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ABSTRACT

We report here evidence of phase-matched optical wave mixing in the extreme ultraviolet (XUV) region. This process has been studied with a collinear two-colour high-order harmonic generation scheme. An 800 nm, 30 fs driving field is used to produce a small bandwidth comb of odd harmonic orders (wavelength around 30 nm) in a long cell filled with argon gas. Mixing frequencies in this spectral range are produced by applying a second weak control-field of 1,400 nm, 40 fs. Low order (third- and fifth-order) nonlinear optical wave mixing is observed to be a phase-matched process. The dependence of the intensity of the harmonic orders and the mixing frequencies on different control-field intensities, gas pressure, and interaction length is analysed to verify the phase matching process.

Keywords: two-colour high-order harmonic generation, phased-matched optical wave mixing, mixing frequencies, extreme ultraviolet, argon

1. INTRODUCTION

High-order harmonic generation (HHG) is a strongly nonlinear process in which the frequency of a fundamental laser is up-converted into its harmonics. HHG was experimentally recorded before appropriate theoretical understandings\textsuperscript{1,2} and coherent control methods\textsuperscript{3,4} were widely studied. One of the breakthrough achievements with one-colour HHG, which has been applied in later studies such as XUV interferometry,\textsuperscript{5} attosecond transient absorption spectroscopy,\textsuperscript{6,7} coherent diffractive imaging,\textsuperscript{8,9} autoionisation of gases,\textsuperscript{10,11} etc., is the generation of phase-matched and coherent soft X-rays.\textsuperscript{12}

Studies of the dynamics of free electron wave-packets in atoms/molecules require ultrashort pulses with temporal duration in the attosecond range. Recently, a two-colour HHG configuration was made possible to produce such a source without the need for state-of-the-art facilities.\textsuperscript{13} In fact, two-colour HHG was already implemented soon after the realisation of the one-colour HHG.\textsuperscript{14,15} Instead of one driving field, a second weak field is additionally applied to the nonlinear medium.\textsuperscript{14–20} Mixing frequencies are then generated when the two constituent fields spatially and temporally overlap. These sources, which are necessary for nonlinear XUV and soft X-ray spectroscopy techniques, are only coherent as long as they are generated with a phase-matched condition. In one of the recent reports, the time-delay-dependence intensity of the phase-matched mixing waves is proposed to provide the evolution information of free electron wave-packets.\textsuperscript{20}

In this paper, studies of the wave-mixing processes in the spectral range around 30 nm are performed. We utilise a collinear scheme in which the HHG is generated by the 800-nm laser pulses in an argon cell. A near-infrared beam at 1,400 nm is then shone on the medium to produce mixing frequencies. The experimental results reveal that the accumulation of these coherent mixing fields can be attributed to the strong low-order nonlinear response of the argon gas. The dependence of the intensities of the harmonic orders and the mixing frequencies on the strength of the control field, the argon gas pressure up to 200 torr and the interaction length are discussed to confirm that the optical wave mixing fields in this spectral region are generated under the phase-matched condition.

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2. EXPERIMENTAL SETUP

Laser pulses of energy 6.0 mJ, time duration of 30 fs and central wavelength of 800 nm (carrier frequency $\omega_1$) are produced by a 1 kHz multi-stage, multi-pass chirped-pulse amplifier system. The laser beam is divided into two separate beams by a 30/70 (R:T) beam splitter. The transmitted beam (4.0 mJ) is truncated with an iris and then focused into a long gas cell with a convex lens ($f = 20 \, \text{cm}$) for the generation of high-order harmonics. Polarisation of the beam is varied with a half-wave plate. The reflected beam from the beam splitter ($\sim 2 \, \text{mJ}$) is directed into an optical parametric amplifier to create a 0.4 mJ signal wave at 1,400 nm (carrier frequency $\omega_2$) and temporal duration 40 fs. This beam co-propagates with the driving beam into the cell which has a glass window. An exit hole on the rear side of the cell is drilled with the focused 800-nm laser pulses. The diameter of the two beams at the focal point is about 150 $\mu$m. The HHG optimised with the 800 nm pulses (effective intensity of $2 \times 10^{14} \, \text{W/cm}^2$) is produced near the exit of the gas cell. The optimised phase-matching condition is maintained during the experiment. The intensity of the 1,400 nm ($< 5 \times 10^{13} \, \text{W/cm}^2$) is varied with neutral-density filters so that it neither produces its harmonic frequencies nor saturates the intensity of the mixing fields. A DC motor with 0.1 fs temporal resolution is used to control the time delay between the two fundamental pulses. The focal plane of the focusing lens is shifted with a translation stage with a spatial resolution of 24 $\mu$m. The output spectrum from the gas cell is a mixture of high harmonic radiation and the fundamental fields. The driving and control fields are blocked by a 300-$\mu$m-thick aluminium foil before the XUV fields enter a diffraction grating and an X-ray CCD camera. These two devices are installed in a vacuum chamber in which the pressure in the vicinity of the CCD chip is held at about $10^{-5}$ torr.

In this report, we discuss the data obtained with parallel polarisation between the two beams. Zero-time delay is assumed to be when the two pulses temporally overlap, and negative time delay implies that the 1,400 nm precedes the 800 nm beam.

3. THEORETICAL BACKGROUND

3.1 Phase-matched high-order harmonic generation

High-order harmonic generation is a highly nonlinear process which occurs as an intense driving laser interacts with a medium. High-intensity driving field ionises the medium which is consequently a superposition of both atomic gas and free-electron plasma. Due to a difference in the frequency between the fundamental ($\lambda_l$) and the harmonic fields ($\lambda_q$), there is a discrepancy in the phase velocities, $v_p = \frac{\omega_p}{c} = \frac{n}{n'}$, of the two beams as they propagate in the interaction volume. The $v_p$, $\omega$, $k$, $c$ and $n$ denote the phase velocity, carrier frequency, propagation constant, speed of light in the vacuum, and the refractive index, respectively. The propagation constant $k$ and the index of refraction $n$ are in turn functions of the wavelength $\lambda$. Therefore, there is a wave-vector mismatch of the $q$th harmonic order

$$\Delta k_q = k_q - qk_l.$$  

(1)

For a partially ionised gas, the phase mismatch $\Delta k_q$ has contributions from four sources: the neutral gas making up the interaction medium, free electrons released from the atoms in the HHG process, the phase-shift incurred by the loosely-focused laser beam and the intrinsic, intensity-dependent dipole phase of the $q$th harmonic. The neutral dispersion

$$\Delta k_{\text{neutral}} = n(\lambda_q) \frac{\omega_q}{c} - qn(\lambda_l) \frac{\omega_l}{c} = \frac{q^2 \pi}{\lambda_l} \delta n(\lambda),$$  

(2)

where $\omega_q = q\omega_1$ and $\delta n(\lambda) = n(\lambda_q) - n(\lambda_l)$. This contribution is negative because the refractive index of the harmonic radiation $n(\lambda_q)$ in the XUV region is always smaller than that of the near-infrared driving field $n(\lambda_l)$.

During the HHG, a large number of free electrons are liberated but most of them do not return to the parent ions. The accumulation of these negatively-charged particles in the interaction medium therefore introduces a polarisability characterised by a plasma frequency $\omega_p = \sqrt{\frac{N_e}{\epsilon_0 m_e}}$, where $\epsilon_0$ is the dielectric constant, $m_e$ the electron mass, $e$ the charge of the electron, and $N_e$ the density of the free electrons. This process induces the refractive index$^{21}$

\[ n_{\text{plasma}}(\omega) = \sqrt{1 - \left(\frac{\omega_p}{\omega}\right)^2} = \sqrt{1 - \frac{N_e}{N_c(\omega)}}, \]  
\hspace{1cm} (3)  

where \( N_c(\omega) = \frac{\epsilon_0 m_e \omega^2}{e^2} \) is the critical plasma density. With a driving field of 800 nm, the free-electron density generated is much lower than the critical density \( (N_e \sim 2 \times 10^{21} \text{cm}^{-3}) \). Therefore,  
\[ n_{\text{plasma}}(\omega) \approx 1 - \frac{1}{2} \left(\frac{\omega_p}{\omega}\right)^2. \]  
\hspace{1cm} (4)  

The plasma contributes to the wave-vector mismatch  
\[ \Delta k_{\text{plasma}} = \frac{q \omega i}{c} [n_{\text{plasma}}(\lambda_i) - n_{\text{plasma}}(\lambda_q)] = N_e \lambda_i r_e (q - \frac{1}{q}), \]  
\hspace{1cm} (5)  

where \( r_e = \frac{e^2}{4\pi\epsilon_0 m_e c^2} \) is the classical radius of the electron. The free-electron plasma makes a positive contribution to the total phase-mismatch for harmonic orders \( q \gg 1 \). In our studies, the Rayleigh length of the driving laser (5 mm to 20 mm) is much longer than the interaction length (\( \sim 2 \) mm). Therefore, the phase-matching condition for efficient HHG can be achieved when the neutral dispersion counterbalances the plasma dispersion without considering dispersion contributed by the Gouy phase shift and the intrinsic harmonic dipole phase;\(^{22}\)  
\[ \Delta k_q = \frac{q 2\pi}{\lambda_i} \delta n(\lambda) + N_e \lambda_i r_e (q - \frac{1}{q}) \approx 0. \]  
\hspace{1cm} (6)  

When the total phase mismatch is minimised, the strength of the harmonic is significantly enhanced. However, the HHG up-conversion efficiency is limited by reabsorption in the nonlinear interaction medium. The intensity of a phase-matched harmonic order in a dispersive and absorbing medium is then estimated to be\(^{23-25}\)  
\[ I_q \approx N^2 L^2 |d^{NL}_q| e^{(-\alpha_q L)} \frac{\sin^2(\frac{\Delta k_q L}{2}) + \sinh^2(\frac{\alpha_q L}{4})}{(\frac{\Delta k_q L}{2})^2 + (\frac{\alpha_q L}{4})^2}. \]  
\hspace{1cm} (7)  

In Eq. (7), re-absorption of the harmonic signal by the gas medium is taken into account, \( N \) is the neutral particle density, \( L \) is the interaction length, \( d^{NL}_q \) is the amplitude of the nonlinear atomic dipole moment, and \( \alpha_q \) is the XUV absorption coefficient of the generating medium. The harmonic intensity \( I_q \) reduces to the familiar \( \text{sinc}^2(\Delta k_q L/2) \)-dependence when the \( \alpha_q \) is small.  

### 3.2 Phase-matched four-wave mixing  
The wave-mixing process in the XUV region involving photons of the HHG, the driving and the control fields is coherent over the interaction length \( L \). Therefore, the intensity of a four-wave mixing field\(^{26}\)  
\[ I_4 \sim I_1 I_2 I_3 |\chi^{(3)}|^2 N^2 L^2 \text{sinc}^2(\frac{\Delta k L}{2}), \]  
\hspace{1cm} (8)  

where \( I_1, I_2, I_3 \) are the intensities of the driving, control and HHG fields. The phase mismatch for the generation of the four-wave mixing process is  
\[ \Delta k = k_4 - k_3 \pm (k_1 - k_2), \]  
\hspace{1cm} (9)  

where \( k_1, k_2, k_3 \) and \( k_4 \) are the wave-vectors of the driving field, the NIR control field, the \( q \)th-order harmonic and the mixing field, respectively. According to Eq. (8), when the wave-mixing process is induced with constant fields and a small phase mismatch, the intensity increment of the mixing frequencies is proportional to the square of the atomic number density and the interaction length. In such a case, this is evidence of phase-matched generation of the four-wave mixing fields.
4. RESULTS AND DISCUSSION

4.1 Phase-matched high-order harmonic generation

In this study, the radius of the focal point can be varied from 45 \( \mu m \) to 100 \( \mu m \) over which the Rayleigh length ranges from 5 \( mm \) to 20 \( mm \). The HHG spectrum with argon gas at 120 torr is shown in the - 150 \( fs \) plot (blue dashed line) of Fig. 1. At a large time delay of - 150 \( fs \) between the two pulses or absence of the second beam, the effect of the control field on the HHG process is negligible. A small numbers of sharp harmonic orders are observed under the current experimental conditions. The spectral width of each order in the harmonic comb is very narrow. The sharpness and spectral width of all harmonics are almost constant at different time delay, see plots 0 \( fs \) (red solid line) and - 150 \( fs \). In the two insets (a) and (b) of Fig. 1, the beam profiles of the harmonics H31 (plot a), H29 (plot b), H27 (plot c) at - 150 \( fs \) and H29 at 0 \( fs \) (plot d) are shown with good spatial coherence.

![Figure 1. HHG spectrum with argon gas at 120 torr at 0 fs (red solid line) and - 150 fs (blue dashed line). The spectrum at - 150 fs only comprises odd harmonic orders (i.e., H23 to H35). However, the spectrum at 0 fs exhibits both odd harmonic orders and four additional mixing peaks on either side of each harmonic. The two insets show beam profiles of (a) H31 (plot a), H29 (plot b) and H27 (plot c) at - 150 fs, and (b) H29 (plot d), H29 + \( \Delta \omega \) (plot e) and H29 - \( \Delta \omega \) (plot f) at 0 fs.](https://www.spiedigitallibrary.org/conference-proceedings-of-spie)

We also record strong and sharp HHG spectra when the focus position of the driving laser pulse is shifted relative to the exit of the gas cell and when the gas pressure is tuned, Fig. 2. In Fig. 2(a), the highest photon counts of all available harmonic orders is initially achieved with argon at 120 torr and the position of the focusing lens set to 0 \( mm \); the position of the focal point is then varied while all other parameters are kept constant. We note that the 0-mm-position is at the exit of the gas cell. Positive and negative values consequently imply that the focal point is inside and outside of the gas cell, respectively. The accumulation distance of all harmonic orders at this pressure is about \( \sim 2 \ mm \). The strength of all harmonics rises when the focal point is scanned from outside toward the exit pin hole of the cell. However, the intensity of the harmonics below H27 (> 30 \( nm \)), e.g., H23 and H25, is seen to quickly decrease when the position of the laser focus moves deeper into the gas cell. This is caused by the re-absorption effect of the interaction medium on the harmonic signal.

In Fig. 2(b), the HHG spectrum is firstly optimised at 120 torr, then the pressure is tuned. The intensity of the harmonic grows between 55 torr and about 120 torr, then decreases as the pressure keeps increasing. However, the spectral aspects, i.e., sharp and small bandwidth, of all harmonic orders is maintained as the gas pressure is varied. We also note that the generation of the H27 to H31 orders is enhanced when the pressure increases up to 120 torr but the intensities of the lower orders are strongly diminished. This is due to a higher transparency of the argon gas in the spectral range < 30 \( nm \).
Figure 2. Development of HHG spectrum taken at time delay - 150 fs with argon gas (a) when the focus position is shifted from outside (negative position) to inside (positive position) the cell filled at 120 torr, and (b) when the focal point is fixed at position 0 mm and the gas pressure is tuned from 55 torr to 200 torr.

In Figs. 3(a) and (b), the data of the most intense harmonic order H29 (black squares and black circles), which are extracted from the Figs. 2(a) and (b), respectively, is shown for a discussion of the phase-matching condition in the HHG production. The development trends of the intensity profile H29 (Fig. 3) fit closely with the model given by Eq. (7) which indicate a quadratic dependence of the harmonic strength on the interaction length \( L \) and on argon gas pressure \( p \). These results are evidence of phase-matched HHG in a strongly absorptive medium. However, when \( L > 0 \) (Fig. 3(a)) and \( p > 120 \) torr (Fig. 3(b)), the efficiency of the harmonic generation surpasses the absorption limit at which the intensity exponentially decays with the interaction length and pressure.

Figure 3. Dependence of the intensity of the harmonic H29 on (a) the focus position and (b) on the argon gas pressure. The experimental data (scattered plots) in the two plots are extracted from the two data sets shown in Figs. 2(a) and 2(b), respectively. The two lines accompanying the scattered plots are the corresponding fitting curves, using Eq. (7).

In conclusion, sharp and narrow bandwidth HHG is produced in our investigation of interaction length-dependence and pressure-dependence. The corresponding interaction length of 2 mm is smaller than the estimated Rayleigh length (from 5 mm to 20 mm), and there is a significant \( L^2 \)-dependence and \( p^2 \)-dependence of the HHG intensities when the experimental condition is optimised at 120 torr. Thus, HHG is generated with phase-matched condition.

4.2 Phase-matched four-wave mixing

In Figs. 1, 2, 4 and 5(b), the orders of all harmonics are labelled with corresponding integer numbers. In addition to the main harmonics, four extra frequencies are recorded on either side of each odd harmonic at zero-time delay.
(Figs. 1, 4 and 5(b)). The spatial profiles of the main harmonic, e.g., H29 (plot d in inset (b) of Fig. 1) and the new frequencies, i.e., H29 ±Δω (plots e and f in inset (b) of Fig. 1) at 0 fs are good. Moreover, the spectra of the new fields are as sharp as those of the main harmonics. The sharpness of both the main harmonics and these new frequencies is also found to be preserved when the interaction length and gas pressure are changed, see Fig. 4. This is clear evidence of wave-mixing processes in the XUV region. The photon energy difference of the driving field and the control field is Δω = ω1 − ω2 ≃ 0.67 eV. On each side of one harmonic, the energy gap between it and the first-order mixing wave and between the first- and a second-order mixing wave is also estimated to be ~ 0.67eV. The energy of the qth-order harmonic is represented by ω3 ≡ qω1; the corresponding energy of the mixing waves ωmix is then approximately ω3 ± mΔω (m = 1, 2), where the “+” and “−” signs of mΔω denote the two closest peaks to the left (shorter wavelength) and to the right (longer wavelength) of the qth-order. These new frequencies are generated with sum-frequency mixing (SFM) and difference-frequency mixing (DFM) processes in the XUV region, respectively.\(^{14}\)

![Harmonic order](image)

**Figure 4.** Illustration of sharp and constant bandwidth of both harmonics and wave-mixing fields generated at zero-time delay with argon gas (a) when the focus position is shifted from outside (negative position) to inside (positive position) of the cell filled at 120 torr, and (b) when the focal point is fixed at the position 0 mm and the gas pressure is tuned from 55 torr to 200 torr. These results are taken from the same data set shown in Fig. 2.

The HHG field ω3 is generated under the phase-matching condition, Section 4.1. The experimental data reveals that the frequency of the mixing wave

$$\omega_{mix} \simeq \omega_3 \pm m(\omega_1 - \omega_2).$$  \hspace{1cm} (10)

The phase of the harmonics remains unchanged over the interaction length because the neutral and plasma dispersion is small for an XUV pulse, and (k1 − k2) ≪ k3, kmix. Thus, the total wave-vector mismatch for the generation of the mixing field

$$\Delta k = k_{mix} - k_3 \pm m(k_1 - k_2) \approx 0.$$  \hspace{1cm} (11)

The SFM and DFM processes occur in the presence of the second field ω2; therefore the intensity of the mixing field should obey Eq. (8). The variation of the intensity of the strongest harmonic order H29 and the mixing frequencies H29 ± mΔω versus the relative intensity of the control field I2 at 120 torr is shown in Fig. 5(a). Similarly, we also illustrate in the inset of Fig. 5(a) the intensity of other first-order mixing fields, i.e., H31 ± Δω (plots e and f) and H27 ± Δω (plots g and h). The data in this plot is extracted at zero-time delay. The highest intensity of the control field, max(I2) ≡ I0, is controlled below the threshold at which the signal of the first-order mixing waves starts to saturate. We observe a similar depletion of the main harmonic orders H23-H35 while the intensity of all corresponding mixing fields increases. Namely, the increasing intensity of the mixing fields and the decreasing intensity of the odd harmonics are observed to scale monotonically with I2 when I2 ≤ 0.2I0. However,
Figure 5. (a) Intensity of the harmonic H29 (plot a), H29 - Δω (plot b), H29 + Δω (plot c) and H29 - 2Δω (plot d) versus the relative intensity of the control field. The inset of (a) illustrates the dependence of the intensity of the first-order mixing fields, i.e., H31 ± Δω (plots e and f) and H27 ± Δω (plots g and h), on the control field. (b) Typical two-colour HHG spectrum (the three strongest harmonic orders H27 to H31 and their corresponding mixing fields) as a function of the time delay between the two fundamental pulses. The data are taken with argon gas at pressure 120 torr.

for 0.2I₀ ≤ I₂ ≤ 0.4I₀, the second-order mixing fields continue to increase linearly with I₂ but the increment of the first-order intensity and the decline of the HHG intensity is no longer linear. In this range of I₂ the intensity of the harmonics is low; therefore there is a decrease of the intensity of the HHG I₃ in Eq. (8). Additionally, the higher-order nonlinear response of the interaction medium also needs to be taken into account. With the power scaling of the mixing fields, the depletion of the original odd harmonics, the correlation between the intensities of the original and new fields (Eq. (8)) and the allowed photon combinations (Eq. (10)), the nonlinear optical wave-mixing processes in the XUV region involving the low-order nonlinear susceptibilities are thus attributed to these observations. We also demonstrate in Fig. 5(b) the time-delay-dependence spectrum with argon gas at 120 torr. The applied intensity of the control field is about 0.3I₀. The observation of mixing waves at large positive time delay might be an indirect signature of cascaded wave-mixing in this spectral region.²⁰

Figure 6. Intensity profiles of H29 and H29 ±Δω as a function of (a) the focus position and (b) the gas pressure. H29 at -150 fs (plots a and e), H29 at 0 fs (plots b and f), H29 + Δω at 0 fs (plots c and g), and H29 - Δω at 0 fs (plots d and h). The data set shown here are extracted from that of Fig. 4.

The influence of the phase-mismatch on the generation of the mixing fields is investigated in detail by
considering the dependence of the output intensities of these fields on the interaction length and on the pressure of the argon gas. The experimental data shown in Figs. 6(a) and (b) are extracted from that illustrated in Figs. 4(a) and (b), respectively. In Fig. 6, we compare the signal of the H29 at - 150 fs, the H29 and the H29 ±Δω at 0 fs in order to see whether the phase-matched condition for the wave-mixing processes is fulfilled. The signal of the harmonic H29 is generated with the phase-matched condition (Section 4.1). As can be seen in Fig. 6(a), the trend of the mixing waves H29 ±ω at 0 fs (plots c and d) closely follows that of the harmonic H29 at - 150 fs and at 0 fs (plots a and b). This indicates that the wave-mixing frequencies are generated with the phase-matching condition. In Fig. 6(b), for p < 120 torr, the increase of the intensities of the H29 ±Δω at 0 fs (plots g and h) also follows that of the main harmonic H29 at - 150 fs (plot e). Therefore, this is additional evidence for phase-matched optical wave mixing processes in our study. However, we note that the phase-matched condition for the H29 at 0 fs (plot f) only holds for p < 100 torr and the phase-matching condition of the H29 - Δω is better than that of the H29 + Δω. Finally, for L > 0 mm and p > 120 torr, the strength of all harmonic orders and mixing waves is dominated by an exponential decay with the interaction length and gas pressure due to reabsorption in the gas medium.

5. CONCLUSION

We have studied phase-matched optical wave mixing processes in the extreme ultraviolet with a collinear two-colour HHG configuration. The experimental data reveals clear evidence of a coherent accumulation of the new frequencies and a high third-order nonlinear response of an argon medium. These results will be useful for future research that requires efficient production of coherent XUV and soft X-ray sources for high energy ultrafast nonlinear spectroscopy.

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