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SAND2019-15325C

All-Dielectric Intersubband Polaritonic Metasurface with Giant Second-Order Nonlinear Response

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Abstract: We demonstrate an extremely nonlinear all-dielectric metasurface that employs intersubband polaritons to achieve a second-harmonic conversion coefficient of 3 mW/W^2, and second-harmonic power conversion efficiency of 0.045% at a modest pump intensity of 6.7 kW/cm^2.

OCIS codes: (190.0190) Nonlinear optics; (160.3918) Metamaterials; (190.4360) Nonlinear optics, devices

1. Introduction

Dielectric metasurfaces have recently gained a lot of interest for their potential in numerous applications ranging from ultrathin optical elements and holography, to nonlinear optical devices with relaxed phase matching requirements. Compared to their plasmonic counterparts, which are intrinsically lossy at optical frequencies and have low optical damage thresholds, dielectric nanostructures exhibit ultralow losses and large optical damage thresholds. They are therefore extremely attractive candidates for use in nonlinear optical devices where loss can be detrimental and fundamental beams need to be intense.

Current design strategies for nonlinear dielectric metasurfaces for second harmonic (SH) generation have focused on fabricating metasurfaces from III-V semiconductor materials such as GaAs and/or AlGaAs which have relatively large bulk second order nonlinearity $\chi^{(2)}$ on the order of 100 pm/V [1]. Since the nonlinear response of such materials cannot be significantly engineered, efforts to increase SH generation efficiency have mostly focused on designing photonic modes in dielectric resonators with high quality factors such as Fano modes or quasi-bound states in continuum [1, 2]. Although fundamentally interesting, these approaches have only increased the SH generation efficiency to ~ 10^{-3} mW/W^2 [2]. In this work, we demonstrate an alternative approach to dielectric nonlinear metasurface design that utilizes a polaritonic dielectric metasurface in which we couple engineered *resonant second order nonlinearities* with *Mie modes* in dielectric resonators. The large nonlinearity is generated using intersubband transitions (ISBT) of III-V semiconductor quantum wells (QWs) embedded within the Mie resonator. Using our approach, we measure a maximum second-harmonic conversion coefficient of 3 mW/W^2, and a maximum second-harmonic power conversion efficiency of 0.045 % at a peak pump intensity of only 6.7 kW/cm^2. The power conversion efficiency that we achieve using our all-dielectric platform is comparable to the record high conversion efficiencies demonstrated using plasmonic metasurfaces coupled to ISBTs [3].

2. Design and Fabrication of the Nonlinear Metasurface

The fabricated nonlinear dielectric metasurface comprises Mie resonators made from III-V semiconductors with *n*-doped QWs (Fig. 1(a)). The approach involved transferring of engineered $In_{0.53}Ga_{0.47}As/Al_{0.52}In_{0.48}As$ quantum heterostructures which are designed to have electronic ISBTs at both the fundamental (~7.8 μm) and second harmonic (~ 3.9 μm) wavelengths (Fig. 1(b)) to a lower refractive index transparent substrate, followed by fabrication of cylindrical Mie resonators using electron beam lithography and etching. Resonators of different radii were fabricated to spectrally scale the magnetic dipole (MD) Mie resonance to the fundamental wavelength (Fig 1(c)). We chose the lowest order MD mode since it can be excited at normal incidence, and it allows us to make the resonators as small as possible. Furthermore, since the MD mode has strong E_z components (Fig. 1(d)), and selection rules dictate that only photonic modes with E_z components can couple to the ISBTs, we can generate the nonlinear response by introducing polaritonic coupling at the fundamental wavelength between the MD and the ISBT.

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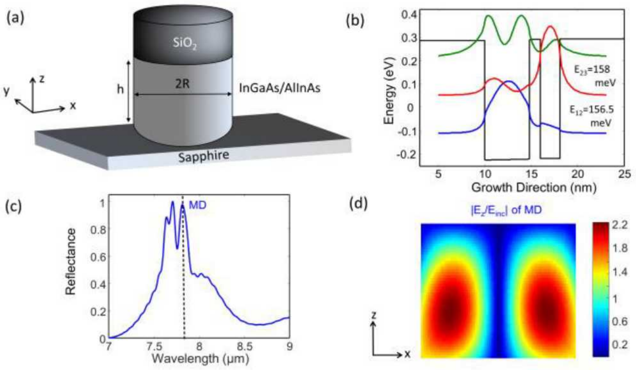

Fig. 1 (a) Schematic of a unit cell of the nonlinear metasurface. (b) 8 band k.p band structure calculation of a single period of the doubly resonant $In_{0.53}Ga_{0.47}As/Al_{0.52}In_{0.48}As$ multi-QW heterostructure. The thicknesses of the layers ($Al_{0.52}In_{0.48}As$ /$In_{0.53}Ga_{0.47}As/Al_{0.52}In_{0.48}As$/ $In_{0.53}Ga_{0.47}As$/ $Al_{0.52}In_{0.48}As$) are 10/5/1.2/2.2/10 (all in nm). (c) Finite-difference-time domain calculation of linear reflectance from a metasurface with cylinder of radius/height (R/h) = 0.87 where h =1.5 µm. This scale factor aligns MD resonance with the ISBT at the fundamental wavelength shown by black dashed line. (d) Numerically calculated magnitude of field enhancement $|E_z/E_{inc}|$ inside the resonator at the MD resonance wavelength.

3. Experimental Results

Figure 2(a) shows experimentally measured normalized linear reflectance spectrum of metasurfaces consisting of resonators of different radii (h is equal to 1.5 µm for all the resonators). When the MD resonance spectrally overlaps with the ISBT, as expected, we observe a splitting of the original MD resonance due to polaritonic coupling. Figure 2(b) shows experimentally measured reflected SH signal of these Mie metasurfaces as a function of pump wavelength. The maximum SH generation efficiency is observed for the metasurface with resonators of scale factor R/h = 0.8. The maximum SH generation efficiency is observed at $\lambda \approx 7.5$ µm which is slightly shorter than the designed resonant wavelength of ISBT. This is likely because the field enhancement inside the resonators is larger at these wavelengths. In Fig. 2(c), we plot the experimentally measured maximum reflected SH power as a function of the square of the peak incident pump power at $\lambda \approx 7.5$ µm for the metasurface with R/h =0.8. The SH nonlinear conversion factor determined from the slopes of the curves can be as large as 3 mW/W^2. Furthermore, at incident peak pump intensity of 6.7 kW/cm^2, the second-harmonic power conversion efficiency defined as the ratio of SH power to the peak pump power is measured to be as large as 0.045 %. This is comparable to the record high conversion efficiencies demonstrated using plasmonic metasurfaces coupled to ISBTs [3]. To conclude, we have demonstrated an all-dielectric polaritonic nonlinear metasurface with nonlinear conversion efficiency that is orders of magnitude larger compared to any all-dielectric metasurface reported till date.

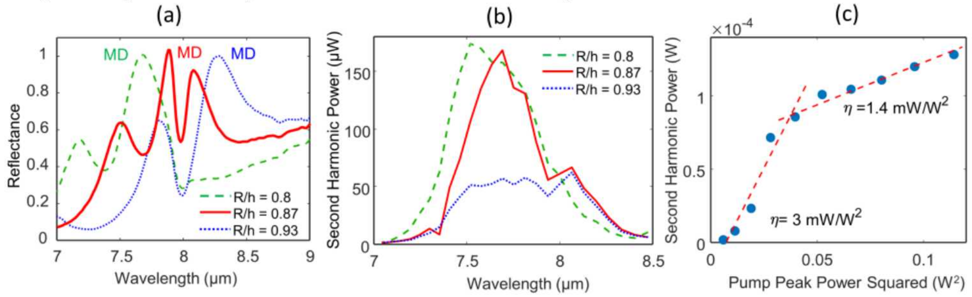

Fig. 2 (a) Experimentally measured linear reflectance spectrum of metasurfaces with Mie resonators of different radii. The magnetic dipole (MD) resonances for the different scale factors are shown in the figure. A splitting of MD resonance is observed when it spectrally aligns with ISBT. (b) Experimentally measured reflected SH signal from metasurfaces with Mie resonators of different radii. The maximum SH signal is observed at $\lambda \approx 7.5$ µm. (c) Experimentally measured reflected SH power as a function of square of the incident peak pump power for the metasurface with R/h = 0.8 at $\lambda = 7.5$ µm (blue dots). The conversion factors given by the slopes of the linear fits (red dashed lines) are stated in the figure.

This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering and performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government. The University of Texas group acknowledges support from the DARPA NASCENT program.

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