Soft X-ray wavelength shift induced by ionization effects in a capillary

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Coherent soft X-rays are produced by high harmonic generation in a capillary filled with argon gas. We demonstrate that the tuning of the harmonic wavelengths with intensity and chirp arises from changes in the argon ionization level. Control over the tuning can be achieved either by changing the average intensity of the laser pulse or by varying the quadratic and cubic spectral phase of the laser pulse. We observe an ionization dependant blue shift of the fundamental wavelength that is directly imprinted on the harmonic wavelengths. The harmonic tuning is shown to depend on nonlinear shifts of the frequency of the fundamental laser pulse due to the plasma created by ionization, rather than directly on any chirp imposed on the fundamental.

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High harmonic generation (HHG) using high-intensity ultrafast laser pulses can be used to produce coherent beams of soft X-rays, opening up new areas of science in both the X-ray and attosecond regimes. However a complete understanding of the harmonic generation process has still not been achieved. The spectrum of X-rays produced in an HHG experiment depends on many parameters, such as the peak laser intensity, the gas absorption, the propagating geometry, phase matching and the gas ionization level. In a capillary guide, phase matching can be affected by variation of the gas pressure within the capillary, choice of the capillary radius and peak laser intensity.[1] Control of the phase profile of the laser pulse[2,3,8] can change the X-ray spectrum considerably. It was shown that the X-ray spectrum can be shifted, relative amplitudes of harmonics can be altered and particular harmonics optimised. Enhancement of the harmonic generation has also been demonstrated through control of the spatial profile of the generating laser pulse.[4] In addition, in gas jets, ionization induced blue shift effects were observed and used to tune the harmonic wavelengths.[5,6]

In this work we show that the major effect on the tuning of the harmonic frequencies is a nonlinear frequency shift of the fundamental laser pulse as it propagates along the capillary and creates a plasma by argon ionization. Changes in the ionization level are achieved either by changing the average intensity of the laser pulse or by stretching the pulse by changing the quadratic and cubic spectral phase of the laser pulse in a precise and measureable way. The laser pulse is characterised before and after propagating through the capillary using second harmonic generation frequency resolved optical gating (SHG-FROG). After propagation through the capillary, we observe a blue shift of the fundamental laser wavelength that is directly imprinted on the measured high harmonics. The amount of blue shift depends on the degree of ionization caused by the laser pulse, and so measured shifts in the harmonics are related to both pulse length and peak intensity.

The laser system is a 1 kHz Ti:sapphire chirped pulse amplifier system producing 1 mJ, 30 fs pulses. An acousto-optic programmable dispersive filter, Dazzler from Fastlite,[7] is positioned before the stretcher to vary the quadratic and cubic spectral phase of the pulse. The laser average power is varied using a half wave plate and polariser after the amplifier. The amplified pulses are focused into a hollow glass capillary of 150 μm internal diameter and 7 cm length with two 300 μm holes 2 cm from the end. The Ar gas from a presure controlled supply flows through the two holes providing a central region of constant pressure. The fundamental light leaving the capillary is blocked by a 200 nm-thick Al filter. The generated X-rays are dispersed using an X-ray spectrometer and detected with a multi-channel plate and CCD. A mirror can be inserted into the beam between the capillary and the Al filter in order to send the fundamental laser pulse, after propagation through the capillary, to the FROG for characterisation. This mirror is also used for the alignment of the laser into the fundamental mode of the capillary. Coupling into the EH_{11} mode is achieved by careful alignment and focusing, resulting in about 70% transmission of the incident beam, with no need for an aperture in the laser beam[1] or a spatial mode profiler.[4]

Harmonics up to the 29th order are seen for input laser pulse energies of 0.6 mJ. The pressure in the capillary is controlled, and the pulse shape is precisely var-

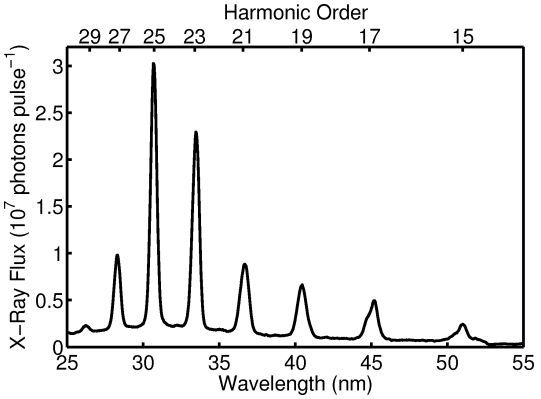

Fig. 1. X-ray spectrum for Ar pressure of 80 mbar, input laser pulse of 40 fs, 0.6 mJ.

ied using the Dazzler. Changing the Ar pressure varies the harmonic output, as has been previously reported,[1] and simple models of phase matching can reproduce the observed variations. Figure 1 shows the output X-ray spectrum for an Ar pressure of 80 mbar where the high energy (short wavelength) harmonics are phase matched. The throughput of the X-ray spectrometer and detector was calibrated by comparison with the total X-ray signal from a calibrated X-ray photodiode after the Al filter. The total conversion efficiency is of the order of 10^{-6}, comparable to the efficiency in the gas jet geometry.[6]

The wavelength of each harmonic can be tuned by changing the quadratic spectral phase and/or the laser intensity. A set of harmonic spectra, displayed as an intensity map for a range of quadratic phase values, is shown in figure 2, indicated by the equivalent group delay dispersion that would produce the actual quadratic spectral phase introduced by the Dazzler. Fig. 2 (a) shows the spectra for constant pulse energy of 0.6 mJ. Fig. 2 (b) shows the spectra for constant peak intensity. In order to maintain constant peak intensity of 10^{14} W/cm^{-2}, pulse energy was adjusted in the range 0.3–0.6 mJ. Enhancement of either the short or long wavelength region of the spectra can be achieved. There is a symmetric wavelength shift as the second order dispersion is changed around the zero dispersion. For the constant pulse energy the zero second order dispersion spectrum shows the harmonics blue shifted relative to the higher dispersion spectra, both for positive and negative dispersion. However for the constant peak power the higher dispersion value spectra are blue shifted, relative to the zero dispersion one. Figure 3 shows the harmonic wavelength shift as a function of second order dispersion, fig. 3 (a), and as a function of average laser power, fig. 3 (b). A shift amounting to almost 50% of the adjacent harmonic spacing is obtained. Similar behaviour is also observed when varying third order dispersion.

The characterisation of the post capillary laser pulse helps to understand the blue shift of the harmonics. Figure 4 shows the spectra of the laser pulse after the capillary as a function of second order dispersion. There is

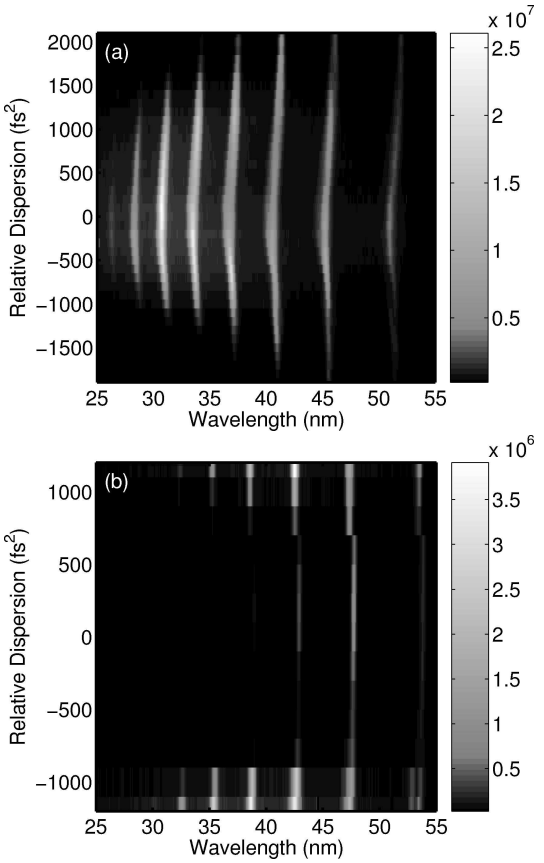

Fig. 2. Spectral intensity map for constant pulse energy of 0.6 mJ (a) and constant peak intensity (10^{14} W/cm^{-2}) at the capillary (b), Ar pressure of 60 mbar.

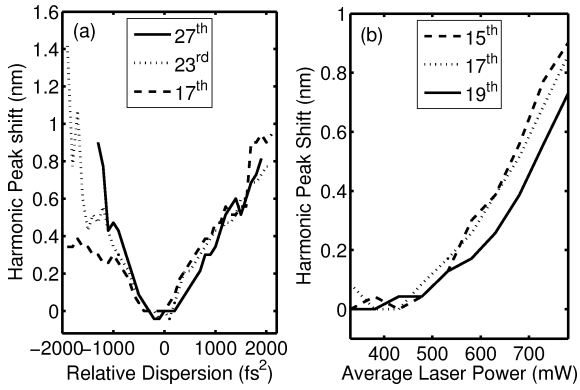

Fig. 3. Wavelength shift as a function of the second order dispersion (a) and average laser power (b).

a clear increase in bandwidth and a blue shift of the pulse spectrum for zero dispersion, at the highest pulse energy. This shift is reproduced on the X-ray harmonic spectra. Calculated harmonics considering the measured laser centre wavelength are compared with the measured harmonic wavelength position in figure 5. There is a very good agreement for the calculated and measured wavelength positions for all of the harmonic orders.

For the range of pulse lengths and energies used in

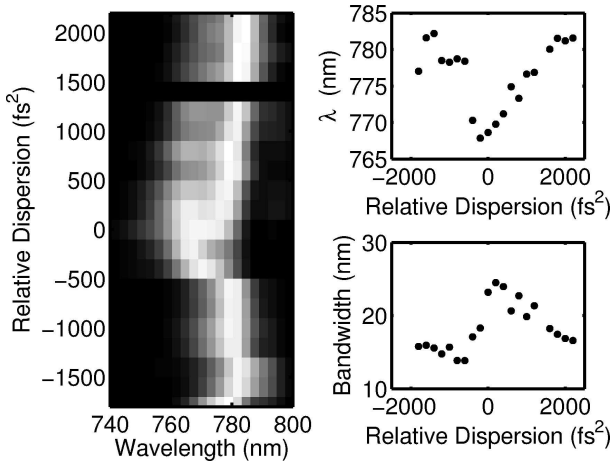

Fig. 4. Spectral intensity map for the laser pulse after the capillary. The centre wavelength, λ, and bandwidth are shown at the top and bottom right graphs, respectively.

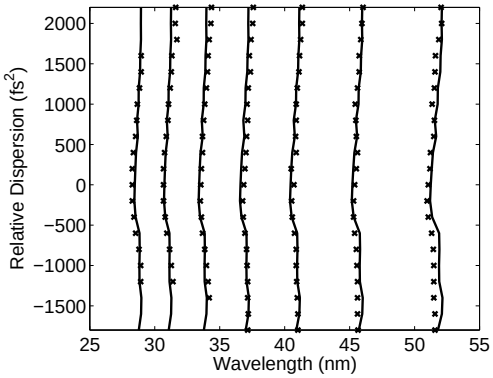

Fig. 5. Measured (crosses) and calculated (solid line) harmonic wavelength peak position as a function of second order dispersion. The measured peak positions are from the data in fig. 2 (a).

this experiment, the degree of ionization on-axis is calculated, using Keldysh theory,[10] to vary from 0 to 100%. Hence both the plasma nonlinearity and phase-matching due to the plasma index must be taken into account. The nonlinearity of the plasma has been previously observed to blue-shift and broaden the fundamental laser pulses,[9] and this effect will be largest at the highest ionization levels. For constant pulse energy, maximum ionization occurs at minimum chirp, producing the observed blue shift of harmonics for minimum chirp. The sign of the chirp does not affect the measured shifts, as the effects here are caused by intensity variations, in contrast to previously-observed effects at low ionization levels.[8] Variation of the pulse length at constant peak intensity requires an increase of pulse energy with width, which in turn produces more total ionization as the chirp is increased, resulting in a blue shift of harmonics as the chirp is increased. In this case, the harmonic intensities are also increased because the overall ionization level starts off below that necessary for phase matching ($\sim$4%), and reaches that required for phase matching as the pulse energy is increased. We note that as the absorption length of argon gas for the harmonics is a few millimeters, all the observed X-rays will be generated in the last few millimeters of the gas-filled region of the capillary. Thus the first few centimeters of the gas-filled region act as a pulse shaper, and the result of the pulse shaping is reflected in the harmonic spectrum generated.

In summary, we have observed a blue shift of the fundamental and harmonic wavelengths that varies with the ionization level of the Ar in the capillary. This was achieved either by changing the average laser intensity or the spectral phase of the laser pulse using the Dazzler. The characterisation of the laser pulse before and after propagating through the capillary permitted the observation of a blue shift of the fundamental laser wavelength that is directly imprinted on the high harmonics. The amount of blue shift is dependent on the argon ionization level in the capillary. The intensities of particular harmonics are enhanced by variations of the ionization levels which produce phase matching. The dependence of harmonic frequency on ionization level in this way provides improved insight into the generation process in capillaries, and provides the ability to modify the X-ray spectrum to produce frequency-agile X-ray tunable sources.

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References

1. A. Rundquist, C. G. D. III, Z. Chang, C. Herne, S. Backus, M. M. Murnane, and H. C. Kapteyn, Science **280**(5368), 1412–1415 (1998).
2. R. Bartels, S. Backus, E. Zeek, L. Misoguti, G. Vdovin, I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Nature **406**(3), 164–166 (2000).
3. T. Pfeifer, D. Walter, C. Winterfeldt, C. Spielmann, and G. Gerber, Applied Physics B **80**(3), 277 – 280 (2005).
4. T. Pfeifer, R. Kemmer, R. Spitzenpfeil, D. Walter, C. Winterfeldt, G. Gerber, and C. Spielmann, Optics Letters **30**(12), 1497–1499 (2005).
5. C. Altucci, R. Bruzzese, C. de Lisio, M. Nisoli, S. Stagira, S. D. Silvestri, O. Svelto, A. Boscolo, P. Ceccherini, L. Poletto, G. Tondello, and P. Villoresi, Physical Review A **61**(2), 021801 (pages 4) (2000).
6. D. H. Reitze, S. Kazamias, F. Weihe, G. Mullot, D. Douillet, F. Aug, O. Albert, V. Ramanathan, J. P. Chambaret, D. Hulin, and P. Balcou, Optics Letters **29**(1), 86–88 (2004).
7. F. Verluise, V. Laude, Z. Cheng, C. Spielmann, and P. Tournois, Optics Letters **25**(8), 575–577 (2000).
8. W. Kornelis, C. P. Hauri, A. Heinrich, F. W. Helbing, M. P. Anscombe, P. Schlup, J. W. G. Tisch, J. Biegert, and Keller, Optics Letters **30**(13), 1731–1733 (2005).
9. N. L. Wagner, E. A. Gibson, T. Popmintchev, I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Physical Review Letters **93**(17), 173902 (pages 4) (2004).
10. V. S. Popov, Physics–Uspekhi **47**(9), 855–885 (2004).