# Femtosecond spectral electric field reconstruction using coherent transients

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We have implemented a new approach for measuring the time-dependent intensity and phase of ultrashort optical pulses. It is based on the interaction between shaped pulses and atoms, leading to coherent transients.

*OCIS codes:* (320.5540) Pulse shaping; (320.7100) Ultrafast measurements



Complete pulse measurement is a central issue to most ultrashort experiments. This is the case both for measuring the shortest "clean" pulses as well as complex shapes produced by pulse shapers. The usual methods can be divided into self-referenced techniques (SPIDER, FROG or derivatives)[1–3] and techniques comparing the unknown pulse with a well-known one. In the first method, non-stationary filter or a non-linear interaction are required. In the second case, the unknown field is compared to a known reference field. A spectacular demonstration was achieved with attosecond pulses used for direct light wave measurement.[4] The method has a linear sensitivity with respect to the unknown pulse, but previous knowledge of the reference pulse can be a severe constraint, particularly in examples such as spectral interferometry where the reference pulse has similar characteristics (same spectral range, and similar or broader spectral width) as the unknown pulse.

We present in this letter a new approach to electric field reconstruction based on an atomic response. In this context, we regard the atomic system as a high spectral resolution probe (compared to spectrometers used in usual methods) of known response to a shaped light field. Contrary to most of the well-known cross-correlation in gases which require no resonant relay-state and leads to intensimetric cross-correlation (just as in frequency mixing processes),[5,6] this method requires resonant bound states and leads to the product of the two electric fields. It is based on a sequence of Coherent Transients (CT) measurements.[7] These CT are oscillations in the excited state population resulting from the interaction between a two-level system and a weak chirped pulse. This scheme takes advantage of the high sensitivity of CT to the pulse shape, in particular, to spectral phase effects.[8,9] In this letter, this new method is first explained then experimental results are presented. Its feasibility is illustrated by measuring glass material dispersion. The limits of the method and its implementation are discussed.

The CT result from the interaction of a two-level system ($|g\rangle$ and $|e\rangle$) with a chirped pulse $E(t)$ of carrier angular frequency $\omega_0$ close to resonance ($\omega_0 \simeq \omega_{eg}$). The transient excited state population is probed towards the $|f\rangle$ level in real time by a second ultrashort pulse $E_{ref}(t)$ which is Fourier transform limited and short compared to the characteristic features of $E(t)$. Its frequency is close to resonance ($\omega_{fe}$). The fluorescence arising from the $|f\rangle$ state is then recorded as a function of the pump-probe delay $\tau$. The probe pulse provides access to the temporal evolution of the population in $|e\rangle$, produced by the pump beam. The result of the interaction is described by first order perturbation theory, and the fluorescence is proportional to

$$\begin{aligned} S(\tau) &= |a_f(\tau)|^2 \qquad (1)\\ &\propto \left| \int_{-\infty}^{+\infty} E_{ref}(t-\tau) \exp(i\omega_{fe}(t-\tau)) a_e(t) dt \right|^2 \end{aligned}$$

with

$$a_e(t) = \int_{-\infty}^{t} E(t') \exp(i\omega_{eg} t') dt' \qquad (2)$$

In the case of a simply chirped pulse $E(t)$, a quadratic phase appears in the integral giving $a_e(t)$ (Eq. 2), leading to oscillations of the probability $|a_f(\tau)|^2$ as already demonstrated.[7,8] These strong oscillations result from interferences between the population amplitude excited at resonance and after resonance. They are extremely sensitive to tiny phase modifications.[9] However, although sensitive to phase effects these CT give access to the excited state probability $|a_e(\tau)|^2$ whereas the probability amplitude is necessary to achieve a complete measurement of the electric field. Moreover, the oscillations are only produced by the second part of the pulse (after resonance).[7]

To overcome these limitations, a new excitation scheme with two pump pulses is used. Two measurements are performed, each with a two pulse sequence with a well defined phase relationship $E_{shaped}(t) = E_1(t) + e^{i\theta} E_2(t)$ where $E_1(t)$ and $E_2(t)$ are two replica of the unknown pulse generated by splitting the same initial pulse $E(t)$ and adding additional spectral phase. These can be obtained either with a Michelson-type interferometer or with a pulse shaper. The first pulse $E_1(t)$ creates an initial population in the excited state. The second pulse $E_2(t)$ is strongly chirped and sufficiently delayed by $T$ so that it does not overlap with the first pulse.

This second pulse creates a population in the excited state which interferes with the initial population created by the first pulse. Thus, oscillations due to CT occur on the whole duration of the second pulse. The final state population during the second pulse can be written as

$$\begin{aligned} |a_{f,\theta}(\tau)|^2 &= \left|a_{f,1}(+\infty) + e^{i\theta} a_{f,2}(\tau)\right|^2 \\ &= |a_{f,1}(+\infty)|^2 + |a_{f,2}(\tau)|^2 \\ &\quad + 2Re\left\{a^*_{f,1}(+\infty) a_{f,2}(\tau) e^{i\theta}\right\} \end{aligned} \quad (3)$$

$|a_{f,1}(+\infty)|^2$ can be obtained from the plateau reached between the two pulses. The last two contributions in Eq. 3 depend on $\tau$ and on the second pulse. For a second pulse of smaller peak power, the crossed term is dominant so that the response is mostly linear with respect to the second pulse. A first measurement for $\theta = 0$ gives $Re\left\{a^*_{f,1}(+\infty) a_{f,2}(\tau)\right\}$. A second measurement for $\theta = \pi/2$ brings the complementary part $Im\left\{a^*_{f,1}(+\infty) a_{f,2}(\tau)\right\}$. For pulse intensities of comparable magnitude, the system of nonlinear equations resulting from both measurements can be solved to extract $a_{f,2}(\tau)$.[11] By derivation one obtains

$$\frac{da_{f,2}}{d\tau} = \int_{-\infty}^{+\infty} \widetilde{E}_2(\omega_{eg} + \Omega) \widetilde{E}_{ref}(\omega_{fe} - \Omega) d\Omega \quad (4)$$

from which $\widetilde{E}_2(\omega)$ can be deduced provided that the reference pulse is known and short enough. Finally, knowing the extra phase added to generate the pulse sequence, $\widetilde{E}(\omega)$ and thus $E(t)$ is obtained. Note that unlike conventional interferometric methods, the reference-field spectrum does not need to overlap that of the unknown field.

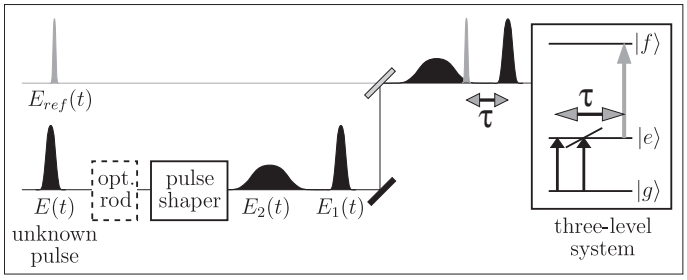


Fig. 1. Set-up Principle: The unknown pulse is sent into a pulse shaper programmed to generate a sequence of two pulses. One FL, the second delayed and chirped. An extra phase can be added. An optional glass rod is added in the front of the set-up. The reference is a pulse shorter than the unknown pulse. Inset: Excitation scheme.

To illustrate the method, an experiment has been performed in an atomic Rb vapor (see Fig. 1). The Rb (5s - 5p ($P_{1/2}$)) transition (at 795 nm) is resonantly excited with the pump pulse sequence. The transient excited state population is probed "in real time" on the (5p - (8s, 6d)) transitions with an ultrashort pulse produced by a home-made NOPA (607 nm, 25 fs). The "unknown" pulse $E(t)$ has initially a duration of 130 fs and can be affected by dispersive materials to demonstrate the measurement capabilities. A phase and amplitude 640 pixels LCD-SLM pulse shaper[10] is used to generate the pump pulse sequence by applying a complex transmission in the spectral domain:

$$H_\theta(\omega) = \frac{\{1 + \exp[i(\theta + \phi'(\omega - \omega_0) + \phi'' \frac{(\omega-\omega_0)^2}{2}]\}}{2} \quad (5)$$

where $\omega_0$ is the carrier frequency of the pump pulse. The first pulse in the pump sequence is identical to $E(t)$. The second one is strongly chirped with $\phi'' = -2.10^5$ fs$^2$ in order to produce CT, and delayed by $\phi' = 6$ ps. An extra phase factor $\theta$ can be added.

In a first experiment, $E(t)$ is close to Fourier limited. Two recordings are performed for $\theta = \theta_0$ and $\theta = \theta_0 + \pi/2$ (with $\theta_0 \simeq -0.2\pi$) as shown in Fig. 2a. Combining these two measurements allows one to determine in-phase and in-quadrature contributions from $E_2(t)$, so that $a_{f,2}(t)$ can be retrieved. The main difference with previous experiments[7,8] is the preparation of a coherent superposition of $|e\rangle$ and $|g\rangle$ by the first pulse. Then the second -strongly chirped- pulse produces large oscillations during its whole duration. These oscillations can be seen as beats between the atomic dipole (which behaves as a local oscillator) and the electric field from the second pulse, as in heterodyne detection. By combining the two measurements, it is therefore possible to retrieve fully the temporal evolution of the excited state probability amplitude due to the second pulse. Figure 2b displays the reconstructed excited state probability amplitude in the complex plane. The expected Cornu spiral[7] is observed. $E_2(t)$ obtained by simple derivation of $a_{e,2}(t)$, is displayed on Fig. 2c. The temporal amplitude and phase are represented. As a comparison, the exact theoretical temporal phase applied by the pulse shaper is shown (dashed line) without any other adjustment than the offset. The agreement is excellent. The quadratic phase added by the pulse shaper is perfectly retrieved.

In a second set of experiments, the dispersion of a SF58 glass rod ($\phi'' = 20492$ fs$^2$) inserted in the pump beam is measured. CT are monitored with and without the rod in the pump beam. This dispersion is sufficiently small so that $E_1(t)$ and $E_2(t)$ do not overlap. Experimental results in the spectral domain are presented on Fig. 3. Figure 3a shows the spectral phase of $\widetilde{E}_2(\omega)$ with and without the dispersive rod. Their difference is plotted on Fig. 3b together with the value calculated from the rod coefficients. The agreement is excellent on the spectral domain where the intensity (dashed line) is significant.

We have thus demonstrated here the capabilities of this method on a simple example. Its main advantage is that the unknown and reference pulses need not have any spectral overlap, thus the method may be easily extended to wavelength regions for which a local oscillator pulse is not usually available. This method provides

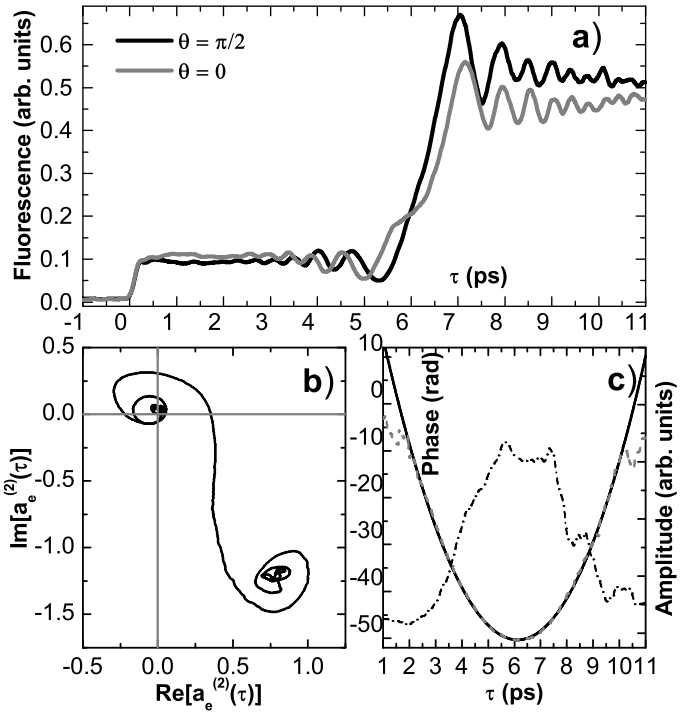


Fig. 2. a) Experimental Coherent Transients resulting from the excitation of the atom by a FT limited pulse (at time $\tau = 0$) followed by a chirped pulse (centered at $\tau = 6$ps), for two different relative phases $\theta = 0$ and $\pi/2$ between the two pulses. b) Probability amplitude $a_{f,2}(\tau)$ reconstructed from the two measurements presented in a) and displayed in the complex plane. The Cornu spiral appears clearly. c) Reconstructed phase (dots), theoretical phase (dashed) and amplitude (dash-dotted) of $E_2(\tau)$.

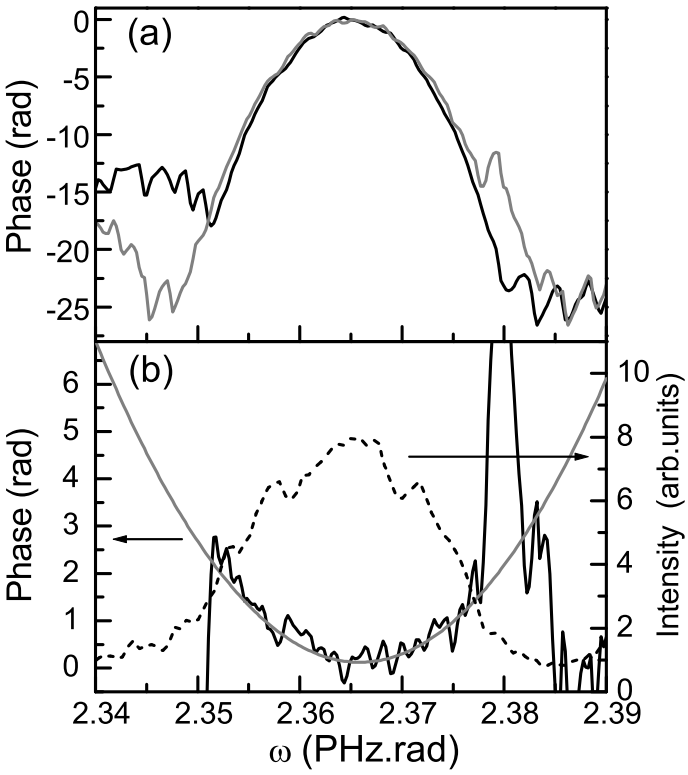


Fig. 3. Electric field reconstruction. a) Spectral Phase retrieved obtained with (gray) and without (black) rod. b) Dashed line: Spectral intensity; Solid lines: Spectral phase due to the rod, (black): Experiment (difference between data shown in a)), (gray): Calculated phase from rod properties.

interferometric-like terms but the two spectral shears remove constraints of usual interferometric technics : same spectral domain, interferometric control of delay, temporal step small enough to resolve fringes. Here the heterodyne beating with the local oscillator provided by the atom shifts the interferometric oscillations around zero frequency which makes them much easier to measure. The requirement of an atomic transition can be seen as a constraint as compared to "all optical" methods for pulse measurements in the visible or near IR. However, some complex pulse shapes (with holes or phase discontinuities) are difficult to measure with standard methods and the present one can be more appropriate (numerical simulations are very encouraging). Moreover this new method can present significant advantages in other spectral range (UV, mid-IR). The condition is to find a three-level system which allows a pump-probe scheme. The pulse sequence could be implemented in a simpler set-up using a modified Michelson interferometer instead of the pulse shaper. The spectral resolution here is not limited by that of the pulse shaper but only by the temporal interval scanned and ultimately by the linewidth of the atomic transition (typically $10^{-6}$), which could be order of magnitude improvements with respect to other methods based on a spectrometer. There is no intrinsic limitation for the bandwidth or the central frequency of the unknown pulse provided that a system with resonant transitions exist in this range. The reference pulse bandwidth should be as large as this of the unknown pulse. We sincerely acknowledge Christophe Dorrer and Manuel Joffre for fruitful discussions and advices.

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