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Article 1 **SESAM mode-locked Yb:Ca3Gd2(BO3)⁴ femtosecond laser** ²

Huang-Jun Zeng 1.2, Zhang-Lang Lin 2, Wen-Ze Xue 2, Ge Zhang 2, Zhongben Pan 3, Haifeng Lin 4, Pavel Loiko 5 **,** 3 **Xavier Mateos 6,#, Valentin Petrov ⁷ , Li Wang ⁷ and Weidong Chen 2,7,*** 4

- ¹ College of Chemistry, Fuzhou University, Fuzhou, 350002 Fujian, China; zenghuangjun@fjirsm.ac.cn (H.J.Z.) 5
- ² Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, 350002 Fu- 6 jian, China; linzl@fjirsm.ac.cn (Z.L.L.)[; xuewenze@f](mailto:xuewenze@)jirsm.ac.cn (W.Z.X.); zhg@fjirsm.ac.cn (G.Z.); chenwei- 7 dong@fiirsm.ac.cn (W.C.) 8
- ³ Institute of Chemical Materials, China Academy of Engineering Physics, Mianyang, 621900 Sichuan, China; 9 pzb8625@126.com (Z.P.) 10
- ⁴ College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen, 518118 Guangdong, 11 China; h.f.lin@szu.edu.cn (H.L.) 12
- ⁵ Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSI- 13 CAEN, Université de Caen Normandie, 6 Boulevard Maréchal Juin, 14050 Caen Cedex 4, France; 14 pavel.loiko@ensicaen.fr (P.L.) 15
- ⁶ Universitat Rovira i Virgili (URV), Física i Cristal·lografia de Materials i Nanomaterials (FiCMA-FiCNA), 16 Marcel.li Domingo 1, 43007 Tarragona, Spain; #Serra Húnter Fellow; xavier.mateos@urv.cat (X.M.) 17
- ⁷ Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2a, 12489 Berlin, Ger- 18 many; petrov@mbi-berlin.de (V.P.); Li.Wang@mbi-berlin.de (L.W.); chenweidong@fjirsm.ac.cn (W.C.) 19
- $\text{Correspondence: } \text{chenweidong@fjirsm.ac.cn}$ 20

Abstract: We report on the first passively mode-locked femtosecond laser operation of a disordered 21 Yb:Ca3Gd2(BO3)⁴ crystal using a SEmiconductor Saturable Absorber Mirror(SESAM). Pumping with 22 a single-transverse mode fiber-coupled laser diode at 976 nm, nearly Fourier-transform limited 23 pulses as short as 96 fs are generated at 1045 nm with an average output power of 205 mW and a 24 pulse repetition rate of ~67.3 MHz. In the continuous-wave regime, high slope efficiency up to 59.2% 25 and low laser thresholds down to 25 mW are obtained. Continuous wavelength tuning between 26 $1006 - 1074$ nm (a tuning range of 68 nm) is demonstrated. Yb:Ca3Gd2(BO3)4 crystals are promising 27 for the development of ultrafast lasers at \sim 1 μ m. 28

Keywords: Ultrafast laser; Solid-state laser; Ytterbium laser; Mode-locking. 29

1. Introduction 31

Ytterbium (Yb³⁺) doped crystals are attractive for the development of power-scalable, 32 wavelength-tunable and ultrafast coherent light sources emitting at the wavelength of 33 \sim 1 μm. Yb³⁺ ions feature a simple electronic level scheme consisting of only two multiplets 34 leading to higher laser slope efficiencies and weaker heat loading (as compared to Nd^{3+} 35 ions) $[1]$. Yb³⁺-doped crystals can be efficiently pumped by commericially available high- 36 power InGaAs diode lasers emitting at 0.94 – 0.98 μm. For generation of ultrashort pulses 37 in the sub-100 fs time domain, it is advantageous to use Yb^{3+} -doped crystals exhibiting 38 structure disorder leading to strong inhomogeneous broadening of the $Yb³⁺$ absorption 39 and emission bands [2-4]. In this way, smooth and broad gain profiles are accessible. It is 40 still interesting to search for novel disordered crystals for Yb^{3+} doping combining large 41 total Stark splitting of the ground-state ²F_{7/2} (leading to broader emission range) and good 42 thermal properties (allowing for power scalable laser operation). 43

Among the laser host crystals suitable for Yb^{3+} doping, double borates [e.g., 44] REAl₃(BO₃)₄, RECa₄O(BO₃)₃, etc., where RE denotes a passive rare-earth ion] are attracting 45 a lot of attention. They are characterized by a relatively large ground-state splitting [~1000 46 cm-1 for GdCa4O(BO3)3] resulting from strong crystal fields, good thermo-optical [5] and 47

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mechanical properties, high Yb^{3+} doping concentration (few tens at.%) that can be 48 achieved and attractive nonlinear optical properties making them suitable, e.g., for self- 49 frequency doubling. Monoclinic calcium RE oxoborates Yb:YCa4O(BO3)³ are a good exam- 50 ple of such crystals that exhibit local structure disorder leading to smooth and broad gain 51 profiles extending up to 1.1 μ m. Such advantages make them very suitable for high-power 52 operation in the continuous-wave (CW) regime [6-8], as well as the design of broadband 53 widely tunable and mode-locked (ML) lasers operation at \sim 1 μ m [9, 10]. A diode-pumped 54 Yb:GdCa4O(BO3)3 (abbreviated: Yb:GdCOB) laser ML by a SESAM generated 90 fs pulses 55 at ~1046 nm corresponding to an average output power of 40 mW at a pulse repetition 56 rate of 100 MHz [10]. 57

Recently, another calcium borate crystal family with a chemical formula 58 Ca₃RE₂(BO₃)₄, where RE = Y, Lu, Gd and La, attracted attention for Yb³⁺ doping [11-14]. 59 These crystals belong to the orthorhombic class with the space group *Pnma* and they are 60 structurally disordered. Thus, Yb^{3+} -doped Ca $sGd_2(BO_3)_4$ crystal (abbreviated Yb:GdCB) 61 with high optical quality has been successfully grown by the Czochralski method and 62 laser performance in the CW regime has been characterized [11, 15, 16]. In the GdCB struc- 63 ture, the Ca^{2+} and Gd^{3+} cations statistically occupy three non-equivalent crystallographic 64 sites forming M-oxygen distorted polyhedrons [17] (the dopant Yb^{3+} ions are expected to 65 replace for the Gd³⁺ ones). This leads to a "glassy-like" spectroscopic behavior of Yb:GdCB 66 crystals at the expense of a relatively low thermal conductivity of 0.92 W/mK at room 67 temperature [18]. Pumping with a multi-transverse mode, fiber-coupled laser diode at 68 976 nm, a maximum CW output power of 1.4 W was obtained at \sim 1060 nm with a rela- 69 tively low slope efficiency of 23.7% and high laser threshold of 900 mW [16]. Self-Q 70 switched operation was also reported [19]. However, there is no report on a passively ML 71 operation of Yb:GdCB laser to date. 72

In the present work, we report on the laser performance of the disordered Yb:GdCB 73 crystal in the CW and the passively ML regimes. Pumping with a single-transverse mode, 74 fiber-coupled laser diode, nearly Fourier-transform limited pulses as short as 96 fs were 75 generated using a SESAM. To the best of our knowledge, this is the first report on the ML 76 operation of the Yb:GdCB crystal. The Second Se

2. Experimental setup 78

The experimental configuration of the Yb:GdCB laser is shown in Fig. 1. A high-qual- 79 ity crystal was grown by the Czochralski method with a measured $Yb³⁺$ concentration of 80 5 at.%.

Figure 1. Schematic of the Yb:GdCB laser. LD: fiber-coupled laser diode; L₁: aspherical lens; L₂: 83 achromatic doublet lens; M_1 , M_2 and M_4 : concave mirrors; M_3 : flat rear mirror for CW operation; 84 DM¹ – DM6: dispersive mirrors; OC: output coupler; SESAM: SEmiconductor Saturable Absorber 85 Mirror. 86

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Yb:GdCB belongs to the orthorhombic class and its lattice constants are $a = 7.1937 \text{ Å}$, 87 $b = 15.5311$ Å and $c = 8.6140$ Å. Here, we use the notations of the standard *Pnma* crystallo- 88 graphic setting. The laser crystal was cut along the *a*-axis with an aperture of 89 4 mm × 4 mm and a thickness of 3 mm. The input and output faces were polished to laser 90 quality and left uncoated. The crystal was mounted on a copper holder without active 91 cooling and placed between two dichroic folding mirrors M¹ and M² (radius of curvature, 92 RoC = -100 mm) with the Brewster angle minimum loss condition fulfilled for both the 93 laser and the pump beams. CW and ML laser operation of the Yb:GdCB crystal were eval- 94 uated with an X-folded astigmatically compensated linear cavity. The crystal orientation 95 determined the laser polarization $E \parallel c$ (due to its higher gain compared to $E \parallel b$). The 96 pump source was a non-polarized fiber-coupled laser diode emitting a nearly diffraction- 97 limited beam with a propagation factor (M²) of ~1.02. The laser diode had a Fiber Bragg $-$ 98 Grating (FBG) for wavelength locking at 976 nm over the entire operation range with an 99 emission bandwidth (full width at half-maximum, FWHM) of 0.2 nm, which well matched 100 the bandwidth of the zero-phonon-line in the absorption spectrum of the Yb:GdCB crystal. 101 Given the transmissions of the pump reimaging lenses and the pump mirror M_1 , the max- 102 imum incident pump power on the laser crystal was 1.29 W. The use of the single-trans- 103 verse mode laser diode with nearly diffraction-limited beam led to higher gain per watt 104 of absorbed pump power due to improved mode-matching and the lowest thermal stress 105 in the laser crystal (as compared to pumping by fiber-coupled diodes with a "top-hat" 106 beam profile). This allowed us to optimize the Yb laser for low threshold and high effi- 107 ciency operation in the CW regime, and also for femtosecond pulse generation in the ML 108 regime. The pump beam was collimated by an aspherical lens L_1 ($f = 26$ mm) and focused 109 into the laser crystal through the M_1 mirror using an achromatic doublet lens L_2 110 $(f=100 \text{ mm})$ resulting in a beam waist radius of 18.7 and 36.9 μ m in the sagittal and tan- 111 gential planes, respectively. 112

3. Continuous-wave laser operation 113

In the CW regime, the laser performance of the Yb:GdCB crystal was evaluated with 114 a four-mirror cavity including a flat rear mirror M_3 and an output coupler (OC) without 115 SESAM, as shown in Fig. 1. 116

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Figure 2. CW laser performance of *a*-cut Yb:GdCB crsytal: (a) input-output dependences for differ- 118 ent transmission of the OC (*T*OC), *η* – slope efficiency; (b) laser spectra. The laser polarization is *E* ‖ *c*. 119

The cavity mode size in the laser crystal was estimated by the ABCD formalism yield- 120 ing a waist radius of 21 and 37 μm in the sagittal and tangential planes, respectively. As 121 shown in Fig. 2(a), a maximum output power of 382 mW was achieved for OC with a 122

transmission at the laser wavelength *Toc* = 0.8% at an absorbed pump power of 725 mW, 123 corresponding to a laser threshold of only 25 mW and a slope efficiency of 55.2%. The 124 maximum slope efficiency of 59.2% was obtained for higher $T_{\text{OC}} = 4.5$ %. The laser thresh- 125 old gradually increased with *T*oc (0.4% - 7.5%), from 25 to 158 mW. The measured single- 126 pass pump absorption under lasing conditions tended to decrease with *T*_{OC} between 56.2% 127 to 43%, indicating a certain ground-state bleaching and its suppression by the recycling 128 effect. The emission wavelength of the Yb:GdCB laser in the CW regime experienced a 129 blue-shift with increasing transmission of the OC, from 1025.5 to 1053.4 nm, see Fig. 2(b). 130 This behavior is due to the quasi-three-level nature of the Yb laser scheme. 131

Figure 3. (a) Caird analysis for the CW Yb:GdCB laser: slope efficiency as a function of the OC re- 133 flectivity *Roc* = 1 - *Toc*; (b) wavelength tuning curve obtained with a SF-10 Brewster prism placed 134 between M_1 and the OC with $T_{OC} = 1\%$. 135

The Caird analysis [20] was applied to estimate the total losses *δ* (including cavity 136 and laser crystal but excluding reabsorption effects) and the intrinsic slope efficiency *η*⁰ 137 through fitting the measured laser slope efficiency as a function of the reflectivity of the 138 OC, $Roc = 1 - Toc$ with the following equation: 139

$$
\eta = \eta_0 \frac{\lambda_p - \ln(R_{OC})}{\lambda_l \delta - \ln(R_{OC})} \tag{1}
$$

where λ_p and λ_l are the pump and laser wavelengths, respectively. The best fit gave round 141 trip cavity losses of *δ* = 0.07% and an intrinsic slope efficiency of *η*⁰ = 59.9%, see Fig. 3(a). 142 The extremely low cavity losses are evidence for good quality of the laser crystal and well- 143 optimized cavity alignment. 144

A SF-10 Brewster prism was inserted close to the OC for wavelength tuning in the 145 CW regime. With a 1% OC, a broad range of continuous wavelength tuning, about 68 nm, 146 was obtained, as shown in Fig. 3(b). 147

4. SESAM mode-locked operation 148

For passively ML operation, the rear mirror M³ was replaced by a plane-concave mir- 149 ror M_4 with RoC = -100 mm to create a second beam waist on the SESAM ensuring its 150 efficient bleaching. The estimated radius of this second beam waist was 60 and 65 μ m in 151 the sagittal and tangential planes, respectively. The SESAM (BATOP, GmbH) applied in 152 this experiment had a relaxation time constant of \sim 1 ps and a modulation depth of 1.2%. 153 The intracavity group delay dispersion (GDD) was managed by implementing four flat 154 dispersive mirrors (DMs, characterized by the following GDD per bounce: $DM_1 = -100$ fs², , 155

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DM2 = -100 fs², DM3 = DM4= -250 fs², DM5 = DM6= -55 fs²) in the other cavity arm. The total 156 negative GDD value was varied by changing the number of bounces on the DMs. 157

Initially, the ML operation was investigated by applying eight bounces (single pass) 158 on the DMs (DM₁ – DM₄), as shown in Fig. 1, yielding a total round-trip negative GDD of 159 -2800 fs². Stable and self-starting ML operation was achieved with the 1.6% OC. The sech $^{\rm 2}$ - 160 shaped spectrum of the soliton pulses was centered at 1052 nm with a FWHM of 8.2 nm, 161 as shown in Fig. 4(a). The measured SHG-based intensity autocorrelation trace gave a 162 pulse duration of 146 fs by assuming a sech²-shape temporal profile, see Fig. 4(b). This 163 results in a time-bandwidth product (TBP) of 0.324 which is slightly above the Fourier 164 transform-limited value for a sech²-shaped temporal profile. The long-scale SHG-based 165 intensity autocorrelation scan of 50 ps indicates a single-pulse operation without any ped- 166 estals or multi-pulses, see inset in Fig 4(b). The maximum average output power 167 amounted to 181 mW at an absorbed pump power of 697 mW. The corresponding peak 168 power was ~17.5 kW at a pulse repetition rate of 62.4 MHz. The optical conversion effi- 169 ciency reached 26% with respect to the absorbed pump power. 170

Figure 4. SESAM ML Yb:GdCB laser with *Toc* = 1.6%. (a) Optical spectrum and (b) SHG-based 172 intensity autocorrelation trace with a sech² fit. *Inset*: simultaneously measured long-scale SHG- 173 based intensity autocorrelation trace for the time span of 50 ps. 174

The pulse duration was further shortened by optimizing the total negative GDD 175 value. The shortest pulse duration was achieved by applying six bounces (single pass) on 176 the DMs (DM³ – DM6), as shown in Fig. 1, yielding a total negative GDD of -1620 fs² . Self- 177 starting ML operation of the Yb:GdCB laser with a 2.5% OC produced soliton pulses at 178 the central wavelength of 1045 nm with a spectral FWHM of 12.1 nm, as depicted in 179 Fig. 5(a). Pulses as short as 96 fs were achieved as shown by the measured SHG-based 180 intensity autocorrelation trace in Fig. 5(b). The corresponding TBP of 0.319 indicates gen- 181 eration of nearly Fourier-transform limited pulses with a sech²-shaped temporal profile. 182 A long-scale SHG-based intensity autocorrelation scan of 50 ps shown in the inset of 183 Fig. 5(b), confirmed a single-pulse ML operation. The maximum average output power 184 amounted to 205 mW at an absorbed pump power of 696 mW, which corresponded to an 185 efficiency of 29.5%. The soliton pulses had a peak power of 28 kW at a pulse repetition 186 rate of 67.3 MHz.

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Figure 5. SESAM ML Yb:Ca3Gd₂(BO₃)₄ laser with *T*_{OC} = 2.5%. (a) Optical spectrum and (b) SHG- 189 based intensity autocorrelation trace with a sech² fit. *Inset*: simultaneously measured long-scale 190 SHG-based intensity autocorrelation trace for the time span of 50 ps. 191

The steady-state pulse train corresponding to the shortest pulses achieved was char- 192 acterized by a radio-frequency (RF) spectrum analyzer. The first beat note was recorded 193 at ~67.3 MHz with a resolution bandwidth (RBW) of 300 Hz, as shown in Fig. 6(a). No 194 spurious modulations could be observed with a signal-to-noise ratio above 76 dBc. This, 195 together with the uniform 1-GHz harmonic beat notes shown in Fig. 6(b) is an evidence 196 for stable CW mode-locking of the Yb:GdCB laser without any Q-switching or multiple 197 pulsing instabilities. 198

Figure 6. Radio-frequency (RF) spectra of the SESAM ML Yb:GdCB laser: (a) fundamental beat 200 note at 67.3 MHz recorded a resolution bandwidth (RBW) of 300 Hz, and (b) harmonics on a 1- 201 GHz frequency span, measured with a RBW of 100 kHz. 202

5. Conclusion 203

In conclusion, we present a comprehensive characterization of a diode-pumped 204 Yb:GdCB laser operating both in the CW and passively ML regimes. Pumped with a sin- 205 gle-transverse mode, fiber-coupled laser diode at 976 nm, the CW laser generated a max- 206 imum output power of 382 mW at 1053. nm corresponding to a very low threshold of 207 25 mW and high slope efficiency of 55.2%. Continuous wavelength tuning between 208

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