Passively mode-locked continuous-wave Rhodamine 110 dye laser

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The passive mode locking of a cw Rhodamine 110 dye laser is reported. In a simple uncompensated linear cavity, subpicosecond pulse generation was achieved over the spectral range 553-570 nm. Pulses as short as 150 fsec were recorded using standard second-harmonic autocorrelation measurement techniques. Two saturable-absorbing species were found to operate successfully in this region.

The passively mode-locked cw dye laser is the source that provides the temporally shortest pulses. In addition to the high amplitude stability typical of this system, there is considerably lower interpulse jitter than in synchronously or hybrid mode-locked cw dye lasers. However, since its inception in 1972, all work has been limited to the spectral region around 615 nm, primarily using a single active-passive dye combination, that of Rhodamine 6G mode locked by DODCI and resulting in the recent attainment of 27-fsec pulses from an optimized and dispersion-compensated cavity. The majority of subpicosecond spectroscopy has correspondingly been confined to the excitation spectral region around 615 nm. Therefore any alternative passively mode-locked systems at additional wavelengths should find immediate application as well as yielding further insights into the pulse-formation mechanism.

We recently reported the generation of 350-fsec pulses from a passively mode-locked Rhodamine 700 cw dye laser at 740 nm in the near infrared. Here we report a further extension of passive mode locking to Rhodamine 110, achieving subpicosecond pulse generation tunable over the range 553 to 570 nm, with highly stable pulses as short as 152 fsec being obtained from a simple linear cavity with no dispersion compensation. Two saturable-absorbing species were found to operate successfully over the above spectral range, and both are reported here.

The experimental arrangement is shown in Fig. 1. A five-mirror cavity arrangement was used, which complied with the criteria laid down by New for stable single-pulse evolution. The active-medium folded section comprised two mirrors (M2 and M3) of 100-mm radius of curvature about a vertical jet stream, while the passive dye jet section consisted of a focusing mirror M4 of 50-mm radius of curvature and a retroreflecting mirror M5 of 25-mm radius of curvature. All the cavity mirrors had broadband dielectric coatings (100% reflectivity at 500-800 nm), wavelength selection was achieved using a dielectric-coated tuning wedge, and there was no dispersion compensation in the cavity. The dye laser was pumped by the all-lines output of a Spectra-Physics argon-ion laser (model 2020). The all-lines output was coupled into the gain medium (1.5 \times 10^{-3} M Rhodamine 110 in ethylene glycol) by a focusing mirror M1 of 50-mm radius of curvature. In these initial tests, since all mirrors had 100% reflectivity, two output beams were derived using reflections off the tuning wedge (TE). One was employed to monitor the laser output using a fast photodiode (BPW 28) in conjunction with a sampling oscilloscope, while the other facilitated pulse-width measurement. A standard collinear second-harmonic-generation autocorrelation technique employing KDP (0.7 mm thick) was used to measure the duration of the dye-laser pulses. Optimization of the output pulse widths was carried out using a scanning autocorrelator, while hard copies of the autocorrelation trace were taken on a single slow scan. In the work reported here sech^2 pulse shapes were assumed throughout.

Two passive mode-locking dyes were used. The first was 2-(p-dimethylaminostyryl)-benzthiazolyl-ethyliodide (DASBTI), which we previously showed to mode lock Rhodamine 6G in the region 570-600 nm. The second dye used was 1,1,3,3,3',3'-hexamethyldocarbocyanine iodide (HICI). The absorption profiles for 3:1 ethylene glycol:ethanol solutions of the dyes are shown in Fig. 2 together with their molecular structures and the pertinent mode-locking range. Peak extinction coefficients of 11 \times 10^4 liters mol^{-1} cm^{-1} and 7.9 \times 10^4 liters mol^{-1} cm^{-1} at 543 and 538 nm were recorded for HICI and DASBTI, respectively.

Fig. 1. Experimental cavity configuration.

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Using passive dye concentrations of the order of $1.5 \times 10^{-4}$ M, subpicosecond pulses were obtained over the range 553–570 nm for each dye. The autocorrelation traces obtained showed the 3:1 contrast ratio indicating complete mode locking. Figure 3 shows autocorrelation traces taken at 553 and 567 nm, practically the extremes of the tuning range of the HICI and DASBTI absorbers, recording pulses of 390 and 443 fsec, respectively. The laser was observed to perform best near the peak of the gain around 560 nm. Figure 4 shows a typical result of 210 fsec obtained using DASBTI as the saturable absorber at 561 nm and the shortest pulse duration obtained of 152 fsec using HICI at 561 nm. To the authors’ knowledge, this is the shortest pulse ever directly generated from a simple linear cavity with no dispersion compensation, although Diels et al. have produced 0.12-ps pulses at 615 nm by external synchronization of relaxation oscillations in a passively mode-locked dye laser. This result may be explained in part by the fact that the ratio of the saturable-absorber cross section to the emission cross section of the gain medium is much higher than for the standard Rhodamine 6G/DODCI combination—leading to a higher value of the $s$ parameter defined by New. The $s$ parameter, which basically is a ratio of the intracavity light flux absorber saturation to gain saturation and gives a measure of the ability of the laser to form discrete stable pulses, had a value two to three times that of the Rhodamine 6G/DODCI system in a similar linear cavity configuration, depending on the operational wavelength.

A spectral investigation revealed that the pulses were not transform limited (the 2.7-nm measured bandwidth should be able to support a 122-fsec pulse). The observed chirp may be due to the broadband double-stack dielectric mirror coatings used.

With each of the saturable absorbers, the optimized laser was generally pumped about 200 mW above threshold (typically 3–4 W all-lines, depending on wavelength), and 25 mW output powers were achieved. When DASBTI was used the laser was non-self-starting when operated just above (~50 mW) threshold and the mode-locking degraded if the pump level was more than 300 mW above threshold. With HICI, however, it was possible to pump well above threshold without significantly affecting the mode locking. Using an HICI concentration of $2 \times 10^{-4}$ M, the threshold was 5 W, and for 7-W pump power a total of 70 mW of laser output was obtained while the pulse duration was of the order of 250 fsec, corresponding to a peak power of 2.8 kW. This stable, short-pulse operation substantially above threshold is in marked contrast to that of other cw passively mode-locked systems.

In conclusion, we have demonstrated the cw passive mode locking of Rhodamine 110 with the saturable...
Fig. 4. Autocorrelation traces of Rhodamine 110 passively mode-locked output pulses at 561 nm: (a) typical trace with DASBTI saturable absorber, (b) shortest pulse recorded with HICI.

absorbers DASBTI and HICI, tunable over the range 553–570 nm. We are confident that in a dispersion-compensated cavity, pulses of less than 100 fs should be readily achievable over this tuning range. We have shown the potential of highly stable pulse trains of low interpulse jitter, high variable repetition rate, and significant average power, suitable for sophisticated pump-and-probe experiments and possibly for pulse compression. This is currently under investigation, as is the extension of purely passive mode locking to further regions of the spectrum.

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References