Frequency-resolved optical-gating measurements of ultrashort pulses using surface third-harmonic generation

Thomas Tsang

Brookhaven National Laboratory, Upton, New York 11973

Marco A. Krumbügel, Kenneth W. DeLong, David N. Fittinghoff, and Rick Trebino

Combustion Research Facility, MS9057, Sandia National Laboratories, Livermore, California 94551-0969

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We demonstrate what is to our knowledge the first frequency-resolved optical gating (FROG) technique to measure ultrashort pulses from an unamplified Ti:sapphire laser oscillator without direction-of-time ambiguity. This technique utilizes surface third-harmonic generation as the nonlinear-optical effect and, surprisingly, is the most sensitive third-order FROG geometry yet.

Techniques for the complete measurement of high-energy ultrashort laser pulses are now well established.[1–3] Frequency-resolved optical gating (FROG), for example, can rigorously and unambiguously measure the intensity and phase of an ultrashort laser pulse over a wide range of wavelengths, pulse lengths, and repetition rates, using the polarization-gate or self-diffraction beam geometry.[4] FROG measurements using third-order processes are limited in sensitivity, however, to a microjoule pulse in single-shot measurements and ~50-nJ pulses in multishot measurements. Indeed, for the measurement of pulse trains from unamplified Ti:sapphire oscillators these third-order processes do not have sufficient strength to yield usable traces. Currently, for the measurement of oscillator pulse trains, it is necessary to use second-harmonic generation (SHG) as the nonlinearity in FROG measurements. Although SHG FROG is simple to set up and has yielded excellent results in numerous situations, including the measurement of pulses as short as 9 fs,[5] SHG FROG has an unavoidable ambiguity in the direction of time.[6] Other intensity-and-phase methods exist that use SHG and are unambiguous. Unfortunately, these methods lack the rigor, generality, and experimental simplicity of FROG. Thus it would be useful to have a strong third-order process that can be used in FROG measurements of ultrashort-pulse laser oscillators.

We have tried many nonlinear media in a search for such a process. For example, heavy-metal-doped glasses have a significantly higher third-order nonlinearity than fused silica, which is usually used for PG FROG measurements. But the scattering that is due to such glasses is too severe. The polymer PPV also appeared promising, and, although it improved measurement sensitivity by an order of magnitude or so, a slow integrated effect prevented multishot measurements.[7] Cascaded second-order effects can appear as third-order effects and hence remove the direction-of-time ambiguity but also lack the sensitivity required for oscillator measurements, although this class of effects is still under consideration.

The purpose of this Letter is to show that a third-order process exists that does in fact succeed in providing unambiguous FROG traces of the Ti:sapphire oscillator. That process is surface third-harmonic generation (THG), which was recently demonstrated to be remarkably strong.[8] Here we demonstrate that its use as the nonlinearity in a FROG device easily yields FROG traces for a Ti:sapphire oscillator. Using input pulses of 300-mW average power and ~100-fs duration in a 100-MHz repetition pulse train, we obtain a few nanowatts of average THG signal power, easily sufficient for the required spectral measurements. Furthermore, this nonlinearity has an additional advantage: The interaction length is extremely short, so in principle one can measure even the shortest pulses by using it without potential distortions caused by geometric, dispersive, and phase-mismatch effects (all proportional to the interaction length).

The experimental setup for surface THG FROG is practically identical to the common SHG FROG setup, and the two geometries are interchangeable (Fig. 1): A pulse from a self-mode-locked Ti:sapphire laser oscillator is divided by a beam splitter. After one replica of the pulse is delayed with respect to the other, a 20× microscope objective is used to focus the two collinearly propagating beams on to the back surface of a 160-μm-thick piece of cover glass. We note that by

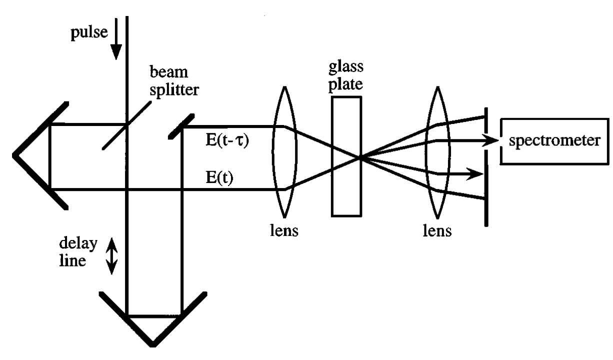

Fig. 1. Experimental setup for surface THG FROG measurements.

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focusing the fundamental beam at the back surface one can use even dielectric materials that are opaque to the third-harmonic radiation. The THG signal is highly localized at the air–dielectric interface and disappears completely when the interface is traversed away from the beam focus. The back surface is then the source of two autocorrelated THG beams, one of which yields a signal field of the form

$$E_{\text{sig}}^{\text{THG}}(t, \tau) = E^2(t)E(t - \tau).$$

[For the second beam the $E(t - \tau)$ term is squared instead of the $E(t)$ term.] The first THG beam is recollimated and sent to a spectrometer equipped with a linear diode array for spectral recording. Spectrograms at various time delays, with a 10-fs interval, are collected and converted into a 256×256 pixel FROG trace. THG FROG traces look much like SHG FROG traces but exhibit some asymmetry, which breaks the ambiguity. Figure 2 gives examples of THG FROG traces for several types of distortion.

The pulse intensity and phase are retrieved from the THG FROG trace by the generalized-projections technique[9] simply modified for the above expression for the THG FROG signal field in terms of the laser pulse field. We tested this algorithm for hundreds of theoretical pulses and found it to be as robust as other FROG algorithms.

Replacing the cover glass with a 50-μm-thick β-barium borate crystal and focusing into the bulk of the material with a 20-cm focal-length lens generates second-harmonic radiation, giving a signal field of the form

$$E_{\text{sig}}^{\text{THG}}(t, \tau) = E(t)E(t - \tau),$$

and a corresponding SHG FROG trace is recorded, thus permitting a comparison of both measurements.

Figure 3 shows the intensity and the phase for a nearly transform-limited pulse, which is retrieved from independent SHG and THG FROG measurements. The insets show the measured traces, which are both nearly symmetric with respect to the time delay. The retrieved intensity and phase agree well for both measurements, showing a nearly Gaussian intensity profile and a nearly flat phase of the pulse.

In general, SHG and THG FROG traces exhibit somewhat different features. As an example, Fig. 4 shows the measured and reconstructed SHG and THG FROG traces for a pulse that is clearly not transform limited. Whereas the measured and the reconstructed traces agree well for both cases, the SHG traces show the typical symmetry with respect to the time delay, giving rise to the time ambiguity in SHG FROG. The THG traces, on the other hand, are asymmetric with respect to the delay axis and, as a result, lack the time ambiguity.

It should be noted that, whereas THG FROG lacks the general direction-of-time ambiguity that occurs for SHG FROG, THG FROG has a direction-of-time ambiguity only for pulses with pure Gaussian intensity profiles and pure linear chirp, i.e., the sign of the chirp cannot be retrieved. This is rarely a problem for experimental data, however, because even small distortions in the pulse shape or phase permit unambiguous retrieval. For pulses that are close to this case we found it nevertheless practical to run the FROG algorithm twice on the experimental data, using a time-reversed version of the original noise as an initial guess for the field in the second run. Comparison of the agreement between the measured and reconstructed FROG traces then identifies the field with the correct sign of the chirp. A potentially more serious ambiguity is that THG FROG can retrieve the relative phase of well-separated double pulses modulo $2\pi/3$ only; i.e., a double pulse consisting of individual pulses of equal strength that are separated by more than approximately twice the FWHM produces the same THG FROG trace when the individual pulses are in phase or have a relative phase difference of $2\pi/3$ or $4\pi/3$. In comparison, it should be noted that SHG FROG can retrieve the relative phase of double pulses only modulo π.

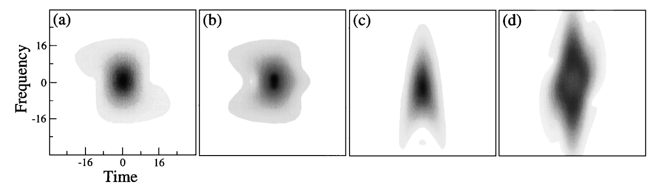

Fig. 2. Examples of simulated THG FROG traces for pulses with Gaussian intensity profiles and different phase distortions: (a) spectral quartic phase, (b) spectral cubic phase, (c) temporal cubic phase, (d) self-phase modulation.

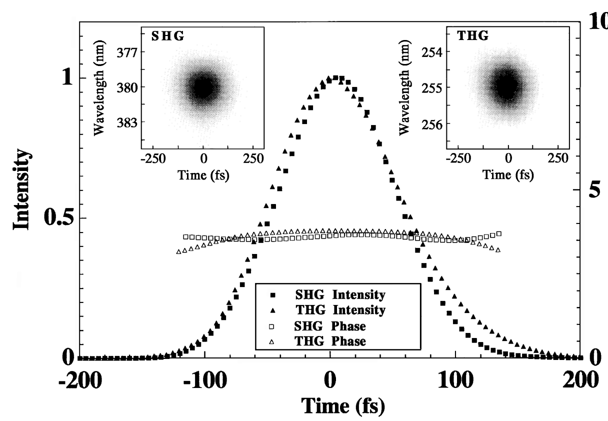

Fig. 3. Retrieved intensity and phase for a nearly transform-limited oscillator pulse. The insets show the corresponding experimental SHG FROG and THG FROG traces, both approximately symmetrical in time.

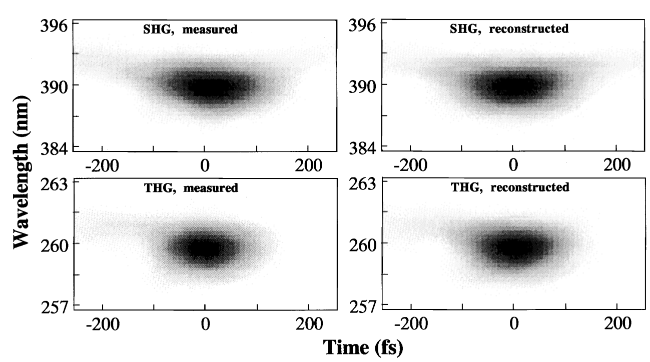

Fig. 4. Measured and reconstructed SHG FROG traces and THG FROG traces for a clearly non-transform-limited pulse.

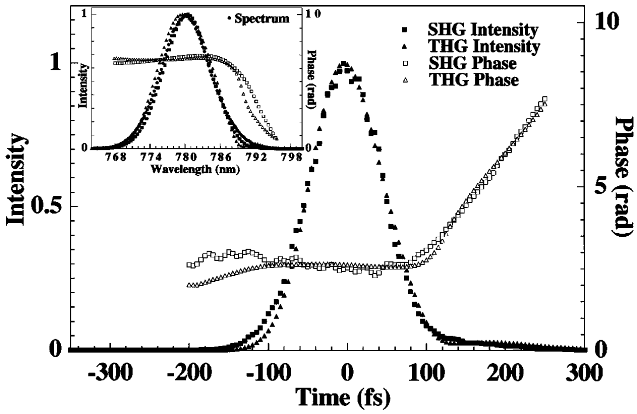

Fig. 5. Retrieved intensity and phase in the time domain for the pulse corresponding to Fig. 4. The inset shows the independently measured spectrum in comparison with the retrieved intensity and phase in the frequency domain.

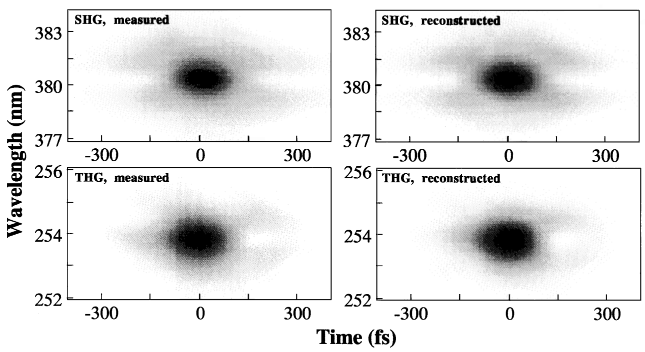

Fig. 6. Measured and reconstructed SHG FROG traces and THG FROG traces for a strongly distorted pulse.

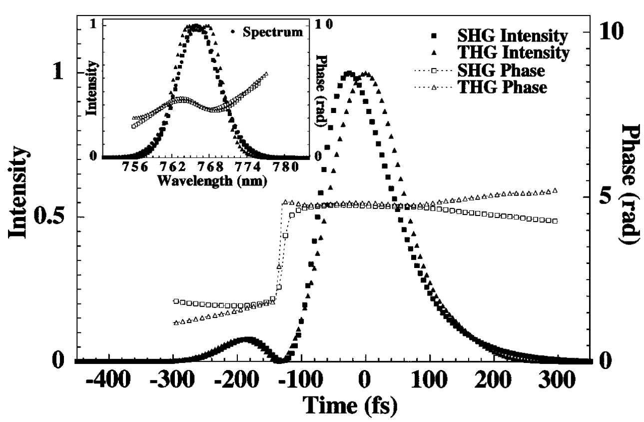

Fig. 7. Retrieved intensity and phase for the pulse corresponding to Fig. 6. The inset shows the independently measured spectrum and the retrieved intensity and phase in the frequency domain.

Figure 5 shows the retrieved intensity and phase of the pulse corresponding to the traces in Fig. 4. The retrieved fields for the SHG and THG FROG measurements agree well, showing the characteristic increase of the phase in the wing and the slow decrease of the intensity for delay times larger 100 fs. The increase of the phase in the time domain corresponds to the decrease in the frequency domain, as shown in the inset of Fig. 5, and both FROG measurements also agree well with the independently measured spectrum.

By reflecting the Ti:sapphire laser beam off a multilayer coated dielectric mirror at an angle of ~50°, we were also able to investigate laser pulses with stronger phase distortions. Figure 6 shows the SHG and THG FROG traces of a pulse that was distorted in this way. Also in this case, good agreement between measured and reconstructed traces is obtained for the SHG FROG measurement as well as for the THG FROG measurement. Figure 7 shows the retrieved fields for this pulse: The characteristic features, i.e., the small satellite pulse and the π-phase jump between the main pulse and the satellite pulse, are clearly reconstructed by both FROG measurements. The slight variation in the position of the intensity maximum of the main pulse is probably caused by a drift of the laser during the measurements, which can also explain the observed deviations between the intensity retrieved from the THG FROG measurement on the one hand and the independently measured spectrum and the retrieved intensity of the SHG FROG measurement on the other. Further evidence of drift is the asymmetry in the SHG FROG trace.

We conclude that FROG employing surface third-harmonic generation as a nonlinear effect is a suitable technique for the measurement of ultrashort pulses directly from laser oscillators. Unlike SHG FROG, THG FROG has no practical direction-of-time ambiguity, making it therefore superior for applications that require pulse characterization without this ambiguity, where the pulse energy is too weak to permit the use of polarization-gate or self-diffraction beam geometries. In addition, surface THG FROG might be preferable for extremely short pulses, which would require extraordinary thin crystals for SHG FROG measurements.

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