Single-shot spectral interferometry with chirped pulses

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We present a method for obtaining time-resolved measurements of the amplitude modulation and the phase shift of a chirped probe pulse interacting with a femtosecond-laser-produced plasma. Based on spectral interferometry, the technique allows for single-shot measurements and keeps the temporal resolution associated with the full bandwidth of the probe pulse. We demonstrate the efficiency of this technique by probing femtosecond-laser breakdown of plastic targets. © 2001 Optical Society of America

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Spectral interferometry, also called Fourier-domain interferometry (FDI), is a well-known linear optical technique\(^1\) for retrieving the phase and the amplitude of an unknown electric field \(E(t)\) in the femtosecond time domain.\(^2,3\) This technique is now widely used in ultrafast laser–matter interaction experiments, including molecular spectroscopy,\(^4\) laser-induced dielectric damage,\(^5\) laser wakefield particle acceleration,\(^6\) femtosecond shock waves,\(^7\) and plasma ionization and hydrodynamics studies.\(^8\)

In spectral interferometry, the perturbed laser pulse to be characterized is set up to interfere with a reference pulse in a spectrometer, yielding spectral fringes of a period that is inversely proportional to the optical path difference between the two beams and of a contrast that is proportional to the perturbation amplitude. Obtaining a complete time history of the perturbation under study requires multiple shots.\(^9\) With high-energy lasers, one way to circumvent the low-repetition-rate problem lies in using a linearly chirped probe, giving a univocal relation between the instantaneous frequency and time to encode the pulse spectrum with the time history of the perturbation. Along these lines, twin-chirped-pulse FDI was implemented with picosecond time resolution in shock wave studies\(^9\) and laser-induced breakdown of air.\(^10\)

In this Letter we discuss the ultimate time resolution that can be achieved with such a technique. This question was raised recently by Chien et al.\(^10\) Those authors said that they are able to achieve a time resolution of a few tens of femtoseconds with this technique, just by improving the spectral resolution. This claim is questionable solely on the basis of their simple interpretation of the time–frequency relation. Here we propose a signal reconstruction procedure that will in fact preserve the time resolution associated with the probe pulse’s spectral bandwidth. The single-shot FDI technique relies on the use of a positive or negative linearly chirped pulse with

\[
\omega(t) = \omega_0 + at, \quad (1)
\]

where \(\omega_0\) is the central frequency of the laser and \(a = 1/\Phi^{(2)}(\omega_0)\) is the inverse of the quadratic phase. Accordingly, the time range over which single-shot measurements can be made is \(\Delta t \approx \Delta\Omega \times \Phi^{(2)}(\omega_0)\), where \(\Delta\Omega\) is the FWHM pulse bandwidth. Because there is a linear relation between frequency and time [see Eq. (1)], a phase and amplitude perturbation of duration \(\delta t\) in the time domain is encoded over a spectral bandwidth \(\Delta\omega = a \times \delta t\) in the frequency domain. For a Gaussian pulse, the FWHM duration \(\Delta T\) (intensity) of the chirped pulse is

\[
\Delta T = \Delta T_0 \left[1 + \frac{4 \ln 2 \Phi^{(2)}(\omega_0)^2}{(\Delta T_0)^2}\right]^{1/2}, \quad (2)
\]

where \(\Delta T_0\) is the Fourier-limited duration of the pulse. By combining the time–bandwidth product and the linear chirp, we obtain a value of the minimum perturbation duration \(\delta t\) for which the linear chirp bandwidth is comparable with the Fourier bandwidth:

\[
\delta \omega = \Delta \omega = a \delta t = \frac{4 \ln 2}{\delta t}, \quad (3)
\]

\[
\delta t^2 = \frac{4 \ln 2}{a}, \quad (4)
\]

or [note that the second term inside the square root of Eq. (2) is much larger than unity]

\[
\frac{\delta t}{\Delta T_0} \approx \left(\frac{\Delta T}{\Delta T_0}\right)^{1/2}. \quad (5)
\]

Indeed, for a short perturbation of duration \(\delta t_p\) much smaller than \(\delta t\), the perturbation spectral bandwidth \(d \omega_p\) becomes much larger than \(\Delta \omega_p\) given by \(\Delta \omega_p = a \times \delta t_p\). There is no longer a direct relation between instantaneous frequency and time because the sharp perturbation transients produce spectral modulations that extend over the entire pulse bandwidth.

In FDI, one determines the phase shift and amplitude modulation of the perturbed probe pulse by comparing the results of a signal shot and of a reference shot. In the twin-probe configuration (two identical time-delayed pulses),\(^3\) the fringe spacing is determined by the temporal delay between the two pulses. To fully exploit the temporal range associated with the bandwidth \(\Delta \Omega\) of the probe pulse, one has to separate the pulses by a time delay much longer than the stretched pulse duration \(\Delta T\). Accordingly, achieving this separation requires the use of a high-resolution spectrometer with a detector that has a large number of pixels.

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of pixels to resolve tiny fringe shifts. We prefer to use a space-shifted implementation of FDI in which we obtain fringes in the spatial domain by shifting and tilting two interfering probe pulse wave fronts in the two arms of a Mach–Zehnder interferometer, one from the unperturbed part of the sample and one from the perturbed part. The spectral axis is parallel to the direction along the fringes, and the fringe separation is now related to the angle of the two beam wave fronts and can be chosen to match the detector’s spatial resolution. A two-dimensional interferogram consists of one spatial dimension \( x \), which is perpendicular to the fringes, and one spectral dimension \( \omega \), which lies along the direction of the fringes. Taking a lineout perpendicular to the direction along the fringes, one gets the spectral power \( I(x) \) (here we use notation that applies to the space-shifted variant of FDI, but the analysis is equally valid for time-shifted FDI):

\[
I(x) = |f(\omega)|^2 \left( 1 + R(x)^2 + 2R(x)\cos \left( \frac{2\pi x}{d} + \Phi(x) \right) \right),
\]

where \( R(x) \) and \( \Phi(x) \) are the reflection coefficient and the phase difference to be determined, respectively, \( f(\omega) \) is the spectral envelope, and \( d \) is the unperturbed interferfringe distance. The inverse Fourier transform of \( I(x) \) shows three peaks in the \( k \) domain; the two interesting (oscillating) peaks are located at \( \pm k_0 = 2\pi d \). Isolating one of the lateral peaks and performing the Fourier transform back into the \( x \) domain on both the signal shot and the reference shot, we get

\[
I_{\text{signal}}(x) = |f(\omega)|^2 [R(x)\exp(ik_0 x + \Delta \Phi(x))],
\]

\[
I_{\text{ref}}(x) = |f(\omega)|^2 [\exp(ik_0 x)].
\]

From the ratio of these quantities, and performing the preceding analysis for many \( \omega \) lineouts, we easily obtain the coefficient perturbation of the complex field reflection:

\[
P(\omega, x) = \sqrt{R(\omega, x)} \exp[i\Delta \Phi(\omega, x)],
\]

where \( R(\omega, x) \) is the amplitude modulation and \( \Delta \Phi(\omega, x) \) is the phase shift induced by the perturbation. With knowledge of chirp parameter \( a \), which can easily be determined experimentally, the original time resolution can be recovered. The field of the chirped pulse can be written in the frequency domain:

\[
E_0(\omega) = \Sigma_0(\omega)\exp[i(\omega - \omega_0)^2/a].
\]

We construct the signal by multiplying Eq. (9) by Eq. (10) such that

\[
E(\omega) = E_0(\omega) \times P(\omega),
\]

where we have dropped the second dimension for the sake of simplicity. Field perturbation \( P(t) \) in the time domain can now be easily recovered by use of an inverse Fourier transform:

\[
P(t) = \mathcal{F}^{-1}[E(\omega)/E_0(t)],
\]

where \( E_0(t) \) is the original chirped pulse in the time domain.

The experiments were performed at the Laboratoire d’Optique Appliquée (Palaiseau) with a titanium:sapphire chirped-pulse amplification laser capable of delivering 10-Hz, 35-fs, 150-mJ pulses at 0.82-µm wavelength. The experimental system has been fully described elsewhere. To summarize, a main beam (\( \lambda = 0.82 \) µm) is focused to an irradiance of \( \sim 10^{13} \) W/cm² upon a plastic (C₃H₈) target with a mirror parabola of 1-m focal length. A weak probe beam, frequency doubled to 0.41-µm wavelength, is focused onto a target such that its focal spot size is much larger than the 60-µm pump focal spot. This difference in size is mandatory to permit interference between perturbed and unperturbed regions of the target. We stretch the 35-fs probe pulse to typical values of 3 ps by slightly detuning the pulse compressor gratings. After reflection from the target, the probe focal is imaged on the entrance separation prism of a Mach–Zehnder interferometer. The two beam wave fronts are slightly tilted and shifted such that perturbed and unperturbed regions of the probe beam can interfere. The entrance prism is image relayed on the entrance slit of a 1-m homemade Czerny–Turner spectrometer. A 16-bit charge-coupled-device camera records the spectrum. Figure 1 shows the normalized...
Fig. 2. Single-shot normalized (a) reflection coefficient and (b) phase shift of a 3-ps chirped probe beam interacting with a plastic breakdown plasma. The signal reconstruction technique was used.

The reflection coefficient ($R_0$ is the unperturbed reflection coefficient of the target) and the phase shift extracted directly from the interferogram by use of the linear relation between wavelength (frequency) and time.

Because plastic breakdown occurs in less than a few tens of femtoseconds, thus strongly violating the criterion of relation (5), many spurious oscillations are observed in the perturbation amplitude and phase. In applying the signal reconstruction technique described above, we obtain the results shown in Fig. 2, where we restrict our attention to the region of transient-time breakdown (around the time 1000 fs). We clearly recover a time resolution that is comparable with the duration of the Fourier-limited probe pulse.

The rapid phase and reflectivity jumps are signature of laser breakdown at the surface of the target. The delay of ~50 fs between the onsets of the phase variation and of the reflectivity variation is easily explained, within the framework of the Drude model, by the greater sensitivity of the phase (real part of the dielectric constant) to the rise in electron density during the transient ionization. The later decay of the phase is the signature of plasma expansion.

We have shown that the ultimate temporal resolution in single-shot chirped-pulse FDI is determined by simultaneous consideration of the Fourier time–bandwidth product and the linear relation between frequency and time. With a simple signal reconstruction procedure based on knowledge of the chirp parameters, we demonstrate a time resolution that is comparable with the duration of the Fourier-limited probe pulse.

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