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Research Article

Generation of Coherent XUV Radiation in N_2 molecule and its Mixture with Ne Gas using Sub-terawatt Laser System

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ABSTRACT

High harmonic generation (HHG) in pure nitrogen (N_2) and its mixture with neon (Ne) gas was produced. The external laser field producing 6mJ optical pulse energy with 50fs pulse duration at 10Hz repetition rate was focused into a gas jet producing high harmonics. The N_2 and Ne gas have different ionization potentials. The strong harmonic signal in pure N_2 was produced, and a weak harmonic signal in a mixture of N_2-Ne was generated compared to that in pure N_2. The increase of Ne contribution in the N_2-Ne mixture resulted in a decrease in harmonic signal. Harmonic signal increase and decrease were observed for different N_2:Ne ratios. The harmonic generation mechanism was discussed in that the ionization of Ne is difficult compared to N_2, and the neutral Ne atom leads to neutral atomic dispersion (phase mismatch), so the harmonic yield decreases.

Keywords: *Extreme ultraviolet radiation, High harmonic generation, Three-step model, Femtosecond laser, Gas mixtures, Absorption*

N_2 Molekülünden ve Bunun Ne Gazı ile Karışımından Sub-terawatt Lazer Sistemi kullanılarak Uyumlu XUV Radyasyonu Üretilmesi

ÖZ

Saf nitrojen (N_2) gazından ve bunun neon (Ne) gazı ile karışımından yüksek harmonik mertebeler (HHG) üretildi. 10Hz tekrarlama hızında 50fs optik darbe süresi ile 6mJ optik darbe enerjisi üreten lazer alanı, yüksek harmoniklerin üretildiği bir gaz jetine odaklandı. N_2 ve Ne gazı farklı iyonlaşma potansiyellerine sahip olduğu için saf N_2 gazında güçlü harmonik sinyali ve saf N_2'ye kıyasla N_2-Ne karışımında zayıf harmonik sinyali elde edildi. N_2-Ne karışımında Ne katkısının artması harmonik sinyalin zayıflamasına neden oldu. Farklı N_2:Ne oranı için harmonik sinyal artışı ve düşüşü gözlendi. Harmonik üretimi mekanizması, Ne iyonlaşmasının N_2'ye kıyasla zor olduğu tartışıldı, ve nötr Ne atomunun, nötr atomik dispersiyona yol açtığı (faz uyumsuzluğu) ve böylece harmonik sinyalin azalmasına neden olduğu açıklanmıştır.

Anahtar Kelimeler: *Aşırı ultraviole radyasyonu, Yüksek mertebeli harmoni üretimi, Üç-adım modeli, Femtosaniye lazer, Gaz karışımı, Soğurma*

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I. INTRODUCTION

There is a strong desire to measure the physical system with high spatial and temporal resolution for accurate measurements. The development of ultrashort laser sources makes it possible to achieve such a desire. The state-of-the-art laser system finds applications from pump-probe spectroscopy [1] to the study of laser beam shape [2-5], and from terahertz science [6, 7] to fragmentation [8, 9]. Moreover, it paves the way for high-intensity physics (high harmonic generation) [10-16]. There are facilities producing high photon energy such as free electron lasers and synchrotron sources [17, 18], but these facilities have some disadvantages such as high cost to operate and not reachable by the researchers working in moderate laboratories. Thanks to the ultrashort tabletop laser system, high harmonic generation (HHG) is an alternative compact radiation source to such immoderate facilities since the HHG emits coherent short wavelength radiation with high brightness [19].

The high harmonic generation arises when an atom is illuminated with short laser pulses ($<10^{-12}$s) at high focused intensities ($>10^{13}$Wcm^{-2}) [20, 21]. HHG produces optical pulses in the extreme ultraviolet spectral region (XUV) [22]. HHG source is a powerful tool in optical spectroscopy. This spectral region allows to study photoionization of many atoms or molecules [23]. In addition, XUV radiation produced by the harmonic generation process releases sub-femtosecond pulses leading to the generation of attosecond pulses (1 as=10^{-18}s) [24-27].

Although high harmonic generation has the potential candidate provide bright short optical pulses, it suffers from its low conversion efficiency [28, 29]. Optimization of harmonic generation yield has been studied by several researchers [12-15, 30-33]. The brightness of such a light source is a benchmark for studying light-matter interaction.

The mechanism of high harmonic generation has been explained by a three-step model; ionization, acceleration, and recombination of an electron, respectively [34]. Ionization: an electron exposed to a strong laser field leaves from the atom through a tunneling process. Acceleration: the freed electron propagates in the continuum with the oscillating laser field and gains kinetic energy. Recombination: the electron is driven back by the laser field, and it recombines with its atomic core. After recombination, the kinetic energy of the electron gained during the laser oscillation is released in the form of high photon energy.

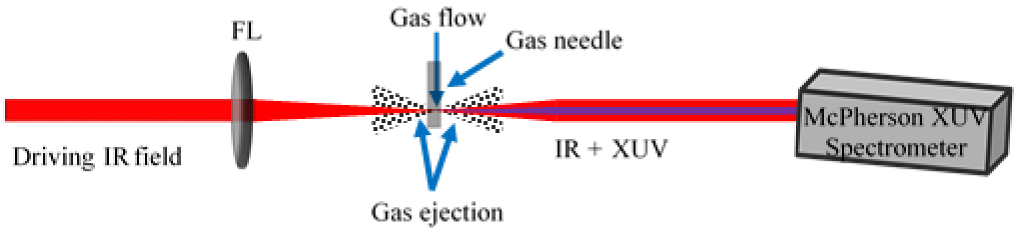

Figure 1. *Schematic of high harmonic generation setup. FL: focusing lens, IR: infrared radiation, XUV: extreme ultraviolet*

Initially, the electron is confined with the Coulomb potential. The strong laser field tilts the potential, and the electron tunnels to the continuum. The electron gains kinetic energy under the oscillating laser field. The kinetic energy of the electron is called the ponderomotive energy, $U_p=e^2E^2/(4m_e\omega^2)$, where e and m_e represent the charge and the mass of the electron. ω and E (E^2 corresponds to the intensity of the laser^t field) are the angular frequency and the electric strength of the laser field, respectively. Recombination of the electron with its parent ion releases the energy in the form of high photon energy. The maximum harmonic photon energy E_{max} is determined by cutoff law $E_{max}=I_p+3.17U_p$ [34, 35], here I_p is the ionization potential of the medium, and U_p is the ponderomotive energy.

In this paper generation of optical pulses in the extreme ultraviolet (XUV) spectral range having photon energies from 26 eV to 48 eV (from 47 nm down to 25 nm in terms of wavelength) has experimentally produced in N_2 and its mixture with Ne gas. The harmonic yield in pure N_2 is stronger than that in the N_2:Ne mixture. The N_2:Ne mixture ratio of 82%-18% gives more output signal compared to that of the 50%-50% mixture ratio.

II. EXPERIMENTAL SETUP

Ti: Sapphire laser system having a pulse energy of 6mJ with pulse duration of 50fs at 10Hz repetition rate is used as a driving laser source. The central wavelength of the driving laser field is centered at 800nm (IR). The radiation is focused by using a 40cm focal length lens (FL), and the focused intensity of the field is 1×10^{15}W/cm^2. The radiation is focused on a gas needle (gas jet) containing pure N_2 gas or its mixture with Ne. The pressure value inside the gas needle is ~40mbar [16].

N_2 gas is provided to the gas tube from a high-pressure N_2 cylinder. The mixture of N_2 with Ne is mixed in a small-size lecture bottle, Fig. 1. The mixture cylinder and the gas line are evacuated by a roughing pump (exhaust) to eliminate residual gas and contamination in the gas line. After the gas mixture is prepared in the small-size lecture cylinder, it flows to a 1mm thickness nickel gas tube. The gas flow intensifies the gas pressure inside the tube. The strong laser field makes holes as an input and an output hole (about 60μm size) on the tube. A detailed demonstration of the experimental setup, Fig. 1, is described in Ref. [36]. The pressurized gas ejects through the holes. The interaction of the laser field with the gas medium results in high order harmonic of the fundamental radiation. Generated harmonics propagate to the McPherson XUV spectrometer.

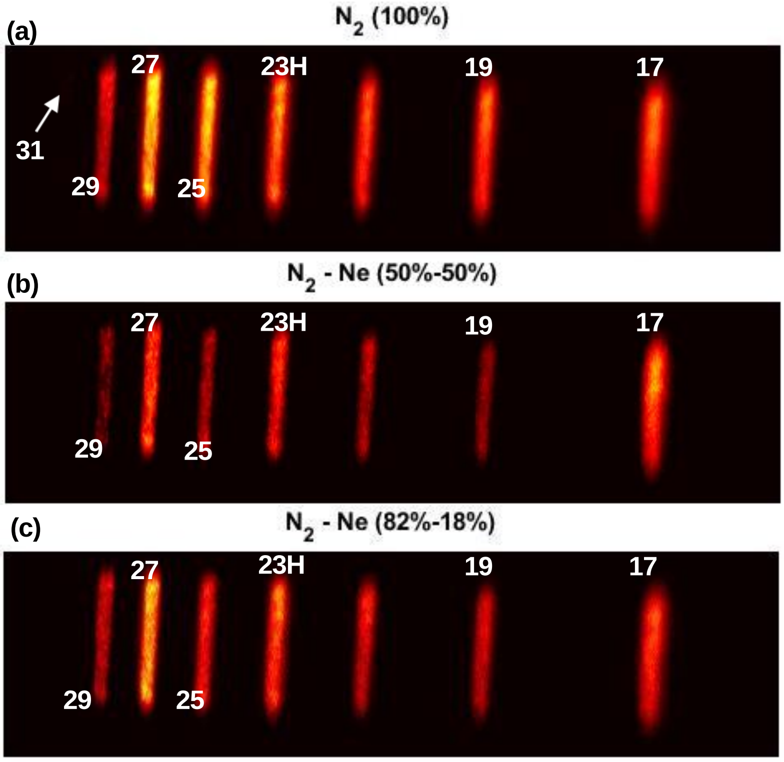

Figure 2. *Raw data of high harmonic spectrum **(a)** in pure N_2 (100%), **(b)** N_2-Ne (50%-50%), and **(c)** N_2-Ne (82%-18%)*

III. RESULTS AND DISCUSSION

A high harmonic spectrum is obtained in N_2 and its mixture with Ne gas. The raw data of high harmonics is presented in Fig. 2. The captured high harmonics (100% N_2, N_2-Ne mixture of 50%-50%, and of 82%- 18% ratio) are presented, and harmonic orders from 17H to 31H are well resolved in Fig. 2. The corresponding backing pressure (gas flow into the gas needle) is ~2.8bar for pure N_2, and ~1.4bar:1.4bar for 50%-50% N_2-Ne mixture, and ~2.2bar:0.6bar for 82%-18% N_2-Ne mixture. The harmonic spectrum in pure N_2 is stronger than that in the mixture at two different concentrations.

The 31H orders are not well resolved, Fig. 2. However, it is observed when the images of Fig. 2 (raw data) are processed (background subtracted), Fig. 3. The lowest harmonic signal is produced in 50%-50% N_2-Ne ratio. The harmonic yield produced in the N_2-Ne (82%-18%) mixture is stronger than the harmonic yield produced in the N_2-Ne mixture (50%-50%), Fig. 3. The greater contribution of Ne gas in the mixture leads to the decrease of the harmonic signal. The HHG in pure Ne is difficult and not observed at used experimental parameters because of the higher ionization potential of Ne gas (I_p=21.5eV [37]), and HHG in pure N_2 is stronger since the ionization of N_2 (Ip=15.7eV for N_2, [38]) is easy compared to ionization of Ne gas.

The harmonic yield produced in the N_2-Ne (50%-50%) mixture has decreased by a factor of 1.7 to 3.9 compared to that produced in pure N_2. The harmonic spectrum in the N_2-Ne (82%-18%) mixture has decreased by a factor of 1.4 to 2.1 compared to the harmonic signal in pure N_2. The harmonic signal in N_2-Ne (82%-18%) mixture is stronger compared to that in N_2-Ne (50%-50%) mixture. The enhancement factors (ratio of harmonic yield produced in N_2-Ne (82%-18%) mixture to harmonic yield produced in N_2-Ne (50%-50%) mixture) are varied from 1.2 to 2.1. Enhancement and decrement factors for each harmonic order (from 17H to 31H) are presented in Table 1.

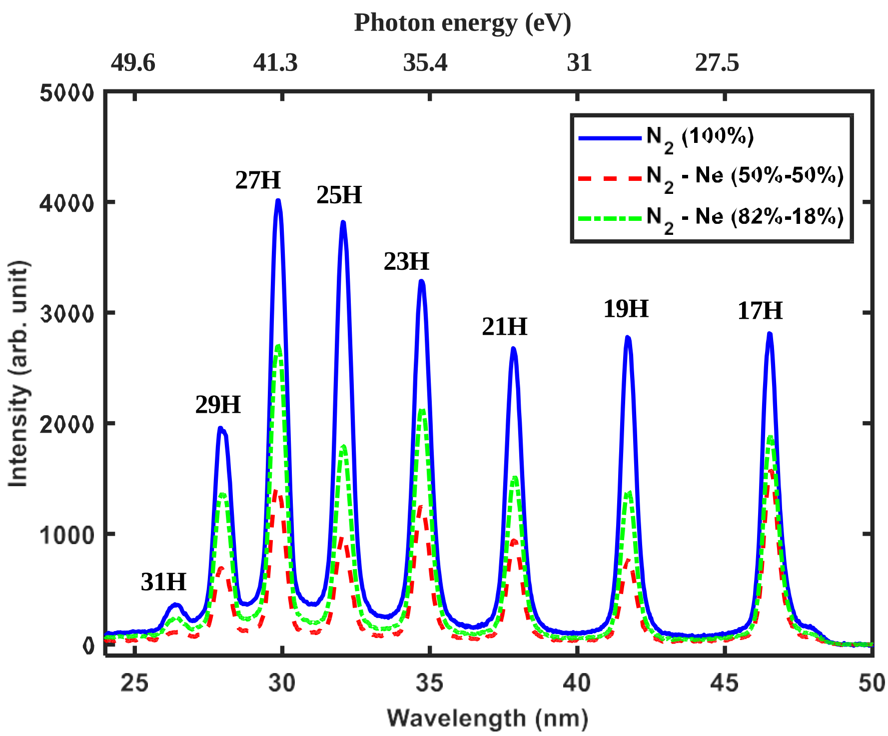

Figure 3. *Harmonic spectrum from 17H to 31H in N_2 and its mixture with Ne gas. The solid blue line is for pure N_2. Dashed red line is N_2-Ne (50%-50%). The dashed-dotted green line is for N_2-Ne (82%-18%)*

Table 1. *Harmonic generation yield decrement and enhancement factors for each harmonic orders*

a= N_2 (100%), b=N_2-Ne (50%-50%), c=N_2-Ne (82%-18%)			
Harmonic orders	**Signal decrement (a/b)**	**Signal decrement (a/c)**	**Signal enhancement (c/b)**
17H	1.7	1.4	1.2
19H	3.8	2.0	1.7
21H	2.7	1.7	1.6
23H	2.5	1.5	1.7
25H	3.9	2.1	1.8
27H	2.6	1.4	1.9
29H	2.9	1.4	2.0
31H	3.2	1.4	2.1

In this experimental work, the harmonic spectrum in pure N_2 and its mixture with Ne gas have been analyzed by using a sub-terawatt laser system producing optical pulses with 50fs pulse duration at a 10Hz repetition rate. High harmonic yield in pure N_2 has been well resolved since the ionization of N_2 gas is easy due to its low ionization potential (I_p=15.7eV) [38]. However, the harmonic spectra produced in Ne gas have not been obtained since Ne gas has a high ionization potential (I_p=21.5eV) [37]. The harmonic signal decreased or increased depending on the Ne contribution in the mixture ratio. When the strong laser fields interact with the medium, not all atoms are ionized. The ionization depends on the ionization potential of the medium. In the current experimental configuration, the medium is a mixture of neutral and ionized atoms. The neutral atom results in the atomic dispersion [12, 14, 16]. An increase of Ne gas concentration in the mixture results in the non-ionized Ne atoms in the medium increase because the ionization of Ne is difficult compared to that of N_2. The neutral Ne atoms increase the neutral atomic dispersion, and the neutral Ne atoms absorb the generated harmonics produced in N_2 gas. Thus, the HHG signal decreases.

When the Ne ratio is 18% in the gas mixture the harmonic signal increases because harmonics produced in N_2 gas with IR radiation contribute to ionization of Ne gas. Thus, a small amount of Ne contribution into N_2 gas results in a harmonic signal increase. However, an increase in the Ne contribution in the gas mixture makes the interaction region have unionized Ne atoms, which absorb the generated harmonics. Thus, the harmonic yield decreases. The absorption of Ne gas has a negative effect on the generated harmonic signal when the Ne contribution is higher in the gas mixture. Non-ionized Ne atoms in the medium result in to increase of the phase mismatch and lead decrement of harmonic signal produced in the gas mixture.

IV. CONCLUSION

Generation of high-order harmonics is produced by using a strong laser field having a pulse energy of 6mJ with a pulse duration of 50fs at a 10Hz repetition rate. Harmonic orders from 17H to 31H are obtained. The strong harmonic signal is obtained in pure N_2. The mixture of N_2-Ne gas leads to a decrement in the harmonic signal compared to that generated in pure N_2. When the Ne contribution increases, the harmonic signal decreases, Fig. 3. The mechanism of this decrement is that non-ionized Ne atoms in the medium have a negative effect on the generated harmonics produced in pure N_2. The neutral atomic dispersion from Ne gas leads to a low HHG signal. The decrease of Ne concentration in the gas mixture increases the harmonic yield. The harmonic signal decreases and the 31H order yield almost disappears when the Ne contribution is 50% in the gas mixture. The small ratio of Ne contribution leads to an increase in the harmonic signal, and the 31H order appears. The factors of signal enhancement and decrement are presented in Table 1. The optimized harmonic source has the potential to reach a short wavelength region from 47nm to 25nm (corresponding photon energy from

26eV to 48eV). The powerful light source has broad application areas, i.e. nonlinear optic in XUV region and enabling imaging of nanoscale objects in coherent short wavelength pulses.

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