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Efficient second harmonic generation from GaAs all-dielectric metasurfaces

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Abstract: We experimentally observe large enhancement of second-harmonic generation (SHG) from GaAs metasurfaces. The SHG polarization when excited at the electric and magnetic dipole resonances is orthogonal and can be attributed to different nonlinear generation mechanisms.
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1. Introduction

Nonlinear optics in nano-structured materials offers possibilities for efficiency enhancement due to strong field confinement and resonant enhancement. Nonlinear optical second- and third-harmonic generations have been extensively studied using plasmonic nanoparticles and their assemblies. Recently, dielectric metasurfaces have emerged as an alternative to metallic metasurfaces due to their much lower loss. Additionally efficient third harmonic generation [1] has been demonstrated recently using metasurfaces created from silicon nanodisk arrays. However, due to the centrosymmetric crystal structure of silicon, second-order nonlinearities were not observed. Here we show efficient second-harmonic generation (SHG) from GaAs metasurfaces by taking advantage of the large second-order nonlinearity in these materials (~200 pm/V) and the resonant enhancement obtained when exciting low order Mie modes. Our demonstration opens up an opportunity for other efficient second order processes such as sum frequency generation, difference frequency generation, parametric down conversion, etc.

2. Resonance enhanced SHG in GaAs dielectric metasurfaces

The nonlinear metasurface consists of a square lattice of GaAs nanodisk resonators sitting on an AlGaO spacer layer. The different GaAs and AlGaAs nanolayers were grown using molecular beam epitaxy. After electron-beam lithography and dry etching, the AlGaAs layer was oxidized to form the low refractive index underlayer (AlGaO, n~1.6) [2]. Figure 1(a) shows a scanning electron microscope image of the GaAs resonators at a 75 degree side view. We measured the reflectivity spectrum of our sample and observed two peaks corresponding to the magnetic and electric dipole resonances (inset of Fig. 1(a)). We then measured the pump wavelength dependent SHG using a tunable Ti:sapphire femtosecond laser as a pump. Fig. 1(b) shows that the SHG power peaks around the magnetic and electric dipole resonances due to the resonantly enhanced electromagnetic field (~40 times at its peak value). Note that the SHG measured when pumping at the magnetic dipole resonance was much stronger than when the pump was tuned to the electric dipole resonance. We attribute this to the combined effect of higher absorption of GaAs at shorter wavelengths and the different SHG mechanisms discussed below.

Pump power dependent SHG measurements (Fig. 1(c)) show a quadratic relationship between the SHG intensity and the pump power, sustained over a wide pump power range until damage of the GaAs resonators occurred at ~5 mW (peak intensity of ~1.5 GW/cm^2). The damage was likely associated with the two photon absorption of GaAs followed by thermal damage due to the largely enhanced electric field intensity inside the resonators. Therefore, scaling this approach to higher pump powers would require the fabrication of bigger resonators so the pump photon energy can be tuned to below half of the GaAs bandgap. At the highest pump power the conversion efficiency of the SHG process exceeds 2×10^{-5} at a pump power of ~12 mW (peak intensity of ~3.6 GW/cm^2). This efficiency is comparable to recently published record high SHG efficiency using mode-matching plasmonic nanoantennas [3]. By optimizing the mode overlap between the fundamental and SH modes, we anticipate further improved conversion efficiency of the GaAs metasurface.

Next, we measured the polarization of the SHG at the magnetic and electric dipole resonances and, surprisingly, observed orthogonally polarized results. Note that the sample was mounted in the xy plane, as illustrated in Fig. 1(a), and the pump polarization was along the x axis parallel to the (100)-direction of GaAs. Fig.1 (e) shows the polar plot of the SH intensity pumped at the electric dipole resonances exhibiting maximum intensity along the y axis (perpendicular to the pump). When the pump is normally incident on the resonators, only the E_x^ω and E_z^ω components of the fundamental field are non-zero due to the symmetry of the cylindrical resonators and the objective used to focus the pump. The bulk second-order susceptibility $\chi^{(2)}_{ijk}$ is nonzero only when $i \neq j \neq k$ due to the zinc-blende crystal structure ($\bar{4}3m$) of GaAs [4]. Therefore, only the $E^{2\omega}_y = 2\chi^{(2)}_{yxz} E^{\omega}_x E^{\omega}_z$ component of the SH beam can be non-zero. Our observation is in a good agreement with this theoretical prediction [5].

In contrast, we observed orthogonal linear SHG polarization (along x axis) when we tuned the pump wavelength to the magnetic dipole resonance (around 1030 nm). This drastic SHG polarization difference cannot be explained by the bulk GaAs nonlinearity since only the E^{ω}_x and E^{ω}_z components of the fundamental field exist inside the resonators at the magnetic dipole resonance. Therefore, $E^{2\omega}_x$ of the SH cannot be generated due to the GaAs bulk nonlinearity. GaAs has been widely shown to also exhibit surface nonlinearity that could contribute to the strong x polarized SH. At the GaAs surface, the bulk symmetry $\bar{4}3m$ is broken, and the new symmetry for the surface is $mm2$ [4]. Therefore, $E^{2\omega}_x = 2\chi^{(2)}_{xxz} E^{\omega}_x E^{\omega}_z$ can be largely enhanced, considering the high electric field enhancement at the surface of the resonators when magnetic dipole mode was excited (inset of Fig. 1(f)). Moreover, the surface enhanced SHG contribution was further enhanced due to the oblique angle of incidence introduced by the objective in our measurements (NA=0.45), as this increased the overlap of the x and z components of the electric field inside the resonators. The y polarized second harmonic beam was weak at the electric dipole resonance because the electromagnetic field enhancement mainly occurs in the center of the resonators (inset of Fig. 1(e)).

Our demonstration paves the road other efficient nonlinear optical processes from III-V semiconductor metasurfaces and may lead to next generation ultra-compact nonlinear optical convertor.

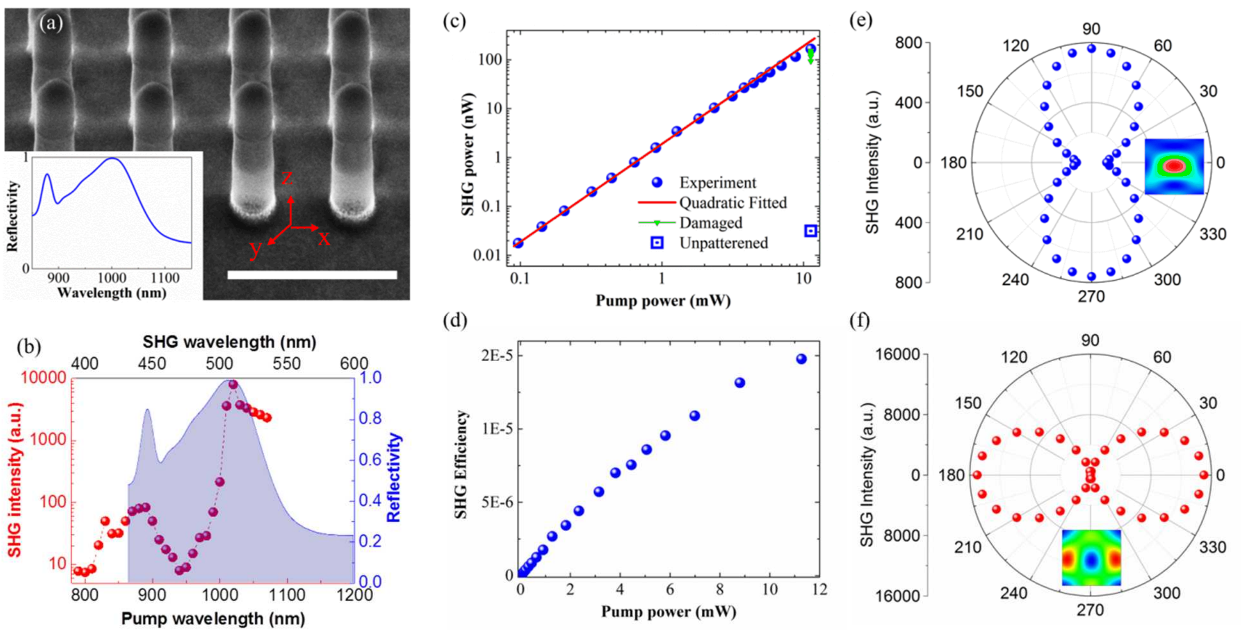

Fig 1. (a) 75 degree side view SEM image of the fabricated GaAs resonators. The height of the resonators is 300 nm and the diameter is ~250 nm. The inset is a reflection spectrum of the GaAs resonators showing well identified magnetic and electric dipole resonances. The scale bars correspond to 1 μm. (b) Experimental results of spectral dependent SHG intensity showing resonance enhanced SHG behavior at the magnetic and electric dipole resonances in a logarithm scale. The experimental linear refraction spectrum is used as a background. (c) Pump power dependent SHG showing quadratic relationship before the GaAs resonators were damaged at higher powers. (d) SHG conversion efficiency at different pump powers. Experimental polar plot of the SHG pumped at the (e) electric and the (f) magnetic dipole resonances (0 and 90 degrees indicate horizontal and vertical polarized light can preferentially transmit through the linear polarizer, respectively).

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