



Intra-cavity frequency-doubled Cr:LiCAF laser with 265 mW continuous-wave blue (395–405 nm) output

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ABSTRACT

We describe continuous-wave (cw) intracavity frequency-doubling experiments performed with a Cr:LiCAF laser. The Cr:LiCAF crystal is home-grown and had passive losses below 0.15% per cm. The laser is pumped by two recently-developed high-brightness tapered diodes, providing a total pump power of 2 W at 680 nm. The Cr:LiCAF laser generated up to 585 mW of cw output power around 800 nm with 43% slope efficiency at an absorbed pump power of 1.4 W. The low passive losses of the crystal enabled storage of up to 380 W of intracavity laser power using a 0.07% transmitting output coupler, demonstrating suitability of Cr:LiCAF gain media for intracavity nonlinear conversion experiments. By performing intracavity frequency doubling with a BBO crystal, cw second-harmonic powers as high as 265 mW around 400 nm have been realized with optical-to-optical conversion efficiencies as high as 13.3%. To our knowledge, these are the highest cw frequency-doubled laser powers and conversion efficiencies obtained from Cr:Colquiriites to date. Moreover, obtained efficiencies are superior compared to what have been achieved with similar Ti:Sapphire systems, due to lower passive losses of Cr:LiCAF crystal. These results demonstrate the appropriateness of Cr:LiCAF gain media as a high-power tunable cw radiation generator in 375–435 nm region.

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1. Introduction

Cr³⁺:Colquiriites are a family of chromium doped laser gain media, in which fluorides of general formula Li Me^{II} Me^{III} F₆ serve as a host (Me^{II}=Ca, Sr, Cd; Me^{III}=Al, Ga, Ti, V, Cr, Fe). In particular Cr³⁺:LiSrAlF₆ (Cr:LiSAF) [1], Cr³⁺:LiCaAlF₆ (Cr:LiCAF) [2] and Cr³⁺:LiSrGaF₆ (Cr:LiSGaF) [3] have been investigated extensively as broadly tunable laser gain media in the near infrared. Favorable properties of Cr³⁺:Colquiriites include (i) broad absorption bands around 650 nm enabling direct diode pumping by low-cost diodes [4–10], (ii) capability of producing high quality crystals with low passive losses (a figure of merit of above 2000) [10,11], (iii) a high fluorescence lifetime and emission cross section product value, enabling lasing thresholds of only a few mW [9,10], (iv) low quantum defect and reasonably low excited state absorption values facilitating efficient laser operation with slope efficiencies above 50% [9,10], and (vi) broad emission bandwidths enabling broadband wavelength tuning [9,10] and sub-10-fs pulse generation [12–14]. These features empowered construction of low-cost and efficient continuous-wave (cw) and

femtosecond laser systems from Cr:Colquiriites in the near infrared region (720–1110 nm).

Blue and ultraviolet regions of the optical spectrum are quite important for many applications, including spectroscopy, optical imaging, remote atmospheric sensing, medical diagnosis, undersea communications and information storage [15]. However, there are only a few sources available in this wavelength range (GaN/GalnN diodes [16,17], Ce-doped lasers [18,19], thulium doped upconversion lasers, etc. [20], second/third harmonic of Nd-based lasers [21], Ti:Sapphire [22] and GaAsP based diodes [23]), and these sources have several shortcomings, including limited tunability, high-cost, complexity, low efficiency, disability in producing cw output, etc. As an attractive alternative, low-cost, broadly tunable cw laser sources in this spectral range can be obtained via frequency doubling of Cr: Colquiriites.

Earlier frequency-doubled cw sources based on Cr:Colquiriites are mostly based on Cr:LiSAF gain medium. Using intracavity frequency doubling, cw blue powers as high as 120 mW have been obtained around 430 nm [24], and cw blue tuning ranges from 427 nm to 443 nm have been demonstrated [25]. Thermal effects in Cr:LiSAF have been shown to be the limiting factor for further power scaling [26]. These earlier findings motivated us to focus on Cr:LiCAF gain media for the development of efficient high-power blue/ultraviolet cw sources. Cr:LiCAF has much better

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thermal specifications than those of Cr:LiSAF (higher thermal conductivity, lower quantum defect, lower Auger rate, etc.) and hence it is more suitable for applications requiring higher power levels [27]. Moreover, the emission spectrum of Cr:LiCAF is blue-shifted compared to Cr:LiSAF, and its second harmonic can reach deeper into the ultraviolet region. Earlier work with Cr:LiCAF gain media reported cw blue tuning in the 375–433 nm range [10]. However, obtained blue powers were limited to only 3.5 mW [10].

In this paper, we will present an intracavity frequency-doubled Cr:LiCAF laser that is pumped by two high brightness 1-W tapered diodes at 680 nm. A high quality Cr:LiCAF crystal with passive losses below 0.15% per cm was used as the gain medium which allowed the construction of a high-Q-cavity that can store intracavity laser powers approaching 400 W around 800 nm. Intracavity frequency doubling with a Beta-barium borate (BBO) crystal enabled generation of cw blue powers up to 265 mW in the 390–410 nm range. The optical-to-optical conversion efficiency of the 680 nm pump beam to the second-harmonic power was above 13%. To our knowledge, demonstrated blue powers and the conversion efficiencies in this work are better than what have been reported from any Cr:Colquiriite based source to date. Moreover, the efficiency levels are higher than what can be achieved by intracavity frequency doubled Ti:Sapphire laser systems due to the advantages of Cr:Colquiriite in terms of passive losses. These results indicate that nonlinear sources based on intracavity frequency conversion within low-cost Cr:Colquiriite laser systems have the potential to efficiently generate broadly tunable radiation at different regions of the optical spectrum, including blue, ultraviolet and near infrared (via optical parametric conversion) [28].

2. Experimental setup

Fig. 1 shows a simplified schematic of the Cr:LiCAF laser that is used in intracavity cw blue generation experiments. Two linearly-polarized tapered diode lasers (TDL-1 and TDL-2) that were developed at Ferdinand Braun were used as the pump source [29]. The TDLs provided up to 1-W of output power at 680 nm, with an output beam quality (M^2) of 1.1 and 5 along the fast and slow axes, respectively. This corresponds to a brightness of around $500 \text{ mW}/\mu\text{m}^2$ ($50 \text{ W}/\text{cm}^2$) that is 2 folds better compared to commercially available diodes. The diode outputs were first collimated using 4.5 mm focal length aspheric lenses. Then cylindrical lenses with a focal length of 50 mm were used to match the divergences of the

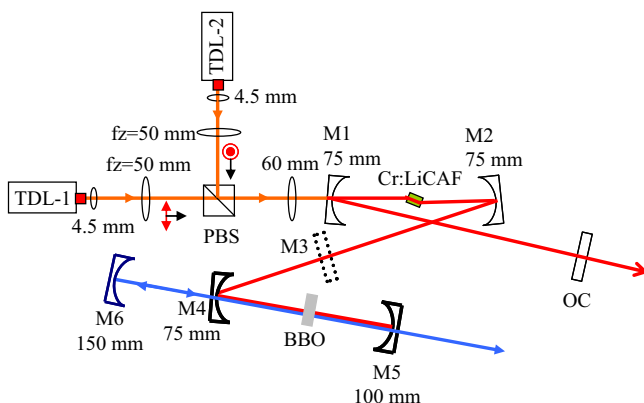


Fig. 1. Schematic of the cw Cr:LiCAF laser setup that was used in intracavity second harmonic generation experiments. The x-cavity is end-pumped by two 1-W tapered diode lasers (TDL-1 and TDL-2) at 680 nm. The dashed mirror (M3) is used in the regular cw laser experiments. For intracavity blue generation experiments, M3 is removed and mirrors M4–M6 are inserted. PBS: polarizing beam splitter cube. OC: output coupler.

pump beams along the fast and slow axes. The diode outputs were combined using a polarizing beam-splitter (PBS) cube. A 60 mm achromatic doublet lens was used to focus the pump beam inside the Cr:LiCAF crystal to a spot size of around $25 \mu\text{m}$. The gain medium was a Brewster–Brewster cut, 4-mm long, 1-mm thick Cr:LiCAF crystal with a chromium concentration of 5%. The crystal was grown using the Czochralski method using slow pulling rates, and had passive losses below 0.15% per cm. It absorbed 85% and 74% of the TM and TE polarized pump light at 680 nm, respectively. Note that around 10% of the TE polarized light is reflected from the Cr:LiCAF crystal surface. With this the total absorbed pump power by the crystal reduced to 1.4 W level.

The Cr:LiCAF laser resonator was a simple x-shaped cavity with astigmatism compensation, and consisted of two curved pump mirrors (M1–M2) with a radius of curvature of 75 mm, a flat high reflector (M3) and a flat output coupler (OC). For intracavity cw blue generation experiments, the cavity was extended, and two more curved high reflective (HR) mirrors (M4 and M5) have been inserted to generate a second focus within the cavity, which is used to place the nonlinear crystal. A 2-mm long BBO crystal has been used for frequency doubling. The crystal was optimized for second harmonic generation at 800 nm (cut angle: 29.2°), and had antireflection coatings in both the fundamental and the second harmonic wavelength regions. Mirrors M4 and M5 transmitted around 90% of the generated blue light, and acted as output couplers at the harmonic wavelength. To obtain a single blue-output, the blue power transmitted from M4 is retro-reflected back using a curved metallic high reflector with a radius of curvature of 150 mm (M6).

3. Experimental results

Fig. 2 shows the measured variation of the Cr:LiCAF laser power as a function of absorbed pump power. The data is taken using three different output couplers with transmissions ranging from 0.07% to 0.75%. The Cr:LiCAF crystal holder temperature was 15°C (see [8] for a detailed discussion on the effect of crystal temperature on cw laser performance). The free running laser wavelength was around 800 nm. Note that the efficiency curves in Fig. 2 are not perfectly linear. We believe that the nonlinearity in the data is possibly due to (i) thermal effects and (ii) changing beam quality/shape of the tapered diode laser with current (beam quality deteriorates with increasing current, which then negatively affects the mode matching between the pump and the laser modes). Using the 0.75% output coupler, a slope efficiency of 43% and a lasing threshold of around 150 mW have been measured from the Cr:LiCAF laser. The output power was as high as 585 mW, at an

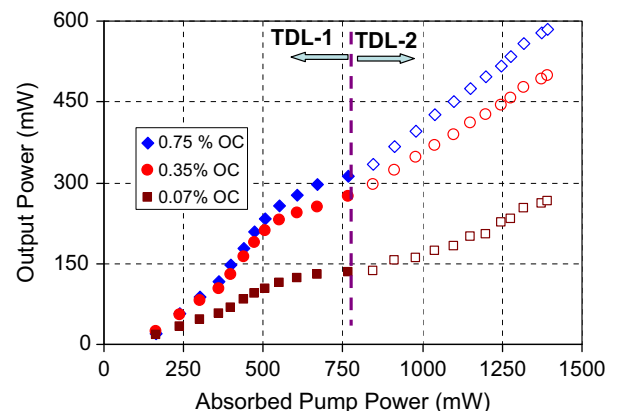


Fig. 2. Measured continuous-wave laser performance of Cr:LiCAF laser taken using different output couplers (OCs) with transmissions ranging from 0.07% to 0.75%.

absorbed pump power level of 1.4 W. Corresponding intracavity power levels reached 75 W level. The obtained output power levels were limited by the available pump power amount. The laser output beam profile was slightly multimodal with an M^2 less than 1.1 along the slow axis and around 3 along the fast axis (at the full pump power level).

We have also measured the cw laser performance at very low output coupling values (0.07%) to investigate the storage capability of the laser oscillator. As expected, the obtainable laser power and slope efficiency for this low output coupling decreased, and were only 265 mW and 19%, respectively. On the other hand, stored intracavity laser powers increased to 380 W level. Acquirable intracavity power level is a good indication that the intracavity losses are at a quite low level for our cavity. As a more direct approach, using the Findlay–Clay [30] and Caird analysis [31], we have calculated the total round-trip passive losses of the cavity to be $(0.15 \pm 0.05)\%$. It is this low passive loss level that enables efficient storage of energy inside the Cr:LiCAF laser cavity using low output coupling. This is mostly the result of the high quality Cr:LiCAF crystal that we have employed in our study, which had passive losses below 0.15% per cm. These values show that, due to their low passive losses, Cr:Colquiriites are quite attractive for intracavity nonlinear frequency conversion experiments [28]. This gives them a significant advantage compared to Ti:Sapphire, which is known to possess higher amount of parasitic losses [32].

As mentioned in the experimental part, for the intracavity cw blue generation experiments, we have removed the flat HR (M3), and used an extended cavity containing a BBO crystal at a secondary focus. The output coupler is also replaced with a HR mirror to boost the intracavity powers. Fig. 3 shows the measured blue power levels as a function of absorbed diode pump power. At the maximum absorbed pump power level of 1.4 W, up to 265 mW of cw second harmonic power has been observed. This corresponds to an optical-to-optical conversion efficiency of 13.3% with respect to incident pump power and 18.9% with respect to absorbed pump power level. Note that estimated intracavity power levels are as high as 200 W (calculated by measuring the leaked laser power amount from the HRs). This is lower than what was obtained in the regular cw cavity due to additional losses from the insertion of BBO crystal and other cavity optics (M4–M5). The conversion efficiency from the fundamental to the second harmonic is also at a reasonable level (0.13%). Note that, according to the intracavity optical second harmonic generation theory presented by R.G. Smith [33], maximum obtainable second harmonic powers could be as high as the maximum available fundamental power

available from the laser. In our case, the obtained second-harmonic powers (265 mW) are around 45% of the fundamental power available from the laser (585 mW). Hence, we believe that, the obtained efficiency in this study could be potentially doubled by using a nonlinear crystal with minimal insertion losses and also by optimizing the nonlinear conversion process (optimizing the focus size, crystal length, laser linewidth, etc.).

At this point, we would like to compare the intracavity cw frequency doubling experimental results obtained in this study with similar earlier studies performed with Cr:Colquiriites. For that purpose, we have prepared Table 1, which gives a summary of earlier results, by listing important key parameters such as pump source/power, second-harmonic (SH) crystal/power/wavelength and the obtained optical-to-optical (o-to-o) conversion efficiency. We have also included a few representative results obtained with other gain media such as Ti:Sapphire and Nd:YAG for comparison purposes. First of all, note that most of the earlier intracavity cw frequency doubling experiments with Cr:Colquiriites have used Cr:LiSAF as the gain medium. The reason might be the lower passive loss level of Cr:LiSAF during those days, which enabled more efficient storage of intracavity power, and hence better performance in nonlinear conversion. However, we have shown that, Cr:LiCAF crystals with passive losses below 0.15% per cm could be produced by optimizing crystal growth parameters [11]. The high quality Cr:LiCAF crystal that was used in our work enabled efficient energy storage within the cavity, which in turn facilitated efficient generation of second harmonic signal. To the best of our knowledge, the obtained second harmonic signal power levels in this study (265 mW) are the highest obtained from Cr:Colquiriite systems to date. Moreover, demonstrated optical-to-optical conversion efficiency (13.3%) is the highest among Cr:Colquiriites and also higher than/comparable with what have been reported with other gain media (Table 1). We believe that our optical-to-optical conversion efficiency can be further scaled up by using a Cr:LiCAF crystal with higher absorption, by using higher power diodes, and/or by optimizing the second harmonic generation process (by using a nonlinear crystal with lower insertion loss, etc.).

Also note from Table 1 that Nd-based intracavity frequency-doubling systems based on well-known hosts such as Nd:YVO₄, are capable of generating more than 50 W of frequency-doubled laser power with efficiencies approaching 30% [39]. However, their narrow gain bandwidths do not allow broad tuning of the frequency doubled radiation, and can produce emission at only fixed lines around 532 nm, 469 nm, 473 nm, etc. On the other hand, Nd-based systems incorporating host lattices with structural disorder can enable broader tuning ranges up to 40 nm; however, demonstrated optical-to-optical conversion efficiencies of these systems are quite low (on the order of 0.1%) [41,42].

Hence, for applications that require broadly tunable coherent light sources in the blue and ultraviolet regions, Cr:Colquiriites and Ti:Sapphire are among the few promising candidates. On the other hand, passive losses of Ti:Sapphire crystals are quite high (2% per cm for reasonably doped crystals [32]) compared to Cr:Colquiriites (0.15% per cm), resulting in lower frequency doubling efficiencies in the intracavity frequency-doubling process (6–7%) even using higher pump power levels reaching 10 W [22,43]. Hence, our results show that Cr:LiCAF is a promising gain medium for efficient generation of broadly tunable radiation in the blue and ultraviolet spectral regions via intracavity frequency doubling. Moreover, with ongoing progress in diode technology, we believe that output power levels from Cr:LiCAF laser systems could be scaled up to several watts level, which is currently limited by the brightness of pump diodes.

We would like to point out that, in our study, the second harmonic emission wavelength could be tuned from about 395 nm to 405 nm by only playing with the tilts of the BBO crystal and cavity optics (typical optical spectrum is shown in the inset figure,

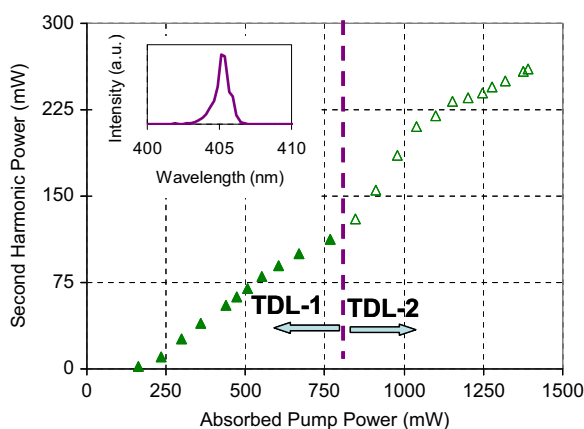


Fig. 3. Measured variation of second harmonic power as a function of absorbed pump power. Inset figure shows typical optical spectrum for the generated second harmonic. The spectrum is centered around 405 nm and has a full width at half maximum of 0.7 nm.

Table 1
Summary of cw intracavity second harmonic generation experiments performed with Cr:Colquiriites.

Material	Pump source	Pump power (W)	SH crystal	SH power (mW)	SH wavelength (nm)	o-to-o efficiency (%)	Year	Ref.
Cr:LiSAF	Multimode diode	1 × 0.5 1 × 0.4	KNbO ₃	13	435 (427–443)	1.4	95	[25]
Cr:LiSAF	Multimode diode (fiber bundle)	4	LBO	20	435 (432–442)	0.5	97	[26]
Cr:LiSAF	Multimode diode	0.35	KNbO ₃	0.37	430	1.1	98	[34]
Cr:LiSAF	Multimode diode	1	LBO	67.8	430	6.8	0	[35]
Cr:LiSAF	Multimode diode	1.2	LBO	120	430	10	1	[24]
Cr:LiSAF	Multimode diode	1	LBO	32	430	3.2	2	[36]
Cr:LiCAF	Single-mode diode	0.145	BBO	3.5	400 (375–433)	2.4	12	[10]
Cr:LiCAF	Tapered diode	2x1	BBO	265	395–405	13.3	13	^a
Nd:YAG	Fiber coupled diode	23	PPKTP	500	473	2.2	3	[21]
Nd:YAG	Fiber coupled diode	23	PPKTP	200	469	0.9	3	[21]
Nd:GdVO ₄	Fiber coupled diode	30	LBO	5300	456	17.7	6	[37]
Nd:YVO ₄	Fiber coupled diode	30	LBO	4600	457	15.3	6	[38]
Nd:YVO ₄	Fiber coupled diode	210	LBO	62,000	532	29	7	[39]
Nd:GdVO ₄	Fiber coupled diode	25	KNbO ₃	300	440	1.2	8	[40]
Nd:SBN	Fiber coupled diode	1.2	SBN	< 1	455–462 525–545	< 0.1	5	[41]
Nd:BNN	Fiber coupled diode	0.8	BNN	1	535 (527–547)	0.1	6	[42]
Ti:Sapphire	Argon ion	7.2	LBO	460	398	6.4	95	[43]
Ti:Sapphire	532 nm	10	BiBO	690	423	6.9	7	[22]

Representative cw intracavity frequency doubling results obtained with other gain media have also been included. Table provides information on the materials used, pump source, pump power, second-harmonic (SH) crystal that is employed, obtained second-harmonic power and wavelength, and resulting optical-to-optical (o-to-o) conversion efficiency from the pump power to the second-harmonic-power.

^a Denotes the results obtained during this work.

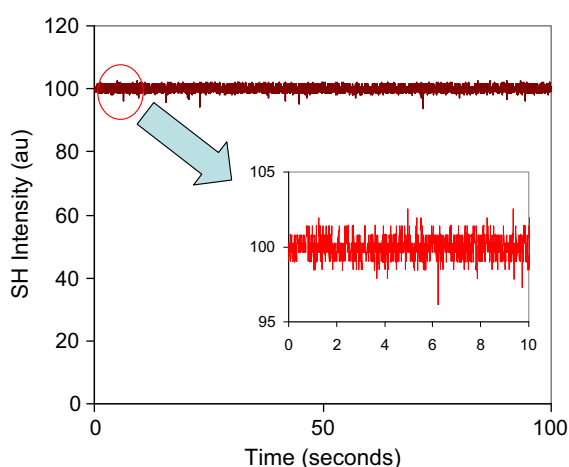


Fig. 4. Measured variation of blue intensity with time. Relative standard deviation is less than 1% in short time scales. Inset figure is a zoomed in version (shows the first 10 ms).

in Fig. 3). In this wavelength range (395–405 nm), the obtained second harmonic power levels were almost the same, due to the relatively flat gain profile of Cr:LiCAF medium in the 790–810 nm range. To obtain a broader tuning range, one can insert a birefringent tuning plate into the Cr:LiCAF cavity, which enables tuning of the fundamental laser wavelength from 745 nm to 885 nm, which in turn allows tuning of the second harmonic wavelength from around 375 nm to 435 nm. However, the obtained power levels at the edges of the tuning (away from 400 nm) are much lower, due to the additional reflection losses from the BBO crystal (optimization of second harmonic generation efficiency requires large tilt angles on BBO, which then creates high passive losses). Hence, besides the 800 nm optimized BBO crystal that was available in our study, usage of other nonlinear crystals with cut angles and antireflective coatings optimized for second harmonic generation at 750 nm, 775 nm, 825 nm and 850 nm could enable efficient second harmonic generation in the full tuning range of Cr:LiCAF medium.

As a final note, Fig. 4 shows the measured fluctuations in the generated second harmonic intensity. The intensity is normalized

to 100 for easier visualization. Inset figure is a zoomed in version. The data is taken with a 1 GHz Si photodiode, and monitored by a 500 MHz oscilloscope at a resolution of 10 ms. Note that the second harmonic intensity is quite stable with a relative standard deviation less than 1% in short time scales (minutes). On the other hand, we have observed larger fluctuations up to 10–20% in longer time scales. We have noticed that these fluctuations in the long time scales were caused by the shift of the Cr:LiCAF laser wavelength due to thermal effects. As a solution to this problem, we have inserted a birefringent tuning plate inside the cavity for wavelength stabilization, which also greatly improved the long term stability of the generated second-harmonic intensity. Enclosing the laser and stabilization of the BBO crystal temperature could provide further improvement in laser noise performance.

4. Summary

In summary, we have investigated cw intracavity frequency doubling effectiveness of a diode pumped Cr:LiCAF laser system. The cw Cr:LiCAF laser was pumped by two 1-W tapered diodes, and produced up to 585 mW of output power at the fundamental wavelength of 800 nm (using 1.4 W of absorbed pump power). Via intracavity frequency doubling in cw regime, up to 265 mW of second harmonic power around 400 nm has been obtained. The optical-to-optical conversion efficiency of the system was 13.3% and 18.9%, with respect to incident and absorbed pump power, respectively. To the best of our knowledge, these are the highest cw frequency-doubled power levels and optical-to-optical conversion efficiencies obtained with Cr:Colquiriite gain media. Compared to similar Ti:Sapphire systems, higher efficiencies could be achieved with Cr:Colquiriites due to lower amount of passive losses. We believe that low-cost, diode-pumped Cr:LiCAF laser based tunable cw sources in the blue and ultraviolet regions are attractive systems for many scientific and technological applications.

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