Kerr-lens mode-locked ytterbium-activated orthoaluminate laser

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We report on the first Kerr-lens mode-locked laser based on an Yb^{3+}-doped perovskite-type orthoaluminate crystal exploiting two different principal light polarizations. The Yb:(Y,Gd)AlO_3 laser delivers soliton pulses as short as 32 fs at 1067 nm with an average output power of 328 mW, a pulse repetition rate of ~84.6 MHz for $E \,||\, a$ polarization. For the orthogonal $E \,||\, b$ polarization, 33-fs pulses are generated at 1057 nm with an average output power of 305 mW. Power scaling to a maximum average output power reaching 2.07 W is achieved at the expense of longer pulse duration (72 fs for $E \,||\, b$), corresponding to an optical efficiency of 43.9% and a peak power of 303 kW.

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Mode-locked (ML) solid-state Yb-lasers emitting few-optical-cycle pulses in the spectral range of ~1 μm represent a hot topic in the field of ultrafast optics. Compared to ML Ti:sapphire lasers operating near 800 nm, they feature the following advantages: i) extremely low quantum defect resulting in very high optical efficiency and reduced thermal issues; ii) power-scalable operation using cost-effective and commercially available high-power fiber-coupled InGaAs laser diodes as pump sources; and iii) compact configurations suitable for robust integration [1].

Recently, efforts towards exploiting the broadband gain spectra of Yb^{3+}-doped disordered crystals for few-optical-cycle pulse generation resulted in entering the sub-20 fs time domain. Y. Wang *et al.* passed this mark for the first time in 2021 using Yb:$CaGdAlO_4$ aluminate as a gain medium and generating 17.8 fs pulses at 1118 nm with an average output power of only 26 mW (optical efficiency $\eta_{opt} = 3.3\%$) via Kerr-lens mode-locking (KLM) [2]. Subsequently, 17 fs pulses at 1080 nm were emitted from a similar KLM laser based on a isostructural aluminate crystal, Yb:$CaYAlO_4$, albeit with ye lower average output power of 18 mW and η_{opt} of only 0.47% [3]. These results indicate quite clearly the existing trade-off between ultimate pulse shortening and output power scalability. To overcome this limitation, a pump scheme allowing one to reduce the quantum defect was employed in a KLM Yb:$CaGdAlO_4$ laser delivering soliton pulses as short as 22 fs at 1040.7 nm with an average output power of 729 mW and a reasonably high $\eta_{opt} = 25\%$ [4]. Ordered host crystals normally possess higher thermal conductivity and are more suited for high output powers/optical efficiencies but classical crystals such as YAG do not offer such broad spectral gain. For instance, a diode-pumped KLM Yb:YAG laser generating 35 fs pulses at 1060 nm with an average output power of 107 mW was demonstrated in 2011 [5]. A shorter pulse duration of 27 fs at 1028 nm for an average output power of 3.3 W was obtained from a KLM thin-disk Yb:YAG laser albeit at an optical efficiency of only 1.05% [6].

Ytterbium (Yb^{3+})-doped yttrium orthoaluminate, Yb:$YAlO_3$ (shortly Yb:YAP), is a structurally ordered laser crystal belonging to the orthorhombic class (sp. gr. P_{nma}, perovskite-type structure). It is very promising for the development of high-power 1 μm lasers due to a combination of excellent thermo-mechanical and spectroscopic properties [7-10]. In particular, it possesses relatively high thermal conductivity, $\kappa_a = 7.1$, $\kappa_b = 8.3$ and $\kappa_c = 7.6$ $Wm^{-1}K^{-1}$ (values along the crystallographic axes for 5 at.% Yb^{3+} doping) [11], which is beneficial for power scalable operation under pumping with commercial high-power InGaAs laser diodes emitting at ~0.98 μm. YAP is optically biaxial and its intrinsic birefringence together with the polarization anisotropy of the Yb^{3+} ion emission properties leads to a linearly polarized laser emission, suppressing thermally induced depolarization losses inherent to cubic laser crystals such as Yb:YAG. The first investigation on its passive mode-locking was reported in 2008: using a SEmiconductor Saturable Absorber Mirror (SESAM), 225 fs pulses were generated at 1041 nm with an average output power of 0.8 W [12]. Subsequently, the same group reported on power scaling of the SESAM ML Yb:YAlO$_3$ laser using a novel off-axis pumping scheme yielding 140 fs pulses at 1009.7 nm with an average output power of 4 W [7]. However, no sub-100 fs ML Yb:YAP laser operation has been reported, yet.

Very recently, we developed a Yb^{3+}-doped yttrium-gadolinium "mixed" orthoaluminate crystal exhibiting compositional disorder, i.e., Yb:Y$_{1-x}$Gd$_x$AlO$_3$ or Yb:(Y,Gd)AlO$_3$ [13]. The estimated mean thermal conductivity of this material is ~6.3 Wm^{-1}K^{-1} [14, 15], indicating a high potential for power scalable laser operation. By implementing a SESAM for starting and stabilizing the soliton pulse shaping, we achieved 43 fs pulses from a diode-pumped Yb:(Y,Gd)AlO$_3$ laser [15]. The superior thermo-optical properties and preliminary mode-locking results motivated us to further explore the potential of the Yb:(Y,Gd)AlO$_3$ crystal for few-optical-cycle generation via soft-aperture KLM.

A high-quality Yb^{3+}-doped Y$_{0.85}$Gd$_{0.10}$Yb$_{0.05}$AlO$_3$ crystal with an Yb^{3+} doping level of 5.65 at.% was grown by the conventional Czochralski method. The laser operation was investigated using an X-shaped astigmatically compensated standing-wave cavity. The schematic of the laser cavity and pumping geometry is presented in Fig. 1.

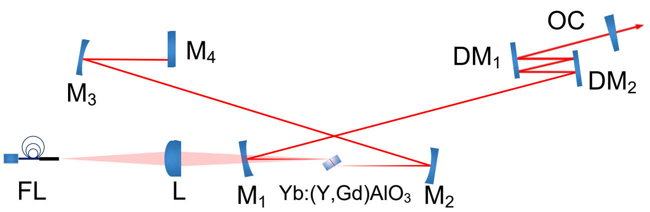

Fig. 1. Schematic of the Yb:(Y,Gd)AlO$_3$ laser. FL: fiber laser; L: spherical focusing lens (f = 75 mm); M$_1$ – M$_3$, dichroic concave mirrors; M$_4$: flat rear mirrors; DM$_1$ and DM$_2$: dispersive mirrors; OC: output coupler.

An uncoated 3-mm thick laser crystal was cut from the as-grown bulk, oriented for light propagation along the crystallographic $\boldsymbol{c}$-axis ($\boldsymbol{c}$-cut) with an aperture of 4 ($\boldsymbol{a}$) × 4 ($\boldsymbol{b}$) mm^2 (sp. gr. P_{nma}). It was mounted in a water-cooled copper holder (coolant temperature: 19°C) and placed at Brewster's angle between two concave dichroic folding mirrors M$_1$ and M$_2$ (radius of curvature: RoC = -100 mm) with the minimum loss condition fulfilled for both the pump and laser wavelengths and polarizations. One arm of the laser cavity was completed by an additional concave mirror M$_3$ (RoC = -100 mm) and a flat rear mirror M$_4$. The other cavity arm was terminated by a pair of flat dispersive mirrors (DMs) and a flat-wedged output coupler (OC). DM$_1$ and DM$_2$ were implemented to compensate the intracavity group delay dispersion (GDD) and balance the self-phase modulation (SPM) induced by the Kerr nonlinearity of the laser crystal for soliton pulse shaping. All DMs had a negative GDD of -200 fs^2 per bounce. A continuous-wave, spatially single-mode fiber laser emitting a nearly diffraction-limited (M^2 of ~1.04) linearly polarized beam was used as a pump source. Its emission wavelength was locked at 979 nm with a spectral linewidth (full width at half maximum, FWHM) of ~0.1 nm. The pump beam was focused into the laser crystal by a spherical lens (f = 75 mm) yielding a beam waist radius of 15.4 µm × 28.6 µm in the sagittal and tangential planes, respectively. By rotating the crystal, we were able to set the laser polarization ($\boldsymbol{E}\,||\,\boldsymbol{a}$ or $\boldsymbol{E}\,||\,\boldsymbol{b}$).

The Yb:(Y,Gd)AlO$_3$ laser was initially characterized for light polarization $\boldsymbol{E}\,||\,\boldsymbol{b}$ corresponding to stronger pump absorption (>99% in ML operation) and an OC transmittance (T_{OC}) of 4.5%. The KLM regime was investigated at an incident pump power of 3.05 W by applying four bounces (single pass) on the flat DMs giving a round-trip GDD of -1600 fs^2. In order to discriminate the CW and KLM regimes, the laser cavity was aligned close to the edge of the stability region through translating the folding mirror (M$_2$) by several hundreds of micrometers away from the pump mirror (M$_1$), which resulted in a significant drop of the CW output power. KLM operation could be initiated by a slight knock on the OC or translating the flat rear mirror M$_4$. This led to an abrupt increase of the output power from 0.535 to 1 W. The spectrum of the femtosecond pulses was centered at 1053.2 nm with a FWHM of 23.2 nm by assuming a sech^2-shaped spectral profile, see Fig. 2(a).

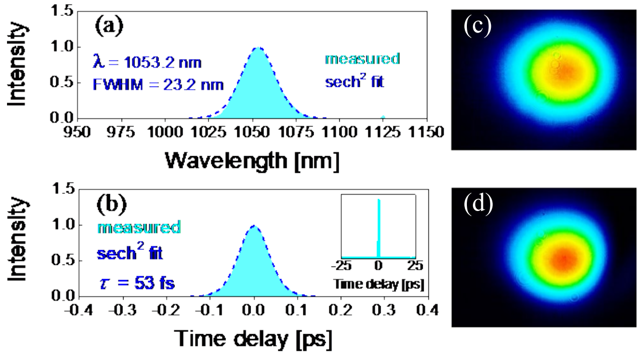

Fig. 2 KLM Yb:(Y,Gd)AlO$_3$ laser with T_{OC} = 4.5% ($\boldsymbol{E}\,||\,\boldsymbol{b}$): (a) Optical spectrum and (b) autocorrelation trace. *Inset* in (b): simultaneously measured long-scale (50 ps) autocorrelation trace. (c,d) Far-field beam profiles: transition from (c) CW to (d) KLM regime.

The recorded background-free intensity autocorrelation trace can be almost perfectly fitted with a sech^2-shaped temporal profile, giving an estimated pulse duration of 53 fs (FWHM), see Fig. 2(b). The corresponding time-bandwidth product (TBP) of 0.332 was only slightly above the Fourier-transform-limited value (0.315). A long-scale intensity autocorrelation scan of 50 ps confirmed single-pulse steady-state ML, see the inset in Fig. 2(b). The total cavity length was ~1.8 m, resulting in a pulse repetition rate of ~83.2 MHz. The peak output power reached ~200 kW. The calculated peak on-axis intracavity intensity in the laser crystal was ~1.23 TW/cm^2. Such a high intensity should lead to a noticeable modification of the spatial mode. This was indeed confirmed by monitoring the far-field beam profiles with a camera placed at ~1.6 m away from the OC. The shrinking of the far-field beam diameter from 2.53 × 2.23 mm^2 to 2.17 × 2.01 mm^2, as shown in Fig. 2(c) and (d), indicated a strong soft-aperture Kerr-lens effect and self-focusing inside the laser crystal.

The pulse duration could be reduced by applying lower T_{OC} of 2.5% while maintaining the same total round-trip negative GDD. Notably shorter pulses of 38 fs were obtained at 1056 nm with a ~31 nm spectral FWHM. In this case, the corresponding TBP was 0.317, again very close to the Fourier-transform-limit. The average output power was 0.763 W (peak power: ~212.1 kW) at a pulse repetition rate of ~83.3 MHz, yielding a peak on-axis laser intensity in the laser crystal of ~2.37 TW/cm^2. The shortest pulse duration for laser polarization $\boldsymbol{E}\,||\,\boldsymbol{b}$ was achieved by further reducing the transmittance of the OC to 1.6%. Soliton pulses as short as 33 fs were generated at 1057 nm with a spectral FWHM of 36 nm, as shown in Fig. 3. The TBP was 0.319, only slightly above the Fourier limit for sech^2-shaped pulses. The peak output power of the laser pulses was 97.8 kW corresponding to an estimated peak on-axis laser intensity of ~1.72 TW/cm^2 in the crystal.

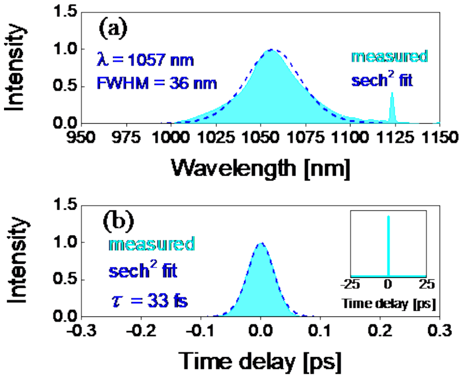

Fig. 3. KLM Yb:(Y,Gd)AlO$_3$ laser with T_{OC} = 1.6% (***E*** || ***b***): (a) Optical spectrum and (b) autocorrelation trace. *Inset* in (b): long-scale (50 ps) autocorrelation trace.

A maximum output power of 305 mW at ~83.2 MHz was obtained for an incident pump power of 3.1 W, corresponding to an optical efficiency of 9.84%. A sharp satellite peak is observed at 1123 nm, which originates from the unmanaged intracavity GDD at the long-wave spectral wing and the non-optimized spectral reflectivity of the cavity mirrors. This phenomenon has been already observed in ML Yb:CaGaAlO$_4$ lasers [16], and explained by F. Druon *et al.* [17]. It could be improved by properly managing the total intracavity GDD over the full spectral band of the ML laser.

Table 1. Characteristics^a of the KLM Yb:(Y,Gd)AlO$_3$ laser *vs* Output Coupling for Laser Polarization *E* || *b*

T_{OC} [%]	P_{pump} [W]	P_{out} [W]	η_{opt} [%]	λ_c [nm]	$\Delta\lambda$ [nm]	$\Delta\tau$ [fs]
1.6	3.1	0.305	9.84	1057	36	33
2.5	3.2	0.763	23.8	1056	30.9	38
4.5	3.05	1	32.8	1053.2	23.2	53
7.5	3.9	1.4	35.9	1053.5	20.7	58
10	4.72	2.07	43.9	1050.1	16	72

$^a T_{OC}$ – output coupler transmittance, P_{pump} – incident pump power, P_{out} – average output power, η_{opt} – optical efficiency refers to incident pump power, λ_c – central laser wavelength, $\Delta\lambda$ – spectral bandwidth (FWHM), $\Delta\tau$ – pulse duration (FWHM).

Power scalable operation for the laser polarization ***E*** || ***b*** was investigated by increasing the output coupling to T_{OC} = 7.5% and 10%. The pump power was adjusted for achieving single-pulse mode-locking with the highest average output power, as well as the highest optical efficiency. Table 1 summarizes the results for various T_{OC} values. The average output power of the KLM Yb:(Y,Gd)AlO$_3$ laser was easily scaled by cranking up the pump power at the expense of the pulse duration. The maximum average output power reached 2.07 W at an incident pump power of 4.72 W. This corresponded to a longer pulse duration of 72 fs, still in the sub-100 fs time range, and a relatively high optical efficiency of 43.9%. The spectrum of the ML laser experienced a slight blue-shift to 1053.5 nm (T_{OC} = 7.5%) and 1050.1 nm (T_{OC} = 10%), as can be expected for a three-level laser system with reabsorption.

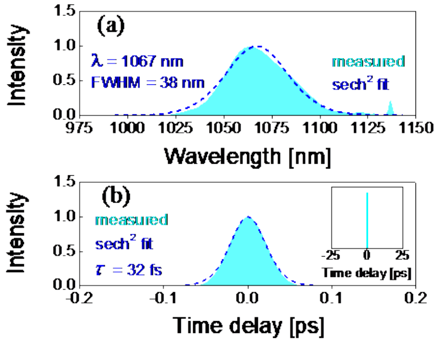

Fig. 4. KLM Yb:(Y,Gd)AlO$_3$ laser with T_{OC} = 1.6% (***E*** || ***a***): (a) Optical spectrum and (b) autocorrelation trace. *Inset* in (b): long-scale (50 ps) autocorrelation trace.

The KLM operation of the Yb:(Y,Gd)AlO$_3$ laser was also studied for the other available light polarization, ***E*** || ***a***, characterized by lower pump absorption (<75% in ML operation). As expected, the shortest pulses with ultimate mode-locking stability were achieved with the 1.6% OC. A maximum average output power of 328 mW was obtained at an incident pump power of 3.75 W and a pulse repetition rate of 84.6 MHz. This corresponded to an optical efficiency of 8.7%. The pulse characteristics are shown in Fig. 4. The laser spectrum had a FWHM of 38 nm at 1067 nm by assuming a sech^2-shaped spectral profile, see Fig. 4(a). The pulse duration was assessed from the recorded background-free intensity autocorrelation trace which is shown in Fig. 4(b). The curve can be almost perfectly fitted with a sech^2-shaped temporal profile yielding a deconvolved FWHM of 32 fs. The TBP was thus 0.320, slightly above the Fourier-transform-limit. A longer time window (50-ps) autocorrelation scan confirmed single-pulse mode-locking without post- or pre-pulses, see the inset in Fig. 4(b). The peak output power reached ~106.6 kW and the estimated peak on-axis laser intensity in the crystal amounted to ~1.87 TW/cm^2.

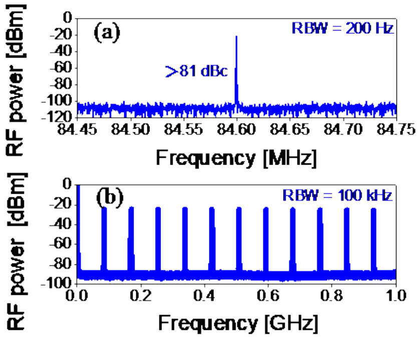

Fig. 5. RF spectra of the KLM Yb:(Y,Gd)AlO$_3$ laser (***E*** || ***a***): (a) Fundamental beat note, RBW = 200 Hz; (b) 1-GHz span with RBW of 100 kHz. RBW: resolution bandwidth.

Figure 5 shows the recorded radio-frequency (RF) spectrum of the steady-state pulse train of the shortest pulses for light polarization $\boldsymbol{E}\,||\,\boldsymbol{a}$. The recorded narrow-band fundamental beat note exhibited a very high extinction ratio of 81 dBc above carrier at 84.6 MHz, see Fig. 5(a). This, together with the uniform harmonic beat notes on a 1-GHz frequency span indicates highly stable KLM operation without any Q-switching instabilities or multi-pulsing, see Fig. 5(b).

Power scaling of the KLM laser for light polarization $\boldsymbol{E}\,||\,\boldsymbol{a}$ was also investigated by increasing the output coupling in the range of $T_{\rm OC}$ = 2.5% - 7.5%, cf. Table 2 summarizing the achieved output characteristics. Due to the relatively low pump absorption compared to $\boldsymbol{E}\,||\,\boldsymbol{b}$, the maximum incident pump power reached 6 W for achieving single-pulse mode locking with higher average output power and optical efficiency. The maximum average output power reached 1.72 W at an incident pump power of 6 W for a pulse duration of 51 fs. This, corresponded to a lower optical conversion of 28.7% compared to $\boldsymbol{E}\,||\,\boldsymbol{b}$.

Table 2. Characteristics[a] of the KLM Yb:(Y,Gd)AlO$_3$ laser *vs* Output Coupling for Laser Polarization $\boldsymbol{E}\,||\,\boldsymbol{a}$

$T_{\rm OC}$ [%]	$P_{\rm pump}$ [W]	$P_{\rm out}$ [W]	$\eta_{\rm opt}$ [%]	$\lambda_{\rm c}$ [nm]	$\Delta\lambda$ [nm]	$\Delta\tau$ [fs]
1.6	3.75	0.328	8.7	1067	38	32
2.5	3.3	0.498	15.1	1065.3	33.3	37
4.5	6	1.24	20.7	1053.5	25.3	50
7.5	6	1.72	28.7	1051.8	22.9	51

[a] $T_{\rm OC}$ – output coupler transmittance, $P_{\rm pump}$ – incident pump power, $P_{\rm out}$ – average output power, $\eta_{\rm opt}$ – optical efficiency refers to incident pump power, $\lambda_{\rm c}$ – central laser wavelength, $\Delta\lambda$ – spectral bandwidth (FWHM), $\Delta\tau$ – pulse duration (FWHM).

To conclude, "mixed" ytterbium-doped yttrium-gadolinium orthoaluminate crystal, Yb:(Y,Gd)AlO$_3$, with compositional disorder, represents an attractive laser material for generation of ultrashort (sub-100 fs) pulses with moderate to high average output powers (up to the watt-level) from Kerr-lens mode-locked solid-state lasers. Owing to its good thermal properties, as well as intense and broad emission bands for polarized light, two regimes of KLM operation were successfully exploited: i) targeting the shortest pulse duration, 32 fs pulses were generated at 1067 nm with an average output power of 328 mW at a repetition rate of 84.6 MHz for light polarization $\boldsymbol{E}\,||\,\boldsymbol{a}$ using low output coupling ($T_{\rm OC}$ = 1.6%), ii) targeting the maximum average output power and laser efficiency, the KLM laser delivered 2.07 W at 1050.1 nm at the expense of longer pulse duration (72 fs) corresponding to an optical efficiency of 43.9% for light polarization $\boldsymbol{E}\,||\,\boldsymbol{b}$ employing high output coupling ($T_{\rm OC}$ = 10%). To the best of our knowledge, this is the first demonstration of Kerr-lens mode-locking employing Yb^{3+}-doped orthorhombic perovskite-type aluminate crystals. Our results pave the way towards the development of high-power (>10 W) KLM ytterbium orthoaluminate lasers pumped by cost-efficient spatially multi-mode InGaAs laser diodes. Heavily-doped Yb:YAlO$_3$-type crystals also look promising for KLM thin-disk lasers at ~1 µm

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