High Quality-factor Kerr-lens mode-locked Tm:Sc_2O_3 single crystal laser with anomalous spectral broadening

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We report on a high Quality-Factor Kerr-lens mode-locked Tm:Sc_2O_3 laser operating around 2.1 µm. Using a simple four mirror Z-shaped cavity consisting of an output coupler of 0.5% transmittance, two folding mirrors, and a chirped mirror, pulses as short as 72 fs with an average power of 130 mW were obtained at a center wavelength of 2108 nm. By replacing the 0.5% output coupler with a ~0.3% one, we observed anomalous broadening of the spectrum into the range between ~2250 and 2350 nm where Tm:Sc_2O_3 shows no gain.

Ultra-short pulse 2 μm lasers are attracting great interest for various applications, including wavelength conversion. This does not only include conversion to longer wavelengths from the mid-infrared to the terahertz region by supercontinuum generation[1] or optical-parametric processes[2-4] in non-oxide nonlinear materials. It also covers the shorter wavelength range, e.g. up to the soft X-ray region by high-harmonic generation[5]. In the last decade, there were many efforts to shorten the available pulse duration directly from Tm^{3+}-doped solid state bulk oscillators[6,7] and in the last few years, sub-100-fs operation was successfully achieved by the combinations of new gain media and saturable absorbers. As an example, 76 fs pulses were obtained from a single-walled carbon nanotube saturable absorber (SWCNSA) mode-locked Tm:$MgWO_4$ laser [8]. Employing Tm:Ho co-doped disordered materials, pulses as short as 52 fs were obtained from a GaSb-based semiconductor saturable absorber mirror (SESAM) mode-locked Tm,Ho:$CaYAlO_4$ [9]. Among the Tm^{3+} doped laser materials, Tm:RE_2O_3(RE=Sc, Lu or Y) materials are promising gain media for high power short pulse 2 μm lasers due to their superior spectroscopic, thermo-optical and thermo-mechanical properties[10]. Their strong crystal field results in fluorescence bands located at wavelengths beyond 1950 nm, enabling laser operation in the wavelength range between 1.95 μm and 2.2 μm. This operation wavelength range suffers less atmospheric water vapor absorption and ground state absorption, which allows for a broad effective gain bandwidth in mode-locked operation. The high thermal-conductivity of these materials can mitigate undesirable thermal influence during high power laser operation. The available pulse duration from pure Tm:RE_2O_3, however, was limited so far. Using an InGaAsSb quantum-well SESAM mode-locked Tm:Sc_2O_3 single crystal laser, the pulse duration was limited to 218 fs[11], whereas 175 fs pulses were reported from an SWCNSA mode-locked Tm:Lu_2O_3 single crystal laser[12]. Very recently, much shorter pulses were generated with mixed ceramics. Pulses as short as 63 fs were demonstrated using an InGaAsSb quantum-well SESAM mode-locked Tm:$(LuSc)_2O_3$ mixed ceramic laser[13] and even shorter pulses of 57 fs durations were demonstrated using a SWCNSA mode-locked Tm:$(LuY)_2O_3$ mixed ceramic laser[14]. Due to their disordered structure, mixed Tm:RE_2O_3 materials exhibit broader gain bandwidths at the cost of a reduced thermal conductivity and often higher scattering losses.

With the aim of the generation of ultrashort pulses at a high average output power, we previously developed a Kerr-lens mode-locked (KLM) Tm:Sc_2O_3 laser and obtained pulses as short as 166 fs with an average output power of 0.44 W[15]. The main advantages of KLM over other mode locking techniques is, that the Kerr lens acts as a fast saturable absorber enabling a large modulation depth and a broad spectral bandwidth at the same time without the penalty of large nonsaturable losses. This advantage is crucial to obtain the shortest pulse durations[16]. In addition, KLM can be very useful to increase the Q-factor of mode-locked laser cavities. In 2019, Kimura *et al.* reported 22 fs pulse duration from a high Q-factor KLM Yb:CALGO laser with anomalous spectral broadening[17]. The spectral broadening overcome the gain bandwidth limitation of Yb lasers. In [19], This broadening was attributed to Raman-assisted spectral broadening that requires a KLM cavity with a high Q-factor.

Here, aiming to further decrease the pulse durations of 2 μm Tm solid state lasers, we developed a simple 4 mirror high Q-factor Kerr-lens mode-locked Tm:Sc_2O_3 single crystal laser. The cavity consisted of an output coupler, two folding mirrors, and a chirped mirror. With an output coupler of 0.5% transmittance, pulses as short as 72 fs with an average power of 130 mW were obtained at a center wavelength of 2108 nm. By further increasing the cavity Q-factor with an output coupler of less than 0.3% transmittance, we observed anomalous spectral broadening into the range from ~2250 to 2350 nm, where Tm:Sc_2O_3 shows no gain. The phenomenon was similar to the result reported by Kimura *et al.*[17] and should allow further pulse shortening of KLM ultrafast Tm solid state oscillators.

The experimental configuration is shown in Fig. 1. The cavity consisted of an output coupler (OC), two folding mirrors (M1 and M2), and a chirped mirror (CM). The gain medium was a Tm^{3+}:Sc_2O_3 single crystal with a doping concentration of 1%, a thickness of 3.7 mm and a group delay dispersion (GDD) of -250 fs^2 per pass. It was mounted in a water-cooled copper heatsink and placed between two folding mirrors at Brewster's angle. We used in-band pumping at 1611 nm ($^3H_4 \rightarrow {}^3F_4$) with a home-built Er:Yb fiber MOPA pump source[18]. The folding mirror's reflectivity in the wavelength range from 1850 to 2200 nm was >99.8%, and the radius of curvature was 100 mm. The pump light was imaged into the gain medium through one of the folding mirrors, which provided a transmittance of 90% for the pump wavelength. The estimated diameters of the pump laser mode and the cavity fundamental mode at the position of the gain medium were 47 × 47 μm^2 and 55 ×57 μm^2 (sagittal × tangential), respectively. The chirped mirror (GDD was -1000 fs^2 from 2000 to 2200 nm, reflectivity was R>99.9% from 1900 to 2200 nm, UltraFast Innovations LLC) was placed at the end of cavity. We used various OCs with different transmittance, 1% (2000-2300 nm), 0.5%(1900-2100 nm) and ~0.3%(1900-2250 nm, GDD~-200 fs^2, 2000-2250 nm). A 3 mm thick silicon plate was employed for external pulse compression.

KLM was obtained with all three different OCs. With the 1% OC, the highest average power of 222 mW with a pulse duration of 80 fs was obtained [Fig. 2(a)] The center wavelength and spectral bandwidth were 2141 nm and 65 nm, respectively [Fig. 2(b)] The repetition rate was 93.8 MHz which is in good agreement with the cavity length [Fig. 2(c)] The maximum pulse energy and peak power were 2.37 nJ and 29.6 kW, respectively. The average output power as a function of the pump power is shown in Fig. 2(d). The KLM was initiated at a pump power of 580 mW by moving the M2. At the onset of KLM the average power doubled, which indicates a large modulation depth. We did not use any hard aperture inside the cavity so the KLM was achieved only by the soft aperture effect.

With the 0.5% OC, the shortest pulse duration of 72 fs with an average output power of 130 mW was obtained [Fig. 3(a)]. The center wavelength and the spectral bandwidth were 2108 nm and 67 nm [Fig. 3(b)] respectively. Due to the narrower bandwidth of the 0.5% OC covering only the range from 1900 to 2100 nm, the center wavelength shifted 33 nm into the shorter wavelength range compared to the 1% OC. The narrow spectral peaks around 2250 nm in Fig. 3(b) are Kelly sidebands. The time-bandwidth product was 0.325, indicating $sech^2$ pulse shape. Pulse energy and peak power were 1.38 nJ and 19.2 kW, respectively.

Using the 0.3% OC, the broadest spectral bandwidth of 75 nm was obtained at a soliton peak wavelength of 2164 nm (Fig. 4 red dashed curve) with a maximum average power of 50 mW. In addition, we found anomalous spectral broadening into the range between ~2250 and 2350 nm. This region is outside of the gain band of the $^3F_4 \rightarrow {}^3H_6$ transition of $Tm:Sc_2O_3$. The $^3H_4 \rightarrow {}^3H_5$ transition could have emission in this range, when pumping with a wavelength of ~800 nm[19-21] or an upconversion pumping process[22] are applied. However, we are using in-band pumping and the measured fluorescence spectrum under pumping at 1611 nm indicates no fluorescence at wavelengths above 2250 nm (Fig.4 green solid curve). The threshold pump power for this spectral broadening was as low as 500 mW. When the pump power was increased, only the spectral components around 2250-2350 nm grew and the peak intensity around 2164 nm was clipped (Fig. 4). The phenomenon and the spectral shape were similar to the result of the 22 fs Yb:CALGO laser reported by Kimura *et al.*[17]. Therefore we attribute the broadening observed here also to Raman assisted spectral broadening. The calculated Raman gain spectrum of our $Tm:Sc_2O_3$ mode-locked laser is also shown in Fig. 4(black dashed curve). The spectral component around 2250-2350 nm is located inside the Raman gain band but not at the Raman gain maximum position. The mismatch can be explained by the reflectivity and GDD of the cavity. The designed reflectivity and GDD of the chirped mirrors used in our experiment are shown in Fig. 5(a). The reflectivity suddenly decreases at wavelengths longer than 2300 nm and the GDD shows strong high order dispersion above 2250 nm that must be also the origin of the dense Kelly sidebands around 2250 nm. The Raman gain decreases when the group velocity (GV) of the pump pulse and the Raman pulse do not match, because in that case they do not overlap in the gain crystal during the cavity round trip. So the position of the anomalous spectral broadening component was governed by a balance of GV and reflectivity. It's worth noting that the main $sech^2$ shape spectral component of our KLM laser was much broader than the fluorescence bandwidth (Fig. 4) and this is the shortest pulse operation mod mode-locked non-disordered Tm solid state laser to our knowledge. Furthermore, assuming the whole spectrum including the Raman gain band at 2250-2350 nm, the corresponding transform limited pulse duration would be ~38 fs [Fig. 5(b)] and thus nearly a factor of two shorter than without the 2250-2350 nm spectral component (~65 fs).

We also tried to measure the pulse duration by intensity autocorrelation. However, the measured autocorrelation trace with the spectral broadening effect was not stable. We explain the unstable autocorrelation trace as follows: The spectral broadening takes place inside the cavity with the very short crystal length. Under these conditions, the coherence of the whole spectrum can be conserved[23)]. However, the GDD value of our cavity around 2300 nm was strongly positive and not flat, and therefore the spectral component around 2300 nm was strongly chirped and/or the phase relationship between the main soliton spectral component and the spectral component around 2300 nm was rapidly changing per round trip. Therefore, the Raman band cannot contribute to pulse shortening. This issue can be solved by using a broader bandwidth chirped mirror, which should then result in close to Fourier limited pulse durations utilizing the whole bandwidth of the oscillator. Furthermore, with a proper GDD mirror, the shape of the Raman component should become smoother and the threshold for the spectral broadening should decrease that mitigates the requirement of a high Q-factor cavity and increases an average output power because the effective Raman gain also broadens and increases with a proper GDD mirror. The other requirements to obtain a short pulse duration with this phenomenon would be following: First the cavity should have broad reflectivity bandwidth to fully cover all spectral components. Second the cavity should have a large modulation depth and/or a high Q-factor as the spectral components outside the gain bandwidth of the gain medium does not contribute to stimulated emission and therefore the gain saturation becomes weaker, which can easily lead to multi-pulse operation or the appearance of narrow CW spectral components[16,24)].

In conclusion, we developed a high Q-factor 4 mirror KLM $Tm:Sc_2O_3$ laser. With an output coupler of 0.5% transmittance, the shortest pulse duration of 72 fs with an average output power of 130 mW was obtained at a center wavelength of 2108 nm. By further increasing the cavity Q-factor by using the output coupler of less than 0.3% transmittance, we observed anomalous spectral broadening into the range between 2250 and 2350 nm, where Tm:Sc_2O_3 shows no gain. This phenomenon is attribute to Raman assisted spectral broadening. With proper design of the total cavity dispersion and reflectivity bandwidths, the additional spectral component can contribute to pulse shortening and the spectrum should support sub-40-fs pulse duration.

Acknowledgments

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References

1) A. M. Heidt, J. H. V. Price, C. Baskiotis, J. S. Feehan, Z. Li, S. U. Alam, and D. J. Richardson, “Mid-infrared ZBLAN fiber supercontinuum source using picosecond diode-pumping at 2 μm,” Opt. Express 21, 24281-24287 (2013)Schliesser, N. Picqué, and T. W. Hänsch, Nat. Photonics **6**, 440 (2012).

2) A. Godard, “Infrared (2–12 μm) solid-state laser sources: a review” C. R. Phys. **8**, 1100-1128 (2007).

3) V. O. Smolski, H. Yang, S. D. Gorelov, P. G. Schunemann, and K. L. Vodopyanov, “Coherence properties of a 2.6-7.5 μm frequency comb produced as a subharmonic of a Tm-fiber laser,” Opt. Lett. **41**, 1388–1391 (2016).

4) P. Liu, F. Qi, W. Li, Y. Wang, Z. Liu, H. Wu, W. Ning, W. Shi, and J. Yao, “Low-threshold terahertz-wave generation based on a cavity phase-matched parametric process in a Fabry–Perot microresonator,” J. Opt. Soc. Am. B **35**, 68-72 (2018).

5) K. F. Lee, X. Ding, T. J. Hammond, M. E. Fermann, G. Vampa, and P. B. Corkum, “Harmonic generation in solids with direct fiber laser pumping,” Opt. Lett. **42**, 1113-1116 (2017).

6) A.A. Lagatsky, S. Calvez, J. A. Gupta, V. E. Kisel, N. V. Kuleshov, C. T. A. Brown, M. D. Dawson, and W. Sibbett, “Broadly tunable femtosecond mode-locking in a Tm:KYW laser near 2 μm,” Opt. Express **19**, 9995-10000 (2011).

7) A. Schmidt, S. Y. Choi, D. Yeom, F. Rotermund, X. Mateos, M. Segura, F. Diaz, V. Petrov, and U. Griebner, “Sub-150 fs Pulses from a Tm:KLuW Oscillator in the 2 μm Wavelength Range,” in Conference on Lasers and Electro-Optics 2012, OSA Technical Digest (online) (Optical Society of America, 2012), paper CM1B.4.

8) L. Wang, Y. Zhao, Y. Wang, L. Zhang, H. Lin, J. E. Bae, S. Y. Choi, F. Rotermund, P. Loiko, X. Mateos, U. Griebner, V. Petrov, and W. Chen, “76 fs SWCNT-SA mode-locked Tm:$MgWO_4$ laser at 2 μm,” in 2019 Conference on Lasers and Electro-Optics Europe and European Quantum Electronics Conference, OSA Technical Digest (Optical Society of America, 2019), paper cf_2_4.

9) Y. Wang, Y. Zhao, P. Loiko, Z. Pan, W. Chen, M. Mero, X. Xu, J. Xu, X. Mateos, A. Major, S. Suomalainen, A. Härkönen, M. Guina, U. Griebner, and V. Petrov, “52-fs SESAM Mode-Locked Tm,Ho:CALGO Laser,” in Laser Congress 2019 (ASSL, LAC, LS&C), OSA Technical Digest (Optical Society of America, 2019), paper AM3A.7.

10) C. Kränkel, „are-Earth-Doped Sesquioxides for Diode-Pumped High-Power Lasers in the

1-, 2-, and 3-μm Spectral Range, IEEE Journal of Selected Topics in Quantum Electronics 21(1), page 250-262 (2015).

11) A. A. Lagatsky, P. Koopmann, P. Fuhrberg, G. Huber, C. T. A. Brown, and W. Sibbett, "Passively mode locked femtosecond Tm:Sc_2O_3 laser at 2.1 μm," Opt. Lett. **37**, 437-439 (2012).

12) A. Schmidt, P. Koopmann, G. Huber, P. Fuhrberg, S. Y. Choi, D.-Il Yeom, F. Rotermund, V. Petrov, and U. Griebner, "175 fs Tm:Lu_2O_3 laser at 2.07 µm mode-locked using single-walled carbon nanotubes," Opt. Express **20**, 5313-5318 (2012).

13) Y. Wang, W. Jing, P. Loiko, Y. Zhao, H. Huang, X. Mateos, S. Suomalainen, A. Härkönen, M. Guina, U. Griebner, and V. Petrov, "Sub-10 optical-cycle passively mode-locked Tm:$(Lu_{2/3}Sc_{1/3})_2O_3$ ceramic laser at 2 μm," Opt. Express **26**, 10299-10304 (2018).

14) Y. Zhao, L. Wang, Y. Wang, J. Zhang, P. Liu, X. Xu, Y. Liu, D. Shen, J. E. Bae, T. G. Park, F. Rotermund, X. Mateos, P. Loiko, Z. Wang, X. Xu, J. Xu, M. Mero, U. Griebner, V. Petrov, and W. Chen, "SWCNT-SA mode-locked Tm:$LuYO_3$ ceramic laser delivering 8-optical-cycle pulses at 2.05 μm," Opt. Lett. **45**, 459-462 (2020).

15) M. Tokurakawa, E. Fujita and C.Krankel, "Kerr-lens mode-locked Tm^{3+}:Sc_2O_3 single-crystal laser in-band pumped by an Er:Yb fiber MOPA at 1611 nm," Opt. Lett. **42**, 3185-3188 (2017).

16) M.Tokurakawa and A. Shirakawa, "Numerical analysis of fast saturable absorber mode-locked Yb^{3+} lasers under large modulation depth," Opt. Express **23**, 26288-26298 (2015).

17) S. Kimura, S. Tani and Y. Kobayashi, "Raman-assisted broadband mode-locked laser," Scientific Reports **9**, 3738 (2019).

18) E. Fujita, Y. Mashiko, S. Asaya, M. Musha, and M. Tokurakawa, "High power narrow-linewidth linearly-polarized 1610 nm Er:Yb all-fiber MOPA," Opt. Express **24**, 26255-26260 (2016).

19) J. Caird, L. DeShazer, and J. Nella, "Characteristics of room-temperature 2.3-µm laser emission from tm^{3+}in YAG and $YAlO_3$" IEEE J. Quantum Electron. **11**, 874 (1975).

20) J. F. Pinto, L. Esterowitz, and G. H. Rosenblatt, "Tm^{3+}:YLF laser continuously tunable between 2.20 and 2.46 μm," Opt. Lett. **19**, 883-885 (1994).

21) F. Canbaz, I. Yorulmaz and A. Sennaroglu, "Kerr-lens mode-locked 2.3-μm Tm^{3+}:YLF laser as a source of femtosecond pulses in the mid-infrared," Opt. Lett. **42**, 3964-3967 (2017).

22) L. Guillemot, P. Loiko, R. Soulard, A. Braud, J. L. Doualan, A. Hideur, R. Moncorgé and P. Camy, "Thulium laser at ~2.3 μm based on upconversion pumping," Opt. Lett. **44**, 4071-4074 (2019).

23) G. Genty, S. Coen, and J. M. Dudley, “Fiber supercontinuum sources (Invited),” J. Opt. Soc. Am. B **24**, 1771-1785 (2007).

24) M. Tokurakawa, A. Shirakawa, and K. Ueda, “Estimation of Gain Bandwidth Limitation of Short Pulse Duration Based on Competition of Gain Saturation,” in Lasers, Sources and Related Photonic Devices, OSA Technical Digest Series (CD) (Optical Society of America, 2010), paper AMB16.

Figure Captions

Fig. 1. Cavity configuration of 4 mirror $Tm:Sc_2O_3$ Kerr-lens mode-locked laser

Fig. 2. (a) Autocorrelation trace, (b) spectrum of the mode-locked pulses, (c) RF spectrum, and (d) average power as a function of pump power using a 1% OC.

Fig. 3. (a) Autocorrelation trace, (b) spectrum of the mode-locked pulses using a 0.5% OC.

Fig. 4. Spectra of pulses with a 0.3% OC under the pump power of 0.5 W (blue solid curve) and 1W (red dashed curve) in log scale, and the fluorescence spectrum of $Tm^{3+}:Sc_2O_3$ (green solid curve) and calculated Raman gain (black dashed curve) in linear scale are shown.

Fig. 5. (a) Mode-locked pulse spectrum (black solid curve), reflectivity (red dashed curve) and GDD (blue dotted curve) of the chirped mirrors. (b) Calculated transform limited pulse with (blue dashed curve) and without (red solid curve) the 2250-2350 nm spectral components.

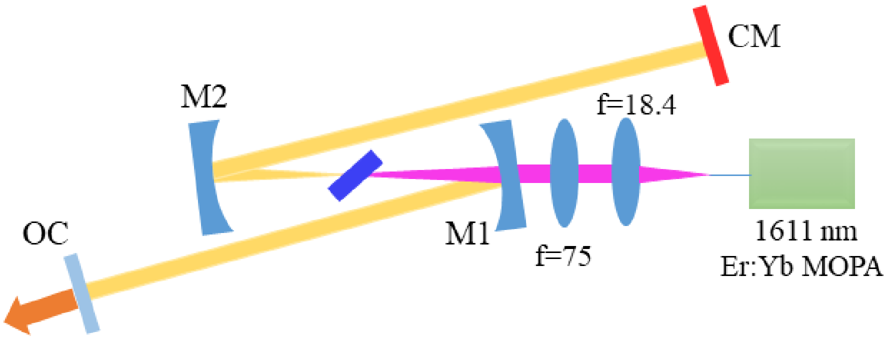


Fig.1.

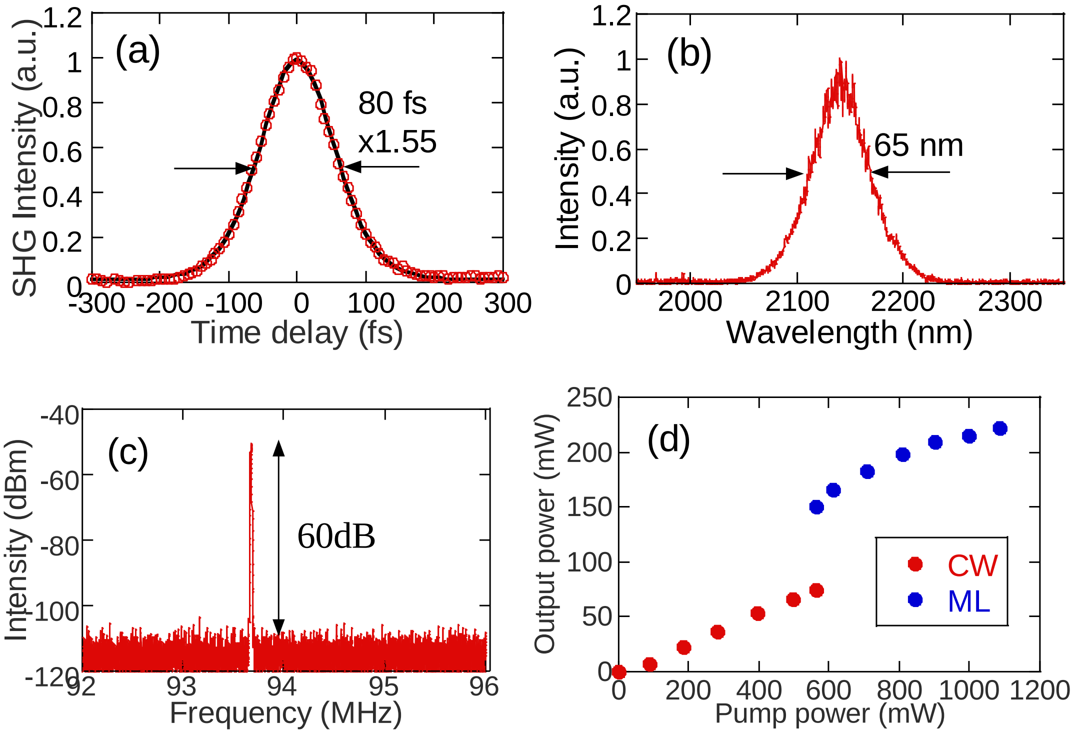
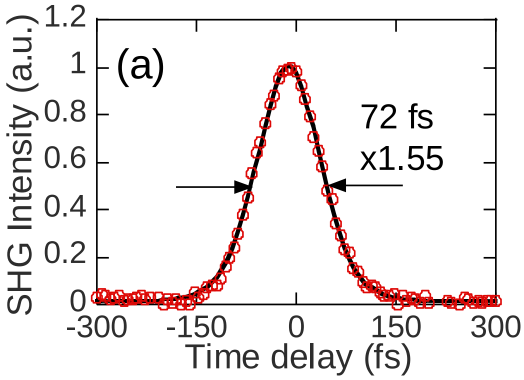

(a)
80 fs
x1.55
SHG Intensity (a.u.)
Time delay (fs)
(b)
65 nm
Intensity (a.u.)
Wavelength (nm)
(c)
60dB
Intensity (dBm)
Frequency (MHz)
(d)
CW
ML
Output power (mW)
Pump power (mW)

Fig. 2.



(a)
72 fs
x1.55
SHG Intensity (a.u.)
Time delay (fs)
1.2
1
0.8
0.6
0.4
0.2
0
-300
-150
0
150
300
(b)
67 nm
Intensity (a.u.)
Wavelength (nm)
2000
2100
2200
2300

Fig. 3.

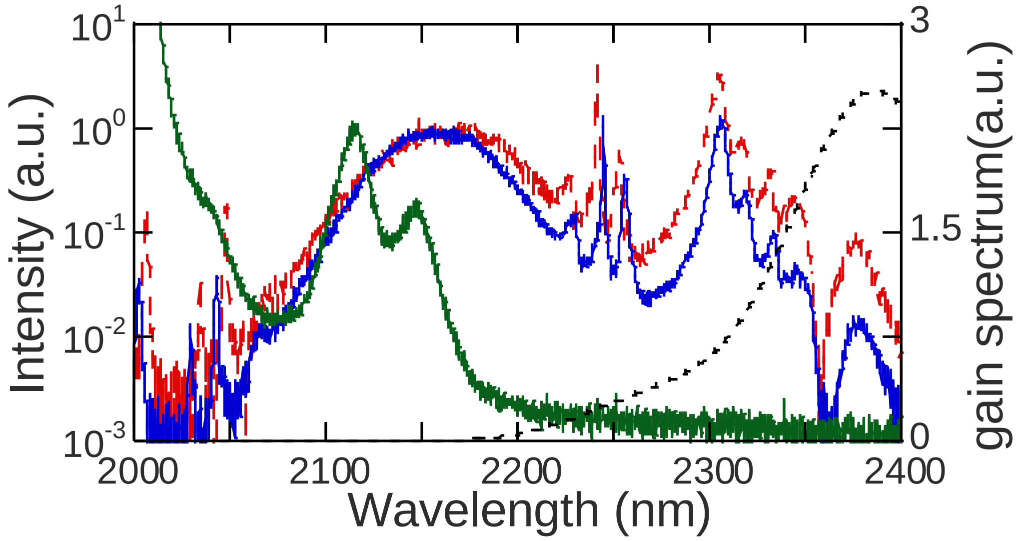

10^1
10^0
10^-1
10^-2
10^-3
Intensity (a.u.)
3
1.5
0
gain spectrum(a.u.)
2000
2100
2200
2300
2400
Wavelength (nm)

Fig. 4.

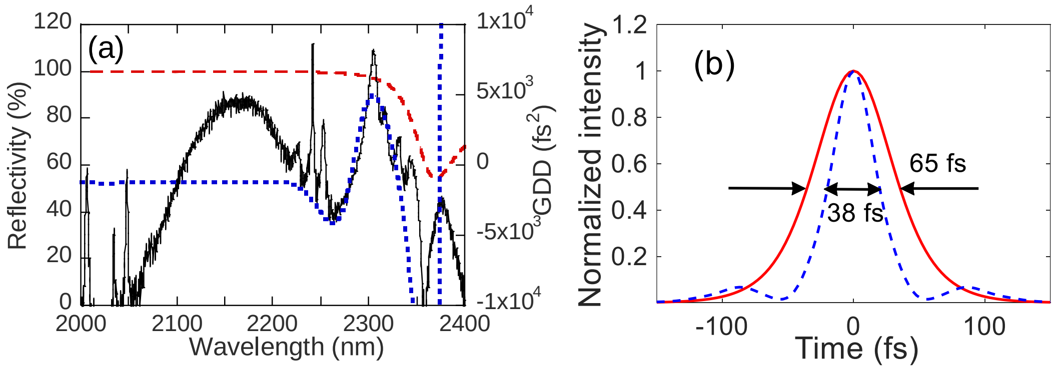

(a)
Reflectivity (%)
GDD (fs^2)
Wavelength (nm)
120
100
80
60
40
20
0
1x10^4
5x10^3
0
-5x10^3
-1x10^4
2000
2100
2200
2300
2400
(b)
Normalized intensity
Time (fs)
1.2
1
0.8
0.6
0.4
0.2
-100
0
100
65 fs
38 fs

Fig. 5