EPJ Web of Conferences **307**, 04076 (2024) https://doi.org/10.1051/epjconf/202430704076
EUROPHOTON 2024

Improved Performance of Frequency Tripling Mirrors: A Promising Approach for Simple and Efficient Third Harmonic Generation

David Zuber,[1,2] Sebastian Balendat,[1,3] Holger Badorreck,[2,3] Marco Jupé,[2,3] Detlev Ristau,[1,2,3] and Uwe Morgner[1,2,3]

[1] *Leibniz Universität Hannover, Institute of Quantum Optics, Welfengarten 1, 30167 Hannover, Germany*
[2] *Cluster of Excellence PhoenixD (Photonics, Optics, and Engineering-Innovation Across Disciplines), 30167 Hannover, Germany*
[3] *Laser Zentrum Hannover e.V., Hollerithallee 8, 30419 Hannover, Germany*
zuber@iqo.uni-hannover.de

Abstract: Frequency tripling mirrors offer a promising alternative to complicated cascaded arrangements for third harmonic generation. By incorporating optimised dielectric layers, we have achieved a remarkable conversion efficiency of 3.5 %. We will give a deep insight into FTMs and discuss applications. © 2024 The Author(s)

Nowadays, the technique of frequency tripling near-infrared lasers has gained importance as a method for generating ultrashort UV laser pulses [1]. Despite its widespread use, direct third harmonic generation (THG) has encountered challenges, particularly in achieving optimal efficiency in single crystal configurations [2, 3], mainly due to stringent phase matching requirements. This problem is particularly pronounced in 800 nm Ti:sapphire systems, where phase-matched THG is unachievable in commonly used crystals such as BBO or KDP [3], necessitating alternative solutions. The generation of the third harmonic via cascaded second-order effects represents a compromise between efficiency and extended phase-matching bandwidth on the one hand and a significantly more complex and adjustment-sensitive setups on the other.

Frequency tripling mirrors (FTMs), which consist of stacks of dielectric layers that are optimised for direct THG, represent a convincing alternative that significantly reduces the complexity of the optical setup and simplifies the conditions for phase matching [4]. In this study, we address the fundamental principles of FTM mirrors and provide both theoretical insights and experimental investigations on FTMs consisting of Hf_xAl_yO and SiO_2. Our experimental results show a conversion efficiency of 3.5 % and emphasise the potential of FTMs for frequency tripling. Furthermore, we investigate possibilities to further improve the THG efficiency and discuss potential applications of this innovative THG approach.

The principle of FTMs is illustrated in Figure 1(a), where conversion layers of high and low third-order nonlinear susceptibility materials (Hf_xAl_yO and SiO_2) are embedded between Bragg reflectors. This basic design serves as the basis for an optimisation algorithm based on a Monte Carlo approach that refines the efficiency of the FTM

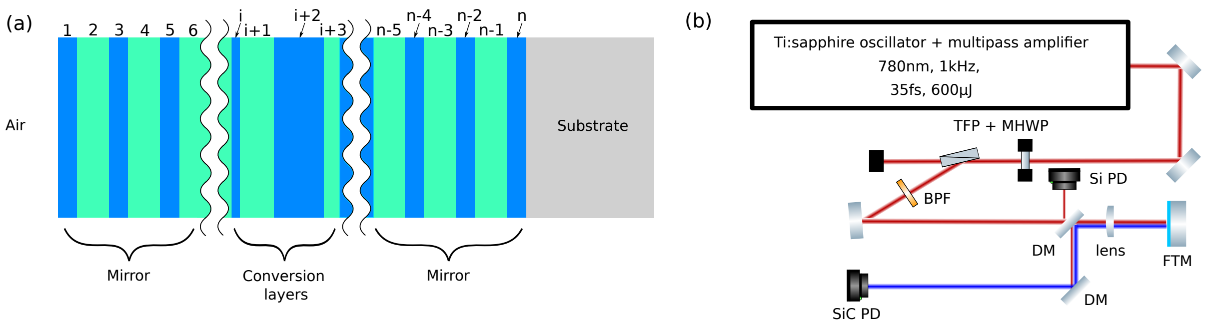

Fig. 1. (a) Basic layout of a THG mirror. A set of conversion layers is surrounded by two Bragg reflectors leading to a field enhancement inside the conversion layers. Blue corresponds to a material with high nonlinear susceptibility while green corresponds to a material with low nonlinear susceptibility. (b) Experimental setup for testing the conversion efficiency. TFP: thin film polarizer, MHWP: motorized half wave plate, BPF: spectral band-pass filter, DM: dichroic mirror, Si PD: silicon photodiode, SiC PD: silicon carbide photodiode, FTM: frequency tripling mirror.

using results from finite-difference time-domain (FDTD) simulations that take THG and dispersion into account. Compared to a single dielectric layer, the efficiency can be increased by up to five orders of magnitude [5].
FTMs are produced using the ion beam sputtering technique [6]. The experimental validation of the performance of the FTMs is performed using the setup shown in Figure 1(b), which employs a 780 nm Ti:sapphire multipass amplifier emitting 35 fs pulses. A combination of a motorised half-wave plate and a thin-film polariser enables pulse attenuation and automatic power sampling. The spectral bandwidth is reduced with a bandpass filter to match the phase-matching bandwidth of the FTMs, with the correct central bandwidth set by filter rotation. The beam is focussed on the FTM and the TH is separated from the fundamental with dichroic mirrors. In-house calibrated photodiodes capture both the fundamental and the third harmonic, allowing the TH efficiency to be measured.
Figure 2(a) shows an optimised FTM design and the electric field distribution of a pulse centred at 788 nm. The blue layers represent Hf_xAl_yO and the green layers SiO_2, while the substrate is shown in grey. The FTM design shown in Figure 2(a) has achieved a striking conversion efficiency of 3.5 %, as can be seen in Figure 2(b). However, at efficiencies around 3 %, saturation becomes apparent. In our discussion, we will address the current limitations of conversion efficiency and spectral bandwidth using insights from simulations. Furthermore, we will analyse the spatio-temporal build-up of the TH in FTMs and discuss the implications of the design shown in Figure 2(a). Moreover, we will present strategies to increase the conversion efficiency further and explore potential applications where FTMs could outperform conventional second-order cascaded approaches, such as their use in laser resonators.

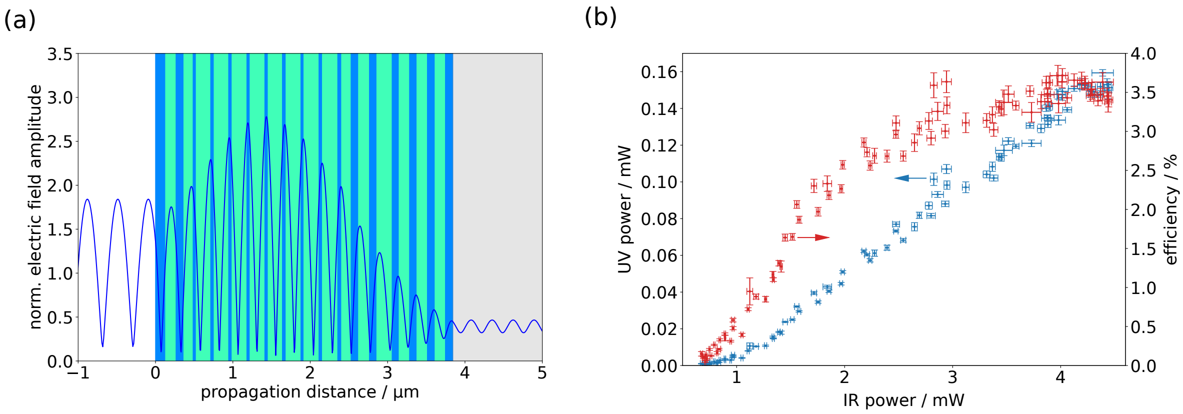

Fig. 2. (a) Electric field distribution of an fundamental pulse at 788 nm in an optimized FTM mirror. Hf_xAl_yO layers are shown in blue and SiO_2 layers in green. The substrate is depicted in grey. The electric field amplitude is normalized to the input electric field amplitude. (b) Experimentally generated UV power (blue) and conversion efficiency (red) for the FTM shown in (a). The arrows indicate the according axis of each data set.

References

1. J. Rothhardt, C. Rothhardt, M. Müller, A. Klenke, M. Kienel, S. Demmler, T. Elsmann, M. Rothhardt, J. Limpert, and A. Tünnermann, "100 W average power femtosecond laser at 343 nm," Opt. Lett. **41**, 1885–1888 (2016).
2. K. Miyata, V. Petrov, and F. Noack, "High-efficiency single-crystal third-harmonic generation in BiB_3o_6," Opt. Lett. **36**, 3627 (2011).
3. P. S. Banks, M. D. Feit, and M. D. Perry, "High-intensity third-harmonic generation," JOSA B **19**, 102–118 (2002).
4. C. Rodríguez, S. Günster, D. Ristau, and W. Rudolph, "Frequency tripling mirror," Opt. Express **23**, 31594 (2015).
5. D. Zuber, S. Kleinert, A. Tajalli, M. Steinecke, M. Jupé, I. Babushkin, D. Ristau, and U. Morgner, "Third and fifth order nonlinear susceptibilities in thin HfO_2 layers," Opt. Express **31**, 19309–19318 (2023).
6. D. Ristau and T. Gross, "Ion beam sputter coatings for laser technology," in *Advances in Optical Thin Films II,* vol. 5963 (SPIE, 2005), pp. 315–326.