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Femtosecond pulse generation around 650 nm in a passive mode-locked Kiton Red dye laser

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Sub 100 fs light pulses have been generated in the spectral region between 640 and 660 nm (the shortest duration being 50 fs at 653 nm) by passive mode-locking of a Kiton Red dye laser with the saturable absorber DQTCI.

Nearly the whole visible and near infrared range (with some gaps) has been covered by various amplifying dye–saturable absorber combinations yielding femtosecond light pulses in cw passive or hybrid mode locked lasers [1–4]. Pulse durations below 150 fs were obtained in some of these combinations by group velocity dispersion balanced laser operation [5].

In the spectral region around 650 nm sub 100 fs pulse generation has not been reported so far. The longest wavelength achieved by passive mode-locking of a Rhodamine 6G laser was 640 nm [6]. In this laser 50 fs pulses at 640 nm have been generated by the saturable absorber TCETI. Energy transfer passive mode-locking with an argon ion pump laser has been applied to cover the 652–694 nm region with Rhodamine 6G+Sulforhodamine 101 mixtures as gain medium and DQTCI or DCCI as saturable absorbers [7,8]. The shortest pulse durations obtained were 58 fs at 685 nm [8] and 120 fs at 666 nm [7]. Applying hybridly mode-locked systems pumped by the second harmonic of Nd:YAG lasers [9,10], pulses of 65 fs duration at 640 nm [9] have been obtained in a Rhodamine 6G/DODCI laser, and pulses of 60 fs duration at 675 nm were generated using Rhodamine 101 or Sulforhodamine 101 as gain media and DQTCI as absorber medium [10]. Subpicosecond Rhodamine B lasers have been operated around 650 nm [10–12]. Cw passive mode-locking with DQTCI was achieved in the wavelength region of 616–658 nm [11]. However, the shortest duration was 220 fs at 635 nm [11]. Hybrid mode-locking of Rhodamin B lasers resulted in pulses of duration down to 320 fs in the spectral region between 604 and 632 nm when DQTCI or DDBCI were used as saturable absorbers [10,12]. Using Oxazine 720 as saturable absorber pulse durations down to 187 fs were obtained around 649 nm [10].

Here we report on passive mode-locking of a Kiton Red laser with the saturable absorber DQTCI [13] producing sub 100 fs pulses in the wavelength region of 640–660 nm. The dyes Kiton Red 620 (Sulforhodamine B, free of sodium) [13,14] and DQTCI (1,3′-diethyl-4,2′-quinolythiacarbocyanine iodide) [13,15] were purchased from Lambda Physik. The fluorescence quantum yield of Kiton Red 620 in ethylene glycol was determined to be $\Phi_F = 0.885 \pm 0.015$ independent of concentration up to about 5×10^{-3} mol/litre. For DQTCI in ethylene glycol $\Phi_F = 0.049 \pm 0.005$ was determined giving a fluorescence lifetime of $\tau_F = 20 \pm 1$ ps (radiative lifetime $\tau_{rad} \approx 4.1$ ns). The fluorescence quantum yield measurement system is described in ref. [16].

Kiton Red 620 has been previously used in hybrid mode-locked systems pumped by the second harmonic of a mode-locked Nd:YAG laser, and pulses as short as 29 fs were obtained at 615 nm when a mixture of DODCI and DQOCI was used as saturable absorber in a dispersion optimized linear cavity

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[17]. In a quasi-linear ring colliding pulse mode-locked femtosecond laser using a binary Rhodamine 6G and Kiton Red energy transfer gain dye mixture, pulses of duration down to 30 fs centered around 623 nm were generated [18].

The linear dispersion balanced laser configuration used here is described in detail elsewhere [19] and we outline the modifications only. The focusing mirrors have smaller radii of curvature, that is 5 cm in the gain section, and 3.8 cm and 2.5 cm in the absorber section. The output mirror reflection band is centered at 640 nm (transmission of 0.8%). The total mirror losses are $\approx 2.4\%$ ($\approx 0.9\%$ for output mirror) at 670 nm, and $\approx 4\%$ ($\approx 1\%$ for output mirror) at 680 nm since the mirrors were designed for a Rhodamine 6G/DODCI laser. At the long wavelength side the mirror losses limit the laser action of Kiton Red 620 without a saturable absorber to about 670 nm. The repetition rate of the laser was 135 MHz and the pulse separation was 7.4 ns.

We used a 3×10^{-3} mol/litre ethylene glycol solution of Kiton Red 620 in a ≈ 100 μm thick gain jet resulting in a small signal transmission of $\approx 20\%$ at 514 nm and $\approx 65\%$ at 488 nm. The concentration of the saturable absorber DQTCI in ethylene glycol was varied in the range from 10^{-4} mol/litre to 5×10^{-4} mol/litre. The absorber jet thickness was ≈ 35 μm. The two fused silica prisms for compensation of group velocity dispersion were 37 cm apart. The Kiton Red laser was pumped by all lines of a small frame argon ion laser (Spectra Physics Model 2016 5W laser). Without saturable absorber the laser threshold pump power was as low as 300 mW and the laser operated at ≈ 600 nm. With saturable absorber added the laser threshold pump power increased to 3 to 5 W depending on the absorber concentration and the wavelength tuning.

Pulses shorter than 100 fs could be generated in the spectral region between 640 and 660 nm by varying the absorber concentration. A tunability of approximately 10 nm was achievable at a fixed absorber concentration by adjustment of the cavity and by changing the velocity of the saturable absorber in the loss jet thus varying the absorber dye thickness. The shortest pulses were obtained around 650 nm. Typical average output powers were 10 mW.

Figure 1 shows the autocorrelation trace (recording time is 10 μs) and the spectrum (recording time

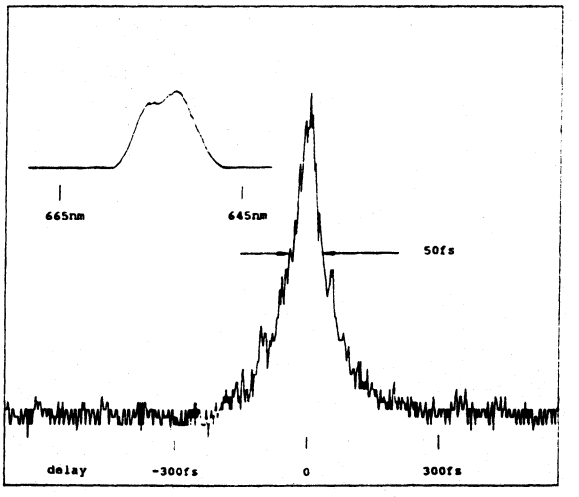

Fig. 1. A noncollinear SHG autocorrelation trace and the corresponding spectrum obtained with the linear passive mode-locked Kiton Red 620-DQTCI dye laser.

is 100 ms) of pulses which have been recorded using a DQTCI concentration of 2.6×10^{-4} mol/litre ($\approx 9\%$ small signal loss due to saturable absorber) and a pump power of 5 W. The pulse duration is $\Delta t = 50$ fs (fwhm) assuming a sech^2-intensity pulse shape ($\Delta\tau/\Delta t = 1.543$ [3,20], where $\Delta\tau$ is the full halfwidth of the autocorrelation trace). The temporal–spectral bandwidth product is $\Delta\nu\Delta t \approx 0.25$ (theoretical value for a sech^2-pulse is $\Delta\nu\Delta t = 0.315$ [3,20]). The wings of the displayed autocorrelation trace are wider than expected for a sech^2 pulse shape. At the 10% level the experimental autocorrelation curve is a factor of 60% broader than the autocorrelation curve of a sech^2-pulse. Autocorrelation traces and spectra of the shape displayed in fig. 1 are obtained in theoretical simulations of femtosecond pulse generation [21] if the negative group velocity dispersion is slightly less than the optimum value necessary to compensate fully the frequency chirp caused by the positive self-phase modulation in the dye jets (laser operation in the higher order soliton-like regime [22] near to the boarder of the stable fundamental soliton-like regime). A slight reduction of the glass path through the prisms (increase of negative group velocity dispersion) resulted in traces fitting reasonably to sech^2-shapes at the cost of a slightly broader pulse duration.

Three additional features of the studied Kiton

Red/DQTCI laser should be mentioned:

(i) Without spectral filtering the laser would operate at the short wavelength side of the absorption peak of DQTCI at 633 nm in the wavelength interval between 600 and 610 nm since the stimulated emission cross section maximum of Kiton Red 620 is around 590 nm. The obtained pulse durations in the spectral region between 600 and 610 nm were > 150 fs. The effective spectral amplification width of the gain and absorber medium seems to hinder the generation of shorter pulses. Laser operation in the 640–660 nm spectral region was achieved by a spectral/spatial filtering using the prism in front of the high reflector. The wavelength separation of more than 40 nm between the two regions resulted in ≈ 2 mm spatial separation of the two spectral components at the prism, and at minimum glasspath through the prism only the long wavelength component survived.

(ii) Normally two pulses are counter-propagating in the linear laser oscillator and the pulses pass the output mirror at times $t = t_R/4$, $3t_R/4$, $5t_R/4$... where t_R is the full resonator round trip time. The output pulse separation is $t_R/2$ (7.4 ns in our case) and the time interval of passing the amplifier medium is $t_R/4$ (3.7 ns in our case). When increasing the pump power the laser jumped readily to a regime where four pulses per round trip were counter-propagating. In the four pulse regime the laser remained very stable and yielded sub-100 fs pulses in the 640–660 nm spectral region. The pulse positions at the output mirror occurred at $t = t_R/4$, $3t_R/8$, $3t_R/4$, $7t_R/8$, $5t_R/4$, ... In this case two pairs of pulses collide in the absorber jet once per round trip and all the four pulses experience equal gain in the amplifying dye jet. The time interval of a pulse passing through the amplifier medium is $t_R/8$ (1.85 ns in our case). Similar observations have been reported in ring lasers containing Rhodamine 700 as an amplifying dye and DDI and HITCI as saturable absorbers [23,24]. There the four-pulse-regime was attributed to the fast recovery time of Rhodamine 700. In our case the fluorescence lifetime of Kiton Red 620 is approximately 4 ns ($\tau_{rad} \approx 4.5$ ns own measurement with apparatus described in ref. [16]). The experimental findings indicate that the pulse separation should be less than twice the fluorescence lifetime (interval of passing the amplifier medium less than fluorescence lifetime) to avoid multiple pulsing [25]. All of the reported measurements here have been performed in the regime with two pulses per round trip which could be achieved by operating the laser only slightly above threshold.

(iii) In both regimes (two or four pulses per round trip) the colliding pulse effect is expected to take place only once per round trip since the absorber was positioned only approximately in the cavity centre and the exact position for colliding pulse mode-locking in each absorber passage has not been searched [19].

In conclusion we report the cw argon ion laser pumped passive mode-locking of a Kiton Red 620 dye laser without applying an energy transfer dye [18]. Using the saturable absorber DQTCI, pulse durations of 50 to 100 fs have been achieved in the spectral region of 640 to 660 nm. It is expected that this region can be extended to longer wavelengths (up to ≈ 675 nm) when mirrors centred around 660 nm are used. The overall efficiency of the laser could be improved by using a thicker gain jet and pumping only by the green line (514 nm) of a larger argon-ion laser.

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