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Highly Nonlinear III-V Semiconductor Metasurfaces

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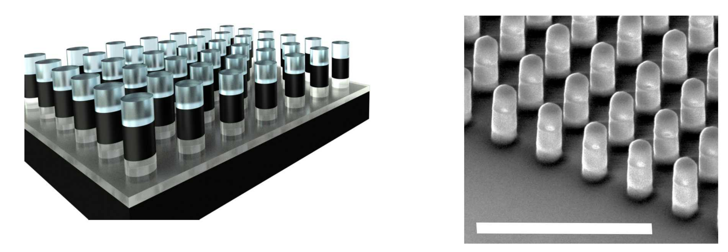
Introduction

The field of nonlinear optics has steadily expanded since it started in 1960, and a rich variety of novel nonlinear phenomena have been demonstrated in a wide range of different systems. It is widely used in fundamental science applications and in industrial products. However, the utilization of nonlinear optic processes in micro- and nanoscale devices is still difficult. A few obstacles such as the requirement of phase matching of the fundamental and generated waves as well as high conversion efficiencies of nonlinear processes are still unsolved. A possible solution is to use nonlinear, all-dielectric metasurfaces [1]. They relax phase matching conditions and increase the conversion efficiencies due to the excitation of Mie resonances. Recent results show that metasurfaces and nanoresonators that are made from III-V semiconductors show record high conversion efficiencies for second harmonic generation (SHG) up to 10^{-5}–10^{-4} in comparison with other nanostructured materials [2],[3]. However, no one has ever tried to generate multiple and simultaneous new frequencies in a GaAs metasurface.

Fabrication of GaAs metasurface

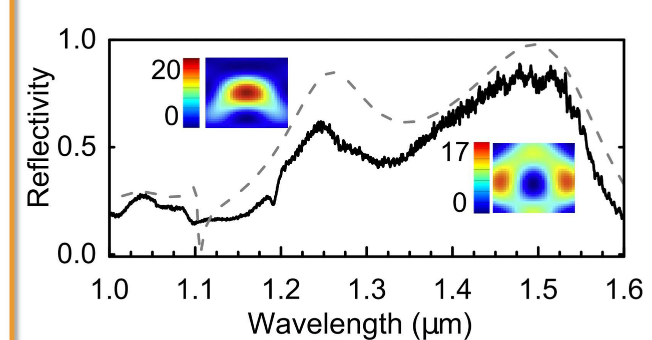
Sample design



SiO_x – 300 nm
GaAs – 400 nm
$(Al_xGa_{1-x})_2O_3$ – 450 nm
Diameter 400 nm
Period 840 nm

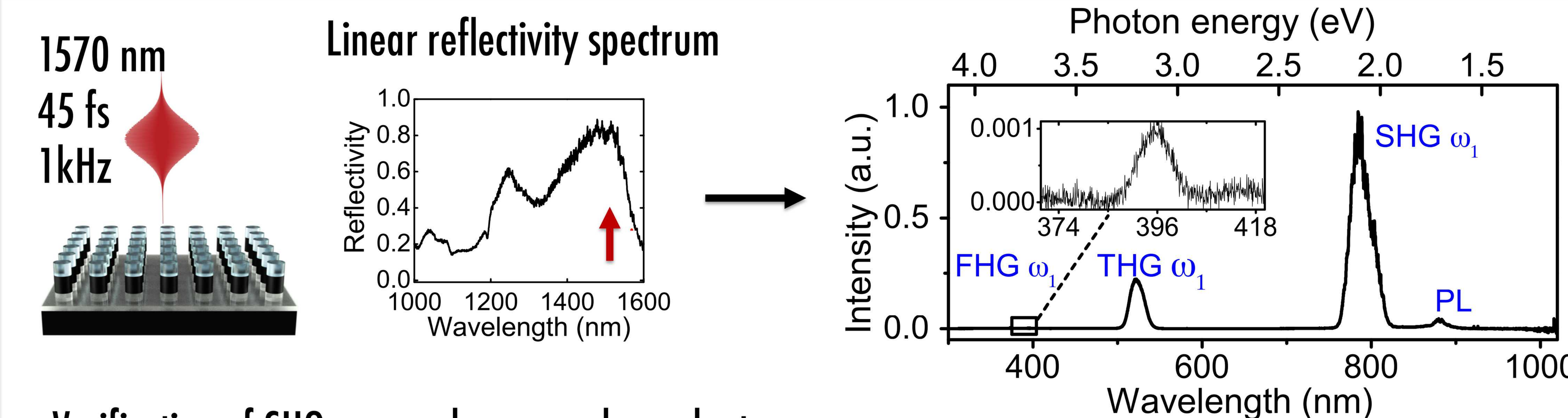
(a) Design of the GaAs metasurface.
(b) A 60° side-view scanning electron microscope image of the GaAs metamixer. The scale bar corresponds to 3 μm.

Linear reflectivity spectrum of the GaAs metasurface: Mie resonances

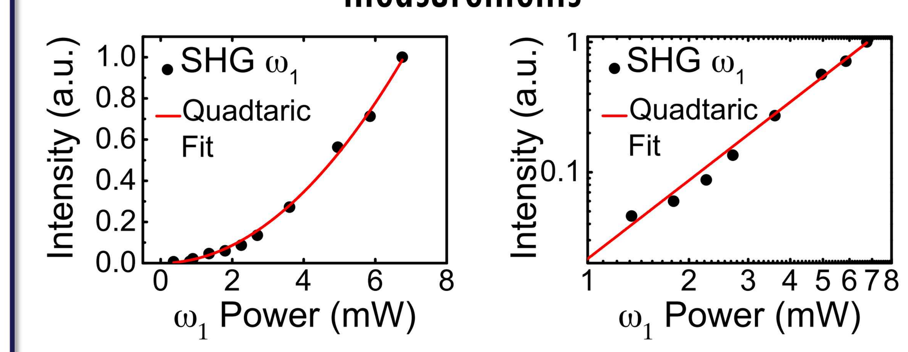


Measured (solid line) and numerically simulated (dashed line) reflectance spectra of the metasurface with two cross-section local electric field distributions at the wavelengths of 1.25 μm and 1.56 μm, which correspond to the maximal electromagnetic field enhancements inside the GaAs nanodisk due to the excitation of the magnetic dipole and electric dipole Mie resonances.

One-beam experiment: SHG, THG, FHG



Verification of SHG process by power dependent measurements

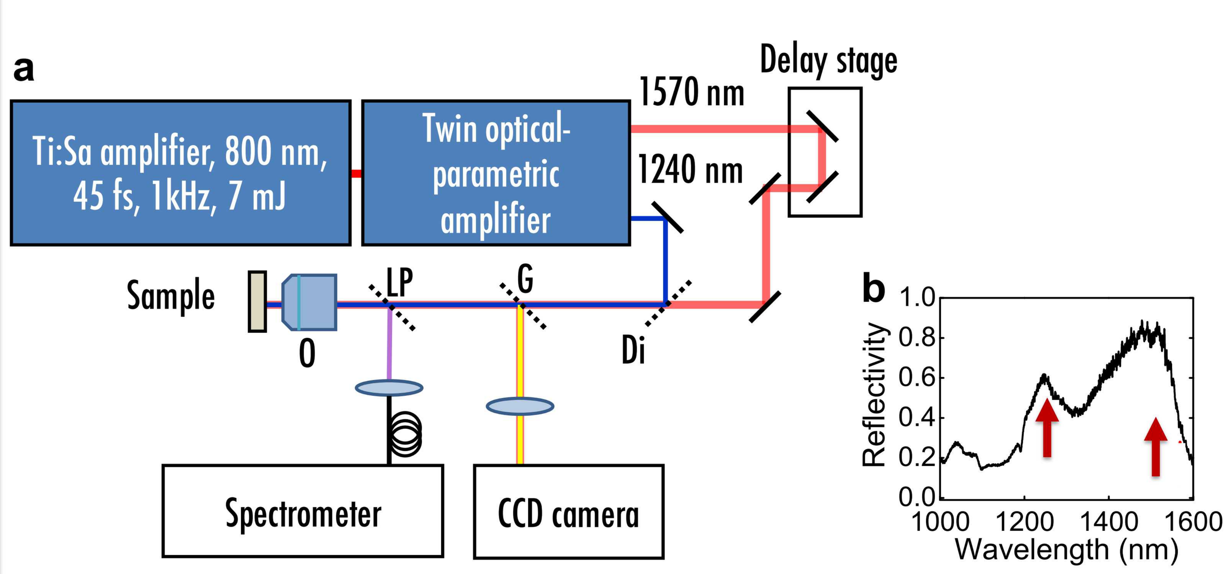


The dependence of the SHG intensity on the pump power shown in linear and logarithmic scales.

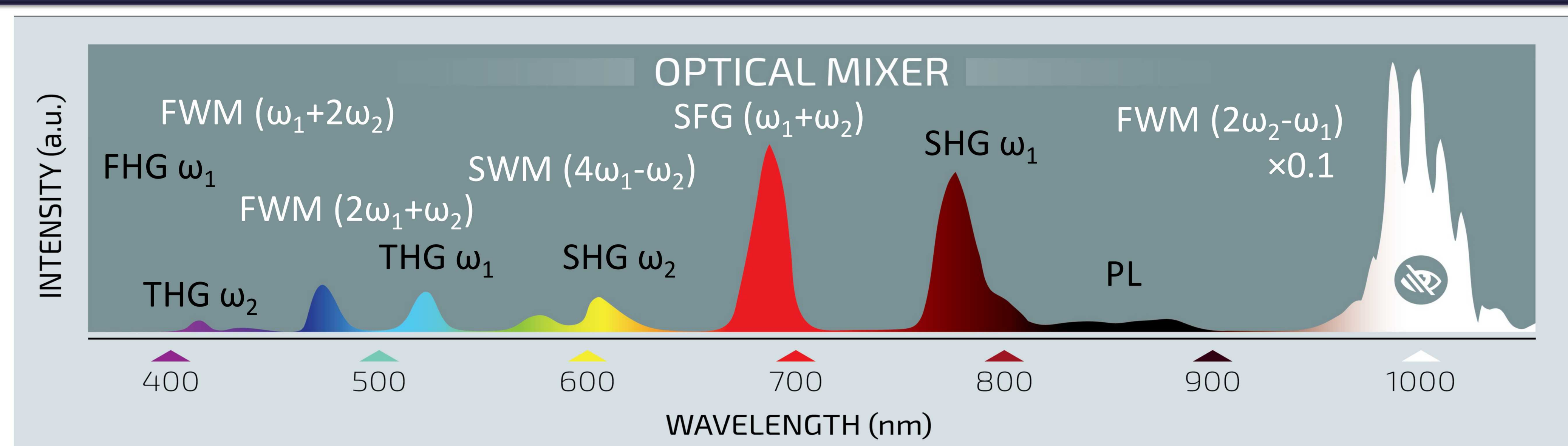
When the GaAs metasurface is pumped by a single near-IR femtosecond beam with a wavelength near magnetic dipole resonance (λ_1~1570 nm), we observe in the nonlinear spectra simultaneous generation of second-, third- and, fourth-harmonics (SHG, THG, and FHG), as well as band-edge photoluminescence (PL) emission.

Two-beam experiment: frequency mixing

Nonlinear frequency mixing measurement setup

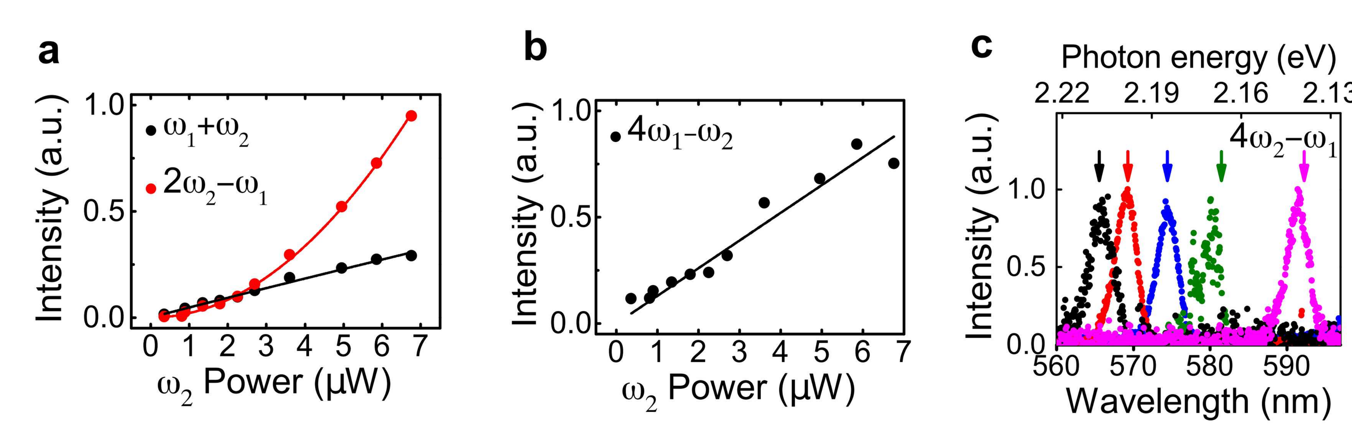


(a) Experimental setup for frequency mixing measurements. Di: dichroic beam combiner, G: glass window, LP: 1064 nm long pass filter, O: near-IR objective. (b) Linear reflectivity spectrum of the GaAs nanocylinder metasurface. Arrows indicate the pumping wavelengths.



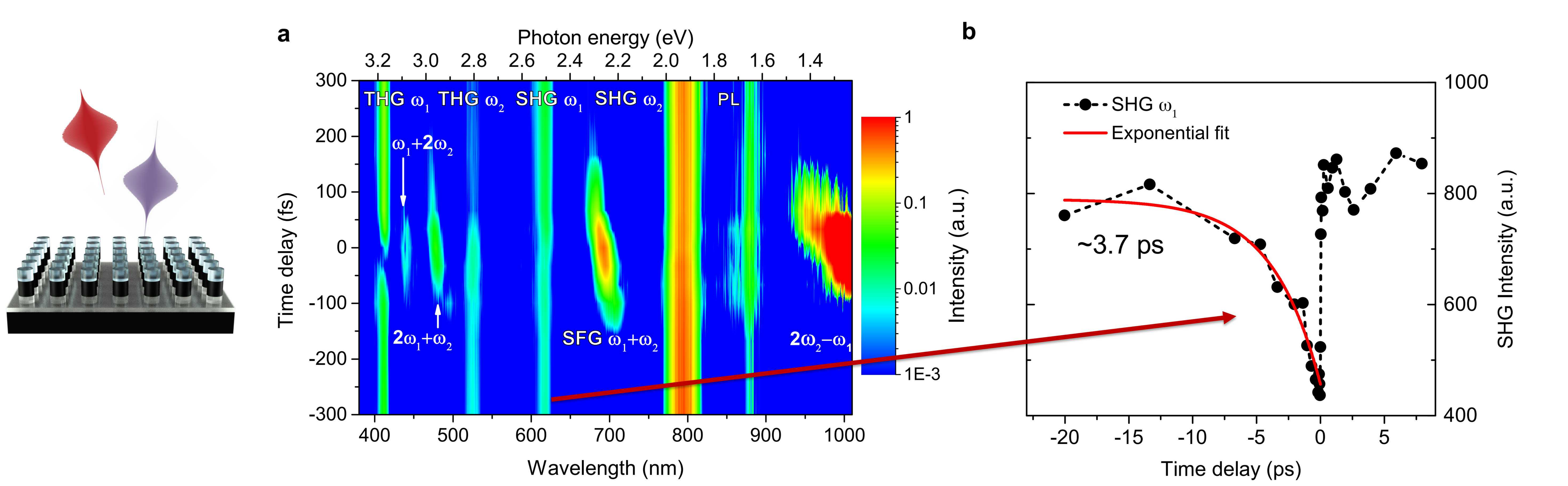
When the two pump laser pulses arrive at the GaAs metasurface at the same time we observe a total of eleven new frequencies spanning the UV to the near-IR. The generated signals can be divided into two groups: six signals rely on only one of the two pump beams:- SHG, THG, FHG and PL emission (labeled in black), and five frequency mixing signals rely on both pump beams (labeled in white) – sum-frequency generation (SFG), six-wave mixing (SWM), and three signals corresponding to four-wave mixing (FWM).

Verification of nonlinear optical processes by power dependent and spectral tuning measurements



(a, b) Dependence of the SFG ($\omega_1+\omega_2$), FWM ($2\omega_2-\omega_1$) and SWM ($4\omega_1-\omega_2$) intensities on the power of the ω_2 pump. (c) Five representative spectra showing the tuning of the normalized six-wave mixing signal when the pump wavelengths are spectrally tuned. The arrows denote the theoretically expected frequencies for the considered six-wave mixing process.

Pump-probe spectroscopy of newly generated frequencies



(a) A 2D contour image of the transient nonlinear signal (logarithmic scale) when the time delay between the two pump pulses is varied. The nonlinear signals that require only one of the pumps do not depend on the delay, while the mixing signals that rely on both pumps occur only when the two pumps overlap in time. (b) The quenching and the recovery of the ω_1 SHG intensity due to the arrival of the second pump ω_2 at the metasurface. The black dots are experimentally measured SHG intensities and the red curve is a single component exponential fit with time constant of ~3.7 ps.

Conclusions

In this work, we experimentally demonstrate simultaneous occurrence of second-, third-, fourth-harmonic generation, sum-frequency generation, four-wave mixing and six-wave mixing processes in III-V semiconductor metasurfaces with spectra spanning from the UV to the near-IR.

More details: Sheng Liu, Polina P. Vabishchevich, Aleksandr Vaskin, John L. Reno, Gordon A. Keeler, Michael B. Sinclair, Isabelle Staude, and Igal Brener. "An all-dielectric metasurface as a broadband optical frequency mixer." *Nature communications* 9, no. 1 (2018): 2507.

References

[1] A. I. Kuznetsov, A. E. Miroshnichenko, M. L. Brongersma, Y. S. Kivshar, and B. Luk'yanchuk, "Optically resonant dielectric nanostructures," Science, vol. 354, no. 6314, p. 2472, 2016.
[2] S. Liu, et al. "Resonantly enhanced second-harmonic generation using III–V semiconductor all-dielectric metasurfaces," Nano Lett., vol. 16, no. 9, pp. 5426–5432, 2016.
[3] S. Kruk, et al. "Enhanced magnetic second-harmonic generation from resonant metasurfaces," ACS Photonics, vol. 2, no. 8, pp. 1007–1012, 2015.

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