Mode-locked Cr:LiSAF laser far off the gain peak: tunable sub-200-fs pulses near 1 μm

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We report, to the best of our knowledge, first mode-locking results of a Cr:LiSAF laser near 1 micrometer region. The system is pumped only by a single 1.1 W high-brightness tapered diode laser at 675 nm. A semiconductor saturable absorber mirror (SESAM) with a modulation depth of 1.5% and non-saturable losses below 0.5% was used for mode-locking. Once mode-locked, the Cr:LiSAF laser produced almost-transform-limited sub-200-fs pulses with up to 12.5 mW of average power at a repetition rate of 150 MHz. Using an intracavity birefringent filter, the central wavelength of the pulses could be smoothly tuned in the 1000–1020 nm range. Via careful dispersion optimization, pulsewidths could be reduced down to 110-fs level. The performance in this initial study was limited by the design parameters of the SESAM used, especially its passive losses, and could be improved with an optimized SESAM design.

1. Introduction

Cr:LiSAF is an attractive broadband solid-state laser medium in the near-infrared region [1–6]. It shows a broad absorption band centered around 650 nm (FWHM: ~ 100 nm) [7], that enables flexible pumping by low-cost red laser diodes or LEDs (light emitting diodes) [8–11]. With the progress in crystal growth process, Cr:LiSAF passive losses are now reduced to 0.15%/cm level from above 0.5%/cm observed in earlier samples [12,13]. Moreover, excited state absorption strength is at a reasonable level in Cr:LiSAF: $\sim 1/3^{\text{rd}}$ of the emission cross section [14]. Combination of these favorable properties enables construction of low-cost and compact Cr:LiSAF laser cavities with mW-level lasing threshold and 50%-level laser efficiency [13]. Furthermore, the broad emission band of Cr:LiSAF (λ_c : ~ 850 nm, FWHM: ~ 200 nm) could potentially enable generation of sub-6-fs pulses [7], and 10–20 fs pulsewidth were already demonstrated [15–18]. In continuous-wave (cw) lasing operation, a tuning range covering the 780–1110 nm region could easily be achieved in systems pumped by simple 100 mW level pump diodes [13]. On the other hand, its relatively low thermal conductivity [19], and presence of rather strong thermal quenching of fluorescence lifetime [20] and Auger upconversion process [14] creates difficulties in power scaling of Cr:LiSAF lasers [21–23].

Compared to the cw case, laser tuning is more challenging in mode-locked operation due to the much more stringent requirements. Despite that, using Kerr-lens mode-locking (KLM), femtosecond (fs) tuning ranges covering 835–910 nm [24], 809–910 nm [25] and 807–919 nm [17] were demonstrated with

Cr:LiSAF. Using single-walled carbon nanotubes for mode-locking, fs pulses in the 868–882 nm range were also achieved [26]. Alternatively, by employing saturable absorber mirrors (SBRs [27], also known as SESAMs [28]) for mode-locking, fs tuning was attained in the 800–905 nm range in [29], in the 803–831 nm, 828–873 nm, 890–923 nm intervals in [30], and between 825–875 nm in [31]. Overall, the achieved mode-locked tuning range of Cr:LiSAF lasers is limited to the 803–923 nm region so far, where the gain of Cr:LiSAF material is rather high [1,14]. On the other hand, it is also interesting to investigate mode-locked operation capability of Cr:LiSAF at wavelengths above 920 nm (far off the gain peak [32]), as applications such as multiphoton-microscopy [33], and spectroscopy [32] could benefit from this wavelength range.

Femtosecond sources around the 1000 nm region are also interesting as seed sources for Yb-based amplifiers. Popular systems such as Yb:YAG could be efficiently seeded by Yb-fiber seeders, whereas for crystals such as Yb:YVO₄ [34], Yb:CaF₂ [35], Yb:KYW [36], Yb:YLF [37–39], Yb:LLF [40,41], the gain spectra cover regions well below 1030 nm, especially at cryogenic temperatures [42–45]. Unfortunately, it is rather difficult to develop Yb-fiber based seeders at these shorter wavelengths [46]. Hence, alternative solid-state laser sources such as Ti:Sapphire have been explored in some of the earlier studies [47–49]. Femtosecond Ti:Sapphire sources are quite well-developed, but their mode-locking at the 1000 nm region is difficult to achieve due to limited figure of merit (FOM) of Ti:Sapphire crystals, as well as the reduced gain cross section of the material in this spectral

region [32,50–52]. To our knowledge, the fs tuning range of diode pumped Ti:Sapphire systems are currently rather limited (775–825 nm from a 2 x 3.5-W diode pumped system [53]), and state-of-the-art fs Ti:Sapphire based sources that could tune to 1000 nm region are still pumped by complex high-power green laser sources.

In this study, in search for a low-cost and compact seed source for Yb:YLF amplifiers [38], we have investigated mode-locking of Cr:LiSAF lasers around the 1000 nm wavelength region. A single 675 nm, 1.1 W tapered diode laser is used as the pump source. In cw lasing experiments, around 150 mW of laser output power could be achieved at 1000 nm. Upon mode-locking with a SESAM with a 1.5% modulation depth, sub-200-fs pulses with up to 12.5 mW of average power was demonstrated at 150 MHz repetition rate. The central-wavelength of the mode-locked pulses could be tuned in the 1000–1020 nm range using an intracavity birefringent tuning plate. Pulses widths down to 110 fs could be achieved via adjusting the dispersion of the cavity. To the best of our knowledge, these are the first mode-locking results obtained from Cr:LiSAF systems around 1000 nm wavelength. We believe that with design of a SESAM that is optimized for mode-locking in this low-gain region, the results acquired in this initial work could be improved significantly, in terms of average power, tuning range, and achievable pulsewidth.

This paper is organized as follows: in Section 2, we describe the experimental setup. In Section 3, cw and cw mode-locked lasing results in the 1000-nm region will be presented. In Section 4, we will conclude with a brief summary.

2. Experimental Setup

Figure 1 shows a simple schematic of the Cr:LiSAF laser. The system is pumped by a 1.1 W tapered diode laser operating at 675 nm (TDL). The TDL was grown and characterized at the facilities of Ferdinand Braun Institute and detailed information on this class of diodes can be found in [54]. The diode output had an astigmatism of 600 μm , and a beam quality factor of around 2.5 and 1.1 in the slow and fast axes, respectively. The brightness of the laser diode is $\sim 1000 \text{ mW}/\mu\text{m}^2$, which is 2–3 times larger compared to typical single-mode laser diodes at this wavelength. Similar high-brightness diodes were already used for efficient pumping of Cr:LiCAF/LiSAF, Alexandrite and Tm:YAG/LuAG lasers [55–57]. The output of the tapered diode laser was first collected by an aspheric lens with a focal length of $f_1 = 4.5 \text{ mm}$. A cylindrical lens with a focal length of 50 mm (f_z) was further used in the fast axis, before the beam is focused down into the Cr:LiSAF crystal using a 75 mm achromatic doublet (f_2). An astigmatically-compensated, X-shaped cavity consisting of two curved pump mirrors (M1 and M2, $R = 75 \text{ mm}$), a flat end mirror (M3), and a flat output coupler (OC) were employed in the cw laser experiments. The pump mirrors (M1–M2) were optimized for laser operation around 1000 nm: they had reflectivity lower than 2% at 675 nm, and higher than 99.98% in the 900–1050 nm range. A 15-mm-long, 0.8% Cr-doped Cr:LiSAF crystal was used as the gain element. The Cr:LiSAF crystal absorbed >99% of the incident TM polarized pump light at 675 nm. The estimated single pass loss of the crystal is around 0.2–0.25%. The length of the crystal was optimized for high-power two-side pumping studies [23], and ideally a shorter crystal with lower passive losses could be used in this 1-W pumped system.

The crystal was 2 mm thick and mounted with indium foil in a copper holder under water cooling at 18 °C. The cw lasing wavelength tuning data is taken using a 3-mm-thick crystal quartz birefringent filter (BRF) with an optical axis 45° to the surface of the plate [58].

Fig. 1. Experimental setup of the tapered diode-pumped Cr:LiSAF laser mode-locked around 1 micrometer. TDL: Tapered diode laser; SESAM: Semiconductor saturable absorber mirror; OC: Output coupler, BRF: Birefringent filter.

For mode-locking experiments, the cavity is extended via addition of another curved mirror (M4) with a radius of curvature of 100 mm. A SESAM is placed at the secondary focus generated by M4. For the cold cavity, the spot size ($1/e^2$ radius) inside the Cr:LiSAF crystal, on the OC and on the SBR are estimated to be around $\sim 15 \mu\text{m} \times 20 \mu\text{m}$, $\sim 125 \mu\text{m}$, and $\sim 25 \mu\text{m}$, respectively. The commercial SESAM (Reflekron, RK177D) had a company specified modulation depth of 1.5%, a non-saturable loss of around 0.5%, a reflectivity range covering the 995–1050 nm region, and a saturation fluence of $35 \mu\text{J}/\text{cm}^2$. The estimated critical intracavity pulse energy for stable cw mode-locking of the system is estimated to be around 10 nJ [59,60]. Mirrors M3–M4 were double chirped mirrors (DCMs) and provided $-80 \pm 20 \text{ fs}^2$ of group delay dispersions (GDD) per bounce in the 900–1070 nm region. Considering the dispersion of the Cr:LiSAF crystal ($15 \text{ fs}^2/\text{mm}$ around 1000 nm) and intracavity air, for the cavity shown in Fig. 1, the total cavity dispersion is estimated to be -550 fs^2 (2x4 bounces on DCMs). Replacing M3 with a regular HR with zero GDD sets the cavity dispersion to the -300 fs^2 level. Including an additional DCM mirror, we could adjust the dispersion to about -800 fs^2 . These additional settings were employed to fine-tune the pulsewidth of the mode-locked laser. A regular 0.2-mm-thick on-surface optical axis crystal quartz birefringent filter was used for tuning of the central wavelength of the mode-locked pulses.

3. Experimental Results

3.1 Continuous-wave lasing results

We have first investigated cw laser performance of the Cr:LiSAF laser around the 1000-nm region in detail. For that, several output couplers with transmission between 0.15 and 1.5% were explored. Figure 2 shows cw efficiency curves taken with different OCs. The reflectivity band of the OCs were centered around 1000-nm region, which shifted the free-running laser cw wavelength to 958–1010 nm band (lasing wavelength is specified for each OC in the figure legend). Note that, the emission cross section of Cr:LiSAF in E//c axis is around $4.8 \times 10^{-24} \text{ cm}^2$ at 850 nm, which decreases to around $2.6 \times 10^{-24} \text{ cm}^2$, $1.4 \times 10^{-24} \text{ cm}^2$ and $0.7 \times 10^{-24} \text{ cm}^2$ at 950 nm, 1000 nm, and 1050 nm, respectively [1]. As an example, at a wavelength of 1000 nm, assuming similar excited

state absorption to emission cross section ratio, the small signal gain is reduced around 3.5 fold. As a result of lower gain, the cavity requires usage of lower output coupling to achieve lasing. On the other hand, usage of lower OC values reduces the slope efficiency of the system (round-trip cavity passive loss is estimated to be 0.5% for the cw cavity). Moreover, compared to regular 850 nm lasing, the quantum defect increases from around 20% to 32%, which increases the thermal load on the crystal. As mentioned earlier, thermo-mechanically, Cr:LiSAF is very sensitive due to effects such as temperature quenching of fluoresce lifetime and Auger upconversion, hence these processes might start to reduce the laser performance as well [14,19–23]. As a result, compared to cw lasing at 850 nm, the cw laser performance we have achieved around 1000 nm is rather limited. For example, we have achieved cw power up to 450 mW and a slope efficiency of 47% from a similar TLD laser pumped system using a 1% transmitting output coupler, at a pump power of 1 W [57]. We see from Fig. 1 that, for the wavelength of 1010 nm, using a 0.3% output coupler, the system only produces around 150 mW of cw output power at a pump power of 1 W, with a slope lower than 20%. The performance is rather limited compared to 850 nm lasing, but this is expected since we are pushing the system far off the gain peak. Also, such performance will not be possible using a Ti:Sapphire laser due to the much larger losses of the system due to the limited FOM of Ti:Sapphire crystals.

Fig. 2. Measured continuous-wave (cw) power efficiency curves of the Cr:LiSAF laser around 1000 nm wavelength using output couplers (OCs) with transmissions ranging from 0.15% to 1.5%. Measured free running lasing wavelength for each OC is provided in the figure legend.

To look at this from another perspective, Fig. 3 shows the cw tuning data taken with the Cr:LiSAF laser at an absorbed pump power of around 0.8 W. Tuning data is taken with 6 output couplers with different output coupling values and reflectivity ranges. We see that, due to the reduced gain of the system, the laser performance decreases sharply as one goes into the longer wavelengths. The reduced gain of the system at longer wavelengths also puts a limit on the long wavelength tuning edge. For output coupling values of 0.15-0.5%, we could tune the laser to around 1100 nm, whereas the long wavelength tuning range was limited to 1050 nm and 1005 nm for the 1% and 1.5% output

couplers, respectively. As a final note, we should mention that it is not easy to increase the gain of Cr:LiSAF crystals by pumping harder due to the Auger upconversion process: when pumping harder to increase inversion and gain, the Auger upconversion process reduces the effective fluorescence lifetime of the crystal and limits the achievable inversion levels [6,19,61].

Fig. 3. Measured cw tuning performance of the Cr:LiSAF laser at 0.8 W absorbed pump power. The data is taken using six different output couplers: the reflectivity range of each OC is given in the figure legend.

3.2 Mode-locked lasing results

Fig. 4. Measured efficiency of the Cr:LiSAF laser in mode-locked regime using 0.15 %, 0.3 % and 0.5% transmitting output couplers. The regions where stable cw mode-locked (CWML) operation could be observed is marked. Inset figure shows a typical beam profile for the mode-locked laser.

We start presentation of the mode-locking results with Fig. 4, which shows the measured efficiency of the extended Cr:LiSAF laser cavity, which now also contains the SESAM. The data is taken with 0.15 %, 0.3 % and 0.5% transmitting output couplers. Comparing with Fig. 2, we see that, due to increased losses of the cavity with the insertion of the SESAM, the lasing threshold of the system increased dramatically. As an example, for the 0.15% transmitted output coupler, the lasing threshold increased from

around 100 mW to around 500 mW. Moreover, the laser slope efficiency with the same OC decreased from around 12% to around 2% (comparing Fig. 2 and 4). Due to increased losses of the system, we could only achieve a mode-locked average power of 12.5 mW. Transverse mode profile of the output beam was symmetric and circular with M^2 below 1.1 (inset picture in Fig. 4). Note that similar performance is observed (in terms of lasing threshold and slope efficiency) for all the output coupling values tested, confirming that the SESAM loss (small signal loss: $\sim 1.5\text{-}2\%$) is significantly higher than output-coupling ($0.15\text{-}0.5\%$) and determines the systems performance. Clearly, the commercially available SESAM used in this study, which had a modulation depth of 1.5% , is not an optimum choice for the low-gain Cr:LiSAF laser, and usage of a SESAM with a lower modulation depth and passive losses could improve the results considerably. We believe that, with an optimized SESAM, the mode-locked average powers could easily be scaled up to 75-100 mW for this 1 W diode pumped system.

Fig. 5. Measured temporal dynamics of the mode-locked Cr:LiSAF laser on different time scales: (a) 100 ns and (b) 500 μs showing stable cw mode-locked operation at around 150 MHz repetition rate.

Fig. 6. Radio frequency spectrum of the cw mode-locked Cr:LiSAF laser around the main RF line confirming clean mode-locked operation. The RF line is centered at 148.45 MHz: data is taken with a span of 2.5 MHz, and a resolution bandwidth of 10 Hz. Inset figure: RF spectrum covering the 0-1.6 GHz range, resolution bandwidth 10 kHz.

As we see in Fig.4, for all the output couplers, the system worked in cw regime for pump powers up to around 700 mW, and beyond that stable cw mode-locked operation could be achieved (indicated by CWML in Fig. 4). Mode-locking required a slight tapping to the SESAM mirror, but once mode-locked the system remained stable for hours. With the saturation of the SBR, the laser power also increased compared to the cw case as we see from the kink in the efficiency curves. The laser repetition rate was around 150 MHz, and the measured pulse train and radio-frequency spectra both confirmed clean cw mode-locked operation (Fig. 5 and Fig. 6).

Figure 7 shows the measured variation of laser pulsewidth and pulse energy as a function of pump power for each output coupling. As expected, with the increasing pump power the obtainable pulsewidths decrease for all output couplers. At the maximum pump power of around 1.1 W, pulsewidths below 150-fs were observed for all output couplings. The maximum achievable pulse energy was around 60 pJ for the 0.15% output coupler and was around 85 pJ for the 0.3% and 0.5% output couplers.

Fig. 7. Variation of measured output pulsewidth and pulse energy with absorbed pump power for 0.15%, 0.3% and 0.5% transmitting output couplers.

Figure 8 shows sample optical spectra acquired when mode-locked. As an example, the optical spectra taken using the 0.3% output coupler at different pump powers will be presented here but the overall trend was similar for other output couplers as well. In Fig. 8, the narrow spectrum corresponds to the cw lasing case, which is centered around 1016 nm. The broader spectra are taken during mode-locking and shows how the optical spectra varies with pump power in the 800-1100 mW absorbed pump power range. As expected, with increasing pump power or pulse energy, the spectra get wider (Fig. 8), and the corresponding pulsewidths get shorter (Fig. 7). On top of this, we have also observed a slight shift of central wavelength of the pulses to longer wavelength (from 1006 nm to 1010 nm) with increasing pump power. We

believe this wavelength shift might be due to residual undesired birefringence induced by the relatively long (15 mm) Cr:LiSAF crystal due a small-tilt error in its placement. Note that, the estimated total intracavity dispersion of the laser is also shown in Fig. 8, and the net cavity dispersion was around -550 fs^2 . Our simultaneous optical spectra and pulsewidth measurements showed that, for all the cases, the pulses are close to transform limited performance. As an example, Fig. 9 shows the measured pulsewidth of the Cr:LiSAF laser with the 0.3% OC, at an absorbed pump power level of 1085 mW. At this setting the laser produced 12.5 mW of output power. The optical spectrum was centered around 1010 nm and had a FWHM of 8 nm. Assuming Sech² pulse shape, this spectrum supports 125 fs level pulses, where the measured autocorrelation trace indicated a 140 fs pulsewidth. This shows that the time bandwidth product of the pulses is around 0.35, slightly above the ideal 0.315 value.

Fig. 8. Sample mode-locked spectra of the mode-locked Cr:LiSAF laser taken at different pump power levels. The free running cw spectrum is also shown for comparison (narrow spectrum). The optical spectra of the pulses get broader and their central wavelength get slightly red-shifted with increasing pump power. Estimated total cavity dispersion is also shown. Data is taken with the 0.3% transmitting output coupler.

Fig. 9. Measured background-free autocorrelation trace for the 140 fs, 80 pJ pulses. Data is taken with the 0.5% transmitting output coupler at an absorbed pump power of 1 W.

We have also checked tunability of the femtosecond pulses via inserting a 0.2 mm thick birefringent filter into the cavity. For the tuning experiments, the negative cavity dispersion is increased to -800 fs^2 via including an additional DCM. With proper adjustment of the BRF rotation angle, the central wavelength of the pulses could be tuned between 1000 and 1020 nm (Fig. 10). The average mode-locked output power stayed around 10 mW in all cases. The pulsewidth stayed mostly below 200 fs except the edges of tuning. Fig. 10 also show the measured small signal reflectivity of the SESAM at 5 degree incidence angle. As we can see, the fs tuning range is limited by the reflectivity edge of the Bragg stack of the SESAM below 1000 nm. Clearly, using SESAMs with reflectivity band covering the 900-1000 nm region: fs tuning of Cr:LiSAF should be possible in this region in future work. On the long wavelength side, tuning is limited by the losses of the SESAM. As we also see earlier in cw tuning experiments (Fig. 3), the losses of the Cr:LiSAF laser should be reduced below 1% to achieve lasing and fs tuning above 1020 nm region. As we discussed earlier, by employing a SESAM with optimized properties (especially reduced losses), this should be feasible in future work.

Fig. 10. Typical spectra from the Cr:LiSAF laser, showing tunability of the central wavelength from 1000 to 1020 nm with sub-200-fs pulse duration. Data is taken with the 0.5% transmitting output coupler at an absorbed pump power level around 1 W, and at an estimated total cavity dispersion of -800 fs^2 . The measured reflectivity of the SBR is also shown.

To check the limitations of the system in terms of obtainable pulsewidths, we have first decreased the net cavity dispersion to the -300 fs^2 level by replacing one of the DCMs in Fig. 1 with a regular high reflector mirror. For this lower dispersion setting, we could achieve pulses as short as 110 fs with 70 pJ pulse energy using the 0.5% transmitting output coupler (Fig. 11). The pulses were centered around 1013.4 nm, and had a FWHM of around 10 nm. Assuming a Sech² pulse shape, the time-bandwidth product of the pulses is estimated to be 0.32, very close to the ideal value. The average mode-locked laser power was 10.3 mW and the corresponding peak power was 560 W for 148 MHz repetition rate. When we tried to reduce the pulsewidth further by pumping the system more, the system started to generate multiple pulses, and this was evident both from the autocorrelation trace and optical spectrum. For these short pulses, the cavity had an

intracavity pulse energy of around 15 nJ, and the fluence on the SBR is estimated to reach $\sim 500 \mu\text{J}/\text{cm}^2$, which is around 15 times higher than the specified saturation fluence of the SESAM ($35 \mu\text{J}/\text{cm}^2$). Hence, for short pulses, presence of double-pulsing instabilities is expected at these conditions due to reverse saturable absorber action of the SESAM via the two-photon absorption process [28,62].

Fig. 11. Measured optical spectrum (a) and autocorrelation trace (b) for the 110 fs 70 pJ pulses from the Cr:LiSAF laser. The estimated total cavity dispersion is also shown. Data is taken with the 0.5% transmitting output coupler at an absorbed pump power level around 1 W.

4. Conclusion

To our knowledge, we have reported first experimental results on mode-locking of Cr:LiSAF lasers around 1000 nm. A SESAM with a modulation depth of 1.5% was used for mode-locking. Stable and robust mode-locked operation with sub-200-fs long pulses in the 1000-1020 nm range was achieved. Multiple pulsing instabilities limited the pulsewidth to the 110-fs level, whereas the relatively high losses of this specific SESAM limited average power performance of the system as well as the fs tuning range.

Due to the lower gain of Cr:LiSAF around 1000 nm, the system is operated using output coupling in the 0.15-0.5% range. Hence, we believe that ideally a SESAM with a

modulation depth of 0.25-0.5% should be sufficient for mode-locking in this spectral region. Reducing the modulation depth of the SESAM could also reduce its passive losses. We believe that, with an improved SESAM design, diode-pumped Cr:LiSAF oscillators have the potential to generate tunable sub-100-fs long pulses with nJ level energies around 1000 nm, which is of great interest in seeding cryogenic Yb:YLF based amplifiers.

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