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Yb-doped mixed sesquioxides for ultrashort pulse generation in the thin disk laser setup

Kolja Beil · Clara J. Saraceno · Cinia Schriber · Florian Emaury · Oliver H. Heckl · Cyrill R. E. Baer · Matthias Golling · Thomas Südmeyer · Ursula Keller · Christian Kränkel · Günter Huber

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Abstract We report on spectroscopic investigations of the mixed sesquioxide laser materials Yb:$LuScO_3$, Yb:$YScO_3$, and Yb:$(Lu,Y,Sc)_2O_3$ as well as mode-locked thin disk laser experiments with Yb:$LuScO_3$ and Yb:$(Lu,Y,Sc)_2O_3$. The disordered crystal structures of these materials result in significantly broader emission spectra than for the pure sesquioxides Yb:Sc_2O_3, Yb:Y_2O_3, and Yb:Lu_2O_3 providing a high potential for ultrashort pulse generation. In SESAM mode-locked thin disk laser experiments, pulse durations of around 100 fs could be obtained exploiting >70 % of the gain bandwidth which is to the best of our knowledge the optimum obtained so far for the mode-locked lasers in this setup.

K. Beil (✉) · C. Kränkel · G. Huber
Institute of Laser-Physics, University of Hamburg,
Luruper Chaussee 149, 22761 Hamburg, Germany
e-mail: kbeil@physnet.uni-hamburg.de

C. J. Saraceno · C. Schriber · F. Emaury · M. Golling · U. Keller
Department of Physics, Institute of Quantum Electronics,
ETH Zurich, 8093 Zurich, Switzerland

O. H. Heckl · C. R. E. Baer
Department of Physics, Institute of Quantum Electronics,
ETH Zurich, 8093 Zurich, Switzerland

T. Südmeyer
Department of Physics, University of Neuchâtel,
2000 Neuchâtel, Switzerland

C. Kränkel · G. Huber
The Hamburg Centre for Ultrafast Imaging,
Luruper Chausee 149, 22761 Hamburg, Germany

1 Introduction

Over the last few years the cubic sesquioxides Sc_2O_3, Y_2O_3, and Lu_2O_3 gained significant attention due to their potential for efficient high-power operation for various laser ions like Yb, Tm, Ho, and Er [1–4]. In particular, Yb-doped Lu_2O_3 offers a very low quantum defect and high absorption cross sections when pumped at the zero phonon line. In combination with the high thermal conductivity of rare-earth doped sesquioxides [5] it is thus an ideally suited material for high-power thin disk lasers (TDLs) [6]. With this material 300 W of pump-limited output power at 74 % optical efficiency could be obtained in multimode cw operation corresponding to the highest efficiency of any high-power TDL so far [5]. In addition, the broad emission bandwidth of Yb:Lu_2O_3 allows for the generation of ultrashort pulse [7] which resulted in pulse durations as short as 738 fs at an average output power as high as 141 W demonstrated in a mode-locked thin disk laser [8]. Quite recently this result was surpassed by Yb:YAG with 583 fs at 275 W using a vacuum environment [9]. In terms of pulse energy 41 μJ was achieved from a mode-locked Yb:YAG thin disk laser with 1.1 ps pulse duration obtained in an active multipass cell [10]. This impressively presents the high potential of the thin disk laser approach utilizing Yb:YAG. Yb:Lu_2O_3 is a promising candidate to demonstrate even shorter pulse durations, higher pulse energies, and higher average output powers under similar conditions thanks to its better thermal and spectroscopic capabilities.

Besides Yb:Lu_2O_3, the related sesquioxide Yb:Sc_2O_3 shows comparable laser performance in cw operation [5]. However, the emission peak around the laser wavelength in the emission spectrum of Yb:Sc_2O_3 is significantly red-shifted compared to Yb:Lu_2O_3. Thus, the combination of both crystals in one disordered crystal structure results in

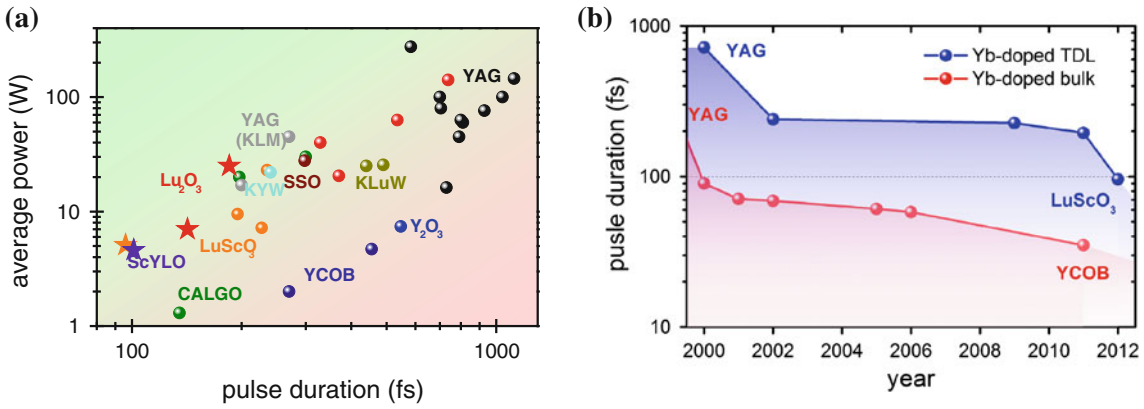

Fig. 1 **a** Output power versus pulse duration of various Yb-based mode-locked TDL oscillators. **b** Timely development of shortest pulse duration of TDLs based on Yb-doped materials

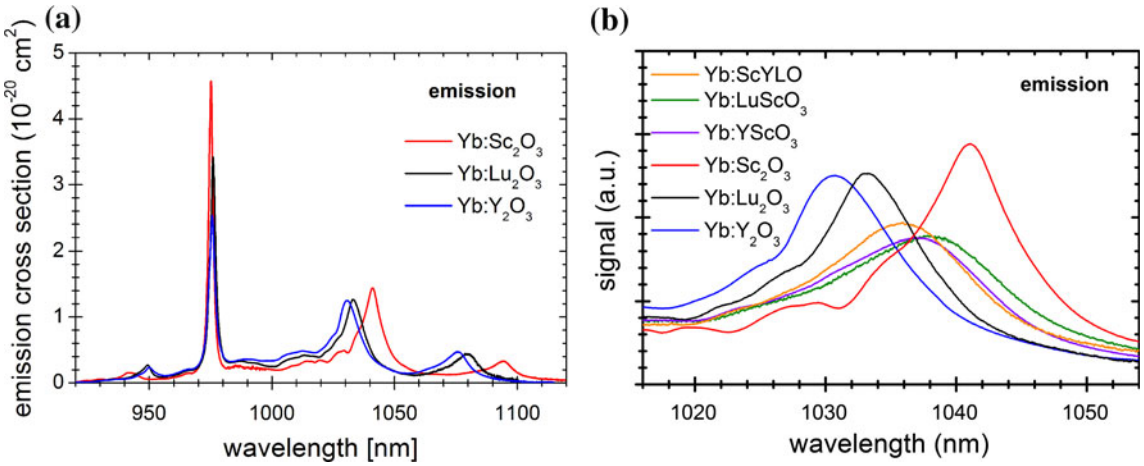

Fig. 2 Emission spectra: **a** pure sesquioxides $Yb:Sc_2O_3$, $Yb:Lu_2O_3$, and $Yb:Y_2O_3$, **b** mixed sesquioxides $Yb:YScO_3$, $Yb:LuScO_3$, and Yb:ScYLO in comparison with pure sesquioxides around the laser wavelength

an inhomogeneously broadened emission bandwidth. The first modelocking experiments with the resulting sesquioxide $Yb:LuScO_3$ delivered already 227 fs [11], which could be improved to 195 fs [12] and recently allowed for sub-100 fs pulse durations [13]. As can be seen from Fig. 1a, to date these are shortest pulse durations ever obtained with a TDL. Figure 1b illustrates the development of the pulse durations since the first mode-locked TDL in 2000 [14].

In this paper, we present a detailed spectroscopic characterization of mixed sesquioxide materials with their tailored gain bandwidths. Among the presented materials, novel hosts such as $Yb:YScO_3$ and $Yb:(Lu,Y,Sc)_2O_3$ (Yb:ScYLO) will be introduced for the first time. All the presented materials are promising candidates for the generation of sub–100 fs pulses at the 100–W average output power level, in particular given the recent demonstration of nearly bandwidth-limited pulse durations of 142 fs with $Yb:Lu_2O_3$, 96 fs with $Yb:LuScO_3$, and 101 fs with Yb:ScYLO.

2 Spectroscopic investigations

The sesquioxides Sc_2O_3, Y_2O_3 and Lu_2O_3 crystallize in cubic bixbyite [15] structure making them suitable gain materials for TDLs where a uniform thermal expansion coefficient is favorable to avoid stress in the crystal. As all cubic sesquioxides form continuous series of solid solutions [16], the mixed sesquioxides $YScO_3$, $LuScO_3$, and ScYLO possess similar crystallographic properties compared to pure sesquioxides containing only one kind of metal ions. Among the rare-earth ions that form cubic oxides, Y has the largest ionic radius (0.9 Å), whereas Sc exhibits the smallest radius (0.75 Å) and Yb_2O_3 (0.87 Å) and Lu_2O_3 (0.86 Å) lie in between. This causes a relatively strong crystal field variance between $Yb:Y_2O_3$ and $Yb:Sc_2O_3$ resulting in a strong shift of the emission peaks which can be found in Fig. 2a.

It can be seen from the spectra that the combination of $Yb:Y_2O_3$ and $Yb:Sc_2O_3$ should result in a material with

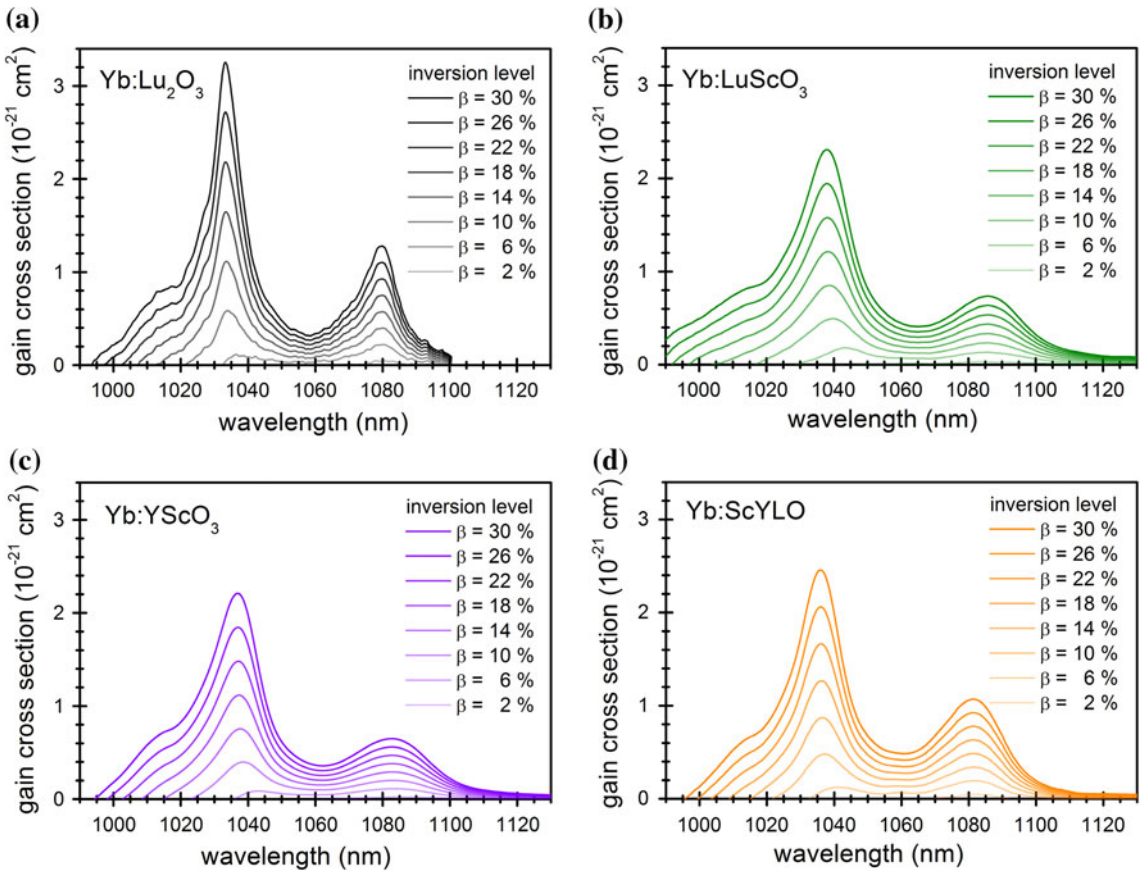

Fig. 3 Gain cross sections of **a** $Yb:Lu_2O_3$, **b** $Yb:LuScO_3$, **c** $Yb:YScO_3$, and **d** Yb:ScYLO

Table 1 Emission bandwidths (FWHM) and bandwidth-limited pulse durations in comparison with experimental results obtained with the investigated sesquioxides

	$Yb:Y_2O_3$	$Yb:Lu_2O_3$	$Yb:Sc_2O_3$	$Yb:LuScO_3$	$Yb:YScO_3$	Yb:ScYLO
Emission bandwidth (FWHM) (nm)	14	13	12	22	22	18
Min. pulse duration (fs)	80	86	93	51	51	62
Exp. pulse duration TDL	430 fs [19]	142 fs	–	96 fs	–	101 fs
Fraction of emission (gain) spectrum TDL (%)	19	65 (75)	–	57 (61)	–	68 (76)
Exp. pulse duration bulk (SESAM)	–	–	230 fs	111 fs [20]	–	–
Fraction of emission spec. bulk (SESAM) (%)	–	–	44	50	–	–
Exp. pulse duration bulk (KLM)	68 fs [21]	65 fs [22]	81 fs [23]	74 fs [20]	–	–

Note that the experimentally demonstrated pulse durations in KLM operation can be shorter than the theoretical limit given above due to spectral broadening caused by nonlinear effects such as self-phase modulation [24]

broadest emission spectrum. However, $Yb:Y_2O_3$ crystals of good quality are more difficult to grow than $Yb:Sc_2O_3$ or $Yb:Lu_2O_3$ crystals. This is due to a phase transition from hexagonal to cubic structure at temperatures in the range of the melting point in Y_2O_3 [15]. Measurements of scattering revealed relatively high losses of 0.18 dB/mm for Y_2O_3 and below 0.01 dB/mm for Lu_2O_3 and Sc_2O_3 [17]. As a result, $Yb:YScO_3$ was expected to show slightly degraded crystal quality. To ensure good crystal quality without scattering, a mixed crystal made of Sc_2O_3, Y_2O_3 and Lu_2O_3 was grown to keep the Y_2O_3 content low. In Fig. 2b, the emission spectra of the mixed crystals $Yb:LuScO_3$, $Yb:YScO_3$, and Yb:ScYLO are presented in comparison to the pure sesquioxides. As expected the emission bandwidths are strongly enlarged. It can be seen that the emission bandwidths of $Yb:YScO_3$ and $Yb:LuScO_3$ are almost the same with 22 nm FWHM each while the emission bandwidth of Yb:ScYLO is just 18 nm FWHM.

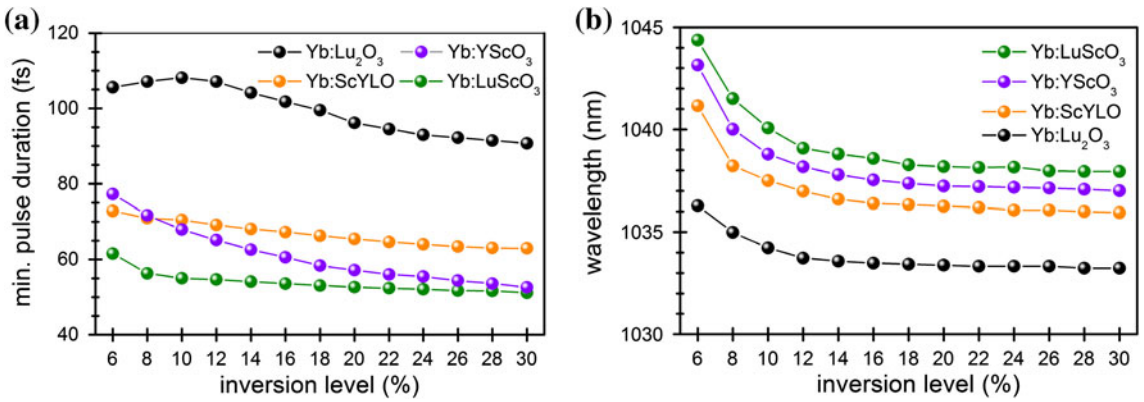

Fig. 4 **a** Bandwidth-limited pulse duration versus inversion level for Yb:Lu_2O_3, Yb:$LuScO_3$, Yb:$YScO_3$, and Yb:ScYLO. **b** Wavelength versus inversion level for the investigated sesquioxide crystals

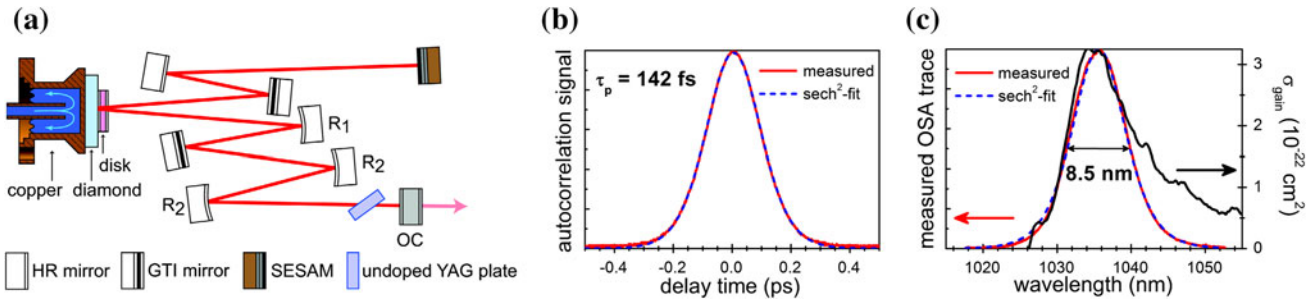

Fig. 5 Yb:Lu_2O_3 **a** Schematic of the cavity. **b** Autocorrelation trace of the pulses. **c** Optical spectrum of the pulses and gain spectrum in comparison

The resulting gain spectra for the well-established Yb:Lu_2O_3 and the mixed sesquioxides Yb:$LuScO_3$, Yb:$YScO_3$, and Yb:ScYLO can be found in Fig. 3 for typical inversion levels of TDLs from 0 to 30 %. From these data, the shortest possible pulse durations for $sech^2$ pulses not considering self-broadening effects are listed in Table 1 and illustrated in Fig. 4a as a function of the inversion level for all investigated sesquioxides. The calculation was done considering the time-bandwidth product limit for $sech^2$-pulses of 0.315 and dividing by the FWHM in frequency domain. The shortest possible pulse duration theoretically accessible by SESAM modelocking would be around 50 fs and could be obtained with Yb:$LuScO_3$ or Yb:$YScO_3$. But given the above-mentioned expectations about the possible scattering in $YScO_3$, Yb:$LuScO_3$ should remain the best candidate for future power scaling of sub-100 fs pulses.

3 Laser experiments

Thin disk laser (TDL) experiments with 1.9 mm of pump spot diameter using a 150 µm thick Yb(3 %):Lu_2O_3 disk glued on a diamond heat sink were carried out in a 24 pump pass module. The disk was HR coated at the backside and AR coated at the front side and was used as a folding mirror in a V-shape type fundamental mode cavity. To avoid residual reflections that can destabilize mode-locked operation, the disk had a wedge of 0.1°. The pump diode was stabilized at 976 nm using a volume Bragg grating to match the narrow zero phonon line absorption of Yb:Lu_2O_3. In Fig. 5a, the schematic cavity setup is shown. Two GTI type mirrors accounting for 2,200 fs^2 of negative dispersion per roundtrip were inserted to achieve soliton modelocking [18]. The necessary self-phase modulation to balance the dispersion was inserted by focusing in a 1.5 mm thick Brewster cut YAG plate. The SESAM used in this experiment was characterized with 1 ps pulses at 1,030 nm. A modulation depth of 3.4 %, nonsaturable losses of 0.8 %, a fast recovery time of 1.9 ps and a saturation fluence of 35 µJ/cm^2 could be evaluated. The output coupling mirror was chosen to transmit 4 % at the central laser emission wavelength of 1,035 nm.

With 47 W of pump power stable modelocking up to an average power of 7 W with pulses as short as 142 fs at a repetition rate of 64 MHz could be obtained. With a time-bandwidth product of 0.337 the laser produces nearly transform-limited pulses. In Fig. 5b, c, the laser spectrum

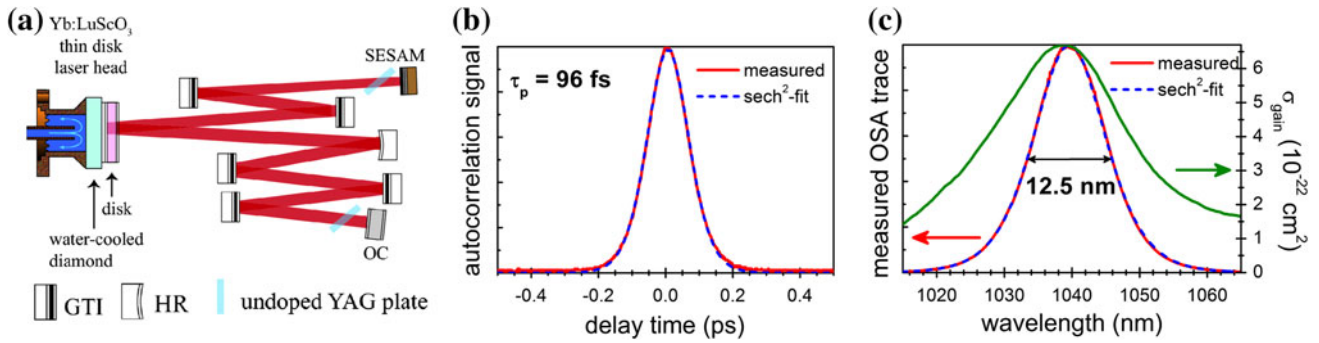

Fig. 6 Yb:$LuScO_3$ **a** Schematic of the cavity. **b** Autocorrelation trace of the pulses. **c** Optical spectrum of the pulses and gain spectrum in comparison

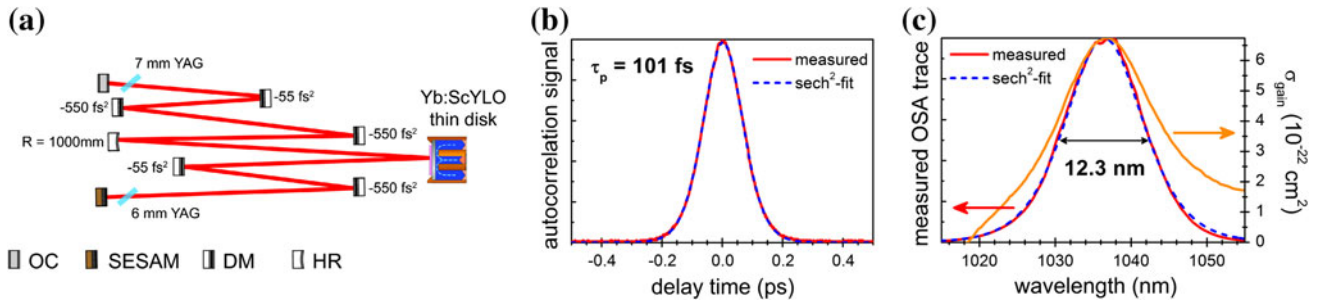

Fig. 7 Yb:ScYLO **a** Schematic of the cavity. **b** Autocorrelation trace of the pulses. **c** Optical spectrum of the pulses and gain spectrum in comparison

and the autocorrelation trace are shown. To evaluate that this is close to the bandwidth limit also the gain spectrum is inserted in Fig. 5c. The inversion level can be estimated from Fig. 4b to be 8 % as the laser wavelength was 1,035 nm. This is confirmed by the fact that in cw operation the laser tended to switch the wavelength between 1,035 and 1,080 nm which is resulting from similar gain at both wavelengths. It can be seen that 75 % of the gain spectrum is locked which is to the best of our knowledge the maximum in the SESAM mode-locked TDL configuration so far. Power scaling to 25 W still delivered 185 fs which is at least 59 % of the gain bandwidth.

In a similar experiment (Fig. 6a) with a 200 µm thick Yb(3 %):$LuScO_3$ disk using 5 GTI type mirrors accounting for $-2{,}800$ fs^2 group delay dispersion in total and an output coupling rate of 2.6 %, 96 fs at 5.1 W of average output power with a repetition rate of 77.5 MHz could be obtained at 1,039 nm. The autocorrelation trace and the laser spectrum can be found in Fig. 6b, c. The fraction of the gain bandwidth modelocked in this experiment was 61 % as shown in Fig. 6b.

First experiments with Yb(3 %):ScYLO (Fig. 7a) using 5 GTI mirrors for $-1{,}760$ fs^2 group delay dispersion and an output coupling rate of 1.8 % led to a pulse duration of 101 fs at 4.6 W of output power and a repetition rate of 75 MHz at 1,037 nm. The autocorrelation and the laser spectrum are shown in Fig. 7b, c. In this case 76 % of the gain spectrum could be locked.

Thin disk laser (TDL) experiments with Yb(3 %):$YScO_3$ could not be performed yet as the processing of the disks is still in progress but in terms of pulse duration it is expected to yield similar results as for Yb:$LuScO_3$.

Compared to the results obtained with these materials in laser setups utilizing bulk crystals, these results are very promising. Although the pulse durations of Kerr-lens mode-locked (KLM) bulk lasers are much shorter, thermal aberrations that occur in the bulk gain medium limit the obtainable output power. In contrast, SESAM mode-locked TDLs are power scalable by increasing the pump spot on the disk and on the SESAM simultaneously keeping the intensity constant. Therefore, this geometry should allow for high-power sub-100 fs pulse generation. Current improvements on SESAM design [25] and the crystal growth technology will probably lead to even shorter pulses at even higher powers in the near future thanks to an increased quality of the disks and SESAMS.

4 Conclusion

In conclusion we grew and spectroscopically investigated the new mixed sesquioxide crystals Yb:$YScO_3$ and

Yb:ScYLO and compared them in terms of ultrashort pulse generation with the already known pure and mixed sesquioxides Yb:Sc_2O_3, Yb:Y_2O_3, Yb:Lu_2O_3, and Yb:$LuScO_3$. It was found that the mixed sesquioxides Yb:$LuScO_3$ and Yb:$YScO_3$ are the best choice for the generation of ultrashort pulses in the TDL setup as both provide an emission bandwidth of 22 nm which is up to 1.8 times larger than for the pure sesquioxide crystals. In SESAM mode-locked experiments, pulse durations of 142 fs with Yb:Lu_2O_3, 96 fs with Yb:$LuScO_3$ and 101 fs with Yb:ScYLO could be obtained by exploiting more than 70 % of the gain bandwidth, which are the shortest pulse durations ever obtained from a mode-locked thin disk laser oscillator.

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