

INVESTIGATION OF HIGH HARMONIC GENERATION IN Ar-Ne GAS MIXTURE

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Abstract. In this study, we experimentally investigate the variation of the phase matching condition of the high harmonic generation (HHG) process with pure argon gas and an argon-neon gas mixture. Phase-matched HHG is generated around the absorption edge of argon gas and then neon gas is added to the original argon gas. The pressure-dependent intensity of the harmonics produced by pure argon gas and the gas mixture is examined. We show that as more neon gas is added to the mixture, the phase matching of the higher order harmonics is less favourable than that of the lower order harmonics. Finally, the total phase mismatch at various gas mixture pressures is discussed. Our experimental results are in agreement with the theoretical calculation.

Key words: high harmonic generation, phase matching, nonlinear optics.

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1. INTRODUCTION

High harmonic generation (HHG) is an extremely nonlinear optical process for producing coherent extreme ultraviolet (XUV) pulses at ultrashort time scales [1, 2]. The high order harmonics emitted in a series of attosecond bursts with high spatial and temporal coherence are of great interest for probing electron dynamics on an attosecond time scale [3] and for coherent diffractive imaging [4]. Harmonics are typically generated in noble gases where an intense laser pulse interacts directly with atoms. When the laser pulse interacts with dielectric or metallic surfaces, a more complex scheme of HHG from surfaces is observed. The three-step model effectively describes the physics of HHG in atomic gases, including the amplitude and phase of harmonics [5, 6]. In this model, the intense laser field modifies the potential barrier so that the initially bound electron is ionised into the continuum. The freed electron wave packet is then driven by the laser field after the field reverses its direction. Under certain conditions, the electron will recombine with

the atom, emitting electromagnetic radiation containing both its kinetic and binding energies. However, in general, an understanding of the HHG process based solely on single-atom dynamics is insufficient to fully explain the experimental data. To achieve a good agreement between theory and experiment, one must also take into account the effects of propagation and phase mismatch between the fundamental field and the harmonic field in the macroscopic medium [7, 8]. The phase mismatch consists of four contributions: the dispersion of the neutral atoms and free electrons, the geometrical wave vector mismatch caused by focusing, and the phase of the atomic dipole [9, 10]. The first two phase mismatches (neutral dispersion and plasma dispersion) are dependent on the gas pressure, while the others are independent of pressure. The major contributions of phase mismatch are pressure-dependent and the phase matching is achieved through a balance between the neutral dispersion and plasma dispersions in a weakly ionised gas. Even if the phase mismatch is minimised, the absorption in the gas medium still limits the maximum efficiency of the harmonic emission [11–13]. In most previous studies, the relevance of the absorption limit on the XUV radiation was investigated by studying the dependence of XUV photon flux on pressure changes. Constant *et al.* [11] did an analysis of the time-dependent factors that control the conversion efficiency, such as the atomic response, phase matching conditions, and absorption of the medium. They found that the overall optimising conditions in the absorbing medium are $L_{\text{med}} > 3L_{\text{abs}}$ and $L_c > 5L_{\text{abs}}$, where L_{med} is the interaction length, L_c is the coherence length, and L_{abs} is the absorption length. By taking into account the factors limiting the HHG efficiency, including phase mismatch, defocusing and absorption, Meyer *et al.* developed the following expression for the intensity of the q th harmonic in an absorptive medium [13].

$$\begin{aligned}
 I_q &= \frac{\pi\omega_q^2}{2cn_q} |d_q^{NL}|^2 N^2 L^2 \exp\left(-\frac{\alpha_q L}{2}\right) \frac{\sin^2\left(\frac{\Delta k_q L}{2}\right) + \sinh^2\left(\frac{\Delta k_q L}{4}\right)}{\left(\frac{\Delta k_q L}{2}\right)^2 + \left(\frac{\Delta k_q L}{4}\right)^2} = \\
 &= \frac{\pi\omega_q^2}{2cn_q} \frac{|d_q^{NL}|^2}{\sigma_q^2} \cdot \frac{1}{1+\delta^2} \left[1 + \exp(-\tau_q) - 2 \cos\left(\frac{\delta\tau_q}{2}\right) \exp\left(-\frac{\tau_q}{2}\right)\right]
 \end{aligned} \tag{1}$$

where L is the medium length, $\sigma_q = \alpha_q/N$ is the absorption cross section, $\tau_q = \alpha_q L$ and $\delta = 2\Delta k_q/\alpha_q$.

We have also shown that when the phase mismatch is minimised, the harmonic yield approaches its maximum value and the harmonic intensity decreases gradually as the gas pressure increases [14–16]. Most previous research on HHG has been conducted in a noble gas medium. Recently, there has been an increase of interest in using a gas mixture as an HHG target [17–20]. Potentially increasing HHG efficiency and extending HHG application in atoms and molecules are two main advantages of HHG from a gas mixture. In particular, Takahashi *et al.* performed

dramatic enhancement of harmonic generation by adding low ionisation potential gas (xenon) to high potential gas (helium) [17]. A key feature of their experiment is the use of mixed gases with large different ionisation energies. Consequently, the harmonic photons generated from xenon promote a transition of electrons to virtual states, which significantly reduces the tunnelling barrier leading to the enhancement of the tunnelling ionisation rate of helium. Therefore, the conversion efficiency of higher order harmonic generation from helium is increased by a factor of 4×10^3 . Using a mixture of helium and neon, Kanai *et al.* also observed constructive interference and destructive interference structures in the HHG spectrum from this gas mixture [18]. Based on these features, the relative phase of harmonics was successfully retrieved. Using a mixture of krypton and carbon dioxide, the same group was able to reconstruct the molecular structure of CO₂.

In this paper, we use pure argon gas and an argon-neon gas mixture to experimentally investigate the harmonic generation in a semi-infinite gas cell geometry. Particularly, we utilise the high XUV photon flux generated from argon to study the influence of neon atoms on the phase matching condition of the harmonics in low photon energy region (< 50 eV), where this gas could not produce XUV radiation efficiently because of the low driving laser intensity. Based on the findings of this study, the effects of the macroscopic phase matching which plays the important role in generation of bright and sharp harmonics are clearly revealed.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. In this study, a laser system delivering 800 nm, 30 fs pulses at 1 kHz repetition rate is used. The driving laser beam is focused by a 300-mm lens into a long gas cell (200 mm) with a glass window at the entrance and a ~ 0.1 mm pinhole in the aluminium plate at the exit. The hole is drilled by the laser pulses allowing straight-forward handling. The length of the gas cell is chosen according to the focusing lens used for the experiment and normally this length is of order ~ 10 cm. Different from most previous studies using mixed gases, our semi-infinite gas cell geometry that provides a long interaction length allows the production of brighter harmonics than do sub-centimeter cells. Moreover, a greater stability is achieved when the laser beam can freely propagate through the nonlinear medium instead of being confined between an entry and an exit gas nozzle [21]. The long cell is filled with mixed gases of argon and neon and the cell's pressure is measured using a pressure gauge Pfeiffer Vacuum D-35614 Asslar. The XUV radiation is created in a range of ~ 3 mm before the exit pinhole of the cell and then it is isolated from the driving beam by means of a 200 nm-thick aluminum (Al) filter before entering a flat-field XUV spectrometer comprising a slit, a concave grating, and a cooled 14-bit CCD camera.

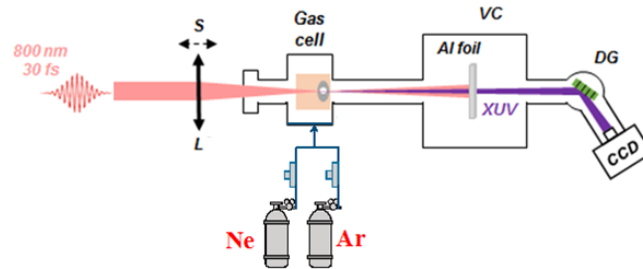


Fig. 1 – (Color online) Experimental setup.

3. RESULTS AND DISCUSSION

3.1. HIGH HARMONIC GENERATION WITH ARGON GAS

Argon gas at 50 Torr in the gas cell is used as the HHG medium. Figure 2 shows a sharp HHG spectrum of argon, which can be obtained by adjusting macroscopic parameters such as the interaction length, laser intensity, and gas pressure. The laser energy is approximately 0.6 mJ and the focus position is about 2 mm inside the gas cell. It is important to note that using an aperture before focusing the driving laser into the gas cell enables us to confine the ionisation fraction of argon to below a critical level ($\sim 5\%$). In addition, the beam profile of H25 (38.74 eV) depicted in the inset of Fig. 2 demonstrates a high spatial coherence that is reflected in the good Gaussian profile. This demonstrates the predominance of short trajectories in the harmonic radiation [22, 23], and the harmonic radiation generated by the atomic ensemble accumulates coherently and is in phase along the effective interaction length [15, 24]. In this measurement, the effective interaction length is inferred from the displacement of the focus position over which the phase matching condition for HHG is satisfied.

The phase matching of HHG is investigated by analysing the variation of the harmonic intensity with respect to argon gas pressure. Figure 3 shows the change of the H25 intensity (red diamonds) as the gas pressure is varied from 20 to 100 Torr while all other experimental parameters remain constant. The H25 intensity is observed to increase between the lowest gas pressure and the optimal pressure (50 Torr), then decrease as the pressure increases. We notice that the development trend of the harmonic intensity closely matches the model given in equation (1). In fact, the strength of the harmonic H25 increases quadratically with argon gas pressure for pressure $p < 50$ Torr. This demonstrates phase-matched harmonic generation in an absorbing medium [11]. As the pressure is higher than 50 Torr, the intensity of the 25th harmonic quickly reduces due to a large dispersion of the medium and re-absorption of the gas medium. When $p > 65$ Torr, the efficiency of the harmonic generation surpasses the absorption limit at which the intensity starts to exponentially decay with pressure.

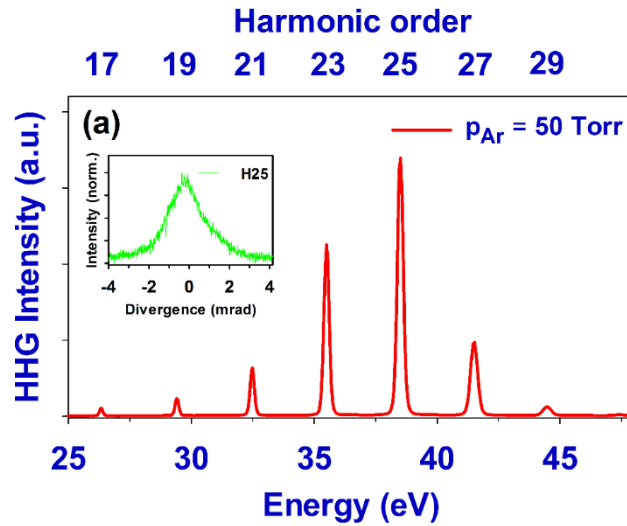


Fig. 2 – (Color online) Phase-matched HHG spectrum from argon gas.

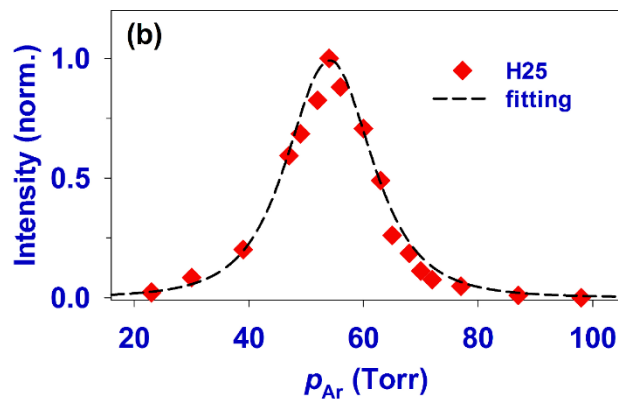


Fig. 3 – (Color online) Dependence of the H25 intensity on the argon gas pressure.

3.2. HIGH HARMONIC GENERATION WITH A MIXTURE OF ARGON AND NEON

In this investigation, neon gas is added in a step-wise manner of 10 Torr to the argon gas, which is initially optimised at 50 Torr. When all other macroscopic parameters remain unchanged, an addition of the neon gas into the mixture causes the measured harmonic photon flux to significantly drop. This is attributed to both the neutral gas dispersion arising from the neon gas and its re-absorption. To minimise such a decrease of the harmonic intensity, we increase the driving laser intensity so that the plasma dispersion can compensate the dispersion of the neutral gas and the Gouy phase shift. Figure 4a shows the harmonic intensity as a function of the total gas

pressure for H23 (35.65 eV) and H27 (41.84 eV), which typically represents the harmonic orders below and above the absorption edge (~ 40 eV) of argon gas, respectively. It is important to note that, as shown in the inset of Fig. 4a, the absorption coefficients of the neon gas for the two harmonic orders H23 (red circle) and H27 (black triangles) are very similar. Figure 4b plots the optimal driving laser intensity (in the order of 10^{14} W/cm²) required to produce sharp HHG spectra, from which the harmonic intensities of H23 and H27 are extracted and shown in Fig. 4a.

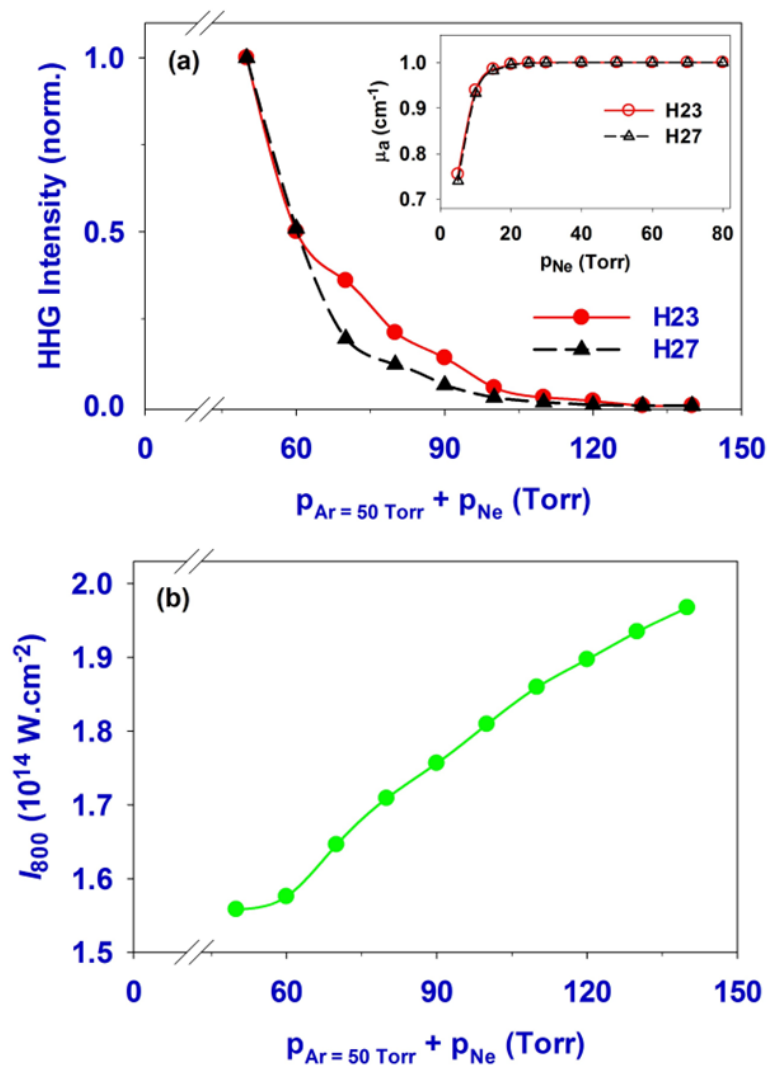


Fig. 4 – (Color online) Normalised H23 (red circles) and H27 (black triangles) intensities (a) and driving laser intensity (b) *versus* gas pressure of the argon and neon mixture. The inset of Fig. 4a shows the absorption coefficient of the neon gas for the two harmonics H23 and H27 [23].

When only 10 Torr of neon gas is added to 50 Torr of argon gas, the harmonic intensity of the two harmonics is suppressed by 50% in comparison to the HHG produced with 50 Torr of argon gas. As more neon gas is pumped into the gas cell, the harmonic strength keeps decreasing. To keep all available harmonics intense and spectrally narrow when injecting neon gas, a higher laser intensity is applied into the interaction medium. The H27 intensity decreases more quickly than that of the H23. Since the absorption coefficients of the harmonics H23 and H27 are the same over the investigating range of the neon gas pressure (inset of Fig. 4a) [23], the absorption effect of the neutral neon atoms is the same for both harmonics. Therefore, under our experimental conditions for such a gas mixture, as the driving laser intensity is increased to minimise the total dispersion, the higher order harmonics in the cut-off region is less favourably phase matched than the lower order harmonics in the plateau region. It is worth noting that the absolute intensity of the 23rd harmonic is always higher than that of the 27th harmonic.

For a better understanding of the data, we perform a calculation of the total phase mismatch of the H23 and H27 at different pressures of the gas mixture. The total phase mismatch can be given by [11]

$$\begin{aligned} \Delta k_q &= \sum_i (k_{q,i} - qk_{l,i}) \\ &\cong \sum_i \left[qp_i(1 - \eta_i) \frac{2\pi}{\lambda_l} \delta n_i + \eta_i p_i N_{atm} r_e \lambda_l \left(q - \frac{1}{q} \right) \right] + \frac{1}{z_R} (1 - q), \end{aligned} \quad (2)$$

where: i is Ar or Ne, q – harmonic order, p_i – pressure of gas i in atmosphere, η_i – ionisation rate of gas i , N_{atm} – gas density at 1 atm ($N_{atm} = 2.5 \times 10^{25} \text{ m}^{-3}$), r_e – classical electron radius ($r_e = 2.8 \times 10^{-15} \text{ m}$), λ_l – the 800-nm driving laser field and $\delta n_i = n_i(\lambda_q) - n_i(\lambda_l)$, and z_R is the Rayleigh length. $n_i(\lambda_j)$ denotes refractive index of the field j in the medium i . The difference of the refractive index between the XUV and the driving laser field in argon and neon are $\delta n_{Ar} = -2.82 \times 10^{-4}$ and $\delta n_{Ne} = -6.7 \times 10^{-5}$ [25, 26]. In Equation (2), the first, the second, and the third term denote the neutral dispersion, the plasma dispersion and the Gouy phase shift. In this study, the contribution of the intrinsic dipole phase into the total phase mismatch is negligible because the Rayleigh length ($z_R \cong 10 \text{ cm}$) is longer than the effective interaction length of about 3 mm.

Figure 5 displays the calculated total phase mismatches of the two harmonics H23 (red circles) and H27 (black triangles). Experimental parameters, *i.e.*, pressure of the gases, driving laser intensity, ionisation rate of each gas, for the data shown in Fig. 4 are adapted to the calculation. Even though stronger driving laser field is applied to counterbalance the dispersion arising from the neutral dispersion and Gouy phase shift, the total phase mismatches of these two harmonics increase linearly with the gas density when more than 10 Torr of neon gas is added to the

original 50 Torr of argon. Additionally, the increasing rate of the Δk_{27} (rad/mm) is higher than that of the Δk_{23} (rad/mm). In other words, the phase matching condition of the harmonic H27 is getting worse more quickly than that of the harmonic H23. This is in good agreement with our experimental data, which demonstrates that the decreasing rate of the intensity of the 27th harmonic is greater than that of the intensity of the 23rd harmonic (Fig. 4a).

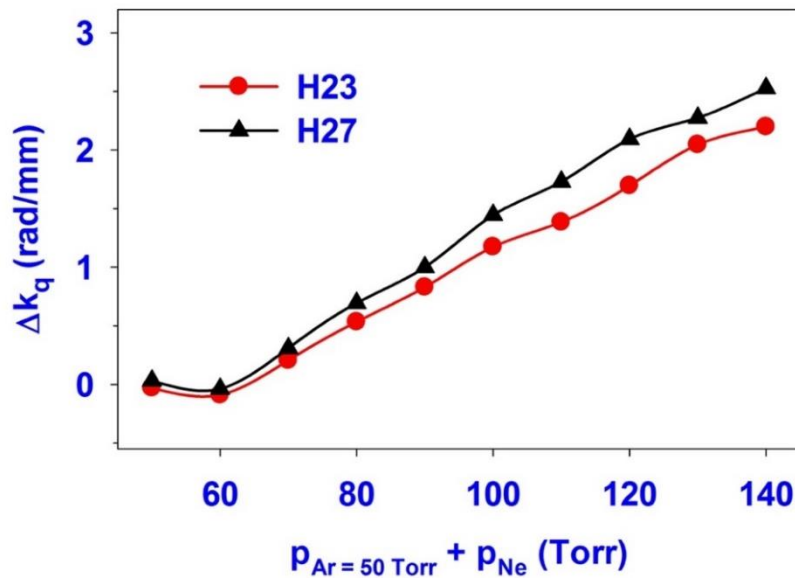


Fig. 5 – (Color online) Calculated total phase mismatch of H23 and H27 when the pressure of the gas mixture is varied by adding neon into the original 50 Torr of argon gas.

4. CONCLUSION

We have experimentally studied the phase matching condition of high harmonic generation with argon gas, and a mixture of argon and neon. The pressure-dependent harmonic intensity in a long gas cell filled with argon gas, and gas mixture of argon and neon is investigated. A linear increment of the total phase mismatches of two harmonics generated in the mixture of these two gases and lying at the absorption edge of argon gas is also examined. We showed that the higher-order harmonic is less favourably phase matched than the lower-order one when more than 10 Torr of neon gas is added into argon gas. Our experimental results agree well with the theoretical calculation.

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