Third Harmonic Generation in Polycrystalline Anatase Titanium Dioxide Nanowaveguides

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Abstract: We experimentally demonstrate third-harmonic generation in polycrystalline anatase titanium dioxide nano-waveguides, using ultrashort optical pulses centered around 1550 nm. Phase matching is achieved using higher order optical modes at the third harmonic wavelength.

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1. Introduction

Over the last few years titanium dioxide (TiO_2) has been gaining popularity as a new material for nonlinear-optical applications around 1550 nm, due to its wide transparency, high linear and non-linear refractive indices, and electronic bandgap that does not support two-photon absorption in the telecommunications C-band range. Recently, nano-waveguides from polycrystalline TiO_2 with losses around 6-8 dB/cm in the C-band have been successfully demonstrated [1]. The optical Kerr coefficient of such waveguides was measured to be 1.6×10^{-14} cm^2/W [2], similar to silicon nitride, which makes TiO_2 an interesting new material for integrated nonlinear optics. In this work we present a first to our knowledge demonstration of phase-matched third harmonic generation in TiO_2 waveguides from telecom wavelengths to the visible.

2. Fabrication/Experimental Setup

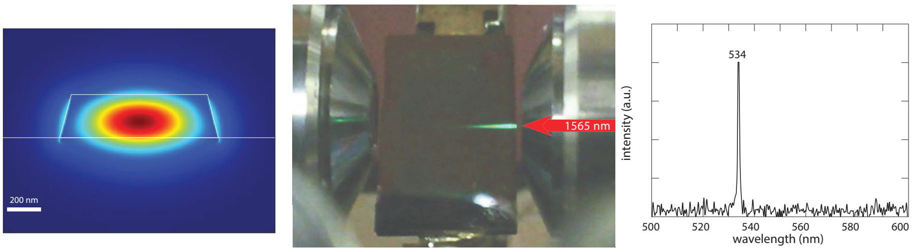
TiO_2 waveguides used for this study were fabricated using standard top-down e-beam lithography in which TiO_2 layer was deposited using reactive RF magnetron sputtering on top of an SiO_2 layer on a silicon substrate, and overclad with transparent fluoropolymer. Several waveguide geometries were fabricated, with dimensions $250 \times 600, 700, 800$ and 900nm, with a sidewall angle of 75°, and overall length of 9.2mm. The pump light source used was an optical parametric oscillator (OPO), with 80 MHz repetition rate, and 180 fs pulse duration at 1550 nm. The central wavelength of the laser was tuned from 1440 to 1560 nm, which also varied its spectral bandwidth from 10-31nm. After passing through a polarizing beam splitter and half-wave plate, the light was coupled into the waveguide using a free-space objective. Third harmonic light emitted upwards from the waveguide was collected using a multimode fiber probe located directly above the waveguide, followed by a spectrometer.

3. Theory

The efficiency of third harmonic generation is a function of the phase mismatch between the fundamental and the third harmonic signals, which is given as $\Delta\beta = \beta_s - 3\beta_p$, where β_s and β_p are the momenta of the signal and the pump photon, respectively. This implies that the effective refractive indices of the fundamental and the third harmonic signals must be the same. However, due to strong material dispersion, the effective index at the third harmonic wavelength is usually significantly lower than that of the fundamental, creating a need for an alternative phase matching method. In this work we use a multimode phase matching scheme, which has been demonstrated for photonic crystal devices [3]. In high index contrast devices, waveguide dispersion dominates over the material dispersion, allowing higher order modes at the third harmonic wavelength to have the same effective index as the fundamental mode at the pump wavelength. In order to check multimode phase matching conditions for our devices, we calculate the dispersion for all guided modes from 400 nm to the cutoff wavelength, using a finite difference eigenmode solver. For each mode pair, we calculate the phase mismatch (details of these simulations are reported elsewhere [4]), and find the strongest THG signal to be expected at 518.5 nm, for 900nm-wide waveguide. For 600, and 700 nm waveguides, the simulations did not show any phase-matched wavelengths corresponding to the available pump spectrum. For a 800nm-wide waveguide, the phase-matched mode was found to be orthogonally polarized to the fundamental one, preventing efficient THG process.

4. Results/Discussion

Figure 1 (b) shows visible third harmonic light emitted from a 900nm-wide TiO_2 waveguide, when pumped with 360mW of average power centered at 1565nm. The spectrum of the third harmonic light is shown in Figure 1 (c). This experiment was repeated for 600, 700, and 800 nm waveguides, where small amounts of green light were visible at the input facet, but no significant third harmonic light was measured, confirming our simulations based on multimode phase matching.



(a) (b) (c)

Fig. 1. (a) Electric field mode profile for anatase TiO_2 waveguide; (b) visible third harmonic generation for 1565 nm pump wavelength; (c) Measured THG spectrum.

Figure 2 shows the third harmonic spectrum vs fundamental wavelength for the 900 nm wide waveguide with a silica top-cladding. Here, we tuned the central OPO wavelength from 1440 to 1560 nm. The third harmonic spectrum is plotted vertically as an intensity plot for each pump wavelength; the energy conservation line (λ_S=λ_p/3) is added for clarity, and the yellow dots indicate the peak of each THG spectrum. We note that, since the OPO power changes greatly as a function of wavelength, we kept the input at 20 mW for each central pump wavelength for consistency. One of the immediate features of Figure 2 is very low intensity of the third harmonic signal for wavelengths below 1500nm, which corresponds to a non-phase-matched THG. For the 1500-1550nm range, we observe a much stronger third harmonic signal, and dlight shifts from the energy conservation line. The strongest signal corresponds to a phase-matched THG, and the offset indicates phase-matched wavelength of 509nm, which is close to our calculated value. The spectral broadening of the pump signal, and fabrication tolerances (5%) may contribute to the differences between expected and experimental values of the THG signal.

In conclusion, we have experimentally demonstrated phase-matched THG from anatase TiO_2 waveguides, where phase matching is achieved using higher order modes at the third harmonic wavelength. This work opens the possibilities to engineer TiO_2-based nanowaveguides for frequency conversion applications.

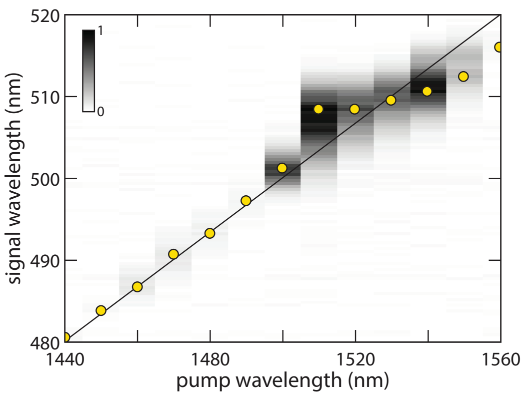


Fig. 2. Third-harmonic signal versus pump wavelength.

References

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