Generation and Characterisation of <10 fs pulses for Ultrafast Spectroscopy in ULTRA

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Abstract

Sub-10 fs pulses for driving 2-dimensional electronic spectroscopy experiments at the ULTRA have been produced and characterised using a combination of hollow core fibre pulse broadening and a commercial dispersion scan system. This source will be initially for 2D electronic spectroscopy, but opens the door for a range of other broadband time-resolved spectroscopies.

1 Introduction

Few-cycle ultrafast pulses in the near infra-red (NIR) have a variety of applications, including 2-dimensional electronic spectroscopy (2DES, as is the intent for this project) [1, 2], as well as high harmonic generation [3], impulsive Raman scattering [4] and ultrashort UV-pulse generation [5]. To generate these pulses, one must first broaden a driving laser field, often on the order of 30 – 100 fs, and then compress the pulse by compensation of the spectral phase [6].

The use of hollow core fibres (HCFs) for this purpose is well established, and most commonly utilises a hollow capillary filled with a noble gas. In these systems, self- and cross-phase modulation alongside four-wave mixing allow the efficient broadening of these pulses while providing passive pointing stabilisation for the emerging pulses [7].

Alternative methods for pulse broadening include filamentation [8], multi-pass cells [9] and multiple plate and bulk glass broadening systems [10]. Compared to these systems, HCF broadening presents a number of benefits. Firstly, the propagation presents a fixed point for the output of the beam, providing passive pointing stabilisation. The gas pressure inside the capillary can be easily tuned to match the spectral broadening required and to compensate for day-to-day spectral phase or pulse energy changes, making these systems flexible. Finally, as the system we have built uses commercially readily available low-cost hollow capillaries, they can be easily rebuilt and replaced in the case of laser damage, or to match new experimental requirements.

To compress the broadened pulses, a commercial dispersion scanning diagnostic and compressor (d-scan) have been installed. The d-scan comprises two main units: an inline pulse compressor which utilises chirped mirrors and wedged glass to compensate higher order phases of the pulse; and a measuring head, which contains a barium borate (BBO) crystal for second harmonic generation [11]. The spectral intensity and phase of the generated second harmonic is dependant on the duration and spectral and temporal phases of the incoming pulse, and the phase imparted by a fixed length of glass in the beam is well defined. As a result, as long as the optimal compression point for the pulse is crossed by scanning the wedges, the spectral phase of the the ultrashort pulse can be reconstructed using a retrieval algorithm.

While a range of other diagnostic techniques for measuring few-femtosecond pulses exist [12–14], the d-scan provides a range of benefits. First, because the technique must necessarily scan the dispersion of the beam through the optimal compression point, it acts as both a diagnostic and an in-line compressor for the system. Furthermore, because the measurement head is independent of the compression unit, the compression can be optimised for any point along the beam path. This is in contrast to FROG or SPIDER systems where the diagnostic arms are independent of the beamline, and so while the retrieved phase can be used to estimate downstream phases using simple phase propagation techniques, they cannot easily be used to measure phases at arbitrary points in an experimental layout.

The few-cycle pulses produced by this system are to be used for few-femtosecond 2DES, which is an ultrafast optical technique that utilises a sequence of pulses in the 400 – 950 nm range to study ultrafast chemistry

[15]. 2DES allows identification of ultrafast relaxation dynamics by extracting electronic dynamics from two-dimensional datasets comprising “excitation” and “detection” axes. In our experiments the excitation axis is determined by the bandwidth of the laser pulse used, and the use of a broadband pulse allows us to observe dynamics instigated by a range of excitation wavelengths [2]. The detection axis is determined by the Fourier transform of the third order response over the time delays between the first two pulses in the system. These delays are scanned over a range of picoseconds in femtosecond steps. Electronic dynamics have characteristic times on the order of tens of femtoseconds, and so to resolve these the driving pulses pulses must have few-femtosecond durations.

2 Methods

In ULTRA we have installed a 30 cm long, 100 µm inner-diameter HCF, filled with 2.5 bar of argon, and are driving the broadening of 35 fs, 200 µJ, 800 nm laser pulses at 10 kHz. The cells used for the fibre mounting were purchased from the Travers group at Heriot-Watt University [5]. A layout schematic for the system on the optical table can be seen in fig. 1

The energy of the pulses used is controlled by a half wave plate and linear polariser, with 1 mm UV fused silica windows on the fibre cells to mitigate b-integral accumulation and the windows at Brewster’s angle to remove back-reflections. The gas pressure safety limits of the system computed to match commercial standards, with the system safe to operate up to 5 bar backing pressures [17].

While the beam is stabilised upstream by two-point stabilisation using a Newport GuideStar II system, the

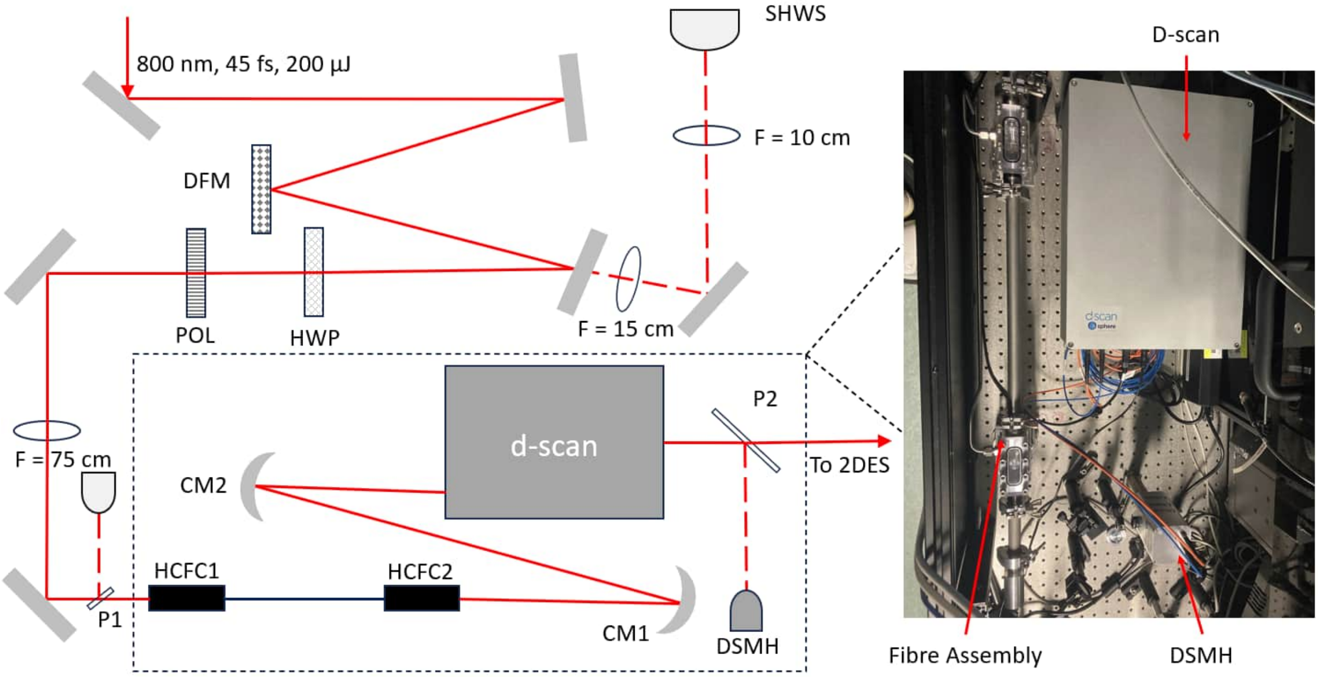

Figure 1: Left: A schematic of the beam path to the HCF and the diagnostics before and after it to prepare the beam for use in 2DES experiments. The solid lines represent the primary beam path, and the dashed lines are leak-through and pickoff beams for diagnostics. DFM is the deformable mirror, HWP and POL are the half wave plate and polariser for pulse energy control, and SHWS is the Shack Hartmann wavefront sensor. P1 is a pickoff to allow monitoring of the focal plane of the beam which couples into the fibre cell HCFC1. The beam leaves the fibre inside the rear cell HCFC2 and is recollimated to a 3 mm beam waist by CM1 and CM2. This beam is then passed through the d-scan compression module and the emerging beam has a small pick off sent to the d-scan measuring head (DSMH) while the main beam is sent to the 2DES experimental setup. Right: The fibre and d-scan as installed in ULTRA-B. The fibre is housed in standard KF tubing with gas fill and vent lines installed by CLF engineering. The 1 mm thick UV fused silica windows are pressure rated up to 5 bar in this system. The d-scan box contains chirped mirrors and a pair of wedges for adjustable bulk compression. The measuring head (DSMH) has two optical fibres for measurement of both the fundamental spectrum and the second harmonic in spectrometers located in the d-scan box.

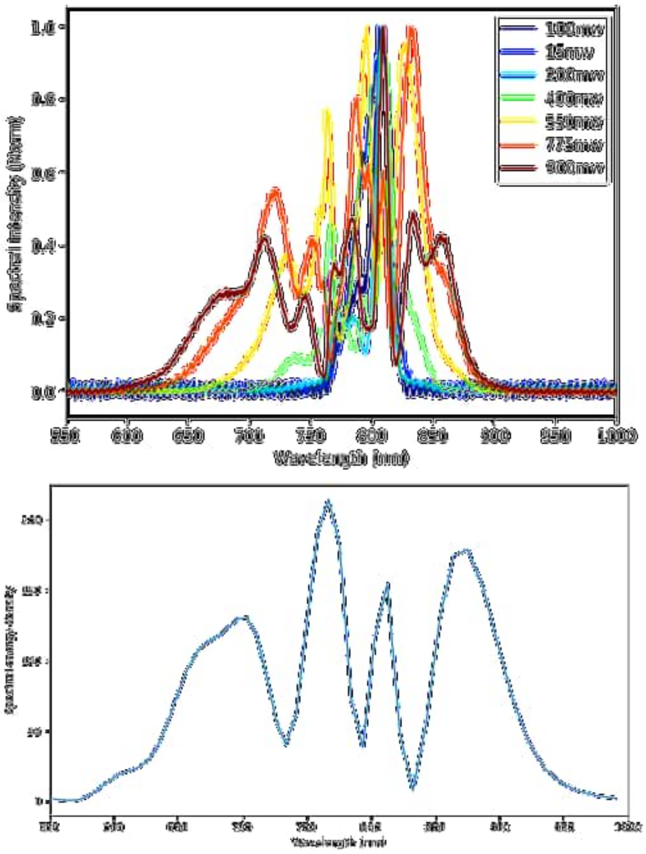

Figure 2: Above: The broadening of the spectrum of the pulse as the pulse energy is increased. The increase in pulse energy drives greater broadening due to third order nonlinearity. Below: Simulated spectra for the broadening of the pulses, calculated using [16].

position of the focal spot on the entrance of the fibre is stabilised using a 32-element deformable mirror (Dynamic Optics PZT Multi), with the leak-through of the beam on the following mirror used to image the the conjugate plane of the deformable mirror on a Shack Hartmann wavefront sensor (Dynamic Optics).

The spectrally broadened pulses seen in fig. 2 were reconstructed and compressed using the commercial d-scan as seen in fig. 3. To do this a partial reflection of the compressed pulse is sent to the d-scan measurement head to generate its second harmonic. Both a linear and second harmonic spectrum are recorded for the purposes of reconstruction. The reconstruction gives us a full-width at half-maximum duration for the pulse of 4.75 fs with <2% error in the reconstruction of the trace.

3 Future Development and Further Applications

As the broadened pulses can now be rapidly and reproducibly retrieved, the comissioning of the 2DES experimental apparatus is now underway. The original experimental system was provided through collaboration with the Meech group at the University of East Anglia to be repurposed as a facility experiment [2, 18, 19]. While the system operates under the same principal, a number of modifications are in progress to allow for the use of >200 nm spectral bandwidth, <10 fs pulses to be used, as this requires adaption due to phase considerations. A number of standard dyes will be used for commissioning experiments in the coming months.

While this particular fibre installation is for use in 2DES experiments, the implementation design has been engineered to provide an easily reproducible platform for similar systems throughout the CLF. All fittings are standard KF, Swagelock or british standard pipe (BSP) where possible, and the Brewster's angle window mounts can easily be altered to fit desired driving wavelengths.

Similar to 2DES, impulsive Raman experiments also require few-femtosecond, broadband pulses to excite a continuum of excited states in the impulsive limit (i.e. duration of the pulse is substantially shorter than that of the instigated dynamics). By scanning the delays between a pair of pump pulses and a final probe pulse, the scanned delays and measured Raman spectra the vibrational states of a system can be recovered from the modulations of its refractive index [4].

HCF systems needn't only be used to broaden pulses, as soliton self-compression allows pulse compression down to single cycle durations within [5]. This is key both for driving HHG, as is done in the Artemis facility, as well as for a variety of time resolved spectroscopies. They can also be used for generating resonant dispersive waves — few-femtosecond UV pulses with sub-femtosecond timing jitter that can be coupled into vacuum systems [20, 21]. These include quantum control experiments, where the timing resolution of three independent pulses must be controlled — often one in the UV to "pump" dynamics, one in the infrared to influence the dynamics, and a final x-ray pulse to "probe" a core state [22, 23].

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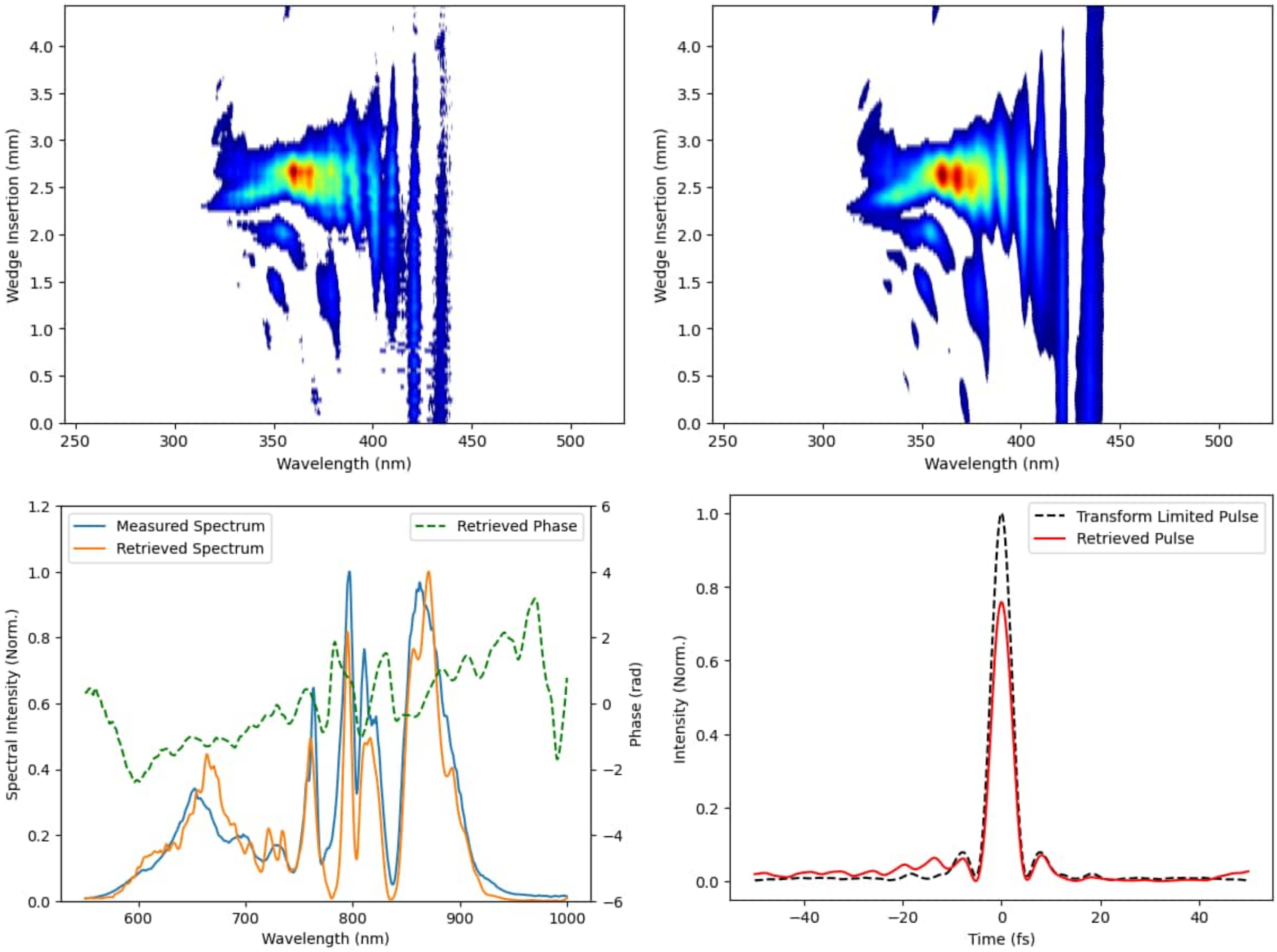

Figure 3: The diagnostic data from measurements taken with the d-scan. Top-left: The measured d-scan trace, showing spectrum vs. thickness of glass insertion. Optimal compression is reflected by highest spectral intensity, seen at 2.8 mm wedge insertion. Top-right: The reconstruction of the measured trace using proprietary reconstruction algorithms provided by Sphere Photonics. Bottom-left: The reconstructed (orange) vs. measured (blue) spectrum at the point of optimal pulse compression, overlayed with the retrieved spectral phase (green, dashed). Bottom-right: The retrieved temporal intensity of the pulse (red) and that of the ideal transform limited pulse (black, dashed). The reconstruction gives a pulse duration of 4.75 fs, with a transform limit of 4.68 fs ad a reconstruction error of 1.9%.

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