Generation of 33-fs pulses from a passively mode-locked Cr\(^{3+}\):LiSrAlF\(_6\) laser

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Femtosecond and picosecond operating regimes of an argon-ion-pumped Cr\(^{3+}\):LiSrAlF\(_6\) laser, passively mode locked using a saturable absorber, are described. At low absorber concentrations, self-starting cw mode locking is demonstrated, which yields pulses as short as 33 fs after extracavity prism compensation. At higher concentrations, both cw mode-locked and Q-switched mode-locked operation is possible. The significance of the recovery time of the absorber is discussed.

The recent demonstration of the Cr\(^{3+}\) lasing transition in a variety of new vibronic solid-state laser media\(^1\)\(^2\) has led to a great deal of interest in these materials as alternatives to Ti:sapphire for the generation of broadly tunable ultrashort pulses. Attractive features of these materials include long upper-state lifetimes, which permit their use as efficient amplifying media, and absorption bands in the red, which allow diode pumping by the new generation of laser diodes near 670 nm.\(^3\)\(^4\) One of these materials is Cr\(^{3+}\):LiSrAlF\(_6\) (Cr:LiSAF), which has a linewidth of ~220 nm and an upper-state lifetime of 67 \(\mu\)s.\(^2\)

A krypton-ion-pumped Cr:LiSAF laser has been actively mode locked to generate pulses of 150-fs duration,\(^5\) and an argon-ion-pumped Cr:LiSAF laser has recently been passively mode-locked to produce sub-50-fs pulses.\(^6\) Cr:LiSAF has also been self-mode-locked, which yielded 50-fs pulses.\(^7\) Here we report the development of a passively mode-locked Cr:LiSAF laser that generates nearly transform-limited pulses as short as 33 fs. We have studied different mode-locked operating regimes as a function of absorber concentration and pump power and have investigated the requirements for absorber lifetimes in self-starting passively mode-locked lasers. A moving mirror has also been demonstrated as an alternative starting mechanism for the laser.\(^8\)

The experimental arrangements used were similar to those in our previous study.\(^6\) An antireflection-coated 10-cm focal-length lens was used to focus the 488-nm line from an argon-ion laser into a 22-mm-long Brewster-angled Cr:LiSAF rod (supplied by Lightning Optical Corporation). The laser rod was oriented such that the \(E\) vector of the laser field was parallel to the \(c\) axis of the crystal, which led to ~2% of the pump radiation's being absorbed. The astigmatically compensated cavity was formed by using single-stack dielectric-coated mirrors and is shown in Fig. 1. Mirrors M1 and M2 had a radius of curvature of 10 cm. All the mirrors used were highly reflecting at the laser wavelength, except mirrors M6 and M7, which were of ~99% reflectivity. The Brewster-angled prisms P1 and P2 were made from Schott F2 glass and were mounted on translation stages to allow the amount of intracavity glass to be varied. The folded section containing the 100-\(\mu\)m-thick flowing jet of the saturable absorber was formed by two 5-cm radius-of-curvature mirrors. Tuning of the laser was achieved either by using a 0.8-mm-thick single-plate birefringent filter or by translating a slit between prism P2 and mirror M7. A slit was placed adjacent to mirror M3 to provide discrimination between the high- and low-intensity modes.\(^9\) The mode-locked train was monitored with a fast photodiode. All pulselength measurements were performed by using a standard, collinear autocorrelator with a 100-\(\mu\)m-thick LiIO\(_3\) frequency-doubling crystal.

Initially, neocyanine ([bis-4-(1-ethylquinoline) \(\gamma'-4'-(1'-ethylquinoline)\)]pentamethine cyanine diiodide), dissolved in ethylene glycol, was used as the saturable absorber. In a simple cavity with no adjustable group-velocity dispersion (i.e., without prisms P1 and P2) and at a neocyanine concentration of 10\(^{-5}\) M, the cw lasing threshold of the laser was 225 mW of absorbed pump power. For pump powers of 200 mW above this threshold, stable self-starting cw mode-locking was achieved, and pulses of 4.8 ps in duration, tunable from 800 to 853 nm, were produced. The only amplitude modulation observed on the mode-locked pulse trains throughout this research (typically ~1%) was attributable to the pump laser. At concentrations below 10\(^{-5}\) M, mode-locked pulses of similar durations were produced, but the laser was not stable. For concentrations between 10\(^{-5}\) and 5 \(\times\) 10\(^{-5}\) M, cw mode locking producing picosecond pulses was maintained for pump powers between 425 mW and 1.2 W. On increasing the concentration further, either cw mode-locked or Q-switched mode-locked operation was observed, dependent on the pump power used. The Q-switched mode-locked envelopes had a typical separation of ~23 \(\mu\)s, and the FWHM of each envelope was 6 \(\mu\)s. The mode-locked pulse durations inside the Q-switched mode-locked envelopes were typically ~6 ps.

At a concentration of 6 \(\times\) 10\(^{-6}\) M, the cw lasing threshold was 310 mW. At pump powers between 420 and 730 mW, stable Q-switched mode-locked...
operation was observed. Above 730 mW, only cw mode locking occurred. As the concentration of the neocyanine was increased, the pump powers at which cw mode locking replaced Q-switched mode-locked operation also increased. At a concentration of \(7.1 \times 10^{-5}\) M, the laser stably self-started with Q-switched mode-locked operation and cw mode locking could not be obtained up to the maximum available pump powers of 1.2 W.

To generate femtosecond pulses from this laser, the prism sequence was inserted into the cavity. In our earlier study we employed SF10 glass prisms. Even with the elimination of second-order dispersion and minimization of third-order dispersion, i.e., by working at the minimum glass path for the smallest possible prism separation, it was possible only to produce pulses of \(~50\) fs in duration, though bandwidths capable of supporting significantly shorter pulses were observed. This was the result of the relatively large third-order dispersion of SF10 glass, which has been estimated to be \(~10\) fs per pass for a pulse having a 22-nm bandwidth.\(^{10}\) It is known that F2 glass has a smaller third-order dispersion by a factor of \(~3\) compared with that of SF10 glass\(^{11}\) and so should permit the generation of much shorter pulses.

At an optimum F2 prism separation of 40 cm and a neocyanine concentration of \(10^{-5}\) M, stable self-starting cw mode-locked pulses of 50-fs duration were routinely produced with average output powers of \(~25\) mW for 1 W of pump power. To compensate for residual chirp on the pulse, an identical pair of F2 prisms was placed extracavity at a separation of 40 cm. These prisms also served to precompensate for the dispersive effects of the subsequent optics and autocorrelator components. After optimizing the extracavity prisms, we routinely generated stable pulses of 33 fs. A typical autocorrelation and associated spectrum of these pulses is shown in Fig. 2, with the FWHM being taken from the baseline. The time–bandwidth product of these pulses was 0.39, reasonably close to the value of 0.315 expected of transform-limited sech\(^2\) pulses.

The mode-locked picosecond output from the laser was used to measure the fluorescence lifetimes of different saturable absorbers. With pulses of \(~6\) ps and average powers of 10 mW, the fluorescence decay of the absorbers was measured by using a Hamatsu optical sampling oscilloscope (Model OOS-01) with a 20-ps resolution. The excellent stability of the laser permitted a large number of averages to be taken (as many as 10,000) for these measurements. The lifetime of neocyanine in ethylene glycol was measured to be \(~120\) ps.

To assess the effects of the saturable absorber lifetime on the mode locking of the laser, we used DTTCI (3,3'-diethylthiatricarbocyanine iodide) instead of neocyanine. The lifetime of DTTCI was measured to be \(~800\) ps in ethylene glycol. With the non-dispersion-compensated cavity, the cw lasing threshold was the same as with neocyanine at DTTCI concentrations of \(10^{-5}\) M. The concentration of DTTCI was increased while the output of the laser was monitored to determine whether it was mode locked. Up to pump powers of 1.2 W and concentrations of \(2 \times 10^{-4}\) M, no mode locking was observed.

Stable mode locking was obtained, however, by using the dye DDCI-4 (1,1'-diethyl-4,4'-dicarbocyanine

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Fig. 1. Schematic of laser cavity.

Fig. 2. (a) Interferometric autocorrelation and (b) associated spectrum of typical output of a dispersion-compensated passively mode-locked laser.
iodide) as the saturable absorber, whose lifetime was measured to be ~250 ps. The laser performance was similar to that observed with neocyanine. With DCCI-4, stable cw mode locking was achieved at a concentration of $10^{-6}$ M, and subpicosecond pulses were generated from 821 to 855 nm.

It appears that for Cr:LiSAF a saturable absorber with a relatively short upper-state lifetime (<800 ps) is required to initiate stable cw mode-locked operation in the passively mode-locked laser. However, we stress that we believe that the saturable absorber dye is required only to start the mode locking. With or without the intracavity prisms, the pulse durations observed are much shorter than would be expected from the mode locking of a (solid-state) laser by a fast saturable absorber dye in the absence of significant gain saturation. Once the intracavity power has reached the necessary level, we speculate that self-focusing or Kerr lens mode locking becomes the primary mode-locking mechanism. This was confirmed by the sensitivity of the mode-locked performance to the cavity alignment, the location of the beam waist, and the intracavity slit. The ultimate pulse duration achieved in the laser is then determined by the interplay between self-phase-modulation and group-velocity dispersion. Also, it appears that too much absorber is detrimental to cw mode locking and leads to Q switching.

We note that in earlier research on passively mode-locked cw dye lasers, we observed that the dye combinations that produced stable passive mode locking (albeit with a slow saturable absorber13) also required the saturable absorber dye to have a relatively short lifetime (<1 ns in either the ground state or the photoisomer). We speculate that in these systems also the fast response of the absorber is required to initiate the pulse evolution, which then proceeds as passive mode locking with a slow saturable absorber. Synchronous pumping should relax this requirement. New and Rea14 discuss the start-up of passively mode-locked lasers, and their Eq. (27) yields a condition for the minimum absorber strength required for self-starting that decreases with decreasing absorber lifetime. The start-up of such lasers is currently being studied numerically and appears to confirm our empirical observation.15

When mirror M6 was mounted on a shaker (Bruel and Kjaer Model 4080), contiguous trains of picosecond pulses were observed. This behavior was similar to that initially reported with Ti:sapphire lasers.3 Further optimization of this technique may provide an alternative approach to starting the Cr:LiSAF laser in the same manner as using a shaking mirror to start and sustain a self-mode-locked Ti:sapphire laser.16

In summary, we have generated nearly transform-limited pulses of 33-fs duration. These pulses are, to our knowledge, the shortest produced directly from any chromium-ion-based solid-state laser. We have also studied the behavior of the laser as affected by the absorber concentration and pump power. We have demonstrated two saturable absorbers and have suggested a requirement for short absorber lifetimes in self-starting passively mode-locked lasers. We have also demonstrated a moving mirror as an alternatively starting mechanism. With proper optimization of the laser demonstrated, including minimization of the intracavity third-order dispersion, this scheme should permit the generation of tunable sub-20-fs pulses from Cr:LiSAF.

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