Wavelet filter for Femtosecond Stimulated Raman Spectroscopy: a new approach brings new horizons

Miroslav Kloz[1,2], Jörn Weissenborn[1], Yusaku Hontani[1], John T.M. Kennis[1]

[1] *Department of Physics, VU University Amsterdam, Boelelaan 1081HV, Amsterdam, The Netherlands, m.kloz@vu.nl*
[2]*Institute of Physics ASCR, v.v.i. (FZU), ELI-Beamlines Project, 182 21 Prague, Czech Republic*

It is now more than 50 years since the discovery of stimulated Raman phenomena and more than 15 years since establishing the femtosecond stimulated Raman spectroscopy (FSRS) technique. Nevertheless, stimulated Raman measurements as an analytical probe have not become a widespread and universal tool, as major problems persisted through unresolved issues of parasitic signals and insufficient sensitivity of applied detection techniques.
Here, we attempt to solve both issues simultaneously by a simple but radical shift in the experimental approach. The traditional way is to generate FSRS signal by a single narrowband picosecond pulse. Such approach is easiest to implement, but in several independent ways not practical for actual Raman data acquisition. We developed a new approach where the FSRS signal is generated by pseudorandom waveforms instead of by a single narrowband pulse. The acquired Raman spectra are then convoluted by the applied waveform, and based on the knowledge of the waveform they can be unambiguously deconvoluted with no loss of spectral resolution. The power of the technique is dual. First, the deconvolution works as a very efficient filter that rejects parasitic signals and offsets automatically and with high fidelity, doing away with human bias in baseline estimation. The second gain is hidden in the delocalization of the signal over a broad interval on the detector array, greatly reducing the so called “fixed pattern noise”. In fact, very similar approaches have for decades been a workhorse in many technologies such as Wi-Fi data transfers. With the Wavelet-Resolved FSRS (WR-FSRS) we managed to record femtosecond transient Raman spectra to a sensitivity approaching units of µOD in only tens of seconds of accumulation time. To our best knowledge WR-FSRS increases the resolving power of FSRS experiments at least one order of magnitude while making data processing entirely automatic, rendering the experiment accessible to researchers without FSRS expertise. Based on the data we acquired, we believe that WR-FSRS can be a breakthrough in the field time-resolved Raman spectroscopy.

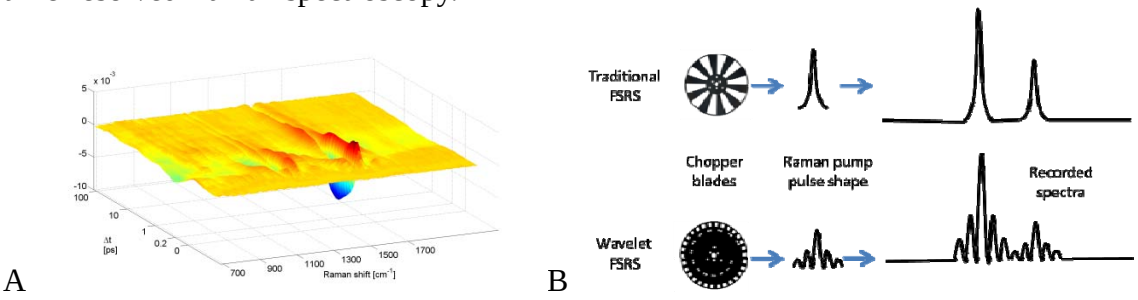

Fig. 1: A: Ultrafast vibrational dynamics of the peridinin-chlorophyll protein (PCP), aphotosynthetic light-harvesting complex of red algae. Each time-resolved FSRS spectrum is the result of only 4 seconds of data accumulation, resulting in baseline noise of ~100µOD. The entire experiment results from only 5 minutes of acquisition time on 1 kHz laser system. B: Wavelet generation through a specially designed chopper blade allows to implement WR-FSRS with minimal extra costs in comparison to traditional FSRS, as no programmable pulse shaper is needed.