Studying the Interplay of Electric and Magnetic Resonance-Enhanced Second Harmonic Generation: Theory and Experiments

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Abstract: We present an experimental study of a metasurface, which exhibits electric and magnetic resonances, in order to understand their independent contributions to second-harmonic generation. A hydrodynamic model framework is used to match experimental results.

OCIS Codes: (190.0190) Nonlinear Optics; (160.3918) Metamaterials; (190.2620) Harmonic Generation and Mixing

1. Introduction

Since the advent of nanofabrication, one area of nonlinear optics that has been gaining significant interest is second harmonic generation (SHG) enhanced by resonant nanostructures. Both electric and magnetic resonances have been studied, though separately, in metallic nanostructures, such as nanoantennas [1], and split-ring resonators [2,3] among others. While it has been widely shown that enhanced local fields can boost SHG, it is of great interest to study a nanostructure where both types of field enhancements are employed simultaneously and direct comparison of their contributions to SHG is possible.

A metasurface – a nanostructured 2D metamaterial film – arranged as a single-period array of coupled metal nanostrips has been shown to support both symmetric and anti-symmetric displacement current flow, hence exhibiting electric and magnetic resonances for TM polarized light [4]. These nanostructures have been termed as "metamagnetics" due to their optical magnetic response. We have already studied these structures to simulate nonlinear optical properties as well as numerically and experimentally explore the effect of roughness [5, 6]. Here, this type of metasurface is taken as a well-suited object for generating and studying SHG in metals.

We have already studied SHG from metal-based nanoresonators, describing the nonlinear optical response by a hydrodynamic model [7]. Modeling the Thomas-Fermi screening length of metal as an electron fluid, we were able to provide a cohesive understanding of the source of SHG and also match previously published experimental results. Using this theoretical framework, we have optimized the metasurface to achieve electric and magnetic resonances simultaneously and exhibit SHG for 45° incidence. Here, we also use the hydrodynamic model framework to confirm our experimental results.

2. Experimental and Simulation Results

Two metasurfaces are used in this study – Figure 1b shows the schematic for the structures and Figure 1a shows an SEM image of one structure. The experimental and simulated transmission spectra for TM polarized normal incident illumination are shown in Figure 1c. Both metasurfaces have an electric resonance at 570 nm, so that an ideal matching

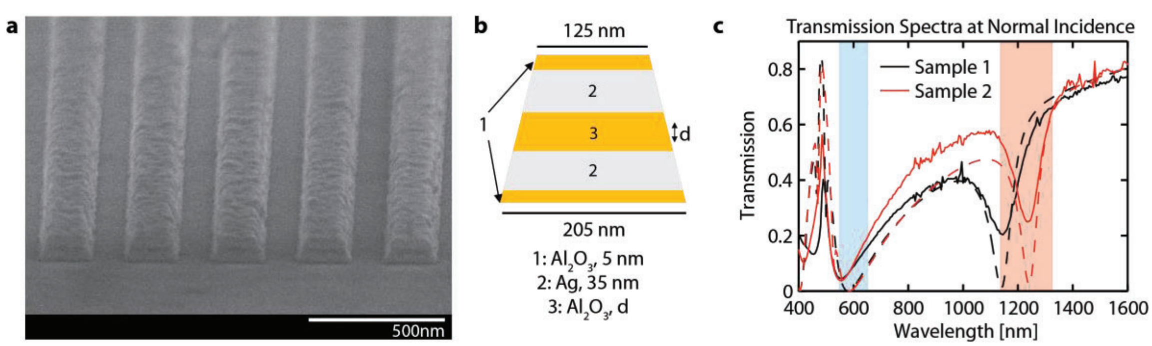

Fig 1. (a) SEM image of metasurface, (b) Schematic of sample cross section, d = 18 nm for Sample 1 and 14 nm for Sample 2, (c) Experimental (solid) and simulated (dotted) transmission spectra of Samples 1 and 2 at normal incidence.

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fundamental wavelength is 1140 nm; the first sample (Sample 1) has a magnetic resonances at 1140 nm, and the second (Sample 2) having a resonance at 1240 nm. While the magnetic resonance of Sample 1 is matched with the fundamental of the electric resonance, the magnetic resonance of Sample 2 is slightly detuned in order to probe each resonance individually. We achieve broad tunability in the magnetic resonance wavelength by simply changing the thickness of the alumina spacer layer between the silver nanostrips. A 5 nm silica layer is used to overcoat the structure to protect silver from oxidation. By using two samples with the same electric resonance, we mitigate any sample-to-sample error in the SH signal, and therefore we can study the interaction of magnetic and electric resonances. The red and blue regions in Figure 1c depict the excitation and emission wavelength ranges respectively.

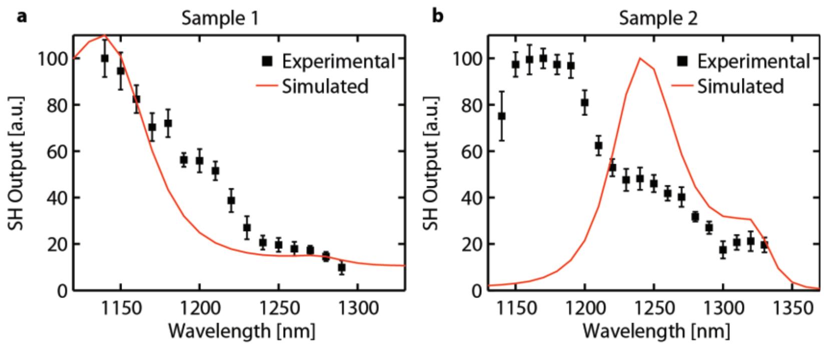

Fig 2. Simulated and experimental results for transmitted second harmonic output from (a) Sample 1 and (b) Sample 2.

Figure 2a and 2b show the experimental and simulated SH output from Sample 1 and Sample 2 as a function of wavelength. Both wavelength scans were conducted with 5mW pump power from a femtosecond Ti:Sapphire laser fed into an Optical Parametric Amplifier (OPA). For Sample 1, we observe a relative maximum at 1140 nm, which is where the fundamental for the electric resonance and magnetic resonances are matched. While experiments were only conducted down to 1140nm due to technical difficulties, simulations confirm that the maximum for SH output is at 1140 nm for Sample 1.

For Sample 2, where the resonances are slightly detuned, we clearly observe peaks for both electric and magnetic resonances in the experimental results. The SH signal observed from the fundamental of the electric resonance is about two times stronger than its magnetic counterpart. We conclude though that SHG has a palpable dependence on the fundamental magnetic resonance. The hydrodynamic model provides a different result for the SH output for Sample 2 - there still are two distinct peaks, but the entire spectrum is redshifted by ~100nm. While this could be attributed to various physical phenomena, such as roughness or charge accumulation at the silver boundaries, we are not certain of the cause of the disparity at this time. By adding such physical phenomena to hydrodynamic model, we intend to understand how they can affect the SHG process in metallic particles.

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