

Femtosecond mode-locked Nd,La:CaF₂ disordered crystal laser

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Abstract: A diode-pumped femtosecond mode-locked Nd,La:CaF₂ disordered crystal laser was reported for the first time. By appropriately choosing the Nd and La-doping concentration, stable mode-locked femtosecond laser pulses were obtained by using a semiconductor saturable absorber mirror (SESAM). The laser produced 633 fs pulses at the central wavelength of 1061 nm with an average output power of 200 mW at 82 MHz repetition rate.

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OCIS codes: (140.3480) Lasers, diode-pumped; (140.4050) Mode-locked lasers; (140.3580) Lasers, solid-state; (140.3380) Laser materials.

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1. Introduction

In recent years, compact, efficient and robust diode-pumped all-solid-state femtosecond lasers in the infrared spectral range are desired for many scientific research and industrial applications, such as ultrafast spectroscopy, medical treatment, and superfine materials processing. So far, neodymium (Nd³⁺) doped laser material has been one of the most widely used gain medium in all-solid-state lasers due to its large emission cross section, long upper level lifetime and efficient four-level system [1, 2]. Among hundreds of Nd³⁺ doped materials, Nd:YAG and Nd:YVO₄ crystals received the most attention in such as laser diode (LD) pumped high power and high efficiency picosecond oscillators and amplifiers, owing to their advantages of low cost, long lifetime and stable performance [3–7]. 27W, 20 ps; 20W, 20 ps pulses were produced from a Nd:YAG laser and a Nd:YVO₄ laser by virtue of SESAM mode-locking in 2000, respectively [8, 9]. Limited by their narrow gain bandwidth, most of the singly doped Nd³⁺ materials can only generate picosecond lasers and only a small portion of which support the generation of femtosecond laser such as Nd:glass [10, 11]. Pulse as short as 60 fs was generated from a Nd:glass laser but the bad thermal property limited its application [12]. As a result it's meaningful to seek new kinds of Nd³⁺ doped laser materials for the generation of high power and high efficiency femtosecond lasers.

Due to the inhomogeneous broadening effect, neodymium (Nd³⁺) in fluoride behaves relatively large emission cross section and a long upper level lifetime. In the meantime, fluoride as the substrate possesses the advantages of large size, low nonlinear refractive coefficient and high thermal conductivity. All the merits make Nd³⁺ doped fluoride crystals promising laser materials for generating high power and high repetition rate ultrashort pulses. However, singly Nd³⁺-doped fluoride crystal also has fatal defect due to a very detrimental concentration quenching effect caused by Nd³⁺-Nd³⁺ clusters [13]. This situation could be released by co-doping with "buffer" ions like trivalent rare-earth ions (Sc³⁺, Y³⁺, La³⁺, Gd³⁺, Lu³⁺) or monovalent metal ions (Li⁺, Na⁺, K⁺, Ag⁺). In the co-doped fluoride crystals, the Nd³⁺-Nd³⁺ clusters could be broken, resulting in improved luminescence efficiency and fluorescence bandwidth. Several experiments on mode-locking of Nd,Y-codoped fluoride crystals have proved the potential of these crystals [14, 15]. Especially near and sub-100 fs pulses have been generated from Nd,Y:CaF₂ and Nd,Y:SrF₂, respectively, suggesting they are promising for the ultrashort pulse generation [16, 17]. It's also a great interest to explore the laser performance of Nd³⁺ fluoride crystals codoped with other trivalent rare-earth ions, like Lu³⁺, La³⁺, and Sc³⁺ [18, 19]. Recently, C. Li et al. studied continuous wave (CW) and mode-locked operation of a Nd,La:CaF₂ laser [20]. They realized picosecond mode-locking with pulse duration of 11 ps and average output power of 110 mW. However, there has been no report on the generation of femtosecond pulses from the Nd,La:CaF₂ laser.

In this paper, we demonstrate a diode-pumped femtosecond passively mode-locked laser based on the Nd,La:CaF₂ disordered crystal. By choosing appropriate La-doping concentration, the maximum CW laser output power of 805 mW was obtained under an incident pump power of 5 W. With 0.5 at.% Nd, 8 at.% La:CaF₂ crystal as the gain medium, 633 fs pulses with an average output power of 200 mW were obtained at a repetition rate of 82 MHz. The central wavelength was at 1061 nm with 2.8 nm bandwidth. This is to the best of our knowledge, the

first demonstration of femtosecond mode-locking operation in a diode-pumped Nd,La:CaF₂ laser.

2. Experimental setups and results

The Nd,La:CaF₂ crystal was grown by temperature gradient technique (TGT) and doped with Nd³⁺ and La³⁺ ions. At room temperature, the absorption and fluorescence spectra of Nd,La:CaF₂ samples with 0.5 at.% Nd³⁺ and different La³⁺ concentration were shown in Fig. 1. As the La³⁺ doping concentration increases, the spectral intensity changes while the absorption and emission peaks remain unchanged. Room temperature absorption and fluorescence spectra show that three absorption peaks located at 736, 791 and 796 nm and two emission peaks at 1050 and 1065 nm, respectively. The main absorption band at 791 nm matches the commercial laser diode as pump and the broad fluorescence bandwidth from 1030 to 1110 nm supports ultrashort laser generation.

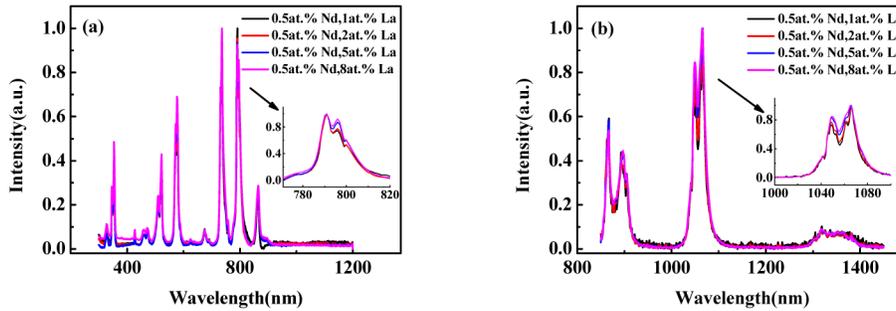


Fig. 1. Absorption (a) and fluorescence (b) spectra of Nd, La:CaF₂ disordered crystal at the room-temperature.

The experimental setup for CW laser operation was shown as Fig. 2. A standard plano-concave laser resonator with output coupler mirrors of different transmissions was designed to investigate the laser performance of different doping concentration of the Nd,La:CaF₂ crystals. A 790 nm fiber-coupled laser diode with 100 μ m fiber core diameter and a numerical aperture of 0.22 was used as the pump source. The 0.5 at.% Nd, 1 at.%, 2 at.%, 5 at.% and 8 at.% La:CaF₂ samples with thickness of 5 mm and cross section of 3 mm \times 3 mm were prepared as the laser mediums. The pump laser was focused into the sample by using a coupling system with a magnification ratio of 1:0.8, resulting in a pump beam waist radius of about 40 μ m in the crystal. To move the heat, the sample was wrapped with indium foil and mounted on a water-cooled copper kept at 12°C. M1 was a dichroic mirror with high transmission at 790 nm and high reflection at 1020 - 1100 nm. M2 was a concave mirror with radius of curvature (ROC) of 200 mm. OC was a plane output coupler mirror with a transmission of 0.8%, 1.6%, and 2.5%, respectively, at 1020 - 1100 nm.

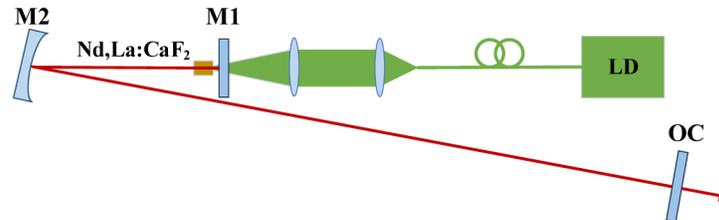


Fig. 2. Experimental setup of continuous wave Nd,La:CaF₂ laser.

The relationship between the average output power and the pump power with different OCs and different La³⁺ doping concentration is shown in Fig. 3. Under the pump power of 5 W, the

maximum output power for 1 at.%, 2 at.%, 5 at.%, and 8 at.% La^{3+} -doped samples are 710 mW, 555mW, 758 mW, and 805 mW, respectively, corresponding to the slope efficiency of 23.7%, 18.5%, 25.27%, and 26.83%, respectively. For all samples the highest output power is obtained with the 0.8% OC, which may be explained by the decreased power density of the oscillator which the pumping rate remained invariant. Figure 3 also indicates there is no saturation phenomenon for all samples. However, we did not further increase the pump power for fear of crystal fracture.

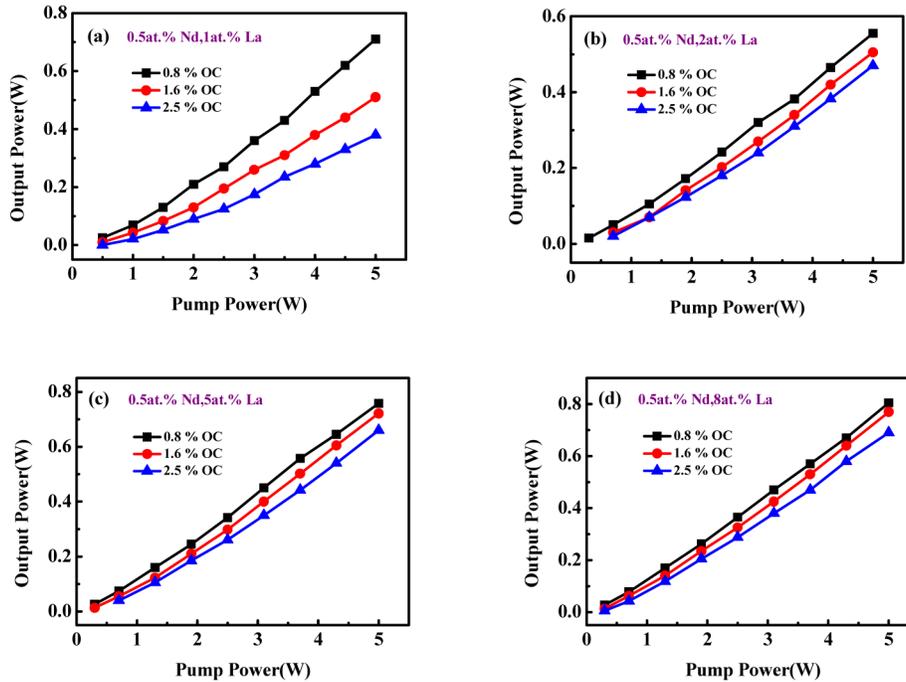


Fig. 3. Continuous wave output power versus the pump power for 0.5 at.% Nd:CaF₂ samples co-doped with (a) 1at.%, (b) 2at.%, (c) 5at.%, and (d) 8at.% La^{3+} for the output couplers of 0.8%, 1.6%, and 2.5% transmissions.

The mode-locking experiment was conducted with the setup shown in Fig. 4. In this case, the 0.5 at.% Nd^{3+} and 8 at.% La^{3+} CaF₂ crystal was utilized to study the mode-locking performance. We inserted in a pair of Gires–Tournois interferometer (GTI) mirrors for chirp compensation, which can provide about -250 fs^2 and -300 fs^2 group delay dispersion (GDD) per bounce at 1064 nm, respectively. In order to realize self-starting mode-locking, a concave mirror with ROC of 300 mm was used to focus the laser on the SESAM (Batop GmbH). The SESAM is designed with a modulation depth of 0.6%, a non-saturable loss parameter as low as 0.4%, and a saturation fluence of $70 \mu\text{J}/\text{cm}^2$. Based on the ABCD matrix calculation, the beam radius on the SESAM was $65.5 \mu\text{m} \times 67.4 \mu\text{m}$. In this setup, an OC with a transmission of 0.8% was used as the folding mirror. The total cavity length is 1.8 m corresponding to a pulse repetition rate of 82 MHz.

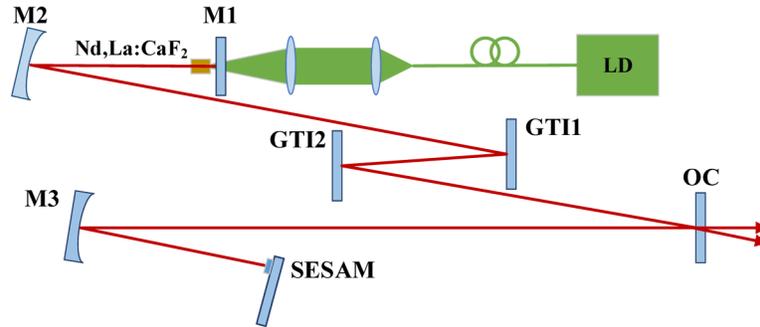


Fig. 4. Experimental setup of the diode-pumped passively mode-locked femtosecond Nd,La:CaF₂ laser.

With increasing the pump power, the laser experiences Q-switching, unstable Q-switching mode-locking till stable CW mode-locking. Figure 5 shows the mode locking pulse train detected by a fast photodiode and recorded with a digital phosphor oscilloscope with 500 MHz bandwidth (Tektronix DPO 3052). Long range scanning shows the mode-locking is clean without obvious Q-switching instability. Under an incident pump power of 4.5 W (the measured absorbed pump power was 2.7 W), we obtained a total 200 mW mode-locked output power from the OC. Figure 6 shows the average output power of the laser with respect to the incident pump power under different operation conditions.

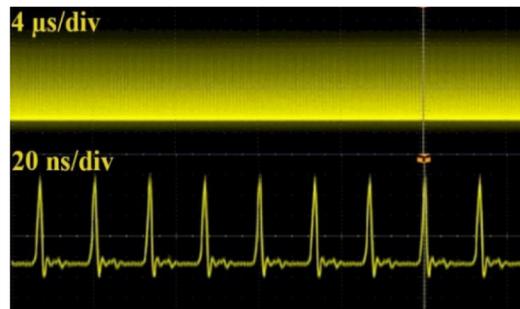


Fig. 5. Mode-locked pulse trains at 4 μ s/div and 20 ns/div, respectively.

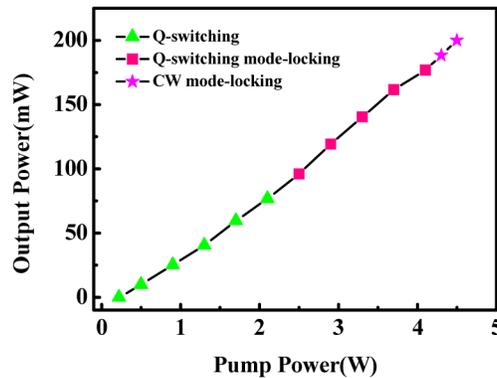


Fig. 6. Average output power as a function of the pump power.

The pulse duration was characterized by an intensity autocorrelator (APE: PulseCheck USB). Shortest pulse duration was obtained by replacing the GTIs with different GDD and bouncing times. Figure 7(a) shows the typical intensity autocorrelation trace when the GTIs

introduced a total GDD of -550 fs^2 . Assuming a sech^2 -pulse shape, we obtained a femtosecond pulse duration of 633 fs. The corresponding mode-locking spectrum was measured by a fiber coupled spectrometer (HR 4000, Ocean Optics, 0.4 nm wavelength resolution) which is shown in Fig. 7(b). The laser spectrum is centered at 1061 nm with a full width at half maximum (FWHM) bandwidth of 2.8 nm. The time-bandwidth product is calculated to be 0.472 which is a little bigger than the Fourier transform limited sech^2 -pulses. Further optimizing the cavity chirp should enable us to generate sub-500 fs pulses by this setup.

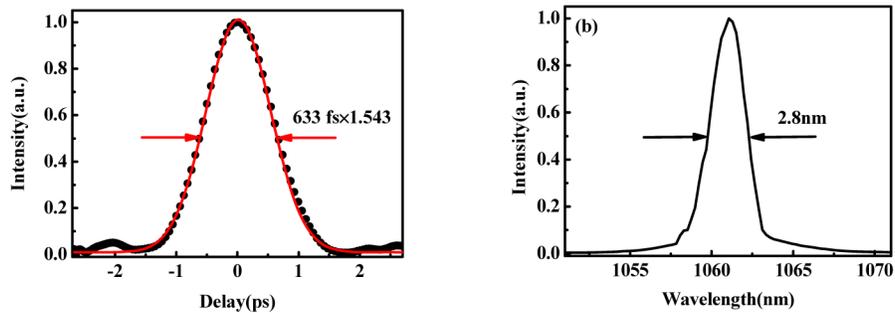


Fig. 7. Intensity autocorrelation trace (a) and laser spectrum (b) of the mode-locked Nd,La:CaF₂ laser.

3. Conclusion

In conclusion, we demonstrated a diode-pumped femtosecond mode-locked operation based on a Nd,La:CaF₂ disordered crystal for the first time. As a representative example, using the 0.5 at.% Nd³⁺ and 8 at.% La³⁺ CaF₂ crystal, pulses as short as 633 fs with a spectral bandwidth of 2.8 nm at the central wavelength of 1061 nm were generated. The average output power of 200 mW was obtained under an incident pump power of 4.5 W. Complete investigation of the femtosecond mode-locking performances of the Nd:CaF₂ disordered crystals with different co-doping ions and concentration will give us better understanding of the laser potential of the co-doping fluoride crystals. By optimizing the chirp compensation and laser efficiency, it is greatly possible to generate high power sub-500 fs pulses and to find potential application in diode-pumped solid state amplifier.

Acknowledgment

This work was supported by the National Major Scientific Instruments Development Project of China (Grant No. 2012YQ120047) and the National Natural Science Foundation of China (Grant Nos. 61205130 and 61422511).