Route to phase control of ultrashort light pulses

L. Xu, Ch. Spielmann, A. Poppe, T. Brabec, and F. Krausz

Abteilung Quantenelektronik und Lasertechnik, Technische Universität Wien, Gusshausstrasse 27, A-1040 Wien, Austria

T. W. Hänsch

Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany

Received July 12, 1996

A feasibility study of controlling the carrier phase in ultrashort light-wave packets emitted by a sub-10-fs laser is reported. An experimental apparatus capable of exploring the phase sensitivity of nonlinear-optical interactions is presented.

The electric field of pulsed radiation can generally be written as

$$E(t,z) = A(t,z)\exp[-i\omega_0 t + i\psi(z)] + \text{c.c.}, \qquad (1)$$

where $A(t,z)$ is the complex amplitude envelope of the pulse and the carrier frequency ω_0 is defined as the center of gravity of the frequency spectrum of $E(t, z = 0)$. The concept of the envelope can by physically meaningful for pulse durations that are comparable with a carrier oscillation cycle. If t is measured in a frame of reference retarded by z/v_g, where v_g is the group velocity, $\psi(z)$ defined by requiring $\mathrm{Im}[A(t = 0, z)]$ has a simple physical meaning: It determines the position of the carrier wave with respect to the envelope. Whereas several techniques have been developed to obtain the complex envelope $A(t,z)$ of ultrashort light pulses, to our knowledge ψ has not been obtained. In light pulses of a duration comparable with the carrier oscillation cycle, a change in ψ significantly affects the evolution of $E(t)$. As a consequence, ψ is expected to become an essential parameter for nonperturbative (strong-field) interactions of few-cycle ultrashort light pulses with atomic systems. In this Letter we report on what is believed to be the first experiment that provides information on the evolution of the carrier phase in laser pulses and permits exploration of the influence of this parameter on atomic processes.

A prerequisite for exploiting the potential of phase control in nonlinear optics and high-field physics is the availability of a stationary train of ultrashort pulses with a fixed ψ. Mode-locked lasers generally fail to meet this requirement because the relative carrier phase ψ picks up a shift of

$$\Delta\psi = \phi_R - \omega_0 T_R \qquad (2)$$

on each round trip in the cavity. Here ϕ_R and T_R stand for the round-trip phase and the group retardation, respectively. We investigated the evolution of ψ in sub-10-fs wave packets emitted by a mode-locked Ti:sapphire ring oscillator.[1] To this end, we constructed a Michelson-type correlator (Fig. 1) to measure the interferometric cross correlation $G(\tau)$ of successive pulses from the laser.[2] $\Delta\psi - 2k\pi$ can be determined from $G(\tau)$ (Fig. 2), where k is a positive integer, which does not affect the physical properties of the wave packet.

First, we investigated the influence of dispersive material on $\Delta\psi$ by varying the path length l_g in a thin fused-silica glass wedge incorporated into the laser.[1] The filled circles in Fig. 3 show the measured variation of $\Delta\psi$ with l_g, and the line depicts $\Delta\psi(l_g) = \Delta\psi_0 + \phi(\omega_0) - \omega_0(\partial\phi/\partial\omega)_{\omega_0} = \Delta\psi_0 + 2\pi(\mathrm{d}n/\mathrm{d}\lambda)_{\lambda_0} l_g$. For fused silica $\mathrm{d}n/\mathrm{d}\lambda = 0.017\ \mu\mathrm{m}^{-1}$ at 800 nm. The small change in l_g that changes $\Delta\psi$ by 2π permits $\Delta\psi$ to be controlled without notably affecting the pulse shape.

Stationary output pulses with a fixed relative carrier phase call for setting $\Delta\psi = 2k\pi$. However, this adjustment will freeze ψ for only a limited period because nonlinear contributions to ϕ_R and T_R [see Eq. (2)] translate fluctuations of the pulse parameters into small fluctuations of $\Delta\psi$, resulting in $\Delta\psi(T) = 2k\pi + \delta\psi(T)$. Over many round trips $\delta\psi$ can lead to a substantial jitter of the relative carrier phase $\psi(T) - \psi_0 = (1/T_R)\int_0^T \delta\psi(T')\mathrm{d}T'$. The rms of this jitter, σ_ψ, can be expressed as

$$\sigma_\psi^2 = \langle[\psi(T) - \psi_0]^2\rangle = \frac{1}{\pi}\int_0^{\omega_R/2} \frac{S_{\delta\psi}(\omega')}{\omega'^2 T_R^2}\,\mathrm{d}\omega', \qquad (3)$$

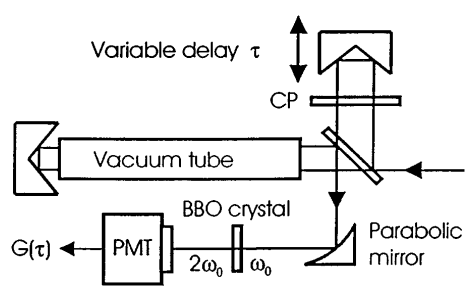

Fig. 1. Schematic diagram of the correlator used to the measure $\Delta\psi$. To balance dispersion in the correlator arms precisely, we evacuate a delay section equal to the resonator round-trip length in the long arm, a compensation plate (CP) is used to introduce the same amount of fused silica into the short arm as the tube windows introduce into the long arm, and pulse splitting and recombination are implemented by identical broadband dielectric coatings on opposite sides of the beam splitter. PMT, photomultiplier tube; BBO, β-barium borate.

0146-9592/96/242008-03$10.00/0

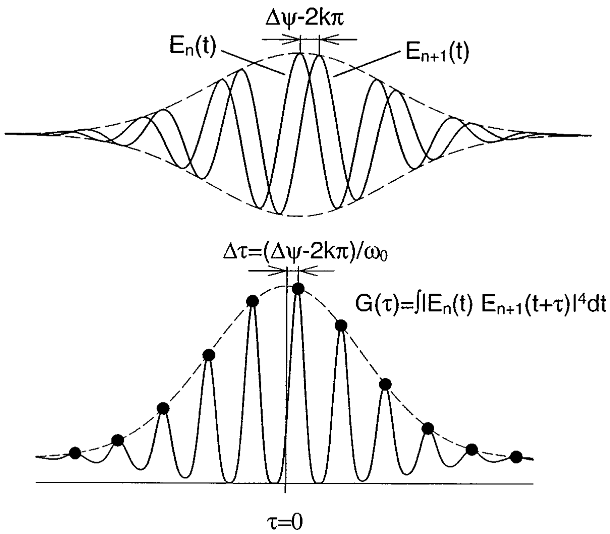

Fig 2. Principle of the measurement of $\Delta\psi$. $E_n(t)$ and $E_{n+1}(t)$ describe successive pulses from the laser. $\Delta\psi - 2k\pi$ can be determined from the position of the fringe peaks on the envelope of $G(\tau)$.

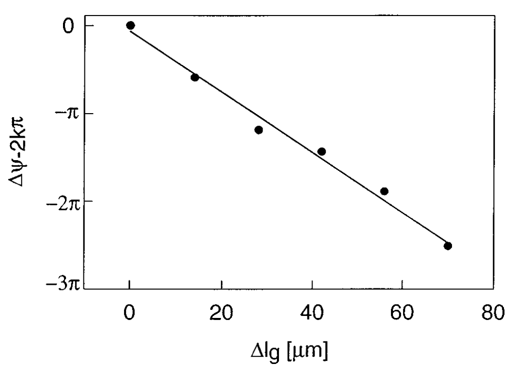

Fig. 3. Measured (filled circles) and calculated (line) changes in the round-trip carrier phase shift with increasing propagation length through the intracavity fused-silica wedge. The initial value of l_g was set to yield $\Delta\psi(\Delta l_g = 0) \approx 2k\pi$.

where the angle brackets denote time average, $\omega_R = 2\pi/T_R$, and $S_{\delta\psi}(\omega)$ is the power spectral density of $\delta\psi(T)$. Replacing the lower boundary of integration by $\omega(0 < \omega < \omega_R/2)$ yields $\sigma_\psi(\omega)$, the rms jitter of ψ that originates from noise components of frequencies higher than ω. $\sigma_\psi(\omega)$ is expected to decrease rapidly with increasing ω. The critical frequency ω_{crit} at which $\sigma_\psi(\omega)$ drops below a fraction of π determines the dephasing rate of the emitted wave packets and hence the speed of optical control required for phase stabilization.

Because $S_{\delta\psi}(\omega)$ cannot be directly measured, we relate $\delta\psi(T)$ to variations of the pulse parameters. This relationship will permit an evaluation of $S_{\delta\psi}(\omega)$ from the noise spectra of the respective pulse parameters. Owing to a short soliton period, the solitonlike pulse circulating in a sub-10-fs Ti:sapphire oscillator adiabatically follows changes in the laser parameters that occur on a long time scale compared with T_R. According to soliton perturbation theory,[3] under these circumstances two invariants, the changes in pulse energy $\delta W(T)$ and in the center frequency $\delta\omega_0(T)$, along with external noise sources, govern the variation of other pulse parameters. This also holds for the relative carrier phase, which can be written as

$$\delta\psi(T) = \phi_{\text{Kerr}}\left(\frac{\mathrm{d}P_{\text{peak}}}{\mathrm{d}W}\right)_{W_0}\delta W(T) - \omega_0 D\,\delta\omega_0(T). \quad (4)$$

The first term is a perturbation to ϕ_R and accounts for the soliton phase shift that is due to the optical Kerr effect in the gain medium, where ϕ_{Kerr} gives the phase shift per unit power and P_{peak} is the peak power of the pulse inside the gain medium. The second term results from a variation of T_R owing to a shift of the laser spectrum and scales with D, the net intracavity group delay dispersion (GDD). The influence of other noise sources (e.g., cavity length fluctuations) on $\delta\psi(T)$ can be shown to be negligible.

The filled squares and triangles in Fig. 4 represent the measured variation of $\Delta\psi$ with intracavity pulse energy for two different values of the intracavity GDD. Our measurements yield a $\delta\psi$ that decreases with increasing pulse energy. This is in apparent contradiction with the positive sign of the soliton phase shift that is due to $\phi_{\text{Kerr}} > 0$. One can resolve the paradox by assuming a coupling between δW and $\delta\omega_0$. As a matter of fact, the evaluation of the center of gravity of the corresponding laser spectra reveals an appreciable variation of ω_0 with W, as shown by the filled circles in Fig. 4.[4] For increasing pulse energy, the negative cavity GDD translates the red shift of ω_0 into a contribution of negative sign that dominates over the positive soliton phase shift.

Integration of Eq. (4) yields

$$\Delta\psi(W) - \Delta\psi(W_0) \approx \phi_{\text{Kerr}}[P_{\text{peak}}(W) - P_{\text{peak}}(W_0)] - \omega_0(W_0)D[\omega_0(W) - \omega_0(W_0)], \quad (5)$$

which permits a comparison of the experimental results with the prediction. Using the measured values of the pulse width $\tau_p(W, D)$, which vary between 8 and 14 fs, one can calculate $P_{\text{peak}}(W, D)$ for both settings of

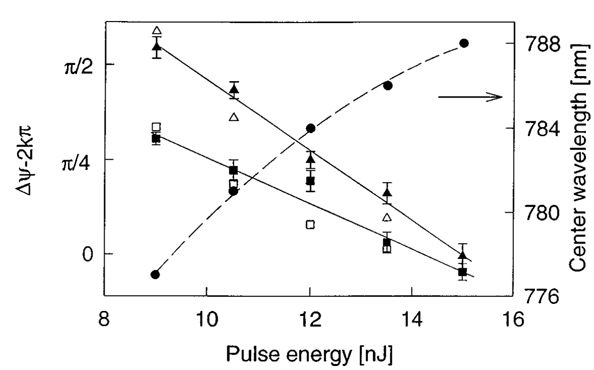

Fig. 4. Measured round-trip carrier phase shift for different values of the cavity GDD (filled squares and triangles) and center of the laser spectrum (filled circles) as a function of intracavity pulse energy. The open squares and triangles depict $\Delta\psi - 2k\pi$ obtained from Eq. (4) as described in the text. The solid lines and the dashed curve are guides to the eye.

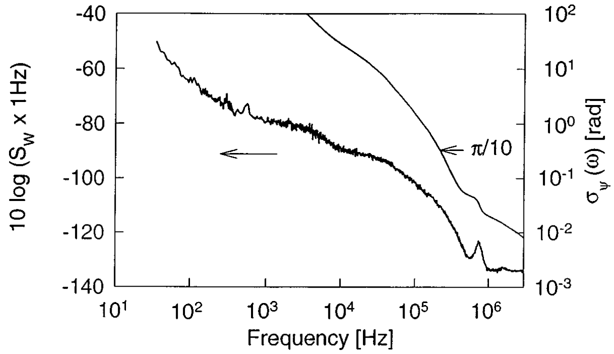

Fig. 5. Power spectral density $S_W(\omega)$ of $\delta W(T)/W_0$ (normalized to a 1-Hz bandwidth) on a logaritmic scale and the calculated $\sigma_\psi(\omega)$ as defined in the text.

the cavity GDD by taking into account the variation of τ_p inside the resonator. $\omega_0(W)$ is directly obtained from $\lambda_0(W)$, as shown in Fig. 4. Inserting $P_{\text{peak}}(W, D)$, $\omega_0(W)$, and $\phi_{\text{Kerr}} = 0.4 \times 10^{-6}\ \text{W}^{-1}$ (Ref. 2) into expression (5) leads to the values represented by the open squares and triangles in Fig. 4 for $D_1 = -17\ \text{fs}^2$ and $D_2 = -24\ \text{fs}^2$, respectively. These values of the cavity GDD were obtained from least-squares fits to the measured data (filled squares and triangles in Fig. 4). They are consistent with previously evaluated values of the cavity GDD,[1] and $D_1 - D_2$ is reasonably close to 5 fs^2, which we obtained from the path-length change $l_{g,1} - l_{g,2}$ in the fused-silica wedge.

Having identified pulse-energy fluctuations as the primary source of $\delta\psi$ by measurement of $S_W(\omega)$, the power spectrum of $\delta W(T)/W_0$, we can determine $S_{\delta\psi}(\omega)$ by using $\delta\psi = (\text{d}\Delta\psi/\text{d}W)_{W_0}\delta W$ in the adiabatic approximation. This relation yields $S_{\delta\psi}(\omega) = \alpha S_W(\omega)$, where $\alpha = [W_0(\text{d}\Delta\psi/\text{d}W)_{W_0}]^2$. We obtained $S_W(\omega)$ by measuring the sideband of the first harmonic of the laser power spectrum with a rf spectrum analyzer[5] and is shown in Fig. 5. For $D = -17\ \text{fs}^2$ and $W_0 = 15$ nJ we evaluate $\alpha = 7.5$ from the filled squares in Fig. 4. Substitution of $S_{\delta\psi}(\omega) = \alpha S_W(\omega)$ into Eq. (3) and replacing the upper boundary of integration by ω yield the rms carrier phase jitter $\sigma_\psi(\omega)$ depicted in Fig. 5. It drops to $\pi/10$ at $\omega_{\text{crit}}/2\pi \approx 2 \times 10^5$ Hz; hence we may conclude that a sub-10-fs Ti:sapphire laser is capable of generating trains consisting of $>10^3$ stationary light pulses with a fixed carrier phase, provided that the nominal value of $\Delta\psi(W_0, l_g)$ is set equal to $2k\pi$. The fact that notable noise-induced dephasing occurs on a time scale longer than 5 μs implies that the 3-μs gain relaxation time of Ti:sapphire allows one to control the intracavity pulse energy at a sufficiently high speed to prevent dephasing of the few-cycle wave packets. As a consequence, our experiments lead us to the conclusion that the physical characteristics of a sub-10-fs Ti:sapphire oscillator potentially permit the generation of phase-stabilized light pulses.

The most important consequence of the above investigations is that the experimental system that is presented offers the possibility of exploring the influence of ψ on light–matter interactions. As noise-induced dephasing occurs on a microsecond time scale, setting $\Delta\psi - 2k\pi$ equal to π/N, i.e., a submultiple of π (with N being currently limited to $\approx$20 by the measurement accuracy of $\Delta\psi$) will result in a periodic modulation of ψ of the output pulses at the corresponding submultiple $1/2NT_R$ of the pulse-repetition rate. Identifying and quantifying a corresponding modulation in any of the physical measurables of the irradiated atomic system will provide direct information on the role of the phase of the light field in the respective interaction. One intriguing example is photoemission in the optical tunneling regime.[6] The observation of a modulation of the photocurrent at $1/2NT_R$ may provide not only the first direct evidence for optical tunneling but also the ψ-dependent signal needed to control the carrier phase in laser pulses. These prospects together with recent progress in approaching the single-cycle limit in optical pulse generation[7] hold out the promise of exploring and controlling atomic processes to an unprecedented depth by use of precisely controlled electromagnetic transients.

We thank R. Szipöcs and K. Ferencz (Research Institute for Solid State Physics, Budapest, Hungary) for providing the special dispersive mirrors used in the Ti:sapphire laser. This research was supported by Fonds zur Förderung der wissenschaftlichen Forschung grants P-09710 and P-10409, and by Österreichische Nationalbank grants 5335 and 5124.

References

1. L. Xu, Ch. Spielmann, F. Krausz, and R. Szipöcs, Opt. Lett. **21,** 1259 (1996).
2. K. L. Sala, G. A. Kenny-Wallace, and G. E. Hall, IEEE J. Quantum Electron. **16,** 990 (1980).
3. H. A. Haus and A. Mecozzi, IEEE J. Quantum Electron. **29,** 983 (1993).
4. Our computer simulations revealed that the coupling between δW and $\delta\omega_0$ relates to the rapidly decreasing magnitude of negative cavity GDD below 0.7 μm (Ref. 1) and to the finite response time of the Kerr nonlinearity.
5. D. von der Linde, Appl. Phys. B **39,** 201 (1986).
6. L. V. Keldysh, Sov. Phys. JETP **47,** 1307 (1964).
7. M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, S. Sartania, Ch. Spielmann, and F. Krausz, "Compression of high energy pulses below 5 fs," submitted to Opt. Lett.