Ultrafast Temporal Shaping
Is Coming of Age

Temporal shaping will play a significant role in the emerging femto revolution, especially for futuristic biomedical applications.

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Thanks to advances in ultrafast laser design, ultrafast laser sources have emerged from the laboratory and are now used for commercial applications ranging from ophthalmic surgical procedures such as lasik to cutting sapphire wafers.

Ultrafast lasers have a long history in the lab environment: They have been used in research laboratories since the '70s (picosecond) and '80s (femtosecond). The early interest in these sources was that they represented a new "stopwatch" that could be used for timing ultrafast phenomena. In fact, the 1999 Nobel Prize in chemistry was awarded to Ahmed H. Zewail for the development of femtochemistry.

Among the many other scientific developments that have derived from ultrafast lasers are frequency combs, which yielded a Nobel Prize in physics (shared by John L. Hall and Theodor W. Hänsch) in 2005, as well as nonlinear optical microscopy, including second- and third-harmonic generation microscopy[1,2] and two-photon excited fluorescence microscopy.[3]

Temporal shaping in the form of adjustable dispersion has been a part of ultrafast lasers since the early femtosecond laser systems were built by Charles V. Shank and co-workers at the AT&T Bell Laboratories.[4] At that point, it was clear that group velocity dispersion control was needed to produce reliable femtosecond laser pulses.

In some cases, introducing group velocity dispersion in the form of linear chirp can be useful, as in chirped pulse amplification (CPA), which depends on stretching the pulses before amplification and compressing them afterward. All commercial lasers include static temporal shaping; in the near future, however, we can expect adaptive temporal shaping that adjusts the laser output to maintain stricter specifications and more reproducible outcomes. We also anticipate on-the-fly pulse optimization that responds to changes in the target.

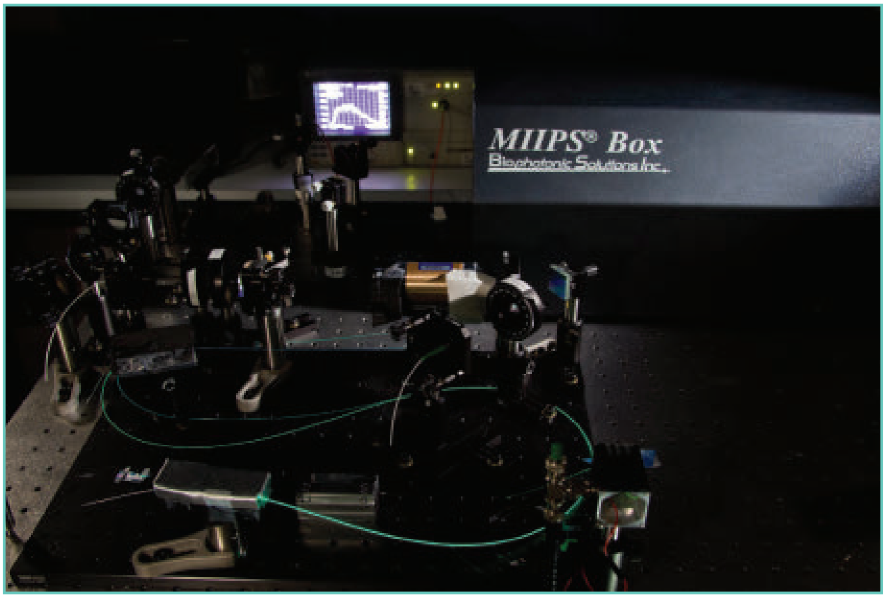
Temporal shaping: background

The seminal work of Edmond Treacy of the United Aircraft Research Labora-

Table 1.

Modulator Type	Wavelength Range	Modulation Rate	Number of Elements	Development Stage
Acousto-Optic Modulators				
Fourier Plane	Germanium: Mid-IR TeO_2: 650-1200 nm SiO_2: 200-400 nm	5 kHz 5 kHz 5 kHz	Depends on function generator	Available commercially for research
In-Line	Hg_2Cl_2: Mid-IR TeO_2: 650-1200 nm SiO_2: 250-400 nm	5 kHz 5 kHz 5 kHz	Depends on function generator	Available commercially for research
Liquid Crystals				
LCD	450-1900 nm	10 Hz	128 or 640	Available commercially for research
LCOS	400-1900 nm	60 Hz	800 × 600	Industrialized
LCOS	532-1550 nm	200 Hz	12,288	For research
Deformable Mirrors				
SiN Membrane	200-2000 nm	8 kHz	140	Available commercially for research
Solid	VUV to Mid-IR	100 Hz	<40	Available commercially for research

LCOS - Liquid crystal on silicon.



Yb fiber laser with adaptive pulse shaping capable of producing 42-fs pulses. Images courtesy of the Dantus Research Group at Michigan State University.

tories on optical stretcher and compressor designs in the late ’60s, together with the theoretical work by Oscar Martinez in the ’80s, laid the groundwork for pulse compression. A 1969 quote from Treacy[5] illustrates how the utility of temporally shaped pulses was not a chief concern: “Although there is probably very little to be gained at the present time from the extension of chirp radar techniques to optical wavelengths, there are some good reasons to explore the physics of chirped pulses in the optical region.”

A decade later, the reason to pursue optical designs to control the chirp in laser pulses was pulse stretching and compression. It is worth noting that at present, CPA and fiber-CPA, based primarily on grating stretcher and compressor, have allowed the design of ultrafast lasers for medical, industrial and all scientific applications using amplified laser sources.

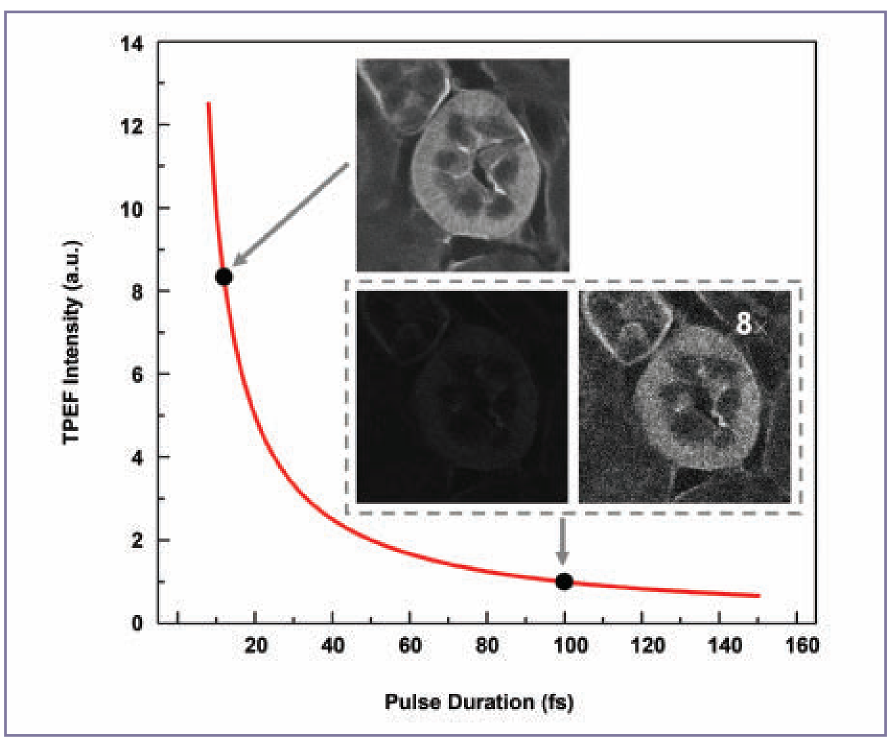
Since the late ’70s and early ’80s, researchers have realized that gratings and prisms alone are not sufficient to control the temporal shape of ultrafast laser sources. This observation led to the early designs for adaptive pulse shapers to control pulses in the time domain[6] and the frequency domain,[7,8] and took advantage of chirped pulses to control their temporal shape in the time-frequency domain.[9,10] In the early days of pulse shaping, it was not unusual to find research groups using paper clips, cut-out aluminum foil and microscope coverslips placed at specific locations to control the phase and amplitude of femtosecond pulses and to study how nonlinear optical processes depended on those changes.

Those early efforts culminated in the development of computer-controlled programmable pulse shapers based on acousto-optic modulators and LCDs. For the first time, a user could control the phase and amplitude of 128 and, later, 640 spectral components of the laser. These programmable pulse shapers allowed researchers to create pulse replicas and eventually to compress the laser pulses. The subsequent commercialization of complete programmable pulse shapers by Fastlite and Biophotonic Solutions Inc. has made temporal pulse shaping available for scientific research since the mid-2000s, and this has resulted in several hundred scientific publications.

Modulators: A range of options

The key element in a programmable pulse shaper is the device that converts voltages into phase and amplitude modulation. Three main categories of modulators have been implemented: acousto-optic modulators[11] (including acousto-optic programmable dispersive filters[12]), LCDs[13] and deformable mirrors.[14] Table 1, which is divided into the three main types of modulators, summarizes the main characteristics of the various devices.

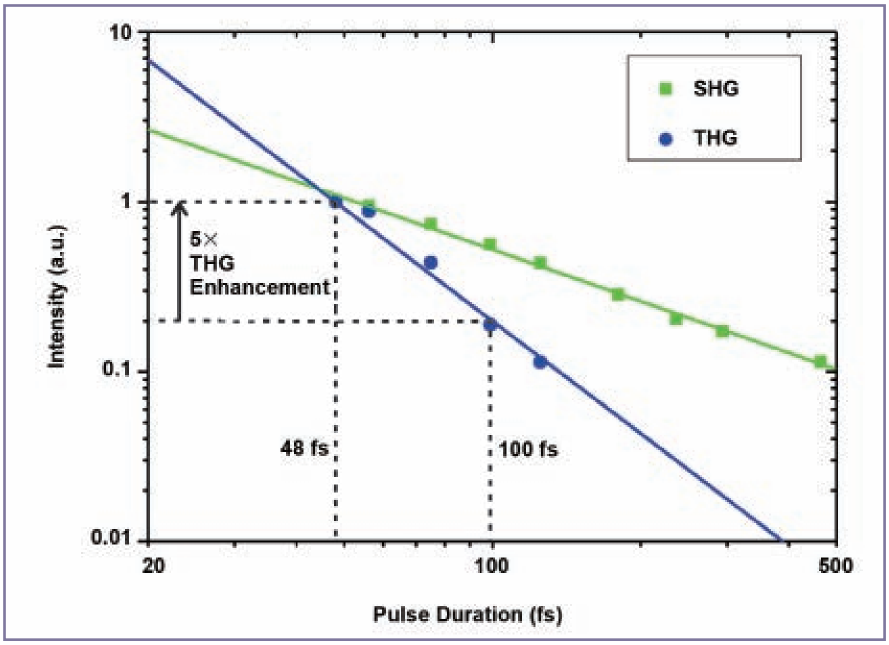
The modulators in Table 1 illustrate creativity in terms of finding methods to control ultrafast pulses. All of these devices are commercially available and have unique areas of strength. Acousto-



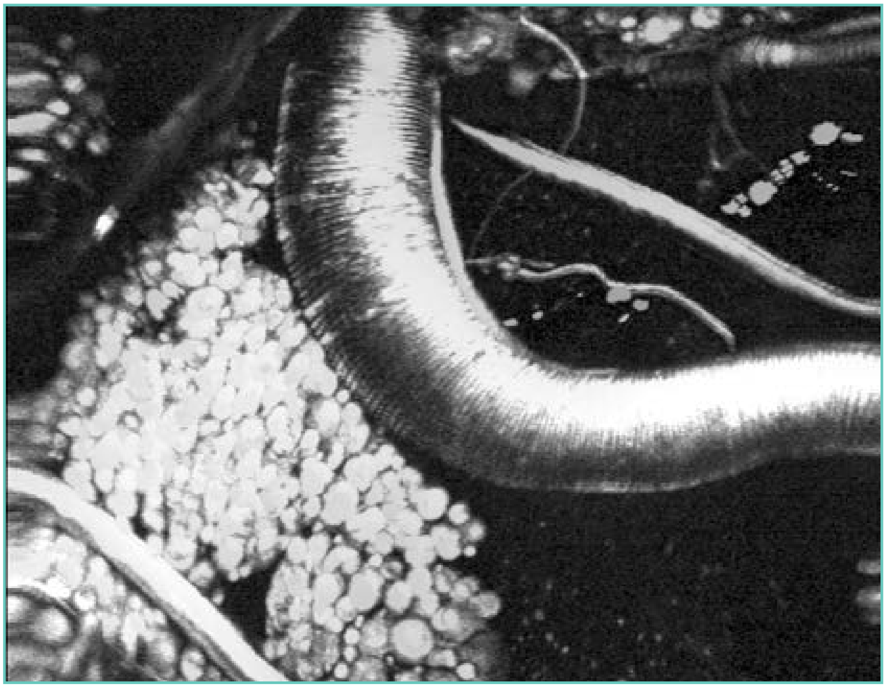
Shorter pulses produce much brighter images. P. Xi et al (2009). Two-photon imaging using adaptive phase compensated ultrashort laser pulses. *J Biomed Opt*, Vol. 14, Issue 1, p. 014002.
TPEF = two-photon excited fluorescence.

optic modulators offer the fastest modulation rates: When used in-line, as opposed to at the Fourier plane, acousto-optic pulse shapers are compact because no gratings or prism dispersion is required. Liquid crystals are some of the most developed modulators because of their use in displays.

Liquid crystal on silicon (LCOS) provides a platform that can be mass-manufactured, and the 800 × 600 unit from Hamamatsu has been industrialized and is already being used for spatial-shaping in industrial laser machining. Some dielectric coated units achieve 95 percent efficiency. Deformable mirrors offer a practical alternative with an extremely broad wavelength range; unfortunately, however, commercially available devices are small, have relatively few elements, and are quite expensive compared with other modulators. Since there is currently no technical reason why deformable mirrors have not been industrialized, it is possible that these devices will find their way into the stretcher or compressor of ultrafast laser amplifiers.



The signal in SHG (second-harmonic generation) microscopy increases as the inverse of pulse duration, while the THG (third-harmonic generation) signal increases as the inverse of pulse duration squared. Therefore, shorter pulses are important for THG microscopy. Adapted from B. Nie et al (2012). Multimodal microscopy with sub-30-fs Yb fiber laser oscillator. *Biomed Opt Exp*, Vol. 3, Issue 7, pp. 1750-1756.



THG imaging of an unstained living *Drosophila melanogaster* trachea, using a sub-50-fs Yb laser compressed at the focal plane of a 40× 1.1-NA microscope objective.

Adaptive pulse shaping

In the early days of pulse shaping, an independent, dedicated instrument such as an autocorrelator achieved pulse characterization. It was difficult to obtain timely and accurate information about the pulses, and to convert that information to a format that would control the pulse shaper. This led to a clever approach in which a nonlinear optical signal – such as second-harmonic generation, or SHG, which becomes more intense when the pulses get shorter – was used as a feedback signal. The closed loop included the laser, the shaper, the SHG detector and the computer running an optimization algorithm.[15,16] This approach led to automated pulse compression and was adopted by a number of research groups. It is a simple approach; however, it is time-consuming because it involves using random phases to find the one that produces the most intense SHG signal.

The discovery that a calibrated reference from a pulse shaper could be used to characterize a pulse revolutionized pulse characterization and adaptive pulse compression. The approach was first demonstrated in a publication discussing pH functional imaging.[17] The pulse measurement approach[18] was compared to the Wheatstone bridge used in electronics, whereby a known resistor is used to characterize an unknown resistor.

The first scientific discussion of the method, which is now known as multiphoton intrapulse interference phase scan (MIIPS), met with great resistance because the technique did not include a beamsplitter, an optical delay line or any other of the standard parts used in pulse characterization equipment.[19]

Once the accuracy of spectral phase measurements made with MIIPS was found to exceed the accuracy of those made with white-light interferometry,[20] it was accepted into the more recognized approaches for pulse characterization.

When the pulse shaper characterizes a pulse, pulse compression becomes highly simplified, because the shaper only needs to compensate for the measured phase distortions. This has led to the introduction of MIIPS-enabled instruments for automated pulse characterization and compression.

Interest in automated pulse characterization and compression using programmable pulse shapers has grown exponentially since the turn of the century. By now, hundreds of systems are being used in laboratories around the world. Industry, which looks for optimum value and reliability, is beginning to look beyond static pulse compression. Although achieving robust automated characterization and compression might seem to be the first priority, the 20 percent to 50 percent reduction in pulse duration may not justify a system's cost.

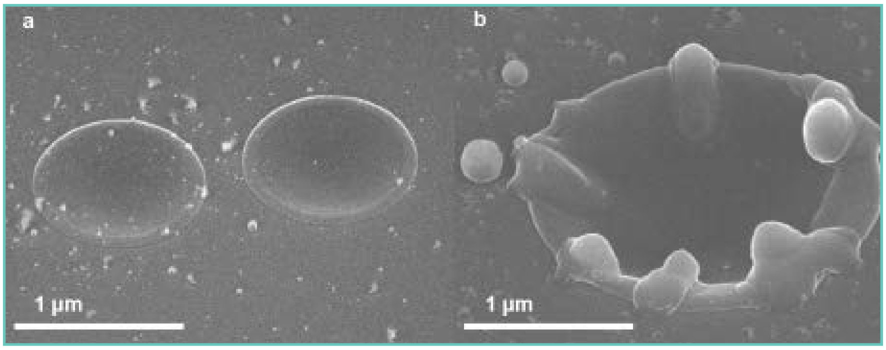
What justifies the cost, however, is that pulse shaper implementation may permit the use of much lower-cost laser sources. Pulse shapers have compressed 240-fs pulses down to sub-7 fs,[21] and 50-ps diode laser output down to 600 fs. These quantum leaps in ultrafast pulse compression promise to broaden the range of available sources for femtosecond laser applications, enabling prices and performance levels that could soon match picosecond laser sources.

Recently, interest has shifted toward on-the-fly optimization of ultrafast lasers through a feedback loop. Ultrafast laser output can be stabilized through real-time chirp adjustment[22] or via real-time chirp, third-order dispersion, and amplitude measurement and compensation.[23] The concept behind these approaches is to monitor the laser pulse characteristics throughout the day and to maintain optimization. Interest in adaptive optimization comes from implementations of ultrafast lasers that involve vibration and temperature changes. These adaptive systems are beginning to enter commercial laser sources and are expected to bring about a new level of reliability and ease of use to ultrafast laser systems.

Beyond pulse compression, temporal shaping has enabled applications such as significantly increased materials removal and standoff explosives detection. One can imagine that, in the future, temporal pulse shapers will be optimizing the output of the laser for particular applications. This may involve creating trains of pulses for materials processing – pulses with a fast rise time and slow decay for cutting transparent materials. Pulse shapers could also involve lasers whose output switches from femtosecond to picosecond pulse duration as the substrate being processed changes in composition from dielectric to metallic, as in modern computer chips. The technology that will enable the lasers of the future could depend on cost-effective and highly reliable temporal shaping based on programmable modulators.

Biomedical applications

Nonlinear optical microscopy has benefited greatly from the development of femtosecond lasers. Given that the signal may be due to two- or three-photon excited fluorescence, as well as second- or third-harmonic generation, shorter pulses yield greater signal. A tenfold



Silicon micromachining with transform-limited **(a)** and chirped **(b)** femtosecond laser pulses. T.C. Gunaratne et al (2009). Influence of the temporal shape of femtosecond pulses on silicon micromachining. *J Appl Phys*, Vol. 106, Issue 12, p. 123101.

reduction in pulse duration results in a tenfold increase in signal for second-order processes and a hundredfold improvement in signal for third-order processes.[24] Adaptive pulse compression has already achieved these gains. The gains in signal imply that bright images can be achieved from unstained samples at very low average laser energies.

Current femtosecond ophthalmic procedures use ~500-fs sources, based on early pulse duration studies, and these are not expected to embrace shorter pulses in the near future. However, advances in pulse shaping are expected to reduce the price of the laser sources and increase their reliability.

Future femtosecond refractive correction, however, may take advantage of shorter laser pulses. Current research efforts by L. Xu and colleagues[25] have shown the ability to modify the cornea, essentially using a sub-90-fs pulse to write a corrective prescription on it. Such procedures may last less than a year, but doctors may be able to fine-tune one's vision beyond 20/20 on a yearly basis.

Further into the future

Ultrahigh-finesse intracellular surgical procedures have already been demonstrated in laboratories, and femtosecond lasers are the only tools capable of delivering the required energy with nanometer precision. Many organs in our bodies are considered too small and fragile for corrective procedures. Femtosecond lasers, however, could soon permit routine polyp removal from vocal cords without affecting the voice. They could also correct hearing and treat many of the ducts that are important for keeping our eyes irrigated and our ears pressure-compensated.

The shorter the pulses used, the less the average energy. This reduces thermal load and allows imaging at greater depth. A "femto-scalpel" could enable manipulation of intracellular organelles, selective elimination of mitochondria, deletion of genes in chromosomes, introduction of genetic material, selective "rewiring" of living neurons, and even – someday – living brains.

Meet the authors

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