Single-step visible continuum for 2D spectroscopy from 130fs Ti:Sapph pulses via hollow-fiber

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Extended Abstract

Optical coherent multi-dimensional spectroscopy (CMDS) is a powerful technique to unravel electronic couplings and energy transfer pathways in natural and artificial light-harvesting systems [1]. The potential of CMDS experiments to unravel physical and chemical phenomena hinges on the properties of the broadband source available for experiments, such as spectral bandwidth, phase properties, and stability. In the visible, the vast majority of CMDS setups are currently based on non-collinear optical parametric amplifiers (NOPAs). However, the community has recently started exploring alternatives based on temporal self-phase modulation, such as self-guided filaments [2] and hollow-core fibers [3]. Both of these techniques are rapidly gaining in popularity. Because of the nature of the nonlinear processes involved, short-input pulses are typically preferred. However, such sources are more costly, harder to maintain and may not be available to all spectroscopists.

Here, we demonstrate a multi-dimensional optical spectrometer based on visible continuum obtained by propagating 130fs, <600µJ Ti:Sapph pulses into a 2.5m long hollow-fiber. With these low input pulse requirements, we show that the generated continuum exhibits excellent properties for multi-dimensional spectroscopy experiments: a spectrum spanning the 520nm to 700nm region, good spatial mode, 45µJ pulse energy in the visible, stability over 24 hours enabling long experiments and a self-compressed output in the visible. We further show the suitability of this source by sending the output to a 2D spectrometer based on acousto-optic modulators (AOM) for coherent pulse train generation. We subsequently perform a comprehensive vibronic analysis on the reference molecular dye Nile Blue.

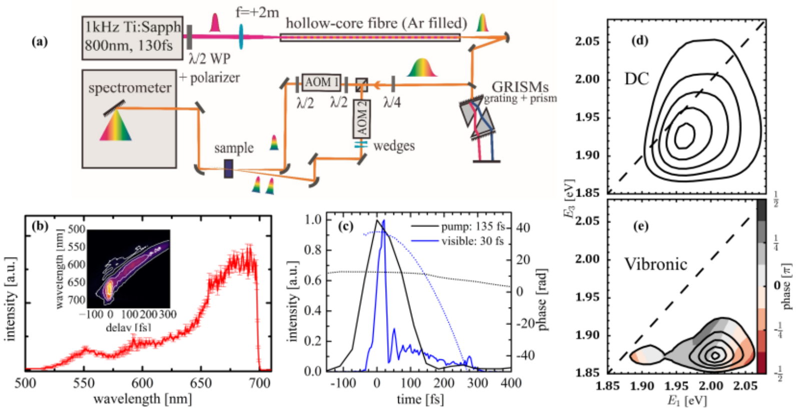

Figure 1. Adapted from Ref [4] (a) Scheme of the spectrometer (b) Fiber output after a shortpass filter at 700nm. Inset: TG-FROG trace (c) temporal profile of pulse shown in panel b (blue), exhibiting a 4.5 folds compression in comparison with input pulse (black) (d-e) vibronic analysis of Nile Blue, enabling to decouple the DC and vibronic parts in the dataset.

4. References

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