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Efficient all-solid-state Ce:LiLuF laser source at 309 nm

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Abstract

We report an all-solid-state laser giving 67 mW of 309 nm output when pumped with a frequency-quadrupled, all-solid-state Raman-shifted Nd:YAG laser. This efficient laser source has an optical-to-optical conversion efficiency of 41%, with respect to 289 nm conversion to 309 nm, and slope efficiencies of up to 62% (based on absorbed power).

1. Introduction

Tunable ultraviolet (UV) lasers are required for many applications including biophotonic sensing, remote sensing of atmospheric species and short pulse generation. Cerium lasers produce tunable output directly in the UV, based on the 5d–4f transition of Ce^{3+} ions doped into fluoride crystals. Ce^{3+}:$LiSrAlF_6$ (Ce:LiSAF) and Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) lasers have been the study of extensive research in recent years, resulting in efficient (up to 46% [1]) and tunable (281–315 nm [2]) devices. By comparison, Ce^{3+}:$LiLuF_4$ (Ce:LiLuF) has received little attention as a UV laser crystal; however in recent years, Ce:LiLuF crystal growth procedures have improved dramatically and now crystals can be obtained with comparable quality to Ce:LiCAF and Ce:LiSAF. Ce:LiLuF has performed well at both low and high repetition rates with slope efficiencies as high as 62% [3] and 51% [4], respectively. Ce:LiLuF has been tuned between 305 and 333 nm [4] with the peak emission being at 309 nm and a weaker emission peak at 327 nm.

Ce:LiCAF and Ce:LiSAF both have a strong absorption peak at 266 nm, which has enabled these crystals to be pumped by frequency-quadrupled Nd:YAG [5,6] and $Nd:YVO_4$ [7] lasers at 266 nm. Ce:LiLuF is strongly absorbing at 210 nm, 243 nm and 294 nm [8], but exhibits very little

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absorption at 266 nm. Ce:LiLuF has been pumped by various UV laser sources including: KrF lasers (248 nm) [6], quintupled Nd:YAG (213 nm) [8], frequency-quadrupled Nd:YAG-pumped Ce:LiSAF (290 nm) [9] or Ce:LiCAF lasers, and the frequency-doubled yellow output of a copper vapour laser (CVL) (289 nm) [4].

It has been found that pumping Ce:LiLuF at the longer wavelengths has produced the most efficient operation due to the minimal colour centre formation (solarization) effects at the longer wavelength [4]. Also, the energy deficit is significantly smaller when pumping at longer wavelengths, being only 6.5% for a pump source at 290 nm, but 31% for a pump source at 213 nm, for laser action at 310 nm.

Although the frequency-quadrupled Nd:YAG-pumped Ce:LiSAF (or Ce:LiCAF) laser and the quintupled-Nd:YAG lasers are all-solid-state pump sources that do provide pump light at an appropriate wavelength for Ce:LiLuF, they are relatively inefficient. In general, to obtain a suitable pump source for Ce:LiLuF using an Nd laser as the all-solid-state pump source, one requires either 5th harmonic generation or 4th harmonic generation with one non-harmonic conversion step to get to a suitable wavelength. In the last few years, Raman-shifted Nd:YAG lasers at 1156 nm [10], and intra-cavity frequency-doubled Raman lasers at 578 nm [11,12] have been developed. By frequency-quadrupling the 1156 nm output of a Raman-shifted Nd:YAG laser, UV output can be obtained at 289 nm which falls near the peak of the 294 nm absorption band for Ce:LiLuF. In contrast to the all-solid-state laser sources used previously to pump Ce:LiLuF, in this Raman laser, both the non-harmonic step (Raman conversion of 1064–1156 nm) and the first harmonic step (frequency conversion of 1156–578 nm) are carried out intra-cavity, where non-converted wavelengths are efficiently recycled. The 10 kHz frequency quadrupled Raman-shifted Nd:YAG laser reported by Pask et al. [12] has a 1064–289 nm conversion efficiency of 8.6% which compares favourably with the most efficient 1 kHz frequency-quadrupled Nd:YAG-pumped Ce:LiCAF laser reported to date, with a conversion efficiency of 7.7% (1064–290 nm) [1].

In this paper, we show the results of using an efficient frequency-quadrupled, Raman-shifted Nd:YAG pump source to pump a Ce:LiLuF laser at 10 kHz pulse repetition frequency. The pump source for the entire system is a fibre coupled laser diode producing 18 W at 808 nm from which 67 mW at 309 nm has been achieved, giving an optical conversion efficiency of ~0.4% with respect to the laser diode output. A Ce:LiLuF slope efficiency of 62% (based on absorbed 289 nm pump power) was obtained with an 80% output coupler. This high efficiency is attributed to the high beam quality and ideal wavelength of the pump source.

2. Experimental configuration

The diode pumped Nd:YAG laser was Q-switched with a repetition rate of 10 kHz, and incorporated a $LiIO_3$ Raman shifter (to generate first Stokes at 1156 nm) and an LBO frequency doubler (to generate yellow at 578 nm), with both nonlinear crystals placed intra-cavity for maximum efficiency (~7% conversion efficiency from diode power to yellow output). This system produced up to 1.4 W of yellow output, with a pulse duration of 12 ns and a measured M^2 of approximately 2.5. Full details of the Raman-shifted Nd:YAG laser have been reported elsewhere [12].

The experimental configuration used for frequency quadrupling step is shown in Fig. 1. Using a Keplerian mirror telescope, the yellow laser was collimated and spot size doubled to approximately 1.5 mm in diameter. A Berek polarization compensator was then used to convert the horizontally polarized yellow light into vertically polarized yellow light to match the 4th harmonic generation geometry. An $f = 50$ mm cylindrical lens was used to line-focus the yellow beam into the 8-mm long BBO crystal used for 4th harmonic generation, and the resulting UV beam was re-collimated using another $f = 50$ mm cylindrical lens. The UV light was separated from the unconverted yellow light using a Pellin Broca prism. The conversion efficiency from 578 to 289 nm was 26% which compares favourably with previous results obtained for frequency doubling the yellow output of a CVL [4] and is thought to be due to the high

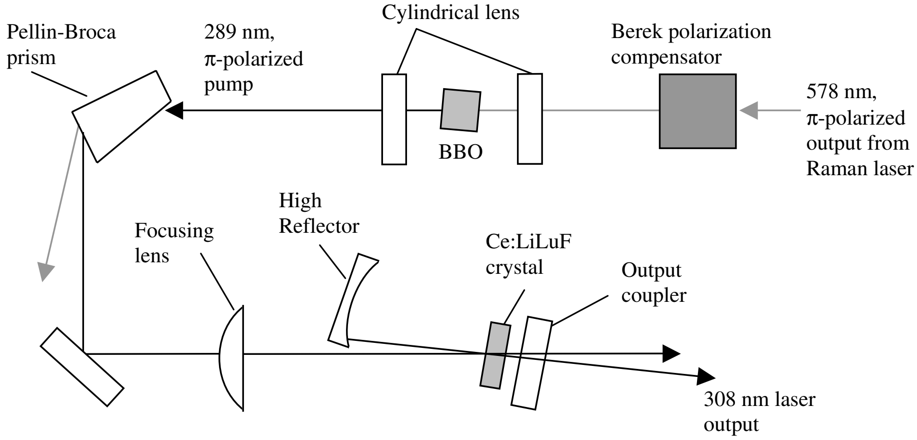

Fig. 1. Schematic of the Ce:LiLuF laser, pumped by a frequency-quadrupled Raman-shifted Nd:YAG laser.

beam quality and high peak power of the yellow laser. The 289 nm output was had an M^2 value of 2.8.

An uncoated, slightly wedged (0.5°), 0.8 mm thick Ce:LiLuF crystal was used, with a <1 at% cerium doping level providing approximately 70% absorption of the π-polarized 289 nm pump. The crystal was grown using the Czochralski method at the Universite du Maine, and cut such that the crystallographic *c*-axis was contained in the horizontal plane of the end face. The temperature of the crystal was controlled by a water-cooled chiller.

The 289-nm pump beam was focussed into the Ce:LiLuF crystal to a spot size of approximately 150 μm in diameter, using an $f = 215$ mm lens (Fig. 1). A near-hemispherical laser cavity was used, consisting of a broadband flat output coupler (with a choice of reflectivities of 50%, 80% and 90%) and a curved high reflector (ROC = 100 mm), with the distance between the output coupler and high reflector adjusted for maximum output, corresponding to a cavity length of 9.2 cm. The laser crystal was placed close to the output coupler to maintain a small laser mode size in the crystal.

The efficiency of Ce:LiLuF lasers can be strongly influenced by detrimental solarization effects [13]. At high repetition rates the extent of solarization in Ce:LiLuF has been shown to depend on the temperature of the crystal, with a cooler crystal lasing more efficiently [14]. (This has not been reported to be the case at low repetition rates and, as such, it is possible that these temperature dependent solarization effects are manifested only at high repetition rates.) Thus to take advantage of this temperature dependence, experiments were undertaken at room temperature and with the crystal cooled to 7 °C.

3. Results

A summary of the laser results obtained is shown in Table 1. Output power characteristics for the Ce:LiLuF laser operating with 80% and 90% reflectivity output couplers are shown in Fig. 2. As expected, higher output powers were obtained at the lower temperature, for example, with the 80% output coupler we measured an output power of 67 mW at 7 °C compared to 40 mW at room temperature. Using the 80% reflectivity output coupler, with the cerium crystal cooled to 7 °C, a maximum slope efficiency of 47% with respect to incident pump power was obtained. Taking into account the fraction of unabsorbed pump light (30%), we calculate the slope efficiency with respect to absorbed power to be 62%.

The maximum output obtained with the 80% reflectivity output coupler was 67 mW from 230 mW of incident pump power, giving a UV-to-UV conversion efficiency of 29% with respect to pump

Table 1
Summary of results for Ce:LiLuF laser pumped by frequency-quadrupled Raman Nd:YAG laser

Output coupler reflectivity	Summary of results			
	Crystal temperature (7 °C)		Crystal temperature (25 °C)	
	Maximum output power	Slope efficiency[a]	Maximum output power	Slope efficiency[a]
90%	55 mW	35%	35 mW	24%
80%	67 mW	47%	40 mW	44%
50%	33 mW	n/a	18 mW	n/a
7% at 309 nm 70% at 327 nm	7 mW (at 327 nm)	n/a	n/a	n/a

[a] Slope efficiency based on total pump power.

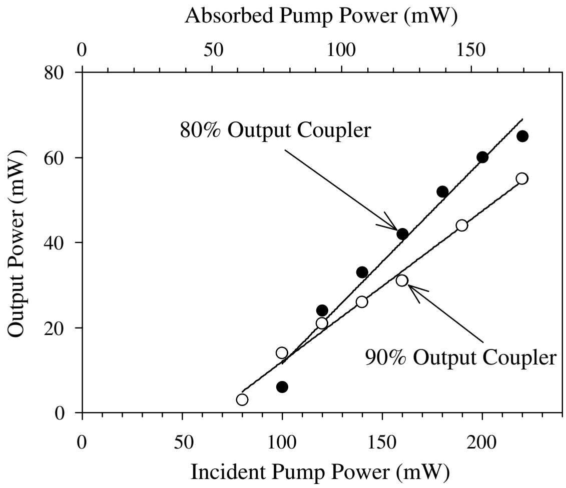

Fig. 2. The 309 nm, π-polarized laser output of Ce:LiLuF laser (cooled to 7 °C) with an 80% and a 90% reflectivity output coupler, pumped by the frequency-quadrupled Raman laser.

power at 289 nm. Taking into account only the absorbed fraction of the pump power we get a UV-to-UV conversion efficiency of 41%. The threshold of the laser with the 80% reflectivity output coupler was approximately 65 mW of absorbed pump power and there was no noticeable saturation at the higher pump powers.

Lasing was also investigated with the 90% and 50% reflectivity output couplers, where maximum outputs were 55 and 18 mW, respectively, with the crystal cooled to 7 °C. A slope efficiency of 35% with respect to incident pump power was measured for the 90% output coupler and the threshold was found to be approximately 50 mW of absorbed pump power.

The output obtained from the Ce:LiLuF laser, using the broadband output couplers, was found to be π-polarised and at 309 nm in all cases. This wavelength and polarization corresponds to the shorter wavelength emission peak of the double peaked emission spectrum of Ce:LiLuF [14]. Lasing on the longer, σ-polarized, 327 nm peak, where the effective emission cross-section is lower, was investigated, using an output coupler with 69% reflectivity at 327 nm and 7% reflectivity for 309 nm to discriminate against the 309 nm transition. A maximum of 7 mW of 327 nm, predominantly σ-polarized output was obtained. The ability to obtain lasing at both 309 and 327 nm, corresponding to the two emission peaks for Ce:LiLuF, suggests continuously tunable output should be obtainable between these wavelengths, since good tuning over this range has previously been obtained with the same crystal [4], with less efficient lasing.

Pump and laser pulse shapes obtained with the 80% output coupler, and for maximum output power are shown in Fig. 3. The roundtrip gain exceeds the loss approximately 10 ns after the start of the pump pulse and a further 5 ns (~16 round trips) is required to produce the output pulse, giving an overall build up time of ~15 ns. Fig. 3 shows the 309-nm laser pulse, with a pulse length of ~5 ns (FWHM), and the fluorescence both with and without laser action (cavity unblocked and blocked). The fluorescence, which decays with a lifetime of ~40 ns, is proportional to the upper level population peaking approximately 10 ns after the peak of the pump. The depletion, measured by comparison of the two fluorescence curves,

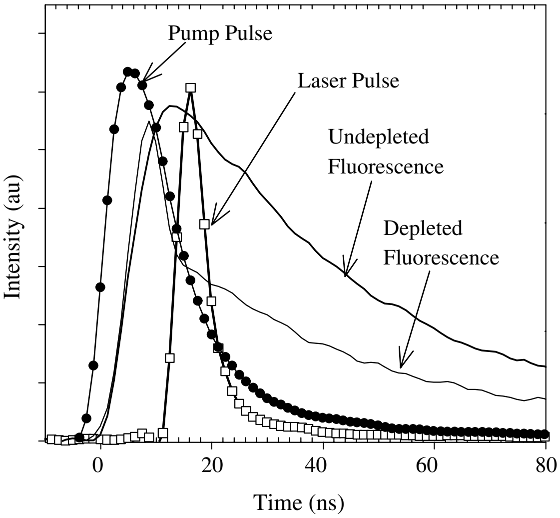

Fig. 3. Fluorescence, laser and pump pulse shapes from the frequency-quadrupled Raman laser pumped Ce:LiLuF laser operating with a 80% reflectivity output coupler at 7 °C.

was found to be ~45%. The stability of the output power of the cerium laser was approximately 5% corresponding to the stability of the pump laser.

4. Conclusion

Improved performance is expected through the use of a longer crystal, or double passing the pump beam, which would provide increased pump absorption in the Ce:LiLuF crystal. In previous work [14], the laser output continued to increase with decreasing crystal temperature well below the minimum temperature of 7 °C achieved in the present study, hence we also expect that reducing the crystal temperature further will increase both the output power and slope efficiency. Anti-reflection coating or using a Brewster-cut Ce:LiLuF crystal would both improve pumping efficiency and reduce intra-cavity losses, thereby increasing the laser efficiency.

We have demonstrated an efficient all-solid-state UV laser giving up to 67 mW of 309 nm output and 7 mW of 327 nm output. We believe that the 62% slope efficiency (with respect to absorbed pump light at 289 nm) obtained here is the highest slope efficiency for a high repetition rate Ce:LiLuF laser to date. Indeed, this is the highest slope efficiency reported to date for any high repetition rate (>1 kHz) cerium laser. Solarization effects have been reported in Ce:LiLuF at high repetition rates [14,15] which have not been reported at low repetition rates. These manifest as a dependence of the Ce:LiLuF lasing efficiency on crystal temperature [14]. Simple cooling of the lasing crystal has enabled us to overcome most of these effects so that Ce:LiLuF can now perform as well at high repetition rates as at low repetition rates.

Further, the UV-to-UV conversion efficiency of 41% with respect to the available power at 289 nm, and 0.4% with respect to the laser pump source for the entire system reported here is comparable to the highest conversion efficiency reported to date for Ce:LiCAF (43%) [1] and Ce:LiLuF (47%) [3]. This is thought to be primarily due to the good beam quality and near-optimum wavelength of the 289-nm pump source. As a compact, all-solid-state Ce:LiLuF laser, the frequency-quadrupled Raman-shifted Nd:YAG pumped Ce:LiLuF laser described in this paper is highly efficient in comparison to previously reported counterparts such as the 213 nm quintupled Nd:YAG pumped Ce:LiLuF laser which had a UV-to-UV conversion efficiency of 0.5% [8] and when pumped with the 290 nm Ce:LiSAF output the UV-to-UV conversion efficiency was 8% [9]. Further research is underway to obtain tunable output (305–333 nm) from the all-solid-state Ce:LiLuF laser, which will complement the all-solid-state Ce:LiCAF and Ce:LiSAF lasers which are tunable between 281–315 nm.

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