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Quasi-linear ring colliding-pulse mode-locked femtosecond laser using binary energy-transfer gain dye mixture

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A new method for femtosecond pulse generation using a binary energy-transfer dye mixture for the gain medium in a novel linear ring colliding-pulse mode-locked dye-laser configuration is reported. Pulses of 30-fsec duration and 18.7-nm spectral width are routinely generated.

A passively colliding-pulse mode-locked (CPM) dye laser[1] is the main source of optical pulses in the femtosecond region. Much theoretical and experimental research has been performed to understand the physics underlying femtosecond pulse generation, such as balancing the self-phase modulation[2] and group-velocity dispersion (GVD) in the laser cavity. Diels *et al.*[3] have used an intracavity glass prism to compress pulses, while a four-prism sequence was used by Valdmanis *et al.*[4] A Gires–Tournois interferometer has been used alone or incorporated into a four-prism sequence to improve chirp compensation.[5,6] Two mirrors in the cavity could be used for adjusting the intracavity second-order dispersion $\ddot{\phi}(\omega)$ without any additional optical elements.[7] Considerable pulse shortening has also been achieved by adding an organic material of high nonlinear refractive index to the saturable absorber solution.[8] It has been suggested that additional pulse shortening could be achieved if the emission wavelength were shifted further into the red and that this may be accomplished by using lower transmission of the output mirror to reduce the losses in the cavity.[5] The desired red shift has also been accomplished using a spatial filter in the four-Brewster-angled-prism sequence.[9] These methods are limited by a bandwidth of less than 16 nm. To extend the laser bandwidth, energy transfer between dye mixtures through Forster dipole–dipole interaction could be utilized.[10,11]

In this Letter we report on femtosecond pulse generation using a binary gain dye mixture of Rhodamine 590 and Kiton Red, whose absorption spectrum partly overlaps the emission spectrum of Rhodamine 590, in a new cavity design. The fluorescence spectrum of the mixture shifts to the red, improving the CPM laser performance characteristics for short-pulse generation. The new cavity design consists of a quasi-linear CPM ring. All this results in better intracavity chirp compensation and a pulse duration of 30 fsec. By these methods a bandwidth of 18.7 nm is achieved, which exceeds the best results so far by 17%, and it has a symmetric shape, which indicates good chirp compensation.[12]

It has been shown that the phase shift $\ddot{\phi}(\omega)$ of the wave reflected from a dielectric mirror is more pronounced for laser frequencies further from the central frequency and for angles of incidence away from the angle for which a quarter-wave layer mirror is coated.[13] In our laser the mirror incidence angles have been greatly reduced, resulting in almost normal incidence. Thus we have avoided astigmatic distortions introduced by oblique angles of incidence that limit the performance of the system.[14] For multilayer dielectric mirrors, the dependence of the reflection coefficient on the angle of incidence is smallest for normal incidence. With the configuration discussed above we have minimized this dependence. This is important since small deviations of the incidence angles from the optimum for which the mirror has been coated for cause chirping of the pulse for frequencies at the bandwidth edge. This effect becomes significant when the bandwidth is large.[13,15]

A schematic of the near-linear six-mirror ring cavity (the angles are not to scale) is shown in Fig. 1. Two Z folds are employed around the saturable absorber jet and gain jet with angles of incidence of less than 2°.

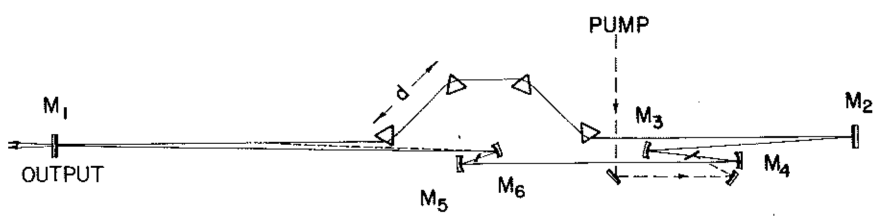

Fig. 1. Schematic diagram of the linear ring cavity CPM dye laser employing an almost-normal incidence angle. The dashed line represents the possibility of adjusting the cavity length, which is easily achieved by moving mirror M_1 and making a minor adjustment to mirror M_6.

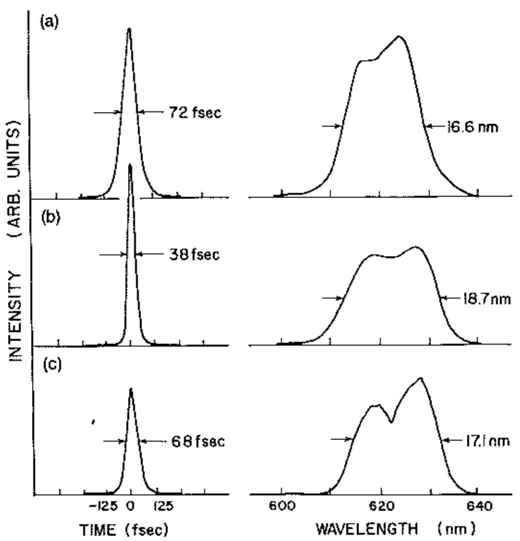

Fig. 2. Autocorrelation functions and their corresponding spectra for three different Kiton Red concentrations in a Rhodamine 590–ethylene glycol solution (Rhodamine 590 concentration of 1.6×10^{-3} *M*). (a) No Kiton Red, (b) 3.9×10^{-5} *M* Kiton Red, (c) 7.1×10^{-5} *M* Kiton Red.

The radius of curvature of the absorber jet mirrors is 5 cm, while for the gain jet a combination of one 1.27-cm- (0.5-in.-) diameter mirror and two half-mirrors (each with a radius of curvature of 10 cm) is employed to obtain a minimum incidence angle. The incidence angles at the end mirrors (mirrors M_1 and M_2) are less than 0.5°. In addition to reducing the possible introduction of uncompensatable nonlinear dispersion, this small angle has enabled us to have a cavity with adjustable length, making it possible to operate the laser at the repetition rate between 82 and 127 MHz. The distance between the end mirrors can be reduced from 1.6 to 1.1 m, requiring only small adjustment of one of the absorber jet mirrors (mirror M_6). All the cavity mirrors are single-stack 19-layer dielectric mirrors with resonance wavelength at 625 nm at normal incidence. The output mirror M_1 has a 3% transmission for optimum operation at the shortest pulses. 3,3′-Diethyloxadicarbocyanine iodide (DODCI) was used as the standard saturable absorber dye, and ethylene glycol was used as the solvent. A mixture of Rhodamine 590 and Kiton Red was used in the gain jet and has produced the best results. The saturable absorber jet thickness is 50–70 μm, and the gain jet thickness is approximately 120 μm. The amount of negative GVD in the cavity is controlled by a four-prism sequence.[16]

The laser output was monitored by a standard autocorrelator driven by a shaker. To minimize the material dispersion in the path of the pulse to be measured, a 0.1-mm-thick KDP crystal was used for second-harmonic generation, and a pellicle beam splitter was employed. The usual assumption of a squared-hyperbolic-secant pulse shape was used in calculating the pulse duration. The typical pump power that gave the shortest output pulses ranged from 2 to 2.3 W depending on the concentration of the gain medium. The cavity path length used for this experiment was 3.66 m, while the average output power for optimum performance was 40 mW. This gave an intracavity pulse energy of 16 nJ.

Laser-pulse autocorrelation functions and corresponding spectra taken as a function of Kiton Red concentration in the gain mixture are displayed in Fig. 2. We started with an optimum concentration of DODCI (6.3×10^{-5} *M*) for pure Rhodamine 590 in ethylene glycol with a concentration of 1.6×10^{-3} *M*. The DODCI concentration was kept constant throughout the measurements. The concentration of Kiton Red in the mixture was increased in small quantities of 7.9×10^{-6} *M* (0.5% of that of Rhodamine 590 starting from no Kiton Red). Minor adjustment of the laser alignment was necessary from time to time to maintain stable conditions as the gain dye concentration was changed. The output power was kept at 40 mW by decreasing the pumping power from 2.6 to 2.0 W. The bandwidth increased and the pulse duration simultaneously decreased to a certain point [Fig. 2(b)]. Further increases in the concentration decreased the bandwidth and increased the pulse width. This, and the fact that the laser exhibits stable operation for net negative GVD (the pulse broadens when negative GVD is added) while the pulse abruptly collapses for net positive intracavity GVD, is an indication of the solition pulse-shaping mechanism.[4,5] The best results

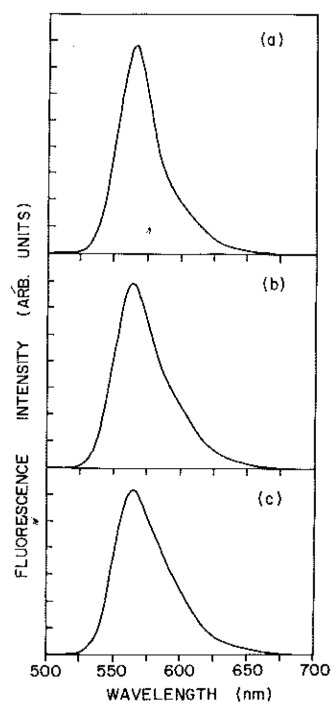

Fig. 3. Fluorescence spectra of the binary energy-transfer gain dye mixture corresponding to the data in Fig. 2. The increase in concentration of Kiton Red in the Rhodamine 590 solution is manifested by a lower peak intensity as more of the Rhodamine 590 fluorescence is absorbed by Kiton Red and reemitted further into the red. (a) Rhodamine 590 (concentration of 1.6×10^{-3} *M*), (b) mixture of Rhodamine 590 (concentration of 1.6×10^{-3} *M*) and Kiton Red (concentration of 3.9×10^{-5} *M*), (c) mixture of Rhodamine 590 (concentration of 1.6×10^{-3} *M*) and Kiton Red (concentration of 7.1×10^{-5} *M*).

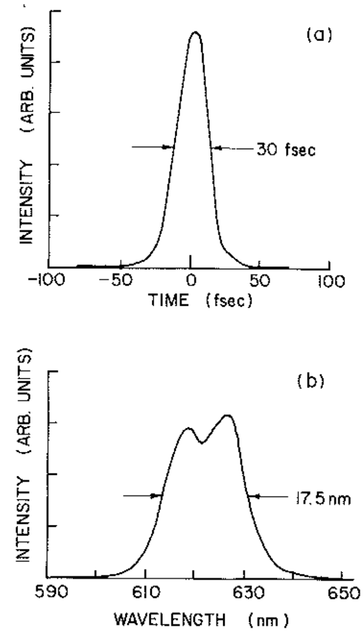

Fig. 4. (a) Autocorrelation trace of pulses having 30-fsec duration and (b) corresponding spectrum obtained for optimum Kiton Red–Rhodamine 590 solution when DODCI concentration was also optimized (DODCI concentration of 1.3×10^{-4} *M*).

were obtained with the mixture of 1.6×10^{-3} *M* of Rhodamine 590 and 3.9×10^{-5} *M* of Kiton Red (2.5% of the Rhodamine 590 concentration). The bandwidth of 18.7 nm is, to our knowledge, the largest achieved in CPM laser stable operation. This invites the possibility of pulses shorter than the corresponding 38 fsec, if all the laser parameters were optimized.

Figure 3 displays the corresponding change in fluorescence spectra of the binary energy-transfer gain dye mixture for the results presented in Fig. 2. The increase in concentration of Kiton Red in Rhodamine 590 solution is manifested by a lower peak intensity. As more of the Rhodamine 590 fluorescence is absorbed by Kiton Red, it is reemitted further into the red where the fluoresence is seen to increase.

A similar intracavity pulse compression experiment was carried out using the binary energy-transfer dye mixture while the DODCI concentration was changed. The net intracavity GVD was monitored by moving one of the prisms normal to its base. The results show that pulses as short as 30 fsec are obtained for a DODCI concentration of 1.3×10^{-4} *M* at a pumping power of 2.0 W (Fig. 4).

It has been shown that an average positive chirp is generated for pulses at energies greater than 10 nJ and shorter than 70 fsec, owing to the prevailing contribution of the positive chirp from the nonlinear refractive index of the solvent over the negative chirp from the absorber saturation. The negative chirp from the absorber saturation shifts to the beginning of the pulse as the saturation parameter $\gamma = E_o/E_s$ increases.[17] This and the fact that the positive chirp from the solvent increases with pulse energy are considered to have a special effect in our experiment. Our DODCI concentration is approximately 10 times smaller than the optimum concentration used by Jacobovitz *et al.*,[18] which increases γ and makes the negative chirp considerably less pronounced by shifting it to the beginning of the pulse, thus affecting an even smaller part of it. At the same time the positive chirp due to the nonlinear refractive index of the solvent is the same as in the study by the same group of Miranda *et al.*[17] Since the part of the chirp from the DODCI jet is reduced, this results in a more successful intracavity compensation. This was possible because high concentrations of DODCI were not necessary to shift the lasing spectrum into the red.

In summary, we have described a uniquely designed stable femtosecond dye laser that generates pulses as short as 30 fsec using a mixture of Rhodamine 590 and Kiton Red in ethylene glycol as the gain medium. Both the design and the gain mixture contribute to reducing the multiple sources of chirp while at the same time considerably broadening the spectral width. The bandwidth of 18.7 nm is to our knowledge the highest obtained so far and could result in a 22-fsec pulse width if transform-limited pulses were achieved.

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