Applied Physics B (2022) 128: 177
https://doi.org/10.1007/s00340-022-07887-8

Applied Physics B
Lasers and Optics

RESEARCH

Second and third harmonic generation from simultaneous high peak- and high average-power thin disk laser

U. Andral[1] · P. Walch[2] · V. Moreno[1] · B. Mahieu[2] · T. Produit[1] · M. Lozano[2] · L. Bizet[2] · C. Herkommer[3,4] · M. Moret[1] · Y.-B. André[2] · P. Krötz[3] · T. Metzger[3] · K. Michel[3] · A. Mysyrowicz[2,5] · J. Kasparian[1,6] · D. Lupinski[7] · A. Houard[2] · J.-P. Wolf[1]

Received: 29 March 2022 / Accepted: 4 August 2022 / Published online: 25 August 2022

Abstract

We report Second Harmonic Generation (SHG) and Third Harmonic Generation (THG) energy conversion efficiencies up to 59% and 27%, respectively, for laser pulses simultaneously delivering high peak power in the sub-TW range and average powers in the sub-kW range. No damage or efficiency decrease is observed after more than 100 h operation time. The resulting high-energy visible and near-UV pulses are suitable for applications, such as lightning control, material analysis and machining, or OPCPA pumping.

1 Introduction

The advent of chirped pulse amplification (CPA) and thin disk technology allowed the development of ultrashort laser systems with unprecedented performance, in terms of either peak- or average powers. However, very few systems [1, 2] have the potential of providing these performances simultaneously, such as terawatt peak power and kilowatt average power. As an exception, a CPA Ytterbium thin disk laser was recently developed, providing a sech^2 pulse with 0.689 TW peak power at 1 kHz repetition rate [1]. In the context of laser filamentation, terawatt peak intensity is required to produce long plasma channels, and simultaneously high average power is needed to heat up the conductive channel and provide a lower density column to guide lightning strikes [3–6]. For lightning control applications [7], shorter wavelengths such as the second or third harmonic of the 1030 nm fundamental radiation could be more advantageous, as ionization of the air molecules requires less photons [8, 9]. Several other applications would also benefit from shorter wavelengths, such as remote laser induced breakdown spectroscopy (R-LIBS, [10, 11]), multiplexed and wide-field micromachining [12], particle acceleration [13] and pumping of OPCPAs [14].

Frequency doubling and tripling laser beams that provide simultaneously TW-peak and kW-average powers constitutes a significant technological challenge. Unlike for nanosecond pulses, SHG conversion efficiencies exceeding 50% are difficult to achieve for intense pulses in the picosecond and femtosecond regimes, due to dispersion, damage caused by ionization or ablation, and self- as well as cross-phase modulation affecting the phase matching between the pump and the harmonic field [15]. A record conversion efficiency of 80% for ultrashort TW pulses was achieved with a Nd:Glass laser at the LLE facility, providing 1 J at 1053 nm, in 500 fs pulses (2 TW) already in 1995, using a type I KDP crystal [16]. The repetition rate was, however, in the order of 1 shot every tens of minutes. More recently, 400 TW of SHG peak power in 500 fs pulses were demonstrated at the Orion facility, with a conversion efficiency of 40% [17]. Similar

---

✉ J. Kasparian
jerome.kasparian@unige.ch

[1] Groupe de Physique Appliquée, Université de Genève, Ch. de Pinchat 22, 1211 Geneva 4, Switzerland

[2] LOA, ENSTA Paris, CNRS, École Polytechnique, Institut Polytechnique de Paris, 828 Bd des Maréchaux, 91762 Palaiseau, France

[3] TRUMPF Scientific Lasers GmbH + Co. KG, Feringastr. 10a, 85774 Unterföhring, Germany

[4] Department of Physics, Technische Universität München, James-Franck-Str. 1, 85748 Garching, Germany

[5] André Mysyrowicz Consultants, 6 Rue Gabriel, 78000 Versailles, France

[6] Institute for Environmental Sciences, Université de Genève, Bd Carl Vogt 66, 1211 Geneva 4, Switzerland

[7] Cristal Laser, P.A. du Breuil, 32 Rue Robert Schuman, 54850 Messein, France

efficiencies (45%) were obtained at fundamental intensities of 50 GW/cm^2 [18], and even nearly 80% from a 150 TW Ti:Sapphire laser delivering 4 J pulses at 800 nm, at a repetition rate of 5 Hz [19]. Conversely, 2 kW average powers have been obtained for SHG [20] and 50 MW for THG [21] of picosecond pulses at MHz repetition rates, but with modest peak powers of 1 GW and 50 MW, respectively. In the present paper, we report the use of large aperture (50–55 mm) LBO crystals for obtaining as much as 295 mJ at 515 nm (0.3 TW peak, 295 W average power, 59% efficiency) for near infrared (NIR) pulses of 495 mJ, 1 ps duration (0.5 TW peak, 500 W average power). We also obtained 96 mJ in 1 ps or 118 mJ in 3 ps at 343 nm (~ 100 GW peak and 100 W average power, up to 27% energy efficiency), from NIR laser pulses of 430 mJ (0.43 TW peak, 430 W average power).

2 Experimental setup

The experimental setup is sketched in Fig. 1. The thin disk Yb:YAG laser system has been described in detail elsewhere [1]. Briefly, it consists of a commercial fiber-based seed laser (Trumpf TruMicro 2000), providing stretched pulses (1 ns) at 1030 nm with an energy up to 100 µJ, a commercial regenerative amplifier (Trumpf DIRA 200-1) boosting the pulse energy up to 240 mJ at 1 kHz, a custom-design multipass amplifier based on 4 thin disk laser heads, pumped each by 2.5 kW diode lasers at 940 nm providing 800 mJ pulses, and a large grating compressor, which recompresses the pulses slightly under 1 ps with an efficiency of 90%. The final output of the Yb thin disk laser system is thus 720 mJ, 920 fs, 1 kHz, although the present work used pulse energies only up to 495 mJ to prevent damages to the optics located downstream. The slightly astigmatic beam delivers on the frequency-doubling crystal a fluence of up to 70 mJ/cm^2 in a diameter of 30 mm ($1/e^2$), with an M^2 of 1.89 (resp. 2.32) for the major (resp. minor) axis (see Fig. 1a).

Large aperture lithium triborate (LBO) crystals were grown by Cristal Lasers SA for frequency doubling and tripling the output of the thin disk Yb laser system. LBO was selected as nonlinear material on the basis of its high damage threshold (18.9 GW/cm^2 at 1053 nm [22]), low dilatation coefficient ($|\theta_L| = 10^5\ \mathrm{K}^{-1}$), excellent transmission (absorption $\mu < 10^{-2}\ \mathrm{m}^{-1}$ at 1.064 µm [23]) and relatively high nonlinear coefficient of ($d_{32} = 1.02$ pm/V [24]). A major aspect was also the technological possibility of producing large aperture crystals (55 mm × 55 mm × 1.8 mm) with excellent optical quality. The thickness of the crystals was selected based on the model of Kobyakov et al. [25] and Sahakyan and Starodub [26]. It is a trade-off between phase matching, group velocity mismatch, efficiency and walk off, that were evaluated with the help of the SNLO Classic software from AS—Photonics [27].

For SHG, the AR coated crystal was cut at angles $\theta = 90°$ and $\phi = 13.8°$ to achieve type I (o–o–e) phase matching. The remaining near-IR was removed from the frequency-doubled beam by using a dichroic mirror, and measured by a Coherent PM1K thermopile sensor. Alternatively, measuring the whole output beam before and after the SHG LBO crystal showed that losses in the LBO crystal remained within the margin of error of the powermeter.

No temperature stabilization was used. No damage was observed over the whole experimental campaign, neither due to overheating nor to nonlinear damages of the surfaces, nor to hot spots/filamentation in the LBO bulk crystal.

A second large aperture LBO crystal was grown for mixing (SFG) the SHG and the fundamental beams to generate the third harmonic (THG) at 343 nm. More precisely, we used a ϕ 50 mm × 2 mm crystal, AR coated at the 3 wavelengths, and cut at $\theta = 90°$ and $\phi = 50.5°$ in the YZ plane, to achieve type II (o–e–o) phase matching. This configuration

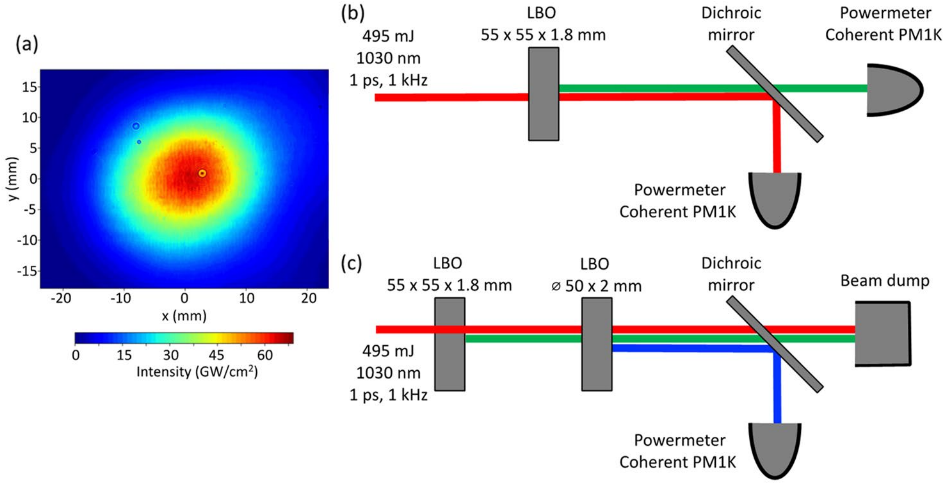

Fig. 1 **a** Fundamental input beam profile; **b**, **c** experimental setup for **b** SHG and **c** THG

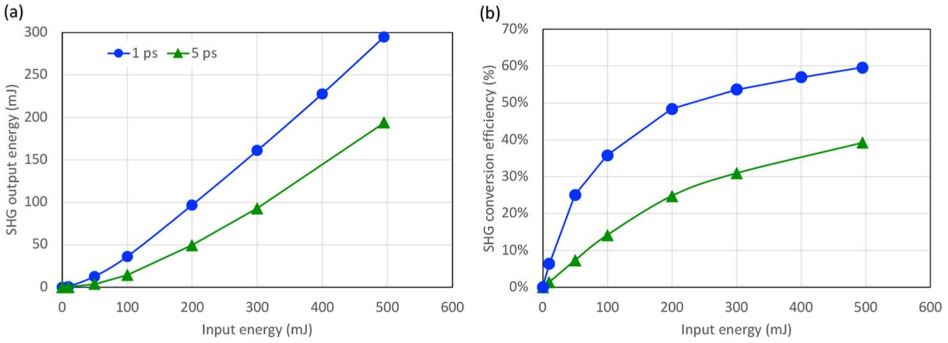

Fig. 2 SHG energy (**a**) and energy conversion efficiency (**b**) of the high peak- and average power Yb thin disk laser system, using a 55 mm aperture LBO crystal

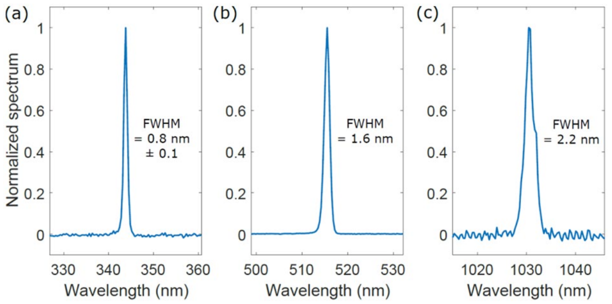

Fig. 3 Output spectra of **a** THG, **b** SHG and **c** fundamental. The spectral resolution is 0.1 nm

allows for a direct SHG–THG crystal stacking, without the need for a half-wave plate. The THG was separated from the beam using a dichroic beam splitter with 95.6% reflection at 343 nm (Thorlabs HBSY23), and measured with the same Coherent PM1K thermopile sensor. The measured 2% reflection of the fundamental and SHG on the beam splitter was subtracted from the measurement.

3 Results and discussion

The SHG conversion efficiency (Fig. 2) exhibits the characteristic tanh^2 behavior of a frequency-doubling process with depleted pump [28]. Even for an intensity of 70 GW/cm^2, the saturation is not reached as the conversion efficiency still rises. Higher intensities would, however, increase the risk of damaging the coating and the crystal. At the maximum investigated pump power (0.5 TW), the 515 nm output reaches 295 mJ, i.e., ~ 0.3 TW peak power and 295 W average power, for a spectral width of 1.6 nm (Fig. 3b). This corresponds to an energy conversion efficiency of 59%. As mentioned in the introduction, simultaneous high peak and high average power in the green spectral region was not reported before. For example, the impressively high yield (80%) and energy (3.2 J) reported in [16] only correspond to an average power of 16 W. The divergence of the SHG beam was minimal, with a beam diameter growing from 3 to 5 cm over 127 m propagation, i.e., a divergence of 0.16 mrad comparable to that of the NIR pump beam. No significant modification of the divergence due to thermal lensing in the crystals was observed.

Figure 4 displays the output energy and conversion efficiency into the third harmonic. The 1 ps pulses reach a peak conversion efficiency of 26%, while the maximum output energy (117 mJ, i.e., 27% conversion efficiency, 117 W average power and 39 GW peak power) is obtained with pulses of 3 ps. The corresponding spectral width is 0.8 nm (Fig. 3a).

The saturation of the conversion efficiency is most likely due to $\chi^{(3)}$-induced phase mismatch effects via cross- and self-phase modulation occurring at higher intensities, and to back conversion [29]. Nonlinear absorption in the crystal and the associated thermal effects may also contribute. Two-photon absorption at 515 nm can be excluded, and two-photon absorption at 343 nm is unlikely since the LBO bandgap lies at 7.78 eV [30]. However, some three

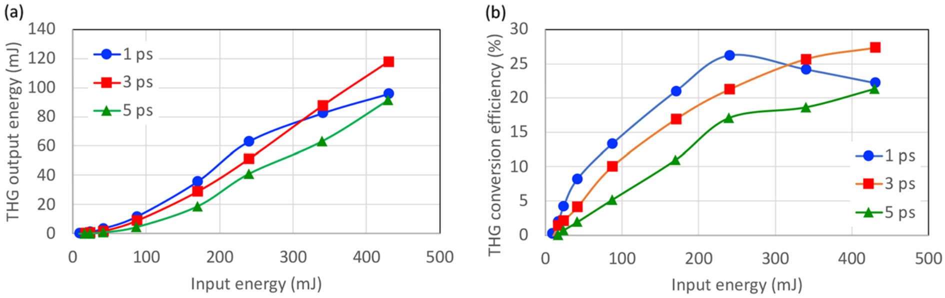

Fig. 4 **a** Output energy and **b** energy conversion efficiency of third harmonic generation by mixing the SHG with the fundamental beam in a type II phase matching configuration

photons absorption may still occur, as already observed in LBO [31]. Slight misalignments may also have contributed. On the other hand, multiphoton absorption in the UV often induces photo-damage or long-term loss of conversion efficiency [32], which was not observed in our case, even after more than 100 h of operation to date. Moreover, the smooth spectral tails show no sharp cut, excluding issues with the angular acceptance of the crystal.

Reducing the peak power by stretching the pulse is a common and efficient way of reducing the *B*-integral and, therefore, shifting the onset of filamentation to longer distances [6, 33, 34]. Indeed, due to nonlinear effects in air, such as Kerr self-focusing, a 96 GW peak power laser pulse at 343 nm would self-collapse as a bundle of filaments after only 33 m of propagation [33, 34]. Chirping the pulse to 3 ps duration shifts the saturation of the conversion efficiency to higher input energies, so that the THG beam reaches 27% of conversion efficiency (118 mJ, 40 GW peak- and 118 W average power). Further chirping the pulse to 5 ps reduces the maximum conversion efficiency to 21%.

In conclusion, we demonstrated SHG and THG conversion efficiencies of up to 59% and 27%, respectively, with a high energy, high repetition rate Yb thin disk laser system. These performances are unprecedented for laser systems simultaneously delivering high peak and average powers, in the sub-kW and sub-TW, respectively.

Acknowledgements We acknowledge the mechanical workshop of the University of Geneva for their contributions to the experimental setup.

Author contributions JPW designed the study; UA, TP, MM conceived and built the experimental setup; CH, PK, TM, KM conceived the laser source; UA, PW, VM, BM, TP, ML, LB, YBA performed the experiments; DL produced the doubling and tripling crystals; TM, KM, AM, JK, AH, JPW supervised the experiment; TP, VM, PW, JK, JPW analyzed the results; JPW and JK drafted the manuscript, All authors were given the opportunity to review the manuscript and approved it.

Funding Open access funding provided by University of Geneva. European Union Horizon 2020 Research and innovation program FET–OPEN under the grant agreement no 737033-LLR.

Data availability Data underlying the results presented in this paper are available upon request to the authors.

Declarations

Conflict of interest The authors declare no conflicts of interest.

References

1. C. Herkommer, P. Krötz, R. Jung, S. Klingebiel, C. Wandt, R. Bessing, P. Walch, T. Produit, K. Michel, D. Bauer, R. Kienberger, T. Metzger, Opt. Express **28**, 30164 (2020)
2. Y. Wang, H. Chi, C. Baumgarten, K. Dehne, A.R. Meadows, A. Davenport, G. Murray, B.A. Reagan, C.S. Menoni, J.J. Rocca, Opt. Lett. **45**, 6615 (2020)
3. P. Rambo, J. Biegert, V. Kubecek, J. Schwarz, A. Bernstein, J.-C. Diels, R. Bernstein, K. Stahlkopf, J. Opt. Tech. **66**, 194 (1999)
4. D. Comtois, C.Y. Chien, A. Desparois, F. Gérin, G. Jarry, T.W. Johnston, J.C. Kieffer, B.L. Fontaine, F. Martin, R. Mawassi, H. Pépin, F.A.M. Rizk, F. Vidal, P. Couture, H.P. Mercure, C. Potvin, A. Bondiou-Clergerie, I. Gallimberti, Appl. Phys. Lett. **76**, 819 (2000)
5. J. Kasparian, R. Ackermann, Y.-B. André, G. Méchain, G. Méjean, B. Prade, P. Rohwetter, E. Salmon, K. Stelmaszczyk, J. Yu, A. Mysyrowicz, R. Sauerbrey, L. Wöste, J.-P. Wolf, Opt. Express **16**, 5757 (2008)
6. J.-P. Wolf, Rep. Prog. Phys. **81**, 026001 (2018)
7. T. Produit, P. Walch, C. Herkommer, A. Mostajabi, M. Moret, U. Andral, A. Sunjerga, M. Azadifar, Y.-B. André, B. Mahieu, W. Haas, B. Esmiller, G. Fournier, P. Krötz, T. Metzger, K. Michel, A. Mysyrowicz, M. Rubinstein, F. Rachidi, J. Kasparian, J.-P. Wolf, A. Houard, Eur. Phys. J. Appl. Phys. **93**, 10504 (2021)
8. X.M. Zhao, J.-C. Diels, C.Y. Wang, J.M. Elizondo, IEEE J. Quantum Electron. **31**, 599 (1995)
9. E. Schubert, A. Rastegari, C. Feng, D. Mongin, B. Kamer, J. Kasparian, J.-P. Wolf, L. Arissian, J.-C. Diels, New J. Phys. **19**, 123040 (2017)
10. K. Stelmaszczyk, P. Rohwetter, G. Méjean, J. Yu, E. Salmon, J. Kasparian, R. Ackermann, J.-P. Wolf, L. Wöste, Appl. Phys. Lett. **85**, 3977 (2004)
11. H.L. Xu, W. Liu, S.L. Chin, Opt. Lett. **31**, 1540 (2006)
12. B. Franta, E. Mazur, S.K. Sundaram, Int. Mater. Rev. **63**, 227–240 (2017)
13 M. Kozlova, I. Andriyash, J. Gautier, S. Sebban, S. Smartsev, N. Jourdain, U. Chulagain, Y. Azamoum, A. Tafzi, J.P. Goddet, K. Oubrerie, C. Thaury, A. Rousse, K. Ta Phuoc, Phys. Rev. X. **10**, 011061 (2020)
14. ELI-beam L1: https://www.eli-beams.eu/facility/lasers/laser-1-allegra-100-mj-1-khz/, last accessed 4 July 2022
15. V. Krylov, A. Rebane, A.G. Kalintsev, H. Schwoerer, U.P. Wild, Opt. Lett. **20**, 198 (1995)
16. C.Y. Chien, G. Korn, J.S. Coe, J. Squier, G. Mourou, R.S. Craxton, Opt. Lett. **20**, 353 (1995)
17. S. Parker, C. Danson, D. Egan, S. Elsmere, M. Girling, E. Harvey, D. Hillier, D. Hussey, S. Masoero, J. Mc Loughlin, R. Penman, P. Treadwell, D. Winter, N. Hopps, High Power Laser Sci. Eng. **6**, e47 (2018)
18. A. Marcinkevicius, R. Tommasini, G.D. Tsakiris, K.J. Witte, E. Gaizauskas, U. Teubner, Appl. Phys. B **79**, 547 (2004)
19. C. Aparajit, K. Jana, A.D. Lad, Y.M. Ved, A. Couairon, G.R. Kumar, Opt. Lett. **46**, 3540 (2021)
20. C. Röcker, A. Loescher, F. Bienert, P. Villeval, D. Lupinski, D. Bauer, A. Killi, T. Graf, M. Abdou Ahmed, Opt. Lett. **45**, 5522 (2020)

21. C. Röcker, P. Weinert, P. Villeval, D. Lupinski, M. Delaigue, C. Hönninger, R. Weber, T. Graf, M. Abou-Ahmed, Opt. Express **30**, 5423 (2022)
22. http://www.redoptronics.com/LBO-crystal.html, last accessed 4 July 2022
23. https://eksmaoptics.com/nonlinear-and-laser-crystals/nonlinear-crystals/lithium-triborate-lbo-crystals/, last accessed 4 July 2022
24. T. Schröder, K.-J. Boller, A. Fix, R. Wallenstein, Appl. Phys. B **58**, 425 (1994)
25. A. Kobyakov, E. Schmidt, F. Lederer, J. Opt. Soc. Am. B **14**, 3242 (1997)
26. A.T. Sahakyan, A.N. Starodub, J. Phys. Conf. Ser. **1189**, 012050 (2019)
27. https://as-photonics.com/products/snlo/ — last accessed 4 July 2022
28. R. Boyd, *Nonlinear optics*, 3rd edn. (Academic Press, Cambridge, 2008)
29. J. Ma, J. Wang, P. Yuan, G. Xie, L. Qian, Chinese Opt. Lett. **15**, 021901 (2017)
30. R.H. French, J.W. Ling, F.S. Ohuchi, C.T. Chen, Phys. Rev. B **44**, 8496 (1991)
31. C. Mühlig and S. Bublitz: Absolute Absorption Measurements in Nonlinear Optical Crystals. In: Laser Congress 2019 (ASSL, LAC, LS&C), OSA Technical Digest (Optica Publishing Group, 2019), paper JTu3A.24. https://doi.org/10.1364/ASSL.2019.JTu3A.24
32. S. Möller, A. Andresen, C. Merschjann, B. Zimmermann, M. Prinz, M. Imlau, Opt. Express **15**, 7351 (2007)
33. A. Couairon, A. Mysyrowicz, Phys. Rep. **441**, 47 (2007)
34. L. Bergé, S. Skupin, R. Nuter, J. Kasparian, J.P. Wolf, Rep. Prog. Phys. **70**, 1633 (2007)