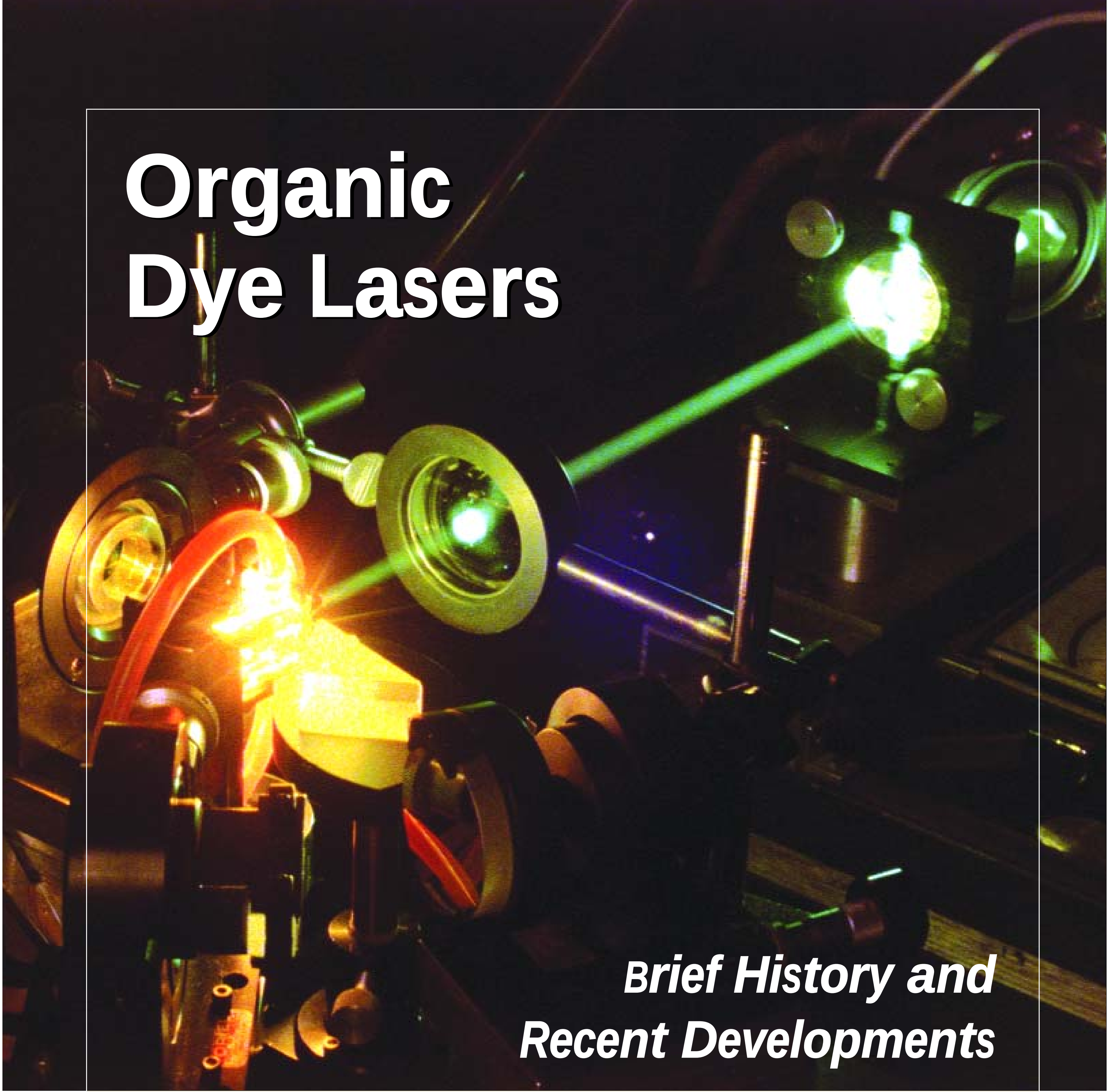
Organic Dye Lasers



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Brief History and Recent Developments

Frank J. Duarte

The organic dye laser has the distinction of being the first broadly tunable laser. It is also capable of providing a wide variety of output forms that range from ultrashort to high energy pulses, and from highly stable continuous wave (CW) narrow linewidth oscillation to high average power emission. Current application areas include medicine, laser isotope separation and basic physics research, including laser cooling. The author provides an overview of the history of tunable dye lasers, a synopsis of recent research and a look at what the future may hold.

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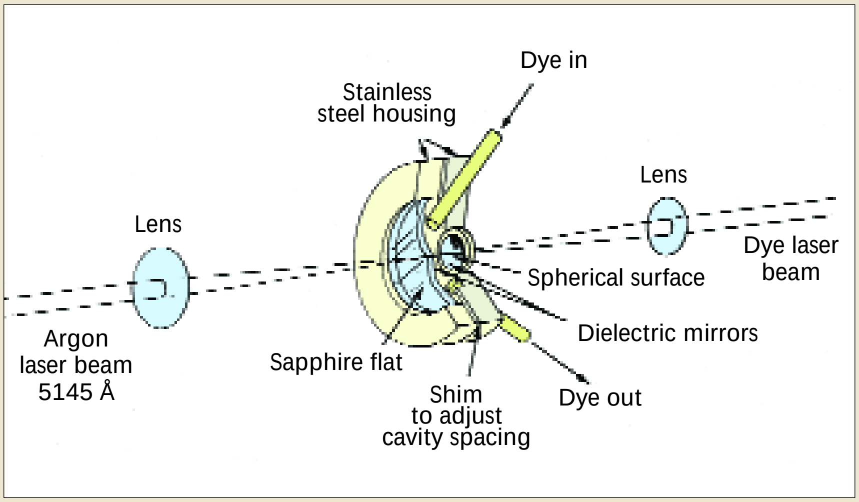


Figure 1. Longitudinal excitation scheme used in the first CW dye laser. [From Peterson et al.[19]]

Organic dye lasers were discovered in 1966 by Sorokin and Lankard[1] and Schäfer et al.[2] This is the simple part of the story, since the field quickly evolved into a variety of subfields described in numerous papers published in the refereed literature. Before I outline the history of these tunable coherent devices, some general comments are in order. First, because of their wide applicability and resulting popularity, dye lasers have given origin to an enormous body of scientific literature. A recent computer search for scientific articles with the words "dye laser" in the title or abstract yielded more than 18,000 references. This simple exercise demonstrates the extraordinary impact of the dye laser in scientific research. One example that stands out in this regard is the field of laser spectroscopy, which was revolutionized by the ability to produce tunable narrow linewidth emission throughout the visible and the near infrared. Additional applications are found in physics, chemistry, Lidar, medicine and industry.

A second issue, one that must be addressed, is that over the years dye lasers have acquired a reputation in some quarters as being "user unfriendly." This perception contributed to the high level of activity in the solid-state laser arena that led to development of several highly successful tunable sources. Today, dye lasers continue to be used in many applications that require unique wavelength agility in the visible or specific output characteristics, such as high average power or high pulse energy. Thus, the dye laser exists as a complementary source of coherent radiation alongside the numerous solid-state lasers developed in the course of the past two decades.

In this article I focus on dye lasers in the high pulse energy, high average power, CW, femtosecond and narrow linewidth categories. I also present a brief survey of developments in the solid-state dye laser subfield. It should be noted that the historical summary provided here is, by definition, incomplete. For comprehensive historical summaries, interested readers should refer to titles such as *Dye Lasers*,[3] *Dye Laser Principles*[4] and *Selected Papers on Dye Lasers*.[5] Note that, in the case of pulsed dye lasers, the term "narrow linewidth emission" is reserved for linewidths $\Delta\nu \leq 2$ GHz, and preferably for single longitudinal mode emission.

High pulse energy dye lasers

The first excitation of dye lasers was performed using pulsed ruby lasers.[1,2] This work was quickly followed by the successful demonstration of laser action using flashlamp pumping.[6,7] Dye laser action using the second harmonic of a Nd:YAG laser was demonstrated in the same time period.[8] Early excitation work using gas lasers included reports on nitrogen-laser,[9] excimer-laser[10] and copper-vapor-laser[11] pumping.

Flashlamp pumped dye lasers have been shown to yield high energy pulses in the microsecond regime at a low pulsed repetition frequency (prf). This category includes dye lasers excited by linear flashlamps capable of delivering 40 J per pulse,[12] dye lasers excited transversely that provided some 140 J per pulse[13] and dye lasers using coaxial flashlamp excitation yielding up to 400 J per pulse.[14] Large excimer laser pumped coumarin dye lasers have been reported to deliver some 800 J per pulse in the blue-green region of the spectrum with 500 ns long pulses.[15] In most cases this output energy was in the form of broadband emission.

High average power dye lasers

Pulsed dye lasers are known to yield coherent tunable radiation at high average powers. An important feature of the laser dye as a liquid gain medium is its ability to flow at linear speeds suitable for the necessary cooling to be achieved. Flashlamp pumped dye lasers operating at a prf in the 850 Hz range have been reported to yield up to 1.2 kW in five second bursts.[16] The previously mentioned 140 J per pulse laser[13] was reported to deliver average powers in excess of 1 kW. An excimer pumped dye laser emitting at $\lambda \approx 400$ nm, developed at Los Alamos National Laboratory,[17] delivered 50 W at a prf of 250 Hz. Copper vapor laser excitation of a dye laser system at the Lawrence Livermore National Laboratory[18] yielded more than 2.5 kW at a prf of 13.2 kHz. The emission was narrow linewidth and tunable in the 550-650 nm region. This laser was developed for atomic-vapor-laser-isotope separation.[18]

Continuous wave dye lasers

A very important development was the introduction of the CW dye laser by Peterson et al.[19] in 1970 (see Fig. 1). This breakthrough provided the necessary infrastructure for the subsequent development of the femtosecond laser. Single longitudinal mode oscillation, using an

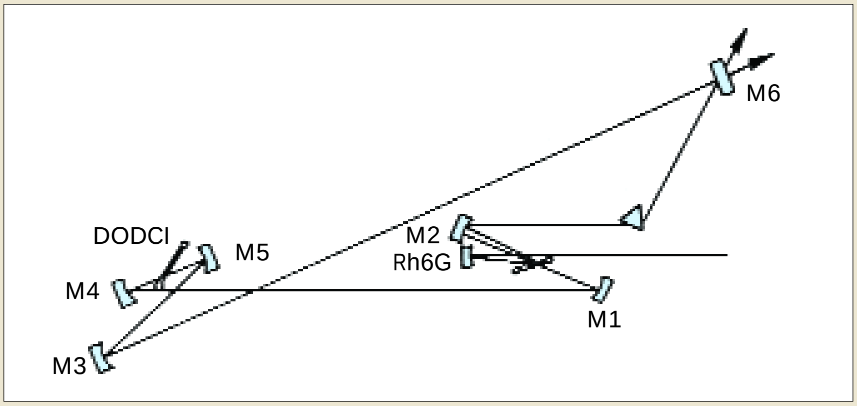


Figure 2. Prismatic pulse compression in ring dye lasers. This particular configuration yielded pulses shorter than 60 fs. [From Dietel et al.[37]]

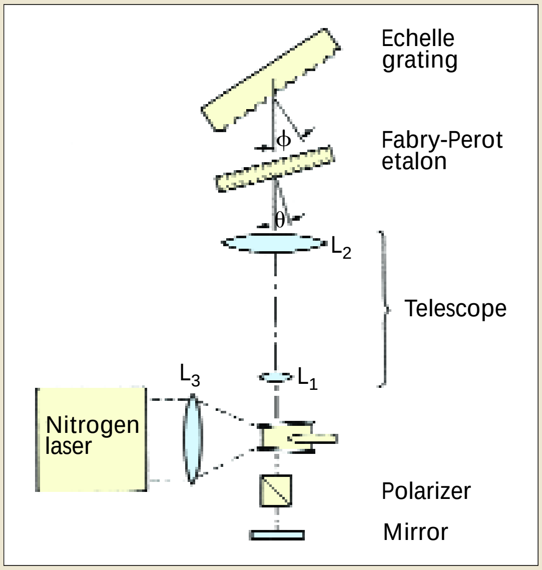


Figure 3. First narrow linewidth broadly tunable pulsed dye laser. This oscillator architecture uses a transmission telescope to illuminate the diffraction grating deployed in the Littrow configuration. [From Hänsch.[44]]

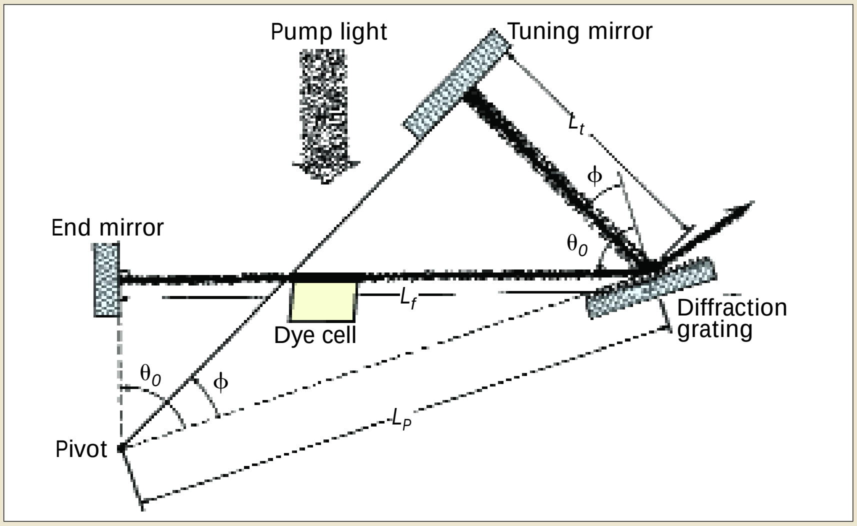


Figure 4. Grazing incidence cavity incorporating synchronous tuning mechanism. [From Liu and Littman.[48]]

intracavity prism and an etalon, was reported by Hercher and Pike,[20] and spatial hole burning in these lasers was demonstrated by Pike.[21] Other significant developments included the introduction of astigmatically compensated cavities[22] and the use of a dye jet.[23] Frequency stabilization of CW dye lasers was demonstrated in 1973 by Barger et al.[24] and the use of an external optical resonator to achieve linewidths in the 100 Hz range was reported by Drever et al.[25] The dual use of electro-optic and acousto-optic modulators in frequency stabilization was described by Hall and Hänsch.[26] The extraordinary wavelength coverage and agility available from these lasers was made evident in a series of papers by several authors.[27-29] CW dye lasers have played a significant role in atomic and molecular physics, including the area of laser cooling.

Ultrashort pulse dye lasers

The use of dyes as intracavity saturable absorbers was first reported in mode-locking experiments in solid-state lasers.[30, 31] Self-mode-locking in dye lasers with a saturable absorber[32] was demonstrated in 1968. An important development in this field was the demonstration of passive mode locking in CW dye lasers by Ippen et al.[33] in 1972. Bandwidth limited subpicosecond pulses[34] were generated in 1976. Shortly thereafter, the "era of the femtosecond"[35, 36] began.

Momentous developments in the area of ultrashort pulse lasers include the demonstration of colliding pulse mode-locking[36] and the introduction of prismatic pulse compression techniques[37-39] (see Fig. 2). Fork et al. reported six femtosecond pulses in 1987 by use of an intracavity multiple prism pulse compressor and an extracavity assembly integrated by a multiple prism compressor in sequence with a multiple grating compressor.[40]

Distributed feedback dye lasers

Distributed feedback configurations are widely used in semiconductor lasers and in the generation of short pulse radiation. Distributed feedback dye lasers were first demonstrated in 1970 by

Kogelnik and Shank.[41] Shortly thereafter, Shank et al.[42] introduced the well known interferometric excitation configuration. The coupled-wave theory of distributed feedback lasers followed these experimental developments.[43]

Narrow linewidth tunable pulsed laser oscillators

The first narrow linewidth tunable laser oscillator (shown in Fig. 3) was introduced by Hänsch[44] in 1972. This telescopic Littrow grating configuration greatly enhanced intracavity dispersion by expanding the beam that illuminated the diffraction grating. Grazing incidence grating cavities[45-47] were introduced in the late 1970s. Synchronous tuning in these cavities (see Fig. 4) was demonstrated soon afterwards.[48] These designs were widely applied to semiconductor lasers in the 1990s.

Prismatic beam expansion,[49-51] introduced in the early 1970s, eventually led to replacement of the telescope by multiple-prism beam expanders.[52,53] Prismatic pre-expansion of near grazing incidence oscillators followed shortly thereafter.[54] In addition to the lasers at Lawrence Livermore National Laboratory,[18] other high prf multiple-prism grating oscillators applicable to multiple step laser excitation were developed in various laboratories around the world.[55-57] Typically these oscillators can deliver single longitudinal mode emission at linewidths in the $450 \leq \Delta\nu \leq 600$ MHz range without the use of an intracavity etalon.[55] These dispersive oscillator architectures (see Fig. 5) have been widely applied to linewidth narrowing in gas as well as solid-state lasers.

Narrow linewidth master oscillators for injection of high pulse energy flashlamp pumped dye lasers include those based on multiple etalon designs[58,59] and multiple-prism grating architectures.[60] The laser described by Duarte et al.[60] is a highly stable and rugged dispersive oscillator that delivers a linewidth of $\Delta\nu \approx 300$ MHz. It was designed and engineered for and with the U. S. Army. Funding restrictions at the end of the Cold War prevented further development, including integration with high energy forced oscillator stages.

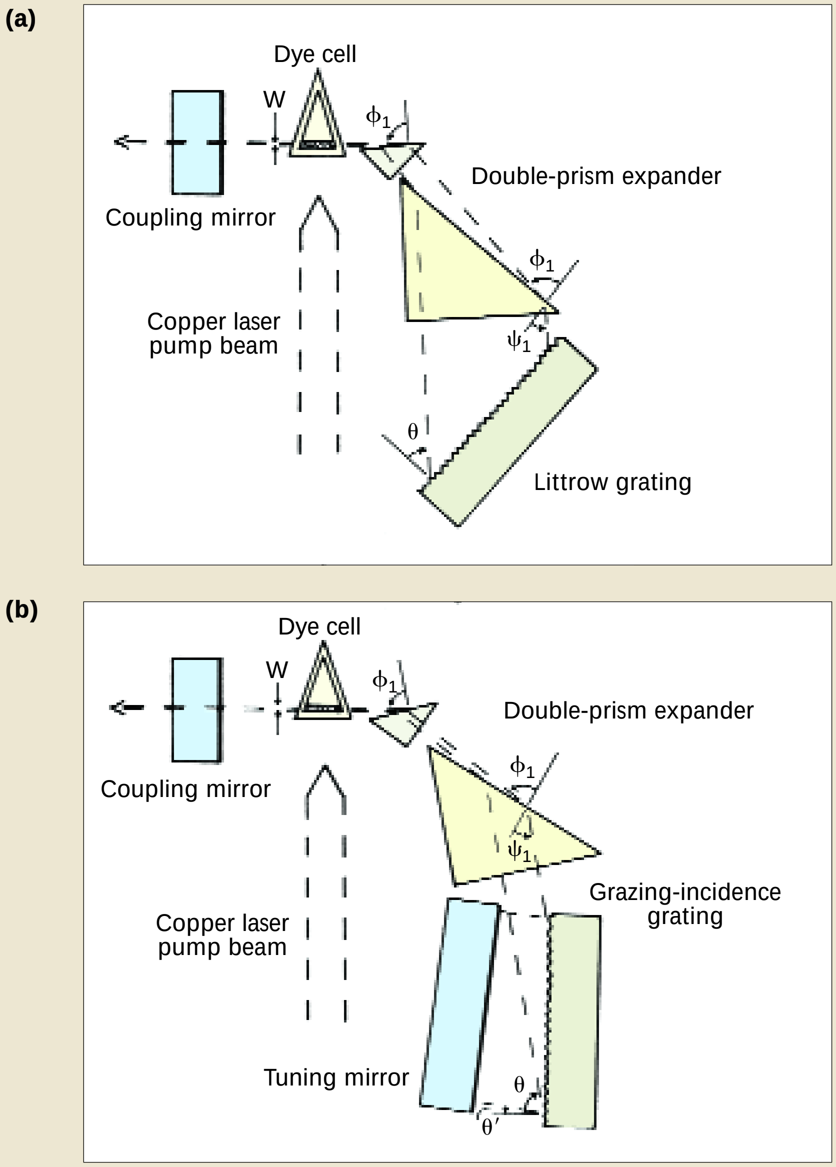


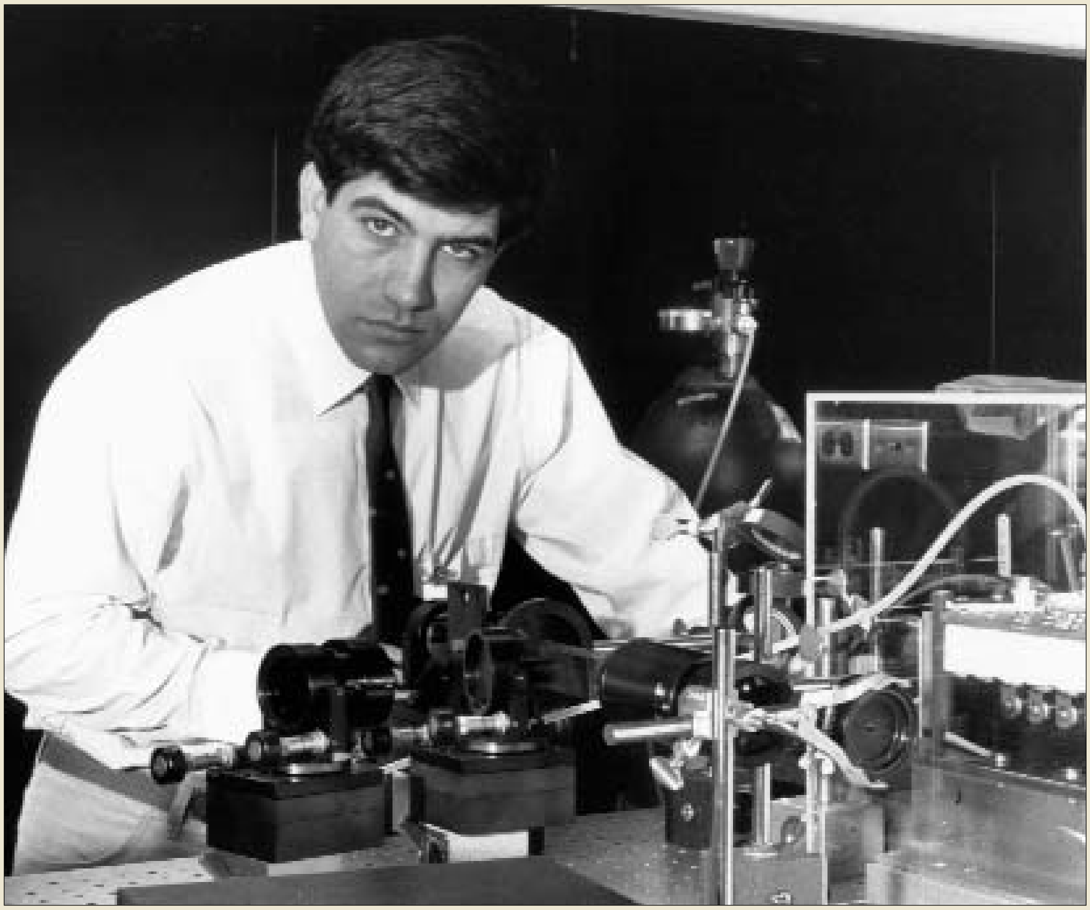
Figure 5. Multiple prism grating oscillators under high prf copper vapor laser excitation. [From Duarte and Piper.[55]]

Dispersion theory for linewidth narrowing and pulse compression

The generalized dispersion theory for multiple-prism arrays was introduced in 1982 by Duarte and Piper.[61] This theory is applicable to linewidth narrowing[61] and pulse compression.[62] The design of achromatic expanders is discussed in additional publications.[63,64] Ray matrices for propagation in multiple prism arrays are also available.[65-67]

Solid-state dye lasers: recent developments

Shortly after the discovery of liquid dye lasers, solid-state dye lasers were introduced by Soffer and McFarland[68] and Peterson and Snavely.[69] Except for sporadic work, however, it was not until the 1990s that activity in the field would be re-energized by the introduction of new and improved gain media.[70,71] In the past few years, work in this field has been



The author, around 1987, next to one of his blue-green high power excimer-laser-pumped dye laser oscillators. It was a period of intense activity in the development of high power tunable lasers.

reported by more than 20 laboratories around the world. Gain media for solid-state dye lasers include dye-doped organic matrices[72] and dye-doped organic-inorganic matrices in which the organic is a polymer.[73] A significant advantage of solid-state gain media is that, once synthesis is perfected, its production costs are relatively low. Recent advances in dye-doped organic-inorganic matrices have demonstrated better conversion efficiencies[73] and improved thermal characteristics, which lead to reduced beam divergences.[74] The reduced laser beam divergences are obtained using a dye-doped polymer-nanoparticle gain medium. The function of the nanoparticles, made of silica and uniformly dispersed in the dye-doped polymer, is to improve the dn/dT coefficient.[74] Work on crystalline laser dye gain media has also been the focus of recent research.[75, 76]

Tunable narrow linewidth solid-state dye lasers were demonstrated by Duarte[77] in 1994. An optimized multiple-prism grating oscillator has been shown to yield single longitudinal mode emission at $\Delta\nu \approx 350$ MHz in near-Gaussian temporal pulses.[78] Distributed feedback laser designs have also been developed.[79, 80]

The future of organic dye lasers

A wide range of industrial applications could benefit from the generation of cost effective tunable laser radiation in the visible. Liquid, high average power dye lasers could make a significant contribution to the field, thanks to the introduction of new or modified highly stable water soluble dyes. However, this might require advances in the design of laser dye molecules.

Although semiconductor laser excitation of dye lasers has been used for a while,[81, 82] this area offers significant promise for both liquid and condensed matter dye lasers. Thanks to the availability of high brightness light emitting diodes, work on small, low cost, solid-state narrow linewidth dye lasers should also proceed, and with a good probability of success. Direct electrical excitation of organic molecular gain media is the focus of a substantial research effort[83] that might eventually lead to a new class of solid-state dye lasers.

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