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Generation of 36-fsec pulses near 775 nm from a colliding-pulse passively mode-locked dye laser

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We report the passive mode locking of a cw Rhodamine 700 dye laser using the saturable absorber DDI. Pulses as short as 36 fsec near 775 nm have been produced in a colliding-pulse mode-locked dye laser with adjustable intracavity group-velocity dispersion.

The passively mode-locked cw dye laser has been proven to be a reliable source of ultrashort optical pulses. By combining the colliding-pulse mode-locked (CPM) technique[1] and intracavity group-velocity dispersion (GVD) control,[2] pulses as short as 27 fsec (Ref. 3) and 19 fsec (Ref. 4) have been produced. Until recently, however, all the passively mode-locked systems have used the active–passive dye combination of Rhodamine 6G and DODCI, operating in the spectral region near 620 nm. In recent years several active–passive dye combinations have been studied, yielding subpicosecond pulses from 550 to 810 nm and from 487 to 508 nm.[5,6] Passive mode locking of Rhodamine 700 in the 730–760-nm range using linear and ring cavities with several saturable absorbers such as DDI, cryptocyanine, DOTCI, and HITCI has been reported.[7,8] The shortest pulse duration obtained in this spectral range is 110 fsec at 750 nm. Langford *et al.* have also reported the generation of femtosecond laser pulses (180 fsec) in the near-infrared spectral region (~850 nm) by the passive mode locking of a color-center ($LiF:F_2^+$) laser[9] in a GVD-compensated configuration. In addition, there is currently great interest in the production of optical pulses with very high peak powers (>1 TW). Terawatt subpicosecond pulses have already been obtained by chirped-pulse amplification in Nd:glass[10] and in complex systems based on dye and excimer amplifiers.[11] However, these systems suffer from the lack of ultrashort pulse sources at the amplifier wavelength. With the recent development of new solid-state laser materials such as alexandrite and $Ti:Al_2O_3$, which have amplification bandwidths larger than the femtosecond pulse spectra, the generation of sub-100-fsec pulses in the near infrared is interesting.

We report here the generation of pulses as short as 36 fsec near 775 nm in a CPM ring cavity with adjustable intracavity GVD. These pulses are the shortest, to our knowledge, ever produced in a passively mode-locked dye laser with a dye other than Rhodamine 6G.

The active dye used in this system was Rhodamine 700 (concentration of 2×10^{-3} *M* in ethylene glycol), which provided good efficiency near 750 nm when pumped with all-red lines (647 and 676 nm) from a krypton-ion laser (Spectra-Physics Model 171). The efficiency of this dye was similar to that of Rhodamine 6G pumped by an argon-ion laser. (The alternative method of exciting Rhodamine 700 is to use an energy-transfer dye mixture[12] pumped by an argon-ion laser. We have tried this method without success because the gain provided was low compared with that of the other solution using a krypton-ion pump laser.) The experimental configuration is shown in Fig. 1. A classical six-mirror ring cavity, similar to that developed by Valdmanis *et al.*,[3] was used. The krypton-ion laser was coupled into the gain medium by a 100-mm radius-of-curvature focusing mirror. Mirrors M1 and M2 had 150-mm radii of curvature, mirrors M4 and M5 had 50-mm radii of curvature, and mirror M3 was plane. A plane output coupler M6, with a 1.5% transmission ratio near 770 nm, provided two output beams. The two jets were horizontal; their thicknesses were approximately 200 μm for the amplifying medium and 100 μm for the saturable absorber. A sequence for four fused-silica Brewster-angled prisms in a vertical plane[4] was used to adjust the intracavity GVD. To reduce the high-order dispersion in the cavity, all the mirrors (except the output coupler) had single-stack dielectric coatings of 100% reflectivity for normal incidence centered at 750 nm. The cavity round-trip time was 10 nsec. The pulse duration was measured using a classical background-free second-harmonic generation autocorrelator with a 300-μm-thick KDP doubling crystal. By using a 50-μm-thick KDP crystal instead of the previous crystal, we have measured the same pulse duration (of the order of 40 fsec). Thus we can say that the thickness of our KDP crystal was not a limitation for the pulse duration measurement. Sech[2] pulse shapes have been as-

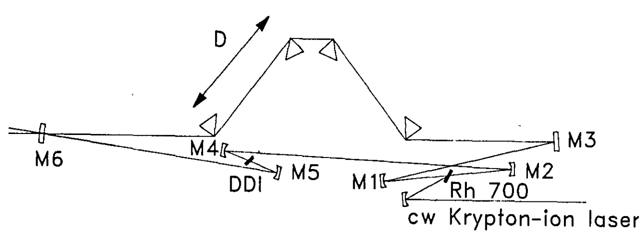

Fig. 1. Schematic of the CPM ring configuration. Rh, Rhodamine.

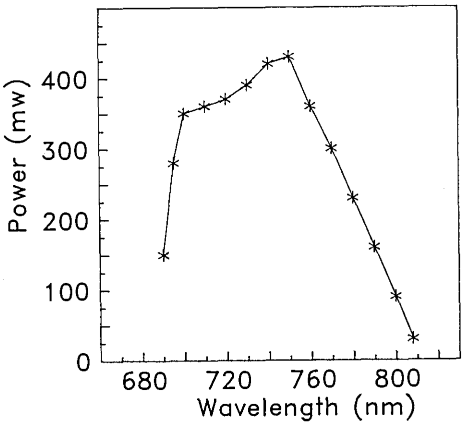

Fig. 2. Tuning range of the cw Rhodamine 700 dye laser pumped with 3-W all-red lines from a krypton-ion laser.

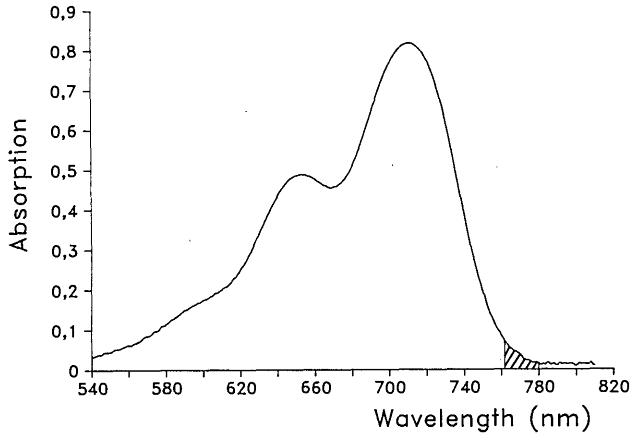

Fig. 3. Absorption profile of DDI. The hatched region represents the bandwidth in which sub-50-fsec pulses were obtained.

sumed. Spectral information was recorded using an optical multichannel analyzer (EGG-PAR OMA III) coupled to a 0.25-m spectrometer. Without a saturable absorber in the cavity, the lasing threshold at 760 nm was 1.1 W. Figure 2 shows a plot of the cw output power versus the wavelength for a pump power of 3 W. A maximum power of 430 mW at 750 nm and a tuning range from 700 to 800 nm were obtained using an intracavity tuning wedge.

The saturable absorber used was DDI, which has a peak extinction coefficient of $20 \times 10^4\ \mathrm{L \cdot mol^{-1} \cdot cm^{-1}}$ at 710 nm for a 1:10 solution of propylene carbonate/ethylene glycol (Fig. 3). We have found that propylene carbonate is the best solvent to dissolve DDI powder. Regardless of the DDI concentration used in the passive mode-locked regime, the laser wavelength was near 775 nm. We have used concentrations between 5×10^{-4} and 10^{-3} M, which give lasing thresholds of 2–3 W. The distance D between the prisms (see Fig. 1) was increased from 300 mm in the Rhodamine 6G/DODCI configuration to 355 mm in this configuration. This fact can be understood by looking at the variation of the negative GVD introduced by the sequence of four fused-silica prisms with the wavelength (Fig. 4). For a fixed distance between the prisms and the same glass pathway, the negative GVD decreases when the wavelength increases. In other words, we must increase the distance D between the prisms to get the same negative GVD. We had already observed such behavior in a CPM laser working at 685 nm.[6] It has been proven that the high-order dispersion in the cavity is one of the main limitations to producing short pulses in CPM dye lasers.[13] The use of single-stack dielectric coatings, which strongly decrease the third-order dispersion, has allowed the reduction of the pulse duration.[3] Figure 4 shows that the absolute value of the third-order dispersion introduced by the prisms (which is the derivative of the curve shown) decreases when the wavelength increases. This means that prisms' high-order dispersion will probably not be a limitation for the generation of short pulses in the mid-infrared or infrared.

By adjustment of the intracavity dispersion, the laser operated routinely below 45 fsec and pulses as short as 36 fsec were obtained (assuming a sech^2 pulse shape). Figure 5 shows the autocorrelation and the corresponding spectrum of the shortest pulses produced. The spectrum, centered at 775 nm, had a 18.5-nm width, which gave a time–bandwidth product of $\Delta t \Delta \nu = 0.333$. For this result the lasing pump threshold was 3.2 W, and the laser operated 200 mW above

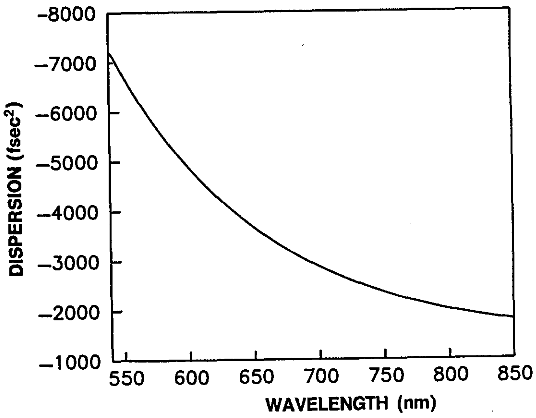

Fig. 4. Theoretical plot of the negative GVD versus the wavelength introduced by a sequence of four fused-silica Brewster-angled prisms for a fixed distance $D = 0.3$ m (see Fig. 1) and a total glass pathway $E = 10$ mm.

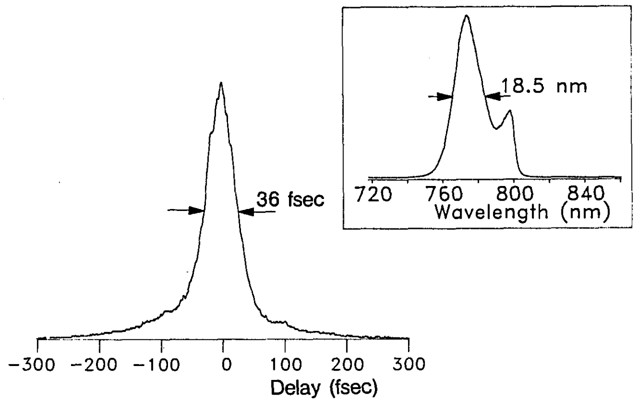

Fig. 5. Autocorrelation trace and the corresponding spectrum of the shortest pulses obtained from a Rhodamine 700/DDI CPM dye laser.

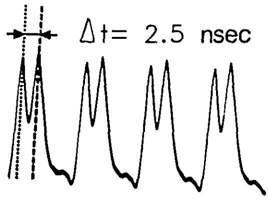

Fig. 6. Oscilloscope trace showing the pulse train.

the threshold, which yielded output powers of up to 7 mW per beam. The autocorrelation trace shows some wings, indicating that the pulses are chirped and cannot be described by a sech^2 function. The spectrum exhibits a double-peak shape, which must introduce a modulation in the temporal profile, and then wings in the autocorrelation. Furthermore, before reaching the KDP the pulse travels through the output coupler of the laser, the beam splitter, and the lens of the autocorrelator. The chirp introduced by the glass may also explain the wings in the autocorrelation. By translating a prism to be in excess of positive dispersion beyond the value corresponding to the shortest pulses produced, we have not observed triple-humped autocorrelation traces as has been previously reported.[5,14,15] With more positive dispersion, the pulse duration increased while the spectrum shortened and shifted to the red wavelengths.

When the laser operated in a stable passive mode-locked regime, regardless of the pulse duration, the cavity supported four pulses (two clockwise and two counterclockwise). Using a fast photodiode we have observed that the energy per beam is equally distributed among two pulses and that they are separated in time by 2.5 nsec (which corresponds to one fourth of the cavity round-trip time and also to the distance between the gain medium and the saturable absorber) (Fig. 6). With such a temporal distribution the two additional pulses also collide in the saturable absorber 2.5 nsec later than the two first pulses. Moreover, each pulse arrives in the gain medium separated by 2.5 nsec. The four pulses are then equally amplified. By decreasing the pump power we have not been able to find a stable mode-locked regime with only two pulses in the cavity. We think that this behavior can be explained by the relatively fast gain recovery time of Rhodamine 700. That means that it should be possible to reduce the cavity round-trip time to approximately 6 nsec in order to have only two pulses in the cavity.

We have seen that the spectrum of the pulses is centered at 775 nm. At this wavelength the absorption of the saturable absorber is low, so we must use high concentrations (of the order of 10^{-3} *M*), which give a pump power threshold of 3 W. To obtain four stable pulses, the pump laser operated only 200 mW above the threshold. With more pump power two additional pulses arrive in the cavity and affect the stability and the pulse duration. Consequently the dye laser was a little more sensitive to fluctuations than in the Rhodamine 6G/DODCI configuration. However, as noted above, we have not observed high-order soliton behavior as seen in lasers operating at other wavelengths. The evolution of the pulse duration with intracavity dispersion does not present a discontinuity near the minimum duration. Therefore the stability remains good even for the shortest pulses. Although we have produced short and stable pulses, we are now trying to find another saturable absorber that would allow the generation of such short pulses, but with the absorption profile moved further toward longer wavelengths.

In conclusion, we have reported the passive mode locking of a cw Rhodamine 700 dye laser using DDI as the saturable absorber. Using a dispersion-compensated CPM dye laser, pulses as short as 36 fsec have been generated near 775 nm. These pulses are, to our knowledge, the shortest ever produced in a CPM dye laser in the near infrared.

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