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High Harmonic Generation in Ar and N_2 Gas Mixture Using Ultrashort High Power Laser System

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ABSTRACT: High harmonic generation (HHG) has been accepted as a tool for tabletop based generation of light source in the XUV and soft x ray region. HHG can produce coherent optical pulses having pulse duration in the femtosecond or even attosecond time region. In this paper, generation of high harmonics are produced by using high power laser system having optical pulses at 6mj pulse energy with pulse duration of 50fs at 10Hz repetition rate. High harmonics in pure Ar, N_2 and mixture of the Ar-N_2 are used as a generation medium to produce high harmonics. The harmonic signal is increased or decreased depending on the experimental condition. Harmonic yield produced in Ar is stronger than harmonic yield produced in N_2 gas. Generation of high order harmonics are observed up to 35H (~54eV corresponding photon energy), and harmonic order from N_2 gas is 33H. The mixture of two gas species cause to enhancement of 35H order, which is weakly observed in pure N_2 gas. The mechanism of high harmonic generation is explained that strong harmonic signal generated in pure Ar gas helps increase the ionization rate of N_2 gas. Thus, the harmonic signal in Ar-N_2 is boosted compared the harmonic signal produced in pure N_2. The enhancement factor of harmonic yield is from ~2 to 5 for per harmonic order.

Keywords: Ultrafast physics, femtosecond laser, high harmonic generation, extreme ultraviolet, attosecond science

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INTRODUCTION

High harmonic generation (HHG) has proved as one of the best approaches to produce coherent light source (Mcpherson et al., 1987) thanks to development of ultrafast femtosecond laser technology. The wavelength range of this source can reach soft x ray region. Harmonic source has opened new research areas such as attosecond science (Paul et al., 2001), imaging for nano size object with coherent XUV radiation (Sandberg et al., 2007), and nonlinear optic in XUV region (Nabekawa et al., 2005). These research areas help to understand background of electron–photon interaction. This coherent short wavelength source makes possible to study electron dynamics (Drescher et al., 2002) and to measure pulse duration of XUV pulses in the attosecond regime (1as=10^{-18}s) (Sekikawa et al., 2004).

General mechanism of harmonic source has been explained by three-step model (Corkum, 1993). In this model an electron exposed to strong laser field is ionized. The ionized electron is accelerated back and forth due to oscillating laser field and gains kinetic energy. If the freed electron recombines with its parent atom, it releases its kinetic energy as a high photon energy. The three-step model semi classically explains the features of harmonic generation process. Harmonic spectrum usually exhibits well-known features, namely plateau and cutoff. Harmonic yield relatively stays same in the plateau region. There is a sudden sharp decrease in the harmonic signal, which is called cutoff region. Maximum harmonic orders are determined by cutoff harmonic. It is given as $E_{cutoff}=I_p+3.17U_p$ (Krause et al., 1992). I_p is the ionization potential of the used gas, and U_p is the ponderomotive energy that refers to free electrons averaged kinetic energy gained in the laser electric field. U_p is proportional to intensity of the laser field. The meaning of the cutoff formula is that an atom having higher ionization potential produces high harmonics than an atom having lower ionization potential. The atom having higher ionization potential experience higher laser field intensities, and it leads to significant harmonic emission (Li et al., 1989).

High harmonic generation is a promising source for generation of coherent XUV source. However, this light source has drawbacks in terms of its conversion efficiency (conversion from fundamental field to high harmonics). The conversion efficiency is on the order of 10^{-6} (Falcão-Filho et al., 2010). There are many approaches studied to increase the efficiency of harmonic light source. For instance, high harmonic generation in gas mixture (Sayrac et al., 2015), the role of the shape of the optical field (Bartels et al., 2000), controlling of the electron trajectories in the optical field (Salières et al., 2001), the pressure optimization of high harmonic generation (Sayrac et al., 2015; Sayrac et al., 2018; Sayrac et al., 2019), generation of even harmonics of the fundamental radiation in two-color laser fields (Sayrac et al., 2019), and the effect of an increasing laser intensity (Gibson et al., 2004). These studies aimed to improve the generation efficiency of coherent XUV radiation.

In this paper, the harmonic spectra from gas species of Ar, N_2, and Ar-N_2 mixture are experimentally produced. The ionization potentials (I_p) of these gases are 15.6eV (Womer, 1931), 15.7eV (Tate and Smith, 1932) for Ar and N_2. The harmonic spectrum in pure Ar and N_2 reaches harmonic order up to 35H (~23nm or ~54eV). The harmonic spectrum from mixture of Ar –N_2 presents that harmonic signal decreases compared to harmonic signal in pure Ar, and it increases compared to harmonic signal in pure N_2. The novelty of this paper is that mixture of atomic Ar with molecular N_2 is used as generation medium for HHG. In addition, high power laser system is used as a driving laser source and enables to reach high order harmonic of 35H, which corresponds to ~23nm wavelength in the XUV region.

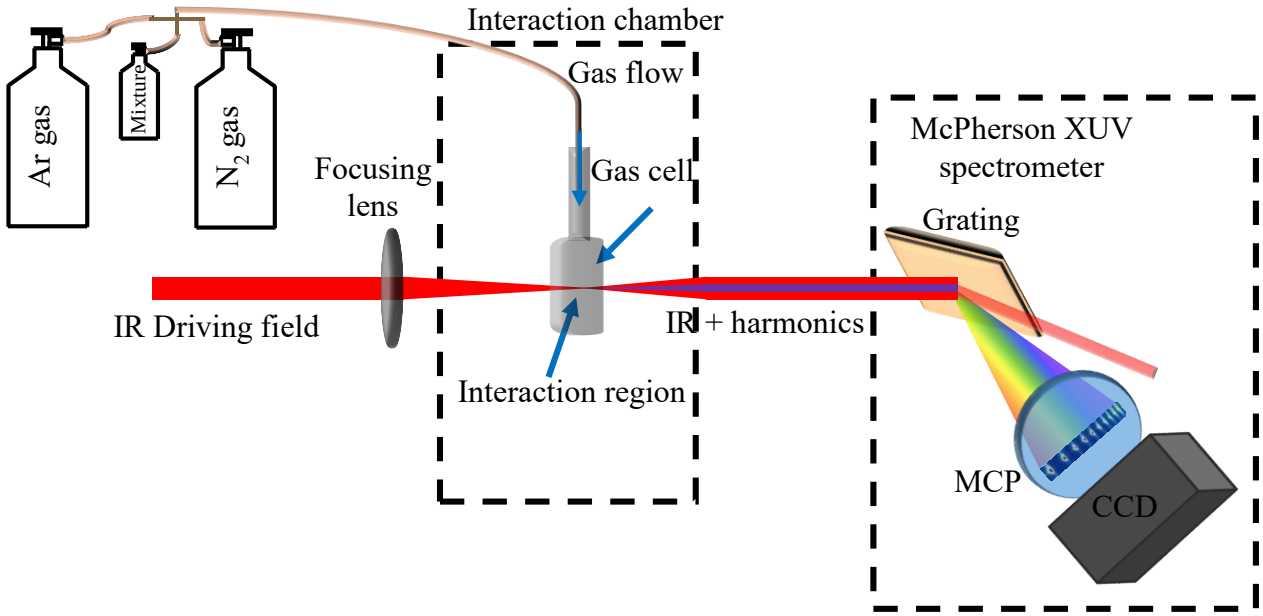

Figure.1. General sketch of the experimental setup for high harmonic generation

MATERIALS AND METHODS

Sketch of the high harmonic generation experimental arrangement is shown in Figure 1. The experiment is performed by using a high power Ti:Sapphire pulse amplifier (CPA) laser system producing optical pulses having 6mJ pulse energy with 50fs (~18 cycle) pulse duration at 10Hz repetition rate. The wavelength of the driving laser field is centered at 800nm (IR). The driving laser field is focused onto a gas cell using a long focal length (~40cm) lens through a thin BK7 glass window into a vacuum chamber (interaction chamber), which is held at low pressure of ~10^{-3}mbar. The intensity of the laser field at the focus is 1×10^{15}Wcm^{-2}. The high energy laser radiation is focused into a gas cell, where harmonic spectrum are generated. High harmonics are generated from a gas cell, which is machined in a machine shop. It has length of 7mm and diameter of 5mm. There is a hole on the cell, and the hole is taped by an aluminum foil tape to increase density of the gas. Pure gas is directly sent to the gas cell to produce harmonic spectrum. Gas mixture is prepared in a small size lecture bottle, and then the gas mixture is sent to the gas cell. The tabletop high harmonic generation setup having length of ~2m size can produce wavelength down to ~23nm. The tabletop femtosecond laser system having about 60mW output power feeds the harmonic generation system in the current experimental design.

In this experiment, high harmonic spectra are recorded for Ar gas, N_2 gas and their mixture in 1:1 ratio. In these three different gas media, the driving laser wavelength is centered at 800nm. The produced x ray pulses in Ar gas with driving laser field enhance the ionization rate of N_2 gas. Especially 35H, which is weakly observed in N_2 gas is boosted in harmonic generation in Ar-N_2 gas mixture. The mixture of gas species helps to increase the harmonics signal and to extend the harmonics to high photon energy region. High harmonic orders are obtained from 17th harmonic to 35th harmonic (corresponding wavelength from ~47nm to ~23nm). The harmonic energy per optical pulse is expected a nanojoule range, and this range corresponds to harmonic power of ~3kW (Sayrac et al., 2018). This high order harmonic radiation can be used for seeding high harmonic generation source in free electron lasers (FEL) (Lambert et al., 2008; Zhukovsky, 2017; Zhukovsky, 2017; Zhukovsky, 2017; Zhukovsky, 2018). The driving laser field has 50fs pulse duration at 800nm central wavelength. The generated harmonic orders having wavelength from ~47nm to ~23nm have the pulse duration shorter than the fundamental field.

The high harmonic source produces attosecond pulses (Paul et al., 2001), making possible to investigate atomic and molecular dynamics occurring on the sub femtosecond time scale (Vodungbo et al., 2012).

The generated harmonics propagate through to the McPherson XUV spectrometer. The harmonics interacting with grating are separated according to their wavelength. The separated harmonics pass through a micro channel plate (MCP) and are amplified. The amplified harmonics are captured by a charge coupled device (CCD) camera. The recorded data are analyzed by using the MATLAB program. The state of the art laser system allows to reach higher order harmonics by extending the cutoff harmonic since the ponderomotive energy is proportional to the intensity of the laser field.

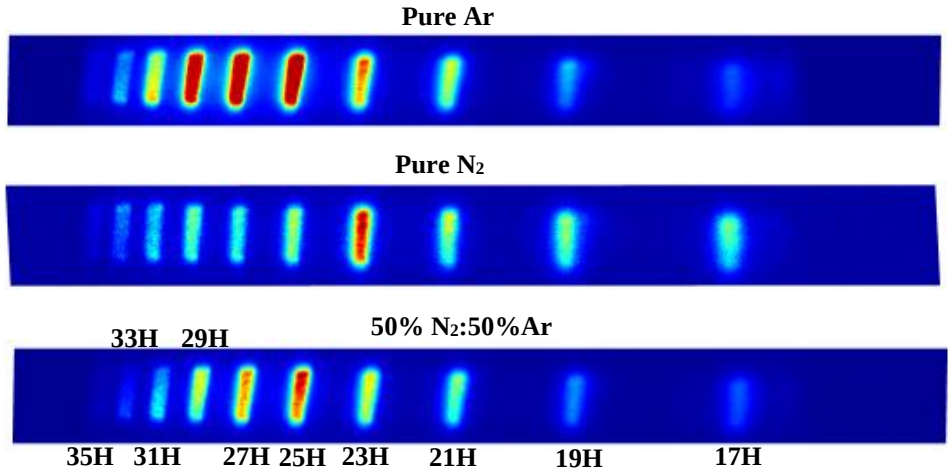

Figure.2. Raw data of harmonic spectrum in Ar, N_2 and N_2:Ar mixture (50%:50%) ratio. Harmonic orders from 17H to 33H are well resolved. The 35H is weakly observed in pure Ar

RESULTS AND DISCUSSION

The generated harmonic spectrum contains a series of well resolved harmonic peaks, which corresponds to the harmonic orders from 17H (~47nm) to 35H (~23nm), (from ~26eV to 54eV). Figure 2 presents raw data of harmonic orders recorded by the CCD camera. The harmonic orders from 17H to 33H are well resolved. The weak 35H is barely observed in Figure 2.

Harmonics spectrum in Ar, N_2 and their mixture in 50%-50% ratio are produced. The pulse energy is 6mJ, and the interaction chamber pressure is set to 4×10^{-3}mbar. The integration time of the spectrum is 10s, Figure 2, 3. Harmonic spectrum in pure Ar gas are obtained, and the harmonics orders reach up to 35H order. In pure N_2 gas the harmonic spectrum are well obtained up to 33H order, and the 35H order is weakly observed in N_2 gas, zoomed in part of Figure 3. The harmonics signal in pure Ar is relatively higher than the harmonic signal in pure N_2. When the mixture of Ar-N_2 in 50%-50% ratio is used as an interaction medium, the harmonic signal is decreased compared to harmonic yield in pure Ar, but the harmonic signal is increased compared to harmonic signal produced in pure N_2. Also the 35H, which is weakly observed in pure N_2 is increased in the mixture of Ar-N_2.

The ionization potential of Ar and N_2 gas species are close to each other, and the calculation of above mentioned cutoff harmonic (highest photon energy) is about 56eV, which is close agreement with experimentally observed cutoff harmonic. Even though the experimentally observed cutoff harmonic is slightly less than the predicted by the intensity of the laser pulse. The discrepancy comes from the miscalculation of the field intensity and the beam loss on the optical components namely mirrors, optical lens, and vacuum chamber window.

In this paper, high power laser system producing optical pulses having energy per pulse 6mJ with 50fs pulse duration at 10 Hz repetition rate is used as a driving laser source. Harmonic spectrum are

produced in three different gas media, namely Ar, N_2, and mixture of Ar-N_2. Harmonic yield in Ar gas has relatively more signal than harmonic signal produced in N_2 gas. Mixture of Ar-N_2 produces more harmonic signal compared to harmonic signal produced in pure N_2. The mixture of two gas species results to increase of harmonic signal compared to harmonic signal in pure N_2.

The mechanism of harmonic signal increase in Ar-N_2 gas mixture can be explained that the strong harmonics produced in Ar gas and the fundamental field increase to ionization of N_2 gas. The mechanism can lead to increase of the harmonic signal in Ar-N_2 gas mixture compared to harmonic signal in pure N_2. However, decrease of the harmonic signal in gas mixture of Ar-N_2 compared to harmonic yield in pure Ar can be explained because of the absorption of the harmonics in gas mixtures. The absorption in the harmonic spectrum leads to decrease of the harmonic signal. The harmonic signal in Ar-N_2 gas mixture is increased as a factor of ~2 to 5 compared to harmonic yield in pure N_2.

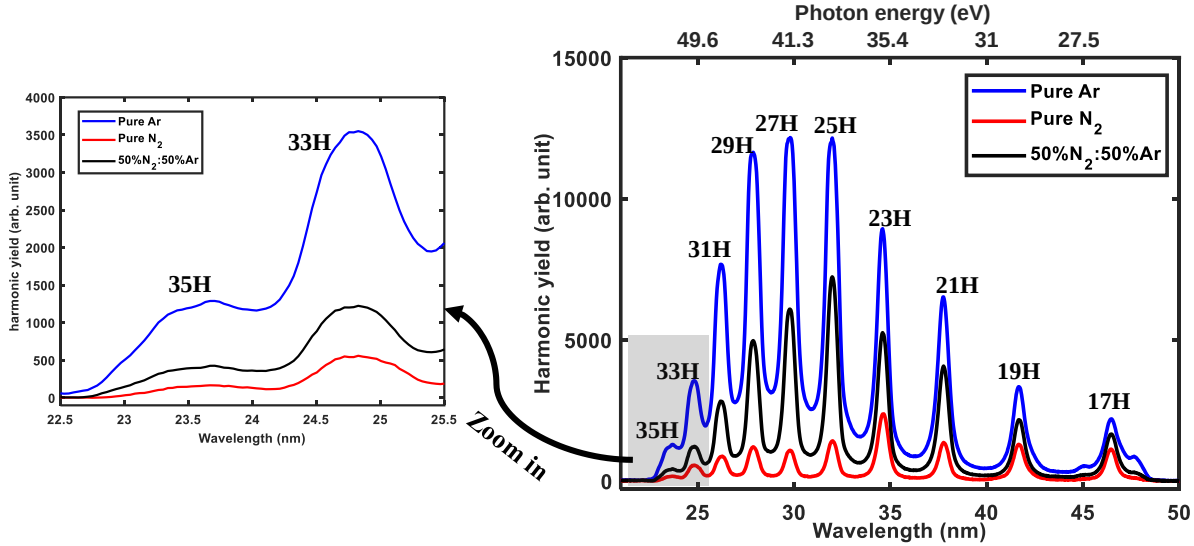

Figure.3. Harmonic spectrum in Ar, N_2 and Ar-N_2 mixture (50%:50%) ratio. The solid blue line is for pure Ar. The solid red line is for pure N_2. The solid black line is for mixture. The right figure presents the complete harmonic spectrum from 17H to 35H orders, while the left figure shows the zoomed in part of the harmonic orders for 33H and 35H

CONCLUSION

Generation of short wavelength (XUV region) is experimentally produced. The laser intensity of 1×10^{15}Wcm^{-2} is focused on to the gas species. High harmonics up to 35H order are generated. The generation of the 35H is increased in the gas mixture of Ar-N_2 compared to harmonic signal in pure N_2. Harmonic spectrums in Ar, N_2, and Ar-N_2 mixture are well resolved. The harmonic signal is increased in the gas mixture compared to harmonic yield in pure gas species of N_2 (Figure 3). The mechanism of harmonic spectrum in gas mixture is explained that the harmonics produced in one gas species and the fundamental field result to increase the harmonic signal from another gas species.

The harmonic yield enhancement stems from the harmonic yield in Ar gas and the driving laser field boost the ionization rate of N_2 gas, so the harmonic yield in gas mixture is increased by the contribution of strong XUV radiation produced in Ar gas. In the nitrogen-argon gas mixture the enhancement of harmonic signal compared to harmonic signal produced in pure N_2 has been observed.

The highest harmonic photon energy is estimated by using the optical field intensity. The experiment and the calculation give highest harmonic energy of ~54eV and ~56eV, respectively. The strong laser field and the gas mixture helps reach high order harmonics. The optimized harmonic source

can be useful for various application areas such as attosecond light source, nonlinear optic in XUV region and imaging of small scale object by using harmonic source.

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