Three envelope approach for ultrafast pulse characterization in a pump-probe experiment

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We demonstrate an approach for the complete characterization of temporally identical ultrashort pulses at the focal point in a pump-probe experiment for potential use of pump-probe data deconvolution. Our approach uses three envelope measurements, autocorrelation, fundamental, and second harmonic spectra, combined with an error minimization pulse retrieval scheme. The three envelope approach is suitable when the measured envelopes have low noise and minimal systematic errors. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2837660]

Ultrashort pump-probe experiments[1] are commonly used to study various dynamics, such as molecular reorientation time constants,[2] wave-packet evolution,[3] quasiparticle dynamics in semiconductor and correlated electron materials,[3] depth by time of flight,[4] etc. Sometimes, short dynamics comparable to the probe pulse width are encountered. For such data, it is desirable to know the exact shape of the pulses at the sample site so that an accurate dynamical response can be obtained through data deconvolution.[5] Most deconvolutions reported in the literature use only an estimate of the ultrashort laser pulse intensity at the sample, as derived from a conventional autocorrelation trace, which is known to yield incorrect results.[6] Complete pulse characterization at the focal spot is necessary in such cases. Several methods exist for amplitude and phase characterization of ultrashort laser pulses.[7–9] However, only a few of these schemes can be easily adapted to the characterization at the sample site. For example, a second harmonic generation frequency resolved optical gating[7] (SHG-FROG) can be performed by replacing the sample with a second harmonic crystal and collecting the FROG trace in a spectrometer as the relative delay between the pump and probe is varied. Another technique that can be implemented in the pump-probe geometry is "phase and intensity correlation and spectrum only."[9] Spectral phase interferometry for direct electric-field reconstruction (SPIDER),[8] another widely used technique, is not easy to implement because it requires an additional chirped pulse. Most pump-probe measurements are done in a focusing geometry with noncollinear beams. This makes several other techniques including SPIDER even harder to implement.

In this article, we demonstrate a complete pulse characterization approach that is simple to implement without requiring any further modification to an existing degenerate pump-probe setup. The results from our approach have the potential for pump-probe data deconvolution. The approach is based on measuring an autocorrelation (pump and probe are identical in time) and the spectra correspond to the fundamental and second harmonic of the pump. Since this method involves less data collection, it is faster and easier to implement. Using the three measured envelopes, we reconstruct the pulse amplitude and phase with the help of a line-minimization-based error reduction method.

In 1989, Naganuma *et al.*[10] proved that the electric field $E(\tau)$ is uniquely determined by the magnitudes of three different spectral envelopes, namely, the magnitudes of the intensity spectrum $|\tilde{I}(\Omega)|$, the second harmonic spectrum $|\tilde{E}_2(\Omega)|$, and the fundamental field spectrum $|\tilde{E}(\Omega)|$. Here, Ω is the frequency with respect to the carrier frequency of the corresponding field. In general, all the three envelopes can be obtained using first and second order interferometric autocorrelations, as were done in experiments before.[10–12] However, interferometric autocorrelations are not possible in the case of a typical noncollinear pump-probe experiment. Here, we demonstrate an approach where the three envelopes mentioned above are directly measured. The fundamental field spectrum is easily measured by directly placing a spectrometer slit close to the focal point. The second harmonic spectrum can also be measured similarly by including a SHG crystal at the focus. The intensity spectrum can be obtained using a background free intensity autocorrelation. The Fourier transform of the autocorrelation is proportional to the magnitude squared of the intensity spectrum. For the first time, we demonstrate complete characterization of an ultrashort pulse using three such separate measurements. For the reconstruction, we use an algorithm that we recently developed.[12]

A common criticism against one-dimensional scheme is that they are not suitable to distinguish pulses that have similar autocorrelation and spectrum.[6] Recently, we showed that it is possible to distinguish such pulses when the signal to noise of measurements is sufficiently high.[13] Our experimental measurements have good signal to noise ratio (>200).

Our experimental arrangement consists of pump and probe pulses derived from a Ti:sapphire oscillator producing pulses that support a spectral bandwidth of approximately 40 at 800 nm. The pulses first go through a prism-based chirp precompensation unit to compensate for the dispersion in the

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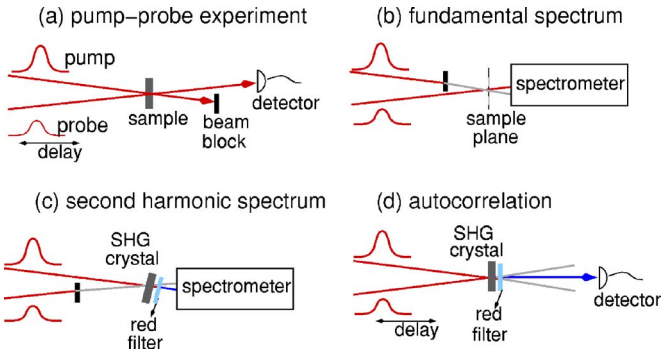

FIG. 1. (Color online) Schematic of a pulse characterization scheme at the sample site in a pump-probe experiment. (a) Typical pump-probe setup, (b) measurement of fundamental spectrum, (c) second harmonic spectral measurement, and (d) intensity autocorrelation.

optics. The prechirped pulses split into two, one weak and one strong, and they go through optics of similar glass thickness, which are used for pulse modulation, polarization control, and beam attenuation. The pulses are perpendicularly polarized and noncollinearly focused at the sample site with different focal length lenses (100 and 175 mm) which have nearly the same thickness. The angle between the pump and probe is approximately 18°. The pulses at the focus can be considered to be nearly identical in their temporal shapes. Our approach for measuring the three envelopes of interest is shown in Fig. 1. The linear spectrum is directly measured with an ocean-optics spectrometer with a resolution of 0.38 nm [Fig. 1(b)]. A 30 μm thick, type-I, beta barium borate (BBO) crystal is used to measure the second harmonic spectrum with the same spectrometer [Fig. 1(c)]. The BBO crystal is placed normal to the incoming beam during the measurement. In order to obtain the intensity spectrum, we measure the intensity autocorrelation with copolarized pulses [Fig. 1(d)], obtained by simply rotating the halfwave plate in the probe line. The BBO crystal is placed in such a way that the normal bisects the two incoming beams. The background free autocorrelation is obtained by scanning the probe pulse in time.

The experimental data are shown in Fig. 2 as solid

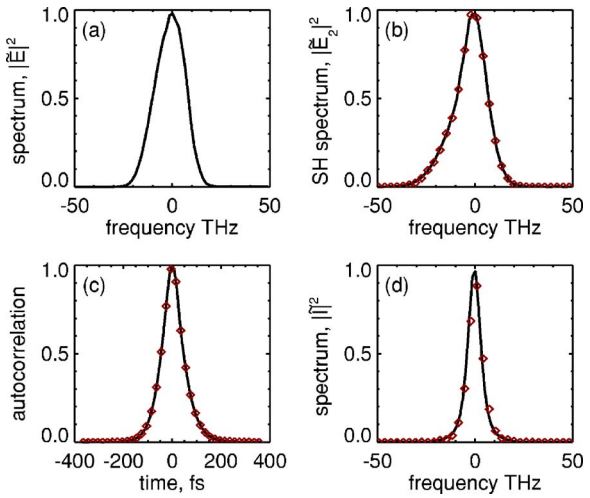

FIG. 2. (Color online) Measured experimental data (solid lines) used for retrieval of the pulse shape. Symbols are calculated using the retrieved electric field. (a) Fundamental spectral intensity, (b) second harmonic spectral intensity, (c) intensity autocorrelation, and (d) Fourier transform of intensity autocorrelation.

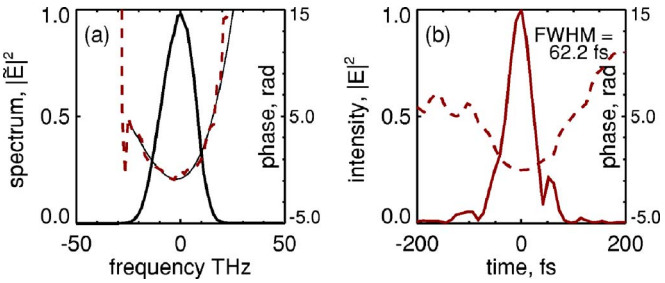

FIG. 3. (Color online) Reconstruction results. (a) Measured spectral intensity (thick solid line) and adapted phase (dotted line). (b) Temporal intensity and phase of the retrieved optical pulse.

curves. The fundamental and second harmonic spectra are shown in Figs. 2(a) and 2(b). The measured autocorrelation and its corresponding Fourier transform are shown in Figs. 2(c) and 2(d), respectively. Our goal is to use the three spectral envelopes to determine the unknown phase of $\tilde{E}(\Omega)$. With a guess for this unknown phase $\phi_g(\Omega)$, we can calculate a guess field $E_g(\tau)$. Using the guess field, intensity $\tilde{I}(\Omega)$ and second harmonic spectra $\tilde{E}_2(\Omega)$ are determined. The phase $\phi_g(\Omega)$ is iteratively updated using a line-minimization scheme until the error Δ between the measured and adapted spectral magnitudes is minimized. Our error metric is defined as $\Delta=(1/N)\Sigma_i^N(|\tilde{I}_g(\Omega_i)|-|\tilde{I}(\Omega_i)|)^2+(1/N)\Sigma_i^N(|\tilde{E}_{2g}(\Omega_i)|-|\tilde{E}_2(\Omega_i)|)^2$. A good starting guess for ϕ_g is to use a Taylor expanded phase that minimizes Δ. With this starting guess, an arbitrary unknown phase can be determined iteratively. The results of reconstruction are shown in Figs. 2 and 3. The adapted phase for the spectrum is shown in Fig. 3(a). Using the adapted phase and measured spectrum, the temporal intensity and phase are determined [Fig. 3(b)]. From this retrieved field, the second harmonic spectrum and autocorrelation are calculated and are shown as diamonds in Fig. 2. There is good agreement between the measured and calculated profiles in Fig. 2. The final error Δ is 0.000 71 after 270 iterations using a Powell line-minimization technique.[14]

The reconstructed spectral phase in Fig. 3(a) is asymmetric about the center frequency, implying that the pulse has a contribution from third order dispersion also. Until this point, the only ambiguity is time direction or sign of spectral phase. We determine that the dispersion is positive by placing a glass with known dispersion in one of the arms and noting that the autocorrelation width increased, thus resolving the time direction. We fit a polynomial (shown as thin solid line) to the frequency phase and find that the second and third order dispersions are +418 (Ref. 2) and +887 fs,[3] respectively. From the determined pulse shape, one can deconvolve pump-probe data[5] to reveal the dynamics at the time scales of pump pulse width.

In principle, the three envelope technique can also reconstruct complicated pulse shapes. Figure 4(a) shows numerically generated complex pulse (solid lines) and reconstructed (symbols) pulse shape using the three envelope method. Figure 4(b) shows the reconstruction results when the three envelopes have ≈12% intensity noise (single sided normal distribution) and 2.5% random frequency calibration noise. This reconstruction resembles the original. Although this example illustrates that complicated pulses can also be reconstructed in the presence of noise, ambiguous reconstructions arising due to noise are not entirely ruled out.

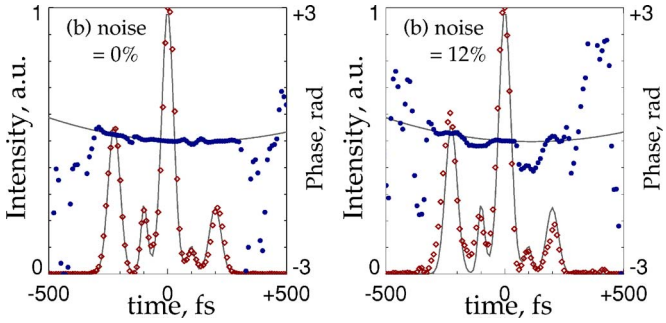

FIG. 4. (Color online) Reconstruction simulations of a complicated pulse in the presence of (a) no noise, and (b) intensity noise=12%. Solid lines correspond to original pulse and symbols (diamonds are intensity, and filled circles are phase) represent reconstruction results.

Some ambiguities are possible with this technique and are considered in a different study.[15]

We have demonstrated a simple three envelope scheme for the complete characterization (no time-direction ambiguity) of the amplitude and phase of temporally identical pump and probe pulses at the sample site in a pump-probe spectroscopy experiment. A line-minimization-based algorithm is used to reconstruct the pulse shape in time. This approach can be used elsewhere for ultrafast pulse characterizations. Some ambiguities of this technique are addressed in a different article.

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[1] *Ultrafast Phenomena XIV: Proceedings of the 14th International Conference* (Springer, Berlin, 2004).
[2] H. E. Lessing and A. V. Jena, Chem. Phys. Lett. **42**, 213 (1976).
[3] J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer, Berlin, 1999).
[4] B. Bonello, B. Perrin, E. Romatet, and J. C. Jeannet, Ultrasonics **35**, 223 (1997).
[5] N. E. Henriksen and V. Engel, J. Chem. Phys. **111**, 10469 (1999).
[6] J. H. Chung and A. M. Weiner, IEEE J. Sel. Top. Quantum Electron. **7**, 656 (2001).
[7] K. W. DeLong, R. Trebino, J. Hunter, and W. E. White, J. Opt. Soc. Am. B **11**, 2206 (1994).
[8] C. Iaconis and I. A. Walmsley, Opt. Lett. **23**, 792 (1998).
[9] J. W. Nicholson, J. Jasapara, W. Rudolph, F. G. Omenetto, and A. J. Taylor, Opt. Lett. **24**, 1774 (1999).
[10] K. Naganuma, K. Modi, and H. Yamada, IEEE J. Quantum Electron. **25**, 1225 (1989).
[11] T.-W. Yau, Y.-Y. Jau, C.-H. Lee, and J. Wang, Tech. Dig. Ser.-Opt. Soc. Am. **1999** 263.
[12] B. Yellampalle, R. D. Averitt, and A. J. Taylor, Opt. Express14, 8890 (2006).
[13] B. Yellampalle, E. E. M. Chia, K. Kim, R. D. Averitt, and A. J. Taylor, Tech. Dig. Ser.-Opt. Soc. Am. **2007**.
[14] W. Press, B. Flannery, S. Teukosky, and W. Vetterling, *Numerical Recepies in C: The Art of Scientific Computing* (Cambridge University Press, Cambridge, 1986).
[15] B. Yellampalle, K. Kim, and A. J. Taylor, Opt. Lett. **32**, 3558 (2007).