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Third Harmonic Generation and $\chi^{(5)}$ Effects in Thin Gradient HfO_2 Layers

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Nowadays, frequency tripling of near infrared lasers is a typical way to generate high power, ultrashort UV pulses [1]. However, third harmonic generation (THG) suffers either from a low efficiency if the TH is driven directly in bulk crystals [2], or from a complicated and alignment sensitive setup if cascaded second order effects are used. Instead of these approaches, third harmonic generation in thin-layered structures feature a much smaller footprint and relaxed phase matching condition [3]. There are also clear indications that their biggest drawback, the low conversion efficiency typically caused by the small interaction length in the layers, will be overcome in near future. Recently, a conversion efficiency of 1.8% had been achieved for a multilayer system with Hafnia as nonlinear material component [4]. Besides a careful optimization of the layer structure, also the chosen materials and their nonlinear properties play an important role. In this work, we present theoretical and experimental studies of the nonlinear properties of ion beam sputtered HfO_2-layers. To get a better insight into the THG effects, the material is deposited as a gradient layer with continuously varying thickness from 200 nm to 1500 nm. A home built femtosecond laser system at 1030 nm [5] is focused onto the sample (see Fig. 1(a)), and the TH is recorded as a function of the layer thickness. In parallel, FDTD simulations based on the software 'Lumerical' are performed, including linear and third order nonlinear effects of the layer and the substrate material. The results are shown in Fig. 1(b). Simulation and experiment show good agreement and correctly reflect the interference effects caused by both, the fundamental and the third harmonic wave, as well as linear and nonlinear influences caused by the substrate.
The experiments were performed at intensities in the range of several hundred GW/cm^2. By increasing the intensity, the TH signal increases stronger than expected for a third-order effect. While this effect can partially be attributed to self-focussing in the substrate, also further effects seems to have an influence on the THG. After careful consideration of different linear and nonlinear effects, THG by fifth order nonlinearity ($3\omega = \omega + \omega + \omega + \omega - \omega$) seems to be the most likely explanation. By comparison of the experimentally measured TH to the simulations, values of $\chi^{(3)} = (1.0 \pm 0.17) \times 10^{-21}\, m^2/V^2$ and $\chi^{(5)} = (3.1 \pm 1.9) \times 10^{-40}\, m^4/V^4$ are obtained. Up to the authors knowledge, this is the first time a value of the fifth order nonlinear susceptibility is derived for dielectric coating materials. We believe that this detailed study of involved effects on the THG in thin layers will lead to a new generation of multi-layered structures for THG with even higher conversion efficiencies.

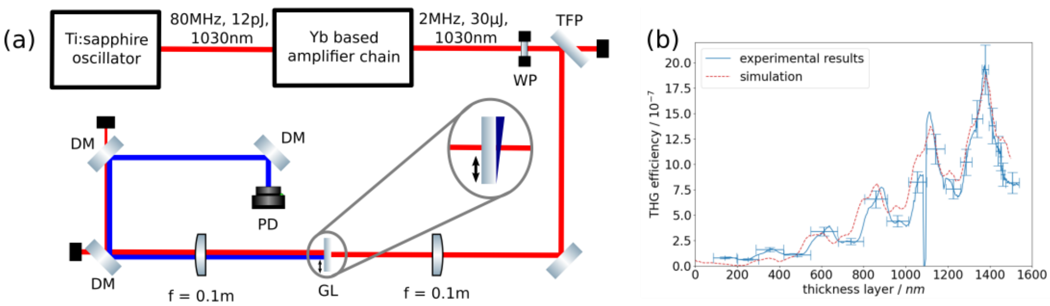

Fig. 1 (a) Experimental setup. WP: waveplate, TFP: thin film polarizer, GL: gradient layer, DM: dichroic mirror, PD: photodiode. (b) Experimentally measured (blue, solid) and simulated (red, dashed) THG efficiencies obtained for different layer thickness under consideration of linear and nonlinear effects in both, the layers material and the substrate.

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