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Micro-pulling-down-method-grown Ce:LiCAF crystal for side-pumped laser amplifier

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ABSTRACT

We demonstrate 290 nm lasing and femtosecond amplification using a micro-pulling-down method-grown Ce:$LiCaAlF_6$ crystal, which was excited using dual-side- and four-side-pumping schemes. These results show that the flexibility and lower cost of this growth technique as well as the multi-side pumping configurations provide good prospects for high-power ultraviolet generation that will enhance the applications of the Ce:$LiCaAlF_6$ laser and amplifier.

The success of fluorides as compact and convenient ultraviolet (UV) laser materials has been remarkable and has, therefore, fueled further research for the improvement of crystal quality and optical techniques. Particularly important are trivalent cerium (Ce^{3+})-doped fluorides, for example, Ce^{3+}:$LiCaAlF_6$ (Ce:LiCAF) [1], Ce^{3+}:$LiSrAlF_6$ (Ce:LiSAF) [2], and Ce^{3+}:$LuLiF_4$ (Ce:LLF) [3] because they can be directly pumped by a Nd:YAG laser [1,4,5]. Among these, Ce:LiCAF is perhaps the most extensively studied as a laser and amplifier material because of the absence of color center formation and solarization effects, therefore enabling high-power UV emission [6,7]. Efficient UV generation through lasing techniques and/or amplification heavily relies on crystal quality and pumping configuration. In this paper, we report on the application of multiple-side-pumping configurations, specifically dual-side pumping and four-side pumping, to generate and/or amplify 290 nm laser radiation. The four-side-pumping configuration is an extension of the prismatic cell pumping technique, which was proposed by Bethune and initially used for dye lasers [8]. These pumping schemes are expected to reduce the risk of damage during multi-watt pumping by distributing the energy load equally throughout the crystal volume while delivering the same amount of total energy as if it was delivered to one side only.

For the purpose of materials screening, a growth technique that can grow laser-quality crystals at the shortest possible time and cost is most favorable. The Czochralski, Bridgman, and floating zone methods are some examples of classic crystal growth techniques that are widely used in research and industry [9,10]. However, typical growth rates of about 0.5–1 mm/h, the need for large quantities of raw material, and post-growth processing such as cutting and polishing tend to make these growth techniques time- and cost-intensive. For this reason, the micro-pulling-down (mPD) method would be the suitable choice. The μ-PD apparatus modified for fluoride crystal growth is shown in Fig. 1. Starting materials are thoroughly mixed and placed inside a graphite crucible. The growth chamber is then evacuated to 10^{-4} Torr using rotary and diffusion pumps. By radio frequency (RF) heating, the crucible is baked at 600 °C for 1 h in order to remove oxygen traces from the moisture of raw materials and adsorbates on the chamber surface. Simultaneously, the chamber is further evacuated to 10^{-5} Torr. After baking, the recipient is filled with a mixture of non-reactive gases, Ar and CF_4, until ambient pressure. The crucible is heated to a melting temperature of about 1450 °C. The crystal is grown with a pulling rate of 0.1 mm/min and with complete solidification of the melt charged in the crucible. Owing to a fast growth speed of 0.05–0.1 mm/min, a high quality crystal can be grown using less than 1 g of raw material in 5–12 h [9–11]. This allows the growth of large crystals in a shorter time and at a lower cost compared with the classical methods mentioned earlier. Merits of this growth method that makes it suitable for materials research can be found elsewhere [6]. Recent improvements of this method have made the quality of mPD method-grown crystals comparable to those prepared by conventional growth techniques [7,10]. The comparison between mPD method and Czochralski method is shown in Fig. 2. The success of this growth method has been extended to

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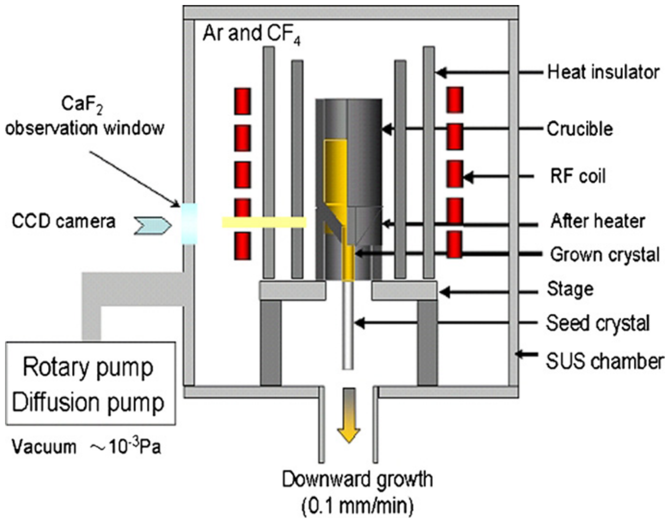

Fig. 1. mPD apparatus modified for fluoride crystal growth.

Growth direction	Upward	Downward
Typical Growth rate	0.05 – 0.1 mm/hr	0.05-1 mm/min
Amount of raw material	plenty	< 1 g
Maximum crystal diameter	400 mm	0.05 - 10 mm
Maximum crystal length	1-2 m	1 m
Stage of development	Grow crystals for actual devices	Research stage for materials screening
Other characteristics	Large crystals, less inclusion of heavy solid particles	Shaped crystal growth, "ready to use" fibers, multi-crystal growth, less inclusion of air bubbles

Fig. 2. Comparison between micro-pulling down method and Czochralski method.

fluoride crystals as well, and has since had a tremendous impact on applications of fluorides in various fields such as optical materials, laser gain media, amplifiers, and scintillators [12]. The Ce:LiCAF crystal used for this work was successfully grown by the mPD method. By pumping this mPD method-grown Ce:LiCAF crystal with the fourth harmonic of a Nd:YAG laser (266 nm), in a dual-side-pumping configuration, we are able to obtain 290 nm laser emission with ~24% slope efficiency. Moreover, using the same crystal in a four-side-pumping (prismatic cell-type pumping) configuration, we demonstrate amplification of femtosecond UV pulses with a net gain of 4. This is the first demonstration of lasing and amplification using a mPD method-grown Ce:LiCAF. These results demonstrate that a combination of using the mPD method to provide a cheaper and more efficient alternative to growing laser-quality fluoride crystals and the choice of an appropriate pumping scheme that would allow multi-watt pumping without damaging the crystal could facilitate the generation of high-power UV radiation. Availability of high-power UV radiation will greatly enhance the application of UV research.

In order to grow Ce:LiCAF by the mPD method, high-purity (>99:99%) CeF_3, AlF_3, CaF_2, and LiF powders (Stella Chemifa) are used as starting materials. The raw materials are placed in a graphite crucible whose temperature can be controlled by radio frequency heating. A seed crystal is touched to the molten material and the crystal is pulled downward at a rate of 0.1 mm/min. The growth atmosphere is a mixture of Ar and CF_4. The as-grown crystal is 30 mm in length and 2 mm in diameter. Both ends of the as-grown crystal did not have any coating but are cut at Brewster angle. Two sides are polished and will serve as the pumping windows. The sample is doped with 1 mol% Ce ions as a nominal composition. Fig. 1 shows an absorption peak at around 270 nm and an absorption coefficient, α, of 2.7 cm^{-1} at 266 nm, therefore making the fourth harmonic of a Nd:YAG laser a suitable pump.

The laser oscillator is composed of a flat high reflector with >99% reflection at the wavelength of the laser emission (290 nm) and an output coupler with 60% transmission at 290 nm. The length of the laser cavity is 6 cm. The Ce:LiCAF crystal is simultaneously pumped by the fourth harmonics of two synchronized Nd:YAG lasers operating at 10 Hz repetition rate and 4 ns pulse duration. Cylindrical lenses with a focal length of 300 mm are set to focus the pump beams onto opposite sides of the crystal. These lenses are placed at a distance of 260 mm from the sample to excite the entire side with sufficient fluence without damaging the crystal. The output energy is measured using a joule meter. Details of the experimental setup, including the schematic diagram, can be found elsewhere [13].

The experimental setup for using the same mPD method-grown Ce:LiCAF as a UV femtosecond amplifier is illustrated in Fig. 4. The pump beam was provided by the fourth harmonics of a Q-switched Nd:YAG laser at 266 nm, 10 Hz repetition rate, and 100 mJ pulse energy. In order to achieve four-side pumping, two high reflecting mirrors, with >99% reflection at the pump wavelength, were positioned at 90° angle with respect to each other, as shown in Fig. 3, to excite the top (1), front (2), rear (3), and bottom (4) surfaces of the crystal. The pump beam diameter was expanded to 2.5 cm and was loosely focused by a cylindrical lens ($f = +100$ cm) in order to uniformly illuminate the whole sample, with a fluence of ~60 mJ/cm^2 at the crystal surface. On the other hand, the seed beam, with 290 nm wavelength and 1 kHz repetition rate, was provided by the third harmonics of a Ti:sapphire laser. It was collimated to a diameter of 1.8 mm in order to minimize coupling loss. The seed beam and the pump beam were synchronized using external delay generators. The pulse duration of the seed and amplified pulse was evaluated from the visible third order intensity autocorrelation [7,14], while the spectral bandwidth was evaluated using an ultraviolet streak camera [15].

Fig. 6 shows the measured 290 nm oscillator output energy as a function of absorbed pump energy. Factoring in scattering and Fresnel reflection losses, the actual absorbed energy is ~10% of the pump energy. This was confirmed by independent transmission measurements. A slope efficiency of ~24% was achieved with the dual-side-pumping configuration. The output energy remained linear with absorbed energy, where an absorbed pump energy of

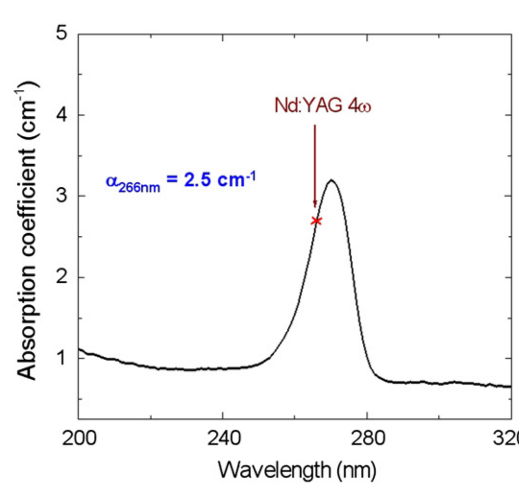

Fig. 3. Absorption coefficient of the mPD method-grown Ce:LiCAF crystal. Absorption at 266 nm makes the fourth harmonic of a Nd:YAG laser a suitable excitation source.

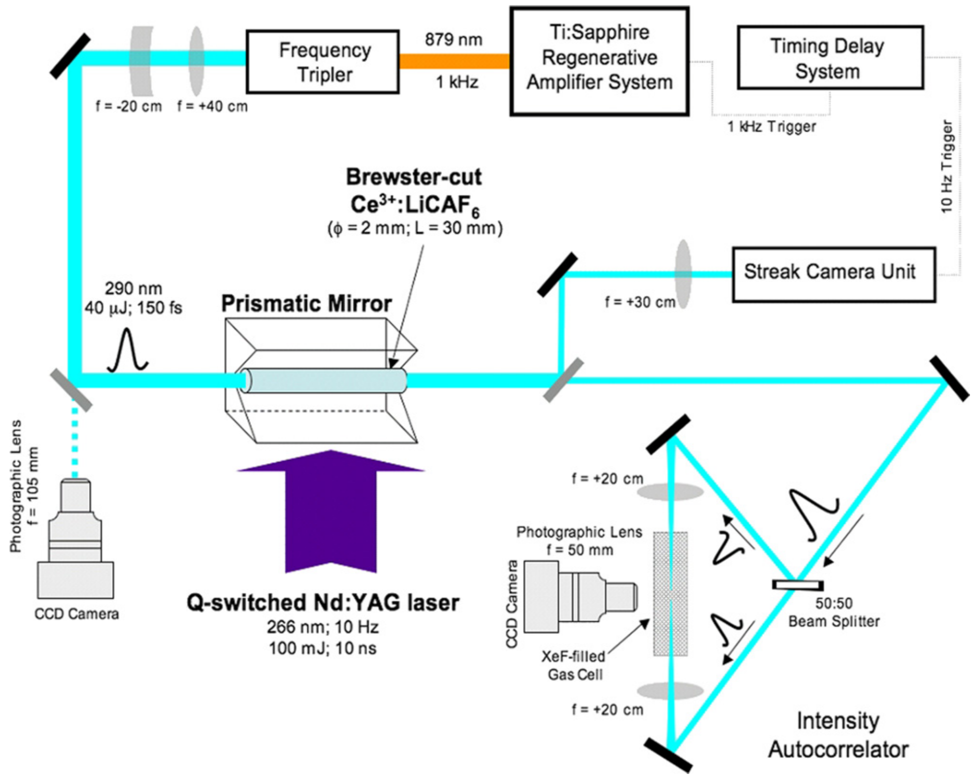

Fig. 4. Schematic diagram for the amplifier system. Seed pulses from a frequency-tripled Ti:sapphire laser are synchronized with the 266 nm pump pulses from a Q-switched Nd:YAG laser. The amplified pulses are measured using a streak camera unit and an autocorrelator.

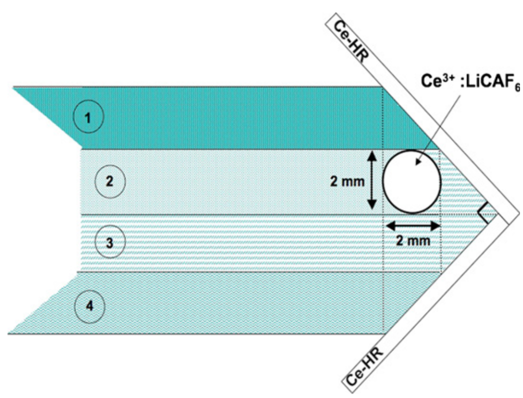

Fig. 5. Prismatic cell-type configuration used to accomplish four-side pumping of Ce:LiCAF. With this scheme, the (1) top, (2) front, (3) rear, and (4) bottom surfaces of the crystal are uniformly illuminated.

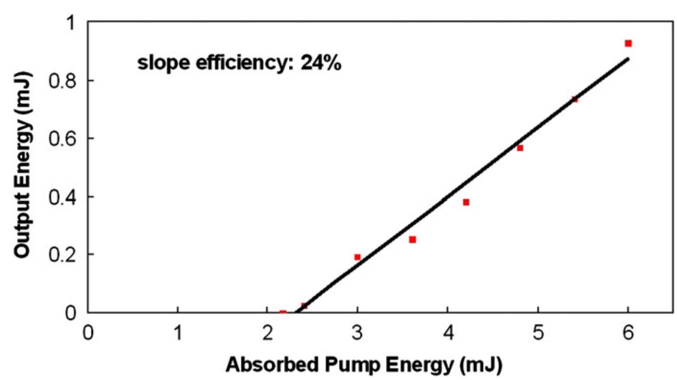

Fig. 6. Oscillator output energy using dual-side-pumping scheme. Laser emission wavelength is 290 nm with 24% slope efficiency.

6 mJ achieved a maximum output energy of 1 mJ. The lasing threshold was determined to be 2.5 mJ. This corresponds to a threshold fluence of $\sim$10 mJ/cm^2. Lasing with four-side pumping was carried out using the same four-side-pumping scheme for the amplifier, which is illustrated in Fig. 5, to facilitate uniform distribution of the pump energy to all the sides of the crystal. Lasing was achieved with a slope efficiency of $\sim$3%. The lower slope efficiency achieved could be due to the energy loss from the two mirrors and the increased scattering loss due to the two unpolished sides. The performance of the mPD-grown Ce:LiCAF is expected to improve by increasing its absorption coefficient through optimizing the crystal growth parameters and polishing all the pumping side windows.

The four-side-pumping configuration detailed in Fig. 5 was used to amplify 290 nm femtosecond seed pulses. Fig. 7 shows a plot of the differential gain as a function of seed pulse energy. Differential gain was determined by the ratio of the transmitted 290 nm seed pulse with and without the 266 nm pump. However, actual measurements showed that $\sim$50% of the seed pulse energy was transmitted through the Ce:LiCAF crystal after considering coupling and reflection losses. Hence, the net gain would be half of the differential gain. For example, with the 40 μJ input seed energy, the energy of the amplified pulse was measured to be 160 μJ, therefore translating to a net gain of 4 and a differential gain of 8. A higher gain is expected on increasing the pump fluence to at least 1 mJ/cm^2 and improving the absorption efficiency. Spectral and temporal profiles of the amplified pulse show a full width at half maximum of 0.68 nm and 240 fs, respectively, with a corresponding time bandwidth product (TBP) of 0.44. For comparison, the spectral bandwidth and pulse duration of the input seed pulse are 0.54 nm and

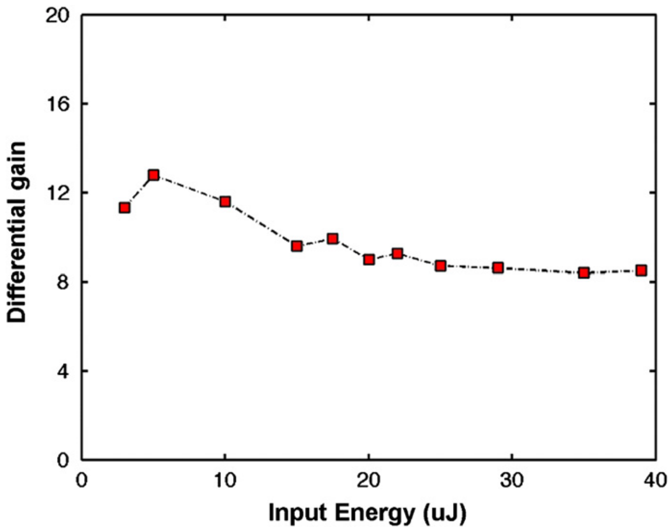

Fig. 7. Differential gain vs. input seed energy. Considering the transmission of 290 nm seed pulses is 50%, the net gain can be determined by dividing the differential gain by a factor of 2.

150 fs, respectively. The corresponding TBP is 0.35, which is in good agreement with the ~0.315 limit of a transform-limited $sech^2$ pulse. The B-integral was estimated to be less than 5, using the Ce:LiCAF parameters discussed elsewhere [7]. This value is comparable to the one achieved previously with a chirp pulse amplifier system. These show that even without dispersion compensation, no significant distortions on the spectral and temporal shapes of the amplified pulse were observed, therefore, highlighting the advantage of using a prismatic cell-type pumping configuration.

In summary, we demonstrate 290 nm lasing and amplification of UV femtosecond pulses in a Ce:LiCAF crystal successfully grown by the mPD method. Using multiple-side-pumping configurations, we are able to obtain 290 nm laser output with a conversion efficiency of ~24% and ~1 mJ pulse energy. Moreover, amplification of nearly transform-limited UV femtosecond pulses was achieved even without using chirped pulse amplification. This is the first demonstration of lasing and amplification using mPD method-grown Ce:LiCAF. Moreover, the proposed pumping configurations promise to be dynamic, scalable, and have a high potential for multi-watt UV generation. Availability of high-power UV radiation will greatly enhance the application of UV research.

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