formerly Optical Spectra magazine

PHOTONICS SPECTRA®

The International Journal of Optics, Electro-Optics, Lasers and Fiber Optics August 1982

A NEW ERA IN FAST SPECTROSCOPY

LASERS ON THE FAST TRACK:

ACHIEVING PICOSECOND AND SUBPICOSECOND PULSES

A NEW ERA IN FAST SPECTROSCOPY

by A.G. Doukas and R.R. Alfano

At the Ultrafast Spectroscopy and Laser Laboratory (USL) of the City College of New York's Department of Physics, we use picosecond and femtosecond lasers and associated techniques to study the dynamical processes in matter. The past ten years have seen the arrival of a new era in spectroscopy, that of ultrafast time-resolved spectroscopy. Each advancement in the technology of ultrafast pulse generation has opened up new areas of research in physics, chemistry and biology, but the race among investigative groups has been particularly keen in the last few years. Currently, pulses as short as 30 femtoseconds (30×10^{-15} second) have been generated by passively mode-locked dye lasers. We will consider the generation of picosecond and femtosecond pulses by passive mode-locking, and the two types of lasers involved, describe solid-state and dyes.

Fourier analysis tells us that any repetitive pulse train in the time domain can be expressed as the sum of many sine waves of frequencies that are integral multiples of some basic frequency and that have fixed phases. The story of the generation of short pulses is the attempt to bring the modes of the laser into fixed phase with each other. Ways of doing this are locking the modes by amplitude or phase modulation.

Consider some theory. A laser resonator will only allow certain discrete frequencies to build up and propagate within the cavity. These frequencies, or modes, are determined by the fact that the field inside the resonator reproduces itself, forming a standing wave. In addition, the fluorescence of the lasing medium helps determine the frequency and bandwidth of the emitted light. The lasing frequencies are those allowed by the resonator inside the fluorescence profile of the lasing medium.

Depending upon the lasing medium and the length of the resonator, the number of modes can be very large. For Nd^{+3}:glass, to take one example, the number of modes under the fluorescence profile of Nd in a one-meter cavity is about 10^4. So the means of producing short pulses are there; all it takes is making the laser's modes oscillate in phase. For a passively mode-locked laser, that means putting a saturable absorber inside the laser cavity.

Figure 1 is a schematic of a solid-state laser. The oscillator consists of an appropriately doped glass rod with both ends cut at the Brewster angle and a cell of bleachable dye between two highly reflective mirrors. A flashlamp provides the energy for exciting the laser's active medium. In a free-running laser, the phase of the differing modes' oscillation is random with respect to each other, and the laser's intensity in the time domain has the characteristic of thermal noise.

The formation of a short pulse has three distinct qualitative phases.[1] Initially there is only spontaneous fluorescence inside the cavity. Because the absorbance of the saturable dye is nonlinear, it will absorb noise pulses that are above the background less than it does the background noise. In the second step the ultimate pulse will be amplified more than the low-intensity noise pulses. When the intensity of the pulse goes above $10MW/cm^2$, the absorber bleaches, and the pulse is amplified even more. But the pulse width increases because of dispersion inside the rod. In passing through the absorber, the wings of the pulse are absorbed more than the peak. The streak camera makes it possible to observe the evolution of a short pulse from the noise.[2] Figure 2 shows a streak microdensitometer trace of the pulse evolution in a ruby laser.[2] The pulse duration depends on the recovery time of the saturable absorber. Pulses as short as 3psec have been reported for Nd:glass lasers.[3]

A unique tool

Mode-locking of dye lasers has made available a new tool for research. To begin with, the extremely large bandwidths of dye lasers can produce ultrashort pulses in the femtosecond regime. In fact, the current record for short pulses is less than 100 femtoseconds[4] and for externally dechirped pulses 30fsec.[5] Pulse duration is only 15 optical wavelengths in space. In addition, dye lasers can operate at very high repetition rates, greater than 10^8 Hz, and can be tunable.[2,6]

Figure 3 is a schematic of a passively mode-locked ring dye laser. The lasing dye (Rh-6G) and the mode-locking dye (DODCI) are flowed through two nozzles that are set at the Brewster angle. The Rh-6G jet stream is located between mirrors 7 and 8, whose radii are 10cm, and the DODCI stream between mirrors 3 and 4, whose radii are 5cm. Thickness of the jet stream is about 160μm. Three other flat mirrors,

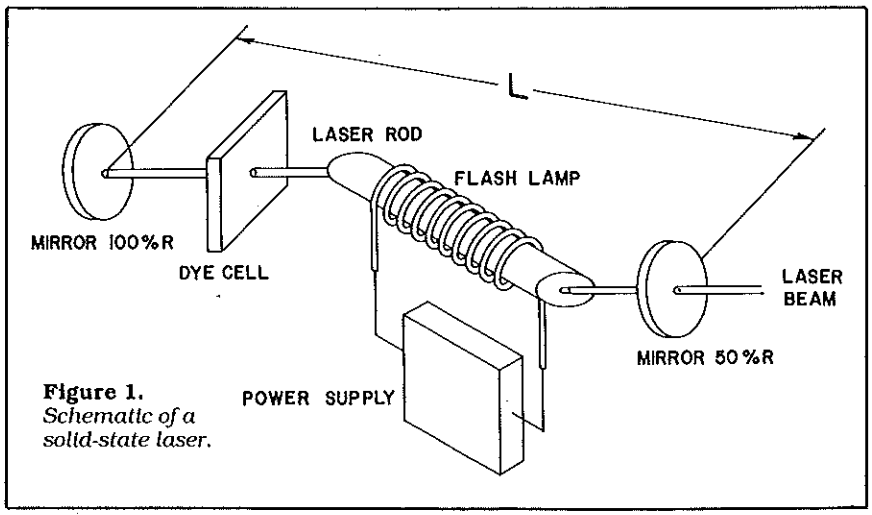

Figure 1. *Schematic of a solid-state laser.*

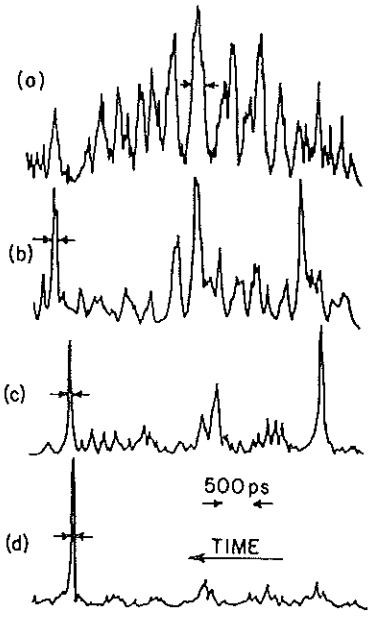

Figure 2.
Streak-camera microdensitometer trace shows the evolution of a short pulse from the noise. The laser is ruby, mode-locked by DODCI. Time resolution is 30psec. Traces a-d correspond to 3μsec, 2.5μsec, 1.5μsec and 1μsec, respectively, before the peak of the giant pulse (see Ref. 2).

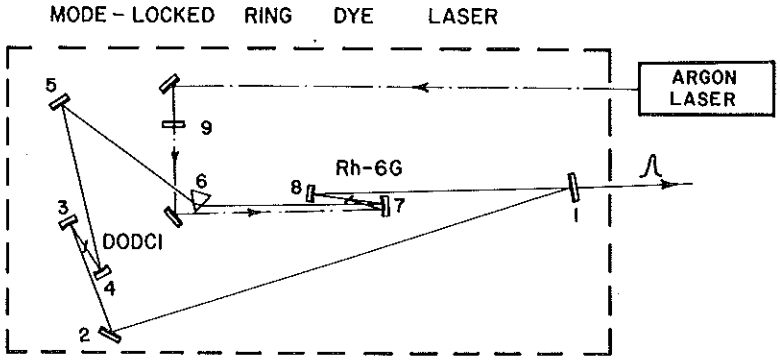

Figure 3.
Schematic of a passively mode-locked ring dye laser.

numbered 1, 2 and 5, complete the cavity. A Brewster prism (6) in the cavity provides the means of tuning the laser's wavelength over 200A.

To obtain more efficient use of pumping energy, a λ/2 retardation plate (9) is placed in the path of the pumping laser beam and changes the polarization of the argon laser's light to match that of the dye laser. When the polarizations match, the threshold pump power of the dye laser is only 900mW. Normally, however, the polarization of the argon laser is "S" and that of the dye laser "P." In this case, the threshold is more than 3W. The decrease in the threshold power arises from the anisotropic absorption and gain cross-sections of the Rh-6G dye, and makes for more efficient pumping.[7] The laser produces pulses of 300-500fsec at the rate of 125MHz.

What limits the pulse duration seems to be the thickness of the DODCI jet stream, which should be less than the pulse width. Shank and his coworkers[4] have recently demonstrated that a DODCI jet stream 10μm thick can produce 90fsec pulses.

With the ring dye laser, the process of pulse formation is quite different from that described for the solid-state laser. For one thing, the recovery of the saturable absorber does not have to be very fast. Because the leading edge of the pulse is more absorbed by the bleacher dye than the peak portion, the absorber will sharpen a pulse propagating through it at the leading edge, and the rest of it goes on through the bleached dye without absorption. At the lasing medium, on the other hand, the fast depletion of the excited state because of the lasing action causes the trailing edge to sharpen. These two effects combine to generate ultrashort pulses.

Dye laser pulses are of low energy (in the picojoule to nanojoule range). Therefore, they must be amplified for most applications. In a typical three-stage amplifier, three dye cells are pumped by the second harmonic (0.53μm) from a Nd:YAG laser, which is operated at 10Hz — that is, only 10 pulses of approximately 125MHz can be amplified. Total boost of the system is 10^{+6} to 10^{+7}, which yields a pulse with energy of about ½mJ, but when the beam is focused, it can produce power densities of 10-100 gigawatt/cm^2.

How it works

The method for time-resolved spectroscopy is a basic one: the molecular system is excited by a strong laser pulse, and the subsequent relaxation of the system is studied by such spectroscopic techniques as fluorescence, absorption and Raman spectroscopy.

Most widely used for fluorescence measurements is the technique that incorporates a streak camera.[8] Photoelectrons emitted by light striking the photocathode at different times are deflected by an applied voltage ramp that causes electrons to be transversely streaked across a phosphorescent screen at the same time that they are accelerated through the anode. Such photoelectrons, released at different times from the photocathode, will strike the phosphorescent screen at different positions, causing a track whose spatial intensity profile is directly proportional to the incident temporal profile of the fluorescence.

A recently developed technique for time-resolved fluorescence spectroscopy is population mixing.[6] The technique does not require a multistage amplifier and streak camera, yet it provides a resolution limited only by the pulse duration. Figure 4 shows a schematic of the apparatus.

Population mixing is accomplished in the following manner. The pulse train is divided by a beamsplitter into two trains modulated at frequencies w_1 and w_2. A variable delay (τ) is added to one of the beams, and both beams are focused by a lens onto the sample. The photoluminescence is then collected, focused on the slit of a monochromator and detected by a photomultiplier and a lock-in amplifier set at the difference frequency $|w_1 - w_2|$. In this way, the lock-in amplifier measures only the photoluminescence signal produced by the mixing of the populations generated by the two pulse trains. The two pulses create free-carrier population n(t) and n(t+τ), respectively. The detected signal is

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

If a single exponential decay time (T) of the carrier is assumed, we obtain

$$I_s(\tau) \propto \exp(-|\tau|/T)$$

The correlation function supplies information about the relaxation of the photogenerated carriers. Similar

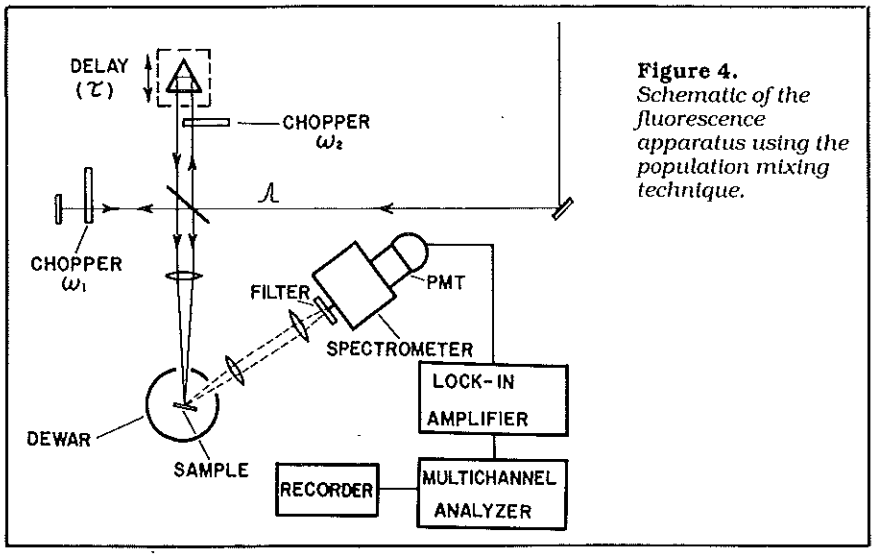

Figure 4. *Schematic of the fluorescence apparatus using the population mixing technique.*

methods developed by von der Linde and coworkers have been used for time-resolved photoluminescence[9] and Raman[10] measurements.

In a schematic of the picosecond absorption apparatus, the timing mechanism of the kinetics device is the speed of light itself. Light takes 3.3psec to travel 1mm in air, and if the pathlength of the pump pulse is decreased 1mm by moving a delay prism, the pump pulse will arrive at the sample site 6.6psec — twice the change of the pathlength — before the probe pulse. The kinetics of any absorption changes can be followed point by point by moving the delay prism across the time domain of interest.

The probe beam, or super-continuum, is generated by focusing the 1060nm pulse into a CCl_4 cell ten centimeters long.[11] The super-continuum of subpicosecond pulse duration can also be produced by focusing the pulses from the subpicosecond dye laser into CCl_4 or optical fibers. Changes in absorbance measured through the difference in optical density at a given delay time between the arrival of the exciting and probe pulses are easily obtained as:

$$\Delta OD\,(t) = -\log\left[\frac{I^e\,(t)\,I^p_0}{I^e_0\,I^p\,(t)}\right]$$

where the superscripts e and p denote the transmitted intensity of the probe with and without the exciting pulse, respectively, at delay time (t). Typically, ΔOD approximately equal to 10^{-3} can be measured. A similar experimental apparatus has been used with a mode-locked dye laser to study absorption kinetics with femtosecond resolution. □

References

1. Letokhov, S.V. (1969). Generation of ultrashort light pulses in lasers with a nonlinear absorber. *SOV. PHYS. JETP.* 28:562-568.
2. Bradley, D.J. (1977). Methods of generation in *Ultrashort Light Pulses, Picosecond Techniques and Applications.* S.L. Shapiro, ed. Springer-Verlag, New York. 17-81.
3. Alfano, R.R., N.H. Schiller, and G.A. Reynolds (1981). Production of picosecond pulses by mode-locking a Nd:glass laser with dye #5. *IEEE JOURN. QUANT. ELEC.* QE-17:290-291.
4. Fork, R.L., B.I. Green, and C.V. Shank (1981). Generation of optical pulses shorter than 0.1psec by colliding pulse mode-locking. *APPL. PHYS. LETTS.* 38:671-672.
5. Shank, C.V., R.L. Fork, R. Yen, R.H. Stolen and S. Tomlinson (1982). Compression of femtosecond optical pulses. *APPL. PHYS. LETTS.* 40: 761-763.
6. Rosen, D.L., A.G. Doukas, Y. Budansky, A. Katz and R.R. Alfano (1981). A subpicosecond tunable ring dye laser and its applications to time-resolved fluorescence spectroscopy. *IEEE JOURN. QUANT. ELEC.* QE-17:2264-2266.
7. Dorsinville, R. (1978). Laser action and orientational relaxation in binary solutions of dyes. *OPTICA ACTA* 25: 375-377.
8. Schiller, N. et al. (1980). An ultrafast streak camera system. *Optical Spectra* 14:6:55-63.
9. Von der Linde, D., J. Kulh, and E. Rosengart (1981). Picosecond correlation effects in the hot luminescence of GaAs. *JOURN. OF LUMINESCENCE* 24/25:675-678.
10. Von der Linde, D., J. Kulh, and H. Klingenberg (1980). Raman scattering from nonequilibrium LO phonons with picosecond resolution. *PHYS. REVIEW LETTS.* 44:1505-1508.
11. Alfano, R.R. and S.L. Shapiro (1970). Emission in the region 4000 to 7000A via four-photon coupling in glass. *PHYS. REV. LETTS.* 24:584-587.

A.G. Doukas, currently Research Associate Professor of Physics and Assistant Director of USL, is a graduate of the University of Thessaloniki (Greece) and received his PhD in physics from the City University of New York in 1977. Robert R. Alfano, professor of physics at CCNY, is the director of the USL.

Research at USL is sponsored by AFOSR, NIH, NASA, NSF, CUNY FRAP and Allied Corporation. The authors would like to thank Hamamatsu Corporation for technical support.