Research Article Vol. 32, No. 23 / 4 Nov 2024 / Optics Express 40507
Optics EXPRESS

Diode-pumped Kerr-lens mode-locked ytterbium-doped compositional mixed calcium aluminate laser

ZHI-QIANG LI,[1] ZHANG-LANG LIN,[1] HUANG-JUN ZENG,[1] HAI-YU NIE,[1] GE ZHANG,[1] PEIXIONG ZHANG,[2] WENJIE WU,[2] ZHENQIANG CHEN,[2,7] ZHEN LI,[2] PAVEL LOIKO,[3] SIMONE NORMANI,[3] XAVIER MATEOS,[4] HSING-CHIH LIANG,[5] VALENTIN PETROV,[6] AND WEIDONG CHEN[1,6,*]

[1] *Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, 350002 Fuzhou, China*
[2] *Department of Optoelectronic Engineering, Jinan University, 510632 Guangzhou, China*
[3] *Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen, 6 Boulevard Maréchal Juin, 14050 Caen Cedex 4, France*
[4] *Universitat Rovira i Virgili, URV, Física i Cristal·lografia de Materials, FiCMA, Marcel·lí Domingo 1, 43007 Tarragona, Spain*
[5] *Institute of Physics, National Yang Ming Chiao Tung University, 30010 Hsinchu, Taiwan*
[6] *Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2a, 12489 Berlin, Germany*
[7] *tzqchen@jnu.edu.cn*
[*] *chenweidong@fjirsm.ac.cn*

Abstract: A diode-pumped Kerr-lens mode-locked laser generating 25 fs pulses at 1080 nm is demonstrated with an Yb^{3+}-doped compositional mixed calcium aluminate crystal. The Yb:Ca(Gd,Y)AlO_4 laser, which operates at a repetition rate of 65.6 MHz via soft-aperture Kerr-lens mode-locking, is pumped by a spatially single-mode, fiber-coupled diode laser. To the best of our knowledge, this is the first Kerr-lens mode-locked laser that utilizes an Yb^{3+}-doped compositional mixed calcium aluminate crystal as the gain medium.

1. Introduction

Ytterbium (Yb^{3+}) doped tetragonal calcium rare-earth aluminate crystals, i.e., Yb:Ca$LnAlO_4$, where Ln^{3+} denotes Y^{3+} or Gd^{3+}, currently represent one of the most attractive laser active media solutions for high-power femtosecond pulse generation via passive mode-locking [1–6]. The Gd^{3+} compound is abbreviated as Yb:CALGO and the Y^{3+} compound as Yb:CALYO. The Ca$LnAlO_4$ compounds crystallize in a K_2NiF_4-type structure with the Ca^{2+} and Ln^{3+} cations statistically distributed over the same Wyckoff site (4*e*, C_{4v}-symmetry) resulting in local structure disorder. When doped with Yb^{3+} ions, their absorption and emission spectral bands experience inhomogeneous line broadening [7]. This nearly "glassy-like" spectroscopic behavior of Yb^{3+}:Ca$LnAlO_4$ is characterized by extremely broad, smooth and flat gain profiles which support sub-50 fs pulse generation from passively mode-locked (ML) lasers [5,8,9], including multi-GHz repetition rates [10]. Despite their disordered nature and the similarity of spectroscopic behavior to that of glasses, the Ca$LnAlO_4$ crystals benefit from high thermal conductivity with weak dependence on the Yb^{3+} doping level [11].

Remarkably, sub-20 fs pulses could be generated from ultrafast lasers based on such crystals via Kerr-lens mode-locking (KLM), albeit at modest average output powers: Wang *et al.* achieved 17.8 fs pulses at 1118 nm from a KLM Yb:CALGO laser albeit with a low average output power

#541656 https://doi.org/10.1364/OE.541656
Journal © 2024 Received 9 Sep 2024; revised 30 Sep 2024; accepted 2 Oct 2024; published 22 Oct 2024

of 26 mW [12]. A similar performance was reported for a KLM Yb:CALYO laser at 1080 nm [13]. In both cases, however, extracavity residual chirp compensation was applied to obtain such short pulse durations. Moreover, as pointed out above, due to their appealing thermal and thermo-optical properties, the Ca*Ln*AlO_4 crystals are ideally suited for high-power laser operation at ~1 µm [14], including thin-disk laser geometry [15]. Thus, power scaling was demonstrated by pumping with a high-brightness Yb-fiber laser at 976 nm; a KLM Yb:CALGO bulk laser generated 729 mW of average output power at 1040.7 nm with a pulse duration of 22 fs [16]. As shown in [16], the naturally strong birefringence of these crystals leads to anisotropic absorption and emission properties, which offers more flexibility in pumping and spectral bandwidth utilization, in addition to the reduced thermal depolarization losses.

Current trends in material engineering of broadband laser gain media include the management of gain profiles via compositional disorder in solid-solution ("mixed") crystals. For calcium rare-earth aluminates, this can be realized by mixing CALGO and CALYO together or by Lu^{3+} codoping one of them [17–19], which may result in further smoothing and flattening of the gain profile of the Yb^{3+} ions. Recently, we reported on the crystal growth, polarized spectroscopy, and laser operation of a novel "mixed" calcium gadolinium-yttrium aluminate crystal, i.e., $CaYb_{0.032}Gd_{0.448}Y_{0.520}AlO_4$, with an Yb^{3+} doping level of 3.2 at%. Mode-locked by a commercially available SEmiconductor Saturable Absorber Mirror (SESAM), the Yb:Ca(Gd,Y)AlO_4 laser generated 35 fs pulses at 1059.8 nm with an average output power of 51 mW at a repetition rate of ~65.9 MHz [20].

Among the passive mode-locking techniques, KLM stands out as one of the most promising methods for generation of extremely short light pulses from solid-state lasers down to a few femtoseconds [21]. This is achieved owing to the action of an artificial "fast" saturable absorber (SA) based on Kerr lensing in the active medium. In the case of soft-aperture KLM, the Kerr lens leads to a better overlap of the pump and laser beams resulting in higher gain at the peak of the pulse. This modulates the gain in the cavity similar to the action of a physical SA but at a much higher speed leading to an intensity-dependent quasi-instantaneous self-amplitude modulation (SAM) [22]. Apart from its intrinsically very fast response, the key advantage of KLM enabling the generation of ultimately short light pulses with extreme bandwidths and high average output powers is the absence of spectral limitations and non-saturable losses being common to physical SAs [23]. The superior spectroscopic properties as well as the preliminary laser results motivated us to explore further pulse shortening in the Yb:Ca(Gd,Y)AlO_4 laser to better exploit its gain bandwidth. In the present work we demonstrate, for the first time to the best of our knowledge, sub-30 fs pulse generation from a diode-pumped Yb:Ca(Gd,Y)AlO_4 laser via soft-aperture KLM.

2. Laser setup

An X-folded astigmatically compensated linear cavity was used to explore the KLM laser performance of the Yb:Ca(Gd,Y)AlO_4 crystal, as shown in Fig. 1.

A fiber-coupled InGaAs laser diode delivering unpolarized continuous-wave (CW) laser radiation with a near diffraction-limited beam quality (measured $M^2 = 1.02$) at 976 nm was employed as a pump source. Its emission wavelength was locked by a fiber Bragg grating (FBG) resulting in a spectral linewidth (full width at the half maximum, FWHM) of 0.2 nm. An uncoated laser element with an aperture of 4 mm × 4 mm and a thickness of 3 mm was cut from the annealed bulk crystal with an actual Yb^{3+} doping level of 3.2 at.% for light propagation along the crystallographic ***c***-axis (***c***-cut). Both its surfaces were polished to laser-grade quality with good parallelism. The laser element was mounted in a copper holder without active cooling (room temperature) and placed at Brewster's angle between two concave mirrors M_1 and M_2 (radius of curvature: RoC = -100 mm) with the minimum loss condition fulfilled for σ-polarization. The pump beam was reimaged into the laser crystal employing an aspherical lens L_1 (focal length: $f = 26$ mm) and a spherical lens L_2 ($f = 75$ mm) through the concave pump mirror M_1,

Optics EXPRESS

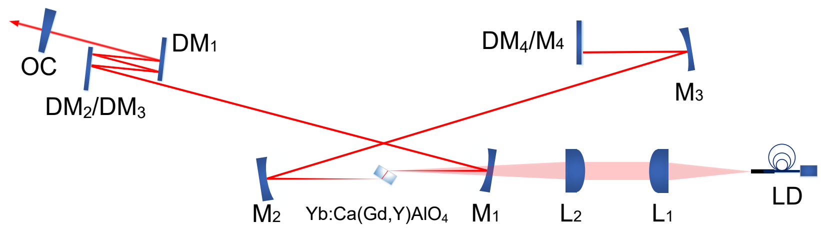

Fig. 1. Experimental setup of the diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser. LD: fiber-coupled InGaAs laser diode; L_1: aspherical lens; L_2: spherical lens; M_1, M_2 and M_3: concave mirrors; M_4: flat rear mirror; DM_1 – DM_4: flat dispersive mirrors; OC: output coupler.

which resulted in a beam waist (radius) of 16.8 µm × 28.8 µm in the sagittal and tangential planes, respectively. The maximum incident pump power amounted to 1.33 W. For KLM operation, flat dispersive mirrors (DMs) were implemented to compensate the material dispersion and to balance the self-phase modulation (SPM) induced by the Kerr nonlinearity of the laser crystal. A round-trip material dispersion of +503.6 fs^2 at 1080 nm was estimated from the reported Sellmeier equations by averaging the linear susceptibility of two parent compounds, CALGO and CALYO [24]. The physical length of the mode-locked laser cavity was ~2.25 m which corresponds to a pulse repetition rate of ~66 MHz.

3. Results and discussion

Initially, the laser performance of the diode-pumped Yb:Ca(Gd,Y)AlO_4 laser was characterized in the CW regime with two DMs providing a round-trip negative group delay dispersion (GDD) of -1600 fs^2 (DM_1 = -150 fs^2 and DM_2 = -250 fs^2) and an OC transmission of $T_{OC} = 4\%$. A maximum output power of 290 mW at a central wavelength of 1052 nm was obtained at an absorbed pump power of 779 mW. The pump absorption was estimated by measuring the single pass pump absorption with consideration of the reflection from the surfaces of the laser crystal.

The realization of KLM operation is normally difficult as the laser resonator must operate close to its stability limit and mode-locking is normally not self-starting but requires an external trigger, e.g., knocking on a resonator mirror to induce intensity fluctuations. The present laser was aligned towards the edge of the stability region through translating the folding mirror M_2 away from the pump mirror (M_1) step by step, which resulted in a significant reduction of the CW output power. After careful cavity alignment, KLM operation could be started by slightly knocking the OC or translating the flat rear mirror M_4. When mode-locked, the Yb:Ca(Gd,Y)AlO_4 laser experienced an abrupt increase of the output power from 89 mW (CW) to 116 mW (KLM). The measured optical spectrum of the laser pulses is shown in Fig. 2(a). It indicates an emission bandwidth (FWHM) of 35 nm at a central wavelength of 1070 nm by assuming a $sech^2$-shaped spectral profile. The recorded second-harmonic generation (SHG) based background-free intensity autocorrelation trace gave a deconvolved pulse duration of 39 fs (FWHM) assuming a $sech^2$-shaped temporal pulse profile, see Fig. 2(b). The corresponding time-bandwidth product (TBP) of 0.357 was slightly above the Fourier-transform limit for soliton pulses (0.315). A long-scale SHG-based background-free intensity autocorrelation scan (50 ps) confirmed the single-pulse steady-state mode-locking, see the inset in Fig. 2(b). These results were achieved at an absorbed pump power of 842 mW, corresponding to an optical efficiency of 13.8%. The calculated peak output power reached 39.5 kW.

A radio-frequency (RF) spectrum analyzer was used for confirming the stability of the KLM operation. The relatively high extinction ratio of >74 dBc above the noise level for the fundamental beat note at 66.48 MHz in combination with the uniform harmonics recorded on a

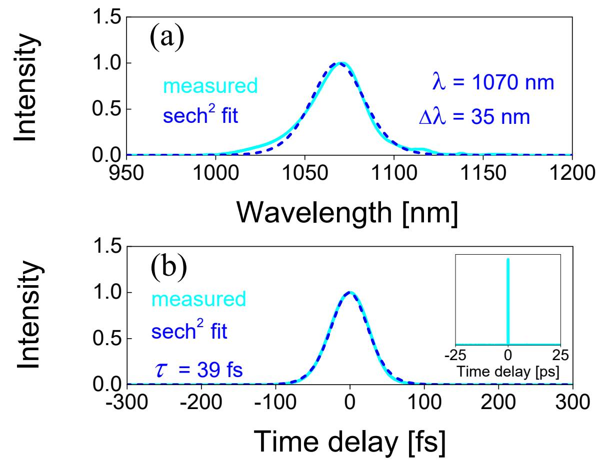

Fig. 2. Diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser with T_{OC} = 4%. (a) Optical spectrum; (b) background-free SHG-based intensity autocorrelation trace. Inset: autocorrelation trace measured on a time span of 50 ps.

1-GHz frequency span are evidence of highly stable CW-ML operation without any Q-switching or multi-pulsing instabilities, see Fig. 3.

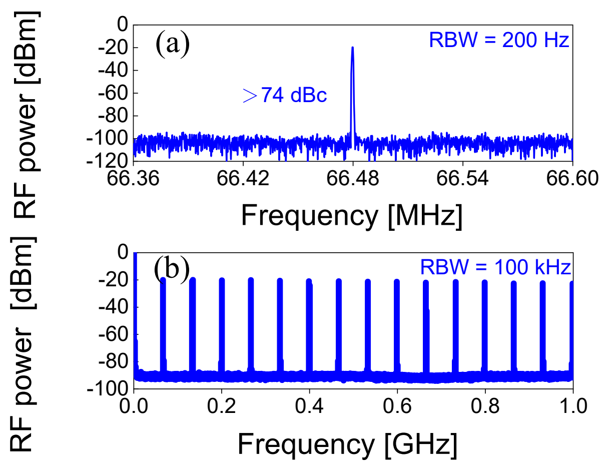

Fig. 3. RF spectra of the diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser with T_{OC} = 4%: (a) Fundamental beat note at 66.48 MHz recorded with a resolution bandwidth (RBW) of 200 Hz, and (b) harmonics on a 1-GHz frequency span recorded with a RBW of 100 kHz.

The shortest pulses with ultimate stability were achieved by reducing the output coupling transmission down to 1.6% for a round-trip negative GDD of -1250 fs^2 provided by three DMs (DM_1 = -150 fs^2, DM_3 = -150 fs^2 and DM_4 = -50 fs^2). Their characterization is shown in Fig. 4. The measured laser spectrum exhibited a broader bandwidth of 53.1 nm at a central wavelength of 1080 nm by assuming a sech^2-shaped spectral profile, as shown in Fig. 4(a). The satellite peaks observed around 1.15 and 1.2 µm are attributed to the unmanageable intracavity GDD at the long-wavelength spectral edge and the non-optimized spectral reflectivity of the cavity mirrors. This phenomenon has previously been observed in Yb:CALGO [25] and explained by F. Druon *et al.* [9]. Optimizing the total intracavity GDD across the full spectral bandwidth of

Optics EXPRESS

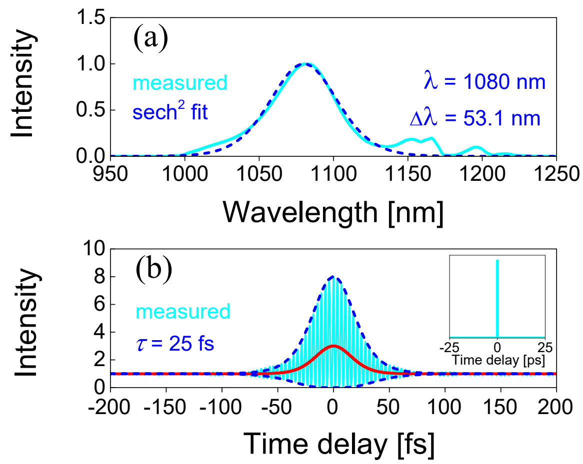

Fig. 4. Diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser with $T_{OC} = 1.6\%$. (a) Optical spectrum. (b) Interferometric autocorrelation trace, the red curve in the autocorrelation corresponds to the intensity autocorrelation profile. Inset: autocorrelation trace measured on a time span of 50 ps.

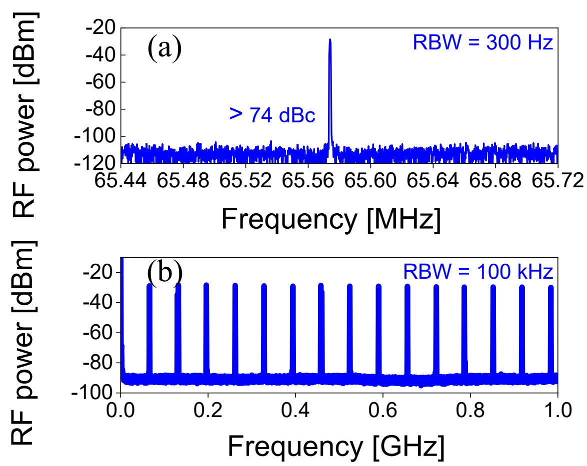

Fig. 5. RF spectra of the diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser with $T_{OC} = 1.6\%$: (a) Fundamental beat note at 65.57 MHz recorded with a resolution bandwidth (RBW) of 300 Hz, and (b) harmonics on a 1-GHz frequency span recorded with a RBW of 100 kHz.

the mode-locked laser could improve this issue. The corresponding pulse duration estimated from the recorded interferometric autocorrelation trace was as short as 25 fs (~7 optical cycles) with a sech^2-shaped temporal profile. The corresponding TBP amounted to 0.341 being slightly above the Fourier-transform limit. The inset in Fig. 4(b) shows the measured SHG-based background-free intensity autocorrelation trace with 50 ps time span indicating single-pulse CW-ML operation free of multiple pulse instabilities. The average output power for the shortest pulses amounted to 47 mW at an absorbed pump power of 836 mW, corresponding to an optical efficiency of 5.6% and a peak power of 25.2 kW. This corresponds to a pump absorption of about 63%, similar to the case of $T_{OC} = 4\%$.

The RF spectra of the shortest pulses were recorded to verify the ultimate stability of the ML operation in different frequency span ranges, as shown in Fig. 5. The recorded first beat

Optics EXPRESS

note located at 65.57 MHz exhibited a similar extinction ratio of >74 dBc above carrier and the harmonics were similarly uniform as recorded on the 1-GHz frequency span.

High peak laser intensities in the laser crystal are expected to introduce a noticeable change of the spatial profile of the laser beam owing to the Kerr self-focusing. This was confirmed by monitoring the far-field beam profiles both for the CW and KLM regimes with an IR camera placed at ~0.8 m from the OC (T_{OC} = 1.6%). The transition from CW to dominating soft-aperture KLM was accompanied by a significant shrinking of the beam diameter from 2.51 mm (x) × 2.20 mm (y) to 2.14 mm (x) × 1.94 mm (y), as shown in Fig. 6. The measured beam propagation factors (M^2) of the shortest pulses were 1.03 × 1.05.

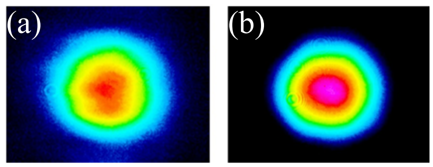

Fig. 6. Measured far-field beam profiles of the diode-pumped KLM Yb:Ca(Gd,Y)AlO_4 laser with T_{OC} = 1.6%: (a) CW and (b) KLM regimes of operation.

4. Conclusion

In summary, we have successfully demonstrated the first KLM operation of an Yb laser employing a "mixed" calcium gadolinium-yttrium aluminate crystal as a gain medium. Using a single-transverse mode, fiber-coupled laser diode at 976 nm as a pump source, 25 fs pulses were directly generated from the Yb:Ca(Gd,Y)AlO_4 laser via soft-aperture Kerr-lens mode-locking. At the expense of a slightly longer pulse duration (39 fs), the average output power reached 116 mW using an output coupler with a higher transmission. The present laser results together with previously reported spectroscopic findings indicate that "mixed" calcium rare-earth aluminates could potentially outperform Yb:CALGO and Yb:CALYO for generating few-optical cycle pulses at 1 µm. By optimizing the doping concentration of Yb^{3+} ions and using high-power Yb-fiber lasers as pump sources, sub-20 fs pulses with more than 100 mW average output powers could be expected.

Funding. National Natural Science Foundation of China (62675263, 62375106); Project of Science and Technology of Fujian Province (2023H0047); Sino-German Scientist Cooperation and Exchanges Mobility Program (M-0040); Ministerio de Ciencia, Innovación y Universidades (PID2022-141499OB-10).

Acknowledgment. Xavier Mateos acknowledges the Serra Húnter program.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

References

1. W. Tian, X. Tian, Q. Li, *et al.*, "Kerr-lens mode-locked femtosecond Yb:CALYO oscillator with more than 20-W average power," Opt. Lett. **48**(18), 4789–4792 (2023).
2. D. Y. Kim, B. J. Park, S. Y. Lee, *et al.*, "High-power 50 fs Kerr-lens mode-locked Yb:CALGO oscillator," Opt. Laser Technol. **159**, 109019 (2023).
3. X. Meng, X. Ban, C. Lv, *et al.*, "95 fs pulses with 1.21 MW peak power from diode-pumped ultrafast Yb:CaGdAlO$_4$ laser using an additional Kerr medium," Opt. Commun. **498**, 127246 (2021).
4. W. Tian, C. Yu, J. Zhu, *et al.*, "Diode-pumped high-power sub-100 fs Kerr-lens mode-locked Yb:CaYAlO$_4$ laser with 1.85 MW peak power," Opt. Express **27**(15), 21448–21454 (2019).
5. W. Tian, G. Wang, D. Zhang, *et al.*, "Sub-40-fs high-power Yb:CALYO laser pumped by single-mode fiber laser," High Power Laser Sci. Eng. **7**, e64 (2019).
6. W. Tian, R. Xu, L. Zheng, *et al.*, "10-W-scale Kerr-lens mode-locked Yb:CALYO laser with sub-100-fs pulses," Opt. Lett. **46**(6), 1297–1300 (2021).

Optics EXPRESS

7. P. Loiko, J. M. Serres, X. Mateos, *et al.*, "Microchip Yb:$CaLnAlO_4$ lasers with up to 91% slope efficiency," Opt. Lett. **42**(13), 2431–2434 (2017).
8. S. Kimura, S. Tani, and Y. Kobayashi, "Raman-assisted broadband mode-locked laser," Sci. Rep. **9**(1), 3738 (2019).
9. P. Sévillano, P. Georges, F. Druon, *et al.*, "32-fs Kerr-lens mode-locked Yb:$CaGdAlO_4$ oscillator optically pumped by a bright fiber laser," Opt. Lett. **39**(20), 6001–6004 (2014).
10. M. Hamrouni, F. Labaye, N. Modsching, *et al.*, "Efficient high-power sub-50-fs gigahertz repetition rate diode-pumped solid-state laser," Opt. Express **30**(17), 30012–30019 (2022).
11. P. Loiko, F. Druon, P. Georges, *et al.*, "Thermo-optic characterization of Yb:$CaGdAlO_4$ laser crystal," Opt. Mater. Express **4**(11), 2241–2249 (2014).
12. Y. Wang, X. Su, Y. Xie, *et al.*, "17.8 fs broadband Kerr-lens mode-locked Yb:CALGO oscillator," Opt. Lett. **46**(8), 1892–1895 (2021).
13. J. Ma, F. Yang, W. Gao, *et al.*, "Sub-five-optical-cycle pulse generation from a Kerr-lens mode-locked Yb:$CaYAlO_4$ laser," Opt. Lett. **46**(10), 2328–2331 (2021).
14. F. Druon, M. Olivier, A. Jaffrès, *et al.*, "Magic mode switching in Yb:$CaGdAlO_4$ laser under high pump power," Opt. Lett. **38**(20), 4138–4141 (2013).
15. S. Ricaud, A. Jaffres, K. Wentsch, *et al.*, "Femtosecond Yb:$CaGdAlO_4$ thin-disk oscillator," Opt. Lett. **37**(19), 3984–3986 (2012).
16. F. Labaye, V. J. Wittwer, M. Hamrouni, *et al.*, "Efficient few-cycle Yb-doped laser oscillator with Watt-level average power," Opt. Express **30**(2), 2528–2538 (2022).
17. Z. Wang, J. Liu, P. Chen, *et al.*, "Growth and characterization of Yb:CALYGLO crystal for ultrashort pulse laser applications," Crystals **14**(2), 120 (2024).
18. Q. Hu, Z. Jia, A. Volpi, *et al.*, "Crystal growth and spectral broadening of a promising Yb:$CaLu_xGd_{1-x}AlO_4$ disordered crystal for ultrafast laser application," CrystEngComm **19**(12), 1643–1647 (2017).
19. Q. Hu, X. Su, Y. Wang, *et al.*, "Spectroscopic properties and ultrafast performance of Yb:$CaLu_xGd_{1-x}AlO_4$ crystal," Laser Phys. Lett. **14**(4), 045809 (2017).
20. P. Zhang, W. Wu, Z.-L. Lin, *et al.*, "Growth, spectroscopy and SESAM mode-locking of a "mixed" Yb:Ca(Gd,Y)AlO4 disordered crystal," Opt. Express **32**(3), 3221–3233 (2024).
21. D. H. Sutter, L. Gallmann, N. Matuschek, *et al.*, "Sub-6-fs pulses from a SESAM-assisted Kerr-lens modelocked Ti:sapphire laser: at the frontiers of ultrashort pulse generation," Appl. Phys. B: Lasers Opt. **70**(S1), S5–S12 (2000).
22. U. Morgner, F. X. Kärtner, S.-H. Cho, *et al.*, "Sub-two-cycle pulses from a Kerr-lens mode-locked Ti:sapphire laser," Opt. Lett. **24**(6), 411–413 (1999).
23. D. H. Sutter, G. Steinmeyer, L. Gallmann, *et al.*, "Semiconductor saturable-absorber mirror–assisted Kerr-lens mode-locked Ti: sapphire laser producing pulses in the two-cycle regime," Opt. Lett. **24**(9), 631–633 (1999).
24. P. Loiko, P. Becker, L. Bohatý, *et al.*, "Sellmeier equations, group velocity dispersion, and thermo-optic dispersion formulas for $CaLnAlO_4$ (Ln = Y, Gd) laser host crystals," Opt. Lett. **42**(12), 2275–2278 (2017).
25. Y. Zaouter, J. Didierjean, F. Balembois, *et al.*, "47-fs diode-pumped Yb^{3+}:$CaGdAlO_4$ laser," Opt. Lett. **31**(1), 119–121 (2006).