Generation and characterization of μJ-level, 10 fs UV pulses

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Abstract: We demonstrate μJ-level 10-fs pulses in the 315-380 nm spectral range generated by broadband sum-frequency generation. The pulses are characterized using 2D spectral shearing interferometry based on difference-frequency with a visible NOPA.
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Femtosecond light pulses in the ultraviolet (UV) spectral range (λ<400 nm) are required to study fundamental chemical and biological processes in biomolecules, which exhibit strong absorption bands in this region. High time resolution and two-dimensional spectroscopy experiments [1], in particular, call for ultrabroadband, few-optical-cycle pulses. The generation of sub-10-fs UV pulses is however not straightforward, since this spectral region poses serious challenges in terms of: (i) broad pulse bandwidth; (ii) pulse energy; (iii) spectral phase handling; (iv) pulse characterization. There are no gain media that produce femtosecond pulses in the UV region, and optical parametric amplification cannot be applied due to the occurrence of two-photon absorption of the short-wavelength pump pulse; therefore, most approaches rely on nonlinear frequency conversion to shift ultrashort visible and infrared pulses to the UV range.

Frequency conversion of broadband pulses to short wavelengths needs to compromise between the competing requirements of phase-matching bandwidth and high efficiency: broadband phase matching in fact requires thin nonlinear crystals, which limit the efficiency of the process. This limitation can be overcome by achromatic phase matching [2], at the expense however of a significant complication of the experimental setup. In addition, linear dispersion by propagation in transparent media is particularly severe in the UV spectral range, and careful control of the spectral phase to achieve transform-limited (TL) pulse duration is difficult due to higher order dispersion introduced by pulse compressors. Finally, pulse characterization in the UV is complicated by the short-wavelength absorption of nonlinear materials and the unfavourable phase-matching conditions.

In this work, we introduce a simple scheme for the generation of ultra-broadband UV pulses which effectively overcomes these challenges. Our system is based on collinear sum-frequency generation (SFG) between broadband visible pulses and narrowband IR pulses, and uses a suitable phase-matching configuration to balance the trade-off between bandwidth and efficiency. This process also allows us to efficiently manipulate the spectral phase of the UV light through parametric transfer. Finally, we introduce a two-dimensional spectral shearing interferometry (2DSI) technique for the UV pulse characterization.

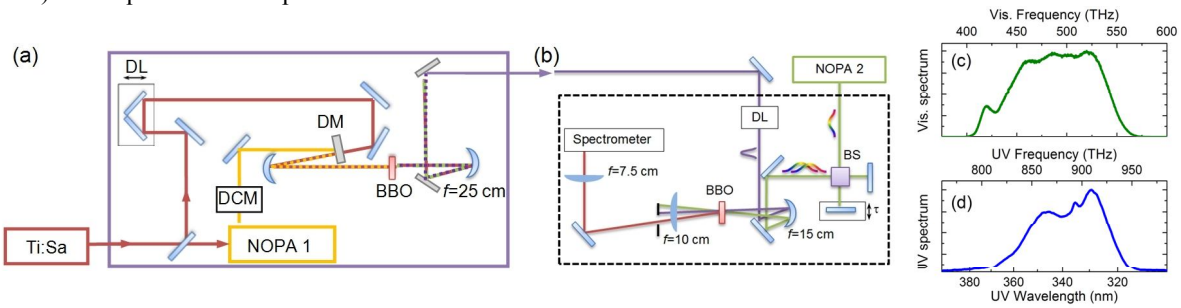

Fig. 1. (a) Experimental setup for the generation of UV pulses; DL: Delay Line; DM: Dichroic Mirror; (b) 2DSI setup based on DF generation with an external pulse; BS: cube Beam Splitter. (c) Spectrum of the Visible NOPA1; (d) Spectrum of the UV pulse

The experimental setup is shown in Fig. 1(a). The system is powered by a regeneratively amplified Ti:Sapphire laser (Libra, Coherent), which delivers 800-nm, 100-fs pulses at 1 kHz repetition rate. A fraction of the laser light drives a visible non-collinear OPA (NOPA), pumped by the second harmonic and seeded by a white-light continuum. This NOPA (NOPA1 in Fig. 1(a)) produces broadband pulses with spectrum extending from 525 to 740 nm (see Fig. 1(c)), corresponding to a TL pulse duration of 6 fs, and with energy higher than 10 J. The spectral phase of the NOPA pulses is then manipulated by a series of reflections onto a pair of dielectric Double Chirped Mirrors (DCMs). The shaped visible pulses are mixed with 30-μJ replicas of the fundamental pulses in a BBO crystal cut for SFG. The interaction Type and the polarization of the beams were chosen in order to optimize the up-

converted bandwidth; the broadest UV pulses are obtained with Type II ($e_{800} + o_{VIS} \rightarrow e_{UV}$) configuration, which has a broad acceptance bandwidth for the visible pulse and a narrow one for the 800-nm pulse, thus allowing to efficiently transfer the broad NOPA bandwidth to the UV. The BBO crystal is 50-µm-thick: this thickness is a good trade-off balancing efficiency and bandwidth of the SFG process. Due to the high energy of the visible and infrared pulses, long focal lengths are necessary in order to prevent self-phase modulation in the BBO crystal. The SFG stage provides UV pulses with energy up to 1.5 µJ. A typical spectrum is shown in Fig. 1(d): it extends from 315 to 380 nm and corresponds to 7.4 fs TL pulse duration. The spectral phase of the UV light is controlled by the mechanism of indirect phase transfer [3]: the SFG process transfers the spectral phase of the broadband visible to the up-converted light. To compensate for the positive chirp acquired by propagation to the measurement point, we impart a slight negative chirp to the visible pulses, resulting in negatively chirped UV pulses.

The measurement of the UV pulses is challenging since, due to lack of transparency, no traditional methods based on second-harmonic or SFG can be applied; for this reason, we developed a system based on difference-frequency (DF) generation with a visible gate pulse, in combination with the 2DSI technique [4], which allows full characterization of ultrabroadband pulses. The 2DSI method relies on spectral shearing interferometry with zero delay; with respect to the SPIDER technique, which encodes phase as a sensitively calibrated fringe in the spectral domain, 2DSI robustly encodes phase along a second dimension.

In traditional 2DSI, two highly chirped replicas of a broadband gate pulse are mixed with the test pulse in a nonlinear crystal, to generate two spectrally-sheared up-converted signals. The delay of one of the chirped auxiliary pulses is scanned over a few optical cycles and the spectrum of the up-converted signal is recorded as a function of this phase delay, yielding a 2D map which encodes the group delay (GD) of the test pulse. In our approach, the 2DSI signal is the DF between the UV pulse and a broadband gate provided by an auxiliary visible NOPA (NOPA2 in Fig. 1(b)). Two collinear replicas of the strongly chirped visible pulse are generated by a Michelson interferometer. They are further mixed with the UV pulses in a 10 µm thick Type-I BBO crystal, using a non-collinear focusing geometry, and the DF signal is detected by a spectrometer. One of the arms of the Michelson interferometer is equipped with a high precision translation stage, which scans the delay between the two gate pulses by +/-5 fs. A preliminary 2DSI map of a UV pulse with slightly reduced bandwidth (Fig. 2(b), solid line) is shown in Fig. 2(a). The GD (white line in Fig. 2(a)) is deduced from Fourier transform along the scanning delay and from the measured spectral shear between the two down-converted spectra. The spectral phase deduced from the GD (Fig. 2(b), dashed line) applied to the measured UV spectrum allows retrieving by Fourier transform the pulse in the temporal domain. The resulting pulse (Fig. 2(c)) has 10-fs width, very close to the 8.9-fs TL value.

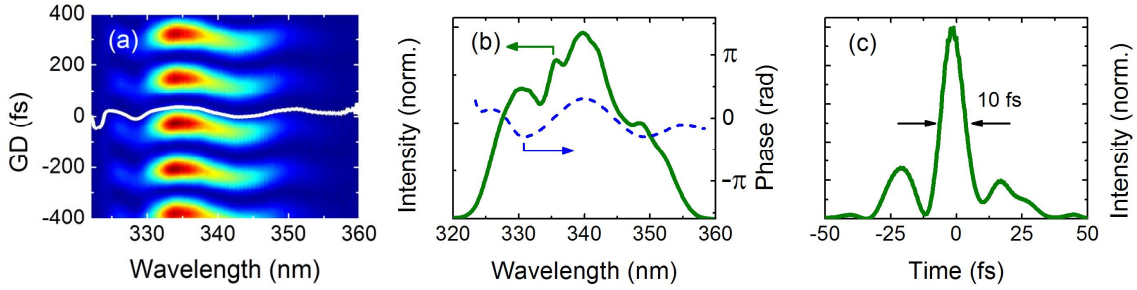

Fig. 2 (a) 2DSI experimental map and retrieved GD (white line). (b) Spectral intensity (solid) and phase (dashed); (c) retrieved pulse profile.

In conclusion, we have introduced a simple and robust scheme for the generation of UV pulses with bandwidth in the 315-380 nm region and with energy in excess of 1 µJ; the system allows easy control of the spectral phase in the UV range. In addition we demonstrated a technique for the characterization of the UV spectral phase and anticipate the generation of UV pulses with duration well below 10 fs.

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[1] J. Jiang and S. Mukamel, "Two Dimensional Ultraviolet (2DUV) Spectroscopic Tools for Identifying Fibrillation Propensity of Protein Residue Sequences", Angew. Chem. Int. Ed. **49**, 9666 (2010).
[2] P. Baum, S. Lochbrunner and E. Riedle, "Tunable sub-10 fs ultraviolet pulses generated by achromatic frequency doubling", Opt. Lett. **29**, 1686 (2004).
[3] H.-S. Tan, E. Schreiber and W. S. Warren, "High-resolution indirect pulse shaping by parametric transfer", Opt. Lett. **27**, 439 (2002).
[4] J. R. Birge, R. Ell and F.X. Kaertner, "Two-dimensional spectral shearing interferometry for few-cycle pulse characterization", Opt. Lett. **31**, 2063 (2006).