Bright Quasi-Phase-Matched Soft-X-Ray Harmonic Radiation from Argon Ions

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Selective enhancement ($>10^3$) of harmonics extending to the water window ($\sim$4 nm) generated in an argon gas filled straight bore capillary waveguide is demonstrated. This enhancement is in good agreement with modeling which indicates that multimode quasi-phase-matching is achieved by rapid axial intensity modulations caused by beating between the fundamental and higher-order capillary modes. Substantial pulse energies ($>$10 nJ per pulse per harmonic order) at wavelengths beyond the carbon K edge ($\sim$4.37 nm, $\sim$284 eV) up to $\sim$360 eV are observed from argon ions for the first time.

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High harmonic generation (HHG) [1,2] using ultrafast laser sources is an attractive route to generating coherent beams of extreme ultraviolet (XUV) radiation in the 20–400 eV spectral region [3–5]. The key advantages of the HHG approach are the high beam quality, ultrashort pulses ranging from femtoseconds (fs) to attoseconds [6], and the comparative ease with which compact, commercially available femtosecond lasers can be converted to the XUV.

The most significant barrier to widespread application of HHG is the low conversion efficiency, and consequently low output that can be achieved. While reasonable conversion efficiencies in the range of $E_{\mathrm{harmonic}}/E_{\mathrm{Laser}} = 10^{-5}$–$10^{-6}$ (E_{harmonic} and E_{Laser} are the energies in a single harmonic and laser pulse, respectively) have been achieved at photon energies $<$40 eV [3], the reported conversion efficiency at wavelengths $>$200 eV has been much lower (10^{-11}) [7]. This is due to the fact that the highest efficiencies can only be achieved by phase-matching the harmonic production process throughout the length of the generating medium, thereby ensuring that the harmonic signals from different parts of the medium interfere constructively. Under these circumstances, the intensity of the qth order harmonic will grow as $I_q \sim L_m^2$ and $I_q \sim N^2$ under phase-matched conditions (where L_m is the medium length and N is the number density of emitters). Phase-matching requires that the propagation vectors satisfy

$$\Delta k = k_q - qk_0, \tag{1}$$

where Δk is the wave vector mismatch, k_q and k_0 the qth harmonic and laser wave vector, respectively. In practice, this condition cannot be met at high photon energies, because generating high order harmonics [2] requires that very high laser intensities be used. This results in a large ionization fraction ($>$0.5) at the peak of the pulse, which leads to a large negative dispersion that cannot be compensated by the positive dispersion of neutral gas [3].

Here we report on a recent experiment performed on the Astra laser at the Rutherford Appleton Laboratories [8] that has demonstrated substantial enhancement ($>10^3$) of the HHG signal—particularly in the water window ($\sim$4 nm). This enhancement is interpreted as being the first observation of a novel scheme for quasi-phase-matching (QPM [9]) of the harmonic signal generated in gas filled capillary waveguides, multimode quasi-phase-matching, in which QPM arises from the strongly modulated intensity profile formed when multiple waveguide modes are excited [10].

QPM [9,11,12] provides a promising alternative to current phase-matching schemes by eliminating the need to achieve $\Delta k = 0$. Instead, QPM relies on periodically suppressing the HHG process in regions, which would contribute destructively to the harmonic signal; i.e., the HHG source term is modulated with a QPM period

$$L_{\mathrm{QPM}} = \frac{2\pi m}{\Delta k}, \tag{2}$$

where m is the order of the process. As a result, the signal can grow substantially above that generated from a single coherence length, albeit more slowly than with true phase-matching. In principle, QPM can be achieved in a variety of ways, e.g., by varying the medium density or by modulating the intensity of the fundamental driving laser. The latter approach is highly promising, since the strong intensity dependence of HHG implies that a relatively small reduction in the intensity of the fundamental results in a substantial reduction of the harmonic signal. Substantial enhancements are possible with periodic variations as low as a few percent [9,11].

One platform for achieving a suitable intensity profile for QPM is via intensity modulations present in a hollow-core capillary with multiple excited modes [10,13]. In the idealized case of only two excited modes, it is easy to see that the resulting on-axis intensity profile in a capillary will display periodic intensity modulations: the two modes, the fundamental mode, $j = 1$, and the jth mode, propagate with different k, vectors k_1 and k_j, respectively, and consequently produce a regular intensity beat pattern of period

$$L_{\mathrm{QPM}} = \frac{2\pi}{k_1 - k_j}. \quad (3)$$

QPM will then occur when $\Delta k = k_1 - k_j = 2\pi/L_{\mathrm{QPM}}$, from Eq. (2), which can be achieved by choosing appropriate values for the peak intensity and the gas density.

In practice it is difficult to achieve excitation of only two waveguide modes owing to the imperfect mode matching at the capillary input plane and mode coupling within the capillary due to ionization [13]. One might assume that the intensity modulation resulting from multiple modes beating in capillary would be detrimental to achieving QPM. However, close inspection of the resulting on-axis intensity profile shows that the spacing of the peaks in the axial intensity are determined by the highest-order significantly contributing mode in the capillary [10]. In simple terms, the main difference between two modes and multiple mode beating is that some peaks are "missing" and hence the overall signal growth is reduced—but still substantial enough to allow significant gains to be made over fairly short medium lengths.

This significantly modulated intensity in the capillary has a major benefit for the quasi-phase-matching scheme—while instantaneously high at points of high intensity, the average ionization, Z^*, is significantly reduced resulting in a reduced Δk, which in turn requires a longer—and crucially, easier to achieve—L_{QPM} to compensate (typically for soft x-ray harmonics $L_{\mathrm{QPM}} < 50\ \mu\mathrm{m}$ in a high ionization limit).

This experiment was performed on the Astra laser at the Rutherford Appleton Laboratory. Astra delivered ~40 fs pulses limited to a maximum of 50 mJ on target for this experiment. An $f = 1$ m focusing lens was used to couple the laser into the capillary entrance (bore radius, a, ~90 μm, up to 15 mm long). The capillary coupling parameter was varied by aperturing the beam from >20 mm to 10 mm diameter. The capillary was filled with argon gas via two laser-machined entrance holes resulting in a constant pressure region of 10 mm length. The harmonic radiation was detected using an ANDOR XUV CCD detector coupled to a flatfield grating spectrometer with an angular acceptance of ~5 mrad × 5 mrad. The conversion efficiency was calculated using the known quantum efficiency of the CCD, the grating efficiency (calibrated on the Daresbury synchrotron), and the transmission function of accurately metrologized filters and mirrors using the known coefficients [14]. The overall uncertainty in the transmission function is estimated to be ~55%.

For a Gaussian beam optimal coupling into the fundamental mode of the capillary is achieved for the vacuum coupling parameter, $\chi = w_0/a$, given by [7,13], where w_0 is the $1/e^2$ radius of the incident intensity profile and a is the capillary bore radius. For $\chi \neq 0.64$, or a non-Gaussian pulse profile, more and more incident energy is excited into higher-order modes. Under typical experimental conditions the transverse profile of the incident beam will more closely resemble an Airy profile, due to the fact that for efficient energy extraction in high power laser

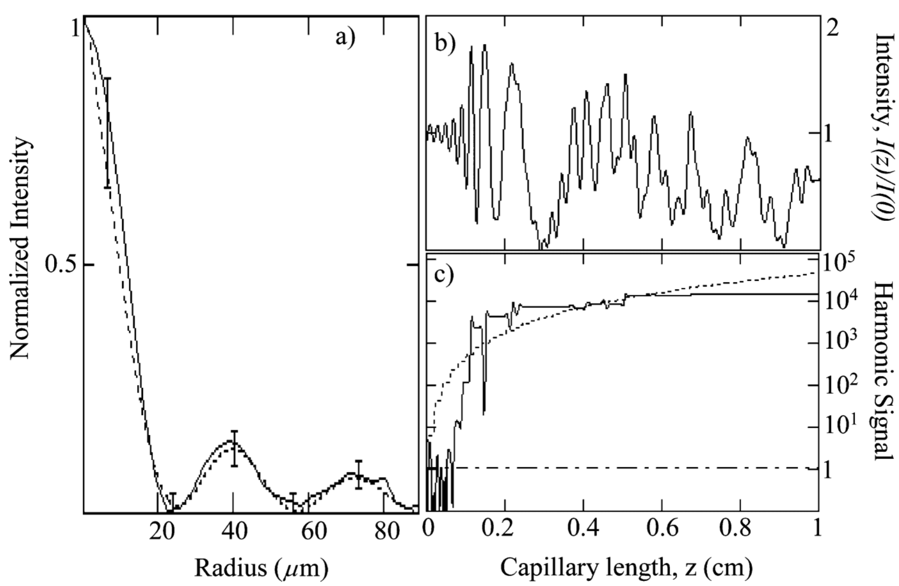

FIG. 1. The experimentally measured incident pulse profile in Fig. 1(a) (solid line) can be approximated by the Airy profile (dotted line) corresponding to vacuum coupling parameter of $\chi = 0.2$ for a capillary with bore radius $a \sim 90\ \mu$m (the error bars on the experimental data are due to radial averaging). The expected on-axis intensity profile, normalized at $z = 0$, for a 1 cm long evacuated capillary (no gas present), assuming the dotted profile in Fig. 1(a), is shown in Fig. 1(b). The resulting harmonic signal growth for $q = 201$ under optimal MMQPM conditions ($p \sim 20$ mbar Ar, $I_{\max} \sim 1 \times 10^{15}\ \mathrm{W\,cm^{-2}}$, solid line) is shown in Fig. 1(c) normalized to that expected for a single coherence length (dashed line). Also shown (dotted line) is the expected HHG signal growth for idealized two mode beating [from Eq. (3)] under optimized MMQPM conditions and $j = 20$ ($L_{\mathrm{QPM}} = 200\ \mu$m).

systems the near-field transverse intensity profile is approximately top-hat pulse.

A lineout of an experimentally measured laser focal spot is shown in Fig. 1(a) (solid line) and the calculated on-axis intensity profile $I(z)$ for a 1 cm long evacuated capillary (no gas present) with $a \sim 90$ μm for coupling, corresponding to the dotted profile in Fig. 1(a), is shown in Fig. 1(b). The rapid fluctuations in the on-axis intensity arise from beating between the fundamental and higher-order capillary modes. The peaks in the intensity profile are spaced by multiples of the beat period, L_B, allowing quasi-phase-matching through the multimode quasi-phase-matching (MMQPM) process [10]. The expected growth of the harmonic signal for $q = 201$, with pressure, $p \sim$ 20 mbar Ar and an incident energy of ~7 mJ, giving peak intensity $I_{\max} \sim 1 \times 10^{15}$ W cm^{-2} in the capillary, is shown in Fig. 1(c).

The signal growth for $q = 201$ from an optimized two mode MMQPM scheme [i.e., pressure and intensity matched to satisfy Eq. (2) and (3) for modes $j = 1$ and $j = 20$ beating with equal intensity and ignoring mode attenuation, giving $L_B = L_{\mathrm{QPM}} = 200$ μm for $a \sim 90$ μm [10]] is represented by the dashed lined in Fig. 1(c). The harmonic order $q = 201$ is generated from Ar ions which, as result of higher ionization potential, has a higher harmonic cutoff than that of neutral Ar [5]. It should be noted that the rapid growth of HHG over the first few mm of the capillary for the intensity profile in Fig. 1(b) compared to that for the two mode MMQPM scheme [Eq. (3)] is due to the constructive interference of multiple modes leading to an increased on-axis intensity above that achieved in the idealized two mode case.

Strong enhancement ($>10^3$) of the HHG signal in the water-window spectral region was observed experimentally for a vacuum coupling parameter of $\chi \sim 0.2$ and $p \sim$ 20 mbar Ar, while for mismatched conditions (42 mbar Ar, $\chi \sim 0.2$) water-window HHG was destroyed (Fig. 2). The maximum harmonic order detected was ~231$^{\mathrm{st}}$ (>360 eV, 3.43 nm second order diffraction). The lower limit of the conversion efficiency (as described in Fig. 2) is estimated to be $>10^{-6}$ per harmonic at 300 eV. This implies a photon flux of $>10^{10}$ per harmonic peak per second (10 Hz system) and a peak brightness of $>10^{21}$ photons/s/mm^2/mrad^2 in 0.1% bandwidth, making this the brightest source of water-window region x rays from HHG observed to date.

One important point of note is that this is the first observation of QPM (albeit imperfect, i.e., peaks missing) in ions with $m = 1$. Under ordinary conditions, exploiting the higher ionization potential of ion species for increased photon energy HHG will result in very large values of Δk due to the high average degree of ionization. Quasi-phase-matching under such conditions requires a very short L_{QPM}, which in practice is difficult to achieve, due high average ionization. In MMQPM the intensity modulations are sufficiently large that Z^* is significantly reduced, increasing L_{QPM} and thereby allowing low-order ($m = 1$) QPM.

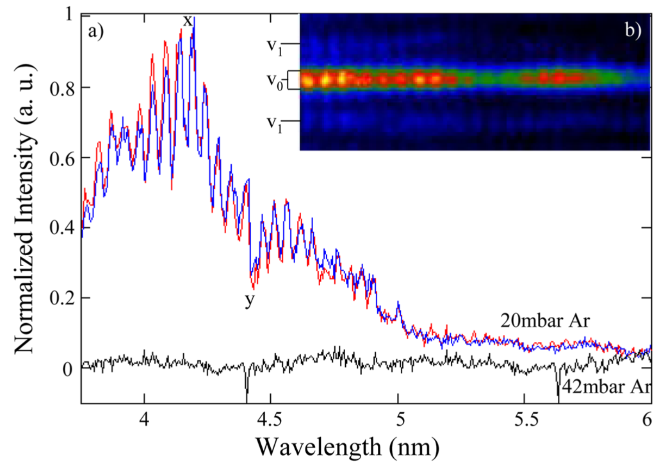

FIG. 2 (color online). Quasi-phase-matched HHG in the water window. The red and blue traces are two separate shots recorded at $p \sim 20$ mbar Ar with the signal corrected for system transmission, while the black trace is obtained for mismatched conditions with $p \sim 42$ mbar Ar. Both harmonic signal traces (red and blue) are normalized to the value of the peak of the blue trace (labeled x) showing the good short term reproducibility of the data (3-minute interval). The dip in harmonic intensity at ~4.4 nm (labeled y) is due to uncertainty in the absolute transmission around the C K edge of the 0.2 μm Al/0.3 μm CH filter used to block optical light. The raw data from the CCD are shown in the inset. A lower limit of the conversion efficiency per harmonic order is calculated by vertical integration of the background subtracted signal in the bright region over the interval "v_0" only and correcting for system transmission. Significant signal with clear harmonic structure is present for one side lobe (marked v_1 in the inset) either side of the bright central order, v_0. Up to six side lobes were observed experimentally and are likely due to off axis QPM.

MMQPM at longer wavelengths was investigated for a vacuum coupling parameter of $\chi \sim 0.3$, with strong enhancement observed for $q = 25$ with an input laser energy of ~6 mJ giving $I_{\max} \sim 9 \times 10^{14}$ W cm^{-2} and $p \sim$ 54 mbar Ar. The HHG spectrum obtained under these conditions is shown in Fig. 3(a). The peak conversion efficiency per harmonic was similar to the water-window case at $>10^{-6}$ at ~32 nm (25th order).

Allowing for filter transmission the full width half maximum (FWHM) of the quasi-phase-matched harmonic comb can be estimated as ~20 harmonic orders for the water-window harmonics and 2–3 orders at the 25th harmonic. This narrow band enhancement is characteristic of successful phase-matching, since the phase mismatch Δk depends inversely on the harmonic order q. As a result, the fractional bandwidth of the enhanced harmonic spectrum ($\Delta\omega_{\mathrm{FWHM}}$) is very similar for the two cases studied here ($\Delta\omega_{\mathrm{FWHM}}/\omega_{191} \sim \Delta\omega_{\mathrm{FWHM}}/\omega_{25} \sim 0.1$, where ω_q is the angular frequency of the qth harmonic order). Intensity tuning of the peak harmonic was observed when $I_{\max}$ was varied (and thus Δk) for otherwise constant conditions. In

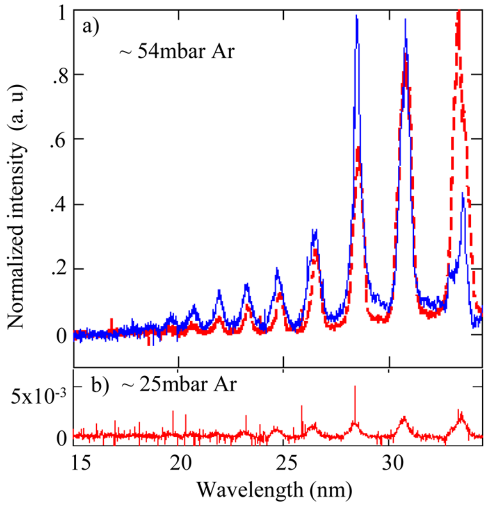

FIG. 3 (color online). Multimode quasi-phase-matching at ~30 nm wavelength. (a) Under matched conditions an enhancement of >200 is observed for 2–3 harmonic orders. Varying Δk by a small change in intensity allows the position of the brightest peak to be tuned from the 25th to the 29th harmonic. $I_{\max} = 9 \times 10^{14}\ \mathrm{W\,cm^{-2}}$ (red) and 7×10^{14} (blue). Mismatched conditions can be seen in (b) which shows the typical "plateau" structure of almost equal intensities typical of HHG in the absence of phase-matching [1].

Fig. 3 the brightest harmonic is shifted from the 25th to the 29th harmonic by varying the incident energy from ~6 mJ ($I_{\max} \sim 9 \times 10^{14}\ \mathrm{W\,cm^2}$, red dashed trace) to ~5 mJ ($I_{\max} \sim 7 \times 10^{14}\ \mathrm{W\,cm^2}$, blue trace). Reducing the laser intensity causes lower ionization and hence requires a higher harmonic order to satisfy the QPM matching condition.

Figure 4 shows the calculated enhancement in the harmonic signal as a function of Ar pressure for $q = 201$ and $q = 25$ for the conditions of Figs. 2 and 3, respectively. Also shown are the measured harmonic signals normalized to the peak of the calculated curve. The calculated pressure dependence is in good agreement with the experimental data which lends support to our interpretation that the observed high efficiency of HHG arises from MMQPM. The small discrepancy between experiment and theory at high pressures is due likely to the fact that the presence of gas in the capillary causes the repartition of energy into higher modes [13] resulting in increased high frequency modulation over longer interaction lengths. This will result in a better MMPQM profile and a correspondingly higher signal even for poorer matching conditions. Importantly, the fact that the pressure tuning curves are not narrow spikes at an optimum pressure for peak enhancement indicates that even random intensity profiles can see enhancement; i.e., an optimal MMQPM intensity profile for a given value of Δk will essentially be random for $\Delta k + \varepsilon$, where ε is a small change in the value of Δk.

In conclusion, a substantial enhancement ($>10^3$) of HHG, which is interpreted as being due to multimode

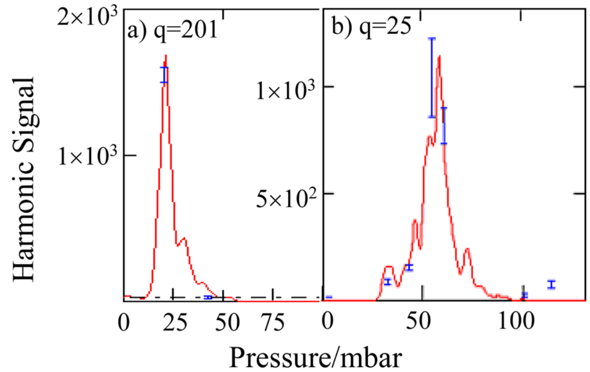

FIG. 4 (color online). Pressure tuning curves (solid lines) calculated for (a) $I_{\max} \sim 1 \times 10^{15}\ \mathrm{W\,cm^{-2}}$ ($q = 201$, $\chi \sim 0.2$) and (b) $I_{\max} \sim 9 \times 10^{14}\ \mathrm{W\,cm^{-2}}$ ($q = 25$, $\chi \sim 0.3$), showing expected harmonic signal enhancements over the signal expected for a single coherence length. The experimental data (points) is normalized to the peak of the pressure tuning curves. The vertical extent of the data points indicates the experimentally observed shot to shot variation in the intensity of the stated harmonic order.

quasi-phase-matching—MMQPM, over a narrow range of harmonic orders in argon has been observed. In principle, this approach should allow the selective enhancement of harmonics across the full spectral range. Modeling suggests that substantial improvements (>10) on the current results can be achieved by optimizing the coupling into the waveguide so as to predominantly excite only two modes.

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