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Efficient femtosecond mode-locked Nd,Y:SrF₂ laserLong Wei¹, Hainian Han^{2*}, Wenlong Tian¹, Jiaxing Liu², Zhaohua Wang², Zheng Zhu³, Yulei Jia³, Liangbi Su⁴, Jun Xu⁴, and Zhiyi Wei^{1,2*}¹School of Physics and Optoelectronic Engineering, Xidian University, Xi'an 710071, China²Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China³College of Science, Chinese University of Petroleum, Qingdao 266580, China⁴Key Laboratory of Transparent and Opto-functional Inorganic Materials, Shanghai Institute of Ceramics, CAS, Shanghai 201800, China

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An efficient femtosecond mode-locked laser using Nd and Y-codoped SrF₂ crystal as the gain medium is presented in this letter. A 332 fs pulse centered at 1057 nm with a repetition rate of 89.8 MHz, a spectral width of 4.3 nm, and a mode-locked output power of up to 395 mW has been obtained under 1 W pump power, corresponding to an optical-to-optical efficiency of 39.5% and a slope efficiency of 69%. To the best of our knowledge, this is the highest optical efficiency in femtosecond Nd-doped crystal lasers. © 2014 The Japan Society of Applied Physics

The scientific and industrial applications of femtosecond lasers continuously promote research on femtosecond lasers with high efficiencies and low costs.^{1,2)} Nd-doped materials are important gain media for generating ultrafast lasers.^{3–7)} To date, most reports on Nd ultrafast lasers describe high output powers or high efficiencies, but long pulse durations at picosecond level because of their narrow emission spectral widths.^{5–7)} There also are some femtosecond Nd lasers based on Nd-doped disordered crystals or Nd:glass.^{8–12)} Nd-doped disordered crystals are potential gain media for femtosecond lasers, but have not shown the advantages of high efficiency or high output power until now.^{8–10)} Because of its poor thermal conductivity and small emission cross section, Nd:glass is not good for application to high average output power level devices.^{9,13)}

In recent years, Nd-doped fluorides have drawn some attention.^{14–16)} Among them, Nd:SrF₂ has been found to be a potentially useful laser-pumped storage medium.¹⁷⁾ Firstly, SrF₂ crystal has a simple cubic structure and good thermal property similar to most fluorides.^{18,19)} Secondly, similarly to most Nd-doped gain materials, Nd:SrF₂ possesses relatively high absorption and emission cross sections.^{14,17)} Thirdly, the broad emission bandwidth of Nd:SrF₂ makes it suitable for generating femtosecond lasers.^{14,17)} Lastly, compared with Nd:CaF₂, Nd:SrF₂ has a small number of clusters that favor energy transfer (cross-relaxation, up-conversion) between the ions.^{15,20)} The clustering consequently lowers the Nd³⁺ fluorescence quantum efficiency. Codoping with nonactive ions such as Y³⁺ or La³⁺ as buffers in fluorides is a method used to restrain clustering to improve the quantum efficiency of Nd³⁺.^{17,21)} This method has been demonstrated to be useful in Y³⁺ and Lu³⁺ codoped Nd:CaF₂ lasers.¹⁵⁾ More recently, the pulsed pumping and continuous-wave (CW) operations of Nd³⁺ and Y³⁺-codoped SrF₂ have been investigated and have shown good performance.²²⁾ In other words, Nd,Y:SrF₂ crystal seems to be one of the best candidates for femtosecond lasers with high output power, high efficiency, and low pumping cost.

In this letter, the femtosecond mode-locked laser based on the Nd- and Y-codoped SrF₂ gain crystal is reported. By appropriately designing the cavity, with a pulse duration of 332 fs, the mode-locking output power of up to 395 mW was obtained at the pump power of 1 W, corresponding to the optical-to-optical efficiency of 39.5% and the slope efficiency of 69%. In terms of the optical-to-optical efficiency in the

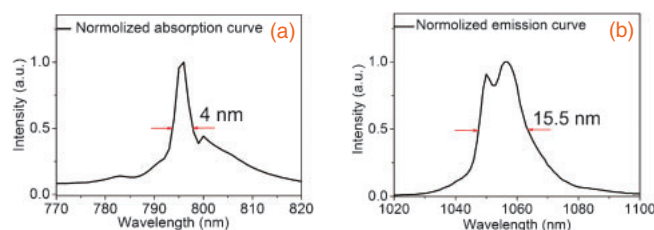


Fig. 1. (a) Absorption spectrum with peak centered at 796 nm. (b) Emission spectrum with peak centered at 1057 nm.

mode-locked operation, the value has exceeded that ever reported for femtosecond Nd crystal lasers, and it approaches the optical efficiencies of 33% (176 fs), 35.7% (418 fs), and 43.8% (738 fs)^{23–25)} obtained from classic Yb femtosecond mode-locked lasers (Yb:KGW, Yb:YAG, and Yb:Lu₂O₃, respectively). Note that the femtosecond Nd,Y:SrF₂ mode-locked laser has the advantage over an Yb system in easily emitting light with low power pumping.

The gain medium used in our experiments was a Brewster-cut 0.4 at. % Nd, 10 at. % Y:SrF₂ crystal with a size of 3 × 3.8 × 6 mm³ and a gain length of 6 mm. The highest absorption peak is at 796 nm with an FWHM of 4 nm and the highest emission peak is around 1057 nm with an FWHM of 15.5 nm, which theoretically support the generation of 76 fs pulses, as shown in Fig. 1. The thermal conductivity of this Nd,Y:SrF₂ crystal is 3.5 W·m⁻¹·K⁻¹, close to the value of 3.6 W·m⁻¹·K⁻¹ for Yb-doped SrF₂.¹⁸⁾ During the experiment, the gain medium was wrapped with indium foil and mounted tightly on a water-cooled copper heat sink block, cooled by the flowing water. A homemade 796 nm CW Ti:sapphire laser was chosen as a pumping source. The pumping laser was polarized and just matched the Brewster-cut crystal to decrease the loss of reflection at the interface of the crystal.

The experimental setup is shown in Fig. 2. L was a focusing lens with a focal length of 125 mm, by which the pumping laser can be focused into a size of 54 × 62 μm² (1/e², radius), measured using the CCD (WinCamD) at 0.76 W pump power. M1 was a pumping mirror with a 200 mm radius of curvature (ROC), antireflection (AR)-coated at 796 nm, and with a high reflectivity in the wavelength region of 1 to 1.1 μm. M2 was a high-reflectivity mirror (HR) with a high reflectivity in the region of 0.9–1.1 μm and an ROC of 150 mm. M3 was a Gires–Tournois interferometer

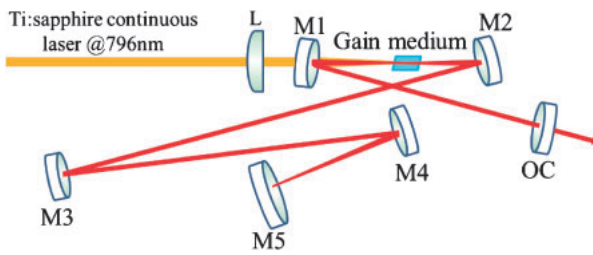


Fig. 2. Schematic of the Nd:Y:SrF₂ femtosecond laser setup.

mirror (GTI; Layertec). The use of the GTI resulted in lower losses than that of prisms and provided a group delay dispersion (GDD) of about -1200 fs^2 around 1057 nm. M4 was an HR reflection-coated in the region of 1.02–1.1 μm with an ROC of 300 mm, used to focus the laser beam onto a semiconductor saturable absorption mirror (SESAM). M5 was an SESAM, used to start the mode-locking or a plane HR in the case of CW operation. The transmittance of the output coupler (OC) was 1.6%. From ABCD matrix calculation, the laser beam had a radius of about 70 μm inside the crystal and that of 57 μm on the SESAM. During the experiment, we adjusted the focal spot of the pumping laser slightly deviating from the center of the crystal to match well the pumping laser with the gain laser in the crystal.

A plane HR was firstly adopted as M5 to align the cavity for the CW operation. By changing the temperature of the flowing water, the output power was experimentally found to be the highest at the temperature at around 12 °C. Thus, 12 °C was chosen as the cooling temperature of the copper heat sink block for all experiments. An output power of 346 mW was obtained at the pump power of 1 W. The corresponding optical-to-optical efficiency was 34.6%, showing the good laser performance of Nd:Y:SrF₂. Then, under the same pump power, M5 was changed to an SESAM (BATOP) to start the passive mode-locking. The SESAM was designed for 0.4% modulation depth at 1064 nm, with a saturation fluence of 90 $\mu\text{J}/\text{cm}^2$ and a relaxation time of 500 fs. By slightly adjusting the position and angle of the SESAM and M2 to optimize the cavity, a mode-locking operation with the highest optical-to-optical efficiency was realized. The average output power was 395 mW at the pump power of 1 W, corresponding to an optical-to-optical conversion efficiency of 39.5%. In our experiments, the SESAM has low loss similar to that of the HR we used, proved by our laser threshold measurements. The threshold for the SESAM was 210 mW, while it was 243 mW for the HR in the cavities with the same parameters. When the laser operated in the mode-locked regime, there were slight reductions in resonator losses associated with the saturation of the SESAM loss as pointed out by Klopp et al.²⁶⁾ Thus, the output power in mode-locking (ML) exceeded that in the CW operation.

The pulse duration of 332 fs (FWHM) was measured using a commercial autocorrelator (Femto Chrome Research FR-103MN), assuming a sech^2 fit, as shown in Fig. 3(a). Figure 3(b) shows a pulse centered at 1057 nm with a spectral width of 4.3 nm (FWHM), recorded using a spectrum analyzer (Yokogawa AQ6370C). We also measured the radio frequency (RF) spectrum with an RF spectrum analyzer (Agilent E4407B) and a fast detector of 1 GHz. Figure 3(c) shows the fundamental beat note at 89.8 MHz, with a high

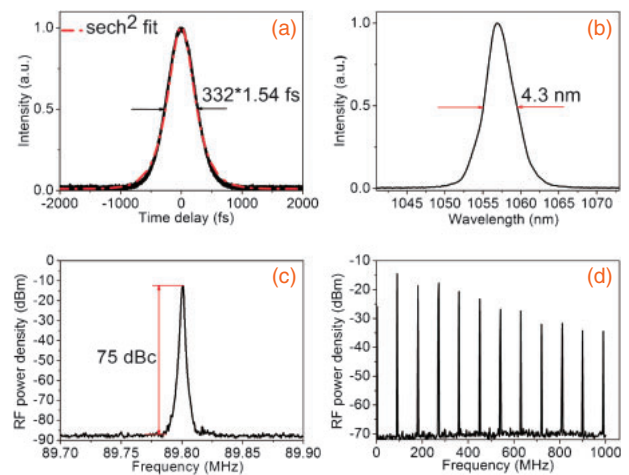


Fig. 3. (a) Intensity autocorrelation trace. (b) Laser spectrum. (c) RF spectrum at the fundamental beat note. (d) RF spectrum of 1 GHz wide-span range.

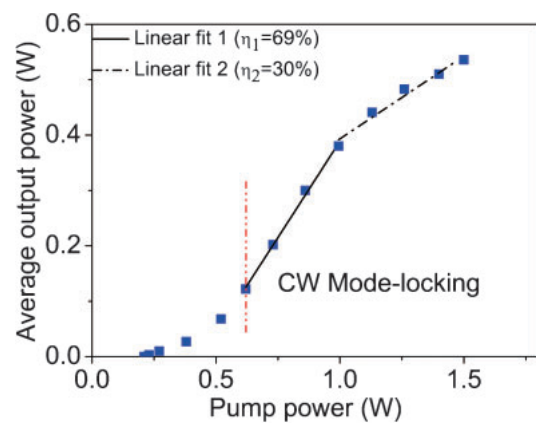


Fig. 4. Dependence of the output power vs pump power in mode-locked operation.

extinction down to 75 dBc, recorded with a resolution bandwidth of 1 kHz and a span of 89.7 to 89.9 MHz. The fundamental beat note showed that the repetition rate of the mode-locking was 89.8 MHz, corresponding to a 1.67-m-long cavity. Figure 3(d) shows the high harmonics of the fundamental beat note measured with a resolution bandwidth of 100 kHz and a span of zero to 1 GHz. There are no obvious noise waves in Figs. 3(c) and 3(d), which illustrates that the mode-locking is under a continuous stable condition.

The dependence of output power versus pump power in ML was observed, as shown in Fig. 4. On the premise of keeping the mode-locking condition, the mode-locked output power could be optimized to the desired value as high as possible by slightly adjusting the mirrors and SESAM while reducing the pump power. When the output power decreased to 122 mW, the mode-locking turned into Q-switching. When the pump power was close to 1 W, the highest optical-to-optical efficiency of 39.5% in mode-locking was obtained. The highest output power was 536 mW at the pump power of 1.5 W. As shown in Fig. 4, the slope efficiency obviously decreased to 30% (linear fit 2) when the pump power was larger than 1 W, which is related to the deprivation of the spot shape and beam quality of the Ti:sapphire laser in a high-

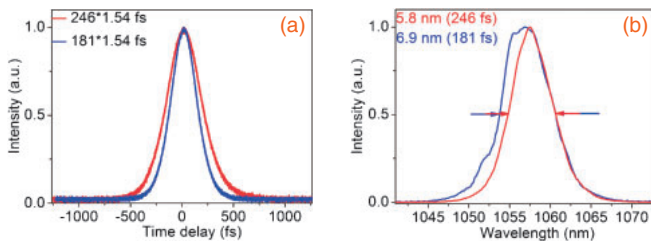


Fig. 5. (a) Intensity autocorrelation traces of the 246 and 181 fs pulses. (b) Corresponding laser spectra.

output-power operation. The focal spot of the pumping laser was found to be $134 \times 102 \mu\text{m}^2$ measured using the CCD at 1.35 W pump power. This leads to the mismatch of the pumping and gain lasers in the crystal, which cannot be completely overcome by adjusting the position and angle of mirrors. The slope efficiency of ML can approach 69% when the pump power is in the region of the linear fit 1, as shown in Fig. 4. Thus, it is reasonable to believe that the output power can be further improved if a better pumping source is used.

By adjusting the angle of the GTI and the pump power further, shorter pulses of 246 and 181 fs (FWHM) were achieved, assuming sech^2 pulse intensity profiles, as shown in Fig. 5(a). The corresponding spectral widths were 5.8 nm (246 fs) and 6.9 nm (181 fs), as shown in Fig. 5(b). The output power was 280 mW at the pump power of 1 W (246 fs), the same as that at 1.5 W (181 fs), corresponding to the conversion efficiencies of 28 and 18.67%, respectively. The time-bandwidth products of three mode-lockings of 332, 246, and 181 fs were 0.383, 0.383, and 0.335, respectively, and gradually approached the Fourier limit of 0.315. These results further experimentally demonstrate that the Nd,Y:SrF₂ is a good candidate for the generation of short pulse in mode-locking. As we know, large self-phase modulation and precise dispersion compensation play important roles in obtaining shorter pulses for femtosecond mode-locked lasers. Therefore, if we choose better dispersion compensation elements, such as prisms or a pair of chirped mirrors, to precisely control the net anomalous GDD and enhance self-phase modulation by optimizing the cavity and power to obtain high power density in the crystal, a sub-100 fs pulse could be obtained from the Nd,Y:SrF₂.

In conclusion, we have demonstrated an efficient femtosecond mode-locked Nd,Y:SrF₂ laser with 395 mW output power under 1 W pump power, corresponding to the highest optical-to-optical efficiency of 39.5% and slope efficiency of 69% while neglecting the decline in laser pumping. The pulse centered at 1057 nm with the duration of 332 fs and the spectral width of 4.3 nm in this mode-locking has been observed. By optimizing the cavity and power further, the shorter pulse of 181 fs has been achieved with the output power of 280 mW at the pump power of 1.5 W. Our results indicate that the Nd,Y:SrF₂ gain crystal is a promising laser medium for generating high-power femtosecond lasers with

low energy cost. With a 4-nm-bandwidth absorption spectrum for Nd,Y:SrF₂ and the appearance of a 796 nm laser diode (LD),²²⁾ it will enable us to realize LD-pumped femtosecond Nd,Y:SrF₂ lasers in the future, and further apply them in optical comb and amplification systems.

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