Controlling the Phase Evolution of Few-Cycle Light Pulses

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Using a coherent nonlinear optical technique, slipping of the carrier through the envelope of 6-fs light wave packets emitted from a mode-locked-oscillator/pulse-compressor system has been measured, permitting the generation of intense, few-cycle light with precisely reproducible electric and magnetic fields. These pulses open the way to controlling the evolution of strong-field interactions on the time scale of the light oscillation cycle and are indispensable to reproducible attosecond x-ray pulse generation.

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Exposing matter to a few oscillation cycles of intense optical radiation permits bound electrons to survive in the vicinity of the nucleus up to unprecedented intensity levels, allowing them to enter previously inaccessible, extreme regimes of nonlinear optics [1]. Observed and predicted consequences include optical-field ionization rates comparable to the light frequency [2], making atoms radiate high-order harmonics of the driving laser up to photon energies exceeding by more than a factor of 300 the energy of the laser photons [3], and the possibility of generating *isolated* x-ray pulses of attosecond duration [4]. These phenomena emerge in the strong-field, low-frequency regime of nonlinear optics, where the electronic motion is directly controlled by the electric and (at relativistic intensity levels also) magnetic fields. In a few-cycle laser pulse, with an electric field

$$E(t) = A(t)\cos(\omega_0 t + \varphi), \qquad (1)$$

the fields sensitively depend on the parameter φ, determining the position of the carrier wave (oscillating at frequency ω_0) with respect to the amplitude envelope $A(t)$. The resultant sensitivity of high-field phenomena to the absolute phase φ is beautifully demonstrated by the dependence of the predicted attosecond time structure of few-cycle-driven high harmonics on φ, as shown in Fig. 1. As a consequence, one of the greatest challenges in ultrafast laser physics and nonlinear optics, the *reproducible* generation of isolated attosecond x-ray pulses, relies on light pulses with reproducible absolute phase.

Rapid changes of φ in the pulse train delivered by mode-locked oscillators [5] and failure of gaining access to this parameter prevented precise control over strong-light-field-driven processes thus far. In this Letter we report on precision measurement of the variation of φ in sub-10-fs light pulses. The demonstrated technique allows selection of pulses with a fixed (but not known) absolute phase from the high-repetition-rate pulse train for the generation of high-intensity few-cycle light pulses with precisely reproducible electric and magnetic fields. Recent measurement of the interferometric cross correlation of successive sub-10-fs laser pulses revealed that φ suffers a shift upon each round trip of the pulse in the resonator due to a difference in the phase and group velocities in the laser components [5]. This round-trip phase shift typically accumulates to several hundred times 2π plus a physically relevant component $\Delta\varphi$, which obeys $0 < \Delta\varphi < 2\pi$ and accounts for a corresponding shift of the carrier with respect to the envelope. As a result, the carrier is offset at the carrier-envelope-offset frequency $f_{\text{ceo}} = (\Delta\varphi/2\pi)f_r$ (f_r is the pulse repetition rate), which leads to an offset of the phase-locked comb of laser modes, yielding a frequency of the nth mode $f_n = f_{\text{ceo}} + nf_r$ [6,7].

In Ref. [5], which addressed φ and possible ways of its control for the first time, $\Delta\varphi$ was shown to be also affected by nonlinear effects, which translate pulse energy fluctuations into a jitter of $\Delta\varphi$. Hence, precise measurement of the temporal evolution of φ in the output pulse train is required for selecting pulses with a fixed absolute phase, i.e., pulses having reproducible waveforms. Triggered by

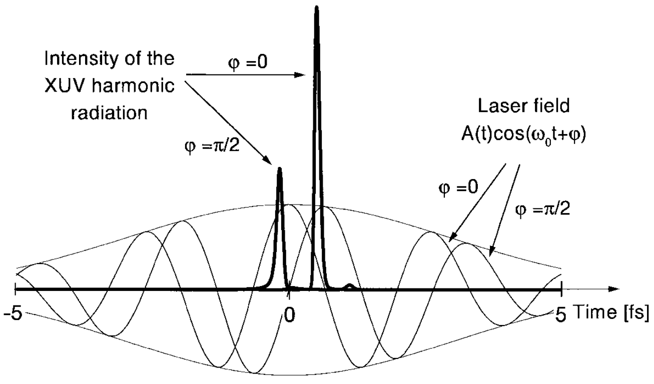

FIG. 1. Calculated temporal evolution of the cycle-averaged intensity of high-harmonic emission within 10% bandwidth at 3.2 nm from helium (interaction length 18 μm, pressure 500 Torr) driven by linearly polarized 5-fs light pulses ($\lambda_0 = 750$ nm, $I_{\text{peak}} = 2 \times 10^{15}$ W/cm^2) for two different values of the absolute phase φ. For the underlying physics, see Ref. [1].

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the recent availability of intense few-cycle laser pulses [8] down to pulse durations as short as 4 fs [1,9] several methods have been proposed for gaining access to either φ or $\Delta\varphi$ [1]. These include techniques drawing on strong-field processes, such as optical field ionization [10–12] or high harmonic generation [13], and on heterodyning different parts of the mode-locked spectrum [6,7,14].

Locking the equidistant frequency comb to stabilized external harmonic frequencies, as recently demonstrated for precision frequency-domain metrology [15–17], constitutes one possible means of implementing the latter concept. In our work, we have opted for another implementation of this approach. By frequency converting *directly* a portion of the spectrally broadened laser spectrum and coherently mixing this converted radiation with the fundamental laser field at the shifted frequency we are able to precisely measure and control the evolution of φ in the high-repetition-rate train of few-cycle pulses emerging from a mode-locked-oscillator/pulse-compressor system. The phase-locked frequency comb of the laser is first broadened by self-phase modulation to span a full octave. In this process, f_r is preserved [17] but f_{ceo} may be affected, as discussed below. The low-frequency wing of the spectrum, $f_{\text{ceo}} + n_{\text{low}} f_r$, is then frequency doubled and recombined with the high-frequency wing, $f_{\text{ceo}} + n_{\text{high}} f_r$. Beating of the modes $n_{\text{high}} = 2n_{\text{low}}$ with the second harmonic of modes n_{low} gives rise to a beat note at $2 \times (f_{\text{ceo}} + n_{\text{low}} f_r) - (f_{\text{ceo}} + n_{\text{high}} f_r) = f_{\text{ceo}}$ [18], yielding direct information about the round-trip phase shift $\Delta\varphi$ and thereby the temporal evolution of φ in the output pulse train. Beat notes of the same origin are also expected at $nf_r \pm f_{\text{ceo}}$, for $n = 1, 2, \ldots$.

The cascaded nonlinear processes (second harmonic generation of a self-phase-modulated pulse) call for high peak powers and very low pulse energy fluctuations. To this end, a specially designed Kerr-lens-mode-locked, mirror-dispersion-controlled Ti:sapphire laser has been developed. The resonator is made up of broadband chirped mirrors and incorporates a 1:1 imaging telescope consisting of two 1-m-radius mirrors and several plane folding mirrors (not shown in Fig. 2) to extend its length to ≈6 m. Pumped with a single-frequency, frequency-doubled Nd:YVO$_4$ laser ($P_{\text{pump}} \approx 4.5$ W), the laser generates a highly stable train of 9-fs, 20-nJ pulses at a repetition rate of 24 MHz (Fig. 2). These megawatt pulses produce substantial spectral broadening by self-phase modulation in a standard single-mode optical fiber (FS-SC-3314, Thorlabs). Figure 3 depicts the broadened spectrum, which spans from less than 500 nm to more than 1100 nm. A 1-mm-thick BBO doubling crystal oriented for type-I phase matching at ≈1080 nm is placed in close proximity of the fiber output to maximize frequency doubling efficiency.

The spectrally broadened pulses are passed through a chirped-mirror compressor, yielding sub-6-fs, 6-nJ pulses in a diffraction-limited beam (see inset of Fig. 3). Chirped mirror CM4 transmits some 30% of incident radiation near 540 nm. The spectral components leaking through this mirror near 540 nm are dispersed by a diffraction grating and directed through a polarizer and a slit onto a photomultiplier. The polarizer has been aligned to permit interference between the orthogonally polarized fundamental and frequency-doubled laser fields. The upper diagram in Fig. 4 shows a typical rf power spectrum of the combined fundamental and frequency-doubled laser radiation entering the detector within the wavelength range of (540 ± 0.8) nm. The sharp features showing up in the spectrum at ≈4 MHz and $\approx f_r - 4$ MHz disappear as the polarizer is rotated to have its principal axes aligned along the polarization directions of the fundamental and

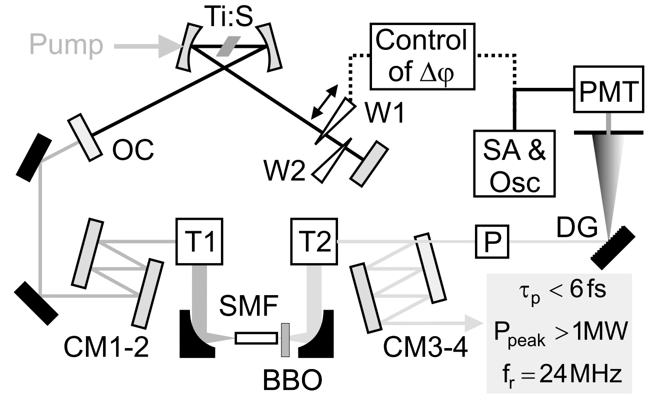

FIG. 2. Schematic of the experimental setup. Ti:S, highly doped ($\alpha_{514} = 5\ \text{cm}^{-1}$) 3-mm-long titanium-doped sapphire crystal; OC, 25% output coupler; W1-2, thin (0.1–0.7 mm) fused-silica wedges; CM1-4, ultrabroad-band chirped mirrors providing precise dispersion control over 630–950 nm; T1-2, 1:5 magnifying and demagnifying telescope, respectively; SMF, 3-mm-long single-mode fiber; P, polarizer; DG, diffraction grating; PMT, photomultiplier; SA & Osc, rf spectrum analyzer and digital oscilloscope.

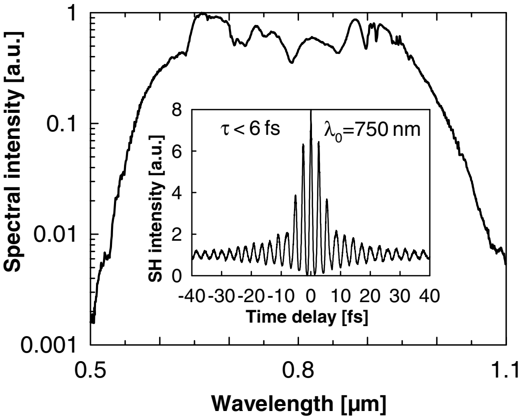

FIG. 3. Spectrum and interferometric autocorrelation (inset) of the pulses exiting the pulse compressor consisting of the single-mode fiber SMF and chirped mirrors CM3-4, respectively (see Fig. 2). The high fringe visibility over the entire autocorrelation trace indicates a pulse close to its bandwidth limit, which would allow a pulse duration of ≈5 fs.

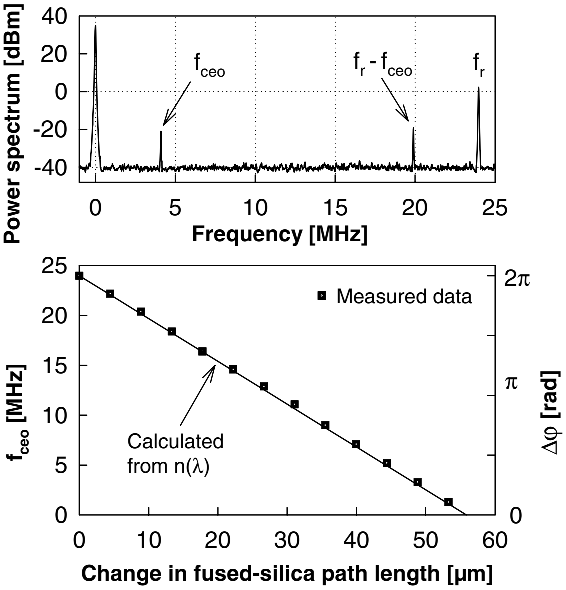

FIG. 4. Upper diagram, rf power spectrum of the combined frequency-doubled and fundamental laser radiation at 540 nm within an ≈1.5-nm bandwidth, as recorded using a 30-kHz rf resolution bandwidth. Lower diagram, carrier-envelope-offset frequency versus change in round-trip path length through the fused silica wedge W1. The accuracy of measuring (and setting) f_{ceo} is merely limited by random shifts of the beat within some ±15 kHz over a convenient measurement time of some 10–15 seconds.

frequency-doubled radiation, indicating that the features originate from a beat between these two fields. With the polarizer aligned for maximum beating signal, the power of these beat notes is some 20 dB below that emerging at f_r, a difference rapidly increasing as the detection bandwidth is increased beyond 2 nm as a consequence of the temporal offset between the frequency-doubled and fundamental wave packet at 540 nm.

To provide additional evidence for the origin of the observed rf spectral features we have varied the round-trip phase shift $\Delta\varphi$ by translating a thin wedged fused silica plate in the laser cavity and measured the frequency of the beat signal as a function of the (round-trip) path length in the fused silica plate. The results are depicted by squares in Fig. 4. The absolute phase φ in Eq. (1) is defined in a retarded frame (traveling at the group velocity v_g), hence it is varying as $\varphi(z) = \varphi_0 + (\beta_0 - \omega_0 v_g^{-1})z$ during propagation through a dispersive transparent medium [5]. Here $\beta_0 = \beta(\omega_0) = \omega_0 n(\omega_0)/c$ is the propagation constant at ω_0 and $v_g = (\partial\beta/\partial\omega)_{\omega_0}^{-1}$ is the group velocity. From these expressions straightforward algebra yields $\delta\varphi = 2\pi(\partial n/\partial\lambda)_{\lambda_0}\delta z$, where $\partial n/\partial\lambda = -0.018\ \mu\mathrm{m}^{-1}$ for fused silica at $\lambda_0 = 790$ nm, the center laser wavelength. The change in f_{ceo} is then predicted as $f_{\mathrm{ceo}}(\delta z) = f_{\mathrm{ceo},0} - 0.018 f_r \delta z\ [\mu\mathrm{m}]$ and depicted by the full line in the lower diagram of Fig. 4 for the starting position ($\delta z = 0$) set to yield $f_{\mathrm{ceo}} = f_r$. In contrast with previously used techniques (tilting and translating the cavity end mirror [15,16]), the present approach allows precise determination of a change in $\Delta\varphi$ and f_{ceo} *independently* and thereby experimental verification of the relationship between these two quantities for the first time. The excellent agreement between the *calculated* slope $\partial f_{\mathrm{ceo}}/\partial z$ and the *measured* variation of f_{ceo} provides conclusive evidence of the origin of the observed beat signal and our ability to measure and control the pulse-to-pulse shift in φ with a high precision.

The stability of f_{ceo} invites to "slowing down" the evolution of φ to frequencies as low as a few kHz and observing it directly in the time domain. To this end, we have tuned the carrier-slip beat to $f_{\mathrm{ceo}} < 10$ kHz and analyzed the photomultiplier signal amplified by a low-pass amplifier ($f_T = 100$ kHz). Figure 5 shows millisecond snapshots of the temporal evolution of the intensity of 540-nm light composed of fundamental laser radiation and frequency-doubled 1080-nm light (with the dc component eliminated), reflecting the evolution of φ in the laser pulse train [19]. The nearly periodic evolution of the carrier-slip beat signal over several cycles in Fig. 3 implies that the round-trip phase shift can be kept "frozen" at values as low as $\Delta\varphi = 2\pi f_{\mathrm{ceo}}/f_r < 10^{-3}$ rad for extended periods as long as milliseconds without active stabilization; i.e., random phase jitter remains less than this value over milliseconds. As a consequence, full electronic control or stabilization of φ can be accomplished with a sub-millisecond-response servo loop adjusting either the path length in the fused silica wedge or the intracavity pulse energy (upon controlling the pump power) [5] such that f_{ceo} is phase locked to a user-defined value.

It is important to note that the generation of phase-stable few-cycle pulses at the output of the fiber/chirped-mirror compressor by locking f_{ceo} to zero in the present scheme does not strictly imply phase stabilization *directly* at the output of the mode-locked oscillator as well. This

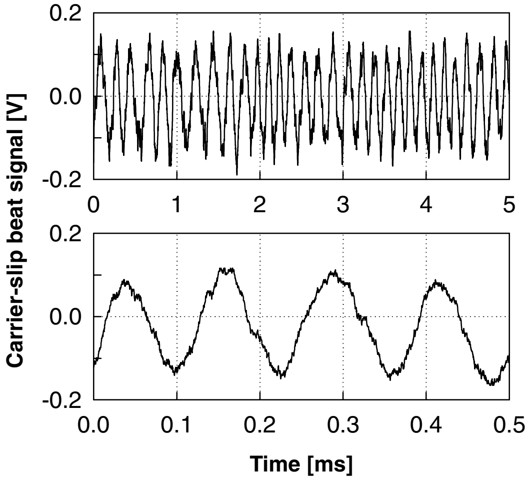

FIG. 5. Snapshots of the temporal evolution of the carrier-slip beat signal reflecting slow quasiperiodic variation of the absolute phase φ in the sub-6-fs pulse train.

is because drift or fluctuations in the pulse parameters at the fiber input and in the fiber parameters (e.g., temperature, refractive index) may be translated into a corresponding change in φ by linear and nonlinear effects [5]. The resultant time-dependent shift of φ then emerges with opposite sign at the output of the oscillator, if φ is stabilized at the fiber output. Nevertheless, in the present case these external fluctuations and drifts in φ are estimated as very low ($<\pi/50$) owing to the high pulse energy stability, moderate spectral broadening in the fiber, and the short fiber length.

Whereas locking both f_{ceo} and f_r is imperative for applications in frequency-domain metrology [7,15–17], neither of them needs to be stabilized if intense phase-stabilized few-cycle light pulses are to be generated. This is a consequence of (i) the very low jitter of $\Delta\varphi$ observed in our experiments and (ii) the fact that the pulse energy can be significantly boosted (by extracavity amplification) only at substantially reduced repetition rates (≤ 1 kHz). Setting f_{ceo} equal to a few hundred kHz and using a tracking oscillator will allow one to produce a virtually noise-free quasisinusoidal signal reflecting the variation of φ in the 24-MHz pulse train even more accurately than do the traces shown in Fig. 5. Triggering the pulse picker to, say, the positive-slope zero transition of the beat signal will select pulses with φ fixed to within $2\pi(f_{\text{ceo}}/f_r) < \pi/10$ for external amplification (and moderate pulse compression). The phase of the resultant intense few-cycle pulses can be fine tuned externally by the same technique used for adjusting $\Delta\varphi$ inside the cavity.

In conclusion, we have demonstrated a sub-10-fs source equipped with a phase-tracing system capable of delivering seed pulses with a locked pulse-to-pulse phase for kHz-rate amplifier/compressor systems without any active stabilization. The presented results open the way to generating few-cycle light pulses focusable to intensities up to 10^{18} W/cm^2 and beyond with precisely reproducible electric and magnetic fields. Exposing matter to these ultraintense electromagnetic transients will allow unprecedented control of optical field ionization of atoms and subsequent motion of the freed electron wave packets in strong fields. Anticipated impacts of this new experimental capability include the controlled generation of isolated attosecond bursts of coherent soft-x-ray radiation and highly collimated relativistic electron pulses with sublaser-cycle duration.

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Note added.—Since the submission of this paper, Jones *et al.* [20] reported full electronic control of f_{ceo} and f_r and similar results have been achieved independently by Holzwarth *et al.* [21], yielding unprecedented frequency rulers for metrology.

Note added.—With a somewhat improved carrier-slip beat signal we have achieved full control of the evolution of φ by phase locking f_{ceo} to an rf local oscillator by fine control of the pump power as described in Ref. [21].

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[18] Many modes within the detection bandwidth may contribute to this signal.
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