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PROGRESS IN OPTICS

VOLUME

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Amsterdam · Boston · Heidelberg · London · New York · Oxford · Paris
San Diego · San Francisco · Singapore · Sydney · Tokyo

Elsevier
Radarweg 29, PO Box 211, 1000 AE Amsterdam, The Netherlands
Linacre House, Jordan Hill, Oxford OX2 8DP, UK

First edition 2007

Library of Congress Catalog Card number: 61-19297

ISBN: 978-0-444-53023-3
ISSN: 0079-6638

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07 08 09 10 11 10 9 8 7 6 5 4 3 2 1

Printed and bound in The Netherlands

Dedication

This volume is dedicated to the memory of two members of the Editorial Advisory Board of *Progress in Optics*

Professor Lorenzo Narducci
and
Professor Herbert Walther

two distinguished scientists and friends of many of our readers. Their recent deaths represent a profound loss to the whole scientific community.

Preface

It is a great pleasure to present to our readers the fiftieth volume of *Progress in Optics*, a series which commenced publication in 1961, close to the time when the laser was invented. Since then optics has been evolving very rapidly. The nearly 300 articles that appeared in these volumes faithfully reflect most of the exciting developments in optics and in related subjects that have taken place since that time.

To celebrate this milestone all the articles contained in the present volume are devoted to historical developments of optics.

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August 2007

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Chapter 1

From millisecond to attosecond laser pulses

by

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ISSN: 0079-6638 DOI: 10.1016/S0079-6638(07)50001-6

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§ 1. From millisecond to nanosecond pulses

Maiman [1960] achieved the first operating laser, utilizing a ruby crystal pumped by a xenon flash lamp. The light flash lasted about one millisecond and the laser output fluctuated during this time interval. The population inversion in the fluorescent levels of the chromium ion changed by the competition between the pumping mechanism and the stimulated emission processes into various laser modes. This led to relaxation-type oscillations and fluctuations in the laser output.

Hellwarth [1961] achieved Q-switched pulses of less than one microsecond duration within the first year of ruby laser operations. The quality factor Q of the laser cavity was kept low by the use of a polarization-switching cell with nitrobenzene, subjected to an electric field causing birefringence by the Kerr effect. A large population inversion in the ruby was established, as the light with orthogonal polarization was switched out of the laser cavity with a Nicol prism. When the electric field on the Kerr cell was suddenly switched off, a powerful laser pulse was created by the high gain from the large inverted population. Reproducible pulses with a duration between ten and one hundred nanoseconds were obtained. In this early period Q-switching was also obtained by mounting a mirror or a cubic totally reflecting glass prism on a turbine-driven dentist drill. The laser oscillation could only build up when the rotating mirror was nearly parallel to the fixed mirror of the laser cavity. Clearly this mechanical method had no mode control and became rapidly obsolete, although it survived longer for the Q-switching of high-power CO_2 lasers operating near 10 μm wavelength.

This cursory overview is intended for non-specialists who wish to become acquainted with the remarkable developments in time-resolved optical techniques. More comprehensive reviews may be found in the references.

The next section outlines the evolution from the nanosecond to the femtosecond regime. It is based on familiar nonlinear optical processes, including saturable absorption, intensity-dependent index of refraction, self-focusing and self-phase modulation. The transition to the attosecond regime is described in the third, final section. It requires "extreme" nonlinear processes, including tunneling and the creation of quasi-free electrons, which enable a transition from the visible to the (soft) X-ray spectral region.

§ 2. From nanosecond to femtosecond pulses

This section is based on an earlier brief overview (Bloembergen [1999]) and on a comprehensive review by Brabec and Krausz [2000]. Mode-locking of a large number of longitudinal modes all activated by the gain profile of the laser medium can lead to one short pulse traveling back and forth in the laser cavity of length l. The frequency spacing between the modes is $c/2l$ and the output is a train of pulses, separated by the time interval $2l/c$. Active mode-locking was first achieved for a helium–neon laser by Hargrove, Fork and Pollack [1964]. They acoustically modulated the index of refraction at the frequency $c/2l$. Since the gain–bandwidth profile of the gas-laser transition is rather small, the pulse duration remained much longer than ten nanoseconds.

Passive mode-locking of a ruby laser was first demonstrated by Mocker and Collins [1965]. They used a saturable absorber in the form of a liquid film or jet with a bleachable dye near one of the mirrors in the laser cavity. The generation of a short pulse output may be qualitatively understood as follows. Initially, spontaneous emission processes are amplified and lead to a stochastic light intensity. A peak in this output is subsequently amplified the most, as it causes more initial bleaching of the saturable absorber. Thus, intervals of higher intensity are attenuated less on passage through the bleachable dye film. After many round trip passages in the laser cavity the energy transfer of the stimulated emission processes is concentrated in a shorter time interval, which is eventually limited by the inverse of the gain–bandwidth product of the laser. For the ruby laser this product was not large enough to break the nanosecond barrier.

Pulses shorter than a nanosecond, marking the entrance into the picosecond domain, were first obtained by De Maria, Stetser and Heinan [1966]. They used a neodymium glass laser. This material as well as neodymium–yttrium–aluminum garnet (Nd-YAG) have sufficient gain–bandwidth product to permit the generation of laser pulses of 10 ps duration. These lasers, operating near 1.06 μm wavelength, were widely used for experiments in time-resolved spectroscopy. Other frequencies could be obtained by harmonic generation, stimulated Raman scattering and self-phase modulation. Shapiro [1977] has edited an early review dedicated to picosecond pulse generation.

The first entry into the sub-picosecond or femtosecond regime was accomplished by Shank and Ippen [1974]. They used a broad gain dye-laser medium in combination with a saturable dye-absorber film. An analysis by New [1972] had shown that it is possible to obtain pulses shorter than the characteristic relaxation times of the dye media, since the pulse duration is determined by the time that the saturable gain exceeds the saturable absorption. The use of a ring

dye-laser system with counter-propagating laser pulses, which cross each other in the passage through a saturable dye film, only a few microns thick, led to even shorter pulses. A limit is then set by the dispersion in the group velocity which causes different Fourier components of the short pulse to propagate at different speeds. This produces chirping and stretching of a short pulse. The chirping produced by self-phase modulation is often more important. As the intensity rises during the leading edge, the index of refraction rises, and the light frequency is shifted toward the red. A blue shift occurs at the trailing edge of the pulse. Chirping can be compensated by introducing negative group-velocity dispersion. This may be accomplished by prism or grating configurations which increase the optical path length for red-shifted light relative to that for blue-shifted light. Fork, Britto Cruz, Becker and Shank [1987] reported pulses of 6 fs duration by a prism configuration in the laser cavity. The linearly polarized light beams were incident near Brewster's angle to avoid reflection losses at the prism surfaces. These femtosecond dye laser systems required frequent adjustments, and alignment of the components was critical. Ippen [1994] has reviewed the theory and experiments of passive mode locking of laser pulses.

A true revolution in femtosecond generation occurred when Spence, Kaen and Sibbett [1991] discovered that a titanium-aluminum oxide (Ti-sapphire) laser crystal would yield very short pulses without the use of a saturable absorber. Self-focusing of the laser beam in the Ti-sapphire crystal occurs because of its intensity-dependent index of refraction. In combination with a suitably placed aperture, the self-focusing can cause a larger fraction of the pulse energy to pass through the aperture at higher intensity. Thus, less absorption and more gain occurs for more intense parts of the stochastic amplified spontaneous emission. This so-called Kerr-lens self-focusing is a purely dispersive effect. It is independent of material relaxation times and can be as fast as the inverse of the frequency detuning from the absorption edge of the material. Compensation of group-velocity dispersion in the laser crystal and the chirping by self-phase modulation is again essential. Besides prism and grating configurations, negative group-velocity dispersion may also be obtained from chirped mirrors. These are layered dielectric films in which red-shifted components penetrate deeper than blue-shifted components, as the thickness of alternating films causing Bragg reflection increases with increasing depth. The compensation of self-phase modulation by negative group-velocity dispersion leads to the formation of soliton-like optical pulses.

Nonlinear effects are essential not only in the generation of picosecond and femtosecond pulses, but also in their measurement and evaluation. The short pulse is split into two pulses and a variable time delay is introduced between the first (or pump) pulse and the second (or probe) pulse. The two pulses are recombined

in a thin nonlinear crystal and the second harmonic generated in the crystal by the combination of the two pulses is observed as a function of the time delay. Armstrong [1967] first used this technique for picosecond pulses. Note that one femtosecond corresponds to a differential of 0.3 μm in the path lengths of the two pulses. The intensity of the second harmonic as a function of path length differential yields the autocorrelation of the intensity of the laser pulse. For a complete characterization of the pulse the temporal behavior of the phase of the light field must also be determined. This information may be obtained by analyzing the temporal behavior of individual Fourier components in the generated second harmonic signal. It is spectrally analyzed as a function of time delay. Trebino and Kane [1993] introduced this technique, which they called Frequency Resolved Optical Gating or FROG. Other variations were subsequently introduced but they are all based on a comparison of spectrally resolved components of the pulse and its delayed or advanced replica.

Conversely, pulses may be generated with prescribed amplitude and phase variation. A femtosecond pulse is spectrally resolved by a grating and different Fourier components may be reflected by different segments of piezoelectric crystals. Voltages applied to the segments will produce different path lengths or phases to individual Fourier components on reflection. These components may be recombined by the same grating to produce a modified pulse with a different temporal phase variation. The different Fourier components may, of course, also be attenuated by adjustable factors. These changes may also be induced by passage through a liquid crystal array with a configuration of electrodes. The latter technique has been used by Shverdin, Walker, Yavuz, Yin and Harris [2005] to generate a half-cycle optical wave form of about 1.5 fs duration. A 5 fs pulse of a Ti-sapphire laser is used to generate several orders of anti-Stokes and Stokes components of vibrational and rotational Raman transitions in hydrogen gas (H_2 or D_2). The components cover a frequency range of more than two octaves. By adjusting the amplitudes and phases of these components before recombining them it was possible to obtain constructive interference over one half cycle, and nearly complete destructive interference outside that time interval.

Femtosecond pump-probe techniques have led to many applications in time-resolved spectroscopy in chemistry, biology and solid state physics, but their discussion falls outside the scope of this review. Zewail [2000] received the 1999 Nobel Prize for chemistry for this work on femtochemistry, the time-resolved spectroscopy of a large variety of chemical reactions. Lobastov, Shrinivas and Zewail [2005] have recently extended femtosecond time resolution to the field of electron diffraction. They call it 4D ultrafast electron microscopy. A weak femtosecond "probe" pulse is used to liberate on the average about one photoelectron

from the cathode of a standard electron microscope with a de Broglie wavelength of 0.335 nm at 120 keV energy. Distortion of the electron orbits due to space-charge effects by a Coulomb repulsion between electron pairs is thus avoided. A pulse train of ten million pulses per second builds up the diffraction of a target that has been excited by a femtosecond "pump" pulse at an adjustable earlier time. Thus, the variation in the diffraction pattern with sub-nanometer spatial resolution and 10 fs temporal resolution can be observed.

The output of a Kerr-lens mode-locked tabletop Ti-sapphire laser with a crystal volume of a few cc typically produces a train of 10 fs pulses with about one nanojoule per pulse with a repetition rate of 10 MHz. Such a pulse train can be amplified by a factor of 10^3 or 10^4 by the chirped pulse amplification (CPA) technique described by Mourou, Barty and Perry [1998]. The pulse is first deliberately lengthened by a factor of 10^3 to 10^4 by an antiparallel grating configuration before being amplified. This is necessary to avoid damage by light-induced dielectric breakdown in the amplifying medium. The amplified pulse is then re-compressed by a matching configuration of grating pairs. The CPA technique can generate a train of 10 fs pulses with a pulse energy of one microjoule, or even higher. If such a pulse is focused by a microscope objective to an area of 10^{-8} cm^2, the resulting power flux density would be 10^{16} W/cm^2, with a corresponding light-field amplitude of 10^9 V/cm. This is approximately equal to the Coulomb field at the Bohr orbit in the ground state of the hydrogen atom, or the field responsible for the binding of a valence electron in a molecule. It is physically obvious that any material subjected to such a field is immediately transformed to a fully ionized plasma. If the light intensity is lowered by one or two orders of magnitude, in the range of 10^{14} to 10^{15} W/cm^2, there is still a probability for ionization by tunneling rather than by multi-photon ionization. The first experiment on two-photon induced photoelectric emission from an alkali-metal cathode was carried out by Teich, Schoer and Wolga [1964] with a ruby laser. The effect of thermionic emission due to heating during the long laser pulse had to be carefully eliminated. Bechtel, Smith and Bloembergen [1975] demonstrated four-photon emission from tungsten by a picosecond Nd-glass laser. The ionization of Kr atoms by an eleven-photon process at 1.06 μm was reported by Mainfray [1978]. The ionization rate and the formation of Kr^+ ions increased as the eleventh power of the laser intensity. This probably represents the limit of high-order nonlinear optical perturbation theory. Keldysh [1965] had already discussed the transition from multi-photon to tunneling ionization. He introduced a dimensionless parameter, the square root of the ratio of the ionization potential I_p to the quiver energy or ponderomotive energy $U_p = e^2E^2/2m\omega^2$ of a free electron oscillating in a laser field of amplitude E and circular frequency ω. For $U_p > I_p$ tunneling predominates. This occurs when a

femtosecond train of pulses is focused on a stream of noble gas atoms emanating from a nozzle at intensities in the range of 10^{14} to 10^{15} W/cm^2. The presence of quasi-free electrons created by tunneling ionization leads to new phenomena, including high-harmonic generation in the soft X-ray regime. The creation of these quasi-free electrons has made it possible to explore the attosecond regime and the measurement of time intervals shorter than one optical cycle.

§ 3. The attosecond regime

Corkum [1993] introduced the recollision model which describes the phenomena occurring in a fraction of an optical cycle. A valence electron tunnels out of its atomic or molecular orbital preferentially when the linearly polarized femtosecond optical field is near its maximum in either direction. The liberated electron is accelerated in the next quarter cycle. When the direction of the laser field reverses at the first zero crossing, the electron is decelerated and reverses direction during the next half cycle and returns to the ion it had left behind. It reaches this ion approximately at the second zero crossing after its liberation. Detailed classical calculations of the equation of motion of the free electron in the optical cycle for a range of moments of its creation by tunneling show that the maximum kinetic energy is $3.2U_p$, where U_p is the ponderomotive energy introduced previously. The electron has a probability to fall back into the valence orbital it had left behind with the emission of a photon with a maximum energy $I_p + 3.2U_p$. If it does not recombine, it may recollide again after half of an optical cycle. If the femtosecond pulse is relatively long and contains many optical cycles, the recollisions spaced by one half optical cycle lead to a time series of photon pulses. In the frequency domain this corresponds to a series of odd harmonics, spaced by 2ω. Such high harmonics, often in the range of ten to a few hundred times the optical frequency, have been observed by many investigators. Kapteyn, Murnane and Christov [2005] have reviewed this generation of extreme UV and soft X-rays in targets which usually consist of a jet of noble gas atoms, into which a train of femtosecond optical pulses are focused. The harmonics are mostly emitted in a narrow cone parallel to the direction of the optical pulse. Phase matching between the harmonics and the infrared pulse is possible because the generated plasma lowers the optical index of refraction. The energy distribution over the harmonic spectrum depends sensitively on the intensity and phase variation in the incident laser pulse. The generation of particular harmonics may be optimized by controlling the phase variation as described previously (Kapteyn, Murnane and Christov [2005]).