Appl. Phys. B (2013) 112:55–60
DOI 10.1007/s00340-013-5396-3

Applied Physics B
Lasers and Optics

Diode-pumped orthogonally polarized dual-wavelength Nd^{3+}:$LaBO_2MoO_4$ laser

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Received: 20 September 2012 / Accepted: 27 February 2013 / Published online: 26 March 2013
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Abstract Polarized spectroscopic properties related to 1.07 μm laser operation of a 1.8 at.% Nd^{3+}:$LaBO_2MoO_4$ crystal grown by the Czochralski method were investigated at room temperature. Using a 2.2-mm-thick, *Z*-cut Nd^{3+}:$LaBO_2MoO_4$ crystal as gain medium, orthogonally polarized dual-wavelength laser at 1,068 and 1,074 nm was first realized in a plano-concave resonator end-pumped by a quasi-continuous-wave 795 nm diode laser. A total output peak power of 1.2 W with slope efficiency of 26 % around 1.07 μm was obtained. The influences of resonator length and pump power on output laser wavelength were also investigated.

1 Introduction

Terahertz (THz) radiation has some unique properties unavailable in other region of electromagnetic spectrum [1], and can be widely used in various applications, such as remote THz image, chemical identification, nondestructive testing, molecular spectroscopy, radio astronomy, biomedical diagnostics, and military [2, 3]. Among many technical methods proposed for the generation of THz radiation [3, 4], the scheme based on difference-frequency generation (DFG) in a nonlinear optical crystal offers the advantages of compactness, simplicity, and high efficiency [5–8]. For this purpose, a simultaneous dual-wavelength laser source operating in orthogonally polarized modes is superior to using two separate lasers.

By a special design of laser system, such as the addition of other intra-cavity optical element [8, 9] or the using of diode-pumping an anisotropic gain medium through optical bifurcated fiber [10, 11], the orthogonally polarized dual-wavelength lasers can be realized in some Nd^{3+}- or Yb^{3+}-doped laser materials. Furthermore, by using the thermal-lensing effect of the host material [12] or the different transitions originated from two inequivalent Nd^{3+} centers [13], a single laser crystal can also be used to generate two output beams with the polarizations being orthogonal to each other, without introducing any optical elements in a single laser resonator. This method is more simple and favorable to construct a compact and portable THz source, as demonstrated in a single Q-switched Nd^{3+}:$LiYF_4$ laser [14].

Lanthanum borate molybdate $LaBO_2MoO_4$ crystal belongs to the monoclinic system and has a non-centrosymmetric structure, which shows the potential of nonlinear optical effects based on $\chi^{(3)}$- and $\chi^{(2)}$-nonlinearity [15, 16]. Nd^{3+}:$LaBO_2MoO_4$ crystal melts congruently and can be grown by the Czochralski method in the air atmosphere. Unpolarized spectroscopic properties of Nd^{3+}:$LaBO_2MoO_4$ crystal have been investigated and weak pulsed stimulated emissions at two wavelengths of 1,058.5 and 1,067.5 nm pumped by Xe-flashlamp has been detected in a (010) plate [16, 17]. Based on its stimulated emission and nonlinear optical properties, Nd^{3+}:$LaBO_2MoO_4$ crystal is considered as a promising laser gain medium with nonlinear optical effects, such as self-frequency doubling and self-Raman lasers.

In this work, using a *Z*-cut Nd^{3+}:$LaBO_2MoO_4$ crystal as gain medium and without introducing any optical elements, simultaneous laser operations at two wavelengths of 1,068

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and 1,074 nm with the polarizations being orthogonal to each other are firstly reported in a 795 nm diode-end-pumped plano-concave resonator. At the same time, the influences of the resonator length and pump power on the output wavelength of the Nd^{3+}:$LaBO_2MoO_4$ laser are also investigated.

2 Spectroscopic property

The Nd^{3+}:$LaBO_2MoO_4$ crystal was grown by the Czochralski method in the air atmosphere [17]. By the inductively coupled plasma atomic emission spectrometry (ICP-AES, Ultima2, Jobin-Yvon), the accurate concentration of the Nd^{3+} ions in the crystal was measured to be 1.8 at.% (about 1.6×10^{20} cm^{-3}). In order to analyze the spectroscopic properties accurately, the crystal was oriented and cut along three optical indicatrix axes *X*, *Y*, and *Z*, which are corresponding to the directions with the minimum, intermediate, and maximum values of refractive index, respectively.

Room-temperature polarized absorption spectra of the Nd^{3+}:$LaBO_2MoO_4$ crystal were recorded using a spectrophotometer (Lambda35, Perkin-Elmer) and are shown in Fig. 1 in a range from 770 to 840 nm. For *E*//*X* polarization, there are two absorption peaks at 798 and 802 nm with similar absorption cross sections (about 4.6×10^{-20} cm^2). For *E*//*Y* and *E*//*Z* polarizations, the absorption peaks are both located at 802 nm and the absorption cross sections are similar, i.e., about (2.5–2.6) $\times 10^{-20}$ cm^2. The full-width at half the maximum (FWHM) of the absorption band for *E*//*X* polarization is estimated to be about 10 nm. The structure analysis has shown that there are two inequivalent La^{3+} sites in the $LaBO_2MoO_4$ crystal [17]. Therefore, the broad FWHM of the crystal is caused by the multisite distribution of Nd^{3+} ions, which substituted the La^{3+} ions in the crystal. The FWHM of the absorption band around 800 nm of the Nd^{3+}:$LaBO_2MoO_4$ crystal is about ten times larger than that (about 1 nm) of Nd^{3+}:YAG crystal [18], which means that the crystal is very convenient for laser diode (LD) pumped operation. However, it must be noted that the multisite distribution of Nd^{3+} ions in the crystal is also a drawback for efficient laser operation, because it leads to a reduced extraction efficiency and an instability of output laser spectrum [19].

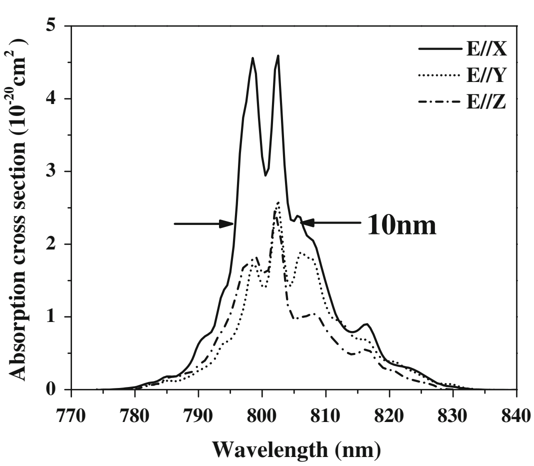

Fig. 1 Room temperature polarized absorption spectra of the Nd^{3+}:$LaBO_2MoO_4$ crystal in a range from 770 to 840 nm

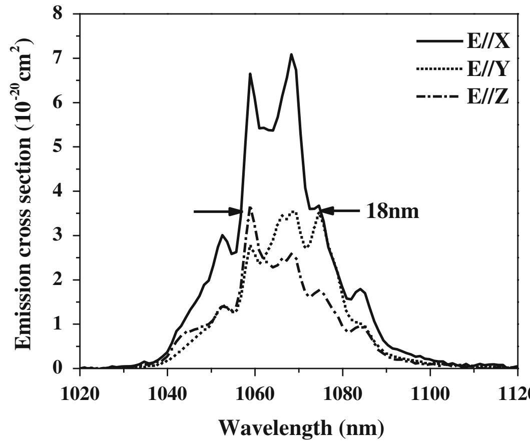

Fig. 2 Room temperature polarized emission spectra of the Nd^{3+}:$LaBO_2MoO_4$ crystal in a range from 1,020 to 1,120 nm

Room-temperature polarized emission spectra of the Nd^{3+}:$LaBO_2MoO_4$ crystal were recorded in a range from 1,020 to 1,120 nm by a spectrophotometer (FL920, Edinburg) when the exciting wavelength was 802 nm. The stimulated emission cross sections were calculated by the Füchtbauer-Ladenburg formula and are shown in Fig. 2. For *E*//*X* polarization, the peak emission cross section at 1,068 nm is 7.1×10^{-20} cm^2. The FWHM of the emission band around 1,070 nm of the crystal is about 18 nm and twenty times larger than that (about 0.8 nm) of Nd^{3+}:YAG crystal [18], which means that the crystal is a promising gain medium for tunable and ultrashort pulse lasers. For *E*//*Y* polarization, there are two emission peaks at 1,068 and 1,074 nm with similar emission cross sections (about 3.5×10^{-20} cm^2). For *E*//*Z* polarization, the peak emission wavelength is 1,058 nm and the emission cross section is 3.7×10^{-20} cm^2. The different peak emission wavelengths at different polarizations of the crystal suggest the potentiality that different laser wavelengths may be realized by using different oriented crystals as gain media.

3 Laser experimental arrangement

End-pumped linear plano-concave resonator was adopted in the laser experiment and the schematic of experimental

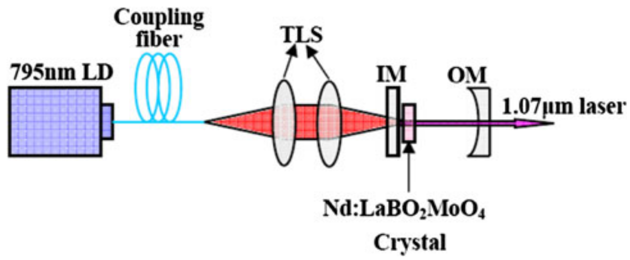

Fig. 3 Experimental setup of the quasi-cw 795 nm diode-pumped Nd^{3+}:$LaBO_2MoO_4$ laser around 1.07 μm

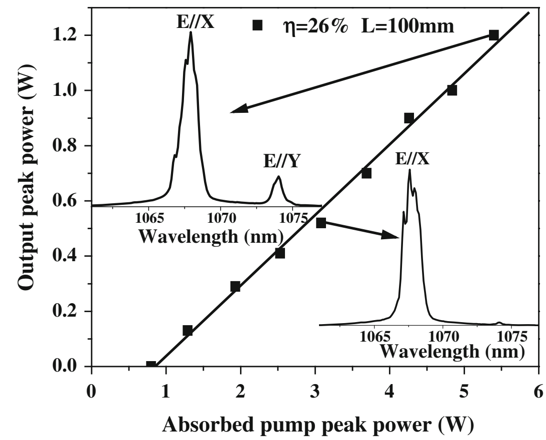

Fig. 4 Output peak power of the Nd^{3+}:$LaBO_2MoO_4$ laser as a function of absorbed pump peak power at 795 nm for OM transmission of 5.1 % and resonator length L of about 100 mm. Laser spectra at different absorbed pump peak powers are also shown

setup is depicted in Fig. 3. Based on the absorption spectra shown in Fig. 1, a *Z*-cut Nd^{3+}:$LaBO_2MoO_4$ crystal was used as gain medium for the utilization of large absorption in *E*//*X* polarization. The thickness of the crystal plate was about 2.2 mm. Because of the lack of LD emitting at 802 or 798 nm in our lab, a 795 nm fiber-coupled LD (100 μm diameter core) was used as pump source. The unpolarized pump light from the LD was firstly passed a simple telescopic lens system (TLS) consisted of a collimating and a focusing lens, and then imaged onto the plate. The plate was positioned at the focal point of the TLS and the waist radius of pump beam in the plate was estimated to be about 60 μm. The uncoated crystal plate was attached on an aluminum slab with heat-conducting adhesive and no other device was used to control the temperature of the plate. There is a hole in the center of the slab to permit the passing of the pump and fundamental laser beams. Because of the low fluorescence quantum efficiency (about 40 % [17]) of the $^4F_{3/2}$ multiplet of Nd^{3+} in $LaBO_2MoO_4$ crystal, a large amount of heat was generated in the crystal pumped by LD [20]. Due to the lack of the suitable crystal cooling apparatus in our lab presently, the LD was operated in quasi-continuous-wave (quasi-cw) mode in order to reduce the influence of pump-induced thermal load on laser performance and avoid the fracture of the plate at high pump power. Pump pulse period was 100 ms and pulse width was 2 ms, which is far longer than fluorescence lifetime (about 0.1 ms) of upper laser level $^4F_{3/2}$ of Nd^{3+} ions in the crystal. The flat input mirror (IM) of the laser resonator had 90 % transmission at 795 nm and 99.7 % reflectivity around 1,070 nm. An output mirror (OM) with curvature radius of 100 mm and transmission of 5.1 % around 1,070 nm was used to complete the resonator. About 55 % of the incident pump power was absorbed by the crystal plate.

4 Results and discussion

When the resonator length was close to the curvature radius of OM, i.e., 100 mm, output peak power of the Nd^{3+}:$LaBO_2MoO_4$ laser as a function of absorbed pump peak power at 795 nm was recorded and is shown in Fig. 4. A maximum output peak power of 1.2 W was achieved when the absorbed pump peak power was 5.4 W. The absorbed pump threshold was about 0.8 W and slope efficiency η was 26 %. The obtained slope efficiency is lower than those (about 50–60 %) realized in some commercial laser crystals, such as Nd^{3+}:YAG and Nd^{3+}:YVO_4 [21, 22]. Previous investigation has shown that the inhomogeneous broadening of fluorescence band of laser materials has a strong influence on the output laser performances, and will reduce the slope efficiency of output laser [19, 21, 23], as demonstrated in some laser crystals with multisite characteristic (about 30–40 % slope efficiency obtained in these crystals [21, 23]). Of course, the optical quality of the Nd^{3+}:$LaBO_2MoO_4$ crystal also influences the output laser performances. The stabilities of output power of the Nd^{3+}:$LaBO_2MoO_4$ laser at absorbed pump peak power of 5.4 W were measured in a short (1 min) and long (60 min) times, respectively, as shown in Fig. 5. It can be found that the fluctuations of output power were $< \pm 2$ and ± 3 % during 1 and 60 min of operation, respectively.

Spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser at different pump powers were recorded by a spectrometer (HR4000, Ocean Optics) and are also shown in Fig. 4. It can be seen that at the maximum pump power of 5.4 W, there are two laser wavelengths at 1,068 and 1,074 nm, and their polarized directions are orthogonal to each other and parallel to *X* and *Y*, respectively. The frequency difference between the two laser lines is close to 1.57 THz. When pump power was reduced to about 3.1 W, laser oscillation at 1,074 nm disappeared. When the pump power was 5.4 W and the cavity length was 100 mm, the spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser were recorded at time interval of 3 min

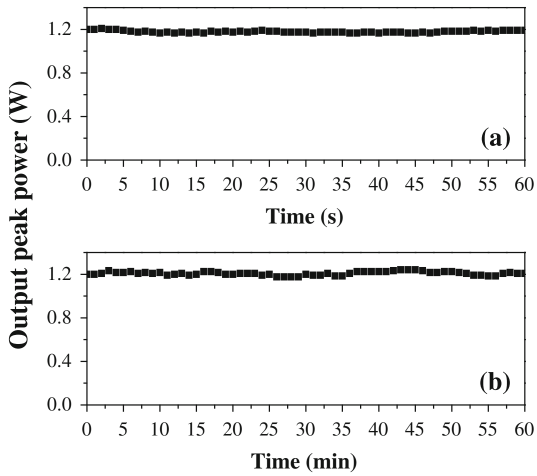

Fig. 5 Output power stabilities of the Nd^{3+}:$LaBO_2MoO_4$ laser in a short (**a**) and long (**b**) times when absorbed pump peak power was fixed at 5.4 W

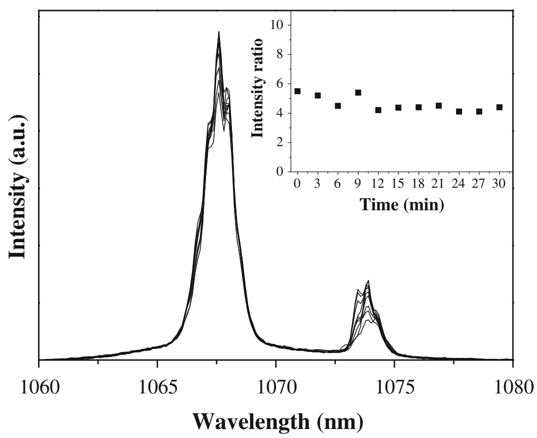

Fig. 6 Spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser recorded at time interval of 3 min during 30 min of operation when the pump power was 5.4 W and the cavity length was 100 mm. The *inset* shows the intensity ratio between 1,068 and 1,074 nm lasers versus operation time

during 30 min of operation, as shown in Fig. 6. It can be seen that the intensity ratio between 1,068 and 1,074 nm lasers is kept at about 4.5 after a certain operating time (about 10 min), which shows a good time stability for both laser lines. Therefore, mode hopping was not observed in the Nd^{3+}:$LaBO_2MoO_4$ laser. Furthermore, the distance between IM and the input face of the crystal was about 1 mm for the Nd^{3+}:$LaBO_2MoO_4$ laser. The etalon effect between IM and the input face of the crystal could occur to modulate the laser spectrum with a peak–peak separation of about 0.6 nm for our experimental condition. This separation is far smaller than that (6 nm) between 1,068 and 1,074 nm lasers. Therefore, two laser lines observed in this work are not caused by any etalon effect [24]. When the distance between IM and the input face of the crystal was

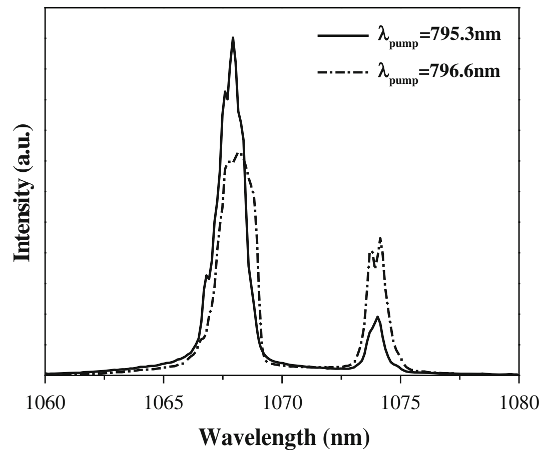

Fig. 7 Spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser pumped at 795.3 and 796.6 nm when absorbed pump peak power was fixed at 5.4 W

increased, i.e., the crystal was moved from the focal point of the TLS (in which the highest pump power density can be achieved in the crystal), laser oscillation at 1,068 nm became more favorable and the ratio of output powers between 1,068 and 1,074 nm increased. This variation was caused by the decrement of pump power density because the gain of 1,074 nm laser is lower than that of 1,068 nm. Dual-wavelength laser oscillation of the Nd^{3+}:$LaBO_2MoO_4$ crystal may be originated from the existence of two inequivalent Nd^{3+} centers, as having been realized in the Nd^{3+}:$La_2CaB_{10}O_{19}$ crystal [13]. This can be confirmed by the spectral change of the Nd^{3+}:$LaBO_2MoO_4$ laser pumped by different LD wavelengths at 795.3 and 796.6 nm achieved by changing the operating temperature of the LD [24], as shown in Fig. 7.

When the resonator length was close to 100 mm, output power at 1,068 nm was far higher than that at 1,074 nm, as shown in Fig. 4. For the efficient generation of THz radiation by DFG, the ratio of the powers between the two orthogonally polarized lasers should be close to unity. We found that the ratio of the laser powers at 1,068 and 1,074 nm changed with the resonator length, as shown in Fig. 8 at the maximum absorbed pump peak power of 5.4 W. The variation of the total output power was less than 10 % when the resonator length was changed from 100 to 90 mm. With the reduction of the resonator length, output power at 1,074 nm increased gradually. When the resonator length was 90 mm, output power at 1,074 nm was far higher than that at 1,068 nm. A similar phenomenon has been observed in some Nd^{3+}-doped self-frequency doubling laser crystals and the influence of the cavity length on the polarization of the output laser can be explained by a walk-off effect [25, 26], which is also existed in the Nd^{3+}:$LaBO_2MoO_4$ biaxial crystal when a Z-cut sample is pumped as a gain medium. At the resonator length of 95 mm, the intensity ratio between 1,068 and

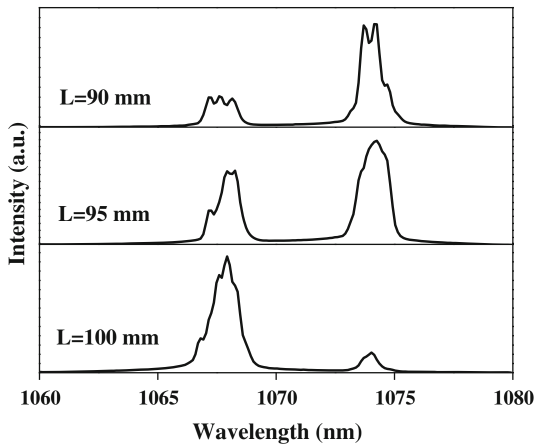

Fig. 8 Spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser at different resonator lengths *L* when absorbed pump peak power was fixed at 5.4 W

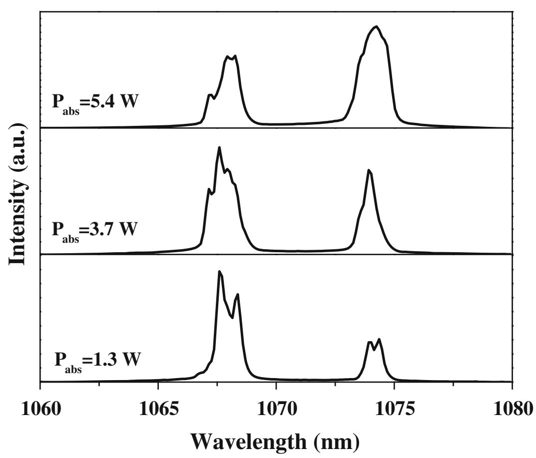

Fig. 9 Spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser at different absorbed pump peak powers P_{abs} when resonator length was fixed at about 95 mm

1,074 nm lasers was near 0.75 and stable during 30 min of operation. Then, when the resonator length of the Nd^{3+}:$LaBO_2MoO_4$ laser was adjusted to a certain value, the ratio of output powers between 1,068 and 1,074 nm lasers can be near unity. At the resonator length of 95 mm, spectra of the Nd^{3+}:$LaBO_2MoO_4$ laser at different pump powers were also recorded and are shown in Fig. 9. With the reduction of the pump power, decrement of the output power at 1,074 nm was more rapid than that at 1,068 nm. It means that laser threshold at 1,074 nm is higher than that at 1,068 nm, which is originated from the smaller emission cross section at 1,074 nm shown in Fig. 2. Although the laser gain at 1,074 nm is lower than that at 1,068 nm, better mode matching between the 1,074 nm laser and the pump beam realized in this cavity configuration, which must be demonstrated by more experiments in the future, may causes a higher slope efficiency and then output power at 1,074 nm laser.

5 Conclusions

Room temperature polarized absorption and emission spectra of 1.8 at.% Nd^{3+}:$LaBO_2MoO_4$ crystal were measured and analyzed. Due to the multisite distribution of Nd^{3+} ions in the crystal, broad FWHMs of absorption around 800 nm and emission bands around 1,070 nm were observed. Pumped by a 795 nm LD, dual-wavelength laser at 1,068 and 1,074 nm with the polarizations being orthogonal to each other was realized in a *Z*-cut Nd^{3+}:$LaBO_2MoO_4$ crystal without introducing any optical elements. The maximum total output peak power of 1.2 W with slope efficiency of 26 % was obtained. Furthermore, the ratio of the laser powers between 1,068 and 1,074 nm changed with the resonator length and pump power. When the resonator length and pump power were adjusted to certain values, the ratio of the laser powers between these two wavelengths can be close to unity. The results shown that the Nd^{3+}:$LaBO_2MoO_4$ crystal may have a potential application in terahertz technique.

Acknowledgments This work has been supported by the National Natural Science Foundation of China (grants 91122033), the Knowledge Innovation Program of the Chinese Academy of Sciences (grant KJCX2-EW-H03-01) and Chunmiao Project of Haixi Institute of Chinese Academy of Sciences (CMZX-2013-005).

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