Discovery of the Tunable Dye Laser

Jeff Hecht

The narrow-emission bandwidth of laser light quickly attracted the attention of spectroscopists in the early 1960s, but that narrow linewidth came at a cost—the wavelength was fixed. Laser researchers found that they could shift the fixed wavelength somewhat by applying magnetic fields to the laser, they developed tunable parametric oscillators, and eventually they found a few laser lines that were tunable. But those arrangements were cumbersome and their range limited. As a student in the mid-1960s, spectroscopist Theodor Hänsch felt "a sense of frustration" that he had no way to tune lasers "to wavelengths that were interesting."

What spectroscopists really wanted was a laser that could be tuned across a broad range of interesting wavelengths. The first such tunable laser, the organic dye laser, was discovered by accident in research on Q-switching ruby lasers. The first Q switches were active devices based on Kerr cells or rotating mirrors, but in early 1964 the first passive Q switches were developed using saturable absorbers. Later that year, Peter Sorokin at the IBM Watson Research Center showed that certain organic dyes dissolved in solvents made simpler and more convenient saturable absorbers.

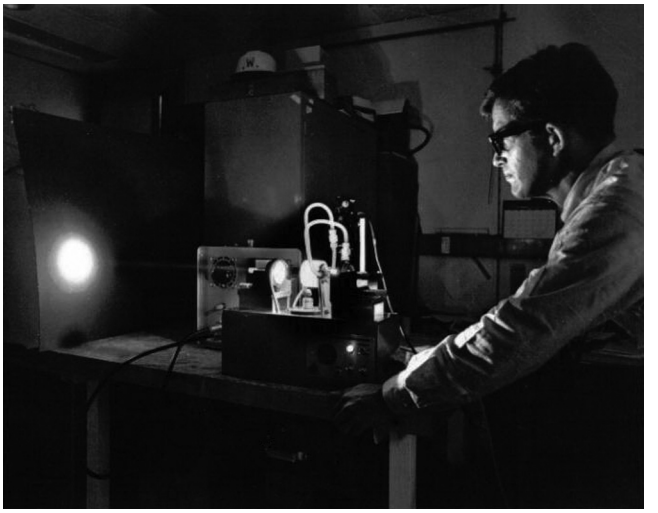
After that success, Sorokin found himself with a large collection of dye compounds that had been prepared for the saturable absorber experiments. The dyes had interesting properties including strong fluorescence, so he decided to try producing stimulated Raman scattering. He fired pulses from a big Korad ruby laser into a dye that had never been tested in Q switching. The first experiment produced a black smudge on a photographic plate, but it was late Friday afternoon and he had to leave. Monday morning, 7 February 1966, he told his assistant Jack Lankard they should try aligning a pair of mirrors with the dye cell before they fired the laser again. "Jack came back from developing the plate with a big grin on his face. There was one place in the plate that the emulsion was actually burnt," Sorokin later recalled. They knew it was laser action because the bright line was at the peak of the dye fluorescence

Word of their experiments traveled slowly; Sorokin chose to publish his results in the March 1966 issue of the *IBM Journal of Research and Development* because he liked the editor, but it was not widely read. That gave two other groups a chance to independently invent the dye laser.

The idea of a dye laser came to Mary Spaeth, then at Hughes Aircraft Co., about the same time Sorokin was working on his experiment. She recalls, "I was sitting on my bed with my two year old daughter on my lap, two months pregnant with my second daughter, and about 20 papers spread out in front of me. I had been studying dyes that had been used for many years for photographic purposes. In particular, I was studying models for how they are excited and how they transfer energy from one molecule to another in the photographic process. The excited states of these dyes have a geometry very similar to their ground states, so they have very strong absorption spectra. I suddenly realized that if a dye could be put in a suitable solvent, you could have an enormous population inversion after illumination by a short-pulse laser. It was just like the light bulb pictures you see in the funny books. Boing! There it was, clear as day."

She also realized that because dyes have huge numbers of rotational states, they should have a broad gain bandwidth, so that placing dispersive optics in a laser cavity with the dye solution should allow wavelength tuning. But first she wanted to try exciting the dye with pulses from a

ruby laser. It was not part of her job, so it took her months to make arrangements to pump dyes with a ruby laser in Dave Bortfeld's lab. As she sat epoxying a dye cell together, Bortfeld entered the room and threw a paper airplane at her. She recalls, "I looked at him to try to figure out why he had done that. As I unfolded the airplane, I found it was a copy of Sorokin's paper," which Bortfeld had just spotted. She knew the dyes, so she instantly realized what it was about. "We decided, what the heck, we were working independently, and we continued on our way."



▲ **Fig. 1.** Peter Sorokin with the flashlamp-pumped dye laser in 1968. (Courtesy of International Business Machines, © International Business Machines Corporation.)

Expecting the dye to emit at a wavelength a little longer than 700 nm, she did not set up a detector, figuring she would be able to see the laser spot on a magnesium oxide block. However she didn't see anything. "I was about eight months pregnant, I had trouble reaching the knobs on the oscilloscope, it was 7 in the evening, and I was very tired," she recalls. Bortfeld told her to go home, while he set up a photodetector and tried again. He called later that evening to tell her it had worked.

In further tests, they changed dye cells and moved their optics and found the oscillation wavelength of one dye changed from 761 to 789 nm when they tried cells from 8 mm to 10 cm long, and mirror spacing from 10 to 40 cm. They sent a paper to *Applied Physics Letters*, which received it 11 July 1966 and published it in the 1 September issue. It was the first report to show that dye laser wavelength could be changed, although it was not yet practical tuning. Spaeth did not get the chance to explore tuning further. Hughes management had no interest in dye lasers, and she had a difficult childbirth, so her immediate priorities became recovering and dealing with two small children.

Fritz Schaefer wrote that his group at the Max Planck Institute in Germany was unaware of either effort when they stumbled upon the dye laser while studying saturation in a different group of organic dyes. A student was testing the effects of increasing the dye concentration by firing ruby pulses into the solution, Schaefer wrote, when "he obtained signals about one thousand times stronger than expected, with instrument-limited risetime[s] that at a first glance were suggestive of a defective cable. Very soon, however, it became clear that this was laser action." They may have learned of Sorokin's work after submitting a paper on their results which *Applied Physics Letters* received on 25 July, two weeks after Spaeth's paper. (After revisions received by APL on 12 September, Schaefer's paper was published in the 15 October 1966 issue, citing Sorokin's paper but not Spaeth's.) Like Spaeth, they reported wavelength changes, in their case arising from changes in dye concentration.

Sorokin soon demonstrated flashlamp pumping, shown in Fig. 1, which proved important because it could pump dyes across a broader range of wavelengths than the ruby laser. In 1967 Bernard Soffer and Bill McFarland at Korad replaced one cavity mirror with an adjustable diffraction grating to make the first continuously tunable dye laser. They tuned across 40 nm and also reduced emission linewidth by a factor of 100. At last, spectroscopists had a broadly tunable laser, and they soon were busy exploring the possibilities.

Triplet-state absorption in the dyes limited pulse duration to nanoseconds in those early pulsed lasers, but in 1969 Ben Snavely from Eastman Kodak and Schaefer found that adding oxygen to the solvent could quench triplet absorption. Snavely then teamed with Kodak colleagues Otis Peterson and Sam Tuccio to develop a continuous-wave (CW) dye laser. They first investigated prospects for pumping with intense plasma light sources, then tried pumping with an argon-ion laser. That required longitudinal excitation and liquid flow to keep the dye solution cool, deplete triplet states, and avoid

▲ **Fig. 2.** Mary Spaeth at Livermore. (Courtesy of Lawrence Livermore National Laboratory.)

thermal lensing. In 1970, they produced CW output of about 30 mW at 597 nm when pumping a dye solution flowing between a pair of dichroic mirrors with a 1-W argon-ion laser.

Further refinements followed. Trying to increase CW dye output by increasing the pump power and focusing it onto a smaller spot tended to burn the coatings off the quartz windows covering the dye. That problem was solved when Peter Runge and R. Rosenberg at Bell Labs developed a way to flow a jet of dye solution through the pump beam in a laser cavity without confining it, so there was no glass or coating to be damaged.

Pulsed dye lasers had launched tunable laser spectroscopy. CW dye lasers and higher powers led to a series of landmark experiments. Conger Gabel and Mike Herscher at Rochester reached tunable single-mode dye power of 250 mW between 520 and 630 nm and used intracavity harmonic generation to produce tunable ultraviolet power of up to 10 mW. Felix Schuda, Herscher, and Carlos Stroud at Rochester stabilized a CW dye laser to 10 to 15 MHz to measure a the hyperfine absorption spectrum of the sodium D line, showing that dye lasers could do important experiments in fundamental physics.

Spectroscopy with CW dye lasers advanced rapidly. Two-photon Doppler free spectroscopy with dye lasers, which allows extremely precise wavelength measurement, was developed independently in 1974 by David Pritchard at MIT and by Arthur Schawlow and Theodor Hänsch at Stanford.

CW operation of broadband dyes also opened the way to ultrashort laser pulses. In 1964, Willis Lamb had showed that mode locking could generate extremely short laser pulses with duration limited by the Fourier transform of the laser bandwidth. As long as laser bandwidth was limited, mode locking could not generate very short pulses. However, with suitable optics a CW dye laser could oscillate across most of the dye's emission bandwidth, allowing mode locking to generate ultrashort pulses. In 1972, Erich Ippen and Charles Shank generated 1.5-ps pulses by passive mode locking of a dye laser, and in 1974 they generated subpicosecond pulses with kilowatt peak power. That launched the growth of ultrafast technology, described in a later section by Wayne Knox.

As Schawlow wrote in the speech he gave when receiving the 1981 Nobel Prize in Physics, "spectroscopy with the new [laser] light is illuminating many things we could not even hope to explore previously." One of the amazing things was the small shifts of transition wavelengths between different isotopes of elements such as uranium. Tunable narrow-line dye lasers could resolve those shifts, offering the possibility of selectively exciting the fissionable isotope U-235. As described in another article in this section, the Lawrence Livermore National Laboratory used banks of dye lasers, pumped by large copper-vapor lasers, to enrich both uranium and plutonium. At Livermore, Spaeth (Fig. 2) found support for her interest in dye lasers, and managed development of massive CW dye lasers that generated kilowatts for Livermore's uranium-enrichment demonstrations.