A multi-shot transient-grating cross-correlation frequency-resolved optical gating (FROG) is implemented for the characterization of nanojoule-scale, few-femtosecond, deep-ultraviolet pulses. In theory, the system can characterize pulses with a bandwidth extending from below 200 nm to above 1.5 μm. It is experimentally shown that a 200 THz (50 nm) wide dispersive wave centered at 275 nm, generated in a gas-filled HC-PCF, has a temporal duration of 4 fs. The numerical simulations agree well with the experiment. The results confirm that dispersive wave emission in a gas-filled HC-PCF can be used as a novel source of ultrashort UV pulses in a range of applications, for example, ultrafast UV pump-probe spectroscopy.

Easily accessible few-femtosecond (fs) vacuum and deep-ultraviolet (UV) pulses would be an indispensable tool in many areas of ultrafast UV spectroscopy. For example, they could be used to probe molecular dynamics on ultrashort time-scales and, hence, allow the study and control of chemical reactions and the excitation of electronic resonances [1]. They could also be used for the investigation of the band structure of surface electrons in different kinds of topological insulators [2]. Few-fs deep-UV pulses are however relatively difficult to generate in the laboratory, mostly requiring millijoule scale pump pulse energies [3,4] or highly specialized pump sources [4,5], and preventing their widespread use in realistic application environments.

Coherent ultrashort pulses, wavelength tunable from 113 to 550 nm [6–9], can be efficiently generated by soliton effects in a gas-filled kagomé-style hollow-core photonic crystal fiber (kagomé-PCF) pumped by 800 nm pulses from a Ti:sapphire amplifier or 1030 nm pulses from an ultrafast fiber or a thin-disk laser. In this way, a spectrally flat vacuum-UV supercontinuum has previously been demonstrated with a bandwidth large enough to support sub-fs pulses [9]. The basic mechanism for frequency upshifting is dispersive wave (DW) emission, which occurs when part of the energy in a self-compressed soliton pulse is transferred to a higher frequency, phase-matched by a higher-order dispersion [10]. An example of such a broad soliton-driven spectrum, measured in this Letter, is shown in Fig. 1 (blue curve). The DW is located at ∼1100 THz (275 nm), with a spectral FWHM at 200 THz (50 nm) and an energy of ∼80 nJ. The ultrashort nature of the DW emission in a gas-filled kagomé-PCF originates from self-compression and self-steepening of the pump soliton, leading to the formation of an optical shock. At the point of maximum compression, pulse durations below 1 fs can be reached [9]. Since the DW is emitted directly after the shock formation, its duration is expected to be ultrashort. Despite strong supporting evidence from comprehensive numerical modeling [9], this has so far not been experimentally confirmed. In this Letter, we use a device based on transient-grating cross-correlation frequency-resolved optical gating (TG-XFROG) to measure the temporal properties of the deep-UV DW pulses with a high sensitivity over the wavelength range of 200 to 1500 nm. The results show that the duration of the deep-UV DW pulses is below 4 fs.

Recently, a two-photon autocorrelator was used to measure the temporal duration of a DW emitted at 270 nm [11]. The autocorrelation does not, however, yield information about the temporal structure and phase, and is not sufficiently reliable for fully confirming the temporal dynamics of DW emission. Spectral phase interferometry for direct electric field reconstruction [12] and frequency resolved optical gating (FROG) [13] are the most...
popular and convenient techniques for full characterization of ultrashort optical pulses. Among the different FROG arrangements, those based on second-harmonic generation (SHG) and other $\chi^{(2)}$ processes are in widespread use for characterizing weak (few pJ) pulses in the infrared and visible [14]. SHG-based FROG, however, cannot be used in the UV due to the difficulty of transmitting and measuring the generated vacuum-UVC wavelengths. Another drawback is the difficulty of maintaining phase matching over the wide bandwidths associated with the ultrashort pulses. These difficulties can be avoided if the $\chi^{(3)}$ processes are used. Transient-grating (TG) [15] and self-diffraction [16] FROG geometries make use of two reference pulses that overlap spatially and temporally in the nonlinear sample, resulting in the creation of an interference pattern that generates (via the Kerr effect) a refractive index grating [17]. The measurand pulse is diffracted by the grating, generating a signal beam. The spectrum of the signal pulse is recorded as a function of the delay between the measurand and the reference pulses, allowing robust reconstruction of the field of the measurand pulse [14]. Usually, the TG-FROG operates with three beams of the same frequency, yielding perfect phase matching in a box-car arrangement [18]. Importantly, the signal is in the same spectral region as the measurand pulse. Both the self-diffracted (SD) and the TG-FROG have been used to measure high-peak-power ultrashort pulses in the deep-UV [3–5]. In TG cross-correlation FROG (TG-XFROG) [19], the two reference pulses can lie in a different spectral region from the signal. This allows the use of energetic near-IR pulses from the pump laser (e.g., a Ti:sapphire amplifier) to characterize weak (few nanojoules) UV pulses, the only requirement being a broad enough phase-matching bandwidth. (Phase matching is no longer automatically satisfied in this case.)

The experimental layout is shown in Fig. 2(a). The deep UV measurand pulses are generated in a kagomé-PCF (Fig. 1 inset). The fiber (F) ends are glued into two gas cells (GC1 and GC2) so that the chambers are connected to each other only by the fiber. The gas cells are connected to a vacuum or a gas supply and can be filled to a maximum pressure of 50 bars. In the experiment, the second cell (GC2) was filled with 27 bar of Ne, and the first cell (GC1) was continuously evacuated to the mbar level. The absence of self-focusing in GC1 creates stable launch conditions and avoids damage to the fiber end-face. Pulses of a duration of 40–50 fs, a wavelength of 800 nm, and a repetition rate of 1 kHz from a Ti:sapphire amplifier (Coherent, Legend Elite) were coupled, using an achromatic lens (AL), into the fiber through a 2 mm thick fused silica window mounted in GC1. To deliver the generated UV light, the second gas cell (GC2) was sealed with a 1.5 mm thick magnesium fluoride window. The divergent output beam was collimated using an off-axis parabolic aluminum mirror (APM1) and reflected off three dielectric separators (UVS, from Layertec) to isolate the UV part of the spectrum. The bandwidth of the separators is ~70 nm with a central wavelength of 280 nm. The signal then passes slightly above the prism mirror (PM) and is focused by the aluminum parabolic mirror (APM2) into a fused silica plate (FS).

Since fused silica has an essentially instantaneous Kerr response for the pulse durations considered, a TG signal will only appear if the reference and measurand pulses overlap in both space and time. The reference pulse is derived directly from the Ti:sapphire amplifier used to pump the kagomé-PCF. It passes through a piezo-driven delay stage (DS1) to control its position relative to the measurand pulse, and is then equally divided at the beam splitter BS (50/50). One part is reflected off the prism mirror (PM) and focused by APM2. The second part is transmitted through an infrasil window so as to introduce the same amount of dispersion as BS (50/50) and then passes through a second piezo-driven delay stage (DS2) before being combined with the measurand pulse and the first reference pulse at PM and APM2. After interacting in the fused silica plate (FS, 25 μm thick), all the beams are blocked except for the signal beam, which is collimated and refocused using the parabolic mirrors APM3 and APM4 onto the CCD spectrometer (Ocean Optics Maya Pro).

The XFROG beam arrangement (a standard boxcar geometry is not required) is shown in Fig. 2(a). After being reflected and focused by a parabolic mirror, the reference pulses propagate in the horizontal plane parallel to the optical table with angular offsets $\alpha_R = \pm 1.5°$ from the normal to the FS, producing interference fringes with spacing $\Lambda = 15.3 \mu m$ and two spots (R, marked red) in the plane of the beam-blocking iris. The measurand pulse (M) propagates downward out of the horizontal plane at an angle $\alpha_M = 1.5°$, forming a spot (M, marked purple) slightly below and exactly half-way between the two reference spots. Many SD orders appear to the left and right of the reference pulses, and the two diffracted orders (S and D) of the measurand pulse are also marked [the light purple dots in the top left corner of Fig. 2(a)]. The signal (S) is taken from one of these diffracted orders.

![Fig. 2.](image-url)
In contrast to the TG-FROG, the TG-XFROG signal is not perfectly phase-matched. For the geometry described above, however, the coherence length \( L_c \approx \lambda^2 n_l / \lambda \), where \( n_l \) is the index of silica, works out to be 1.7 mm at \( \lambda = 0.2 \) μm and 217 μm at \( \lambda = 1.5 \) μm. Since \( L_c \) is much greater than the plate thickness (\( L = 25 \) μm), the entire spectral range is phase-matched. The relatively large grating pitch [see the sketch in Fig. 2(b)] means that diffraction occurs in a Raman–Nath or thin-grating regime [17], as confirmed by the multiple diffraction orders seen in the experiment. In this regime, the measurable conversion efficiency into the two first diffracted orders (S and D) is given by the expression [17,20]

\[
I_S/I_M = I_D/I_M = f_1^2(4\pi n_2I_R L/(3\lambda)) \approx (2\pi n_2I_R L/(3\lambda))^2,
\]

where \( I_R \) and \( I_M \) are the intensities of the reference and measurand pulses, and \( n_2 \) is the nonlinear index of fused silica. If 60 fs reference pulses of up to 6 μJ of energy are used, the TG-XFROG diffraction efficiency is greater than \( 10^{-4} \) from the deep UV to the near IR [Fig. 2(c)], reaching almost 1% below 300 nm. Indeed, in the experiments reported here, the reference pulse energy had to be significantly attenuated so as to avoid distortion of the signal beam by multiple diffractions.

In comparison to a standard FROG, which typically is not sensitive enough to measure all wavelengths, the use of energetic reference pulses in the cross-correlation geometry provides an easily detectable signal across the entire bandwidth, despite the efficiency drop at longer wavelengths [Fig. 2(c)]. The main disadvantage of the XFROG geometry is the angular dispersion of the different signal wavelengths. This can be side-stepped if, for example, an integrating sphere is used. In the following experiments, however, we directed attention to a sub-octave-spanning portion of the UV spectrum, simply by coupling the signal directly into the UV spectrometer. Overall, the TG-XFROG arrangement can be very sensitive, allowing the characterization of extremely broadband (multiple octave) and very weak (few nanojoules) ultrashort pulses.

In XFROG, reliable retrieval requires the reference pulse to be fully characterized, for example, using SHG-based FROG. In our case, this is conveniently done by scanning the relative delay of the reference pulses [using stage DS2 in Fig. 2(a)] and collecting the FROG signal produced by self-diffraction. The SD beams are denoted by orange colors [Fig. 2(a), top left] and appear on the both sides of the reference pulses. Since the SD signal was always present under the same conditions as those needed to produce the measurand TG signal, a separate setup was not necessary to characterize the reference pulses, greatly reducing the overall experimental complexity.

Figure 3(a) shows the SD-FROG trace of the reference pulse. The temporal resolution was 1.3 fs, and the spectral resolution 0.4 nm. Although SD scattering is not phase-matched [14], the coherence length is longer than the thickness of the FS plate so that a good signal can be obtained provided the reference pulses are energetic enough (6 μJ in this case). Although the angle between the reference pulses is relatively large (3°), and the SD signal is isolated by the iris, the trace was contaminated by reference light scattered into the spectrometer. To reduce this effect, the delay was set to a position where there is no temporal overlap between the reference pulses and the measurand; the resulting signal was recorded and used for background subtraction.

Figure 3(b) shows the retrieved spectrogram calculated using commercial software (Femtosoft Technologies), with a FROG error of 0.003 [13]. Because the SD-FROG is unambiguous in time, a negative chirp is clearly recognizable in the trace. The exact amount of the chirp can be derived from the temporal and spectral phases of the reconstructed pulse [Figs. 3(c) and 3(d)]. The temporal FWHM is 60 fs with a FWHM spectral bandwidth of 12 THz (25 nm). An independent spectral measurement of the reference pulse [black curve in Fig. 3(d)] was compared with the retrieved spectrum [blue curve in Fig. 3(d)]. The good agreement shows that the retrieval algorithm is accurate. The temporal resolution of XFROG is not limited by the width of the reference pulse [14], so that a longer pulse with some amount of chirp is acceptable as a reference for characterizing ultrashort UV pulses. This measured negative chirp is actually the result of optimizing the laser compressor for the almost transform-limited pump pulse at the fiber input.

Figure 4(a) shows the measured TG-XFROG trace of the deep-UV pulses emerging from the gas-filled HC-PCF, with temporal and spectral resolutions of 0.9 fs and 0.55 nm. Figure 4(b) shows the corresponding retrieved TG-XFROG trace reconstructed with an error of 0.015. There is a large and almost linear positive chirp across the pulse, the temporal FWHM of which is 280 fs [Figs. 4(c) and 4(d)]. This is the result of the propagation of the DW pulse in air and the optical elements, which are highly dispersive in the deep UV. By numerically subtracting the known dispersion of the window and the air path, the pulse parameters at the fiber end-face were calculated. The results [Figs. 4(e) and 4(f)] show that the emitted deep-UV pulse has a transform-limited temporal FWHM of 4 fs, confirming the predictions of the few-fs nature of the DW. The temporal intensity distribution has an oscillation characteristic of a pronounced cubic spectral phase. We believe that this arises from residual higher order dispersion in the optical elements (e.g., the UV separators).

We have also numerically modeled the experimental results using the multimode, carrier-resolved, unidirectional propagation equation reported in [21]. The model includes dispersion, the Kerr effect, self-focusing, and photoionization. We used the SD-FROG measurements from Fig. 3 to obtain the correct input electric field for the numerical simulations. As mentioned above,
the main beam from the Ti:sapphire amplifier is split into the reference arm for TG-XFROG and the arm pumping the kagomé-PCF [not shown in Fig. 2(a)]. The wavelength-dependent phase shift, introduced by each optical element between the input face of the kagomé-PCF and the point where the SD-FROG measurements were performed, was calculated. The resulting pump pulse, which we used in the numerical simulations, had a FWHM of 44 fs.

Figure 1 compares the experimental and simulated spectra, showing good agreement when the pump energy was set 2 μJ lower in the simulations than in the experiment. We attribute this to a strong loss band in the kagomé-PCF between 400 and 500 THz, which is not accounted for in the numerical simulations and would result in increased pump energy requirement. This is supported by the fact that, in the spectral region around the pump frequency, the higher frequency self-phase modulation sideband is suppressed in the experiment. This may also explain the small discrepancy in the DW spectral position, since the phase-matched DW frequency is reduced if the pulse peak power is decreased [22]. Figures 5(a) and 5(b) compare the experimental and simulated spectrogram of the DW after numerical back-propagation. The modeling yields a FWHM pulse width of 3 fs and has no residual quadratic chirp. This discrepancy is due to the bandwidth reduction caused by the UV separators [compare the Fig. 1 blue curve and the Fig. 4(d) black curve] and their residual higher order dispersion, leading to a longer measured pulse duration. In future work, we plan to replace the separators with broadband mirrors so as to measure the full supercontinuum and fully utilize the extremely broadband capability of the TG-XFROG.

In conclusion, TG-XFROG can be used to measure broadband and relatively low-energy (∼100 nJ) UV pulses emitted from a hollow core kagomé-PCF through DW generation. DW pulses with FWHM durations of 4 fs, in reasonable agreement with numerical simulations, could be characterized. The use of high-energy reference pulses makes it possible to characterize measurable pulses of only a few nanojoules of energy and, theoretically, the TG-XFROG setup supports multi-octave bandwidths in the wavelength range from below 200 nm to beyond 1500 nm.

REFERENCES