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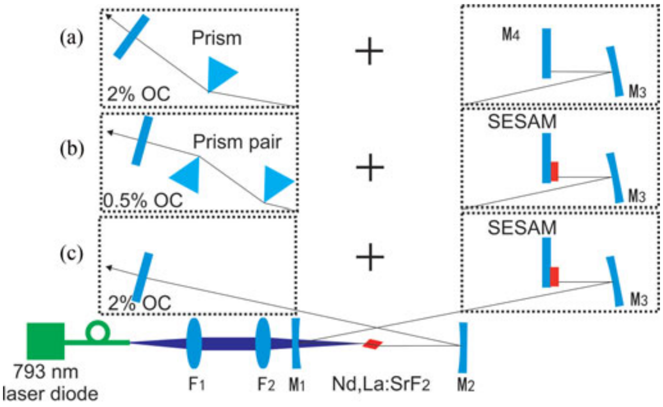

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Femtosecond and Dual-Wavelength Picosecond Operations of Nd,La:SrF$_2$ Disordered Crystal Laser

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Abstract: Both femtosecond and dual-wavelength picosecond operations of Nd,La:SrF$_2$ disordered crystal laser have been demonstrated for the first time. In femtosecond operation, the laser generated mode-locked pulses with pulse duration of 156 fs, which is an average power of 90 mW, and a repetition rate of 108 MHz at the central wavelength of 1057 nm. In dual-wavelength picosecond operation, the laser generated synchronously mode-locked pulses with an average power of 500 mW and pulse duration of 9.9 ps. The mode-locked spectrum clearly shows two peaks at 1050.8 and 1059.0 nm, respectively. A periodic beat frequency of 2.2 THz is observed from the intensity autocorrelation trace, indicating the synchronous dual-wavelength mode-locking. Our experimental results show that Nd,La:SrF$_2$ disordered crystal is an excellent laser material for femtosecond and dual-wavelength lasers near 1 μm wavelength.

Index Terms: Laser crystals, ultrafast lasers, diode-pumped lasers, mode-locked lasers.

1. Introduction

Ultrafast laser sources at 1 μm wavelength have shown wide applications in nonlinear spectroscopic techniques, ophthalmology, ultrafast micromachining, femtosecond chemistry, and so on [1], [2]. Neodymium (Nd)-doped laser gain media are generally a favorable choice to produce ultrashort pulse at 1 μm wavelength because of their large emission cross-section and moderate lifetime of upper energy level. Especially, the four-energy-level characteristic of Nd^{3+} ion makes it easy to realize population inversion and yield high gain. High-average-power mode-locked lasers have been demonstrated based on mature Nd:YAG and Nd:YVO$_4$ crystals [3], [4]; however, their gain linewidths only support picosecond pulse generation. Although Nd:glass has a wide gain linewidth to support femtosecond pulse generation and amplification, its poor thermal conductivity prevents production of high average power [5]. Therefore, much attention has been paid to develop new Nd-based broadband laser materials for ultrashort pulse generation.

Nd-doped disordered crystals have attracted wide interest due to their broad gain linewidth arising from inhomogeneous spectral broadening and splitting of disordered structure. In 2009 and 2010, Xie *et al.* realized 900 fs and 534 fs mode-locked pulse generation from Nd:CLNGG and Nd:CNGG-CLNGG disordered crystal lasers, respectively [6], [7]. In the following years, femtosecond mode-locked pulses were generated in Nd-doped melilite, langasite, fluoride, and borate disordered crystal [8]–[12]. The shortest pulse as short as 79 fs was achieved from the Nd:$Ca_3La_2(BO_3)_4$ disordered crystal laser [12]. Besides the advantage of femtosecond pulse generation, disordered crystals are also excellent gain media for dual-wavelength or triple-wavelength operations due to their multiple emission centers [13]–[18].

Nd,La:SrF_2 is a typical disordered crystal, which belongs to the family of MeF_2-LnF_3-NdF_3 ($Me^{2+} = Ca^{2+}$, Sr^{2+}, Ba^{2+}, Cd^{2+}; $Ln^{3+} = Y^{3+}$, La^{3+}). As we know, the cluster effect of Nd^{3+} in Nd:SrF_2 results in fluorescence quenching even at low Nd-doping level. However, the addition of La^{3+} buffer ion to Nd:SrF_2 significantly alleviates this problem [19], [20]. In the Nd,La:SrF_2 crystal lattice, the Sr^{2+} ion is replaced by the Nd^{3+} or La^{3+} ion, and the heterovalent charge of substitution is compensated by the additional F^- ion, resulting in various optical centers [21]. As each optical center possesses individual emission line, the fluorescence spectrum of Nd,La:SrF_2 is a collection of different optical center emission, which results in a significant inhomogeneous broadening and splitting. Therefore, Nd,La:SrF_2 crystal is a potential alternative as gain medium of femtosecond laser and dual-wavelength laser. So far, only sub-picosecond mode-locking operation has been reported in the Nd,La:SrF_2 disordered crystal laser, which generated 739-fs mode-locked pulses [22].

In this paper, we demonstrate both femtosecond mode-locked and dual-wavelength synchronous picosecond mode-locked operations of the Nd,La:SrF_2 disordered crystal laser for the first time. In femtosecond operation with fine intracavity dispersion compensation, the laser generated mode-locked pulses with pulse duration of 156 fs, the average output power of 90 mW, and the repetition rate of 108 MHz at the wavelength of 1057 nm. Without the dispersion compensation, stable dual-wavelength synchronous mode-locked was realized with pulse duration of 9.9 ps and maximum average output power of 500 mW at the wavelength of 1050.8 and 1059.0 nm. A clear beat frequency modulation was observed in the autocorrelation (AC) trace, indicating that the dual-color pulses were temporally synchronous in the laser. Such temporally synchronous dual-color ultrashort pulses are very useful for generation of coherent terahertz radiation and ultrahigh-repetition-rate pulses [23], [24].

2. Wavelength Tuning Experiment

First, we performed the wavelength tuning experiment of Nd,La:SrF_2 crystal. The experimental setup for wavelength tuning is shown in Fig. 1(a). The Nd,La:SrF_2 crystal has a maximum absorption cross-section of 1.32×10^{-20} cm^2 at 795 nm. We chose a fiber-coupled 793-nm laser diode (LD) as the pump source. The fiber had a numerical aperture (NA) of 0.15 and a core diameter of 105 μm. After collimated and focused by the F_1 and F_2 lenses with a same focal length of 10 cm, the pump beam was imaged into the crystal. The fluorescence spectrum was measured for different Nd,La:SrF_2 samples with the fixed Nd doping concentration (0.5 at. %) and variable La doping concentration of 2 at.%, 5 at.%, and 8 at.%. It is found that the introduction of buffer ion increases the fluorescence intensity and broadens the emission bandwidth markedly [22]. Here, we chose the optimal sample with 0.5 at.% Nd and 8 at.% La doping concentration. The peak emission cross-section and fluorescence lifetime were measured to be 2.73×10^{-20} cm^2 and 484.5 μs, respectively. Although the thermal conductivity decreases from 9.3 W/mK in undoped SrF_2 crystal to 3.5 W/mK in our sample, it is still three times as large as that of phosphate glass [25], [26]. The Nd,La:SrF_2 crystal with a length of 6.5 mm was placed at Brewster angle to minimize the Fresnel loss for p-polarization light and avoid the formation of parasitic cavity which is detrimental to mode-locked operation. In order to remove the generated heat while pumping, the laser crystal was wrapped with indium foil and tightly mounted in a water-cooled copper holder, and the circulating water temperature was sustained at 15 °C. The three plano-concave mirrors M_1, M_2 and M_3 had the same radius of curvature (ROC) of 10 cm and all of them were coated for high reflectivity at

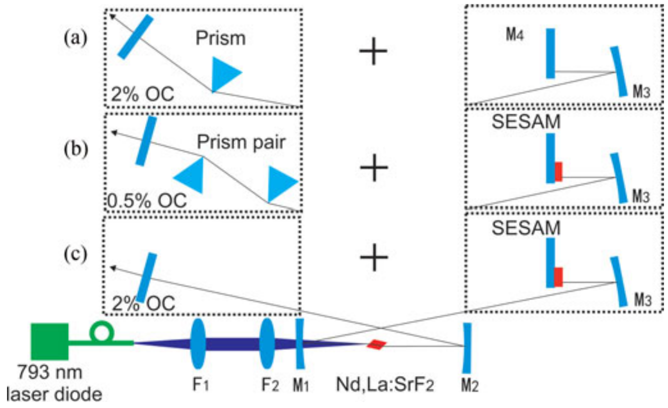

Fig. 1. Experimental setup for (a) wavelength tuning, (b) femtosecond mode-locking, and (c) dual-wavelength picosecond mode-locking. SESAM: semiconductor saturable absorber mirror; OC: output coupler.

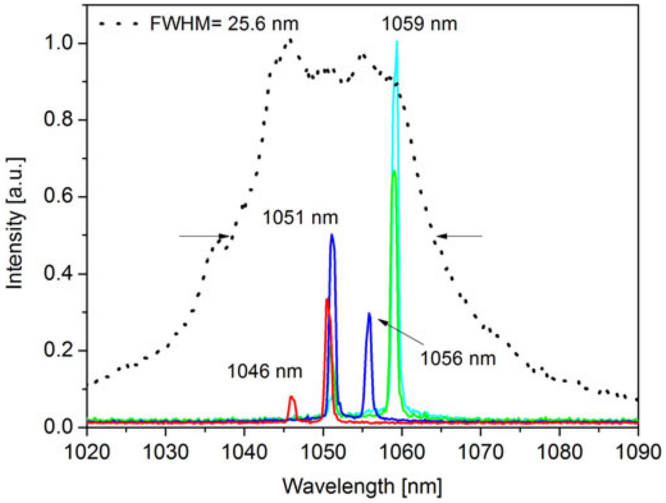

Fig. 2. Fluorescence spectrum (black dot line) and wavelength tuning spectra (colorful solid line).

the laser wavelength and high transmittance at the pump wavelength. The output coupler (OC) had a transmittance of 2%. A single SF10 prism was inserted in the cavity as the wavelength tuning element.

For comparison, the fluorescence spectrum of the Nd,La:SrF_2 disordered crystal was recorded by the optical spectral analyzer (Ocean Optics, USB4000) with a resolution of 1.2 nm, as shown in Fig. 2. The pump wavelength was 793 nm. As expected, Nd,La:SrF_2 disordered crystal shows a broad emission bandwidth of 25.6 nm (full-width at half-maximum (FWHM)), which is even comparable to that of Nd:glass [27]. Under the absorbed pump power of 2.3 W, the wavelength tuning spectra could be obtained by slightly adjusting the angle of OC, as shown in Fig. 2. The laser wavelength cannot be tuned continuously. In the tuning curve there exist four peaks at 1046, 1051, 1056, and 1059 nm, which is the typical characteristic of disordered crystals due to multiple emission centers. The discrete emission peaks of Nd,La:SrF_2 make it possible to achieve multiple wavelength operation.

3. Femtosecond Operation of the Nd,La:SrF_2 Laser

To obtain femtosecond operation, we replaced the mirror M_4 by a semiconductor saturable absorber mirror (SESAM). The employed SESAM had a modulation depth of 1.8% and non-saturable loss

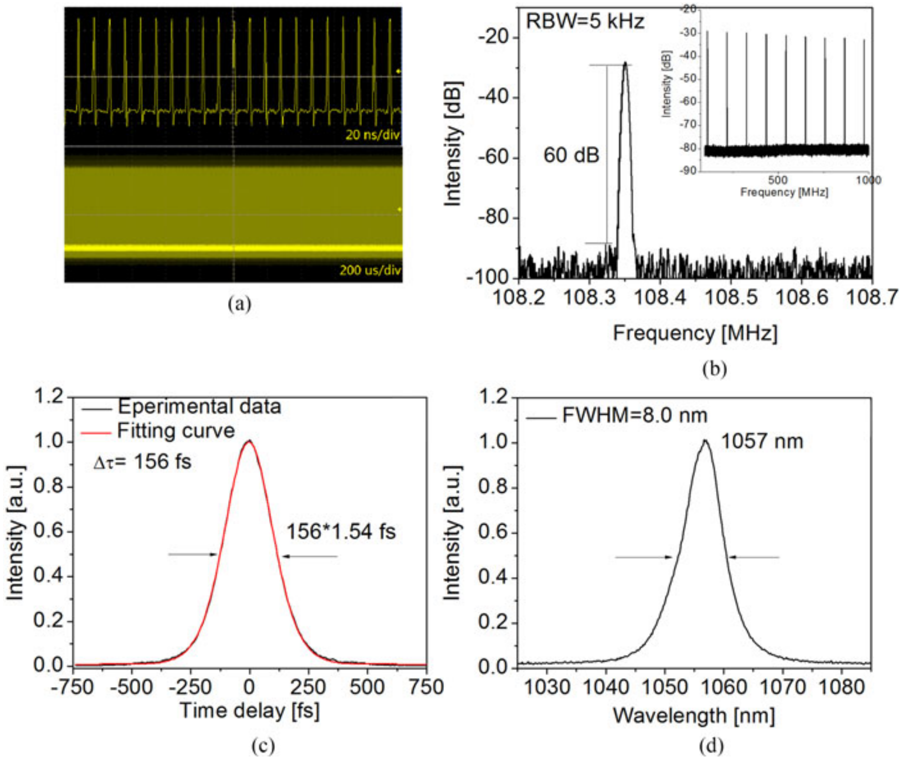

Fig. 3. (a) CW mode-locked pulse trains in nanosecond and microsecond scales. (b) Radio frequency spectrum with a span of 0.5 MHz and a resolution bandwidth of 5 kHz. (Inset) Radio frequency spectrum with a span of 1 GHz and a resolution bandwidth of 300 kHz. (c) Autocorrelation trace of the mode-locked pulses. (d) Mode-locked pulse spectrum.

of 1.2%. An OC with a lower transmittance of 0.5% was used, which is beneficial to increase the intracavity power and enhance self-phase modulation (SPM) effect for broadening the laser spectrum. Otherwise, a pair of SF10 prism was used to finely compensate for the intracavity dispersion.

In order to get shortest femtosecond mode-locked pulses, firstly, it is necessary to compensate the intracavity group delay dispersion (GDD). Compared with GTI mirrors and chirped mirrors, the prism pair can provide adjustable negative GDD by changing the insertion amount. Therefore, the shortest mode-locked pulse can be achieved with the optimal net GDD in the cavity. Second, a large modulation depth of SESAM is beneficial to shorten the mode-locked pulses. We compared the mode-locked results by employing SESAMs with modulation depth of 0.6%, 0.9% and 1.8%, and found that with the SESAM of modulation depth of 1.8% we could achieve the shortest mode-locked pulse. Third, we adopted an OC with a low transmittance (0.5%) to increase the intracavity power and enhance the SPM effect. The enhanced SPM effect can broaden the laser spectrum and shorten the mode-locked pulse.

In the experiment, stable CW mode-locking was realized under the absorbed pump power of 2.6 W (corresponding incident pump power of 3.7 W), delivering an average output power of 90 mW. The low optical-to-optical conversion efficiency ($\sim$2.4%) can be attributed to the low transmittance (0.5%) of the output coupler, the insert loss of prism, and the large non-saturable loss (1.2%) of the SESAM. When we increased the incident pump power beyond 3.7 W, the output power tended to saturate. The stable CW mode-locking can be sustained for several hours. Typical CW mode-locked pulse trains are shown in Fig. 3(a). The radio frequency (RF) spectrum in Fig. 3(b) shows a high signal-to-noise ratio of 60 dB, indicating the clear mode-locking operation. From the RF spectrum, it shows a pulse repetition rate of 108.35 MHz, which corresponds to the cavity length of $\sim$1.38 m. In mode-locking operation, the output laser beam had a round TEM_{00} mode.

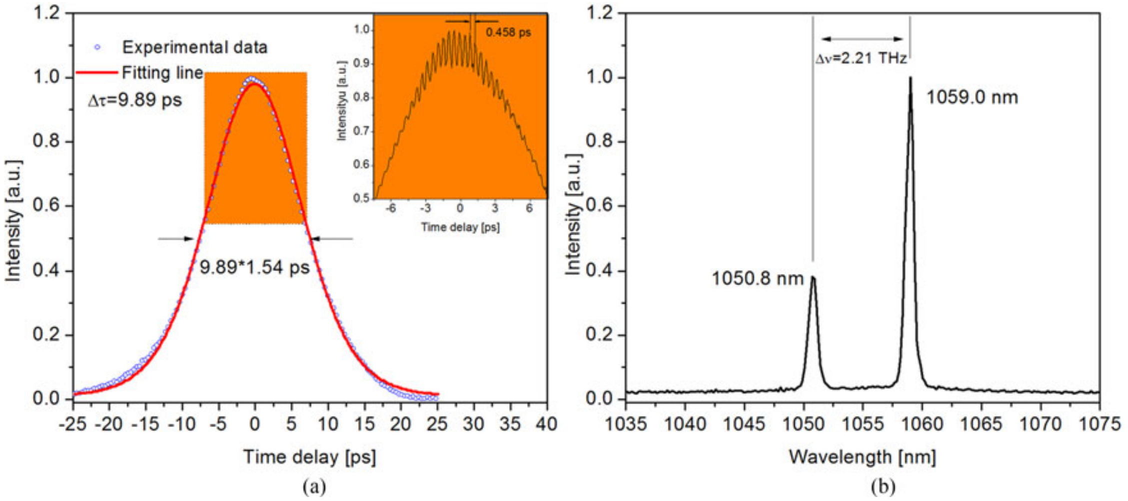

Fig. 4. (a) Autocorrelation trace of dual-wavelength synchronously mode-locked pulses. (Inset) Autocorrelation trace with higher resolution. (b) Mode-locked pulse spectrum.

In the experiment, we carefully optimized the intracavity dispersion for achieving the shortest mode-locked pulses. The tip-to-tip distance of the two prisms was fixed at 25 cm. While the prism pair provided a GDD of -720 fs^2, the measured pulse duration was 383 fs. As we increased insertion amount of prism, the pulse duration gradually shortened and the pulse spectrum gradually broadened. When a GDD of -350 fs^2 is provided by the prism pair, we obtained the shortest mode-locked pulse of 156 fs and the corresponding intensity AC trace is shown in Fig. 3(c), in which a Sech^2 pulse shape is assumed. The mode-locked spectrum has a FWHM of 8 nm with a central wavelength of 1057 nm, as shown in Fig. 3(d). The time-bandwidth product is 0.33, which is close to the transform limit value of 0.315 for Sech^2 pulses. With further increasing the insertion amount of prism, the CW mode-locking became unstable near zero dispersion region. If we continued to increase the insertion amount of prism, the unstable Q-switched mode-locking operation turned into stable picosecond pulse output, suggesting that the net cavity dispersion became positive. The transformation of femtosecond and picosecond mode-locking operations could be repeated by adjusting the insertion amount of prism.

4. Dual-wavelength Picosecond Operation of the Nd,La:SrF$_2$ Laser

Since multiple emission peaks exist in the Nd,La:SrF$_2$ disordered crystal as shown in the tuning experiment, dual-wavelength synchronous mode-locking was realized with the setup in Fig. 1(c). For the dual-wavelength mode-locking experiment, we pulled out the prisms and employed a SESAM with a modulation depth of 0.6%. In addition we adopted an OC with a transmittance of 2% in order to improve the average output power. Stable CW mode-locking was also obtained and the maximum average output power was 500 mW at the absorbed pump power of 2.08 W. The output laser beam had a good TEM$_{00}$ mode. While we further increased the pump power, the dual-wavelength mode-locking operation would switch into single-wavelength mode-locking operation. Fig. 4 shows the AC trace and the optical spectrum of the mode-locked pulses. The mode-locked pulse spectrum clearly shows two peaks at 1050.8 nm and 1059.0 nm with a frequency interval of $\sim$ 2.21 THz. The stable dual-wavelength mode-locking operation could be sustained for several hours and the intensity and wavelength interval in the mode-locking spectrum kept unchanged. Assuming a Sech^2 pulse shape, the AC trace shows the mode-locked pulse duration of 9.89 ps. The optical beating between 1050.8 nm and 1059 nm is observed in the AC trace (see the inset of Fig. 4(a)), which indicates that the dual-color pulses are temporally synchronous and spatially superimposed. The low visibility of the

AC trace results from the big intensity difference between two wavelengths. In fact, if the intensities at two wavelengths are equal, a high beating visibility can be achieved in the AC trace [28]. The beating pulses have a period of 0.458 ps, corresponding to a beating frequency of ∼2.2 THz. The dual-wavelength synchronously mode-locked laser can be used to generate coherent THz wave by difference frequency process.

5. Conclusion

Both femtosecond mode-locking and dual-wavelength synchronous picosecond mode-locking operations of the Nd,La:SrF_2 disordered crystal laser have been experimentally demonstrated for the first time. Benefiting from the inhomogeneous spectral broadening and splitting of the disordered crystal, the Nd,La:SrF_2 mode-locked laser generated femtosecond pulses with a pulse duration as short as 156 fs, an average output power of 90 mW and a repetition rate of 108 MHz at 1057 nm. By utilizing multiple emission peaks in the disordered crystal, dual-wavelength synchronous mode locking was also realized with mode-locked pulse duration of 9.89 ps and an average output power of 500 mW at the wavelengths of 1050.8 nm and 1059 nm. To our best knowledge, it is the first time to demonstrate both femtosecond operation and dual-wavelength synchronous picosecond operation from a disordered crystal laser, which will meet various applications.

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