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ABSTRACT

In the context of ultrafast spectroscopy, the availability of few-femtosecond UV pulses is key to disclose the role of electron dynamics in photo-activated biochemically relevant processes. Here, we present an optical setup for the generation of UV pulses spectrally tunable between 270 and 350 nm with transform limited durations of 3.0 and 2.9 fs, respectively, and repetition rates up to 50 kHz, using resonant dispersive wave (RDW) emission in an argon-filled hollow-core fiber. The RDW emission is driven by 1030 nm sub-20 fs pulses produced by post-compressing an Yb-based laser with a dispersion-engineered multi-pass cell (MPC). The combination of an MPC with a capillary constitutes a compact source to deliver few-femtosecond UV pulses at high-repetition rates, which is ideal for statistically demanding experiments in molecular physics.

The investigation of electron dynamics in matter requires extremely short pulses, with durations from a few femtoseconds to a few tens of attoseconds. Since the shortest achievable pulse duration is ultimately limited by the carrier wavelength, state-of-the-art attosecond and few-fs sources typically operate in the extreme ultraviolet (XUV) or vacuum ultraviolet (VUV).[1] Although ultrafast XUV and VUV sources have paved the way for the study of photophysical and photochemical phenomena on the electron time scale, their characteristic photon energy above the typical ionization threshold of organic species prevents the study of valence shell electron dynamics in molecules. Conversely, UV light pulses enable light induced excitation without ionization and allow electronic processes to be studied in neutral molecules.[2] However, efficient generation of few-optical-cycle UV pulses is technologically challenging and has only been achieved in the past decade.[3] In particular, ultrashort UV pulses have been generated by nonlinear frequency upconversion of ultrashort near-infrared (NIR) pulses using numerous approaches: high-order achromatic phase matching in doubling crystals,[4] harmonic generation in noble gases,[5] four-wave mixing in gases,[6] or in laser-machined high-pressure glass cells reaching down to sub-2 fs durations[7] with μJ-level pulse energy.[8] Another promising avenue for the production of few-fs UV pulses is resonant dispersive wave (RDW) emission in hollow-core fibers (HCFs), which compared to the above-mentioned techniques enable a remarkable spectral tunability. Deep UV RDW emission was pioneered in anti-resonant hollow-core fibers[9–11] but more recently has been optimized in flexible hollow-capillary fibers, achieving generation of bright pulses tunable down to 110 nm with energies in the microjoule range, corresponding to conversion efficiencies up to 15%.[12,13] This scheme has been applied to produce sub-3 fs pulses in the deep UV.[11,14] Recently, RDW sources have been combined with high-average power Yb driving lasers to scale the UV source repetition rates. Since Yb-based sources deliver relatively long pulse durations, above 100 fs, which are incapable of directly driving coherent RDW emission, significant reduction of the driving pulse duration is necessary, so far only achieved via the use of hollow-core fibers (HCFs).[15] In recent years, multi-pass cell (MPC) pulse compression has emerged as a promising alternative to HCF. This alternative has been shown to provide better energy scalability, higher efficiencies, and lower sensitivity to beam pointing instabilities.[16,17]

In this work, we use a high-repetition rate ytterbium laser system to demonstrate a compact table-top setup for the generation of few-fs UV light through RDW emission driven by a single-stage dispersion-engineered multi-pass cell (MPC) pulse-post-compressor. The latter was previously reported in detail in Ref. 18, providing advantages for the compression stage in terms of compactness, throughput, and power scalability.[19,20] By driving a hollow-core fiber (HCF) with 15 fs pulses centered at 1030 nm from an MPC, deep UV is generated through RDW emission with a tunability from 270 nm up to 340 nm with μJ pulse energy, scalable in repetition rate from 1 kHz up to 50 kHz.

Figure 1 shows the experimental setup made of two stages: (i) a pulse compression stage using an MPC followed by (ii) DUV-generation through RDW emission in a gas-filled hollow capillary. A commercial ytterbium-doped fiber laser system (Tangerine, Amplitude Lasers), delivering 250-μJ, 150-fs pulses with tunable repetition rate between 1 and 200 kHz (average power from 250 mW to 50 W), was used. The MPC is composed of two concave mirrors, with a radius of curvature (ROC) of −200 mm and a diameter of 50 mm. These two mirrors are separated by a distance $L \approx 400$ mm, which gives an L/R ratio of ~1.98. The MPC is filled with krypton gas, with a pressure of 1.4 bar, to achieve nonlinear spectral broadening through self-phase modulation (SPM). In order to partially compensate for the linear dispersion, one of the MPC mirrors is coated with a group delay dispersion (GDD) of −30 fs^2. The light is mode-matched to the eigenmode of the MPC using a lens telescope resulting in a circular Herriott pattern with 15 equally spaced reflections on each cell mirror, as illustrated in Fig. 1 (inset).

The MPC output is then collimated using a single curved mirror with a radius of curvature (ROC) = −1500 mm and sent to a broadband double chirped mirror (DCM) pair in combination with 1 mm of fused silica for subsequent compensation of the phase, acquired through SPM nonlinearity inside the MPC. The total throughput of the MPC post-pulse compression stage is 86% (93% MPC throughput).

Figure 2 illustrates the complete pulse characterization after the multi-pass cell pulse-post compression stage. The spectra generated in this first stage are measured with two different spectrometers, Ocean FX and NirQuest from Ocean Insight Inc.,[17] and merged to capture the full spectral range. At 50 kHz repetition rate, the spectrum supports a transform limited (TL) pulse duration of 13 fs at full width at half maximum (FWHM). The post-compressed pulses are then characterized using a home-built second harmonic generation frequency resolved optical gating (SHG FROG) setup, yielding a temporal pulse duration of 14.7 fs with a FROG error of 0.05% using a grid size of 512 × 512.

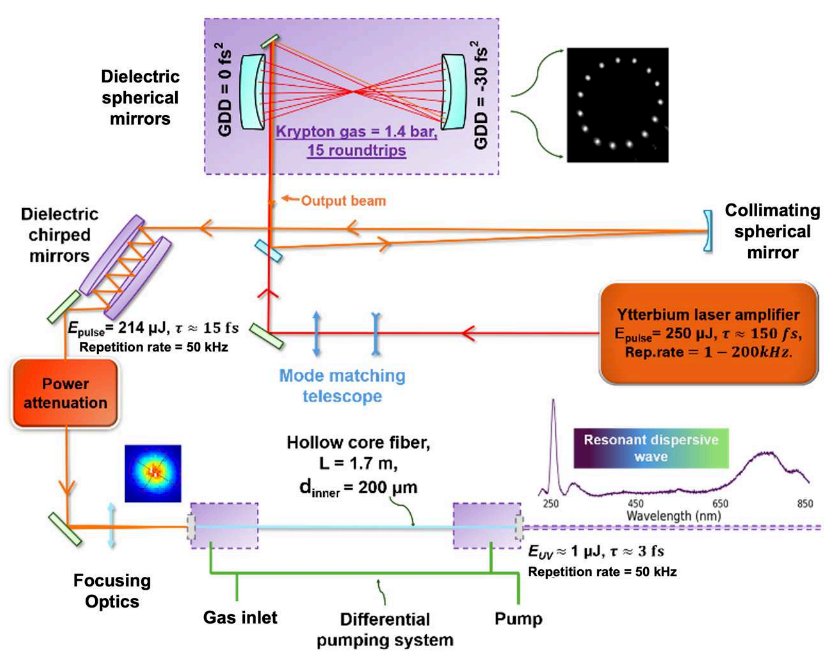

FIG. 1. Overall experimental layout consisting of a dispersion-engineered MPC used to drive RDW emission in a gas-filled HCF. The inset shows the MPC pattern to monitor the mode-matching for the MPC as well as input beam profile of the driving IR field for the RDW fiber. The fiber is evacuated at the output, while Ar gas is injected from the input, therefore creating a differential pumping scheme.

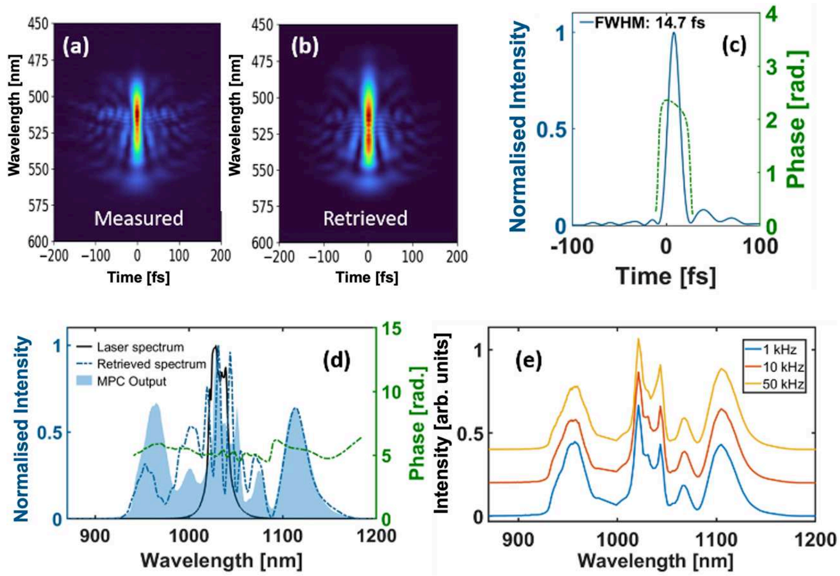

FIG. 2. Characterization of MPC output. (a) Measured and (b) retrieved FROG map. (c) Retrieved temporal profile (blue) and phase (dashed green). (d) Spectral comparison between laser spectrum (MPC input, black) vs MPC output (filled blue) and the corresponding FROG retrieved spectrum (dashed blue) and phase (dashed green). (e) Measured MPC output spectra at different repetition rates.

The compressed pulses then pass through a variable attenuator consisting of a broadband λ/2-wave plate and a broadband thin-film polarizer (TFP). The attenuator controls the input pulse energy at the entrance of the hollow-core fiber (HCF) setup, which, together with the gas pressure, is critical to define the wavelength of the RDW emission. A mirror telescope with radii of curvature (ROC) of ROC_1 = +1000 mm for M1 and ROC_2 = −1000 mm for M2 separated by a distance of ~600 mm is mounted to match a beam diameter at $1/e^2$ of 134 × 132 μm, i.e., of ~66% of the 200 μm HCF core diameter. The 1.7-m long HCF is filled with a negative pressure gradient (high pressure at the input of the fiber, vacuum at the output of the fiber) of Ar gas as a nonlinear medium, and the gas pressure is adjusted to tune the phase matching condition of the RDW for a given wavelength. The total effective pulse energy available after the pulse compression stage is 214 μJ, resulting in a maximum energy of ~180 μJ at the fiber input, which is used to drive the RDW emission. The total transmission through the fiber is measured to be ~40% at low gas pressure (<300 mbar) and decreases slightly to 38% at high gas pressure (>500 mbar) for all three repetition rates. This corresponds to a coupling efficiency of over 78% after taking into account the linear transmission of the fiber (46% at 1030 nm). A one inch 0.5 mm thick CaF_2 window is used to seal the HCF output after differential pumping system. Figure 3 shows the tunability range of the dispersive wave at different repetition rates as a function of different argon gas pressures at the input to the gradient configuration. The generated spectra are acquired with an integrating sphere connected to the Avantes (*AvaSpec-2048XL*) spectrometer. The combined acquisition setup has been calibrated using two separate lamps (Bentham CL3 and CL6).

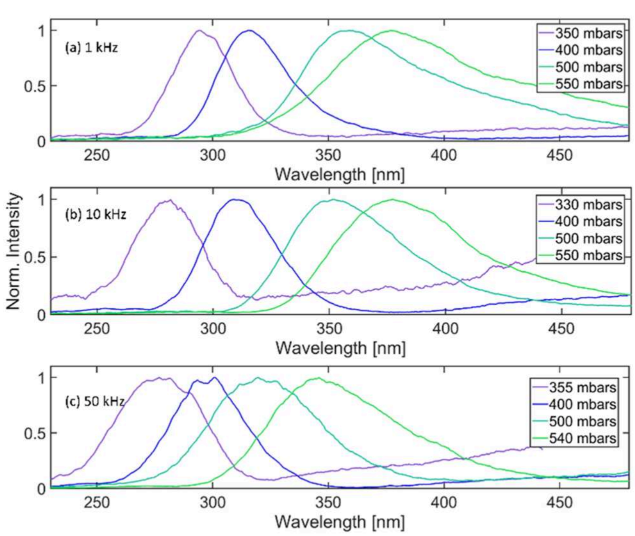

FIG. 3. Tunable UV spectra measured after the HCF as a function of input gas pressures at different repetition rates (a) 1 kHz, (b) 10 kHz, and (c) 50 kHz. Shorter wavelengths are generated with higher pump pulse energy and lower gas pressures.

In this experiment, the pulse-to-pulse fluctuation of the MPC output, incorporating the compressor and coupling optics for the fiber, was measured to be 0.12% at 50 kHz. The RDW stability is expected to be equal to or even superior to the one of the driver lasers, as thoroughly documented in Ref. 21. However, as the repetition rate of the laser is increased to 50 kHz, the coupling into the HCF becomes less stable. This observed instability can be attributed to thermal-induced fluctuations in beam pointing, which could be circumvented by installing an active beam pointing stabilization system. In addition, further instabilities may arise from the photoionization-induced gas heating and subsequent inter-pulse thermal dynamics as observed in microstructured fibers.[22]

The length of the hollow core fiber (HCF) for RDW generation is calculated by the soliton self-compression length encoded by the

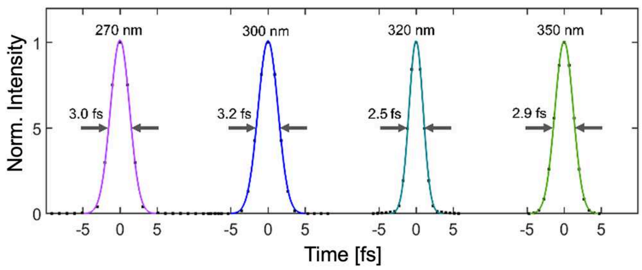

FIG. 4. Calculated transform limited temporal profiles of generated UV light at 50 kHz in HCF with FWHM duration and central wavelength indicated.

fission length.[12,23] The scaling rule for a fixed driving wavelength, which approximately defines the RDW, can be written as

$$L_f \propto \frac{\tau a^2}{\sqrt{I_0}},$$

where a is the core radius of the HCF, τ is the pulse duration of the driving field, and I_0 is the intensity. Since the core of the HCF cannot be too small to utilize all the available energy and to avoid ionization,[12] the duration of the driving laser must be short to keep the overall setup compact. However, for RDW generation, our driving laser pulses are longer than those using 800 nm lasers,[12,23] which can be circumvented by the intrinsic wavelength scaling of the RDW. For the longer driving wavelength of 1030 nm, higher gas pressure is required for a fixed HCF core size to phase match the emitted RDW at a given wavelength. Increasing gas pressure increases the nonlinearity in the HCF, while at the same time, the negative contributions to the group velocity dispersion (GVD) from the hollow capillary become more pronounced at longer wavelengths, as explained in Ref. 15.

The RDW range generated by the HCF extends from 270 nm at low gas pressure (330 mbar) to ~350 nm at high gas pressure (540 mbar). As the RDW phase matching pressure increases for longer wavelengths, the nonlinearity also increases, requiring lower pump energies to drive clean RDW emission while avoiding modulational instability and nonlinear mode coupling.[23]

To measure the pulse energy of the generated RDW emission from the soliton at the output of the capillary, four spectral separators (*Optoman PAN2532*) with reflectivity >70% in the wavelength range of 210–360 nm were used. At 50 kHz, UV pulse energy measured for the four RDW wavelengths shown in Fig. 4 remained above 1 μJ. The maximum UV energy of 1.2 μJ was measured at 270 nm, taking into account the losses introduced using the spectral separators. The Fourier transform of each RDW spectrum is performed, resulting in a transform limited pulse duration with FWHMs of 3.0 fs at 270 nm and 2.9 fs at 350 nm wavelength as shown in Fig. 4. One of the major advantages of generating ultraviolet pulses through the RDW emission process is that the resulting duration is nearly equal to the transform limit, thereby obviating the need for additional compression. However, direct measurement of these broadband pulses at these wavelengths poses significant challenges. Nonetheless, earlier studies have demonstrated that the pulse duration measured is nearly equivalent to transform-limited duration without compression for the entire spectral tuning region.[14]

In summary, we have demonstrated a multi-kHz tunable UV source generated through RDW in a hollow core fiber driven using an Yb laser source compressed via a single stage MPC. In comparison with capillaries, MPCs provide more favorable peak-power vs system length scaling properties, especially if large compression factors are considered. Our results show a tunable source of UV light pulses with μJ-level energy and up to 50 kHz repetition rate, while the spectral shape remains invariant to the average power scaling. The high losses observed in the separation of the RDW from the fundamental can be circumvented by improving the coating design of the spectral separators. This table-top setup provides high-throughput and repetition-rate, overcoming the temporal resolution and signal-to-noise ratio limitations of state-of-the-art UV setups. When scaling the power of the laser source, our approach will constitute a necessary step to preserve compactness. This work contributes to the development of the next generation of ultrafast spectroscopy measurements with tunable wavelengths in the DUV region to study time-resolved dynamics in bio-relevant molecules.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Laura Silletti and Ammar bin Wahid contributed equally to this work.

Laura Silletti: Formal analysis (equal); Investigation (equal); Validation (equal); Visualization (equal). **Ammar bin Wahid**: Formal analysis (equal); Investigation (equal); Validation (equal); Visualization (equal); Writing – original draft (equal). **Teodora F. Grigorova**: Formal analysis (equal); Investigation (equal); Writing – review & editing (equal). **Lorenzo Pratolli**: Formal analysis (equal); Investigation (equal); Validation (equal); Visualization (equal). **Ana Oliveira e Silva**: Formal analysis (equal); Investigation (equal); Validation

(equal); Visualization (equal). **Nans Hermellin**: Formal analysis (equal); Validation (equal). **Terry Mullins**: Supervision (equal); Writing – review & editing (equal). **Andrea Trabattoni**: Writing – review & editing (equal). **Christoph M. Heyl**: Writing – review & editing (equal). **Christian Brahms**: Conceptualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Vincent Wanie**: Conceptualization (equal); Funding acquisition (supporting); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **John C. Travers**: Conceptualization (equal); Funding acquisition (supporting); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **Francesca Calegari**: Conceptualization (equal); Funding acquisition (lead); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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