Phase-Matched Second-Harmonic Generation from Metasurfaces Inside Multipass Cells

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Abstract. We demonstrate a scalable approach to increase conversion efficiencies of nonlinear metasurfaces by incorporating them into multipass cells and allowing the pump beam to pass several times through the metasurfaces. As a proof of principle, we achieve phase matching of the second-harmonic generation (SHG) signal with superlinear dependence on the number of passes. We measure an order of magnitude enhancement in the SHG signal when the incident pump traverses the metasurface up to 9 passes. The generic nature of our approach holds promise for diverse applications in nonlinear optics.

1 Introduction

Nonlinear metasurfaces and metamaterials have recently emerged as a promising solution to realize thin nonlinear devices. These artificial structures, composed of subwavelength building units, can exhibit properties not (easily) achieved in natural materials. Despite their advantages, and tremendous progress of the field, nonlinear metamaterials have not yet found real-world nonlinear applications, where they could outperform existing nonlinear material systems, such as nonlinear waveguides [1], fibers [2], or crystals [3]. This limitation primarily stems from their still-insufficient conversion efficiencies.

In this work, we introduce a novel and straightforward approach to enhance nonlinear responses of metamaterials, thereby improving their conversion efficiencies. Our method involves incorporating metamaterials inside multipass cells, allowing the pump beam to pass through the metamaterial several times. We experimentally demonstrate a remarkable tenfold enhancement in the SHG signal from a plasmonic metasurface after 9 passes through the metasurface [4]. Notably, our results show a superlinear dependence of the SHG signal on the number of passes *N*, confirming successful phase matching. This demonstrated approach is quite generic, offering compatibility with various existing enhancement techniques and metamaterials.

2 Measurements

In our experimental setup, the metasurface is positioned between two mirrors, forming the multipass cell configuration as illustrated in **Fig. 1**. This configuration enabled successive interactions of the pump beam with the metamaterial, enhancing the nonlinear response. A tunable titanium sapphire femtosecond laser (Chameleon

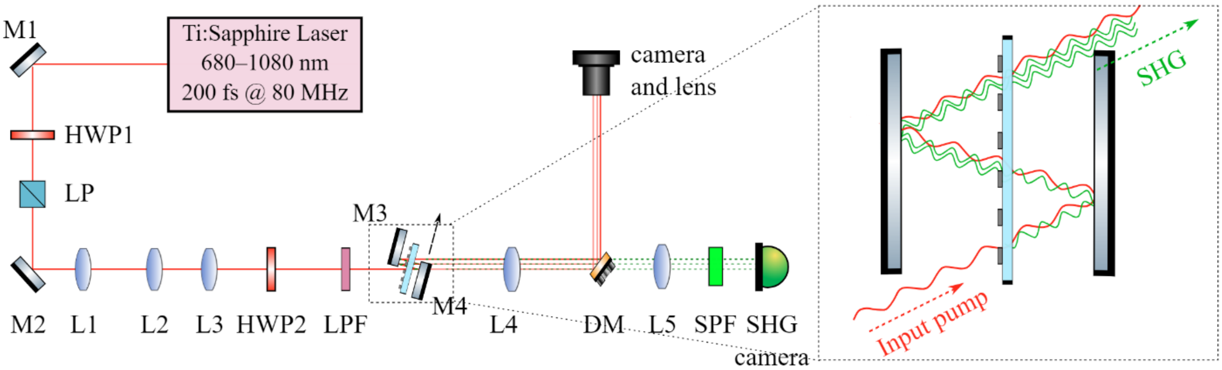

Fig. 1. A schematic of the setup used to measure SHG response of the sample at different number of passes *N*. The inset on the right shows the phase-matched SHG emission in case of 3 passes of the pump beam through the metasurface device.

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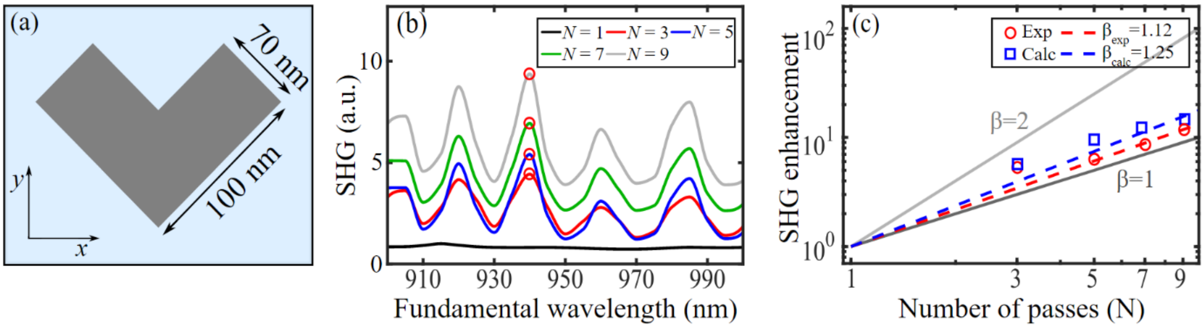

Fig. 2. (a) A schematic representing a single nanoparticle from the metasurface. (b) Measured SHG signal for different number of passes *N*. (c) The enhancement factor of the SHG signal as a function of the number of passes *N*.

Vision II), operating at an 80 MHz repetition rate and offering a spectral range of 680–1080 nm with pulse duration of 140 fs (near 800 nm), was used as the pump source.

We used a sample consisting of V-shaped aluminium L-shaped nanoparticles (illustrated in **Fig. 2** (a)) deposited on a 0.5 mm-thick SiO_2 substrate. The nanoparticles were positioned randomly (being however similarly oriented) with a particle density of 11.11 particles $/\mu m^2$. The sample design was optimized to exhibit localized surface plasmon resonance (LSPR) around 420 nm for x-polarized incident light, which was confirmed by transmission measurements [4]. We utilized the optical phase shifts induced by the LSPR to phase match SHG signal inside the multipass cell. The phase-matching condition was fulfilled for numerous pump wavelengths in our pump wavelength range from 900 to 1000 nm.

3 Results and discussions

Our semi-analytical model predicted an enhanced SHG response at specific wavelengths [5]. This trend was validated by experiments [**Fig. 2** (b)], where we measured the SHG response at varied pump wavelengths (900–1000 nm) and number of passes (up to N = 9).The observed phase-matched wavelengths were found to be 904 nm, 920 nm, 940 nm, 960 nm, and 985 nm, closely aligned with the calculated values, confirming the accuracy of our model. Notably, the SHG response exhibited a superlinear dependence ($\mathrm{SHG} \propto N^{\beta_{\mathrm{exp}}}$, where $\beta_{\mathrm{exp}} = 1.12$) on the number of passes N as illustrated in **Fig. 2** (c), confirming successful phase matching. In the ideal scenario of a lossless and perfectly phase-matched sample, the SHG signal would exhibit a quadratic dependence on the number of passes N ($\mathrm{SHG} \propto N^{\beta_{\mathrm{ideal}}}$, where $\beta_{\mathrm{ideal}}= 2$) [6]. However, various loss mechanisms, including transmission losses and beam intensity changes during propagation, result in a measured scaling factor below 2. Moreover, the bandwidth of the input pump ($\Delta\lambda_{\mathrm{FWHM}}$ = 9.5 nm near 950 nm) affects the SHG power dependence. Since the sample is not phase matched for all wavelengths within the pump bandwidth, the observed SHG response is affected.

4 Conclusions

We have performed a proof-of-principle demonstration how nonlinear responses of metasurfaces could be enhanced through implementation of a straightforward multipass configuration. By allowing the pump beam to interact with the metasurface multiple times, we achieved an order of magnitude enhancement in SHG signal. Notably, our study introduces straightforward strategy for achieving phase matching of the SHG signal within nonlinear metamaterials, eliminating the need for complex fabrication procedures associated with traditional phase-matched stacked metasurfaces.

In future, we plan to fabricate metasurfaces directly on top of the mirrors or replace the mirrors with highly-reflecting metasurfaces. These approaches aim to reduce the normal dispersion, simultaneously increasing the achievable enhancement factor β. Additionally, we plan to incorporate curved (convex) mirrors within our multipass cell to replace the flat mirrors, effectively realizing a configuration similar to a conventional Herriot cell [7]. This adjustment aims to extend the Rayleigh range of the laser beam, thereby allowing more passes of the beam through the metasurface. Such demonstrations hold promise for the development of ultra-thin, tunable, and multifunctional nonlinear devices, eliminating the need for complex fabrication procedures.

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