Measurement of the intensity and phase of attojoule femtosecond light pulses using Optical-Parametric-Amplification Cross-Correlation Frequency-Resolved Optical Gating

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Abstract: We use the combination of ultrafast gating and high parametric gain available with Difference-Frequency Generation (DFG) and Optical Parametric Amplification (OPA) to achieve the complete measurement of ultraweak ultrashort light pulses. Specifically, spectrally resolving such an amplified gated pulse vs. relative delay yields the complete pulse intensity and phase vs. time. This technique is a variation of Cross-correlation Frequency-Resolved Optical Gating (XFROG), and using it, we measure the intensity and phase of a train of attenuated white light continuum containing only a few attojoules per pulse. Unlike interferometric methods, this method can measure pulses with poor spatial coherence and random absolute phase, such as fluorescence.

OCIS codes: (190.4410) Nonlinear Optics, parametric processes; (190.7110) Ultrafast nonlinear optics; (300.6420) Spectroscopy, nonlinear; (300.6530) Spectroscopy, ultrafast.

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1. Introduction

The past decade has seen great progress in the development of techniques for measuring the intensity and phase vs. time of femtosecond laser pulses. Techniques such as Frequency-Resolved Optical Gating (FROG) and Cross-correlation FROG (XFROG) [1, 2] allow the measurement of a wide range of pulses. While these techniques have achieved fairly high sensitivity, they are not sensitive enough to measure extremely weak ultrashort light pulses—pulses with only a few photons—whose measurement would often help to elucidate important fundamental light-emission processes [3].

Previously we and others showed that trains of ~1-photon pulses could be measured using spectral interferometry [4]. Unfortunately, all interferometric methods carry extremely stringent coherence requirements, including the need for precise mode-matching, nearly perfect spatial coherence, and highly stable absolute phase (carrier-envelope phase) from pulse to pulse in the train. While these conditions can be met by *laser* pulses, they are rarely met by *light* pulses (pulses not directly emitted by a laser), such as fluorescence, Raman scattering, and super-continuum. Thus, interferometric methods can be useful for aligning lasers and studying high-optical quality media, but they are of little value in more practical situations.

So a non-interferometric technique lacking such restrictive coherence requirements and which achieves few-photon sensitivity is needed to help to elucidate fundamental weak-light emitting processes in many fields.

In this letter, we present a non-interferometric method capable of measuring trains of few-photon spatially incoherent light pulses. It is a variation on the XFROG method and hence involves measuring a time-gated pulse spectrum vs. delay to yield a visually intuitive spectrogram of the weak pulse. Unlike previous FROG methods, however, the gating process involves *gain*. Using either Optical Parametric Amplification (OPA) or Difference Frequency Generation (DFG) with an intense, higher-frequency, shorter gate pulse, it is possible to gate in time with a simultaneous gain of up to ~10^6. Like previous FROG and XFROG techniques, OPA and DFG XFROG do not require mode-matching, spatial coherence, or stability of the absolute phase.

OPA XFROG and DFG XFROG have additional advantages over interferometric methods. For example, another obstacle to the use of spectral interferometry for measuring weak pulses in many applications (even when the coherence requirements are met) is the lack of a well-characterized reference pulse with the same spectrum as the unknown weak pulse.

Fortunately, appropriate reference pulses are much more readily available for OPA and DFG XFROG. This is because the creation of ultraweak fluorescence or Raman scattering pulses requires its own excitation pulse, which itself must have three characteristics: 1) it must have a shorter wavelength, 2) it must be shorter in duration, and 3) it must be relatively intense. Coincidentally, these are precisely the three conditions for the reference pulse in OPA and DFG XFROG! Thus, for this wide range of cases, an ideal reference pulse for OPA and DFG XFROG is guaranteed to be available! OPA XFROG and DFG XFROG are thus ideal for measuring such ultraweak ultrashort light pulses as luminescence or Raman scattering.

OPA, DFG and stimulated Raman scattering (SRS) processes have been used for gating or gain in many situations previously. [5-9]. One particularly relevant case has been the simultaneous use of gating and gain in ballistic-imaging techniques [10, 11] where few-photon unscattered pulses containing the desired image must be time-gated.

Ultrafast fluorescence measurements are usually carried out by fluorescence up-conversion [12-14]. This involves the gating of the emitted fluorescence in a nonlinear medium (e.g. BBO) with the output 800-nm pulse of a Ti: Sapphire laser producing the sum frequency signal in the ultraviolet spectral range [12], but without gain. Also, streak cameras [14, 15] are commonly used to measure the fluorescence spectrum as a function of time, but their time-resolution is limited to a few picoseconds.

In contrast, OPA and DFG XFROG involve a nonlinear-optical process (OPA or DFG instead of sum-frequency generation, SFG) that has high gain, which allows us to significantly amplify ultraweak fluorescence pulses, increasing sensitivity by many orders of magnitude. Here we also use a modified FROG algorithm to retrieve the intensity and phase of the ultraweak pulse from the measured OPA XFROG spectrogram.

Specifically, we demonstrate OPA XFROG by measuring trains of pulses as weak as 50 aJ (a few hundred photons) per pulse. Indeed, because our repetition rate is 100,000 times lower in this work than in the previous spectral-interferometry work [4], the total number of photons required for our measurement here is actually less than in the previous interferometric measurement of ultraweak pulses.

2. OPA/DFG XFROG

The FROG technique first requires a gate pulse to gate the unknown pulse. In standard FROG, the gate pulse is the pulse itself. However, when a well-characterized reference pulse is available, it is generally better to use it. The expression for such an XFROG trace is:

$$I_{XFROG}(\omega,\tau) = \left| \int_{-\infty}^{\infty} E(t) E_{gate}(t-\tau) \exp(-i\omega t)\, dt \right|^2 \qquad (1)$$

where the gate function, $E_{gate}(t)$, can be any function (i.e., pulse) that happens to be available and which has temporal structure on the order of that of the pulse to be measured, $E(t)$. All that is required is a signal field that is a function of time and delay, an example of which is a product of the form, $E(t) E_{gate}(t-\tau)$, which can then be spectrally resolved.

A schematic of the apparatus for both OPA XFROG and DFG XFROG is shown in Fig. 1. In OPA XFROG, the weak pulse (shown in red) is parametrically amplified in the crystal by the more intense reference-gate pulse (shown in blue). The DFG XFROG signal pulse is also shown (in brown). Either pulse can be spectrally resolved to yield an OPA XFROG or DFG XFROG trace.
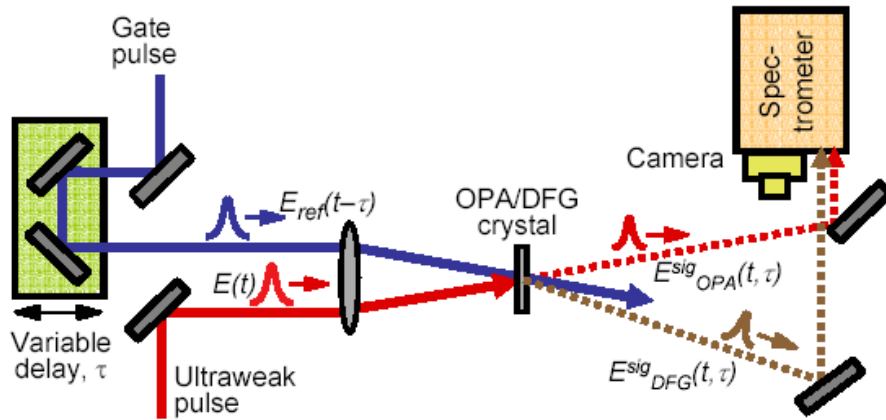
Fig. 1. Schematic of the experimental apparatus for OPA/DFG XFROG. The gate pulse is characterized using a GRENOUILLE (not shown) before it enters the XFROG setup.

The coupled-wave equations for the generation of both the signal and idler (which we will refer to here as the OPA and DFG pulses, respectively, because the term, "signal," is already used in FROG and has a different meaning) are:

$$\frac{\partial E_{OPA}}{\partial z} = i\kappa E_{ref} E_{DFG}^* \tag{2}$$

$$\frac{\partial E_{DFG}}{\partial z} = i\kappa' E_{ref} E_{OPA}^* \tag{3}$$

where $\kappa = \dfrac{4\pi d_{eff}}{n_{OPA}\lambda_{OPA}}$ and $\kappa' = \dfrac{4\pi d_{eff}}{n_{DFG}\lambda_{DFG}}$.

Assuming negligible pump depletion, the electric field of the signal pulse emerging from the crystal in an OPA XFROG apparatus has the form:

$$E_{sig}^{OPA}(t,\tau) = E(t) E_{gate}^{OPA}(t-\tau), \tag{4}$$

with $E(t)$ is the unknown input pulse. The second factor is the gate pulse and is given by

$$E_{gate}^{OPA}(t) = \cosh(g\left|E_{ref}(t)\right|z). \tag{5}$$

Here, the gain parameter, g, is given by:

$$g^2 = \frac{16\pi^2 d^2_{eff}}{n_{OPA}n_{DFG}\lambda_{OPA}\lambda_{DFG}}. \tag{6}$$

Thus the pulse to be measured undergoes exponential gain during OPA and retains its phase during the process of OPA.

Note that, unlike other FROG methods, OPA XFROG has a background even at large delays due to the transmission of the input pulse. Our equations (and the resulting algorithm) take this into account. In the limit of high gain, however, this background is small and unnoticeable.

The setup for DFG XFROG is similar to that of OPA XFROG, except that now the idler is imaged onto the slit of the spectrometer to yield a DFG XFROG trace. Although it is known that DFG XFROG is a sensitive technique for measuring fairly weak pulses [16], the method has never been demonstrated with gain. Here we consider the effect of possible gain, so that the electric field is given by:

$$E_{sig}^{DFG}(t,\tau) = E(t) E_{gate}^{DFG^*}(t-\tau).$$ (7)

This has the same unknown input pulse and a gate function of the form:

$$E_{gate}^{DFG}(t) = \exp[i\phi_{ref}(t)]\sinh\left(g\left|E_{ref}(t)\right|z\right),$$ (8)

where $\phi_{ref}(t)$ is the phase of the reference pulse. In the limit that the reference pulse is weak, the net gain is small, and the above expression reduces to $E_{gate}^{DFG}(t) = E_{ref}(t)$.

The measured XFROG trace is simply the magnitude-squared Fourier transform of the various signal fields.

In both OPA and DFG XFROG, the unknown pulse is easily retrieved from the measured trace using the iterative XFROG algorithm, modified for the above expressions for the gate pulse. Note that, in the presence of high gain, the gate pulse experiences shortening in time, which is generally desirable.

In this treatment we have neglected the effect of group velocity mismatch (GVM) between the interacting pulses [17-19]. GVM between the pump and signal pulses constrains the interaction length over which parametric amplification occurs. The larger the GVM, the shorter the interaction length will be. GVM depends on the crystal type, pump wavelength and the type of phase-matching. Defining a pulse-splitting length l_{sp} as the propagating distance after which the pump and signal pulses separate from each other, it is known that GVM effects can be neglected in light-generation experiments to a first approximation for the cases where the crystal lengths are shorter than the pulse splitting length:

$$l_{sp} = \frac{\tau_p}{GVM_{sp}},$$ (9)

where τ_p is the length of the longer of the signal or pump pulse. In using OPA or DFG for pulse measurement, however, one must be careful to avoid allowing the gate pulse to walk more than one coherence time (usually less than one pulse length) with respect to the pulse to be measured, or the gate pulse will sample a range of pulse phases, not the correct phase for a particular delay. Our experiments measuring ~250-fs pulses are close to this limit, but we imagine that this method will find far greater use in the few-ps regime for measuring ultrafast fluorescence, where GVM effects are less detrimental.

3. Experimental results

Our experimental setup for OPA/DFG XFROG is shown in Fig. 1. In our experiments, specifically, the output from a femtosecond KM Labs Ti:Sapphire oscillator is amplified using a kilohertz repetition rate regenerative amplifier. The amplified 800 nm pulse is characterized using a Swamp Optics GRENOUILLE. The pulse is then split into two. One pulse is used to generate a white-light continuum (with poor spatial coherence) in a 2-mm thick sapphire

plate, which is then spectrally filtered to yield a narrow slice of the spectrum about 3 nm wide around 600 nm. This pulse is attenuated using a 3 ND filter by a factor of 1000 and has ~ 80 femtojoules of energy. It is the "unknown" pulse in our experiments. The other pulse is frequency-doubled in a 1-mm thick BBO crystal using Type I phase matching and passed through a variable time delay to act as a gate (pump) pulse (with 5.8 μJ) for the OPA/gating process. The two pulses are focused by a 75-mm spherical mirror at a small angle (3°) in a 1-mm thick BBO crystal, again using Type I phase matching. The thickness of the crystal is chosen so that the pulse splitting length is about two times the crystal length, and therefore the effects of GVM are small enough that we can use the simple gate functions described above [17-19]. This is also sufficient to cover the phase-matching bandwidth of the weak pulse. The signal at the CCD array is integrated over a few seconds. The OPA signal emerging from the BBO crystal sees an average gain (G) of about cosh(5.75) ≅ 150, which, in view of the weak pulses involved, easily satisfies the condition of negligible pump depletion. This value of the gain was, however, more than sufficient to record the spectrally dispersed signal at the camera, i.e., the OPA spectrum vs. relative delay, which is the OPA XFROG trace.

Using OPA XFROG, we first measured the above-described 80 fJ pulse. Fig. 2 shows the measured and retrieved OPA XFROG trace for this pulse, along with the retrieved intensity and phase vs. time and the spectrum and spectral phase vs. wavelength. The FROG error was 0.01 for this 128 x 128 pixel trace. The retrieved pulse had a temporal intensity FWHM of 266.5 fs. The spectral FWHM was 2.428 nm, so that the FWHM time-bandwidth product (TBP) was 0.4976.

To verify the results of the above OPA XFROG measurement, we measured the same continuum pulse using a well established, but less sensitive, method: SFG XFROG (see Fig. 2). The experimental setup was identical to that of OPA XFROG, except that a 100 micron thick Type I BBO crystal was used to phase-match the sum frequency; the 800-nm gate pulse was attenuated to 400 nJ; and the sum-frequency signal was imaged onto the spectrometer. In this case, the retrieval on a 128 x 128 trace had a FROG error of 0.014. The temporal intensity FWHM was 284.4 fs and the spectral FWHM was 2.056 nm, so that the FWHM TBP was 0.4498. The two measurements of the same pulse agree reasonably well, as does the independently measured spectrum.

There is another limit on the available gain in OPA XFROG. If the gating pump pulse is too intense, it results in Optical Parametric Generation (OPG) in the nonlinear crystal, which is generated from noise and is an unwanted background for the OPA signal. Since, for very high gain, OPG can rival OPA in intensities, this could result in distortion in the XFROG retrieved pulse. Thus, in our experiments, we kept the pump power low enough to avoid distortions due to potential OPG background. OPG also places a limit (a few photons per pulse) on how weak the unknown signal can be and still be measured accurately.

In order to test the limits of the method, we measured, in an additional experiment, a train of attojoule pulses (only a few hundred photons). The OPA process was induced in a 2-mm thick BBO crystal. Our OPA XFROG measurement of a slice of the continuum 10 nm wide--attenuated to only 50 attojoules--is shown in Fig. 3, for which the gain was about 10^5. The OPA signal in this trace was only ~5 times stronger than the OPG background. In this measurement, the OPA gain was high enough to saturate the CCD array and so the signal also had to be attenuated before entering the spectrometer. In practice, the gain should not be so high that such attenuation is required, but allowing it to occur here provided an additional test of the method. As a result of the very high gain, the OPA XFROG trace showed large fluctuations in signal strength from one step to the next due to variations in the gate-pulse energy and from the inherent instability of a single pass OPA process with high gain. Fortunately, the OPA XFROG retrieval algorithm sees through this artifact (which cannot correspond to real trace structure) and smoothes it out during the retrieval. The retrieved pulse had a temporal intensity FWHM of 170 fs and a spectral FWHM of 5.165 nm, with a FROG error of 0.0146 on a 128 x 128 grid. The corresponding TBP was 0.731.
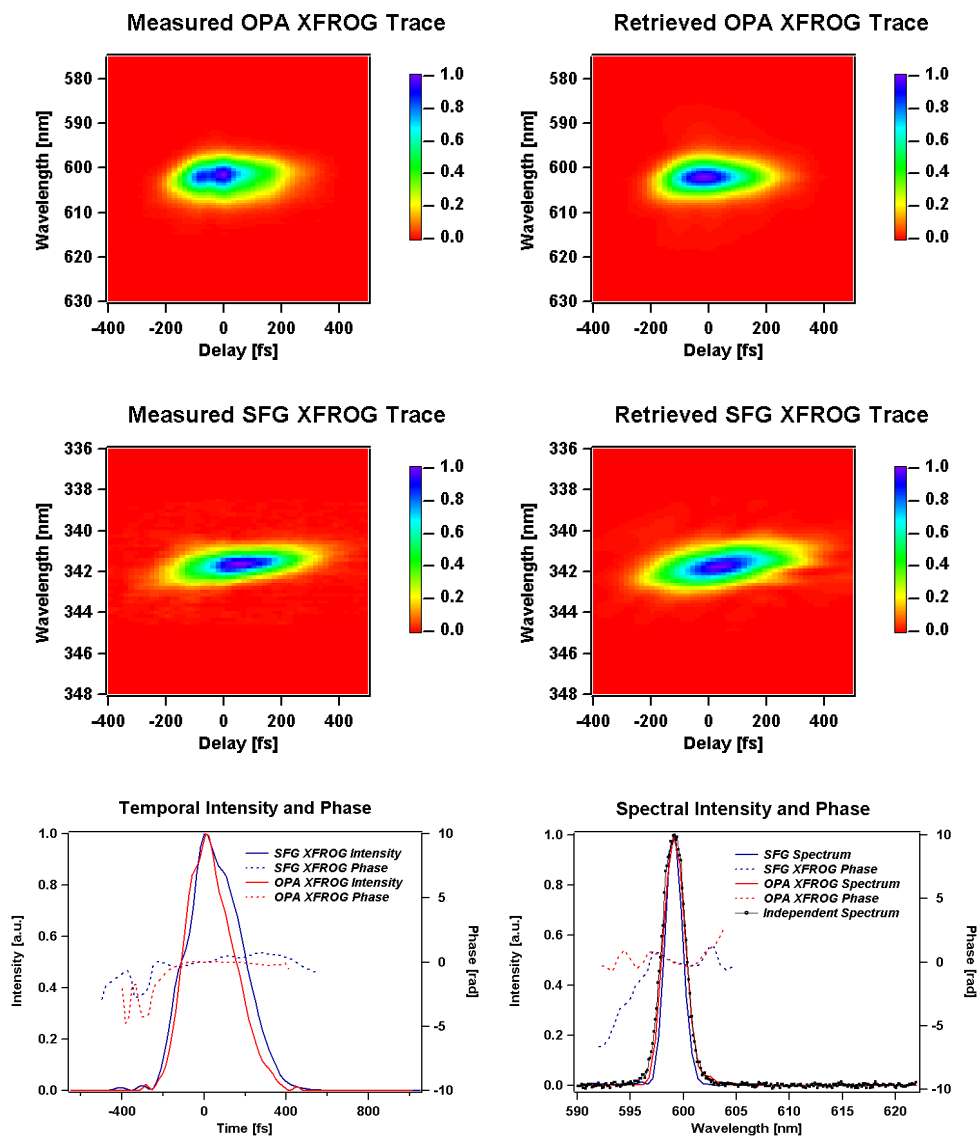
Fig. 2. The measured and retrieved traces and retrieved intensity and phase vs. time and the spectrum and spectral phase vs. wavelength of a spectrally filtered continuum from a sapphire plate. The retrieved intensity and phase from the OPA XFROG measurement of 80fJ pulses agrees well with the retrieved intensity and phase of unattenuated continuum of 80pJ using the established technique, SFG XFROG as well as the independently measured spectrum.

We should point out that this particular measurement violates the GVM constraints. But the experiment was performed to test the limits on how weak a pulse can be measured using this technique. Since fluorescence pulses of interest generally tend to be a few hundred femtoseconds to a few picoseconds long, GVM should prove less problematic in such

measurements. However the measurement shows that it is possible to measure extremely weak pulses by using a thicker crystal.

In all of these experiments, we checked for beam distortions due to high-intensity nonlinear-optical processes in the OPA crystal, such as small-scale self-focusing (which, if present, could indicate that the FROG measurement distorted the pulse in the measurement process), and have found none. Also, we calculated the total phase shift due to the nonlinear refractive index—about 0.11 radians per millimeter—which is insufficient to affect our measurements.

DFG XFROG, in principle, yields similar results with the same magnitude of gain. It would be a convenient technique to measure weak signal pulses at shifted wavelengths.
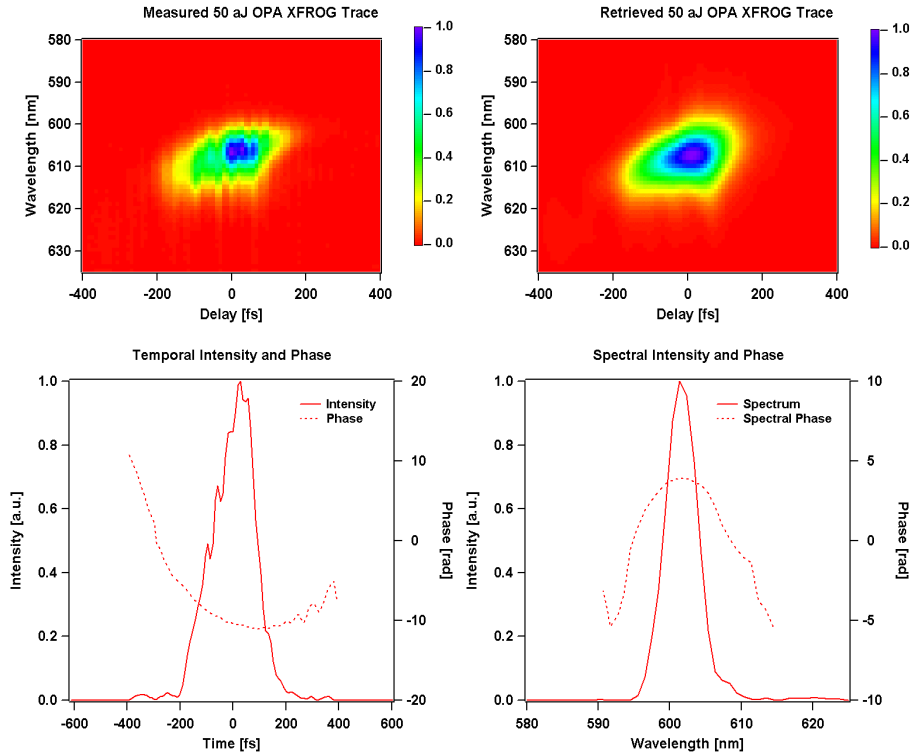

Fig. 3. OPA XFROG measurement of a 50 aJ attenuated and filtered continuum generated using a sapphire plate.

4. Conclusions

We have introduced a new variation of the existing XFROG technique called OPA XFROG which, along with its cousin DFG XFROG, is the most sensitive light-pulse-measurement technique now available. Unlike interferometric methods, it does not carry prohibitively restrictive requirements, such as mode-matching, perfect spatial coherence, and highly stable absolute phase. While care must be taken to avoid GVM effects in such measurements for fs pulses, it should not pose a prohibitive problem for most ps measurements. This makes OPA and DFG XFROG powerful tools for measuring non-laser ultrashort light pulses. We have demonstrated that OPA XFROG can measure the intensity and phase vs. time of pulses with only a few attojoules per pulse and with pulse widths on the order of 250 fs; the measured results with OPA XFROG agree well with those measured by using a well-known method, SFG XFROG. By increasing the pump power (despite the limits imposed by competing OPG processes), it should be possible to measure ultraweak pulses of the order of a few hundred

zeptojoules (i.e., just a few photons per pulse). DFG XFROG has the same sensitivity and should be ideal for measuring light pulses in the infrared. We plan to use OPA XFROG to measure ultraweak ultrafast fluorescence from biologically important media.

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