# Measurement of the intensity and phase of ultraweak, ultrashort laser pulses

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We show that frequency-resolved optical gating combined with spectral interferometry yields an extremely sensitive and general method for temporal characterization of nearly arbitrarily weak ultrashort pulses even when the reference pulses is not transform limited. We experimentally demonstrate measurement of the full time-dependent intensity and phase of a train of pulses with an average energy of 42 zeptojoules ($42 \times 10^{-21}$ J), or less than one photon per pulse. 

Despite the rapid progress that has recently occurred in the development of techniques for characterizing ultrashort laser pulses,[1–3] essentially no progress has occurred in extending these techniques to lower pulse energies. All full-characterization techniques require a nonlinear-optical process and hence are inherently no more sensitive than autocorrelation. Indeed, all require at least a few picojoules for a multishot pulse measurement and significantly more for single-shot measurement (if single-shot measurement is even possible).

Significant benefits would result, however, from an extremely sensitive full-characterization technique for ultrashort laser pulses. For example, most ultrafast nonlinear-optical material-characterization experiments yield signal-pulse energies of femtojoules or less. Currently, only the signal-pulse energy is usually measured in such experiments. Many researchers, however, have realized that a well-characterized signal pulse provides significantly more material information. Several groups are now spectrally resolving the signal beam versus delay in such experiments. Chemla *et al.*[4] and Patkar *et al.*[5] use several pulse-characterization methods to characterize the signal pulse in four-wave-mixing experiments performed on semiconductors. Consequently, they have significantly improved our understanding of the physics of multiple quantum wells. A technique that provides the full time-dependent (or frequency-dependent) intensity and phase of an ultraweak, ultrashort pulse would be quite useful.

By itself, this task is daunting.[6] Fortunately, ultraweak, ultrashort laser pulses do not exist "in a vacuum." The processes that create them generally involve a much stronger pulse as input. Indeed, the intracavity processes that produce ultrashort laser pulses are nonlinear and necessarily yield only fairly intense pulses at the output of the laser. Such pulses are easily measurable with a technique such as frequency-resolved optical gating (FROG).[1] Thus a well-characterized reference pulse is available in essentially all cases.

In this Letter we demonstrate a method for measuring arbitrary ultraweak coherent fields (ultrashort laser pulses in our case) that takes advantage of this fact. The method is simply the combination of two well-known techniques, which were not previously combined to our knowledge. We use FROG to characterize the reference pulse directly from the laser [any method that yields the reference-pulse phase versus frequency, $\varphi_{\mathrm{ref}}(\omega)$, will suffice]. By using FROG, we avoid the necessity of assuming a transform-limited pulse. We then use spectral interferometry (SI), which is a simple and linear, and hence very sensitive, method for measuring the frequency-domain phase difference between two pulses. SI simply involves measuring the spectrum of the sum of two pulses, which easily yields the phase difference between the two pulses. Here SI provides $\varphi_{\mathrm{unk}}(\omega) - \varphi_{\mathrm{ref}}(\omega)$, where $\varphi_{\mathrm{unk}}(\omega)$ is the ultraweak unknown pulse phase versus frequency. Knowledge of $\varphi_{\mathrm{ref}}(\omega)$ from the FROG measurement will then yield $\varphi_{\mathrm{unk}}(\omega)$. And because the unknown pulse spectrum is easily measured (with the same SI apparatus), the FROG and SI measurements together yield the full intensity and phase of the unknown ultraweak pulse. Thus the combination of FROG and SI provides a nearly general technique for measuring even the weakest ultrashort laser pulses. Here we demonstrate this combination technique, which we call temporal analysis, by dispersing a pair of light *e* fields (TADPOLE). Using TADPOLE, we measure pulses as weak as 42 zeptojoules (zJ), or $42 \times 10^{-21}$ J. This represents an 8-order-of-magnitude improvement in sensitivity in intensity-and-phase measurement.

SI was first introduced by Froehly *et al.*,[7] and it has been used for several applications.[8,9] Its use has been limited, however, to situations in which only a phase difference is required or to the measurement of complex pulses, compared with which a pulse directly from a laser has a constant phase. This study is believed to be the first demonstration of SI with a fully characterized reference. SI involves simply directing the two pulses collinearly into a spectrometer (see Fig. 1). The spectrum of the two pulses is

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$$I_{\mathrm{SI}}(\omega) = I_{\mathrm{ref}}(\omega) + I_{\mathrm{unk}}(\omega) + 2\sqrt{I_{\mathrm{ref}}(\omega)}\sqrt{I_{\mathrm{unk}}(\omega)} \times \cos[\phi_{\mathrm{unk}}(\omega) - \phi_{\mathrm{ref}}(\omega) - \omega\tau]. \quad (1)$$

Here $I_{\mathrm{ref}}(\omega)$ and $I_{\mathrm{unk}}(\omega)$ are the spectra of the reference and the unknown pulses, respectively, and $\tau$ is the delay between the two pulses. $\tau$ is chosen to yield fringes in the sum spectrum. We measure the two individual pulse spectra by blocking one beam and measuring the spectrum of the other. The SI spectrum, $I_{\mathrm{SI}}(\omega)$, determines the phase difference, $\varphi_{\mathrm{unk}}(\omega) - \varphi_{\mathrm{ref}}(\omega) - \omega\tau$, the only remaining unknown. The spectrum is easily extracted noniteratively with one of several well-known fringe-inversion techniques.[9,10] The magnitude of the relative delay also emerges from the analysis (it is the linear term) and hence does not need to be independently measured except to determine its sign and avoid an ambiguity in the sign of the cosine argument. In fact, $\sqrt{I_{\mathrm{ref}}(\omega)}\sqrt{I_{\mathrm{unk}}(\omega)}$ can also be extracted from the analysis, so there is no need to measure the spectrum of the unknown pulse as long as the spectrum of the reference is precisely known. An advantage of Si is that it is a type of heterodyne technique and can act to amplify the weak pulse. For example, choosing the reference pulse to be $M$ times more intense than the unknown pulse produces fringes that are $4M^{1/2}$ times as intense as the spectrum of the unknown pulse. The only requirement of SI is that the spectrum of the unknown pulse lie within that of the reference pulse.

To demonstrate TADPOLE for the measurement of ultraweak ultrashort pulses, we used second-harmonic generation (SHG) FROG to measure a train of linearly chirped 145-fs reference pulses directly from a Ti:sapphire oscillator. The oscillator ran at 859 nm with a repetition rate of 96 MHz. The sensitivity required for measuring the reference pulse is a limitation of TADPOLE. We used a multishot SHG FROG because it relies on a second-order nonlinearity and is the most sensitive of the FROG geometries. Typical multishot SHG FROG devices can measure pulse trains with peak powers of less than 200 W and can easily measure a fraction of the 35-kW (peak-power) train from our oscillator. There is a temporal ambiguity in SHG FROG, but we note that it reverses the spectrum as well as the time axis. Thus, as long as the spectrum is asymmetric, an independent measurement determines the correct direction of time. Figure 2 shows the SHG FROG trace, the retrieved spectrum and phase of the reference pulses, and the measured spectrum for comparison.

The train of pulses was then passed through attenuators and 16 cm of fused silica, which lengthened the pulse to 250 fs to simulate the performance of a material-characterization experiment (see Fig. 3). The weak 250-fs pulse was then combined with an attenuated piece of reference pulse in a spectrometer. We chose a delay that yielded approximately 12 fringes across the spectrum. A thermoelectrically cooled CCD camera then recorded the SI spectrum for a 0.5-s exposure. Even though the experiment is interferomeric, we did not need to stabilize the interferometer over this time scale. We were able to make a clean measurement for average powers of 4 and 36.4 pW (168-nW and 2.61-$\mu$W peak powers) in the unknown and the reference arms of the experiment, respectively. Thus the average energy per unknown pulse was only 42 zJ, or 1/5 of a photon. Such sensitivity is partly due to the high repetition rate but is also due to the high sensitivity of cooled CCD cameras, the linear lossless nature of the technique, and the heterodyne effect discussed above.

Figure 4 shows the resulting SI spectrum and the unknown pulse spectrum obtained by blocking the reference beam. We extracted the phase difference by Fourier transforming the spectrum, filtering out the negative and zero-frequency components, frequency shifting the positive-frequency component to dc (to remove the delay term), and inverse Fourier transforming back to the frequency domain. The phase of the resulting spectrum is then the phase difference between the reference and the unknown spectra.[10] The unknown pulse's spectrum and the phase obtained are

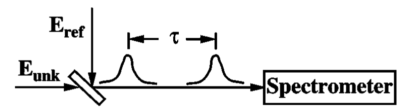


Fig. 1. Apparatus for SI measurements of the frequency-domain phase difference between two pulses.

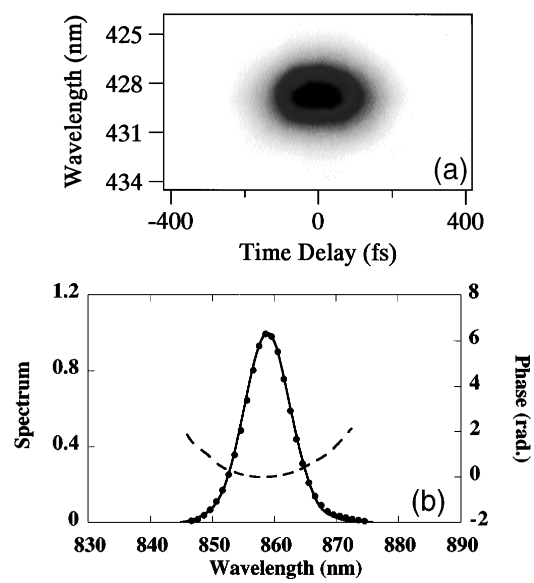


Fig. 2. (a) FROG trace of the reference pulse used in TADPOLE experiments. (b) Reference-pulse measured spectrum (filled circles) and the retrieved spectrum (solid curve) and spectral phase (dashed curve) from the FROG trace. When a 128 × 128 grid was used for the FROG trace, the FROG algorithm produced a FROG error of 0.0038.

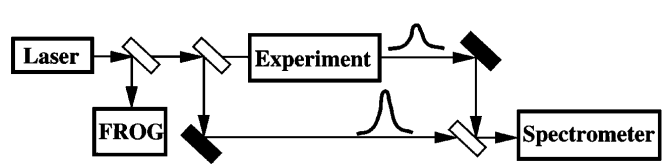


Fig. 3. Apparatus for TADPOLE measurements of ultraweak pulses generated in an experiment consisting of 16 cm of fused silica used to stretch and chirp the pulse and attenuation by a factor of $10^9$.

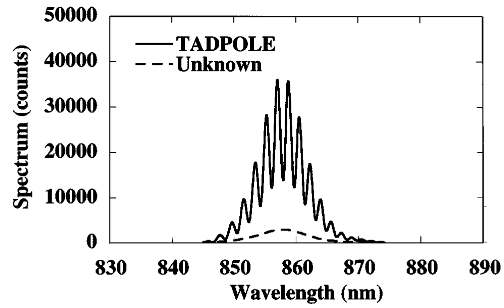


Fig. 4. SI spectrum (solid curve) and the unknown pulse spectrum (dashed curve).

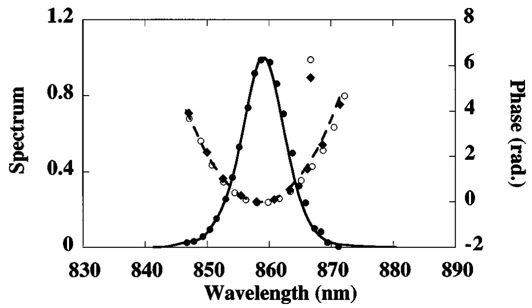


Fig. 5. Spectrum and phase of the unknown ultraweak pulse train measured with TADPOLE (filled and open circles, respectively) and with FROG (solid and dashed curves, respectively). Filled diamonds, calculated phase predicted by adding the phase change that is due to the known dispersion of quartz to the reference phase.

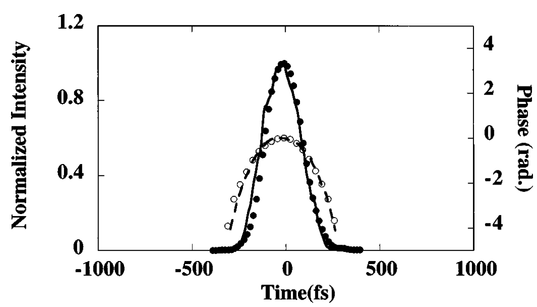


Fig. 6. Temporal intensity and the phase of the unknown ultraweak pulse train measured with TADPOLE (filled and open circles, respectively) and with FROG (solid and dashed curves, respectively).

shown in Fig. 5. For comparison, the figure shows the independently measured spectrum and phase obtained by use of FROG on the unattenuated pulses. It also shows the phase calculated from the dispersion of quartz and the phase of the reference phase. Figure 6 shows the unknown pulse's intensity and phase versus time obtained with TADPOLE and FROG. The FROG and TADPOLE measurements agree, and the change in the phase is also consistent with that predicted by the known dispersion curve of quartz. Because the reference pulse is appreciably chirped, a measurement assuming a transform-limited pulse would have been inaccurate.

The ultimate sensitivity of TADPOLE is extremely high: our measurement of zeptojoule pulses still involved on average 5000 counts per pixel. Additional attenuation by 100 or so is therefore possible, yielding sensitivity for pulse trains in the yoctojoule ($10^{-24}$ J) range, or a small fraction of a photon per pulse. Single-shot measurement of an individual pulse in the subfemtojoule range should also be possible.

TADPOLE also appears to be an excellent method for measuring shaped ultrashort pulses.[11] Because they are often spread out in time, such pulses can be too weak to yield sufficient signal in a FROG measurement. Shaped pulses can also be so complex that they would require an inconveniently large number of data points in a FROG trace. The SI spectrum, on the other hand, has the advantage of being one dimensional, thus requiring significantly fewer data points for a given level of pulse complexity than FROG. Because the shaped pulse is usually constructed from a nearly transform-limited pulse, the latter pulse provides an ideal reference pulse, easily measured with FROG.

Thus the combination of FROG and SI, which we call TADPOLE, extends the sensitivity of rigorous full characterization of ultrashort laser pulses by many orders of magnitude, which should be useful for many applications. As a final note, the experimental arrangement bears a resemblance to the homodyne detector used to characterize quantum fields.[12] Indeed, in a slightly modified form this technique can in principle be used to measure quantum field statistics of pulsed fields.

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