Section 3 ADVANCED TECHNOLOGY DEVELOPMENTS

3.A Synchronously Pumped, Colliding-Pulse, Mode-Locked Dye Laser

Remarkable advances in the development of ultrashort pulse dye lasers and amplifiers have been made in the past few years, permitting the study of ultrafast nonlinear phenomena on the subpicosecond time scale. In particular, the development of colliding-pulse mode-locking (CPM),¹ combined with direct control of the dye oscillator dispersion and phase modulation properties, has enabled the generation of stable optical pulses shorter than 100 fs.

Normally, the CPM laser is cw-pumped by an Argon-ion laser, and the pulses are amplified with pump pulses of duration of the order of 10 ns, usually from a Q-switched Nd:YAG laser or a copper vapor laser. There are advantages to be gained, however, by synchronous amplification with pump pulses shorter than 100 ps.^{2,3} These advantages include increased efficiency, stability, and energy contrast between the amplified pulse and amplified spontaneous emission (ASE). Amplification with short pump pulses requires a strict (~ 20-ps) synchronization between the pump and oscillator pulses. This requirement necessitates that the dye oscillator be synchronously pumped. We report here the development of a dye laser that combines the advantages of synchronous pumping, colliding-pulse mode-locking, and tunable intracavity group velocity dispersion (GVD).

A necessary condition for the dye laser to be both synchronously pumped and colliding-pulse mode-locked is that the critical position of the saturable absorber be unaffected by adjustments of the cavity length. The use of an antiresonant ring as one end mirror of a linear cavity⁴ enables this condition to be satisfied. An antiresonant ring consists of a 50% splitter and two mirrors to return the beams to the splitter. An incoming pulse from the linear part of the cavity is split into two equal parts by the 50% splitter; when the two pulses recombine on the splitter, they interfere so that the entire pulse is returned to the cavity. As suggested by Siegman⁵ and subsequently demonstrated with Q-switched Nd:YAG⁶ and Nd:glass⁷ lasers, colliding-pulse mode-locking may be achieved by placing a saturable absorber exactly halfway around the antiresonant ring from the 50% splitter.

A diagram of the dye laser is shown in Fig. 22.23. The laser consists of a four-mirror linear cavity, where one end mirror is a 5%-output coupler mounted on a translation stage, and the other is an antiresonant ring. The gain medium is a 200- μ m jet of Rhodamine 6G in ethylene glycol, and the saturable absorber is a 20- μ m jet of 3,3'-diethyloxadicarbo-cyanine iodide (DODCI) in ethylene glycol.



Fig. 22.23

Synchronously pumped, colliding-pulse, mode-locked dye laser. The laser consists essentially of a four-mirror linear cavity, where the antiresonant ring acts as one end mirror. Four Brewster prisms are included to control the intracavity dispersion. The dye laser is synchronously pumped at 100 MHz by the frequency-doubled output of a cw mode-locked Nd:YAG laser; the pump power is typically 1.5 W, and the pump pulse width is about 70 ps. Both the Nd:YAG pump laser and the dye laser are mounted on super Invar slabs to minimize cavity length fluctuations. The shortest pulses and greatest stability are obtained when the cavity lengths are properly matched and the Nd:YAG laser cavity length is adjusted to minimize the phase jitter of the Nd:YAG output. In this case, the dye laser stability is comparable to that of the pump, which is approximately 1% rms for frequency-doubled Nd:YAG.

The four Brewster prisms are placed in the linear part of the cavity to control the cavity GVD. As first demonstrated by Fork *et al.*,⁸ the

angular dispersion of the prisms causes negative GVD. The dispersion of the prism glass, however, is positive (normal). By translating one of the prisms normal to its base, as shown by the arrows in Fig. 22.23, the amount of glass in the cavity — i.e., the amount of positive GVD — may be continuously adjusted without changing the negative GVD. In this way it is possible to tune the cavity GVD through zero. This is important, because the GVD, due to dielectric mirror coatings and the gain and absorber media, becomes extremely critical for pulses shorter than about 200 fs. Furthermore, theoretical studies of ultrashort pulse lasers have shown that soliton-like pulse shaping, where positive self-phase modulation (SPM) is balanced by negative GVD, can occur in an ultrashort pulse laser, yielding shorter and more stable pulses than are otherwise obtainable.⁹

With a DODCI concentration of about 2×10^{-3} M, the dye laser output is 60 mW at 616 nm. Pulse widths as short as 70 fs can be obtained; a pulse autocorrelation is shown in Fig. 22.24. The usefulness of the Brewster prism arrangement is apparent since the minimum pulse width we have attained without the prisms is 85 fs, and the long-term stability of the laser is greatly improved.



Fig. 22.24

Autocorrelation of the dye laser output. The correlation width is 106 fs, corresponding to a real pulse width of 70 fs, assuming a sech² pulse shape.

The dependence of the laser pulse width on the net cavity GVD (i.e., on the amount of intracavity glass relative to the zero-GVD point) is displayed in Fig. 22.25. Note that not only is there a sharp minimum in the pulse width, but also the pulse shape and spectrum differ qualitatively in two different regions of the graph. When there is too little glass in the cavity, the pulse has negligible wings, the spectrum tails to the yellow, and the time-bandwidth product $\tau_p \Delta \nu$ is about 0.35. When there is too much glass in the cavity, the pulse has broad wings, the spectrum tails to the red, and $\tau_p \Delta \nu$ is about 0.5. Near the minimum of

the curve in Fig. 22.25, the spectrum is symmetric. The minimum pulse width occurs with approximately 50 to 100 μ m less glass in the cavity than with the prism position, which yields a symmetric spectrum. This is consistent with the picture of soliton-like pulse shaping mentioned above.

Future improvements to the laser will include the use of dielectric mirror coatings with controlled dispersion properties, and control of the intracavity SPM (principally through more careful control of the gain and absorber saturation characteristics).



Fig. 22.25

Pulse width versus relative prism position. The pulse width decreases to a sharp minimum and then increases as the amount of glass in the cavity is increased.

In conclusion, we have developed a synchronously pumped, colliding-pulse mode-locked dye laser that produces pulses of duration down to 70 fs suitable for synchronous amplification. The system is based on a cw mode-locked Nd:YAG laser, so that kilohertz-repetition-rate synchronous amplification is possible using a recently developed regenerative Nd:YAG amplifier.¹⁰ This oscillator-amplifier system produces ultrashort, microjoule pulses at a high repetition rate suitable for white-light continuum generation and subpicosecond time-resolved spectroscopy.

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