QUANTUM ELECTRONICS

Sub-picosecond spectroscopy

Examples of ultrafast photoprocesses now being studied include vibrational dynamics of molecules, primary photobiological mechanisms and electronic processes in semiconductors.

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Ever since Man first became interested in movements and events that occurred too rapidly for his eye to follow, short flashes of light have been used to isolate moments of time. Each time shorter light pulses have become available, new areas of research have opened up, and new types of ultafast processes have become amenable to study. Spark photography can freeze the most rapid movement of macroscopic objects with flashes a tenth of a microsecond in duration. High-speed flashlamps and electronics play an important role in the study of fast photophysical and photochemical reactions with a resolution of a tenth of a nanosecond—10^{-10} sec. The last decade has seen dramatic advances in the development of short laser pulses and their application to the study of picosecond phenomena. Now, the extension of this technology into the sub-picosecond (10^{-13} sec) regime offers exciting possibilities for accurate studies of previously unresolved, ultrafast photoprocesses in physics, chemistry and biology. Investigations are being extended to such diverse and important topics as the vibrational dynamics of molecules and lattices, primary photobiological mechanisms and picosecond electronic processes in semiconductors.

Picosecond optical pulses became a reality about five years after the invention of the laser. In 1966, pioneering work at United Aircraft (now United Technologies) by Anthony DeMaria, D.A. Stetster and H. Heynau[1] with a nonlinear absorber inside a flashlamp-pumped Nd:glass laser first led to the generation of pulses less than 10 picosec in duration—too short to measure by conventional electronic means. This work provided the stimulus for the rapid development of a variety of new methods for pulse measurement and diagnostics. In the decade that followed, these techniques were refined and new pulse sources were invented to provide even shorter pulses.[2] The dramatic progress made during this period is illustrated by the plot in figure 1 of available pulse duration versus year. The Nd:glass laser remained the workhorse of the picosecond field during this period. A well-designed system can provide pulses in a range about 4 picosec with a certain degree of reliability. This laser provides the high peak powers important to many applications; but it is inherently a very low repetition-rate system, and it is subject to statistical fluctuation from pulse to pulse.

The passively-modelocked, flashlamp-pumped dye laser, invented by W. Schmidt and Fritz Schäfer[3] at the Max Planck Institute at Göttingen and successfully developed by Dan Bradley and his co-workers at Queen's University, Belfast[2] and later at Imperial College, London, provides wavelength-tunable picosecond pulses with durations down to about 2 picosec. It is still a flashlamp system with fluctuation and low repetition rate, but it generates pulses in a way that is fundamentally different from that of the Nd:glass laser. It was this difference that made possible the next generation of picosecond pulse sources: passively modelocked, continuously operated dye lasers. With the first operation of a continuous (cw) system[4] at Bell Labs, we reported pulses as short as 1.5 picosec. Subsequent improvements have led to systems that can produce pulses as short as 0.3 picosec.[5,6]

Perhaps even more important than the sub-picosecond pulse durations are the reproducibility from pulse to pulse and the very high repetition rates (greater than 10^5 pulses per second) obtained with cw systems. Powerful signal-averaging techniques can now be applied to picosecond studies. Measurements can be made with low-intensity pulses that do not distort the process under investigation. Recent experiments in our laboratory also indicate that a cw sub-picosecond pulse generator can provide the foundation for a high-power pulse source. Pulses from the generator can be amplified to high intensity in a series of laser-pumped dye amplifiers without significant change in pulse duration. It will become increasingly difficult for conventional flashlamp lasers to compete with such a system. In this paper we will therefore concentrate on the characteristics of the cw sub-picosecond laser, techniques being developed to take advantage of it, and some applications.

Sub-picosecond pulse generation

The process of sub-picosecond pulse generation in a dye laser is an interesting phenomenon in itself. With a proper adjustment of parameters it can occur without the presence of any picosecond recovery time in the dye media. Geoffrey New[7] first described the conditions under which dramatic pulse shortening occurs, and Hermann Haus[8] has obtained ana-

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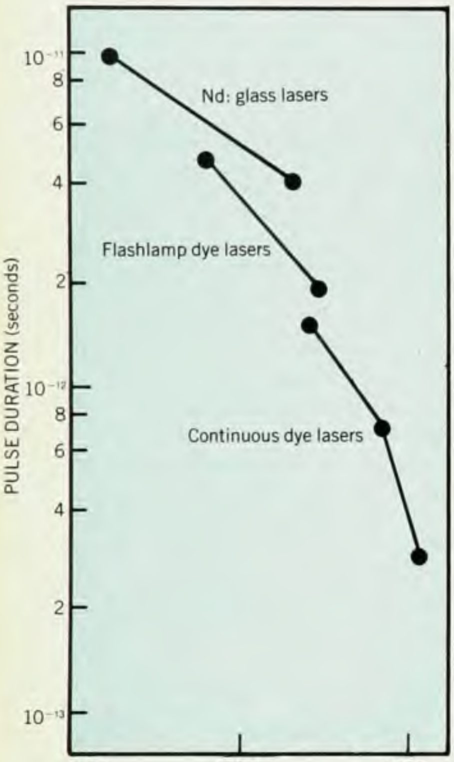
lytical solutions for steady-state pulse shape and stability in such systems.

Pulse narrowing in a single pass through the laser may be discussed qualitatively with the help of figure 2. Two dyes are involved in the process. A slowly recovering saturable absorber acts to steepen only the leading edge of the pulse. Saturation of the gain dye in combination with linear loss discriminates against the trailing edge. One condition for short pulse formation is simply that the absorber saturate more easily than the gain; this allows net gain at the peak of the pulse and loss on either side. The process is repeated as the pulse makes multiple passes through the laser. Both dyes substantially recover during the several nanosec between passes. Shortening continues until dispersive effects prevent the pulse from becoming narrower. This situation contrasts with the transient build-up in the Nd:glass laser, in which the gain cannot recover in a pulse round-trip time and a fast recovering saturable absorber is required to select the pulse from noise. Operation of the dye laser is continuous, with the result that the pulses always reach a reproducible, steady-state shape.



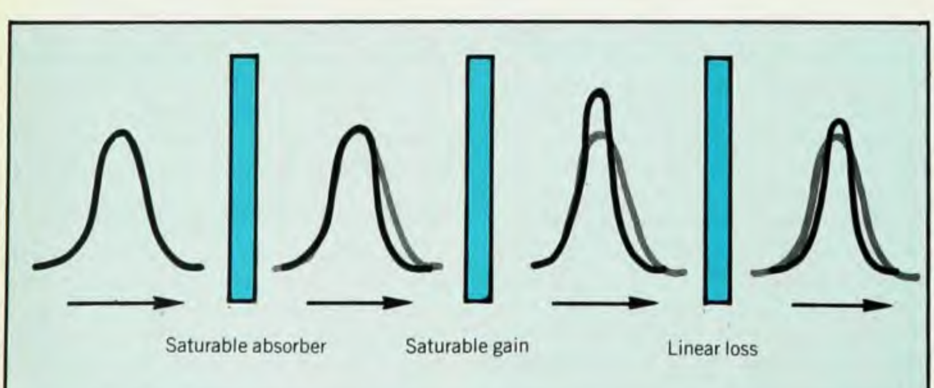
Decrease in laser-pulse duration during the last decade. In this article we consider primarily the production of sub-picosecond pulses with continuous lasers. Figure 1

Figure 3 is a schematic illustration of a sub-picosecond laser system that was used in the experiments described here. The system is pumped by several watts of continuous power from a commercial argon-ion laser. The dye gain medium, Rhodamine 6G in a free-flowing thin stream of ethylene glycol, is located at a focal point approximately in the center of the resonator. Near one end of the resonator is a second, free-flowing ethylene glycol stream containing the saturable absorber dyes. The laser mode is focussed more tightly in the absorber stream than in the gain stream.

Sub-picosecond pulses can be obtained with the saturable dye known as DODCI alone in the absorber stream. DODCI has a measured recovery time of 1.2 nanosec, so that pulse formation must occur in the manner described above. Experiments show that the addition of a second dye, malachite green, improves the overall stability and allows operation well above laser threshold. Malachite green alone will not induce modelocking, but it does provide supplementary absorption recovery on a picosecond time scale. With this combination of absorbers the laser in figure 3 is a reliable tool for sub-picosecond spectroscopy.

An acousto-optic deflector positioned near the other end of the resonator is used to "dump" single pulses from the laser. This scheme is preferable to using the pulse output from a partially transmitting mirror—the individual pulse energy is greater and the pulse repetition rate is adjustable. Single pulses can be dumped at rates greater than 10^5 pps to maximize the extent of signal averaging. Alternatively, the repetition rate can be reduced if the system being studied needs more time between pulses for complete recovery.

The sub-picosecond pulses produced by this laser each have an energy of approximately 5×10^{-9} joule. Beam quality is such that the pulses can easily be focussed to energy densities of several millijoules/cm^2 or photon densities greater than 10^{16}/cm^2.



Formation of sub-picosecond pulses in a dye laser. The saturable absorber steepens the leading edge of the original pulse, and the gain dye and the linear loss combine to discriminate against the trailing edge. The process is repeated during multiple passes through the laser. Figure 2

Pulse measurement

New experimental techniques are necessary before these sub-picosecond laser pulses can be used to study ultrafast processes. The fastest photodetectors and oscilloscopes are simply not capable of recording events with sub-picosecond resolution. The only sensors with this resolution are the pulses themselves; so measurement techniques have been developed that use one pulse to detect another, or one pulse to probe the response of a material to excitation by another pulse. Many of the techniques that were used in conjunction with flashlamp picosecond pulse sources can now be modified and improved for high resolution studies with continuous sub-picosecond sources. Reference 2 gives a chronology and summary of these techniques.

The first step in any sub-picosecond study is a careful characterization of the pulses themselves. We show an experimental pulse-measuring arrangement particularly suited for use with high repetition rate systems in figure 4. The pulse train from the laser is divided into two beams by a partially reflecting beam-splitter. The two beams then follow different paths in a modified interferometer and emerge parallel but not quite colinear. A relative delay between the two beams is obtained by changing the length of one path with a stepping-motor controlled translation stage. The stepping-motor drive also controls a multi-channel signal averager. Data from the measurements are then stored and averaged as a function of delay between the two beams. Notice that no high speed is required of the photo-electronic detection system; all the temporal resolution in these experiments is obtained by short optical pulses and accurate mechanical control of delay. The delay time τ is related to a stage translation Δz by $\tau = 2\Delta z/c$ where c is the speed of light. Delay reproducibility can be better than 10^{-14} sec.

In our pulse measurement, we focus the two beams by a simple lens through the same region of a thin crystal of potassium dihydrogen phosphate (KDP). This nonlinear crystal is oriented in such a way that phase-matched second harmonic generation occurs at an angle bisecting the two beams only when pulses from each beam are present simultaneously in the crystal. There is no background signal from each beam individually, and accurate measurement of overlap in the pulse wings is possible. In mathematical terms, one measures the autocorrelation function $G(\tau)$ of the pulse intensity $I(t)$:

$$G(\tau) \propto \int_{-\infty}^{\infty} I(t)I(t+\tau)\,dt$$

Although the pulse shape $I(t)$ cannot be deduced uniquely from $G(\tau)$, very good estimates are possible. Furthermore, analysis of time-resolved spectroscopic measurements generally requires accurate knowledge of only the correlation function and not of $I(t)$.

The efficiency of this second-harmonic generation scheme for pulse measurement is high. An accurate trace can usually be obtained by taking the average result of very few scans. Nevertheless, an important feature, for many applications, of the experimental arrangement of figure 4 is that it allows averaging over many rapid scans to improve the ratio of signal to noise. This guards against possible distortion due to system drift during a single, slow scan.

The pulses produced by the cw generator of figure 3 have by now been accurately characterized.[5] The shortest pulses are produced at wavelengths near 615 nanometers. When they are dumped from the laser they have a duration just under a picosecond; they are asymmetric in temporal shape and are frequency swept ("chirped") in a way consistent with normal dispersion. For experimental purposes they may be compressed in time and filtered to produce Fourier transform-limited pulses with a duration of about 0.3 picosec (as shown in figure 5).

Pump-probe experiments

With slight modification, the system shown in figure 4 can be applied to a great variety of sub-picosecond studies. The sample under investigation replaces the KDP crystal, and the detector is moved to monitor the transmission of one of the beams (the probe beam) through the sample. The probe beam is made considerably weaker than the other (pump) beam. At negative delay the probe pulses precede the pump pulses through the sample and are unaffected by any action of the pump. At positive delay, the probe pulses experience increased or decreased transmission due to the excitation. The change in transmission describes the dynamical behavior of the sample. With high-repetition-rate pulses and automated delay scanning, one has in effect a sub-picosecond optical sampling oscilloscope.

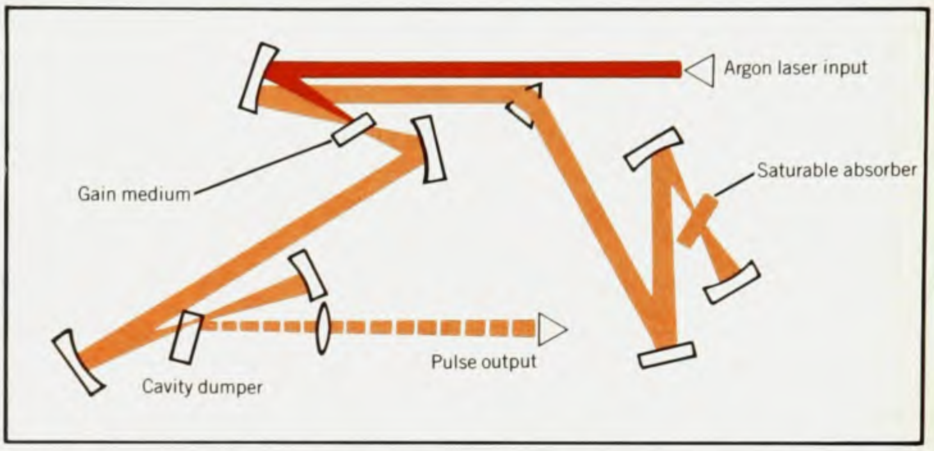
Pump-probe measurements can monitor saturation, depletion and recovery of the original species. They can be applied to studies of induced absorption due to excited states, energy acceptors or photochemical products. With the addition of polarization analysis, one can evaluate the dynamic contributions of induced dichroism or birefringence. The experimental arrangement is also easily extended to measure amplitude and polarization changes in the surface reflectivity of solids.

A measurement made over several minutes with a cw system allows averaging over more than 10^7 pulses. Changes in sample transmission as small as 10^{-4} are easily detected. With such small perturbations one avoids the distorting effects of stimulated emission and other nonlinear processes that often accompany high-power laser measurements.

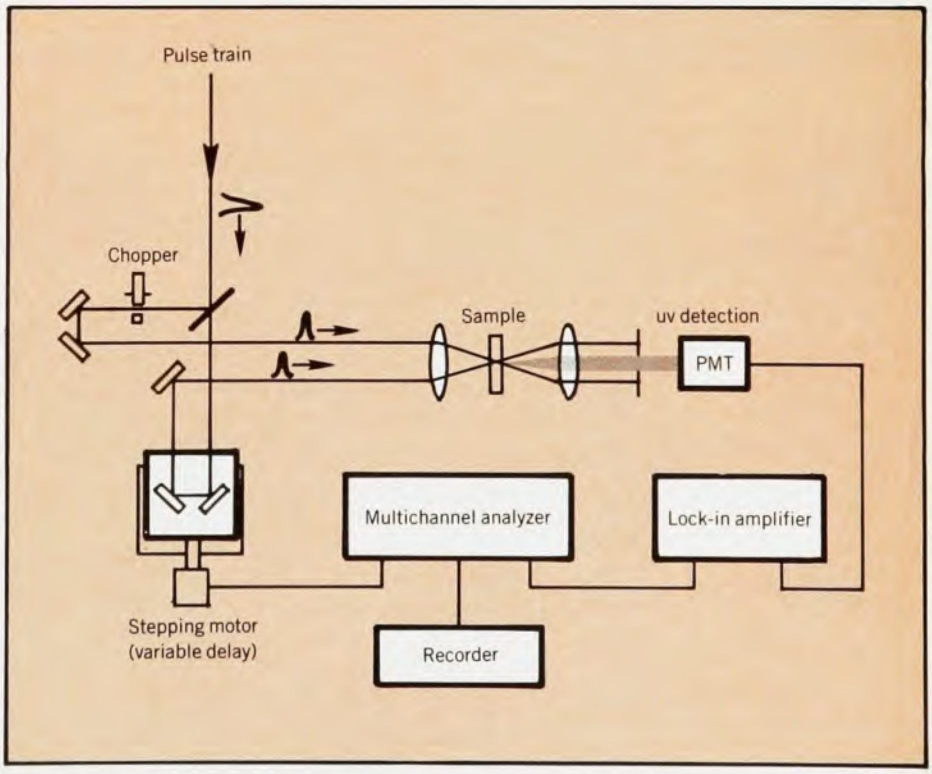
Accurate interpretation of pump-probe measurements near zero delay also requires that one analyze the effects of pulse coherence. Consider the response expected from an ideal, instantaneously responding system. The transmission change $\Delta T(t)$ produced by the pump pulse $I(t)$ alone is given by the proportionality

$$\Delta T(t) \propto \int_{-\infty}^{t} I(t')\,dt'$$

if we assume that the system does not recover on the time scale of interest. After convolution with a probe pulse that has the same shape and is coherently related



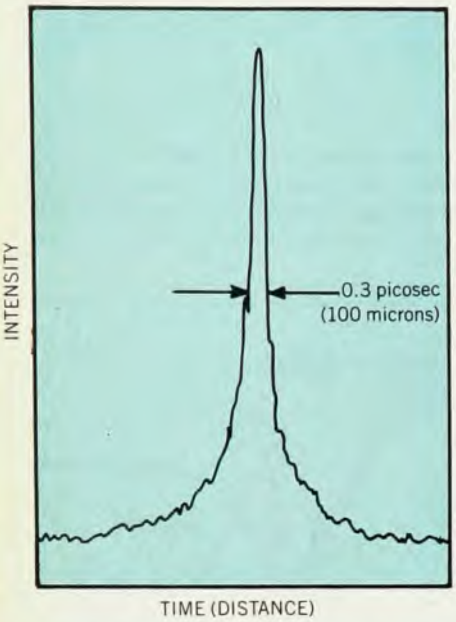
A continuously operated sub-picosecond pulse generator. Pumping (top right) is by a commercial argon-ion laser. The two dyes are Rhodamine 6G in ethylene glycol (labeled "gain medium") and the saturable absorber, which contains a mixture of DODCI and malachite green. The "cavity dumper" (lower left) is an acousto-optic deflector positioned to dump single pulses out of the laser at high repetition rate. Figure 3



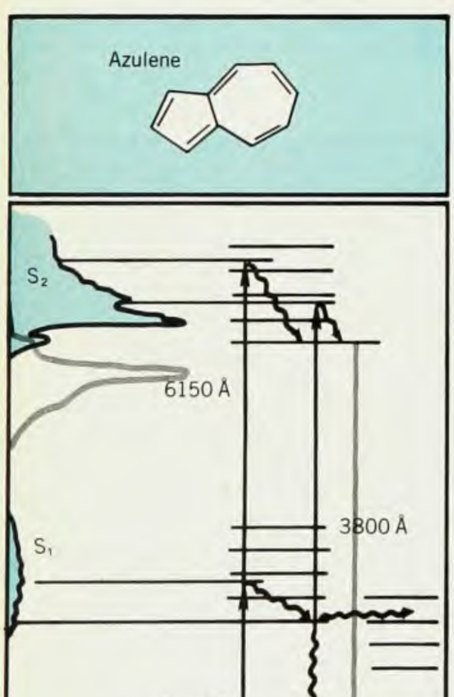
Pulse diagnostics for high-repetition-rate, sub-picosecond pulses. The stepping motor moves a translation stage to provide variable delay between two beams. Pulse measurements are made by nonlinear correlation in a sample of potassium dihydrogen phosphate, KDP. Figure 4 to the pump, the measured response is

$$M(\tau) \propto \int_{-\infty}^{\tau} G(\tau')\, d\tau' + E(\tau)$$

The first term is the expected response if



Autocorrelation measurement of a sub-picosecond pulse produced by the generator shown in figure 3 and measured by the arrangement of figure 4. Figure 5



Azulene, $C_{10}H_8$, has unusual fluorescence properties that can be studied in the experimental arrangement of figure 4. Figure 6

the pulses were not coherently related. It can be obtained simply by integration of the measured pulse autocorrelation function $G(\tau)$. The second term is a coherence artifact inherent in nonlinear spectroscopy. It is significant only when the pump and probe pulses overlap in time and interfere, so that $E(\infty) = 0$. One can also show that

$$E(0) = \int_{-\infty}^{0} G(\tau')\, d\tau' = \frac{1}{2} M(\infty)$$

Thus, $M(\tau)$ has already reached its eventual level at $\tau = 0$ and does not suffer the delay that one might expect from the finite pulsewidth. In a more general case the coherence contribution near $\tau = 0$ is affected by the dynamic response of the system, but its contribution can still be calculated for comparison with experiment.[2]

Optical gating

The concept of gating adds an additional dimension to ultrafast spectroscopic studies, allowing time resolution of emission due to fluorescence or light scattering. The most widely used picosecond gate is the optical Kerr shutter developed by Michel Duguay and his coworkers at Bell Labs.[7] In this device a picosecond light pulse induces a transient birefringence in a Kerr active material, situated between two polarizers set initially for extinction. The induced birefringence permits momentary passage of the light signal under investigation. Relative timing of the gate and the signal is controlled in the manner outlined above for pump-probe experiments.

We have operated a carbon-disulfide Kerr shutter with pulses from the cw sub-picosecond generator. High repetition rate allows a new mode of operation with polarizations biased for maximum incremental transmission, but gate resolution is limited by the 2 picosec (orientational) response time of CS_2.[8] With amplified pulses, efficient Kerr gating will be possible with faster (electronic) Kerr materials.

An alternative to the Kerr shutter for gating is frequency up-conversion. The optical signal to be sampled is mixed in a nonlinear crystal with the picosecond gating pulse. Conversion to the sum frequency occurs only in the presence of both optical fields. The advantage over the Kerr shutter is that there is less background signal in the absence of the gating pulse. Gating efficiencies can be comparable. Herbert Mahr and Mitchell Hirsch of Cornell recently employed up-conversion successfully in conjunction with a cw picosecond system.[9] More widespread application to time-resolved spectroscopy can be expected in the near future.

Frequency conversion

To expand the potential applicability of sub-picosecond spectroscopy even further, one would like to generate pulses at a variety of wavelengths—not only to have new excitation frequencies, but also to do simultaneous probing at different wavelengths. Advances in this area rely primarily on nonlinear optical frequency conversion.

Sub-picosecond pulses from the cw dye laser can be converted into sub-picosecond ultraviolet pulses near 307 nm by phase-matched second-harmonic generation in lithium iodate. Because of the short pulse duration one must guard against pulse spreading by group-velocity mismatch between the two pulse frequencies. In $LiIO_3$ a transit-time mismatch of 1 picosec/mm between the red and the uv requires the use of a thin crystal. With a crystal thickness of about 0.2 mm, a conversion efficiency of 15% is still possible. The sub-picosecond uv pulses produced can be used in conjunction with visible probe pulses to study relaxation from highly excited states to lower intermediate levels.[10]

With high power pulses a greater number of nonlinear optical techniques[2] become possible for generating frequencies from the vacuum ultraviolet to the infrared region of the spectrum. Particularly useful is the technique known as continuum generation.[11] Recently, in our laboratory, multiple-stage amplification of sub-picosecond pulses from the stable cw oscillator has resulted in peak powers approaching a gigawatt at a repetition rate of 10 pps. Focussing these pulses into water produces efficient conversion to a continuum of light extending from the near infrared to the near ultraviolet. Measurements at selected wavelengths indicate that these continuum pulses, like the pulses producing them, have durations less than a picosecond.

Recent applications

It is worth reiterating that sub-picosecond pulse generation is reproducible with the system we have described. This reproducibility, combined with the high repetition rate, greatly extends the potential of time-resolved spectroscopy. To illustrate the use of some of the different measurement techniques, we describe now some recent experimental applications. The particular examples, by necessity limited in number, have been chosen to emphasize the interdisciplinary nature of work in this area.

Nonradiative relaxation in molecules. The kinetics of energy redistribution and excited-state relaxation play an important role in the chemistry and photochemistry of molecules. Nonradiative processes can be especially fast, and they are not always amenable to study by conventional techniques. This is particularly true of the molecule azulene, which theoretical chemists find interesting because of its unusual fluorescence properties. The molecular structure and a schematic energy diagram of azulene is given in figure

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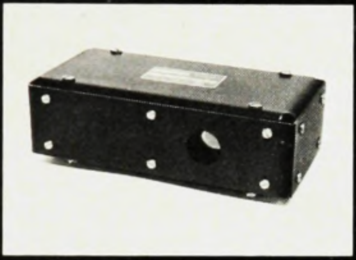

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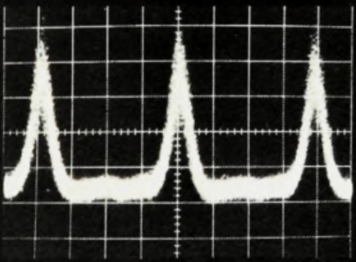

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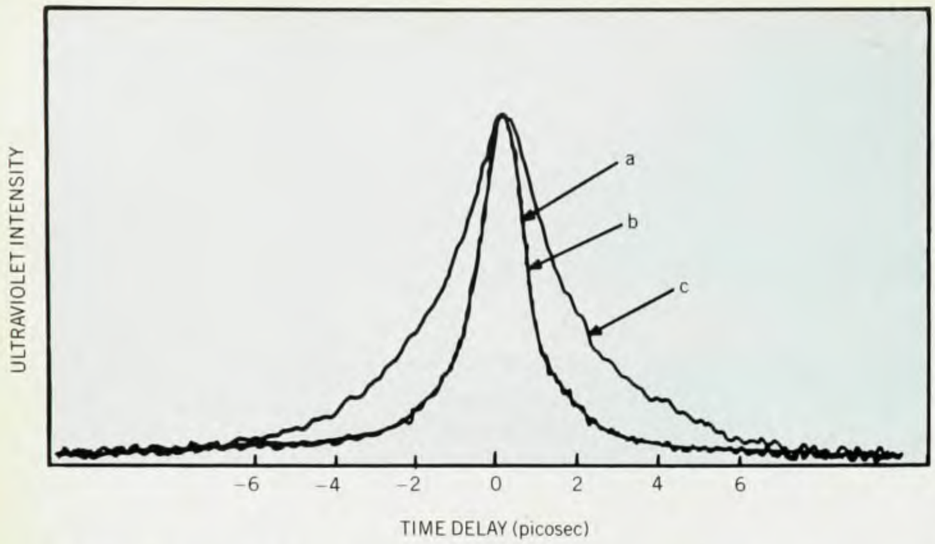


Pulse correlations. Curve **a** is by second-harmonic generation, curve **b** by two-photon fluorescence and curve **c** by two-step induced fluorescence in azulene. Curve c is broadened by the picosecond lifetime of the S_1 state. Figure 7

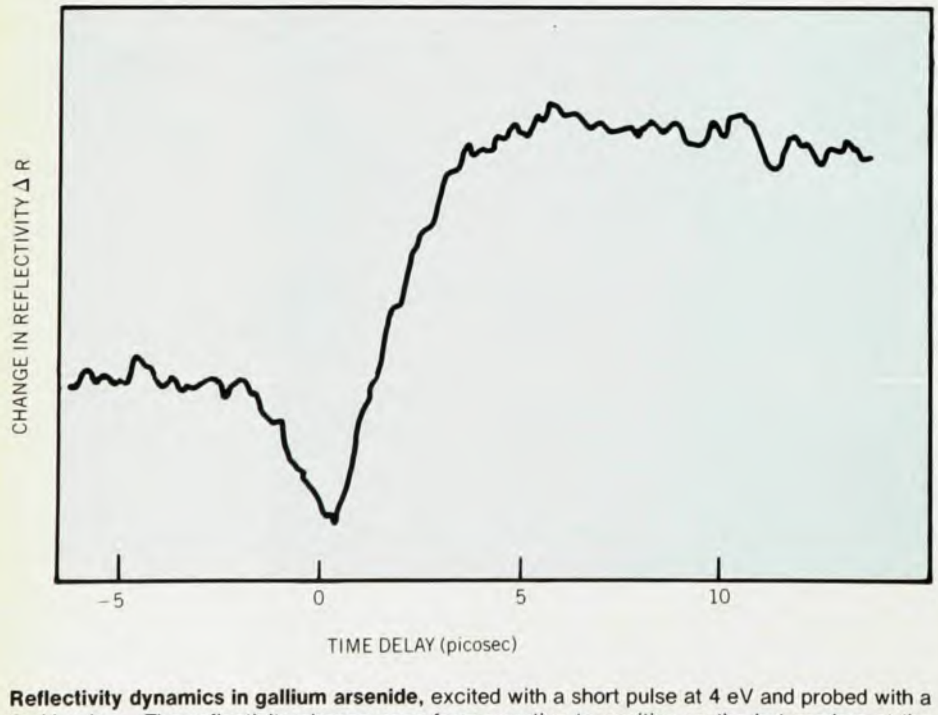
6. Of particular interest is the lifetime of the lowest excited single state, S_1.[12] A very low quantum efficiency of fluorescence indicates that this level is rapidly depopulated by nonradiative decay. In this case, the fact that fluorescence can be observed following excitation to high states suggests a method for monitoring the population in S_1.

The experimental arrangement of figure 4 lends itself well to this purpose. With the KDP in place, ultraviolet light is detected only when the two pulses arrive at the same time. If the KDP is replaced by a thin cell containing a solution of azulene, the situation is different. A single pulse can excite molecules to S_1 where they remain for some short time τ. If the second (delayed) pulse arrives within this time, some molecules are elevated further to a state from which fluorescence can occur. Each individual pulse can also induce two-step excitations, but this process only gives a constant background independent of delay. By measuring the average fluorescence as a function of delay, one can deduce τ. A reference curve (one for which $\tau = 0$) is obtained by substituting a dye (α − NPO) that has no intermediate state; the experiment then becomes simply an autocorrelation measurement of the pulses by two-photon fluorescence.

Typical experimental curves we obtained in this study appear in figure 7. Two reference curves ($\tau = 0$) one by second-harmonic generation autocorrelation and the other by two-photon fluorescence, are indistinguishable on a 0.1-picosec scale. The third curve is clearly broadened by the S_1 lifetime. Proper analysis and deconvolution yield a simple exponential behavior with a time constant of 1.9 ± 0.2 picosec. That we obtained the same result in a variety of solvents indicates, not surprisingly, that the surrounding solvent molecules have little influence on this time scale. More recently, we found that deuteration of the azulene has no dramatic effect. Ground-state recovery measurements made with amplified sub-picosecond pulses have also yielded a time of 1.9 ± 0.5 picosec and point to the conclusion that the lifetime of state S_1 is limited by direct coupling to the ground state. These experiments and others like them provide a basis for comparing theoretical models and will, we hope, lead to a better understanding of nonradiative relaxation.

Carrier dynamics in semiconductors. Short optical pulses provide a unique way to observe, directly, the dynamics of hot-carrier distributions in semiconductors. The experimental method is to generate a distribution of hot carriers at the surface of a semiconductor by band-to-band absorption of an optical pulse of frequency much greater than the bandgap. A second, less intense, optical pulse then probes the incremental reflectivity change at a delayed time relative to the generation. While the carriers relax by phonon emission the reflectivity is altered (due to the continuously changing occupation probabilities of different electronic states) until the carrier distribution is in thermal equilibrium with the lattice. Details of the relationship between the optical reflectivity and the carrier distribution depend on the frequency of the probing pulse and the band structure of the particular semiconductor.

The case of gallium arsenide excited with a short pulse at 4 eV and probed with a 2-eV pulse is shown in figure 8. In this example the sign of the reflectivity changes goes from negative to positive as the carriers relax through the probing pulse energy. The energy loss rate for electrons and holes can be estimated from the zero crossing to be 0.4 eV/picosec.

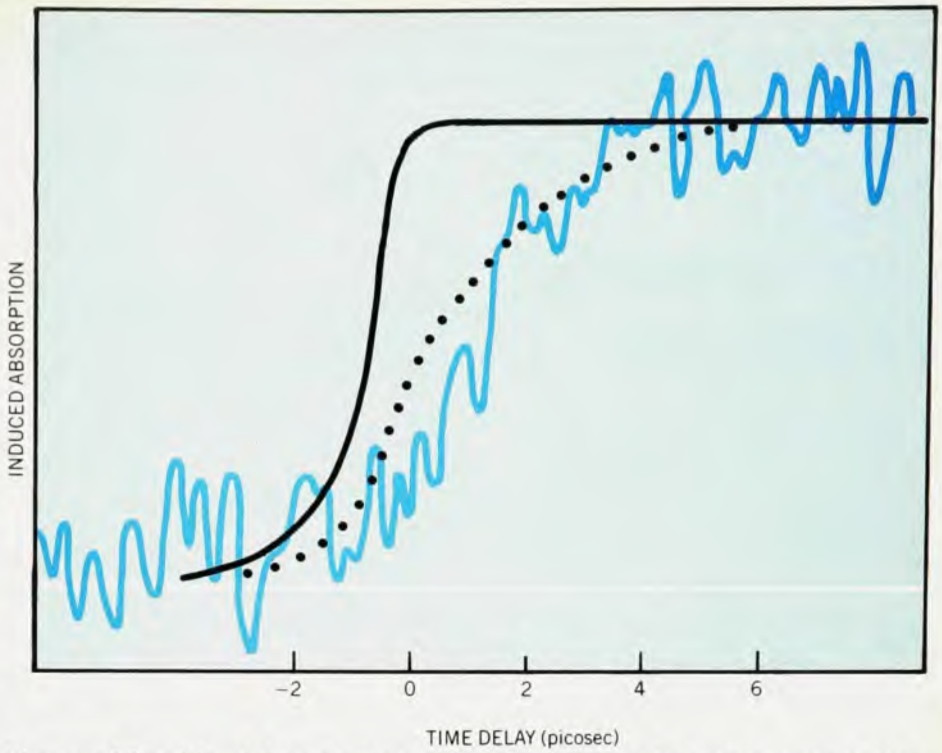


Reflectivity dynamics in gallium arsenide, excited with a short pulse at 4 eV and probed with a 2-eV pulse. The reflectivity change goes from negative to positive as the hot carriers at the semiconductor surface relax through the probing pulse energy. Figure 8

Biological processes. It is becoming apparent that time-resolved spectroscopy can make important contributions to the better understanding of biological molecules.[2] Although conventional measurement techniques can determine that a biological reaction has taken place, the complexity of these molecules often masks the actual atomic or molecular rearrangement that has occurred. Recent experiments indicate that many of these processes do occur with picosecond speeds. Dynamic measurements may be used to distinguish between the different possible reaction pathways; for example, we have used sub-picosecond pulses to investigate the rate of photodissociation of hemoglobin compounds,[13] and, most recently, the ultrafast dynamics of bacteriorhodopsin's photochemistry.

Bacteriorhodopsin is the object of intense research interest because of its similarities to visual pigment rhodopsins and because of its biological role as an energy converter. The absorption of light is known to induce very rapid formation of a more energetic species with a red-shifted absorption maximum.[14] We have applied the pump-probe technique discussed above to resolve the dynamics of this primary step: bacteriorhodopsin→bathobacteriorhodopsin. The pump excitation is expected to result in an increase in absorption of a probe at 615 nm. The colored curve in figure 9 shows the experimental result, which is consistent with the expectation. The black curve is the calculated (and experimentally verified) response of the measurement system to an instantaneous rise. It includes the effects of coherence near zero delay. The dotted line shows an ideal 1.0-picosec response. The actual curve deviates somewhat from this simple curve, but nevertheless approaches its final value with a time constant of 1.0 ± 0.5 picosec.

This result argues against any major change in molecular configuration. It does, however, support a proposal of Aaron Lewis[15] in which the instantaneously excited electron distribution in the chromophore induces a conformational transition in the neighboring protein. Further sub-picosecond measurements are now planned to distinguish specific conformational changes.



Picosecond time-delayed absorption increase in bacteriorhodopsin. Excitation by a "pump" pulse increases the absorption of a "probe" pulse at 615 nanometers. The colored curve is experimental data, resolving the delay in the photochemical reaction. The black curve is the calculated response to an instantaneous rise, and the dotted line shows a 1.0-picosecond response. Figure 9

Future directions

Sub-picosecond pulse lasers similar to that shown in figure 3 are now being put into operation in a number of different university and industrial laboratories. As they begin to be used in increasingly varied applications, existing techniques will certainly be improved and new sub-picosecond techniques will be developed. Lasers with different dye combinations will be found to generate sub-picosecond pulses in other regions of the spectrum, and we expect that pulses even shorter than 0.3 picosec will eventually be produced.

Several other sub-picosecond pulse sources based on the cw dye laser are in the early stages of development. Workers at the University of California at Berkeley[16] have obtained pulses as short as 0.6 picosec from a doubly modelocked laser. In this mode of operation the saturable absorber dye is allowed to lase in synchronism with the gain medium; short pulses are produced at two wavelengths simultaneously. Workers at Bell Labs have recently obtained independently tunable pulse outputs at two wavelengths by synchronously locking two dye lasers to the same modelocked dye laser.[17] The same group produced tunable, sub-picosecond pulses by operating two synchronously modelocked lasers in tandem.[18]

Amplification of sub-picosecond pulses to high peak powers will be important for many applications. We have already demonstrated that amplified pulses can be converted efficiently to new wavelengths and that they can be used to generate a sub-picosecond continuum for studies encompassing a broad spectral range. High-power pulses can now be applied to the study of low density materials and low cross-section interactions. Even more important may be new investigations of weak nonlinear processes and highly excited systems. Because the threshold for material breakdown is higher for shorter pulses, amplified sub-picosecond pulses can be focussed to optical intensities of record magnitude.

The importance of picosecond processes and devices to both fundamental and applied research is becoming increasingly apparent. This month the first international Topical Conference on Picosecond Phenomena is being held in Hilton Head, South Carolina. Topics to be discussed include photosynthesis, vision processes, electron solvation, nonradiative molecular dynamics, hot electron effects in semiconductors, short wavelength generation, laser-induced fusion, and picosecond electronics. Important advances will certainly be made and new areas of importance uncovered now that these investigations can be pursued on a time scale of 10^{-13} sec.

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