Amplitude and chirp characterization of high-power laser pulses in the 5-fs regime

Z. Cheng, A. Fürbach, S. Sartania, M. Lenzner, Ch. Spielmann, and F. Krausz

Institut für Angewandte Elektronik und Quantenelektronik, Technische Universität Wien, Gusshausstrasse 27, A-1040 Vienna, Austria

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Frequency-resolved optical gating (FROG) based on second-harmonic generation has been demonstrated to be capable of high-fidelity measurement of the electric-field envelope and of the temporal evolution of the instantaneous carrier frequency of 0.1-TW 5-fs pulses without the need for any correction for systematic experimental errors. At a 1-kHz repetition rate, pulse energies of a few microjoules are sufficient for reliable FROG characterization of pulses with durations down to the single-cycle regime. The results obtained reveal that carefully designed hollow-fiber chirped-mirror compressors are able to deliver high-power sub-10-fs pulses with a smooth Gaussianlike leading edge that has an intensity contrast of $\approx 10^{-2}$.

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With the advent of chirped multilayer dielectric mirrors[1–4] precise dispersion control over unprecedented bandwidths became feasible in femtosecond laser systems. As a result of this progress, near-infrared laser pulses self-phase modulated in single-mode optical fibers[5] or gas-filled hollow waveguides[6] could be compressed to pulse durations shorter than 5 fs at a carrier wavelength of $\approx 0.8\ \mu$m characterized by an optical period of ≈ 2.7 fs. These pulses with durations approaching the light oscillation cycle are now available with nanojoule energies at megahertz repetition rates[5] as well as at millijoule energy levels and kilohertz rates.[7] Just as in the case of longer pulses, accurate knowledge of the pulse shape and the possible chirp that is carried is desirable in many spectroscopic as well as high-field applications.

For their characterization and theoretical description, light-wave packets are decomposed into a carrier and an envelope, $E(t) = A(t)\exp[-i(\omega_0 t + \varphi_0)] + \text{c.c.}$, where the carrier frequency ω_0 is, by definition, the first moment of the spectral intensity distribution of the wave packet and $A(t) = |A(t)|\exp[-i\varphi(t)]$ is the complex envelope of the pulse incorporating full information on both the real amplitude envelope $|A(t)|$ and a possible time-dependent frequency shift, $\Delta\omega(t) = \mathrm{d}\varphi/\mathrm{d}t$, referred to below as chirp. It was recently shown that this decomposition, which provides the basis for currently used pulse-characterization techniques, is physically meaningful down to the single-cycle regime,[8] justifying the extension of these characterization techniques down to pulse durations τ_p approaching the carrier oscillation period $T_0 = 2\pi/\omega_0$.

Frequency-resolved optical gating[9] (FROG) is one of the most widely used techniques for measuring $A(t)$ and $\Delta\omega(t)$ of ultrashort pulses. FROG based on second-harmonic generation (SHG) has been extended to the 10-fs regime[10] and more recently to pulse durations as short as 4.5 fs at megahertz repetition rates and nanojoule pulse energy levels.[11] In this Letter we present, for what is to our knowledge the first time, FROG measurements of submillijoule pulses in this time domain. Our results demonstrate that (i) careful design of the SHG-FROG apparatus allows measurement of the amplitude and chirp of high-energy near-infrared optical pulses down to ≈ 5 fs without the need for corrections for systematic errors and down to ≈ 3 fs with corrections for time smearing and the finite phase-matching bandwidth and (ii) reliable pulse characterization in this time domain can be performed at average power levels of a few milliwatts at a 1-kHz repetition rate. Most importantly, (iii) the data presented provide what is believed to be the first evidence for the feasibility of generating 0.1-TW pulses with good quality in the few-cycle regime. In fact, our FROG measurement yields 5-fs pulses exhibiting a smooth Gaussianlike intensity envelope on the leading edge with a contrast of $\approx 10^{-2}$.

SHG-FROG measures the intensity spectrum $S(\omega, \tau)$ of second-harmonic (SH) radiation produced in a thin nonlinear crystal by two identical replicas of the pulse as a function of the delay τ between them. To obtain the expression for the SHG-FROG signal, we first solve the slowly evolving wave master equation,[8] which is valid down to the single-cycle regime, for the SH pulse, using the nonlinear polarization source term introduced by Weiner.[12] Assuming a thin crystal, changes in the complex envelope $A(t)$ of the laser pulse during propagation (owing to depletion and dispersion) are neglected. We also neglect the frequency dependence of the second-order nonlinear susceptibility $\chi^{(2)}$. Furthermore, the frequency dependence of the (nearly parallel) wave vectors of the fundamental and the SH waves are approximated with linear functions, $k_f(\omega) \approx k_f(\omega_0) + k_f'(\omega_0)(\omega - \omega_0)$ and $k_s(\omega) \approx k_s(2\omega_0) + k_f'(2\omega_0)(\omega - 2\omega_0)$, respectively. For type I phase-matching SHG these simplifications yield

$$S(\omega, \tau) \propto \frac{|\chi^{(2)}|^2\omega^2}{{n_s}^2}\,\mathrm{sinc}^2\left\{[\Delta k_0 + (\omega - 2\omega_0)\Delta k_1]\frac{L}{2}\right\} \times \left|\int_{-\infty}^{\infty}\mathrm{d}\omega' A(\omega - \omega')A(\omega')\exp(i\omega'\tau)\right|^2, \quad (1)$$

where n_s is the refractive index of the SH wave at $2\omega_0$, $\Delta k_0 = 2k_f(\omega_0) - k_s(2\omega_0)$, $\Delta k_1 = k_f'(\omega_0) - k_s'(2\omega_0)$, L

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is the length of the nonlinear medium, and $A(\omega)$ is the Fourier transform of the complex envelope $A(t)$ of the laser pulse. As revealed by relation (1), a limited phase-matching bandwidth appears owing to group-velocity mismatch between the fundamental and the SH waves, restricting the bandwidth of the SHG-FROG apparatus. The approximations made above are justified as long as the spectral width $\Delta\omega$ (FWHM of the spectral intensity) of the laser pulses satisfies $\Delta\omega \leq 1/\Delta k_1 L$. This condition ensures that the SH spectrum is not significantly distorted by the finite phase-matching bandwidth (PMB).[13]

Figure 1 shows the calculated SH spectrum (for infinite PMB) of a pulse with a Fourier-limited duration of 5 fs that was produced with an energy of 0.5 mJ at a repetition rate of 1 kHz by the system described in Ref. 7. The solid curve depicts the frequency dependence of the function in front of the integral in relation (1) for a 20-μm-thick KDP crystal cut for type I phase matching, which is used in our FROG correlator. Group-delay mismatch between the fundamental and the SH waves appears to introduce little distortion of the FROG signal even at 5 fs for this crystal. For comparison, Fig. 1 shows the same function for a 20-μm BBO crystal (type I phase matching), which has a much smaller PMB than KDP. It is interesting to note that the tuning wavelength (at which the phase-velocity mismatch is zero) was set equal to 950 nm, and the peaks of the FROG spectral transmittivity curves are blueshifted to $\approx$390 nm by the ω^2 factor in relation (1) for these broad PMB's.

In addition to the PMB, the finite beam size in combination with the noncollinear geometry also modifies the FROG signal, as was pointed out in Ref. 10. For a Gaussian beam profile and pulse shape the noncollinear SHG-FROG signal is modified by replacement of τ with $\tau/(\delta + 1)$, where the error introduced by time smearing is $\delta = (1 + \delta t^2/\tau_p{}^2)^{1/2} - 1$, τ_p is the pulse duration, $\delta t \approx \sqrt{2 \ln 2}\, d/\omega_0 w_m$, where w_m and d are the Gaussian beam radius and the separation of two laser beams on the focusing mirror, respectively. As shown by Baltuska *et al.*,[11] the minimum value of this time-smearing error, which is realized when the two laser beams are separated by $2w_m$ on the focusing mirror, depends only on the (angular) carrier frequency ω_0 as $\delta t_{\min} \approx 2\sqrt{2 \ln 2}/\omega_0$. In our experiments $\delta t = 1.6$ fs.

Figure 2 depicts the relative systematic errors introduced by time smearing and the finite PMB in our FROG apparatus, as obtained for Gaussian pulses. For pulse durations longer than 5 fs the relative errors are less than 5%; hence both the SHG spectral transmittivity function in relation (1) and the time-smearing effects can be neglected. For $\tau_p < 5$ fs, the full expression given in relation (1) must be considered and corrected iteratively for the time-smearing effect.

Our SHG-FROG correlator uses the same optical components and geometry (apart from a slight change from collinear to noncollinear recombination of the laser beams) as those used in our interferometric autocorrelator designed for high-fidelity measurement of sub-10-fs pulses.[14] The beams (impinging at near-normal incidence) were focused by a spherical mirror with a focal length of 7.5 cm onto the 20-μm KDP crystal. The SH signal was carefully collected and imaged onto the entrance slit of a spectrometer equipped with a 12-bit 1024-pixel CCD array. The recorded SH spectra were corrected for the spectrometer response. The time delay between the pulses was changed in 0.4-fs steps by a translation stage driven by a linear motor. The delay was calibrated with an interferometric setup employing a He–Ne laser. The overall delay scan range was 240 fs. The data were processed by a commercially available retrieval algorithm (Femtosoft Technologies).

The pulses characterized by this SHG-FROG apparatus were delivered by the hollow-fiber chirped-mirror high-power pulse compressor seeded with 25-fs 1.2-mJ pulses at a 1-kHz repetition rate.[7] The energy of the compressed pulses was ~0.55 mJ, 1% of which was directed into either the FROG system or an interferometric autocorrelator by the Fresnel reflection off a thin glass plate. The spectrum of the compressed pulses was also monitored with a spectrometer. The contour plots of the measured and the retrieved FROG traces are shown in Fig. 3. To resolve the inherent time-reversal ambiguity of SHG-FROG we subsequently passed the pulses through a thin fused-silica plate and broadened them by some 50%. FROG measurement of these chirped pulses allowed us to recognize the leading and trailing edges of the unchirped pulses, yielding the temporal and the

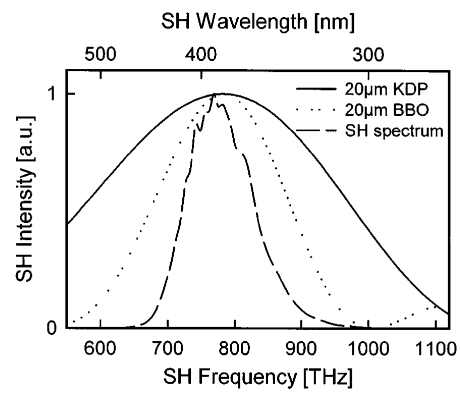

Fig. 1. SH spectrum $S(\omega, 0)$ calculated from the measured spectrum of the 5-fs pulses, assuming the absence of chirp on the pulse and infinite PMB in the SHG crystal (dashed curve), and $S(\omega, 0)$ originating from 20-μm-thick KDP and BBO crystals in the case of an infinite laser bandwidth.

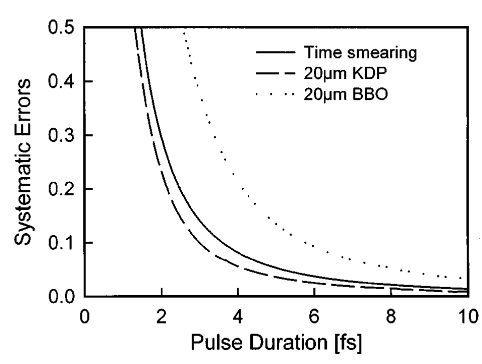

Fig. 2. Relative systematic errors introduced by time smearing and the finite PMB for 20-μm-thick KDP and 20-μm-thick BBO crystals.

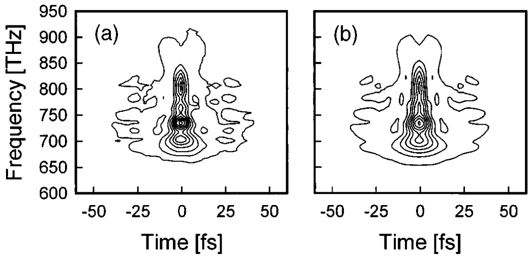

Fig. 3. (a) Measured and (b) retrieved FROG traces, $S(\omega, \tau)$, of the high-power pulses. The contour plots depict isointensity lines at the levels of $0.01 \times S_p$ and integer multiples of $0.1 \times S_p$, where S_p is the peak of $S(\omega, \tau)$.

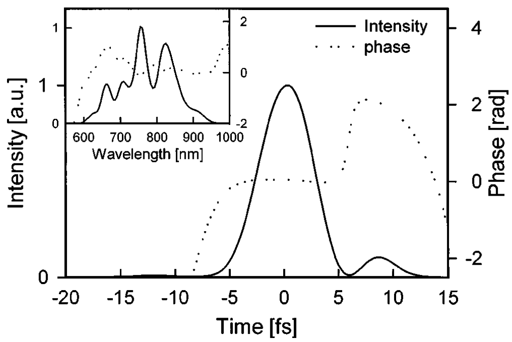

Fig. 4. Pulse shape, spectrum, and phases retrieved from the FROG trace shown in Fig. 3(b).

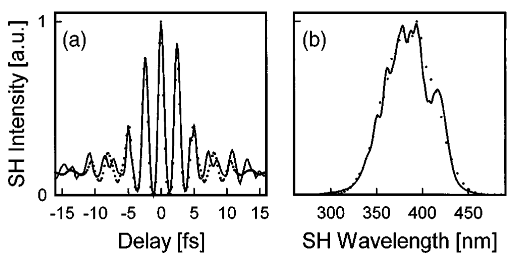

Fig. 5. (a) Measured interferometric autocorrelation trace (solid curve) and the calculated trace (dotted curve) from the retrieved FROG trace. (b) FROG frequency marginal (dotted curve) and the autoconvolution (solid curve) of the measured fundamental spectrum.

spectral intensities and phases of the measured pulses, as depicted in Fig. 4. Note that the phases were set arbitrarily equal to zero at the temporal and spectral center of the pulse. The excellent pulse quality on the leading edge makes these pulses ideally suitable for high-field experiments (e.g., high-order harmonic generation).

We obtained the results of the FROG characterization by disregarding the SHG spectral transmittivity function (i.e., the finite PMB) and neglecting time smearing, i.e., without any correction for systematic errors, obtaining a pulse duration of 5.5 fs. The good agreement between the autoconvolution of the fundamental spectrum and the FROG marginal[15] in Fig. 5(a) as well as those of the measured and the calculated interferometric autocorrelations [Fig. 5(b)] justify neglecting transmittivity and time smearing, in accordance the results shown in with Fig. 2. Correction for these small systematic errors results in a hardly recognizable change in the pulse shape and a pulse duration of 5.3 fs. Correcting for the finite PMB and geometric time smearing allows this system to characterize pulses down to 3 fs in duration, i.e., in the single-cycle regime.

It is important to note that these data do not constitute a complete characterization of these few-cycle laser pulses, because the absolute carrier phase φ_0 is not accessible. This parameter plays a significant role in the interaction of few-cycle intense laser pulses with matter[16] and hence needs to be measured if high-field processes induced with few-cycle light pulses are to be controlled. Unfortunately, a change in the carrier phase affects only the low-frequency long-wavelength spectral components far off the carrier wavelength, even in the 4–5-fs regime. As a consequence, access to the absolute phase would require extraordinarily broad bandwidth and high dynamic range in the FROG measurement. Seeking alternative ways of determining the phase therefore appears to be desirable.

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References

1. R. Szipöcs, K. Ferencz, Ch. Spielmann, and F. Krausz, Opt. Lett. **19,** 201 (1994).
2. F. X. Kärtner, N. Matuschek, T. Schibli, U. Keller, H. A. Haus, C. Heine, R. Morf, V. Scheuer, M. Tilsch, and T. Tschudi, Opt. Lett. **22,** 831 (1997).
3. R. Szipöcs and A. Köhazi-Kis, Appl. Phys. B **65,** 115 (1997).
4. G. Tempea, F. Krausz, Ch. Spielmann, and K. Ferencz, IEEE J. Sel. Topics Quantum Electron. **4,** 193 (1998).
5. A. Baltuska, Z. Wei, M. S. Pshenichnikov, D. A. Wiersma, and R. Szipöcs, Opt. Lett. **22,** 102 (1997).
6. M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, Ch. Spielmann, S. Sartania, and F. Krausz, Opt. Lett. **22,** 522 (1997).
7. S. Sartania, Z. Cheng, M. Lenzner, G. Tempea, Ch. Spielmann, F. Krausz, and K. Ferencz, Opt. Lett. **22,** 1562 (1997).
8. T. Brabec and F. Krausz, Phys. Rev. Lett. **78,** 3282 (1997).
9. D. J. Kane and R. Trebino, IEEE J. Quantum Electron. **29,** 571 (1993).
10. G. Taft, A. Rundquist, M. M. Murnane, I. P. Christov, H. C. Kapteyn, K. W. DeLong, D. N. Fittinghoff, M. A. Krumbügel, J. N. Sweetser, and R. Trebino, IEEE J. Sel. Topics Quantum Electron. **2,** 575 (1996).
11. A. Baltuska, M. S. Pshenichnikov, and D. A. Wiersma, Opt. Lett. **23,** 1474 (1998).
12. A. M. Weiner, IEEE J. Quantum Electron. **19,** 1276 (1983).
13. To within the approximation described in the text, the expression given by relation (1) is more accurate than a similar simplified expression derived in Ref. 12, in that it accounts for all sum-frequency combinations $\omega_1 + \omega_2$ yielding contributions to a given detected spectral component $S(\omega, \tau)$, in contrast with the expression summarized by Eqs. (3)–(5) in Ref. 12
14. Ch. Spielmann, L. Xu, and F. Krausz, Appl. Opt. **36,** 2523 (1997).
15. K. W. DeLong, D. N. Fittinghoff, and R. Trebino, IEEE J. Quantum Electron. **32,** 1253 (1996).
16. F. Krausz, T. Brabec, M. Schnürer, and Ch. Spielmann, Opt. Photon. News **9**(7), 46 (1998).