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 Ar gas. We demonstrate that the tuning of the harmonic wavelengths with intensity and chirp arises from changes in the Ar ionization level. Control over the tuning can be achieved either by changing the average intensity of the laser pulse or by varying the quadratic spectral phase of the laser pulse. We observe an ionization-dependent blueshift of the fundamental wavelength that is directly imprinted on the harmonic wavelengths. The harmonic tuning is shown to depend on nonlinear spectral shifts of the fundamental laser pulse that are due to the plasma created by ionization, rather than directly on any chirp imposed on the fundamental wavelength.

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High-harmonic generation 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 the attosecond regimes. However, complete understanding of the harmonic generation process has still not been achieved. The spectrum of x rays produced in a high-harmonic generation experiment depends on many parameters, such as peak laser intensity, gas absorption, propagating geometry, phase matching, and gas ionization level. In a capillary guide, phase matching can be affected by variation of the gas pressure within the capillary, the choice of the capillary radius, and the peak laser intensity.[1,2] Control of the phase profile of the laser pulse[3–5] can change the x-ray spectrum considerably. It has been shown that the x-ray spectrum can be shifted, relative amplitudes of harmonics can be altered, and particular harmonics optimized. Enhancement of harmonic generation has also been demonstrated through control of the spatial profile of the generating laser pulse.[6] In gas jets, ionization-induced blueshift effects have been observed and used to tune the harmonic wavelengths.[7,8] In capillaries, ionization has been shown to produce several important effects, including self-focusing and quasi-phase matching, that can be beneficial in extending the wavelength region accessible for harmonic generation.[9,10]

In this work we directly measure the nonlinear frequency shift of the fundamental laser pulse as it propagates through the capillary and correlate it to the measured tuning of the harmonic frequencies. Changes in the ionization level are achieved either by changing the average intensity of the laser pulse or by stretching the pulse by changing its quadratic spectral phase in a precise and measurable way. Before and after propagating through the capillary, the laser pulse is characterized by use of a second-harmonic generation frequency-resolved optical gating (FROG). After propagation through the capillary, we observe a blueshift of the fundamental laser wavelength that is directly imprinted on the measured high harmonics. The amount of blueshift 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[11] (AOPDF) is positioned before the stretcher to vary the spectral phase of the pulse. The laser average power is varied by using a half-wave plate and polarizer after the amplifier. The amplified pulses are focused into a hollow glass capillary of 150 μm internal diameter and 7 cm length with 300 μm holes 2 cm from each end. The Ar gas from a pressure-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 by using an x-ray spectrometer and are detected with a multichannel plate and CCD. The entire x-ray beamline from capillary to spectrometer is placed within a vacuum system with base pressure 10^{-6} hPa. 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 characterization. This mirror is also used to aid 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.[6]

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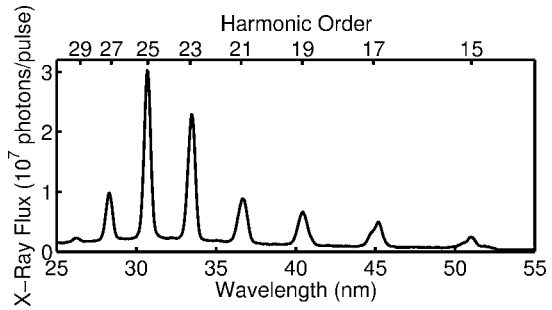

Fig. 1. X-ray spectrum for Ar pressure of 80 hPa, input laser pulse of 40 fs, 0.6 mJ.

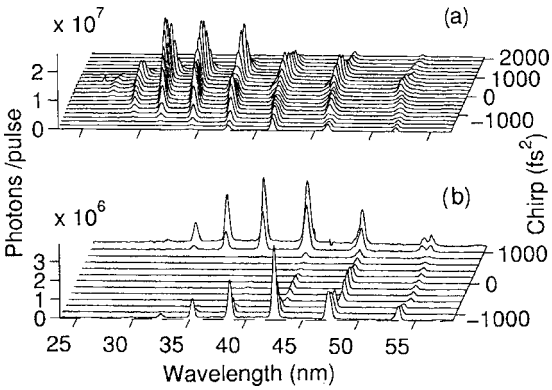

Fig. 2. X-ray spectra as a function of chirp: (a) constant laser pulse energy of 0.6 mJ; (b) constant peak laser intensity (1.15×10^{14} W/cm^2). Ar pressure of 60 hPa.

Harmonics up to the twenty-ninth 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 varied by using the AOPDF. 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 hPa, 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 with the efficiency in the gas jet geometry.[8]

The wavelength of each harmonic can be tuned by changing the quadratic spectral phase or the laser intensity. Figure 2 shows the variation of the x-ray spectra as a function of the quadratic spectral phase (chirp). The numerical values indicate the equivalent group delay dispersion that would produce the actual chirp introduced by the AOPDF. Figure 2(a) shows the spectral variation at a constant pulse energy of 0.6 mJ. The increase in pulse length with chirp produces a reduction in peak intensity from 2.9×10^{14} W/cm^2 to 1.4×10^{14} W/cm^2. Figure 2(b) shows the spectral variation at constant peak intensity. In order to maintain a constant peak intensity of 1.15×10^{14} W/cm^2, the pulse energy was adjusted in the range 0.3–0.6 mJ. A shift amounting to almost 50% of the adjacent harmonic spacing is obtained. In both Figs. 2(a) and 2(b) the wavelength shift is almost symmetric as the laser chirp is changed around zero. However, the direction of shift with increasing chirp in the two cases is opposite. At constant pulse energy the spectrum at zero chirp shows the harmonics blueshifted relative to those produced by a large input chirp, for both positive and negative chirps. Conversely, at constant peak intensity the harmonics produced by unchirped laser pulses are redshifted relative to those produced at large chirps.

Figure 3 plots the harmonic wavelength shift as a function of laser pulse chirp at constant pulse energy, Fig. 3(a), and as a function of average laser power in the absence of any chirp, Fig. 3(b), for comparison. Stretching the constant energy pulse by chirping decreases the degree of Ar ionization, but increasing the average power increases the degree of ionization. Hence they have opposite effects on the wavelength shift of the fundamental and also the harmonics, as shown in Fig. 3. Stretching the pulse by adding cubic spectral phase variation produces a result similar to Fig. 3(a).

The characterization of the precapillary and postcapillary laser pulse helps to explain the blueshift of the harmonics. Figure 4 shows the spectra of the laser pulse as a function of chirp, Figs. 4 a precapillary and 4 b postcapillary. The precapillary spectrum is at the same center wavelength and bandwidth for all

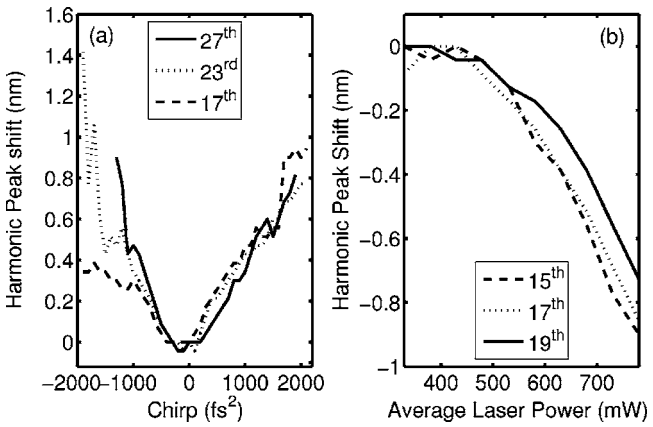

Fig. 3. Harmonic wavelength shift as a function of (a) chirp and (b) average laser power.

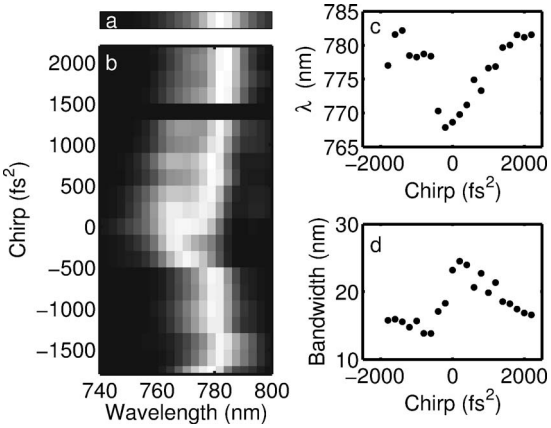

Fig. 4. a, Precapillary laser pulse spectrum; b, postcapillary laser pulse spectra; c, center wavelength λ; d, bandwidth as a function of chirp.

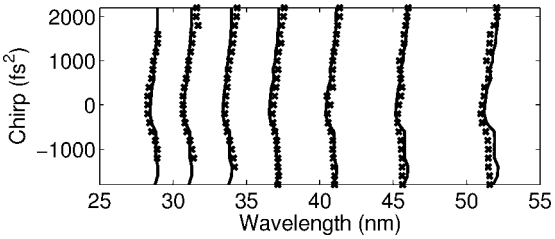

Fig. 5. Measured (crosses) and calculated (solid curve) harmonic wavelength peak position as a function of chirp.

chirps. However, there is a clear blueshift of the post-capillary pulse spectra and an increase in bandwidth for zero chirp at the highest pulse energy. This shift is reproduced on the x-ray harmonic spectra. The calculated harmonics, considering the measured laser center wavelength, are compared with the measured harmonic wavelength position in Fig. 5. There is a very good agreement for the calculated and measured wavelength positions for all harmonic orders.

For the range of pulse lengths and energies used in this experiment, the degree of ionization on axis is calculated, by using the Keldysh theory,[12] to vary from 0 to 100%. Hence both the plasma nonlinearity and the phase-matching due to the plasma index must be taken into account. The nonlinearity of the plasma has been previously observed to blueshift and broaden the fundamental laser pulses,[13] and this effect will be largest at the highest ionization levels. For constant pulse energy, maximum ionization occurs at minimum chirp, producing the observed blueshift 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.[5] 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 blueshift of harmonics as the chirp is increased. Higher harmonics are seen with higher x-ray intensities as the pulse length and energy are increased, because the increase in overall ionization level improves phase matching for higher harmonics. We note that, as the absorption length of Ar 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 blueshift 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 by using an AOPDF. The characterization of the laser pulse before and after it propagated through the capillary permitted the observation of a blueshift of the fundamental laser wavelength that is directly imprinted on the high harmonics. The amount of blueshift is dependent on the Ar ionization level in the capillary. The intensities of particular harmonics are enhanced by variations of the ionization levels that produce phase matching. The dependence of the harmonic frequency on the 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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