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DEVELOPMENT OF SHORT PULSE BROADBAND AND TUNABLE NARROW- LINEWIDTH ULTRAVIOLET LASERS USING Ce:LiCAF CRYSTAL

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Abstract. *We report the development of all-solid state lasers based on Ce^{3+}:$LiCaAlF_6$ (or Ce:LiCAF) crystal as gain medium. These Ce:LiCAF lasers are pumped by 7 ns pulses at 10 Hz from the fourth harmonics (266 nm) of a Q-switched Nd:YAG laser. The effects of output coupler reflectivity, resonator length and pump energy on the Ce:LiCAF laser characteristics were explored. With the broadband laser configuration, the Ce:LiCAF laser achieved a maximum output pulse energy of 3.4 mJ and a laser slope efficiency of about 33%. Single UV laser pulses of 450 ps were generated by controlled resonator transient. With the narrow linewidth laser configuration, tunability of the Ce:LiCAF laser emission from 281 nm to 299 nm is obtained maintaining a linewidth narrower than 0.2 nm. The laser emissions are suitable for spectroscopic and environmental sensing applications.*

Keywords: ultraviolet laser; short pulse laser; tunable laser; resonator transient; rare earth-doped fluoride.

Classification numbers: 42.55.-f, 42.60.Da, 42.60.-v, 42.60.Fc, 42.60.Lh, 42.72.Bj.

I. INTRODUCTION

Solid-state ultraviolet (UV) lasers have received a great deal of interest for numerous applications in science and technology. The most established applications include environmental sensing, engine combustion diagnostics, semiconductor processing, optical micro machining, medicine and biology [1, 2]. Short laser pulses in the UV region are important as a pump source for ultrashort laser pulse generation and as an excitation source for photochemistry and bio-molecular spectroscopy [3, 4]. Of particular interest is measurement of ozone in the atmosphere using Light Detection and Ranging (LIDAR) techniques [5,6]. For this purpose, spectrally tunable and narrow linewidth laser pulses are essential. Existing commercially available tunable UV laser sources consists of subsequent steps of nonlinear frequency conversion such as doubling, tripling, and/or mixing of tunable radiation obtained from traditional tunable visible or near infrared lasers. However, higher harmonic generation is known to have low conversion efficiency, in addition to the complexity of ensuring that phase matching conditions are met [7, 8]. The best candidate laser materials in the UV region are still trivalent cerium ion (Ce^{3+})-doped fluoride crystals. Among the reported Ce^{3+}-doped fluoride laser crystals, Ce:LiCAF is most successful. It can be pumped directly by the fourth harmonics (266 nm) of a Q-switched Nd:YAG laser [9–11]. It also has a potential tuning range from 280 nm to 320 nm giving the ability to generate up to 3 fs laser pulses [12, 13]. Ce:LiCAF also has a large saturation fluence and damage threshold, making it attractive for designing power amplifiers [13, 14]. Most importantly, color center formation or solarization was not observed from Ce:LiCAF, giving it the edge over Ce:LiSAF, which exhibits a lower laser efficiency due to excited state absorption and color center formation [12, 15, 16].

This paper presents the development of broadband, short pulse UV lasers and tunable, narrow linewidth lasers using Ce:LiCAF crystal as gain medium. In previous reports, the focus is only either short pulse [17] or narrow linewidth [18]. The experimentally obtained results indicate that direct and simple generation of the UV laser emissions which possess spectrally tunability, narrow linewidth and/or short pulse duration, are feaseble at a modest laboratory for spectroscopic and environmental sensing applications. The paper is divided into three parts. Firstly, the Ce:LiCAF lasers that use a non-collinear and de-focusing pumping configuration are analyzed. In the second part, we study the broadband laser and produce possible shortest laser pulse basing on controlled resonator transient [19]. The third part presents tunability of the Ce:LiCAF laser emission, using a Littrow grating as a dispersive element, from 281 nm to 299 nm maintaining a linewidth of less than 0.2 nm (FWHM).

II. EXPERIMENT

Figure 1 shows the schematic diagram of the Ce:LiCAF broadband UV laser. The Ce:LiCAF crystal is grown using the Czochralski method with 1% Ce^{3+} doping concentration in the melt [20]. The crystal has dimensions of 1.0 x0.5x0.5 cm. Both its end faces are Brewster-cut and polished. The crystal is optically pumped at 266 nm by the fourth harmonic of a Q-switched Nd:YAG laser (Quanta-Ray INDI, Spectra-physics, Model INDI – HG10S) delivering 7 ns pulses at 10 Hz repetition rate. A non-collinear and de-focusing pumping configuration is used for the Ce:LiCAF lasers. The angle between the pump beam and the optical axis of the laser resonator is about 10°. The smaller angle is limited by the pump beam hitting one of the resonator mirrors. A 40-cm focal length lens is used to focus the pump pulses onto the Ce:LiCAF crystal that is positioned 30-cm from the lens to excite the side of the crystal with sufficient fluency, without ablating the crystal. The spot size of the pump laser beam at the surface of the crystal is 0.1 cm.

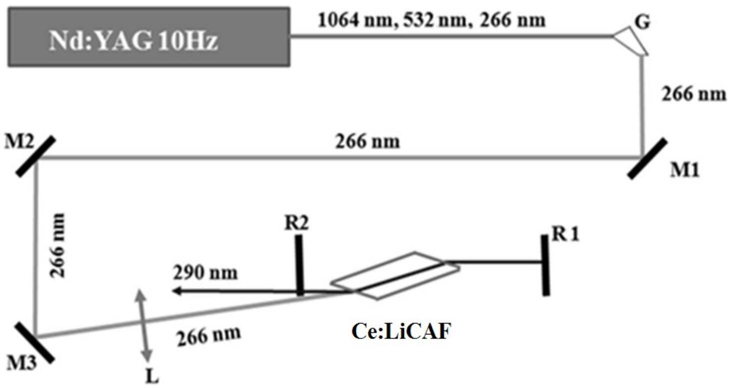

Fig. 1. Schematic diagram of the Ce:LiCAF laser configuration.

In order to investigate experimentally the influences of resonator and pumping parameters on the broadband Ce:LiCAF laser characteristics, the reflectivity of the output coupler R_2 (Fig. 1) can be varied from 14% to 30% (this is limited to the mirrors available in the laboratory), the resonator length (L) and the pump pulse energy can be changed.

In order to achieve spectrally tunable and narrow linewidth pulses, the end mirror (R_1 in Fig. 1) is replaced by a holographic grating (2400 lines/mm), which is used at Littrow operation and has the first diffraction order efficiency of 30% at 290 nm. The tunability of the Ce:LiCAF laser emission at different wavelengths is obtained by rotating the grating.

The spectral profile of the laser output was recorded using a spectrometer (Princeton Instruments SP2500) with a grating of 1900 lines/mm. The resolution of the spectrometer is 0.2 nm. The temporal profile was obtained using a photodiode (Hamamatsu S9055) with a response time of about 250 ps coupled to a 1.5 GHz digital oscilloscope (Tektronix TDS7154B). Laser energy is measured by a power/energy meter (LabMax – Top Coherent).

III. RESULTS AND DISCUSSIONS

III.1. Broadband laser emission

The damage threshold and the saturation fluence at 266 nm pumping wavelength for a Ce:LiCAF crystal are previously reported to be 5 J/cm^2 and 115 mJ/cm^2, respectively [14]. Fig. 2 presents the dependences of the damage threshold and saturation pump energy on the pump spot radius at the surface of the crystal. The results show that the Ce:LiCAF crystal reaches saturation pump energy before it is damaged using the non-collinear and de-focusing pumping configuration (Fig. 1) where the pump pulse is de-focused by placing the crystal away from the focal point. When performing the lasing experiments, care was taken so that the crystal was pumped with energies less than the damage threshold as shown in Fig. 2. The spot size of the pump laser beam at the surface of the crystal is 0.1 cm (or a pump beam radius of 0.05 cm) which means that the pump pulse energy should not be larger than 40 mJ.

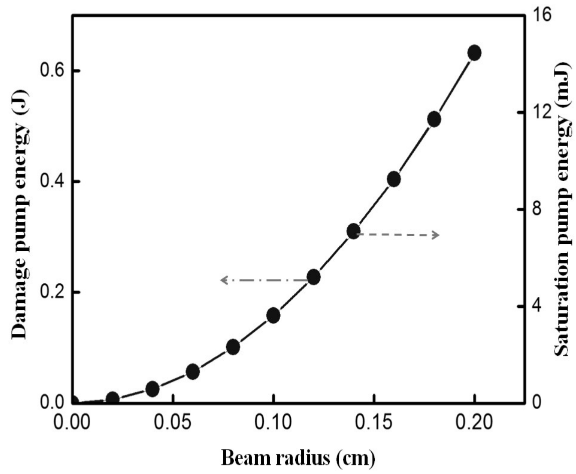

Fig. 2. Saturation pump and damage energy of the Ce:LiCAF crystal as a function of pumping beam radius at 266 nm pump wavelength.

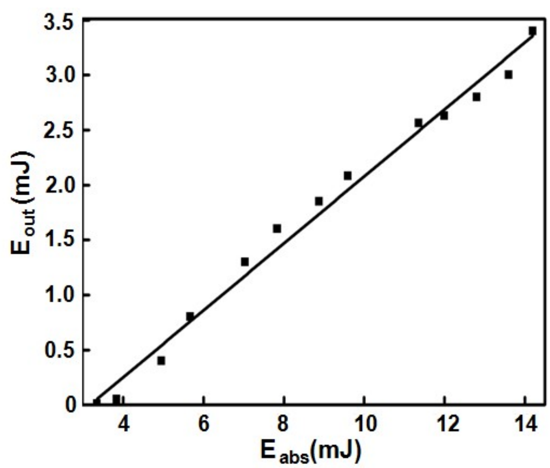

Fig. 3. Output energy of the broadband Ce:LiCAF laser emission as a function of absorbed pump energy. The laser slope efficiency is 33%.

Using the laser oscillator parameters described in Section 2.1 (96.7% end mirror reflectivity, 30% output coupler reflectivity, and 2 cm resonator length), we obtained a laser slope efficiency of around 33% for the broadband laser output at 290 nm as shown in Fig. 3. The maximum laser output pulse energy is 3.4 mJ at the absorbed pump energy of about 14 mJ. The absorbed pump energy was determined by subtracting the transmitted and reflected energy from the incident pump energy. The lasing threshold is at absorbed pump energy of 3.2 mJ.

The spectral profile of the broadband Ce:LiCAF laser output is shown in Fig. 4a. The broadband laser emission has a peak at around 290 nm, as expected from the 5d to 4f allowed dipole transition in Ce^{3+}. The spectral bandwidth measured is about 2.2 nm (FWHM) of a Gaussian fit to the spectral profile. The temporal profile, as shown in Fig. 4b, presents a pulse duration of about 4 ns (FWHM), which was also obtained from a Gaussian fit to the temporal profile.

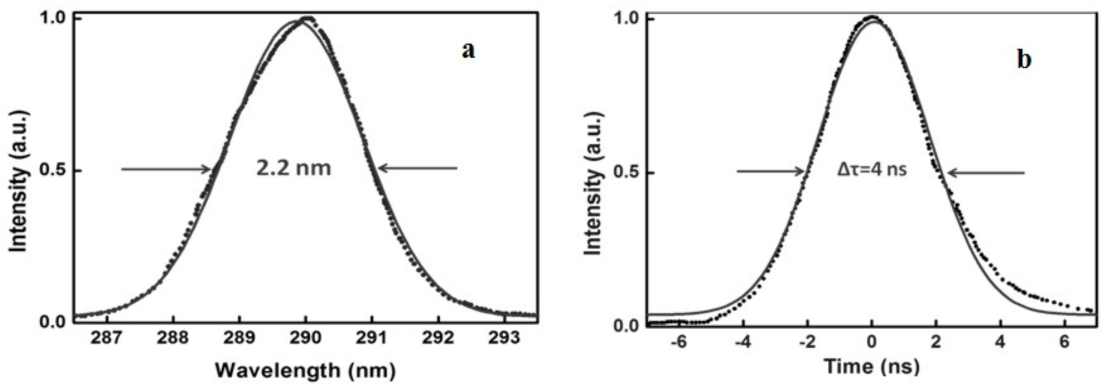

Fig. 4. (a) Spectral and (b) temporal profiles of the broadband Ce:LiCAF laser output from the resonator having 96.7% end mirror reflectivity, 30% output coupler reflectivity, 2 cm resonator length and 10 mJ pump energy pulses.

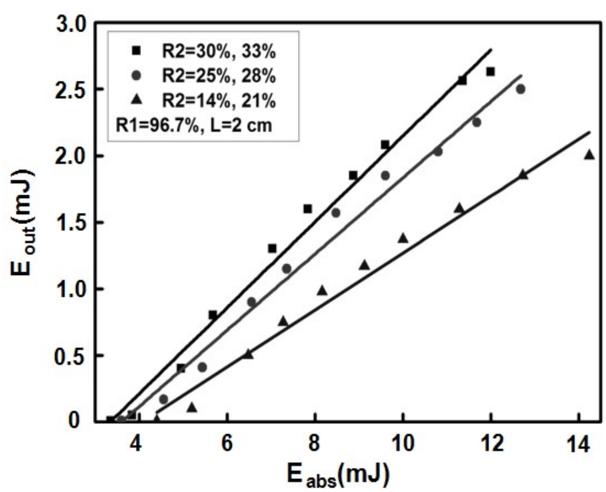

Fig. 5. Dependence of the laser slope efficiency on the reflectivity of the output coupler mirror (R_2). The resonator length is 2 cm.

The effect of output coupler reflectivities on the slope efficiency of the Ce:LiCAF laser output was investigated while the resonator length (L) was kept constant at 2 cm. Fig. 5 shows that a higher output coupler reflectivity results in a higher laser slope efficiency and a lower threshold pump energy. A laser slope efficiency of about 33% was achieved with a 2 cm resonator length and a 30% output coupler reflectivity.

To determine the effect of the resonator length on the laser slope efficiency, the resonator length was then varied from 2 cm to 4 cm while keeping the reflectivity of the output coupler constant at 30%. Figure 6 shows that the laser resonator of 2-cm length has a laser slope efficiency that is better than the others, yielding the slope efficiency of 33%.

One of the simple and effective methods to generate single short laser pulses with a nanosecond pump laser was based on resonator transient in which the laser used a low-Q and short resonator and a near-threshold laser opration [19]. For this purpose in the experiment, we created a 2 cm length and low-Q resonator of the Ce:LiCAF laser using the two mirrors with reflectivities R_1 = 25%, and R_2 = 14%, available in our laboratory. Therefore, the resonator round-trip time between the two mirrors and the cavity photon lifetime of this Ce:LiCAF laser resonator were calculated to be about 150 ps and 48 ps, respectively.

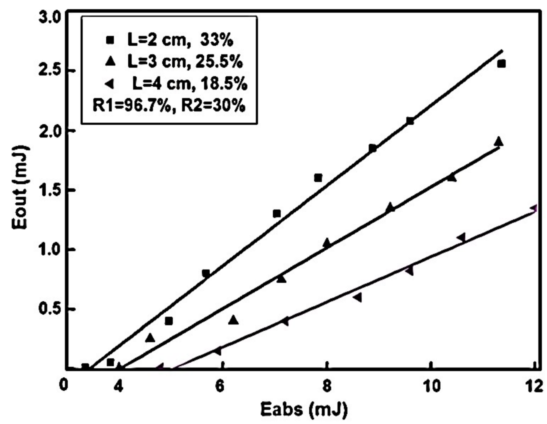

Fig. 6. Dependence of the laser slope efficiency on the resonator length (L). Reflectivity of the output coupler mirror (R_2) is 30%.

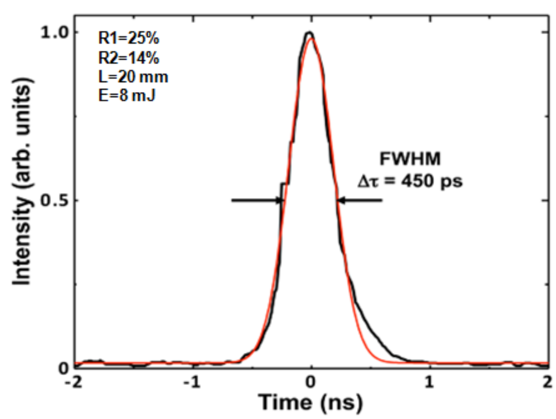

Fig. 7. Single 450 ps laser pulses at 290 nm were generated from the Ce:LiCAF laser by controlled resonator transient, corresponding to a pulse-shortening factor of 14 times.

Single shortest pulse of Ce:LiCAF laser emission was measured to be about 450 ps (FWHM) as shown in Fig. 7. Hence, the pulse shortening factor (ratio of the pumping pulse duration to that of the output laser pulse) was about 14 times. In the case, an output pulse energy of 1.2 mJ at 290 nm was achieved under an 8 mJ pump pulse energy which corresponds to 1.4 times higher than the laser threshold. It is clear that the shortest Ce:LiCAF laser emission was built up after about 3-5 resonator round-trip times and that the calculated cavity photon lifetime (48 ps) is about an order of magnitude shorter than the shortest laser pulse duration. Therefore, it is possible to produce shorter Ce:LiCAF laser pulse duration with a shorter length of the laser resonator.

III.2. Generation of spectrally tunable and narrow linewidth UV laser emission

The spectrally tunable and narrow linewidth Ce:LiCAF laser was simply constituted when the end mirror R_1 of the broadband laser resonator (Fig. 1) is replaced with a holographic grating

(2400 lines/mm, 2 cm $\times$2 cm), because the reflectivity of the diffraction grating is meadsured to be about 30% at 290 nm, the output coupler R_2 is kept with the mirror of 14% reflectivity. In this case, using a the pump pulse energy of 14 mJ, the tunable Ce:LiCAF laser is pumped well above laser threshold. The laser output emission is tunable from 281.5 nm to 299 nm. The laser achieved the conversion efficiencies from 8%–10% depending on the output laser wavelength. The linewidth within the tuning range is narrower than 0.2 nm (FWHM). It is noted that such laser conversion efficiencies will be considerably improved with new UV-optimized grating.

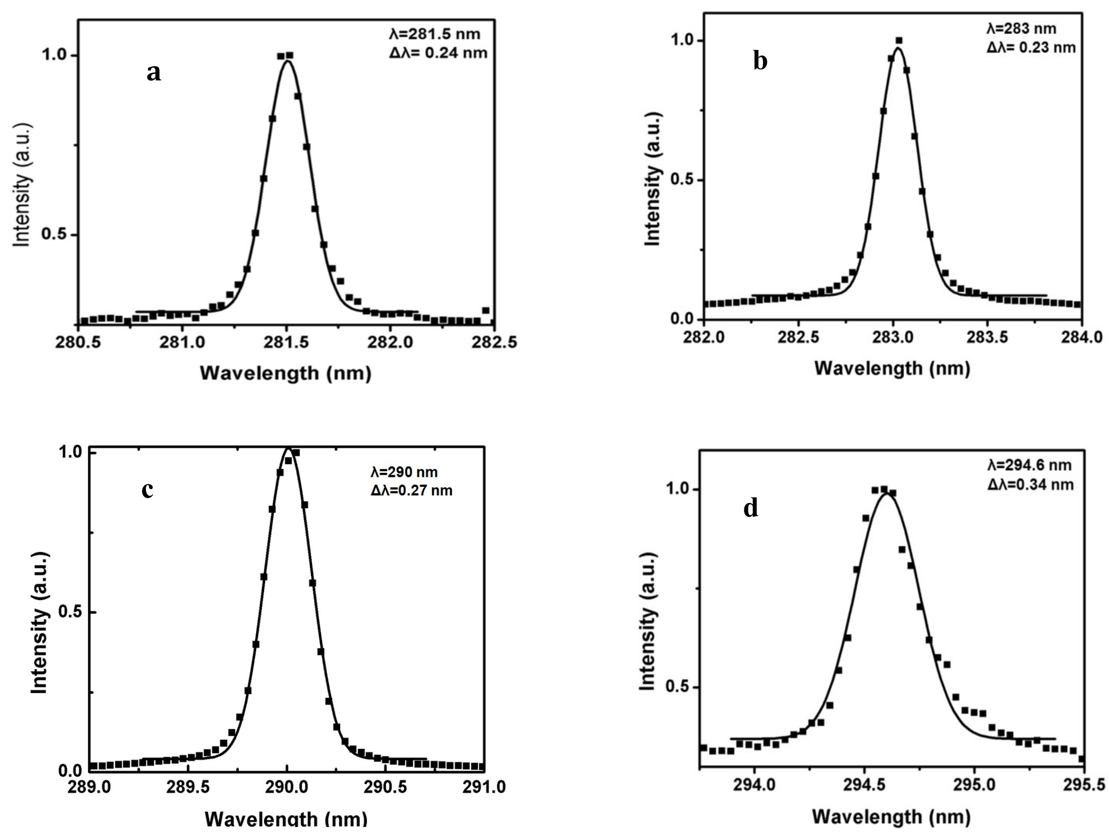

Fig. 8. Spectral profiles and linewidths of the tunable Ce:LiCAF laser emission for different wavelengths.

The spectral linewidths were also calculated according to Eq. (36) [21] to be less than 0.1 nm at difference laser wavelengths. The experimental results as shown in Fig. 8 are limited by the resolution of the spectrometer, which is 0.2 nm. These obtained results in spectral tunability and linewidth are quite better than those reported previously [22], where a tuning range from 284 nm to 294 nm and spectral linewidths of about 0.7 nm were obtained. Such continuously tunable UV laser emissions of spectral linewidth less than 0.2 nm are suitable for selectively optical excitation of many spectroscopic and environmental sensing applications.

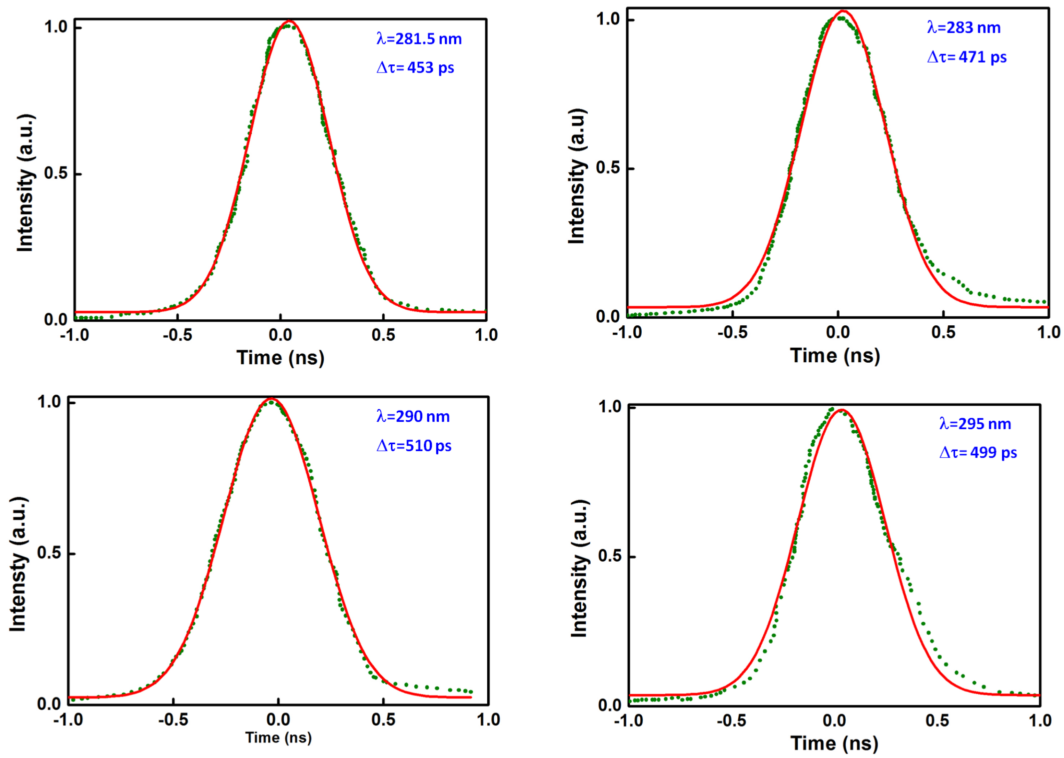

Fig. 9. Temporal profiles of single short (sub-nanosecond) laser pulse generated at different wavelengths at the near-threshold laser operation.

With this tunable laser configuration, we also studied the single short (sub-nanosecond) laser pulse generation at different wavelengths at the near-threshold laser operation regime. In the case, the tunable Ce:LiCAF laser was pumped by a pump pulse energy of 8 mJ, corresponding to 1.4 times higher than the laser threshold. The temporal profiles for single Ce:LiCAF laser laser pulses at different emission wavelengths are shown in Fig. 9. The laser pulse duration ranged from 453 ps (Fig. 9a) to 510 ps (Fig. 9c). To the best of our knowledge, the tuning range from 281 to 299 nm is the broadest tuning range for sub-nanosecond laser pulse emission from Ce:LiCAF. The obtained results demonstrate that short (subnanosecond) laser pulse generation in the UV region are available as optical excitation sources for time- resolved spectroscopy and measurements.

IV. CONCLUSION

In conclusion, we demonstrate that modest laboratories could successfully develop different pulsed UV laser sources using Ce:LiCAF crystal as gain medium and the fourth harmonics (266 nm) of a nanosecond Q-switched Nd:YAG laser as a pump laser. With the single–grating Ce:LiCAF laser resonator configuration, we produced continuously tunable laser emission from 281 nm to 299 nm maintaining a linewidth narrower than 0.2 nm (FWHM) and a pulse energy of about 1 mJ. Furthermore, single sub-nanosecond laser pulses (450 ps) were generated at any wavelengths in the tuning range from 281 nm to 299 nm controlled resonator transient. Such UV laser emissions are suitable for many spectroscopic and environmental sensing applications.

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