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Femtosecond pulses at 800 nm by passive mode locking of Rhodamine 700

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We report the passive mode locking of a Rhodamine 700 dye laser in the near infrared. Using a short cavity in order to avoid multiple pulses and by adjusting the intracavity dispersion, we have produced 50-fsec pulses at 800 nm with HITCI as the saturable absorber.

It is now well established that cw passive mode locking with an intracavity group-velocity dispersion compensation is the most interesting technique for producing short and stable femtosecond pulses.[1-3] However, passive mode locking was, until recently, restricted to the single active–passive dye combination of Rhodamine 6G/DODCI that yielded pulses in the spectral region near 620 nm. To extend the spectral range of the femtosecond pulses, expensive and complex amplifiers have been developed to produce a spectral continuum of ultrashort pulses. On the other hand, several active–passive dye combinations and color-center lasers have been studied in order to obtain subpicosecond pulses from 487 to 850 nm.[4,5] We recently reported the passive mode locking at 775 nm of a cw Rhodamine 700 krypton-ion-pumped dye laser using DDI as the saturable absorber.[6] In this Letter we present a further extension of the passive mode locking of Rhodamine 700 near 800 nm by using another saturable absorber.

The active medium was Rhodamine 700 (2×10^{-3} M in ethylene glycol), which provided good efficiency near 760 nm and a tuning range of 700–810 nm. Figure 1 shows the cavity configuration that yielded femtosecond pulses. It is based on the design of Valdmanis *et al.*[3] and is similar to the cavity that generated pulses of 36-fsec duration near 775 nm from the Rhodamine 700/DDI laser reported in Ref. 6. Mirrors M1 and M2 had 150-mm radii of curvature, and mirrors M3 and M4 had 50-mm radii of curvature, with the other mirrors (M5, M6) being plane. A 100-mm radius-of-curvature mirror focused the pump radiation of a krypton-ion laser into the active dye jet, which was ~200 μm thick. All the cavity mirrors, excepted the output coupler, supported single-stack dielectric coatings of 100% reflectivity. The transmission of the output coupler was approximately 1.5% near 800 nm. The distance D between the prisms was increased from the 300 mm of the Rhodamine 6G/DODCI configuration to 380 mm. This is due to the decrease of the negative group-velocity dispersion introduced by the prisms as the wavelength increases.[6] Pulse duration was measured by using a standard noncollinear second-harmonic-generation autocorrelator with KDP as the doubling crystal and by assuming a sech^2 profile. Spectra were recorded using a 0.25-m spectrometer coupled with an optical multichannel analyzer.

The saturable absorber used was HITCI, which has an absorption cross section of 10^{-15} cm^2 at 760 nm for an ethylene glycol solution (Fig. 2). Passive mode locking was observed for concentrations from 3×10^{-5} to 10^{-4} M, which gave lasing thresholds between 3 and 4 W. The best results in stability and pulse width were obtained for high concentrations. With a classical cavity round-trip time of 10 nsec, four stable pulses were observed: two clockwise and two counterclockwise. Owing to the distance between the saturable absorber and the gain medium (a quarter of the cavity round trip), the two additional pulses collided in the saturable absorber. Moreover, the four pulses were equally amplified in the amplifying medium because they arrived every quarter of the cavity round-trip time. We think that this multiple-pulse behavior is due to the fast gain recovery time of Rhodamine 700. Such behavior has already been observed in our Rhodamine 700/DDI femtosecond dye laser[6] and by Knox[7] with Rhodamine 800 as the amplifier medium (which is a dye similar to Rhodamine 700). In order to avoid these multiple pulses, we have reduced the cavity round-trip time to 7.2 nsec, which is close to the minimum cavity length possible if we take into account the

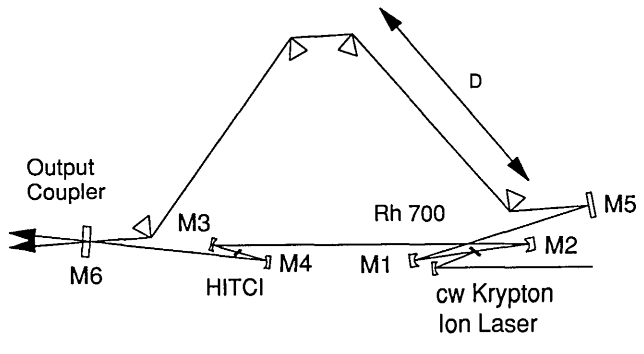


Fig. 1. Laser configuration.

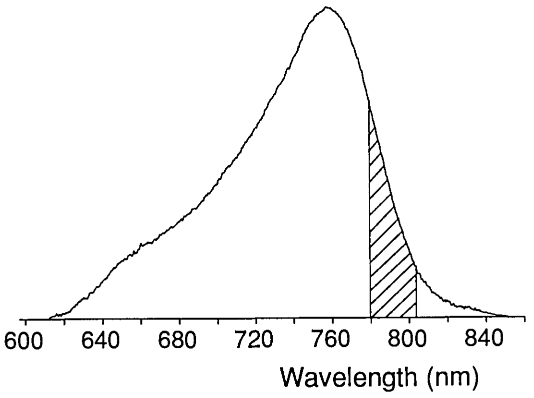


Fig. 2. Absorption profile of HITCI. The hatched region represents the bandwidth in which the pulses were obtained.

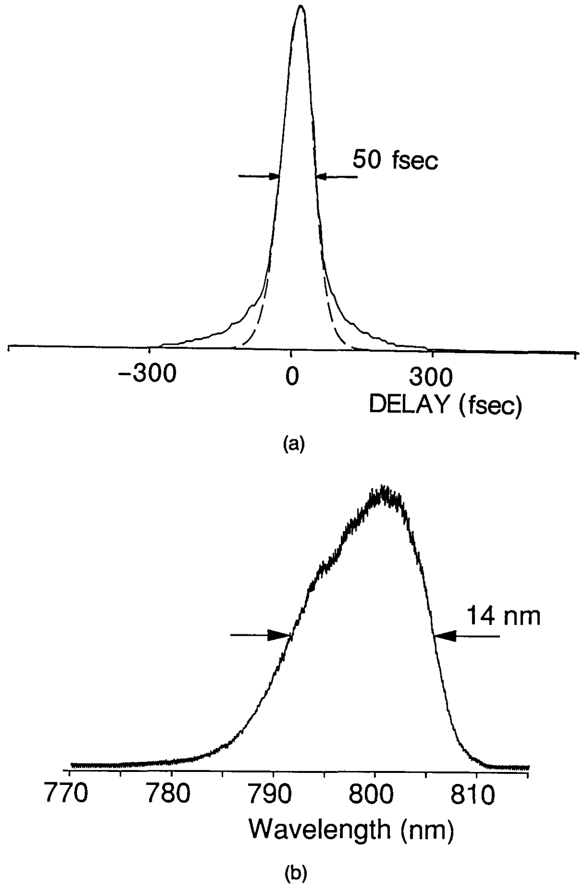
overall dimensions of the sequence of prisms and mirrors mounts. When the distance between the gain medium and the saturable absorber was maintained at a quarter of the cavity length, we always observed four pulses. However, it was possible to obtain only two pulses just above the lasing threshold, but this behavior was not stable and either the laser stopped or four pulses appeared again. To make these additional pulses disappear, we decided to decrease the distance between the two dye jets. We expected that the gain would be asymmetrical and that two of the four pulses would be less amplified. Since the saturable absorber is saturated only for an intensity corresponding to the coherent sum of the two-pulse amplitudes, this asymmetry would suppress the two additional pulses. In fact, we have decreased the distance between the saturable absorber and the amplifier medium from 540 to 400 mm and have observed that the laser operated in a stable regime with only two pulses in the cavity. Of course, with a pump power greater than 300 mW above the threshold the double-pulse regime appeared again. Moreover, the two-pulse behavior was easier to obtain when the laser spectrum was shifted to the red by moving the absorber dye jet away from the focus point.

In these conditions, and by adjusting the intracavity group-velocity dispersion, the laser routinely operated below 60 fsec, and pulses as short as 50 fsec were obtained (with the assumption of a sech^2 pulse shape). Figure 3 shows the autocorrelation and the corresponding spectrum of the shortest pulses produced. The spectrum, centered at 800 nm, had a 14-nm width [Fig. 3(b)] and presented a relative symmetrical shape. For a laser pump power 300 mW above the threshold, the laser was stable over hours, and two output beams of 20-mW power were available. With the 140-MHz repetition rate, this corresponds to a peak pulse power of approximately 2.5 kW. As in the visible configuration, the shortest pulses for a saturable-absorber jet were obtained not at the focus point of the passive folded section but for a position that tended to shift the pulse spectrum more into the infrared. For the absorber jet at the focus point, the shortest pulses obtained had a duration of 75 fsec and a spectrum centered at 795 nm.

When the laser operated at the shortest pulses with a spectrum shifted more into the red, and when we translated a prism to be in excess of positive group-velocity dispersion, no soliton behavior, as reported previously,[8] was observed. We only observed that the pulse broadened and the spectrum became symmetrical and shifted toward the long wavelengths. We think that this behavior is due to the spectrum filtering introduced by the dielectric coatings of the cavity mirrors whose cutoff wavelength is at 810 nm.

The stability of the laser is better than in the version with DDI as the saturable absorber. In fact, the use of an absorber (HITCI) with an absorption profile shifted into the infrared increases the efficiency of the mode locking.

In conclusion, we have reported the passive mode locking of a cw Rhodamine 700 dye laser with HITCI as the saturable absorber. In a dispersion-compensated ring cavity, pulses as short as 50 fsec have been produced near 800 nm. The system is simpler than the laser developed by Knox,[7] which consists of a colliding-pulse mode-locked dye laser pumped by a cw Ti:Al_2O_3 argon-pumped laser. Moreover, their performances are similar. We think that this infrared femtosecond laser will find applications in time-resolved spectroscopy and in the generation of high-peak-power pulses. In fact, with the development of new solid-state materials that have a large amplification bandwidth, such as Ti:Al_2O_3, this infrared femtosecond laser could be the seeding oscillator of a high-



(a)

(b)

Fig. 3. (a) Autocorrelation and (b) the corresponding spectrum of the shortest pulses produced. The dashed curve in (a) represents the best fit of the autocorrelation if we assume a sech^2 pulse shape.

power amplifier that uses the chirped amplification technique.[9]

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