Passive mode locking of a cw energy-transfer dye laser operating in the infrared near 800 nm

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The cw tuning range, and subsequent passive mode locking of a new argon-ion-pumped energy-transfer dye laser operating in the near IR, is reported for the first time to our knowledge. The gain medium, consisting of an energy-transfer mixture of Pyridin 1 as the donor dye and Rhodamine 800 as the acceptor dye, has been found to lase continuous waves from 742 to 823 nm. Passively mode locked with bis-4-(1-ethylquinoline) [y-4'-(1'-ethylquinoline)] pentamethine cyanine diiodide dye, this active medium yielded sub-500-fsec pulses from 783 to 815 nm in a linear cavity, with pulses as short as 260 fsec being generated.

Since its first demonstration in 1972,\(^1\) the cw passively mode-locked Rhodamine 6G/DODCI active–passive dye combination has been shown to be a reliable source of ultrashort optical pulses, capable of directly generating pulses as short as 27 fsec (Ref. 2) in dispersion-optimized colliding-pulse ring cavities. Until 1985 passive mode locking was limited to this dye combination, which operates in the spectral region near 620 nm. Advances using various other active–passive dye combinations have extended the technique to cover most of the visible and near-IR spectral regions.\(^3\) Although a krypton-ion-pumped Rhodamine 700 dye laser has been passively mode locked in the near IR,\(^4\) a system employing an argon-ion-pumped energy-transfer dye mixture has been shown to give improved performance in the same spectral region.\(^5\) We report here the performance of a new argon-ion-pumped passively mode-locked dye-laser system that extends the covered spectral region further into the IR, to near 800 nm.

Rhodamine 800 has a structural form similar to that of Rhodamine 700, with the exception of a CN grouping attached to the 9 position of the xanthene ring instead of the CF\(_3\) group.\(^6\) With absorption coefficients and lifetime and lasing efficiencies similar to those of Rhodamine 700, an energy-transfer system employing Rhodamine 800 would be expected to give operational characteristics similar to those of the previously demonstrated Rhodamine 700 system.\(^5\)

An active medium comprising a mixture of the two dyes Pyridin 1 and Rhodamine 800 was investigated in a simple three-mirror cavity as a possible candidate to exhibit an efficient energy-transfer mechanism.\(^7\) The argon-ion pump beam (up to 7.5-W all-lines output from a Spectra-Physics model 2020 laser) was coupled into a 100-µm vertical free-flowing active dye jet (2.6 × 10\(^{-3}\) M Pyridin 1 with 2.3 × 10\(^{-3}\) M Rhodamine 800 in a 1:9 propylene carbonate:ethylene glycol mixture) using a mirror of 50-mm radius of curvature. A mirror of 100-mm radius of curvature acting as a retroreflector and another, also of 100-mm radius of curvature, formed the active folded section. The cavity was completed by a plane retroreflector. All the cavity mirrors had a single-stack dielectric coating of 100% reflectivity at normal incidence centered near 760 nm. Two output beams were provided by the reflections from the surfaces of an intracavity dielectric tuning wedge (Spectra-Physics), which was used for wavelength control.

Figure 1 shows a plot of the output power versus wavelength for a pump power of 7.2 W. A maximum of 18.5-mW average output power at 782 nm was obtained, and laser operation occurred from 742 to 823 nm. Such a generated tuning range is evidence of the

![Fig. 1. Tuning range of the cw Pyridin 1–Rhodamine 800 energy-transfer dye laser.](image-url)
Fig. 2. Schematic of five-mirror passively mode-locked dye-laser cavity.

Fig. 3. Structural formula and absorption profile of neo-cyanine. The hatched region represents the tuning range in which sub-500-fsec pulses were generated.

The new active medium was then passively mode locked in a simple five-mirror cavity (Fig. 2) configured to meet New's criteria for stable single-pulse evolution. The pump, mirror coatings, active dye mixture, tuning wedge, and jet thicknesses were as in the three-mirror cw cavity. The active folded section, however, employed tighter focusing to achieve higher efficiencies from the gain medium. Mirror \( M_1 \) was of 45-mm radius of curvature, and mirrors \( M_2 \) and \( M_3 \) were of 50-mm radius of curvature. Mirrors \( M_4 \) and \( M_5 \), which formed the second folded section around the absorber dye jet, were each of 25-mm radius of curvature. The cavity round-trip time was 7.5 nsec. One of the output beams from the tuning wedge was directed onto a fast photodiode (BPW28), while pulse-width measurements using the collinear second-harmonic-generation autocorrelation technique were performed on the other beam. Lithium iodate was used as the frequency-doubling crystal for autocorrelation, and sech\(^2\) pulse profiles are assumed throughout this research.

The principal saturable absorber investigated in this laser system was bis-4-(1-ethylquinoline) [\( \gamma -4'-(1'-ethylquinoline) \)] pentamethine cyanine diiodide. This is commonly known as neocyanine and had a measured peak extinction coefficient of \( 19.2 \times 10^4 \text{ L mol}^{-1} \text{ cm}^{-1} \) in a 1:9 ethanol:ethylene glycol solution. Figure 3 shows the structural formula and recorded absorption profile of this dye. Absorber concentrations in the laser were varied between \( 2 \times 10^{-8} \) and \( 2 \times 10^{-4} \text{ M} \), giving lasing thresholds of 2.8-6 W. Passive mode-locking action was observed for pump powers 0.1-0.3 W above threshold, and average output powers of about 1 mW per beam were obtained. Sub-500-fsec pulses were generated from 783 to 815 nm, and representative autocorrelation traces at the shortest and longest wavelengths are given in Fig. 4. The shortest pulses obtained with the system were 260 fsec at 814 nm; Fig. 5 shows an autocorrelation trace of such a pulse. In this cavity no intentional intracavity group-velocity dispersion compensation was present, and measurements of the generated spectra showed considerable excess bandwidth. For example, the 400-fsec pulses measured at 783 nm exhibited a 2.0-nm asymmetric bandwidth extending to the longer wave-
length, which, assuming sech² pulse profiles, would have been capable of supporting 320-fsec pulses.

In conclusion, we have reported a new active lasing medium consisting of an energy-transfer dye mixture of Pyridin 1 and Rhodamine 800, which has a demonstrated cw tuning range of 742 to 823 nm, excited using the all-lines pump from an argon-ion laser. This active medium has been passively mode locked using neocyanine as the saturable absorber to produce pulses of less than 500-fsec duration tunable from 783 to 815 nm, with pulses as short as 260 nm within this region. This spectral region was previously unobtainable using passively mode-locked dye-laser systems. Since the pulses have been shown to have excess bandwidth, shorter bandwidth-limited pulses could perhaps be obtained using a prism sequence⁹ to control intracavity dispersion. Initial results from a colliding-pulse mode-locked system have been encouraging.

This relatively simple source of femtosecond pulses near 800 nm may find application in measurements of the relaxation dynamics of semiconductor and doped-glass materials.

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References