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Frontier of Femtosecond and Picosecond Optical Measuring Techniques[a]

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INTRODUCTION

Over the past two decades, we have seen dramatic advances in the generation of ultrafast laser pulses.[1-12] It is now commonplace to produce picosecond (10^{-12} sec) pulses. New developments have extended this technology into the femtosecond (10^{-15} sec) time region. Soon pulses consisting of 1 cycle will be produced (i.e., 2 fs at 600 nm). These ultrafast pulses permit novel investigations to study phenomena in many fields. The design of sophisticated techniques based on these laser pulses has given rise to instruments with extremely high temporal resolution. Ultrafast laser technology offers the possibility of studying and discovering key processes unresolved in the past. A new era of time-resolved spectroscopy has emerged, with pulses so fast that one can now study the nonequilibrium states of matter and test quantum and light models.

Today, ultrafast laser technology is one of the most active areas of science because it can be used in a diverse number of fields: solid-state physics, biology, and chemistry. TABLE I lists the primary relaxation times in condensed matter. This report highlights my presentation on the fs and ps optical measuring techniques available today.

MEASURING TECHNIQUES

It has been over twenty years since picosecond laser pulses were first produced in glass lasers (DeMaria and co-workers) by mode locking.[6] During this time, the solid-state laser has become the workhorse. However, it is slowly being replaced by dye and Yag lasers. A new type of light source on the horizon was discovered in 1970 by Alfano and Shapiro—the ultrafast supercontinuum laser that spans from UV to IR. TABLE 2 lists the currently available ultrafast laser sources.[1-5]

Currently available electronic equipment does not have the time response required to directly measure events that take place in picoseconds or femtoseconds. The fastest oscilloscope, for example, equipped with a sampling head has a resolution of 100 psec. Indirect methods, however, are available for time measurements. The constant speed

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TABLE 1. Primary Relaxation Time Scales in Physics, Chemistry, and Biology

	Time (sec)	
Photosynthesis	10^{-8}	Acoustic phonon
	10^{-9}	Excitons, Vision
Semiconductors		Spin, Magnon
Dyes	10^{-10}	Rotational
nonradiative		Polaritons
singlet-triplet	10^{-11}	
Charge transfer complex		Energy and temperature relaxation of carriers
Photodissociation	10^{-12}	Vibrational, optical phonon
Photoionization		Momentum relaxation of carriers
Solvent caging, H-bonding	10^{-13}	Electron-scattering times electron-hole plasma
Electron-proton transfer	10^{-14}	Vision, libration, hemoglobin

of light traveling over a known distance provides the basis for the time-measuring mechanism on a picosecond and femtosecond time scale. It takes light 10 fs to travel 3μ in the air. A length of 3μ can be easily and accurately measured with a stepping motor.

Two-Photon Fluorescence and Second Harmonic Methods

The two methods that will be described and that are most often used for pulse width measurements are two-photon fluorescence (TPF) and second harmonic generation (SHG) spatial width.

The TPF method was described first by Giordmaine, Rentzepis, and Shapiro.[8] In this method, a beam is split into two beams and directed into a cell that contains a dye that does not absorb at the laser wavelength. The dye molecules, however, may absorb two photons simultaneously and be raised to an excited state. Then, fluorescence takes place in the normal way. The cross section for two-photon absorption is small, so TPF is not ordinarily observed. The probability of a two-photon transition is proportional to the square of the incident light intensity. The size of the TPF is related to the pulse

TABLE 2. Ultrafast Lasers Available

Oscillator	Wavelength (nm)	Pulse Duration
Ruby	694.3	30 ps
Yag	1,064	30 ps
Silicate glass	1,060	8 ps
Phosphate glass	1,054	6 ps
Dye	tunable (syn or flashlamp)	5–10 ps
Dye	610 (passive)	100 fs
Dye + Pulse compression	tunable	300 fs
Dye + Pulse compression	620	27–90 fs
Dye + 4 Prisms	625	27 fs
Dye + SPM[a]	continuum	80 fs
Dye + SPM[a] + Pulse compression	620	8 fs

[a]SPM = self-phase modulation (supercontinuum laser source).

width through the autocorrelation function, $G^{(2)}(\tau)$:

$$I_{2f} = C[1 + 2G^{(2)}(\tau)],$$

where

$$G^{(2)}(\tau) = \int_0^{t_0} I(t)I(t+\tau)dt \Big/ \int_0^{t_0} I^2(t)dt.$$

From the autocorrelation function and the pulse shape, the pulse duration can be estimated. The rapidly varying interference times average out using a course stepping motor of 2μ steps. TABLE 3 gives the real values of the pulse width for three different pulse shapes. Typical dye solutions used are Rh6G in methanol for 1060 nm, and coumarin in methanol or BBOT in chlorohexane for 530 nm.

The first measurements of picosecond duration were made using second harmonic generation. In the SHG method, two pulses enter the nonlinear crystal of KDP in either a collinear or noncollinear geometry. One beam is time-delayed using a stepping motor with accuracy of $\frac{1}{2}\mu$. The correlated signal, $G^2(t)$, at harmonic frequency, 2ω, is

TABLE 3. Relation between Pulse Shape and Pulse Duration and Autocorrelation Durations[a]

Pulse	$\gamma = \Delta t_G/\Delta t$	$\Delta t \Delta f$
$\exp(-4\ln 2\, t^2/\Delta t)$	1.44	0.441
$\mathrm{sech}^2\lvert(1.76t)/\Delta t\rvert$	1.55	0.315
$\exp\lvert-1n2t/\Delta t\rvert$	2	0.11

[a] Δt_G and Δt are (FWHM) of the pulse width as measured by TPF or SHG and the true pulse width, respectively; γ is the form factor.

highly directional and is measured as a function of time delay. TABLE 3 relates the pulse duration to correlation durations.

The time resolution of the SHG and TPF methods is electronic in origin; however, experimental problems limit these methods to about 5 to 10 fs time resolution.

The Streak Camera

The technique most widely used for luminescence measurements in the picosecond range incorporates a streak camera. Photoelectrons emitted by light striking the photocathode are deflected by an applied voltage ramp that causes the electrons to be transversely streaked across a phosphorescent screen. Photoelectrons released at different times from the photocathode strike the phosphorescent screen at different positions, thus causing a track with a spatial intensity profile directly proportional to the incident temporal intensity profile of the fluorescence. The phosphorescent track may be analyzed by sampling by a video system. The ultrashort pulse from the laser is divided by a beam-splitter. Part of the beam is sent to a photodiode that triggers the streak. The other pulse excites the sample. The pump pulse has to be delayed for a few nanoseconds before the streak camera is ready. To produce that long delay, the pump

pulse travels through a white cell. Before the pump strikes the sample, a wedge reflects part of the beam directly into the slit of the streak camera. This prepulse is used for the calibration of the zero time. The fluorescence from the sample is collected and passes through filters to eliminate the excitation pulse. It is then focused onto the slit of the streak camera. A modification of this arrangement allows one to measure the temporal profiles of the fluorescence at both polarizations simultaneously.

The temporal resolution of the streak camera has four components: the range of initial kinetic energies of the electrons emitted from the photocathode; the width of the slit; the scanning velocity; and various edge effects, such as electric-field fringing, optical diffraction, and space-charge effects (Alcock).[11] The temporal resolution of the streak camera is about 1 psec with a jitter of 50 psec. Recent improvements in tube design have reduced the time resolution to 200 fs.

The Optical Kerr Gate

The optical Kerr gate (Duguay and Hansen,[7] and Shimizu and Stoicheff[12]) consists basically of a Kerr active liquid, such as CS_2, situated between two polarizers. Under the intense electric field associated with the laser pulse ($\sim 10^5$ V/cm), the molecules of the Kerr active liquid experience an induced birefringence. The light passing through the liquid becomes generally elliptically polarized and passes partially through the second polarizer. Light can only pass through the gate while it coincides with the intense laser pulse that opens the gate. Thus, the intense laser pulse can be used to carve out successive portions of the temporal profile of the emitted luminescence, $I_L(t)$, to yield the decay time, τ_D.

The resolution of the optical Kerr gate depends on the duration of the gate pulse and the reorientation time of the molecules of the active medium. For CS_2, one of the fastest Kerr-active media available, the reorientation time is 2 ps. The pulse duration of the glass laser (6–10 psec) has in the past been the limiting factor of the resolution of the Kerr gate. The subpicosecond dye laser requires even faster Kerr-active media (polymers, i.e., PDA) based on the electronic distortion rather than the induced orientation of the molecules. Such a gate can have a resolution of 10^{-14} sec.

The theory of the operation of the Kerr gate shows how its time resolution is limited by both the pulse width, T_G, and the lifetime of the birefringence, T_B, of Kerr-active media (T_B is usually limited to over 2 psec by the slow reorientation time of molecules in the liquid). When the lifetime, T_B, of the birefringence ($\sim$2 psec for CS_2) is much shorter than the pulse duration, T_G ($\sim$6 psec for solid-state lasers), the birefringence, $\Delta n(t)$, is given by

$$\Delta n(t) = \Delta n_{\parallel} - \Delta n_{\perp} = \frac{1}{2} n_2 E_G^2(t) = \frac{1}{2} n_2 I_G(t),$$

where the nonlinear coefficient, n_2, of the index of refraction is about 2×10^{-11} esu for CS_2. The corresponding phase retardation, $\Delta\phi(t)$, in a cell of length, z, is given by

$$\Delta\phi(t) = \omega_L \Delta n(t)(z/c),$$

where ω_L is the frequency of the luminescence and c is the speed of light. These equations apply only when the duration of the pulse, T_G, is much longer than the

relaxation time of the Kerr medium, T_B. When this condition is not fulfilled, the more general expression must be used:

$$\Delta n(t) = \frac{1}{2}\frac{n_2}{T_B}\int_{-\infty}^{t} I_G(t') \exp\left[\frac{t-t'}{T_B}\right]dt'.$$

The phase retardation causes the linear polarization of the luminescence to become elliptically polarized and to pass through the second polarizer. The intensity of the transmitted signal with phase retardation, $\Delta\phi$, is proportional to $\sin^2(\Delta\phi/2)$, that is,

$$I_s(\tau, t) \alpha I_L(t) \sin^2\left[\frac{1}{2}\Delta\phi(t-\tau)\right],$$

which gives the instantaneous signal. The response, however, of the photomultiplier is much slower than the duration of the laser pulses. The detection system records the total energy transmitted:

$$I_s(\tau)\alpha \int_{-\infty}^{\infty} I_s(t, \tau)dt = \int_{-\infty}^{\infty} I_L(t) \sin^2\left[\frac{1}{2}\Delta\phi(t-\tau)\right]dt.$$

When the induced retardation is small, $\sin\Delta\phi \approx \Delta\phi$; I_s is simplified as follows for $T_G > T_B$:

$$I_s(\tau)\alpha \int_{-\infty}^{\infty} I_L(t) I_G^2(t-\tau)\, dt.$$

The ultimate time resolution in the fs regime depends on finding a material with a large and electronic fast n_2.

The Parametric Upconversion Gate

The process of parametric upconversion has been used as a mechanism in ultrafast optical shutters (Mahr, Topp).[9] In this technique, the luminescence, $I_L(t)$, from the sample is collected, collimated, and combined with part of the excitation pulse in a noncentrosymmetric crystal (such as ATP, $LiIO_3$, or KDP). The angle of the crystal is set to phase-match the frequency of the gating pulse with a selected frequency of the luminescence. A signal whose frequency is the sum of the laser and luminescence frequencies is generated in the crystal and detected by a photomultiplier. The laser beam is divided by a beam-splitter. One of the beams is directed to a delay prism and reflected back. The other is modulated by a mechanical chopper and excites the sample. The luminescence from the sample is collected and combined with the original laser pulse from the delay prism inside the crystal. When the crystal is tuned to satisfy the matching condition, the sum wave is generated. The signal frequency is separated by a monochromator and detected by a photomultiplier. The lock-in amplifier discriminates whatever background remains. Varying the delay of the gate pulse, we can obtain the temporal profile of the luminescence without the background luminescence. Because upconversion involves virtual electronic transitions, this gate has a response time equal to the pulse width of the pumping laser. To achieve a high efficiency of conversion, the gate has to be tuned to satisfy the phase-matching

conditions of

$$\omega_s = \omega_L + \omega_G$$

and

$$\bar{k}_s = \bar{k}_L + \bar{k}_G.$$

The intensity of the generated signal is given by the convolution function of I_L and I_G:

$$I_s(\tau) \alpha \int_{-\infty}^{\infty} I_G(t - T) I_L(t)\, dt.$$

The time resolution is an electronic response of $\sim 10^{-14}$ sec.

Pump and Probe Gate

In this technique, an intense pulse excites a sample. A weaker probe pulse derived from the initial pulse is reflected along a different and variable optical delay path in order to examine a parameter of the system such as light absorption or Raman scattering as a function of delay time after excitation. Probe pulses can be obtained from harmonic generation, stimulated Raman scattering, and self-phase modulation. The latter greatly expands the bandwidth to the so-called "ultrafast supercontinuum" (Alfano, see next section).[1-5]

The timing mechanism is the speed of light itself. It takes light 3.3 psec to travel 1 mm in the air. If the path length of the pump pulse is decreased by 1 mm, by moving the delay prism, the pump pulse will arrive at the sample site at 6.6 psec (twice the change in the path length) before the probe pulse. Using a stepping motor, delays on the order of 10 fs are possible. The kinetics of any absorption changes can be followed point by point by moving the delay prism across the time domain of interest, usually about 300 psec. The probe beam (supercontinuum) is generated by focusing the 1060-nm pulse into a CCl_4 cell (Alfano and Shapiro).[1-5] At the sample site, the probe beam is divided into two pulses of approximately equal intensity, designated $I^e(t)$ and $I^r(t)$. $I^e(t)$ is transmitted through the same area of the sample that is excited by the 530-nm pulse. The intensity of the transmitted probe beam depends, of course, on the changes initiated by the exciting pulse. $I^r(t)$ is the reference beam and is transmitted through a different part of the sample.

The change in optical density (OD) for a given delay time is obtained as

$$\Delta \mathrm{OD}(t) = -\log\{[I^e(t)/I^r(t)](I^r_o/I^e_o)\},$$

where the ratio, I^r_o/I^e_o, is the normalization factor for the two beams before the sample. The pump and probe method can have a resolution on the order of pulse duration ~ 10 fs.

Supercontinuum Source (SPM)

A most important nonlinear process with application in absorption spectroscopy and pulse compression into the fs regime is self-phase modulation (SPM) (Alfano and Shapiro).[1-5] SPM can be described by a simple model as follows: an intense optical

pulse traveling through a medium can distort the atomic configuration of the material, thus resulting in a change in the refractive index. The electric field of the laser beam in the time domain after traveling a distance, z, in the material is given by

$$E(t) = 1/2E_o(t) \exp\{-i(\omega_L t - n(t)z\omega_L/c)\} + \text{c.c.},$$

where $E_o(t)$ is the envelope of the pulse, ω_L is the laser angular frequency, and n is the total index of refraction. For a medium having inversion symmetry, the first nonlinear coefficient is $X^{(3)}$ and the index of refraction, n, becomes intensity-dependent:

$$n = n_o + n_2E^2.$$

The intensity-dependent term in the index of refraction modulates the spectral intensity given by the Fourier transform. It does so by modulating the instantaneous phase. This leads to a broadening of the pulse both on Stokes and anti-Stokes sides. The pulse becomes positively chirped (red in front and blue in the rear of the pulse). The broadening $\Delta\omega$ is given by

$$\Delta\omega = -\frac{\omega_L z}{c}\frac{\delta(n_2E^2)}{\delta t} \sim \frac{\omega_L n_2 E_o^2 z}{\tau_p}.$$

The pulse modifies its own spectra through a change in phase and envelope. These

TABLE 4. Supercontinuum Generation in Liquids, Glasses, and Semiconductors

Material	Pumping Pulse (nm)	Broadening Useful Continuum (nm)
CCl_4	1,060	440–900
	530	400–900
BK-7	530	400–800
Water	620	300–1,500
GaAs	9,300	3,000–15,000

processes are SPM and self-steepening, respectively. The broadening in liquids and solids is over several thousand wave numbers. TABLE 4 lists typical materials used in SPM. The duration of the broadened emission is comparable to the laser pulse. SPM is the key process behind femtosecond pulse generation by compression.

Pulse Compression

When an intense pulse propagates in a condensed medium, its shape and frequency change due to group velocity dispersion and induced nonlinearity polarization (n_2). The frequency sweep (chirp) is caused by SPM. In most cases, n_2 is positive, causing a positive chirp; that is, it causes a necessary frequency with time that leads to the red part in the leading edge and the blue part in the trailing edge. It is possible to eliminate the linear part of chirp and achieve pulse shortening using a medium with negative $\delta^2n/\delta\lambda^2$ curvature of dispersion or with a pair of gratings. The degree of compression can be varied by changing the distance between gratings. The central portion of SPM pulse (linear chirp) can be compressed with smaller secondary pulses positioned in the

wings. Using long optical fibers, the combination of linear dispersion and n_2 cause the pulse to become rectangular in shape with a chirp. Therefore, the entire pulse can be compressed using a grating pair. Pulse compression from 30 fs down to 8 fs has been achieved by Shank and co-workers using this technique.[10] The key processes in the production of short fs pulses are SPM and a grating pair.

FUTURE DIRECTIONS

The future research trends in ultrafast technology will be in the following areas:

Light Sources:
- —to produce the ultimate shortest light pulse—1 cycle (for 600 nm → 2 fs);
- —to generate a femtosecond pulse with high powers up to 10^{12} watts;
- —to extend the spectral region covering 100 Å to 1 mm;
- —to produce UV and X-ray picosecond lasers;
- —to produce a high-power supercontinuum source.

Detection Techniques:
- —to develop a streak camera with femtosecond time resolution;
- —to develop a neutron and gamma ray picosecond streak camera;
- —to develop optoelectronics with femtosecond resolution;
- —to build an excite and probe device with femtosecond resolution with high sensitivity (1 photon) and spatial resolution (1μ).

Interaction with Matter:
- —to measure the momentum and energy relaxation in semiconductors and microstructures;
- —to investigate ballistic propagation;
- —to induce chemical relaxation and new chemicals;
- —to study effects of laser-induced shock waves in condensed matter;
- —to study precursor propagation;
- —to study damageless propagation;
- —to investigate coherent excitation and coherence of light;
- —to investigate memory effects in matter;
- —to study transparency in matter;
- —to investigate primary steps in biological and medical media;
- —to investigate the nonequilibrium state of matter;
- —*and* now to test models of quantum theory, relativity, and light.

The field of ultrafast technology in science and engineering is just at its beginning.

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