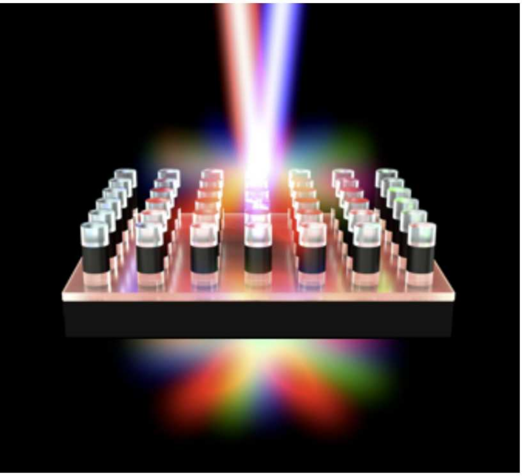
SAND2018-3743C

Nonlinear III-V semiconductor metasurfaces



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Exceptional service in the national interest

Polina Vabishchevich

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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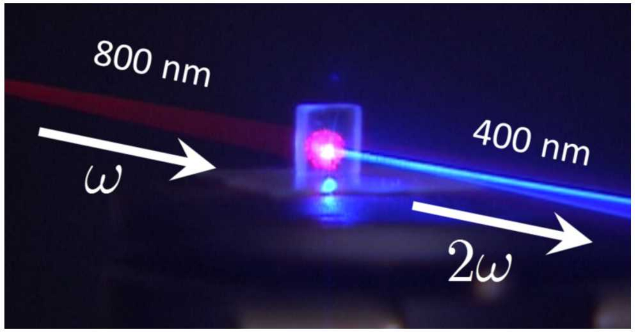
Outline

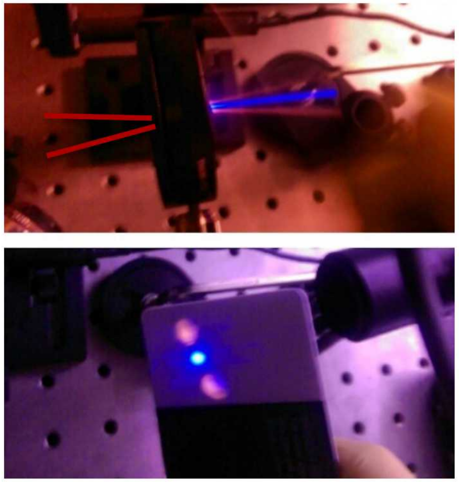
- Nonlinear optics
- Metasurfaces: All-dielectric metasurfaces with Mie resonances
- Nonlinear meatsurfaces
- Frequency mixing in III-V semiconductor metasurface: an optical metamixer
- Enhanced Second-Harmonic Generation Using Broken Symmetry III-V Semiconductor Fano Metasurfaces.

Nonlinear optics: frequency mixing

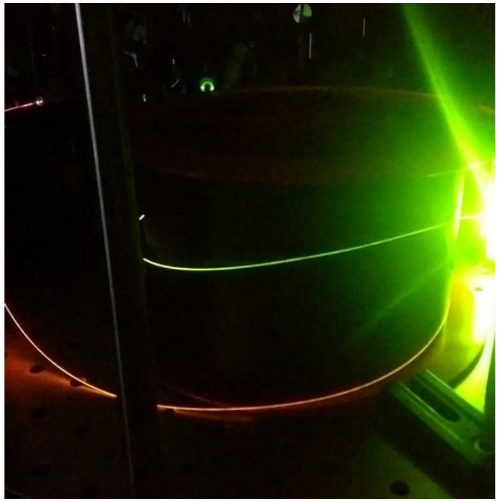
Second-harmonic generation (SHG)



Sum-frequency generation



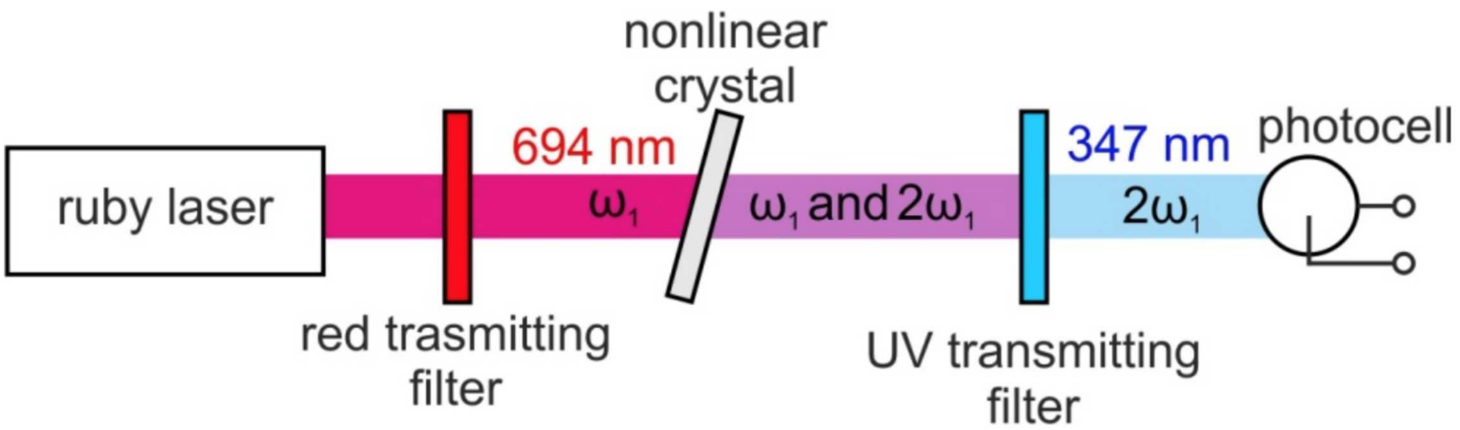
Supercontinuum generation



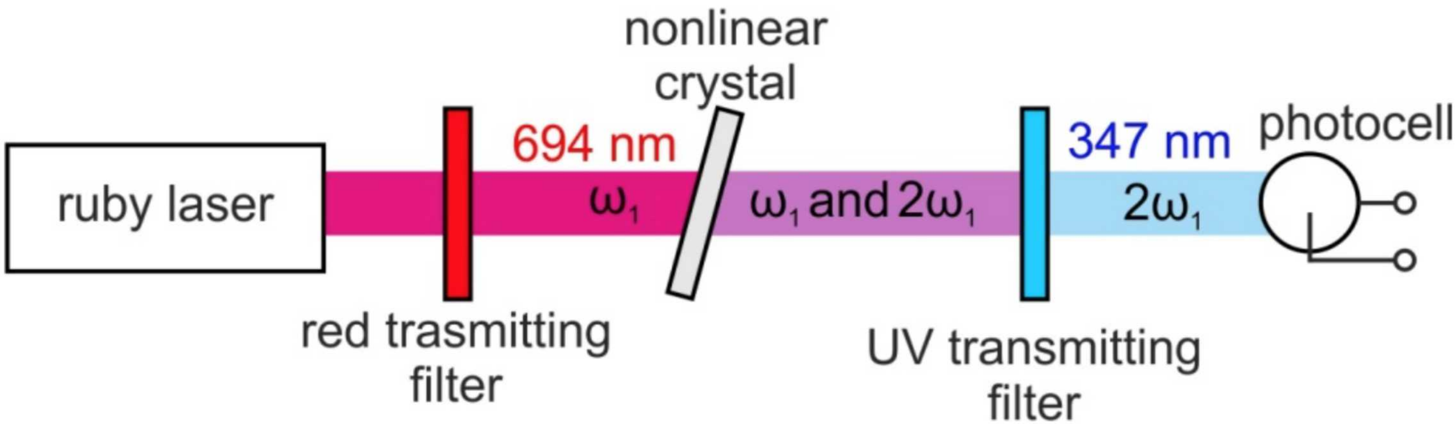
Green laser pointers – SHG!

Historical Background

First demonstration of second-harmonic generation by P. A. Franken et. al. in 1961 started nonlinear optics



Where is nonlinearity?



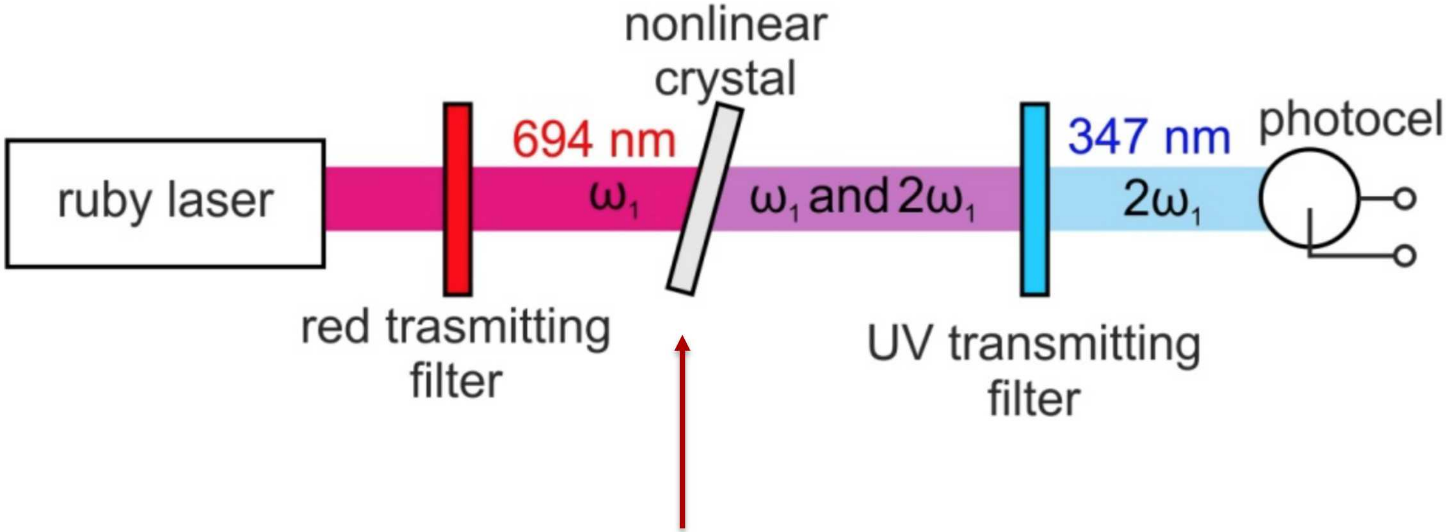
Strong electromagnetic field → Distorted electronic cloud → Induced dipole

Dielectric polarization density **P**(t)
– dipole moment per unit volume

$$\mathbf{P}(t) = \varepsilon_0 (\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \ldots),$$

where the coefficients $\chi^{(n)}$ are the n-th-order susceptibilities of the medium,

Where is nonlinearity?



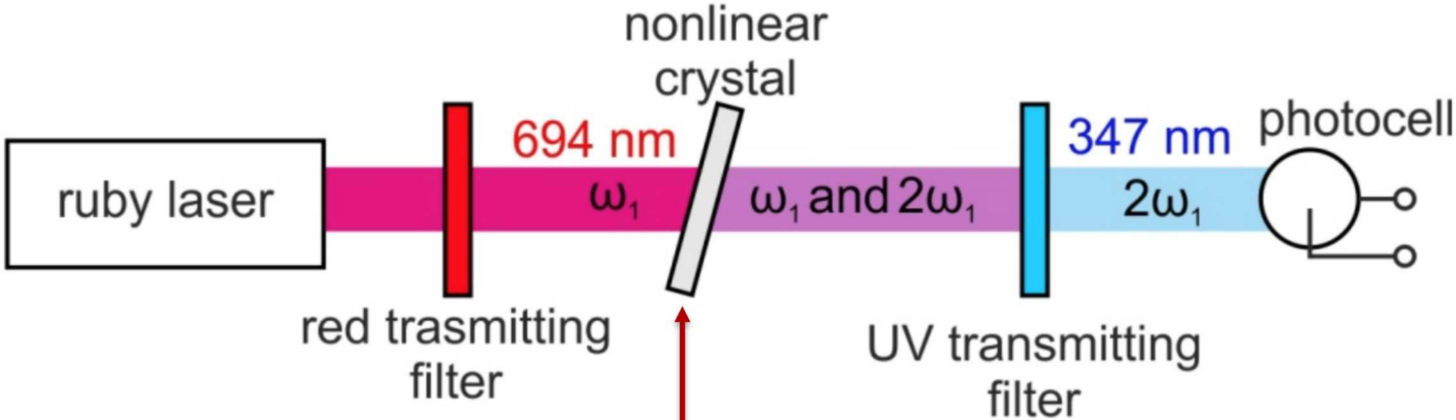
Dielectric polarization density $\mathbf{P}(t)$
– dipole moment per unit volume

Linear optics

$$\mathbf{P}(t) = \varepsilon_0(\chi^{(1)}\mathbf{E}(t) + \chi^{(2)}\mathbf{E}^2(t) + \chi^{(3)}\mathbf{E}^3(t) + \ldots),$$

$$\chi^{(1)} = n^2 - 1$$

Where is nonlinearity?



Dielectric polarization density **P**(t)
– dipole moment per unit volume

Nonlinear optics

$$\mathbf{P}(t) = \varepsilon_0 (\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \ldots),$$

where the coefficients $\chi^{(n)}$ are the n-th-order susceptibilities of the medium,

Nonlinear processes

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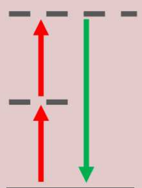
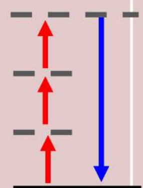
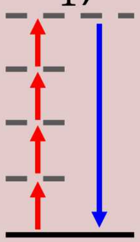
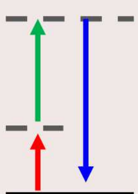
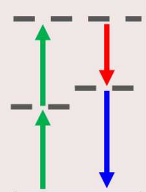
$$\mathbf{P}(t) = \varepsilon_0(\chi^{(1)}\mathbf{E}(t) + \chi^{(2)}\mathbf{E}^2(t) + \chi^{(3)}\mathbf{E}^3(t) + \ldots)$$

$\chi^{(2)}$	$\chi^{(3)}$	$\chi^{(4)}$	$\chi^{(5)}$
Second harmonic generation $(2\omega_1)$ 	Third harmonic generation $(3\omega_1)$ 	Fourth harmonic generation $(4\omega_1)$ 	Fifth harmonic generation $(5\omega_1)$
Sum-frequency generation $(\omega_1+\omega_2)$ 	Four wave mixing $(2\omega_1-\omega_2)$, $((2\omega_1+\omega_2)$ 		

etc.

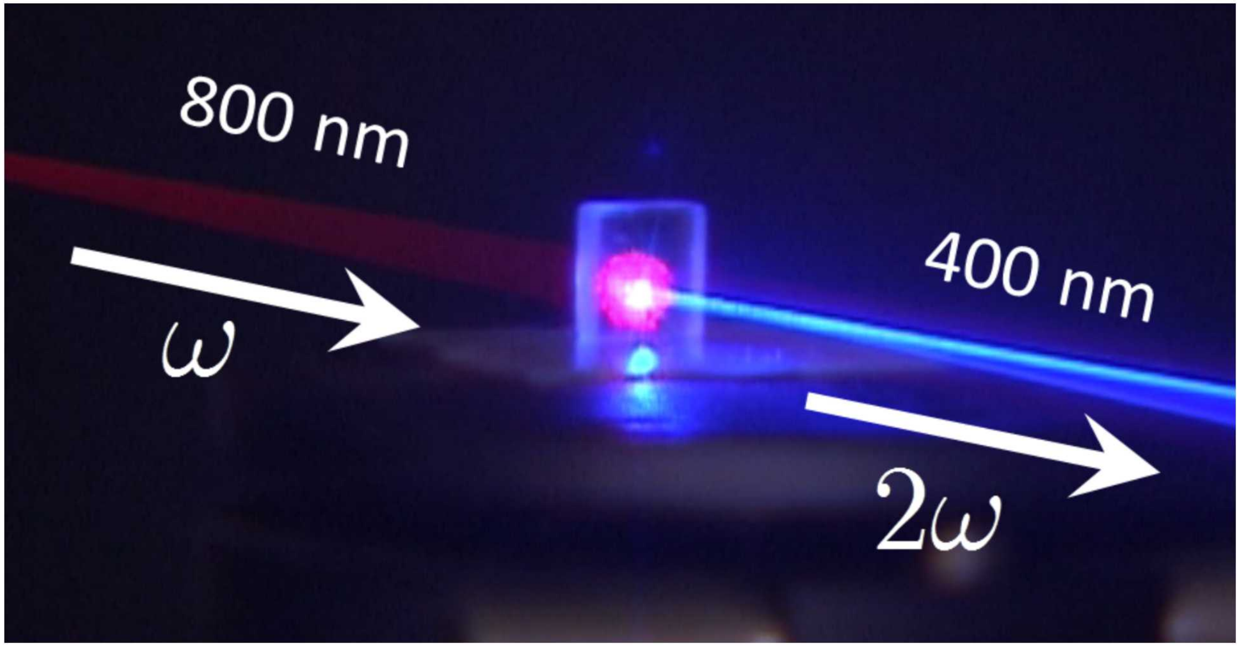
Restrictions

$$\mathbf{P}(t) = \varepsilon_0(\chi^{(1)}\mathbf{E}(t) + \chi^{(2)}\mathbf{E}^2(t) + \chi^{(3)}\mathbf{E}^3(t) + \ldots),$$

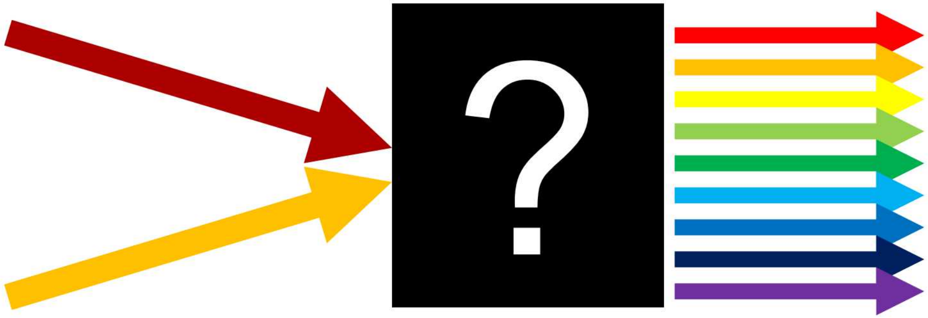
- Strong electromagnetic fields
- Phase matching (dispersion) is critical:

  nonlinear crystal dimensions >> wavelengths

- Special conditions for each nonlinear process



“Super” frequency mixing?

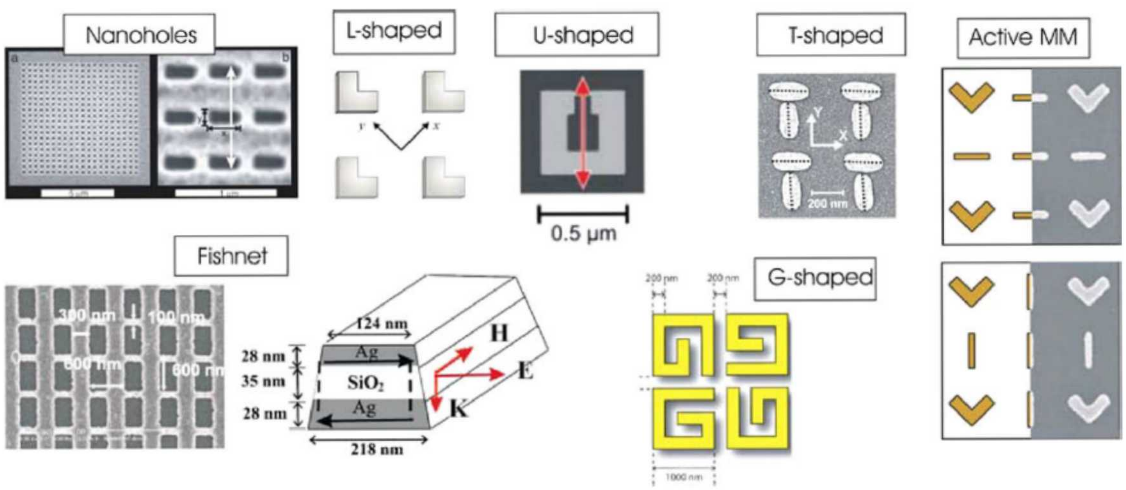


Can we see all of these mixings at the same time?

Nonlinear optical frequency mixing in metamaterials and metasurfaces

Engineered structures that have specific optical properties on demand by choosing materials, shapes and dimensions of the metaatoms.

Dimensions < Wavelength



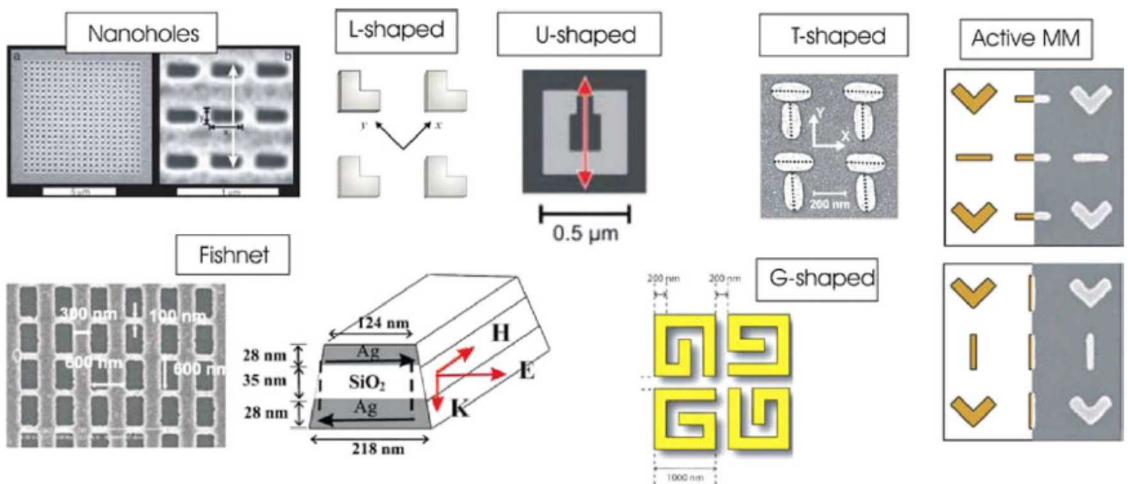
Y. Kivshar et al, Laser Photonics Review, **9**, 195 (2015)

Nonlinear optical frequency mixing in metamaterials and metasurfaces

Engineered structures that have specific optical properties on demand by choosing materials, shapes and dimensions of the metaatoms.

Dimensions < Wavelength

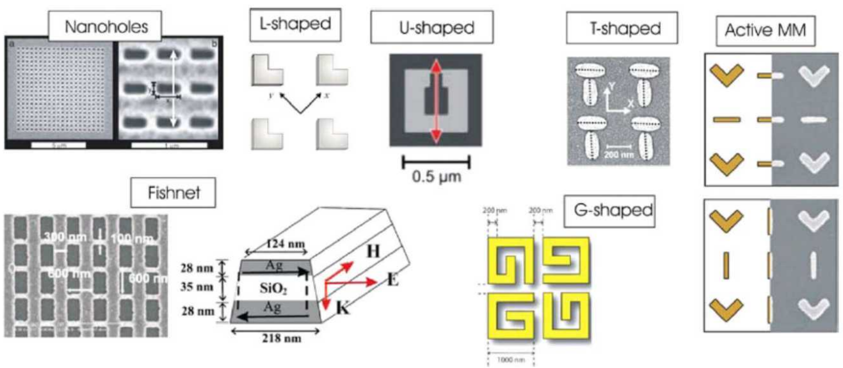
- **Relaxed phase matching conditions**
- **Resonant enhancement of EM field**



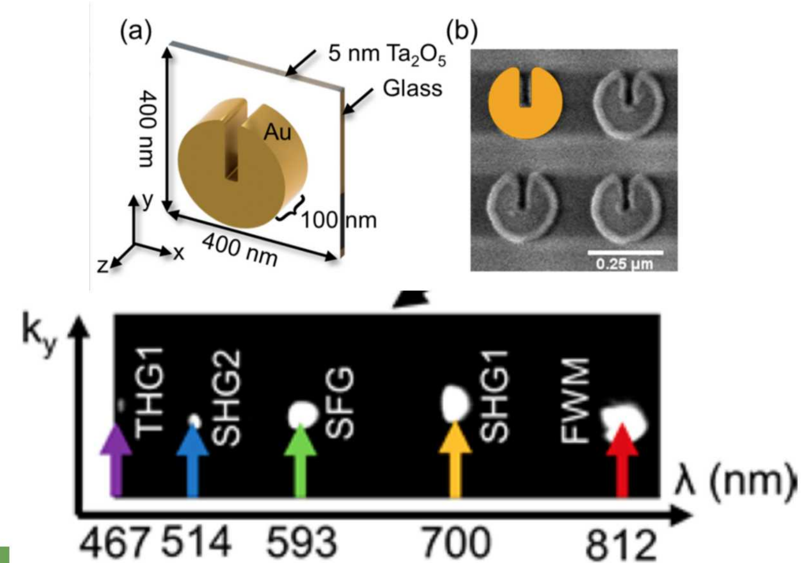
Y. Kivshar et al, Laser Photonics Review, **9**, 195 (2015)

Nonlinear plasmonics

- **Relaxed phase matching conditions**
- **Resonant enhancement of EM field induced by Plasmon excitation**



Y. Kivshar et al, Laser Photonics Review, **9**, 195 (2015)



Sartorello, et al. *ACS Photonics* 3.8 (2016): 1517-1522.

nature photonics FOCUS | REVIEW ARTICLES
PUBLISHED ONLINE: 31 OCTOBER 2012 | DOI: 10.1038/NPHOTON.2012.244

Nonlinear plasmonics

Martti Kauranen[1] and Anatoly V. Zayats[2]

SHG efficiency ~ 10^{-9}

Plasmonic structure: small modal volume
(usually only use surface nonlinearities)

Mie resonances in dielectric metasurfaces

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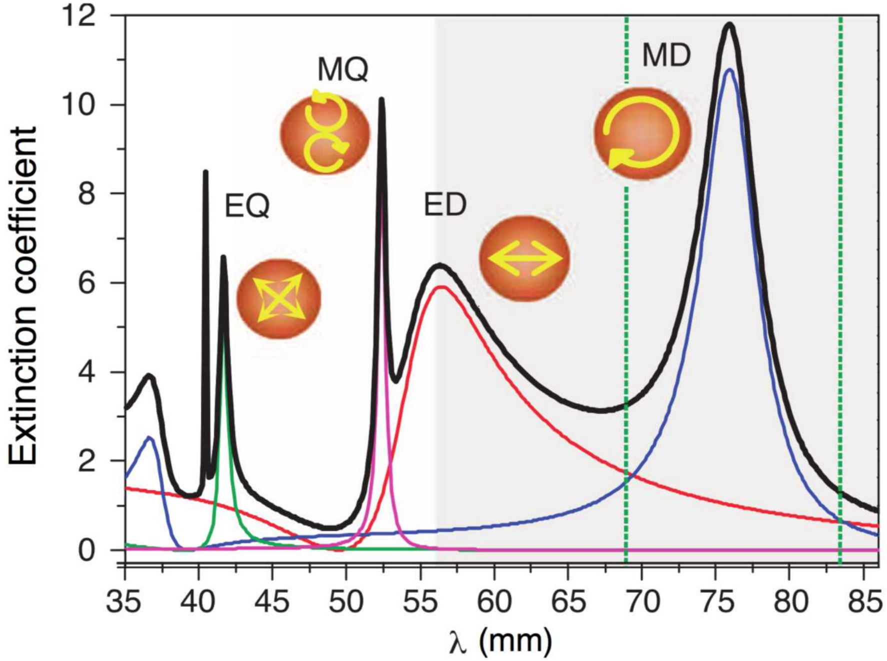
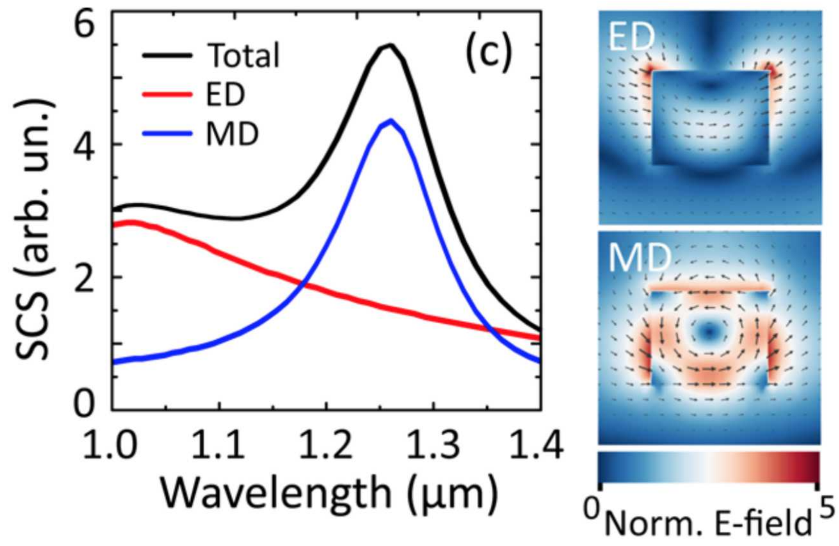
Mie resonances in Dielectric spheres

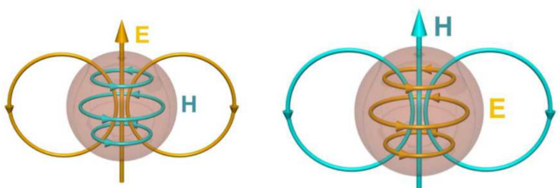


Jean-Michel Geffrin et. al. / Nat. Comm.– 2012.– Vol.3,p.11



M .R. Shcherbakov, et. al. *Nano Letters* **14,** 11, 6488–6492, 2014

Electric Dipole **Magnetic Dipole**



Images: A. Miroshnichenko

Mie resonances
- Low losses
- Much larger mode volume
- High nonlinear coefficient materials (GaAs: ~200pm/V)

Optical frequency mixing in dielectric metasurfaces

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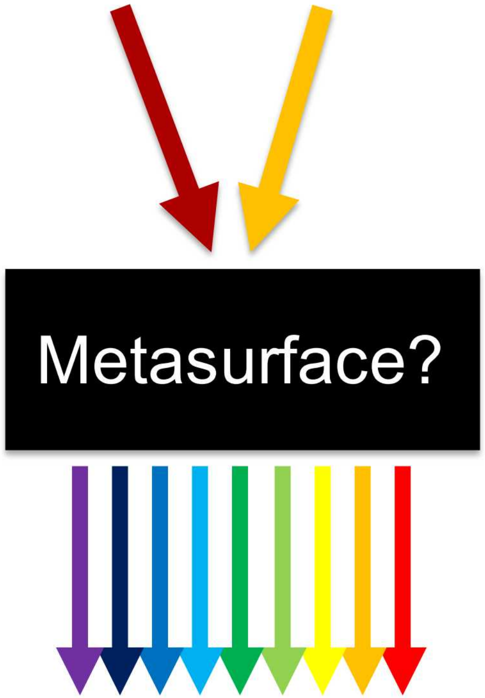


Second Harmonic Generation (10^{-5})

AlGaAs – Carletti, L., et al. *Optics express* 23.20 (2015): 26544-26550.
Kruk, S.et al. *Nano Lett.*, **2017**, *17* (6), pp 3914–3918
GaAs – Liu, S. et. al. *Nano letters* 16.9 (2016): 5426-5432.

Third Harmonic Generation (10^{-7})

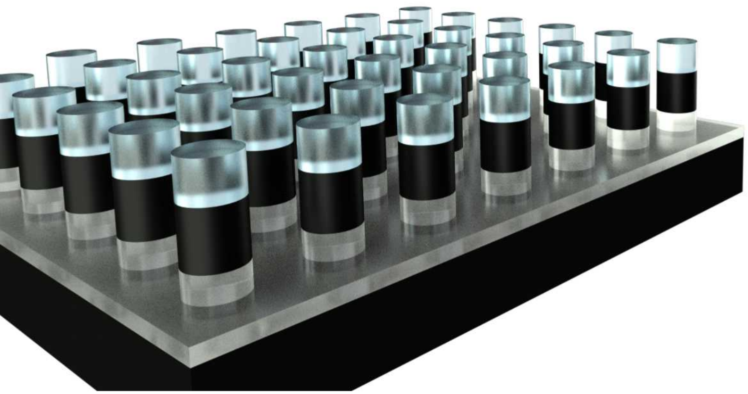
Si *– Shcherbakov et.al. ACS Photonics*, **2015**, *2* (5), pp 578–582
a-Si *– Shorokhov et.al. Nano Lett.*, **2016**, *16* (8), pp 4857–4861

Four-Wave Mixing and THG

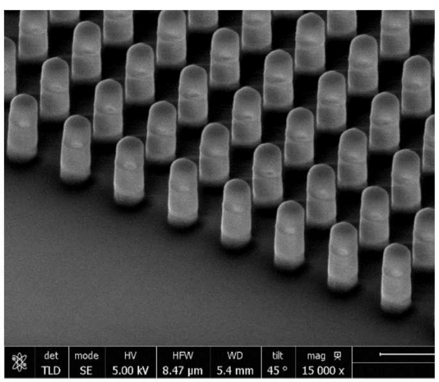
Ge – Grinblat, G., et al, ACS Photonics 4, 2144-2149

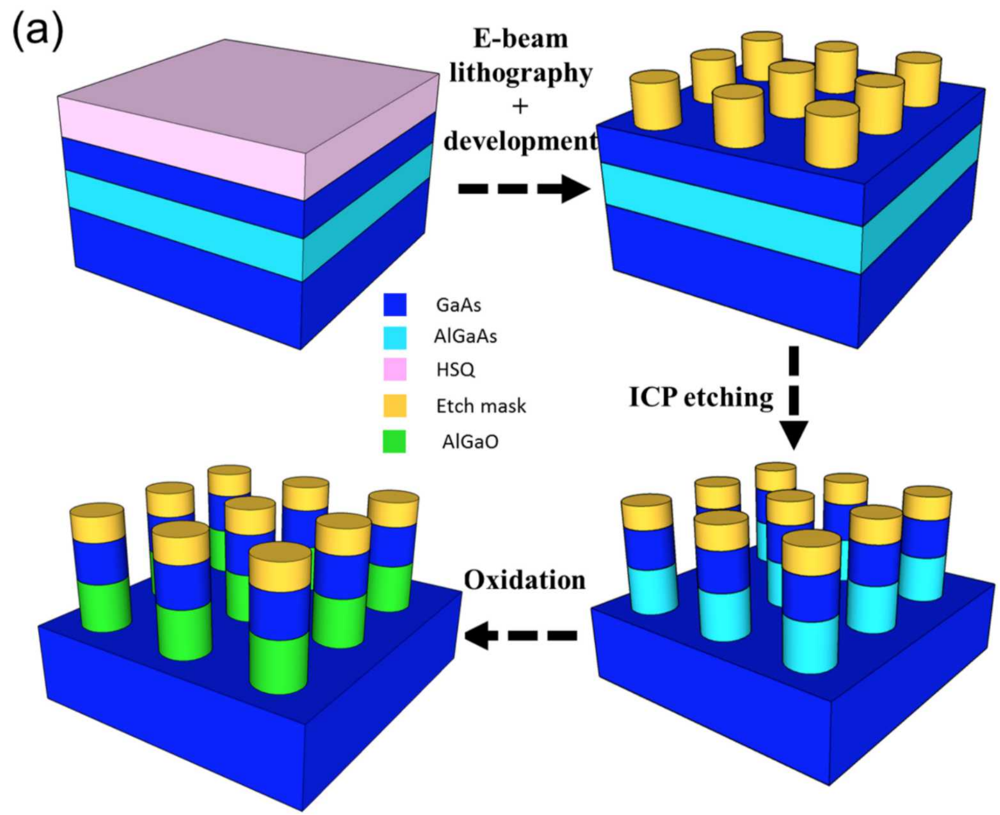
Multiple frequency mixing in dielectric metasurfaces

GaAs based dielectric metasurface



SiO_x – 300 nm
GaAs – 450 nm
$(Al_xGa_{1-x})_2O_3$ – 400 nm
Diameter 430 nm
Period 840 nm

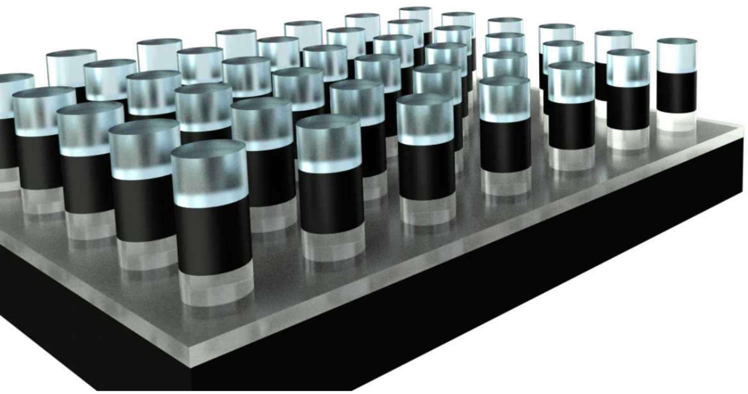




Liu, S. et. al. *Nano letters* 16.9 (2016): 5426-5432

- **High nonlinear coefficient material**

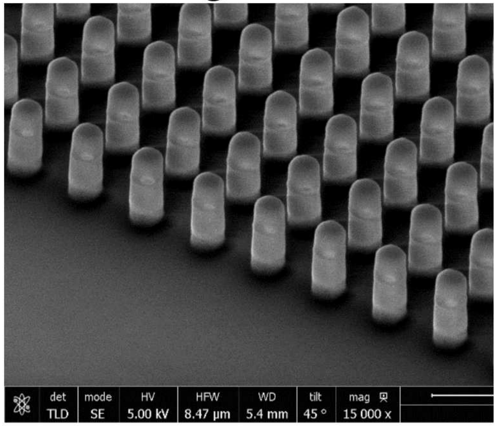
GaAs based dielectric metasurface



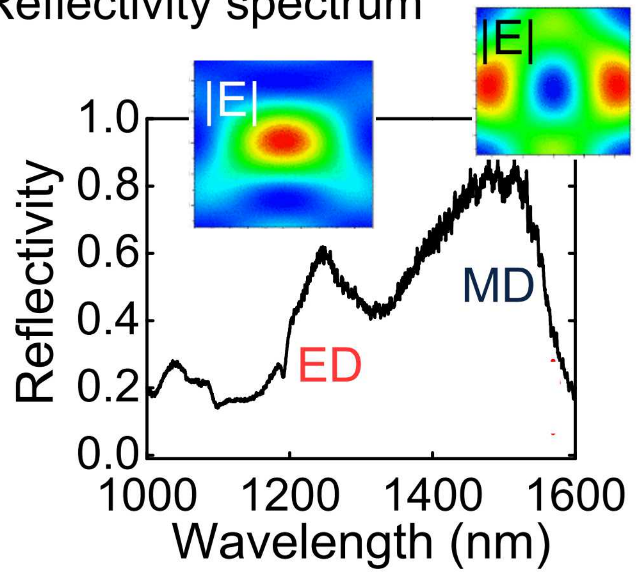
SiO_x – 300 nm
GaAs – 450 nm
$(Al_xGa_{1-x})_2O_3$ – 400 nm

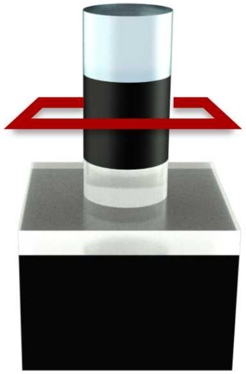
Diameter 430 nm
Period 840 nm

SEM image



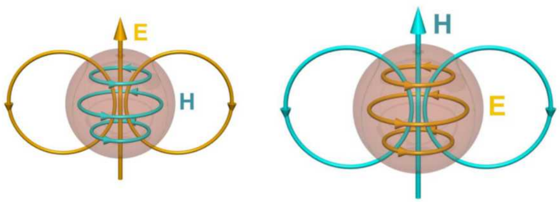
Reflectivity spectrum





Electric Dipole

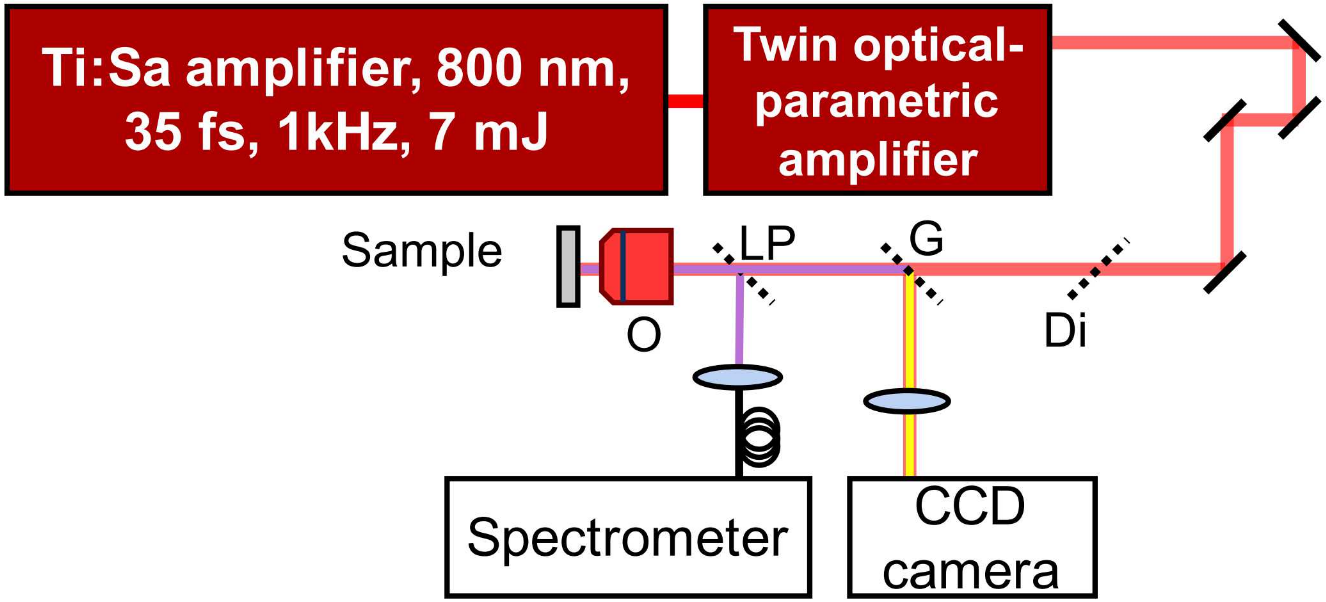
Magnetic Dipole



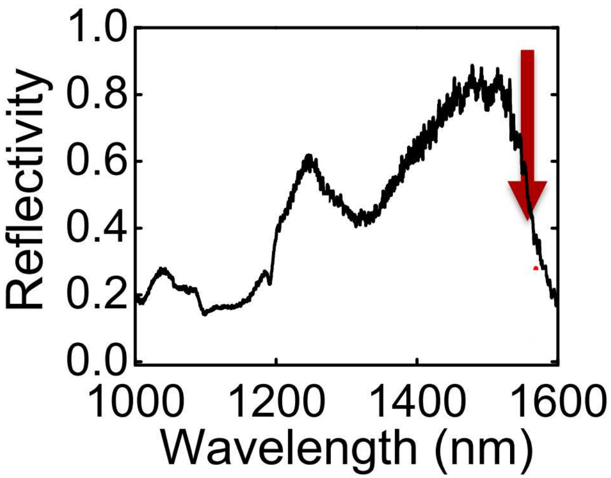
Images: A. Miroshnichenko

- **Resonance enhancement of EM field**

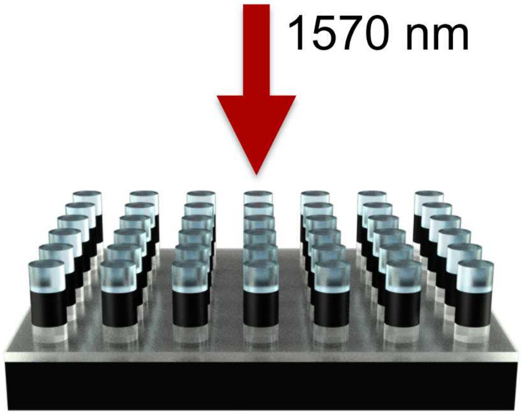
Single beam experiment



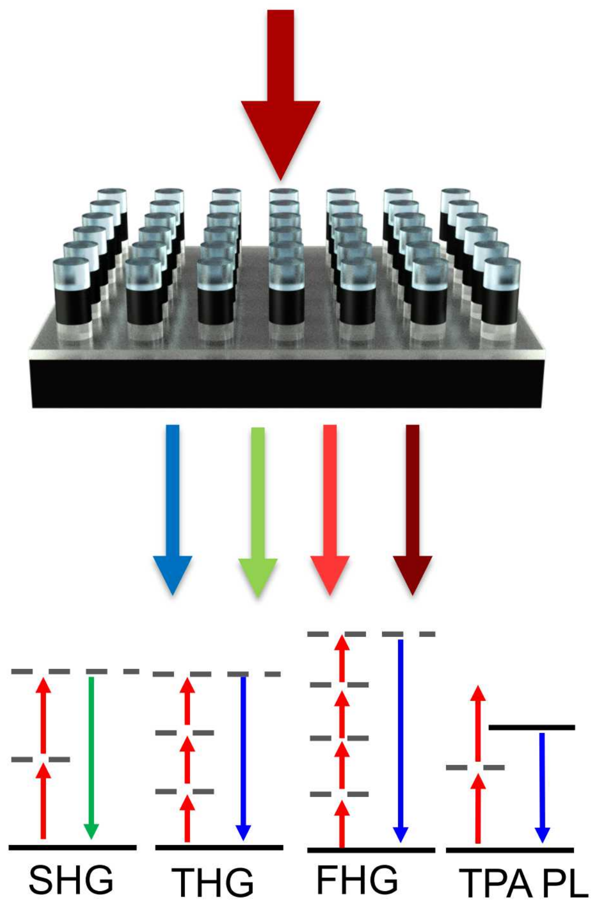
50 fs
1 kHz
Power 4.5 mW
0.4 NA objective

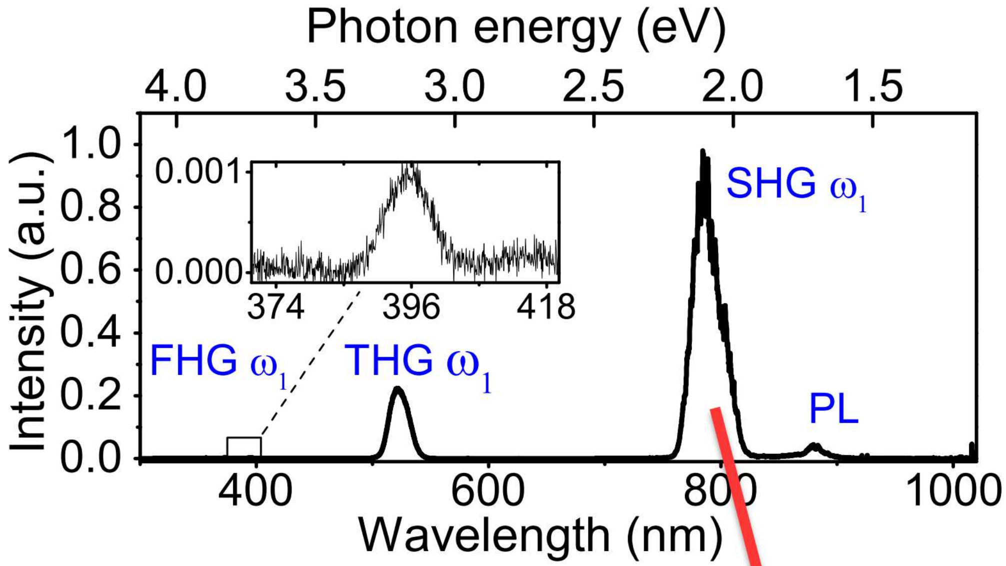


Magnetic dipole resonance

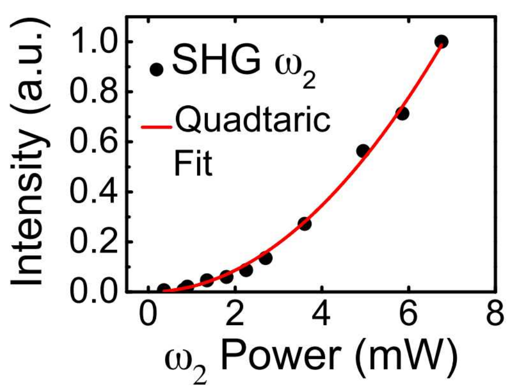


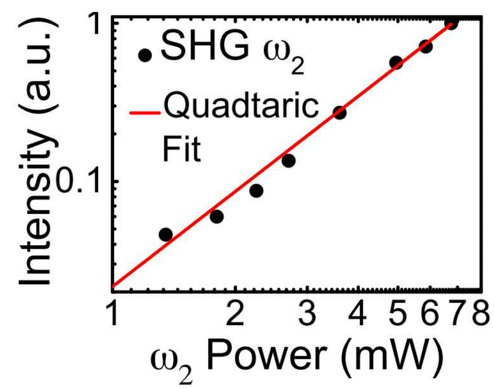
2^{nd}, 3^{rd}, 4^{th} harmonic generation spectra





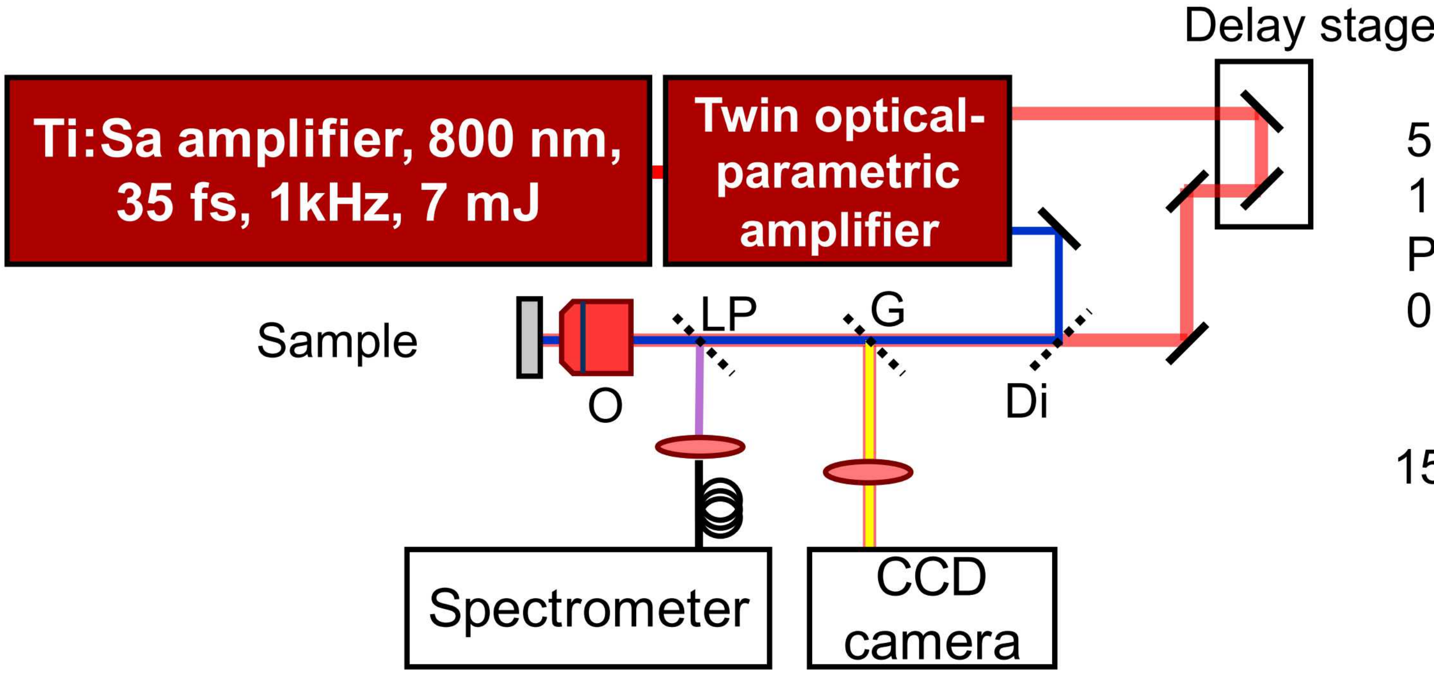
SHG power dependence





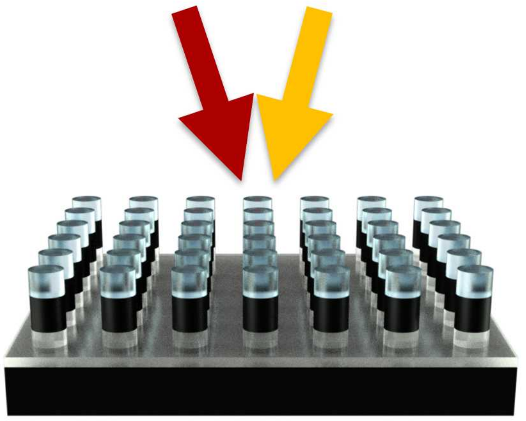
SHG efficiency ~ 2.3×10^{-6}

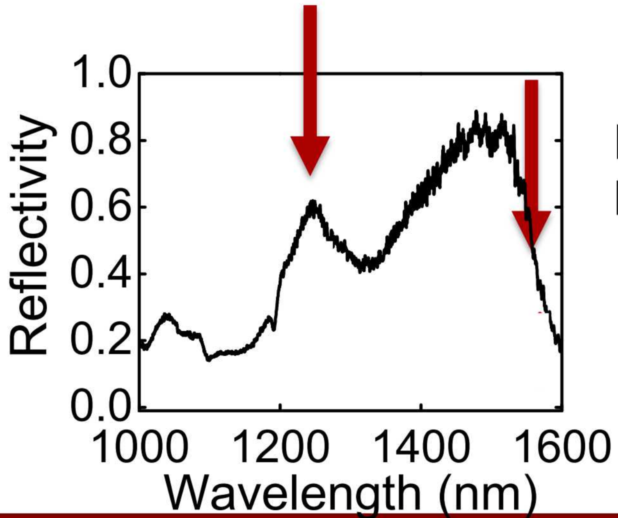
Two-beam experiment



50 fs
1 kHz
Power 3.6 mW, 5 mW
0.4 NA objective

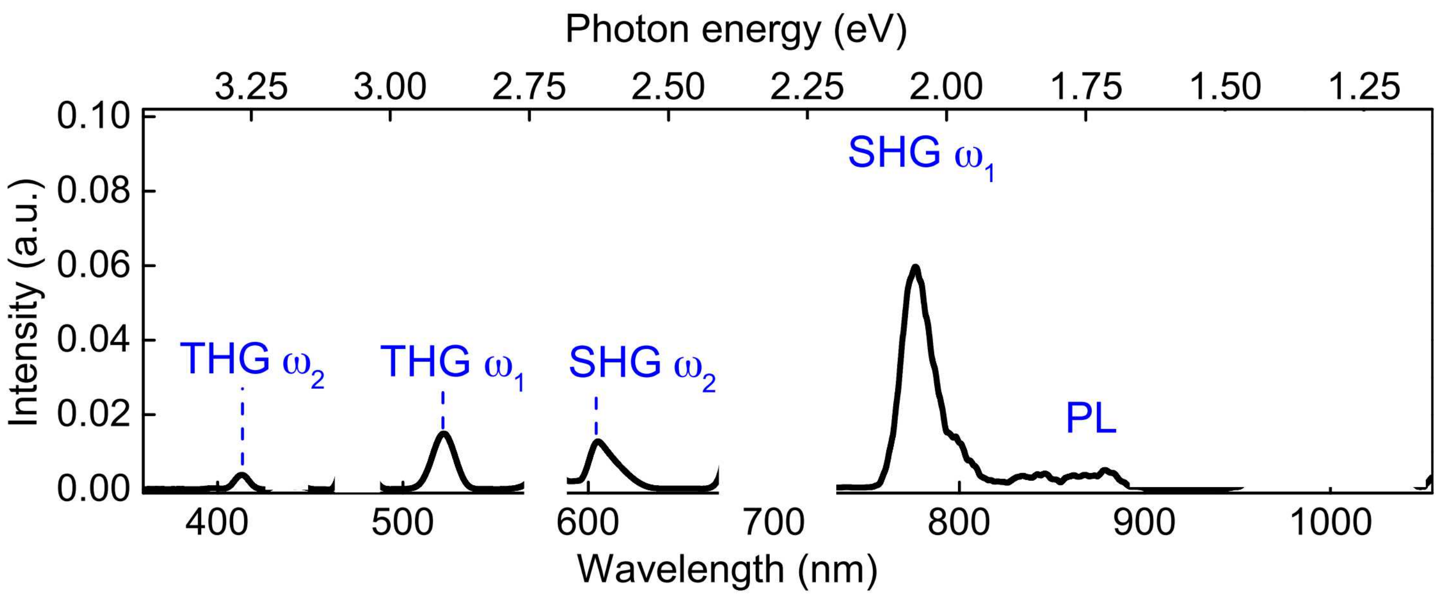
1570 nm 1240 nm

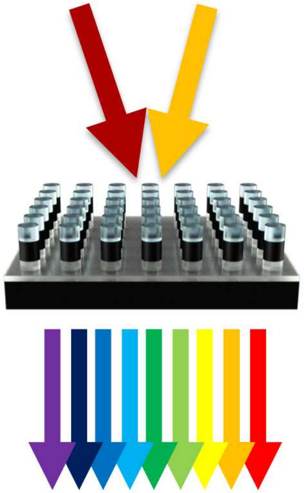


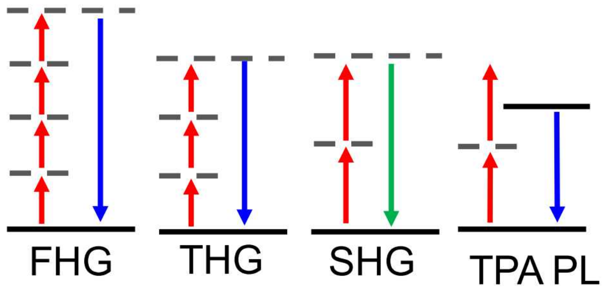


Electric dipole resonance
Magnetic dipole resonance

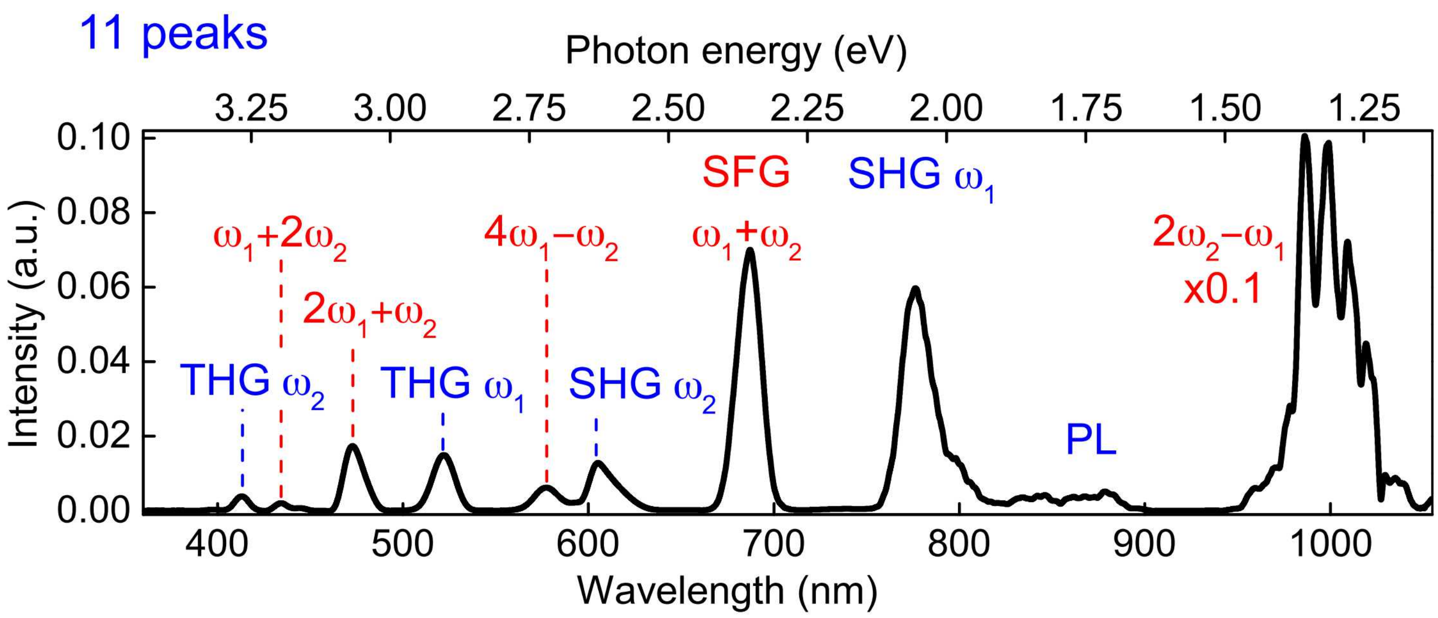
Frequency mixing spectra

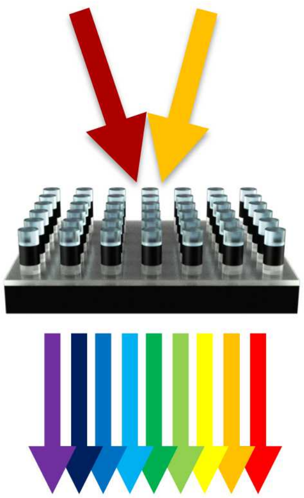


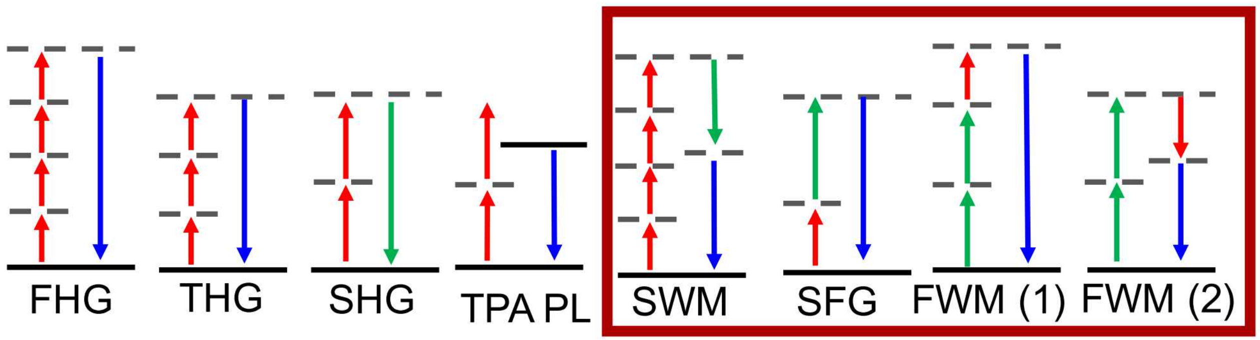




Frequency mixing spectra



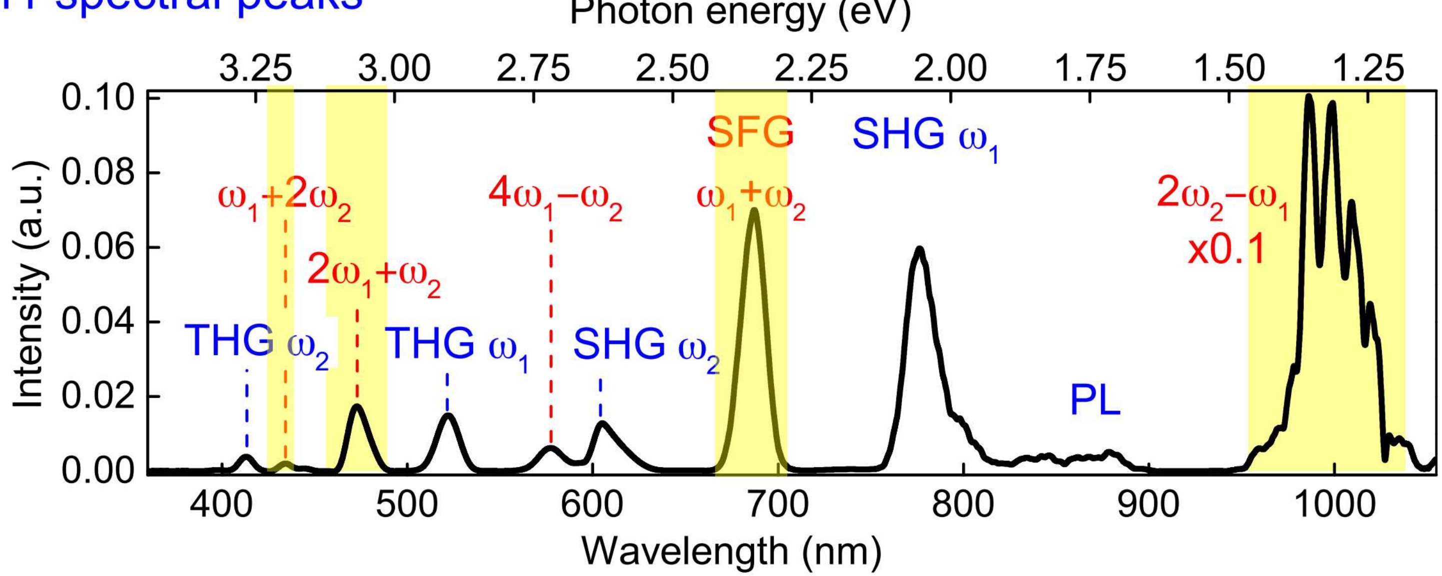


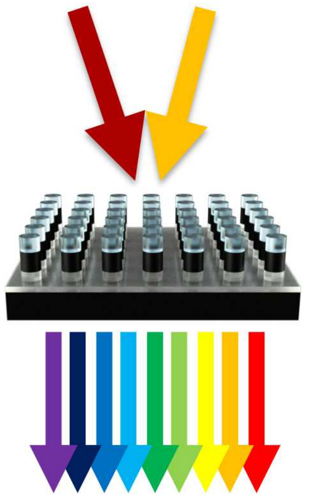


7 different nonlinear processes: 2nd, 3rd, 4th harmonics, sum frequency generation, 4 wave-mixing, six-wave mixing, PL induced by two-photon absorption

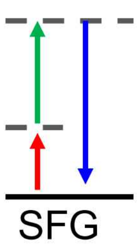
Frequency mixing spectra

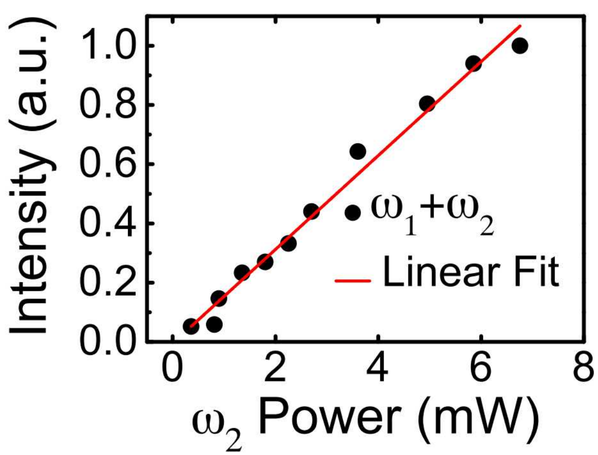
11 spectral peaks

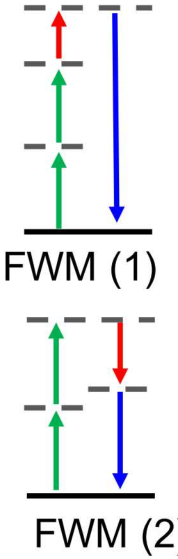




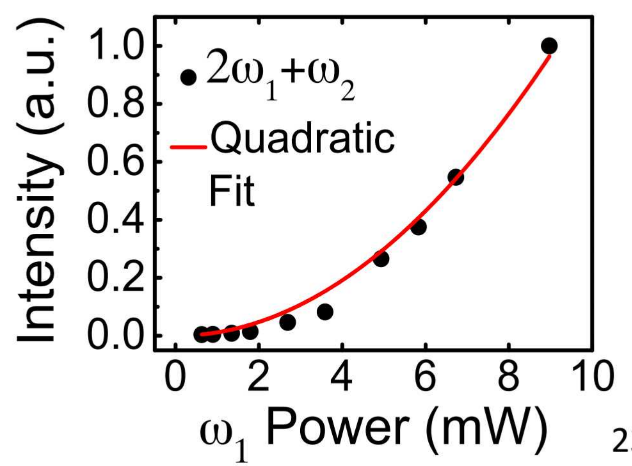
SFG power dependence





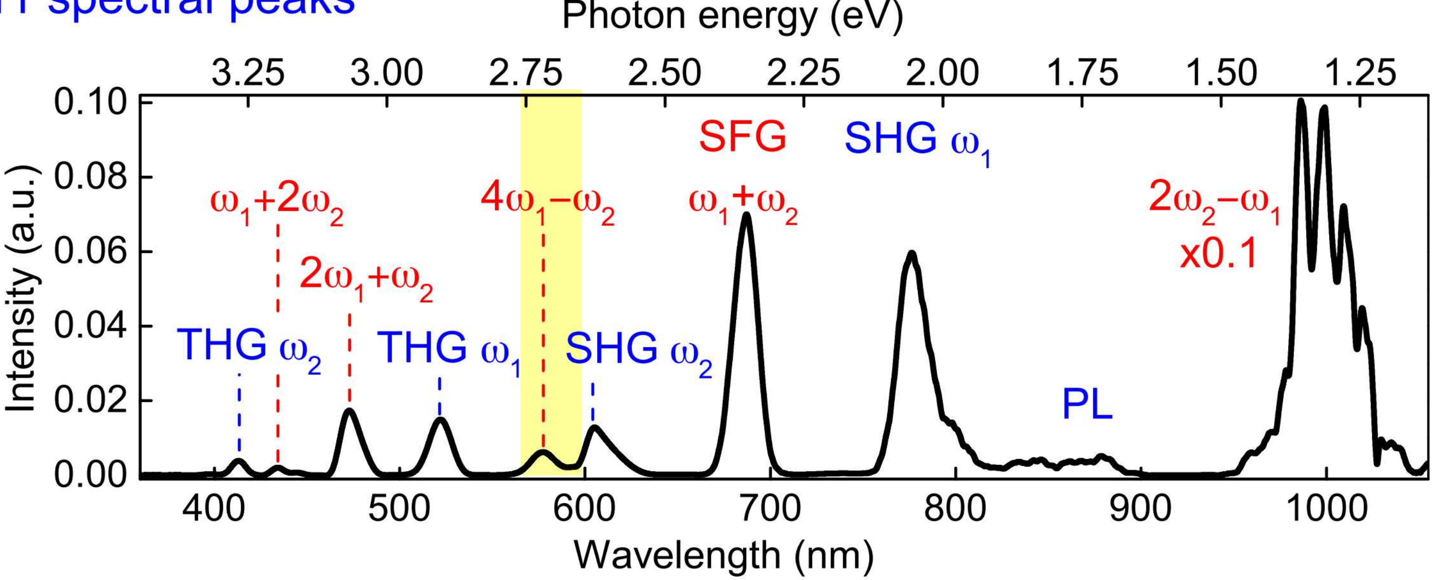


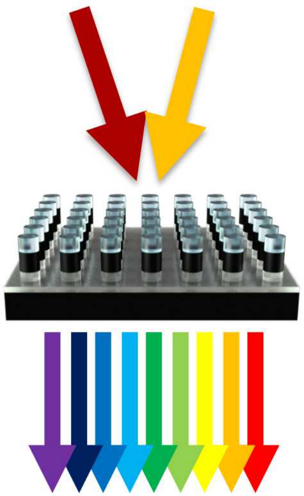
FWM power dependence



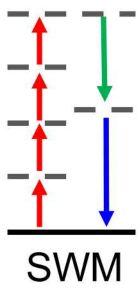
Frequency mixing spectra: six wave mixing

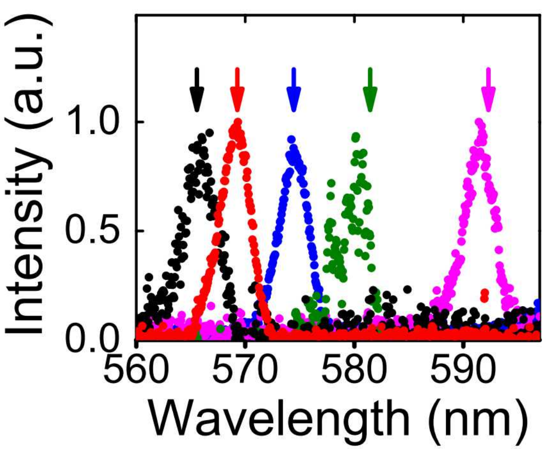
11 spectral peaks



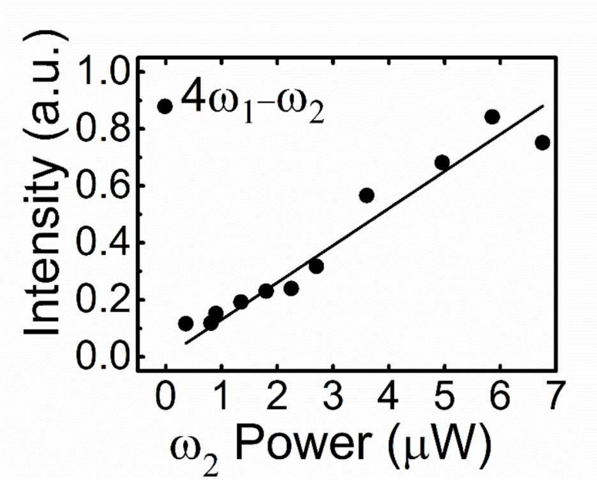


Wavelength shift



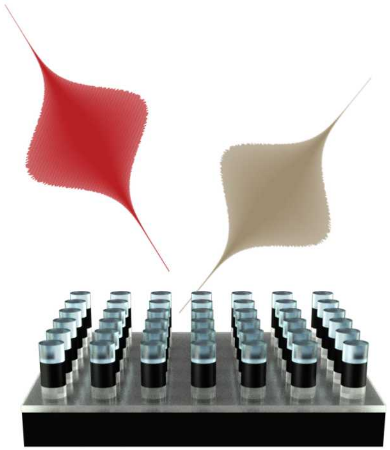


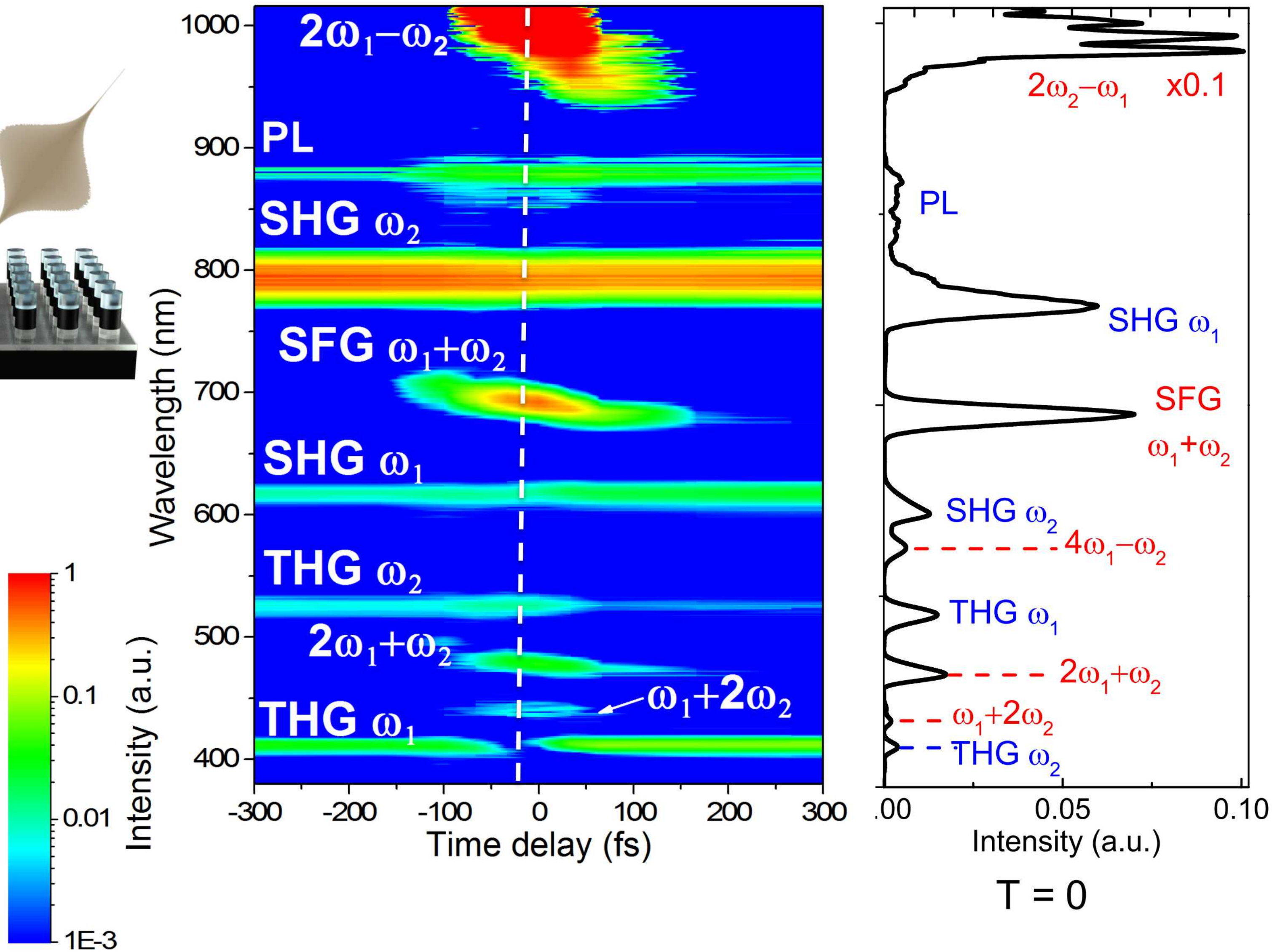
SWM power dependence



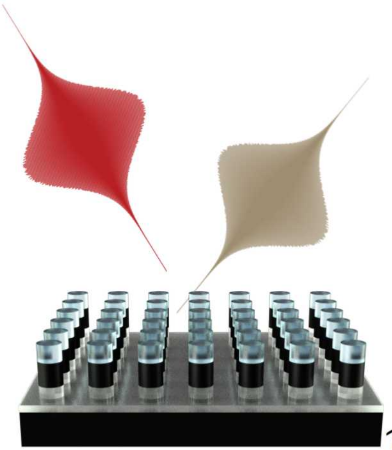
Temporal dynamics of frequency mixing

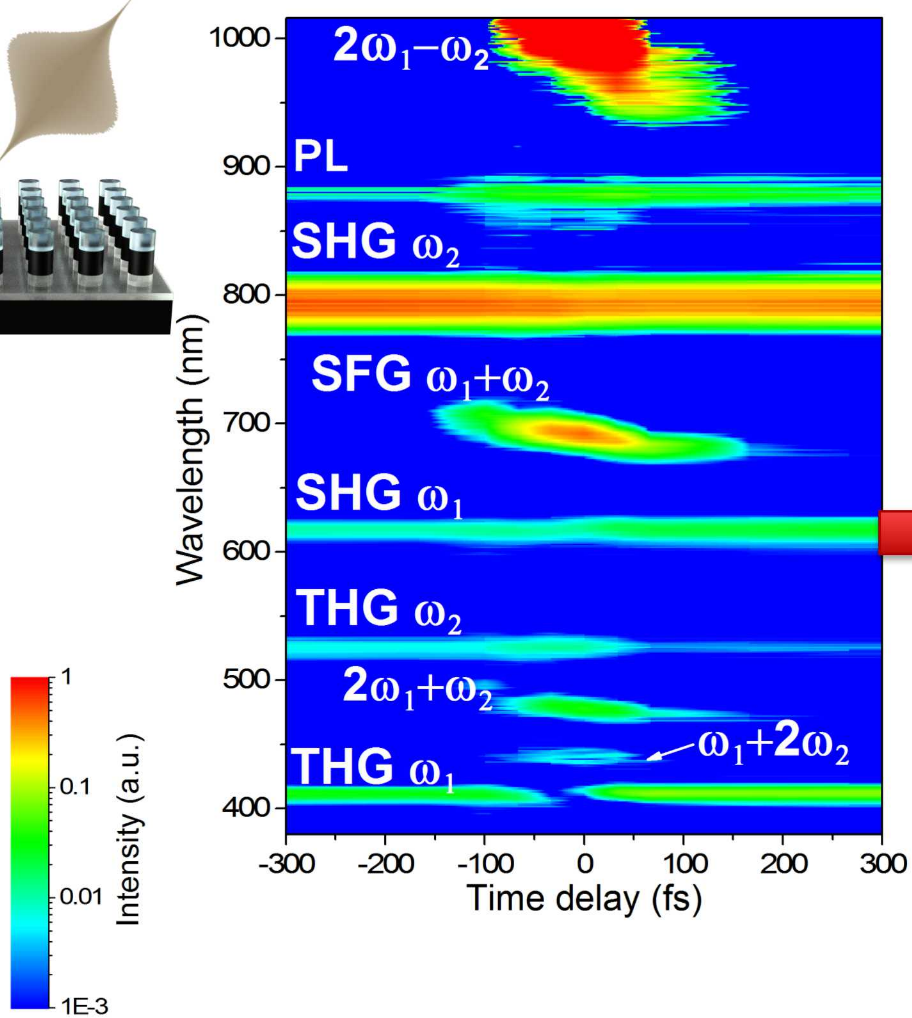
Sandia National Laboratories

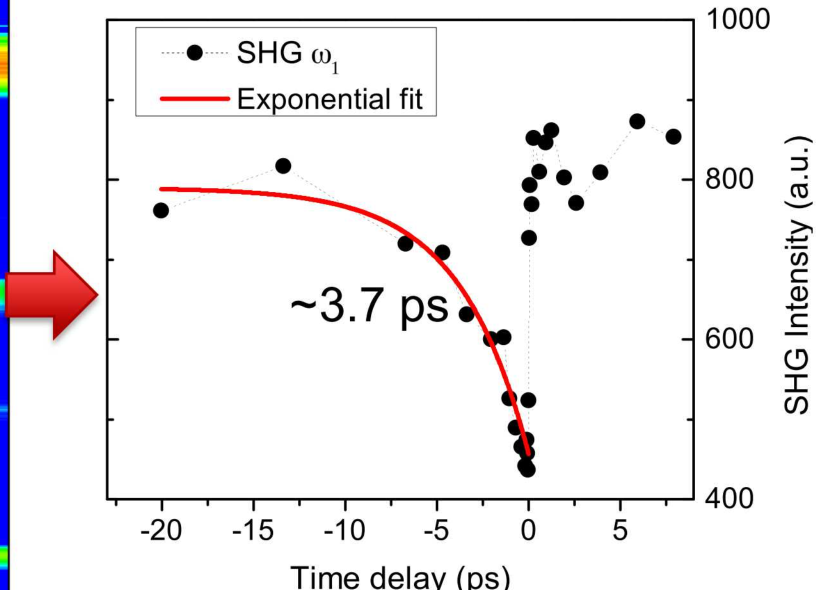




Temporal dynamics of frequency mixing

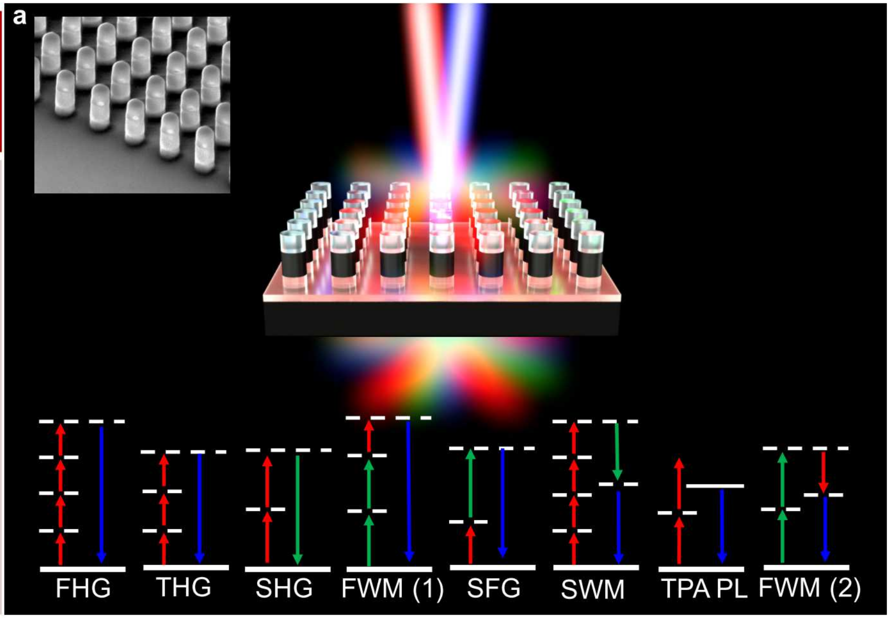






Conclusions I

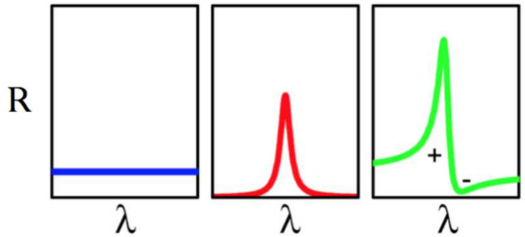
Our work	Other Dielectric metasurfaces	Plasmonic Nanotructures
SHG	SHG	SHG
THG	THG	THG
FHG	SFG	SFG
SFG	FWM	FWM
FWM (1)		
FWM (2)		
SWM		

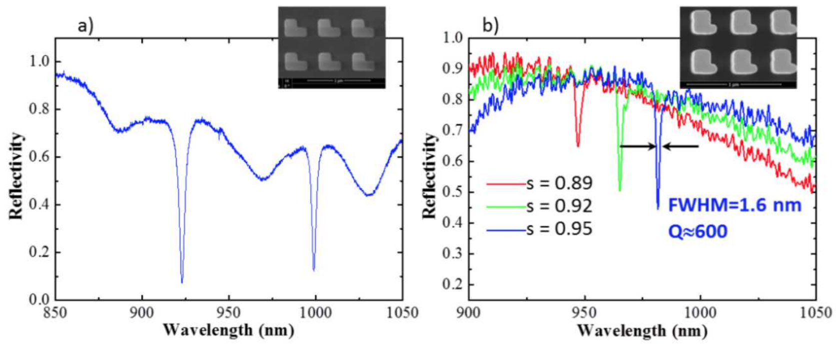


- The even and odd order nonlinearities of GaAs enable our observation of second-, third- and fourth-harmonic generation, sum-frequency generation, two-photon absorption induced photoluminescence, four-wave mixing, and six-wave mixing.

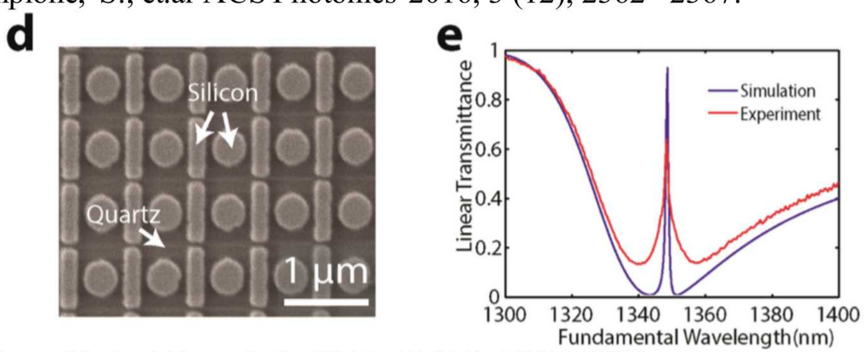
Liu, S.*, Vabishchevich, P. P.*, Vaskin, A., Reno, J. L., Keeler, G. A., Sinclair, M. B., ... & Brener, I. (2017). An optical metamixer. *arXiv preprint arXiv:1711.00090*.

High-Q Fano resonances in metasurfaces

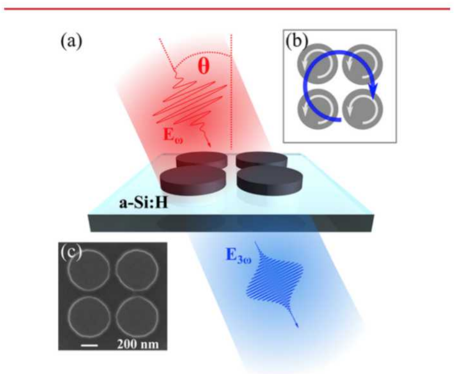


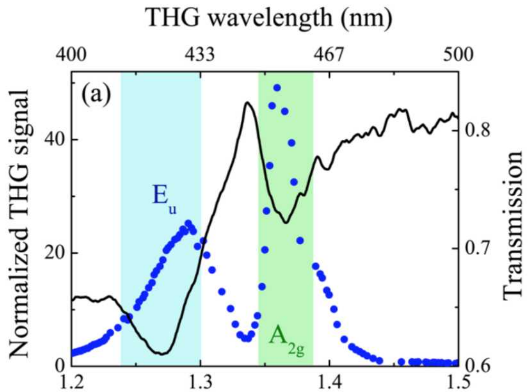


Campione, S.; et.al ACS Photonics 2016, 3 (12), 2362−2367.



Yang, Y et. al.Nano Lett. 2015, 15 (11), 7388−7393.

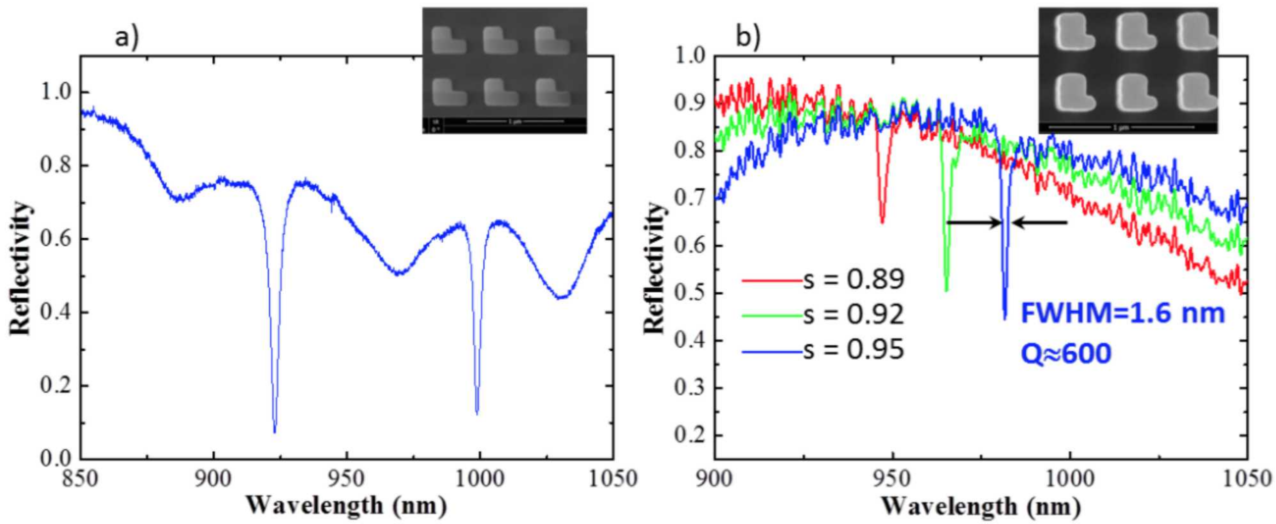




Shorokhov, A. S et al Nano Lett. 2016, 16 (8), 4857−4861.

10-fold enhancement of Third-Harmonic Generation
in Si metasurfaces with Fano resonances

Fano resonances in metasurfaces

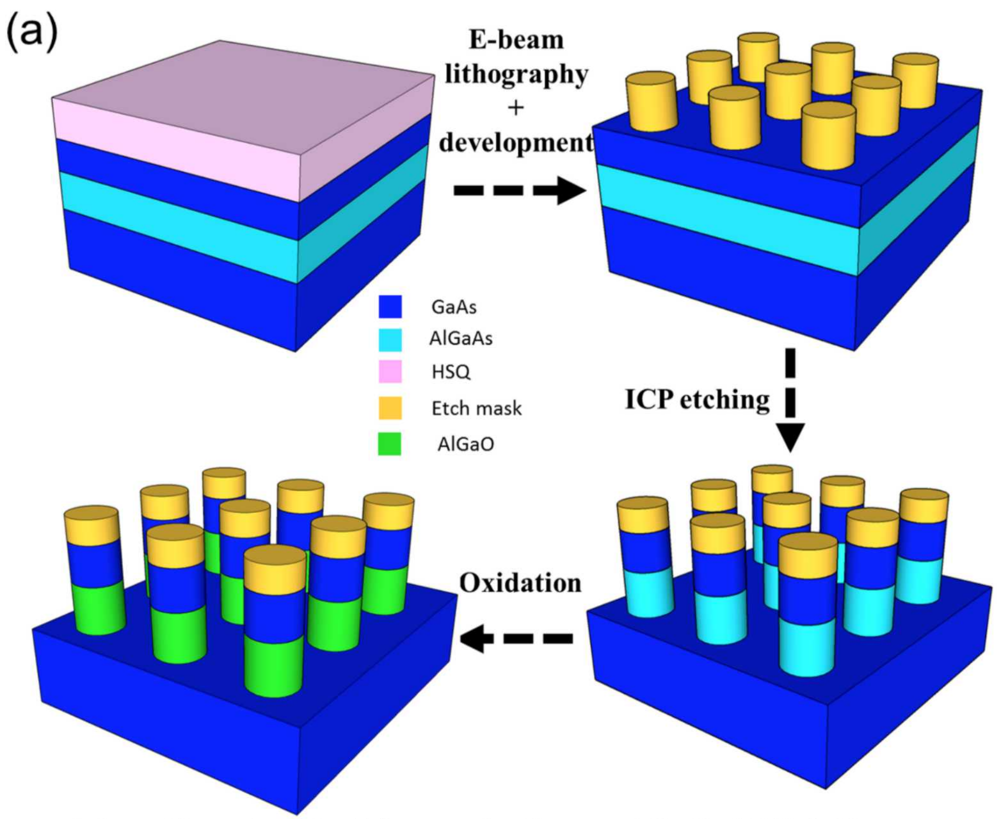


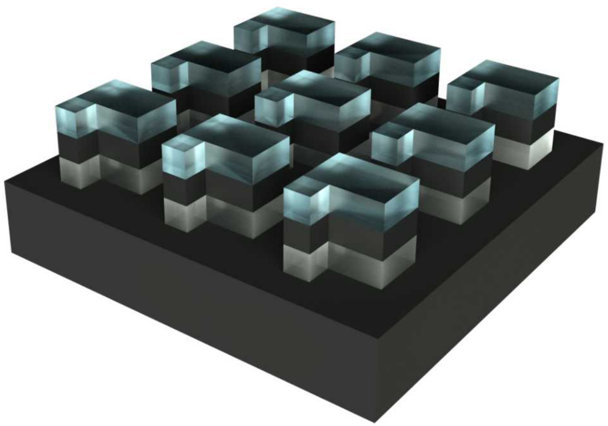
Q ~ 600

Campione, S.; et.al ACS Photonics 2016, 3 (12), 2362− 2367.

Resonantly Enhanced Second-Harmonic Generation Using Broken Symmetry III−V Semiconductor All-Dielectric Metasurfaces

GaAs Fano metasurface



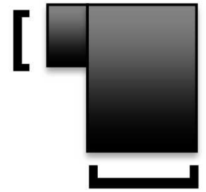


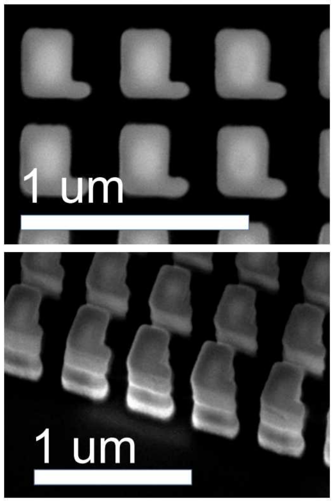
SiO_x – 300 nm

GaAs [100]– 300 nm

$(Al_xGa_{1-x})_2O_3$ – 300 nm

Period 470 nm

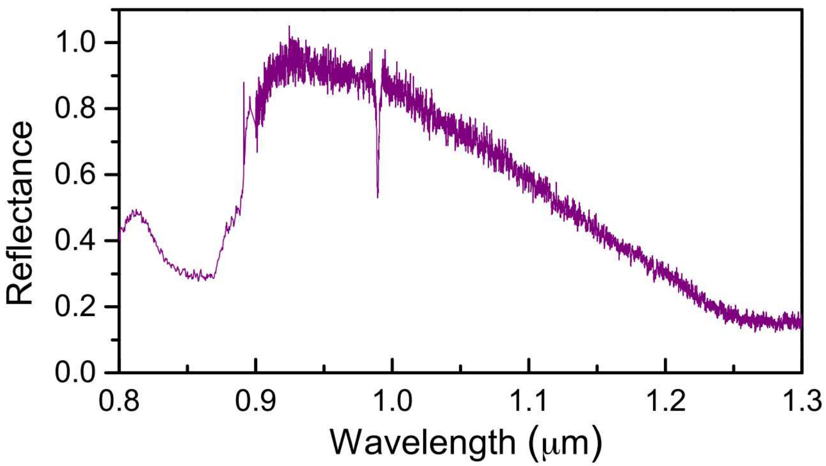


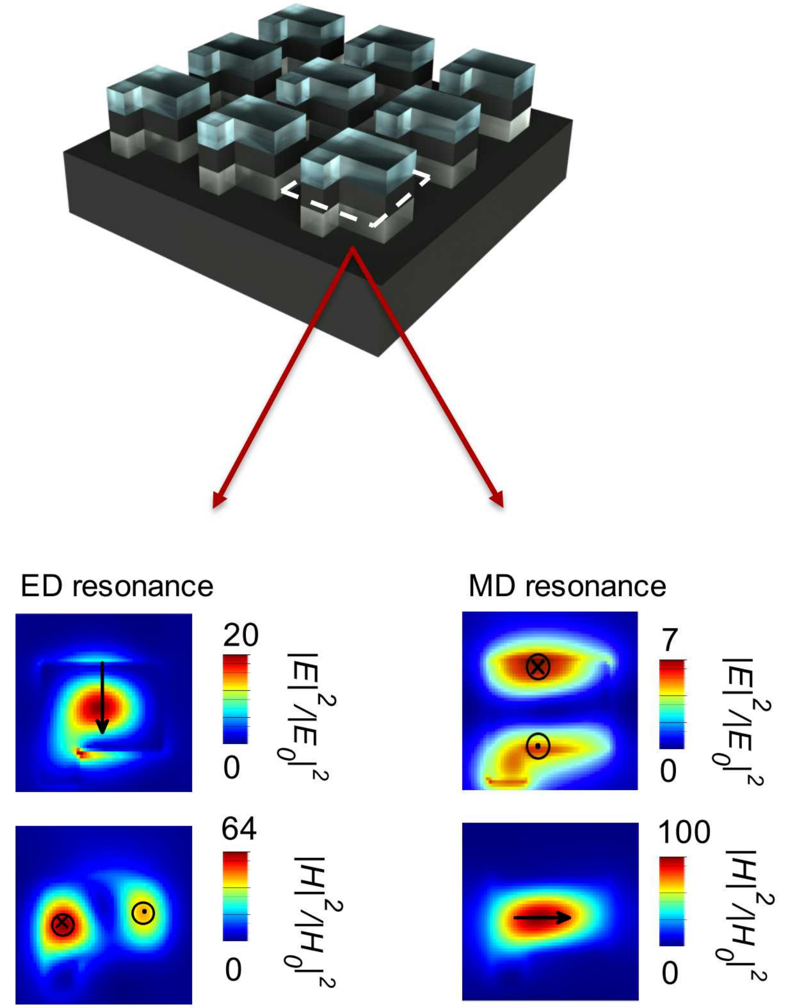


Liu, S. et. al. *Nano letters* 16.9 (2016): 5426-5432

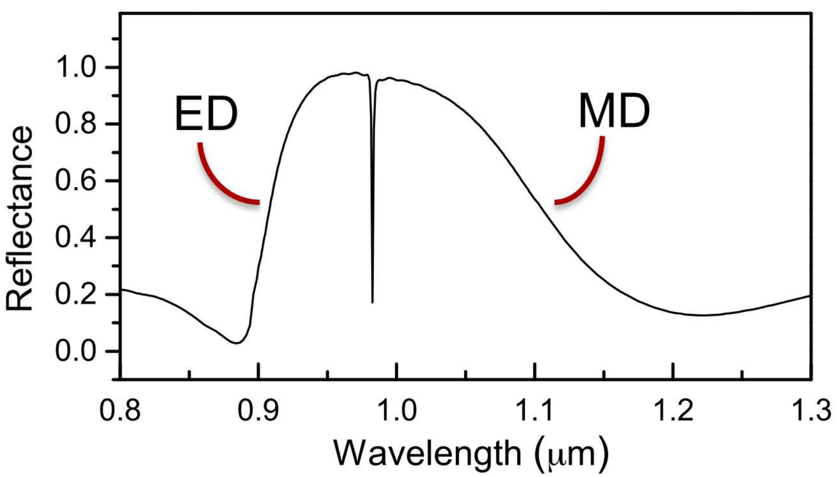
Fano metasurface reflectance spectrum

Experimental measurements



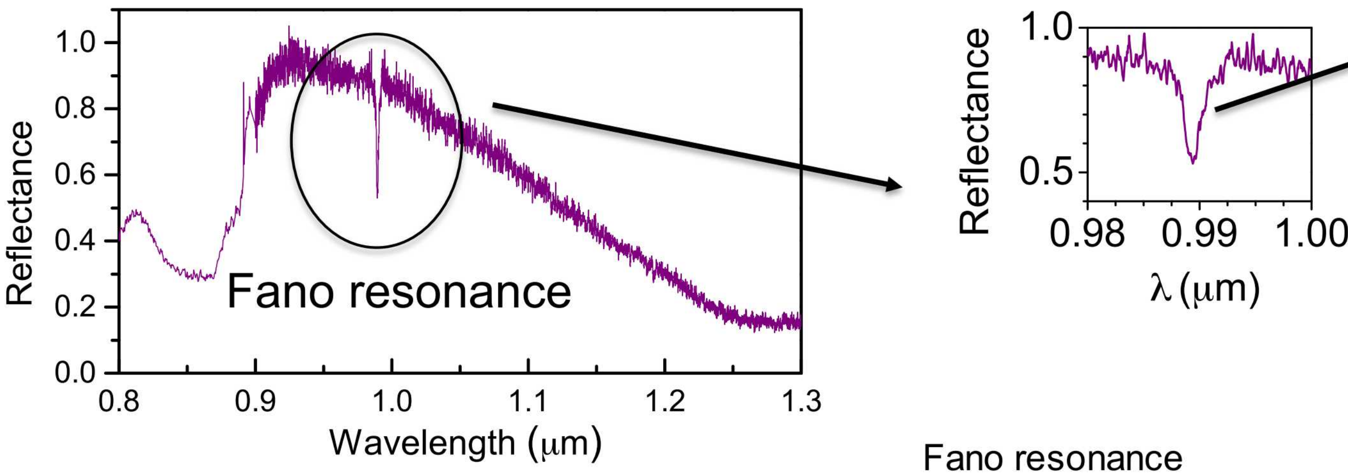


FDTD simulations



Fano metasurface reflectance spectrum

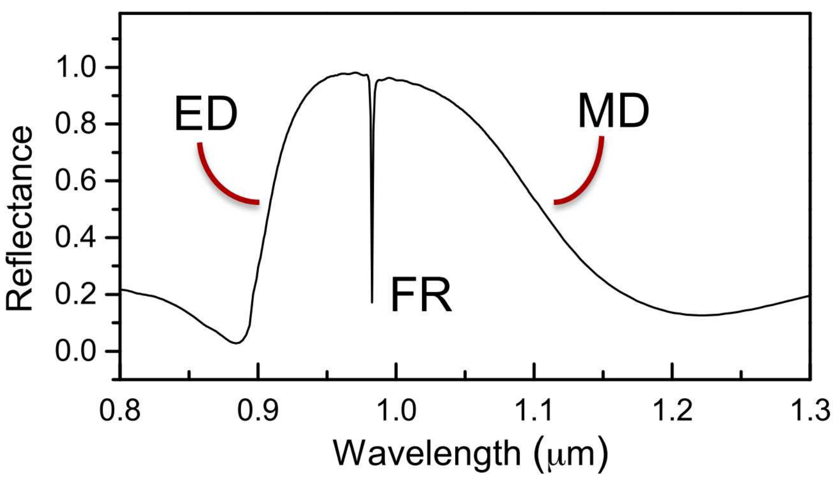
Experimental measurements



$\Delta\lambda$ ~ 2 nm

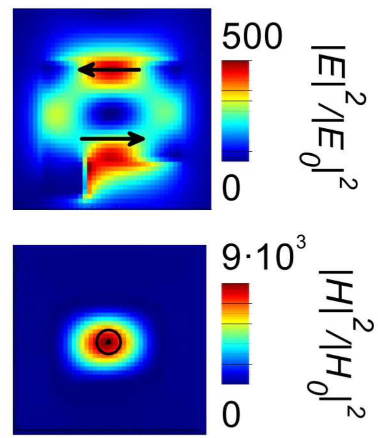
Q factor
$\lambda_F/\Delta\lambda$ ~ 500

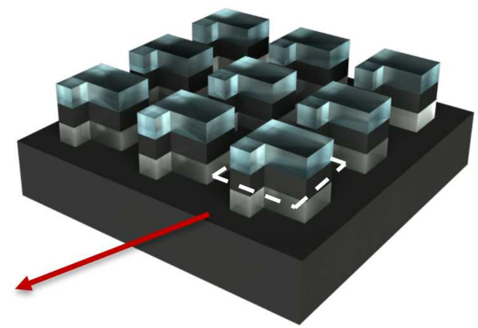
FDTD simulations



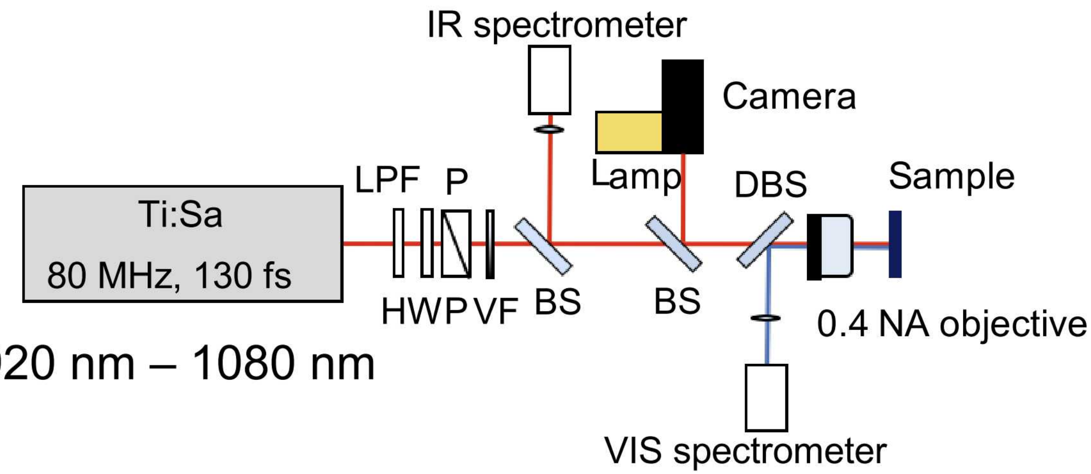
Fano resonance

λ_R~ 0.98 nm



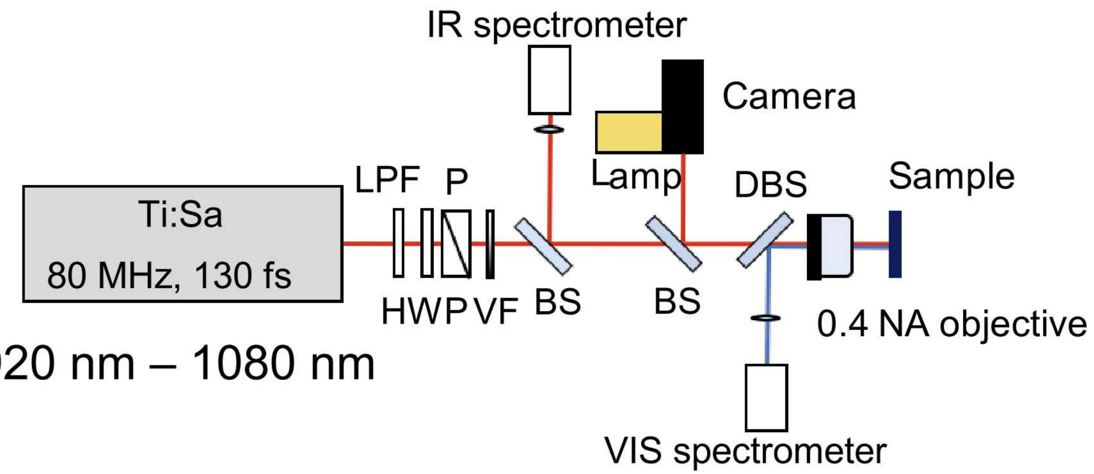


SHG in Fano metasurface

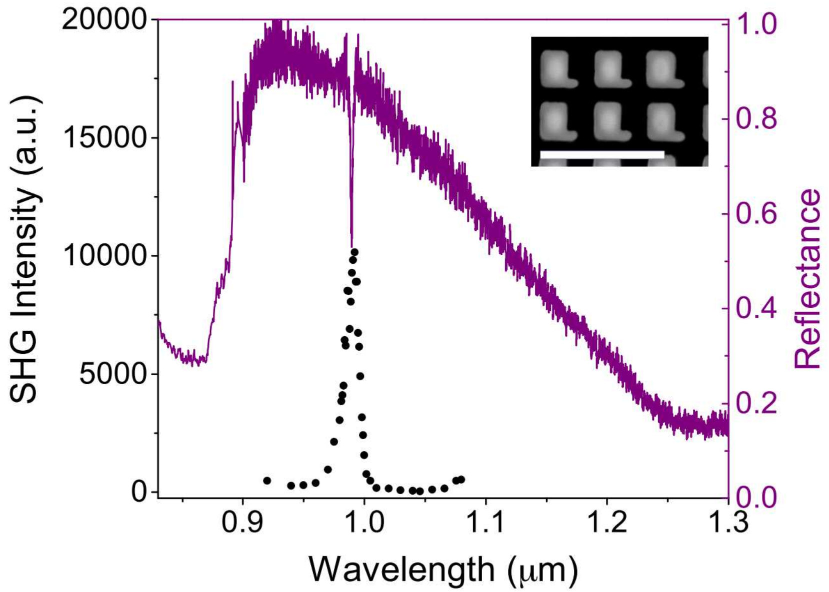


920 nm – 1080 nm

SHG in Fano metasurface



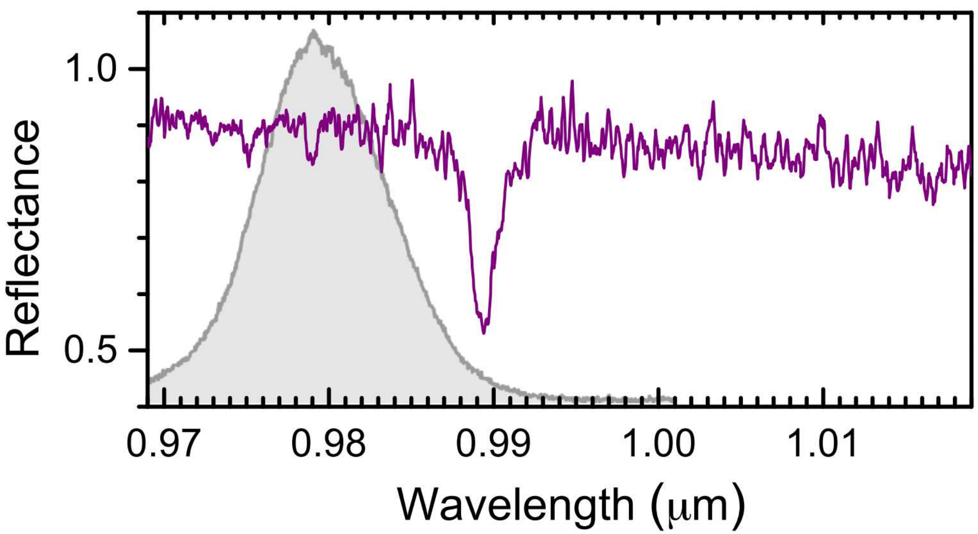
920 nm – 1080 nm

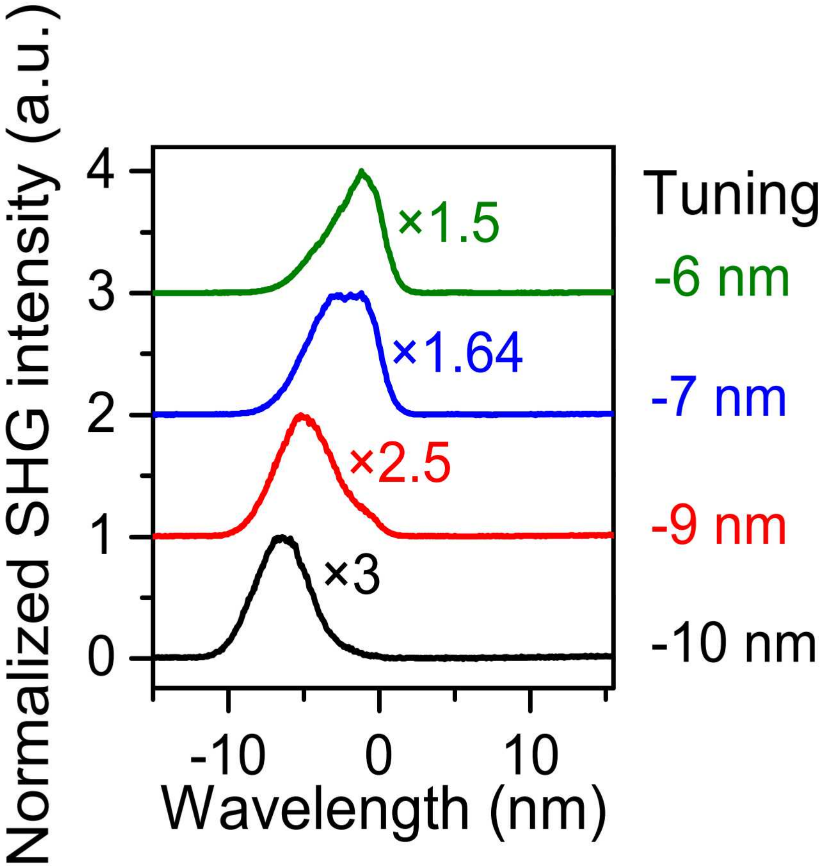


SHG intensity enhancement in the vicinity of the Fano resonance

SHG spectra modification

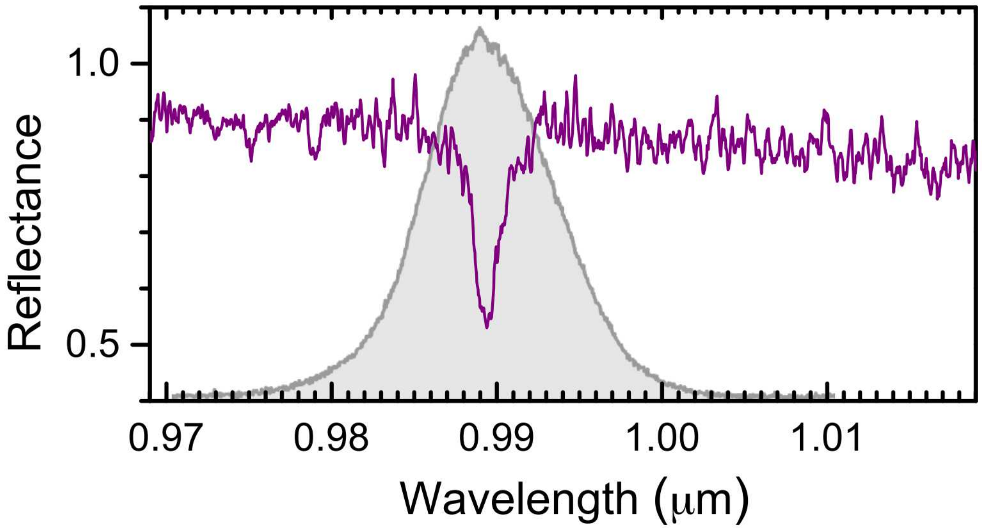
- The modification of a SHG spectrum when the pump carrier wavelength is detuned from the Fano resonant wavelength

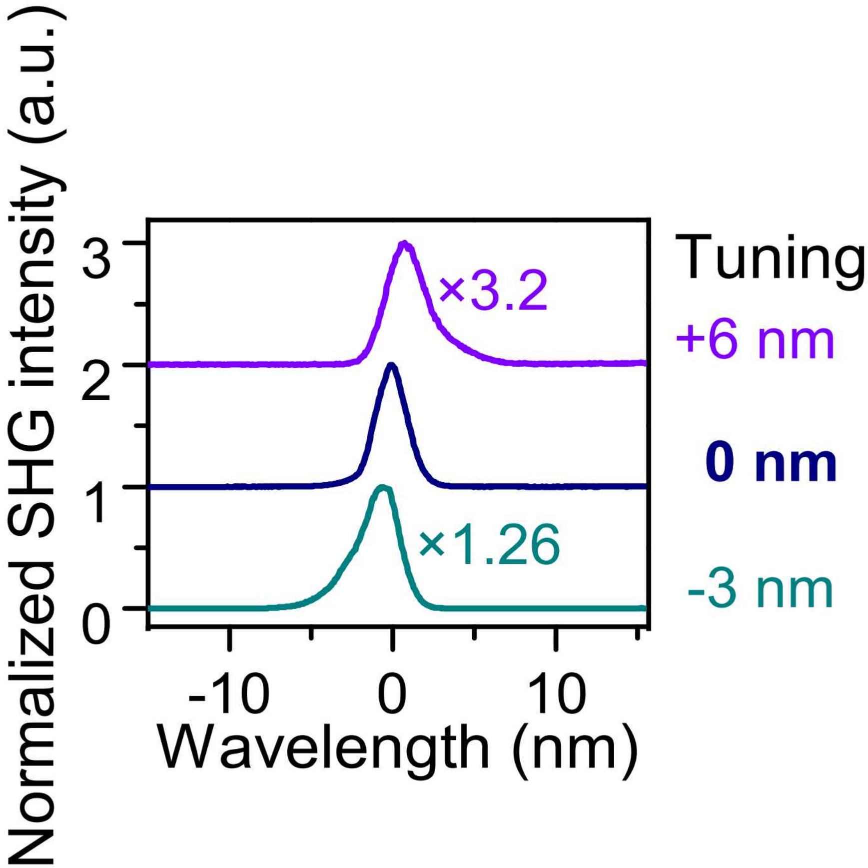




SHG spectra modification

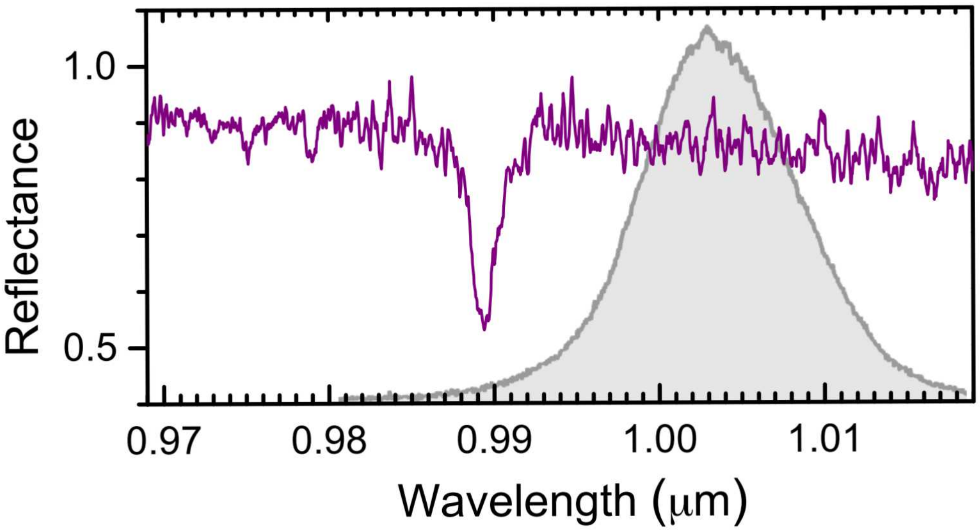
- The modification of a SHG spectrum when the pump carrier wavelength is detuned from the Fano resonant wavelength

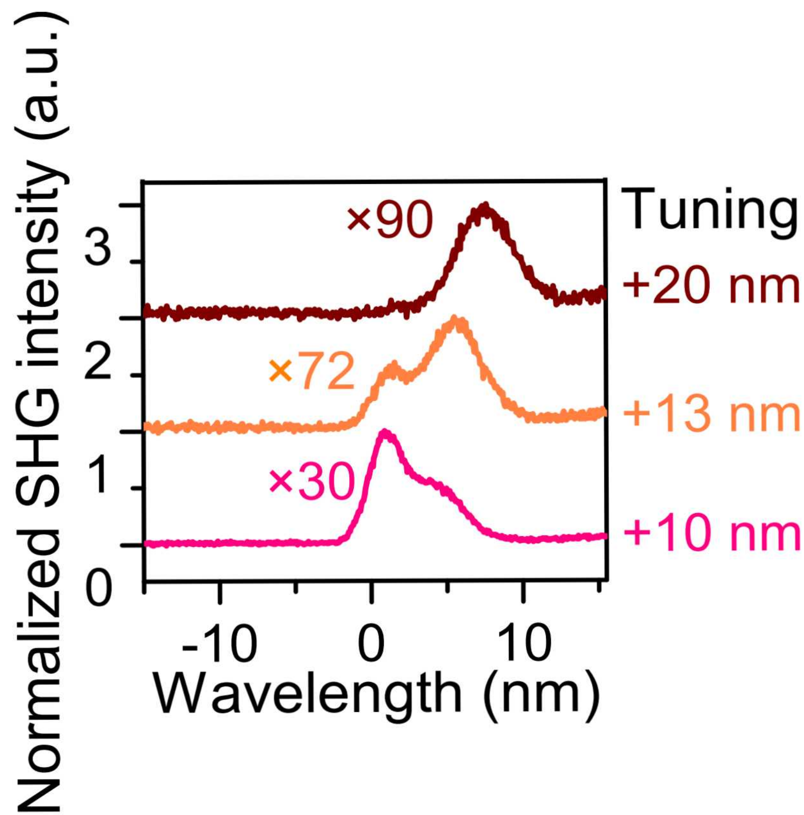




SHG spectra modification

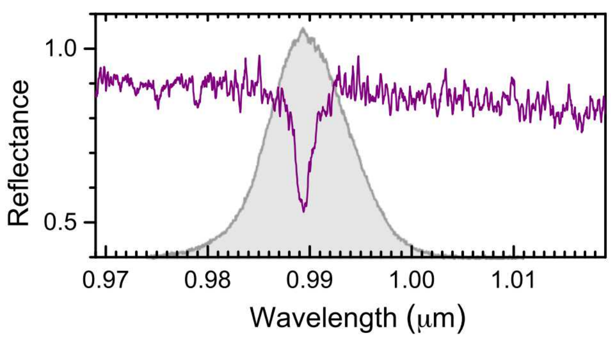
- The modification of a SHG spectrum when the pump carrier wavelength is detuned from the Fano resonant wavelength



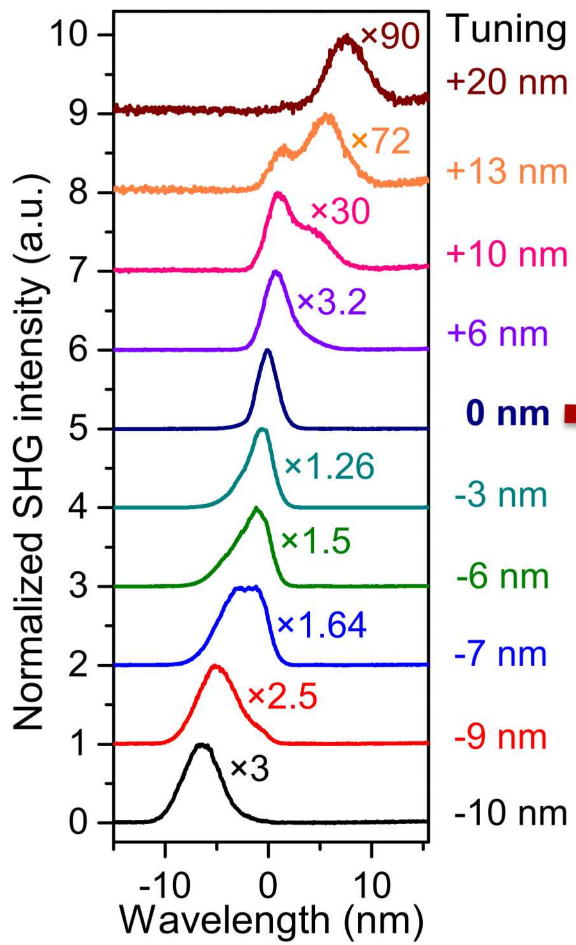


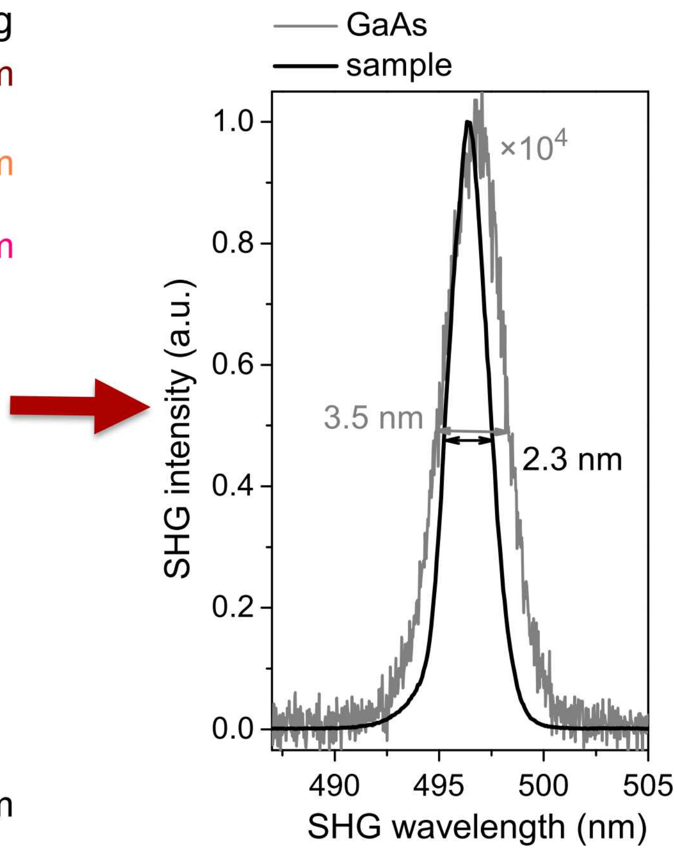
SHG spectra modification

- Narrowing of the second harmonic spectra at resonant wavelength

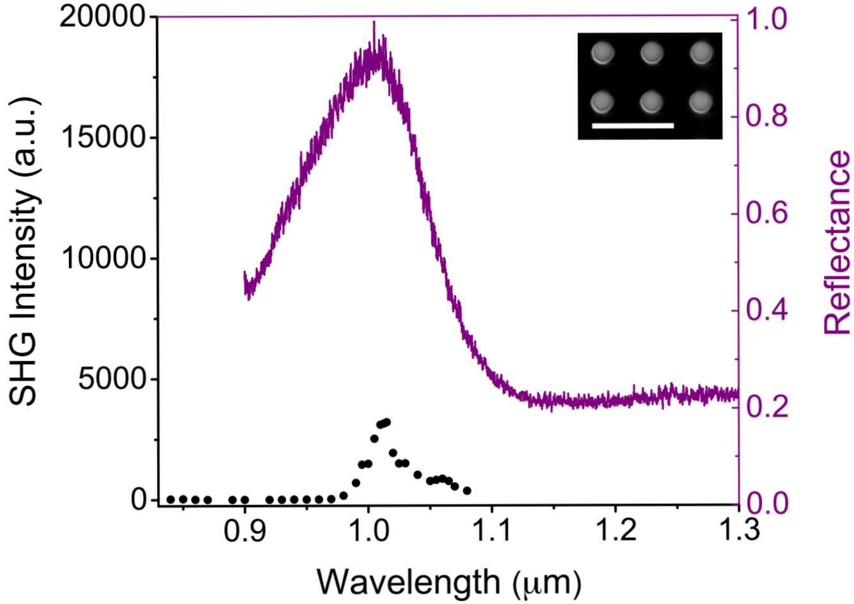


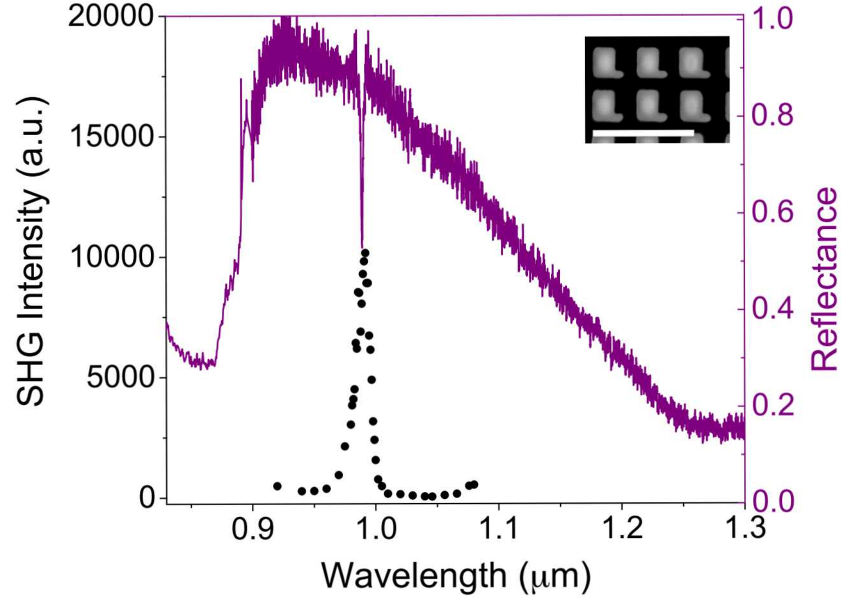
2 nm vs 11 nm

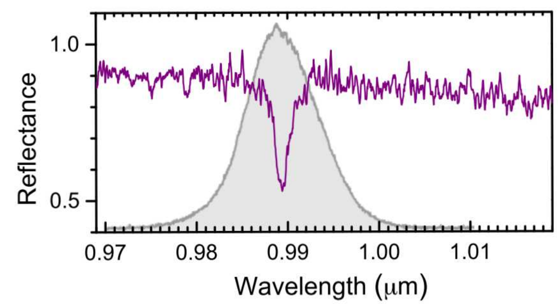




Nanodisk metasurface vs Fano metasurface





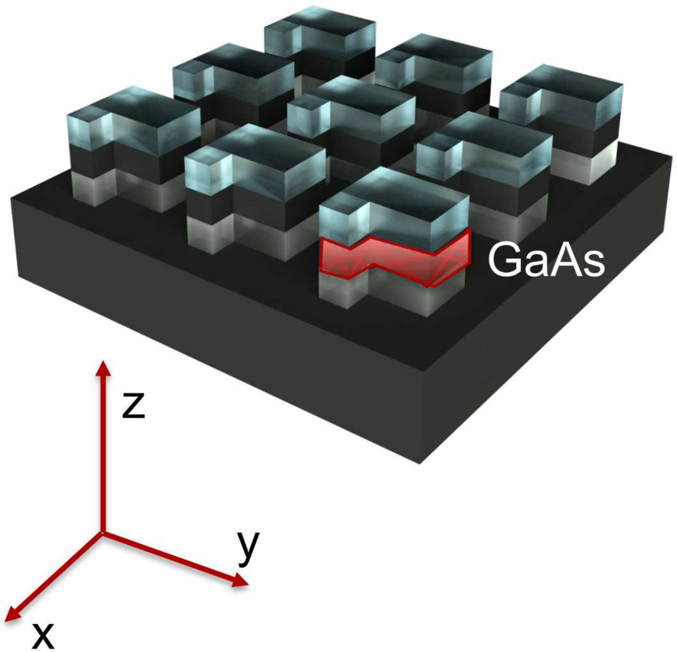


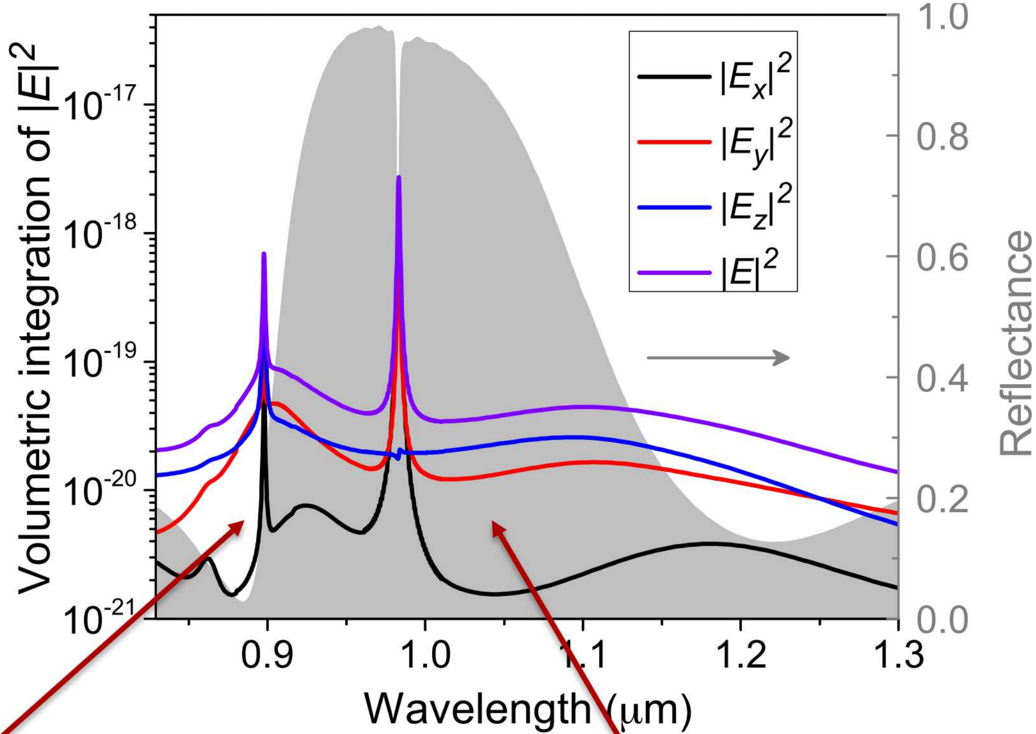
~30% of pump power is used

13-fold enhancement of SHG in Fano metasurface in comparison with the nanodisk metasurface

Numerical calculations

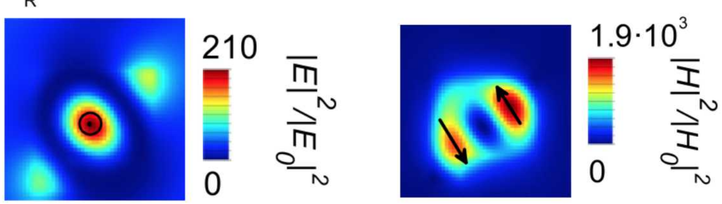
Efficiency ~ electromagnetic field enhancement





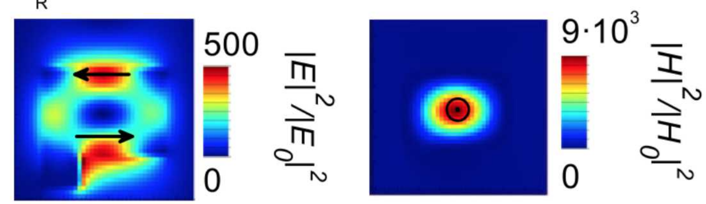
Fano resonance

λ_R~ 0.9 nm

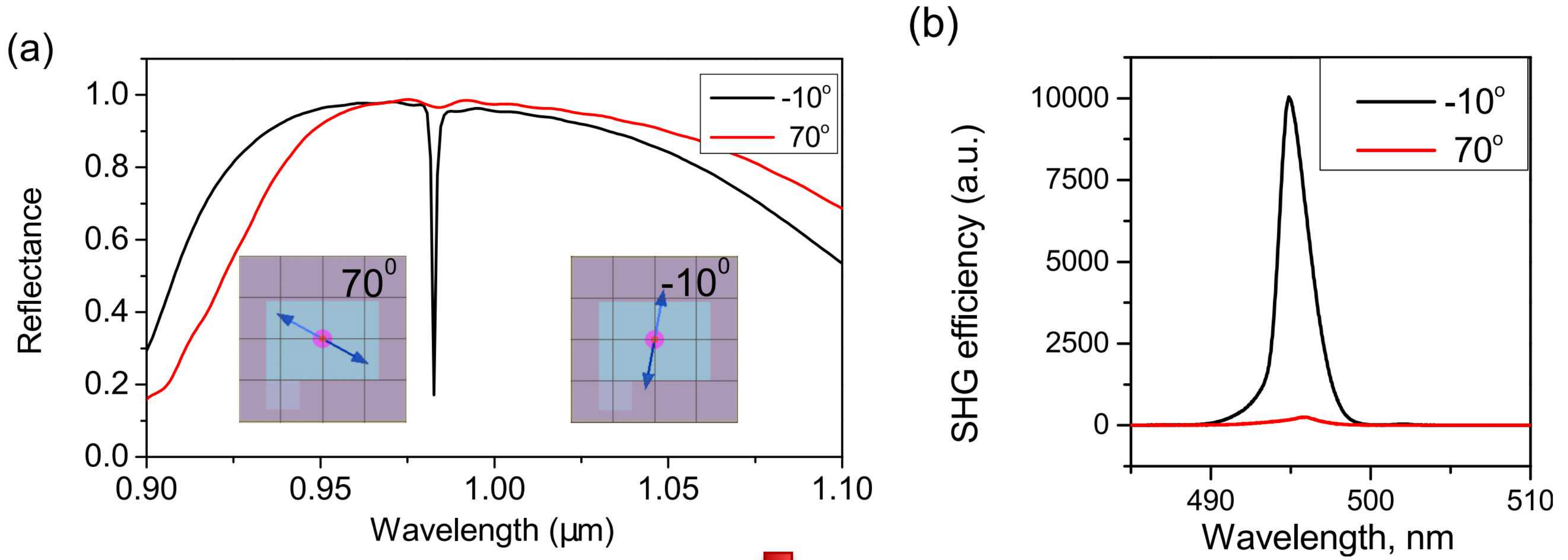


Fano resonance

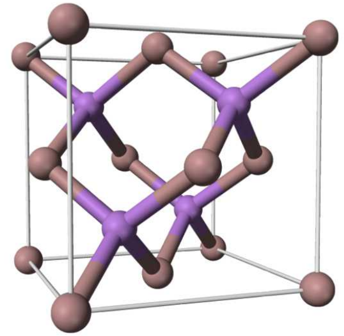
λ_R~ 0.98 nm



Polarization dependence of SHG

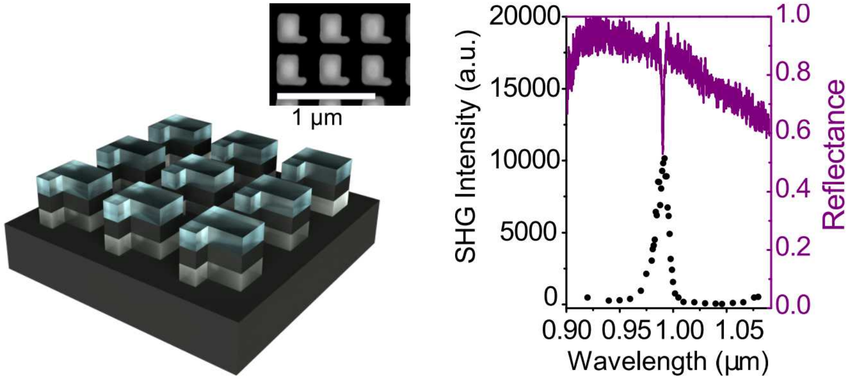


Match the asymmetric shape of the sample with the crystalline axis of GaAs



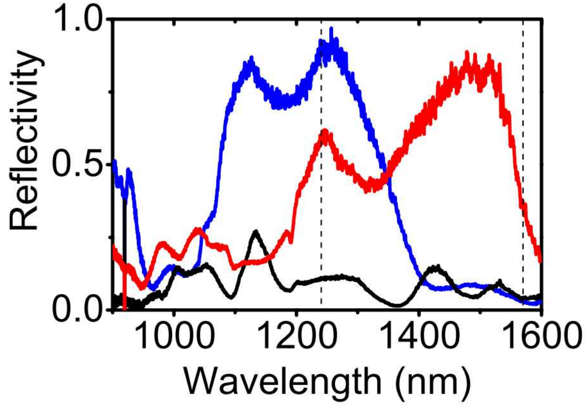
Conclusions II

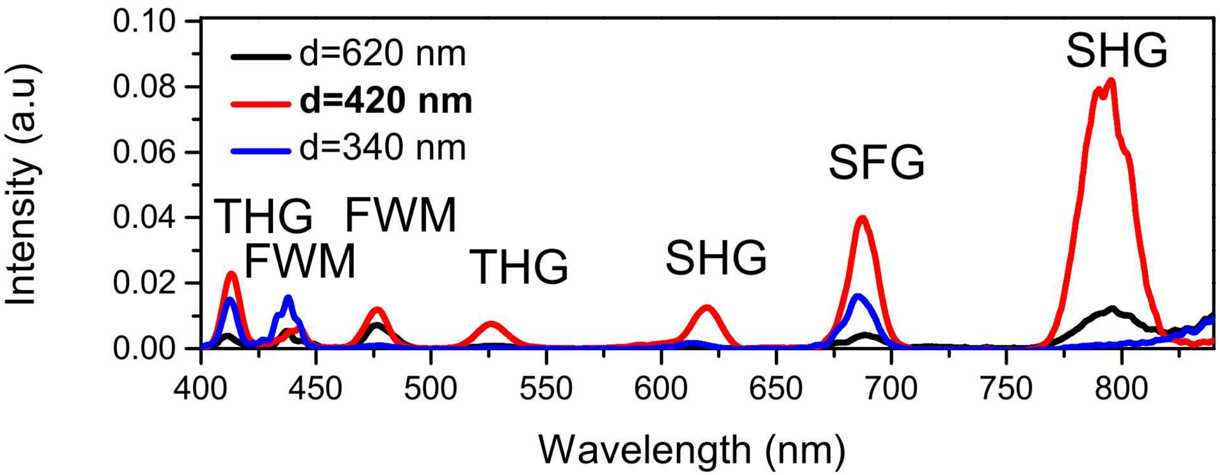
- Nontrivial spectral shaping of second-harmonic spectra in symmetry broken III-V semiconductor metasurface
- Multifold efficiency enhancement induced by high electromagnetic field localization and enhancement inside the nanoresonators.



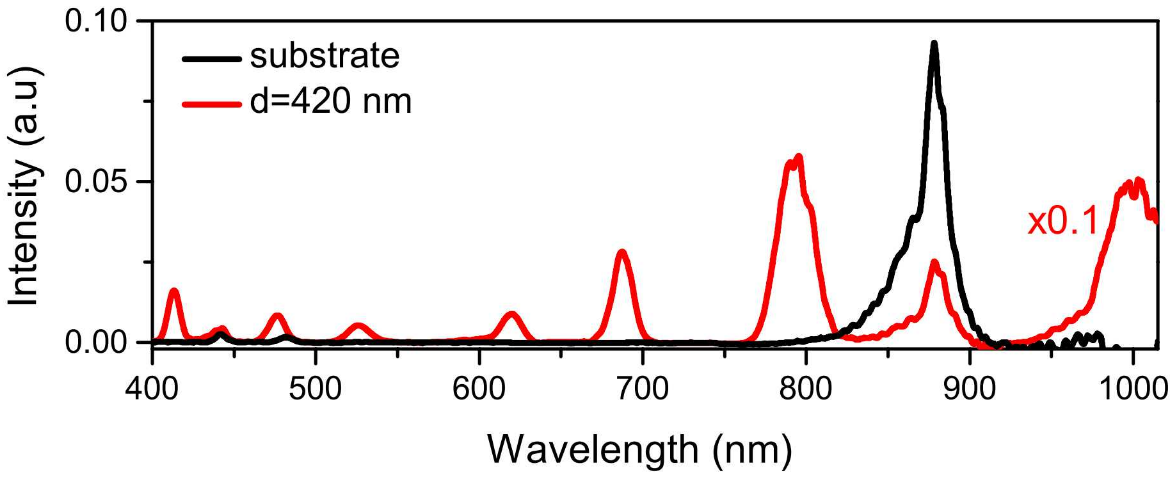
Vabishchevich, P., Liu, S., Sinclair, M. B., Keeler, G. A., Peake, G. M., & Brener, I. (2018). Enhanced Second-Harmonic Generation Using Broken Symmetry III-V Semiconductor Fano Metasurfaces. *ACS Photonics*. Article ASAP
DOI: 10.1021/acsphotonics.7b01478

Resonant enhancement – other dimensions

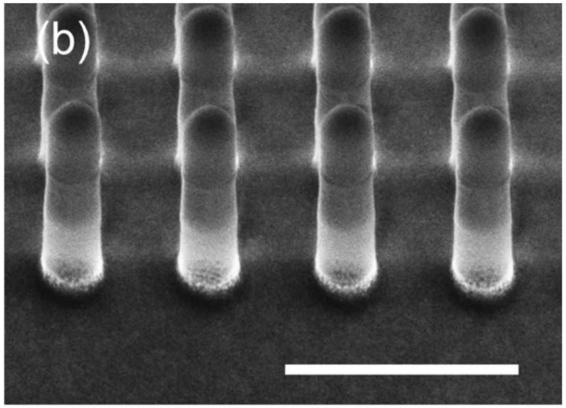


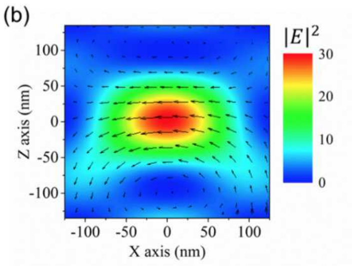


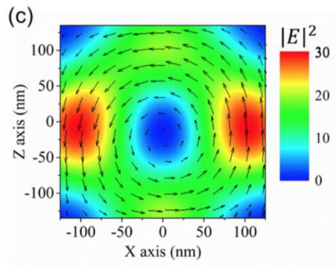
- Signal was optimized for SFG at Diameter = 420 nm.
- Mixing signal is much stronger at D=420 for most of the frequencies.

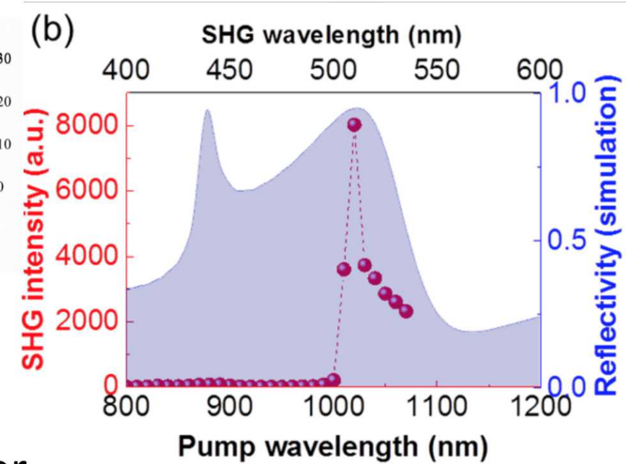


Nonlinear semiconductor metasurfaces









Efficiency:

power of generated SHG/pump power

Surface nonlinearity
Bulk nonlinearity

Second Harmonic Generation (10^{-5})

AlGaAs – Carletti, L., et al. *Optics express* 23.20 (2015): 26544-26550.
Kruk, S.et al. *Nano Lett.*, **2017**, *17* (6), pp 3914–3918
GaAs – Liu, S. et. al. *Nano letters* 16.9 (2016): 5426-5432.

Third Harmonic Generation (10^{-7})

Si – *Shcherbakov et.al. ACS Photonics*, **2015**, *2* (5), pp 578–582
a-Si – *Shorokhov et.al. Nano Lett.*, **2016**, *16* (8), pp 4857–4861