Can We Throw Away Our OPA? Two-Dimensional White Light Spectroscopy at 100 kHz

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We report on a two-dimensional white light spectrometer using a 100 kHz Spirit laser, pulse shaper, and supercontinuua for both pump and probe. We show that high repetitions rates coupled with shot-to-shot delay scanning gives sufficient signal-to-noise such that spectra can be collected rapidly on a variety of interesting systems.

White light has seen extensive use in nonlinear spectroscopy, most commonly as a broadband probe pulse in pump probe spectroscopy. Our lab has recently demonstrated that white light superconinuua generated in YAG windows can be used as both the pump and probe in two-dimensional white light (2D WL) spectroscopy of carbon nanotubes[1]. Carbon nanotubes are strong absorbers and thus provide good signals despite the weak intensity of a supercontinuum pump. In this abstract, we report a second-generation 2D WL spectrometer in which we implement a 100 kHz laser system (Spectra Physics Spirit) and 4f pulse shaper to modulate the pump pulses on a shot-to-shot basis. Our pulse shaper gives phase control over the pump pulses which allows for rapid delay scanning, phase cycling, and pulse compression. It has been previously shown that shot-to-shot data collection at 100 kHz improves signal-to-noise (S/N) over point averaging in transient absorption experiments[2]. Here, we show that there is an additional improvement that is accompanied by the Fourier Transform when collecting 2D spectra (Figure 1). Signals are measureable at 100kHz that are not observable at 1 kHz even when averaging 100 times longer. The S/N is superior due to rapidly scanning the delays, which moves the data away from the noise spectrum. Slow scanning methods that use translation stages do not show this improvement. The improvement in S/N opens the door for experiments using a weak continuum pump. We demonstrate our next-generation 2D WL spectrometer on several interesting systems in the chemical and energy sciences, calling into question the need for OPAs.

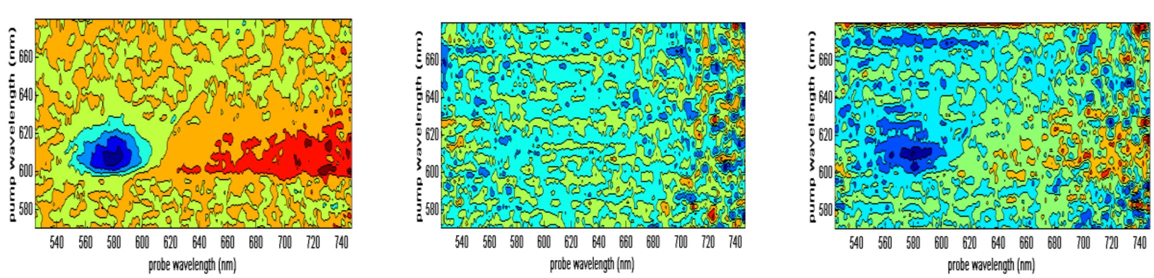

Figure 2. 2D WL spectra collecting using a supercontinuum pump when scanning delays at 100 kHz versus 1 kHz. (left) 2D spectra collected at 100 kHz for 54 sec acquisition time (10,000 averaged spectra); (center) 1 kHz, 54 sec acquisition time (100 averaged spectra); and (right) 1 kHz, 90 min acquisition time (10,000 averaged spectra). The comparison between 100 kHz and 1 kHz spectra illustrates the drastic improvement in signal-to-noise that occurs when delay scanning at high repetition rates, making possible data collection with a weak pump source.

[1] R.D. Mehlenbacher *et al.*, Nat. Comm. **6**, 6732 (2015)
[2] F. Kanal *et al.*, Optics Express **22**, 16965 (2014)