Generation of Coherent XUV Radiation in N\textsubscript{2} molecule and its Mixture with Ne Gas using Sub-terawatt Laser System

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\textbf{ABSTRACT}

High harmonic generation (HHG) in pure nitrogen (N\textsubscript{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\textsubscript{2} and Ne gas have different ionization potentials. The strong harmonic signal in pure N\textsubscript{2} was produced, and a weak harmonic signal in a mixture of N\textsubscript{2}-Ne was generated compared to that in pure N\textsubscript{2}. The increase of Ne contribution in the N\textsubscript{2}-Ne mixture resulted in a decrease in harmonic signal. Harmonic signal increase and decrease were observed for different N\textsubscript{2}:Ne ratios. The harmonic generation mechanism was discussed in that the ionization of Ne is difficult compared to N\textsubscript{2}, and the neutral Ne atom leads to neutral atomic dispersion (phase mismatch), so the harmonic yield decreases.

\textbf{Keywords:} Extreme ultraviolet radiation, High harmonic generation, Three-step model, Femtosecond laser, Gas mixtures, Absorption

N\textsubscript{2} Molekülünden ve Bunun Ne Gazı ile Karışımından Sub-terawatt Lazer Sistemi kullanılarak Uyumlu XUV Radyasyonu Üretilmesi

Öz

Saf nitrojen (N\textsubscript{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\textsubscript{2} ve Ne gazı farklı iyonlaşma potansiyellerine sahip olduğu için saf N\textsubscript{2} gazında güçlü harmonik sinyali ve saf N\textsubscript{2} ye kıyaslı N\textsubscript{2}-Ne karışımında zayıf harmonik sinyali elde edildi. N\textsubscript{2}-Ne karışımında Ne katkısının artması harmonik sinyalin zayıflamasına neden oldu. Farklı N\textsubscript{2}:Ne oranları için harmonik sinyal artış ve düşüşü gözlemdi. Harmonik üretim mekanizması, Ne iyonlaşmasını N\textsubscript{2} ye kıyaslara zor olduğu tartıştı, ve nötr Ne atomunun, nötr atomik dispersiyona yol açtığı (faz uyumsuzluğu) ve böylece harmonik sinyalin azalmasına neden olduğu açıklanmıştır.

\textbf{Anahtar Kelimeler:} Açlık ultraviyole radyasyonu, Yüksek mertebe harmoni üretim, Üç-adım modeli, Femtosaniye lazer, Gaz karışımı, Soğurma
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.

\[ E_{\text{max}} = I_p + 3.17U_p \]

where \( I_p \) 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\textsubscript{2} and its mixture with Ne gas. The harmonic yield in pure N\textsubscript{2} is stronger than that in the N\textsubscript{2}:Ne mixture. The N\textsubscript{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 $1 \times 10^{15}$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.

\[\text{Figure 2. Raw data of high harmonic spectrum (a) in pure N}_2(100\%), (b) N}_2-\text{Ne (50\%-50\%}, \text{and (c) } N}_2-\text{Ne (82\%-18\%) }\]
III. RESULTS AND DISCUSSION

A high harmonic spectrum is obtained in N\textsubscript{2} and its mixture with Ne gas. The raw data of high harmonics is presented in Fig. 2. The captured high harmonics (100\% N\textsubscript{2}, N\textsubscript{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 \(\sim\)2.8bar for pure N\textsubscript{2}, and \(\sim\)1.4bar:1.4bar for 50\%-50\% N\textsubscript{2}-Ne mixture, and \(\sim\)2.2bar:0.6bar for 82\%-18\% N\textsubscript{2}-Ne mixture. The harmonic spectrum in pure N\textsubscript{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 corresponding harmonic yield produced in the N\textsubscript{2}-Ne (82\%-18\%) mixture is stronger than the harmonic yield produced in the N\textsubscript{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 \(\left(I_\text{p}=21.5\text{eV} \ [37]\right)\), and HHG in pure N\textsubscript{2} is stronger since the ionization of N\textsubscript{2} \(\left(I_\text{p}=15.7\text{eV} \ [38]\right)\) is easy compared to ionization of Ne gas.

The harmonic yield produced in the N\textsubscript{2}-Ne (50\%-50\%) mixture has decreased by a factor of 1.7 to 3.9 compared to that produced in pure N\textsubscript{2}. The harmonic spectrum in the N\textsubscript{2}-Ne (82\%-18\%) mixture has decreased by a factor of 1.4 to 2.1 compared to the harmonic signal in pure N\textsubscript{2}. The harmonic signal in N\textsubscript{2}-Ne (82\%-18\%) mixture is stronger compared to that in N\textsubscript{2}-Ne (50\%-50\%) mixture. The enhancement factors (ratio of harmonic yield produced in N\textsubscript{2}-Ne (82\%-18\%) mixture to harmonic yield produced in N\textsubscript{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\textsubscript{2} and its mixture with Ne gas. The solid blue line is for pure N\textsubscript{2}. Dashed red line is N\textsubscript{2}-Ne (50\%-50\%). The dashed-dotted green line is for N\textsubscript{2}-Ne (82\%-18\%).](image-url)
A strong harmonic signal is obtained when the Ne contribution is higher in the gas mixture. Thus, the harmonic yield decreases. The absorption of Ne gas has been found to increase of Ne gas concentration in the medium have a negative effect on the harmonic signal. The decrease of Ne concentration in the medium has an effect on the harmonic signal. The decrease of Ne concentration in the medium has been observed in pure N₂ gas. Thus, the HHG signal decreases.

When the Ne ratio is 18% in the gas mixture the harmonic signal increases because harmonics produced in N₂ gas with IR radiation contribute to ionization of Ne gas. Thus, a small amount of Ne contribution into N₂ gas results in a harmonic signal increase. However, an increase in the Ne contribution in the gas mixture makes the interaction region have ionized 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 an increase of the phase mismatch and lead decrement of harmonic signal produced in the gas mixture.

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

<table>
<thead>
<tr>
<th>Harmonic orders</th>
<th>Signal decrement (a/b)</th>
<th>Signal decrement (a/c)</th>
<th>Signal enhancement (c/b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>17H</td>
<td>1.7</td>
<td>1.4</td>
<td>1.2</td>
</tr>
<tr>
<td>19H</td>
<td>3.8</td>
<td>2.0</td>
<td>1.7</td>
</tr>
<tr>
<td>21H</td>
<td>2.7</td>
<td>1.7</td>
<td>1.6</td>
</tr>
<tr>
<td>23H</td>
<td>2.5</td>
<td>1.5</td>
<td>1.7</td>
</tr>
<tr>
<td>25H</td>
<td>3.9</td>
<td>2.1</td>
<td>1.8</td>
</tr>
<tr>
<td>27H</td>
<td>2.6</td>
<td>1.4</td>
<td>1.9</td>
</tr>
<tr>
<td>29H</td>
<td>2.9</td>
<td>1.4</td>
<td>2.0</td>
</tr>
<tr>
<td>31H</td>
<td>3.2</td>
<td>1.4</td>
<td>2.1</td>
</tr>
</tbody>
</table>

In this experimental work, the harmonic spectrum in pure N₂ 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 N₂ has been well resolved since the ionization of N₂ gas is easy due to its low ionization potential (Iₚ=15.7eV) [38]. However, the harmonic spectra produced in Ne gas have not been obtained since Ne gas has a high ionization potential (Iₚ=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 N₂. The neutral Ne atoms increase the neutral atomic dispersion, and the neutral Ne atoms absorb the generated harmonics produced in N₂ gas. Thus, the HHG signal decreases.

**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 N₂. The mixture of N₂-Ne gas leads to a decrement in the harmonic signal compared to that generated in pure N₂. 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 N₂. 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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**V. REFERENCES**


