

# Passively mode-locked femtosecond laser with an Nd-doped $\text{La}_3\text{Ga}_5\text{SiO}_{14}$ disordered crystal

Jiaying Liu,<sup>1</sup> Zhaohua Wang,<sup>1,\*</sup> Kunna He,<sup>2</sup> Long Wei,<sup>3</sup> Zhiguo Zhang,<sup>1</sup> Zhiyi Wei,<sup>1,5</sup> Haohai Yu,<sup>4</sup> Huaijin Zhang,<sup>4</sup> and Jiyang Wang<sup>4</sup>

<sup>1</sup>Beijing National Laboratory for Condensed Matter Physics and the Institute of Physics, Chinese Academy of Science, Beijing, 100190, China

<sup>2</sup>Department of Applied Physics, China Agricultural University, Beijing, 100083, China

<sup>3</sup>School of Physics and Optoelectronic Engineering, Xidian University, Xi'an, 710071, China

<sup>4</sup>State Key Laboratory of Crystal Material and Institute of Crystal Material, Shandong University, Jinan, 250100, China

<sup>5</sup>zywei@iphy.ac.cn

\*zhwang@iphy.ac.cn

**Abstract:** We experimentally prove the broad emission band of Nd:LGS disordered crystal and demonstrate a diode-pumped passively mode-locked femtosecond Nd-doped  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  (Nd:LGS) laser for the first time. With a birefringent filter inserted into the cavity, the tunable continuous wave (CW) laser of over 60 nm from 1045.2 nm to 1105.3 nm is achieved, which is the widest tuning range with Nd-doped crystals to our knowledge. Further in mode-locked operation, femtosecond pulses with pulse duration of 381 fs, average output power of 75 mW and repetition rate of 134.4 MHz are obtained at the central wavelength of 1066 nm. It is suitable to be a compact seed for femtosecond laser amplifiers.

©2014 Optical Society of America

**OCIS codes:** (140.3480) Lasers, diode-pumped; (140.3530) Lasers, neodymium; (140.4050) Mode-locked lasers.

---

## References and links

1. D. Strickland and G. Mourou, "Compression of amplified chirped optical pulses," *Opt. Commun.* **55**(6), 447–449 (1985).
2. E. W. Gaul, M. Martinez, J. Blakeney, A. Jochmann, M. Ringuette, D. Hammond, T. Borger, R. Escamilla, S. Douglas, W. Henderson, G. Dyer, A. Erlandson, R. Cross, J. Caird, C. Ebbers, and T. Ditmire, "Demonstration of a 1.1 petawatt laser based on a hybrid optical parametric chirped pulse amplification/mixed Nd:glass amplifier," *Appl. Opt.* **49**(9), 1676–1681 (2010).
3. J. H. Sung, S. K. Lee, T. J. Yu, T. M. Jeong, and J. Lee, "0.1 Hz 1.0 PW Ti:sapphire laser," *Opt. Lett.* **35**(18), 3021–3023 (2010).
4. Z. H. Wang, C. Liu, Z. W. Shen, Q. Zhang, H. Teng, and Z. Y. Wei, "High-contrast 1.16 PW Ti:sapphire laser system combined with a doubled chirped-pulse amplification scheme and a femtosecond optical-parametric amplifier," *Opt. Lett.* **36**(16), 3194–3196 (2011).
5. D. C. Brown, J. M. Singley, K. Kowalewski, J. Guelzow, and V. Vitali, "High sustained average power cw and ultrafast Yb:YAG near-diffraction-limited cryogenic solid-state laser," *Opt. Express* **18**(24), 24770–24792 (2010).
6. D. W. E. Noom, S. Witte, J. Morgenweg, R. K. Altmann, and K. S. E. Eikema, "High-energy, high-repetition-rate picosecond pulses from a quasi-CW diode-pumped Nd:YAG system," *Opt. Lett.* **38**(16), 3021–3023 (2013).
7. W. F. Krupke, M. D. Shinn, J. E. Marion, J. A. Caird, and S. E. Stokowski, "Spectroscopic, optical, and thermomechanical properties of neodymium- and chromium-doped gadolinium scandium gallium garnet," *J. Opt. Soc. Am. B* **3**(1), 102–114 (1986).
8. Y. X. Fan, J. L. He, Y. G. Wang, S. Liu, H. T. Wang, and X. Y. Ma, "2-ps passively mode-locked Nd:YVO<sub>4</sub> laser using an output-coupling-type semiconductor saturable absorber mirror," *Appl. Phys. Lett.* **86**(10), 101103 (2005).
9. G. Q. Xie, D. Y. Tang, W. D. Tan, H. Luo, H. J. Zhang, H. H. Yu, and J. Y. Wang, "Subpicosecond pulse generation from a Nd:CLNGG disordered crystal laser," *Opt. Lett.* **34**(1), 103–105 (2009).
10. G. Q. Xie, L. J. Qian, P. Yuan, D. Y. Tang, W. D. Tan, H. H. Yu, H. J. Zhang, and J. Y. Wang, "Generation of 534 fs pulses from a passively mode-locked Nd:CLNGG-CNGG disordered crystal hybrid laser," *Laser Phys. Lett.* **7**(7), 483–486 (2010).
11. A. Agnesi, F. Pirzio, L. Tartara, E. Ugolotti, H. Zhang, J. Wang, H. Yu, and V. Petrov, "378 fs pulse generation with Nd<sup>3+</sup>:SrLaGa<sub>3</sub>O<sub>7</sub> (Nd:SLG) disordered crystal," *Laser Phys. Lett.* **10**(10), 105815 (2013).

12. Z. P. Qin, G. Q. Xie, J. Ma, W. Y. Ge, P. Yuan, L. J. Qian, L. B. Su, D. P. Jiang, F. K. Ma, Q. Zhang, Y. X. Cao, and J. Xu, "Generation of 103 fs mode-locked pulses by a gain linewidth-variable Nd:Y:CaF<sub>2</sub> disordered crystal," *Opt. Lett.* **39**(7), 1737–1739 (2014).
13. A. Agnesi, F. Pirzio, L. Tartara, E. Ugolotti, H. Zhang, J. Wang, H. Yu, and V. Petrov, "Tunable femtosecond laser based on the Nd<sup>3+</sup>:BaLaGa<sub>3</sub>O<sub>7</sub> disordered crystal," *Laser Phys. Lett.* **11**(3), 035802 (2014).
14. J. Hou, L. H. Zheng, J. L. He, J. Xu, B. T. Zhang, Z. W. Wang, F. Lou, R. H. Wang, and X. M. Liu, "A tri-wavelength synchronous mode-locked Nd:SYSO laser with a semiconductor saturable absorber mirror," *Laser Phys. Lett.* **11**(3), 035803 (2014).
15. A. A. Kaminskii, B. V. Mill, G. G. Khodzhabayyan, A. F. Konstantinova, A. I. Okorochkov, and I. M. Silvestrova, "Investigation of trigonal (La<sub>1-x</sub>Nd<sub>x</sub>)<sub>3</sub>Ga<sub>5</sub>SiO<sub>14</sub> crystals," *Phys. Status Solidi A* **80**(1), 387–398 (1983).
16. A. A. Kaminskii, I. M. Silvestrova, S. E. Sarkisov, and G. A. Denisenko, "Investigation of trigonal (La<sub>1-x</sub>Nd<sub>x</sub>)<sub>3</sub>Ga<sub>5</sub>SiO<sub>14</sub> crystals," *Phys. Status Solidi A* **80**(2), 607–620 (1983).
17. J. Stade, L. Bohaty, M. Hengst, and R. B. Heimann, "Electro-optic, piezoelectric and dielectric properties of Langanite (La<sub>3</sub>Ga<sub>5</sub>SiO<sub>14</sub>), Langanite (La<sub>3</sub>Ga<sub>5</sub>Nb<sub>0.5</sub>O<sub>14</sub>) and Langataite (La<sub>3</sub>Ga<sub>5.5</sub>Ta<sub>0.5</sub>O<sub>14</sub>)," *Cryst. Res. Technol.* **37**(10), 1113–1120 (2002).
18. Y. G. Yu, J. Y. Wang, H. J. Zhang, Z. P. Wang, H. H. Yu, and M. H. Jiang, "Continuous wave and Q-switched laser output of laser-diode-end-pumped disordered Nd:LGS laser," *Opt. Lett.* **34**(4), 467–469 (2009).
19. Y. Y. Ren, J. R. Vázquez de Aldana, F. Chen, and H. J. Zhang, "Channel waveguide lasers in Nd:LGS crystals," *Opt. Express* **21**(5), 6503–6508 (2013).
20. Q. Wang, Z. Y. Wei, J. X. Liu, Z. H. Wang, Z. G. Zhang, H. J. Zhang and J. Y. Wang, "Mode-locked Nd:LGS laser with femtosecond pulse duration," in *CLEOPR* (2013), ThA3-7.
21. R. E. Slocum, L. D. Scheerer, P. Tin, and R. Marquedant, "Nd:LNA laser optical pumping of 4He: Application to space magnetometers," *J. Appl. Phys.* **64**(12), 6615–6617 (1988).
22. H. R. Verdun and L. M. Thomas, "Nd:CaYAlO<sub>4</sub>—a new crystal for solidstate lasers emitting at 1.08 μm," *Appl. Phys. Lett.* **56**(7), 608–610 (1990).
23. Z. H. Cong, D. Y. Tang, W. De Tan, J. Zhang, C. W. Xu, D. Luo, X. D. Xu, D. Z. Li, J. Xu, X. Y. Zhang, and Q. P. Wang, "Dual-wavelength passively mode-locked Nd:LuYSiO<sub>5</sub> laser with SESAM," *Opt. Express* **19**(5), 3984–3989 (2011).
24. K. Miyamoto, H. Minamide, M. Fujiwara, H. Hashimoto, and H. Ito, "Widely tunable terahertz-wave generation using an N-benzyl-2-methyl-4-nitroaniline crystal," *Opt. Lett.* **33**(3), 252–254 (2008).

## 1. Introduction

Since large emission cross-sections and long upper level lifetimes, neodymium (Nd)-doped laser media have been widely used in laser systems [1–6]. So far, the Nd-doped glass has become one of the most commercial materials for the femtosecond laser and chirped pulse amplification (CPA) [2–4]. Nd-doped YVO<sub>4</sub> and YAG have been prominent gain materials in low, moderate and high power laser oscillators and amplifiers [5,6]. However, the Nd:glass has a poor thermal conductivity so that it is hard for high power high repetition lasers and Nd-doped commonly used crystals generally have narrow gain linewidth, which limit for generating femtosecond pulses [7,8].

In recent years, the crystals with disordered structure have attracted wide attentions for their inhomogeneous broadening spectra induced by party replacement of the ions in their lattices. Nd-doped disordered crystals possess better thermal conductivities than Nd:glass and broader spectra than Nd-doped commonly used crystals, which are potential candidates to generate high repetition lasers and mode-locked femtosecond pulses. Up to now, only a few femtosecond operations have been reported with Nd-doped disordered crystals [9–14]. G. Q. Xie et al firstly obtained 900 fs and 534 fs pulses with Nd:CLNGG and Nd:CLNGG-CNGG hybrid crystal [9,10]. And more recently, Z. P. Qin et al obtained the shortest pulses of 103 fs with an Nd, Y-codoped CaF<sub>2</sub> crystal [12]. Table 1 shows femtosecond operations with some disordered crystals up to now. The La<sub>3</sub>Ga<sub>5</sub>SiO<sub>14</sub> (LGS) disordered crystal is a multifunctional material, which attracted a widespread attention because of properties such as piezoelectric, electro-optic and dielectric properties [15–17]. Doping with Nd<sup>3+</sup> ion, Nd:LGS becomes a laser medium [18,19]. Figure 1 presents the fluorescence and absorption spectra of Nd:LGS. Its broad absorption band at 809 nm is suitable for laser diode pumping. The emission bandwidth (FWHM) peaked at 1066 nm is broader than 10 nm, which could support femtosecond pulse generation. Additionally, the thermal conductivity of Nd:LGS is about 1.7 W/m/K, much higher than Nd:glass. Since the above characteristics, Nd:LGS is a potential gain media for compact femtosecond laser oscillators. In previous work, we have demonstrated mode-locked femtosecond laser pumped by a CW Ti:sapphire laser with

Nd:LGS [20]. However, the Ti:sapphire laser has disadvantages of higher cost and more complexity. So it is expected to use laser diodes as pump sources for compactness and applications.

**Table 1. Femtosecond Operations with Some Disordered Crystals**

Crystal	Pump laser	Emission wavelength	Pulse width	Output power
Nd:CLNGG [9]	Laser diode	1061 nm	900 fs	486 mW
Nd:CLNGG-CNGG [10]	Laser diode	1061.5 nm	534 fs	60 mW
Nd:SLG [11]	Ti:sapphire laser	1061 nm	378 fs	33 mW
Nd:BLG [12]	Ti:sapphire laser	1076 nm	290 fs	30 mW
Nd,Y:CaF <sub>2</sub> [13]	Laser diode	1064 nm	103 fs	89 mW
Nd:SYSO [14]	Laser diode	1075.5 nm, 1076.8 nm, 1078.2 nm	646 fs (best)	620 mW (tri-wavelength)

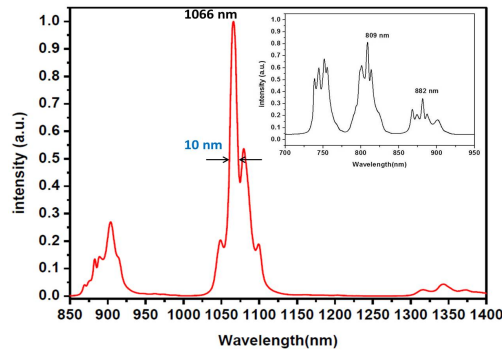


Fig. 1. Fluorescence and absorption (inset) spectra of Nd:LGS.

In this letter, we demonstrate a diode-pumped tunable CW laser of over 60 nm from 1045.2 nm to 1105.3 nm and 381 fs mode-locked laser at the central wavelength of 1066 nm at 134.4 MHz with a Nd:LGS disordered crystal. To the best of our knowledge, it is the widest tunable CW laser among Nd-doped crystals and the first demonstration of femtosecond operation with diode-pumped Nd:LGS laser.

## 2. Experiments and results

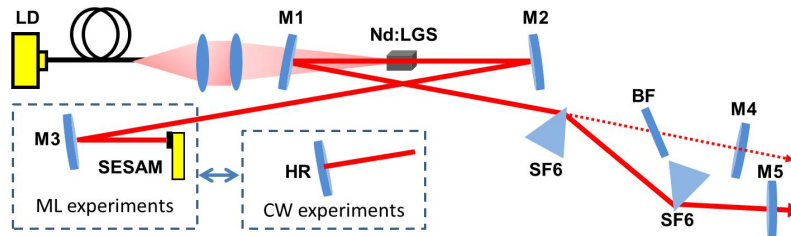


Fig. 2. Experimental setup of a CW and mode-locked (ML) Nd:LGS laser.

To experimentally investigate the laser properties of Nd:LGS, we implemented an tunable CW laser experiment. The setup is designed as Fig. 2. The laser medium we used was a 3x3x8 mm<sup>3</sup>, Z-cut, and 1 at. % doped Nd:LGS disordered crystal, which was water-cooled at 12 °C and antireflection coated at 808 nm and around 1 μm. Then it was end-pumped by a fiber-coupled diode laser with 100 μm core diameter (LIMO, Germany). The pump beam into the crystal was focused by an imaging system with a magnification of 1. An X-folded resonator was employed in our experiment. Both M1 and M2 were plane-concave pump mirrors with the curvature radiuses of 75 mm. A high-reflection mirror (HR) and an output coupler M4 were used as end mirrors. They were coated with high reflection and transmittance of 0.8% or

2.5% at the wavelength range from 1020 nm to 1100 nm, respectively. We adjusted the laser cavity to obtain the maximum CW output power of 726 mW with 0.8% output coupler and 953 mW with 2.5% output coupler at 1066 nm under the pump power of 5.8 W as shown in Fig. 3(a). And the threshold pump powers were as low as 71 mW and 161 mW. Then a birefringent filter was inserted into the cavity with 0.8% output coupler to tune the central wavelength of the CW laser by rotating it, which was placed with Brewster angle for minimizing the reflection loss. Consequently, 60.1 nm continuously tuning range from 1045.2 nm to 1105.3 nm was obtained as shown in Fig. 3(b). The broad emission band makes Nd:LGS a promise crystal to generate femtosecond pulses.

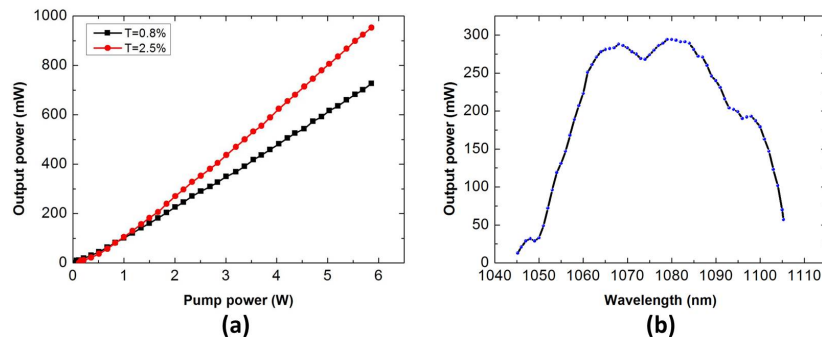


Fig. 3. Output power of the CW Nd:LGS laser. (a) curves with different output couplers; (b) CW output power versus wavelength with output coupler of 0.8% transmittance under the pump power of 5.8 W.

In order to achieve femtosecond mode-locked operation, the HR mirror was replaced by a curved highly reflected mirror M3 with the curvature radiuses of 100 mm and a semiconductor saturable absorber mirror (SESAM), which was employed as an end mirror with a modulation depth of 0.4%, a saturation fluence of  $90 \mu\text{J}/\text{cm}^2$  and a recovery time of less than 500 fs at  $1.06 \mu\text{m}$  (BATOP, Inc.). The dimension of laser spot onto the SESAM was calculated to be about  $70 \mu\text{m}$ . For dispersion compensating, a pair of SF6 prisms were inserted into another arm of the resonator with a tip-to-tip distance of 31.5 cm. The transmittance of the output coupler M5 was 0.8%. The cavity length was 1.12 m, corresponded to a repetition rate of about 134 MHz.

Under the pump power of 5.9 W, the mode-locked operation was achieved after optimizing of mirrors in the cavity and dispersion compensation. However, the mode-locking trains were not stable enough for dual-wavelength and high order transverse mode oscillating in the cavity. From the spectrum of CW laser in Fig. 4(a), we could find dual-wavelength laser centered at 1063 nm and 1066 nm. When we achieved mode-locking at the central wavelength of 1066 nm, the longitudinal mode centered at 1063 nm was also existed as shown in Fig. 4(b), which made significant effect on mode-locking stability and the quality of spectrum. To solve this problem, we inserted a slit between the SF6 prisms in the cavity. After carefully optimizing the position and size of the slit, the longitudinal mode centered at 1063 nm and high order transverse mode oscillation could be suppressed. Finally, stable mode-locked pulses centered at 1066 nm were successfully obtained and the spectrum of mode-locking laser is described in Fig. 4(c). But the average output power decreased from 120 mW to 75 mW for insertion loss of the slit. Figure 5 describes the stable mode-locked pulse trains on the oscilloscope and the output power in mode-locked operation. When the pump power increased to be 4.54 W, stable CW mode-locking was self-starting. The maximum output power was 75 mW under the pump power of 5.9 W.

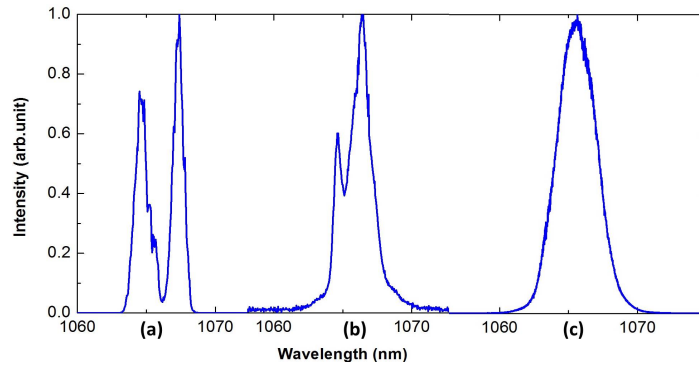


Fig. 4. The spectra of CW laser and mode-locked pulses. (a) is the spectrum in CW operation; (b) is the spectrum of mode-locked pulses without inserting a slit; (c) is the spectrum after optimizing with a slit.

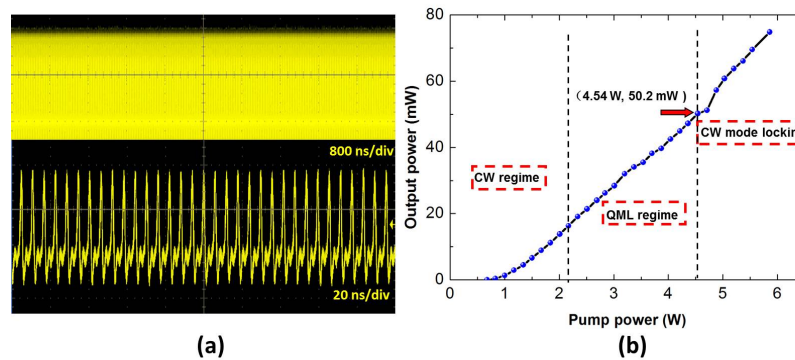


Fig. 5. The CW mode-locked pulse trains on the oscilloscope (a) and the output power curve of the mode-locked Nd:LGS laser with a 0.8% output coupler (b).

A commercial intensity autocorrelator (Femtochrome, FR-103MN) was used to measure the intensity autocorrelation trace of the mode-locked pulses shown in Fig. 6. The width (FWHM) of the autocorrelation trace was about 587 fs. If a  $\text{sech}^2$ -pulse shape was assumed, pulse duration was 381 fs. The inset in Fig. 6 shows the spectrum of the mode-locked pulses measured by an optical spectrum analyzer (AQ6315A, Ando Inc.), which indicates a spectrum FWHM of 3.28 nm at the central wavelength of 1066 nm. The time-bandwidth product was estimated to be 0.33, very close to the Fourier-transform limit of 0.315 for  $\text{sech}^2$  shaped pulses.

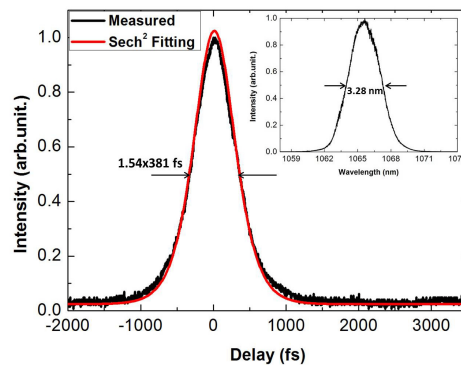


Fig. 6. Intensity autocorrelation trace of the mode-locked pulses measured and  $\text{sech}^2$  fitting; Inset: Spectrum of the mode-locked pulses with a spectrum FWHM of 3.28 nm at the central wavelength of 1066 nm.

Figure 7 shows the radio frequency (RF) spectrum of the mode-locked pulses with a RF spectrum analyzer (Agilent E4407B). On a small span with a resolution bandwidth (RBW) of 1 kHz, the repetition rate was displayed at 134.4 MHz with a high extinction down to over 60 dB. The inset shows the RF spectrum on a wide span of 1 GHz with a RBW of 100 kHz. From the RF spectrum, we could clearly know that the mode-locking was stable. Once the pump laser diode was turn off and restarted, the mode-locking was self-starting and kept stable with the pump power increased.

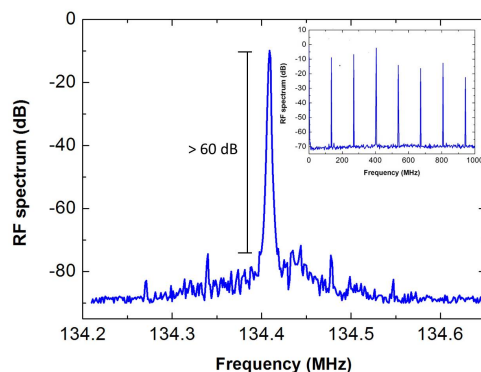


Fig. 7. RF spectrum of pulse train measured by an RF spectrum analyzer with a small span and a RBW of 1 kHz; Inset: RF spectrum on a spectral span of 1 GHz with a RBW of 100 kHz.

### 3. Discussion and conclusions

We experimentally proved the broad emission band of Nd:LGS disordered crystal and obtained the femtosecond pulses. In CW experiment, we got 60.1 nm continuously tuning range from 1045.2 nm to 1105.3 nm. From the CW tunable curve in Fig. 3(b), we can find that there are two main peaks around 1.06  $\mu\text{m}$  and 1.08  $\mu\text{m}$ . Both of them have FWHM of more than 30 nm, which means the possibility to generate femtosecond pulses. We believe femtosecond pulses around 1.08  $\mu\text{m}$  could be obtained by tuning the central wavelength, which provides a suitable laser pump source for helium [21,22]. And if effective methods are implemented, the dual-wavelength mode-locked laser will be also obtained, which has potential applications for the generation of coherent terahertz radiation [23,24].

In mode-locked operation, we successfully suppressed the longitudinal mode centered at 1063 nm and high order transverse mode oscillation by inserting a slit. Stable and self-starting CW mode-locking was achieved. The 381 fs laser pulse with output power of 75 mW is suitable to be a seed for femtosecond laser amplifiers.

In conclusion, we have obtained femtosecond pulses with a Nd:LGS disordered crystal, which possesses a broad emission band. The CW tuning range is more than 60 nm around 1.06  $\mu\text{m}$ . By using a SESAM and SF6 prisms, a stable 381 fs mode-locked laser at the central wavelength of 1066 nm with an average output power of 75 mW has been achieved after optimizing with a slit. Considering the excellent laser properties of Nd:LGS, we believe it will be a promise crystal for compact ultrafast oscillators.

### Acknowledgments

The authors appreciate helpful discussion with Xinkui He and Wenjun Liu. This work is partly supported by the National Major Equipment Development Project of the Ministry of Science and Technology of China (2012YQ120047) and the National Natural Science Foundation of China (11174361).