Ultrashort-Pulse Lasers

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Introduction

A particularly remarkable aspect of lasers is their ability to emit shorter flashes (pulses) of light than achievable with any other means. This ability has, over the years, advanced the observation and measurement of events from the nanosecond timescale down to the picosecond (10^{-12}), femtosecond (10^{-15}), and even attosecond (10^{-18}) timescales. To use such pulses has required the development of new methods for measuring and characterizing the pulses themselves on ultrafast timescales beyond the reach of electronics. These methods have, in turn, made it possible to study ultrafast phenomena in ways that produced completely new insights into the evolution of such phenomena in physics, chemistry, and biology [1]. As ultrashort-pulse laser technology has developed, its other characteristics such as the high peak power and ultrabroad bandwidth packed into a short pulse have also found important applications. The compression of even very modest amounts of pulse energy into femtosecond durations produces sufficiently high peak power for precision machining and micro-surgery without unwanted damage and for nondestructive nonlinear methods of microscopy that produce three-dimensional (3D) biological imaging with micrometer resolution. The ultrabroad bandwidths associated with femtosecond pulses have made possible 3D medical imaging via optical coherence tomography (OCT), simultaneous creation of many wavelength-multiplexed optical communication channels with only one source, and major advances in precision spectroscopy and optical clocks [2,3].

The Optical Society (OSA) played a major role in supporting the field, starting with its creation of the first International Conference on Picosecond Phenomena in 1978 (name changed in 1984 to Ultrafast Phenomena to reflect the emergence of femtosecond science and technology). Held every two years since then (for the 19th time in 2014, the year of this writing) with continuing OSA support, this successful conference has provided perhaps the greatest testament to the continuous technological development and widespread impact of the field with its 19-volume series of hardcover proceedings [4]. OSA journals became the primary source of publications on ultrafast optics and photonics. Multiple sessions on ultrafast optics and its applications every year at conferences like CLEO, QELS, IQEC, and OFC have been essential to advancing the technology and its applications to science and engineering.

Flashlamp-Pumped Picosecond Systems

Nd:glass Lasers

The era of ultrashort pulses began in earnest with the demonstrations in the mid-1960s, by DeMaria and co-workers at United Aircraft, of passive (self) mode-locking in a Nd:glass laser. Mode locking was achieved with a cell of absorbing dye inside the laser that was designed to bleach (saturate) sufficiently and rapidly enough to favor transmission of high intensity peaks over continuous emission and, therefore, the development of short pulses. The passive, saturable absorber technique, in various forms, remains the basis for ultrashort-pulse

generation today. The mode-locked Nd:glass laser pulses, too short to measure at first, were later verified to be on the order of 5–10 picoseconds in duration. For almost a decade, this laser system dominated and drove the development of ultrashort-pulse technology and its applications. For the second decade and a half, mode-locked dye lasers reigned and pushed pulse durations into the femtosecond domain. Finally, with the emergence of new techniques in the late 1980s, passive mode-locking of solid state lasers regained importance and led to the wide range of compact, robust, femtosecond laser systems we have today.

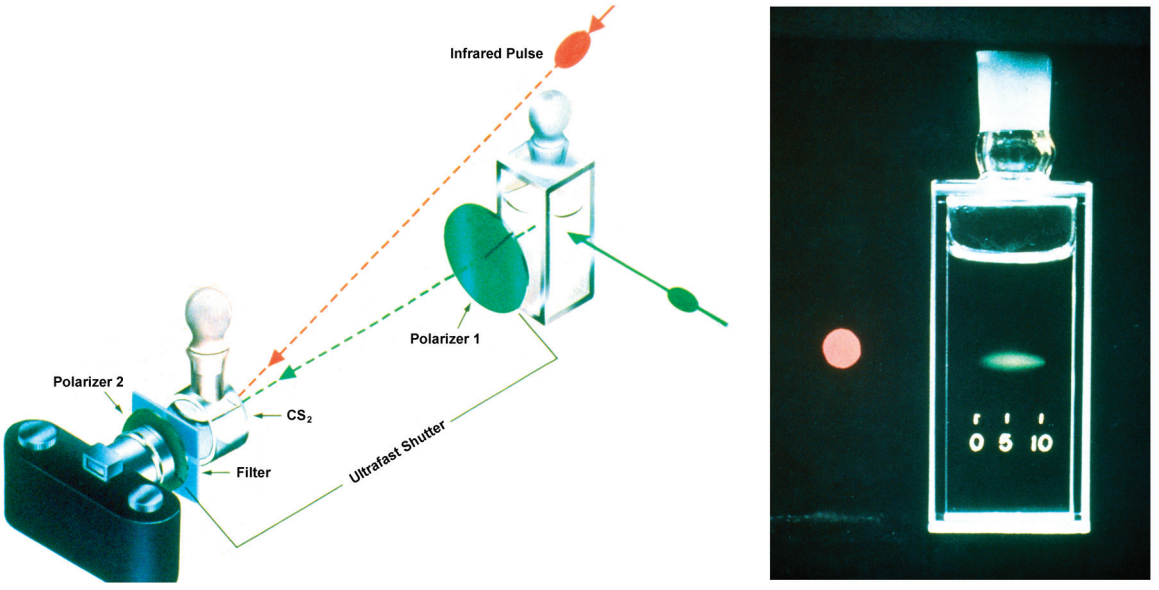
Ultrafast Measurement Techniques and Applications

Stimulated by mode-locked Nd:glass laser demonstrations, many of the ultrashort-pulse characterization, manipulation, and application methods still in use today were invented and developed in the 1960s [5]. Within a year of the invention of the passively mode-locked Nd:glass laser, several methods for pulse measurement with sub-picosecond resolution had been proposed and demonstrated. These techniques essentially use optical pulses to measure themselves. The laser output beam is split into two, one is delayed with respect to the other, and they are combined in a nonlinear crystal to generate second harmonic light (SHG). SHG is a maximum when the two pulses exactly overlap and decreases with delay in either direction. A plot of SHG versus delay yields the second-order autocorrelation function of the pulse intensity I(t). Fitting the observed intensity autocorrelation function to that expected for the pulses requires some assumptions about pulse shape, as this simple method is inherently insensitive to pulse asymmetry. Nevertheless, information about substructure and frequency chirp within the pulse can be deduced by comparing the assumed fit with that expected from the optical frequency spectrum. Methods for complete pulse characterization via frequency-resolved optical gating (FROG) were not developed until the early 1990s. The relatively slow repetition rate of flashlamp-pumped systems made the requirement of repetitive measurements at variable delay somewhat tedious at first. More rapid progress was permitted by the invention of a single-shot method in which two identical copies of a pulse are passed through a two-photon absorbing medium in counterpropagating fashion. The two-photon-induced fluorescence (TPF) intensity pattern, viewed from the side, provides another direct measure of the second-order autocorrelation function. Although widely used and valuable in early work, the TPF method subsequently gave way again to SHG-based methods with the advent of high repetition-rate continuous wave (CW) systems in the mid-1970s.

Most other present-day methods for manipulating pulses and applying them also developed rapidly during this period. It was shown that pairs of gratings can compensate for the chirp produced by linear dispersion in a laser. It followed that pulses could be shortened further by external self-phase modulation followed by a grating pair. Ultrafast responses in materials were observed by splitting a pulse beam into two, an excitation (pump) and a probe, and varying the time delay between them. Continuum generation, discovered by Alfano and Shapiro, made possible the simultaneous probing of changes over broad spectra. The ultrafast optical Kerr shutter, invented by Duguay and co-workers, was used as a picosecond camera to captured dramatic images of light pulses in flight (see Fig. 1 [6,7]) and to carry out the first demonstrations of 3D imaging via variable delay optical gating that later inspired the development by Jim Fujimoto of OCT for medical imaging (see Fig. 2). Other still-useful techniques such as up-conversion gating and transient grating spectroscopy were also demonstrated during this era. Scientific applications expanded to wide-ranging studies of nonlinear optics, picosecond interactions in liquids, and ultrafast processes in chemistry and biology [5].

Pulsed Dye Lasers

Known to have even more broadband potential than the Nd:glass laser, dye lasers were pursued shortly thereafter. The first experiments utilized picosecond pulses from frequency-doubled Nd:glass lasers to generate similarly short pulses from dye lasers. Passive mode-locking of the flashlamp-pumped Rhodamine 6G laser with a saturable dye soon followed. Within a few years the wavelength coverage of ultrashort-pulse dye lasers ranged from almost 400 nm to 1150 nm and amplified peak powers in the



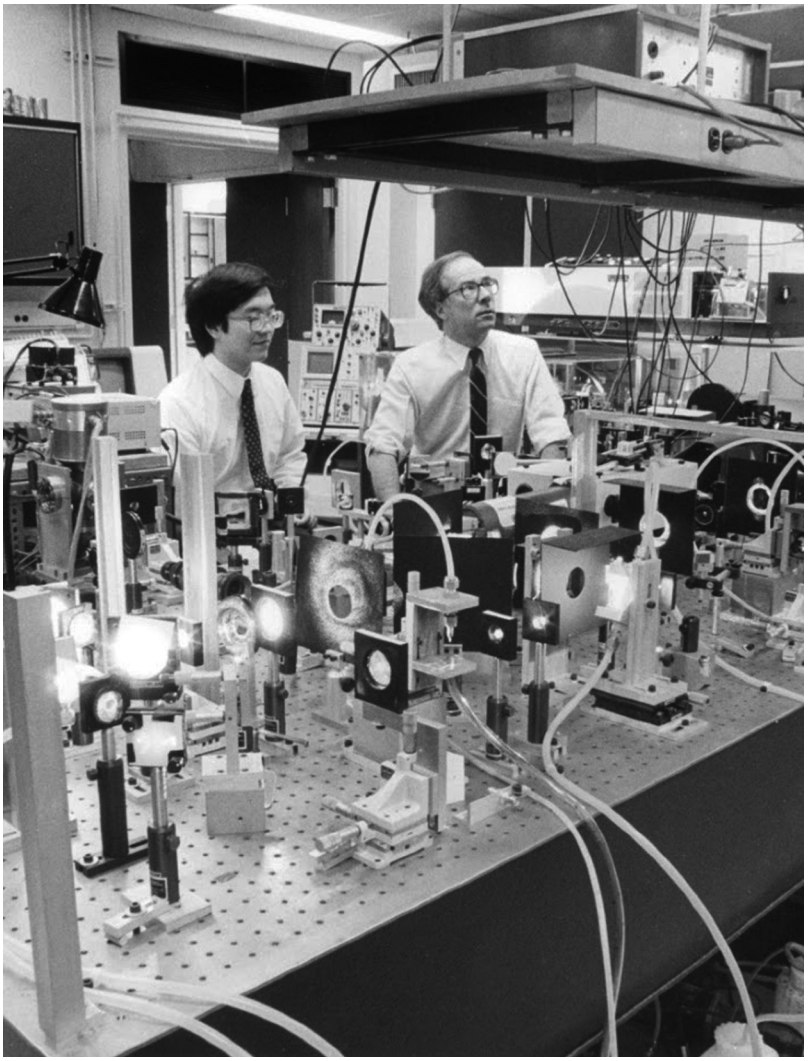
▲ **Fig. 1.** Light in flight. An optical Kerr effect shutter, operated by a picosecond infrared pulse, is used to capture the image of a picosecond pulse passing through a lightly scattering liquid. (a) experimental arrangement (b) the photo. Reprinted with permission from M. A. Duguay and J. W. Hansen, Appl. Phys. Lett. **15**, 192–194 ©1969, AIP Publishing LLC.

gigawatt range had been demonstrated, to a great extent by the Bradley group at Imperial College. As the pulse-forming dynamics of dye systems began to be studied in detail, the following question arose: How were such short pulses generated with saturable absorber dyes having much longer recovery times? In Nd:glass lasers, pulses were shown in studies to build up from noise, with the saturable absorber selecting the most intense pulse and determining the final duration by its recovery time. Dye-laser pulses were getting much shorter. This could happen, according to the insight of G. H. C. New, because, although bleaching the saturable absorber could only shape the leading edge of the pulse shorter than its recovery time, the trailing edge could be shaped by rapid saturation (depletion) of the dye gain medium. By 1975 all of these analyses were put into the subsequently very influential steady-state analytical descriptions, by Haus, of "fast" and "slow" saturable-absorber mode-locking that predicted shapes, durations, and stability [8,9,10].

Continuous-Wave Femtosecond Systems

CW Dye Lasers

Mode-locking of CW dye lasers offered a range of new possibilities for ultrashort-pulse generation. The continuous sources of high-repetition-rate pulses greatly facilitated measurement and the optimization of pulse characteristics via cavity alignment and saturable absorber concentration. With the first reports, in 1972, of passive mode-locking of a CW dye laser, pulses as short as 1.5 ps were reported. Within a year, the first pulses shorter than a picosecond had been produced by Shank and Ippen at Bell Labs (Fig. 3). The femtosecond era had begun. Pulses of 300 fs duration were soon achieved, and application of this new femtosecond capability to studies of ultrafast dynamics in physics, chemistry, and biology followed rapidly. Novel up-conversion pump-probe methods were developed, pulses of 500 fs in duration were amplified to peak powers of gigawatt intensities, and synchronized continuum generation made possible sub-picosecond time-resolved spectroscopy with greatly improved sensitivity and signal-to-noise ratio. Invention of the colliding-pulse

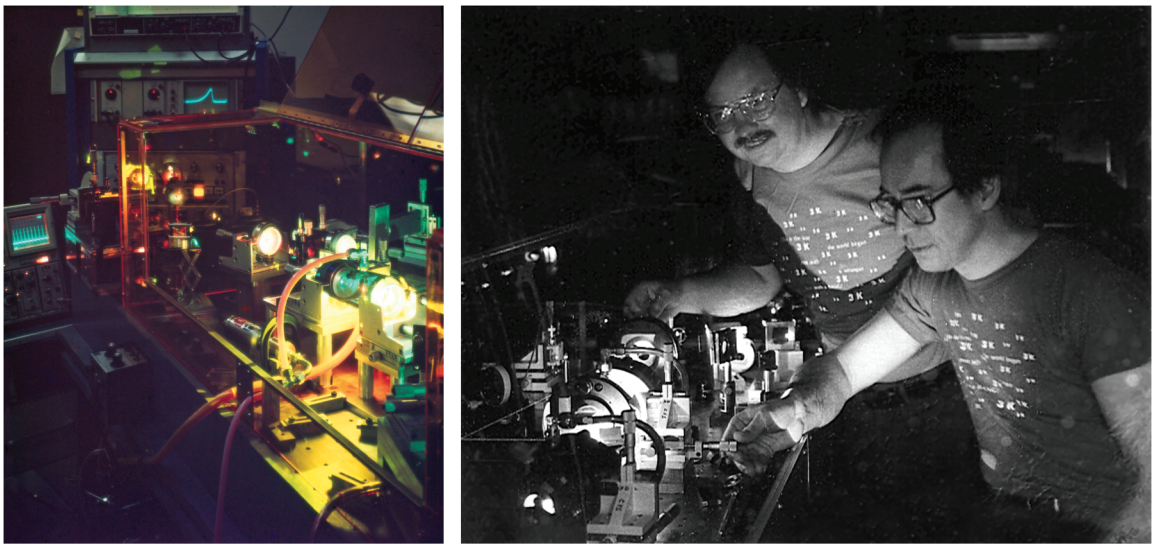


◀ **Fig. 2.** MIT Ultrafast Optics Lab 1985. Erich Ippen and student James Fujimoto view experiment achieving the first demonstration of optical ranging through skin, prelude to the development of Optical Coherence Tomography by James Fujimoto.

mode-locked (CPM) geometry in 1981 at Bell Labs reduced pulse durations to the 100-fs level and further improved stability. The interplay between self-phase modulation and internal dispersion was analyzed theoretically and optimized experimentally via prism pairs to reduce durations further to below 30 fs. Rapid progress was made by several groups, and with amplification and external compression, a record duration of 6 fs, a record that lasted more than a decade, was achieved. Amplified systems, pumped by either 10-Hz frequency-doubled Nd:YAG lasers (Fig. 4) or by kHz copper-vapor lasers, further extended the capability of femtosecond technology and its range of applications. The experiments leading to the 1999 Nobel Prize for chemistry [1] were achieved with this early femtosecond dye-laser technology.

Semiconductor Diode Lasers

Recognized as having gain response times very similar to those of dye lasers, semiconductor diode lasers also became the subject of mode-locking attempts. Shortly after active mode-locking was first demonstrated at MIT in 1978, passive mode-locking of a GaAlAs diode laser in an external cavity produced 5-ps pulse durations at a repetition rate of 850 MHz. Sub-picosecond pulses were later achieved at higher repetition rates, and integrated CPM geometry devices produced pulses as short as 640 fs at a repetition rate of 350 GHz. Impressive demonstrations of high-power, sub-picosecond pulses



(a) (b)

▲ **Fig. 3.** The first femtosecond laser, a Rhodamine 6G dye laser passively mode-locked by a DODCI saturable absorber dye. (a) Instruments record the pulse train and a sub-picosecond-resolution pump-probe trace of a molecular response. (b) Chuck Shank and Erich Ippen with their laser.

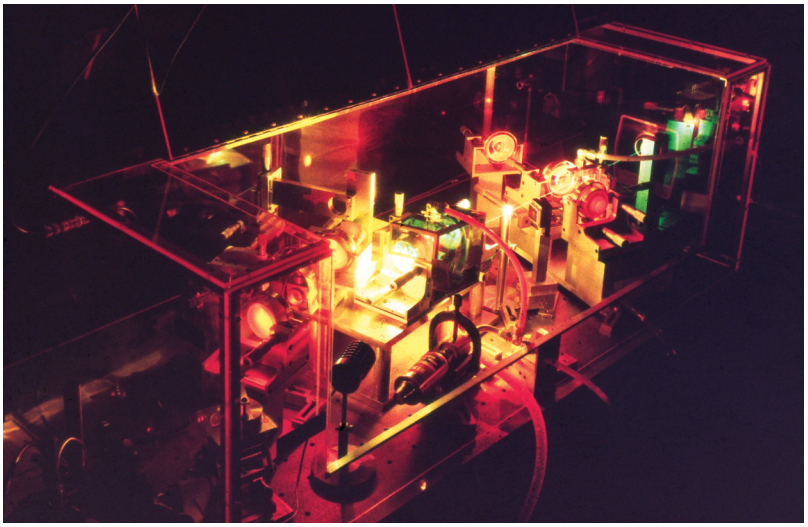
were achieved by Delfyett and co-workers with pulse compression and semiconductor optical amplification. Stable, transform-limited pulse generation with semiconductor diodes has, however, for the most part depended on external-cavity-controlled picosecond sources. Pump-probe investigations revealed that ultrafast nonequilibrium carrier dynamics in a semiconductor make the generation of pulses shorter than 1 ps problematic.

Color-Center Lasers

An important capability for early 1.5-μm-wavelength ultrafast research was provided by the CW color-center laser. First mode-locked by synchronous pumping, the KCl color center laser was thrust into further prominence by Mollenauer's demonstration at Bell Labs that it could produce femtosecond pulses by operating as a "soliton laser." This was achieved by coupling the laser output into an anomalously dispersive, soliton-shaping, optical fiber, the output of which was then coupled back into the laser. It was soon discovered, however, that soliton formation in the fiber was not necessary since this coupled-cavity approach also worked with normal-dispersion fiber. Experiments at MIT further revealed the underlying pulse-shortening mechanism to be the interference of each pulse with a copy of itself that had been self-phase modulated in the fiber. This method, dubbed additive-pulse mode locking (APM), was shown to be compatible with the Haus fast-absorber model. Recognized as a means of creating an "artificial" fast absorber out of reactive nonlinearity in a lossless dielectric, APM then stimulated the application of this technique to a variety of other lasers [11].

Fiber Lasers

Interest in fiber lasers developed rapidly after demonstrations at Southhampton of efficient optical amplification in low-loss fibers doped with rare earths. The key mechanism for ultrashort-pulse generation in fiber lasers—nonlinear polarization rotation—was also found to be describable by the fast-absorber model of Haus developed in the context of APM analysis. Earliest progress was made using Nd:fiber lasers, in both actively mode-locked and passively mode-locked configurations. By 1992 pulse durations as short as 38 fs had been generated at 1.06 μm in a Nd:fiber laser utilizing nonlinear polarization rotation and prism pairs for dispersion compensation. By the turn of the century, however,



◀ **Fig. 4.** High power, 3-stage, femtosecond dye laser amplifier pumped by frequency-doubled Nd:YAG laser at 10 Hz.

development of the much more efficient Yb:fiber laser led to considerably higher powers at 1 μm wavelengths, with similarly short pulses and more compact geometries. In the late 1980s the attention of researchers also turned to Er:fiber lasers for wavelengths being used for optical fiber communications and where fibers were anomalously dispersive, permitting soliton pulse shaping and shortening. Sub-picosecond pulses were first achieved, at NRL and at Southhampton in figure-eight geometries that used a nonlinear loop mirror for intensity modulation and pulse stabilization, and then, at MIT and Southhampton, in the ring geometry stabilized by nonlinear polarization rotation that achieved common usage. The MIT stretched-pulse laser achieved shorter pulses and higher pulse energies and was soon commercialized. Although not geared to the high-power applications of Yb:fiber lasers, Er:fiber lasers continue to be pursued for silicon photonics, fiber-based communications, and a variety of eye-safe applications.

Free-space Solid-state Lasers

The discovery of APM and the prospect it offered for CW mode-locked solid-state lasers led to its application to Nd:YAG, Nd:YLF, and Ti:sapphire systems. To permit amplification to high power, Strickland and Mourou in 1985 demonstrated the chirped-pulse amplification (CPA) scheme that would ultimately open the door to attosecond and petawatt optical physics. With the discovery of the Kerr-lens mode-locked (KLM) Ti:sapphire laser in 1991 by the Sibbett group in St. Andrews, KLM became the dominant ultrashort-pulse generation mechanism in free-space solid-state lasers. Femtosecond science and technology entered a new era, one with a wider variety of femtosecond-laser media, shorter pulses, extreme powers, ultrabroad bandwidths, and, quite dramatically, the convergence of ultrashort-pulse lasers with ultranarrow-linewidth lasers, precision spectroscopy, and optical clocks. This modern era is the subject of a following article.

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