Ultrashort optical waveform measurements using frequency-resolved optical gating

Greg Taft, Andy Rundquist, Margaret M. Murnane, and Henry C. Kapteyn

Department of Physics, Washington State University, Pullman, Washington 99164-2814

Kenneth W. DeLong and Rick Trebino

Combustion Research Facility, Sandia National Laboratories, Livermore, California 94551

Ivan P. Christov

Faculty of Physics, Sofia University, Sofia, Bulgaria

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We measure the intensity and phase of ultrashort pulses from a self-mode-locked Ti:sapphire laser using the recently developed technique of frequency-resolved optical gating. These results represent to our knowledge the shortest complete optical waveform characterization measurements performed to date. We also verify recent theoretical calculations that predict that the main limitation on the pulse duration from these lasers is the presence of uncompensated higher-order dispersion.

In recent years there has been very rapid progress in the generation of ultrashort pulses directly from mode-locked solid-state lasers.[1–4] Stable pulses of 5-nJ energy and durations of <10 fs can be generated routinely from a simple laser oscillator.[5] Efforts to describe the ultimate limits of operation of these lasers,[6] and the pulse-formation processes, have produced different models and predictions for the pulse electric field, that is, the time-dependent pulse intensity and phase.[7,8] Unfortunately, these models are difficult to distinguish experimentally because the predicted electric fields have nearly identical intensity autocorrelations and spectra. However, the newly developed technique of frequency-resolved optical gating[9–12] (FROG) can be used to distinguish between the models. In this Letter we present measurements of the electric field of pulses in the range of 10–15 fs, using second-harmonic generation (SHG) FROG. SHG FROG is experimentally similar to conventional autocorrelation, except that in SHG FROG the autocorrelation signal is spectrally resolved.[9–12] The signal energy is plotted as a function of delay and wavelength to form the so-called FROG trace, a type of spectrogram of the pulse. Our measurements have allowed us to verify model predictions and thus the predicted limits on the output of a Ti:sapphire laser.[5,7]

Because the bandwidth of the pulses in this study could be extremely large—as much as 177 nm—their measurement presents a considerable challenge to any technique, including simple autocorrelation as well as FROG. For example, the phase-matching bandwidth of any nonlinear medium must be at least as large as that of the pulse. Also, the efficiency of any dispersive elements and the response of the detector must be flat or well characterized over a large spectral range. Nonlinear scaling of the frequency interval over this broad range must also be taken into account. In addition, the beam spot size and crossing angle at the nonlinear medium must both be sufficiently small that geometrical temporal smearing effects of even a few femtoseconds do not occur. Finally, group-velocity dispersion must also be negligible to within a few femtoseconds.

Fortunately, SHG FROG, like its simpler version, conventional SHG autocorrelation, involves very little material dispersion, especially since dispersion after the doubling crystal does not affect the measurement. To minimize dispersion, we used a 40%/40% near-IR beam splitter on a thin (5-μm) pellicle substrate. As a result, the dispersion in our measurement apparatus was negligible, even for a 10-fs pulse. A diagram of the SHG FROG apparatus is shown in Fig. 1. To attain a broad doubling bandwidth, we used a thin 35 ± 5 μm KDP crystal. A fused-silica prism, rather than a grating, dispersed the doubled autocorrelation signal, for efficient operation at short wavelengths. Minimal temporal smearing was achieved by using a beam spot size at the SHG crystal of 30 μm and a crossing angle of 2 deg. However, our CCD camera response was not flat at short wavelengths; it decreased significantly in the near UV at

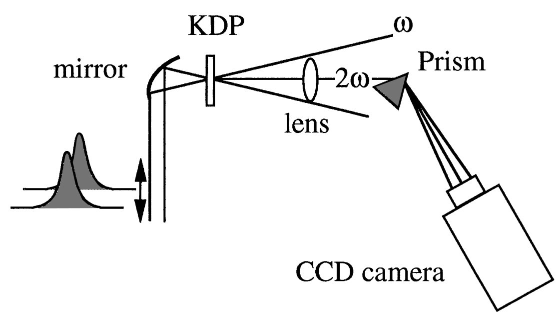

Fig. 1. Experimental setup for the SHG FROG technique.

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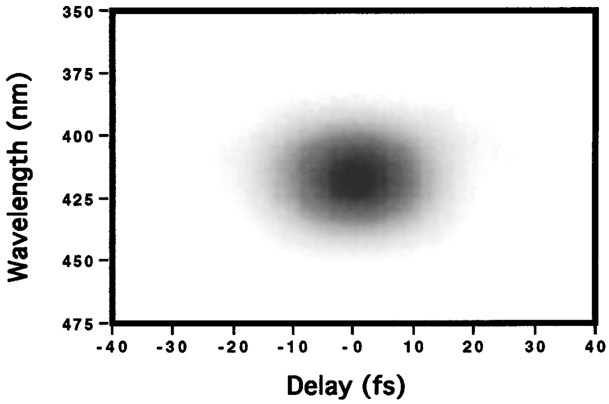
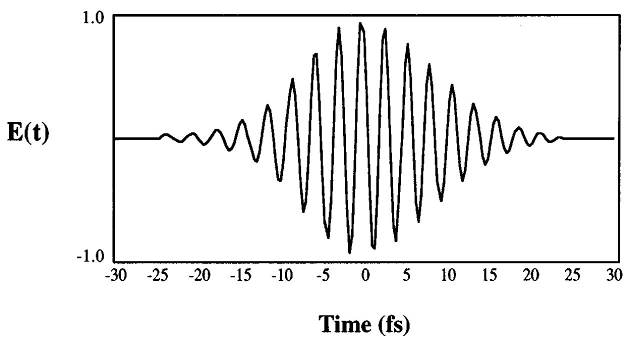


(a)



(b)

Fig. 2. (a) FROG trace of a 13-fs near-transform-limited pulse; (b) retrieved electric field of the pulse measured in (a).

≈390 nm, and we corrected the data for nonuniform spectral response. One arm of the autocorrelator was moved continuously by an encoded micrometer (Oriel Corporation). A 20-cm relay lens focused the SHG signal onto a CCD camera (Cohu 4900), with the entrance window removed. We acquired the SHG spectrum at ~5-Hz repetition rate, using a frame grabber and Macintosh IIci computer running LABVIEW. With the micrometer running at 0.5 μm/s, spectra could be taken at 0.67-fs time delay intervals. We spectrally calibrated the homemade spectrometer by comparing the second-harmonic signal at the CCD camera with that observed by a scanning spectrometer, as the peak of the spectrum was moved by spectrally clipping the output beam with a razor blade.

A self-mode-locked Ti:sapphire laser, operating with a center wavelength of 850 nm, was used to generate the ultrashort pulses, as described in a previous Letter.[5] For these measurements, we found it most convenient to extract the output of the laser through a thin (3-mm) output coupler at the spectrally dispersed end of the laser.[3] The pulses were then recompressed to their shortest pulse width by being passed through a second prism pair, placed at an identical separation to that inside the laser, before being sent to the measurement apparatus.

In previous studies the SHG FROG technique had been shown to work well and to reconstruct faithfully pulses as short as 90 fs.[11] In this study we made two pulse measurements: a 13-fs transform-limited pulse, and a sub-10-fs pulse with a two-peaked spectrum. The FROG trace in the first case is shown in Fig. 2(a), with the retrieved intensity and phase shown in Fig. 2(b). The pulse has an intensity FWHM of 13 fs and is to our knowledge the shortest optical pulse (or pulse of any kind) ever to be fully characterized. As a check on the retrieved waveform, we compared the independently measured spectrum of the pulse, along with the spectrum and phase of the pulse retrieved by the FROG algorithm. These spectra are in close agreement, with only a 3% difference in their FWHM's (88.9 nm). The FWHM of the intensity autocorrelation of the retrieved pulses (18.6 fs) also matches the independently measured intensity autocorrelation of the laser to within 5%. The pulse is near transform limited: the spectral phase of the retrieved pulse is relatively flat, and the time–bandwidth product is 0.5.

We also acquired data for the limiting pulse width obtainable from the laser, which corresponds to a spectral FWHM of 177 nm. For this pulse, the spectrum changed from a smooth single-peaked shape to a characteristic double-peaked profile, centered at 850 nm. Figure 3 shows an SHG FROG trace obtained for this mode of operation of the laser. The small-intensity lobes on both sides of the main trace are interference features that are due to the double-peaked spectrum. The extremely wide bandwidth of this pulse posed considerable difficulties in our measurement. This is shown by the fact that the frequency marginal of the FROG trace did not match the autoconvolution of the spectrum of the original pulse, as it should.[12] The frequency marginal is the spectrum of the FROG signal integrated over all time delays. This lack of agreement may be due to the spectral response of the camera, inaccurate spectral calibrations, fluctuations in the laser spectrum during the measurement, or distortions in the SHG process with such short pulses. An attempt to retrieve the pulse from these distorted data yielded a pulse with a FWHM of 9 fs. As the spectrum of this retrieved pulse was narrower than the actual spectrum, we suspect that the true pulse is even shorter.

Despite the imperfect nature of the data, the SHG FROG trace still provides valuable insight into the operation of the laser at these short pulse durations.

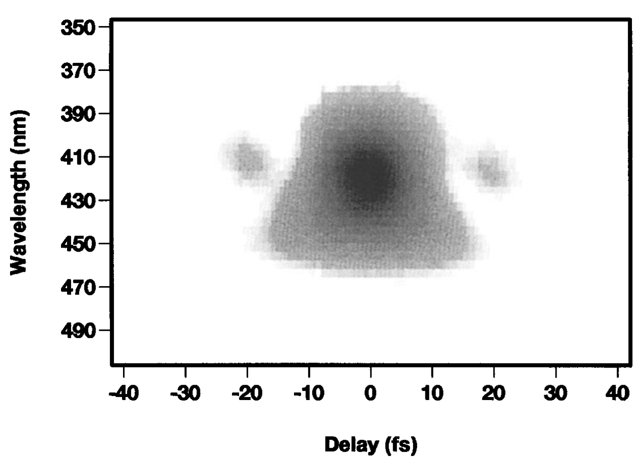


Fig. 3. Experimental FROG trace of a sub-10-fs pulse with a double-peaked spectrum.

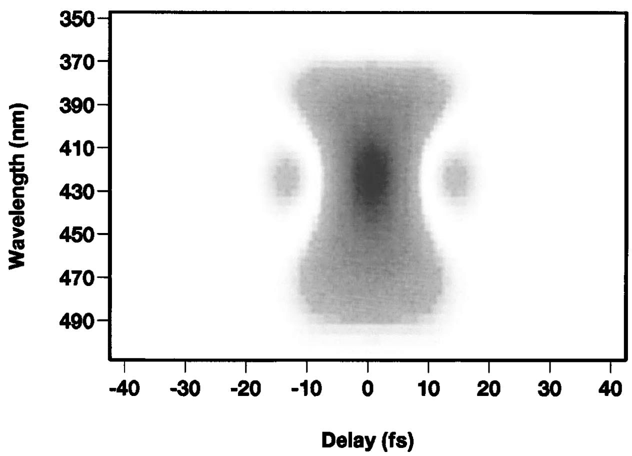


Fig. 4. Predicted FROG trace of a sub-10-fs pulse with a double-peaked spectrum, limited by higher-order dispersion.

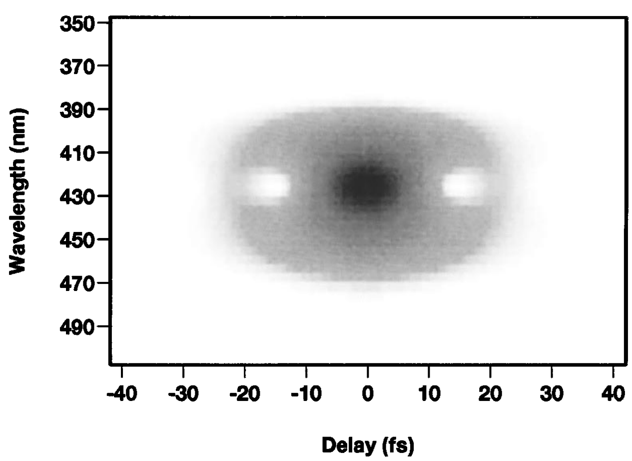


Fig. 5. Predicted FROG trace of a sub-10-fs pulse with a double-peaked spectrum, limited by coherent ringing.

Specifically, two different numerical theories have recently been developed to explain the characteristic broad double-peaked spectra that can be generated from Ti:sapphire lasers.[6–8] The first predicts that this shape is due to the presence of uncompensated dispersion in the laser.[7] The intracavity prism glass can be adjusted to change the intracavity dispersion, which simultaneously changes the output spectrum from a smooth single-peaked profile to a double-peaked profile. The single-peaked spectrum corresponds to the case of uncompensated fourth-order dispersion only, while the double-peaked spectrum corresponds to a combination of both second- and fourth-order dispersion. A numerical model showed that stable pulses can be self-consistently obtained from a cavity with such dispersion characteristics. The second model suggests that the double-peaked spectrum arises from coherent ringing in the laser gain medium, which gives rise to a pulse with postpulses.[8] If the latter theory were correct, it would also represent a fundamental limit to the pulse width that could be generated from a Ti:sapphire laser. Interestingly, while these theories predict quite different pulse electric fields, the predicted spectra and autocorrelations are nearly identical, and both agree well with the experimentally measured quantities.

To compare the measured pulses with these predictions, we used the electric fields predicted by these models to calculate the predicted FROG traces. For the case of a fourth-order dispersion limit on the laser, the FROG trace is shown in Fig. 4. For comparison, in Fig. 5 we also show the FROG trace predicted by the coherent ringing theory.[8] Our experimental measurements, shown in Fig. 3, are in qualitative agreement with the predictions shown in Fig. 4, with both theory and experiment showing islands of higher intensity on either side of the main peak. In contrast, the FROG trace predicted by the coherent ringing theory, shown in Fig. 5, has lobes of lower intensity on either side of the main peak. We note that in the case of Fig. 4 the spectral bandwidth of the predicted pulse is larger than we observe experimentally, because the models do not include spectral clipping by the interval prism pair[5]; thus the reduced spectral width in Fig. 3 compared with that in Fig. 4 is expected. Therefore our results confirm that the pulse width in Ti:sapphire is limited by uncompensated higher-order dispersion in the cavity.

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