Effect of dye concentration on picosecond and femtosecond cw passively mode-locked Ti:sapphire/DDI lasers

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The role of the saturable absorber dye DDI in the cw passively mode-locked Ti:sapphire/DDI laser both with and without compensation prisms is investigated. This laser self-starts when the concentration of the dye is greater than \( \approx 2 \times 10^{-4} \) M for both picosecond and femtosecond pulses. With the prism pair, the laser generates Gaussian pulses 150 fs in duration. The corresponding spectrum is sech\(^2\) in shape with a FWHM of 8 nm. These parameters are independent of the DDI concentration. Without the prism pair, the laser generates picosecond pulses with the FWHM decreasing from 8 to 3 ps as the dye concentration is increased from \( 2.5 \times 10^{-4} \) to \( 1 \times 10^{-3} \) M. Concurrently the pulse shape changes from a Lorentzian to a nearly symmetric two-sided exponential shape. The picosecond spectra for different dye concentrations are also presented.

Recently there has been considerable interest in the generation and amplification of ultrashort optical pulses from the solid-state Ti:sapphire laser. Much effort has also been devoted to the study of its mode-locking mechanisms, both theoretically\(^1\)-\(^5\) and experimentally\(^6\)-\(^\uparrow\).\(^7\)\(^\uparrow\) Recently we reported the pulse-forming dynamics of a picosecond cw passively mode-locked Ti:sapphire laser\(^7\) with DDI dye as the saturable absorber. In this Letter we present further experimental data concerning the effect of dye concentration on the generation of picosecond and femtosecond pulses from such a laser. Both the pulse shapes and the spectra of the laser output are examined.

Our experimental setup is shown in Fig. 1. The laser consists of a six-mirror cavity with a 2-cm-long Brewster-angle-cut Ti:sapphire crystal as the gain medium, an organic dye (DDI) as the saturable absorber (standard Coherent nozzle), and a 5% output coupler. The gain and absorber folding sections of the resonator have focusing mirrors of 15- and 5-cm radii of curvature, respectively. No bandwidth-limiting or tuning elements are employed inside the cavity. The two Brewster-angled prisms are made of SF10 glass and are separated by 66 cm tip to tip. The repetition rate of the laser is 77 MHz. The pulse width and spectrum are measured by a background-free autocorrelator and a monochromator, respectively.

The passively mode-locked laser self-starts when the concentration of the saturable absorber dye is greater than \( \approx 2 \times 10^{-4} \) M for both picosecond and femtosecond pulses. If the dye concentration is greater than \( 1.2 \times 10^{-3} \) M, the output of the mode-locked laser becomes unstable, i.e., it self-Q-switches. To examine the role of the saturable absorber, we record the pulse width and spectrum of the laser output for various concentrations of DDI, either with or without the prism pair. Typical autocorrelation traces of the laser are shown in Fig. 2. We find that the shape of the autocorrelation trace of the femtosecond pulse is Gaussian [Fig. 2(a)] and that of the picosecond pulse is Lorentzian at lower dye concentrations [Fig. 2(c)] and a nearly symmetric two-sided exponential function at higher concentrations [Fig. 2(d)]. The transition of the pulse shapes with dye concentration is not abrupt. The width of femtosecond pulses is \( \approx 150 \) fs (FWHM) and nearly the same for all dye concentrations. This is shown in Fig. 3. This behavior is identical to that of the femtosecond passively mode-locked Ti:sapphire/HITCI laser.\(^6\) The picosecond pulses become narrower as the dye concentration is increased. For the range of dye concentration studied, the pulse durations vary...
Fig. 2. (a) Autocorrelation trace (Gaussian shape) and (b) spectrum (sech^2 shape) of femtosecond pulses, and autocorrelation traces of the picosecond pulses (c) for a dye concentration of \(3 \times 10^{-4}\) M (Lorentzian shape) and (d) for a dye concentration of \(1.1 \times 10^{-3}\) M (symmetric two-sided exponential shape). The open squares are experimental data, and the solid traces are fitted curves.

from \(8\) to \(3\) ps (FWHM). Previously, Sarukura et al.\(^\text{14}\) showed a streak camera trace of asymmetric picosecond pulses with a slowly decaying tail generated by a similar passively mode-locked Ti:sapphire/DDI laser. Since absorber saturation steepens the leading edge of the pulse, the higher the dye concentration is, the sharper the leading edge becomes. This explains why we have experimentally observed different shapes for the picosecond autocorrelation traces at different dye concentrations. It also follows that narrower picosecond pulses and symmetric two-sided exponential autocorrelation traces are observed for higher dye concentrations. The peak intensity in the laser rod (\(1\ GW/cm^2\) for our laser) is significantly higher than the saturation intensity for Ti:sapphire (\(0.2 MW/cm^2\)). The gain saturation effect in addition to absorption saturation could conceivably be significant as a pulse-shortening mechanism. We thus propose that DDI, with an excited-state lifetime of \(\tau_{\text{ext}} = 17\) ps, behaves like HITCI (\(\tau_{\text{ext}} = 1.2\) ns), as a slow absorber in the picosecond passively mode-locked Ti:sapphire laser. In the femtosecond regime, the self-phase-modulation effect owing to the Kerr effect inside the laser cavity together with the negative group-velocity dispersion contributed by the Brewster-angled prism pair provide additional pulse-shaping mechanisms. The widths of the femtosecond pulses in our laser are essentially independent of dye concentration. It is apparent from our data that non-linear pulse-shortening forces are dominant for femtosecond pulses but not for picosecond pulses. The dependence of picosecond and femtosecond pulse duration on the pumping power has also been investigated for a dye concentration of \(6 \times 10^{-4}\) M. The width of picosecond pulses broadens monotonically from \(4.6\) to \(6.5\) ps as the pumping power increases from 4 to 7 W, whereas that of femtosecond pulses is almost independent of the pumping power.

Figure 3 illustrates the spectra of the picosecond pulses as a function of the dye concentration. The pumping power is kept constant at 5 W. The average output spectrum reveals multiple clusters of the longitudinal modes (unresolved). The main peaks in each spectrum are separated by \(3.3\) nm. Similar spectra with clusters separated by \(3.6\) nm have also been observed in a cw Ti:sapphire laser during the early stage of its spectral evolution to the steady state.\(^\text{16}\) The latter observation has recently been identified by us as the spectral windowing effect of

![Figure 3](image-url)

**Fig. 3.** Pulse width of the mode-locked Ti:sapphire/DDI laser as a function of dye concentration with and without a compensation prism pair.
the Brewster-angle-cut Ti:sapphire crystal as a birefringent filter in the cavity.16 We believe that the clusters observed here are also due to the birefringent filtering effect, with spectral broadening contributed by self-phase-modulation and cross-phase-modulation effects. This topic is still under investigation.

The peaks of the absorption spectrum of DDI and the gain spectrum of Ti:sapphire are ≈710 and ≈800 nm, respectively. When the dye concentration is increased, the level of loss in the cavity, particularly the blue side of the gain spectrum, is also elevated. This effect causes the red-shift trend and an increasingly asymmetric and narrower lasing spectrum for higher dye concentrations, as shown in Fig. 4. Previously, Sarukura et al.17 showed that the lasing spectrum of a picosecond mode-locked Ti:sapphire/HITCl laser shifts to the blue and broadens as the pumping power is increased. Their results can also be explained by the physical picture presented above.

The shape of the spectral distribution of the femtosecond pulse is close to $\text{sech}^2$ with bandwidth (FWHM) of $=8$ nm. This is shown in Fig. 2(b). The shape of the femtosecond pulse is, however, Gaussian. As a result, the pulse–bandwidth product, $\gamma_p \Delta \nu \approx 0.58$, is larger than that of the transform-limited product for either Gaussian (0.44) or $\text{sech}^2$ (0.32) pulses. The central wavelength of the lasing spectrum for femtosecond pulses is sensitive to the position of prisms but not the dye concentration. It shifts from $\approx789$ to $\approx796$ nm when a prism is translated by 2 mm for more negative dispersion at a dye concentration of $3 \times 10^{-3}$ M. Nevertheless the femtosecond pulse width is scarcely changed.

In summary, we have investigated the effect of dye concentration on the passively mode-locked Ti:sapphire/DDI laser both with and without the chirp-compensation prism pair. Our experimental results show that the pulse width and the spectrum of femtosecond pulses are not affected by the concentration of the saturable absorber dye, whereas those of picosecond pulses are. The pulse shape and spectrum in the femtosecond regime are Gaussian and $\text{sech}^2$, respectively. For picosecond pulses, the spectrum exhibits multiple clusters. A red-shift trend is also observed for increasing dye concentration. The pulse width decreases from $=8$ to $=3$ ps as the dye concentration is increased from $2.5 \times 10^{-4}$ to $1 \times 10^{-3}$ M. Concurrently the pulse shape changes from a Lorentzian to a nearly symmetric two-sided exponential shape. These results provide additional information on the role that the saturable absorber may play in cw passively mode-locked picosecond and femtosecond Ti:sapphire/DDI lasers.

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References