-C. Chen, J. Miao, H. C. Kapteyn, and M. M. Murnane, "Ultrahigh 22 nm resolution coherent diffractive imaging using a desktop 13 nm high harmonic source," *Opt. Express*, 19, 22470-22479, 2011.

[13] A. Ravasio, D. Gauthier, F.R.N.C. Maia, M. Billon, J. P. Caumes, D. Garzella, M. Géléoc, O. Gobert, J. F. Hergott, A.M. Pena, H. Perez, B. Carré, E. Bourhis, J. Gierak, A. Madouri, D. Mailly, B. Schiedt, M. Fajardo, J. Gautier, P. Zeitoun, P. H. Bucksbaum, J. Hajdu, and H. Merdji, "Single-shot diffractive ımaging with a table-top femtosecond soft x-ray laser-harmonics source," *Phys. Rev. Lett.*, 103, 028104, 2009.

[14] P.M. Paul, E.S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, "Observation of a train of attosecond pulses from high harmonic generation," *Science*, 292, 1689-1692, 2001.

[15] M. Lewenstein, "Resolving physical processes on the attosecond time scale," *Science*, 297, 1131-1132, 2002.

[16] M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, "Attosecond metrology," *Nature*, 414, 509-513, 2001.

[17] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, "Theory of high-harmonic generation by low-frequency laser fields," *Phys. Rev. A*, 49, 2117-2132, 1994.

[18] P. B. Corkum, "Plasma Perspective on Strong-Field Multiphoton Ionization," *Phys. Rev. Lett.*, 71, 1994-1997, 1993.

[19] N. Ben-Tal, N. Moiseyev, and A. Beswick, "The effect of Hamiltonian symmetry on generation of odd and even harmonics," *J. Phys. B At. Mol. Opt. Phys.*, 26, 3017-3024, 1993.

[20] P. Salières and M. Lewenstein, "Generation of ultrashort coherent XUV pulses by harmonic conversion of intense laser pulses in gases: towards attosecond pulses," *Meas. Sci. Technol.*, 12, 1818-1827, 2001.

[21] P. Salières, A. L'Huillier, and M. Lewenstein, "Coherence control of high-order harmonics," *Phys. Rev. Lett.*, 74, 3776-3779, 1995.

[22] M. Sayrac, "High Harmonic Generation in Ar and N2 Gas Mixture Using Ultrashort High Power Laser System," *J. Inst. Science and Technol.*, 10, 1659-1665, 2020.

[23] T. Kanai, S. Minemoto, and H. Sakai, "Quantum interference during high-order harmonic generation from aligned molecules," *Nature*, 435, 470-474, 2005.
[24] M. Murakami, "High Harmonic Generation by Short Laser Pulses: Time-Frequency Behaviour and Applications to Attophysics," PhD thesis, Dept. Phys. Astronomy, Louisiana State University, Louisiana, USA, 2006.
[25] J. L. Krause, K. J. Schafer, and K. C. Kulander, "High-Order Harmonic Generation from Atoms and Ions in the High Intensity Regime," *Phys. Rev. Lett*., 68, 3535-3538, 1992.
[26] I. V. Litvinyuk, K. F. Lee, P. W. Dooley, D. M. Rayner, D. M. Villeneuve, and P. B. Corkum, "Alignment-dependent strong field ıonization of molecules," *Phys. Rev. Lett*., 90, 233003, 2003.
[27] M. Sayrac, A. A. Kolomenskii, and H. A. Schuessler, "Pressure optimization and phase matching of high harmonics generation in CO_2 and C_2H_2 molecular gases," *J. Electron Spectrosc. Relat. Phenom*., 229, 1-6, 2018.
[28] K. V. Zhukovsky, "Generation of x-ray radiation in free-electron lasers with two-frequency undulators," *Russ. Phys. J*, 60, 1630-1637, 2018.
[29] G. Lambert, T. Hara, D. Garzella, T. Tanikawa, M. Labat, B. Carre, H. Kitamura, T. Shintake, M. Bougeard, S. Inoue, Y. Tanaka, P. Salieres, H. Merdji, O. Chubar, O. Gobert, K. Tahara, and M. E. Couprie, "Injection of harmonics generated in gas in a free-electron laser providing intense and coherent extreme-ultraviolet light," *Nature Phys*., 4, 296, 2008.
[30] O. Culfa, "Laser-driven particle acceleration at near critical density plasmas," *Eur. Phys. J. D*, 75, 194, 2021.
[31] J.T. Tate and P. T. Smith, "The efficiencies of ıonization and ionization potentials of various gases under electron impact," *Phys. Rev*., 39, 270-277, 1932.
[32] M. Sayrac, A. A. Kolomenskii, and H. A. Schuessler, "Pressure dependence of high order harmonic generation in nitrogen molecular gas and atmospheric air," *Optik*, 179, 994-1000, 2019.
[33] B. L. Henke, E. M. Gullikson, and J. C. Davis, "X-Ray interactions: Photoabsorption, scattering, transmission, and reflection at E = 50-30,000 eV, Z = 1-92," *Atom. Data Nucl. Data*, 54, 181-342, 1993.