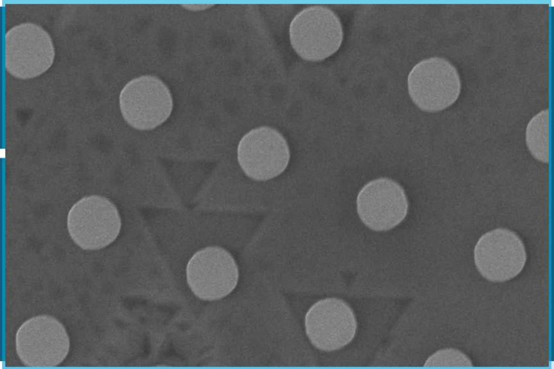
SAND2019-8954C

Enhanced Optical Nonlinearities in All-Dielectric Metasurfaces



Polina Vabishchevich [1,2], A. Vaskin[3], S. Addamane[4], S. Liu[1,2], N. Karl[1,2], A. P. Sharma[4], G. Balakrishnan[4], J. L. Reno[1,2], G. A. Keeler[1], M. B. Sinclair[1], I. Staude[3], Igal Brener[1,2]

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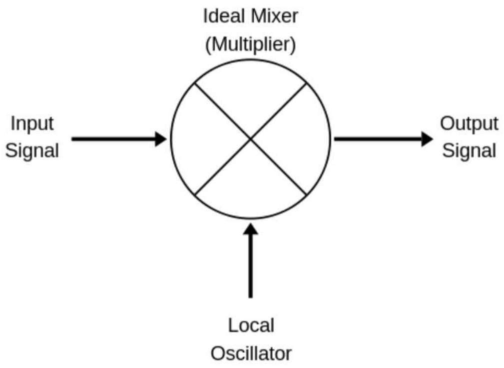
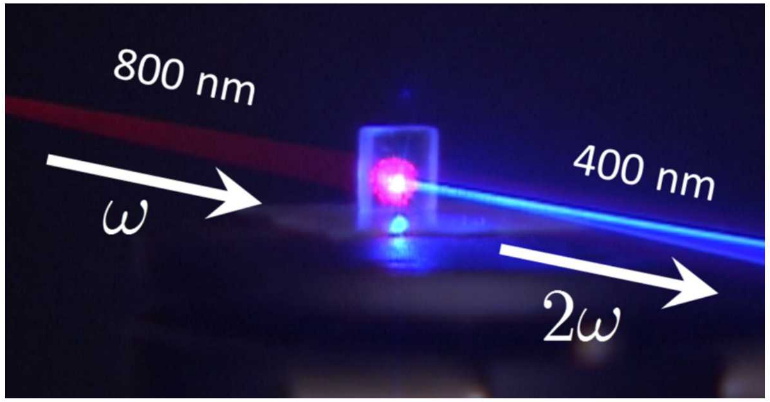
Frequency mixing – from electronics to optics

Electronics – signal processing: modulators, phase detectors, frequency synthesizers, heterodyne, etc.



- Electronics component dimensions << wavelengths

- Nonlinear optics



Variety of frequency conversion effects:
Second Harmonics Generation (SHG)
Third Harmonics Generation (THG)
Sum frequency Generation (SFG)
etc.

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

Phase matching (dispersion) is critical

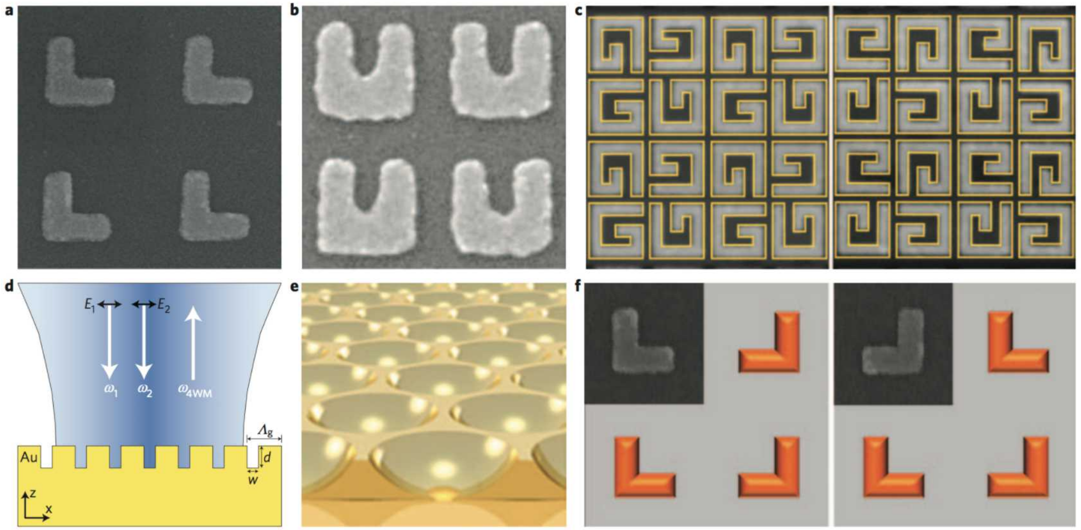
Nonlinear crystal dimensions >> wavelengths

"Super" frequency mixing?

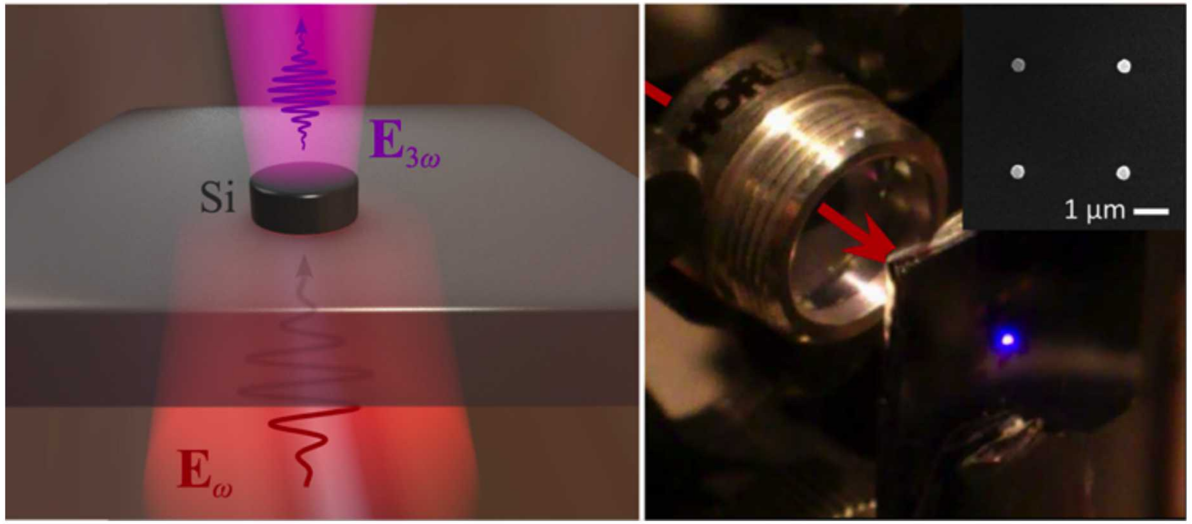
Can we see all of these mixings at the same time?

Require:
- Relaxed phase matching
- Strong nonlinear materials
- High field intensity

Nonlinear Photonic Metasurfaces



Kauranen, M., and Zayats, A. V. *Nature Photonics*, *6*(11), 737, **(2012)**



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

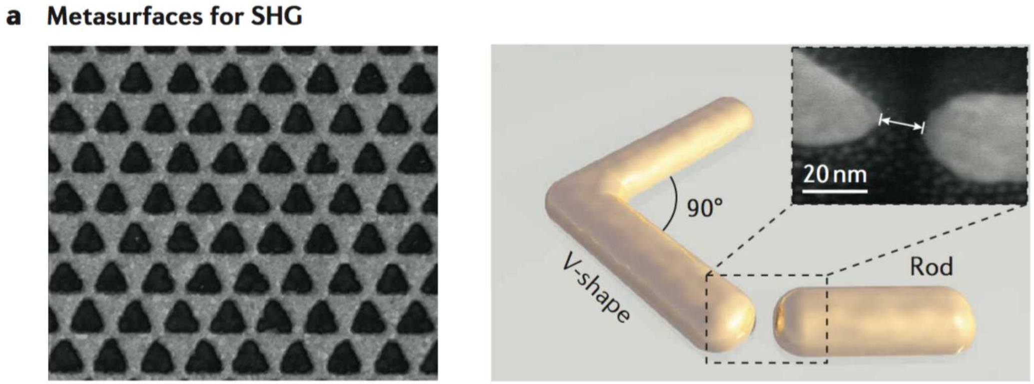
- Surface/Local Plasmon resonances
- Mie resonances

Resonant enhancement of EM field

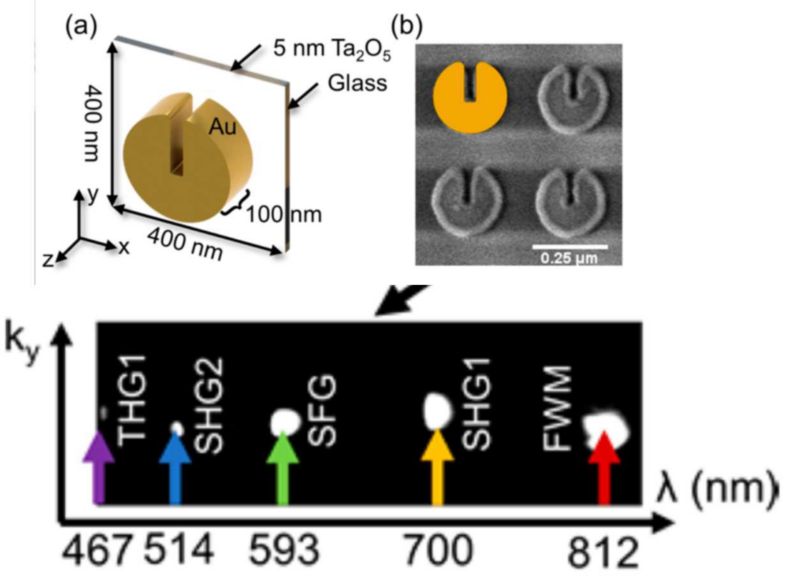
Dimensions < wavelength

Relaxed phase matching conditions

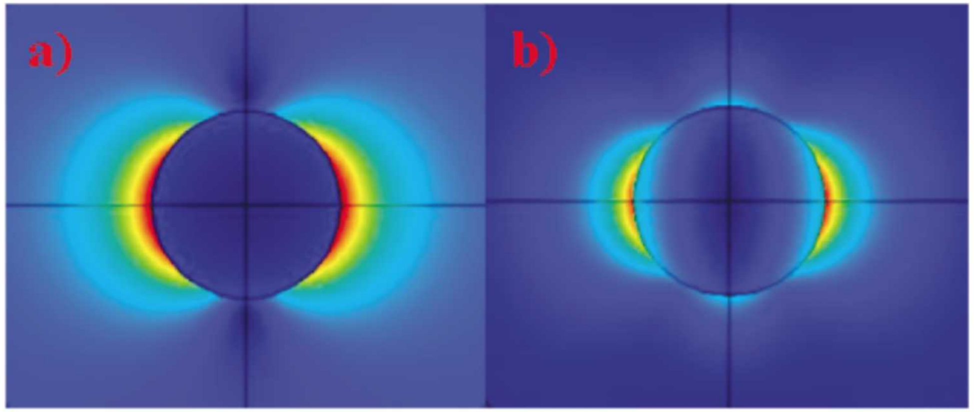
Nonlinear plasmonics



Li, G., Zhang, S. and Zentgraf, T., “Nonlinear photonic metasurfaces” *Nature Reviews Materials*, (2017), *2*(5), p.17010



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

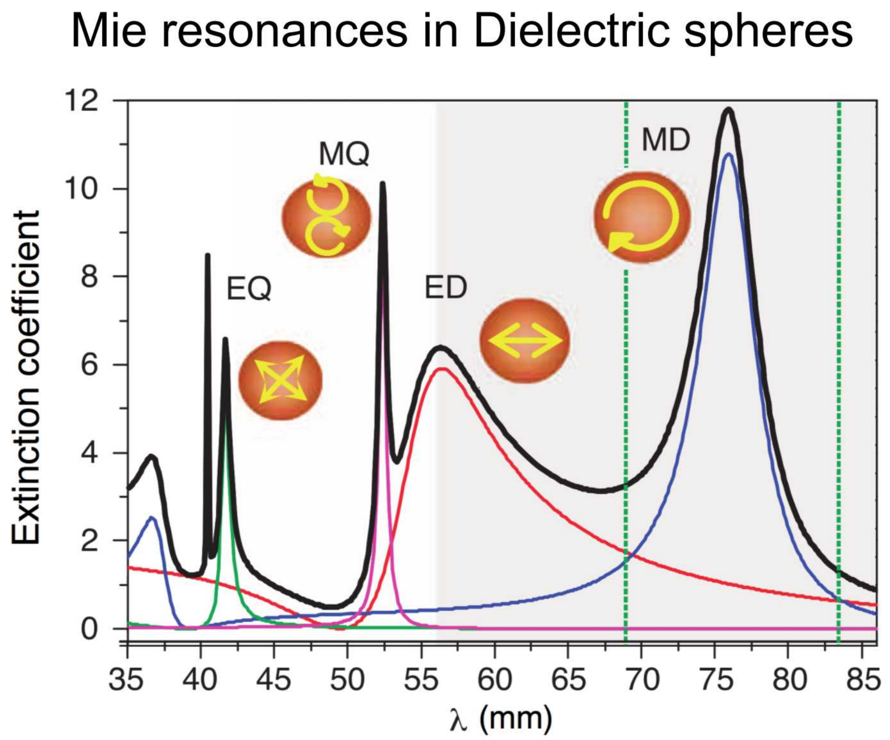


Butet, Jérémy, et al. *Nano letters* 10.5 (2010): 1717-1721.

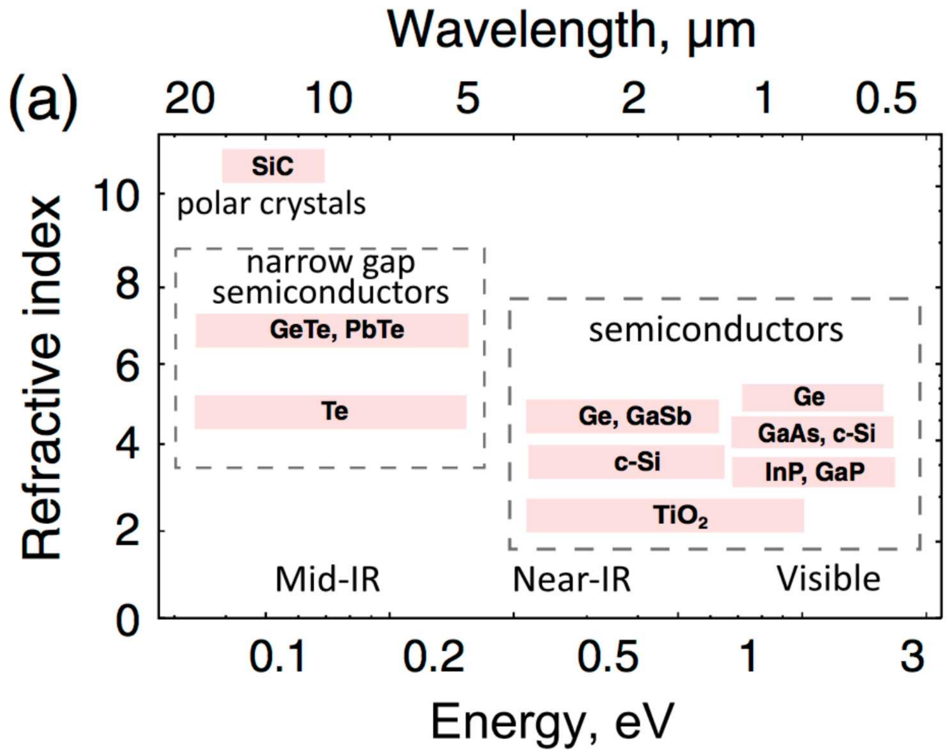
Plasmonic metasurfaces:
- Ohmic losses
- Plasmonic structure: small modal volume (surface nonlinearities)

SHG efficiency ~ 5.1×10^{-10} W/W^2

Mie resonances in dielectric spheres



Geffrin, J.-M., et al. *Nat. Commun.* 3, 1171, **2012**



Baranov, D.G., et al. *Optica* 4.7, 814-825, **2017**

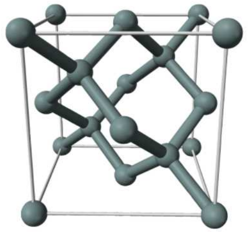
Mie resonances

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

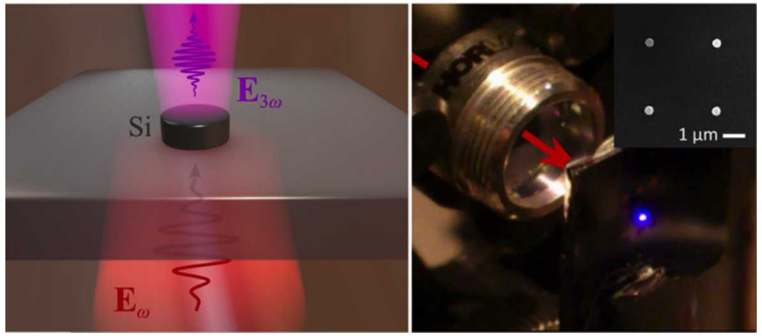
Nonlinear dielectric metasurfaces

centrosymmetric

Si
Ge

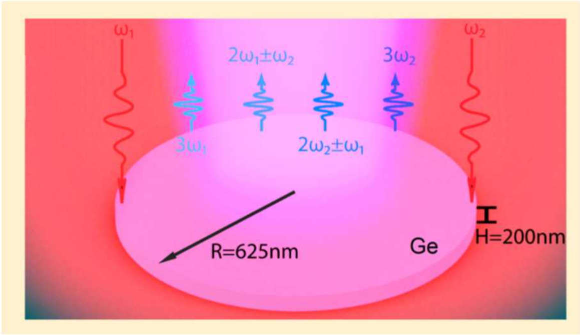


Third Harmonic Generation



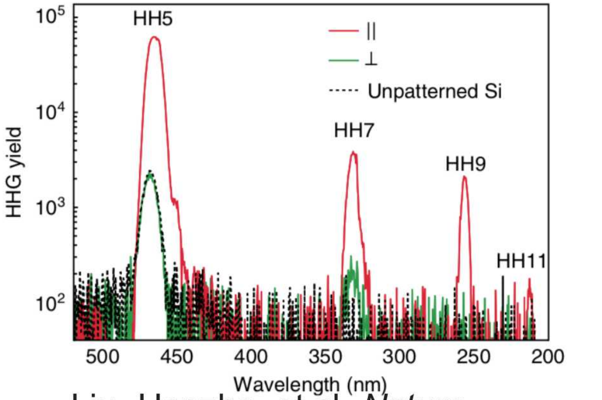
M .R. Shcherbakov, et. al. *Nano Letters* **14,** 11, 6488–6492, **2014**

Four-Wave Mixing



Grinblat, G., et al, ACS Photonics 4, 2144-2149, **2017**

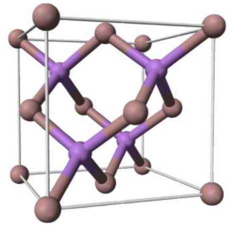
High-Harmonic Generation



Liu, Hanzhe, et al. *Nature Physics* 14.10,1006, **2018**

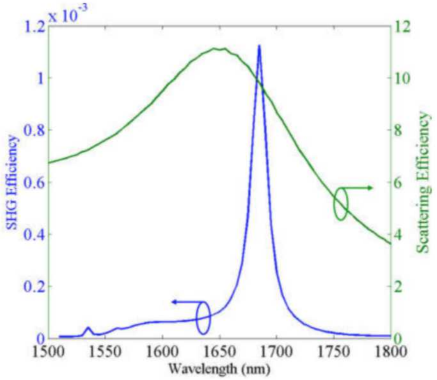
Non-centrosymmetric

GaAs
AlGaAs



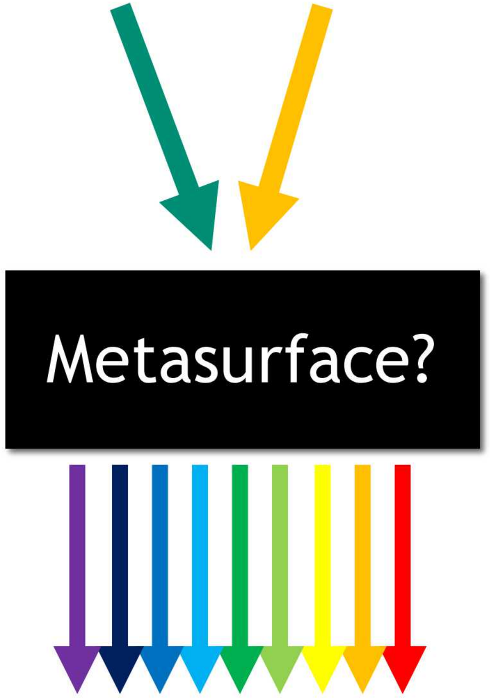
https://www.wikipedia.org

Second Harmonic Generation (10^{-8} W/W^2)



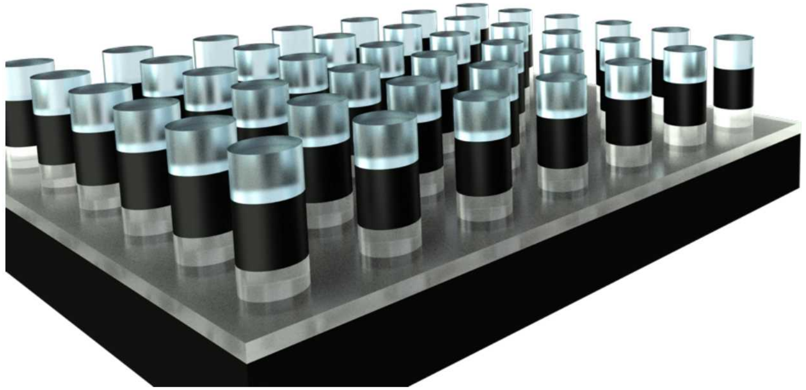
L. Carletti et al., *ACS Photonics*, *3*(8), 1500-1507, **2016**

AlGaAs – Carletti, L., et al. *Optics express* 23.20, 26544-26550**, 2015**
Camacho-Morales, R., et al. *Nano letters* 16.11, 7191-7197, **2016**
Kruk, S.et al. *Nano Lett.*, *17* (6), pp 3914–3918, **2017**
GaAs – Liu, S. et. al. *Nano letters* 16.9, 5426-5432, **2016**



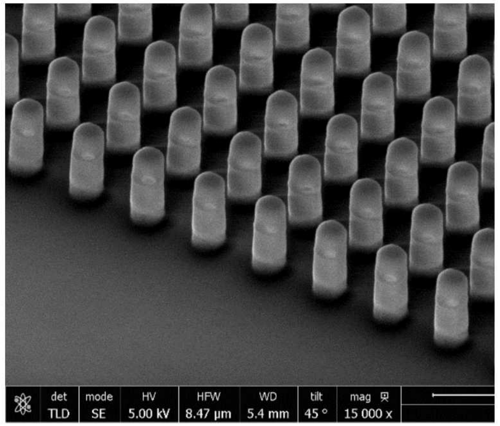
Multiple frequency mixing in dielectric metasurfaces

GaAs based dielectric metasurface

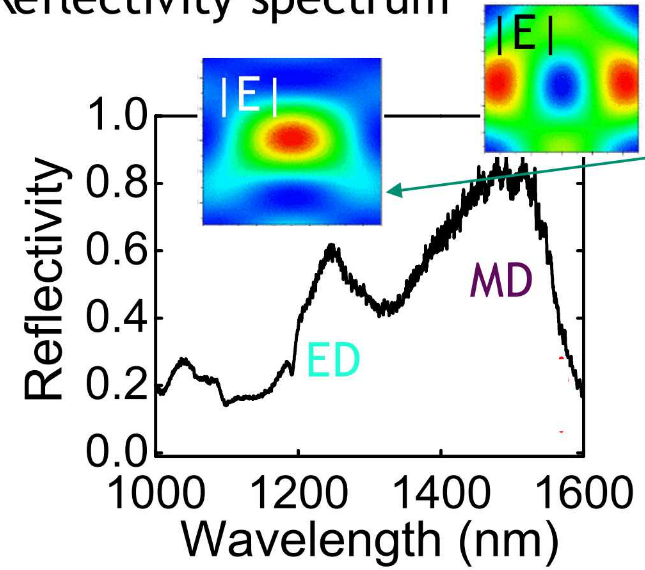


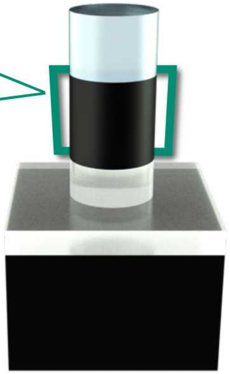
SiO_x – 300 nm
GaAs - 400 nm
$(Al_xGa_{1-x})_2O_3$ – 450 nm
Diameter 400 nm
Period 840 nm

SEM image

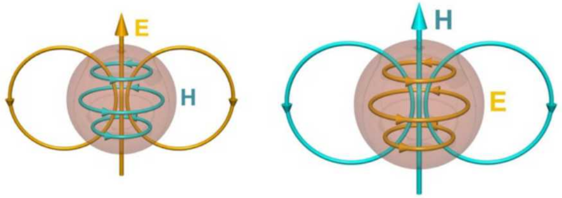


Reflectivity spectrum





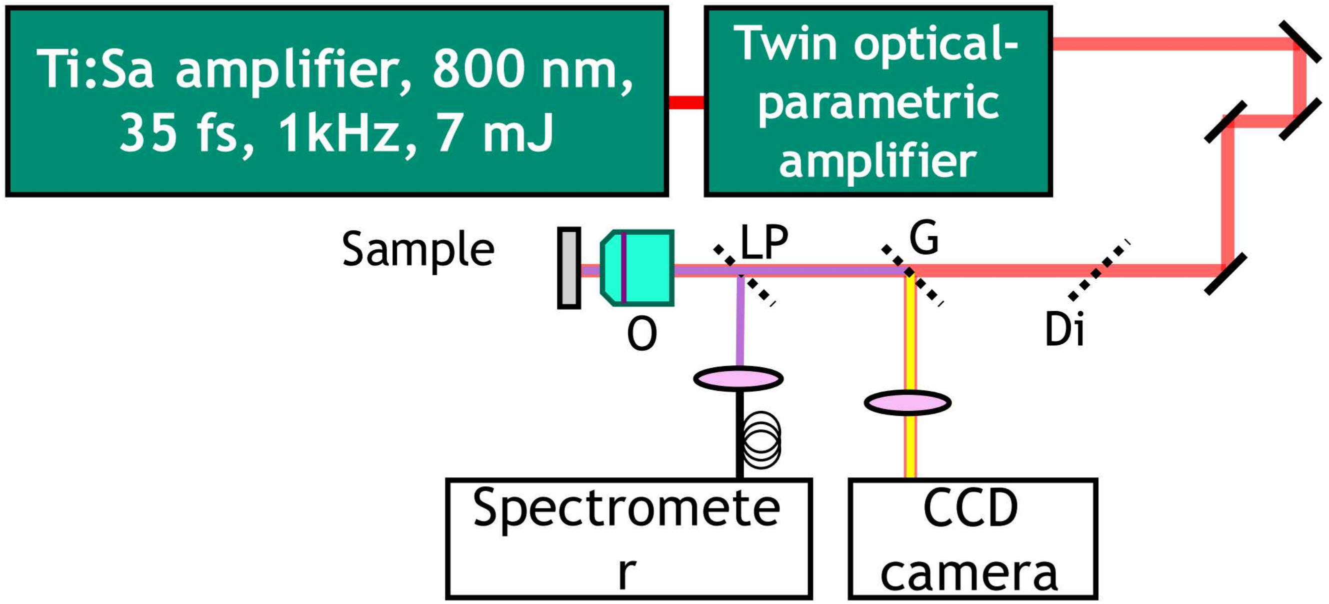
Electric Dipole **Magnetic Dipole**



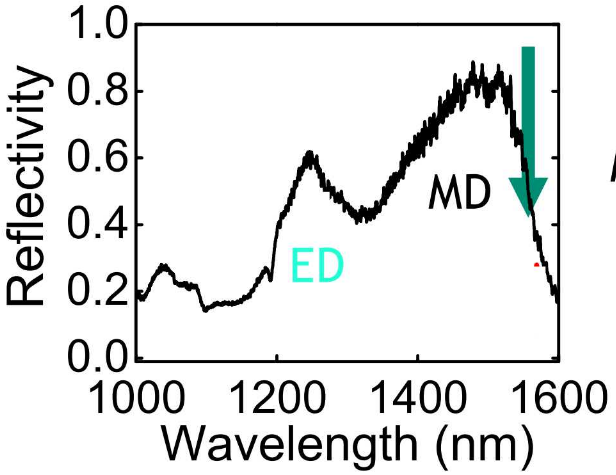
Images: A. Miroshnichenko

- **High nonlinear coefficient material**
- **Resonance enhancement of EM field**

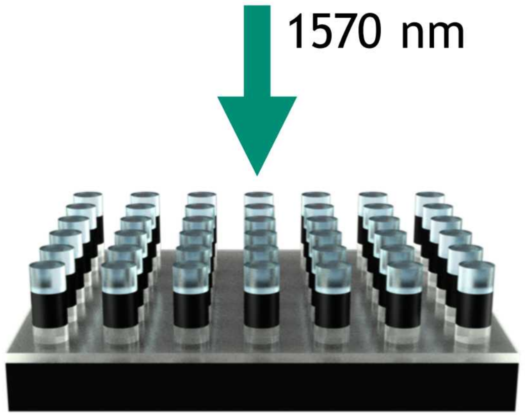
Single beam experiment



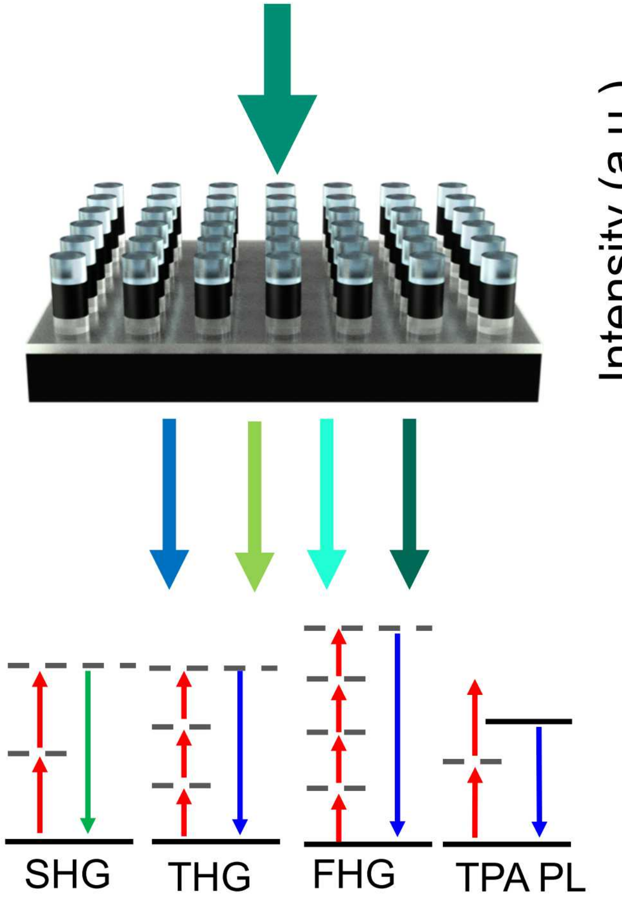
50 fs
1 kHz
0.4 NA objective

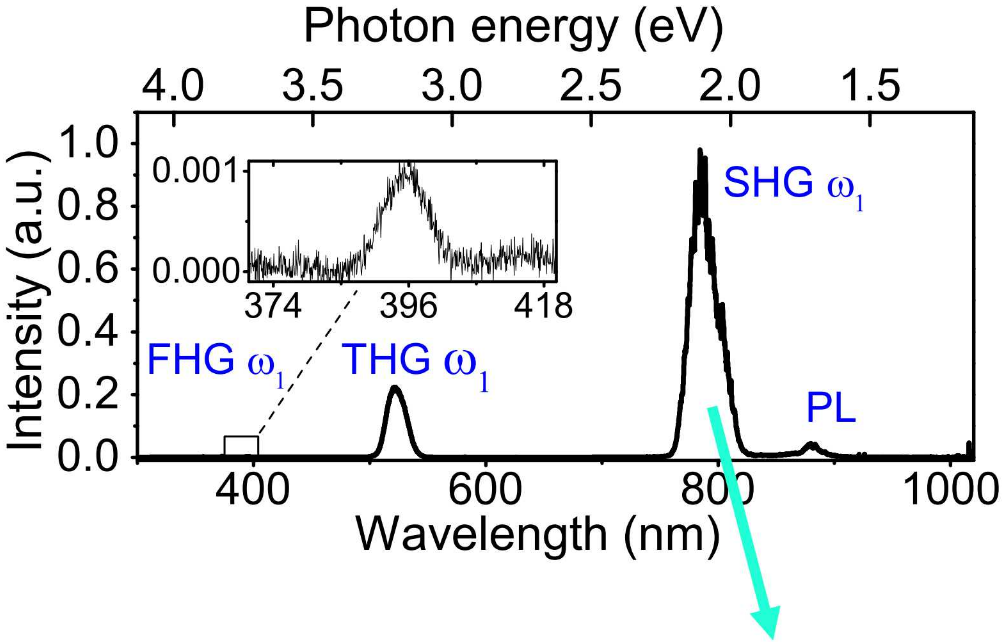


Magnetic dipole resonance

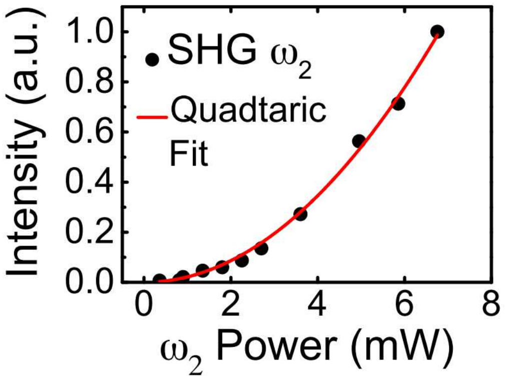


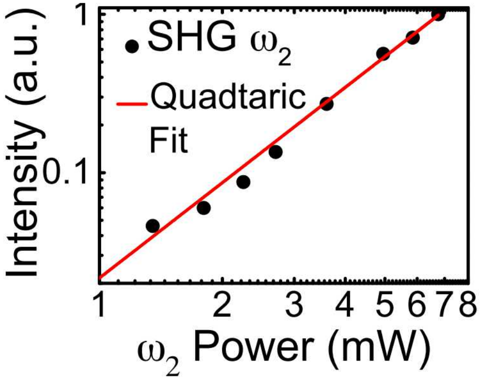
2nd, 3rd, 4th harmonic generation spectra



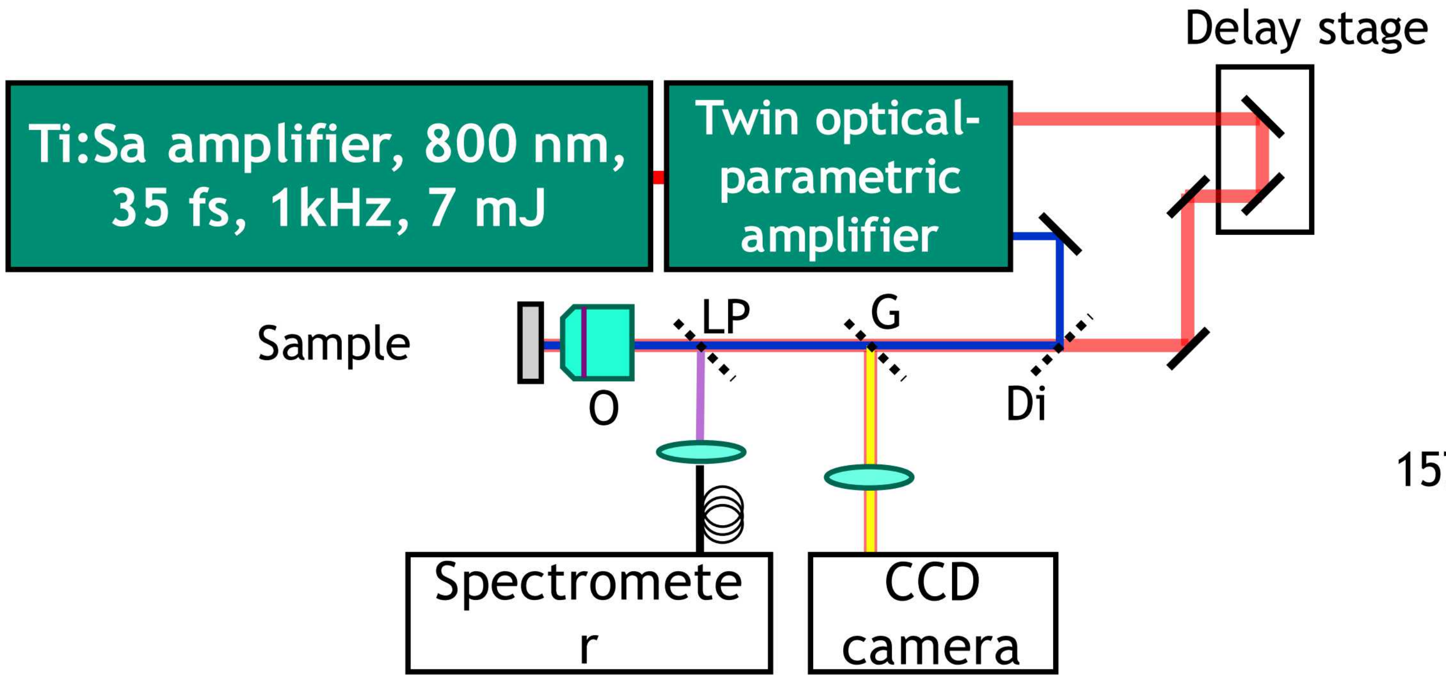


SHG power dependence

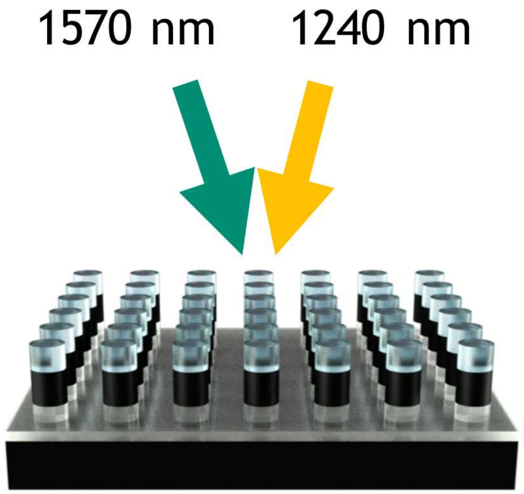


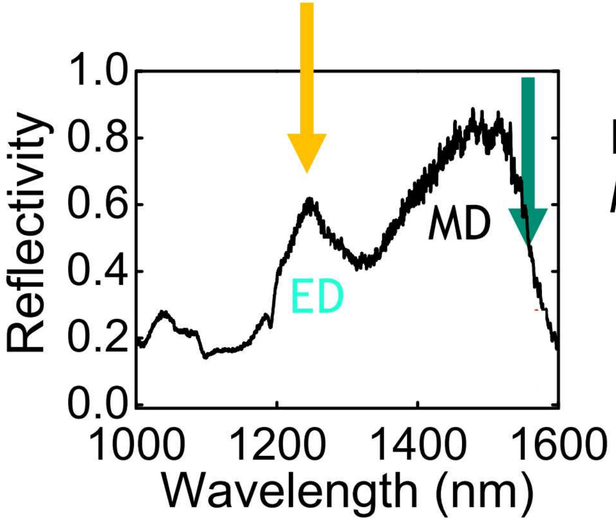


Two-beam experiment



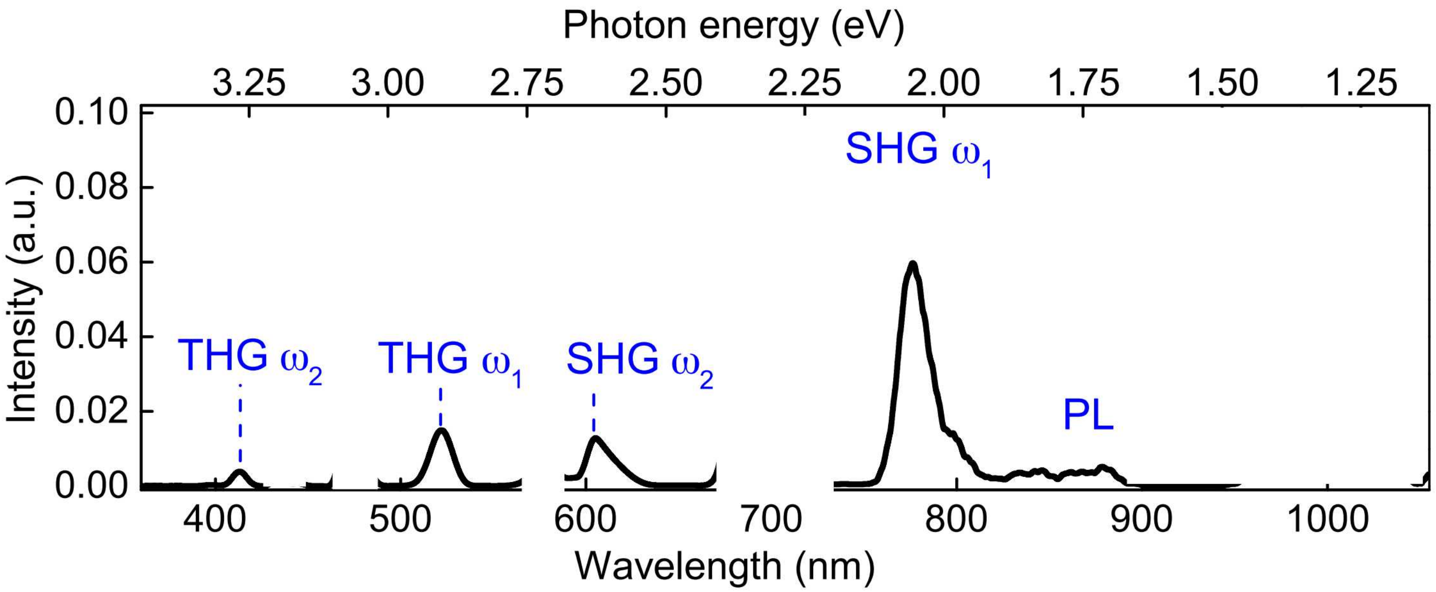
50 fs
1 kHz
0.4 NA objective

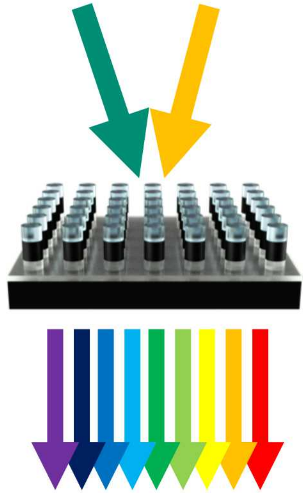


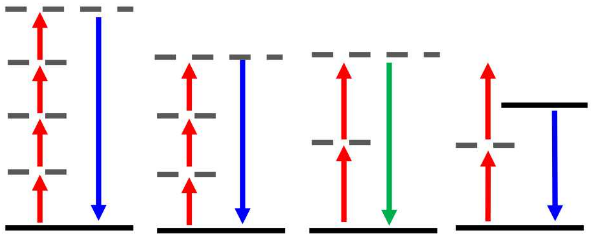


Electric dipole resonance
Magnetic dipole resonance

Frequency mixing spectra

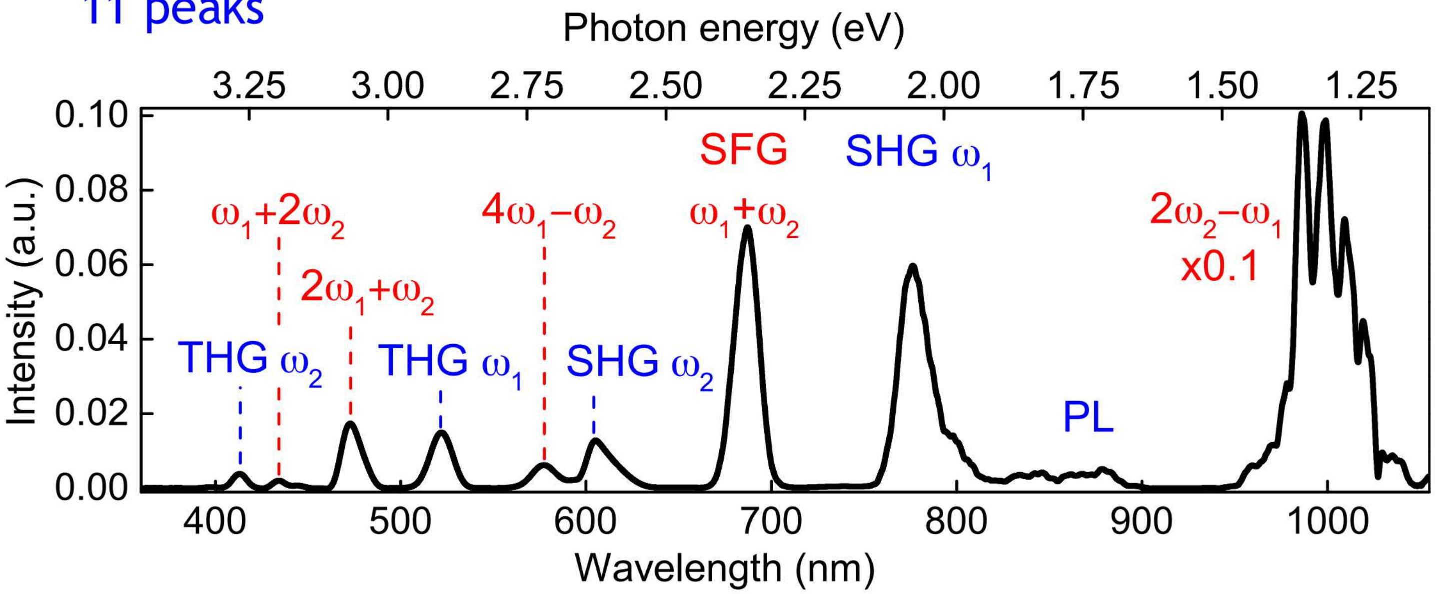


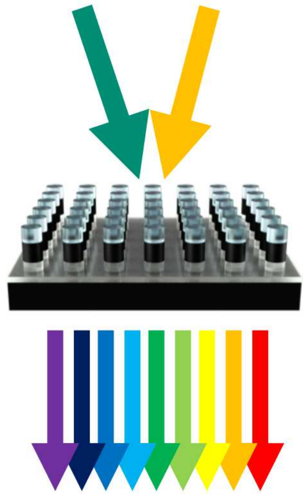


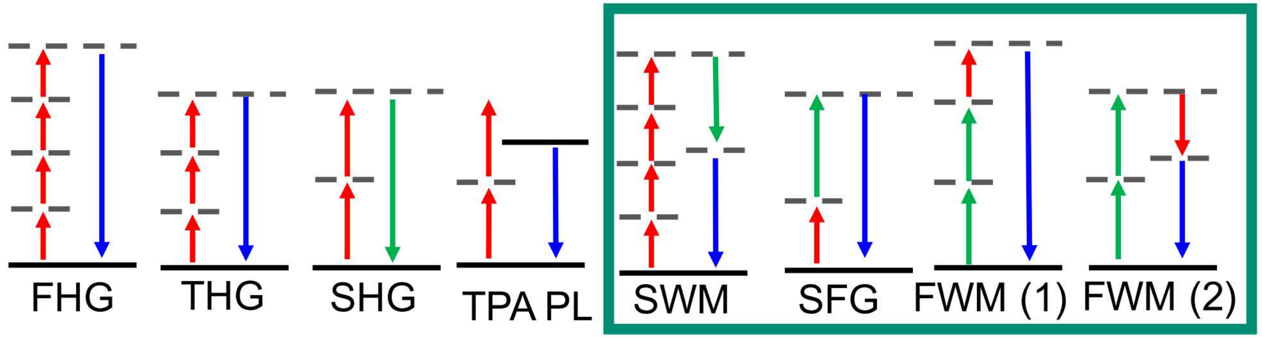


Frequency mixing spectra

11 peaks



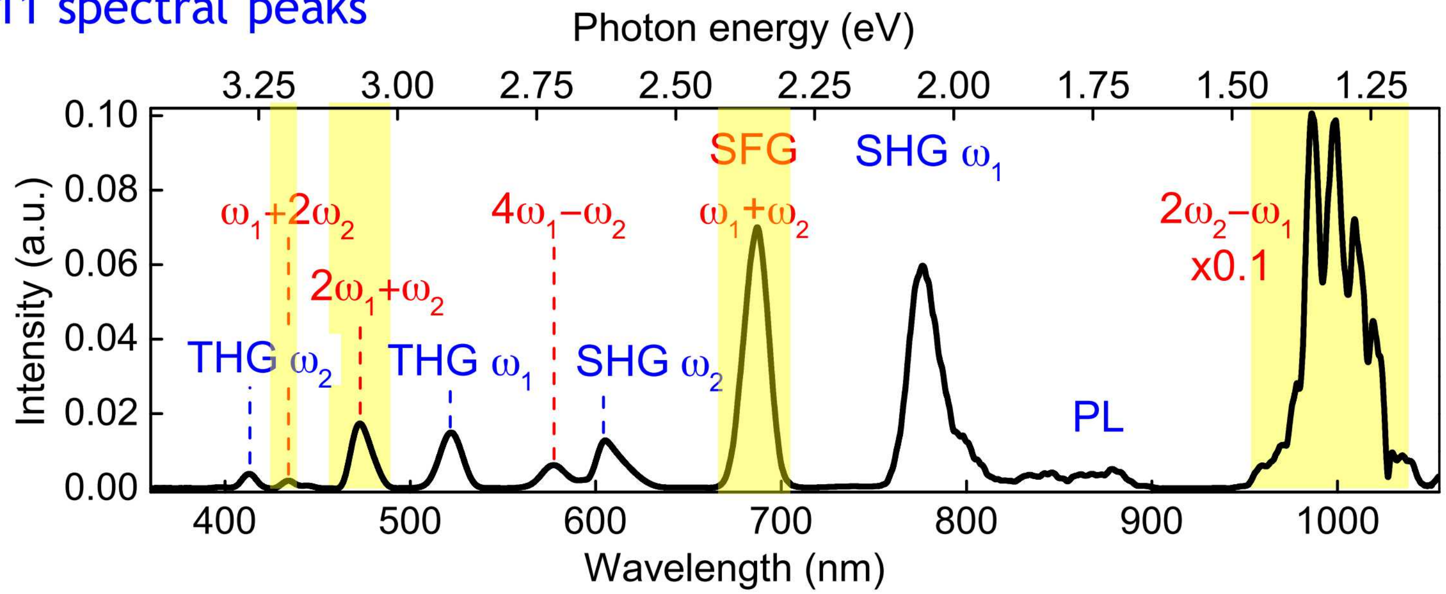


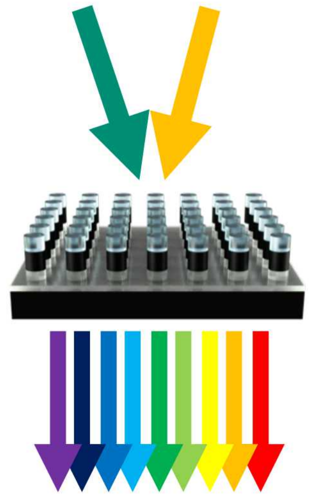


7 different nonlinear processes: 2nd, 3rd, 4th harmonics, sum frequency generation, 4 wave-mixing, six-wave mixing, PL induced by two-photon absorption

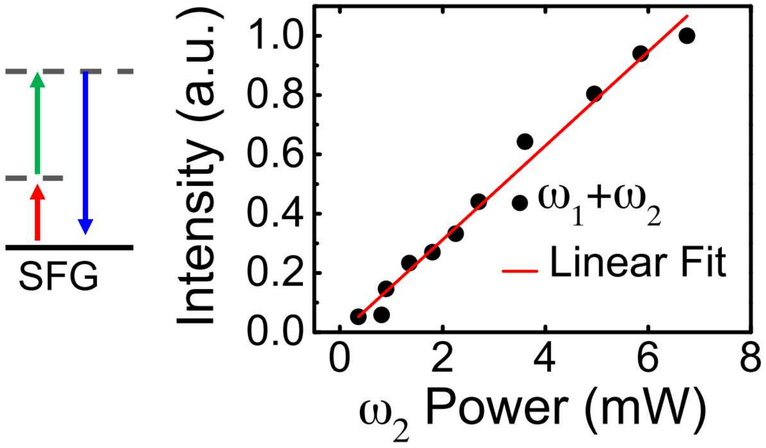
Frequency mixing spectra

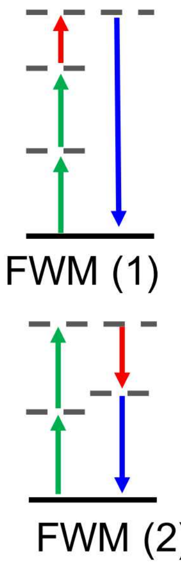
11 spectral peaks



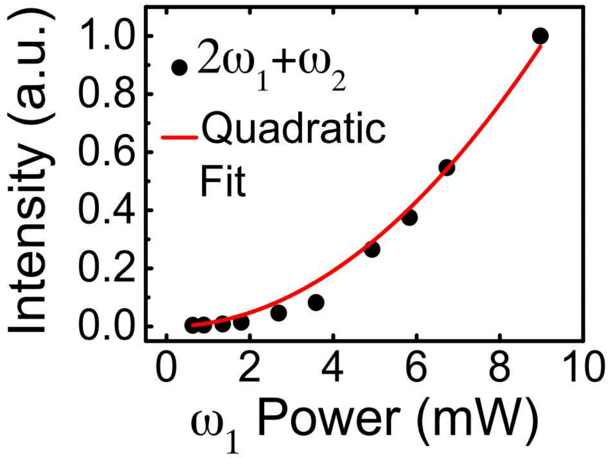


SFG power dependence



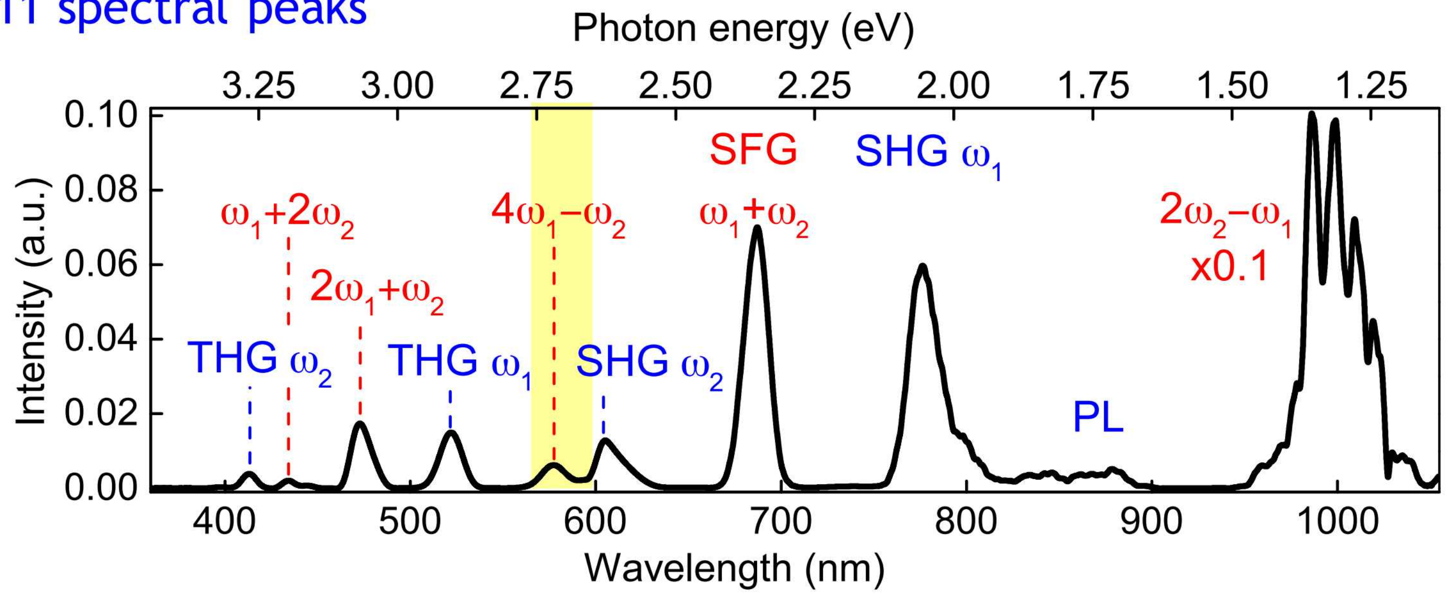


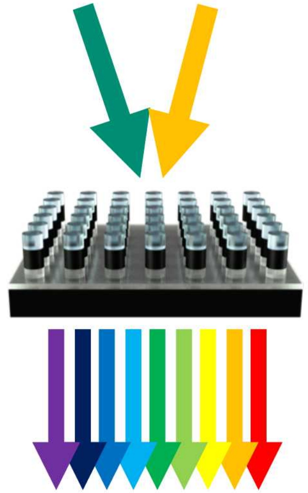
FWM power dependence



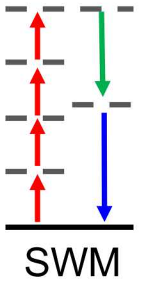
Frequency mixing spectra: six wave mixing

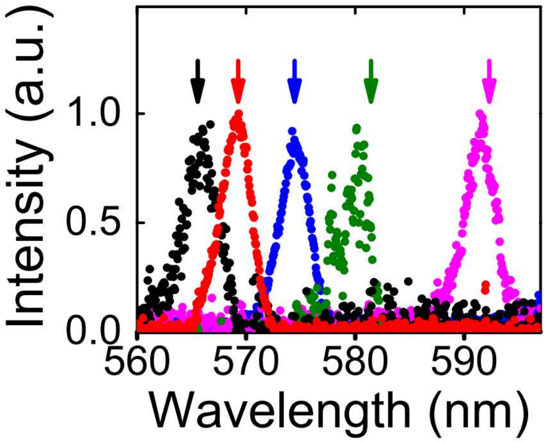
11 spectral peaks



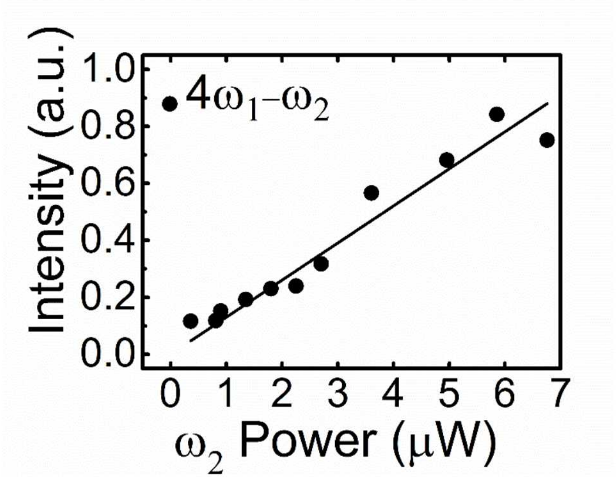


Wavelength shift

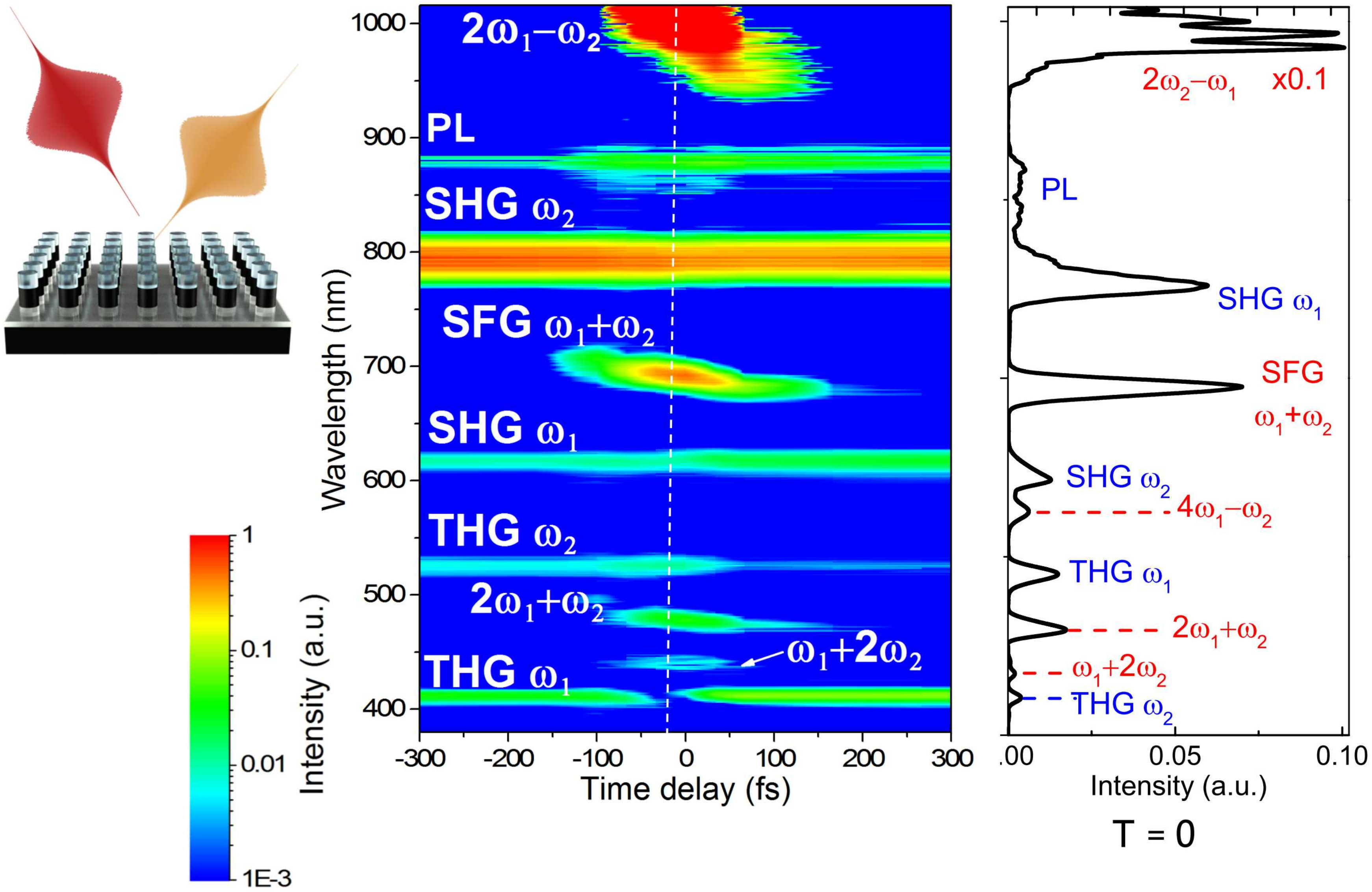




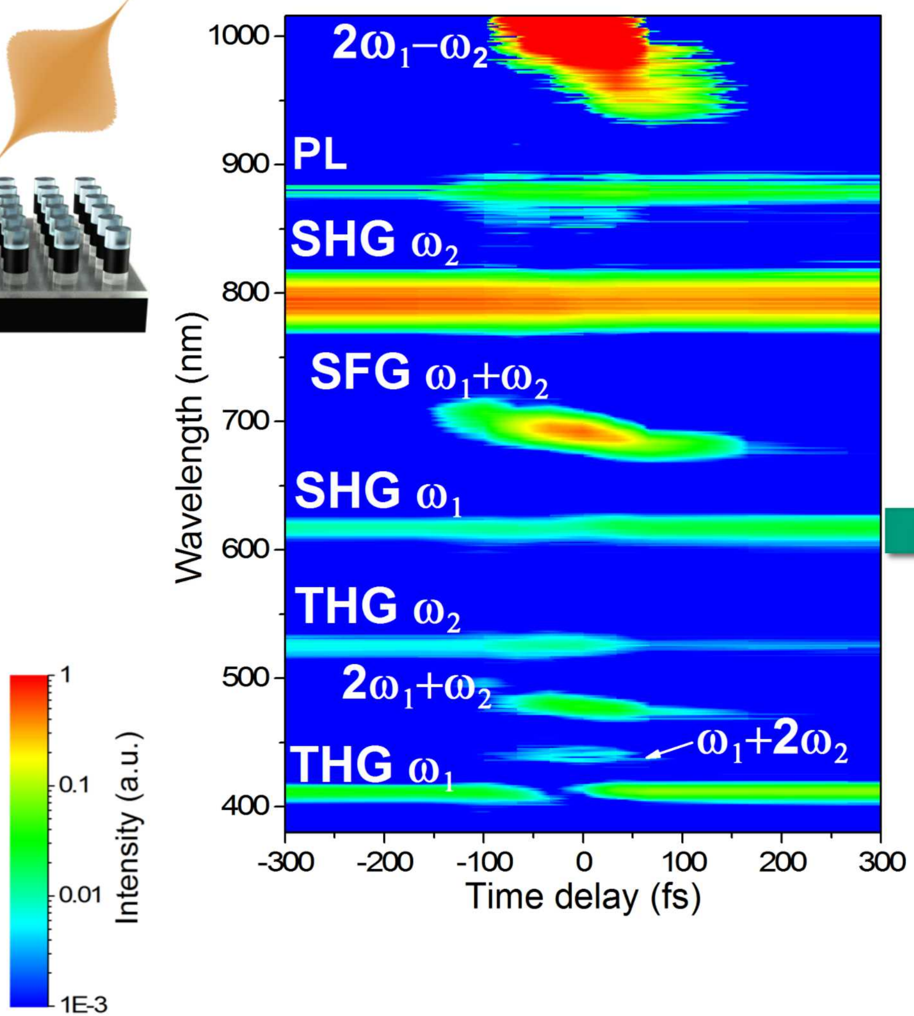
SWM power dependence

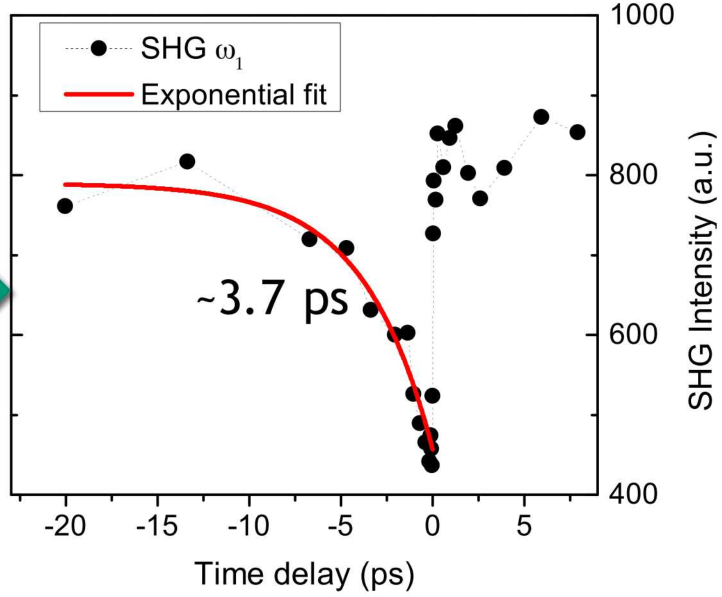


Temporal dynamics of frequency mixing



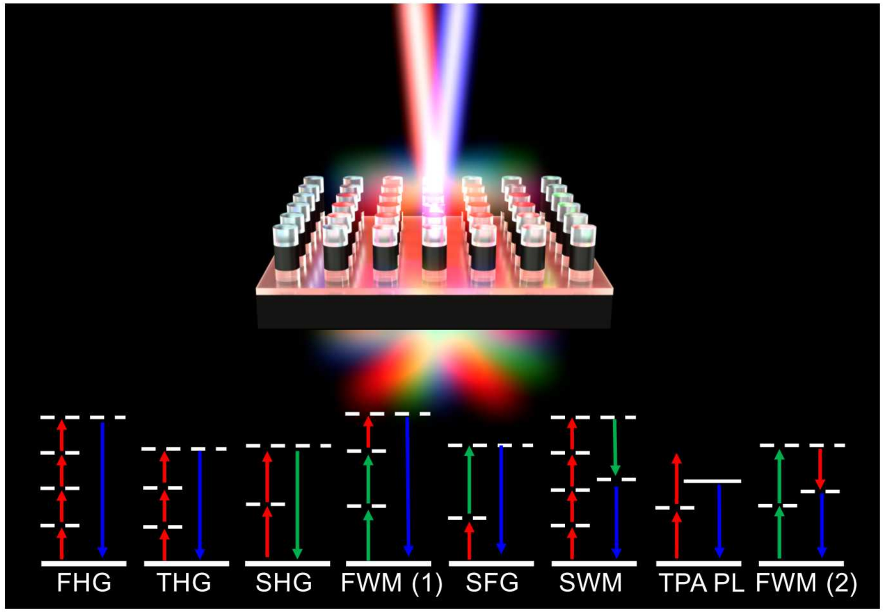
Temporal dynamics of frequency mixing





Shcherbakov, Maxim R., et al.
Nature communications 8.1 (2017): 17

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

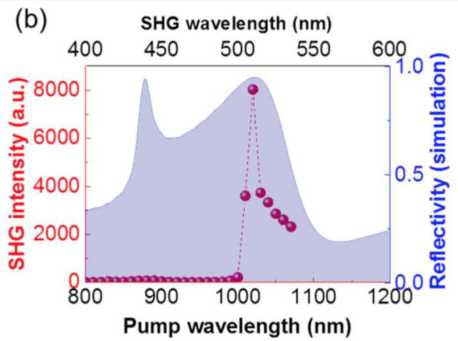


- The even and odd order nonlinearities of GaAs enable our observation of second-, third- and fourth-harmonic generation, sum-frequency generation, two-photon absorption induced photoluminescence, four-wave mixing, and six-wave mixing.

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*.

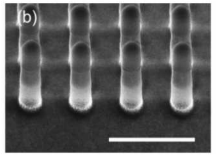
Enhancement of nonlinear processes in metasurfaces

SHG efficiency



Efficiency: power of generated SHG/pump power

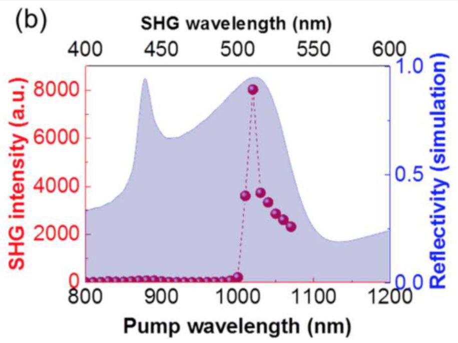
Second Harmonic Generation (10^{-4}-10^{-5})

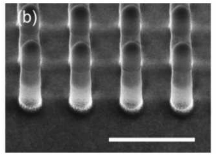


Carletti, L., et al. *Optics express* 23.20, 26544-26550**, 2015**
Camacho-Morales, R., et al. *Nano letters* 16.11, 7191-7197, **2016**
Kruk, S.et al. *Nano Lett.*, *17* (6), pp 3914–3918, **2017**
Liu, S. et. al. *Nano letters* 16.9, 5426-5432, **2016**

Enhancement of nonlinear processes in metasurfaces

SHG efficiency





Efficiency: power of generated SHG/pump power

Second Harmonic Generation (10^{-4}-10^{-5})

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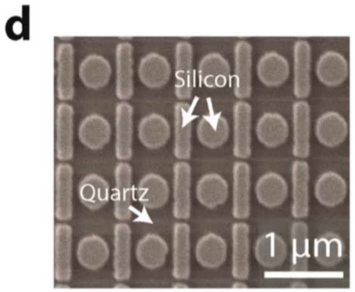
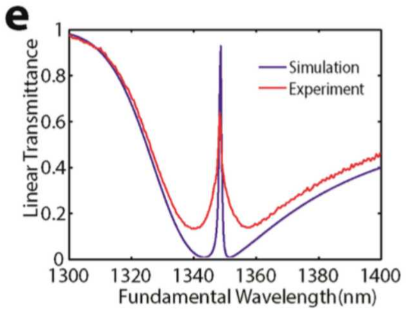
Kruk, S.et al. *Nano Lett.*, *17* (6), pp 3914–3918, **2017**
Liu, S. et. al. *Nano letters* 16.9, 5426-5432, **2016**

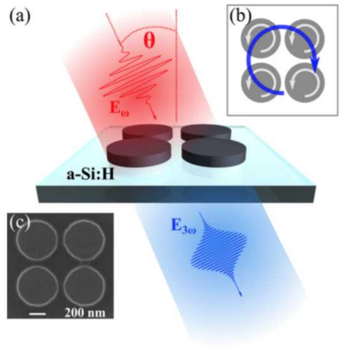
Fano resonances and BIC modes:

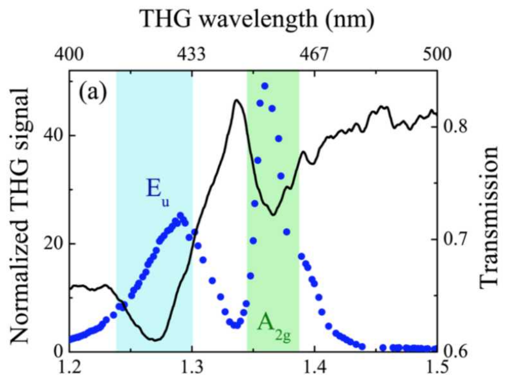
Slit and bar metasruface design





Oligomer metasurfaces



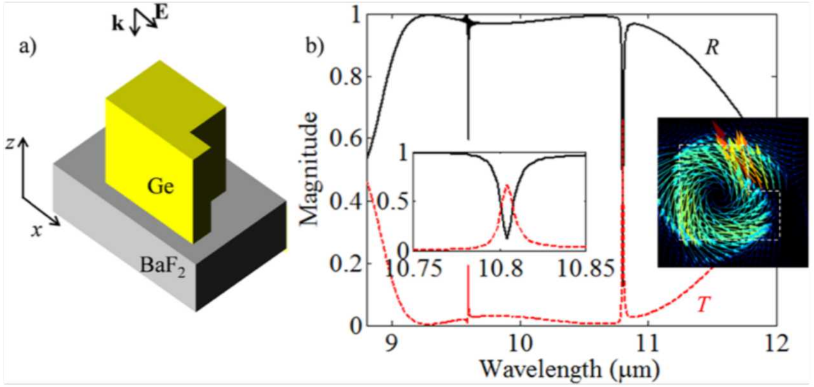


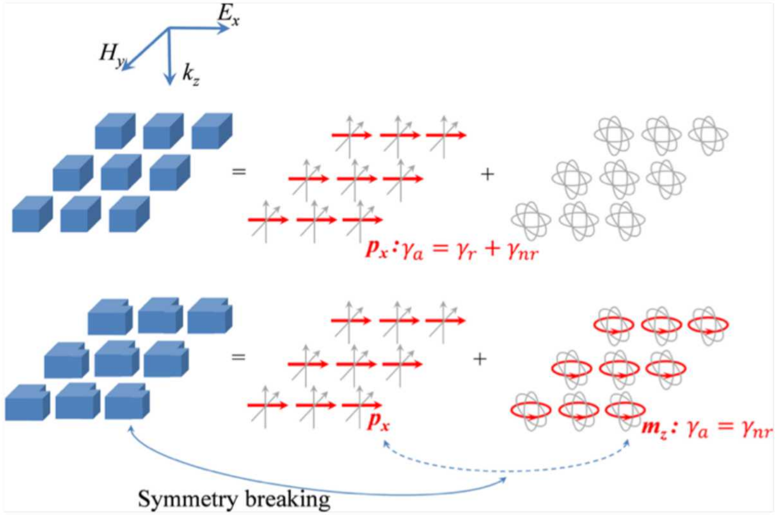
Yang, Y et. al Nano Lett. 2015, 15 (11), 7388−7393.

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

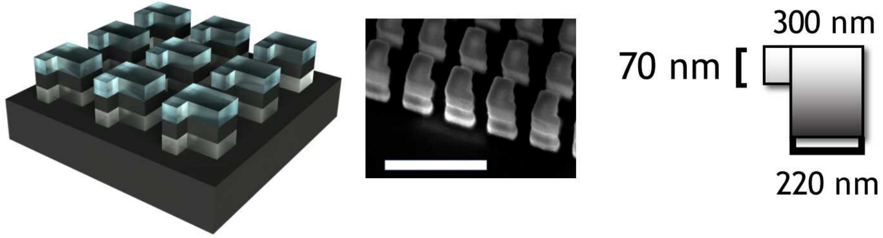
Carletti, Luca, et al. *Physical review letters* 121.3 (2018): 033903.

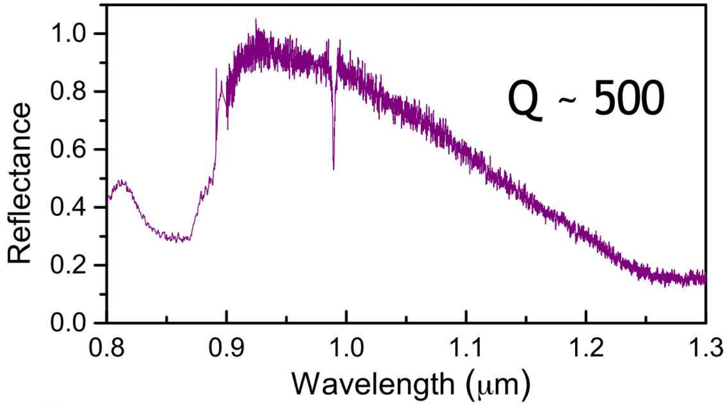
Metasurface with broken symmetry design resonators: High-Q factor resonance



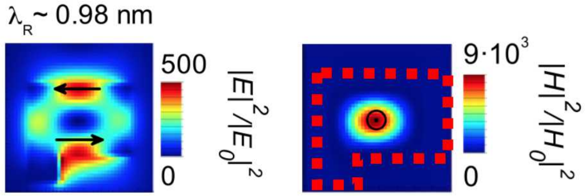


Campione, S. et.al ACS Photonics 3 (12), 2362−2367, **2016**





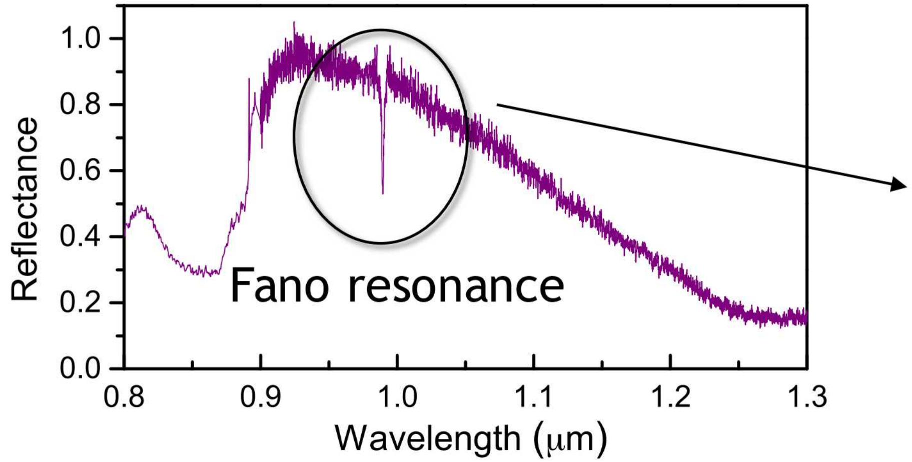
Fano resonance

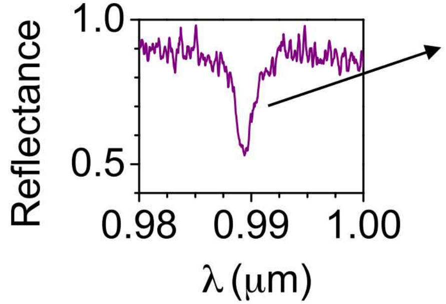


Vabishchevich, P., et al. *ACS Photonics*, *5* (5), pp 1685-1690, **2018**

Fano metasurface reflectance spectrum

Experimental measurements

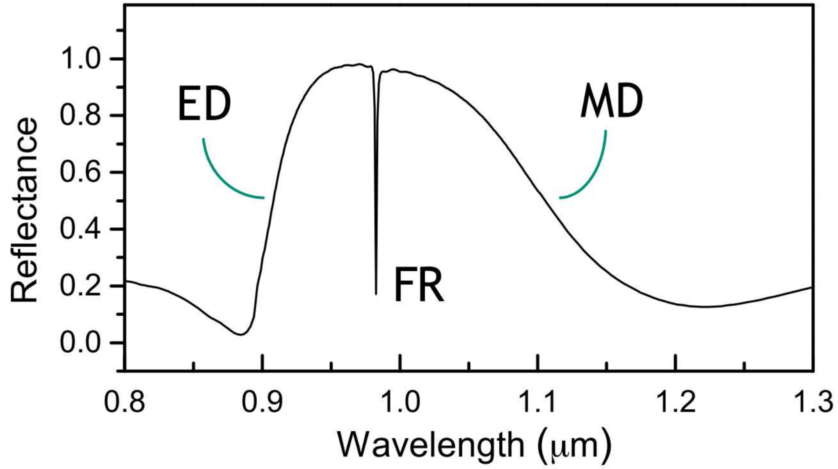




Δλ ~ 2 nm

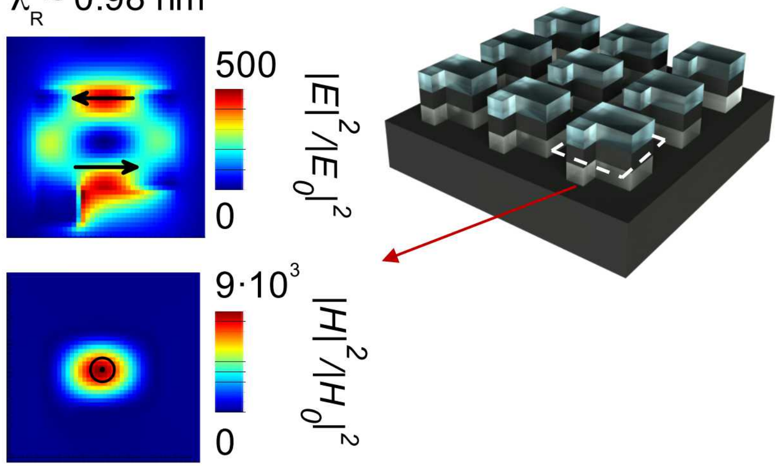
Q factor
$\lambda_F/\Delta\lambda$ ~ 500

FDTD simulations

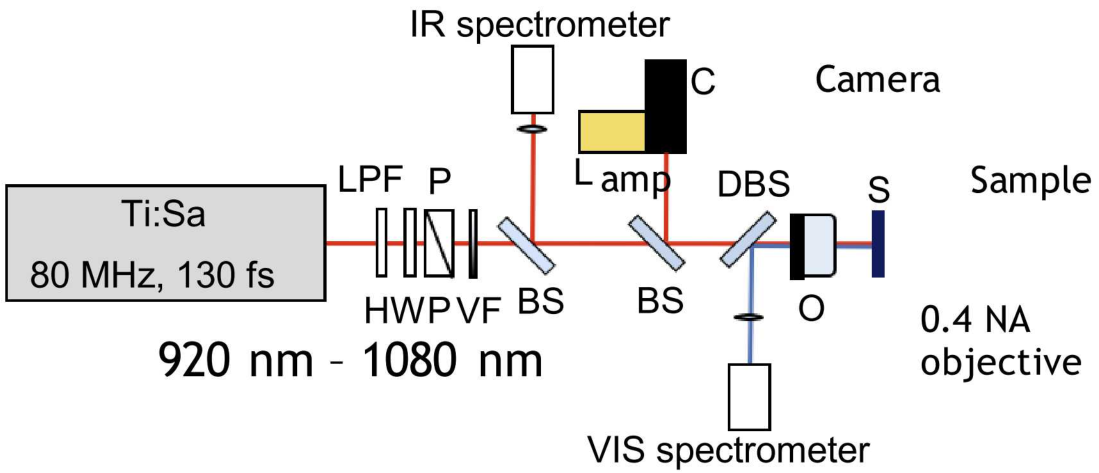


Fano resonance

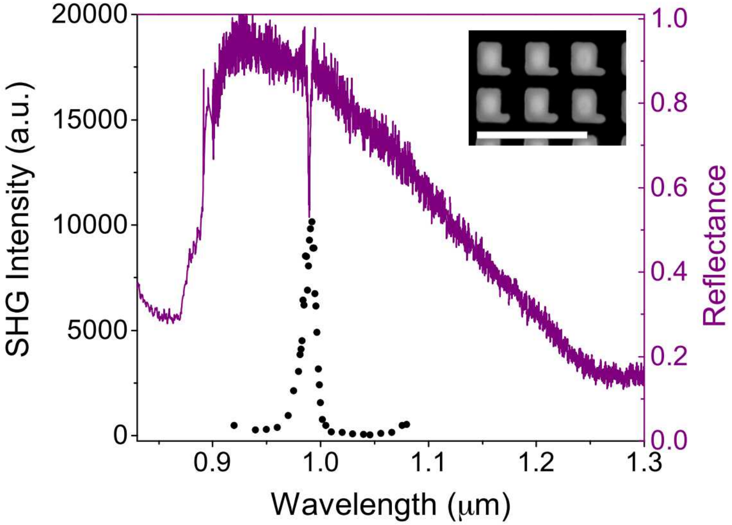
λ_R~ 0.98 nm

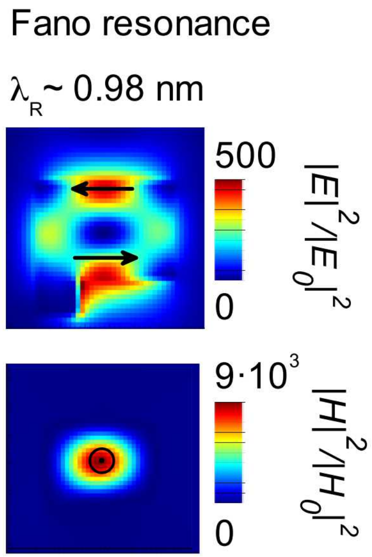


SHG in Fano metasurface



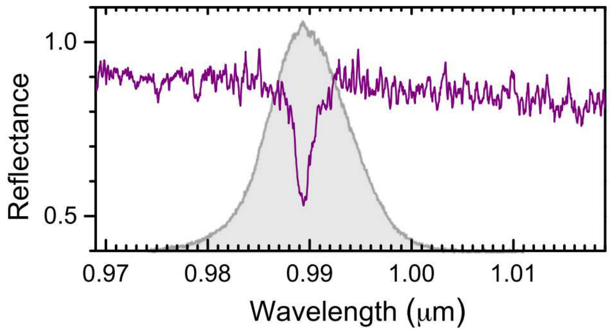
SHG intensity enhancement in the vicinity of the Fano resonance x300



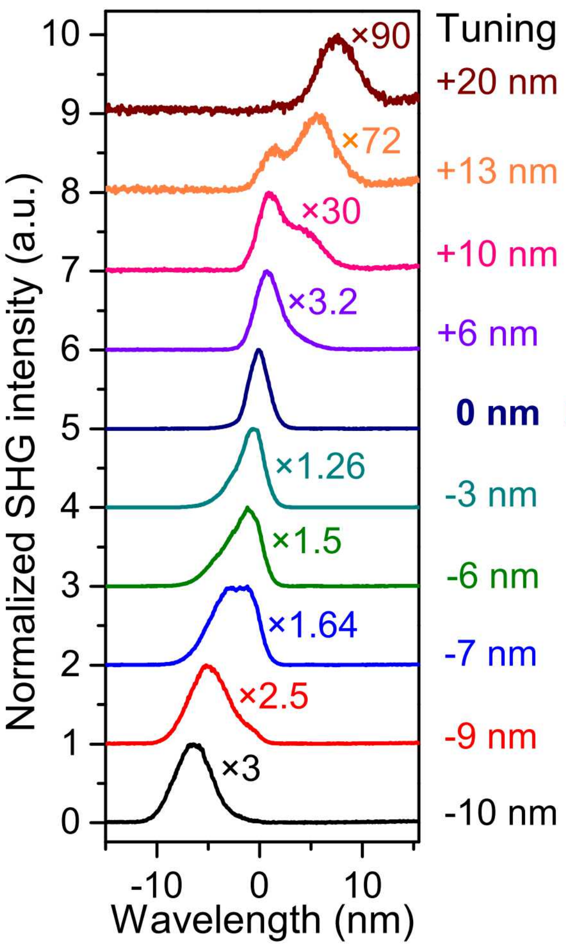


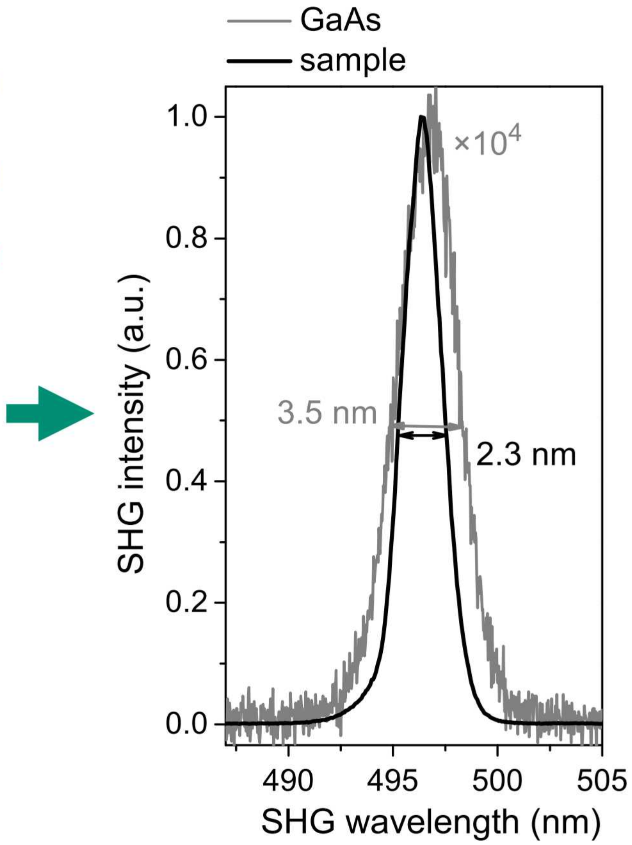
SHG spectra modification

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

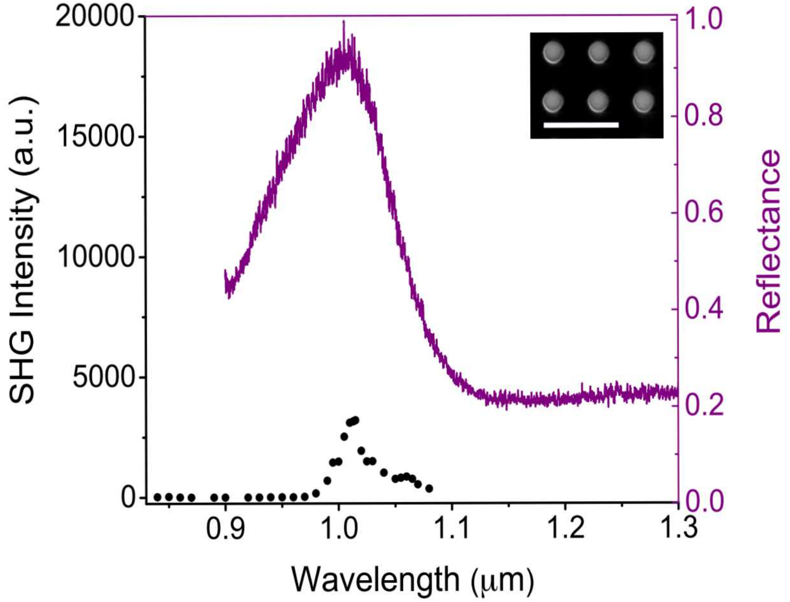


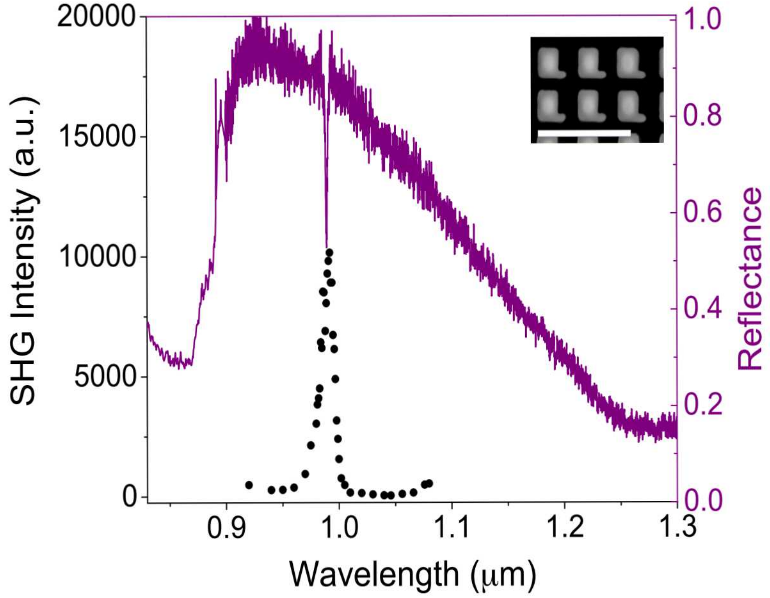
2 nm vs 11 nm

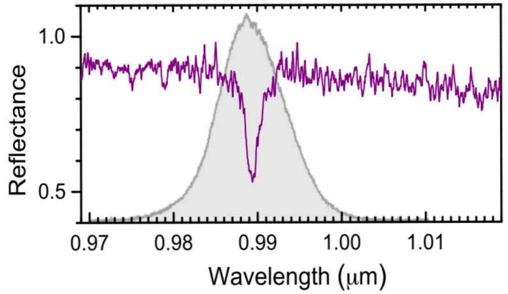




Nanodisk metasurface vs Fano metasurface



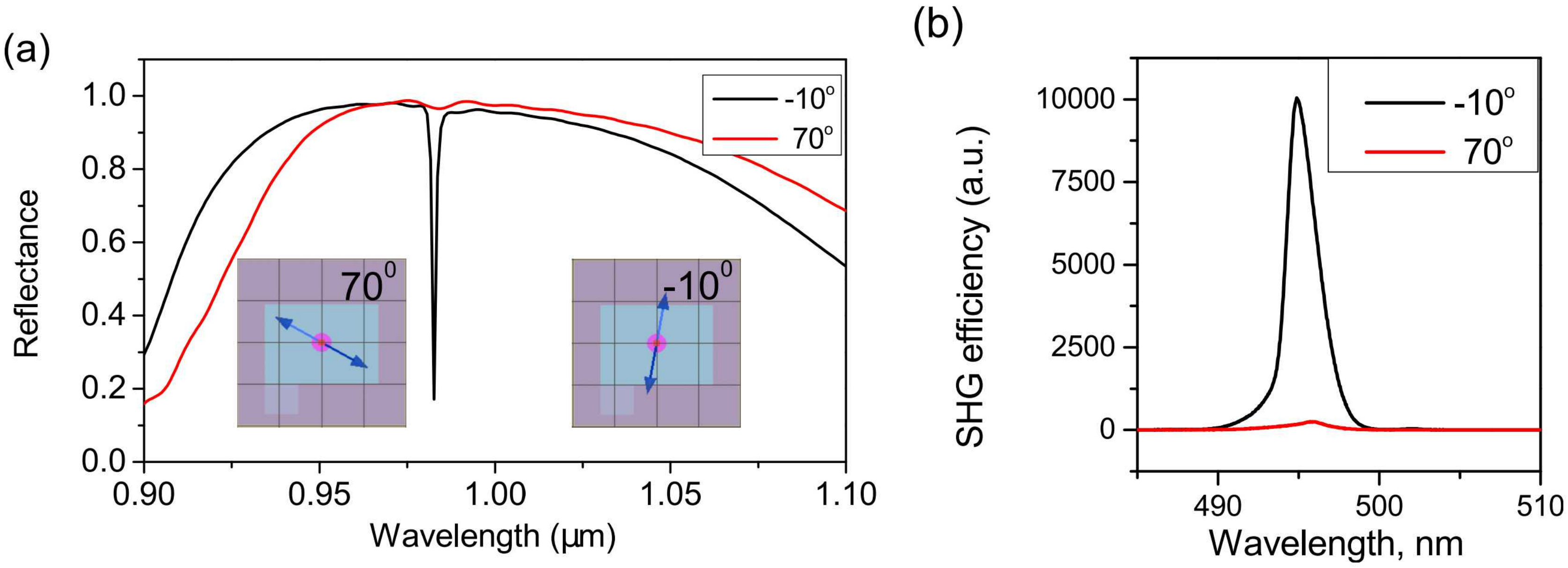


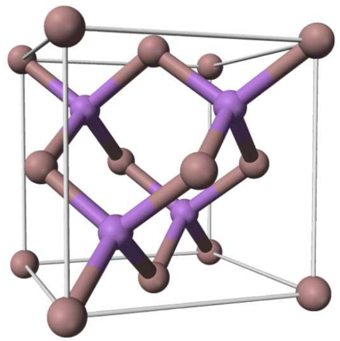


~30% of pump power is used

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

Polarization dependence of SHG

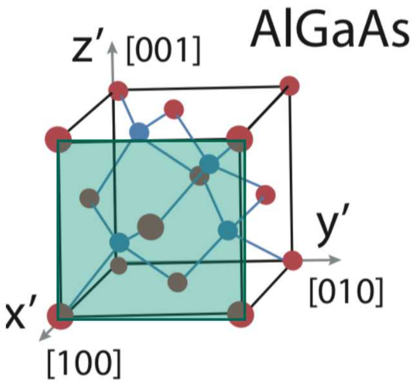




Match the asymmetric shape of the sample with the crystalline axis of GaAs

https://www.wikipedia.org

GaAs (100) lattice plane

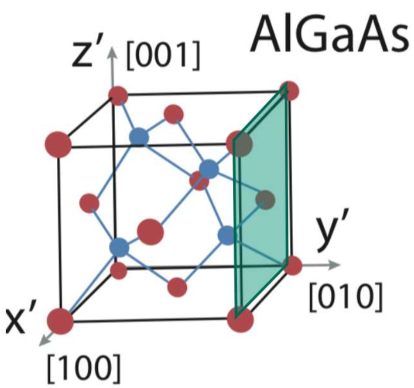


$$\begin{pmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{pmatrix} = 2\varepsilon_0 d_{eff} \begin{pmatrix} E_x(\omega)^2 \\ E_y(\omega)^2 \\ E_z(\omega)^2 \\ 2E_y(\omega)\cdot E_z(\omega) \\ 2E_x(\omega)\cdot E_z(\omega) \\ 2E_y(\omega)\cdot E_x(\omega) \end{pmatrix}$$

$$d_{eff} = \begin{pmatrix} 0 & 0 & 0 & d_{41} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{41} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{41} \end{pmatrix}$$

Absence of SHG in normal direction

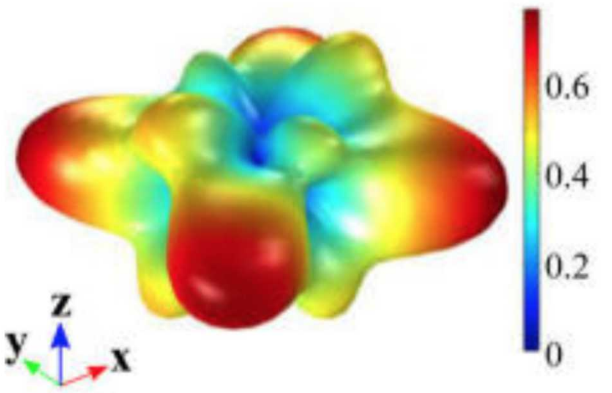
GaAs (100) single disk and metasurface emission patterns

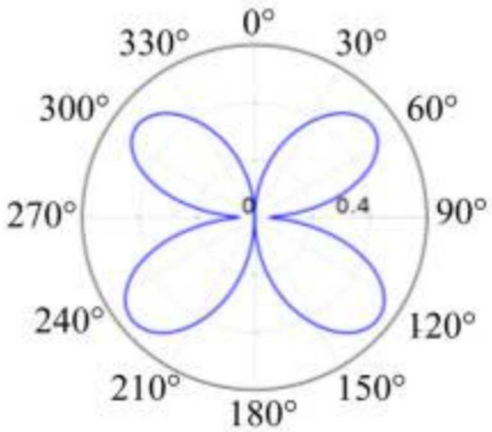


$$d_{eff} = \begin{pmatrix} 0 & 0 & 0 & d_{41} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{41} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{41} \end{pmatrix}$$

Absence of SHG in normal direction

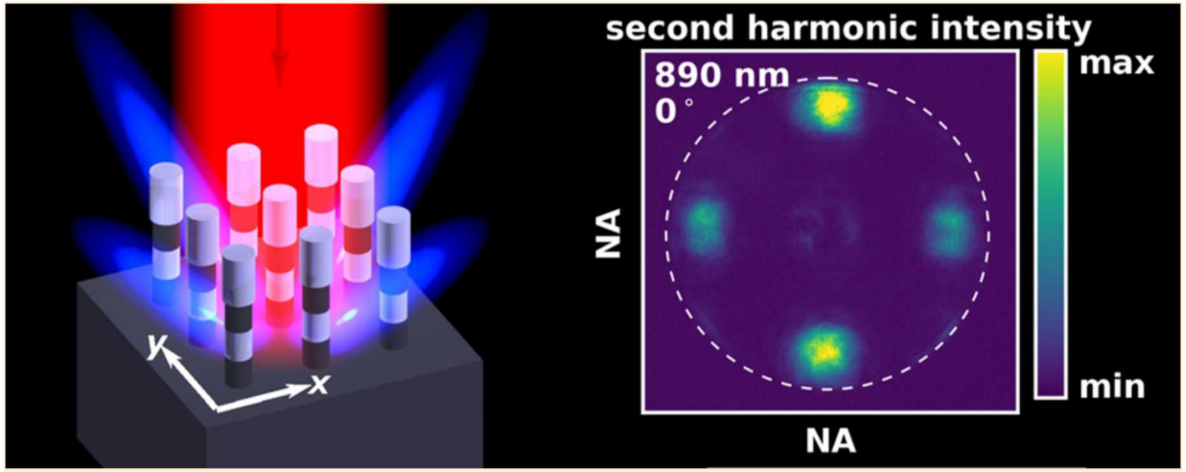
SHG radiation pattern in AlGaAs (100) nanodisks at MD resonance





L. Carletti et al., *ACS Photonics*, *3*(8), 1500-1507, **2016**

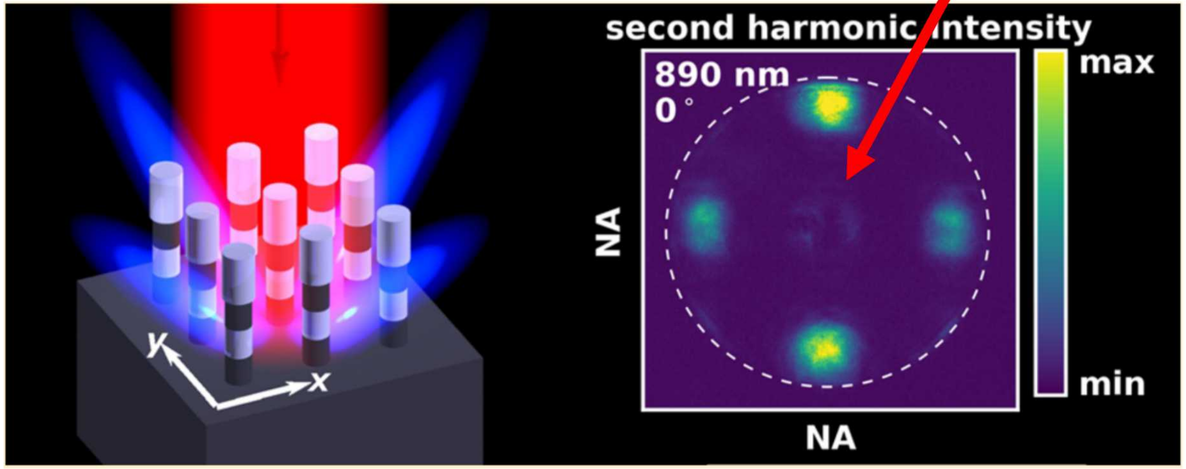
SHG diffraction in the GaAs (100) metasurface



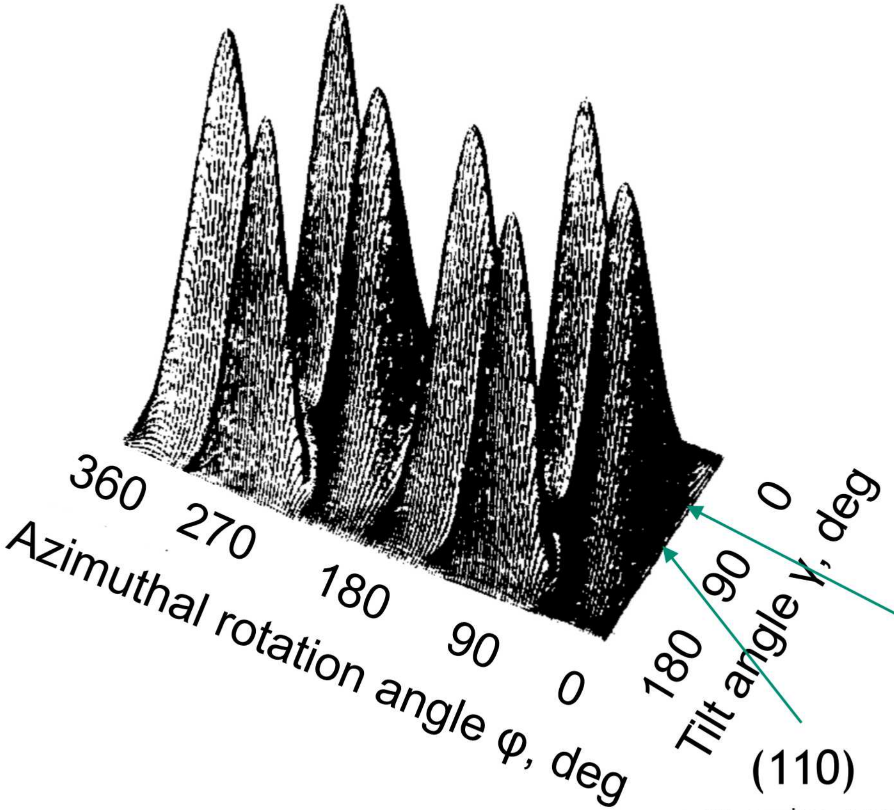
Lŏchner, F. J., *ACS Photonics*, *5*(5), 1786-1793, **2018**

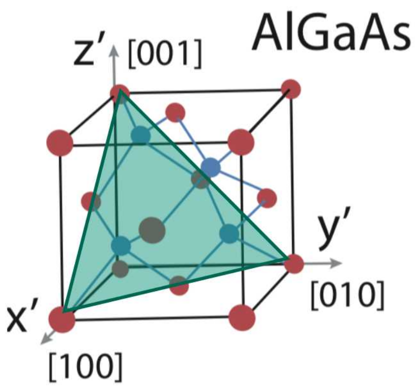
$\boxed{\textbf{Absence of the zeroth-order}}$

Goal: Redirection of the SHG emission to zeroth diffraction order in GaAs metasurfaces



SHG in GaAs for different orientations of the crystal, (111), (110)





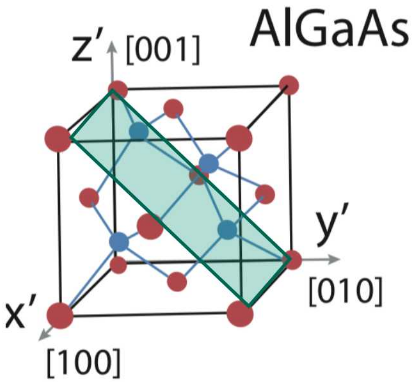
$$d_{eff} = \begin{pmatrix} -\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 \\ 0 & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 & -\frac{1}{\sqrt{6}} \\ -\frac{1}{2\sqrt{3}} & -\frac{1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} & 0 & 0 & 0 \end{pmatrix} \cdot d_{41}$$

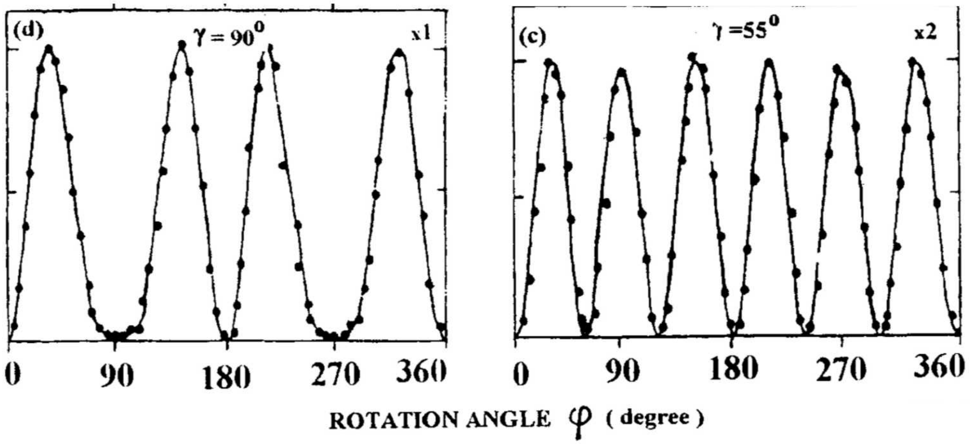
Non-zero SHG signal for under normal incidence

$$P_x(2w) = 2\varepsilon_0 d_{eff}(-\frac{1}{\sqrt{6}})E_x(w)^2 + \frac{1}{\sqrt{6}}E_y(w)^2 - \frac{1}{\sqrt{3}}E_x(w)E_z(w)$$

$$P_y(2w) = 2\varepsilon_0 d_{eff}(-\frac{2}{\sqrt{3}})E_y(w)E_z(w) - \frac{1}{\sqrt{6}}E_x(w)E_z(w)$$

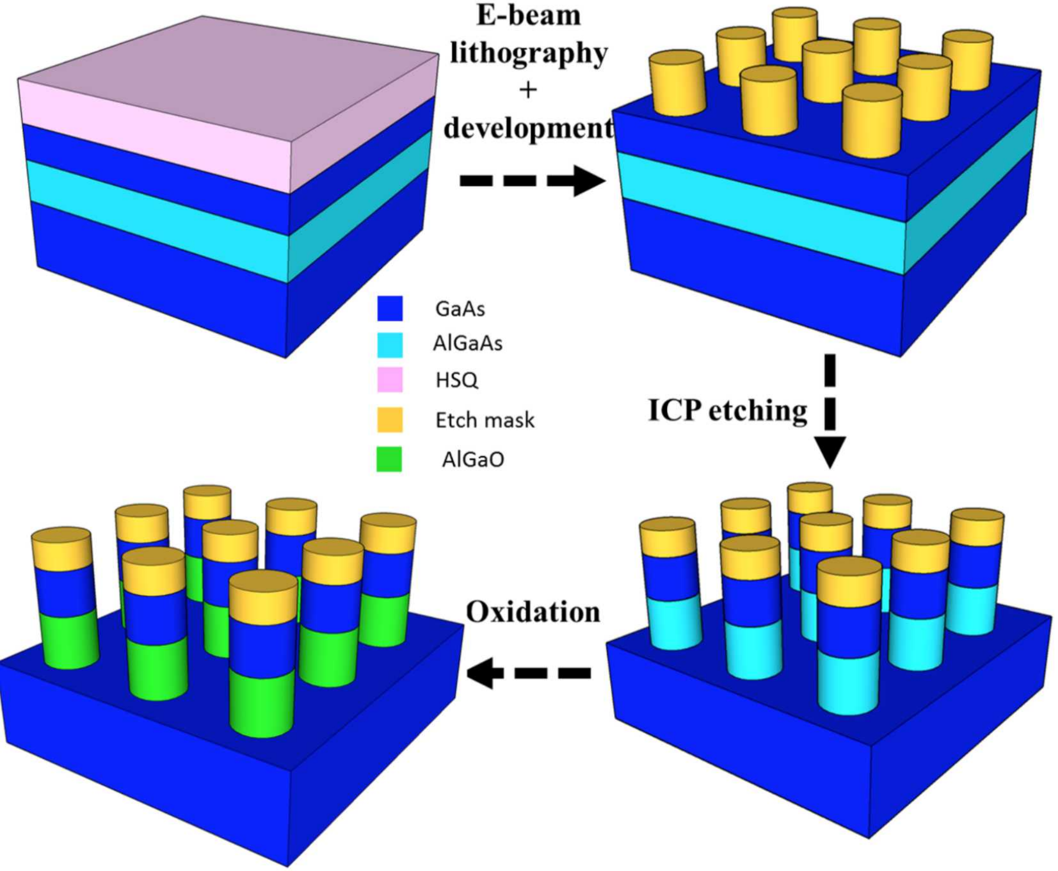
$$P_z(2w) = 2\varepsilon_0 d_{eff}(-\frac{1}{2\sqrt{3}})E_x(w)^2 - \frac{1}{2\sqrt{3}}E_y(w)^2 - \frac{1}{1\sqrt{3}}E_z^2$$





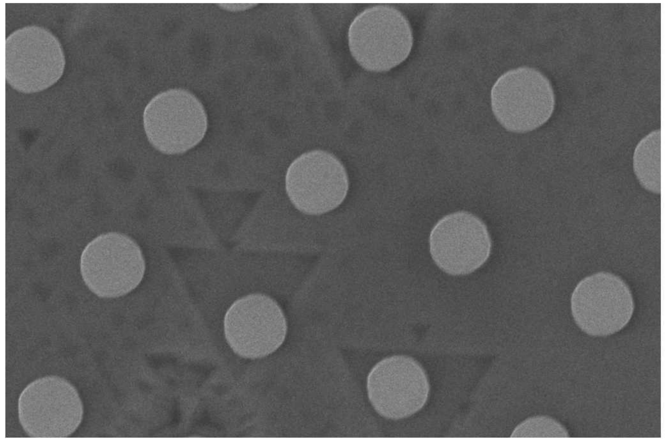
Kulyuk, L L., *Nonlinear Optics of Low-Dimensional Structures and New Materials*. Vol. 2801. International Society for Optics and Photonics, **1996.**

Fabrication of GaAs metasurface

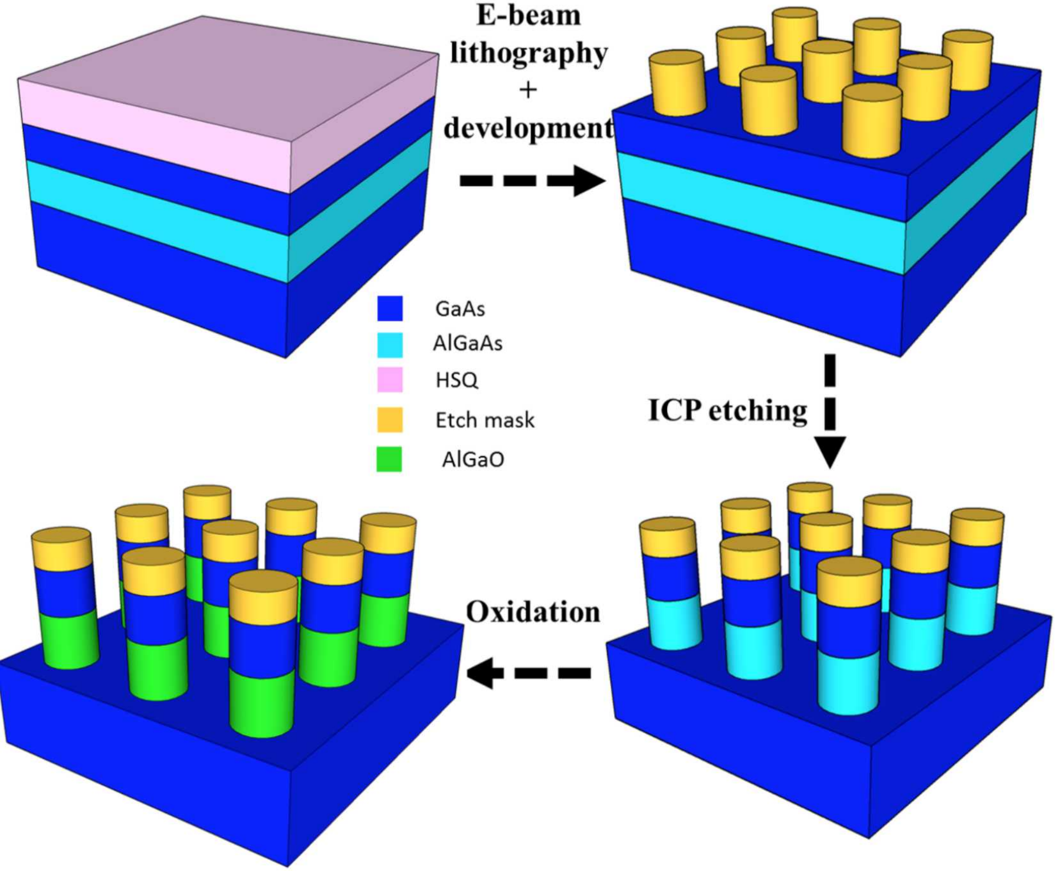


Liu, S. et. al. *Nano letters* 16.9: 5426-5432, **2016**

SiO_x – 400 nm
$Al_{0.1}Ga_{0.9}As$ – 400 nm
$(Al_xGa_{1-x})_2O_3$ – 500 nm
Diameter – 340 nm
Period – 1200 nm



Fabrication of GaAs metasurface



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

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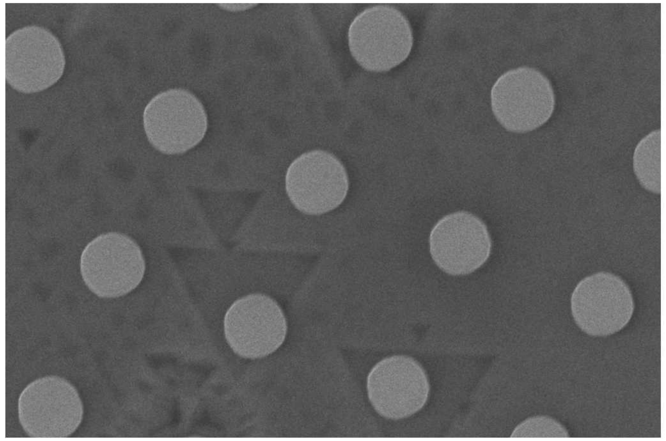
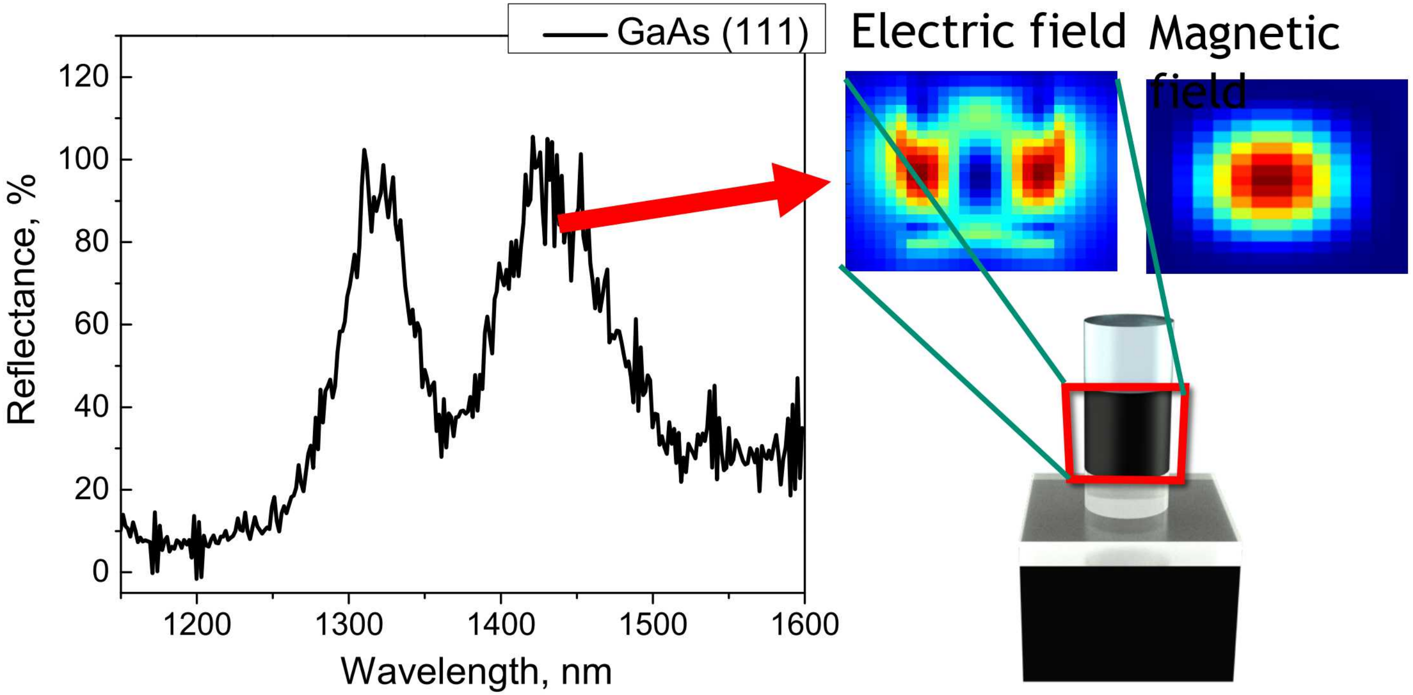
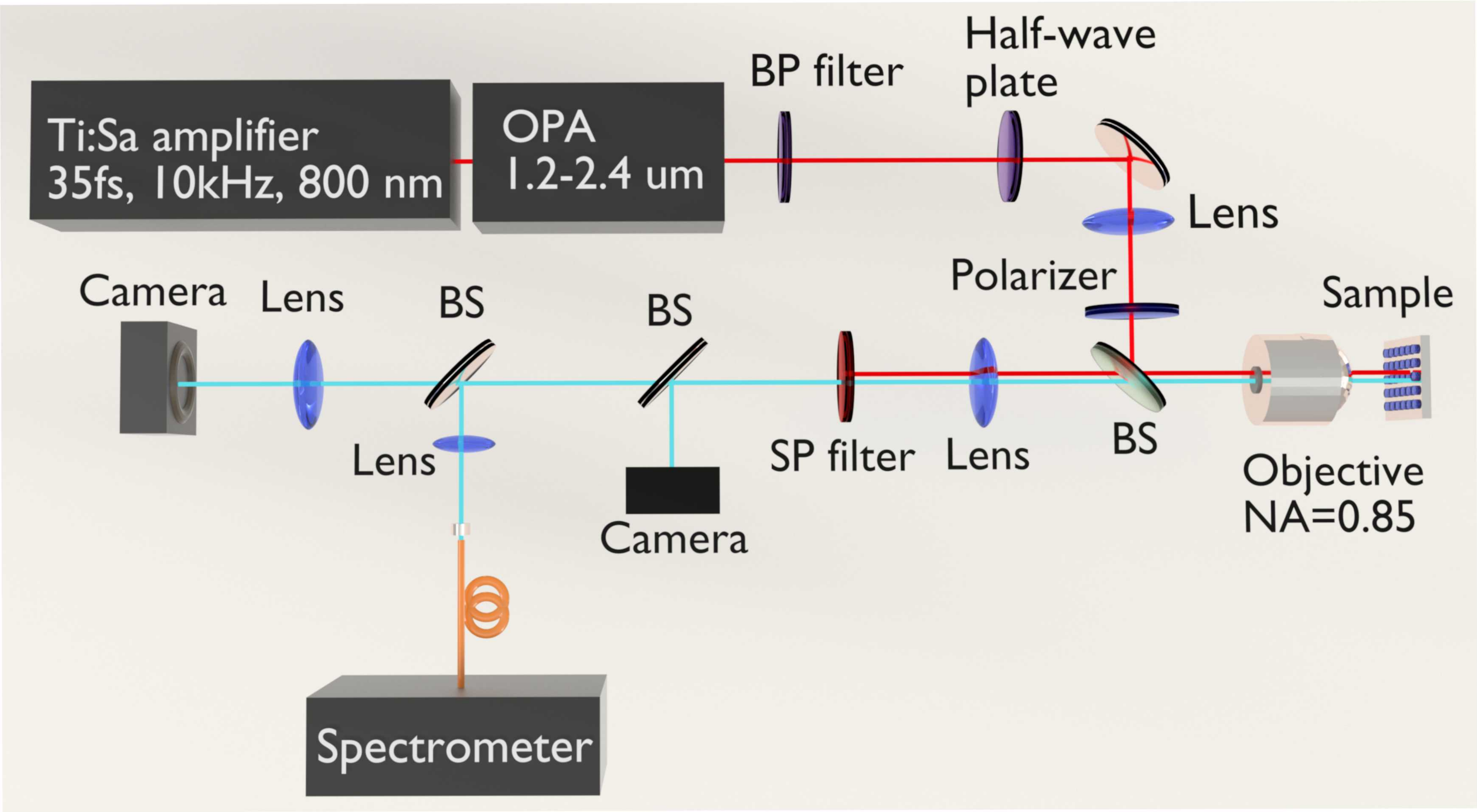


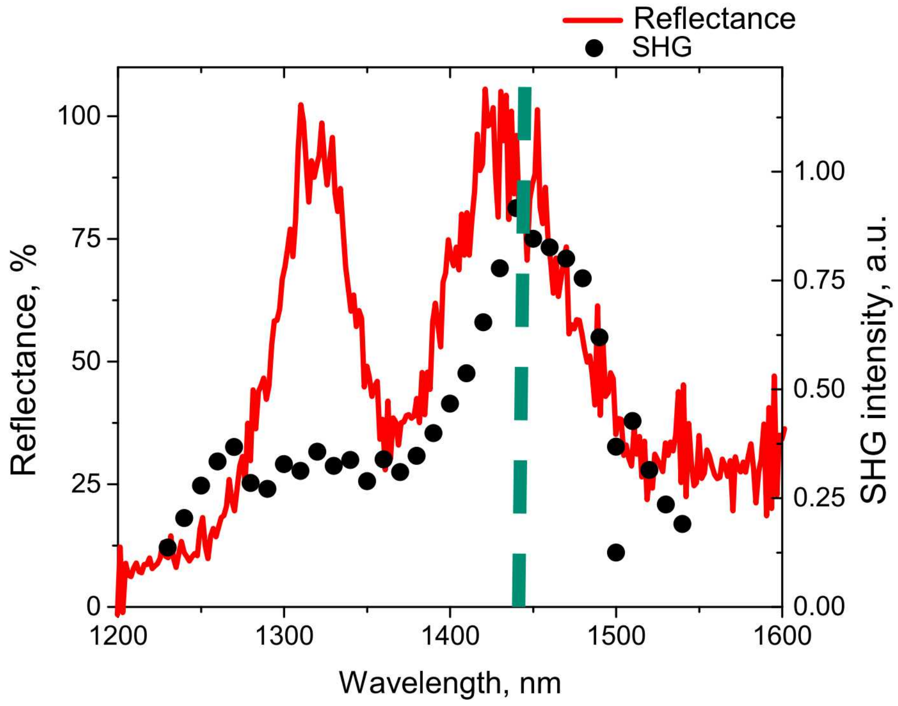
Reflectance spectrum



Experimental setup: Fourier-space imaging

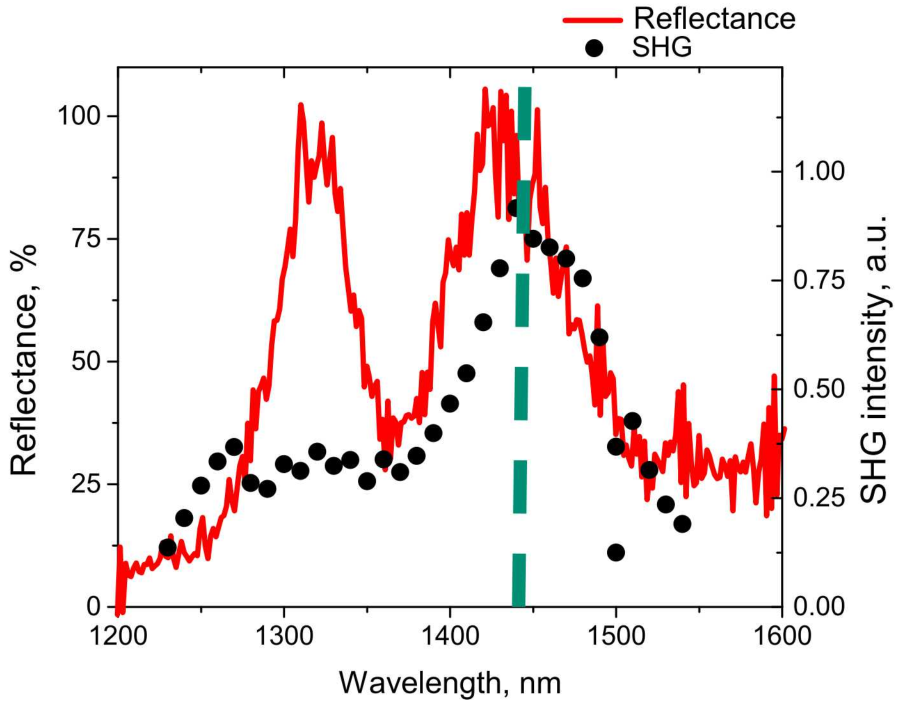


SHG in GaAs (111) metasurface

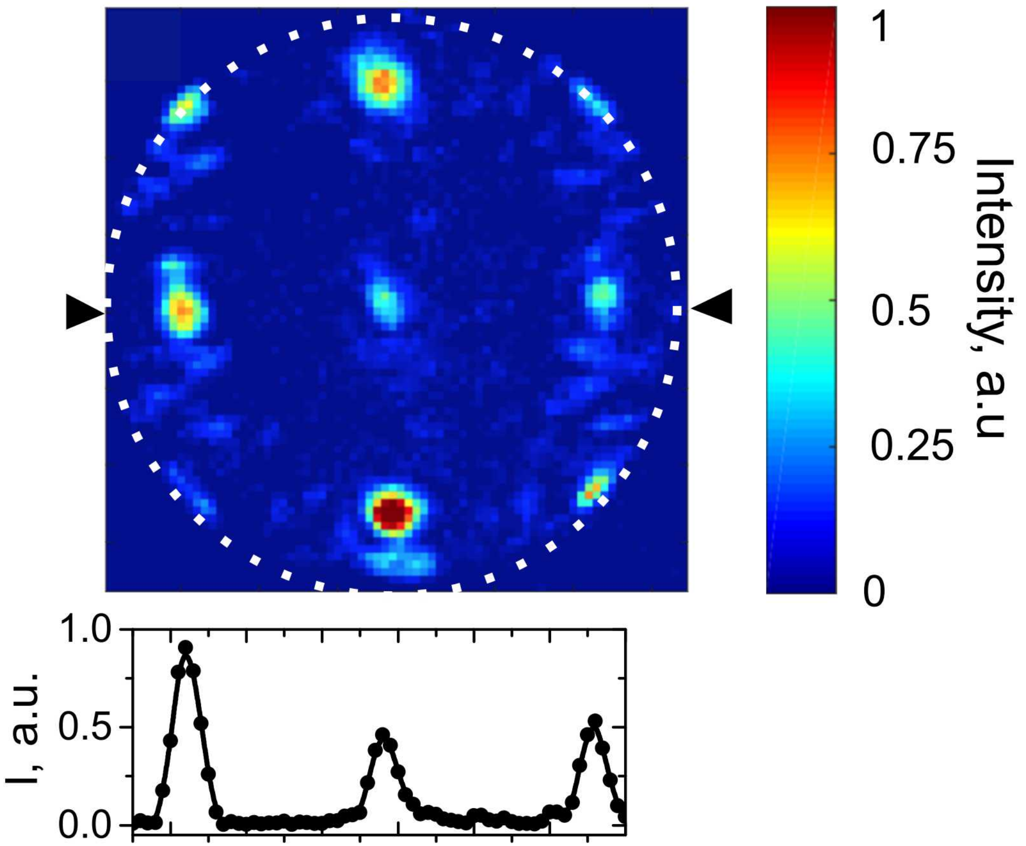


SHG enhancement at the wavelength of MD resonance

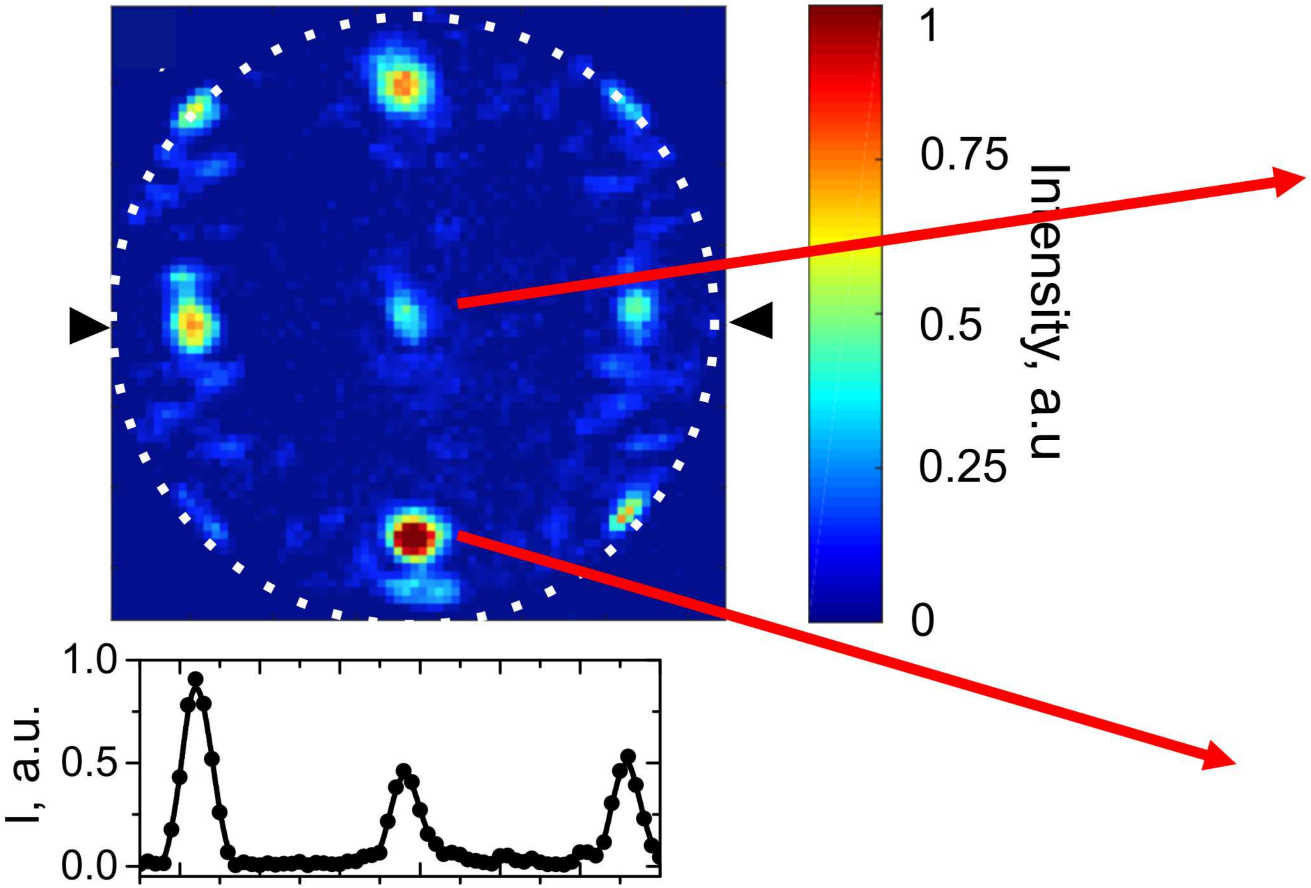
SHG in GaAs (111) metasurface: Fourier-space image



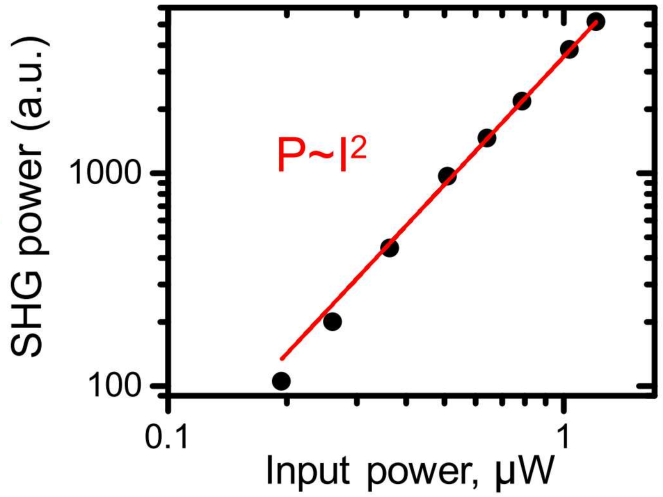
SHG enhancement at the wavelength of MD resonance



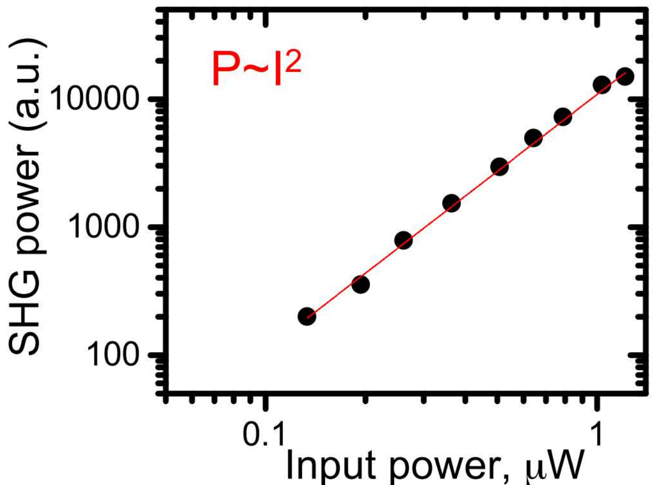
SHG in GaAs (111) metasurface: Fourier-space image



Power dependence for the zeroth-order diffraction

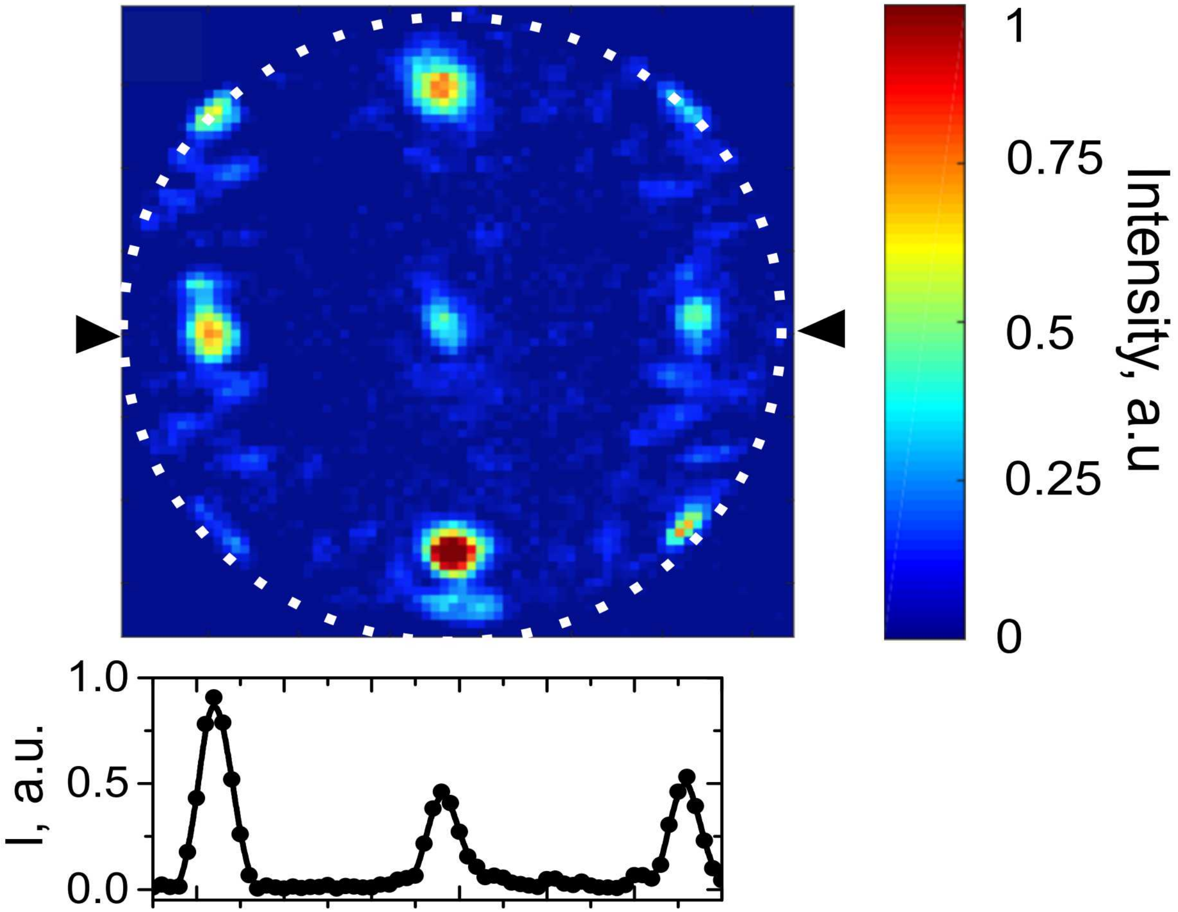


Power dependence for the bottom diffraction peak

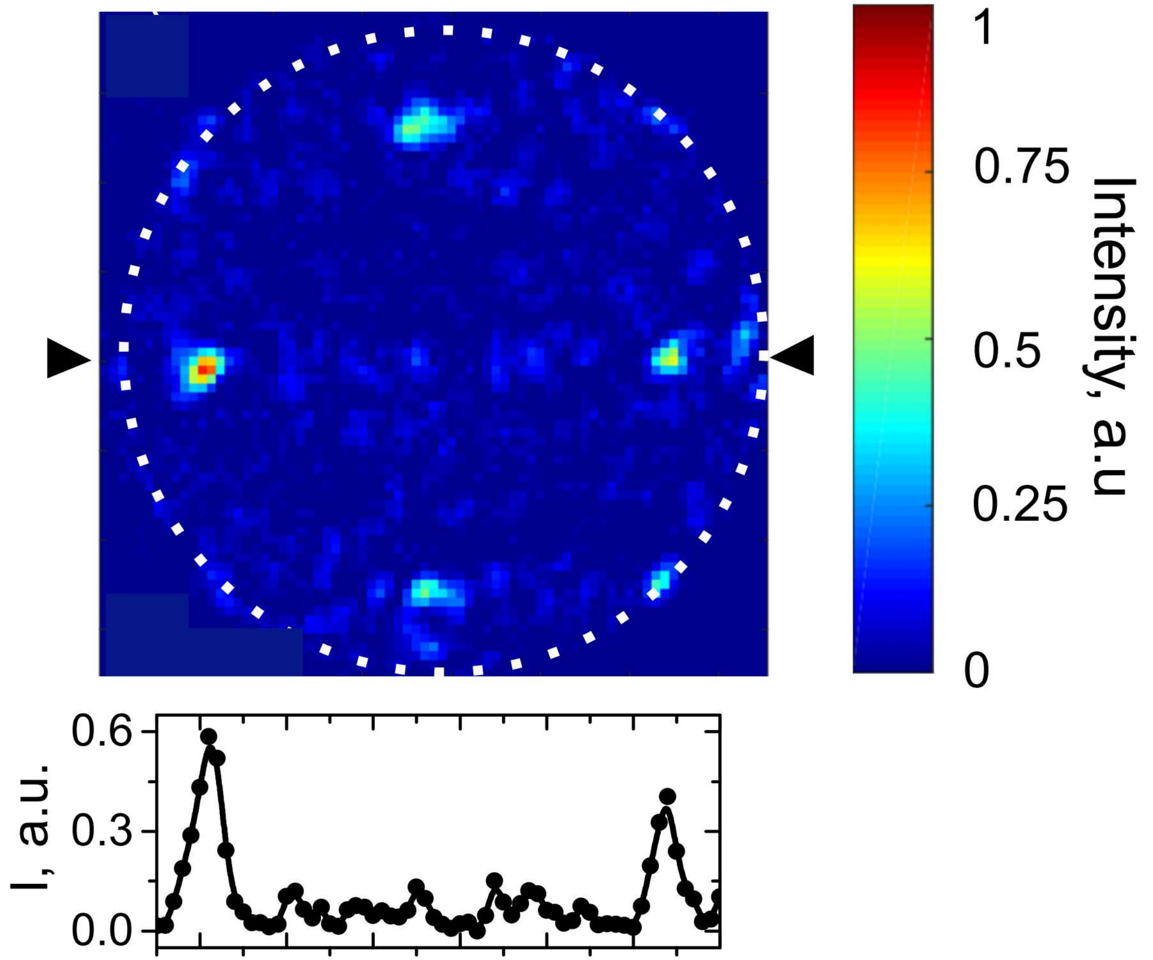


Comparison: Fourier-space images for metasurfaces with different crystallographic planes

(111)

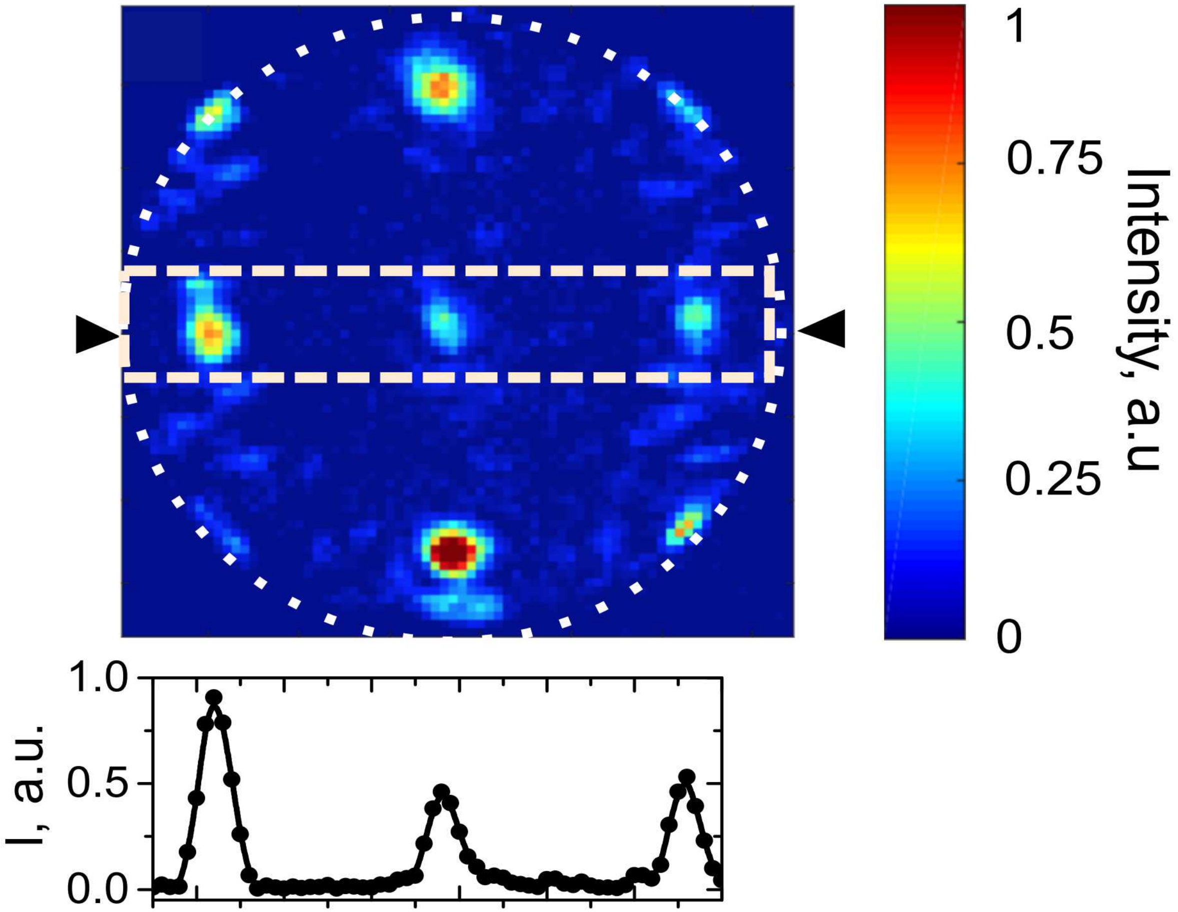


(100)

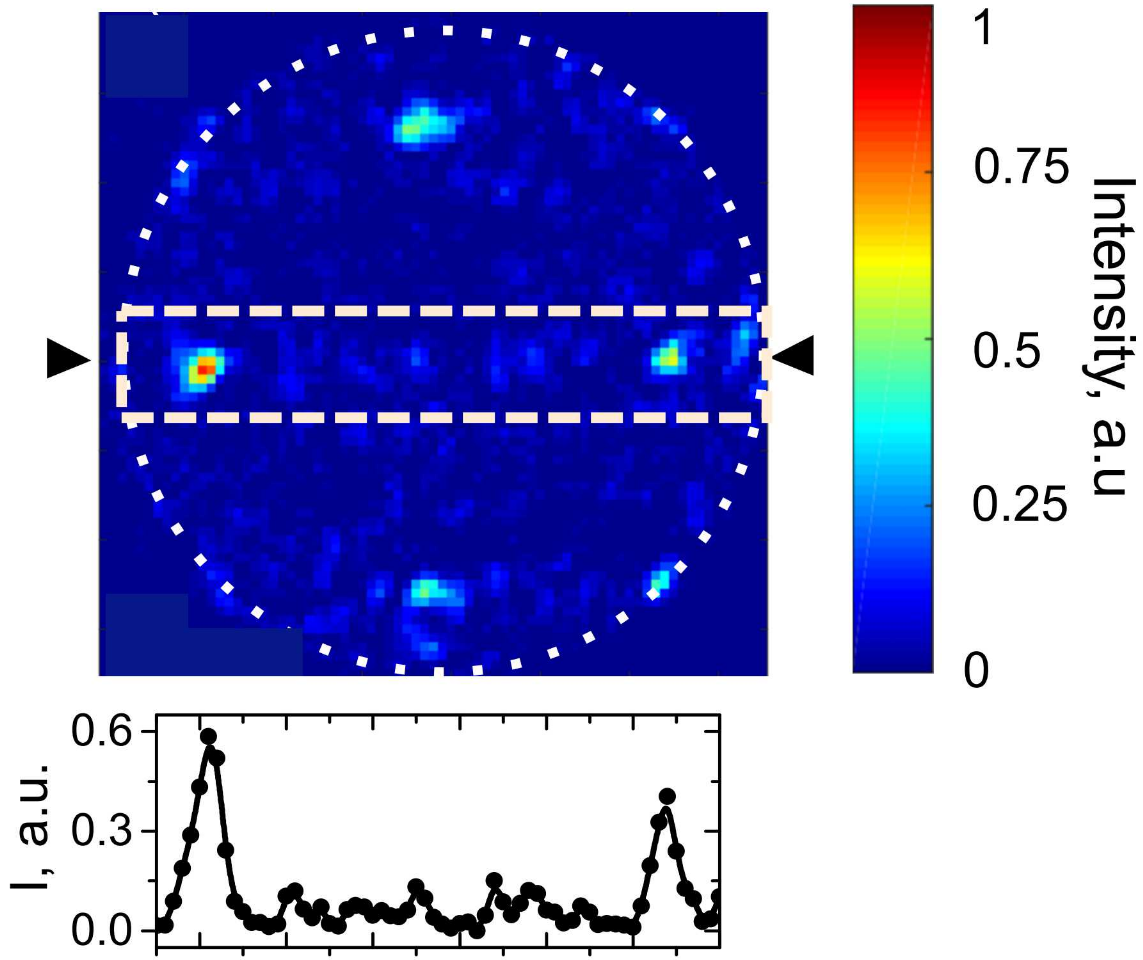


Comparison: Fourier-space images for metasurfaces with different crystallographic planes

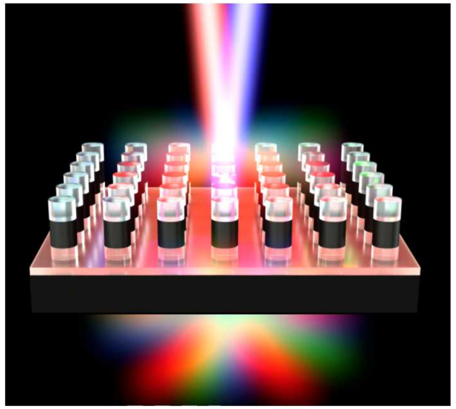
(111)



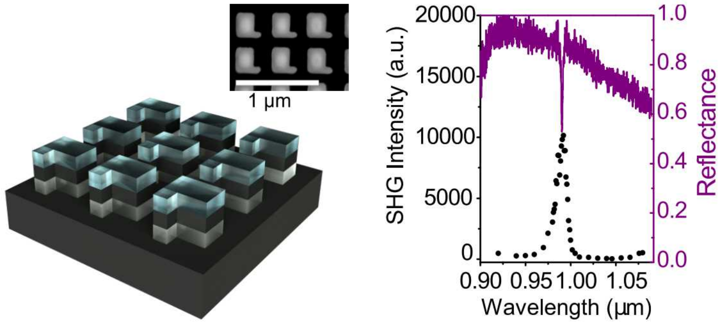
(100)



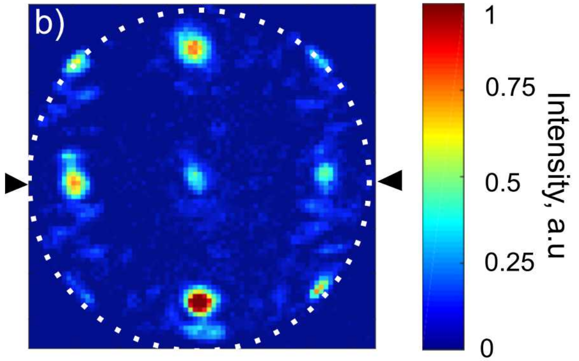
Conclusions



- 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.



- Nontrivial spectral shaping of second-harmonic spectra in symmetry broken III-V semiconductor metasurface.

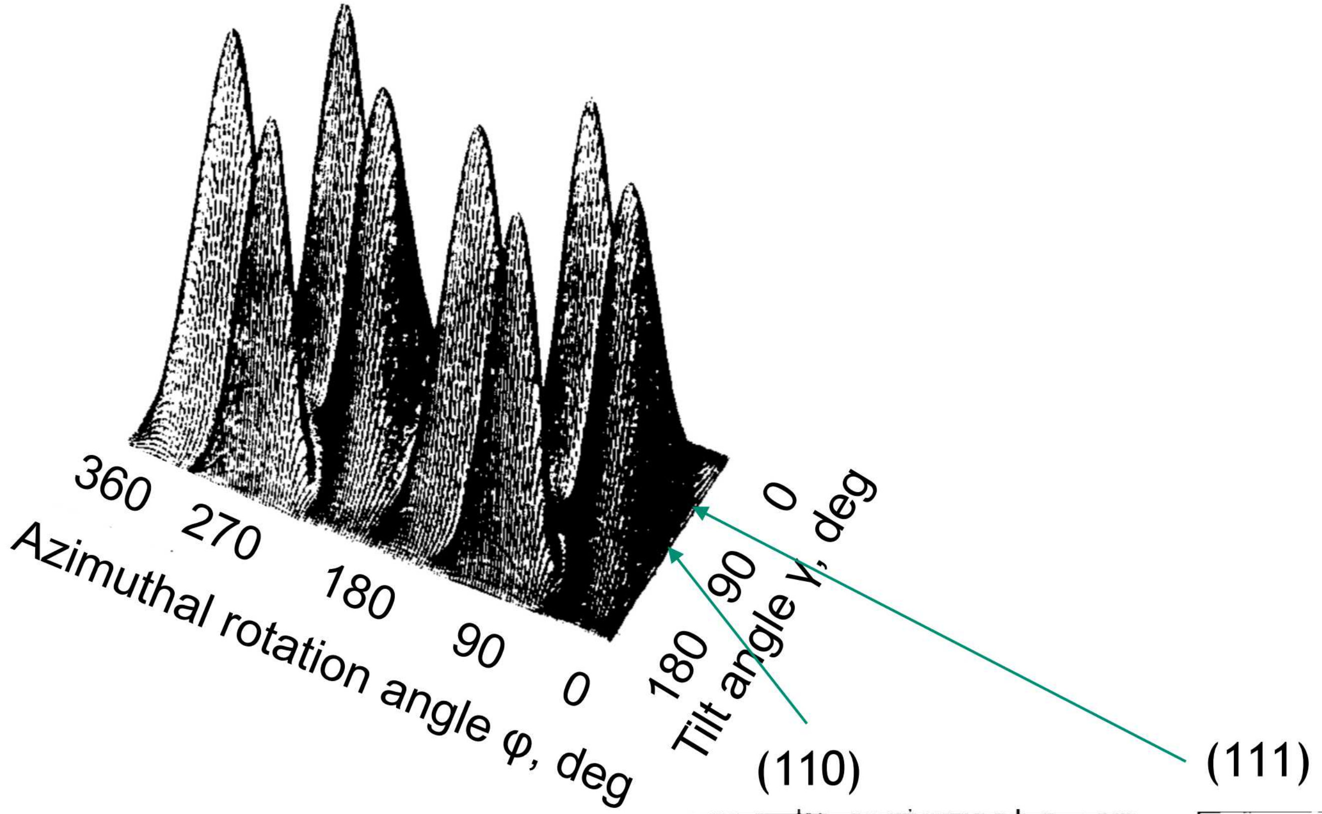


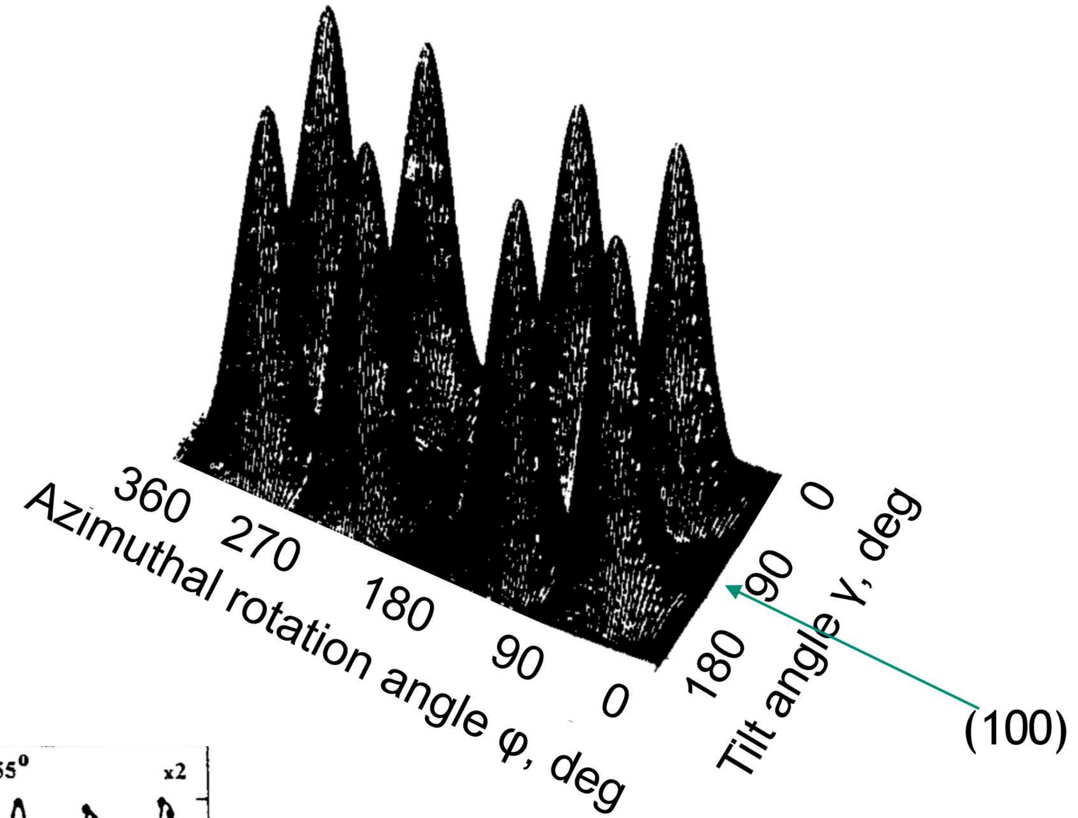
- We utilize III-V semiconductor metasurface fabricated from GaAs grown on (111) lattice plane to control the spatial distribution of the SHG intensity. By changing the orientation of the GaAs crystal from (100) to (111) plane we were able to redistribute the SHG generation to the zeroth diffraction order.

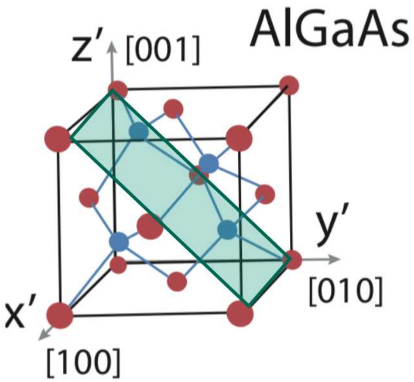
Liu, S.*, et. al, An optical metamixer. *arXiv preprint arXiv:1711.00090*, **2017**

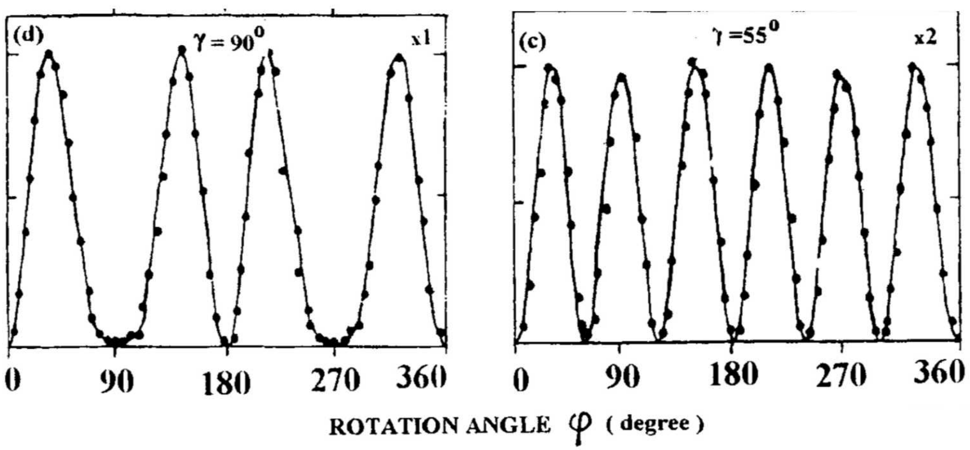
Vabishchevich, P, et. al *ACS Photonics. 5* (5), pp 1685–1690, **2018**

SHG in GaAs for different orientations of the crystal, (111), (110), (100)



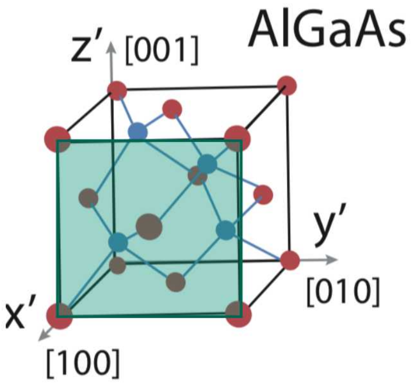






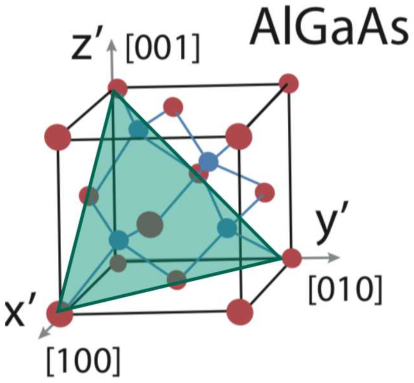
Kulyuk, Leonid L., et al. *Nonlinear Optics of Low-Dimensional Structures and New Materials*. Vol. 2801. International Society for Optics and Photonics, 1996.

Use of (111) GaAs/AlGaAs



$$d_{eff} = \begin{pmatrix} 0 & 0 & 0 & d_{41} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{41} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{41} \end{pmatrix}$$

→ Absence of SHG under normal incidence



$$d_{eff} = \begin{pmatrix} -\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 \\ 0 & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 & -\frac{1}{\sqrt{6}} \\ -\frac{1}{2\sqrt{3}} & -\frac{1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} & 0 & 0 & 0 \end{pmatrix} \cdot d_{41}$$

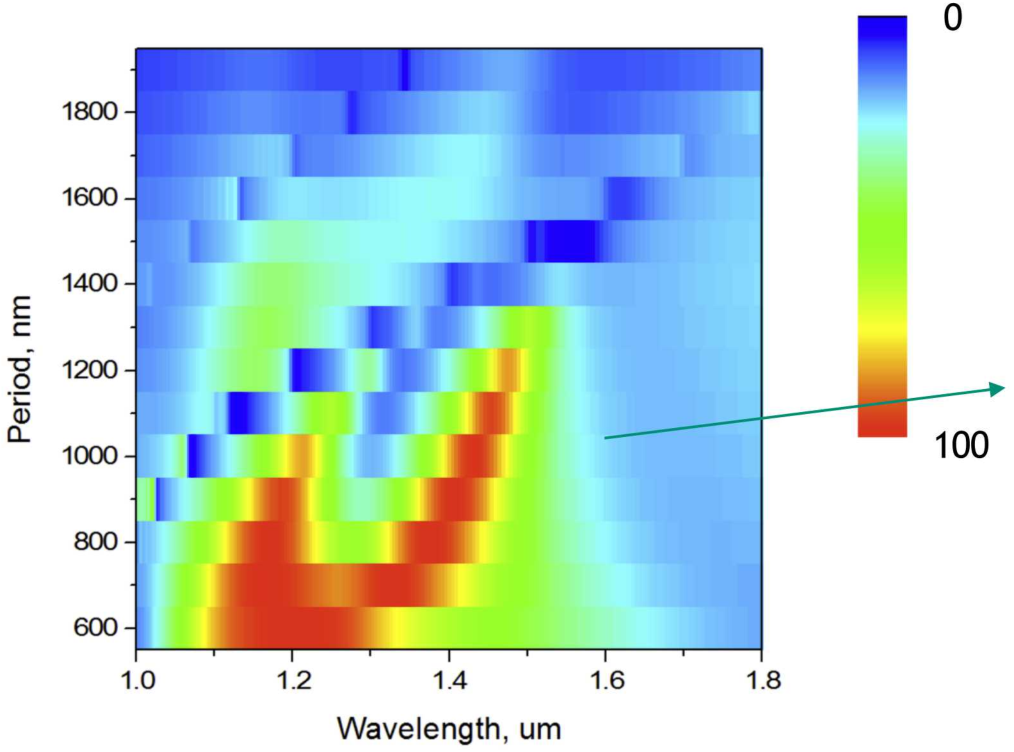
Non-zero SHG signal for (111) Lattice plane under normal incidence

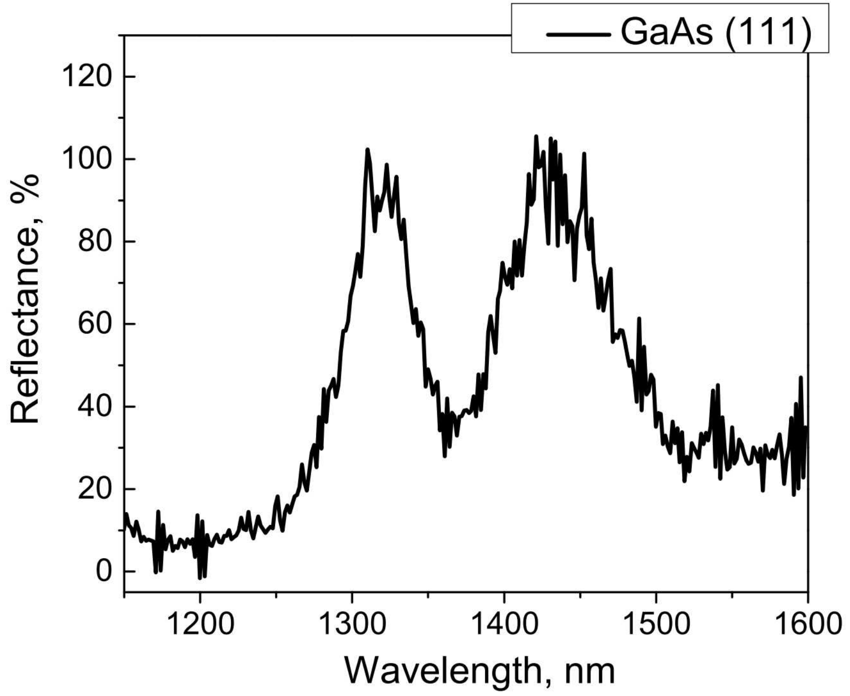
$$P_x(2w) = 2\varepsilon_0 d_{eff}(-\frac{1}{\sqrt{6}})E_x(w)^2 + \frac{1}{\sqrt{6}}E_y(w)^2 - \frac{1}{\sqrt{3}}E_x(w)E_z(w)$$

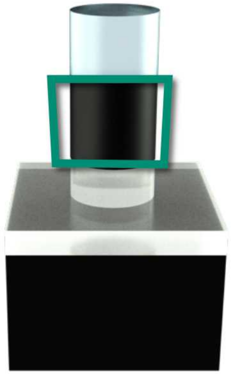
$$P_y(2w) = 2\varepsilon_0 d_{eff}(-\frac{2}{\sqrt{3}})E_y(w)E_z(w) - \frac{1}{\sqrt{6}}E_x(w)E_z(w)$$

$$P_z(2w) = 2\varepsilon_0 d_{eff}(-\frac{1}{2\sqrt{3}})E_x(w)^2 - \frac{1}{2\sqrt{3}}E_y(w)^2 - \frac{1}{1\sqrt{3}}E_z^2$$

Reflectance spectrum

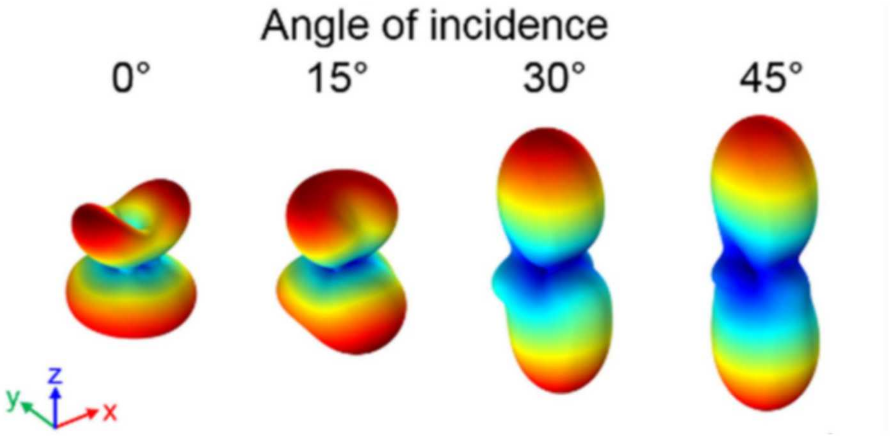






Tailoring of SHG in normal direction

Use of non-normal incidence



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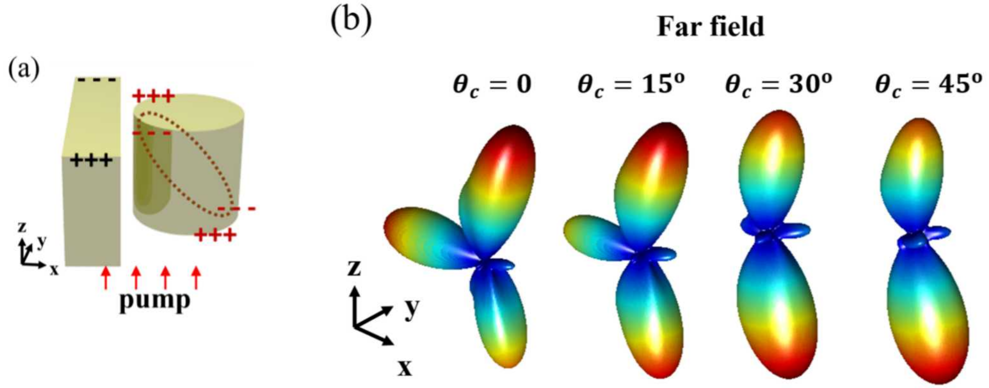

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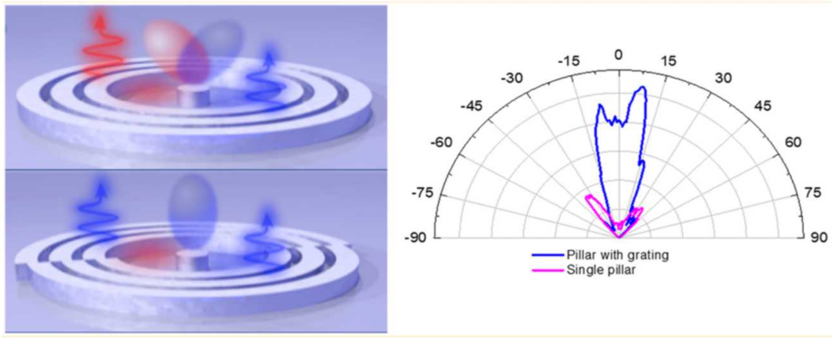
Use of additional gratings



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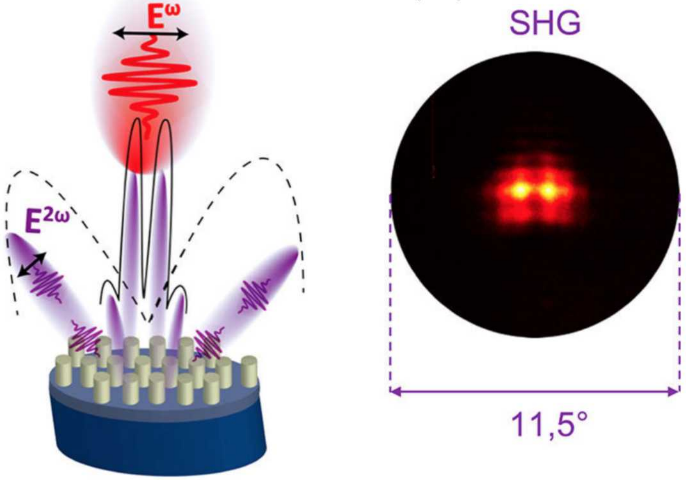

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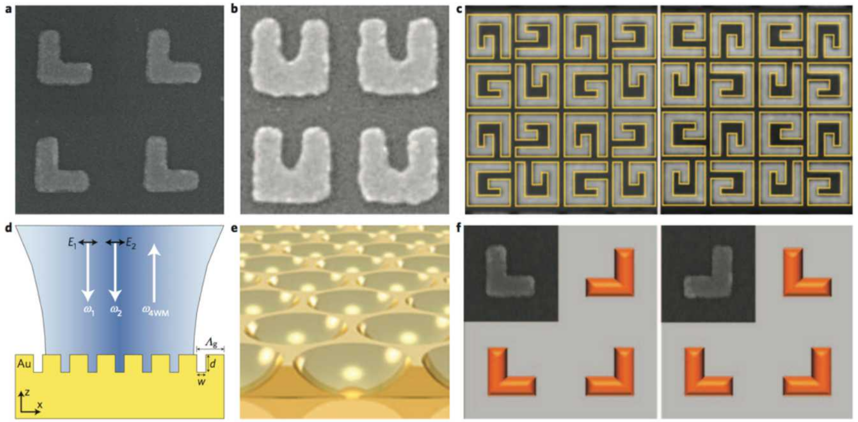
Nonlinear optical frequency mixing in nanostructures

Engineered structures that have specific optical properties on demand by choosing materials, shapes and dimensions of the meta-atoms

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



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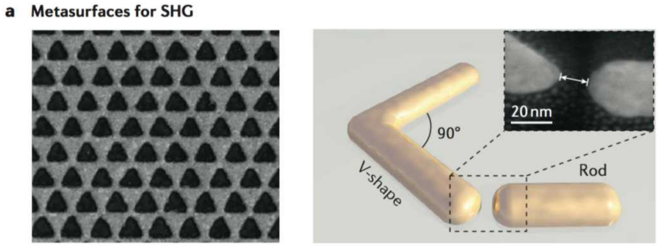

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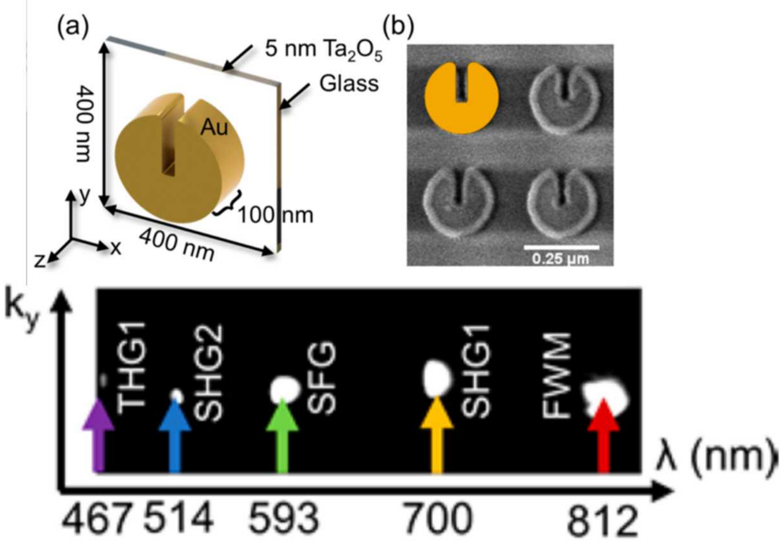

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- Ohmic losses
- Plasmonic structure: small modal volume (surface nonlinearities)

SHG efficiency ~ 5×10^{-10} W/W^2