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All-dielectric metasurfaces: optical nonlinearities and emission control

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Outline

Nonlinear optics in GaAs metasurfaces

- Frequency mixing
- SHG with GaAs (111) metasurface into zeroth diffraction order
- SHG in Fano metasurface

Emission engineering in semiconductor metasurfaces

Outline

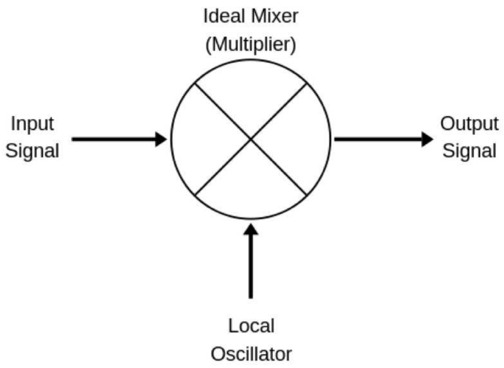
Nonlinear optics in GaAs metasurfaces

- Frequency mixing
- SHG with GaAs (111) metasurface into zeroth diffraction order
- SHG in Fano metasurface

Emission engineering in semiconductor metasurfaces

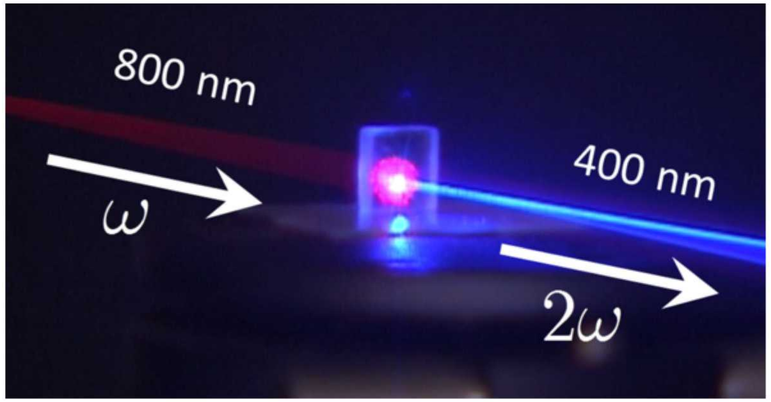
Frequency mixing – from electronics to optics

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

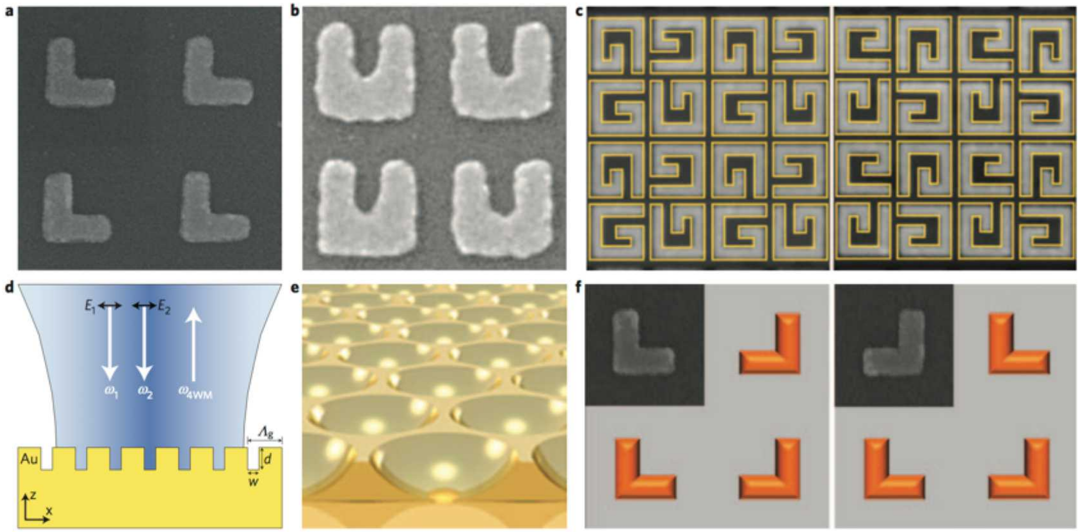
Nonlinear crystal dimensions >> wavelengths

“Super” frequency mixing?

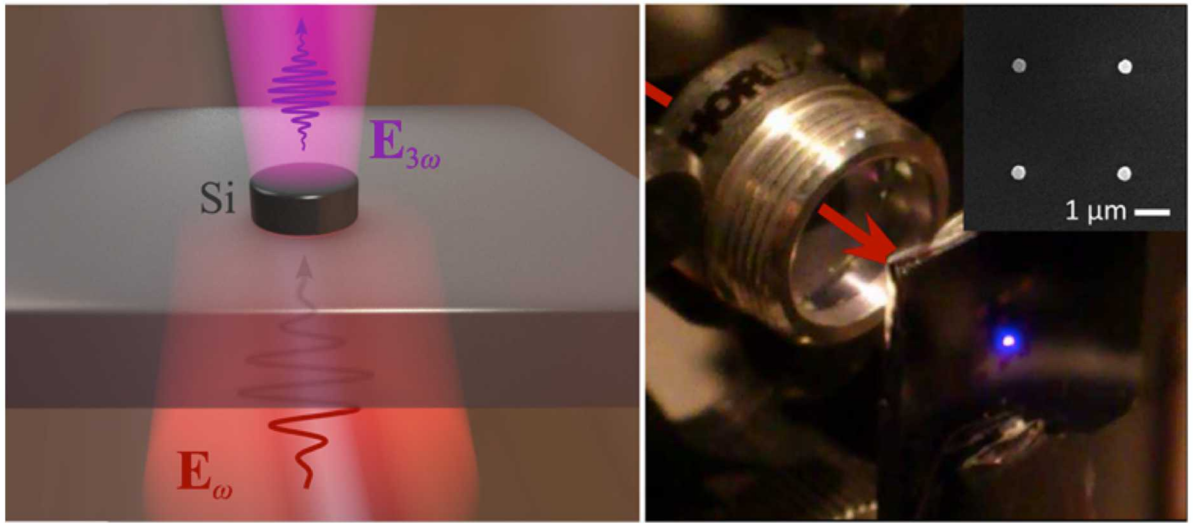
Can we see all of these mixings at the same time?

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

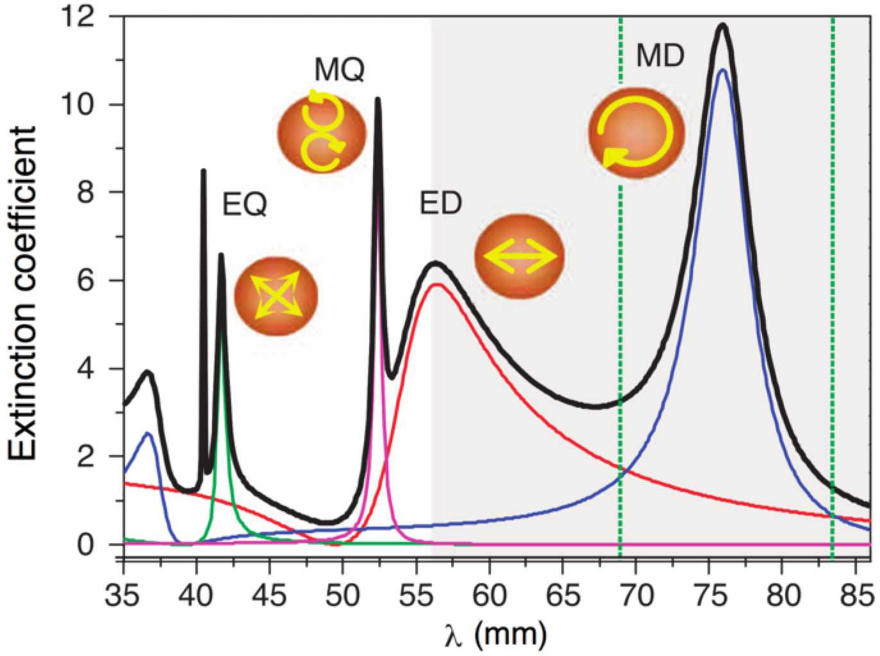
Resonant enhancement of EM field

Dimensions < wavelength

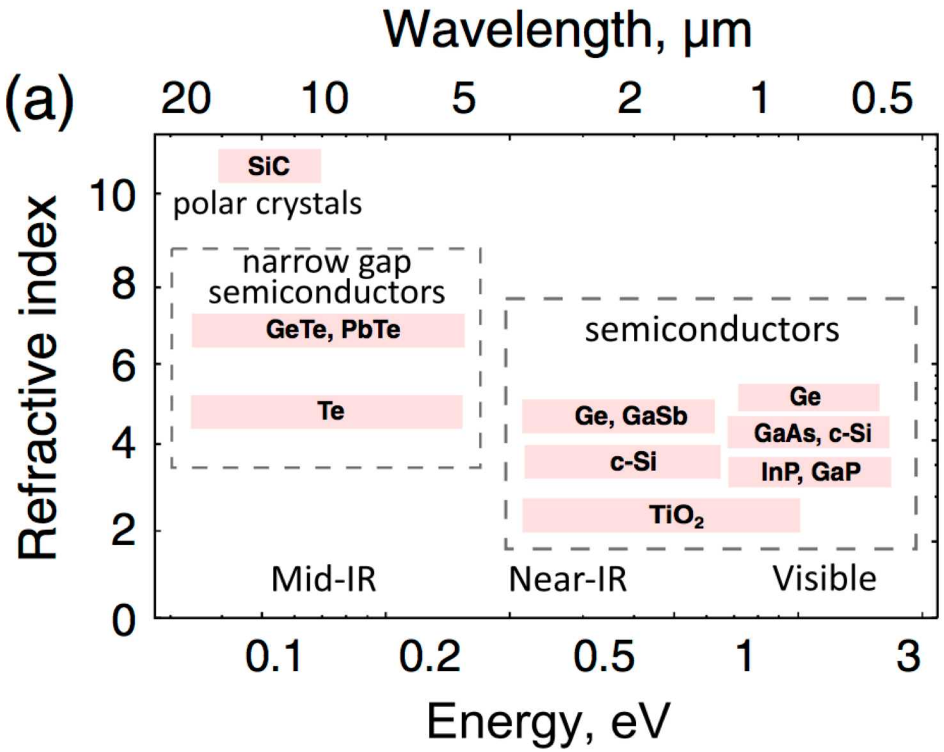
Relaxed phase matching conditions

Mie resonances in all-dielectric metasurfaces

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**

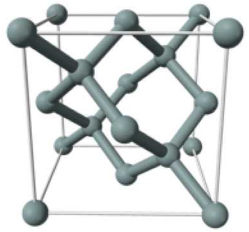
Mie resonances

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

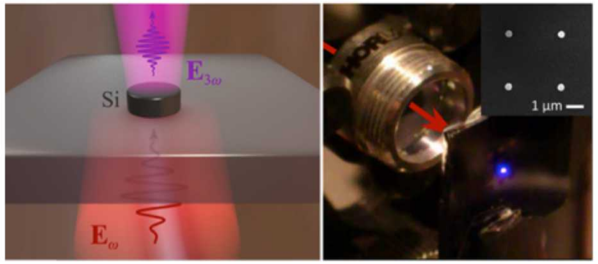
Nonlinear dielectric metasurfaces

centrosymmetric

Si
Ge

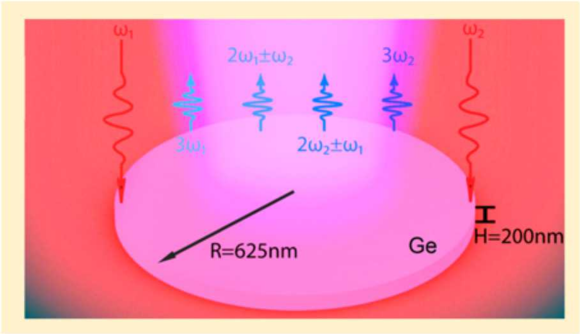


Third Harmonic Generation



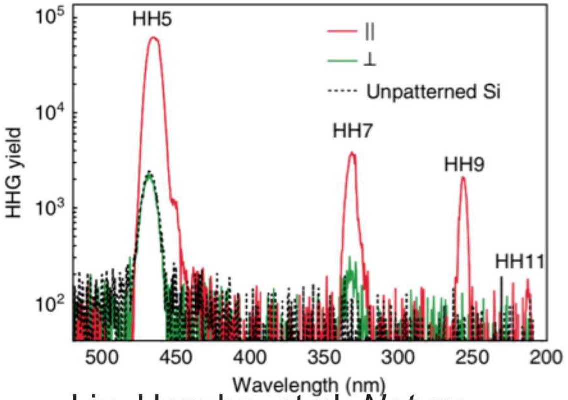
M .R. Shcherbakov, et. al. *Nano Letters* **14,** 11, 6488–6492, **2014**

Four-Wave Mixing



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

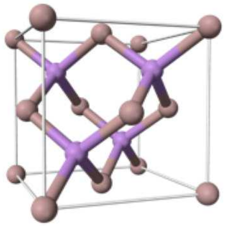
High-Harmonic Generation



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

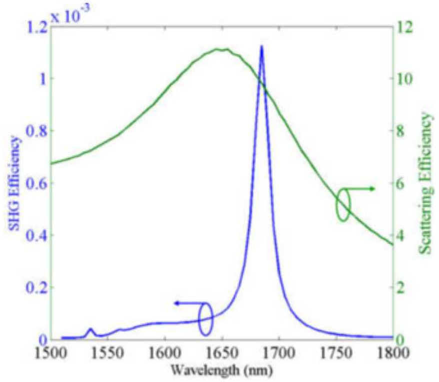
Non-centrosymmetric

GaAs
AlGaAs



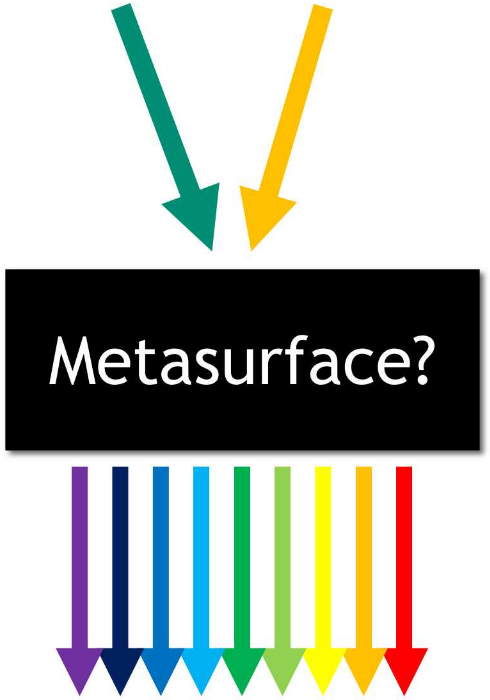
https://www.wikipedia.org

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



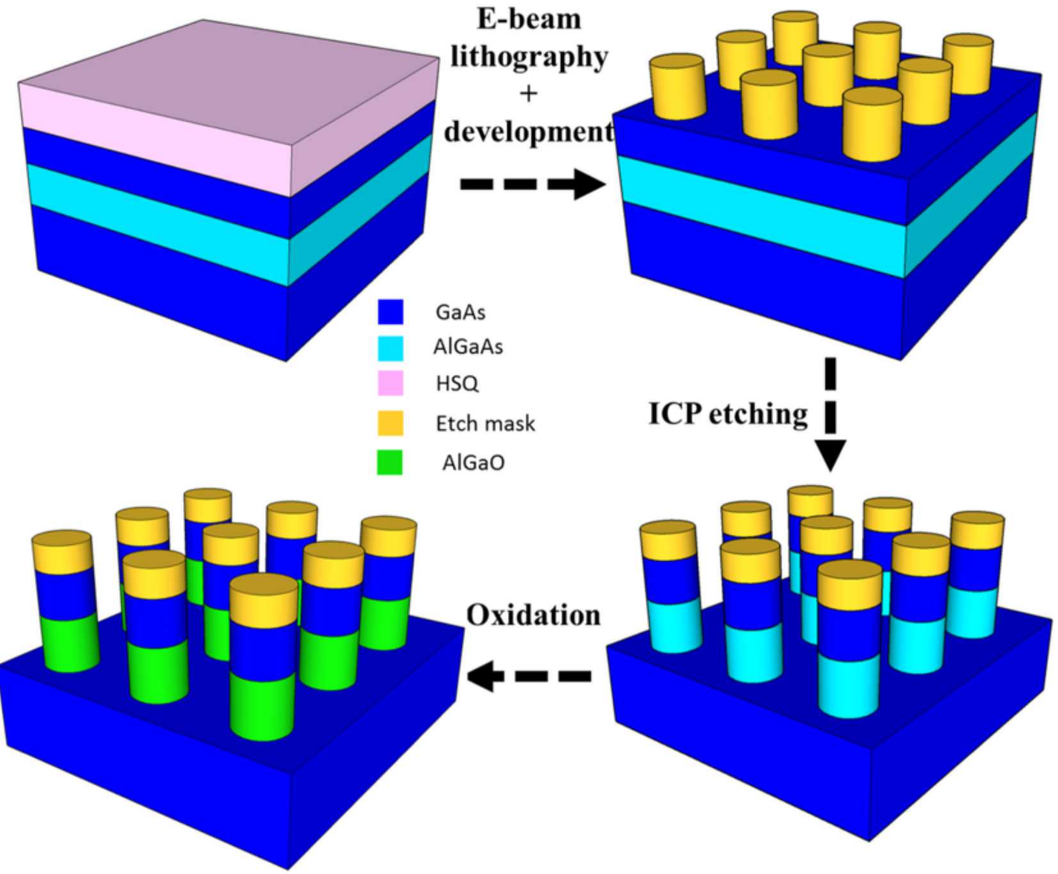
L. Carletti et al., *ACS Photonics*, *3*(8), 1500-1507, **2016**

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

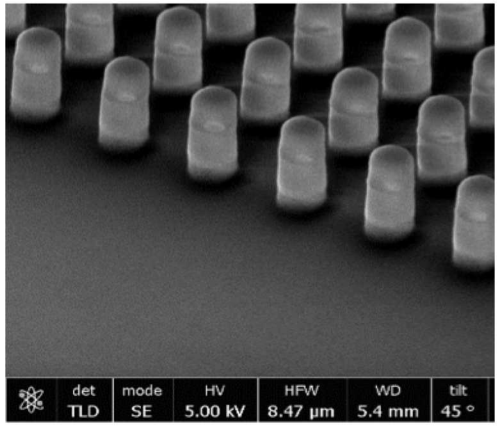
Fabrication of GaAs metasurface



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

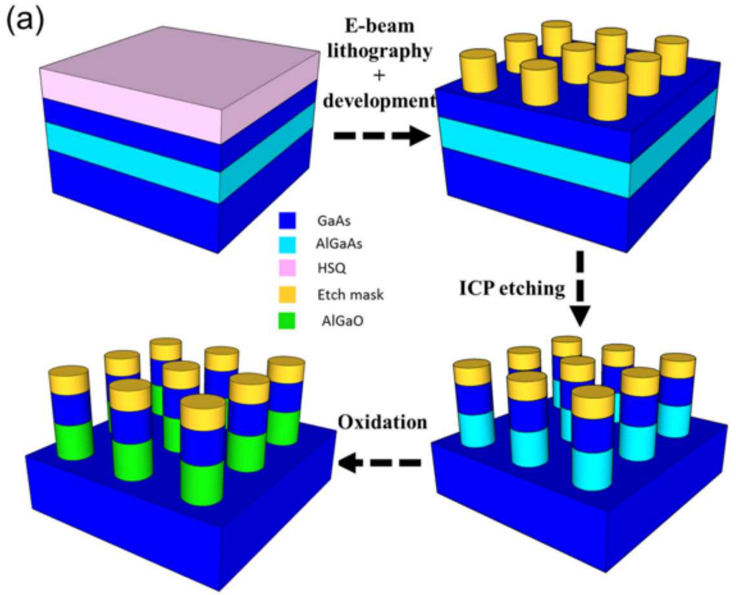
Diameter 400 nm
Period 840 nm

SEM image



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

GaAs-based metasurfaces



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

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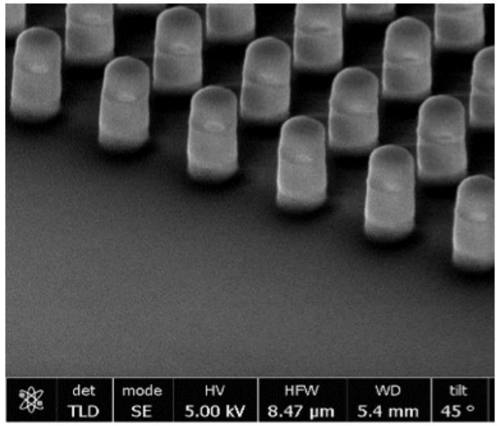
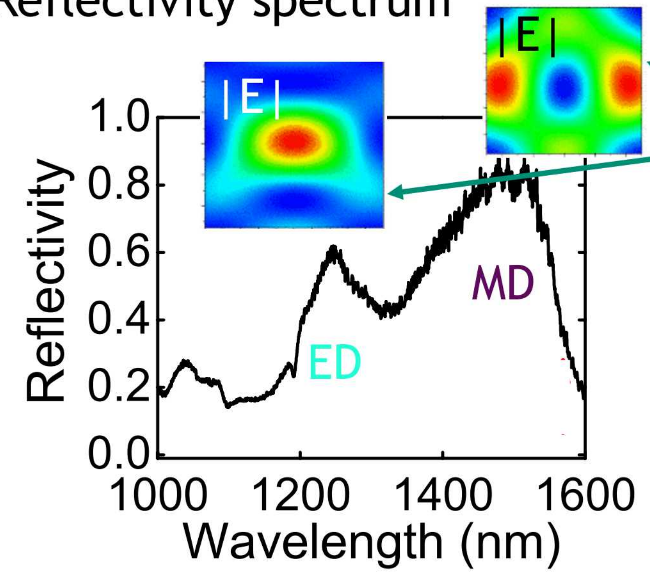
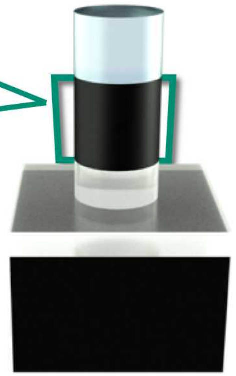
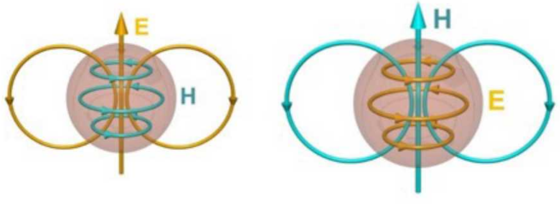
SEM image



Reflectivity spectrum



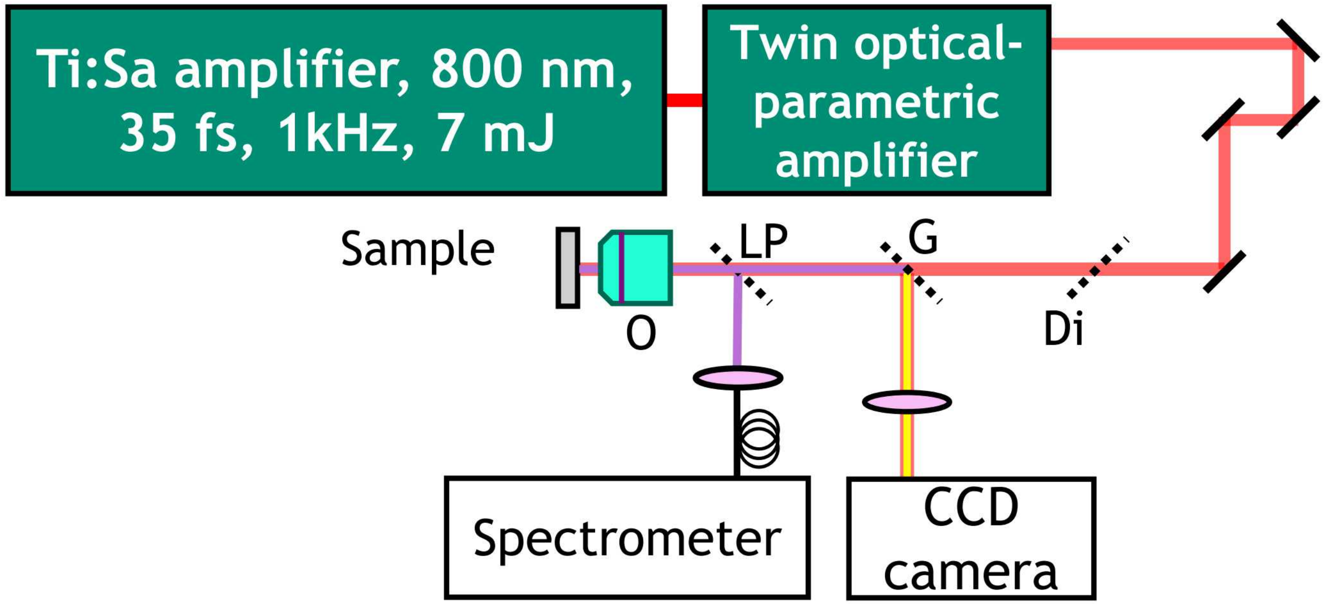




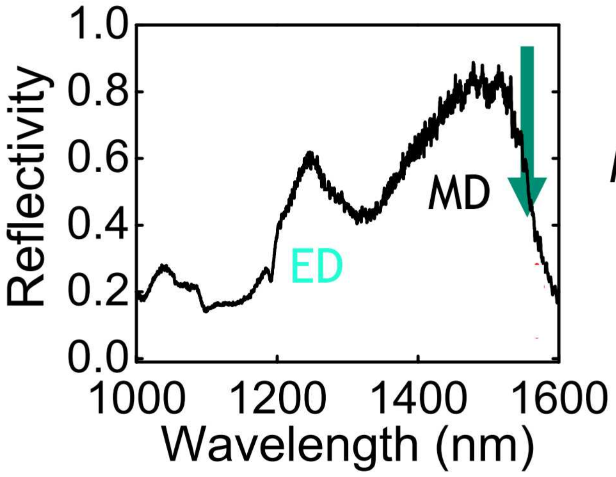
Images: A. Miroshnichenko

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

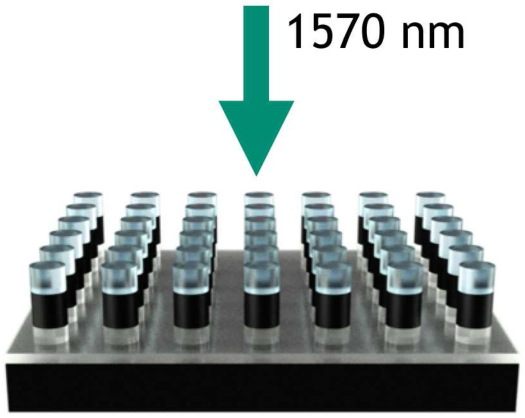
Single beam experiment



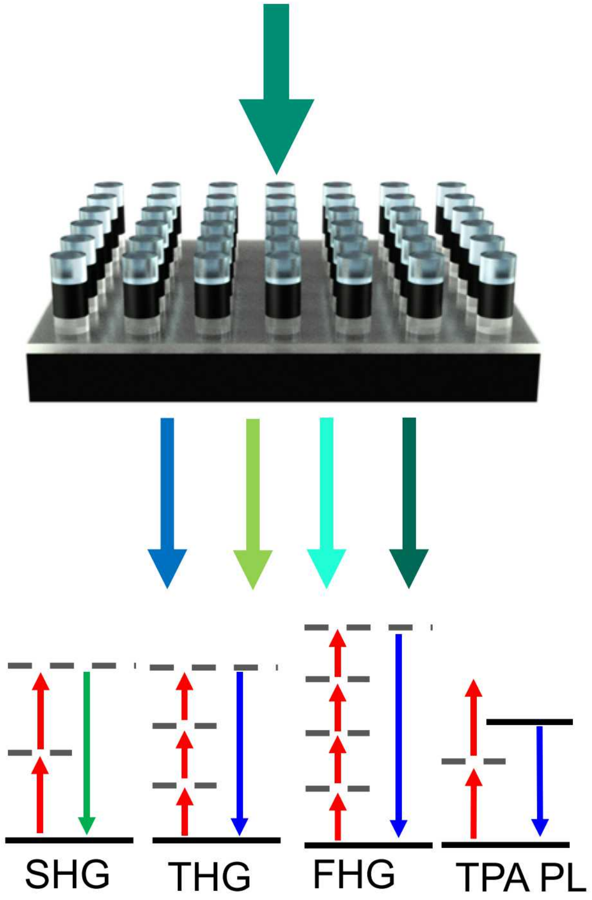
50 fs
1 kHz
0.4 NA objective

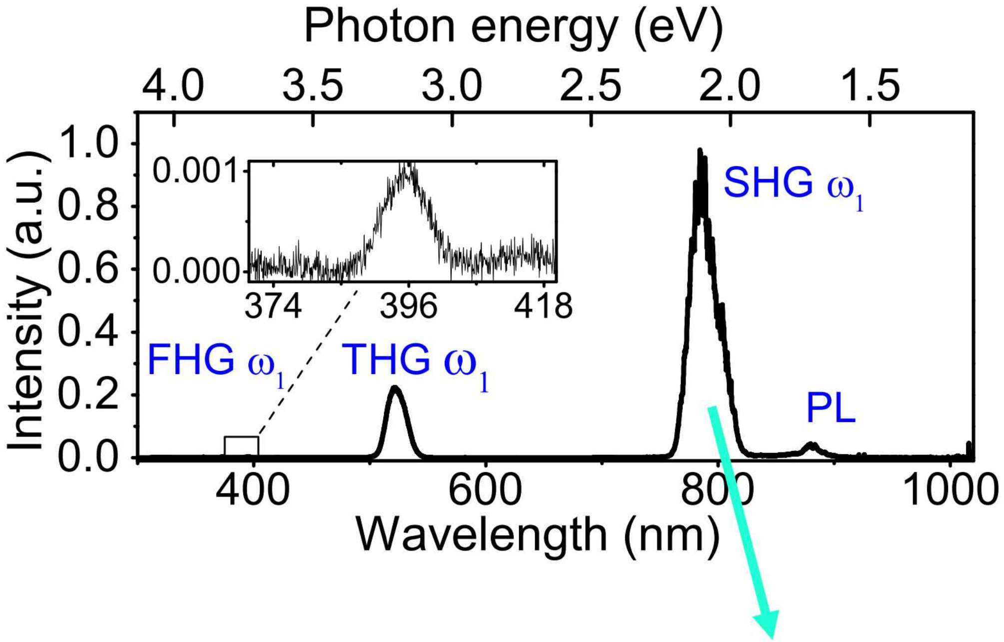


Magnetic dipole resonance

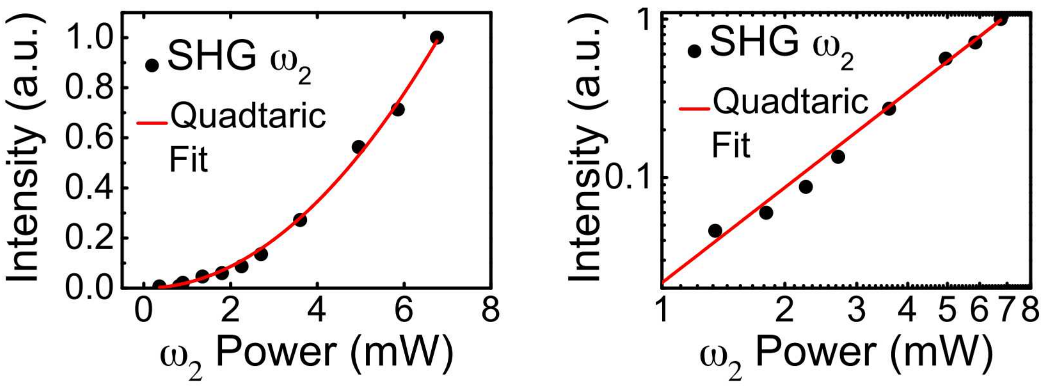


2^{nd}, 3^{rd}, 4^{th} harmonic generation spectra

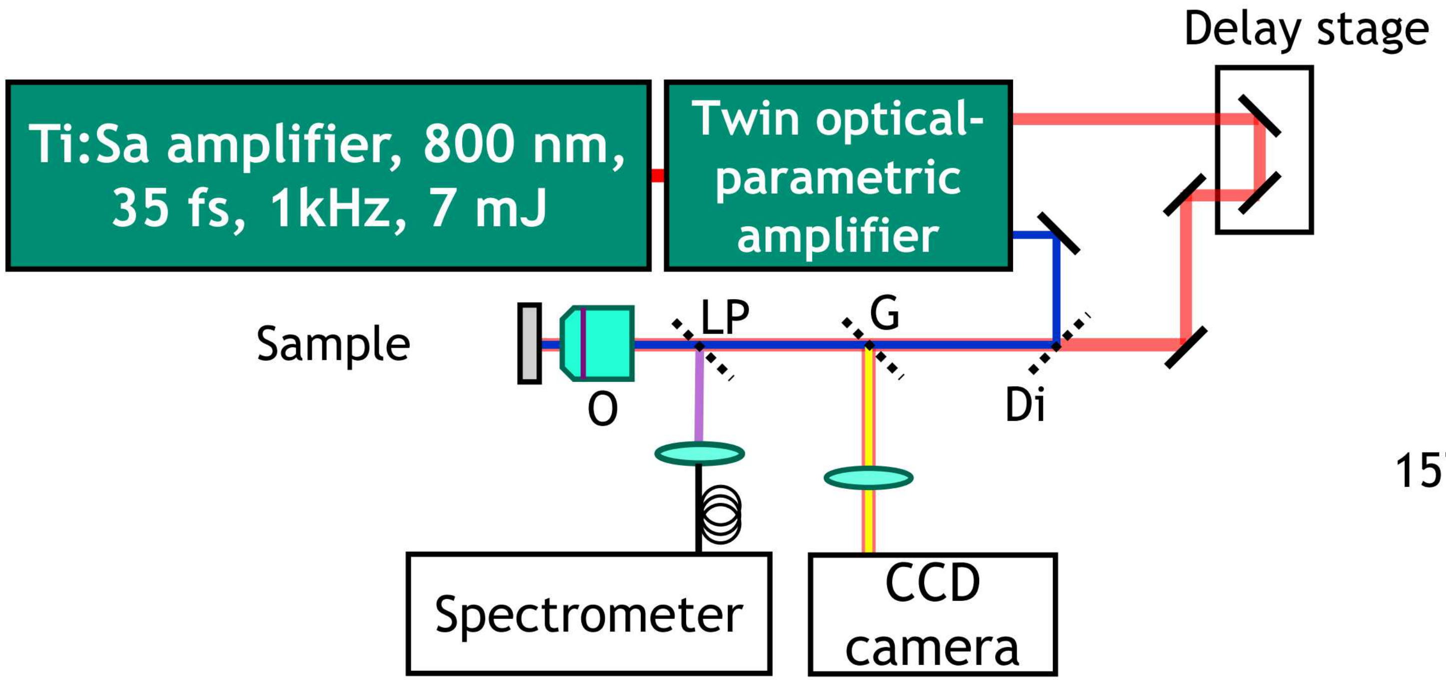




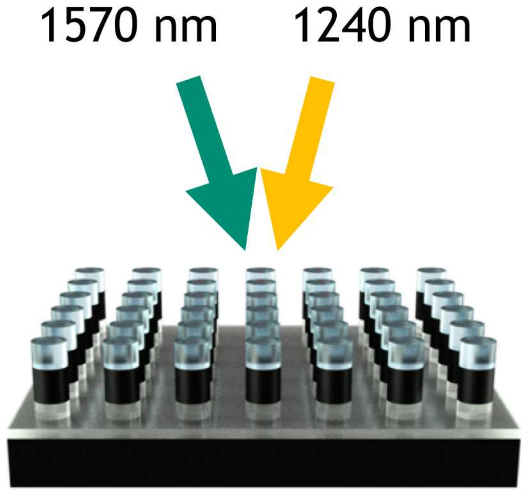
SHG power dependence

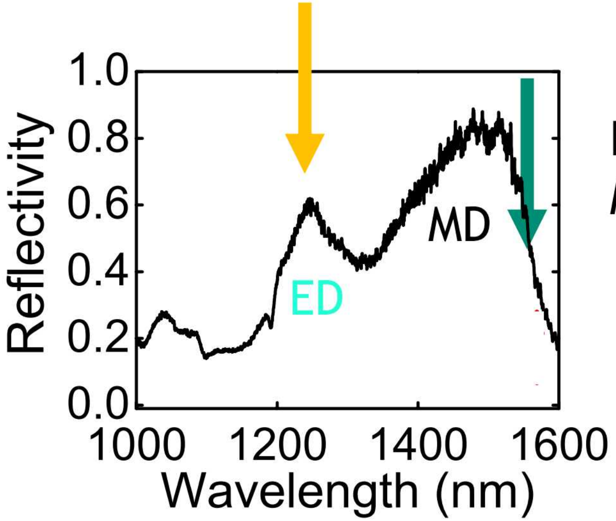


Two-beam experiment



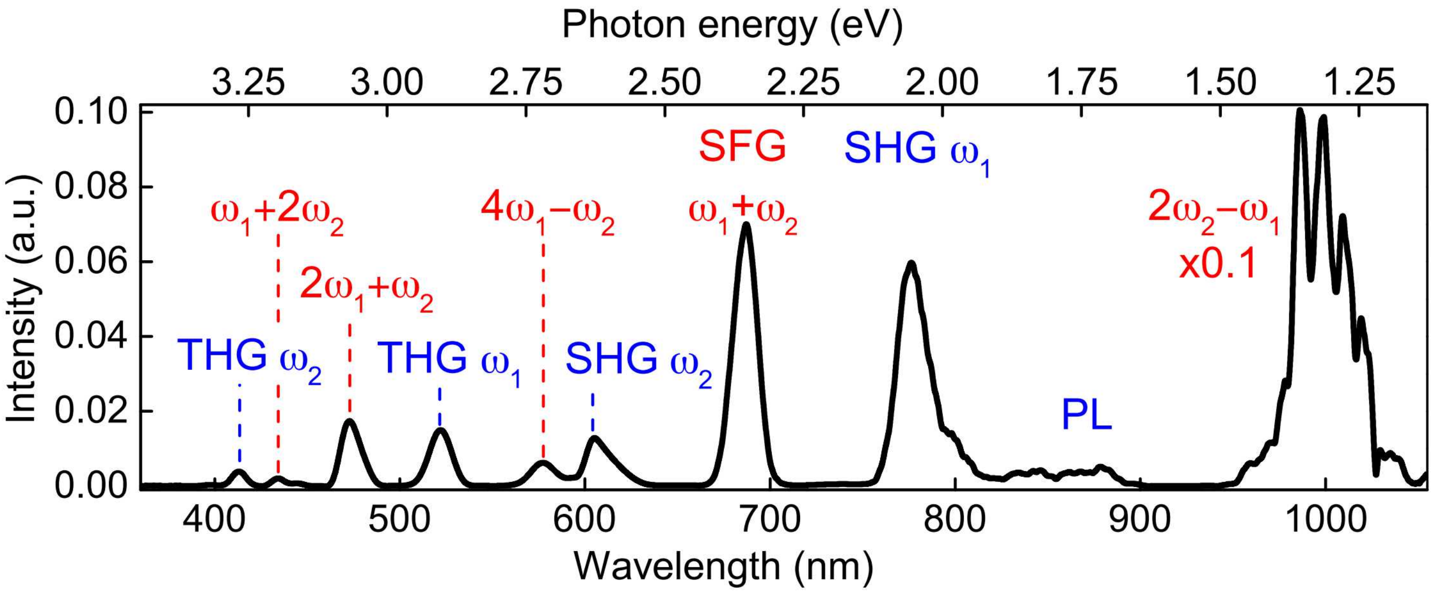
50 fs
1 kHz
0.4 NA objective

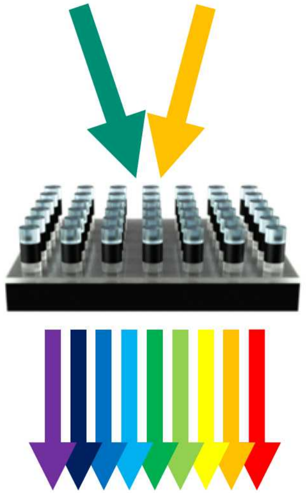




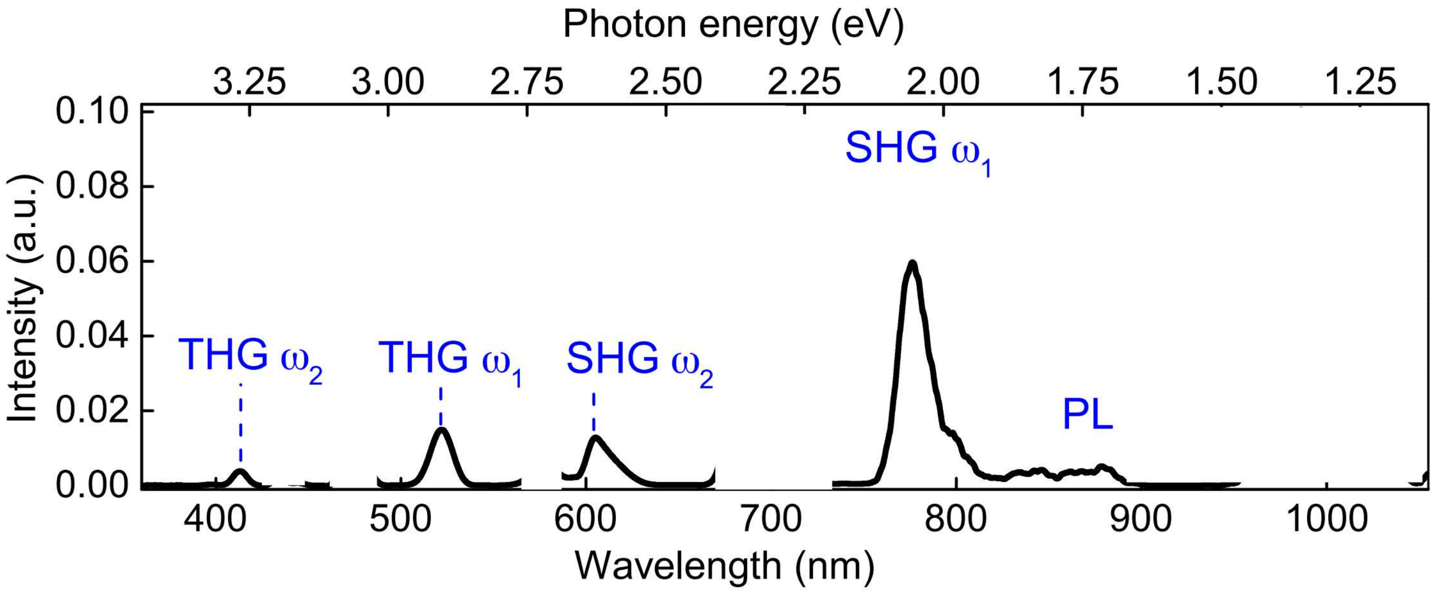
Electric dipole resonance
Magnetic dipole resonance

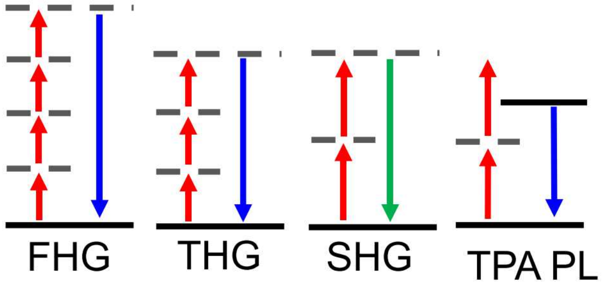
Frequency mixing spectra

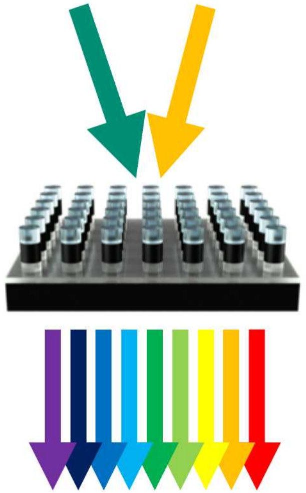




Frequency mixing spectra

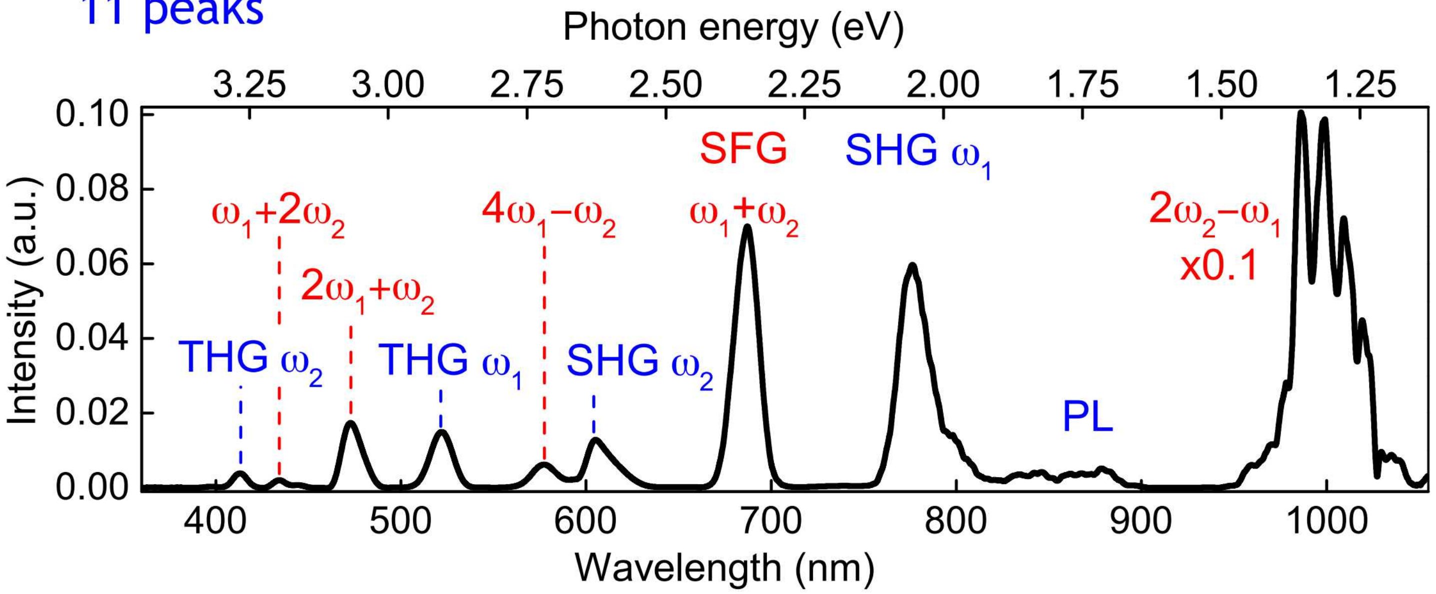


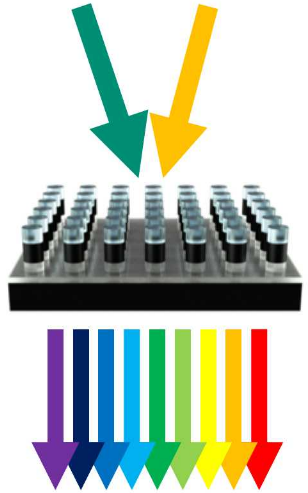


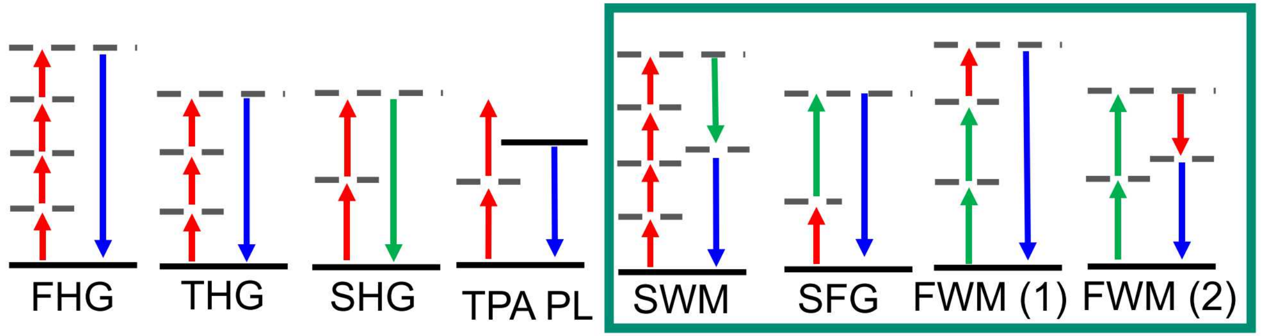


Frequency mixing spectra

11 peaks



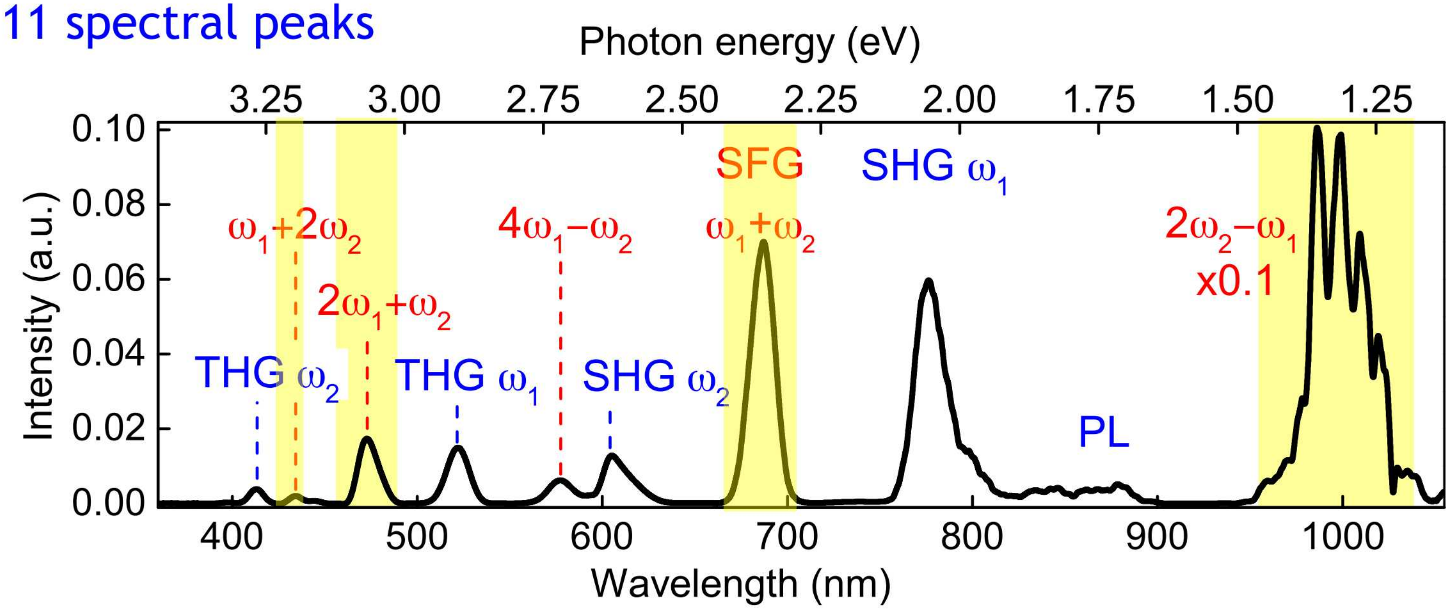


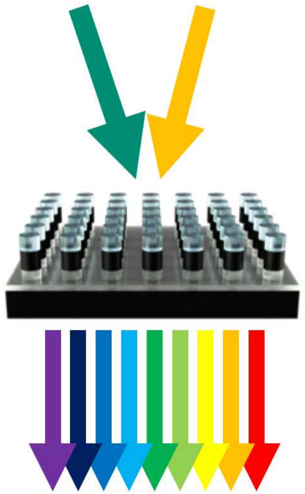


7 different nonlinear processes: 2nd, 3rd, 4th harmonics, sum frequency generation, 4 wave-mixing, six-wave mixing, PL induced by two-photon absorption

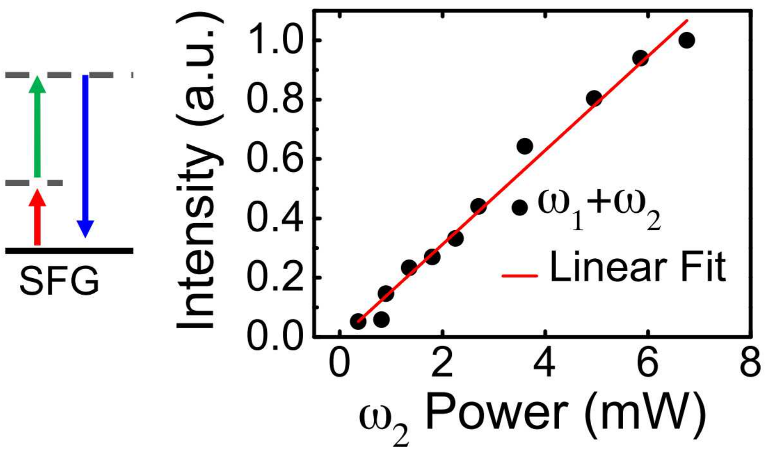
Frequency mixing spectra

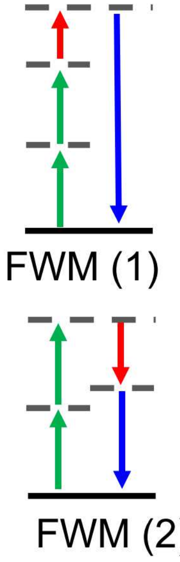
11 spectral peaks



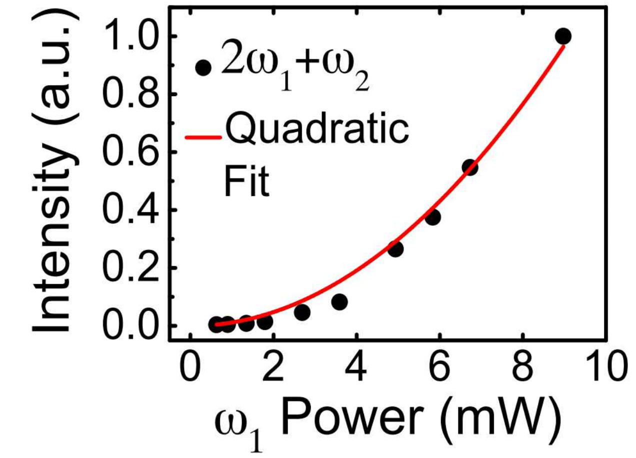


SFG power dependence



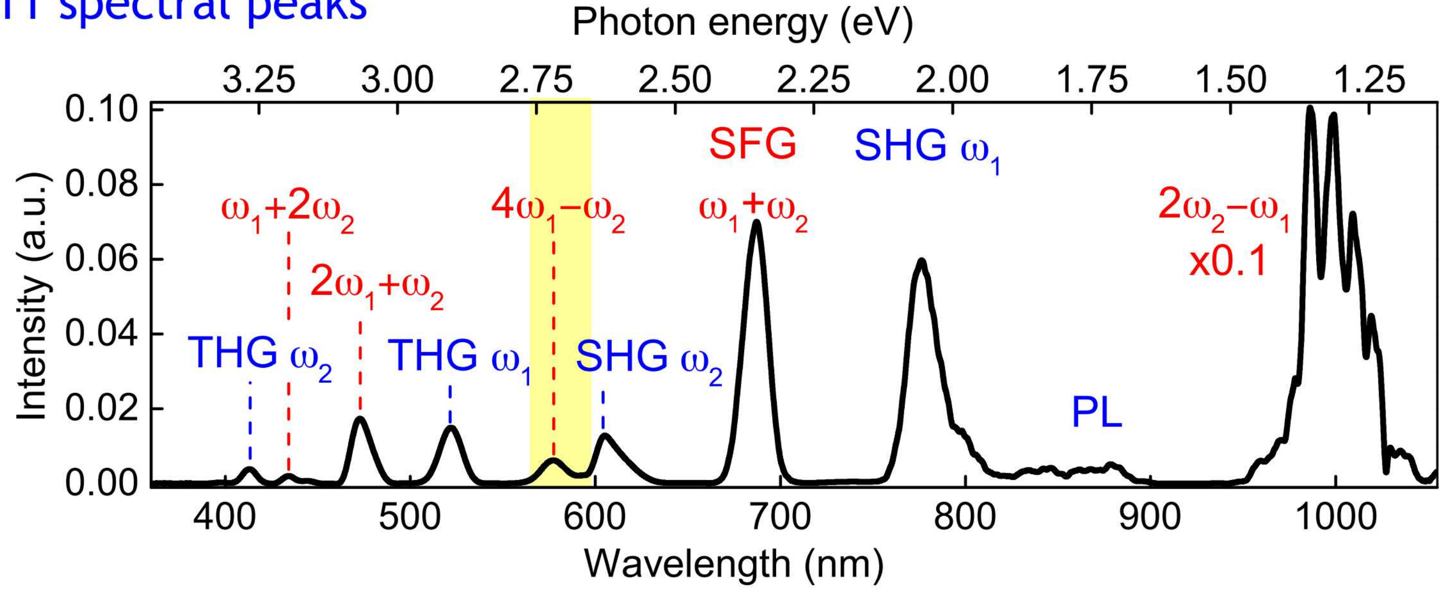


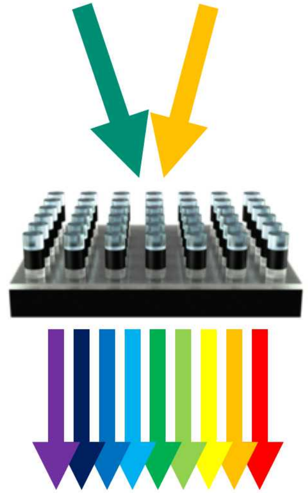
FWM power dependence



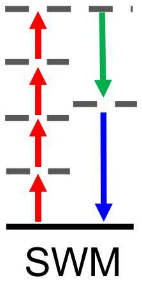
Frequency mixing spectra: six wave mixing

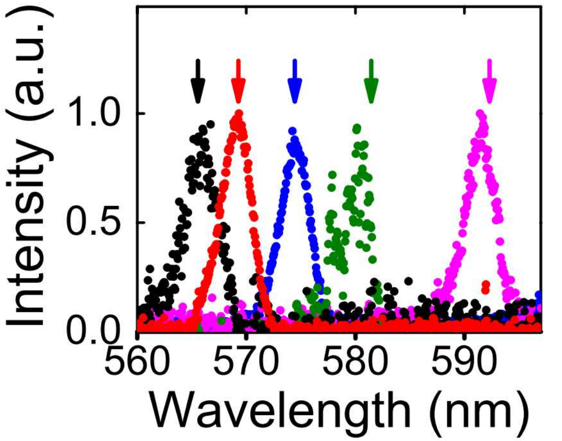
11 spectral peaks



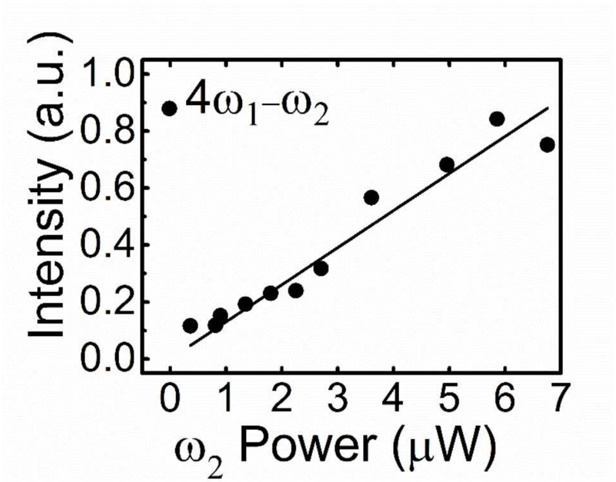


Wavelength shift

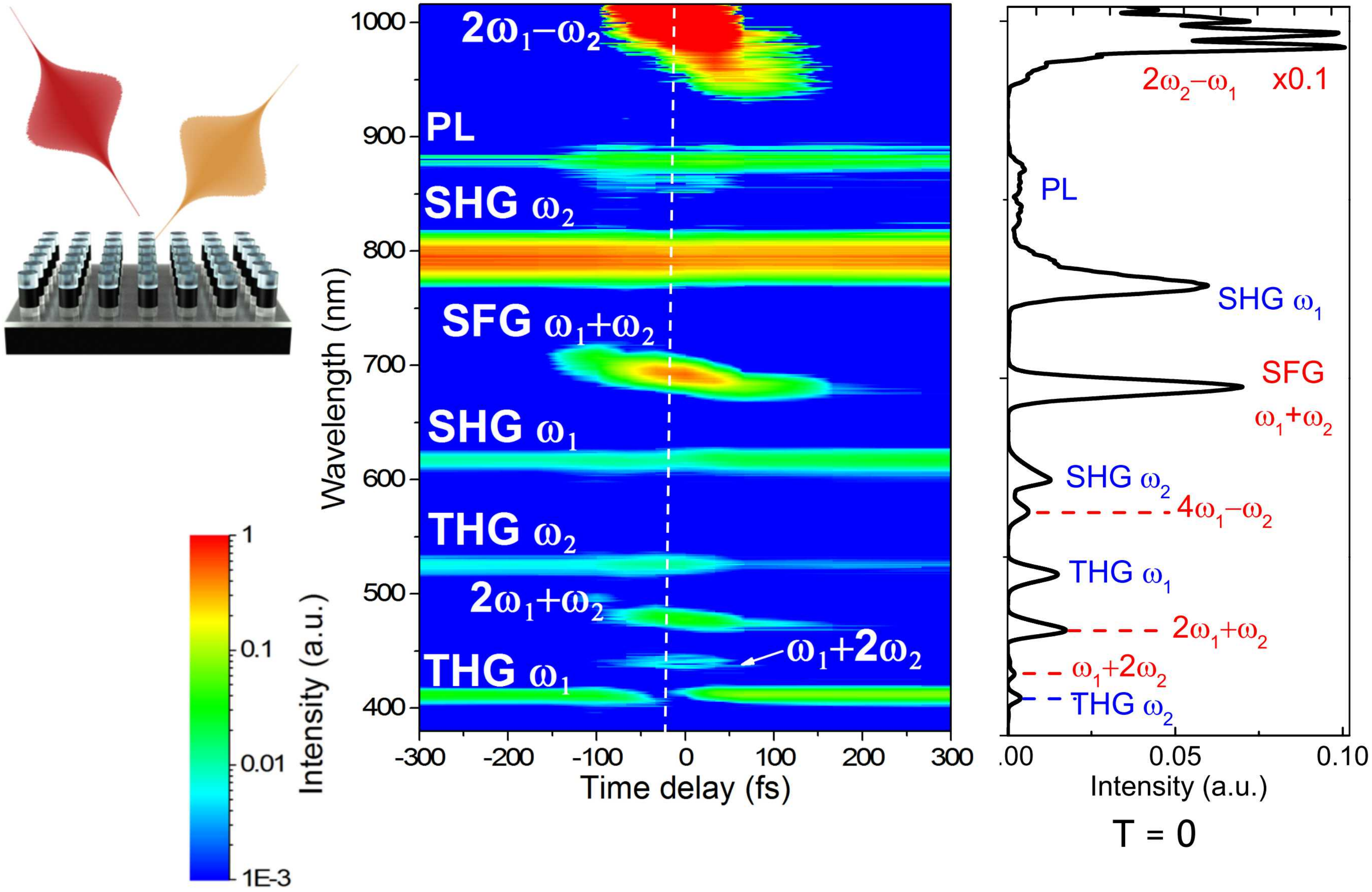




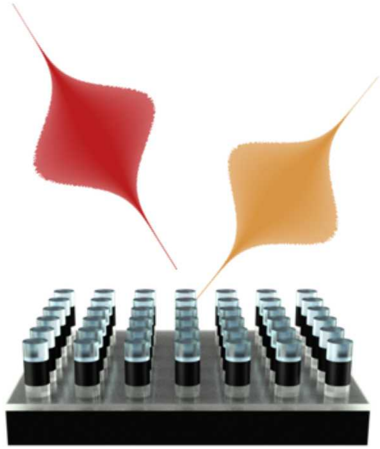
SWM power dependence

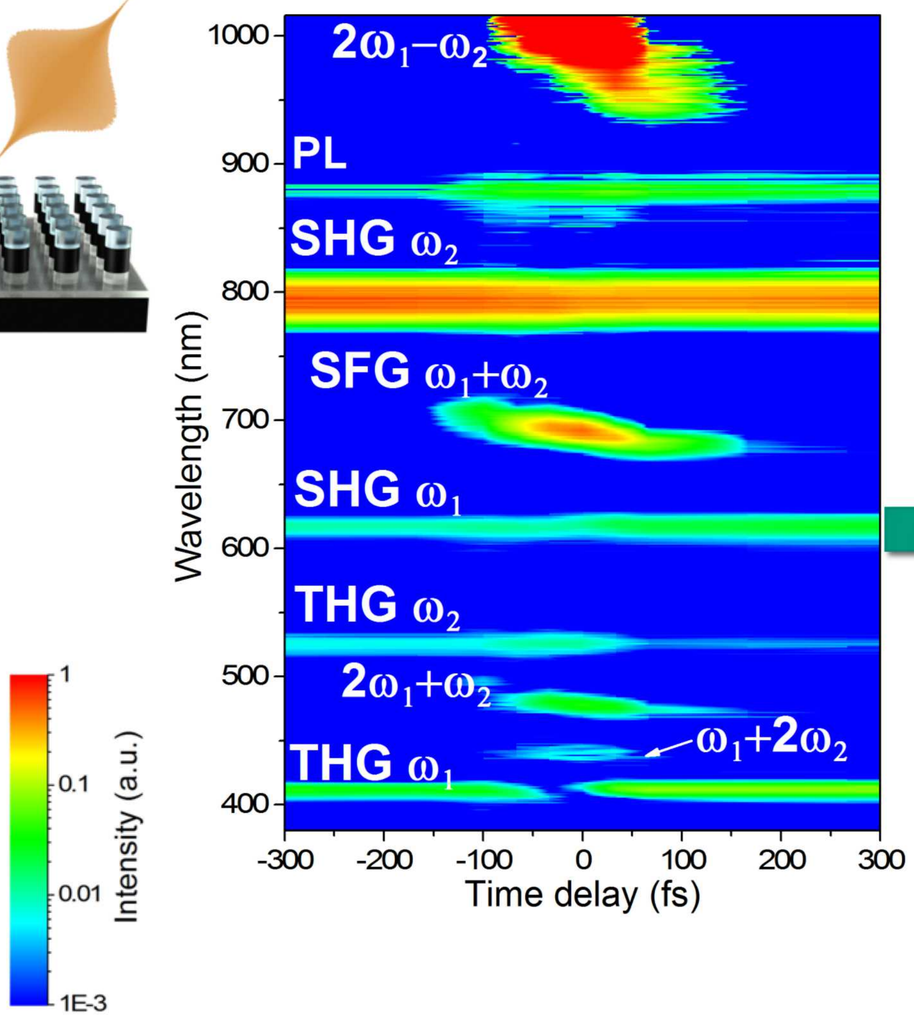


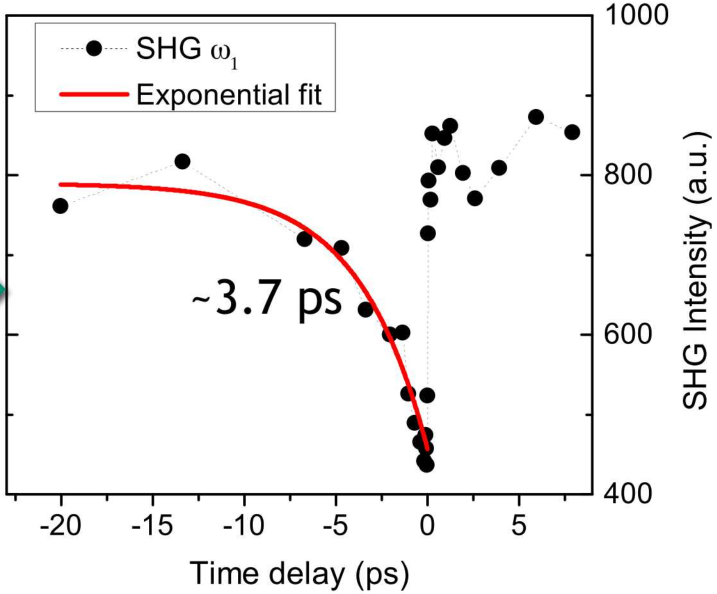
Temporal dynamics of frequency mixing



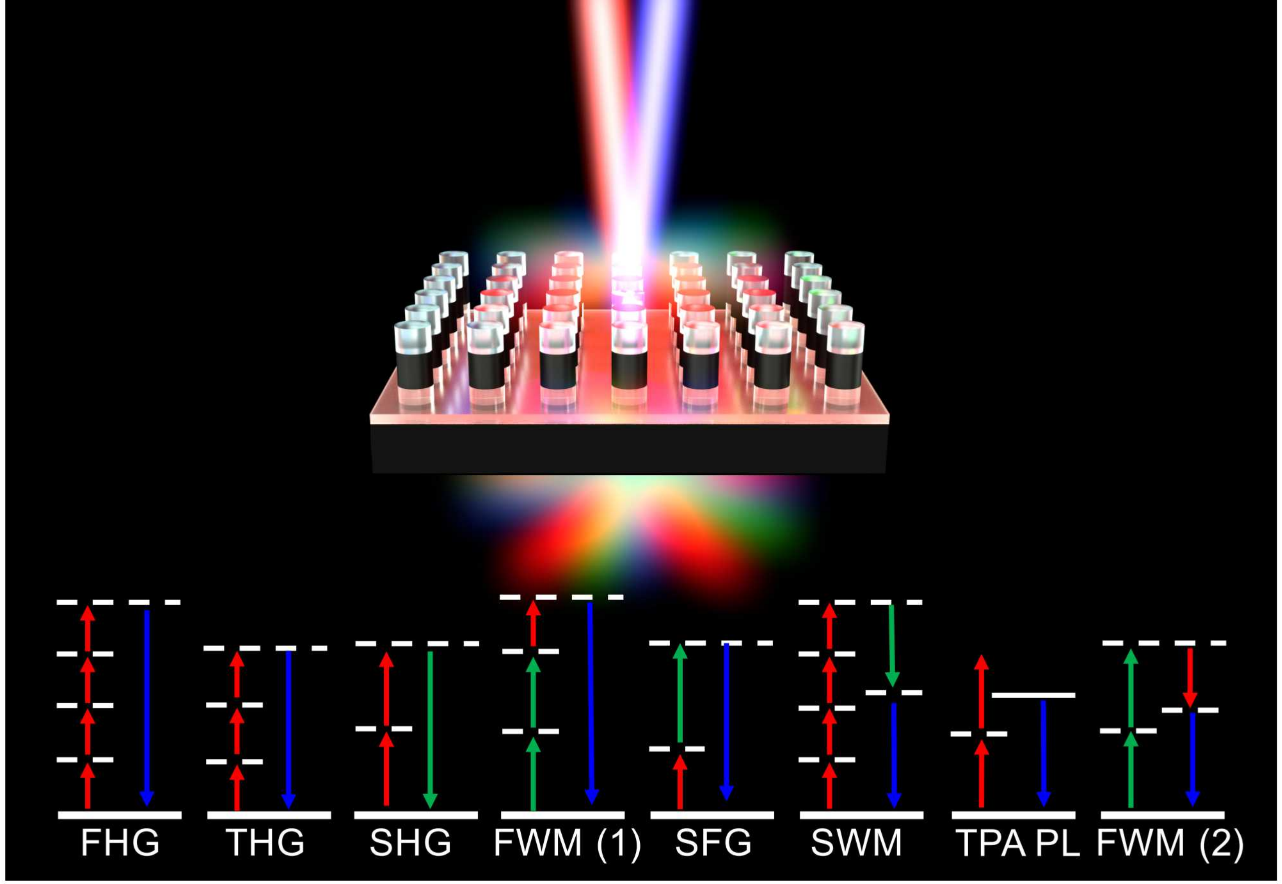
Temporal dynamics of frequency mixing







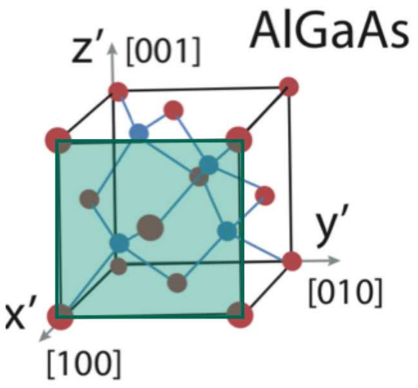
Shcherbakov, Maxim R., et al.
Nature communications 8.1 (2017): 17



- 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.
Nature communications 9 (1), 2507

GaAs (100) lattice plane

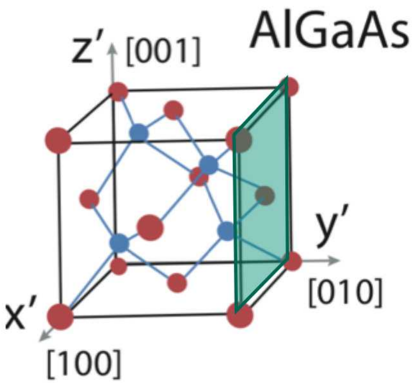


$$\begin{pmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{pmatrix} = 2\varepsilon_0 d_{eff} \begin{pmatrix} E_x(\omega)^2 \\ E_y(\omega)^2 \\ E_z(\omega)^2 \\ 2E_y(\omega)\cdot E_z(\omega) \\ 2E_x(\omega)\cdot E_z(\omega) \\ 2E_y(\omega)\cdot E_x(\omega) \end{pmatrix}$$

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

Absence of SHG in normal direction

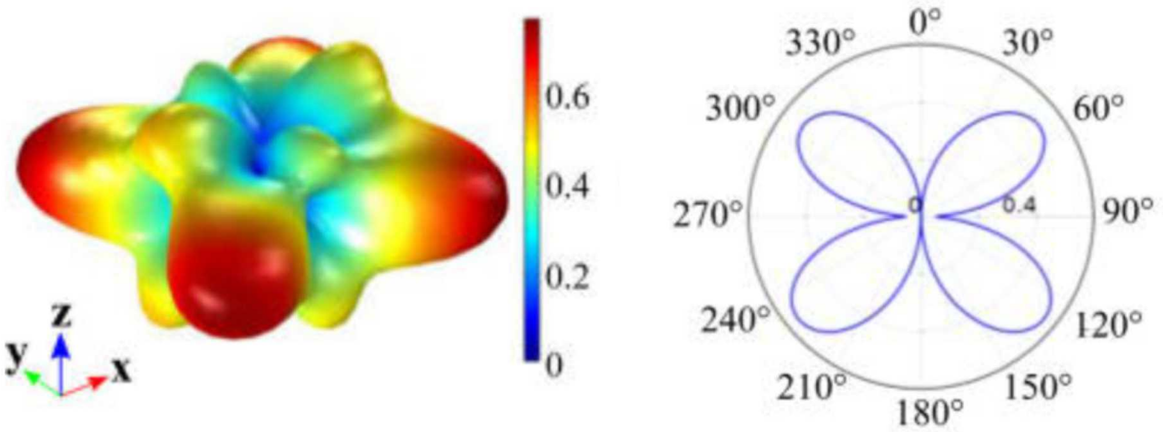
GaAs (100) single disk and metasurface emission patterns



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

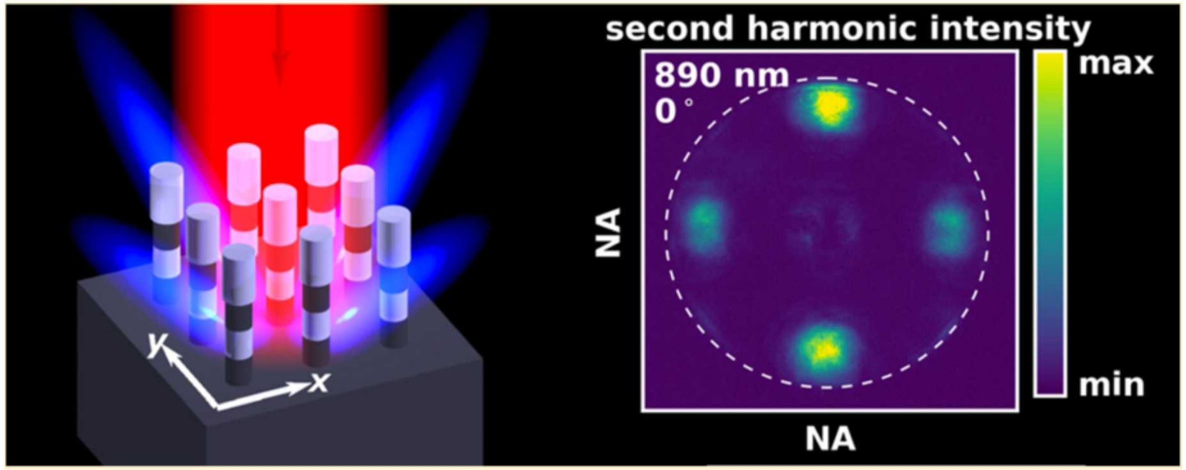
→ Absence of SHG in normal direction

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



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

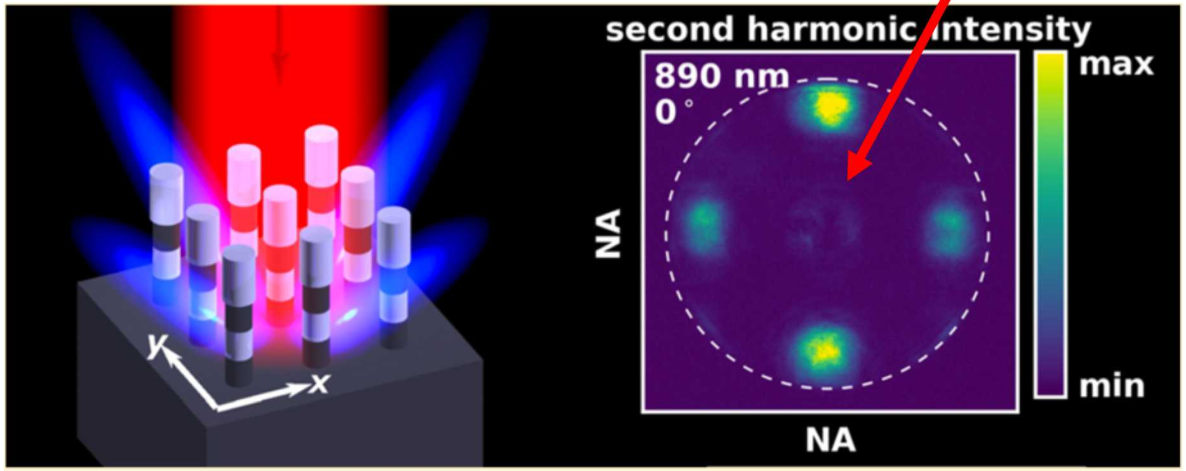
SHG diffraction in the GaAs (100) metasurface



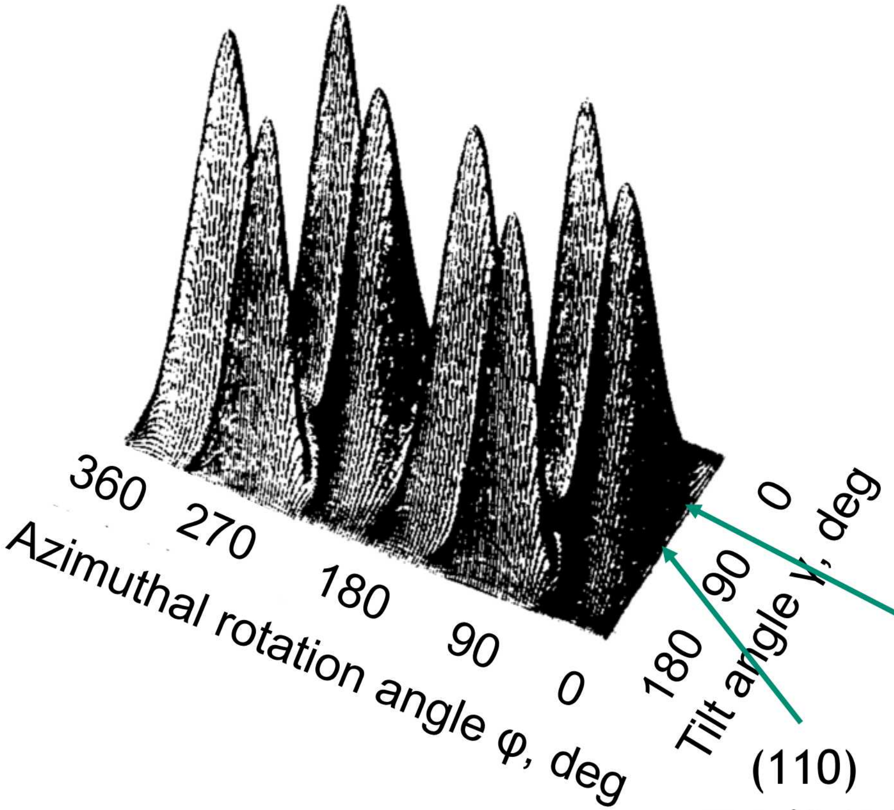
Löchner, F. J., *ACS Photonics*, 5(5), 1786-1793, **2018**

Absence of the zeroth-order

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

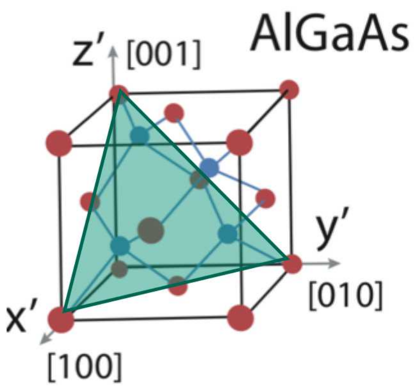


SHG in GaAs for different orientations of the crystal, (111), (110)



(110)

(111)



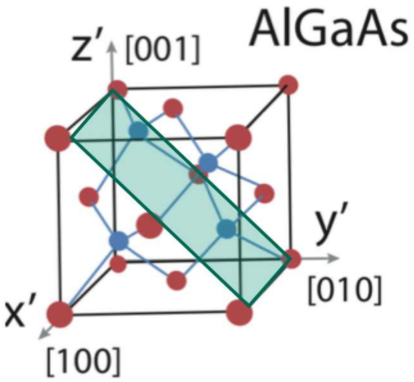
$$d_{eff} = \begin{pmatrix} -\frac{1}{\sqrt{6}} & \frac{1}{\sqrt{6}} & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 \\ 0 & 0 & 0 & -\frac{1}{\sqrt{3}} & 0 & -\frac{1}{\sqrt{6}} \\ -\frac{1}{2\sqrt{3}} & -\frac{1}{2\sqrt{3}} & \frac{1}{\sqrt{3}} & 0 & 0 & 0 \end{pmatrix} \cdot d_{41}$$

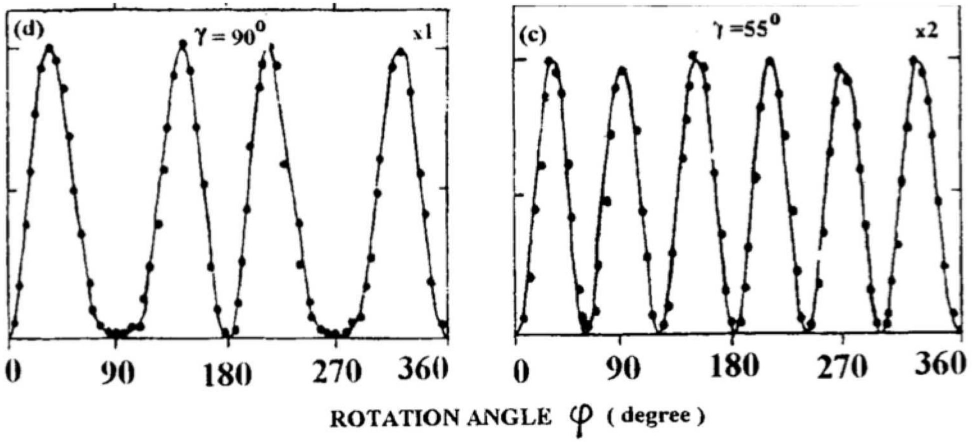
Non-zero SHG signal for under normal incidence

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

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

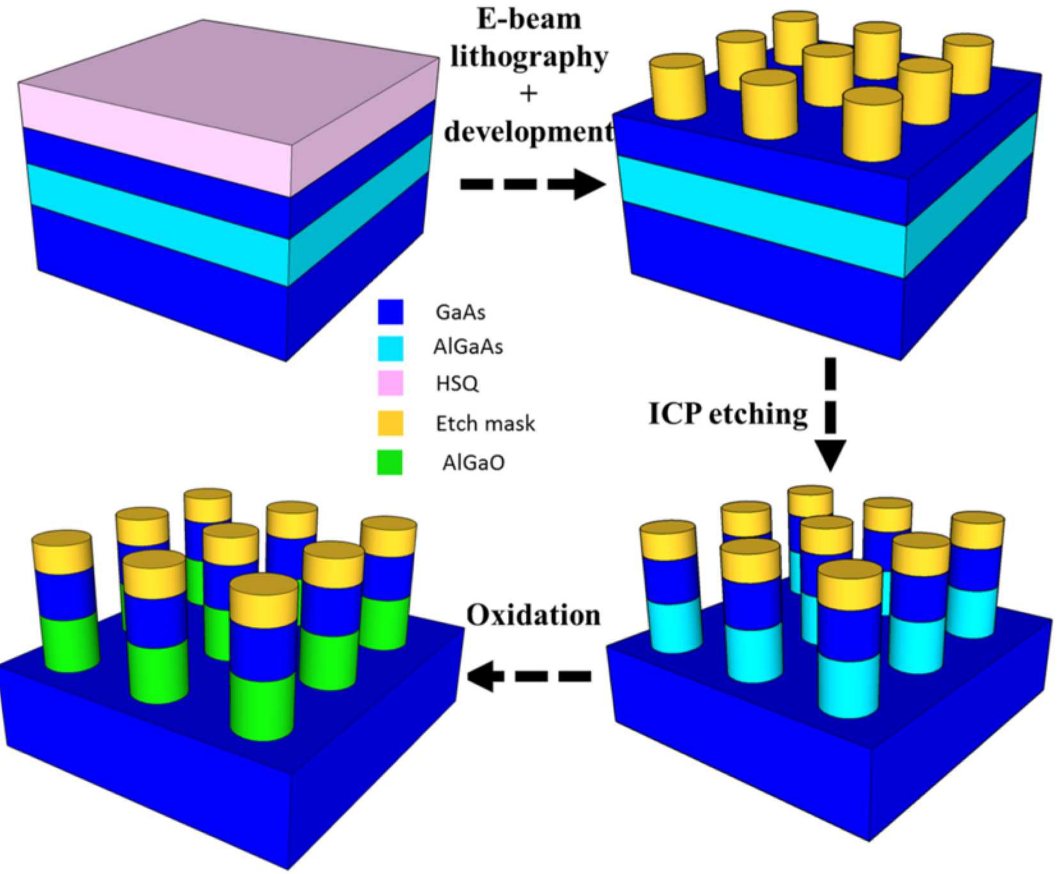
$$P_z(2w) = 2\varepsilon_0 d_{eff}(-\frac{1}{2\sqrt{3}})E_x(w)^2 - \frac{1}{2\sqrt{3}}E_y(w)^2 - \frac{1}{1\sqrt{3}}E_z^2$$





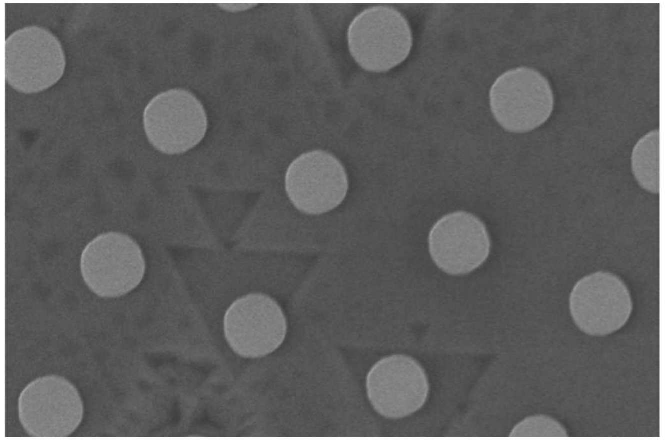
Kulyuk, L L., *Nonlinear Optics of Low-Dimensional Structures and New Materials*. Vol. 2801. International Society for Optics and Photonics, **1996.**

GaAs (111) metasurface: reflectance spectra

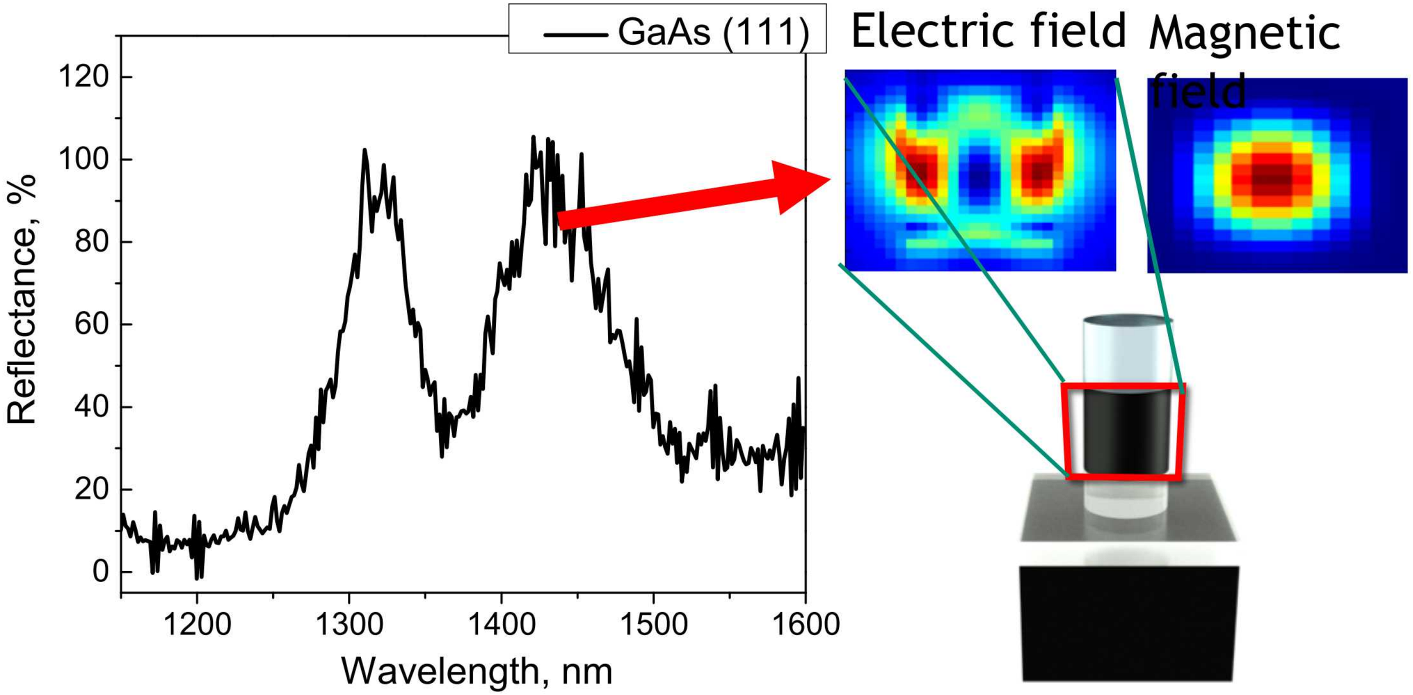


Liu, S. et. al. *Nano letters* 16.9: 5426-5432, **2016**

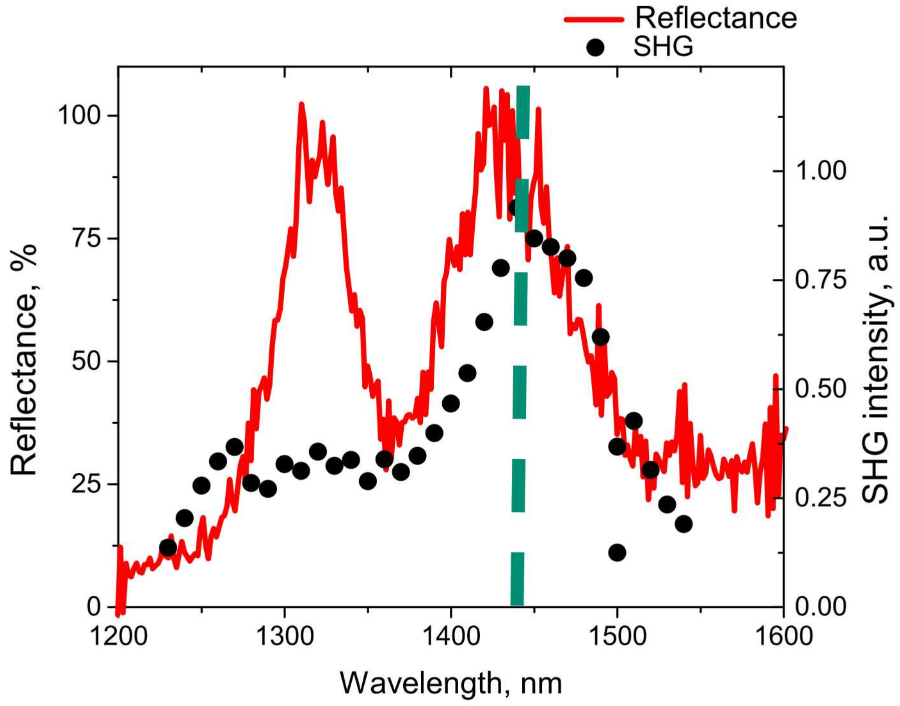
SiO_x – 400 nm
$Al_{0.1}Ga_{0.9}As$ – 400 nm
$(Al_xGa_{1-x})_2O_3$ – 500 nm
Diameter – 340 nm
Period – 1200 nm



Reflectance spectrum

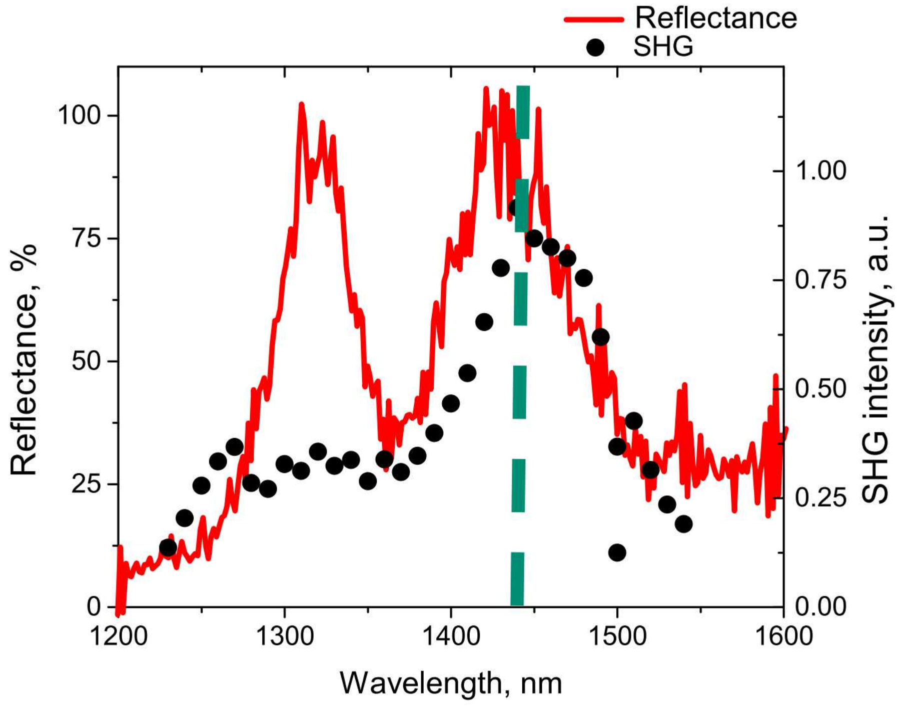


SHG in GaAs (111) metasurface

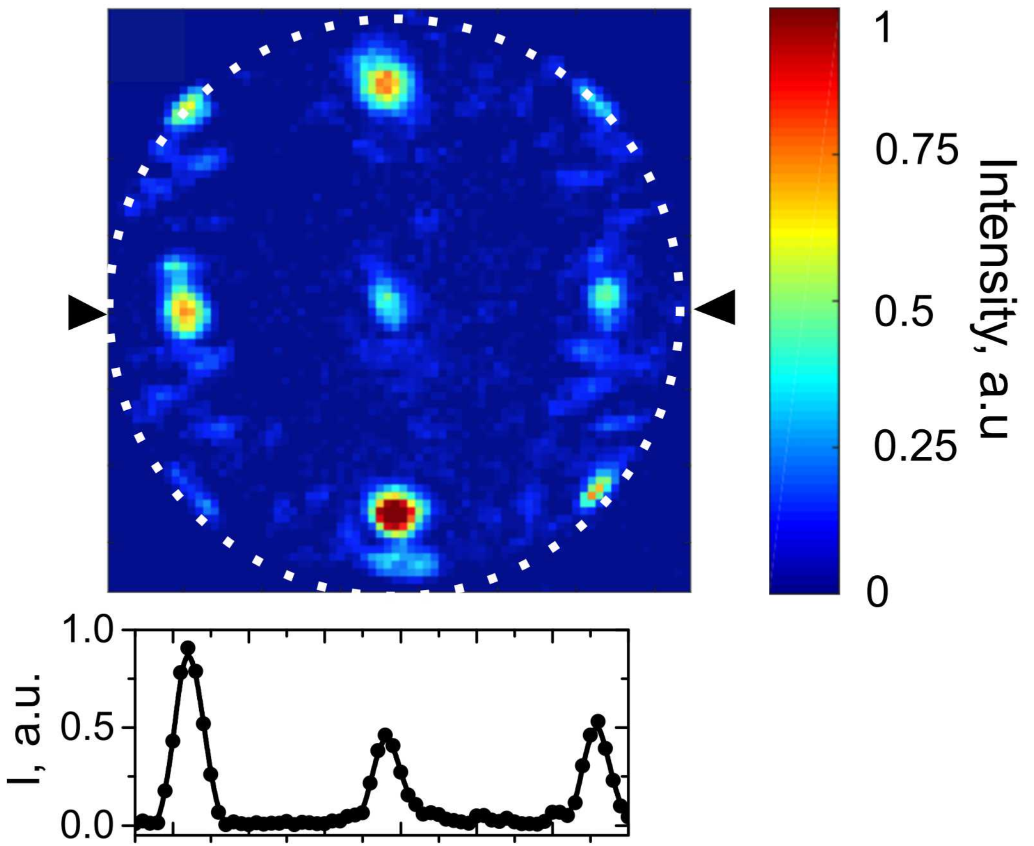


SHG enhancement at the wavelength of MD resonance

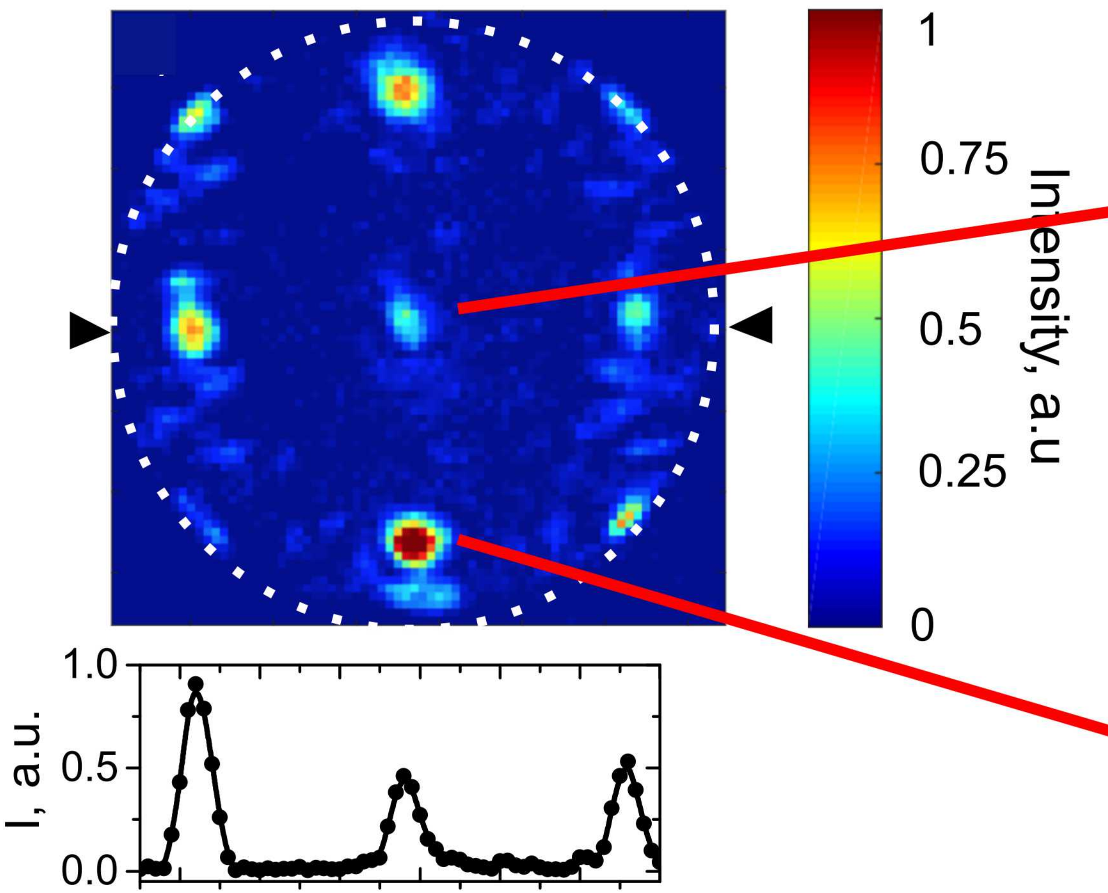
SHG in GaAs (111) metasurface: Fourier-space image



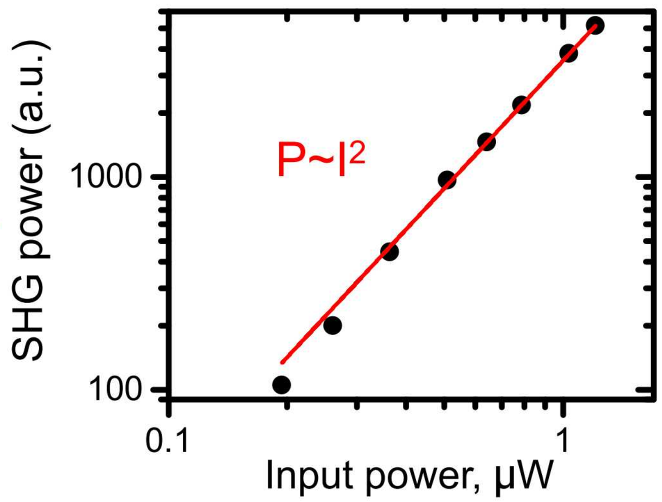
SHG enhancement at the wavelength of MD resonance



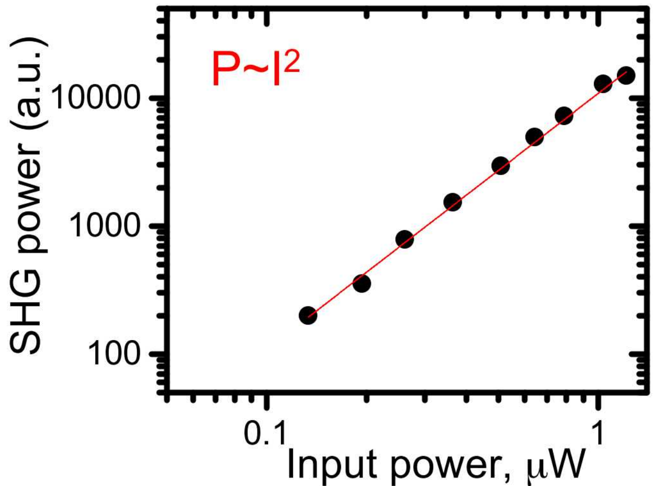
SHG in GaAs (111) metasurface: Fourier-space image



Power dependence for the zeroth-order diffraction

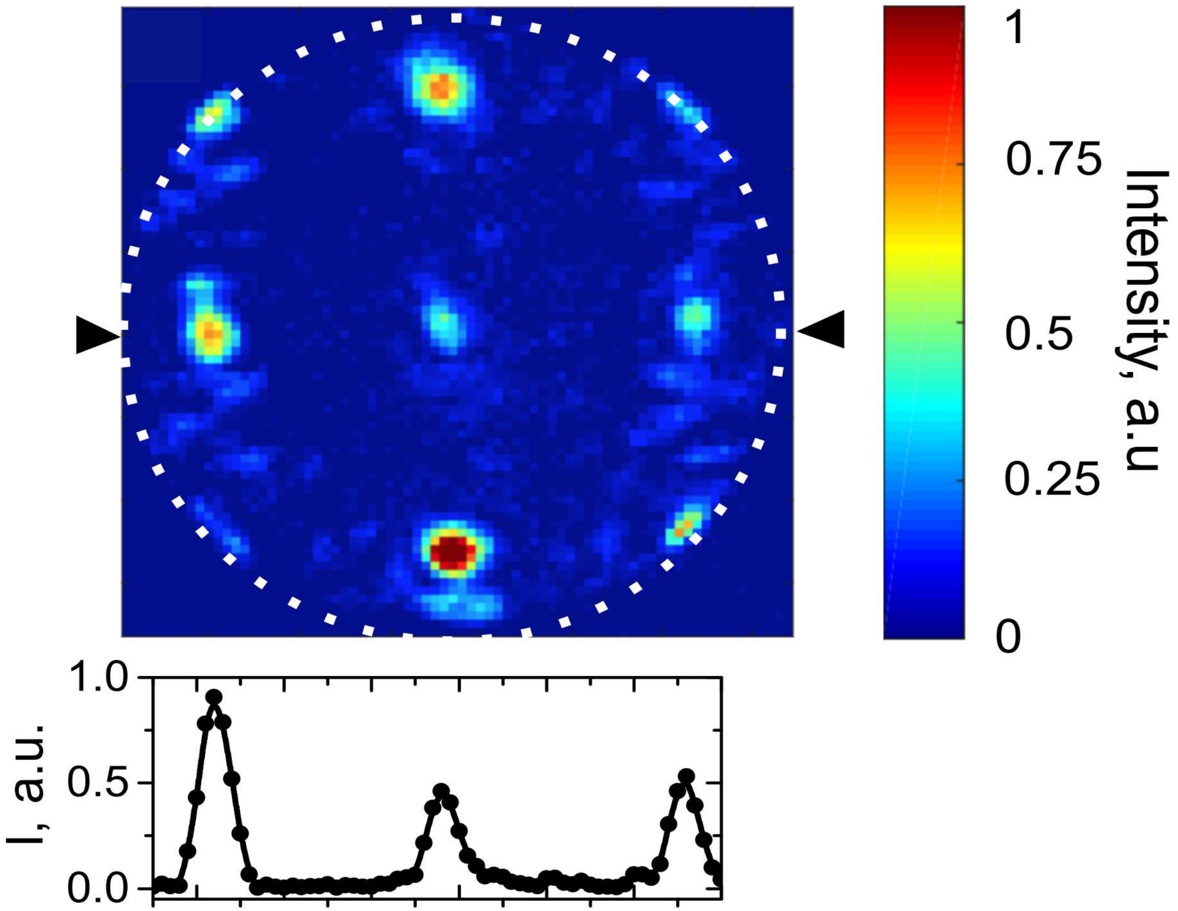


Power dependence for the bottom diffraction peak

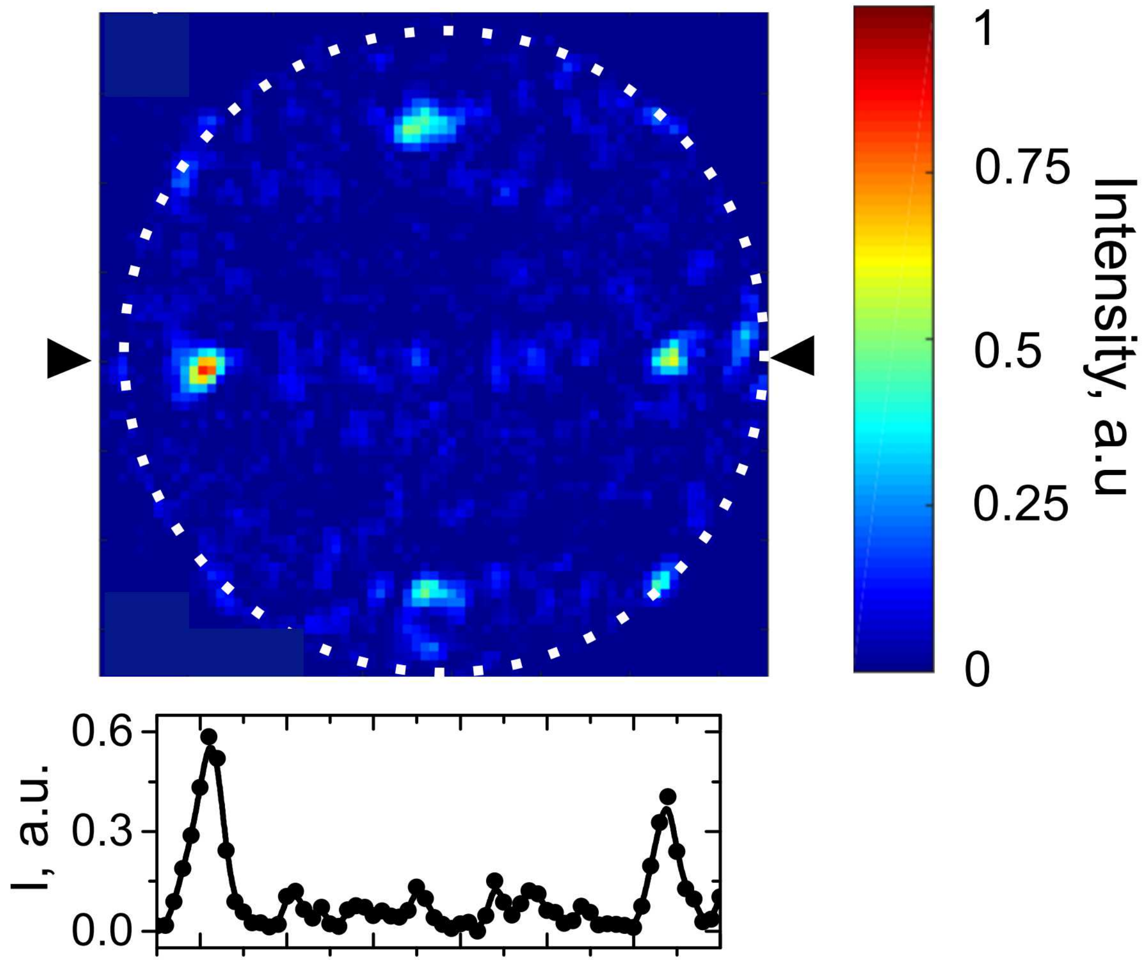


Comparison: Fourier-space images for metasurfaces with different crystallographic planes

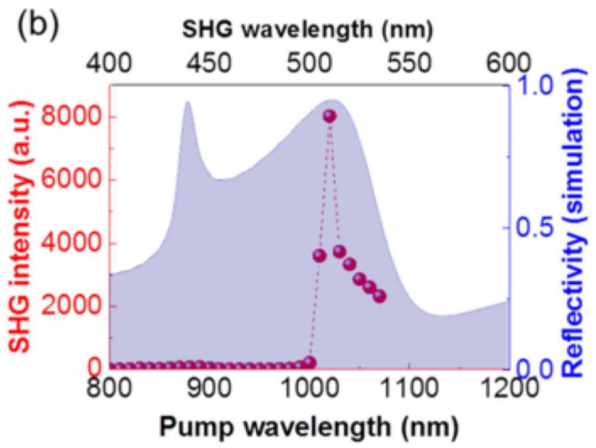
(111)

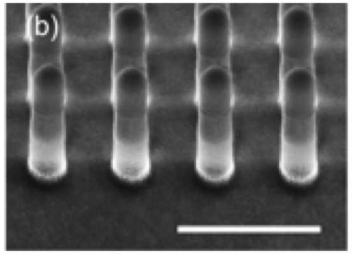


(100)



SHG in GaAs metasurfaces

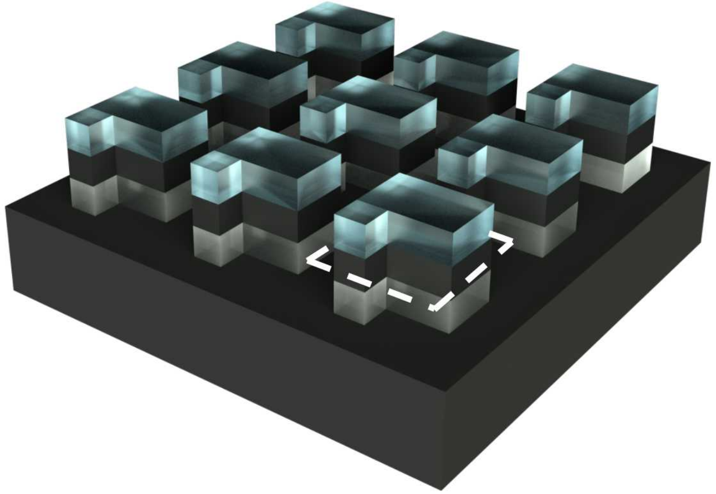




Efficiency: power of generated SHG/pump power

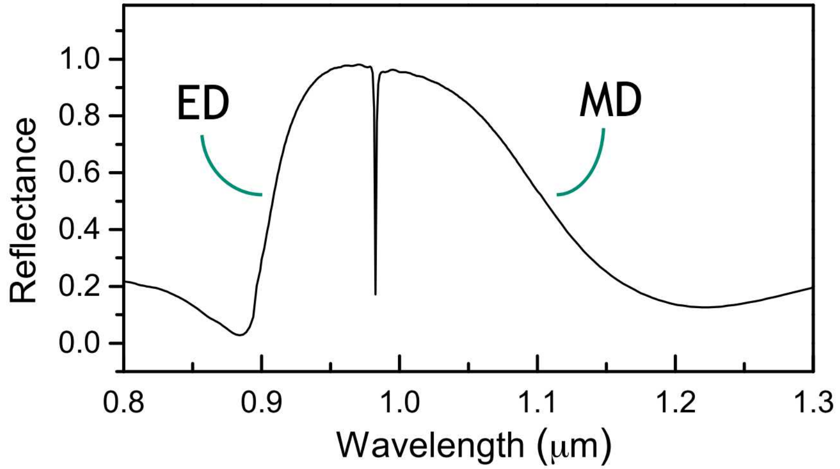
Carletti, L., et al. *Optics express* 23.20, 26544-26550**, 2015**
Camacho-Morales, R., et al. *Nano letters* 16.11, 7191-7197, **2016**
Kruk, S.et al. *Nano Lett.*, *17* (6), pp 3914–3918, **2017**
Liu, S. et. al. *Nano letters* 16.9, 5426-5432, **2016**

Symmetry-broken metasurface:
High-Q resonances



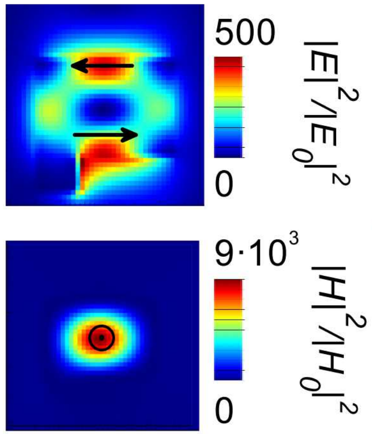
Metasurface with broken symmetry design resonators: High-Q factor resonance

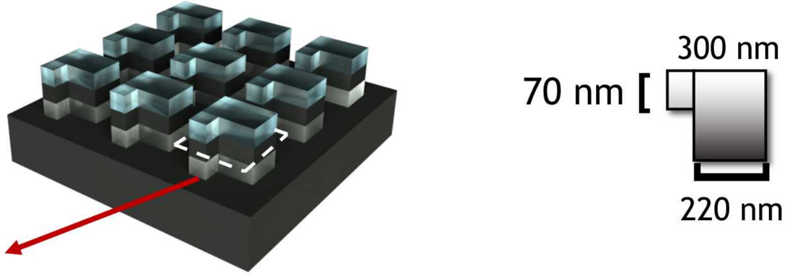
FDTD simulations



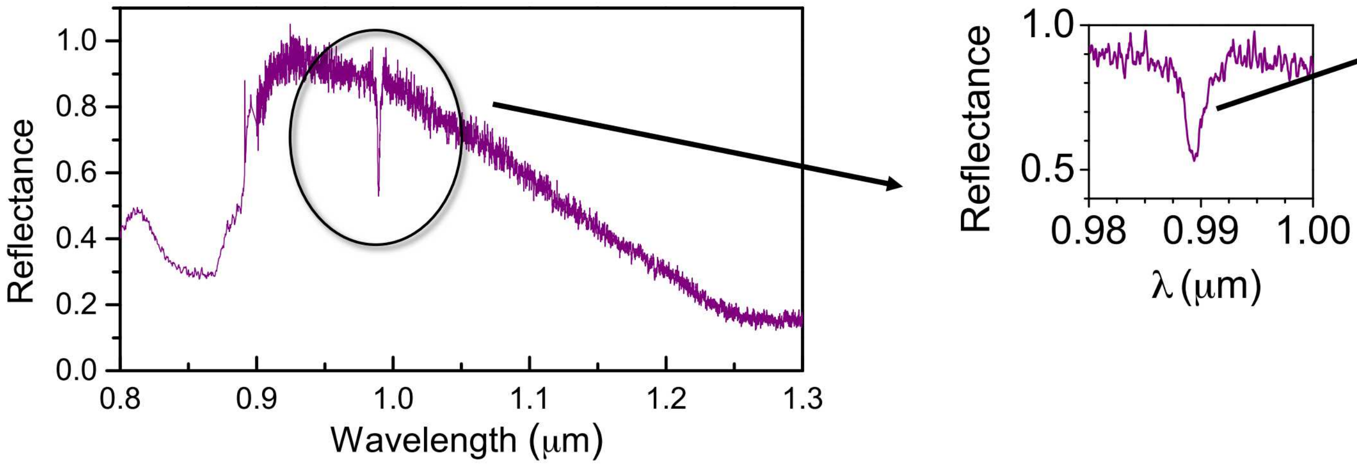
Fano resonance

$\lambda_R \sim 0.98$ nm





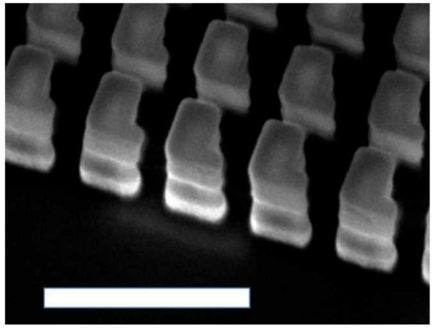
Experimental measurements



$\Delta\lambda \sim 2$ nm

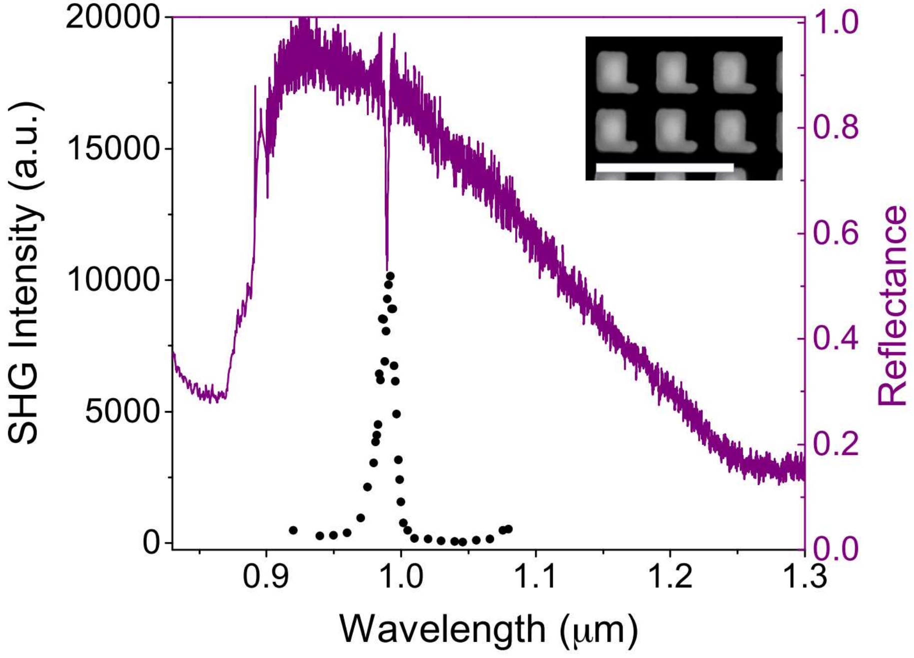
Q factor

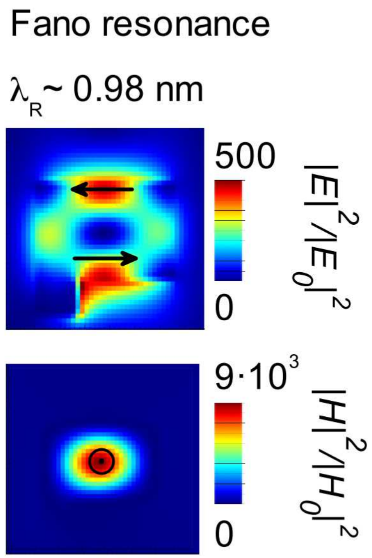
$\lambda_F/\Delta\lambda \sim 500$

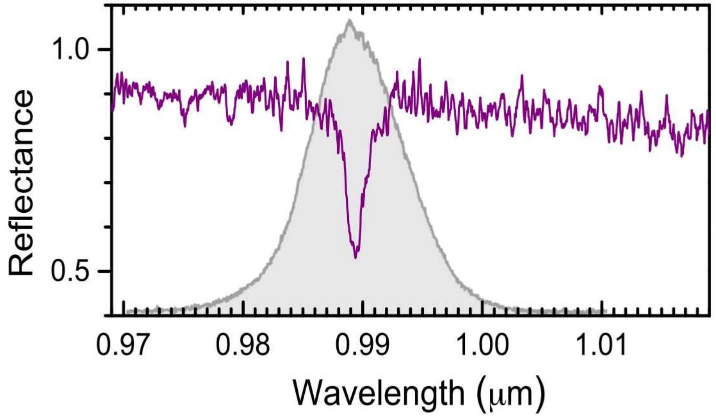


SHG in broken symmetry design metasurface

SHG intensity enhancement in the vicinity of the High Q resonance x300







~30% of pump power is used

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

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

Outline

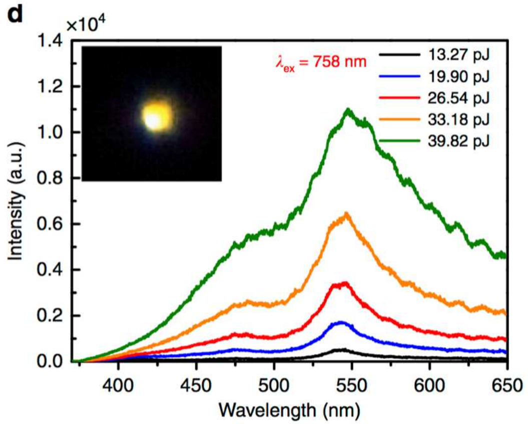
Nonlinear optics in GaAs metasurfaces

- Frequency mixing
- SHG with GaAs (111) metasurface into zeroth diffraction order
- SHG in Fano metasurface

Emission engineering in semiconductor metasurfaces

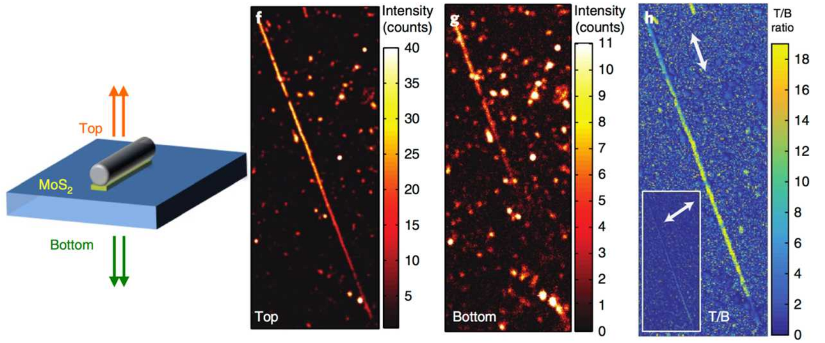
Emission + All-dielectric Metasurfaces: Recent work

White-light emission from silicon nanospheres



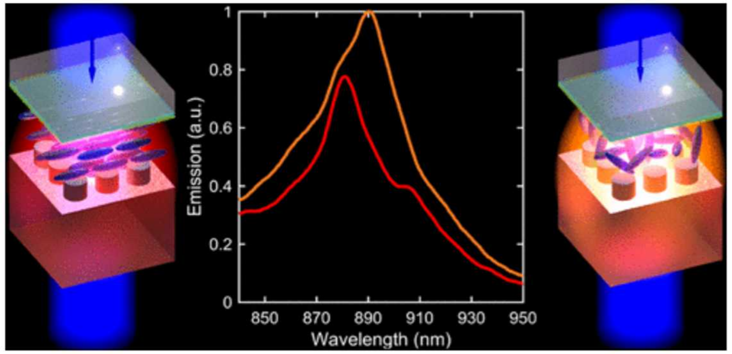
Zhang, C., et al.
Nature communications 9 (2018).

Directional light emission from monolayer MoS2



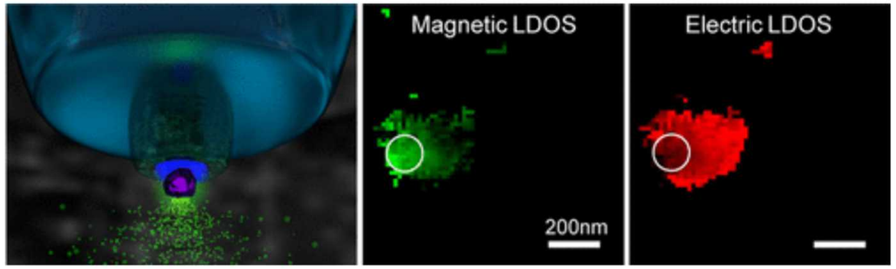
Cihan, A. F., et al.
Nature Photonics 12.5 (2018): 284.

Active Tuning of Spontaneous Emission by Mie-Resonant Dielectric Metasurfaces



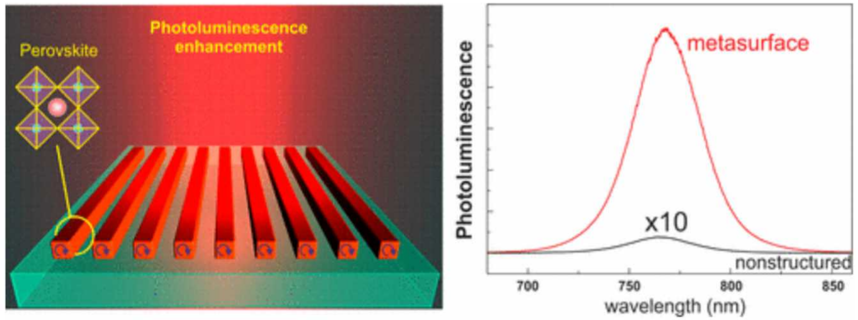
Bohn, J., et al. *Nano letters* 18.6 (2018): 3461-3465.

Enhanced emission of luminescent nanocrystals



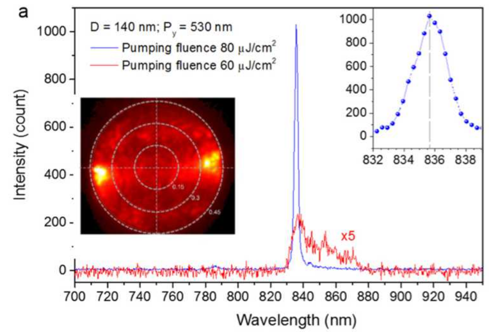
Sanz-Paz, M., et al.
Nano Letters 18.6 (2018): 3481-3487.

Emission enhancement in hybrid perovskite metasurfaces



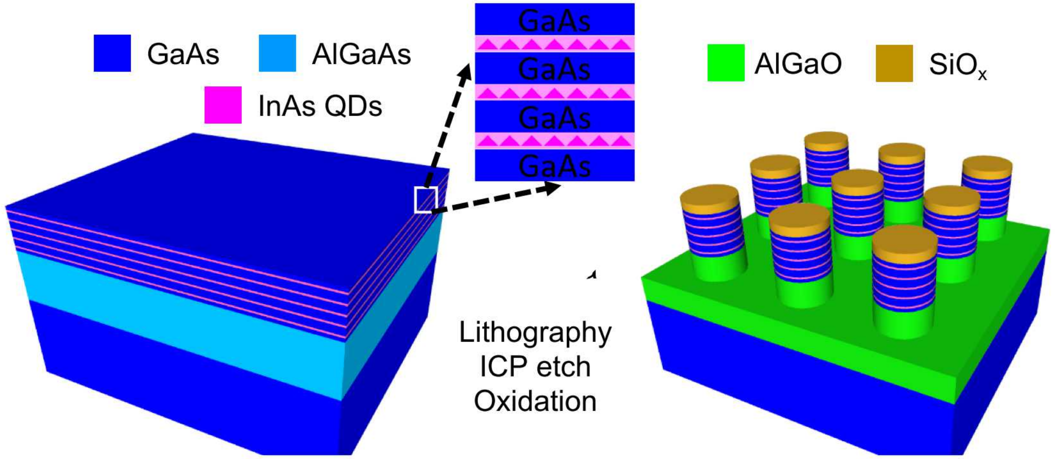
Makarov, S. V., et al. *ACS Photonics* 4.4 (2017): 728-735.

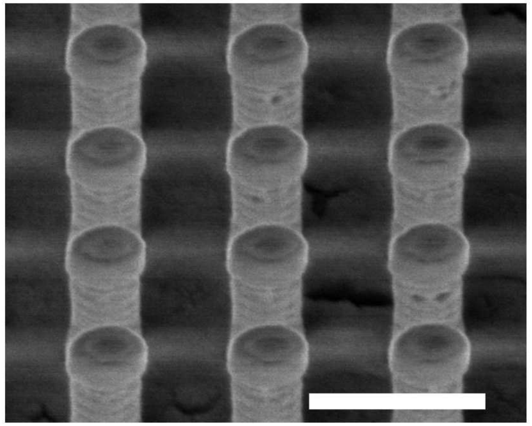
Lasing action in active dielectric nanoantenna arrays



Ha, S. T., et al. *Nature Nanotech* (2018).

Embedding Emitters in Metasurfaces

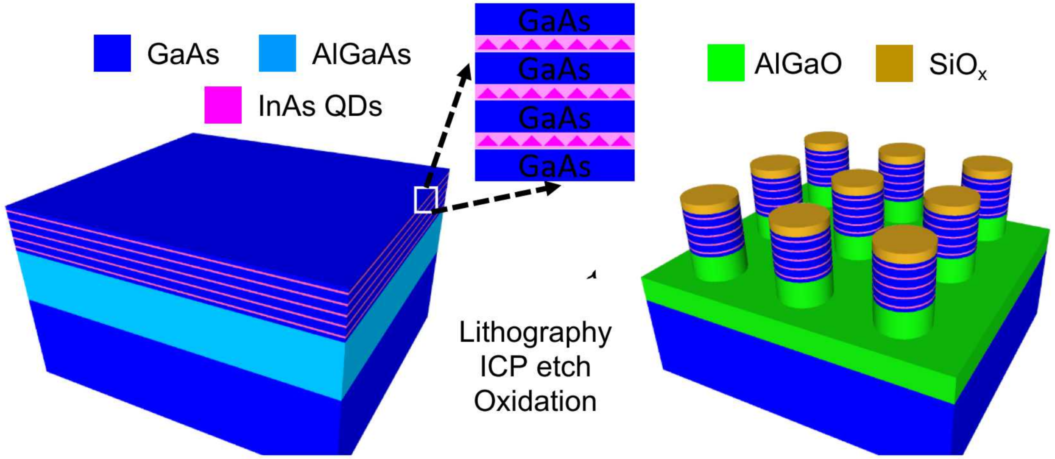


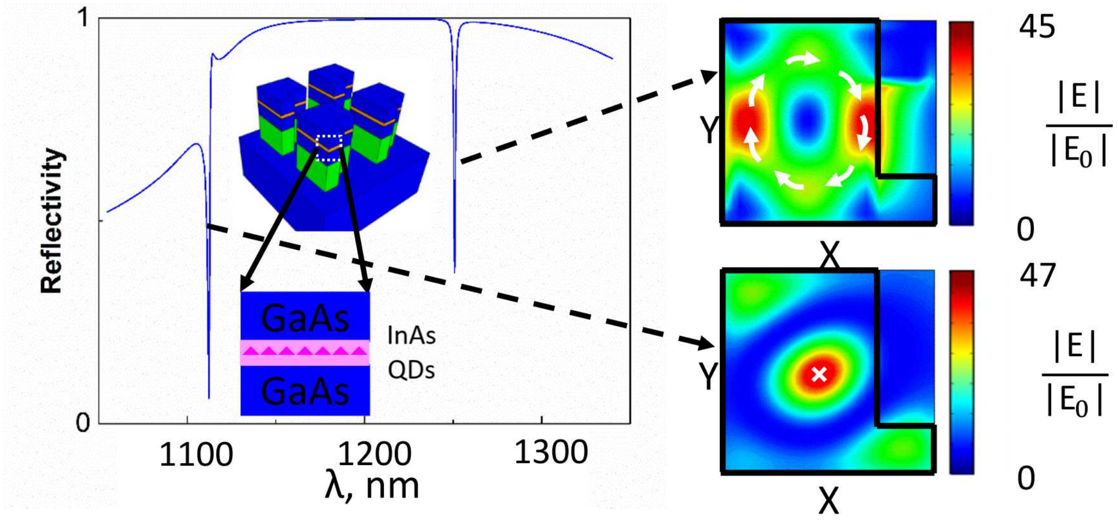


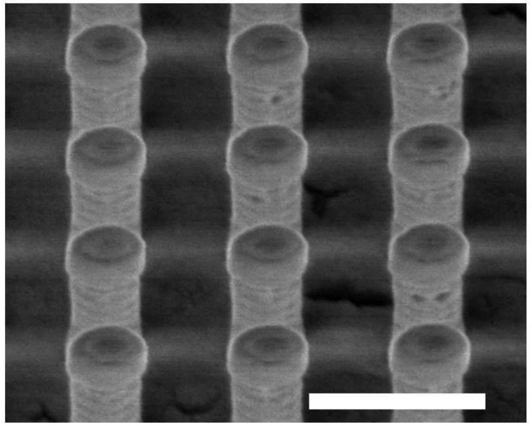
1 μm

Liu. et al. Nano Lett. 18 (11) p.6906-6914 (2018)

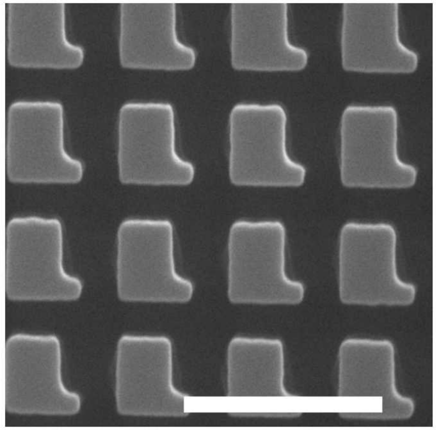
Embedding Emitters in Metasurfaces







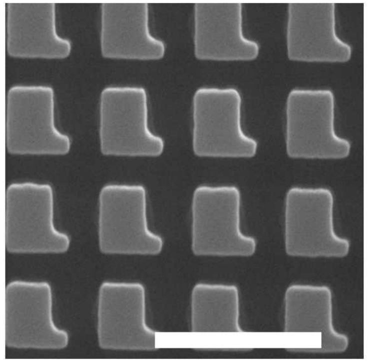
1 µm

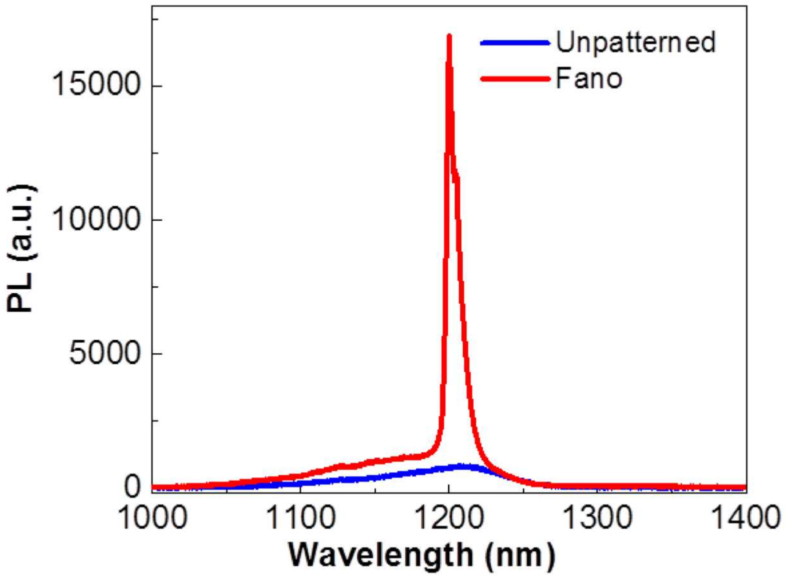


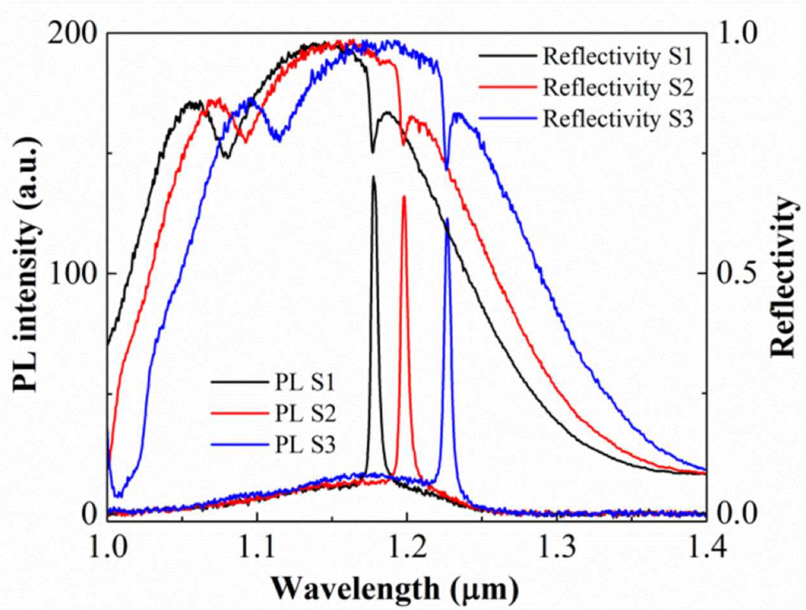
1 µm

Liu. et al. Nano Lett. 18 (11) p.6906-6914 (2018)

Light Emitting Metasurfaces: Quantum Dot emission enhancement





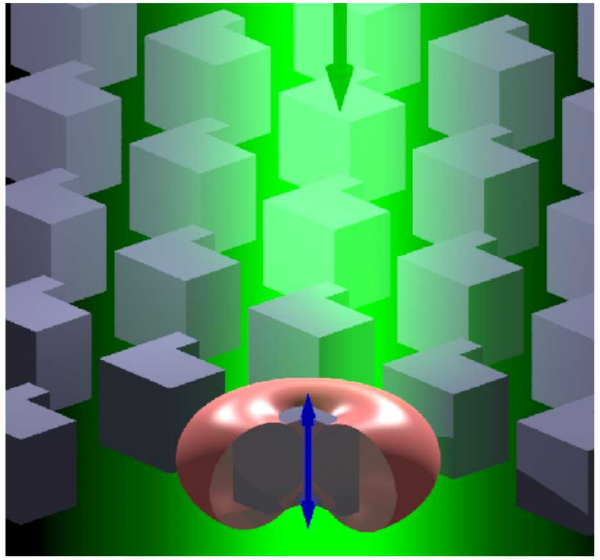


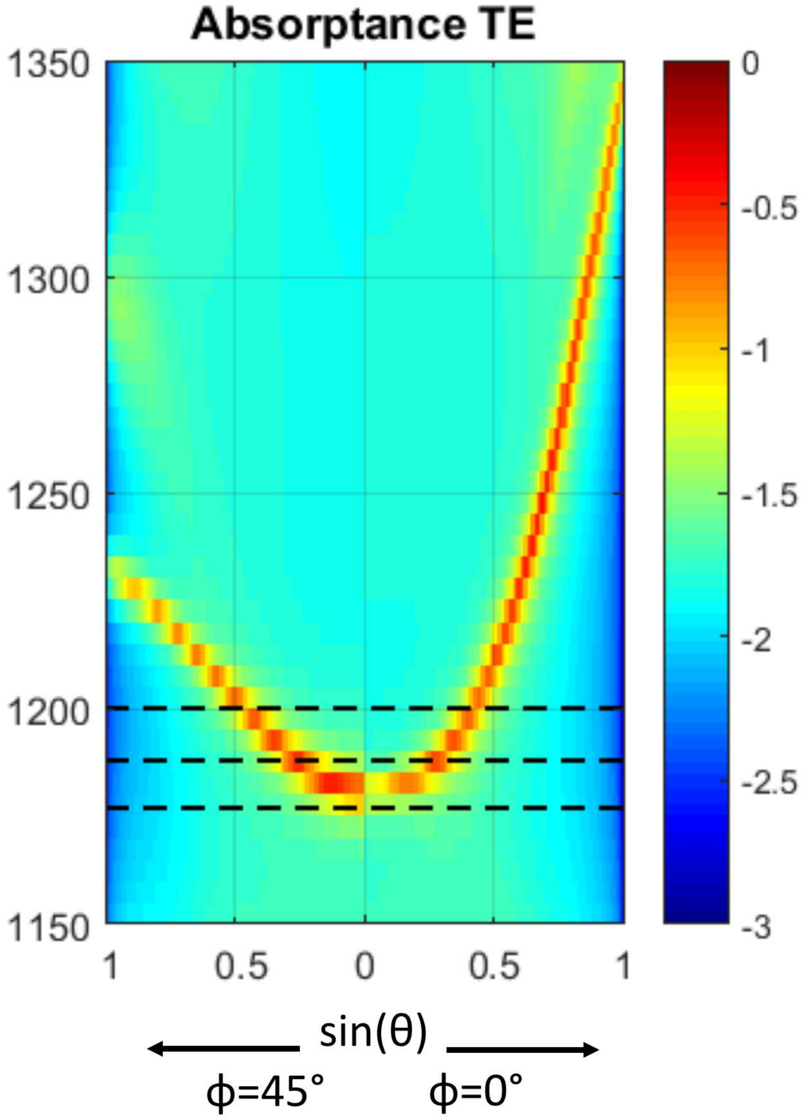
PL spectrum, obtained when Fano resonance overlaps with the maxima of QD PL band, shows ~110-fold enhancement of the PL peak intensity in comparison with unpatterned sample.

Liu. et al. Nano Lett. 18 (11) p.6906-6914 (2018)

Emission with Out of Plane Dipole Modes: angular dependence

Broken-symmetry Resonators



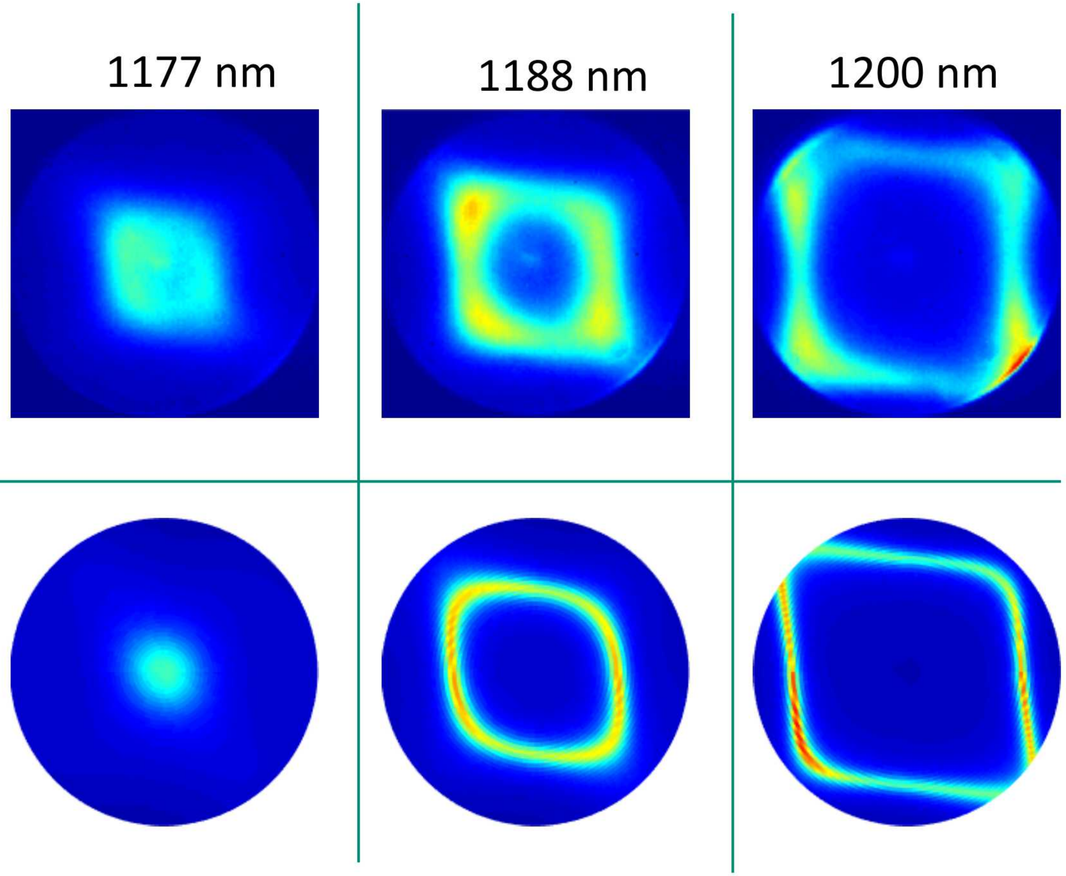


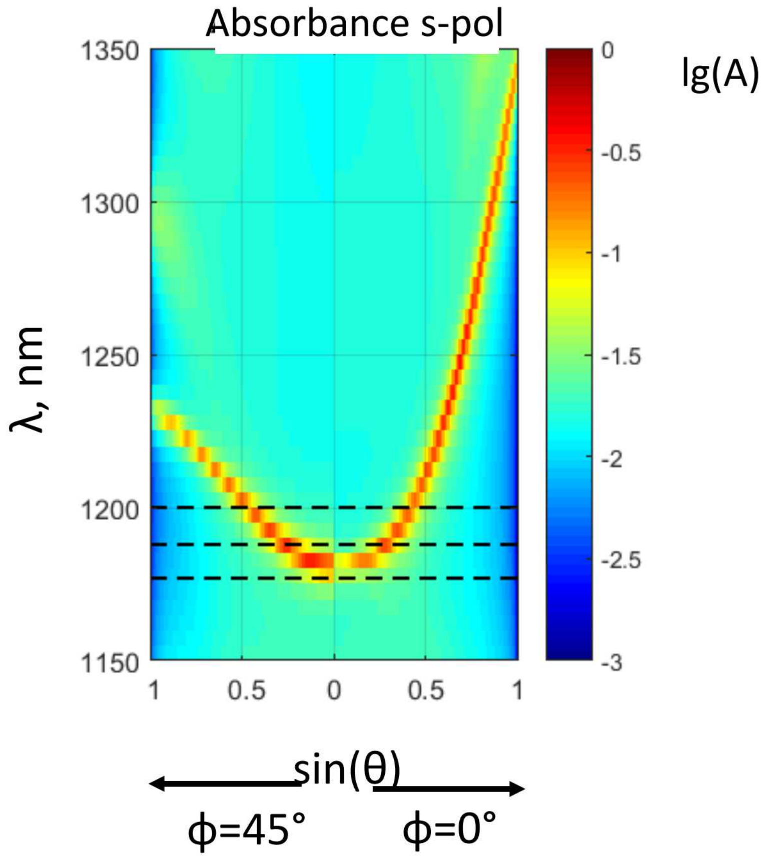
Liu. et al. Nano Lett. 18 (11) p.6906-6914 (2018)

Controlling Far-Field Angular Spread

Measured Fourier-space images

Numerically calculated





By changing the array design, we can change the angular distribution of the PL in the far-field

Liu. et al. Nano Lett. 18 (11) p.6906-6914 (2018)

Conclusions

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

- 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.
- By utilizing symmetry broken design of III-V semiconductor metasurface we were able to achieve 13 fold enhancement of the second-harmonic generation.
- By controlling the symmetry of the resonant modes in the Mie-resonant metasurfaces made from semiconductor heterostructures containing epitaxial quantum dots we were able to enhance the PL brightness by two orders of magnitude.