Measuring fluorescence-like pulses using Optical-Parametric-Amplification (OPA) XFROG

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Knowledge of the time-dependent intensity and phase of luminescence in ultraweak fluorescing biological molecules would yield important information about molecular dynamics, not available from the simple spectrum or intensity. Unfortunately, its measurement is very difficult: such ultrafast luminescence is extremely weak, complex, and broadband. It also has poor spatial coherence and random absolute phase, which defeat interferometric methods. Frequency-Resolved Optical Gating (FROG) and the usual Cross-Correlation FROG (XFROG) using Sum Frequency Generation (SFG) or frequency-upconversion lack the sensitivity to make such measurements.

We've recently shown that a new type of XFROG that uses the nonlinear process of Optical Parametric Amplification (OPA) solves this problem beautifully. OPA not only gates the pulse in time (as required for XFROG), but *it also amplifies it by up to* 10^6 *without distorting the phase*. And use of a non-collinear geometry easily yields bandwidths of ~100 nm. Also XFROG in general ignores the irrelevant absolute phase and spatial incoherence and can easily measure extremely complex pulses. The only requirements for OPA XFROG are that the gate pulse be measurable, bluer than, shorter than, synchronizable with, and much more intense than the pulse to be measured —the precise requirements for the pulse that excites the luminescence in the first place!

Here we demonstrate OPA XFROG for broadband, spatially incoherent, complex, extremely weak pulses with random absolute phase using an attenuated and filtered white-light continuum. We gate the continuum with a variably delayed 5.8-μJ second harmonic of a Ti:Sapphire amplified pulse in a 1- or 2-mm-thick BBO OPA crystal in a non-collinear geometry. Measuring the parametrically amplified continuum pulse spectrum vs. delay yields the OPA XFROG trace. The pulse is then retrieved using the XFROG algorithm—modified to incorporate this new nonlinearity: the gate function is now $G(t) = \cosh[g\,|E_{gate}(t)|]$, where g is the parametric gain. For comparison, we measure the same continuum pulse (but without attenuation) using the less sensitive technique of SFG XFROG. Fig. 1 shows measured OPA XFROG traces for an 80 fJ continuum pulse.

We have also extended OPA XFROG to very large bandwidths and extremely low pulse energies. An OPA XFROG measurement of broadband continuum with ~100 nm bandwidth and attenuated to ~ 50 fJ is shown in Fig. 2. Spatial incoherence from multiple filaments in the continuum generation and fluctuations in white light continuum due to pulse-to-pulse intensity variations wash out structure in the measured trace, but the XFROG algorithm still sees the structure in the pulse, which is expected for broadband continuum.

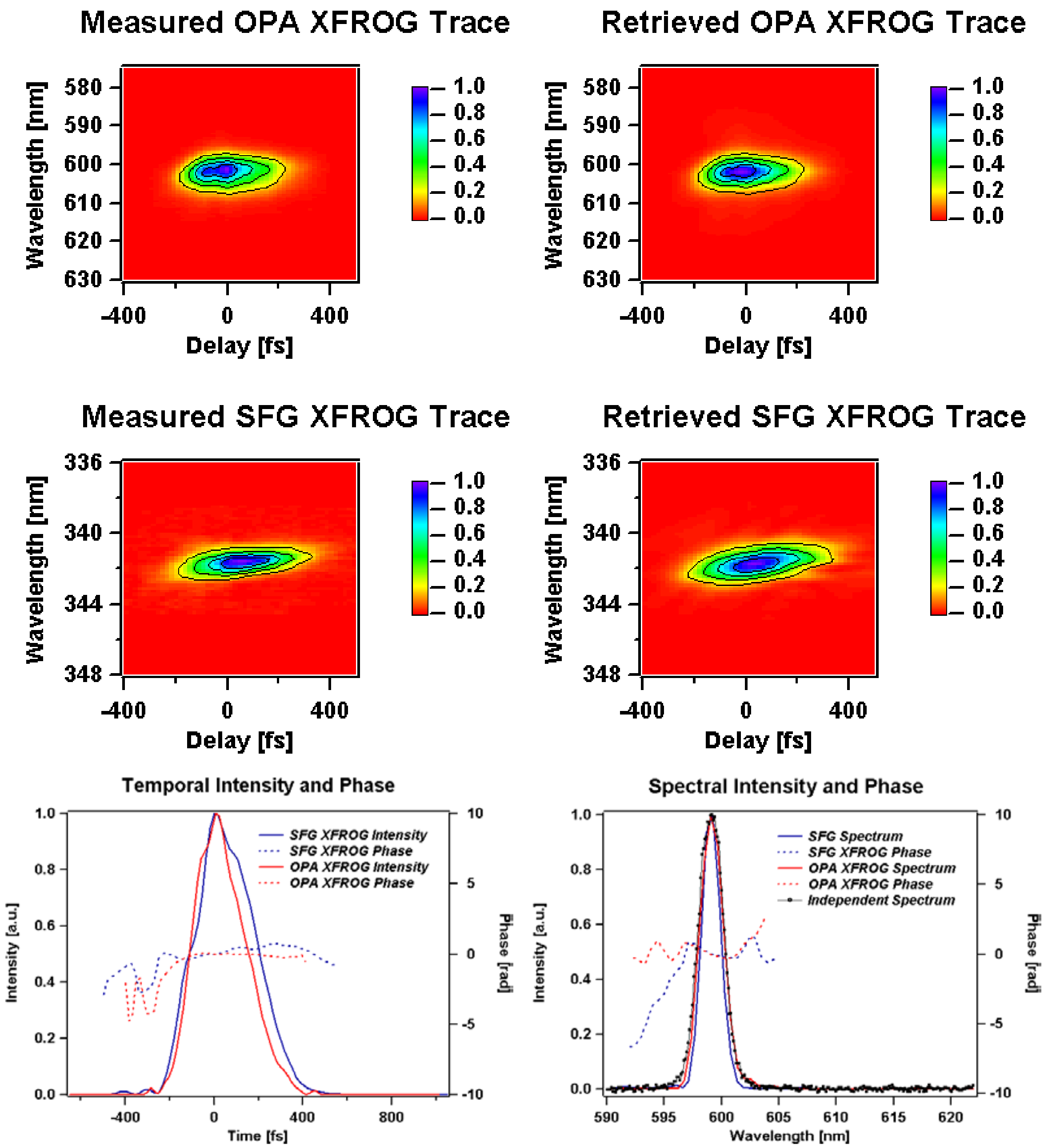

Figure 1. The measured and retrieved traces and retrieved intensity and phase vs. time and the spectrum and phase vs. wavelength of the slice of an attenuated and spectrally filtered 80-fJ continuum from a sapphire plate. The retrieved pulses from OPA XFROG agree well with the retrievals from the established technique, SFG XFROG. Noise is due to shot-to-shot jitter in the continuum and gate pulse.

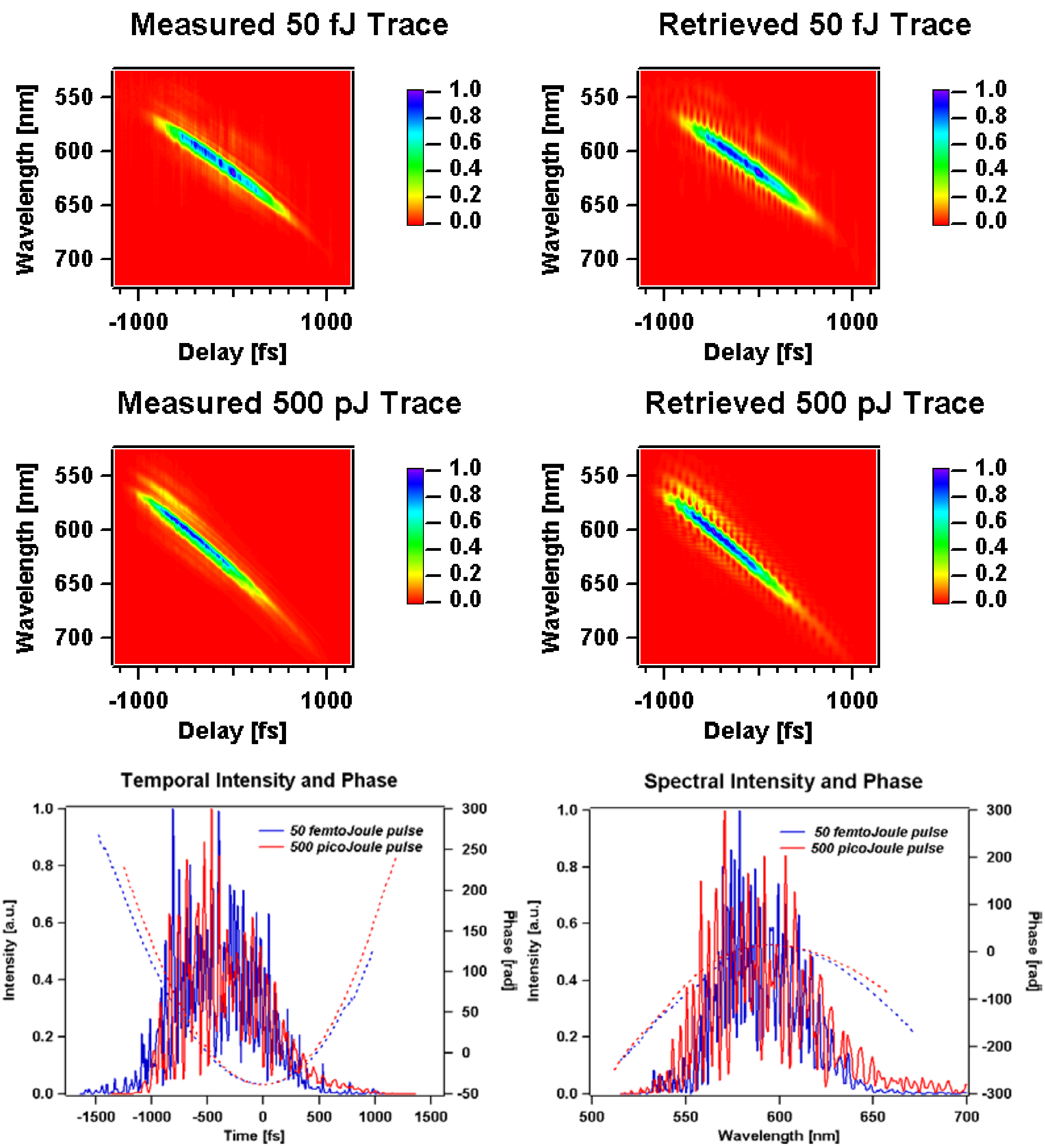

Figure 2. Comparative OPA XFROG measurements of 50 fJ (Gain ~10^3) and 500 pJ (Gain ~ 50) continuum generated using a sapphire plate and filtered using OG555 and BG40 filters. The traces show broadband phase matching due to non-collinear geometry. The fine-scale structure in the spectrum and intensity is real.

An OPA XFROG measurement of filtered continuum, attenuated to only 50 attojoules (i.e., 150 photons per pulse) is shown in Fig. 3. The trace shows fluctuations due to the instability of the OPA process using amplified pulses. These fluctuations do not impede the retrieval of intensity and phase because they cannot correspond to real pulse shape features and the algorithm ignores them.

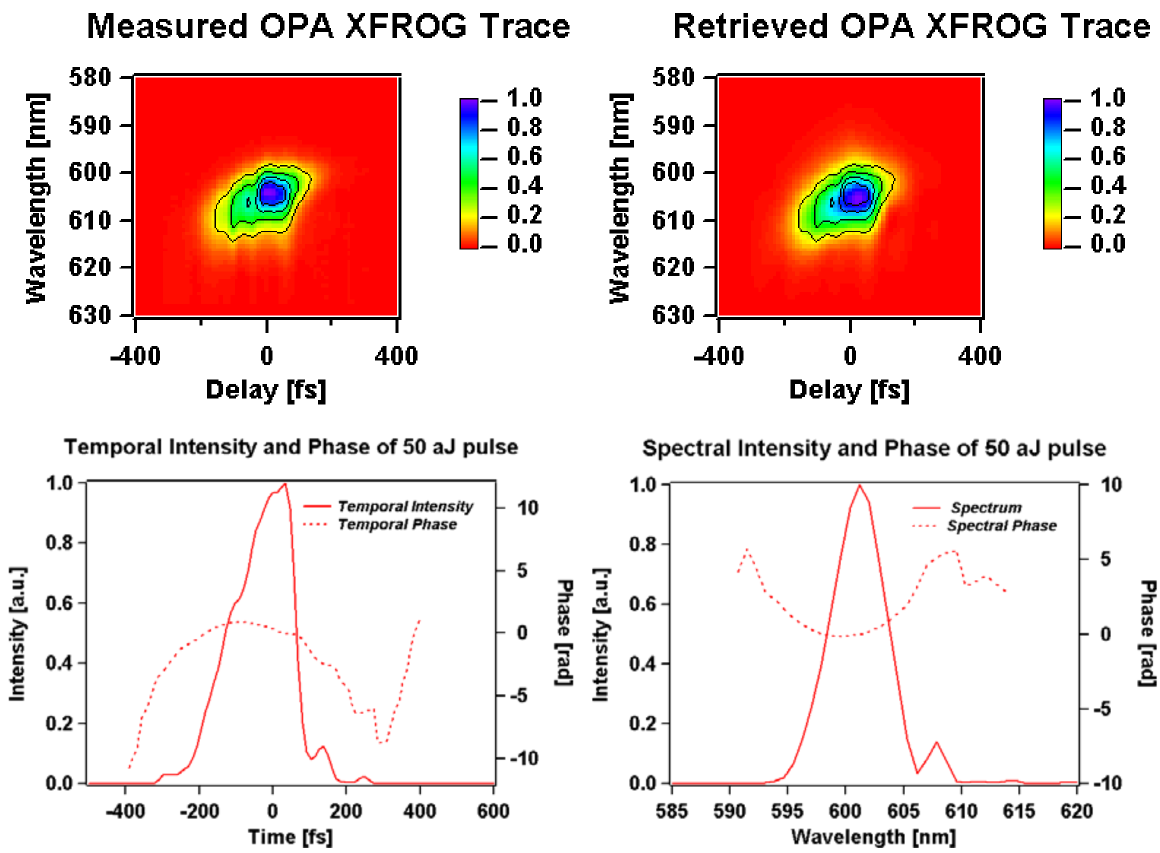

Figure 3. OPA XFROG measurement of a 50-aJ attenuated and filtered continuum generated using a sapphire plate.

The process of broadband phase matching is limited by Group Velocity Mismatch (GVM) conditions between the large range of signal wavelengths and the blue pump. Optical Parametric Generation (OPG) or superfluorescence limits how weak the signal to be measured can be, in order to retrieve the intensity and phase accurately.

In summary, we use the simultaneous gating and gain of the OPA process to measure the field of extremely weak, broadband, complex, spatially incoherent pulses with random absolute phase. This technique, which we call OPA XFROG, should be able to measure trains of fluorescence or spontaneous Raman pulses as weak as a few attojoules (i.e., a few photons).

You can learn more about this technique from:

J. Zhang, A. P. Shreenath, M. Kimmel, E. Zeek, R. Trebino, and S. Link, "Measurement of the intensity and phase of attojoule femtosecond light pulses using Optical-Parametric-Amplification Cross-Correlation Frequency-Resolved Optical Gating", *Opt. Express* 11(6), 601-609 (2003).

Keep a look out at this website for more exciting updates on OPA XFROG.