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Spatio-temporal coupling to create sub-femtosecond pulses

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Abstract

We use the confluence of two intensity-dependent processes—low-order harmonic generation and self-phase modulation—to spatio-temporally couple and compress few-cycle pulses to be sub-cycle in duration. These half-cycle transient fields self-focus in space and in time (increasing the peak field strength) and can generate isolated attosecond pulses. We explore some of the parameter spaces available to create these half-cycle transients and methods of improving the half-cycle contrast. We also discuss how these pulses can increase high harmonic generation efficiency in the water window.

Keywords: ultrafast nonlinear optics, space-time coupling, nonlinear pulse compression, high harmonic generation

(Some figures may appear in colour only in the online journal)

1. Introduction

In order to measure the dynamics of ultrafast processes, we must first create a ‘camera’ with an ultrafast shutter. In our research, we use femtosecond ($1\text{ fs} = 10^{-15}\text{ s}$) and attosecond ($1\text{ as} = 10^{-18}\text{ s}$) electromagnetic pulses to act as a camera shutter. These electromagnetic pulses transiently change the state of matter in their interaction. The goal of attoscience is to measure and understand attosecond timescale processes, which are typically electronic in nature [1, 2]. Experiments with attosecond resolution can explore all forms of matter where we study the electron dynamics [3, 4].

However, these ultrashort pulses require phase control over a broad bandwidth. The creation of coherent supercontinuum spectra has been an area of active research for decades. Although supercontinuum spectra are available directly out of an oscillator [5, 6], strong field physics and attosecond science typically require laser amplifier stages, which limit the spectral bandwidth due to gain narrowing. To overcome this gain narrowing, we use a hollow-core fibre filled with a noble gas. The fibre guides the field, improving the beam quality while increasing the interaction length of the field with the gas. The high intensity of the many-cycle field

interacts with argon gas to increase its bandwidth to support a few-cycle pulse through self-phase modulation [7, 8]. We compress the pulse down to nearly its transform limit through anomalously dispersive optics [9, 10].

This hollow-core fibre technology has advanced such that it has become routine to create two-cycle pulses in the lab. We have used these high-power ultrashort pulses for isolated attosecond pulse generation in the gas phase through various techniques, such as polarization gating [11, 12] and the attosecond lighthouse [10, 13, 14]. The highly nonlinear response of attosecond pulse generation is field-dependent [15–17]. The phase of the few-cycle driving field—called the carrier envelope phase (CEP)—strongly affects the peak field strength, where this peak field dictates many properties of the attosecond pulse such as amplitude, pulse duration, and maximum photon energy [18]. Thus, we must ensure that the CEP is stabilized and that it is maintained when we spectrally broaden and compress the ultrashort pulse.

We seek to push beyond the few-cycle regime to produce single-cycle and even sub-cycle pulses. These shorter pulses can more efficiently generate isolated attosecond pulses [19], increase the maximum photon energy in high harmonic generation [20], and cause less damage in condensed matter.

Figure 1. Low order harmonic generation in quartz. (a) Narrow spectrum from initial 60 fs pulse duration shows distinct SHG and THG from different quartz thicknesses. (b) Spectrally integrated non-phase matched SHG and THG efficiency for various quartz thicknesses; solid curves theory.

To this end, we must increase the easily achievable quarter-octave of bandwidth (from a standard hollow-core fibre) to half an octave or further, while simultaneously controlling the phase over the entire spectrum.

In this paper, we describe our technique to simultaneously exploit several perturbative nonlinear effects to further compress these two-cycle pulses and more efficiently generate isolated attosecond pulses. First, we use a high-energy optical parametric amplifier using the idler output. The idler has a passively stable CEP allowing us to carefully control the laser phase. We increase the spectrum by loosely focussing the few-cycle pulse on a thin quartz optic. We exploit the anomalous dispersion of quartz to maintain the phase of the newly created spectral components [21]. We also take advantage of the fact that quartz is a non-centrosymmetric crystal that generates second harmonic with polarization parallel to the driving field [22]. Although this broadened spectrum can sustain a pulse with sub-cycle duration, we further increase the half-cycle contrast through spatio-temporal coupling, where we can use self-focussing in space and time to further compress the pulse [23].

This spatio-temporal coupling technique is limited in that we cannot independently control the spectral phase of all the frequency components, but we find its simplicity to be an attractive alternative for half-cycle pulse creation. Other methods of creating and controlling octave spanning amplified pulses include separating the spectral components and individually controlling the phase [24, 25], or synthesizing a half-cycle pulse out of several coherently combined lasers [26–29]. Although these field synthesizers allow for the control of amplitude and phase of several spectral branches, the active phase stabilization of each spectral component (in addition to the global phase) is cumbersome and has limited this technology from becoming ubiquitous in laser labs.

2. Discussion

The experimental data presented here is from an optical parametric amplifier (OPA) with the idler wavelength centred at 1.8 μm . This OPA is pumped by a 7 mJ, 100 Hz Ti:Sapphire system, and produces 1 mJ, 60 fs pulses, CEP stable to 350 mrad that are coupled into a 1 m long 400 μm diameter hollow core fibre. When the fibre is filled with 1–1.2 bar argon (depending on laser operating conditions), the resulting broadened spectrum supports sub-two-cycle pulses; the fibre output is compressed using 3 mm of fused silica. The maximum available pulse energy at the target chamber is 450 μJ .

We place a thin quartz optic at the loosely focussed beam. Throughout this work, we use X-cut quartz crystals and the laser polarization is perpendicular to the optic axis (unless otherwise stated). In this configuration, quartz has non-centrosymmetry and can generate second harmonic. The second-order susceptibility is relatively low compared to common frequency-doubling crystals such as beta barium borate, lithium triborate, and potassium dideuterium phosphase, where $d_{11} = 0.3 \text{ pm V}^{-1}$ and the second order susceptibility is $\chi^{(2)} = 2d_{\text{eff}}$. However, the second harmonic polarization is parallel to the driving field and this polarization dependence will be crucial in generating half-cycle transients.

The large quartz bandgap of 9 eV allows us to impinge high intensities on the quartz optic before observing damage, enabling us to increase the second harmonic efficiency. We show in figure 1 the low-order harmonic generation from 60 fs pulses (the hollow-core fibre is evacuated) with pulse energy 450 μJ , and focus waist $w_0 = 200 \mu\text{m}$ for a peak intensity of $6 \times 10^{12} \text{ W cm}^{-2}$. We insert different thicknesses of quartz ranging from 80 to 160 μm in 20 μm steps, and record the visible spectrum. (a) The long pulse duration allows for the separation of the harmonic orders. We integrate the spectra to get a total harmonic count for each quartz

thickness. (b) The coherence lengths of the second and third harmonics and their relative amplitudes (third harmonic $30 \times$ weaker than second harmonic) agree well with the simulation (solid curves).

To simulate the low-order harmonics, self-phase modulation, and self-steepening, we use the Forward Maxwell's equation (FME) [30]. Although we use this equation here to demonstrate that it is able to compute the relative harmonic intensities for narrow bandwidth pulses, this simple equation is able to also accurately calculate the evolution of few-cycle pulses, as we demonstrate below.

The FME, in the frame moving at the pulse group velocity $v_g = c/n_g$ is written as

$$\frac{\partial E(\omega)}{\partial z} = i\frac{\omega}{c}[n(\omega) - n_g]E(\omega) + i\frac{\mu_0\omega c}{2n(\omega)}P_{NL}(\omega). \quad (1)$$

In quartz, we have found that the polarization of the generated spectrum is the same as the incident polarization. The nonlinear response is calculated in the frequency domain from an expansion of the dominant perturbative terms,

$$P_{NL} = \epsilon_0\chi^{(2)}E^2 + \epsilon_0\chi^{(3)}E^3. \quad (2)$$

Although there are higher order terms, we find that the nonlinear mixing of the driving field with these orders dominates the generated optical spectral components. For the third order susceptibility, we use the nonlinear index of refraction $n_2 = 3.2 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$, where $\chi^{(3)} = 4n_2\epsilon_0cn^2(\omega)/3$. The broad frequency dependence allows the FME to model few-cycle pulses accurately. Because of the resulting multi-octave spanning spectrum, we also incorporate the frequency dependence of the nonlinear susceptibilities [31],

$$\chi^{(n)}(\omega) = \chi^{(n)}(\omega_0) \left[\frac{n^2(\omega) - 1}{n^2(\omega_0) - 1} \right]^{n+1}. \quad (3)$$

We can test this 1D model on the supercontinuum generation from the fibre to create the two-cycle pulse. To this end, we compare the results of the FME to our recorded spectrum in figure 2(a), where we assume an initial peak intensity of 6 TW cm^2 , matching the experimental conditions listed above. We find that the resulting calculated spectrum shows the expected spectral broadening and the asymmetry from self-phase modulation, in agreement when including higher order terms in the Nonlinear Schrödinger equation [32].

To temporally characterize the pulse, we use the PHz optical oscilloscope technique [33]. We are able to measure broadband, few-cycle pulses with this technique and measure the electric field. A sample recorded trace is shown in figure 2(b), where the white line is the peak of the trace at each time step, including a digital filter from 400 nm to $4 \mu\text{m}$. To compress the pulse out of the fibre, we use the anomalous dispersion of 3 mm of fused silica. We integrate this signal, shown in (c), to retrieve the electric field. We measure a two-cycle pulse with a small post-pulse. We compare the calculated pulse envelope (blue) to the square of the electric field (black), showing good agreement in (d), where we have included the linear propagation through 2.8 mm of fused silica to ideally compress the pulse. These recorded spectra and

temporal traces are in good agreement with the FME results, including higher order terms such as self-steepening and broadband dispersion effects.

We operate in the regime such that the nonlinear phase shift within the fibre, $\phi_{NL} = kn_2 \int_0^L I(z) dz < \pi$. We find that in our setup, increasing the pressure beyond this value—and thus increasing the nonlinear phase shift—creates a filament within the gas-filled fibre. This filament is highly sensitive to environmental conditions such as air currents, pointing instabilities, and pump power fluctuations. These perturbations degrade the CEP stability; recall that stable CEP is crucial for acquiring reproducible data when using few-cycle pulses. However, if one were able to operate in a higher pressure regime, it would be possible to further compress the pulse.

We use the FME to test the limits of pulse compression with only the anomalous dispersion of glass. We find that it is possible to generate near single-cycle pulses from a variety of initial pulse parameters and we demonstrate two results here. In figure 3(a), we show the calculated spectral content (logarithm of intensity) as the pulse propagates through the fibre. Using 30 fs pulses and only slightly increasing the argon gas pressure to 1.5 bar , we are able to compress the fibre output to a near single-cycle pulse with 1.5 mm fused silica. The resulting compressed pulse is shown in (b), blue curve. We obtain a similar temporal result using our experimental parameters, where the initial pulse is 60 fs in duration, but the argon pressure is increased to 3.0 bar (black curve). Although the initial spectrum is narrower in the latter case, the increased pressure compensates via self-phase modulation to create a similar final spectrum and spectral phase. Both of these calculated fields display large post-pulses, where nearly 50% of the energy is in this second pulse [34, 35]. The transform limited case, shown as the red curve, is less than one-cycle in duration. Because it is difficult to fully compensate for the spectral phase over two octaves of bandwidth, the resulting spectrum would likely need to be separated into discrete branches to achieve this transform limit. Additionally, as the infrared spectrum now spans many small molecule resonances, temporal reshaping of the sub-cycle pulse is inevitable when the beam propagates in air [36].

We find that with our experiments, it is simpler to use a near transform limited pulse of two-cycles in duration in the infrared, and compress the pulse immediately before the experiment. To this end, we transmit the two-cycle pulse into our experimental vacuum chamber. The experimental setup is shown in figure 4. We loosely focus the beam down to $200 \mu\text{m}$, where the peak intensity reaches $2 - 3 \times 10^{13} \text{ W cm}^{-2}$, impinges on a thin quartz sample. We do not observe damage in this regime with a two-cycle pulse, but damage does begin when the peak intensity is approximately twice this intensity. We define damage as we observe that a filament forms within the quartz sample and that the resulting spectrum slowly changes as a function of time over the course of tens of seconds to minutes. Inserts show the field evolution from the OPA and the output of the hollow-core fibre. The pulse is strongly chirped, and 3 mm of fused silica

Figure 2. Comparison of simulation to compressed two-cycle pulse from hollow core fibre. (a) Measured (black) and simulated (blue) spectra from fibre; (b) PHz optical oscilloscope trace of two-cycle pulse; (c) retrieved electric field, and (d) square of electric field (black) and simulated intensity envelope (blue) of compressed pulse.

compressed the pulse to near its transform limit. The two-cycle pulse is incident on the thin quartz optic and the spatio-temporal coupling compresses the pulse to a half-cycle transient.

In figure 5, we calculate the spectral and temporal evolution of a two-cycle pulse through quartz. The initial pulse is 11 fs in duration with a Gaussian envelope, centred at $1.8 \mu\text{m}$ with a CEP of 0. The initial peak intensity is $2.5 \times 10^{13} \text{ W cm}^{-2}$. Spectrally, shown in (a), we can see that the second harmonic is created within the first $25 \mu\text{m}$, but the phase mismatch quickly attenuates it. The contribution from the self-phase modulation now dominates and broadens the

spectrum. At $70 \mu\text{m}$ there is a revival of the second harmonic that beats with the fundamental to create the third harmonic. This interference of low order harmonics continues until the output. We compare this coherence length with that shown in figure 1, for the low intensity long pulse regime. This high intensity short pulse case shortens the coherence length through the nonlinear phase shift and results in spectral component interference.

We can observe the effect of the second harmonic interference on the pulse intensity envelope evolution, shown in (b). The initially Gaussian envelope compresses, increasing the peak intensity at $25 \mu\text{m}$. As the pulse propagates through

Figure 3. Simulation of near single-cycle pulse creation from hollow core fibre. (a) Resulting spectrum from propagation in hollow core fibre with initial 30 fs, 1.5 bar argon. (b) Compression using 1.5 mm fused silica creates near single-cycle pulses from 30 fs pulses in 1.5 bar argon (blue) or 60 fs pulses in 3.0 bar argon (black); transform limited pulse in either case is approximately a single-cycle in duration (red).

Figure 4. Experimental setup for pulse compression. (a) Using a hollow-core fibre filled with 1 bar argon broadens the spectrum to support two-cycle pulses. These pulses can be easily compressed through the anomalous dispersion of fused silica (FS). (b) We take these two-cycle pulses and loosely focus the beam onto an 80 μ m thick quartz plate to further compress the pulse to a half-cycle. Inserts: field evolution from OPA output, through fibre, and compression to two-cycle pulses; spatio-temporal compression for half-cycle transient.

Figure 5. Propagation of two-cycle pulse through quartz dependent on propagation length and intensity. (a) The spectrum broadens to cover two octaves. The second harmonic coherence length is modified by the interference of other spectral components. The dispersion above the fourth harmonic limits the maximum bandwidth. (b) The pulse intensity envelope temporal evolution within the quartz. There is a temporal compression at 25, 70, 110, and 160 μm , given by the increased peak intensity. Intensity dependence of (c) spectrum, and (d) square of field (colourbar is normalized to peak field squared) through 80 μm thick quartz. Just below $2 \times 10^{13} \text{ W cm}^{-2}$ the broadened spectrum leads to a half-cycle transient.

the quartz, this compression recurs near where the second harmonic is maximized. Through the interference of the second harmonic, we observe a breathing of the pulse duration and peak intensity.

We can also observe a ‘coherence length’ as a function of initial intensity for the pulse passing through 80 μm thick quartz. We normalize each (c) spectrum and intensity (d) to its peak. In the spectrum, we can see that the second harmonic is

maximized near $1.5 \times 10^{13} \text{ W cm}^{-2}$, but that the third harmonic maximizes at $2 \times 10^{13} \text{ W cm}^{-2}$. In the intensity plot (d), we square the field to show the contrast of each half-cycle. We see that there is a half-cycle transient when the initial peak intensity is just below $2 \times 10^{13} \text{ W cm}^{-2}$. The field pulse duration is shorter than the envelope, where the full width at half maximum (FWHM) duration of the field squared is 1.2 fs. These are optical near-sub-femtosecond

Figure 6. Spatio-temporal coupling leading to (a) increased peak intensity, and (b) pulse compression. The self-focussing spatially compresses the pulses, to a maximum intensity $8 \times 10^{13} \text{ W cm}^{-2}$ 20 mm downstream from the quartz. The temporal compression leads to an envelope with FWHM of 2.5 fs (colourbar is envelope FWHM). (c) The phase dependence from vacuum propagation from immediately after the quartz (bottom) to 20 mm downstream (top).

Figure 7. Simulation of the CEP dependent visible and near-IR spectrum. The second and third harmonic orders interfere to increase the spectrum to the optical and give rise to spectral fringes that represent the relative CEP.

pulses. When we further increase the initial intensity, the half-cycle contrast decreases even though we observe spectral broadening.

In figure 6, we calculate the spatio-temporal coupling of the resulting pulse. To perform this calculation, we propagate the 1D FME through the quartz for 35 different intensities, which account for the initial Gaussian beam profile from the beam axis to five times the initial beam radius. We use the same parameters as above, with an initial beam waist (radius) $w_0 = 200 \mu\text{m}$. We assume that there is no radial cross-coupling within the quartz, $\Delta_{\perp} E(r, \omega) = 0$, because the large beam does not significantly change size through the thin optic. We then calculate the vacuum propagation accounting for this radial dependence via the Hankel transform.

Due to self-focussing, the peak intensity is four times the initial intensity after the beam has propagated in vacuum by 20 mm. We find that the resulting peak intensity is dependent not only on the initial intensity and beam size, but also the

CEP. Temporally, the pulse compresses as well, also displaying a CEP dependence. We are able to measure a pulse envelope with FWHM of 2.5 fs, which is less than one half-cycle in duration.

Because the pulse duration and peak intensity are CEP sensitive, we also expect that to be the case with the spectrum. In figure 7, we show the spectrum as a function of CEP at 20 mm from the quartz plate. In the fundamental portion of the spectrum, around $1 \times 10^{15} \text{ rad s}^{-1}$, there is little amplitude modulation. However, in the higher energy portion of the spectrum, we see an obvious amplitude modulation, showing a spectral breathing modulo 2π . Additionally, we observe spectral fringes near the third harmonic, which are due to the interfering second and third harmonic pathways and is the measurement of the CEP [37, 38]. Thus, not only does the quartz optic compress the pulse and increase the peak intensity, but it also gives us a measurement of the laser CEP [15]. This CEP information gives us a direct out-of-loop

Figure 8. PHz optical oscilloscope measurement of the half-cycle transient. (a) PHz trace of the measured transient; (b) retrieved field and (c) intensity. The contrast of the main half-cycle to the satellite half-cycles is nearly 3:1.

measurement on the field that is used to drive the strong field processes [39]. We expect that this will benefit high energy, low repetition rate systems where stabilizing the CEP remains a challenge [40].

We measure the half-cycle transient field in figure 8, again using the PHz optical oscilloscope technique [33]. Comparing the measured field here with that shown in figure 2, we see that the preceding and following half-cycles are suppressed relative to the peak half-cycle to create the half-cycle transient field. This half-cycle transient has an intensity contrast of peak to satellite half-cycles of approximately 3:1.

We use these spatially and temporally compressed pulses to generate isolated attosecond pulses in the XUV through high harmonic generation. To deliver the gas at the nonlinear focus 20 mm from the quartz plate, we use a 250 μm diameter pulsed nozzle with 2.4 bar backing pressure of xenon. The resulting spectra are shown in figure 9. Without the quartz plate, we observe that the two-cycle pulse generates a series of high-order harmonics that extend up to 35 eV. Blue/green curves are for two values of CEP. Below 25 eV, these harmonics are easily distinguishable and are not affected by the change in the CEP. Above 25 eV, there is a slight decrease in

Figure 9. XUV spectra from xenon without (green, blue) and with (red, black) quartz 20 mm before gas jet for two different CEP values.

the modulation and the position of the harmonic peaks change due to the laser CEP. These spectra are consistent with the strong-field approximation (SFA) calculation that the attosecond pulse contrast is very poor, approximately 1.3:1.

Conversely, when we insert the 80 μm thick quartz plate 20 mm before the gas jet with the laser polarization perpendicular to the optic axis, we see an obvious change in the XUV spectra. The XUV flux significantly increases and the spectrum is now highly sensitive to the laser CEP. There are two reasons for the increase in XUV flux. First, the spatio-temporal self-focussing increases the driving field peak intensity. The second factor comes from the second harmonic generated in parallel polarization from the quartz. The flux increase from the second harmonic is consistent with a relative intensity of $I_{2\omega}/I_{\omega} = 2\%$.

The observed CEP dependence is consistent with the driving field's having a sub-cycle pulse duration. The continuum spectrum, with the CEP = 0 (red curve), shows an increase in flux by a factor of 55 at 21 eV (taken at the peak of the harmonic). We can change the XUV photon energy by nearly an octave as we change the CEP by π , creating a continuum spectrum now up to nearly 40 eV (black curve). These strongly CEP dependent spectra are consistent with a sub-cycle duration driving field. The SFA calculation predicts an attosecond pulse contrast of nearly 1000:1.

3. Applications and future outlook

These half-cycle pulses may benefit biological imaging two-fold: by more efficiently producing high energy attosecond pulses in the water window, and these sub-cycle infrared pulses could be used in multiphoton microscopy.

We predict that this simple method of half-cycle transient field synthesis can be used to create bright isolated attosecond pulses in the soft x-ray region. In the energy range around 300 eV—called the water window—it is possible to distinguish between carbon and oxygen *K*-shell absorption edges [41]. In this energy range, we can send in an attosecond pulse as a source for coherent diffraction imaging to image a biological sample while it is in an aqueous solution, or study sub-cellular structures in the cytosol. With the sub-100 attosecond resolution that such broad pulses could provide, we could also observe the electron transport through these cellular structures. We now describe how it may be possible to generate these bright high energy attosecond pulses.

We cannot place the quartz optic immediately before a focus that reaches intensities of $5 \times 10^{14} \text{ W cm}^{-2}$, but we can place the quartz several cm before the focus of a loosely focussed beam. We simulate a two-cycle pulse centred at 1.8 μm with an initial beam waist of 6.5 mm focussed with a 75 cm focal length lens, shown in figure 10. First, we calculate the CEP dependent XUV spectra from a 0.9 mJ two-cycle pulse, where the maximum cutoff is 400 eV, shown in (b). Again, this two-cycle pulse predicts a train of four attosecond pulses, where the unique bandwidth of two successive pulses is approximately 100 eV. Second, we simulate the insertion of an 80 μm thick quartz optic 706 mm downstream from the lens, as shown in figure 9(a), where the peak intensity is $2.5 \times 10^{13} \text{ W cm}^{-2}$ and is sufficient to create the broadened spectrum and compress the pulse. With a pulse energy of 1.3 mJ, the spatio-temporal reshaping is optimized 30 mm

downstream of the quartz optic, where the self-focussing decreases the focal length by 14 mm. In (c), the intense half-cycle transient produces an isolated attosecond pulse with a bandwidth of nearly 250 eV, and the XUV peak flux is increased by a factor of 65.

Conversely, it may be possible to use these half-cycle pulses directly. Multiphoton microscopy benefits greatly from decreased pulse duration. However, as many femtosecond lasers are Ti:Sapphire based, three photon processes damage biological tissue and limits the multi-photon signal [42]. Here, by using an infrared source centred at 1.8 μm , we should be able to reach seven photon processes before inducing damage, at which point we are likely beyond the perturbative regime. By reducing the pulse duration to a half-cycle centred at 1.8 μm , we may be able to reach regimes that were not accessible before for multiphoton imaging.

The signal enhancement (SE) of ultrashort pulses in nonlinear processes is given by $\text{SE} = g/(\tau f_R)^{n-1}$, where g is a unitless factor depending on the pulse shape (on the order of unity), τ is the pulse duration, f_R is the laser repetition rate, and n is the degree of nonlinearity [43]. Changing from a three photon process with a Ti:Sapphire laser—the workhorse for many ultrafast labs—to a seven photon process from 1.8 μm with a 1 fs pulse duration, $\text{SE} > 10^{64}$.

Additionally, the benefits of using an infrared laser source rather than a visible half-cycle laser field is that photodamage decreases with increasing wavelength [44]. In this way, we can impinge a high peak intensity pulse in the infrared on a biological sample. The penetration of the infrared could allow for deeper *in situ* probing of biological tissues, while the ultrashort pulse will allow for multiphoton processes to map out a sample.

The limitations of this source for half-cycle transient fields depend on the application and the required half-cycle contrast. In the present work, the half-cycle contrast is limited to approximately 3:1. We are investigating the parameter space that can optimize this pulse contrast. The parameter space includes not only the driving field, but also engineering the quartz optic where we can put in interstitial layers of material with higher $\chi^{(2)}$ or $\chi^{(3)}$, similar to periodic poling used to increase second harmonic generation.

In regards to the source, the driving wavelength, and initial pulse duration, the shorter the initial pulse the more efficiently the pulse can be compressed. We found that according to our calculations, pulses shorter than 15 fs (2.5 cycles) will create a half-cycle pulse for a central wavelength of 1.8 μm . For sub-two-cycle initial pulses, the central wavelength can be from 1.6 to 2.5 μm to create a half-cycle pulse. The short wavelength limitation is the normal dispersion below 1.3 μm in quartz. In this regime, the dispersion of the second and third harmonics is too large to maintain the pulse duration after the propagation through tens of μm . However, it may be possible to limit the quartz plate to one coherence length, approximately 20 μm , and still create a half-cycle pulse although with limited contrast. The long wavelength limit is due to the oxygen absorption near 4 μm in quartz.

Figure 10. Setup for generating bright soft x-ray isolated attosecond pulses. (a) Placing the quartz far before the focus limits the damage on the optic, while downstream the peak intensity is sufficient to generate coherent soft x-rays. The self-focussing decreases the focal length by 14 mm. (b) Using a two-cycle pulse generates several attosecond pulses; the half-cycle cutoff is visible, but with poor contrast. (c) Using the half-cycle transient field generates a much brighter, isolated attosecond pulse.

Finally, it may be possible to find new optical materials with a higher second harmonic efficiency to create these half-cycle pulses [45]. These materials may allow us to more efficiently compress the pulses, or extend the wavelength range of the sources to create these half-cycle pulses.

4. Conclusion

In conclusion, we discuss the spatio-temporal compression of a few-cycle near infrared pulse as it propagates through a quartz plate at high intensity. We model the pulse evolution in a hollow-core fibre and through a thin quartz plate using the FME, quantitatively predicting the parameter space that can optimize pulse compression in a hollow-core fibre and thin optic. We place a thin quartz optic in a moderately high intensity regime, approximately $2 \times 10^{13} \text{ W cm}^{-2}$, but well below damage for a two-cycle pulse, to create half-cycle transients. We use these half-cycle transients to produce isolated attosecond pulses, increasing the generated flux and attosecond pulse contrast. We predict a simple way to implement this technique to create bright isolated attosecond pulses in the soft x-ray region.

The bandwidth of these isolated attosecond pulses is controlled through the CEP of the driving laser. Because the peak field strength controls the bandwidth and the time of recollision, the attosecond pulse duration is also CEP dependent. We compress the generated attosecond pulses

with the anomalous dispersion of ultrathin metal filters [46], but unfortunately, this method is not finely tuneable. By tuning the attochirp—that is the XUV spectral phase—through the driving field CEP, it may be possible to finely control the initial XUV spectral phase to produce transform limited isolated attosecond pulses.

We expect that this technique—placing a thin optic that can generate second harmonic and with anomalous dispersion of the fundamental frequency upstream of an experiment—will allow for the increase in XUV flux from many infrared sources. Attosecond groups are beginning to push infrared amplifier technologies to extend high harmonic generation to the x-ray region. The increased flux from the spatio-temporally coupled pulse should allow for faster data acquisition and better signal-to-noise in a region where the photoelectron cross-section is significantly decreased. Simultaneously, research into strong-field condensed matter physics is opening up attosecond technologies to semiconductors. At high field strengths, using a half-cycle infrared pulse can drive currents, but avoid optically induced damage.

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References

- [1] Corkum P B and Krausz F 2007 Attosecond science *Nat. Phys.* **3** 381–7
- [2] Krausz F and Ivanov M 2009 Attosecond physics *Rev. Mod. Phys.* **81** 163
- [3] Shiner A D, Schmidt B E, Trallero-Herrero C, Woerner H J, Patchkovskii S, Corkum P B, Kieffer J-C, Legaré F and Villeneuve D M 2011 Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy *Nat. Phys.* **7** 464–7
- [4] Vampa G, Hammond T, Thiré N, Schmidt B E, Légaré F, McDonald C R, Brabec T and Corkum P B 2015 Linking high harmonics from gases and solids *Nature* **522** 462–4
- [5] Fortier T, Jones D J and Cundiff S T 2003 Phase stabilization of an octave-spanning Ti:sapphire laser *Opt. Lett.* **28** 2198–200
- [6] Krauss G, Lohss S, Hanke T, Sell A, Eggert S, Huber R and Leitenstorfer A 2010 Synthesis of a single cycle of light with compact erbium-doped fibre technology *Nat. Photon.* **4** 33–7
- [7] Nisoli M, Stagira S, Silvestri S D, Svelto O, Sartania S, Cheng Z, Lenzner M, Spielmann C and Krausz F 1997 A novel-high energy pulse compression system: generation of multigigawatt sub-5-fs pulses *Appl. Phys. B* **65** 189
- [8] Suda A, Hatayama M, Nagasaka K and Midorikawa K 2005 Generation of sub-10-fs, 5-mJ-optical pulses using a hollow fiber with a pressure gradient *Appl. Phys. Lett.* **86** 111116
- [9] Schmidt B E *et al* 2010 Compression of 1.8 μm laser pulses to sub two optical cycles with bulk material *Appl. Phys. Lett.* **96** 121109
- [10] Zhang C, Vampa G, Villeneuve D M and Corkum P B 2015 Attosecond lighthouse driven by sub-two-cycle, 1.8 μm laser pulses *J. Phys. B: At. Mol. Opt. Phys.* **48** 061001
- [11] Sola I J *et al* 2006 Controlling attosecond electron dynamics by phase-stabilized polarization gating *Nat. Phys.* **2** 319–22
- [12] Kim K T, Zhang C, Shiner A D, Kirkwood S E, Frumker E, Gariépy G, Naumov A, Villeneuve D M and Corkum P B 2013 Manipulation of quantum paths for space-time characterization of attosecond pulses *Nat. Phys.* **9** 159–63
- [13] Kim K T, Zhang C, Ruchon T, Hergott J-F, Auguste T, Villeneuve D M, Corkum P B and Quéiré F 2013 Photonic streaking of attosecond pulse trains *Nat. Photon.* **7** 651–6
- [14] Hammond T, Kim K T, Zhang C, Villeneuve D M and Corkum P B 2015 Controlling attosecond angular streaking with second harmonic radiation *Opt. Lett.* **40** 1768–70
- [15] Baltuska A *et al* 2003 Attosecond control of electronic processes by intense light fields *Nature* **421** 611–5
- [16] Kienberger R *et al* 2004 Atomic transient recorder *Nature* **427** 817
- [17] Ishii N, Kaneshima K, Kitano K, Kanai T, Watanabe S and Itatani J 2014 Carrier-envelope phase-dependent high harmonic generation in the water window using few-cycle infrared pulses *Nat. Commun.* **5** 3331
- [18] Hammond T J, Brown G G, Kim K T, Villeneuve D and Corkum P B 2016 Attosecond pulses measured from the attosecond lighthouse *Nat. Photon.* **10** 171
- [19] Sansone G, Poletto L and Nisoli M 2011 High-energy attosecond light sources *Nat. Photon.* **5** 655–63
- [20] Schmidt B E, Shiner A D, Giguère M, Lassonde P, Trallero-Herrero C A, Kieffer J C, Corkum P B, Villeneuve D M and Légaré F 2012 High harmonic generation with long-wavelength few-cycle laser pulses *J. Phys. B: At. Mol. Opt. Phys.* **45** 074008
- [21] Hammond T J, Villeneuve D M and Corkum P B 2017 Producing and controlling half-cycle near-infrared electric-field transients *Optica* **4** 826–30
- [22] Franken P A, Hill A E, Peters C W and Weinreich G 1961 Generation of optical harmonics *Phys. Rev. Lett.* **7** 118–9
- [23] Hammond T J, Monchocé S, Zhang C, Vampa G, Klug D, Naumov A, Villeneuve D M and Corkum P 2017 Integrating solids and gases for attosecond pulse generation *Nat. Photon.* **11** 594–9
- [24] Wirth A *et al* 2011 Synthesized light transients *Science* **334** 195
- [25] Hassan M T *et al* 2016 Optical attosecond pulses and tracking the nonlinear response of bound electrons *Nature* **530** 66
- [26] Huang S *et al* 2011 High-energy pulse synthesis with sub-cycle waveform control for strong-field physics *Nat. Photon.* **5** 475
- [27] Cox J A, Putnam W P, Sell A, Leitenstorfer A and Kaertner F X 2012 Pulse synthesis in the single-cycle regime from independent mode-locked lasers using attosecond-precision feedback *Opt. Lett.* **37** 3579
- [28] Chia S H, Cirmi G, Fang S, Rossi G M, Muecke O D and Kaertner F X 2014 Two-octave-spanning dispersion-controlled precision optics for sub-optical-cycle waveform synthesizers *Optica* **1** 315
- [29] Fattahi H *et al* 2014 Third-generation femtosecond technology *Optica* **1** 45
- [30] Husakou A and Hermann J 2001 Supercontinuum generation of higher-order solitons by fission in photonic crystal fibers *Phys. Rev. Lett.* **87** 203901
- [31] Ettoumi W, Petit Y, Kasparian J and Wolf J-P 2010 Generalized miller formulae *Opt. Express* **18** 6613–20
- [32] Béjot P, Schmidt B E, Kasparian J, Wolf J-P and Légaré F 2010 Mechanism of hollow-core-fiber infrared-supercontinuum compression with bulk material *Phys. Rev. A* **81** 063828
- [33] Kim K T, Zhang C, Shiner A D, Schmidt B E, Légaré F, Villeneuve D M and Corkum P B 2013 Petahertz optical oscilloscope *Nat. Photon.* **7** 958
- [34] Johnson A S, Miseikis L, Wood D A, Austin D R, Brahms C, Jarosch S, Strueber C S, Ye P and Marangos J P 2016 Measurement of sulfur $L_{2,3}$ and carbon K edge xanes in a polythiophene film using a high harmonic supercontinuum *Struct. Dyn.* **3** 062603
- [35] Austin D R, Witting T, Weber S J, Ye P, Siegel T, Matia-Hernando P, Johnson A S, Tisch J W and Marangos J P 2016 Spatio-temporal characterization of intense few-cycle 2 μm pulses *Opt. Express* **24** 24788
- [36] Hammond T J, Monchocé S, Zhang C, Brown G G, Corkum P B and Villeneuve D M 2016 Femtosecond time-domain observation of atmospheric absorption in the near-infrared spectrum *Phys. Rev. A* **94** 063410
- [37] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis *Science* **288** 635–9
- [38] Apolonski A, Poppe A, Tempea G, Spielmann C, Udem T, Holzwarth R, H'ansch T W and Krausz F 2000 Controlling the phase evolution of few-cycle light pulses *Phys. Rev. Lett.* **85** 740
- [39] Fuji T *et al* 2005 Monolithic carrier-envelope phase-stabilization scheme *Opt. Lett.* **30** 332–4
- [40] Cunningham E, Wu Y and Chang Z 2015 Carrier-envelope phase control of a 10 Hz, 25 TW laser for high-flux extreme ultraviolet quasi-continuum generation *Appl. Phys. Lett.* **107** 1108
- [41] Chen M-C, Arpin P, Popmintchev T, Gerrity M, Zhang B, Seaberg M, Popmintchev D, Murnane M M and

- Kapteyn H C 2010 Bright, coherent, ultrafast soft x-ray harmonics spanning the water window from a tabletop light source *Phys. Rev. Lett.* **105** 173901
- [42] Cruz J M D, Lozovoy V V and Dantus M 2006 Coherent control improves biomedical imaging with ultrashort shaped pulses *J. Photochem. Photobiol.* **180** 307–13
- [43] Sheetz K E and Squier J 2009 Ultrafast optics: imaging and manipulating biological systems *J. Appl. Phys.* **105** 051101
- [44] Nimmerjahn A, Theer P and Helmchen F 2008 Two-photon laser scanning microscopy *Ultrashort Laser Pulses in Biology and Medicine* ed M Braun, P Gilch and W Zinth (Berlin: Springer) p 45
- [45] Haertle D, Jazbinsek M, Montemazzani G and Günter P 2005 Nonlinear optical coefficients and phase-matching conditions in $\text{Sn}_2\text{P}_2\text{S}_6$ *Opt. Express* **13** 3765–76
- [46] López-Martens R *et al* 2005 Amplitude and phase control of attosecond light pulses *Phys. Rev. Lett.* **94** 033001