Abstract: A high-quality Nd<sub>0.01</sub>:Gd<sub>0.89</sub>La<sub>0.1</sub>NbO<sub>4</sub> (Nd:GLNO) crystal is grown by the Czochralski method, demonstrating wide absorption and fluorescence spectra and advantage for producing ultrafast laser pulses. In this paper, the tunable and passively mode-locking Nd:GLNO lasers are characterized for the first time. The tuning coverage is 34.87 nm ranging from 1058.05 to 1092.92 nm with a maximum output power of 4.6 W at 1065.29 nm. A stable continuous-wave (CW) passively mode-locking Nd:GLNO laser is achieved at 1065.26 nm, delivering a pulse width of 9.1 ps and a maximum CW mode-locking output power of 0.27 W.

Keywords: Nd:GLNO crystal; tunable laser; mode-locking

1. Introduction

Ultrafast lasers have been applied in various fields, such as high-precision micro machining, aerospace, and medical diagnostics [1,2]. Benefiting from their low quantum defects, wide gain bandwidth, and simple three-level electronic structure, Yb<sup>3+</sup>-doped laser mediums attract widespread attention in the 1 µm band [3–5]. However, the overlap of absorption and emission bands can bring re-absorption loss, resulting in high laser threshold. Compared with Yb<sup>3+</sup>-doped gain mediums, Nd<sup>3+</sup>-doped crystals have no re-absorption loss and are used in low-threshold and high-efficiency ultrafast laser. As is known, the typical gain bandwidth of the Nd<sup>3+</sup>-doped laser materials is narrow, e.g., the gain bandwidth of the Nd:YVO<sub>4</sub> and Nd:YAG crystals were measured to be only 0.96 and 0.89 nm, respectively [6,7]. For this reason, considerable efforts have been made to explore novel Nd<sup>3+</sup>-doped laser materials with a broad gain bandwidth. The pulse duration of 19.2 ps at 1064 nm was achieved in a passively mode-locked Nd:YVO<sub>4</sub> laser in 2008 [8]. Mohammad et al. [9]. reported pulse duration of 16 ps generation in a Nd:GdVO<sub>4</sub> crystal in 2017. He et al. [10]. obtained 3.8 ps pulse duration at a repetition rate of 112 MHz in a Nd:GdYVO<sub>4</sub> crystal. Previously, theoretical and experimental results have demonstrated that Nd<sup>3+</sup>-doped disordered crystals possess broad emission spectra and are suitable for generating ultrashort lasers [11–13].

In the last decade, researchers have invested tremendous enthusiasm into extending Nd<sup>3+</sup>-doped disordered crystals family and exploring their excellent properties. In 2017, a novel disordered crystal Nd<sub>0.01</sub>:Gd<sub>0.89</sub>La<sub>0.1</sub>NbO<sub>4</sub> (Nd:GLNO) was successfully grown by Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences [14].
Owing to La\(^{3+}\) having a relatively large ionic radius in the lanthanide system, the La\(^{3+}\)-doped disordered crystals exhibit a wider fluorescence bandwidth [15]. Moreover, the difference in ionic radii between La\(^{3+}\) and Gd\(^{3+}\) ions is small, denoting the Nd:GLNO crystal possesses excellent lattice matching and thermal property [16,17]. The fluorescence lifetime and the radiative lifetime of Nd:GLNO crystal was obtained to be 176.1 µs and 184.5 µs, respectively. The luminescent quantum efficiency of the \(\text{4F}_{3/2}\) level was estimated to be 95.4% [18]. Ma et al. presented the CW and passively Q-switched Nd:GLNO lasers with Cr\(^{4+}:\text{YAG} crystal and PdSe\(_2\) as saturable absorbers (SAs), respectively, in 2018 and 2020 [15,19]. Unfortunately, the tunable and CW mode-locking Nd:GLNO crystal lasers have not been studied to date.

In this paper, the absorption and fluorescence spectra of the Nd:GLNO crystal were systematically investigated demonstrating a wide absorption and emission band. A tunable operation Nd:GLNO crystal laser was realized with a tuning range of 34.87 nm from 1058.05 to 1092.92 nm. By employing a semiconductor saturable absorber mirror (SESAM) as SA, a stable CW mode-locking Nd:GLNO crystal laser was achieved, generating the shortest pulse duration of 9.1 ps and the maximum mode-locking output power of 0.27 W.

2. Experimental Setup

Figure 1 demonstrates schematic setups of the Nd:GLNO lasers. The 808 nm laser diode was chosen as a pump source with a core diameter of 400 µm and a numerical aperture (NA) of 0.22. The size of the c-cut Nd:GLNO crystal was \(2 \times 2 \times 5\) mm\(^3\). To effectively reduce the influence of thermal effects, the laser crystal was covered with indium and embedded into a copper block. The cooling temperature of the copper block was controlled at 15.5 °C. The total laser cavity length of the mode-locking and tunable lasers was 1.94 m and 0.33 m, respectively. Mirrors M\(_1\), M\(_2\), M\(_4\), M\(_5\) and M\(_6\) were all processed with anti-reflection (AR) coating around 808 nm and high-reflection coating (HR, \(R > 99.9\%\)) at 1030–1100 nm. The curvature radii were \(R = \infty\), \(R = 200\), \(R = 150\), respectively. The output mirror M\(_3\) was partially transmittances (T) coated at 1030–1100 nm (T = 1, 10, 15%, 25% are available). A quartz birefringent filter (BF) was employed in tunable laser cavity to achieve laser tuning operation. The parameters of the SESAM are as follows: saturable fluence is 90 µJ/cm\(^2\), absorptance is 1.5%, a modulation depth is 0.8%, damage threshold is 30 mJ/cm\(^2\), and a relaxation time is 1 ps. A laser power meter (Fieldmax-II, PM10) was used for measuring laser power. The laser output spectra and pulse width of mode-locked Nd:GLNO laser were measured by a spectrometer (Avantes, AcaSpec-3468-NIR256-2.2) and a commercial autocorrelator (APE Pulse Check, 150), respectively. The typical pulse profile and pulse train were recorded by a digital oscilloscope (R&S, RTO 2012) together with a fast InGaAs photon detector (New Focus, 1611).

![Figure 1. Schematic setups of the Nd:GLNO laser, (a) tunable operation; (b) CW mode-locking operation.](image)

3. Results and Discussion

Figure 2 presents the absorption and fluorescence spectra of the c-cut Nd:GLNO crystal at room temperature. As shown in Figure 2a, the absorption peak is at 808 nm and FWHM is 13 nm. Based on the equation \(\sigma = \alpha(\lambda)/Nc\), where \(\alpha\) is the absorption coefficient (8.97 cm\(^{-1}\)) and \(Nc\) is the concentration of Nd\(^{3+}\), the maximum absorption cross-section of the Nd:GLNO crystal was calculated to be \(10.49 \times 10^{-20}\) cm\(^2\). Moreover, the stimulated
emission cross-section ($\sigma_{em}$) can be estimated from the fluorescence spectra using the Füchtbauer–Ladenburg equation:

$$ \sigma_{em}(\lambda) = \frac{\hbar^2 I(\lambda)}{8\pi^2 c \tau_m n_c} \int \frac{n(\lambda) d\lambda}{\lambda} $$

where $\tau_m$, $c$, $n$, $I(\lambda)$ are the fluorescence lifetime, velocity of light, reflective index and fluorescence intensity, respectively. The calculated stimulated emission cross-section of $18 \times 10^{-20} \text{cm}^2$ was relatively large, which was suitable for generating ultrafast laser pulse.

A V-type laser cavity was designed to investigate the CW laser output properties of the Nd:GLNO crystals. Figure 3 displays the relationship between output power and absorbed pump power at different transmittances of output couplers. The maximum CW output power of 4.60 W was achieved with the output mirror of $T = 15\%$, corresponding to an optical-to-optical efficiency of 37.90\% and a slope efficiency of 49.67\%. Furthermore, the laser output wavelength could be flexibly tuned by carefully varying the angle of the BF. Table 1 records the tuning wavelength and the corresponding output power with the output couplers of $T= 1\%$, 10\% and 15\%, respectively. As the transmittance increased, the longitudinal mode oscillation in the cavity was suppressed. Therefore, the tuning range was further reduced. The total tuning coverage of the Nd:GLNO crystal laser was 34.87 nm ranging from 1058.05 to 1092.92 nm. Figure 4 demonstrates the typical single wavelength and multi-wavelength spectra of the Nd:GLNO crystal tunable laser.

![Absorption and fluorescence spectra of the Nd:GLNO crystal. (a) Absorption spectra; (b) Fluorescence spectra.](image1)

![Output power versus absorbed pump power.](image2)
Figure 4. Typical output spectra of the tunable Nd:GLNO laser with T = 1%.

Table 1. Output parameters of the tunable Nd:GLNO crystal laser.

| T (%) | Wavelength (nm) | Output Power (W) |
|-------|-----------------|------------------|
|       |                 |                  |
| 1     | 1058.05         | 0.83             |
|       | 1065.29         | 1.11             |
|       | 1065.61         | 0.86             |
|       | 1091.98         | 0.58             |
|       | 1092.29         | 1.00             |
|       | 1092.61         | 0.98             |
|       | 1092.92         | 0.79             |
| 10    | 1065.29         | 4.13             |
|       | 1092.29         | 3.03             |
|       | 1092.61         | 2.83             |
| 15    | 1065.29         | 4.60             |
|       | 1092.29         | 0.43             |
|       | 1092.61         | 0.31             |

To realize the CWML Nd:GLNO laser operation, a Z-type laser cavity was employed as shown in Figure 1b. Ultrafast laser pulse output was achieved using a SESAM. To reduce the intracavity loss and make the SESAM easily saturated, the CWML laser output characteristics were obtained experimentally at the output mirror of $T_{oc} = 1\%$. As shown in Figure 5, the minimum absorbed pump power to suppress Q-switched mode-locking laser was 3.05 W. The maximum CWML laser output power 0.27 W was achieved. The CWML pulse train was measured using a detector and 1 GHz bandwidth oscilloscope. Figure 6 presents the stable mode-locking pulses recorded at nanosecond and microsecond time scales, respectively. The pulse repetition rate (PRR) is 51.6 MHz corresponding to the cavity length of 1.94 m. Figure 7 demonstrates the signal-to-noise ratio of the first beat. The radio frequency spectrum was clean and stable, indicating excellent stability of the mode-locking ultrafast laser. The signal-to-noise ratio was up to 72.3 dB at the fundamental frequency of 51.6 MHz. The FWHM bandwidth of the autocorrelation trace was about 14.0 ps, corresponding to a pulse duration of 9.1 ps by a sech²-shape pulse fitting. The mode-locking pulse spectrum was shown in the inset of Figure 8. The central wavelength of the measured pulse was located at 1065.26 nm with a FWHM of 0.9 nm.
Figure 5. The CW mode-locking output power versus the absorbed pump power.

Figure 6. Pulse train of the CW mode-locking Nd:GLNO crystal laser.

Figure 7. Recorded RF trace.
4. Conclusions

In conclusion, the Nd:GLNO crystal was grown by the Czochralski method and the spectral characteristics at room temperature were discussed. The maximum CW output power of 4.60 W was obtained with the output mirror of T = 15%, corresponding to an optical-to-optical efficiency of 37.90% and a slope efficiency of 49.67%. The tuning coverage of the tunable Nd:GLNO laser was 34.87 nm at T = 1% ranging from 1058.05 to 1092.92 nm. To the best of our knowledge, a picosecond CWML Nd:GLNO laser at 1065.26 nm was experimentally demonstrated using a SESAM as saturable absorber for the first time. The maximum CWML laser output power of 0.27 W was achieved. The Nd:GLNO crystal ultrafast laser produced 9.1 ps mode-locked pulses with pulse repetition rate of 51.6 MHz and a signal-to-noise ratio of 72.3 dB. The results indicated that the Nd:GLNO crystal is a promising Nd\textsuperscript{3+}-doped gain medium for generating ultrafast laser pulses.

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