Si-on-Sapphire Metasurfaces for High Harmonic Generation and Laser Machining

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Abstract:

We used a silicon-on-sapphire (SOS) platform to fabricate several metasurfaces for nonlinear optical experiments in the mid-infrared (MIR). One series of experiments aims to use the metasurfaces for high-efficiency third harmonic generation (THG) and the generation of higher-order harmonics (HHG). Another one aims to use high laser irradiance and the localized hotspots that form within nanostructures to perform nanoscale machining by accurately controlling laser-induced damage. All samples were fabricated by electron-beam lithography (EBL) performed with the JEOL 9500, e-beam deposition of a chromium mask in one of the SC-4500 evaporators, and HBr etching in the Oxford Cobra ICP etcher.

Summary of Research:

We work in nonlinear optics with sources in the MIR, and our aim is the efficient generation of optical harmonics — light with a multiple of the original MIR frequency [1]. Harmonics generation in solid-state materials is commonly used in laser systems, and has promising applications both in the sciences (such as for solid-state attosecond lasers) and in everyday life (for example in telecommunications) [2].

Nonlinear metasurfaces, planar arrays of thin subwavelength structures, have emerged as a platform for nonlinear generation and nonlinear light control [3,4]. Their parameters, such as geometric properties, refractive index and local phase profile can be adjusted to perform, in a compact form factor, functions that ordinarily require bulky materials or gases [5].

Our metasurfaces are based on dielectric resonators, rectangular silicon structures of dimensions w_{y}, w_{y}, w_{z} arranged in a lattice of period p_{y} , p_{y} . Adjusting these dimensions controls the resonance spectral position as well as its width, both of which must match experimental requirements. At resonance, greatly increased coupling of the fundamental light to the metasurfaces increases the amount of energy available for nonlinear processes, resulting in much increased nonlinear generation. It also changes the energy distribution of the electrons in the material, potentially with a time dependence, which, if correctly exploited, can further enhance the emission of harmonics. The result is, potentially, not only highly efficient HG in a compact, ultrathin system, but also the emission of higher-order harmonics that are normally obtained in gases.

Usually, suitable periods are a little less than the wavelength, x/y dimensions are smaller than that, and the thickness of the structures is even smaller. Deeply subwavelength thicknesses relax phase matching conditions for the generation of harmonics and prevent the harmonics themselves from being excessively reabsorbed.

We fabricate our samples starting from commercially available silicon-on-sapphire substrates. Our structures are arranged in square or rectangular arrays a few hundred μ m to 1 mm to the side. We fabricate several such arrays in a grid on each sample, each with a slightly differing pattern or rescaled by a certain amount, to give a distribution of resonance wavelengths, allowing us to compare resonant and nonresonant behavior and account for slight deviations during fabrication.

Each substrate, etched down to the correct thickness in the Oxford Cobra ICP etcher with HBr, was coated in PMMA 495 A4 on a spinner, baked on the hot plates, and exposed in the JEOL 9500 EBL machine. The pattern was developed with 1:3 MIBK:IPA and a subsequent IPA wash. A Cr mask 40-50 nm thick was then deposited with one of the SC-4500 evaporators. Lift-off followed with sonication in acetone, leaving a patterned Cr mask on the bare Si. Etching around the mask down to the sapphire was again done with the Oxford Cobra. The mask was finally removed with chromium etchant in a wet bench.

This year, we fabricated several series of samples, which have been used to study THG with chirp-dependent



Figure 1: SEM detail of a Si nonlinear metasurface, a $1.16 \times 1.16 \mu m^2$ array of $0.99 \times 0.75 \times 0.3 \mu m^3$ structures.



Figure 3: Transmittance FTIR spectra of the laser machining metasurface. The array detail in Figure 3 is "B1" in the legend. Inset: polarization of the incident wave.

behavior induced by tuning the chirp of the incident pulses as well as HHG experiments. An example of a metasurface for HG is shown in Figure 1.

All HG metasurfaces are simple rectangular resonators, because there is no particular need for a more complex shape for the purposes of the project. The resonator shape, however, can be used to increase field concentration in a particular spot of the resonator. Sharp features or restriction points create hot spots, which are used extensively in nanoscale resonators. The high field concentration may cause damage, usually an unwanted problem, which we want to exploit to machine coupled resonators separated by very small gaps.

We fabricated square resonators with notches (Figure 2), then used laser pulses to cause the structures to break at the notch position, where field concentration is the highest. The resonant behavior of these structures pre-machining, which exemplifies that of all samples in this report, is shown in Figure 3.

The intended outcome is shown in a preliminary result, separately obtained on an older sample, in Figure 4.



Figure 2: SEM detail of a Si laser machining metasurface, a $1.94 \times 1.95 \ \mu\text{m}^2$ array of $1.08 \times 1.15 \times 0.3 \ \mu\text{m}^3$ structures. The notch is 0.72 $\ \mu\text{m}$ deep.



Figure 4: Early example of laser machining shown on a previously fabricated sample. Image credit: Melissa Bosch.

Conclusions and Outlook:

We have successfully used a SOS platform to fabricate a number of Si resonator meta-surfaces, which we have used for a variety of nonlinear experiments in the MIR. We will continue to develop the platform to reach higher harmonic orders and greater efficiency. We have also developed a promising platform for nanoscale laser machining, whose behavior we will study and improve as necessary to achieve small, well defined and controlled features.

References:

- [1] Boyd, R. W., "Nonlinear Optics", Academic Press, 2008.
- [2] Garmire, E., "Nonlinear optics in daily life", Opt. Express, 21(25), 30532, 2003.
- [3] G. Li, S. Zhang, and T. Zentgraf, "Nonlinear photonic metasurfaces," Nat. Rev. Mater., vol. 2, no. 5, p. 17010, 2017.
- [4] A. Krasnok, M. Tymchenko, and A. Al "Nonlinear metasurfaces: a paradigm shift in nonlinear optics," Mater. Today, vol. 21, no. 1, pp. 8-21, Jan. 2018.
- [5] M. R. Shcherbakov, K. Werner, Z. Fan, N. Talisa, E. Chowdhury, and G. Shvets, "Photon acceleration and tunable broadband harmonics generation in nonlinear time-dependent metasurfaces," Nat. Commun., vol. 10, no. 1, p. 1345, 2019.