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Author(s): F. Omenetto
            B. Luce
            C.W. Siders
            A.J. Taylor

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Observation of Optical Pulse and Material Dynamics on the Femtosecond Time Scale

F. G. Omenetto,
B. P. Luce, C. W. Siders(*) and A. J. Taylor

Los Alamos National Laboratory, Materials Science and Technology Division, MST-10
Ms K764, Los Alamos, NM 87545, USA

(*) University of San Diego, Dept. of Chemistry, La Jolla, Ca. 92210, USA

Abstract---The widespread availability of lasers that generate pulses on the femtosecond scale has opened new realms of investigation in the basic and applied sciences, rendering available excitations delivering intensities well in excess of $10^{21}$ W/cm$^2$, and furnishing probes capable of resolving molecular relaxation timescales.

As a consequence and a necessity, sophisticated techniques to examine the pulse behavior on the femtosecond scale have been developed and are of crucial importance to gain insight on the behavior of physical systems. These techniques will be discussed with specific application to guided pulse propagation and ionization dynamics of noble gases.

I. INTRODUCTION

Any significant progress is dictated by the ability to carefully observe the experimental outcome of the process under investigation, which becomes most challenging at time scales that approach the fastest events in nature.

Ultrafast optics has significantly impacted science and technology over the course of the last three decades. Experiments using short pulse lasers have shed light on the nature of dynamical phenomena in the fields of chemistry, biology, electrical engineering, and physics. Generation of 100-fs optical pulses across a wavelength spectrum reaching from the vacuum ultraviolet to the far infrared is now routinely achieved, with the shortest pulses lasting only 4.5 fs [1]. Laser technology, pulse characterization strategies, and techniques for physical measurement are now so mature and reliable that applications of ultrafast technology are being realized in the areas of optical communications, material processing, imaging techniques, and medicine, on top of basic research.

The discovery of new physical phenomena has always accompanied the increasingly high intensities that were made available from shortening laser pulses and re-engineering lasers to furnish more energy.

While there might be an engineering limit or possibly an upper limit for the power deliverable by a single laser pulse, there are still ways to add great versatility by shaping an ultrafast pulse[2], thus delivering a preconfigured excitation pattern at the timescales of an electron-electron interaction, opening a new realm of investigations which have been recently referred to in the literature as coherent control experiments[3,4]. It is of the utmost importance to be able to understand in detail the behavior of pulsed light and systems under investigations at these short timescales, and diagnostic methods have been developing and refining as the laser pulses have shortened and become complex.

Recently, the techniques of Frequency Resolved Optical Gating (FROG)[5], Multipulse Interferometric Frequency Resolved Optical Gating (MIFROG) [6] and variants on the autocorrelation techniques [7], have furnished a full characterization of the electric field, which in turn has been of great help in understanding the underlying dynamics of physical phenomena.

FROG has impacted ultrafast diagnostics by combining the temporal and spectral information of short pulses into one signal. The result is a FROG trace (or spectrogram) which can be reconstructed by numerical methods to furnish the temporal profile and the phase of the pulse. This technique has been crucial in the determination of the role of higher phase terms in short-pulse modelocked oscillators, on accurate characterization of pulsed laser sources at different wavelengths and intensities, and has found application as a tool to analyze the nonlinear effects that occur during propagation in bulk media and guided propagation and nonlinear effects in plasmas.

The capacity of resolving an optical phase shift on the femtosecond scale gives access to the underlying physics on a multitude of phenomena such as multi-terabit communications, Terahertz optoelectronics, generation of ultrafast ionization-based sources of VUV and X-ray radiation and laser-driven coherent processes in plasmas, to name a few.
II. EXPERIMENTS: GUIDED PROPAGATION AND IONIZATION

Optical solitons have been a topic of great interest since the suggestion of their existence by Hasegawa and Tappert in 1971 [8] and their first experimental observation by Mollenauer in 1980 [9]. This interest has intensified in recent times given the great promise that solitons hold as carriers of information for telecommunication systems. Yet, a full experimental description of perturbed soliton propagation and soliton-soliton interaction on femtosecond time scales has been elusive. This is partially due to the practical limitations of conventional measurement techniques and partially to the complexity of third-order effects on pulse propagation. An understanding of these effects acting on ultrashort pulse propagation in single-mode optical fibers is, however, crucial for the development of the next generation of high capacity communication networks. Recently, detailed phase characteristics of soliton evolution have been obtained for the first time, by using FROG [10]. The experiment was performed by propagating $\lambda=1.55 \mu m$, $E=3.5 \text{ nJ}$, $\tau=158 \text{ fs}$ linearly chirped pulses generated by an optical parametric oscillator (OPO) (Spectra-Physics Opal) through a 10-meter link of conventional single-mode optical fiber (Corning SMF-28) and by detecting the output pulses from the fiber with FROG. The pulse energies are carefully varied before being launched into the fiber with combinations of neutral density (ND) optical filters.

With increasing pulse peak power, temporal narrowing is expected as the nonlinearities increasingly counteract the linear dispersion that dominates at low input powers. As the input energy increases, the temporal width of the spectrograms systematically decreases due to the increasing effect of the Kerr nonlinearity. The amplitude and phase profiles reconstructed from the experimental FROG trace corresponding to the observation of the soliton is shown in figure 1. It is evident that the phase profile has flattened across the center of the pulse, which is a signature of soliton formation. The phase behavior not observable directly with other techniques and this measurement confirms the theoretical inferences that are available in the literature. Further examination of the dynamics leading to soliton formation has furnished more insight on their behavior. Comparing the asymptotic profiles (i.e. the profiles of a soliton predicted to form by theory over an infinitely long fiber) to those occurring at 10 meters, the phase dynamics of soliton formation can be deduced. The phase profiles do not flatten instantaneously until the energy of the pulse renders the magnitude of the Kerr effect large enough to fully balance the dispersion and produce the linear phase characteristic of the soliton. It is also quite interesting that $\pi$-phase are seen to be forming, representing a different behavior between the wings of the pulse (less intense thus less prone to balance the linear and nonlinear dispersion contributions) and the central more intense portion of the pulse (where the compensation of the linear and nonlinear terms is more efficient. In the case shown in Fig. 1, the energy of the pulse is 318 pJ and analysis of the pulse behavior through simulation agrees perfectly with the reconstructed data, showing two fully formed $\pi$-phase slips.

![Figure 1](image_url)

**FIGURE 1** Characterization of soliton formation in a single-mode optical fiber for telecommunications. The duration of the pulse is 233 fs at a wavelength of 1.55 microns.

Note also that the phase is now almost perfectly linear across the pulse revealing that features of the asymptotic soliton have become strongly evident. The ability to measure the phase profiles strongly cross-checks and completes the comparison between theory, simulation, and experiment, allowing us to assert with some confidence that we understand the pulse dynamics. The application of this experimental technique to fiber-based propagation studies at this crucial wavelength for telecommunication applications offers the potential for greater understanding of the
physical mechanisms which govern the dynamics of ultrashort pulses in guided propagation. Phase dynamics are quite important, and not fully understood, in soliton-soliton collisions, polarization mode dispersion effects, and so forth. The application of this method promises to be of key importance in exploring mechanisms that can enhance the capacity of optical communication systems, pushing the frontier of optical communications into the femtosecond regime.

When short pulse excitations are applied to materials, and sensitivity to constant and slowly varying terms in the phase are required, FROG is not sufficient. To this end, a variation that combines spectral interferometry and FROG called Multipulse Interferometric FROG (MI-FROG) [6,12] can be employed. The unique abilities of MI-FROG center around its inherently differential nature: ultrafast changes in the time-domain phase of a laser pulse are directly measured with interferometric accuracy to all orders, unlike standard FROG, while shot-to-shot fluctuations in pulse structure are discriminated against.

Additionally, a time-domain phase shift can be measured, over a time-scale much longer than the pump pulse which induces it, unlike similar measurement techniques which adopt spectral (longitudinal) interferometry. The real-time nature of MI-FROG allows immediate and direct observation of gate-pulse time scale dynamics without computational analysis, a significant advantage over any previous FROG technique, while iterative computational techniques combined with interferogram analysis can elucidate femtosecond detail from the MI-FROG trace. A tool of this kind, which allows real-time observation of the phase, would clearly be useful in mapping numerous ultrafast phenomena while the immediate, direct viewing of temporal phase would be advantageous in the optimization, adaptive feedback, and control of, for example, laser-driven plasma-based accelerators or ionization-front based sources of tunable radiation. Among those phenomena being studied with MI-FROG are high-field atomic and molecular ionization fronts, laser driven wakefields in plasmas, and ultrafast electro-optic dynamics in technologically relevant materials. MI-FROG can also be used to measure fast (fs) or slow (ps) time-scale changes in refractive index caused by atomic or molecular response to a copropagating pump pulse.

An experiment that confirms this statement is illustrated in figure 2. A standard chirped pulse amplified laser system was used to provide 1 mJ, 175 fs, 802-nm pulses at one kHz. Twenty percent of this pulse was used to generate, in a Michelson interferometer, the multi-pulse probe and gate sequences while the remainder was frequency doubled in a 300 μm BBO crystal, providing a 30 μJ, 401-nm pump pulse. After dumping the residual IR, the UV pump and attenuated IR probes, all of the same linear polarization, were collinearly focused in a 10-cm focal length telescope. The gate pulses traveled a separate path to a modified FROG apparatus while the probes were dichroically separated from the pump and entered the same FROG apparatus. A 500 μm fused silica plate, thinner than the 650 μm walkoff length, was translated axially near the focus to induce an adjustable amount of cross-phase modulation (XPM).

![Figure 2](image-url)

**Figure 2** MI-FROG (bottom) and trailing pulse FROG (TREEFROG) traces (top) measured in the case of cross phase modulation in fused silica for a ±100 fs (sides), and 0 fs (center) delay between the pump and the probe.

Three sets of MI-FROG traces were recorded both with and without the pump pulse for 0–fs, and 100 fs delay between the pump and trailing probe pulse (the illustrations in fig.2 show the pumped case). The pulses show characteristic blue/red shifts for the delayed cases and a symmetric broadening for the overlapped case, with shifts of ~2.5 nm. When the...
probe rides the leading/trailing edge of the pump, the probe is red/blue-shifted, as expected from the $n_2 l(t)$ nature of the XPM. The MI-FROG traces, all of which have straight fringe patterns with the pump blocked, display a characteristic fringe tilt, indicative of the clean blue/red shift, while the coincident case evidences a downward curvature. That is to say, the fringe pattern follows the time-domain phase shift $\varphi(t) = -n_2 l(t) \alpha z / c$. Analysis [11] of the reconstructed pulse profiles further reveals excellent agreement with the data.

Also present is a significant change in the intensity profiles of the probe pulse near the peak of the pump, which is attributable to the lens-like transverse profile of the XPM. It should be stressed that both the DC component of the phase shift as well as the relative temporal offset between the pulses are obtained from the MI-FROG data with interferometric accuracy and no adjustable parameters are used, unlike standard FROG techniques.

III. CONCLUSION

Characterization of the phase dynamics of a pulse on the femtosecond timescale provides a very powerful tool for the analysis of physical phenomena induced by ultrashort pulsed fields of light. The detail available through observation of the behavior of short pulses sheds unprecedented light on the dynamics of the physical phenomena under observation. The future outlook will draw from the power of observation leading towards further understanding and a more pointed experimental implementation.

Through control of the electric field on such short timescales, phase sensitive experiments can open the way to an unprecedented degree of control allowing tailored excitations at various levels of focused intensity.

The implications of this approach are of fundamental importance because they allow to interact with extreme power and equally extreme finesse on a physical system opening new pathways for science.

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