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Phase-matching for generation of few high order harmonics in a semi-infinite gas cell

Lap Van Dao *, Sven Teichmann, Peter Hannaford

ARC Centre of Excellence for Coherent X-Ray Science, Centre for Atom Optics and Ultrafast Spectroscopy, Faculty of Engineering and Industrial Sciences, Swinburne University of Technology, Melbourne, Victoria 3122, Australia

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ABSTRACT

Small phase-mismatch between the fundamental and high order polarization in the cut-off region, allowing macroscopic phase-matching to be satisfied for these harmonics can be used for generation of extreme ultraviolet radiation with high flux, high coherence and few harmonic orders in a semi-infinite argon gas cell.

1. Introduction

Coherent XUV radiation can now be produced by laser-based high-order harmonic generation (HHG) in noble gases [1–3]. This table-top set-up will complement larger installations such as X-ray free-electron lasers (XFELS) currently under development and can provide a compact source of coherent emission extending down to the water window region [4], between 2 and 5 nm, a region of great interest for biological applications. Such HHG sources open opportunity for new applications in atomic and molecular spectroscopy, condensed matter physics, imaging on the nano- and subnano-scale, and plasma physics [5–8]. Significant enhancements of the efficiency of HHG have been achieved by several groups through quasi phase-matching [9–14], and the "absorption limit" for which reabsorption limits the effective length of the medium has now been reached [10] but the conversion efficiency depends on the HHG configuration even within the absorption limit and the ultimate optimization conditions are still far from being fully characterized. Using self-guided laser beams [15], hollow waveguides [12] or gas cells [16] previous studies report that the efficiency of the high order harmonics near the cut-off region can be enhanced while that of the harmonics in the plateau region is reduced.

The flux, the photon energy and the spatial coherence of the radiation are important properties of the source from the point of view of potential applications. In principle the spatial and temporal coherence properties of the incident laser are transferred to the generated HHG field. But the nonlinear polarization induced in the medium is a complex function of space and time and this can alter the coherence of the harmonic beam generated by the high-order nonlinear processes. For many applications, e.g., coherent diffractive imaging [17], a monochromatic beam is a further important requirement of the source. A spectrometer is typically used to select a single order of the HHG radiation; however the transmission of spectrometers in the XUV range is low. The generation of a few harmonics, or preferably just a single harmonic that can be used directly, without a spectrometer, would be a significant advantage for such applications.

In this Letter we report high harmonic generation in a semi-infinite gas cell. We also show that a high photon flux (about 10^{11}–10^{12} photon/s), high coherence can be achieved for just a few harmonics (23rd to the 33rd), which provides a suitable XUV source in applications such as coherent diffraction imaging or ultrafast spectroscopy.

2. Experiments

The femtosecond laser pulses are generated by a 1 kHz multi-stage multi-pass chirped-pulse amplifier system, which provides 5 mJ pulses with a duration of 30 fs centred at 805 nm. The laser

* Corresponding author at: Swinburne University of Technology, PO Box 218, Hawthorn–Melbourne VIC-3122, Australia. Tel.: +61 3 9214 4317; fax: +61 3 9214 5160.
E-mail address: dvlap@swin.edu.au (L. Van Dao).

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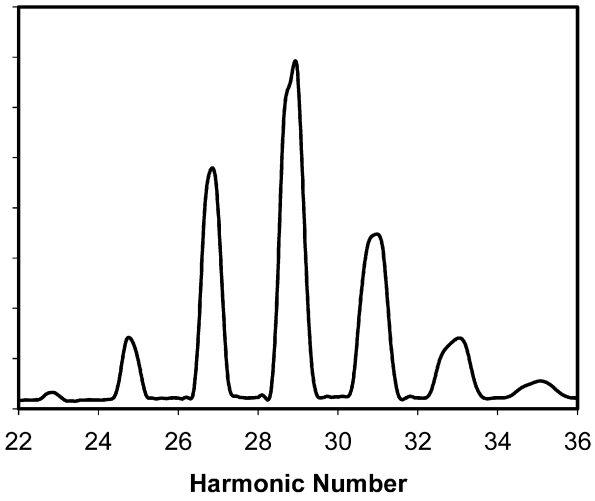

Fig. 1. XUV spectra emitted from argon at 25 Torr for a focus positions at the exit pinhole of the gas cell.

pulses are focussed by a lens with a focal length of 500 mm or 750 mm into a 300 mm long gas cell with a glass window at the entrance and a 200 μm pinhole at exit. The small exit pinhole is used to isolate the vacuum chamber from the argon gas-filled cell. We use argon gas for this experiment because it is transparent and has a relatively large dispersion for harmonics higher than the 27th order (29.6 nm). In the pressure range 10–50 Torr the profile of the transmitted fundamental beam does not change significantly. Harmonics are not generated when the gas cell is evacuated below 10^{-2} Torr. The pressure in the vacuum chamber outside the gas cell is kept at $< 2 \times 10^{-3}$ Torr. The position of the focus point relative to the exit pinhole can be varied. The diameter of an aperture, which is placed in the path of the laser beam before the focussing lens, can be controlled for optimal HHG intensity. A 200 nm-thick aluminium filter with high transmission in the wavelength range of 17–60 nm is used to remove the fundamental beam. The high harmonics pass through a 0.5 mm-wide, 20 mm-high entrance slit of the spectrometer before being dispersed by a 300 lines/mm diffraction grating at grazing incidence and detected by a XUV CCD camera. The overall spectral resolution of the system is about 0.1 nm. To record a large range of HHG orders, a slit is placed at the exit focal plane of the grating and the angle of the grating is scanned. The HHG photon number N_{ph} is calculated from the CCD detector signal (PIXIS-XO-1024B, Roper Scientific) directly and after dispersion by the spectrometer using the expression $N_{\mathrm{ph}} = (N_c \eta / Q\,E) \times (3.65/E_{\mathrm{ph}})$, where N_c is the number of counts per CCD pixel, η depends on the setting of the hardware gain given by the factory, QE is the quantum efficiency of the detector which is nearly constant over the range of photon energies 20–200 eV, and E_{ph} is the photon energy.

3. Results and discussion

Fig. 1 show the XUV spectrum emitted from argon at 25 Torr for focus positions at the exit pinhole of the gas cell. The intensity of the fundamental laser beam is sufficiently high ($5–10 \times 10^{14}$ W/cm^2) to produce harmonics in the plateau region [9,16], but in our experiment less than six harmonic orders are detected and no harmonics in the plateau region are observed. It has been shown that for harmonic generation in different length gas cells [16] the conversion efficiency decreases rapidly with decreasing wavelength because of the phase-velocity mismatch. The phase-velocity mismatch is sensitive to the Gouy phase shift, the density of the gas atoms and the intensity of driving field, and therefore the intensities of the harmonics depend on the focus position, the gas pressure and the intensity of the laser. Each harmonic is red-shifted due to propagation effects [18] and due to the self-phase modulation in the leading edge of the driving pulse and, similar red shifts are also observed when a hollow fibre is used [5,19]. The radial variation of the phase for different quantum pathways [16,20] leads to an effective selection of trajectories where the path with the slowly varying phase survives longest during the propagation and provides a clearly resolved spectrum of harmonics.

The absorption coefficients for the 25th (H25) and the 29th (H29) harmonics differ by more than two orders of magnitude [16], but we still observe an enhancement of both of these harmonics, which is dependent on the size of the fundamental laser beam and the position of the focus. In the pressure range 25–50 Torr we do not observe a strong pressure dependence of the intensity of H25 and H29 when the diameter of an aperture is changed accordingly. This indicates that the absorption effect behaves quite differently in the strong field regime and shows that it does not influence the phase-matching process.

Fig. 2 shows the intensity of H25 and H29 versus distance between the focus point and the exit pinhole. Positive distance indicates that the focus is in front of the pinhole on the side of the gas cell. The intensity of the generated harmonic depends on the gas pressure and the interaction length and is given by $I_q(z) \sim N_{\mathrm{at}}^2 \sin^2(\Delta k_q z/2)/\Delta k_q^2$ where N_{at} is the atom gas density and z is interaction length. This provides positive and negative energy transfer between the fundamental wave and the harmonic wave of order q that depends on the phase mismatch Δk_q [21]. The total phase mismatch of the qth harmonic is $\Delta k_q = (2\pi\omega/c)N_{\mathrm{at}}(q n_{q\omega} - n_\omega)$, where $n_{q\omega}$ and n_ω are the refractive indices of the harmonic wave and the fundamental wave, respectively. If the two wavelengths are phase-matched ($\Delta k \sim 0$) the HHG intensity increases with the length of the interaction medium. When the focus point is changed from a position which is localised in the vacuum region (negative distance) to a position inside the gas cell (positive distance) the interaction length increases. For a gas pressure of 25 Torr (Fig. 2(b)) the intensity of H29 increases quadratically with the interaction length which is consistent with the presence of phase-matching. The harmonics are also generated efficiently if the phase mismatch is small and the medium is long enough for the field to develop.

Guiding the pump laser with a hollow fibre can ensure a constant intensity over the length of the fibre and avoids the Gouy phase shift in the interaction region. The positive dispersion due to the plasma and the negative dispersion due to the neutral argon indicate that the strong enhancement can be attributed to a macroscopic phase-matching in the hollow fibre, but the generated HHG spectra will be broad because of the enhancement of the plateau region [16]. In free space the propagation term induced by the phase shift of a Gaussian fundamental field is proportional to $-q \arctan(2z/b)$, where z is the propagation distance (with $z = 0$ at the focus position), b is the Rayleigh length and q is the harmonic number [11]. Therefore in the case of propagation in a long gas-filled cell, the Gouy phase shift in the fundamental wave over the confocal length can lead to destructive interference between the harmonics created before and after the focus [21]. However, self-guiding of the femtosecond pulses occurs as a result of beam convergence due to self-focussing and beam divergence due to multiple photo-ionization and then the variation of the fundamental wave phase is more complex around the focus point.

Variation of the diameter of the input beam can be used to choose the effective f-number, the spatial quality of the laser beam and the peak intensity in the focus area. The variation of the size of the laser beam also introduces phase variation in the laser wavefront that can be used to minimize the mismatch between the laser phase and the intrinsic phase of the harmonics.