Broadband 2D Electronic Spectroscopy by Hollow-Fiber Compression

Xiaonan Ma*, Jakub Dostál, and Tobias Brixner

Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany, *xiaonan.ma@phys-chemie.uni-wuerzburg.de

We demonstrate broadband diffractive-optic-based 2D electronic spectroscopy (500–700 nm). Pulses are generated in an argon-filled hollow fiber pumped by a Ti:Sa laser and compressed to sub-7-fs duration at the sample position using dispersive mirrors. The fiber provides a clean spatial profile and thus avoids problems arising from spatial chirp.

In recent years hollow fibers filled with noble gases and gas filaments have provided new opportunities for nonlinear optics and ultrafast spectroscopy. In the case of 2D spectroscopy, many groups employ noncollinear optical parametric amplification (NOPA) where one has to face the challenge of spatial chirp. We show that using a hollow-core-fiber compressor allows broadband 2D spectroscopy with clean spatial profiles and very short pulse duration. In order to suppress phase-matching distortions from broad bandwidth [1], we employ diffractive optics, where amplitude fronts are tilted with respect to the propagation direction.

A 35-fs Ti:Sa laser (1 kHz) pumps a hollow fiber (1.05 bar Ar) to produce continuum in the range of 420–950 nm. Customized dispersive mirrors compress the pulses and remove the strongly amplitude-modulated part. The resulting spectrum is quite smooth and allows generation of 7-fs pulses as measured by TG-FROG at the sample position (Fig. 1a). The 2D setup [2] uses a double-chopper scheme for fast data acquisition and scattering removal.

We demonstrate the technique on a solution of cresyl violet. The absorptive 2D spectrum at 800 fs population time (Fig. 1b) was obtained by phasing and comparison with pump-probe spectra measured in the same setup (Fig. 1c). The population dynamics (Fig. 1d) show clear oscillations (585 cm^{-1}) that agree with previous literature results [3]. The technique is especially suited to investigate multi-chromophore systems with broad absorption spectra.

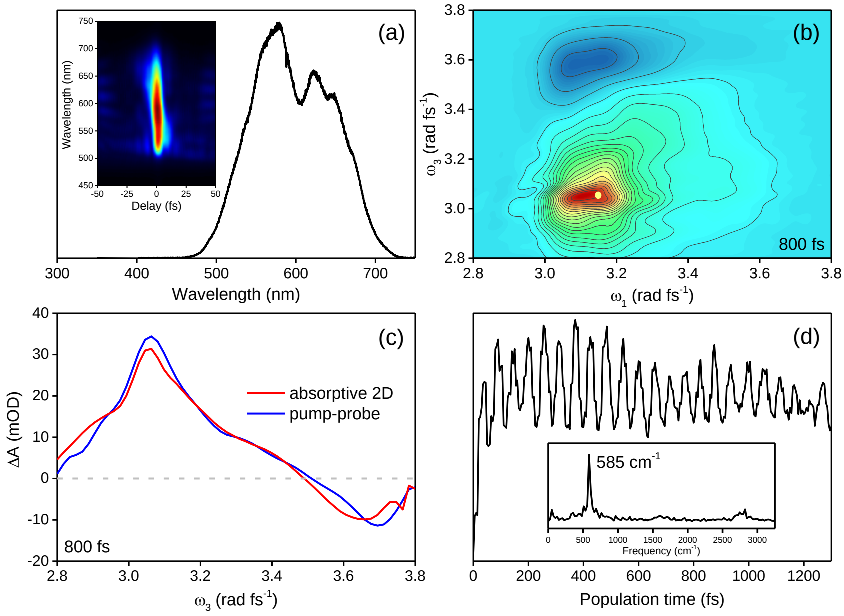

Fig. 1. (a) Spectrum at sample position and TG-FROG trace measured in the same setup. (b) Absorptive 2D spectrum of cresyl violet at 800 fs population time. (c) Comparison of projection of purely absorptive 2D spectrum (red) with pump-probe spectrum (blue). (d) Dynamics at yellow point of absorptive 2D spectrum and inserted Fourier-transformed power spectrum of corresponding oscillations giving an intense peak at 585 cm^{-1}.

[1] M. K. Yetzbacher *et al.*, J. Chem. Phys. **126**, 044511 (2007)
[2] U. Selig *et al.*, Opt. Lett. **35**, 4178 (2010)
[3] B. Spokoyny *et al.*, Opt. Lett. **40**, 1014 (2015); V. I. Prokhorenko *et al.*, Opt. Express **17**, 9764 (2009)