Generation of broadband circularly polarized deep-ultraviolet pulses in hollow capillary fibers

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We demonstrate an efficient scheme for the generation of broadband, high-energy, circularly polarized femtosecond laser pulses in the deep ultraviolet through seeded degenerate four-wave mixing in stretched gas-filled hollow capillary fibers. Pumping and seeding with circularly polarized 35 fs pulses centered at 400 nm and 800 nm, respectively, we generate idler pulses centered at 266 nm with 27 µJ of energy and over 95% spectrally averaged ellipticity. Even higher idler energies and broad spectra (27 nm bandwidth) can be obtained at the cost of reduced ellipticity. Our system can be scaled in average power and used in different spectral regions, including the vacuum ultraviolet.

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High energy and broadband circularly polarized pulses of ultrashort duration in the deep ultraviolet (DUV, 200-300 nm) and vacuum ultraviolet (VUV, 100-200 nm) are a crucial tool for time-resolved spectroscopy of chiral molecules and nanocrystals [1–3], attosecond pulse generation and metrology [4], circularly polarized high harmonic generation [5] and optically pumped ultrafast magnetism studies [6]. Such pulses are most commonly generated in a two-step process: a nonlinear up-conversion stage in a bulk medium to generate linearly polarized UV pulses followed by a polarization conversion stage using quarter-wave plates or shallow-incidence reflection from metal surfaces [7], reported with low (0.6%) energy conversion efficiency and few-cycle input pulse requirements [8]. The order of the generation steps is dictated by the fact that efficient frequency conversion to the DUV in nonlinear crystals requires birefringent phase-matching, precluding direct up-conversion of circularly polarized laser pulses. This approach has two important limitations: low bandwidth and difficulty in reaching the VUV. Bulk nonlinear crystals have narrow up-conversion bandwidth [9] and strong material dispersion, limiting the achievable pulse duration. While gases have much lower dispersion, their lower density severely limits the conversion efficiency [10]. Material dispersion also prevents achromatic phase retardation in the DUV and VUV, with currently available birefringent phase plates inducing non-uniform phase retardation for different spectral components.

In this work, we extend our previous investigations of efficient frequency up-conversion to the DUV based on seeded four-wave mixing (FWM) in stretched gas-filled hollow capillary fibers (HCF) [11]. We demonstrate for the first time, to the best of our knowledge, the direct generation of broadband, ultrashort, circularly polarized DUV pulses (0.95 ellipticity) with pulse energies above 25 µJ and a pump-to-idler energy conversion efficiency of 27%. The FWM scheme does not require birefringent phase-matching. Therefore, we can pump and seed the process with independently converted circularly polarized pulses at 400 nm and 800 nm, where high-quality achromatic phase retarders are available, and directly generate circularly polarized pulses in the DUV.

The use of hollow waveguides overcomes the aforementioned limitations of bulk media as it does not require a polarization conversion stage for the DUV pulse. It also provides octave-spanning phase-matching bandwidth while dramatically enhancing the light-matter interaction as compared to gas-based techniques in free space. The excellent pump-to-idler frequency up-conversion efficiency provided by FWM schemes has been previously demonstrated with linear polarization in hollow-core photonic crystal fibers (PCFs) [12] and in HCFs [13–15], reaching a record high of 50% [11]. While both PCF and HCF allow for precise control over the generated spectral bandwidth and phase [16], stretched HCFs are a fully energy-scalable platform, suitable for pulse compression [17], FWM interactions [11], and optical soliton dynamics [18], among other applications. They offer broad transmission free from resonances and birefringence [19] due to the absence of any micro-structured lattice, which allows access to the VUV and increases the waveguide lifetime.

In our experiments, the output of a 1 kHz, 35 fs FWHM (full width at half maximum), 800 nm Ti:sapphire laser source (Fig. 1) is split into two replicas (BS1). The first acts as the fundamental seed (signal) while the second is frequency-doubled to 400 nm in a 100 µm thick β-Barium borate (BOO) crystal and acts as the degenerate pump. Both pump and seed beam lines are equipped with a motorized half-wave plate (HWP) and a thin-film polarizer (TFP) for power control and conversion to vertical linear polarization with a polarization extinction ratio of 10^{-4}. A quarter-wave plate (QWP) is placed in each beam line to convert to circular polarization. The pump QWP is slightly tilted off of normal incidence to optimize phase retardation at the central wavelength of the pump. Concave mirrors (CMs) are used as focusing optics to avoid temporal dispersion and chromatic aberration, and a dichroic mirror in the pump line filters out the fundamental 800 nm pulse. The converging beams are combined by a dichroic beam-splitter (BS2) and then propagate to the input of the capillary fiber. A motorized delay stage placed in the seed line enables us to perform delay scans and optimize the temporal overlap of the two pulses.

Stretched hollow capillary fibers provide a substantially improved performance compared with rigid fibers [17, 18, 20]. We use our own stretching technique to position a 1.3 m long HCF with a 150 µm core diameter in a sealed gas system that allows for both evacuation and gas fill. Optical access to the waveguide and pressure sealing is provided by
The conservation of the spin angular momentum requires the two input pulses to have the same state of polarization (SOP) for FWM to occur [23]. For the case of circular polarization, this corresponds to both input pulses being left or right circularly polarized. If the input pulses have opposite-circular SOP, no FWM takes place and idler generation is inhibited (Fig. 2b) despite spatio-temporal overlap of the input pulses. The ellipticity of the output UV pulse shown in Fig. 2 is 0.95, very close to purely circularly polarized, with a flat, uniform polarization profile (blue line in Fig. 2d) across its spectrum (black line). As shown by the difference between the black and grey dashed lines in Fig. 2d, the frequency conversion process reshapes the spectrum of both the pump and seed pulses due to Kerr-induced nonlinear propagation phenomena such as self- and cross-phase modulation (SPM, XPM) [24, 25] in addition to strong pump energy depletion (∼ 40%, Fig. 2a). Despite these spectral shifts, we measure pump and seed output ellipticities of 0.93 and 0.95, a negligible change from their input values of 0.945 and 0.95.

The driving energies shown in Fig. 2 are chosen to simultaneously optimize the conversion efficiency and polarization purity of the idler pulse. Increasing the pump and seed energies increases the strength of nonlinear effects. For imperfectly circularly polarized light (ε ≠ 1.0), it has previously been shown that nonlinear phenomena can modify the polarization state by inducing birefringent nonlinear phase shifts [26]. This results in non-uniform polarization profiles and decreased ellipticities. This polarization evolution affects not only the two input
pulses but also the generated idler. In Fig. 3 we present the response of our FWM system when varying the energy of either the pump (left column) or the seed (right column) while keeping the other fixed at 27 µJ (seed) and 80 µJ (pump).

In the pump energy scan, a maximum CE of 27% is obtained for 100 µJ yielding 27 µJ in the DUV (the conditions shown in Fig. 2), whilst the idler energy continues to increase beyond 100 µJ pumping, reaching 34 µJ for 160 µJ pump energy. In the seed energy scan, CE and idler energy reach 30% and 34 µJ, respectively, and slowly decrease for seed energies above 120 µJ. The ellipticities of pump and seed increase with the pump energy, from 0.925-0.94 and from 0.935-0.965, respectively (Fig. 3c). The idler ellipticity remains relatively constant around 0.95 and decreases to 0.91 beyond 100 µJ pump energy, following the CE trend. In the seed energy scan (Fig. 3d), both driving pulses exhibit only minor changes in their ellipticity, with the pump ellipticity constant at about 0.93 and the seed at 0.94-0.95. The idler ellipticity, on the other hand, decreases from 0.95 at low seed energies to 0.86 at 160 µJ.

Figures 3e-j show the pump, seed and idler spectra and their corresponding polarization distributions at specific energy points of interest. From the pump scan, we select the pump energy yielding the highest idler energy (34 µJ in the DUV, obtained for 160 µJ pump energy) and from the seed scan the maximum seed energy. Strong spectral modifications of the pump (depletion) and seed (shifts associated with amplification and pump-induced XPM) are evident in both cases, as compared to their input profiles (grey dashed). The spectra associated with the strong-seeding regime (Fig. 3j) show a more dramatic enhancement of the nonlinear effects. The presence of the strong seed affects the polarization uniformity of both the pump (Fig. 3i) and the seed (Fig. 3j) as compared to the ones for the weak-seeding regime, shown in Fig. 2d, resulting in a decreased ellipticity (0.89) for the pump. The idler generated in the strong-pump regime (Fig. 3e) maintains a flat polarization profile with 0.91 ellipticity. In the strong-seeding case (Fig. 3h), a broadband (27 nm FHWM) DUV idler with 0.86 ellipticity is generated with ~30% conversion efficiency and pulse energy above 20 µJ. Although direct temporal characterization is challenging, this bandwidth is sufficient to support a 4 fs pulse duration.

The slight idler ellipticity decrease mainly observed in the strong-seeding regime cannot be solely attributed to a single phenomenon due to the coupled nature of nonlinear effects taking place in FWM. However, the nonlinear polarization evolution induced by each pulse upon itself can be isolated by launching the input beams separately in 1.8 bar of helium and measuring the output polarization. By evacuating the fibre (to avoid nonlinearity) we can also estimate the polarization of the input pulses from measurements at the fibre output, due to the negligible birefringence of the HCF and the CaF2 windows.

In Fig. 4(a, b) we compare the polarization evolution of the input pulses (in vacuum, gray circles) and under nonlinear conditions (1.8 bar He, black circles) for the pump and seed by themselves. It can be observed that both the pump and the seed exhibit negligible nonlinear polarization evolution by themselves compared to the input pulse measurements. Indeed, the pump is measured to have an ellipticity of 0.94 in both cases, and the seed exhibits a slightly decreasing ellipticity from 0.95 at low energies to about 0.92 at higher energies, similar to the input pulse trend. In Fig. 4(c, d) we show the averaged pump and seed spectra for both high (solid black) and low energy (dashed gray) with the corresponding averaged ellipticities (solid and dashed blue, respectively) for the two pressure sets. In the vacuum setup (Fig. 4c), the spectral broadening of the pump indicates the presence of nonlinearity along the beam path (possibly at the CaF2 windows due to the high intensity of the converging beams), which may explain the slight change of input seed ellipticity with varying energy (Fig. 4b). Finally, in the 1.8 bar helium set (Fig. 4d), the high-energy pulses maintain both polarization uniformity and high ellipticity values, despite the broadened spectral profiles.

In conclusion, we have demonstrated an efficient method for the generation of ultrashort DUV pulses with a uniform spectral distribution of circular (0.95 ellipticity) polarization, high pulse energy (above

![Fig. 3.](image-url)
(b) 25 µJ and 27% pump-to-idler conversion efficiency, via weakly seeded FWM in stretched HCFs. Despite the slight idler ellipticity decrease (0.86) in the strong-seeding regime due to nonlinear propagation effects, we have also generated broadband (27 nm FWHM) circularly polarized DUV spectra supporting 4 fs pulse durations. The system described can be readily adapted to other spectral regions, including the VUV, by proper selection of the driving frequencies, and scaled in average power by increasing the repetition rate.

DISCLOSURES

The authors declare no conflicts of interest.

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