Bright quasi-phasematched soft X-ray harmonic radiation from Argon ions

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Introduction

High Harmonic Generation (HHG) using ultra-fast laser sources is a very attractive route to generating coherent beams of extreme ultra-violet (XUV) radiation in the 20-400eV spectral region. The key advantages of the HHG approach are the high beam quality, ultra-short pulses ranging from femtoseconds (fs) to attoseconds and the comparative ease with which compact, commercially available femtosecond lasers can be converted to the XUV - the most basic HHG set-up simply involves focusing a fs-laser into a gaseous medium. Here we report on a recent experiment performed on Astra that has demonstrated substantial enhancement of the HHG signal – particularly in the water-window (~4nm).

The most significant barrier to widespread application of HHG is the fairly low conversion efficiency and therefore the pulse energies that can be achieved. While reasonable conversion efficiencies in the range of $E_{harmonic}/E_{Laser} = 10^{-5} – 10^{-6}$ have been achieved at photon energies <40 eV[1], the reported conversion efficiency at wavelengths >200eV has been much lower (10^{-11})[2]. In close analogy to the generation of optical laser harmonics in non-linear crystals, the highest efficiency can only be achieved by phasematching the harmonic production process throughout the length of the generating medium, thereby ensuring that the harmonic signal from different parts of the medium interferes constructively. Consequently, the intensity of the q-th order harmonic will grow as $I_q \sim L^2$ and $I_q \sim N^2$ under phasematched conditions (L: Medium length, N: density). Phasematching requires that the propagation vectors meet the condition:

$$\Delta k = k_q - qk_0 = 0, \qquad (1)$$

where Δk is the phase mismatch, k_q and k_0 the q-th harmonic and laser wavevector respectively. In practice this condition is hard to meet at high photon energies, because the underlying mechanism[3] requires that very high laser intensities are used. This results in a large ionization fraction (e.g. >0.5) at the peak of the pulse and consequently a refractive index that is dominated by the free electron contribution.

Quasi-phasematching (QPM)[4] provides a promising alternative to current phasematching 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 period $L_{QPM}=2\pi/\Delta k$. As a result the signal grows somewhat more slowly than in an ideally phasematched scenario, but still results in very substantial gains over an un-matched process. 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. 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[4].

Experimental

The strong enhancement observed in the Astra experiment is thought to be likely due to the intensity modulations present in a hollow-core capillary with multiple excited modes. In the case of only two excited modes, it is easy to see that the resulting on-axis intensity profile in the capillary will display periodic intensity modulations: the two modes are propagating with different k-vectors k_1,k_n and consequently produce a regular beat pattern with periodicity $L_{QPM}=2\pi/(k_1-k_n)$ (Fig 1). QPM will then occur when $\Delta k=2\pi/L_{QPM}$ (2), which can be achieved by choosing appropriate values for the peak intensity and the gas density.

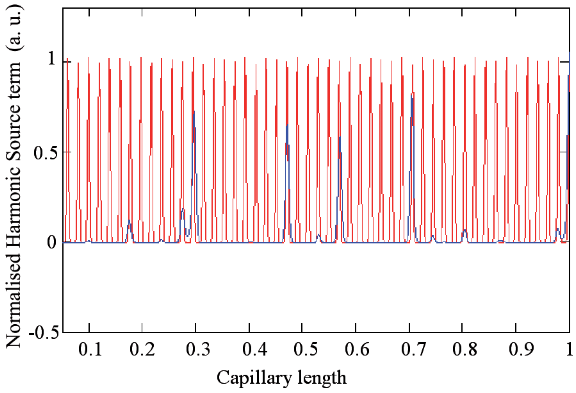

Figure 1. Capillary mode beating pattern for two modes 1 and 6 (red) and modes 1 through 6 (blue). Note that the peaks for 1-6 occur only at positions also occupied by the pattern generated by modes 1 and 6 only. The result of the multimode compared to two-mode beating is therefore simply to slow signal growth, but not to prevent it.

While a situation with only two excited modes is ideal for QPM, it is, in practice, difficult to achieve due to the imperfect mode matching at the capillary input plane and mode coupling due to ionization[5]. At first glance, one might assume that the intensity modulation resulting from multiple modes beating in capillary would be detrimental to achieving QPM. Close inspection of the resulting on-axis intensity profile shows that the periodicity of the intensity peaks are determined by the highest order excited mode in the capillary (fig1). In simple terms, the main difference between two modes and multiple mode beating is that some peaks are 'missing' and hence the signal growth is reduced – but still substantial enough to allow significant gains to be made over fairly short medium lengths.

The highest order mode that is excited can be controlled by varying the coupling parameter $\chi=w_0/a$ (w_0: beam waist, a: capillary radius) and the gas pressure (due to ionization defocus[5]). The number of excited modes increases as the coupling parameter is decreased or the gas pressure increased. The coupling parameter was varied by aperturing the beam to 10-20mm diameter, delivering a maximum energy of up to 50mJ on target in ~50fs duration pulse. An f=1m focusing lens was used to couple the laser into the capillary entrance (~90µm bore radius, 15mm long). The capillary was filled with Argon gas via two laser-machined entrance holes resulting in a constant pressure region of 10mm length. The harmonic radiation was detected using an ANDOR XUV CCD detector coupled to a flatfield grating spectrometer with a ~5mrad×5mrad angular acceptance.

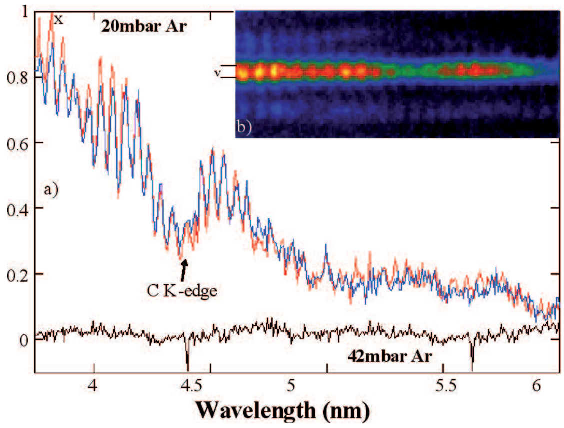

Figure 2. Quasi-phasematched HHG in the water-window. The red and blue trace are two separate shots recorded at p=20 mbar Ar, the black trace is for mismatched conditions with p=42mbar Ar. The recorded spectrum is shown in the inset. The conversion efficiency per harmonic order is calculated from the bright region marked 'v' only .

Phasematching in the water-window was observed for a vacuum coupling parameter of $\chi\sim0.2$ and 20 mbar Ar and $I\sim1\times10^{15}Wcm^{-2}$ peak intensity. Figure 2 compares the signals obtained for phasematched conditions (20mbar Ar, red and blue traces) and mismatched conditions (42mbar Ar, black trace). The dip in harmonic intensity around 4.4 nm is attributed to the transmission curve of the 0.1µmAl/0.2µmCH filter used to block the laser light. The maximum harmonic order detected was ~230th (>360eV, 3.43nm second order diffraction). The short-term reproducibility of the data is also shown in Figure 3 where the red and blue curves correspond to data taken within a 3-minute interval. No signal was observed in the water-window, when the pressure was changed to mismatched conditions. The lower limit of the conversion efficiency is estimated to be $>10^{-6}$ per harmonic at 300eV after correcting for spectrometer response and including only the central, bright signal (figure 2).

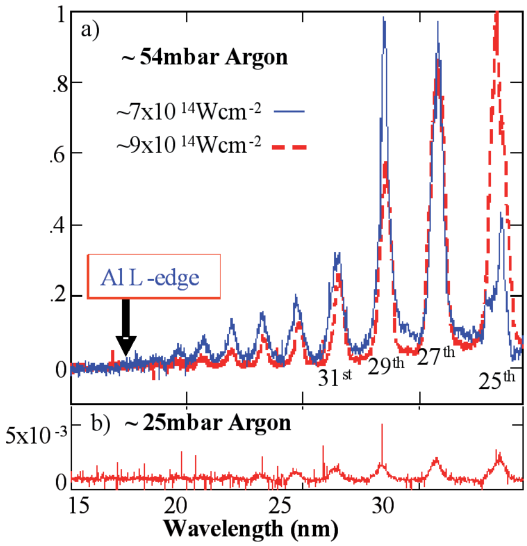

Figure 3. Quasi-phasematching at ~30nm wavelength. a) Under matched conditions an enhancement of >200 is observed for 2-3 harmonic orders. Varying Δk by a small change in intensity allows the position of the brightest peak to be tuned from the 25th to the 29th harmonic. $I=9\times10^{14}$ Wcm^{-2} (red) and 7×10^{14} (blue). Mismatched conditions can be seen in b) exhibiting the 'plateau' structure of almost equal intensities typical of HHG in the absence of phasematching.

Phasematching at longer wavelengths was investigated with a vacuum coupling parameter of $\chi\sim0.3$. Phasematching was observed for q=25 with a laser intensity of $I\sim9\times10^{14}Wcm^{-2}$ and 54mbar Ar. The HHG spectrum obtained under these conditions is shown in Figure 3a. The peak conversion efficiency per harmonic was similar to the water-window case at $>10^{-6}$ at ~32nm (25th order).

Correcting for filter transmission the FWHM of the phasematched 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 phasematching, since the phase mismatch Δk depends linearly on the harmonic order q. As a result, the fractional bandwidth of the enhanced harmonic spectrum is very similar for the two cases studied here ($E_{FWHM}/E_{190}\sim E_{FWHM}/E_{25}\sim0.1$). Tuning of the peak harmonic was observed when the intensity was varied (and thus Δk) for otherwise constant conditions. In figure 3 the brightest harmonic is shifted from the 25th to the 29th harmonic by varying the incident intensity from $\sim9\times10^{14}Wcm^2$ to $\sim7\times10^{14}Wcm^2$. This results in lower ionization and hence requires a higher harmonic satisfy the matching condition (1).

In conclusion, substantial, enhancement of harmonics due to phasematching has been observed over a narrow range of harmonic orders. This approach has yielded the highest conversion efficiencies in the water-window to date with an enhancement over previous results of several orders of magnitude.

References

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