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Phase-stabilized 4-fs pulses at the full oscillator repetition rate for a photoemission experiment

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ABSTRACT Compressing pulses of a mode-locked extended-cavity Ti:sapphire laser using a standard single-mode fiber and tilted-front-interface chirped mirrors yields phase-stabilized 4-fs, 3-nJ light pulses at the full, 24-MHz, repetition rate. The demonstrated source paves the way towards exploring interactions sensitive to the carrier-envelope-offset phase, such as, for example, photoemission from solid targets at moderate intensities ($\sim 10^{12}$ W/cm^2).

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So far, pulses closely approaching the single-cycle limit (i.e. 2.7 fs for a carrier wavelength of 800 nm) have only been demonstrated at reduced repetition rates, either with kHz chirped pulse amplifier systems [1] or with cavity-dumped oscillators [2]. It was shown that the carrier-envelope phase (CEP) of ultra-short pulses changes randomly on a microsecond time scale [3] and thus can only be stabilized in MHz-repetition-rate systems. Control of the CEP in MHz-repetition-rate lasers was restricted so far to the evolution of the CEP [4, 5] while the absolute, instantaneous value of this parameter could not be accessed. Theoretical predictions presented in this article indicate convincingly (and in full agreement with previous calculations [6, 7]) that the phase sensitivity of strong field interactions [8] increases dramatically for sub-5-fs pulses. Thus, the availability of sub-5-fs pulses at repetition rates of tens of MHz (i.e. repetition rates at which the stabilization of CEP is feasible) is a prerequisite for the measurement of the absolute value of the CEP. A source that closely fulfills these requirements is described in this paper. The 25-MHz laser system first presented in [5] (consisting of a powerful Ti:sapphire oscillator and a single-mode fiber) was equipped with a tilted-front-interface chirped mirror (TFICM) [9, 10] compressor that allows compressing the octave-spanning CEP-stabilized pulses to 4 fs. The capability of this source to provide access to the absolute value of the CEP is substantiated by calculations of phase-sensitive photoemission from metal surfaces.

Chirped mirrors are currently used as standard components in ultra-broadband compressors. However, as the relative bandwidth $\Delta\omega/\omega_0$, where ω_0 denotes the central frequency, is increased above 0.3, they exhibit large group-delay dispersion (GDD) oscillations [11]. Recently, an approach has been found that allows us to significantly increase the bandwidth of chirped mirrors. The impedance mismatch at the front interface of the mirror (that represented so far the bandwidth-limiting factor) can be suppressed almost completely using a thin, non-plane-parallel dense optical material as the medium of incidence. Two slightly different implementations of this concept have been dubbed 'back-side-coated' (BASIC) mirrors [9] and 'tilted-front-interface' (TFI) mirrors [10].

The core of the laser system is an extended-cavity, mirror-dispersion-controlled Kerr-lens mode-locked oscillator [12]. The Ti:sapphire laser generates a train of 9-fs, 16-nJ pulses at a repetition rate of 24 MHz. These pulses are spectrally broadened in a 1.5-mm standard single-mode optical fiber ('3M', core diameter 2.8 μm). The fiber length is chosen according to design criteria derived elsewhere [13]. The chirp introduced by self-phase modulation in the fiber together with fiber and air dispersion can be compensated up to the third order by a mirror compressor consisting of 12 identical TFI mirrors manufactured in one coating run. These mirrors [10] have an overall reflectance exceeding 97% between 500 and 1050 nm and controlled GDD over the same range. The compact all-chirped-mirror compressor imposes virtually no CEP jitter on the pulses, in contrast to a compressor containing prisms and/or gratings.

In contrast to our previous work [12] where the full pulse energy was needed to stabilize the CEP, we now couple only 15% of the oscillator output into a photonic crystal fiber and then send the spectrally broadened output to a phase-sensitive, non-linear interferometer. The stabilization loop is implemented in the same way as in [12]. The major part of the pulse energy (85% of the oscillator output, i.e. 3 nJ) is contained in phase-stabilized 4-fs pulses available for experiments.

Characterization of octave-spanning pulses is a challenging problem. Methods like FROG and SPIDER are

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difficult to implement with weak sub-5-fs pulses since retrieving the spectral phase is plagued by the poor signal to noise ratio, particularly in the spectral wings. We rely on a somewhat less involved and more robust technique: pulse reconstruction from the power spectrum and the second-order autocorrelation function (ACF) only. It has been shown that the spectral phase is uniquely determined by the ACF and the power spectrum [14]. Although there are debates on the accuracy of this approach [15], successful pulse reconstruction has been demonstrated even in the presence of noise [16].

We used a dispersion-free autocorrelator for recording the second-order interferometric ACF. Polarization filtering was employed for separating the second harmonic from the fundamental field, since the spectra of the two signals overlap. The second harmonic is generated in a 9-μm BBO crystal phase-matched for 800 nm. Computer simulations show that propagation effects (phase matching, group-velocity mismatch, and dispersive broadening) can be neglected in the evaluation of the pulse duration from the measured autocorrelation trace. The power spectrum and the autocorrelation function of the compressed pulses are shown in Figs. 1 and 2 respectively.

The algorithm we use for pulse reconstruction is analogous to the PICASO algorithm [16, 17], where the spectral phase is retrieved by means of minimization of the difference between the measured ACF and that calculated from the measured power spectrum. This approach leaves some freedom in the mathematical representation of the spectral phase being optimized, since there are several ways of mapping a function of frequency onto a set of optimization parameters. In the original PICASO algorithm the phase is represented by a polynomial expansion and the coefficients of this expansion are optimized in order to retrieve the spectral phase. We compare this representation with two other procedures. In one approach spline interpolation is used to construct the spectral phase as a smooth function from discrete values of the phase known at fixed frequencies; these values play the role of optimization parameters. In the second optimization scheme the optimized parameters are the second-order derivatives of the phase. In both cases ACFs calculated from the retrieved pulses match the measured ACF better than the polynomial representation of the spectral phase (Fig. 3). This can be explained by the fact that the uncompensated GDD contains fast oscillations introduced by chirped mirrors, which the polynomial expansion of the spectral phase may not be able to track. The FWHM of the retrieved pulses varies in these three cases from 3.7 to 4.3 fs, while the FWHM of the bandwidth-limited pulse with the measured spectrum would be equal to 3.4 fs. Figures 1, 2, and 4 depict the reconstruction that provides the best agreement between measured and computed ACFs. The pulse duration evaluated from this best fit was 3.8 fs.

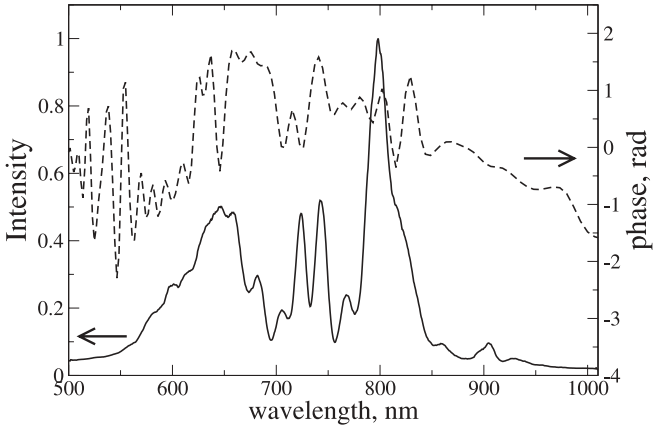

FIGURE 1 Spectrum of the pulses exiting the compressor (*solid line*) and the retrieved spectral phase (*dashed line*)

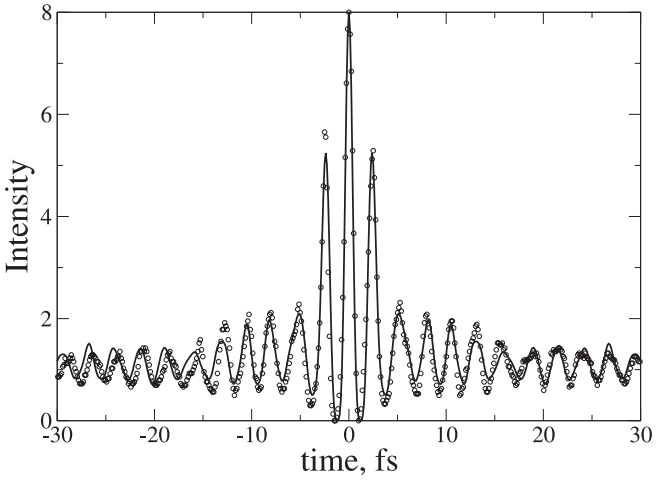

FIGURE 2 The measured ACF (*circles*) and the ACF calculated from the reconstructed pulse (*solid line*)

Having stabilized the temporal evolution of the CEP φ, it would be desirable to measure and tune its instantaneous value. To this end the measurement of photocurrents from metal surfaces (as initially proposed by Poppe et al. [18]) provides a promising approach. Although certain simplifications (e.g. ideally flat surfaces, use of the jellium model for the metal) currently necessary for a theoretical treatment of the problem may result in uncertainties of the absolute size of the effect, our present calculations based on time-dependent density functional theory [19, 20] indicate a dramatic increase of the phase-dependent modulation of the photocurrent from a gold surface (Fig. 5) for decreasing (sub-5-fs) pulse durations. A considerable phase dependence (see Fig. 5) together with sufficiently high electron yields at intensity levels well below the damage threshold (some 10^{13} W/cm^2) suggest that this approach holds the promise to provide a relatively simple way to determine and control φ. Another promising

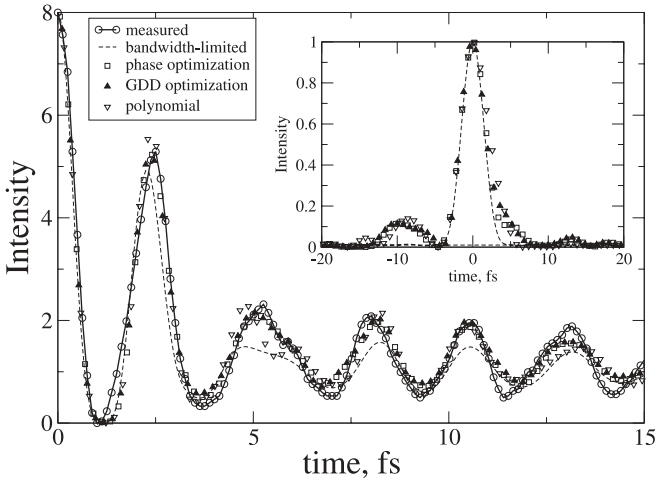

FIGURE 3 Comparison of pulse reconstructions with different representations of the spectral phase. We used the RMS error to quantify differences between the measured ACF and those calculated from the retrieved pulses. Corresponding normalized pulse intensities are shown in the *inset*. The bandwidth-limited pulse (*dashed line*) would have the FWHM equal to $\tau = 3.4$ fs, and the ACF-matching quality RMS $= 0.28$. Other curves correspond to the representation based on the spline interpolation of the spectral phase (*empty squares*, $\tau = 3.8$ fs, RMS $= 0.20$), spline interpolation of the GDD with further integration in order to obtain the spectral phase (*filled triangles*, $\tau = 3.7$ fs, RMS $= 0.22$), and the case when coefficients of a polynomial expansion of the spectral phase were taken as a set of optimization parameters (*open triangles*, $\tau = 4.3$ fs, RMS $= 0.24$). All the reconstructed pulse intensities show similar structure

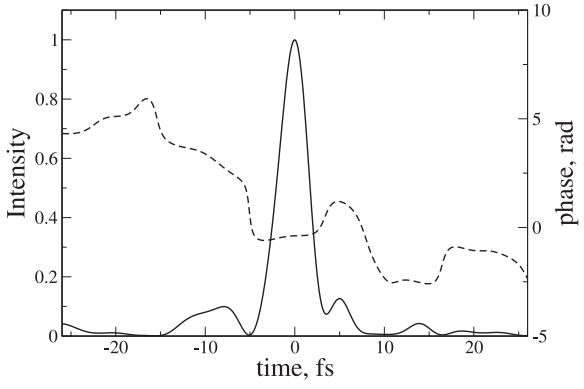

FIGURE 4 The intensity of the retrieved pulse (*solid line*) and its phase (*dashed line*). The main part of the pulse (between −4 fs and 9 fs) contains 80% of the pulse energy. The time direction is not unambiguously defined

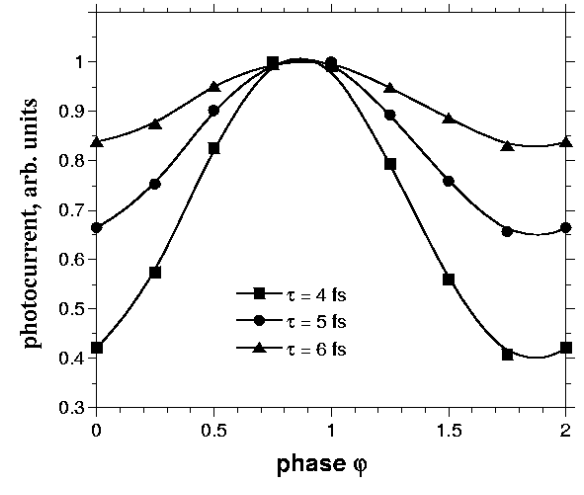

FIGURE 5 Variation of the time-integrated photoemission with the instantaneous phase. The peak intensity for all pulse durations was $I_0 = 10^{12}$ W/cm^2

experiment that (after slight improvement of the output energy) could be enabled by the source we developed relies on above-threshold ionization with linearly polarized (rather than circularly polarized [21]) ultra-short pulses. According to recent theoretical investigations [7], the momentum distribution of electrons ejected from hydrogen atoms irradiated with 4-fs linearly polarized pulses is predicted to exhibit a large phase-induced asymmetry. Theoretical investigations are thus unanimous in predicting a dramatic increase of the phase dependence of light–matter interactions at moderate field strength with decreasing sub-5-fs pulse durations. Combining control of the CEP evolution with extreme pulse parameters (4-fs duration, MW power, and high-25-MHz-repetition rate) the source described in this paper might become the key tool for measuring the absolute, instantaneous value of the CEP.

In conclusion, compression of the spectrally broadened single-mode fiber output of the Ti:sapphire oscillator by TFI mirrors yields 3-nJ phase-stabilized pulses at the full 24-MHz repetition rate, whose duration retrieved by different algorithms is in the range 4 ± 0.3 fs. Different theoretical models show that these pulse parameters are perfectly tailored for experiments aiming to measure and control the absolute phase of the pulse. Based on our own calculations we propose a compact phase-sensitive detector based on the measurement of photoelectrons emitted from solid (metal) surfaces. Our preliminary experiments demonstrate phase dependence of emitted electrons from a gold target and will be published elsewhere.

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