

Broadly tunable ultraviolet miniature cerium-doped LiLuF lasers

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Abstract: Ultraviolet (UV) miniature cerium fluoride lasers have been demonstrated using a low cost, frequency-quadrupled microchip Nd:YAG pump laser. The use of miniature laser cavities was shown to significantly improve the laser performance in the low pump power region. We have achieved slope efficiencies up to 70% and pump thresholds as low as 100 nJ. Continuous tuning from 306 nm to 338 nm was achieved using a Brewster angle prism.

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1. Introduction

Tunable lasers in the UV wavelength range are very useful tools in scientific and industrial fields including UV spectroscopy, remote sensing, and fluorescence sensing in biological research. Cerium doped fluoride lasers have attracted intensive studies in the last two decades [1-4]; they are the only type of solid-state laser directly emitting in the UV wavelength region. Solid-state cerium lasers have many advantages in terms of tunability, simplicity, robustness and long life time, compared to traditional, complex, high cost and inconvenient tunable UV laser sources, such as excimer gas lasers, frequency doubled dye lasers, or solid-state visible or near infrared laser sources converted to the UV through non-linear frequency conversion processes.

$\text{Ce}^{3+}:\text{LiCaAlF}_6$ (Ce:LiCAF), and $\text{Ce}^{3+}:\text{LiLuF}_4$ (Ce:LiLuF) are the two most-studied and used cerium laser crystals, with tunable output from 282-315 nm, and 305-335 nm, respectively [3-5]. Ce:LiCAF has been operated with a slope efficiency of 46% at a power of 600 mW pumped by a 1 kHz frequency-quadrupled Nd:YLF laser [5]. Unlike Ce:LiCAF that can be readily pumped at 266 nm from frequency-quadrupled Nd:YAG 1064 nm lasers, Ce:LiLuF has no absorption at 266 nm. The maximum output power achieved from a Ce:LiLuF laser reported to date was 380 mW operating at 7 kHz pumped by the frequency-doubled yellow output of a copper vapour laser [3]. The highest reported slope efficiency was 62% using an Yb^{3+} co-doped Ce:LiLuF crystal (pumped by a 289 nm Ce:LiCAF laser itself pumped by a 266 nm frequency-quadrupled Nd:YAG laser) where the Yb^{3+} co-doping is reported to reduce colour centre formation [6]. Johnson *et al.* also demonstrated 62% slope efficiency by pumping with a high-beam-quality frequency-quadrupled Raman-shifted Nd:YAG laser at 289 nm [7]. To date, a major limitation to widespread uptake of cerium laser technology in areas such as biophotonics sensing is the high cost of the large-scale UV pump sources usually used.

Previously we reported a miniature architecture for scaled down Ce:LiCAF lasers pumped by low-power microchip Nd:YAG lasers, aimed at developing low cost and compact cerium laser systems suitable for practical applications [8]. Low-cost microchip Nd:YAG lasers represent ideal small-scale pump lasers as they have output energies of order tens of microjoules but with sub-nanosecond pulse durations, producing sufficient peak power (10s of kW) to allow efficient non-linear conversion from 1064 nm to the required 266 nm pump wavelength for Ce:LiCAF lasers. Scaling down of cerium lasers provides an additional benefit in that the performance of conventional cerium lasers is adversely affected by the comparable timescales of the crystal fluorescence decay (25 ns and 45 ns for Ce:LiCAF and LiLuF respectively) and the cavity build-up time (~ 10 ns) for typical cavity lengths (~ 10 cm) reported in previous work [9,10]. By miniaturising the cerium laser, the resulting short cavity reduces the build-up time, allowing more efficient extraction of the inversion owing to reduced fluorescence losses. We have already demonstrated a miniature Ce:LiCAF laser pumped using a frequency quadrupled Nd:YAG microchip laser, demonstrating a threshold as low as 200 nJ, and an output pulse energy of 550 nJ at 289 nm with a slope efficiency of 31% [8]. Very strong improvements in performance of Ce:LiLuF lasers has also been observed when scaling down pump pulse durations and cavity build-up times (cavity lengths), leading to efficient and low-threshold operation [11].

In this paper we report a broadly tunable miniature Ce:LiLuF laser. Our goal was to scale down the threshold so that the Ce:LiLuF laser could be pumped by a miniature Ce:LiCAF laser itself being pumped by a low power microchip Nd laser, while maintaining sufficient efficiency to realise continuous tuning of the combined Ce:LiCAF/ Ce:LiLuF system through the 280 nm to 340 nm spectral region. These convenient tunable UV wavelength sources with sub-nanosecond pulse-widths and kilowatt peak powers will be highly attractive for practical applications, particularly in the bio-sensing field.

2. Experimental configuration and results

2.1 Experimental configuration

To generate a suitable pump wavelength for the Ce:LiLuF laser we built a Ce:LiCAF laser pumped by a frequency-quadrupled Nd:YAG microchip laser (Alphas GmbH). The 266 nm frequency quadrupled Nd:YAG laser provided 12 μJ pulses at 1 kHz with a pulse duration of 550 ps. The Ce:LiCAF laser cavity contained of an uncoated 2.2 mm plane-parallel Ce:LiCAF crystal grown by VLOC Inc.. The pump and cavity configuration was similar to that previously reported by us [8]. To ensure maximum Ce:LiCAF laser output power using the full power available from the microchip pump laser, the cavity mode radius was optimised to be $\sim 65 \mu\text{m}$ using a concave-plane cavity consisting of a dichroic high reflector ($R=100 \text{ mm}$) and a plane output coupler ($T=30\%$) separated by 30 mm. The Ce:LiCAF crystal was placed adjacent to the plane mirror where the mode size was smallest. The pump spot radius formed by a 90 mm focal length lens was measured to be $\sim 70 \mu\text{m}$. With this setup, we obtained a maximum output of 3.5 μJ at 287 nm from the Ce:LiCAF laser, with a pulse duration of 500 ps at a pulse repetition rate of 1 kHz. The optical to optical (266 nm to 287 nm) efficiency was 32%.

The output from the Ce:LiCAF laser was used to pump a miniature Ce:LiLuF laser, shown schematically in Fig. 1. We employed this cavity design with various cavity lengths and output couplers in order to optimize the laser in two different ways – firstly for lowest threshold operation (section 2.2) and secondly for highest efficiency operation (section 2.3). The uncoated Ce:LiLuF crystal was plane cut, and aligned with the c-axis parallel to the pump polarisation for the optimum performance [11]. The Ce:LiLuF was 1.3 mm long giving 85% absorption at 287 nm. The crystal sample used here was grown at Tohoku University using the Czochralski technique. In order to achieve lowest threshold operation, we used a near hemispherical cavity consisting of a plane dichroic high reflector (90% transmission at 287 nm, $> 99\%$ reflection from 304 nm to 350 nm) and a 25 mm curved output coupler (10% transmission at 304 nm to 350 nm) separated by $\sim 25 \text{ mm}$. The crystal was placed adjacent to the plane mirror resulting in an extremely small cavity mode of approximately $15 \mu\text{m}$ in the crystal. This cavity could only be operated up to a pump energy of 1 μJ without damage to the dichroic mirror coating. In order to achieve efficient laser operation with maximum output energy using the full power available from the Ce:LiCAF pump laser, the cavity length was reduced to 14 mm to form a larger cavity mode ($\sim 35 \mu\text{m}$). For tunable operation with the low threshold cavity or high power cavity, a Brewster angle fused silica prism was inserted adjacent to the output coupler.

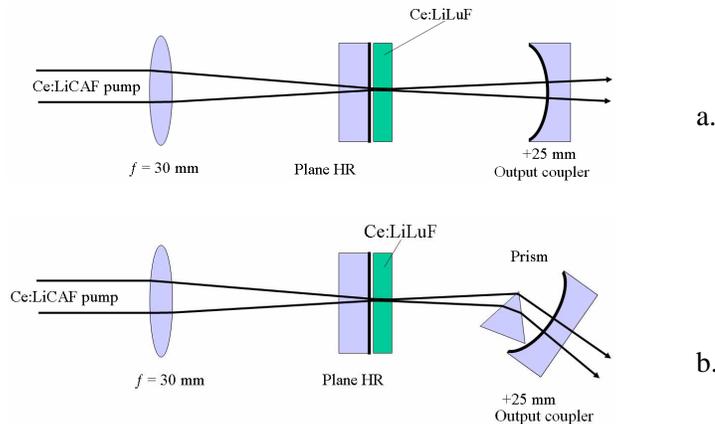


Fig.1. Experimental layout for Ce:LiLuF lasers pumped by a Ce:LiCAF laser. (a) Linear cavity without tuning unit; (b) Bent cavity with Brewster-angle prism as a tuning element.

2.2 Low threshold operation using a small cavity mode ($\sim 15 \mu\text{m}$)

Both free running and tunable operation of the Ce:LiLuF laser was investigated using a $T=10\%$ output coupler and a $15 \mu\text{m}$ cavity mode to achieve the lowest pump threshold. The measured output powers are shown in Fig.2, where the stated pump pulse energy was measured after the focusing lens. The laser output was found to be π polarized (parallel to the crystal c-axis and the pump polarisation). The lowest threshold of $\sim 100 \text{ nJ}$ was achieved in an untuned cavity; the threshold increased to 160 nJ for the prism tuned cavity. More than 150 nJ of output pulse energy was obtained with $\sim 500 \text{ nJ}$ pump in both un-tuned and prism tuned cavities. We have examined the laser pulse profile and cavity build-up time for the low threshold cavity, finding that the 900 ps -duration Ce:LiLuF output pulse was emitted 3 ns after the Ce:LiCAF pump pulse, corresponding to a build-up time of approximately 18 round trips. These results suggest the Ce:LiLuF laser was running with relatively low gain compared to the Ce:LiCAF laser that had a build-up time of less than 1 ns .

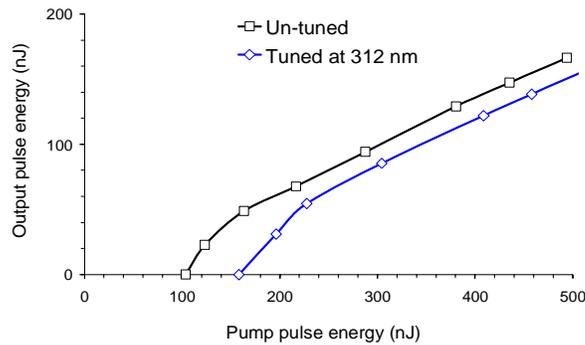


Fig. 2. Output characteristic of Ce:LiLuF lasers with small cavity mode of $\sim 15 \mu\text{m}$.

It is seen in Fig 2 that apparent slope efficiency of the laser varied with pump pulse energy in two distinct regions. When the pump energy is close to the threshold, the laser output quickly increases with pump pulse energy due to a rapid decrease of the build-up time of the Ce:LiLuF laser pulse, allowing earlier extraction of the population inversion and consequent reduction of the losses from fluorescence decay. This effect is typical for gain-switched laser systems [12]. The slope of the curve for higher pump powers, for which the build-up time of the cavity is reduced to much less than the crystal upper level life time (45 ns for Ce:LiLuF), gives a true indication of the slope efficiency of the laser. The high power slope efficiency was 37% , and corresponding optical to optical efficiency was 33% .

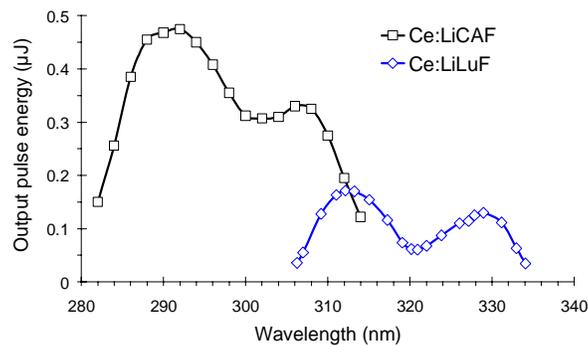


Fig. 3. Output pulse of the prism-tuned Ce:LiLuF laser as a function of wavelength together with the Ce:LiCAF laser tuning curve reprinted from Spence et al. [8]. The 266 nm pump pulse energy required for this combination is $2 \mu\text{J}$.

In order to realize full wavelength tunability of the Ce:LiLuF laser, a Brewster angle silica prism was inserted into cavity close to the output coupler. The T=10% output coupler was used to achieve lowest loss, and consequently the widest tuning. The tunability of the low threshold Ce:LiLuF laser is shown in Fig. 3, together with the Ce:LiCAF tuning curve from our previous work on low threshold Ce:LiCAF lasers [7]. The Ce:LiCAF laser from that work had a maximum output of 550 nJ at 287 nm using only 2 μ J of 266 nm pump energy. The maximum Ce:LiLuF laser output pulse energy was 180 nJ at 312 nm also using 550 nJ of 287 nm pump energy. The total optical to optical efficiency from 266 nm (fourth harmonic Nd:YAG microchip laser) to 312 nm (Ce:LiLuF laser) was 9%. The laser could be continuously tuned from 306 nm to 334 nm with two peaks corresponding to the peaks of the fluorescence bands around 312 nm and 329 nm.

2.3 High power efficient laser operation using a large cavity mode ($\sim 35 \mu\text{m}$)

By reducing the cavity length from 25 mm to 14 mm, the laser cavity mode was increased from $\sim 15 \mu\text{m}$ to $\sim 35 \mu\text{m}$ enabling a higher pump energy to be used without damage to the dichroic mirror coating. Two different output couplers with transmission T=30% and T=10% were investigated. Fig. 4 shows the laser output energy and slope efficiency with the two output couplers, with no tuning prism in the cavity. The best performance was obtained with the T=30% output coupler. The maximum laser output was 1.25 μ J using 3.1 μ J of pump energy giving a slope efficiency up to 68% and optical to optical efficiency of 40%. The threshold pump energy was measured to be 1.35 μ J. In contrast to the low threshold cavity laser, the build-up time of the large mode cavity laser was reduced to less than 1 ns. The laser pulse duration was also shorter, measured to be 500 ps.

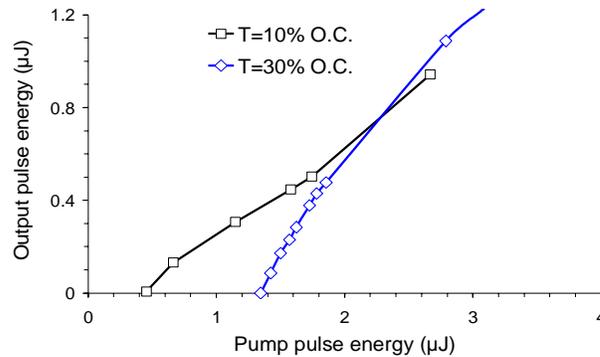


Fig. 4. Output characteristic of the Ce:LiLuF laser with a cavity mode of $\sim 35 \mu\text{m}$

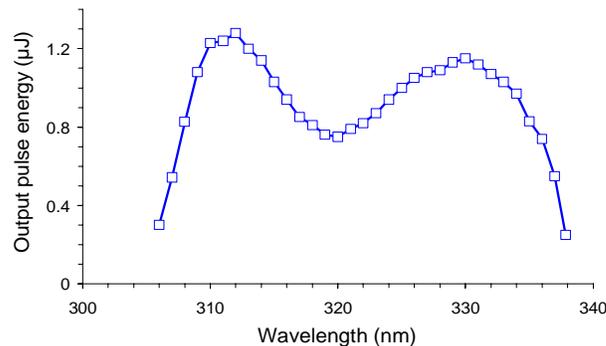


Fig. 5. Tuning curve of Ce:LiLuF laser with large mode pumped by a 3.5 μJ Ce:LiCAF laser

The tuning curve shown in Fig. 5 was measured using the maximum pump energy of 3.5 μJ using the T=10% output coupler and a large cavity mode configuration, but with the cavity extended to 22 mm. This allowed better cavity control of the laser mode direction and hence continuous tuning from 306 nm to 338 nm with pulse energy of up to 1.22 μJ at the 312 nm peak, but at lower slope efficiency (40%). The output pulse was found to be π polarized through the whole tuning range.

3. Discussion

In the present work, we found that miniature Ce:LiLuF lasers pumped by a miniature Ce:LiCAF laser were extremely efficient with a broad tuning range. The maximum slope efficiency of 68% for the miniature Ce:LiLuF laser is the highest reported to the best of our knowledge. The continuous tuning from 306 nm to 336 nm using this 14 mm cavity, together with the extended tunability (306-338 nm) obtained using the same type of cavity but extended to 22 mm in length while maintaining a high efficiency of 40%, are the best results for any Ce:LiLuF laser. This tuning range is significantly extended from the previous best report [4] which had a long wavelength limit of 333 nm.

Comparing our laser efficiency with previous work, it indicates that losses are much lower than in previous reports that also used a high quality crystal but with a much longer cavity [2]. We suggest that the reduced cavity loss is due to reduced color centre absorption inside the crystal. The short cavity with a fast cavity build-up time can significantly reduce the color centre effects, which results in high laser efficiency.

The 100 nJ minimum pump threshold for the miniature Ce:LiLuF laser is also a record for any cerium laser. A similar cavity configuration we reported for a Ce:LiCAF laser, produced a 200 nJ threshold [8]. The 1.3 mm long Ce:LiLuF crystal allowed better mode matching than the longer 2.2 mm Ce:LiCAF crystal when using a cavity mode of $\sim 15 \mu\text{m}$ to achieve low threshold operation.

The 3 ns build-up time of the low threshold Ce:LiLuF laser is still relatively long, compared to 0.8 ns build-up time for the LiCAF pump laser cavity; this results from the lower pump intensity and the resulting lower round-trip gain for the LiLuF laser. The relatively long cavity needed to accommodate the tuning prism prevented scaling to even lower thresholds.

4. Conclusion

We have demonstrated very efficient and compact UV tunable laser sources using a miniature cerium laser configuration. Up to 70% slope efficiency was achieved when Ce:LiLuF was pumped by a miniature Ce:LiCAF laser itself pumped at 266 nm, the 4th harmonic from a low power microchip 1064 nm Nd:YAG laser. The Ce:LiLuF laser output pulse duration was shorter than 500 ps, and, using a prism tuned cavity, continuous tuning from 306 nm to 338 nm was demonstrated.

The lowest pump threshold achieved was just 100 nJ, and more than 150 nJ output was obtained when Ce:LiLuF was pumped by a 500 nJ Ce:LiCAF pump laser. Together with the low threshold Ce:LiCAF laser reported in [7], we can demonstrate a unique UV laser source with full wavelength coverage from 282 nm to 334 nm, pumped using just 2 μJ pulses at 266 nm, which can simply be generated using a multi-kilohertz commercially-available microchip Nd pump laser.

We are currently performing experiments using wedged etalons that should permit tuning of even more compact Ce:LiLuF and Ce:LiCAF lasers. This tuning method will also permit operation of monolithic tunable microchip UV lasers, bringing all the advantages of monolithic lasers along side the ability to tune the output wavelength in the UV [13].

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