A 380-mW 7-kHz Cerium LiLuF Laser Pumped by the Frequency Doubled Yellow Output of a Copper–Vapor–Laser

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Abstract—We report a high average power Ce:LiLuF laser pumped by the second harmonic of the yellow output from a copper vapor laser. This cerium laser yielded up to 380 mW at a pulse repetition frequency of 7 kHz at the peak lasing wavelength of 309.5 nm, with a slope efficiency of 50%. In addition, single prism tunability was obtained from 305.5 to 316 nm and from 322 to 331.8 nm. Preliminary investigation into color center behavior has been performed involving crystal cooling, He–Ne pump probe experiments and antisolvent pumping.

Index Terms—Copper materials/devices, laser crystals, laser tuning, second-harmonic generation, ultraviolet generation.

I. INTRODUCTION

UNABLE UV laser sources in the 250–350-nm spectral range are very attractive tools for applications as diverse as remote sensing of atmospheric species, combustion diagnostics, medical, and biological applications and photolithography. There is now a wide spectrum of methods used to achieve tunable UV output including optical parametric oscillators, frequency-tripled titanium sapphire and Cr:LiSAF lasers and frequency-doubled dye lasers. An alternative and extremely attractive approach to obtaining tunable coherent UV radiation is by means of cerium-doped fluoride lasers that are directly tunable in the UV [1]. Not only does this approach offer the attraction of direct generation of UV output at the wavelength region of interest, but also such systems are extremely simple and compact in comparison to alternative methods of tunable UV laser generation. A further advantage of cerium lasers is their potential for the generation and amplification of short pulses [1], [2].

The prospect of direct UV tunable laser operation based on interconfigurational $5d \rightarrow 4f$ transitions in rare-earth ions such as cerium was identified more than 20 years ago by Yang and DeLuca [3]. It was not long after this that the first cerium laser (Ce:YLF) based on this scheme was practically realized [4]. Since then a large number of cerium activated fluorides and oxides have been investigated, however prior to 1992 only two of these media were found to be laser-active [4], [5]. This lack of success was attributed to excited state absorption (ESA) at both the pump and laser wavelengths and the formation of transient and stable color centers [6], [7], so consequently it was felt for many years that this approach to generating tunable UV laser output offered little promise. However, recently with the discovery of Ce:LiCaAlF$_6$ (Ce:LiCAF) [8], Ce:LiSrAlF$_6$ (Ce:LiSAF) [9], [10] and Ce:LiLuF$_4$ (Ce:LiLuF) [11] it has been shown that given the right choice of crystal host and high crystal purity, efficient and stable operation of cerium-doped lasers can be achieved with minimal loss from ESA and solarization.

So far, interest has been centered on Ce:LiCAF and Ce:LiSAF due to the mature level of the crystal growth procedures developed for Cr:LiCAF and Cr:LiSAF and the ready availability of pump sources in the form of frequency quadrupled Nd$^{3+}$ lasers [12], [13]. Due to these factors, to date Ce:LiCAF and Ce:LiSAF, rather than Ce:LiLuF, have been developed for applications in the areas of remote sensing of atmospheric species [14] and combustion diagnostics [15]. Ce:LiLuF has three main absorption bands in the 200–300 nm spectral region with absorption peaks at around 210, 245, and 290 nm [16]. Ce:LiLuF lasers have been pumped in the past by KrF excimer lasers at 248 nm [1], [2], [11], and more recently by frequency-quintupled Nd:YAG lasers at 213 nm [16] and frequency-quadrupled Nd:YAG pumped Ce:LiSAF lasers operating at 290 nm [17].

An excellent alternative Ce:LiLuF pump source is the frequency doubled copper vapor laser (CVL) at 289 nm [18], [19]. In the past, we have demonstrated the suitability of the frequency doubled CVL as a Ce:LiLuF pump source, generating up to 12 mW at a PRF of 6.2 kHz [20]. More recently, though we have improved on these results by developing a Ce:LiLuF laser that yielded 300 mW at a PRF of 7 kHz [21]. The green and yellow CVL fundamental lines (511 and 578 nm, respectively) can be frequency-doubled or sum frequency mixed to yield three UV wavelengths: 255 nm (frequency-doubled green), 271 nm (sum frequency mixed) or 289 nm (frequency-doubled yellow). The advantages of the frequency-doubled CVL as a Ce:LiLuF laser pump source are threefold. 1) Frequency doubling the CVL is a far simpler procedure than operating a frequency-quadrupled Nd:YAG pumped Ce:LiSAF laser and a very much more efficient operation than obtaining the fifth harmonic of a Nd:YAG laser (due to 213-nm absorption in the nonlinear crystal). 2) Pumping at the relatively long wavelength of 289 nm provides a very low quantum defect for efficient lasing and less potential for ESA and solarization problems. 3) The frequency-doubled CVL is a high average power UV
source providing typical UV output powers of 1–2 W at pulse repetition frequencies (PRF's) of tens of kilohertz. In addition to this, the wavelength agility of the frequency-doubled CVL means that a single CVL pump source can be used to pump a variety of cerium lasers. For instance, we have recently demonstrated a 530-mW 7-kHz Ce:LiCAF laser pumped by the CVL sum frequency mixed line at 271 nm [22]. While the frequency-doubled CVL may not necessarily be the best pump source for some applications where a compact all-solid-state system may be required, it is certainly an excellent source to characterize the Ce:LiLuF laser medium and thus to assist further development of high PRF cerium lasers.

In this paper we present results of high power, high PRF Ce:LiLuF lasers pumped by the frequency-doubled yellow output of a copper vapor laser.

II. EXPERIMENTAL CONFIGURATION

The experimental configuration for the frequency doubled CVL pumped Ce:LiLuF laser is shown in Fig. 1. An in-house built CVL operating at a PRF of 7 kHz of active volume 1 m long with a 25 mm diameter was used. This CVL was operated with an on-axis negative branch unstable resonator of magnification \( M = 128 \) consisting of a 3.2-m radius of curvature (ROC) high reflector and a 2.5-cm ROC concave reflector. A polarizing cube was placed next to the high reflector to ensure linearly polarized output. The optical geometry used to deliver the CVL pump beam into the nonlinear crystal is similar to that reported in [19]. A Keplerian mirror telescope was used to reduce the beam diameter from 25.4 to 3.75 mm. In order to boost the yellow output available for frequency doubling, a green to yellow converting dye cell [23] was placed near to the telescope focus.

Up to 7 W of yellow output was derived from this dye cell from 14.2 W of combined green and yellow input power. The compressed CVL beam was focused into the sBBO nonlinear crystal using a \( f = 50 \) mm cylindrical lens. The UV output was recollimated by another \( f = 50 \) mm cylindrical lens and a Pellin–Broca prism was used to separate the UV beam from the fundamental. The maximum UV power generated from the crystal was 1.63 W. We determined this by measuring the maximum power of 1.3 W after the turning mirror and then correcting for the measured losses associated with the recollimating lens, Pellin–Broca prism and turning mirror. In this case the average second-harmonic conversion efficiency was 23%. The pulselength of the 289-nm pulses was 7-ns full-width at half-maximum (FWHM) and the divergence of the 3.75-mm UV beam was found to be approximately 1 mrad.

The Ce:LiLuF crystal (Kazan State University) was 2 mm long, with a 0.8-wt% cerium-doping level, providing \( \sim 95\% \) pump absorption at 289 nm. The uncoated crystal was plane cut with the \( c \) axis contained in the plane of the end faces and the polarization of the pump laser was aligned to the \( c \) axis of the crystal. A brass mount, that could be cooled down to \(-3°C\), was used to conductively cool the Ce:LiLuF crystal.

The cerium laser cavity consisted of a flat output coupler and a curved (ROC = 250 mm) high reflector, with a cavity length of 50 mm. A range of output couplers were used of reflectivities in the range of 30% to 90% at the Ce:LiLuF laser wavelengths. The 289-nm pump beam was focused past the high reflector into the laser crystal using a \( f = 200 \) mm lens to form a focus in the crystal with a diameter of approximately 200 \( \mu \)m. A quasi-longitudinal pumping geometry was adopted due to the difficulty of manufacturing a dichroic mirror coating of high transmission at 289 nm and high reflectivity in the wavelength range of 306–331 nm and with a sufficiently high damage threshold. Prism tuned operation was achieved by inserting a fused silica Brewster cut prism into the cavity and rotating the high reflector to attain wavelength selectivity. The cavity length was also increased slightly to 65 mm to accommodate this prism.

III. RESULTS

A. Output Power Characteristics

The output power characteristics of the untuned Ce:LiLuF laser are shown in Fig. 2. Note that all pump powers quoted in this paper are those measured after the pump focusing lens, directly before the crystal. Output couplers in the reflectivity range \( R = 30\%–90\% \) were tried and the maximum output power of 380 mW (pulse energy of 54 \( \mu \)J) at a wavelength of 309.5 nm was obtained from the \( R = 50\% \) output coupler from 1.25 W of pump power. In this case, the threshold was 0.26 W and the slope efficiency was found to be 50%. The
output from the laser began to saturate above pump powers of 0.9 W. When the \( R = 80\% \) output coupler was utilized, the laser threshold was reduced to 0.15 W. However, the maximum output power obtained was only 365 mW (pulse energy of 52 \( \mu \)J) from a pump power of 1.07 W and the maximum slope efficiency was 45\%. The Ce:LiLuF laser beam was predominantly \( \pi \) polarized when the laser was operated with either the \( R = 50\% \) or \( R = 80\% \) output coupler. However, when the \( R = 90\% \) output coupler was used and when the laser was operating near threshold, it was observed that the laser was simultaneously lasing at 309.5 nm and at 327 nm and that the output at 309.5 nm was predominantly \( \pi \) polarized whilst the output at 327 nm was predominantly in the \( \sigma \) polarization. At higher pump powers, the laser only operated at 309.5 nm and the output was almost completely \( \pi \) polarized.

The output power from the Ce:LiLuF laser was found to decay with a timescale of around 100 s to a steady-state value of around 60\% of the initial output power when the crystal was not actively cooled. The original output power could always be recovered by blocking the pump beam for a period of 100 s before reapplying the pump beam. This decay in output power and the saturation and rollover in output power observed in Fig. 2 are likely to be related to the effects that the pump-induced crystal heating has on the formation and destruction of transient color centers. By actively cooling the crystal, the decay in Ce:LiLuF laser power was reduced and the steady state output power of the laser was increased. When the crystal was cooled to \(-3\) \( ^\circ \)C there was no observed decay in output power for pump powers of less than 0.9 W, although for higher pump powers decay in the output power was still evident. For instance, when the cerium laser was pumped with 1.24 W, the output power decreased from its initial value of 400 mW to the steady-state value of 335 mW shown in Fig. 2 when the laser was operated with the \( R = 80\% \) output coupler. Fig. 3 depicts how the steady-state output power from the cerium laser varied with the actively cooled crystal temperature when the crystal was pumped with 1 W. The Ce:LiLuF fluorescence lifetime (40 ns) was measured between \(-3\) \( ^\circ \)C–40 \( ^\circ \)C and was found to be independent of temperature in this temperature range. It should be noted that the output power values recorded in Figs. 2 and 5 were all steady-state values taken when the laser crystal was cooled to \(-3\) \( ^\circ \)C and that these output power values were all stable over a few hours of continuous operation.

The laser pulse shapes and crystal fluorescence, when the Ce:LiLuF laser (80\% reflectivity output coupler) was pumped with 0.9 W of pump power (6 times threshold), are shown in Fig. 4. The pulse length of the Ce:LiLuF laser pulse was 3 ns, corresponding to as much as 18 kW of peak power, and the pulse build up time was 2.4 ns. Upon lasing, the crystal fluorescence was depleted by 62\% relative to the fluorescence value when lasing was blocked.

### B. Prism Tunability

In the case of the prism tuned Ce:LiLuF laser, the \( R = 90\% \) output coupler was used in order to broaden the laser tuning curve as much as possible. The resulting tuning curve is shown in Fig. 5. Fig. 5 also shows the unpolarized fluorescence spectra of this Ce:LiLuF sample when it was excited by the 289-nm frequency-doubled yellow line from the CVL. It should also be noted that a Ce:LiLuF fluorescence spectrum...
was also taken when the crystal was pumped at 255 nm and that these two fluorescence spectra were identical.

The Ce:LiLuF laser was tunable from 305.5 to 315 nm and from 322 to 331.8 nm with peaks in the tuning curve at 309.5, 323, and 330.5 nm. The shorter wavelength section of the tuning curve was taken when pumping with 1.1 W and the maximum output power at 309.5 nm was 195 mW. The output power from the longer wavelength range in the tuning curve was found to be a maximum for 0.7 W of pump power yielding 50 mW at 323 nm and 90 mW at 330.5 mW, as is shown in Fig. 5. The linewidth of the prism-tuned Ce:LiLuF laser was measured to be of the order of 0.4 nm. Whilst the peak in the fluorescence spectra at around 310 nm corresponds well to the peak in the laser tuning curve at 309.5 nm, no such coincidence exists for the longer wavelength fluorescence peak around 327 nm.

In fact, at 327 nm, it was observed that there was a dip in output power and that in the vicinity of this dip that the laser oscillation was in the σ polarization even though the Brewster prism was aligned in a plane so as to favor π-polarized oscillation. There is no ground state absorption feature in this spectral region that could account for this dip in output power. The laser output around the tuning curve peaks at 309.5, 323, and 305.5 nm was found to be π polarized.

This polarization switching behavior was further investigated by rotating the prism through 90° and realigning the whole cavity to favor σ-polarized oscillation. The resulting tuning curve (σ-polarized output) is also shown in Fig. 5. This tuning curve was found to peak at 327 nm, the peak of the fluorescence curve, and yielded an output power of 18 mW at this wavelength. Note that the σ-polarized cavity configuration produced π-polarized output when tuned about the shorter wavelength peak (305.5 nm) even though the losses from the intracavity tuning prism were significantly higher for the π-polarization. These effects are currently being experimentally analyzed by doing pump probe polarization gain measurements across the spectral gain region to try and characterize the nature of the competition between the polarized modes.

C. Pump-Probe Experiments and Antisolant Pumping

Preliminary investigations have been made studying the transient absorption of a He–Ne probe beam at 633 nm in the pumped region of the Ce:LiLuF crystal at different temperatures. Whilst pump induced absorption of the He–Ne laser was observed when the crystal was at room temperature, cooling the crystal down to −3 °C virtually eliminated this absorption. The He–Ne absorption was found to be more pronounced when the He–Ne beam was σ polarized than when it was π polarized.

In the past, antisolarant pumping experiments using Nd:YAG lasers have been shown to dramatically increase the efficiency of Ce:LiSAF lasers [24], by bleaching the color centers with an additional 532-nm laser beam. In an attempt to see what effect antisolarant pumping would have on this Ce:LiLuF laser, we pumped the Ce:LiLuF laser crystal with the residual σ polarized 578-nm CVL yellow line left over from the frequency doubling while pumping with the 289 nm harmonic. The 578-nm beam was focused into the pumped volume of the Ce:LiLuF crystal using a f = 140-mm lens. Fig. 6 shows the output pulse shapes from the Ce:LiLuF laser (80% reflectivity output coupler) when it was pumped with 1.3 W of pump power, both with and without the antisolarant pump beam. It can be clearly seen that the effect of the antisolarant beam is significant, increasing the peak pulse amplitude by 30%. Further experiments are underway to try and further quantify this effect.

IV. Discussion

In this paper, we have demonstrated that the frequency doubled CVL is an excellent pump source for high power multikilohertz PRF Ce:LiLuF lasers. In fact, the Ce:LiLuF laser reported here has, we believe, the highest output power and the broadest tunability of any Ce:LiLuF laser ever operated. To the best of our knowledge, the highest slope efficiency reported from any cerium laser is 55%, obtained from a 20-Hz PRF Ce:LiSAF pumped Ce:LiLuF laser [17]. The highest slope efficiency that we recorded of 50% compares very favorably with this, given that we are working at elevated (multikilohertz) PRF’s where color center effects are likely to be considerably more pronounced than they are at 20 Hz (transient color centers with a lifetime of the order of 50 ms have been measured for our Ce:LiLuF crystal [25]). The effect of the high Ce:LiLuF fluorescence yield (88%) and the high quantum efficiency (93% for 289 nm pumping) should combine to produce high slope efficiencies. However, ESA, the relatively short upper laser level lifetime (40 ns), losses from the uncoated crystal faces, and the 4.4% measured single-pass passive absorption/scattering losses, all act in addition to the effects of transient color center absorption to reduce the slope efficiency to the experimentally measured values.

The rollover in laser efficiency at high pump powers, that occurs on a time scale of a few tens of seconds, suggests that the formation and buildup of color centers comprises the major output power limiting mechanism at high powers. Evidence for the existence of the color centers is apparent from the absorption at 633 nm of the He–Ne probe beam in the pumped region of the Ce:LiLuF laser crystal. The improvement in laser performance with the addition of a 578-nm antisolarant
pump is further evidence for the existence of deleterious color centers (note that antisolarant pumping is expected to have no effect on ESA). It is not clear yet what role the temperature plays in the formation and deactivation of the transient color centers. Lim and Hamilton [26] observed that heating Ce:YLF reduced color center lifetimes via a thermal deactivation process, whereas we observed enhanced performance of our Ce:LiLuF laser at reduced temperatures. It is possible that a temperature dependence in the formation of transient color centers dominates over any temperature dependence in color center lifetime. This saturation and output power rollover are in direct contrast to what we have observed when we have used the sum frequency mixed CVL line at 271 nm to pump a 7-kHz PRF Ce:LiCAF laser [22]. This Ce:LiCAF laser yielded up to 530 mW from a pump power of 1.9 W (eight times above threshold), with no observed saturation of output power.

The untuned Ce:LiLuF laser with the 90% reflectivity output coupler lased simultaneously at 309.5 nm (σ-polarization) and at 327 nm (π-polarization), for low pump fluences yet only at 309.5 nm for higher pump powers. The threshold output polarizations follow the observation that the ratio of σ-polarized emission to π polarized emission is far greater for the long-wavelength peak in the fluorescence spectrum, than it is for the shorter wavelength peak [17]. He–Ne pump probe experiments that we performed revealed a color center-induced absorption at 633 nm, that was more pronounced in the σ polarization than in the π polarization. This anisotropy in the absorption at 633 nm has also been observed when this Ce:LiLuF crystal was pumped at 248 nm by a KrF excimer laser [25]. If this polarization dependent color center absorption is preserved at the cerium laser wavelengths, as is the case in Ce:LiCAF and Ce:LiSAF [10], then this will explain why the untuned Ce:LiLuF laser with the 90% reflectivity output coupler only oscillates at 327 nm for low pump fluences. At higher pump powers, the pump induced color centers quench the gain on the predominantly σ-polarized 327-nm transition far more powerfully than they do on the 309.5-nm transition which usually lases π-polarized.

Tunability in both wavelength ranges was greater than previously reported [16], [17]; however, the long-wavelength tuning curve region for the π-polarized cavity configuration had a dip at around 327 nm, corresponding to the long-wavelength maximum in the fluorescence spectrum. This dip in the tuning curve was because the laser oscillation switched to the σ-polarization. It is as yet unclear why maximum output power was obtained from the longer wavelength section of the tuning curve when pumping with only 0.7 W when the output power from the shorter wavelengths around 309.5 nm was increased by pumping with 1.1 W.

V. Conclusion

We have demonstrated efficient (50% slope efficiency) high power (380 mW) operation of a high pulse repetition rate (7-kHz PRF) Ce:LiLuF laser pumped by the frequency-doubled yellow output from a copper vapor laser. By inserting a prism into the cavity, tunability was demonstrated from 305.5 nm to 315 nm and from 322 nm to 331.8 nm. To the best of our knowledge, this laser has the highest output power and broadest tunability yet demonstrated from any Ce:LiLuF laser. These results complement the recent report of a low pulse rate (20 Hz PRF) Ce:LiSAF pumped Ce:LiLuF laser, with a slope efficiency of 55% [17], in demonstrating that Ce:LiLuF is a more efficient laser medium than the more familiar Ce:LiCAF or Ce:LiSAF.

These results are very encouraging even given the evidence for the formation of color centers that are absorbing at the Ce:LiLuF laser wavelengths, that are thus deleterious to laser performance. Use of an antisolarant pump at 578 nm (the CVL fundamental) was found to increase the cerium laser output power considerably. It is the authors belief that, with improved crystal growth to try and minimize color center formation and with the appropriate operating conditions, i.e., with an antisolarant pump, Ce:LiLuF could be a very effective practical tunable UV laser source, for extending the wavelength coverage of practical tunable cerium lasers.

REFERENCES
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