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24 fs Kerr-lens mode-locked Yb:YAlO₃ laser

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We report on a Kerr-lens mode-locked Yb:YAlO₃ laser generating soliton pulses as short as 24 fs at 1085 nm with an average output power of 186 mW and a pulse repetition rate of 87.5 MHz representing the shortest pulses ever achieved from any mode-locked laser based on Yb³⁺-doped structurally ordered crystal. Optimized for power scalable operation, the Yb:YAlO₃ laser delivers 1.9 W at 1060 nm at the expense of longer pulse duration of 44 fs, corresponding to a peak power of 462 kW and an optical efficiency of 43.2%. © 2022 Optical Society of America.

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Recent progress in Kerr-lens mode-locked (KLM) ytterbium (Yb³⁺) solid-state lasers has led to routine generation of sub-30 fs pulses in the spectral range of ~1 μm [1-6]. Among the crystalline laser gain media exploited for the design of ultrafast KLM Yb lasers, the milestones in terms of shortening the pulse duration have been achieved using materials with a structure disorder. Such disordered crystals usually feature a significant inhomogeneous broadening of the spectral bands of the dopant Yb³⁺ ions both at low and ambient temperature leading to broad, smooth and relatively flat gain profiles extending well above 1 μm (a property called a “glassy-like” spectroscopic behavior). Such gain spectra can support the generation of sub-20 fs pulses via soft-aperture Kerr-lens mode-locking. In 2021, pulses as short as 17.8 fs were obtained from a diode-pumped KLM Yb:CaGdAlO₄ laser at a central wavelength of 1118 nm with an average output power of only 26 mW [3]. Subsequently, a similar pulse duration of 17 fs was reported employing the isostructural Yb:CaYAlO₄ crystal [4]. By using a cross-polarized pumping which utilizes better the characteristics of the dichroic cavity mirrors, power-scalable operation of a sub-30 fs KLM Yb:CaGdAlO₄ laser was achieved with an average output power of 729 mW and a pulse duration of 22 fs [5]. The tetragonal CaREAlO₄ (RE = Gd, Y) crystals used exhibit a structure disorder due

to a random distribution of Ca²⁺ and RE³⁺ cations over the same lattice sites

A clear advantage of structurally ordered host matrices is the higher thermal conductivity which favors high-power laser operation. However, Yb³⁺ ions in such crystals exhibit narrower gain profiles determined by well-resolved electronic transitions which can hardly support sub-30 fs pulse generation. Featuring excellent thermo-mechanical and thermo-optical properties, Yb³⁺-doped cubic Y₃Al₅O₁₂ (Yb:YAG) is by far the ultimate “working horse” for kW-class high-power lasers emitting at ~1 μm. However, the gain spectrum of Yb:YAG exhibits a relatively narrow peak at 1030 nm with a bandwidth of ~10 nm (full width at half maximum, FWHM) supporting only >100 fs pulse generation. For achieving shorter pulses, special efforts are needed to broaden the Yb³⁺ gain spectrum. A KLM Yb:YAG laser delivered 35 fs pulses at 1060 nm due to an intracavity filter suppressing the above mentioned strong gain peak at 1030 nm [7]. Using a 2-mm thick sapphire plate as a separate Kerr medium to enhance the self-phase modulation (SPM), a KLM Yb:YAG thin-disk laser generated 27 fs pulses at 1028 nm with an average output power of 3.3 W and an optical efficiency of 1.1% [6]. The measured laser spectrum was five times broader than the Yb:YAG gain bandwidth. This result highlights the feasibility of generating sub-30 fs pulses at ~1 μm from ML lasers based on Yb³⁺-doped structurally ordered crystals intrinsically exhibiting narrow gain spectra while operating in a regime of strong SPM induced spectral broadening.

Yb³⁺-doped perovskite-type orthoaluminates, i.e., Yb:REAlO₃ (where RE = Y, Gd or Lu), are structurally ordered laser crystals belonging to the orthorhombic class (sp. gr. *Pnma*). They crystallize in the same binary system (RE₂O₃ – Al₂O₃) as cubic Y₃Al₅O₁₂. The REAlO₃ crystals are optically biaxial and their intrinsic birefringence naturally results in linearly polarized laser emission and weak depolarization losses, which favors high-power laser operation. Recently, we demonstrated sub-50 fs pulse generation using an Yb³⁺-doped “mixed” (Y,Gd)AlO₃ crystal as a gain medium. The motivation for “mixing” the host-forming Gd³⁺ and Y³⁺ cations was to induce an additional spectral broadening for the dopant Yb³⁺ ions

by compositional disorder. Employing a commercial Semiconductor Saturable Absorber Mirror (SESAM), the diode-pumped Yb:(Y,Gd)AlO₃ laser delivered 43 fs soliton pulses at 1053.2 nm with an average output power of 103 mW [8]. Further pulse shortening was achieved with a soft-aperture KLM Yb:(Y,Gd)AlO₃ laser: pulses as short as 32 fs were obtained at 1057 nm with an average output power of 328 mW [9]. Such short pulse durations were achieved owing to the broad and smooth long-wave part of the emission spectrum of Yb³⁺ extending above 1.1 μm due to multiphonon-assisted emission. This spectral selection was achieved by a low level of inversion in the gain medium (leading to strong reabsorption at the wavelengths of electronic transitions corresponding to narrow gain bandwidths) owing to the low passive and outcoupling losses of the cavity.

The passive losses in the gain medium shall be, however, lower in one of the parent compounds, e.g., Yb³⁺-doped yttrium orthoaluminate YAlO₃. Large volume and high optical quality Yb:YAlO₃ crystals can be more easily grown by the Czochralski method. This crystal also exhibits high thermal conductivity ($\kappa_a = 7.1$, $\kappa_b = 8.3$ and $\kappa_c = 7.6$ Wm⁻¹K⁻¹, for 5 at.% Yb³⁺) [10]. The superior thermo-mechanical properties of the Yb:YAlO₃ crystal make it extremely suitable for developing high-power femtosecond ML lasers [11, 12]. In addition, benefiting from the relatively high nonlinear refractive index ($n_2 = \sim 7.3 \times 10^{-20}$ m²/W) of YAlO₃ [13], the Yb:YAlO₃ crystals can be simultaneously used as a laser gain medium and a Kerr-active medium in a KLM laser without using a real saturable absorber.

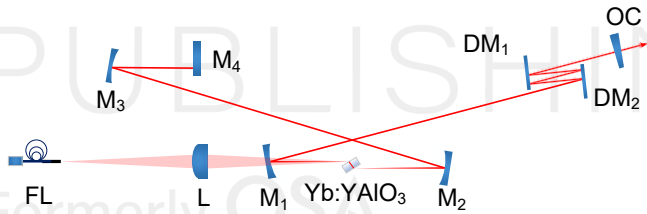


Fig. 1. Schematic of the KLM Yb:YAlO₃ laser. FL: Yb fiber laser; L: spherical focusing lens ($f = 75$ mm); M₁ – M₃, dichroic concave mirrors; M₄: flat rear mirror; DM₁ and DM₂: dispersive mirrors; OC: output coupler.

A 5 at.% Yb³⁺-doped YAlO₃ crystal was cut for light propagation along the crystallographic *c*-axis (*c*-cut) having an aperture of 3(*a*) × 3(*b*) mm² and a thickness of 3 mm. KLM operation of the Yb:YAlO₃ laser was realized using an X-folded astigmatically compensated linear cavity, as shown in Fig. 1. The pump source was a continuous-wave (CW) Yb-fiber laser delivering linearly polarized output at 979 nm with a linewidth of ~ 0.1 nm (FWHM). It provided a nearly diffraction-limited spatial intensity profile with a beam propagation factor (M^2) of ~ 1.04 . The uncoated sample was mounted in a water-cooled copper holder (coolant temperature: 19°C) and placed at Brewster's angle between two concave dichroic folding mirrors M₁ and M₂ (radius of curvature: RoC = -100 mm) with the minimum loss condition fulfilled for both the pump and laser wavelengths. The orientation of the laser crystal was selected to ensure high pump absorption for light polarization $E \parallel b$. The pump radiation was focused into the laser crystal with a spherical focusing lens L (focal length: $f = 75$ mm) resulting in a beam waist (radius) of 15.4×28.6 μm² in the sagittal and tangential planes, respectively. One cavity arm of the Yb:YAlO₃ laser contained an

additional focusing mirror M₃ (RoC = -100 mm) and a flat rear mirror M₄. The other arm contained a pair of flat dispersive mirrors (DMs) and was terminated by a flat-wedged output coupler (OC). The round-trip material dispersion due to the laser crystal was estimated from the dispersion curves of YAlO₃ [14] to be +534 fs² at 1.08 μm for $E \parallel b$, was provided by implementing Two DMs (DM₁ and DM₂) with a group delay dispersion (GDD) of -150 fs² per bounce introduced a roundtrip negative GDD of -1200 fs² as shown in Fig. 1 for soliton pulse shaping.

The Yb:YAlO₃ laser was initially characterized in the CW regime with an OC transmittance (T_{OC}) of 2.5%. The maximum CW output power amounted to 1.36 W at 2.67 W of incident pump power, corresponding to an optical efficiency of 51%. In order to initiate the KLM operation, the laser was aligned close to the edge of the stability region through translating the folding mirror (M₂) by ~ 600 μm away from the pump mirror (M₁). Under this condition, the CW output power of the Yb:YAlO₃ laser significantly dropped down to 348 mW. The KLM operation could be started by slightly knocking one of the cavity mirrors. This led to an abrupt increase of the output power to 670 mW. Once started, the ML laser showed ultimate stability for hours without a hard aperture.

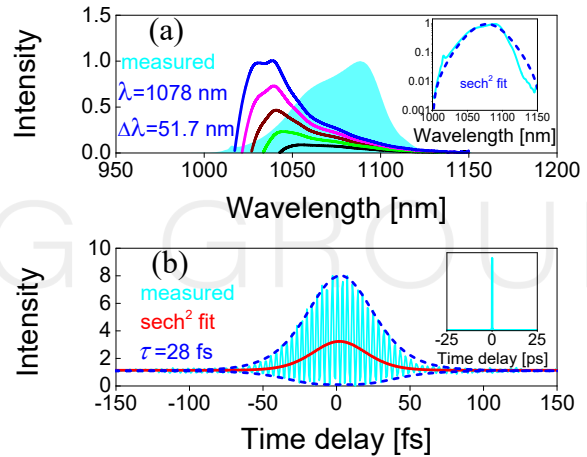


Fig. 2. KLM Yb:YAlO₃ laser with $T_{OC} = 2.5\%$: (a) Optical spectrum plotted together with normalized gain profiles for inversion ratios β in the range from 0.04 to 0.12 for light polarization $E \parallel b$, *Inset*: spectral profile in logarithmic scale with a sech² fit; (b) interferometric autocorrelation trace. *Inset*: simultaneously measured background-free long-scale (50 ps) autocorrelation trace.

The measured optical spectrum of the ML laser plotted both in linear and logarithmic scales is shown in Fig. 2(a). It had a bandwidth (FWHM) of 51.7 nm at a central wavelength of 1078 nm by assuming a sech²-shaped spectral profile. Note that this wavelength is notably longer than the central wavelength of 1056 nm for the KLM Yb:(Y,Gd)AlO₃ laser with the same OC [9]. The measured interferometric autocorrelation trace gave a deconvolved pulse duration of 28 fs (FWHM) by assuming a sech²-shaped temporal profile, see Fig. 2(b). The corresponding time-bandwidth product (TBP) of 0.373 was only slightly above the Fourier-transform-limited value (0.315). A long-scale background-free autocorrelation scan (50 ps) confirmed single-pulse steady-state mode-locking, see the inset in Fig. 2(b). The total cavity length was ~ 1.8 m, yielding a pulse repetition rate of ~ 85.1 MHz. The

normalized gain cross-section spectra of Yb:YAlO₃ for light polarization $E \parallel b$ are also plotted in Fig. 2(a) for inversion ratios $\beta = 0.04, 0.06, 0.08, 0.10, \text{ and } 0.12$. Note that the spectral bandwidth of the laser pulses is notably broader than the gain bandwidth ($\sim 46 \text{ nm}$) of the Yb:YAlO₃ crystal for this polarization. In this configuration, the peak power of the KLM Yb:YAlO₃ laser reached $\sim 247 \text{ kW}$ which yields a relatively high peak on-axis intensity in the crystal of $\sim 761 \text{ GW/cm}^2$. Such a high intracavity intensity in the Kerr medium should indeed lead to a very strong SPM and additional nonlinear spectral broadening exceeding the available gain bandwidth of the laser crystal [6].

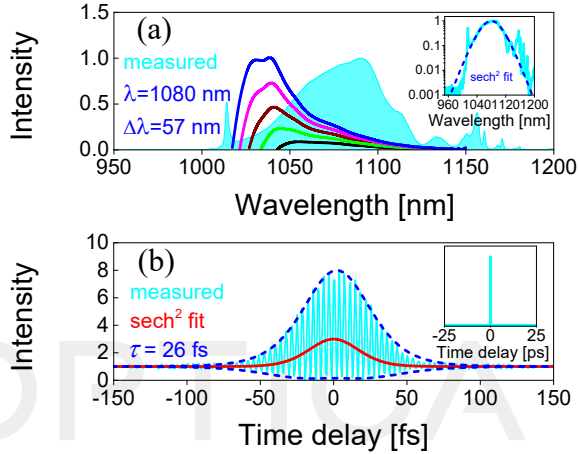


Fig. 3. KLM Yb:YAlO₃ laser with $T_{OC} = 1.6\%$: (a) Optical spectrum plotted together with normalized gain profiles, with inversion ratios $\beta = 0.04 - 0.12, E \parallel b$, *Inset*: spectral profile in logarithmic scale with a sech^2 fit; (b) interferometric autocorrelation trace. *Inset*: simultaneously measured background-free long-scale (50 ps) autocorrelation trace.

The observed spectral broadening beyond the gain bandwidth became more significant when applying lower transmittance OC ($T_{OC} = 1.6\%$) while maintaining the same amount of total round-trip negative GDD. The measured laser spectrum (plotted again both in linear and logarithmic scales and centered at 1080 nm) and the normalized gain profiles of Yb:YAlO₃ are shown in Fig. 3(a). Figure 3(b) shows the recorded interferometric autocorrelation trace which is well fitted with a sech^2 -shaped temporal profile, yielding an estimation of 26 fs for the pulse duration. With a spectral bandwidth of 57 nm (FWHM), the corresponding TBP was 0.345. The inset in Fig. 3(b) shows the measured background-free intensity autocorrelation trace on a long-time span of 50 ps indicating single-pulse steady-state ML operation free of multiple pulse instabilities. The average output power for this OC was 500 mW at an incident pump power of 2.7 W at a pulse repetition rate of $\sim 85.1 \text{ MHz}$. The peak power reached $\sim 199 \text{ kW}$, yielding a peak on-axis intensity in the laser crystal of $\sim 960 \text{ GW/cm}^2$. As mentioned above, the very strong SPM inside the Kerr medium eventually resulted in a significant spectral broadening: the measured laser spectrum exceeded in width the calculated gain profile of the Yb:YAlO₃ crystal and clearly extended towards longer wavelengths even beyond $1.15 \mu\text{m}$ where no gain from purely electronic transitions of the Yb³⁺ ion is expected, see Fig. 3(a).

The shortest pulse duration was achieved by further reducing the

transmittance of the OC to 1%. The pulse characteristics are shown in Fig. 4. The laser spectrum had a FWHM of 63 nm and was centered at 1085 nm, see Fig. 4(a). The pulse duration was assessed from the recorded interferometric autocorrelation trace which is shown in Fig. 4(b). The curve could be almost perfectly fitted with a sech^2 -shaped temporal profile yielding a deconvolved FWHM of 24 fs. The TBP was thus 0.385. A longer scale (50-ps) background-free autocorrelation scan confirmed single-pulse mode-locking without post- or pre-pulses, see the inset in Fig. 4(b).

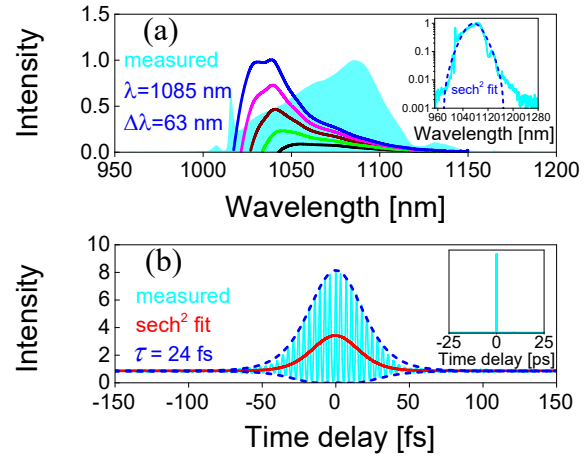


Fig. 4. KLM Yb:YAlO₃ laser with $T_{OC} = 1\%$: (a) Optical spectrum plotted together with normalized gain profiles, $\beta = 0.04 - 0.12, E \parallel b$, *Inset*: spectral profile in logarithmic scale with a sech^2 fit; (b) interferometric autocorrelation trace. *Inset*: simultaneously measured background-free long-scale (50 ps) autocorrelation trace.

The maximum average output power corresponding to the shortest pulses amounted to 186 mW at an incident pump power of 2.55 W for a pulse repetition rate of $\sim 87.5 \text{ MHz}$. The peak output power reached $\sim 77.9 \text{ kW}$ with an estimated peak on-axis intensity in the laser crystal of $\sim 604 \text{ GW/cm}^2$. Such a high intensity should lead to a noticeable modification of the spatial beam profile. This was indeed confirmed by monitoring the far-field beam profiles with a camera placed at $\sim 0.7 \text{ m}$ away from the OC. The transition from CW to dominating soft-aperture KLM, i.e., strong self-focusing inside the Yb:YAlO₃ crystal with a soft-aperture Kerr-lens effect, was accompanied by shrinking of the beam diameter from $2.17 \text{ mm (x)} \times 2.09 \text{ mm (y)}$ to $1.85 \text{ (x)} \text{ mm} \times 1.86 \text{ (y)}$. The measured far-field beam profiles both in CW and the KLM regimes are shown in Fig. 5. The KLM Yb:YAlO₃ laser generating the shortest pulses exhibited a nearly diffraction-limited beam profile with a measured beam propagation factors of $M_x^2 = 1.06$ and $M_y^2 = 1.02$.

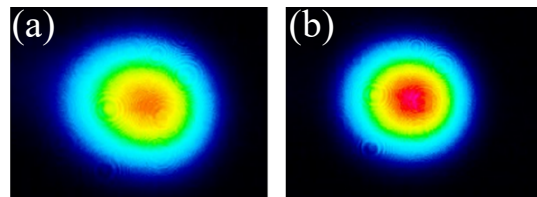


Fig. 5. Measured far-field beam profiles of the KLM Yb:YAlO₃ laser with $T_{OC} = 1\%$: (a) CW and (b) KLM regimes.

Figure 6 shows the recorded radio-frequency (RF) spectrum of the steady-state pulse train of the shortest pulses. The narrow fundamental beat note exhibited a very high extinction ratio of 80 dBc above the carrier at ~ 87.51 MHz, see Fig. 6(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. 6(b).

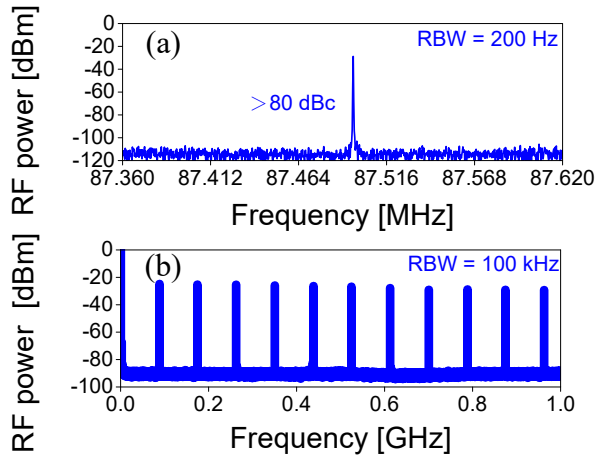


Fig. 6. RF spectra of the KLM Yb:YAlO₃ laser with $T_{OC} = 1\%$: (a) Fundamental beat note, RBW = 200 Hz; (b) 1-GHz span with RBW of 100 kHz. RBW: resolution bandwidth.

The power scaling of the KLM Yb:YAlO₃ laser was investigated by increasing both the output coupling in the range of $T_{OC} = 4.5\% - 10\%$ and the introduced negative GDD, cf. Table 1 summarizing the achieved output characteristics. The total round-trip negative GDD was increased to -1600 fs² (replacing DM₂ with a flat DM₃ with GDD = -200 fs² per bounce) for achieving single-pulse mode locking with higher average output power and optical efficiency. For the highest $T_{OC} = 10\%$ tested, the maximum average output power reached 1.9 W at an incident pump power of 4.4 W for a pulse duration of 44 fs and a pulse repetition rate of ~ 84.4 MHz. This corresponded to a maximum optical efficiency of 43.2%.

Table 1. Characteristics^a of the KLM Yb:YAlO₃ 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]	TBP
1	2.55	0.186	7.4	1085	63	24	0.385
1.6	2.7	0.500	18.5	1080	57	26	0.345
2.5	2.67	0.670	25.1	1078	51.7	28	0.373
4.5	3.2	0.732	22.9	1071	35.6	36	0.335
7.5	3	0.9	30	1061	34.5	37	0.340
10	4.4	1.9	43.2	1060	27.6	44	0.324

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

In conclusion, Yb³⁺-doped yttrium orthoaluminate, i.e., Yb:YAlO₃, represents one of the most remarkable structurally ordered laser crystals which is extremely suitable for developing high-power femtosecond lasers at ~ 1 μ m. Benefiting from its excellent thermo-

mechanical properties, and strongly polarized, broad emission bands, we demonstrate a Kerr-lens mode-locked Yb:YAlO₃ laser generating soliton pulses as short as 24 fs at 1085 nm with an extremely broad spectrum (63 nm, FWHM) exceeding the gain bandwidth of the Yb:YAlO₃ crystal (~ 46 nm) and extending towards longer wavelengths where no gain from electronic transitions of the Yb³⁺ ion is expected. Such nonlinear spectral broadening indicates that the Yb:YAlO₃ laser operated in a strong SPM regime which favors few-optical-cycle pulse generation from passively mode-locked lasers. To the best of our knowledge, this result represents the shortest pulses ever achieved from any Yb³⁺-doped structurally ordered crystal. When employing higher transmission of the output coupler and larger total intracavity negative GDD, the KLM Yb:YAlO₃ laser output power was scaled up to 1.9 W still with sub-50 fs pulses. This result paves the way towards the development of sub-100 fs power scalable mode-locked lasers using Yb:YAlO₃ crystals as a gain medium pumped by cost-efficient spatially multi-mode InGaAs laser diodes in bulk or thin-disk geometries.

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Disclosures. The authors declare no conflicts of interest

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