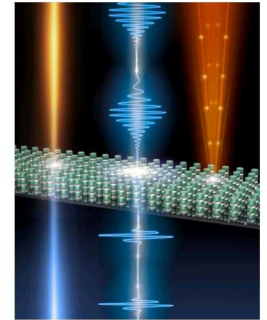
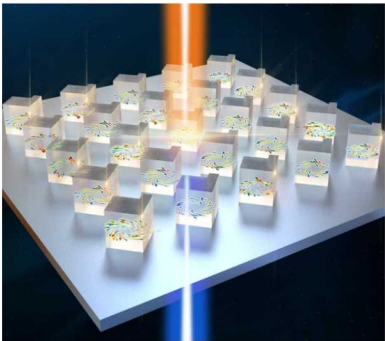




Recent Advances in Linear and Nonlinear All-Dielectric Metamaterials



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SPIE 2018, San Diego

Sandia National Laboratories is a multi mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia L.L.C., a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525



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Jena (Germany): Isabelle Staude, Aleks Vaskin

UT Austin: Nish Nookala, Misha Belkin

Moscow State (Russia): Maxim Shcherbakov, Andrey Fedyanin

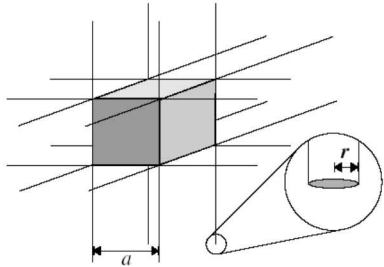


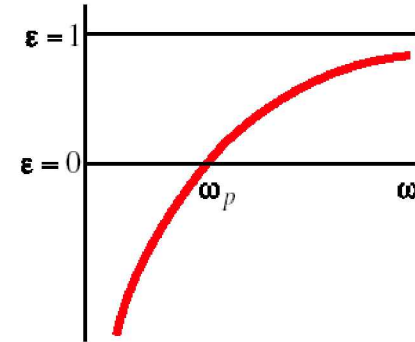
Outline

- **Metamaterials & Metasurfaces: refresher, definitions, etc.**
- **Linear Metasurfaces:**
 - Optical magnetic mirror
 - Huygens meta-optics
- **Emitters + Metasurfaces**
- **Nonlinear optics with all-dielectric metasurfaces:**
- **Outlook on future 3D fabrication**

Refresher: Metamaterials

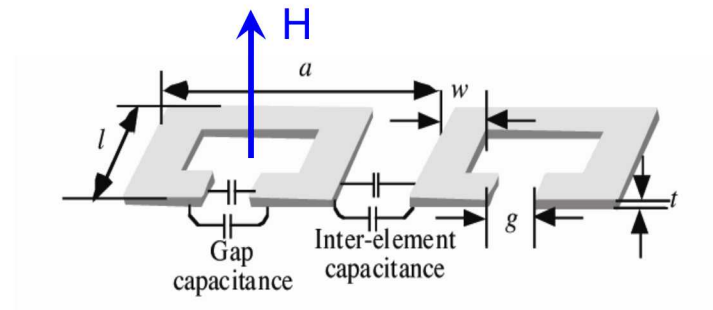
Controlling ϵ

$$\omega_p = \sqrt{\frac{ne^2}{m^* \epsilon_0}}$$


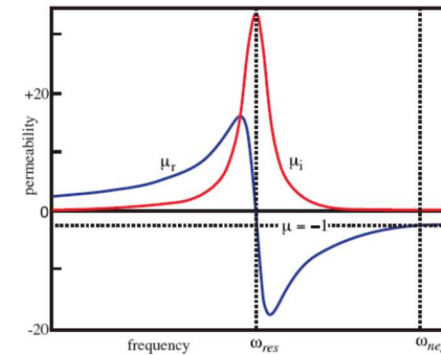


“Diluted Drude” or
some electric resonance

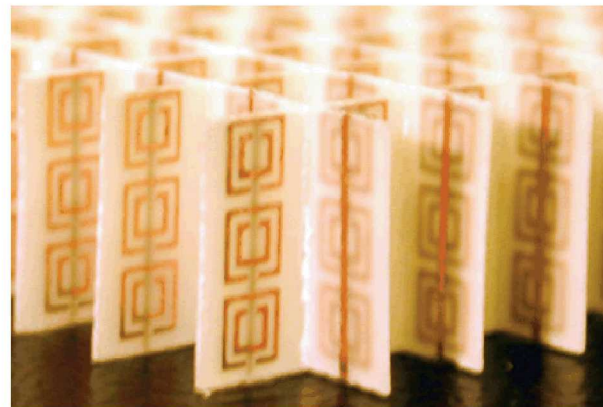
Controlling μ



from; S. McMeekin, Proc. Of SPIE, Vol. 6581



Some magnetic
resonance

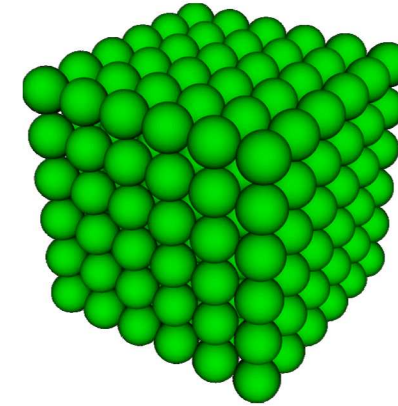


Dielectric Metamaterials

Clausius-Mossoti or
Lorentz-Lorenz

Effective medium approximation

$$\epsilon_{\text{eff}} = \epsilon_m \frac{2\delta_i(\epsilon_i - \epsilon_m) + \epsilon_i + 2\epsilon_m}{2\epsilon_m + \epsilon_i + \delta_i(\epsilon_m - \epsilon_i)}$$



3. Zur Optik kolloidaler Metallösungen; von R. Gans und H. Happel.

Annalen der Physik, 1909

ARTICLES*

(Ack: Ed Kuester,
CU Boulder)

RIAL LOADED WITH

Setzen wir (40'), (41) und (43) in die Gleichungen (44) und (45) ein, so ergibt sich die komplexe Dielektrizitätskonstante und Permeabilität der kolloidalen Lösung

sed form 27th September, 1946.)

$$(46) \quad \epsilon = \epsilon_0 \frac{1 + \frac{4\pi N}{3} \left(\frac{\lambda'}{2\pi}\right)^3 (a_1 - a_2)}{1 - \frac{2\pi N}{3} \left(\frac{\lambda'}{2\pi}\right)^3 (a_1 - a_2)}$$

$$(47) \quad \mu = \frac{1 - \frac{4\pi N}{3} \left(\frac{\lambda'}{2\pi}\right)^3 p_1}{1 + \frac{2\pi N}{3} \left(\frac{\lambda'}{2\pi}\right)^3 p_1}$$

$$F(\theta) = \frac{2(\sin \theta - \theta \cos \theta)}{(\theta^2 - 1) \sin \theta + \theta \cos \theta}$$

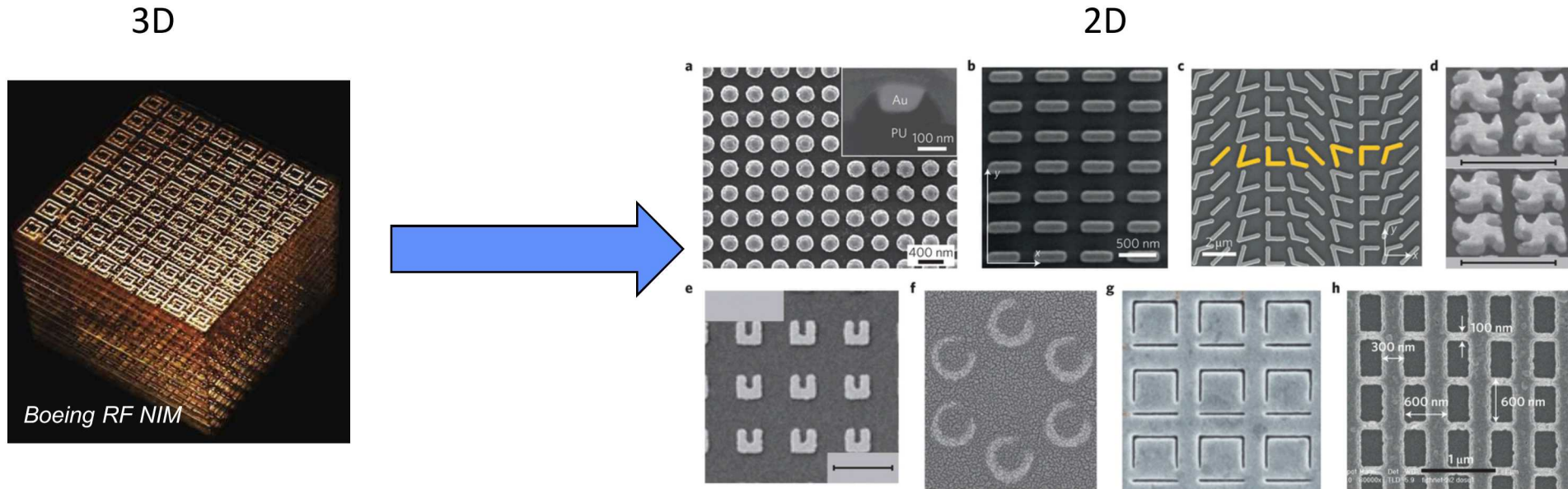
$$\theta = k_0 a \sqrt{\epsilon'_{r2} \mu'_{r2}}$$

$$b_e = \frac{\epsilon_1}{\epsilon_2}, \quad b_m = \frac{\mu_1}{\mu_2}$$

What is a “Metasurface”

The 2D version of a 3D Metamaterial

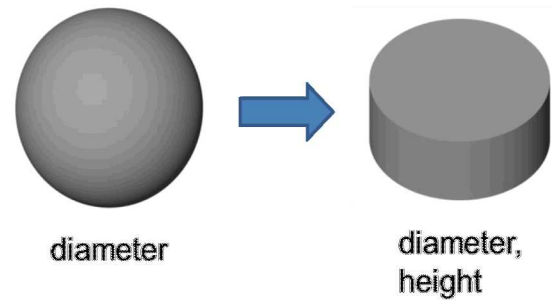
- Metallic or Dielectric
- Resonant or non-resonant



A collection of structures $< \lambda$ where electric/magnetic reflection, transmission & scattering can be controlled by each element

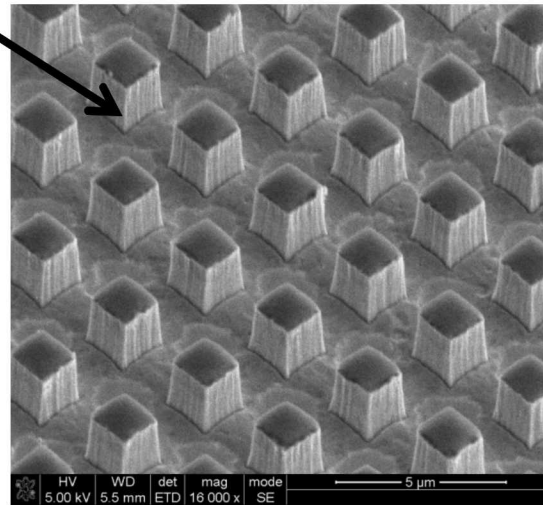
How to Fabricate Dielectric Metasurfaces

- Desired: arrays of high index, wavelength scale “particles”



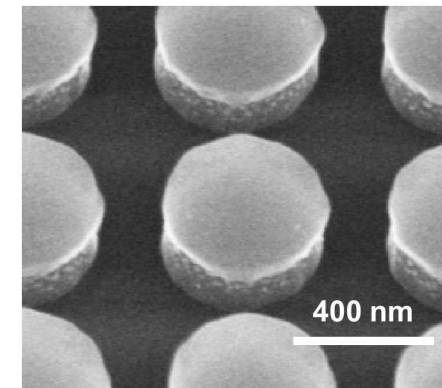
Tellurium
 $n \sim 5$ in the IR

$1.53 \times 1.53 \times 1.7 \mu\text{m}^3$



for 10 μm

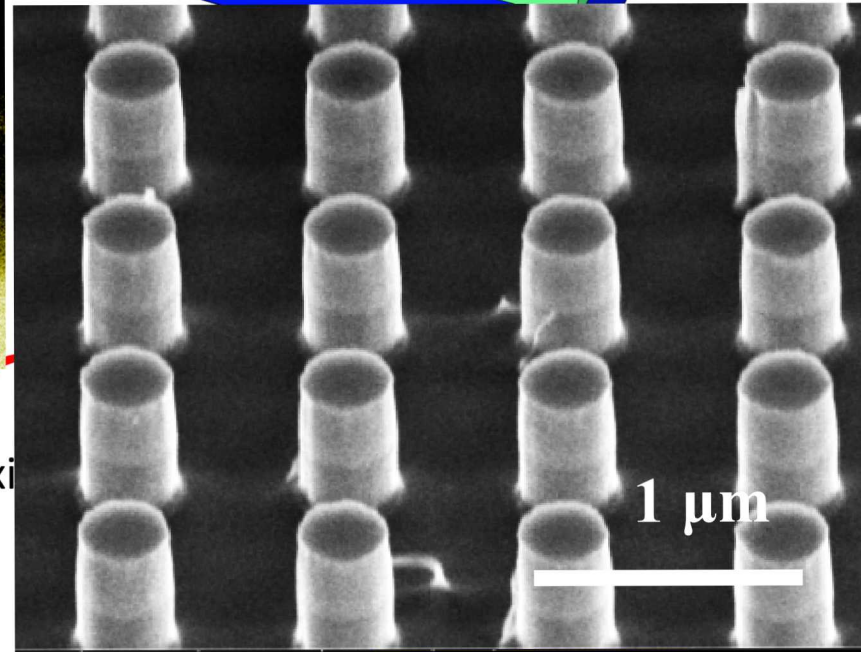
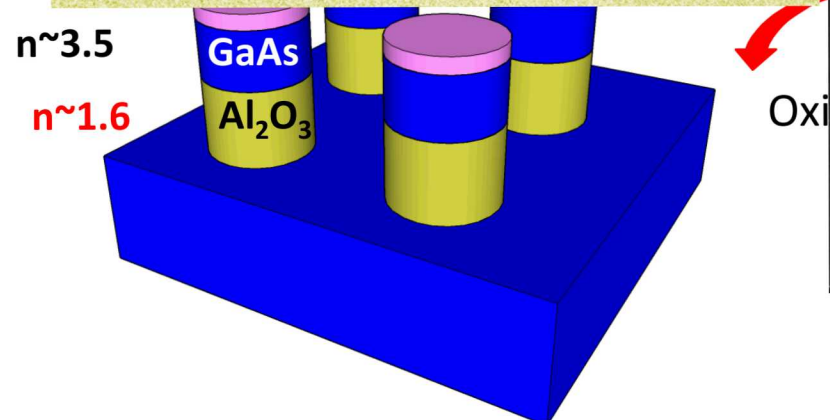
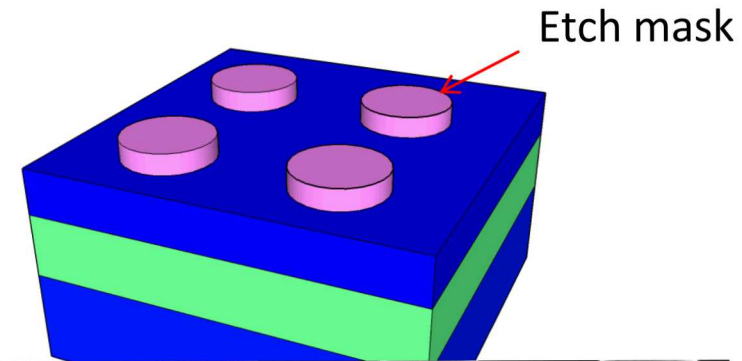
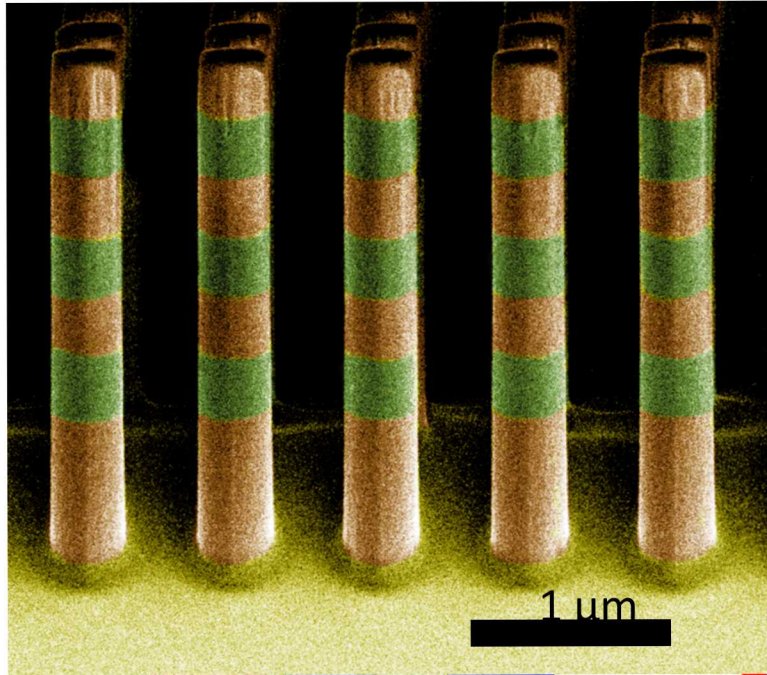
Silicon ($n \sim 3.8$) on glass



for 1.5 μm

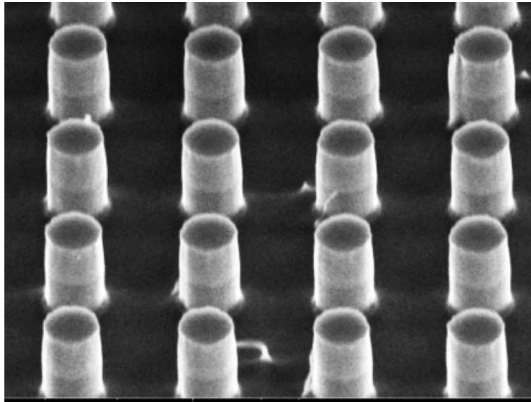
Dielectric Metasurfaces using III-V Semiconductors ($n \sim 3.5$, near IR)

Epitaxially grown: MBE, MOCVD

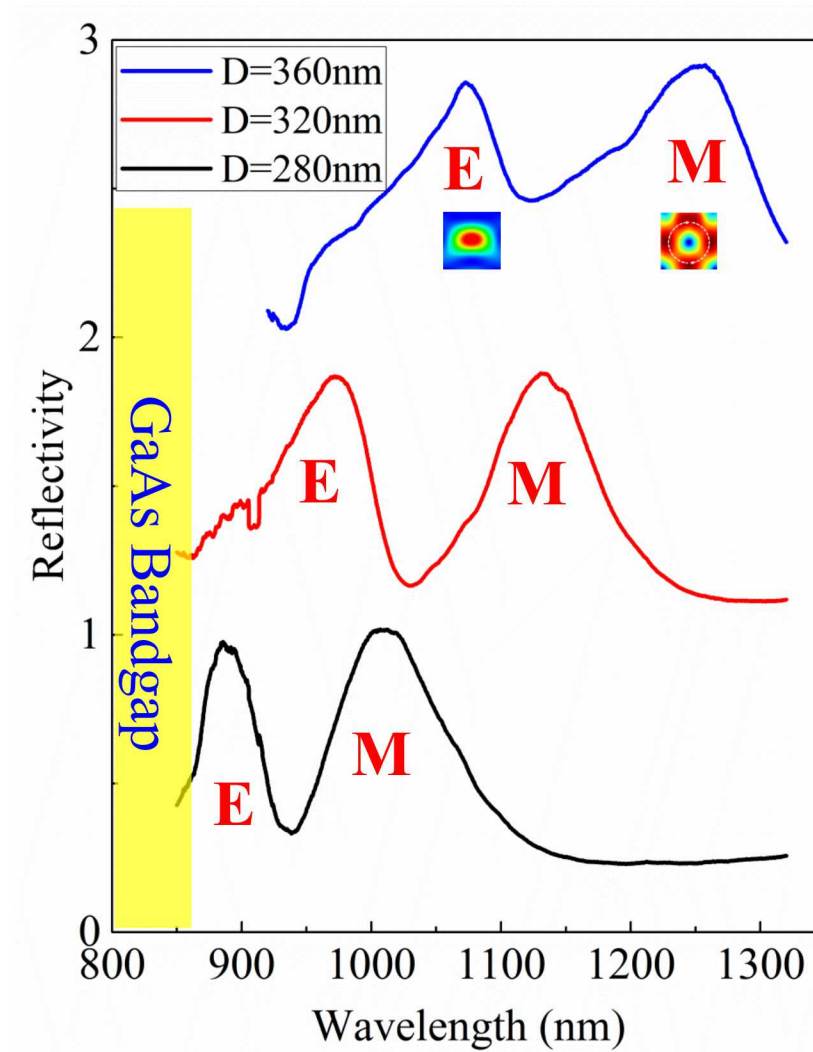


GaAs Dielectric Metasurface

GaAs disk height $\sim 300\text{nm}$
Different diameters

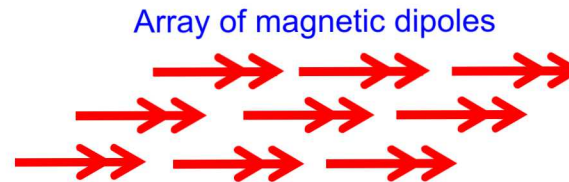


- Extremely low loss below bandgap
- Crystalline



Using Magnetic Resonances: The “Magnetic Mirror”

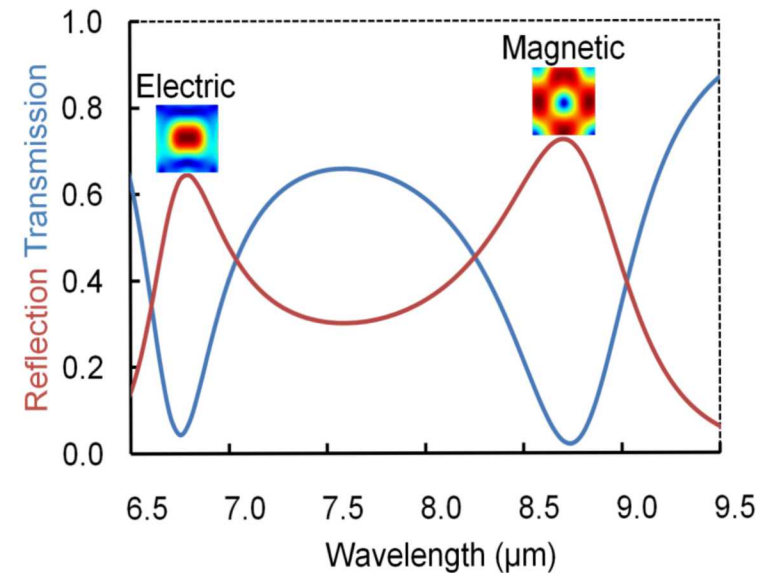
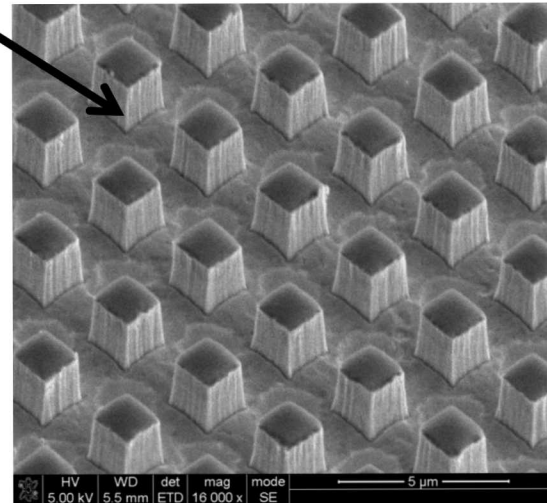
it does not exist in nature



Magnetic dipole responds in phase with the electric field (in microwave language, this represents an **artificial magnetic conductor**)

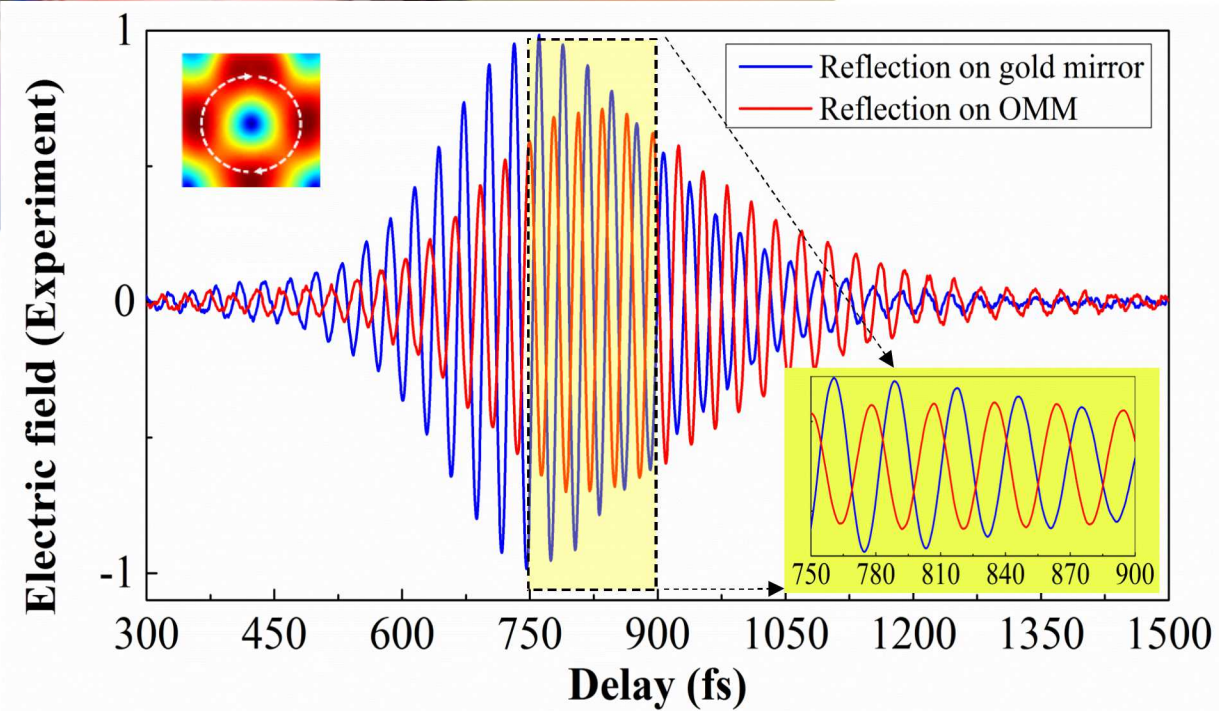
Tellurium
 $n \sim 5$ in the IR

$1.53 \times 1.53 \times 1.7 \mu\text{m}^3$



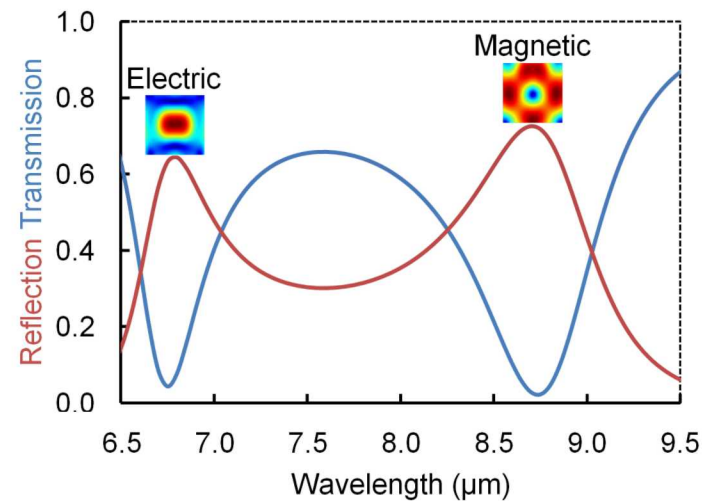
Optical Magnetic Mirror

Absolute Phase of Reflected Wave

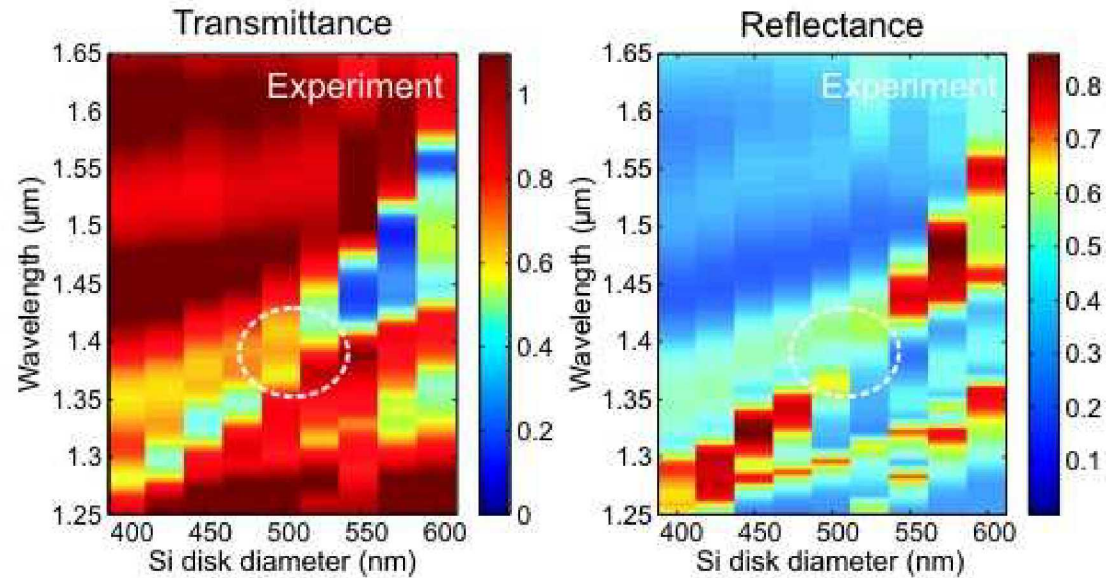
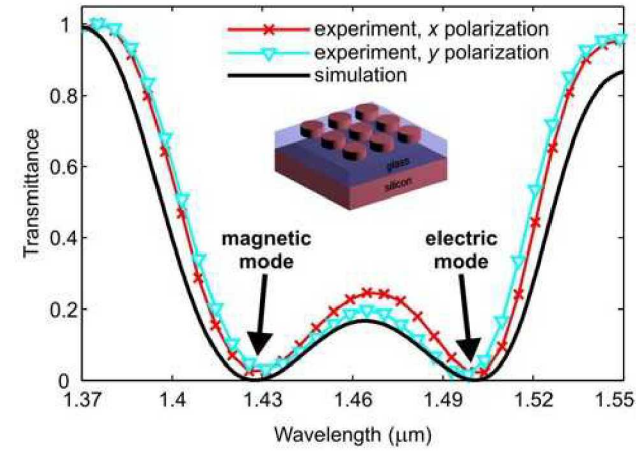
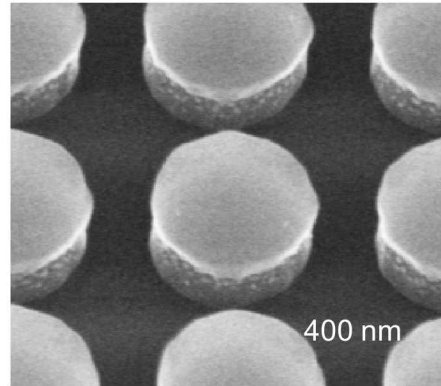


Controlling Spectral Location of Mie Modes

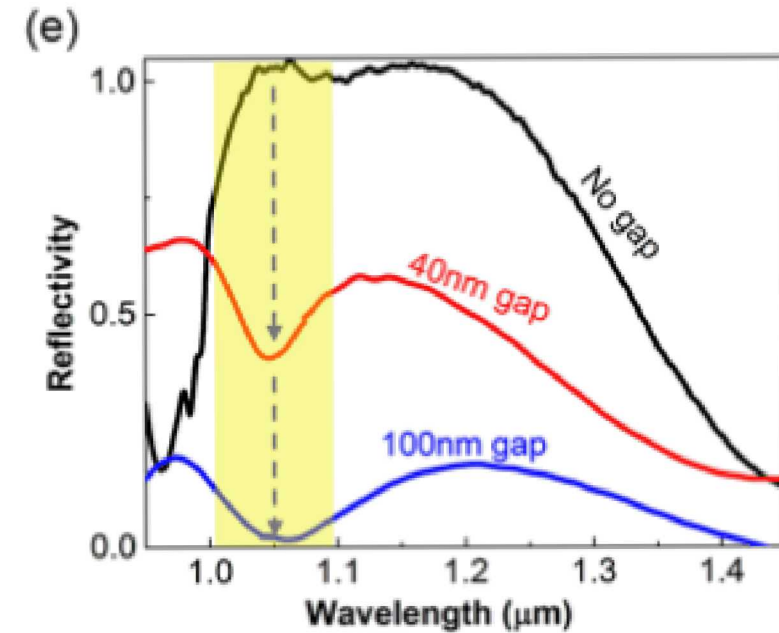
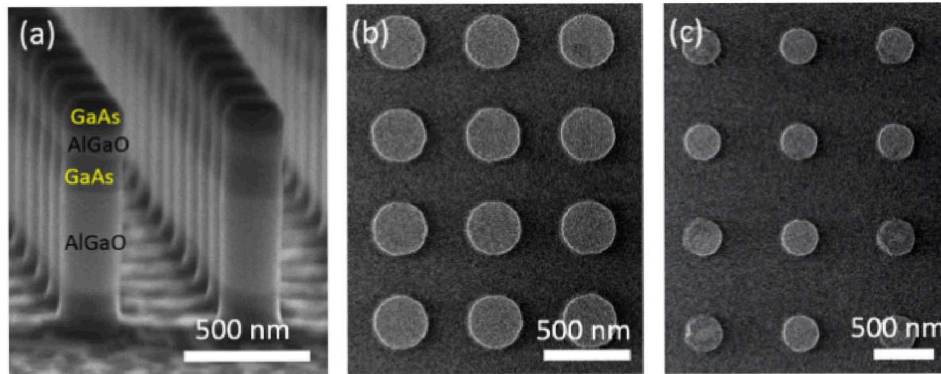
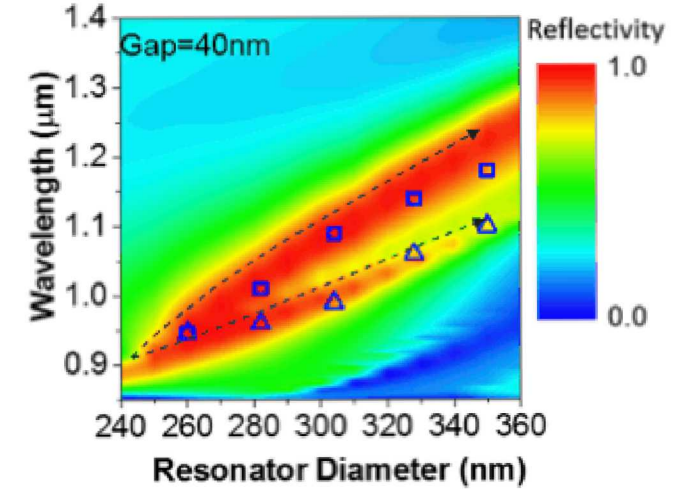
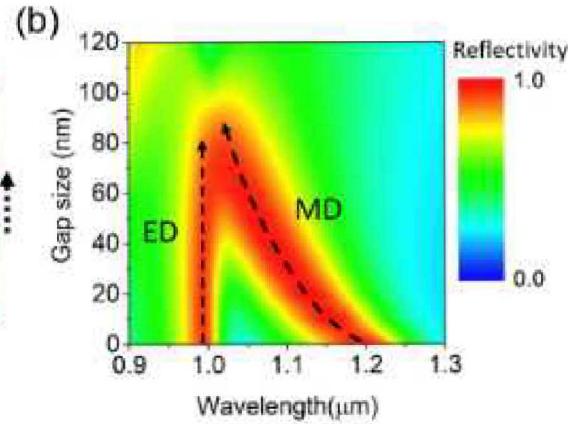
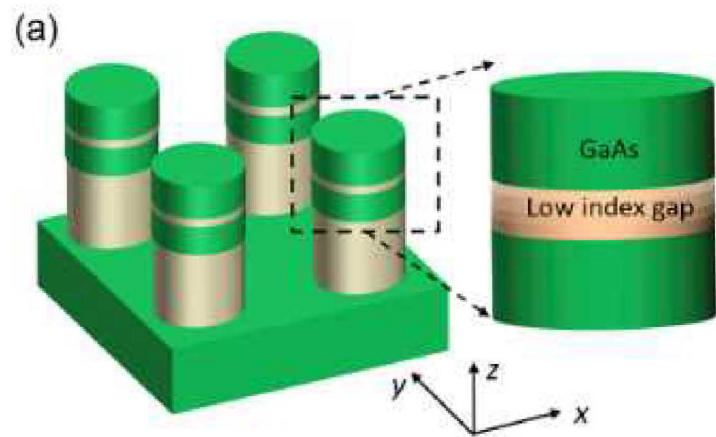
- Change the dielectric environment (liquid crystals: *ACS Nano* 9, 4308 (2015))
- Change aspect ratio of cubes or cylinders
- Introduce a “perturbation” (cut)



Controlling Spectral Location of Mie Modes: Changing the Aspect Ratio



Controlling Mie Modes: introducing a cut



Huygens Metasurfaces: Control of Scattering

- Huygens' Principle:
 - Every point on a wavefront is a secondary source of outgoing waves

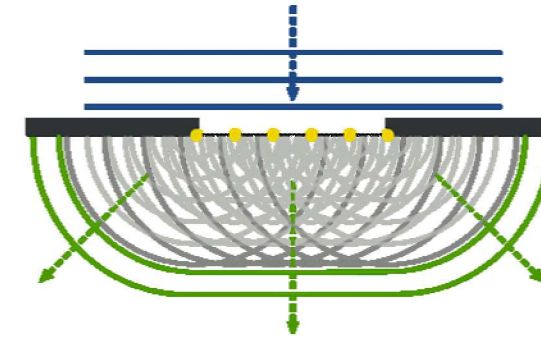
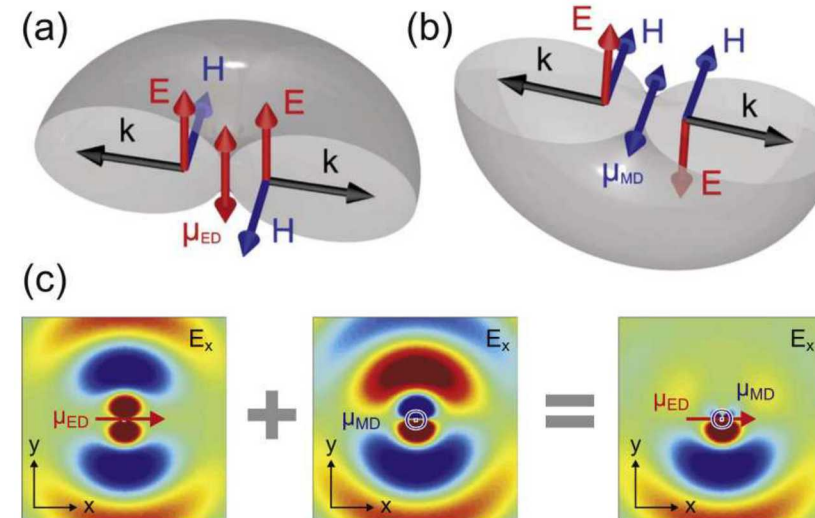


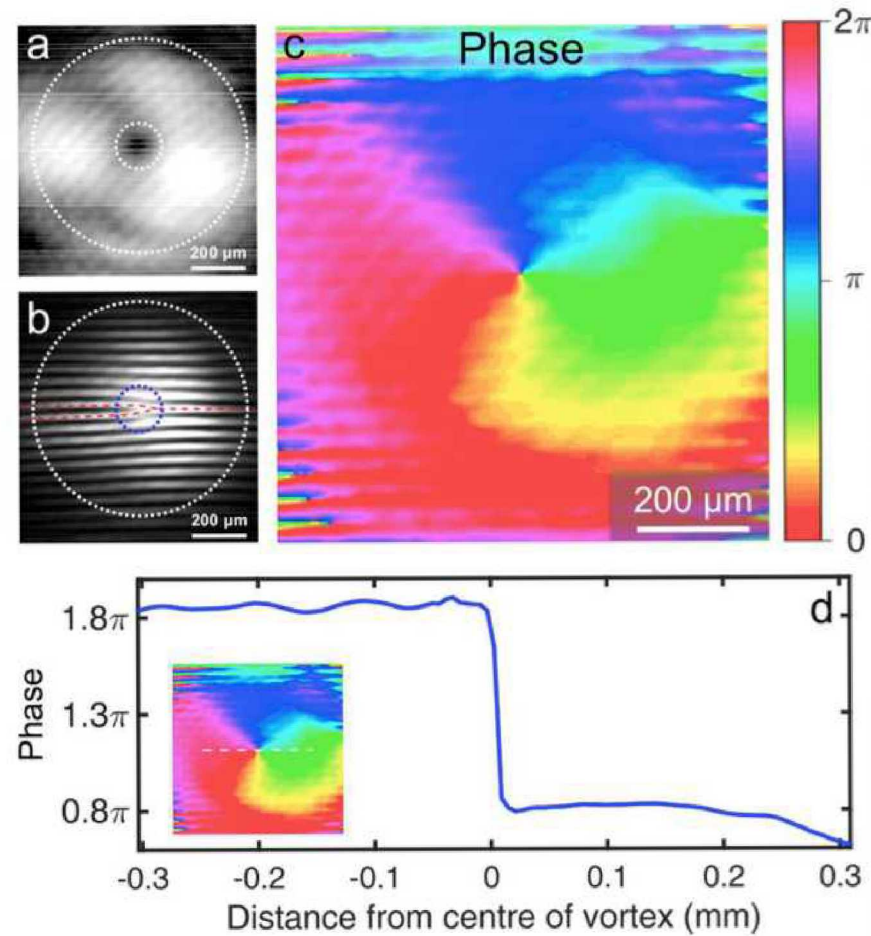
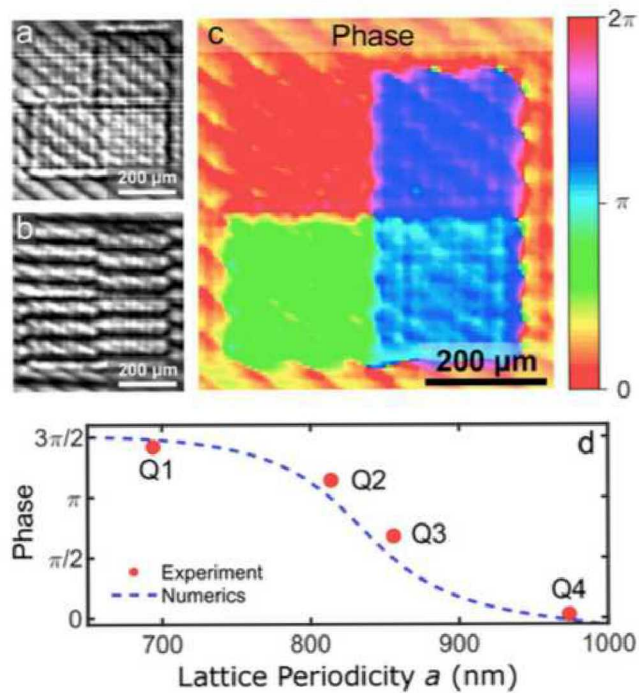
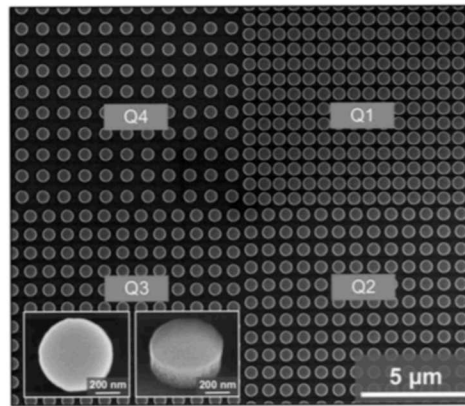
Image: Wikipedia
 Huygens, *Traité de la Lumière*, (1690)
 Love, *Phil. Trans. R. Soc. Lond. A* **197**, 1
 (1901)

- Huygens' sources
 - **Crossed E & H dipoles (Love's formalism)**
 - Produce **forward-only** propagating elementary waves

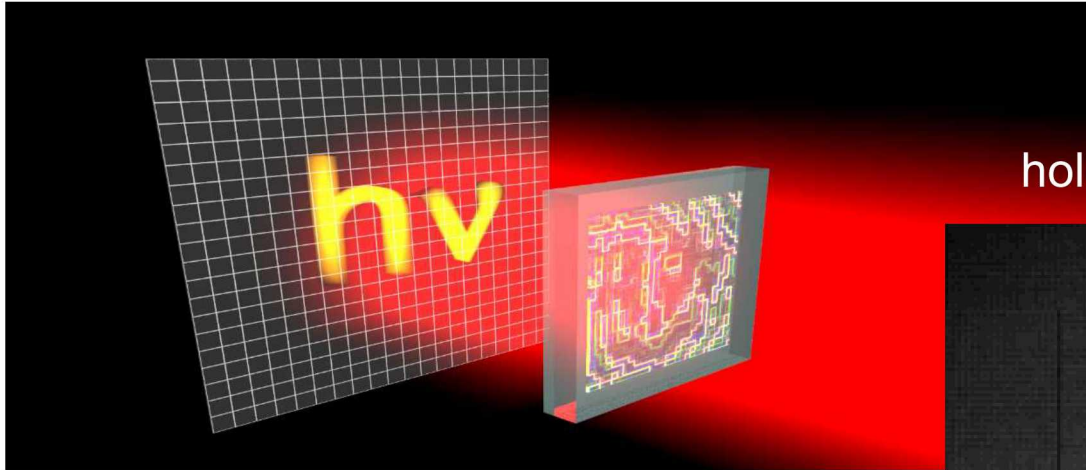


(R. Zia)

Huygens Metasurface Phase Plate: Vortex Beam

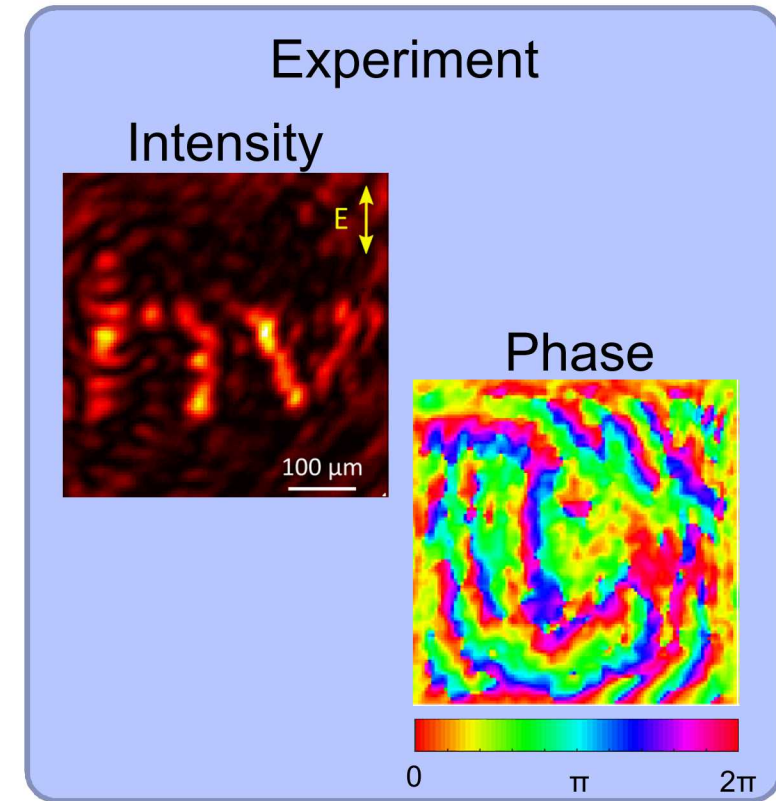
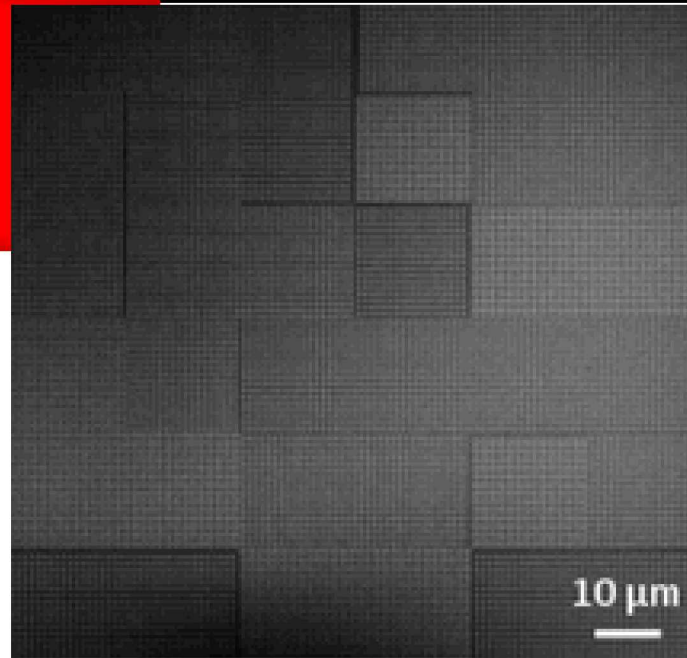


Huygens metasurface hologram demonstrates complex wavefront control



Normal incident 1477nm laser passes through the metasurface to create a 4-phase-level hologram 12 mm behind the sample.

Section of our holographic metasurface

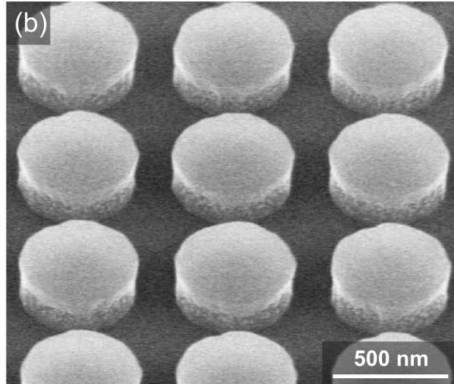


82% transmittance & 40% imaging efficiency

Metasurfaces for Flat Optics: Summary

School #1: “Huygens” metasurfaces

(ANU, Sandia, Argonne, Jena,...)

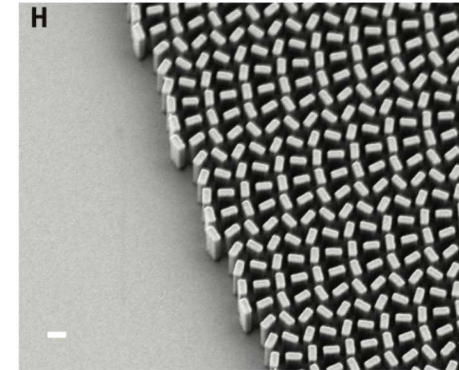


Pros: Easy fabrication

Cons: Needs small array for a single phase “element”

School #2: High aspect ratio elements

(Harvard, Caltech, Technion,...)

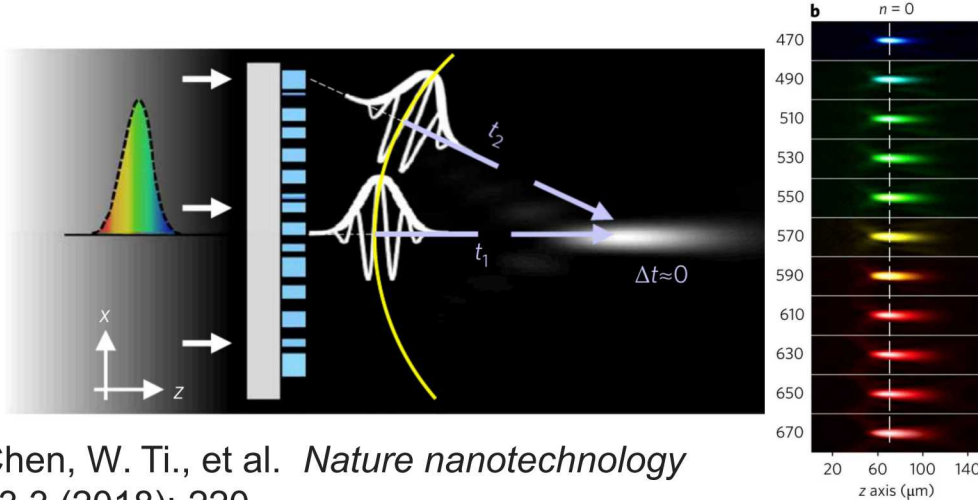


Phase determined by
single element

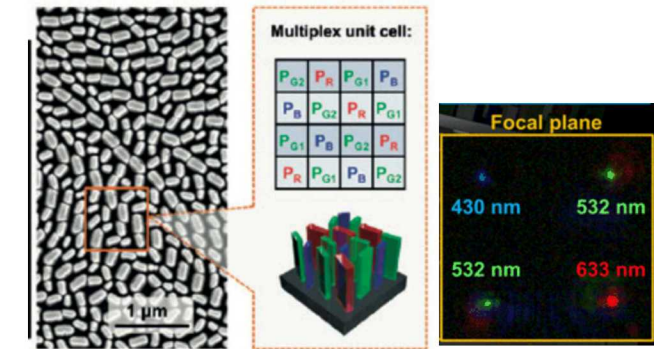
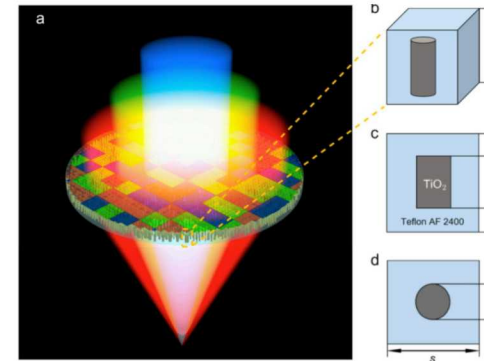
Difficult fabrication

All-dielectric Metalenses: Recent Literature

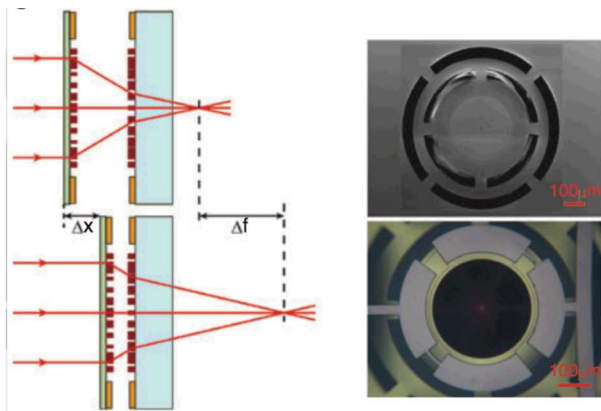
Broadband achromatic metalens



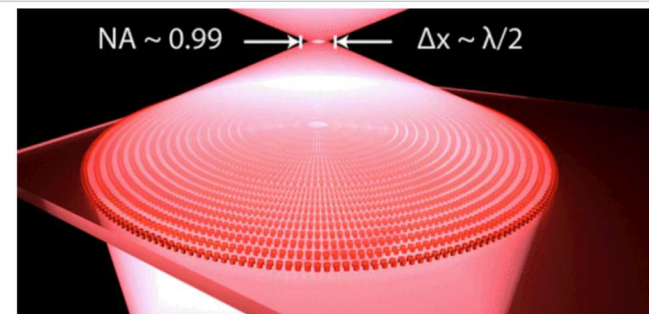
Multiwavelength metasurface lenses



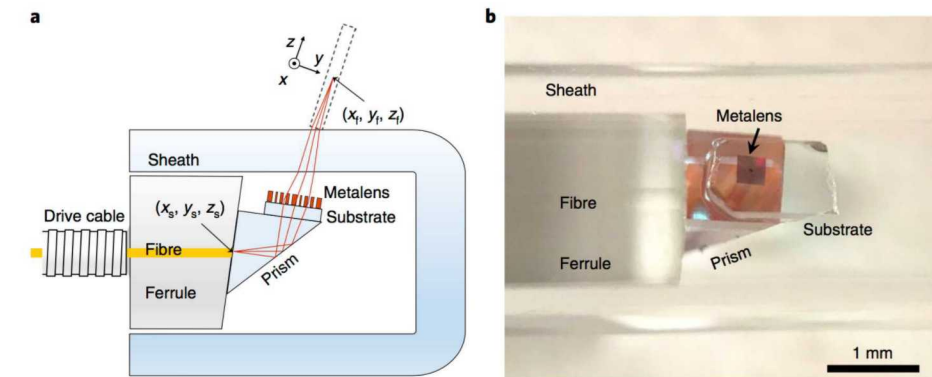
Tunable Metalenses



A Metalens with a Near-Unity Numerical Aperture



Nano-optic endoscope



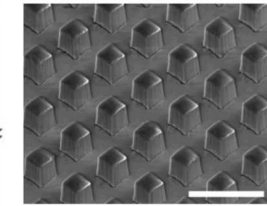
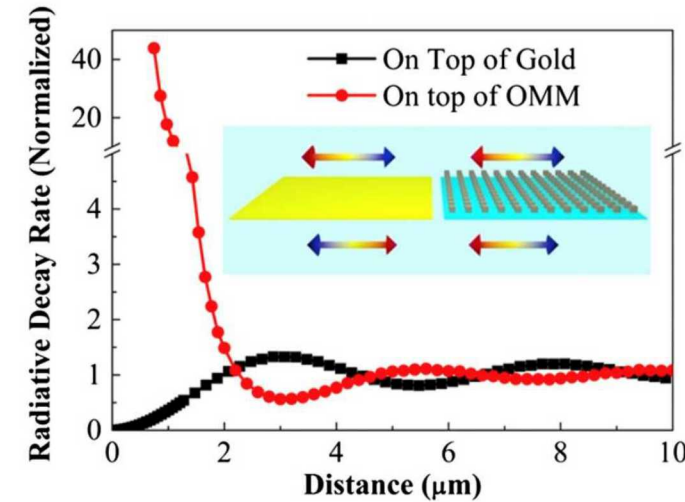


Outline

- Metamaterials & Metasurfaces: refresher, definitions, etc.
- Linear Metasurfaces:
 - Optical magnetic mirror
 - Huygens meta-optics
- **Emitters + Metasurfaces**
- Nonlinear optics with all-dielectric metasurfaces:
- Outlook on future 3D fabrication

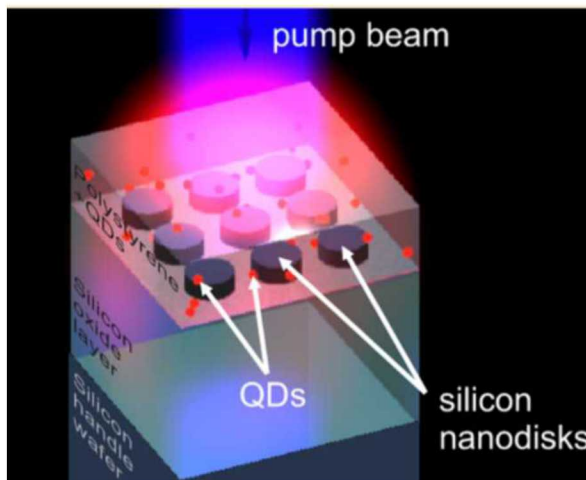
Emitters Coupled to Metasurfaces

Simulation of emitter very close ($\sim \lambda$) close to dielectric optical mirror showed large radiative enhancement.



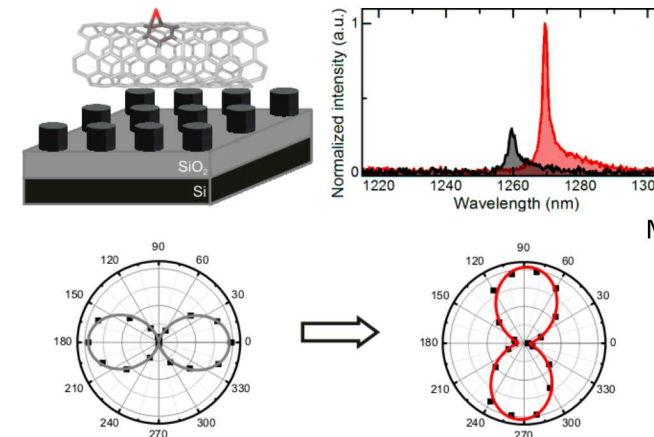
Liu *et al*, Optica 2014

Ensembles of colloidal QDs on top of Si-metasurface: PL reshaping



Staude *et al*, ACS Photonics 2015

Single photon emitter (dopant in CNT) on top of Si-metasurface: rotation of PL polarization



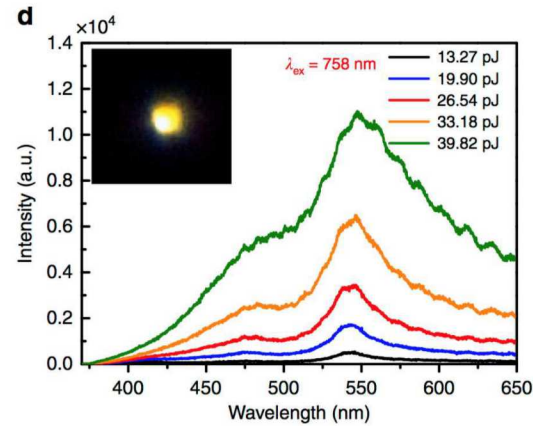
Ma *et al*, ACS Nano 2017

See also work by Hong, Maier, Bonod, Belov, Krasnok, etc.

Emission + All-dielectric Metasurfaces:

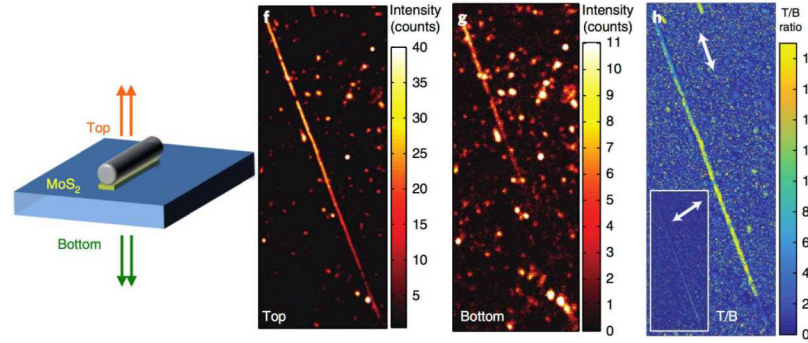
Recent work

White-light emission from silicon nanospheres



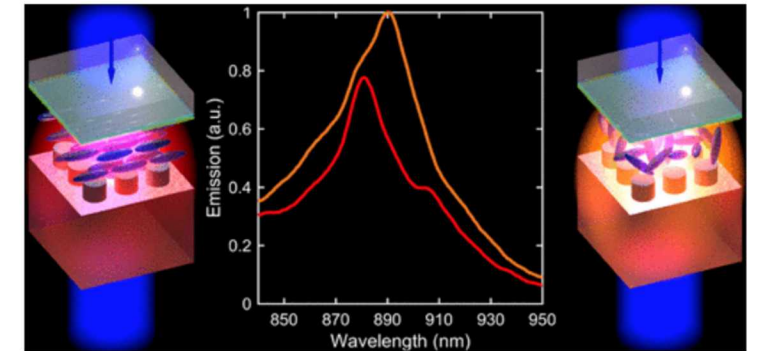
Zhang, C., et al.
Nature communications 9 (2018).

Directional light emission from monolayer MoS₂



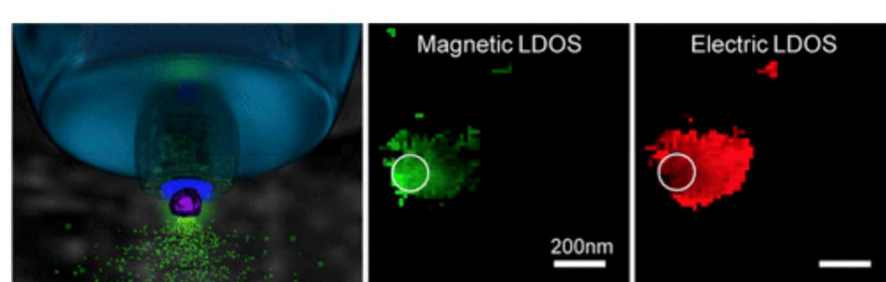
Cihan, A. F., et al.
Nature Photonics 12.5 (2018): 284.

Active Tuning of Spontaneous Emission by Mie-Resonant Dielectric Metasurfaces



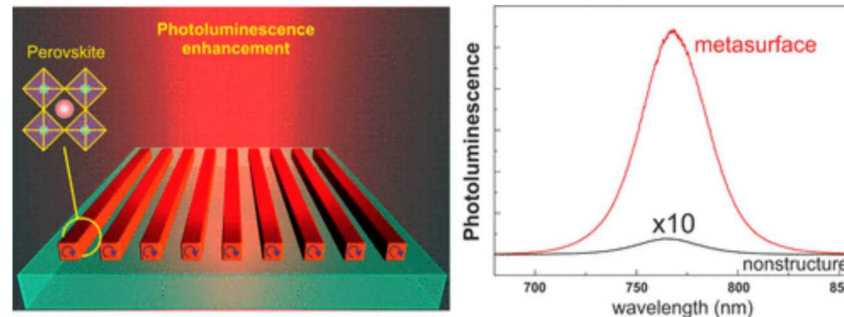
Bohn, J., et al. *Nano letters* 18.6 (2018): 3461-3465.

Enhanced emission of luminescent nanocrystals



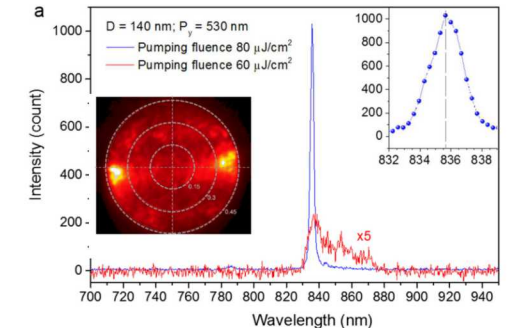
Sanz-Paz, M., et al.
Nano Letters 18.6 (2018): 3481-3487.

Emission enhancement in hybrid perovskite metasurfaces



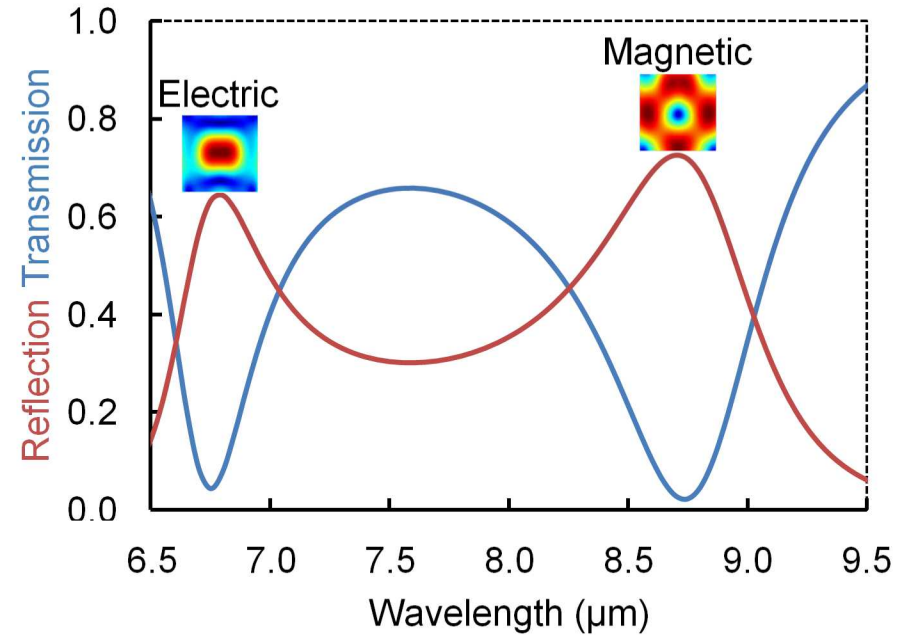
Makarov, S. V., et al. *ACS Photonics* 4.4 (2017): 728-735.

Lasing action in active dielectric nanoantenna arrays



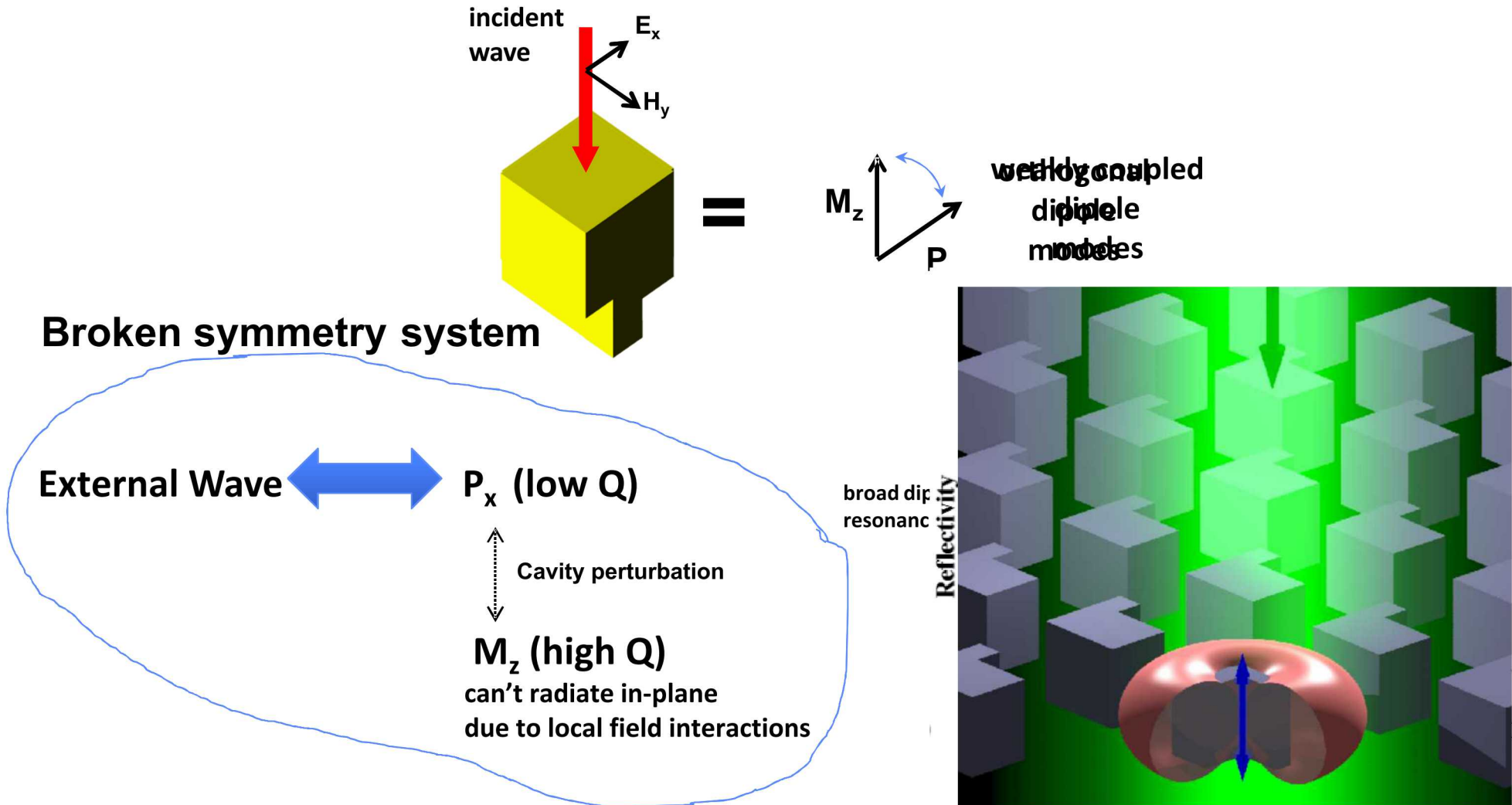
Ha, S. T., et al. *arXiv preprint arXiv:1803.09993* (2018).

Towards High Q: Broken Symmetry Resonators

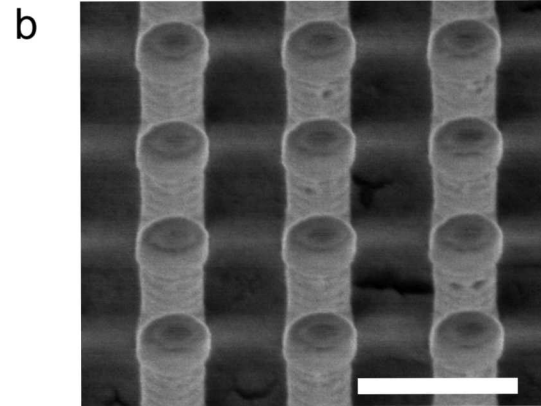
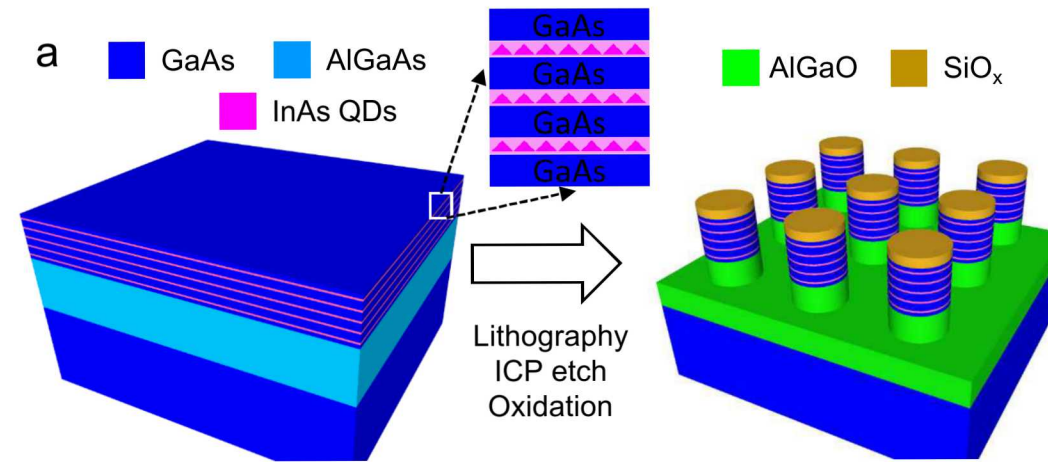


low Q because these modes couple to free space

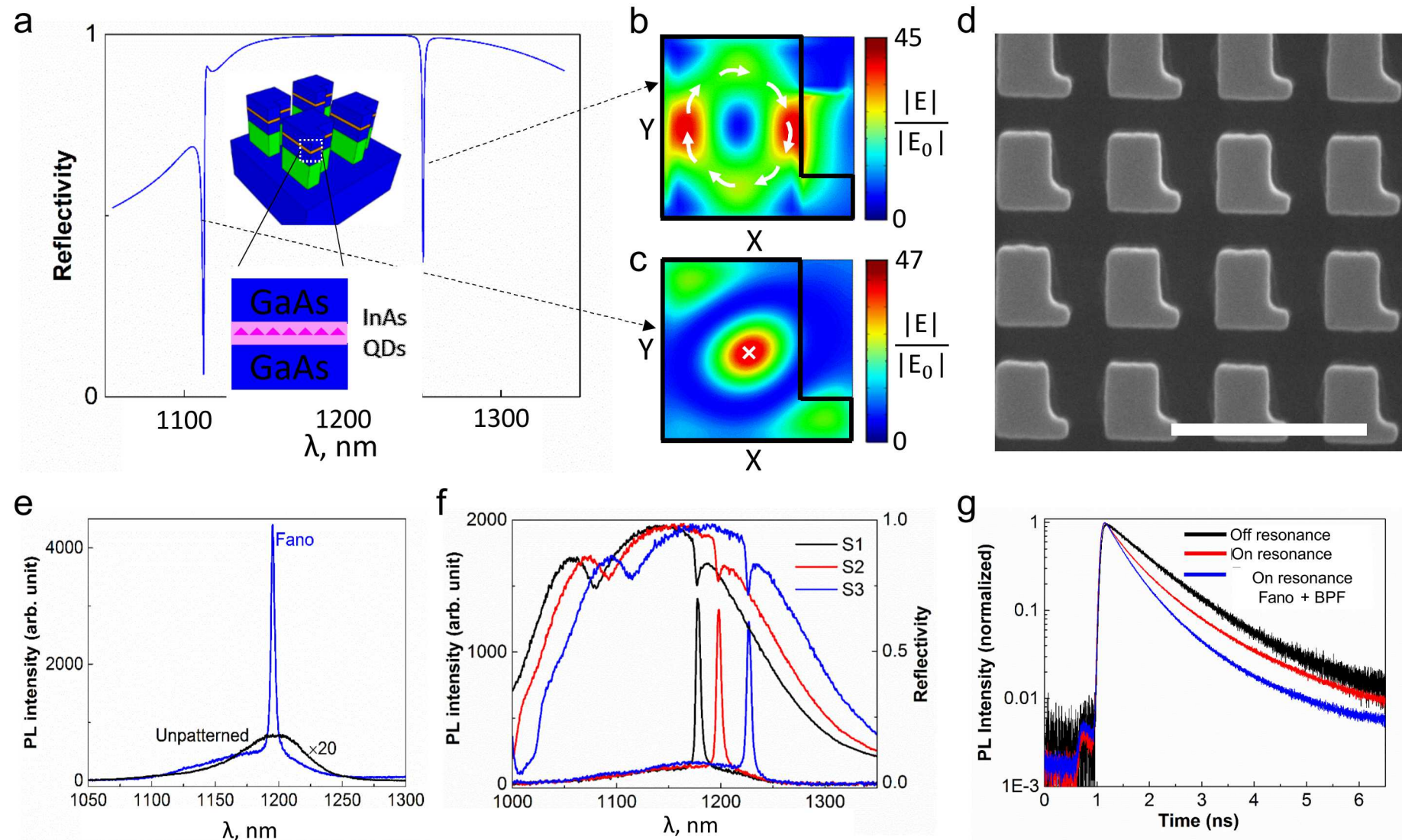
High Q modes Using “Broken Symmetry” Resonators



Embedding Emitters in Metasurfaces



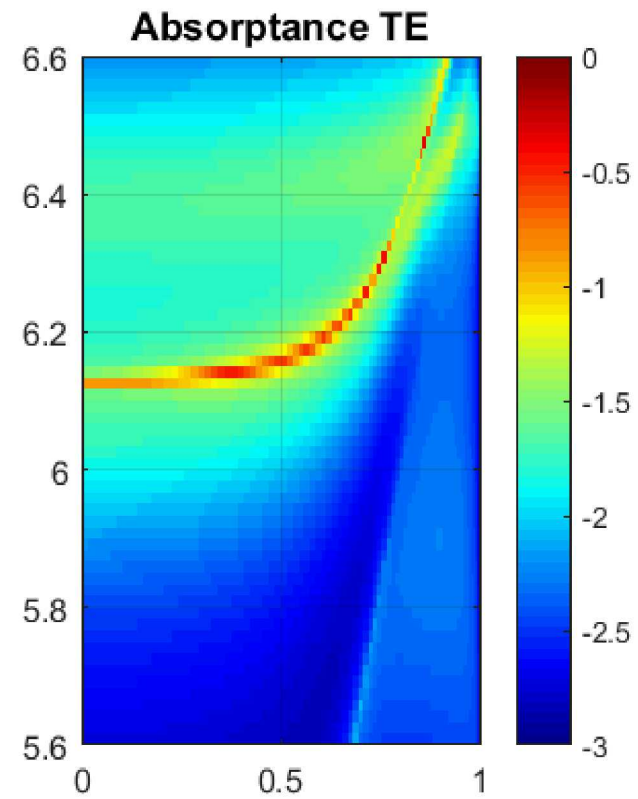
PL: Broken-symmetry Resonators





Back-Focal Plane Emission

(Broken Symmetry Resonators)





Outline

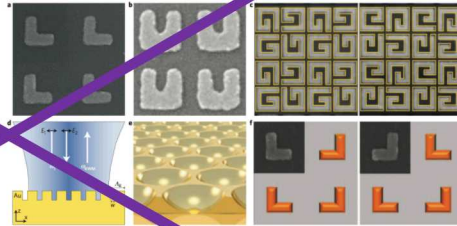
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- Outlook on future 3D fabrication

Paths to Nonlinear Metasurfaces

Metallic/Plasmonic

Using metal nonlinearity:

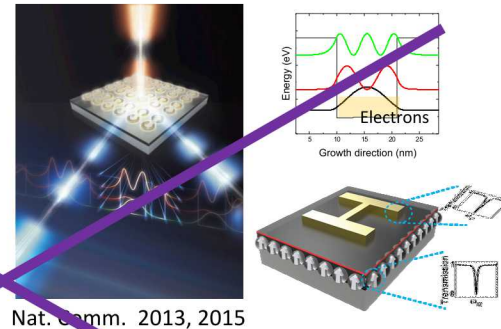
Nature Photonics, 6(11), 137



- + Small mode volume
- Low efficiency

Combined with an “engineered” nonlinearity.

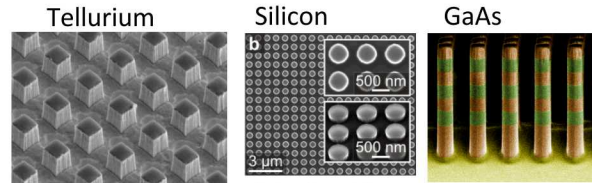
Metallic metasurfaces on III-V heterostructures



Nat. Comm. 2013, 2015

- + Record high SHG
- Saturation

Dielectric



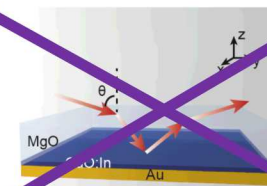
PRL 2012

Adv. Opt. Mat. 2015, NL 2015, ACS Phot. 2016

Adv. Opt. Mat. 2016, NL 2016

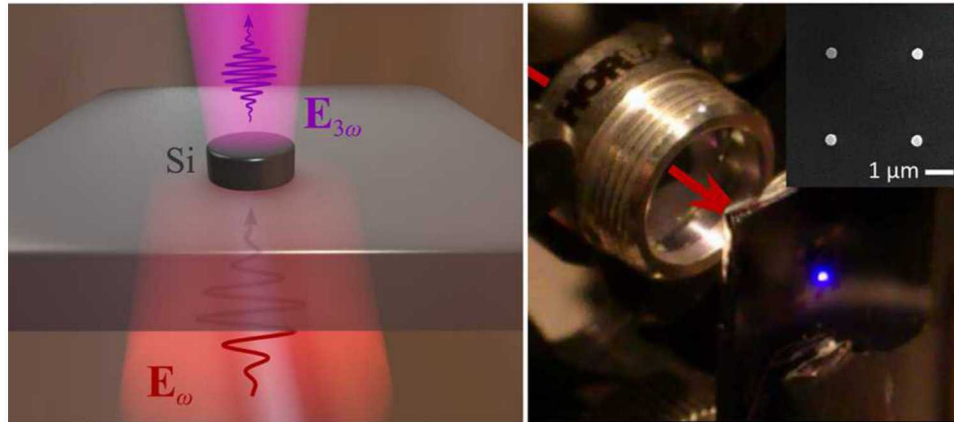
- + High nonlinear coefficient materials (GaAs: $\sim 200 \text{ pm/V}$)
- + large mode volume
- medium to low efficiency

Epsilon Near Zero



- + uniform nonlinearity, high field
- ??

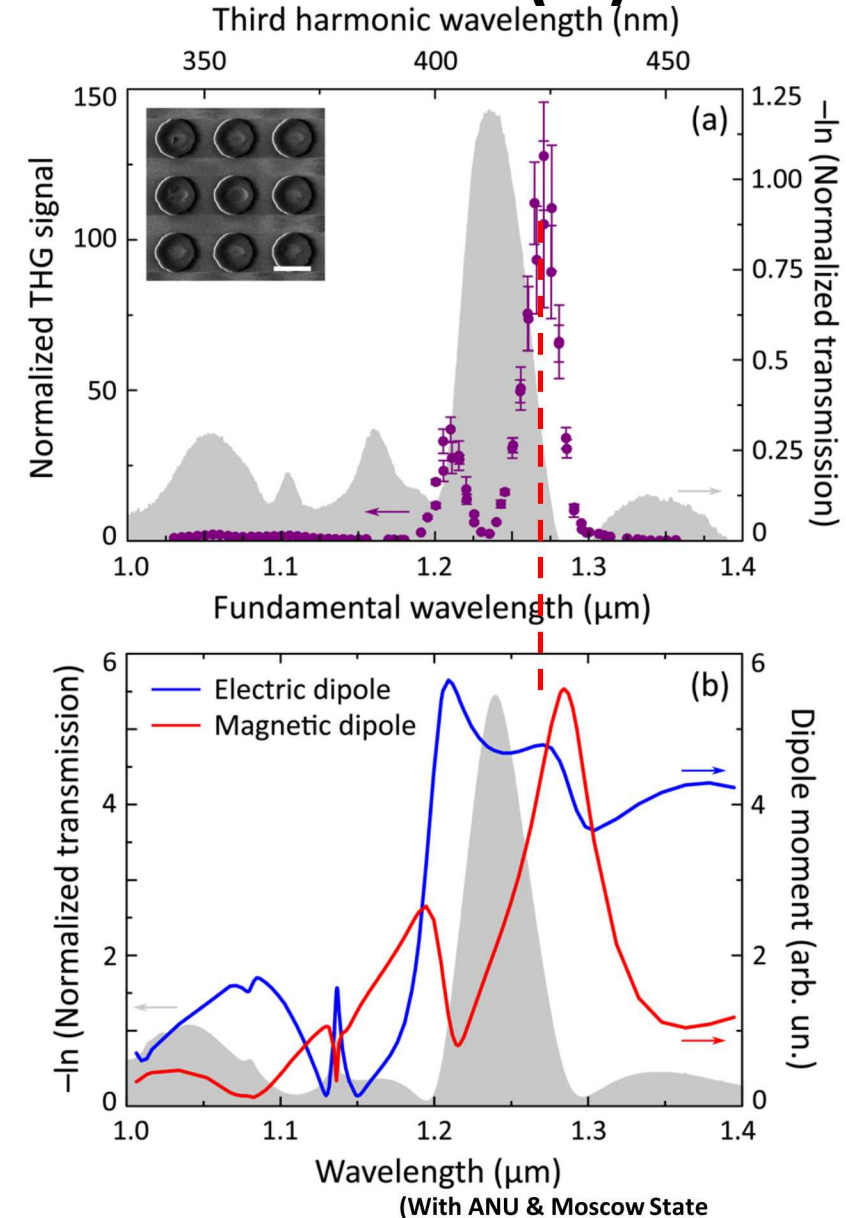
Optical Nonlinearities from Dielectric Metasurfaces: Third Harmonic Generation (Si)



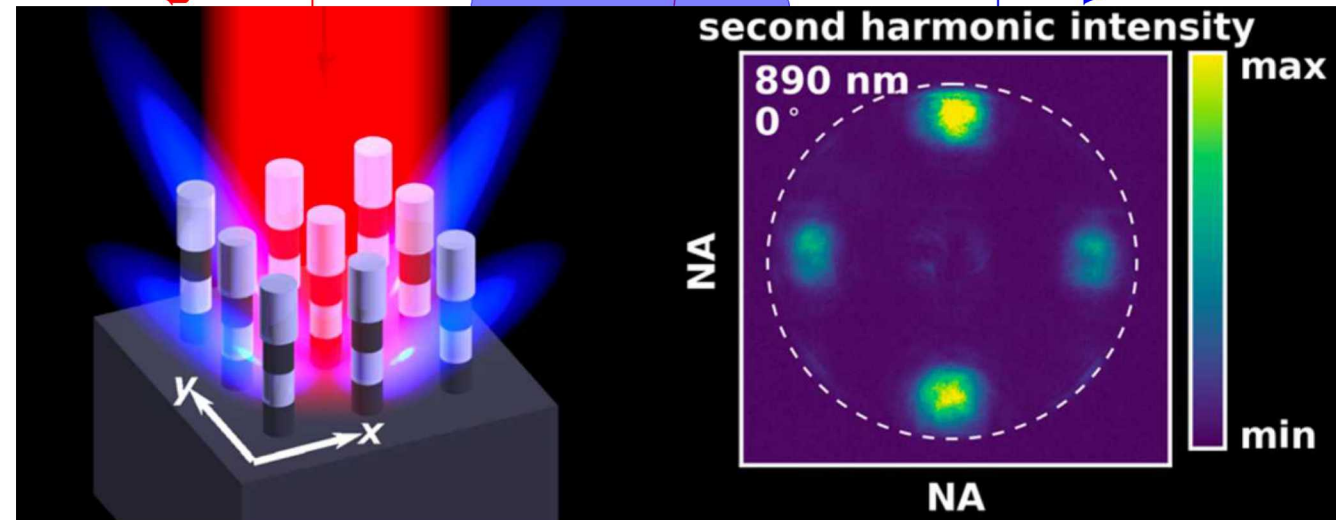
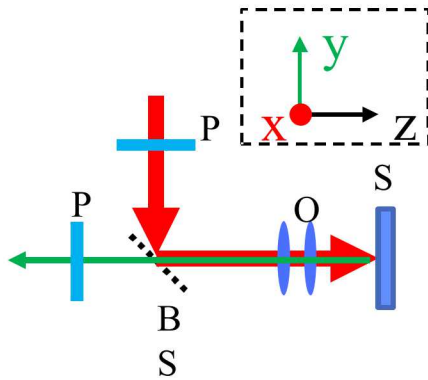
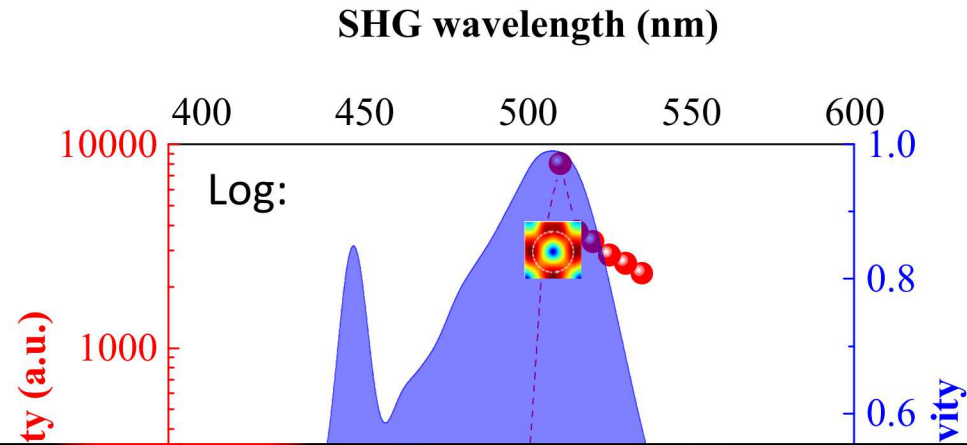
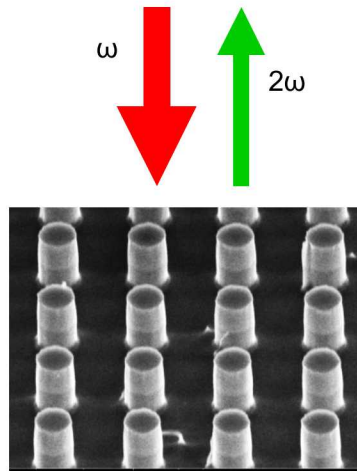
Strong enhancement near magnetic dipole resonance

Silicon is passive & has no $\chi^{(2)}$

Nano Letters (2015)



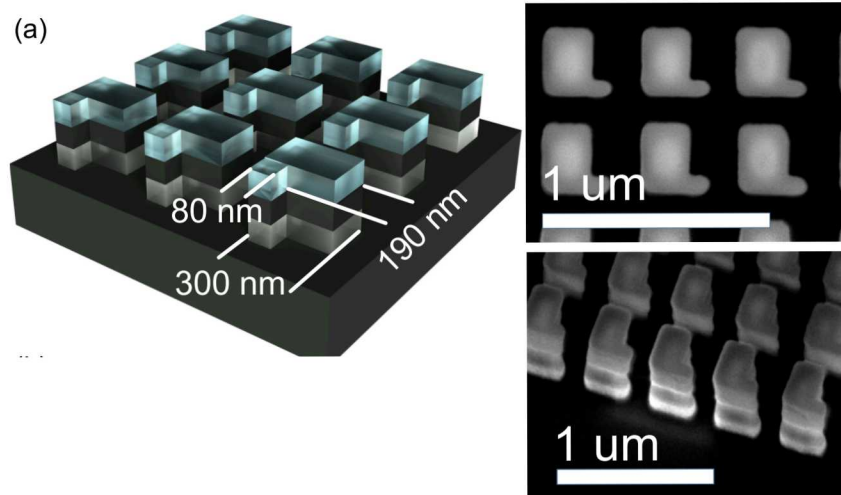
Second Harmonic Generation from GaAs Metasurfaces



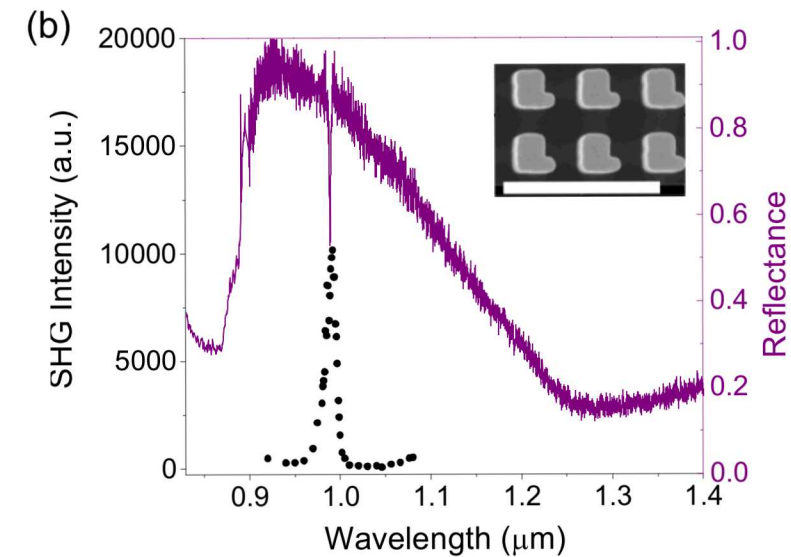
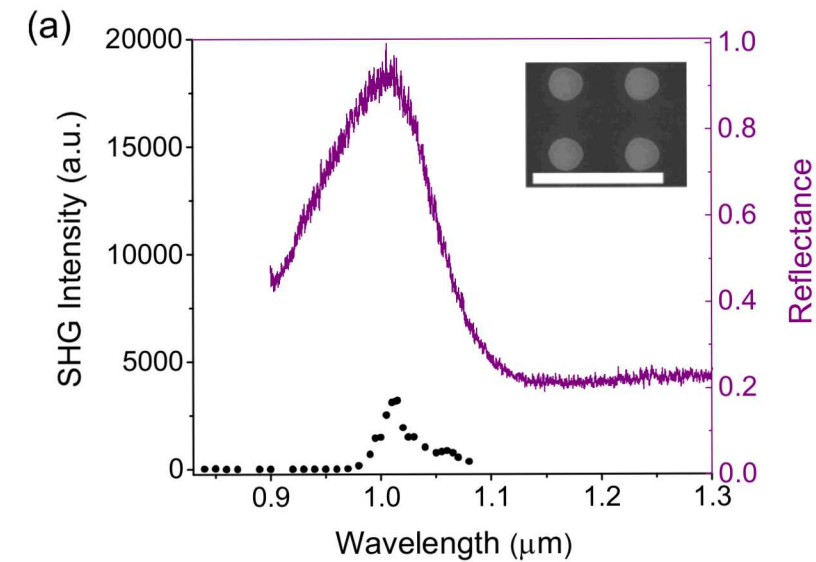
ACS Photonics 2018, 5, 1786–1793

Nano Letters 2016
Also ANU, Costa (Italy)

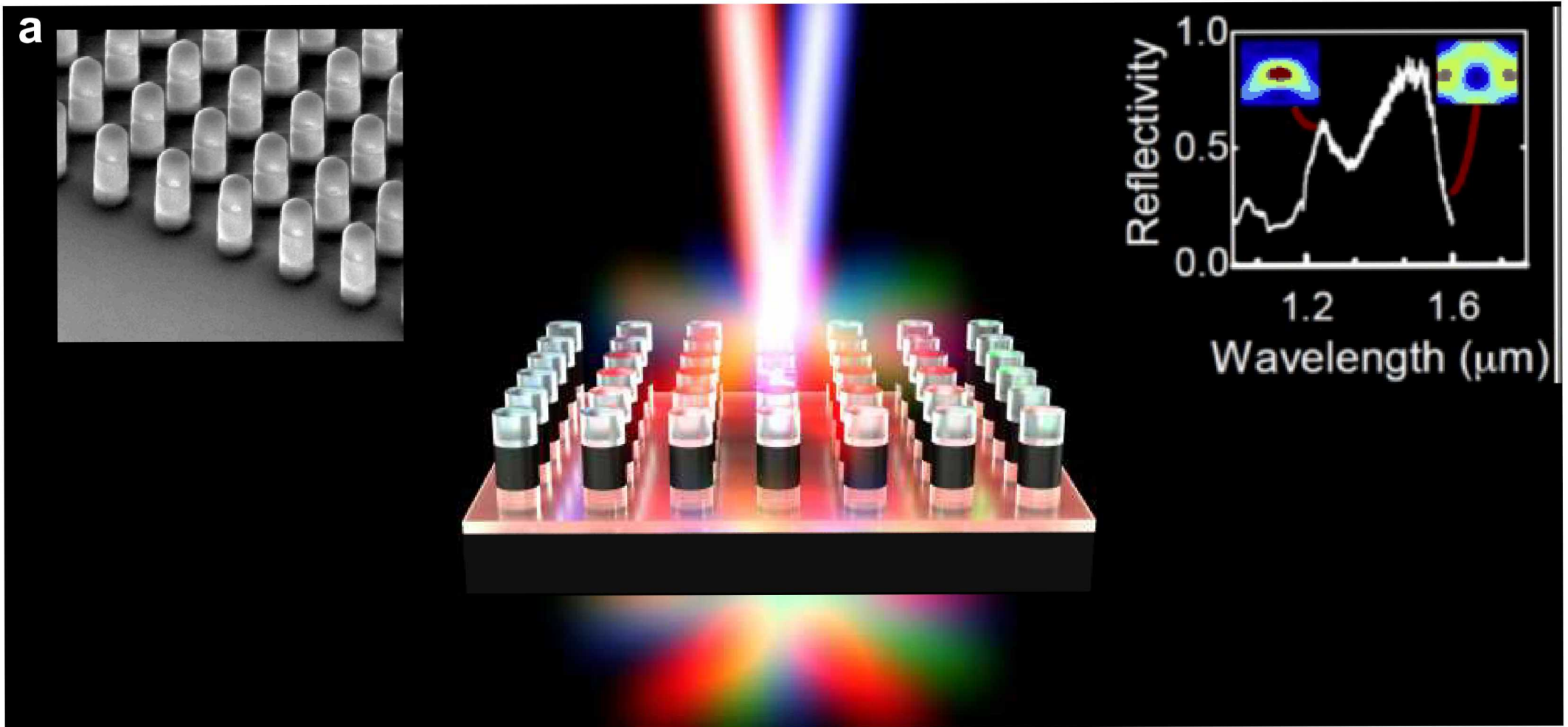
Higher Q: SHG from Broken Symmetry Metasurfaces



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



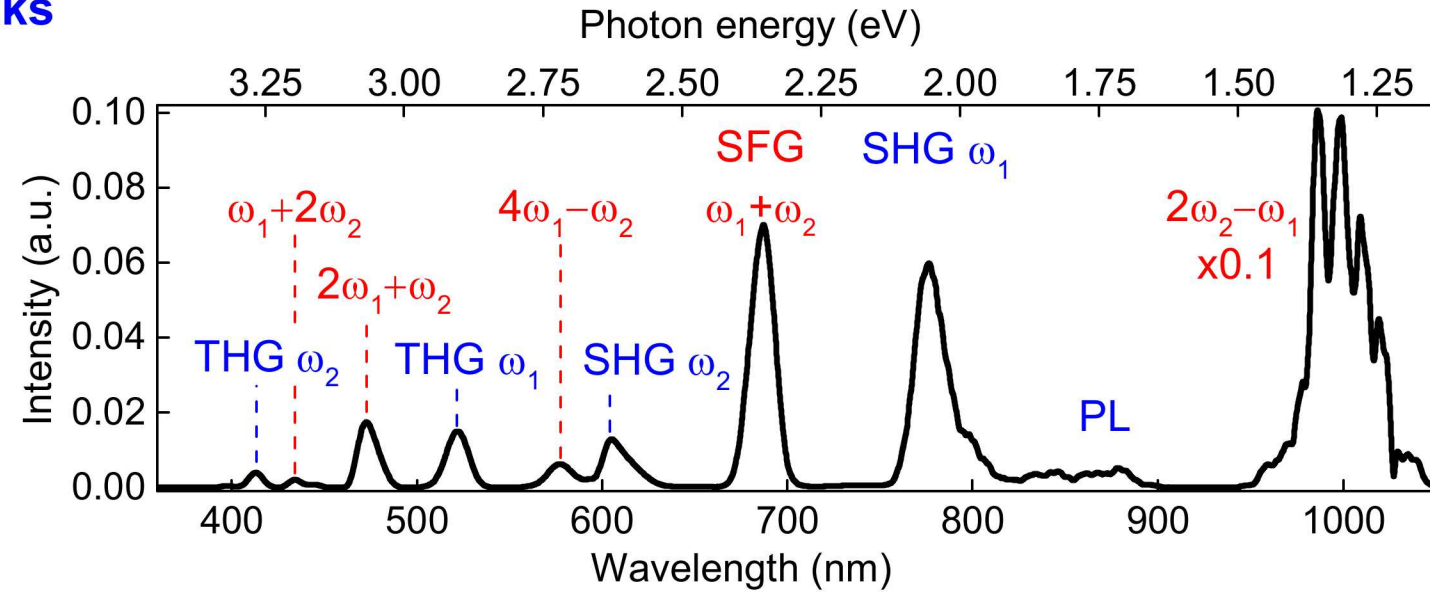
Higher Harmonics: The “Optical Metamixer”



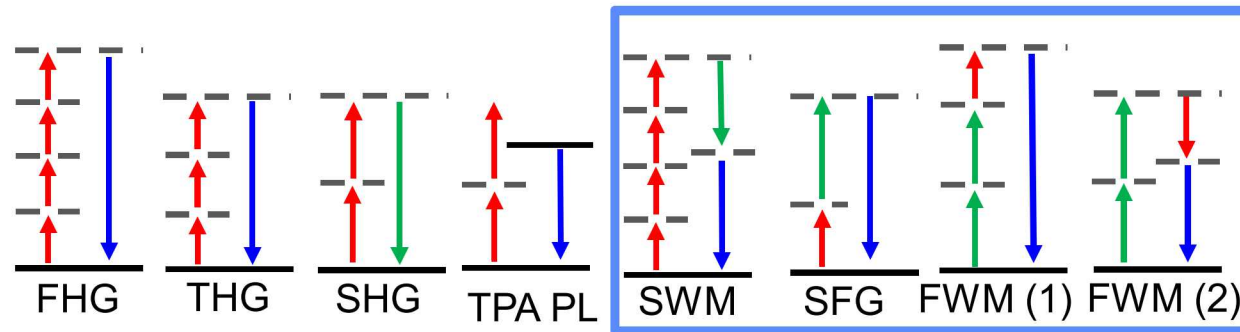
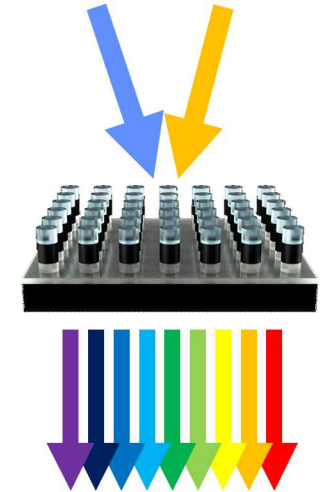
Nature Comm. 2018

Frequency mixing spectra

11 peaks



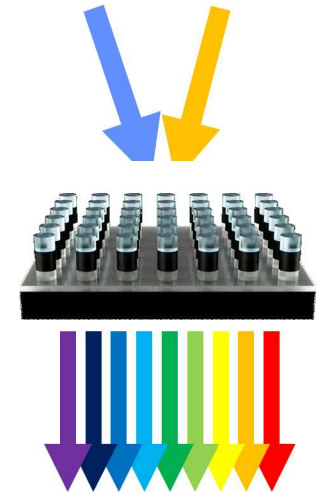
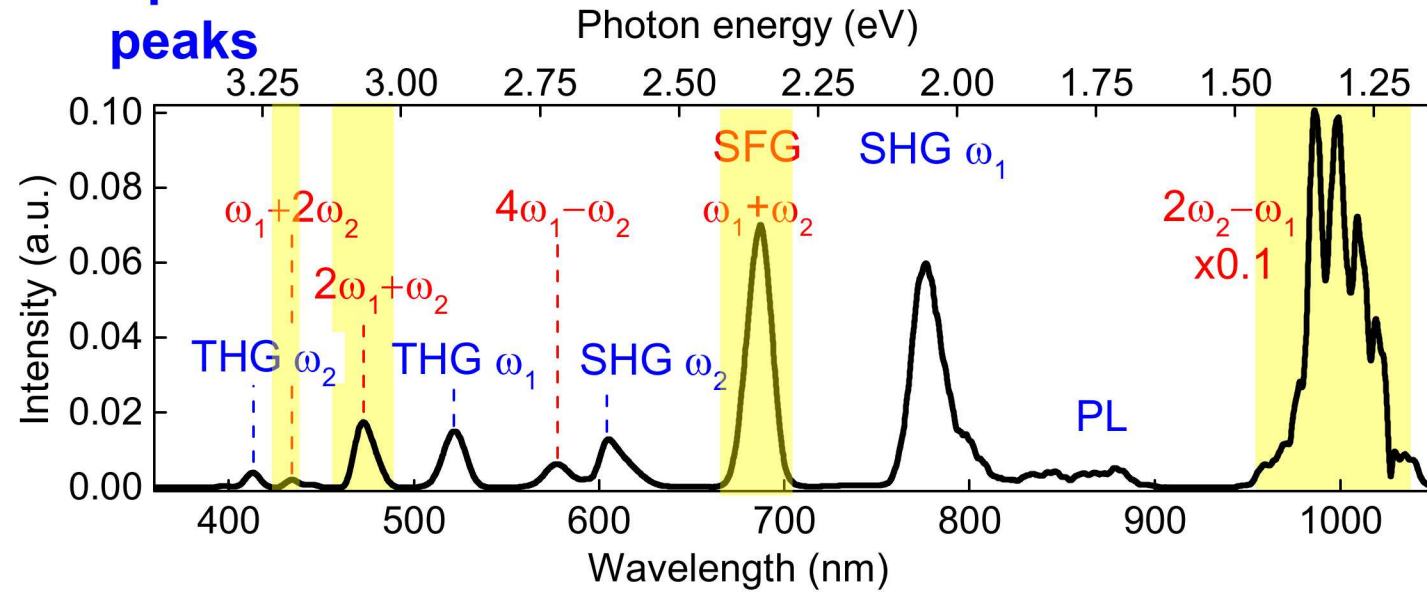
1570 nm 1240 nm



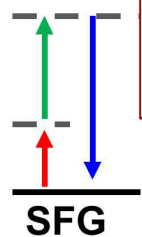
7 different nonlinear processes: 2nd, 3rd, 4th harmonics, sum frequency generation, 4 wave-mixing, six-wave mixing....

Frequency mixing spectra

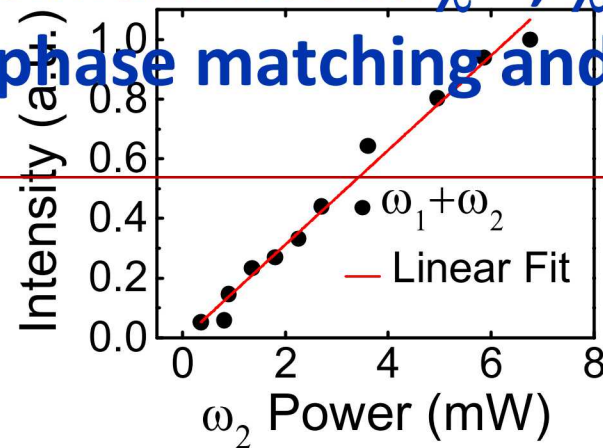
11 spectral peaks



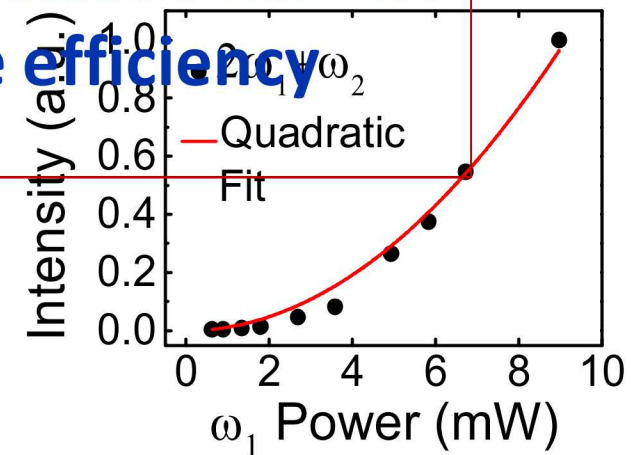
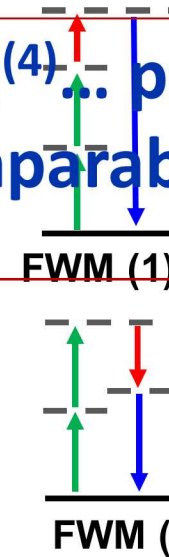
SFG power dependence



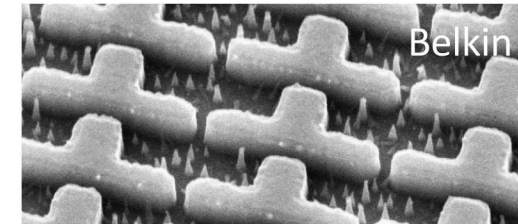
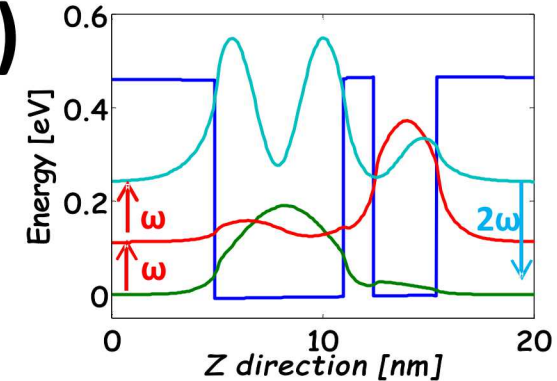
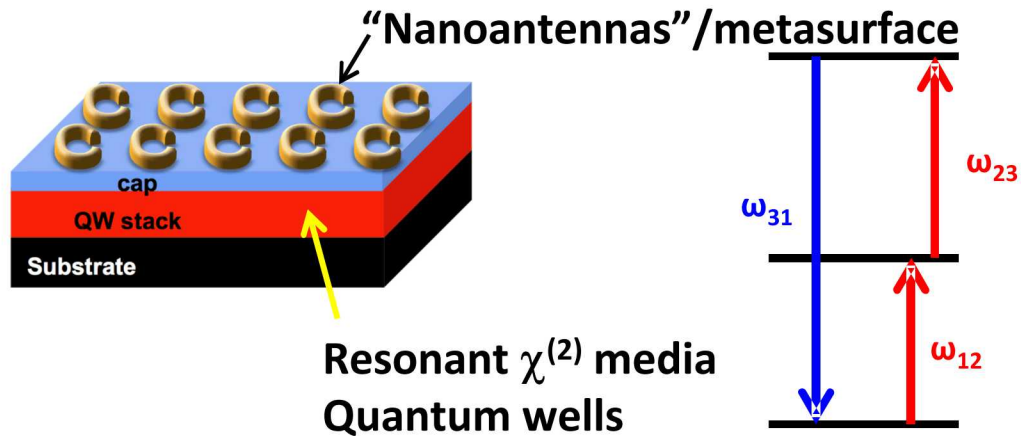
Simultaneous $\chi^{(2)}$, $\chi^{(3)}$, $\chi^{(4)}$ processes with no phase matching and comparable efficiency



FWM power dependence



Interlude: The Highest Nonlinearity from Metasurfaces (SHG)



Max efficiency:

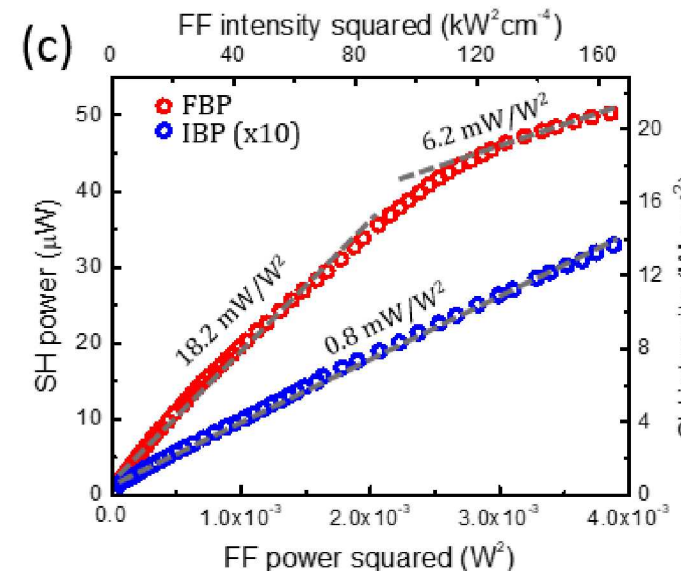
Brener (transmission): 2.3 mW/W²

Belkin (reflection, etched resonators): 17 mW/W²

Belkin & Brener 2018, 18.2 mW/W²

$$\chi_{\text{eff}}^{(2)} \sim 1.31 \times 10^6 \text{ pmV}^{-1}$$

Brener: Nature Communications 6, 7667 (2015)
Belkin & Alu, Nature 511, 65-69 (2014).





Nonlinear Metasurfaces: Comparison

	Metallic MS + Quantum Wells	Mie Resonator MS	
Reported nonlinearity	$\sim 10^{-2} \text{ W/W}^2$	$\sim 10^{-8} \text{ W/W}^2$	(PPLN $\sim 10^{-1}$)
“Material” $\chi(2)$	$\sim 200 \text{ nm/V}$	$\sim 200 \text{ pm/V}$	$I_{\text{SHG}} \sim (\chi^{(2)})^2$
→ Mode Depth	$\sim \lambda/20$	$\sim \lambda/n$	
Mode overlap and $\chi(2)$ utilization	**	* (**)	
→ Q	~ 10	~ 10 (1000)	$I_{\text{SHG}} \sim Q^2 \text{ or } Q^4$
→ Saturation and damage threshold	Low	High	
→ Wavelength scaling	$> 3\text{-}5 \text{ }\mu\text{m}$	visible to far IR	
→ $> 3^{\text{rd}}$ order	No (in mid IR)	Yes	

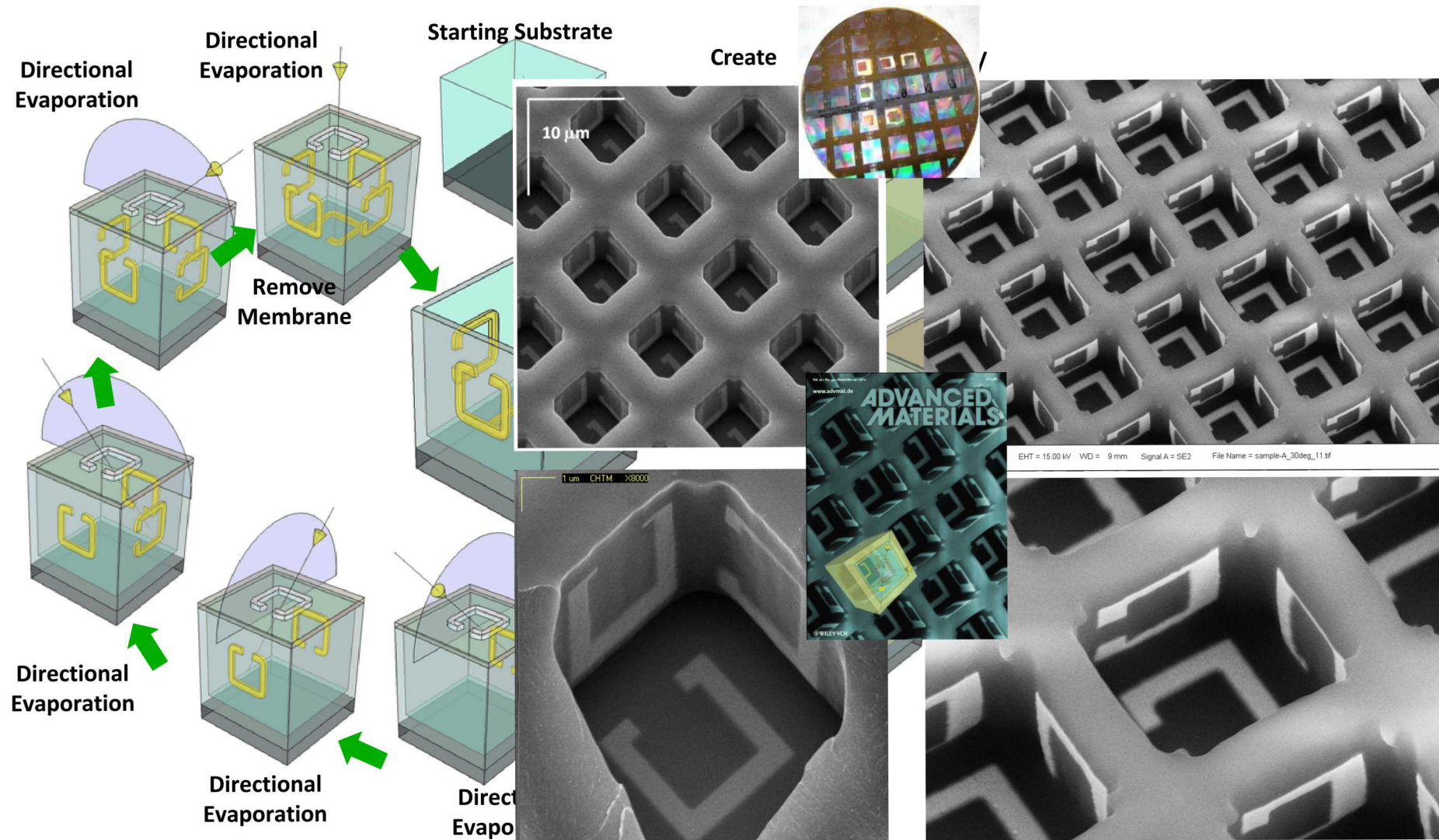


Outline

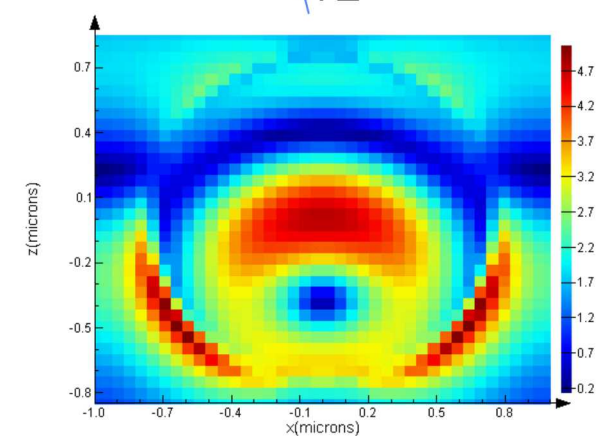
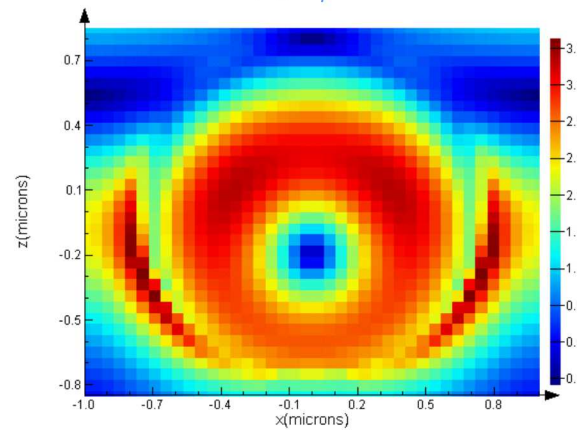
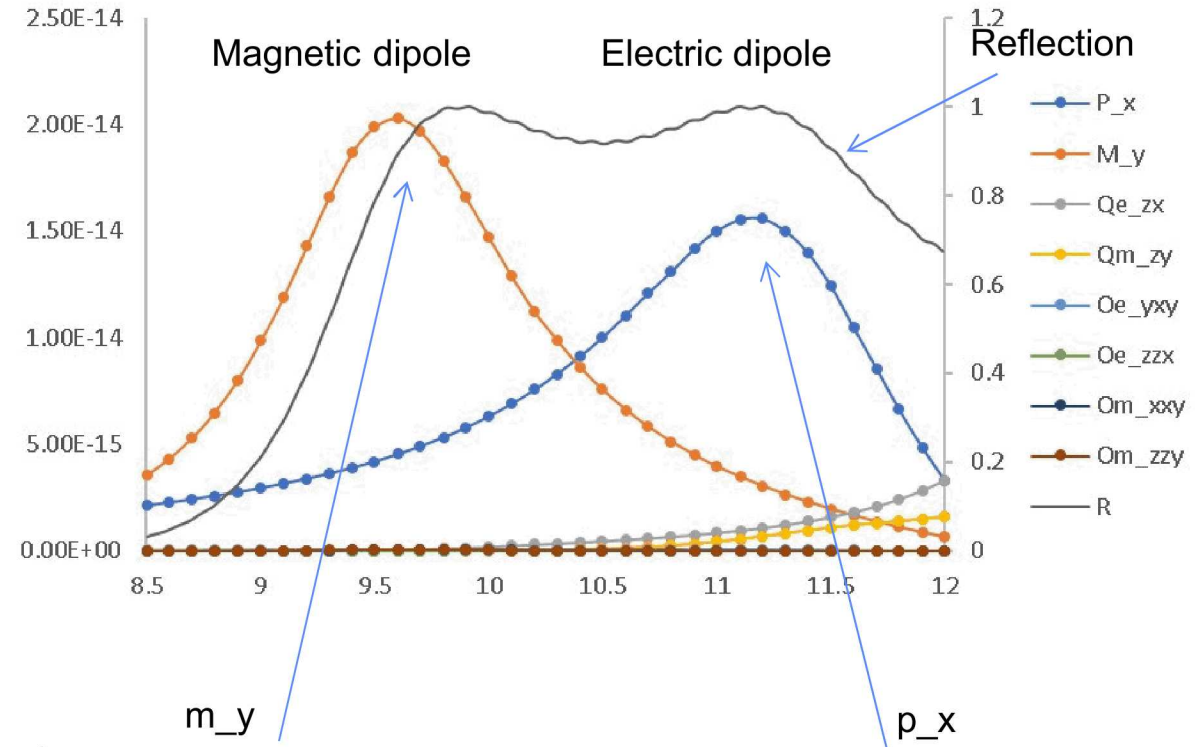
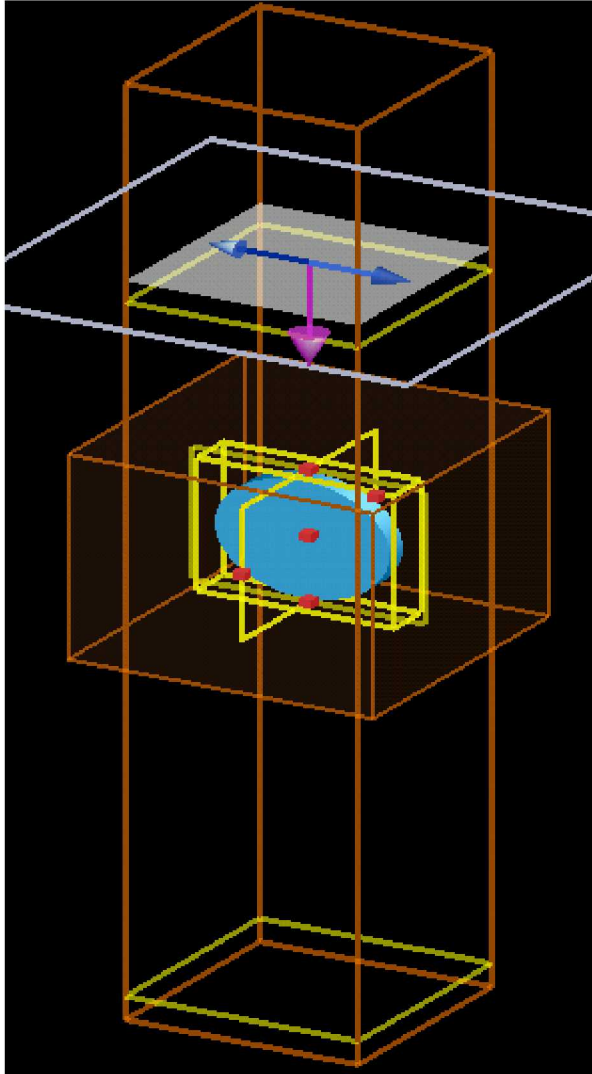
- Metamaterials & Metasurfaces: refresher, definitions, etc.
- Linear Metasurfaces:
 - Optical magnetic mirror
 - Huygens meta-optics
- Emitters + Metasurfaces
- Nonlinear optics with all-dielectric metasurfaces
- **Outlook on future (quasi) 3D fabrication**

Fabrication of 2D Metamaterials

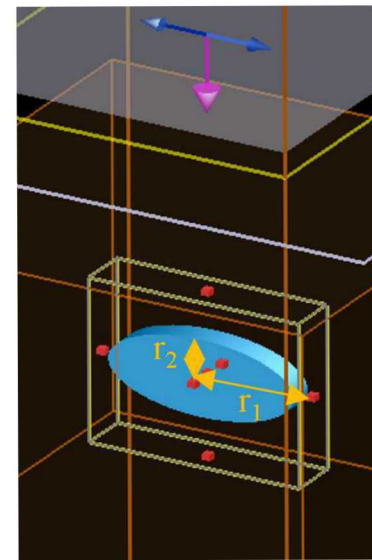
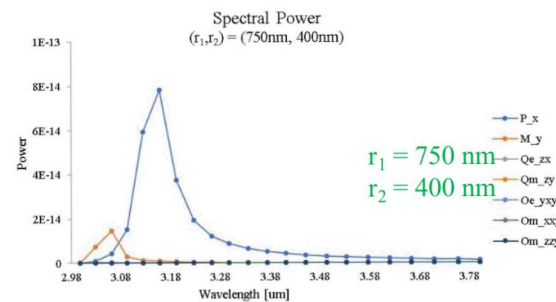
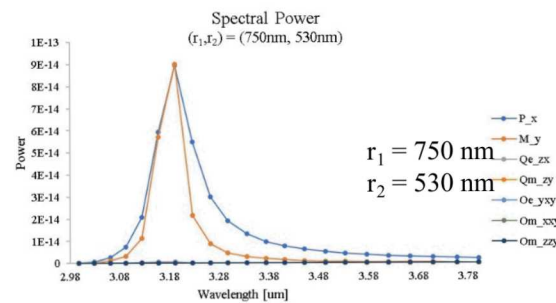
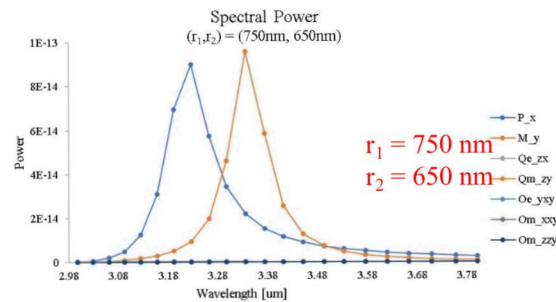
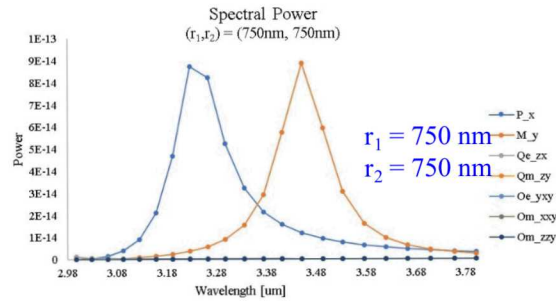
Membrane Projection Lithography



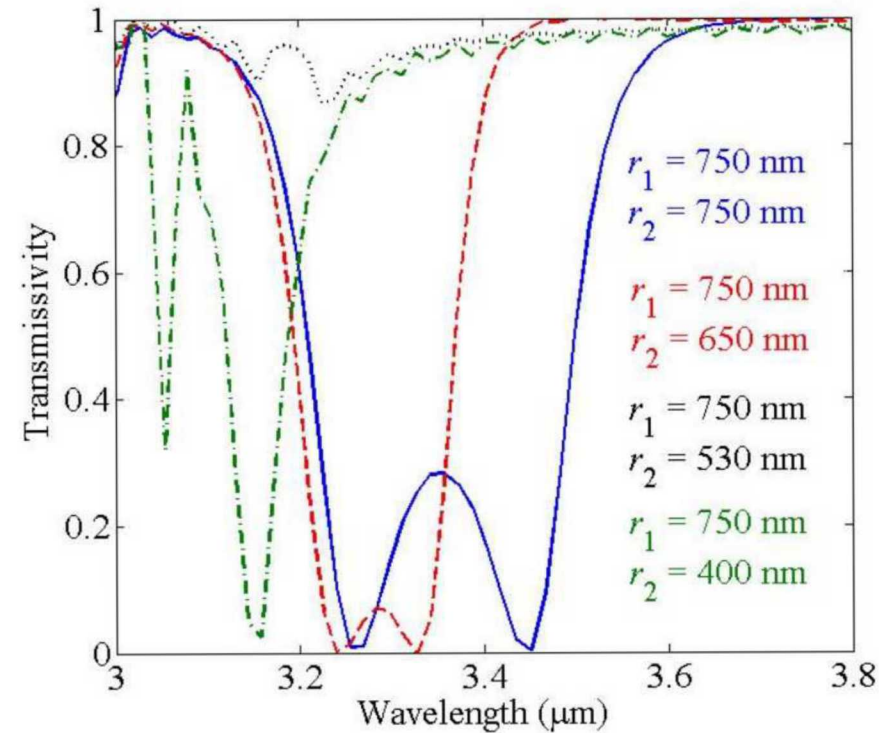
“Vertical” Dielectric Resonators



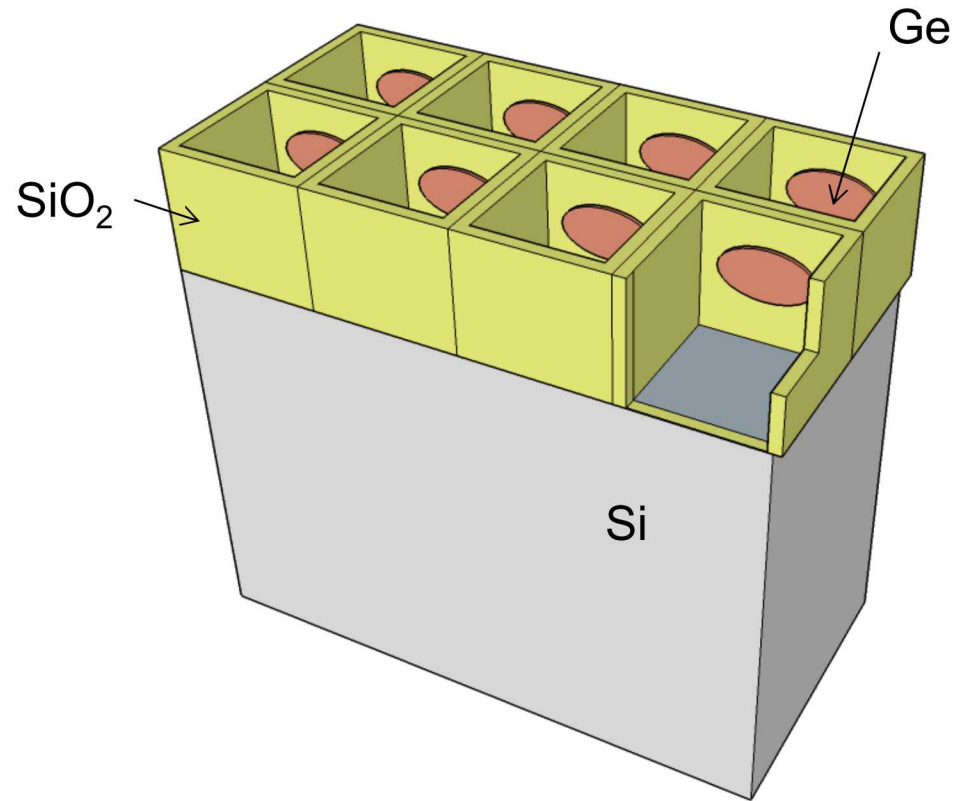
Achieving Huygens Point



3 um period
Resonator in free
space



Huygens Metasurfaces Using Out of Plane Resonators



- Elliptical resonators: Huygens behavior.
- Maybe we can stack several vertical resonators: $>2\pi$ phase shift
- All CMOS compatible, 1st devices in ~1 month



Summary

- **Metamaterials & Metasurfaces: refresher, definitions, etc.**
- **Linear Metasurfaces:**
 - Optical magnetic mirror
 - Huygens meta-optics
- **Emitters + Metasurfaces**
- **Nonlinear optics with all-dielectric metasurfaces:**
- **Outlook on future 3D fabrication**



Acknowledgments

Departed postdocs: Omri Wolf

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CINT + Sandia: Salvo Campione, Sheng Liu, John Reno, Greg Peake, John Klem, Mike Sinclair, Paul Clem, Jon Ihlefeld, Bruce Burckel, Jason Dominguez...

ANU (Australia): Yuri Kivshar, Dragomir Neshev, Katie Chong

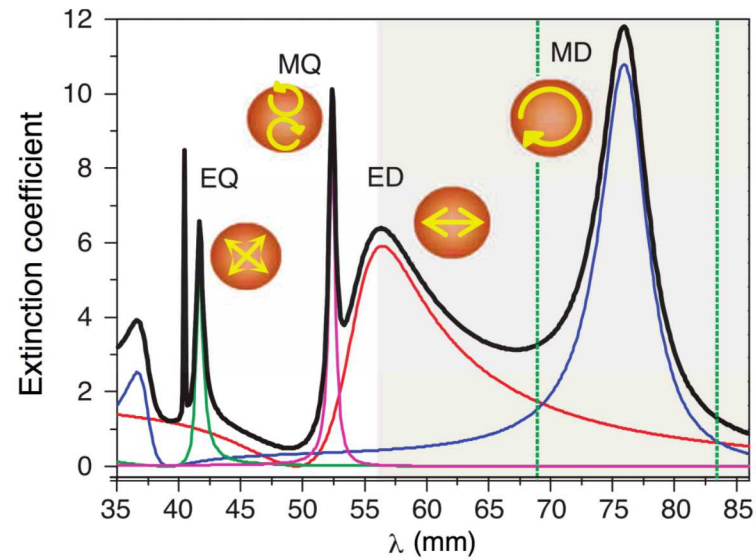
Jena (Germany): Isabelle Staude, Aleks Vaskin

UT Austin: Nish Nookala, Misha Belkin

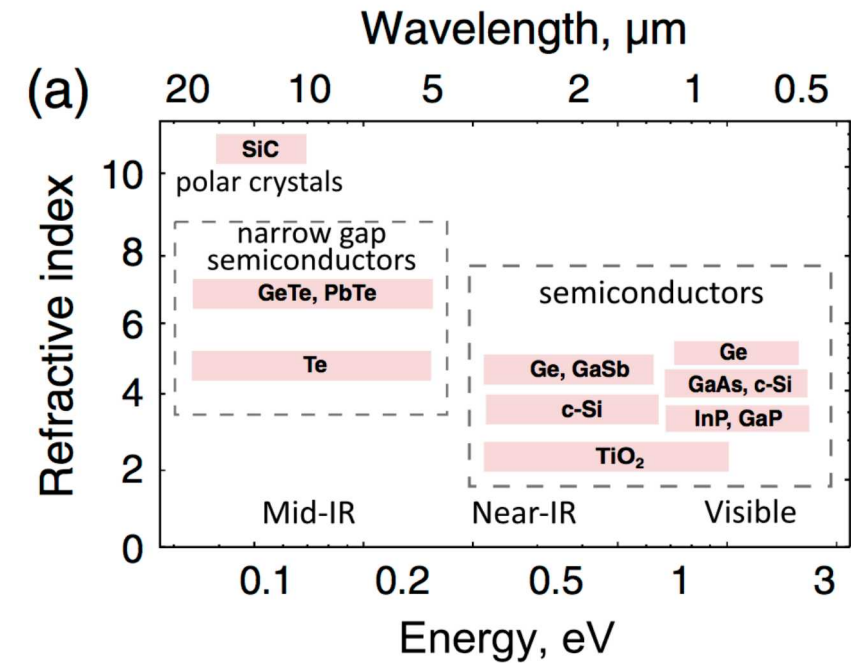
Moscow State (Russia): Maxim Shcherbakov, Andrey Fedyanin

Mie-resonance Dielectric Metasurfaces

Mie resonances in Dielectric spheres

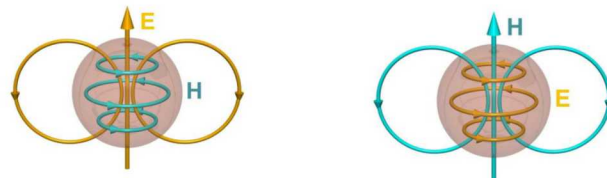


Geffrin, J.-M., et al. *Nat. Commun.* 3, 1171, **2012**



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

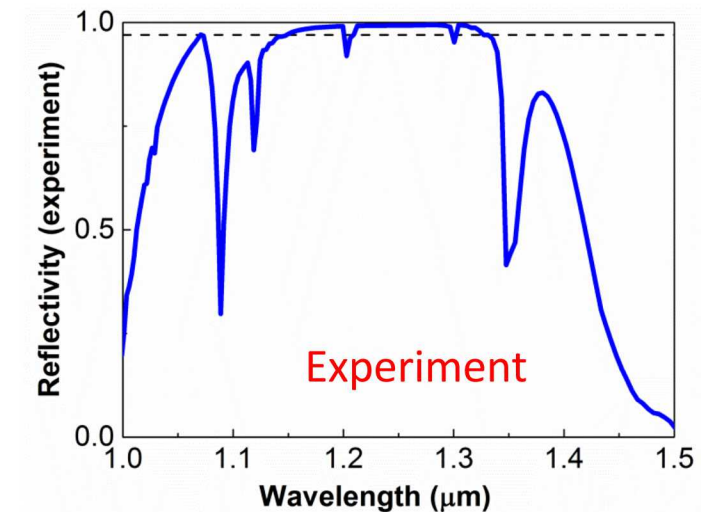
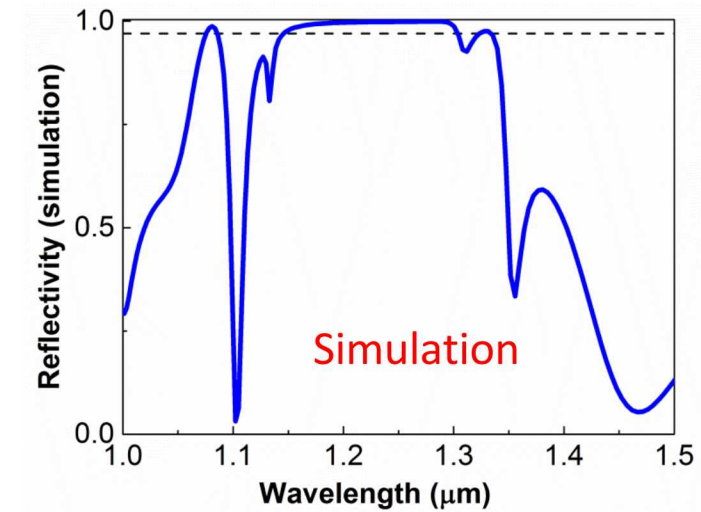
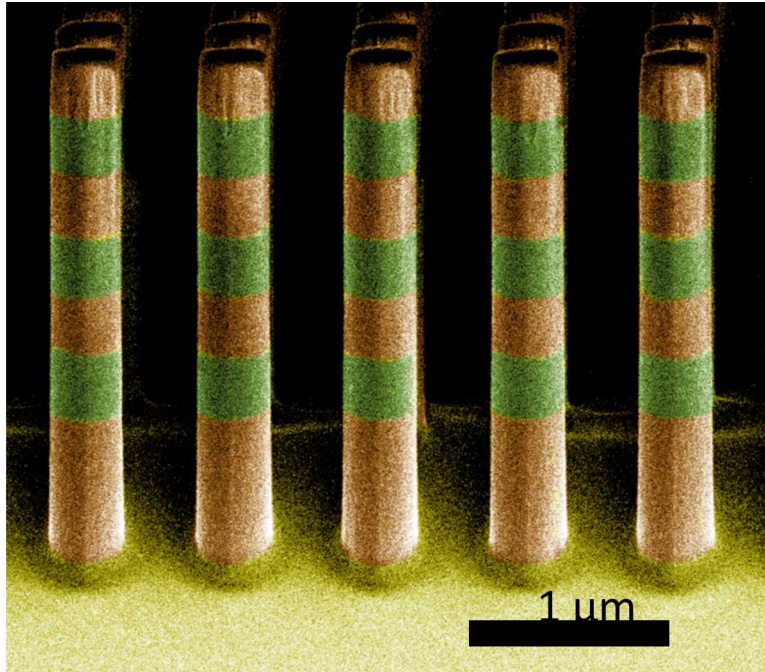
Electric Dipole Magnetic Dipole



Images: A. Miroshnichenko

Multi-layer GaAs Dielectric Metasurfaces

- Epitaxially grown 3X (AlGaAs + GaAs)
- Same fabrication steps as 1 layer

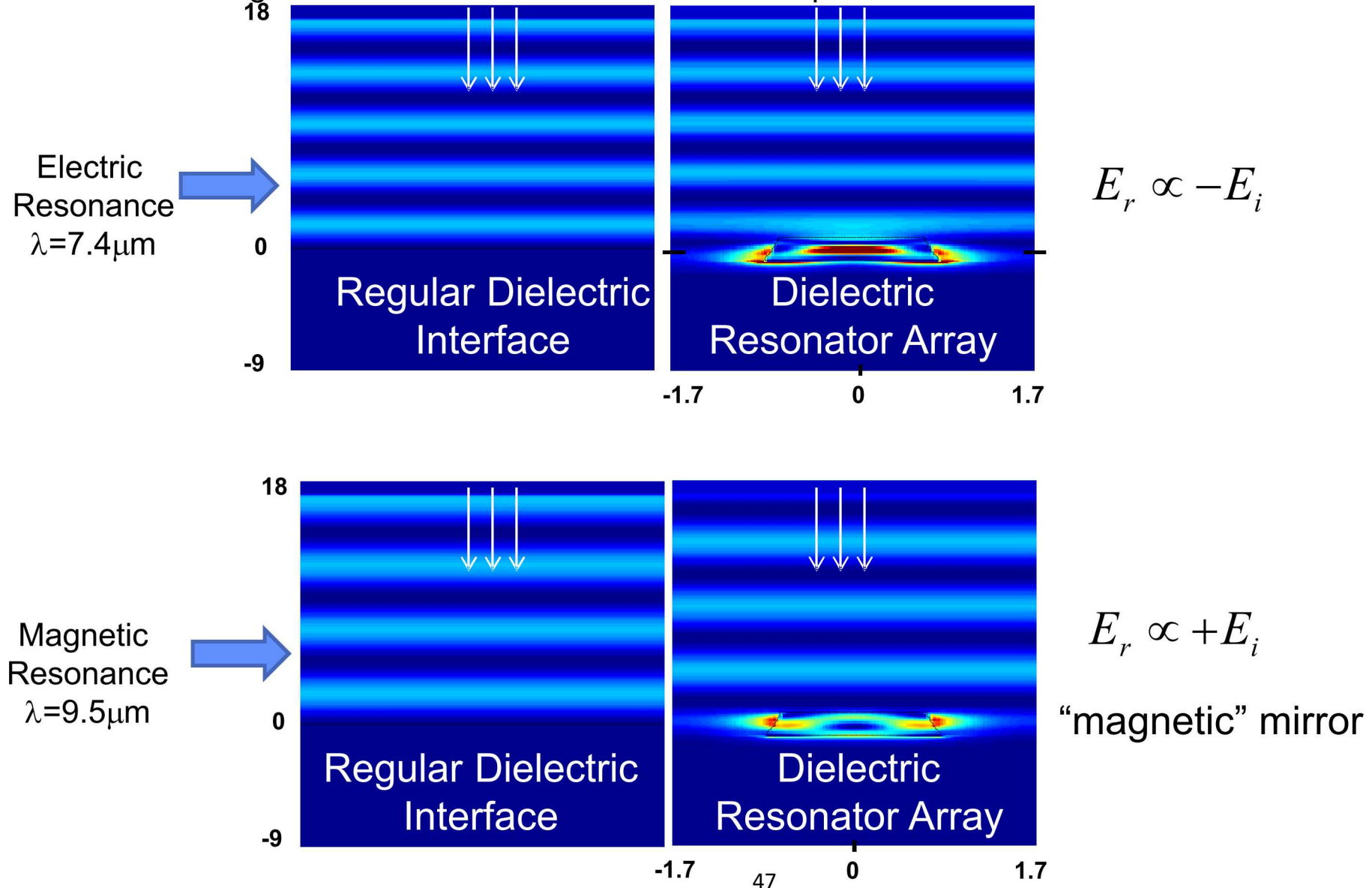


➔ Potential path to 3D dielectric metamaterials

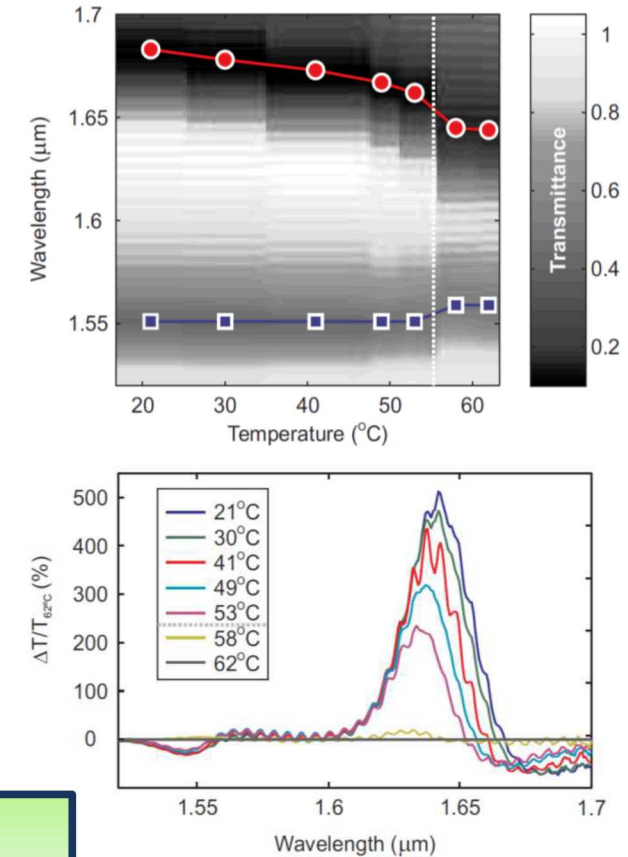
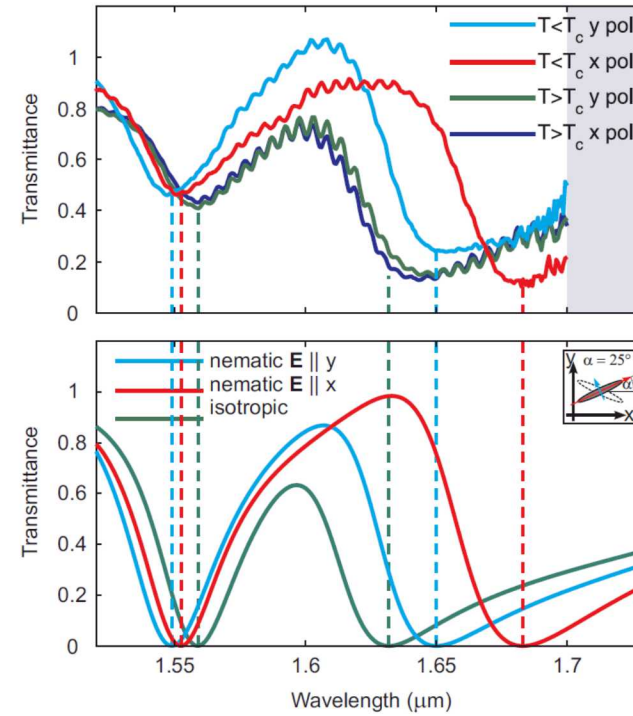
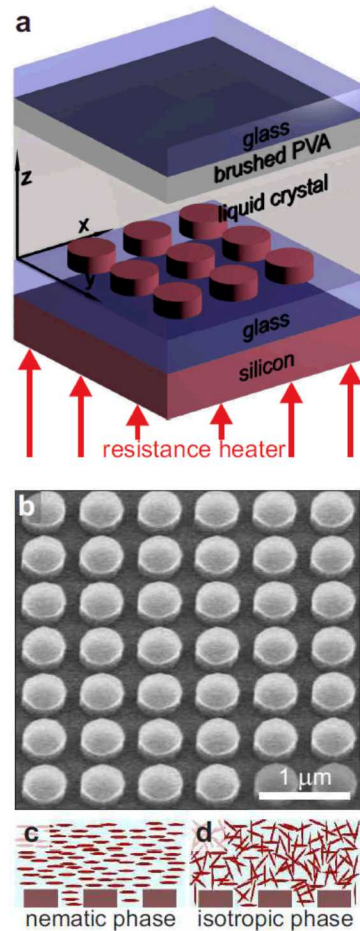


Tellurium Cubic Resonators: FDTD Simulation of the Reflected Wave

Simulated using as-made dimensions and measured optical constants

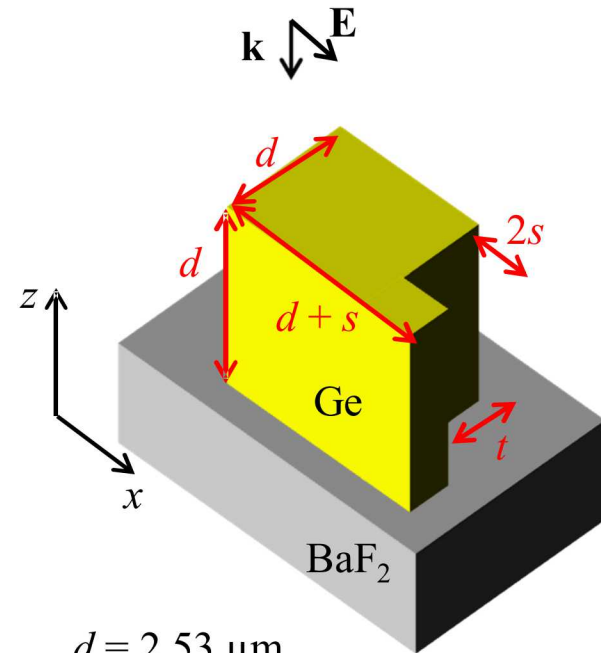


Dynamic Control Using Liquid Crystals



- Tuning range 40 nm
- Tuning contrast 500%

Ge-based Fano metasurface design



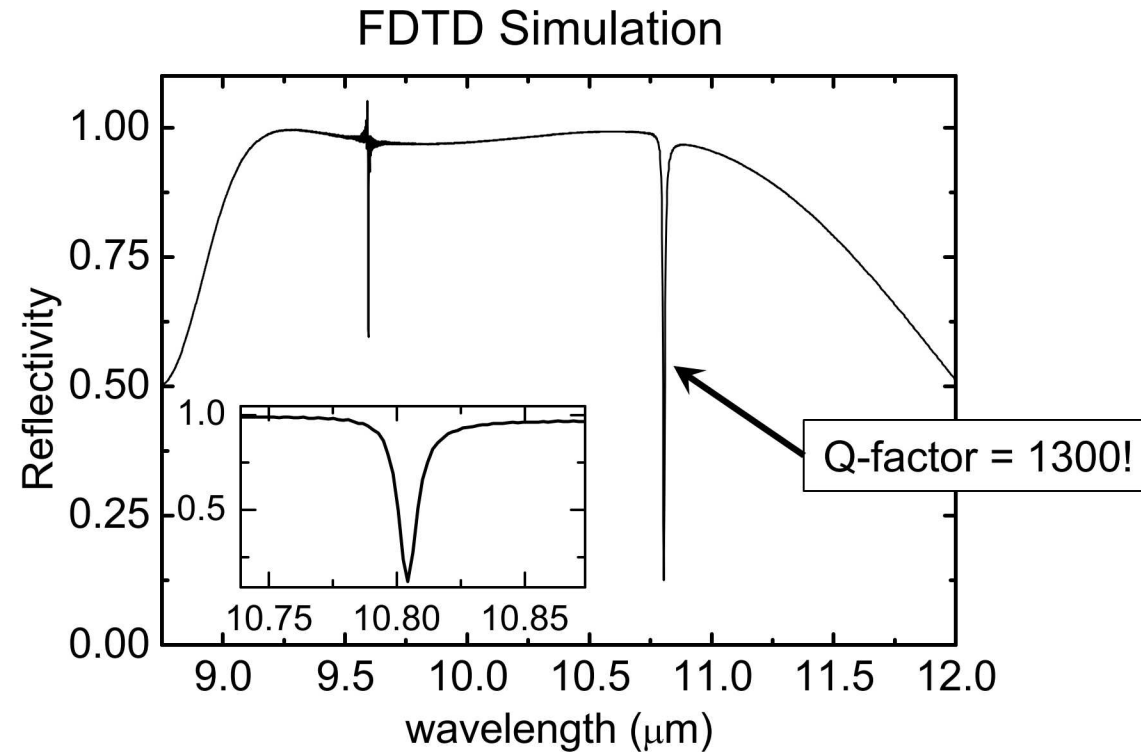
$$d = 2.53 \mu\text{m}$$

$$s = d / 5$$

$$t = 1.5 \mu\text{m}$$

Array Pitch:

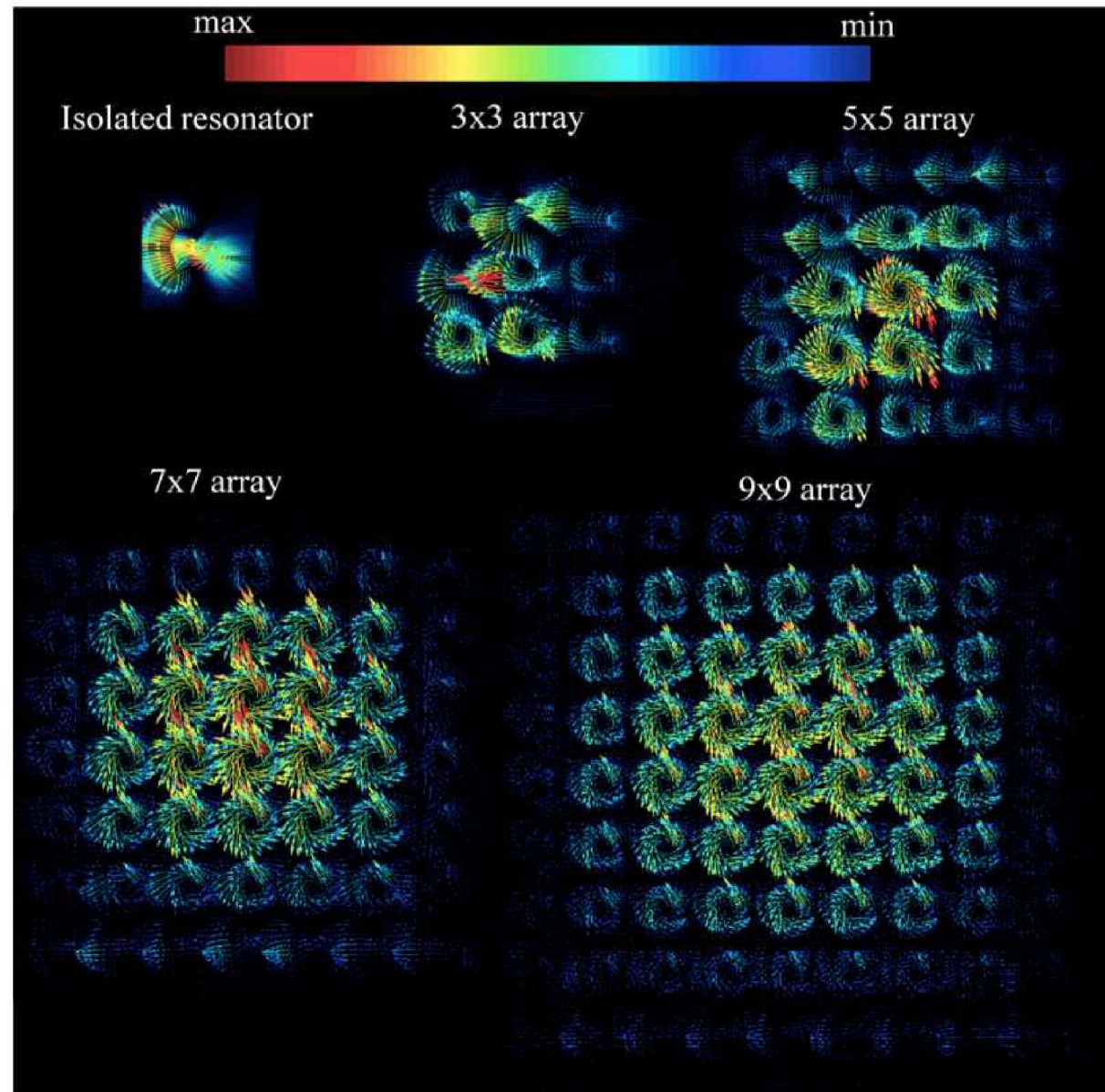
$$a = b = 4.2 \mu\text{m}$$



Design is scalable from near infrared through RF

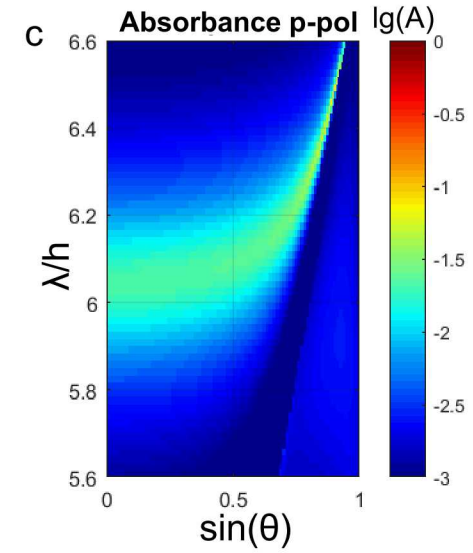
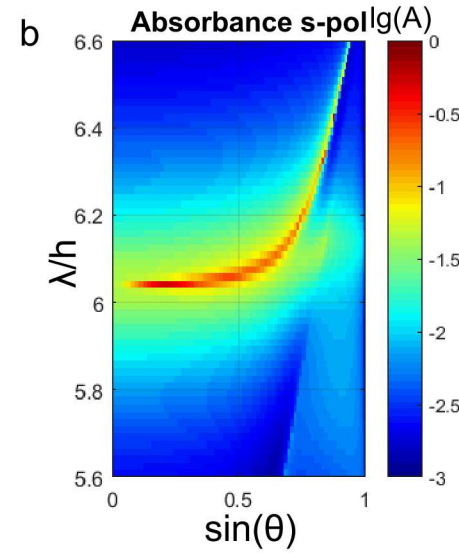
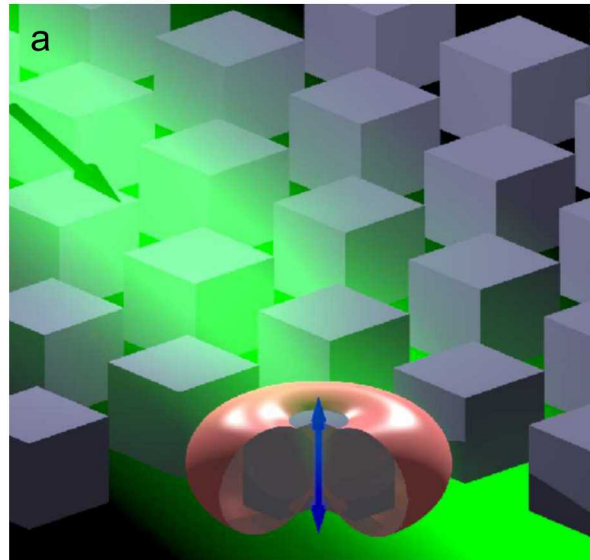


High Q: Only a small array is needed

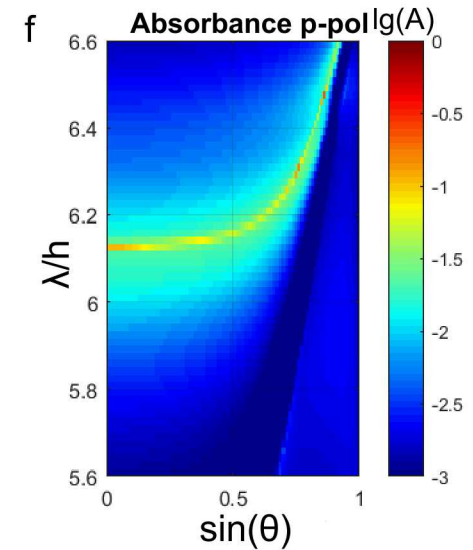
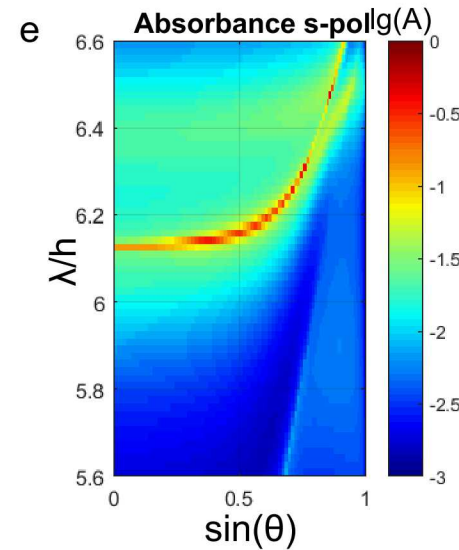
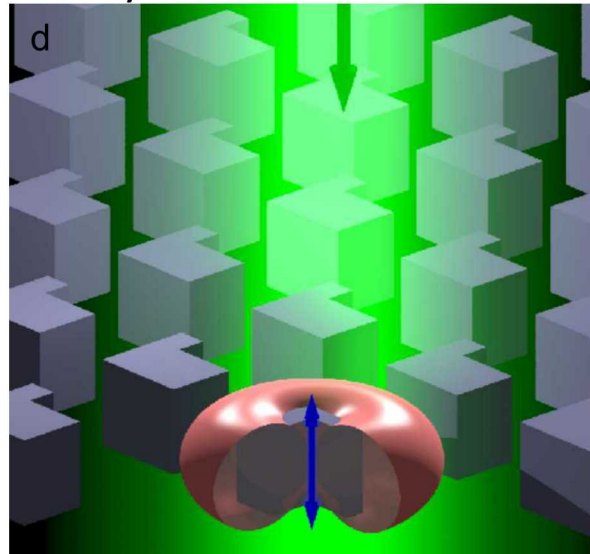


Out of Plane Dipole Modes

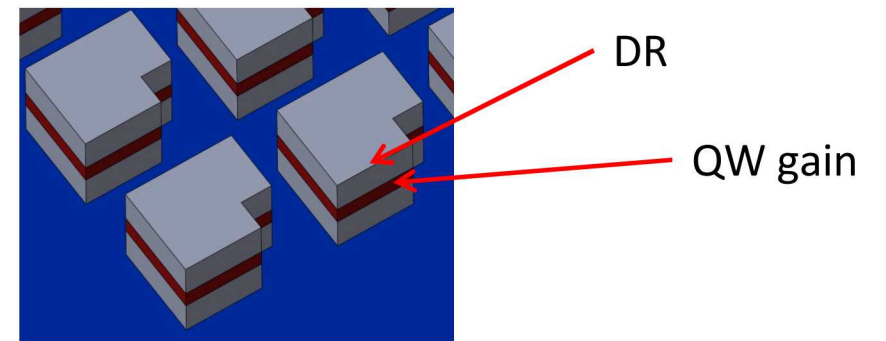
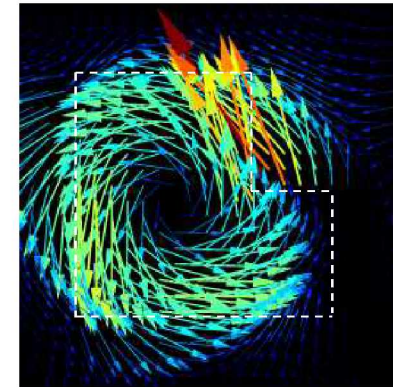
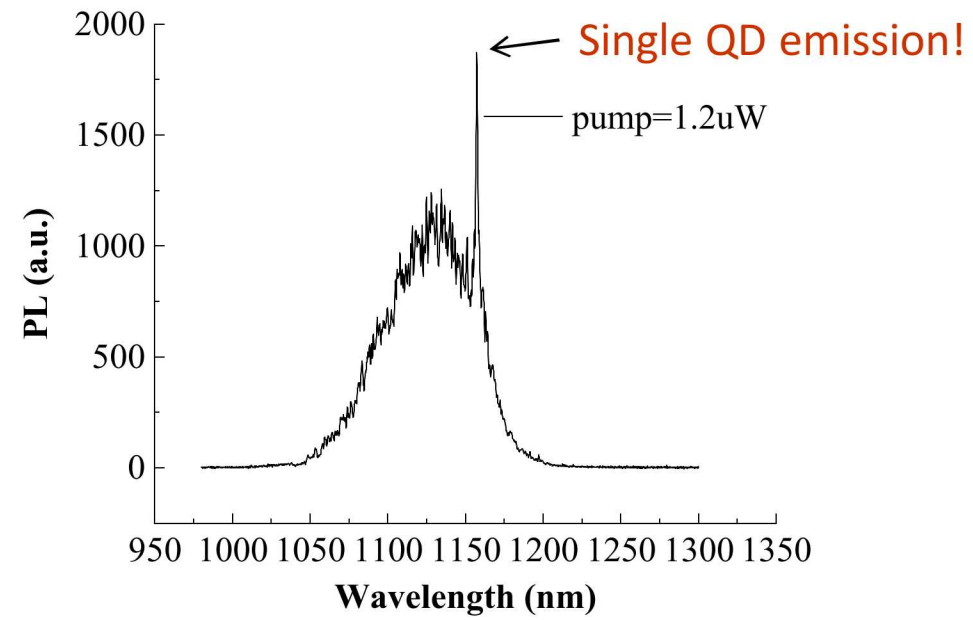
Symmetric Resonators



Broken-symmetry Resonators

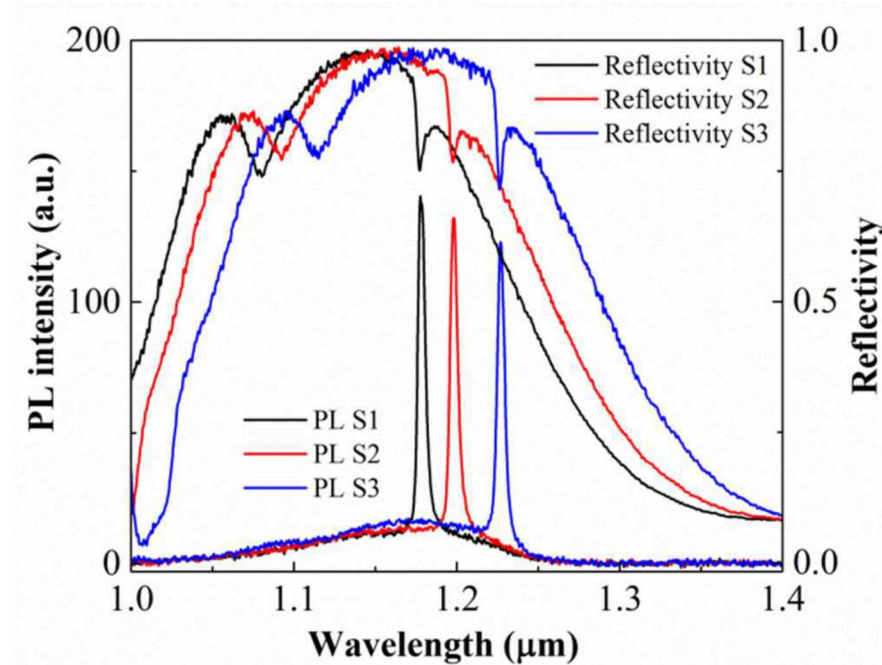
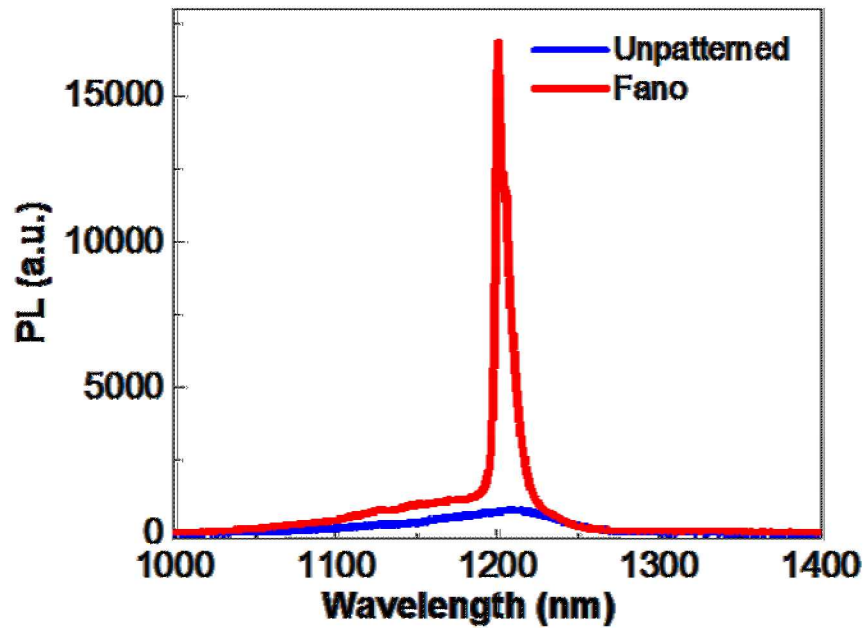
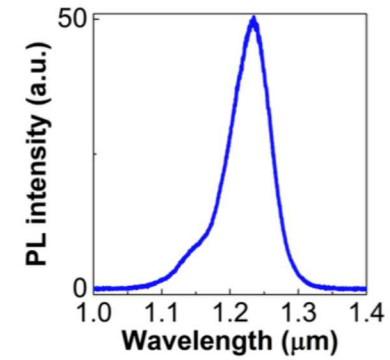
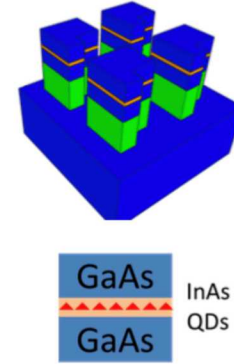
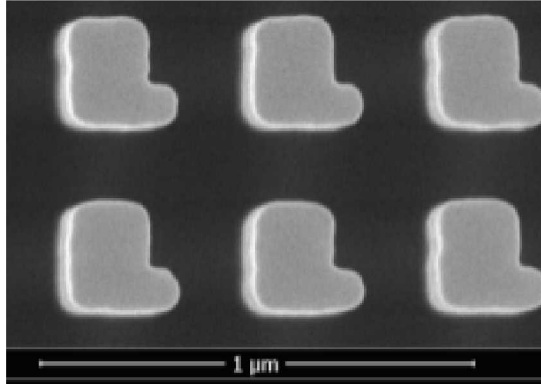


Low temperature PL



Emitters (QDs) Coupled to “Fano” Metasurfaces:

QDs inside, Emission enhancement

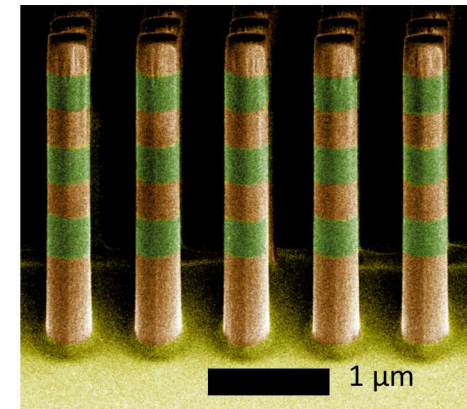
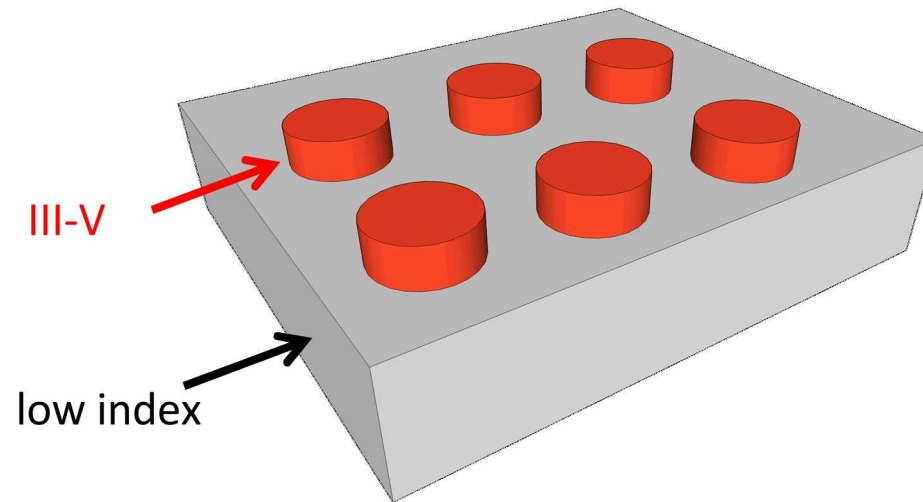


Nonlinear Metasurfaces: Silicon vs III-V's

- Silicon is centrosymmetric → NO second order nonlinearity
- Lots of III-V semiconductors have $\chi^{(2)}$

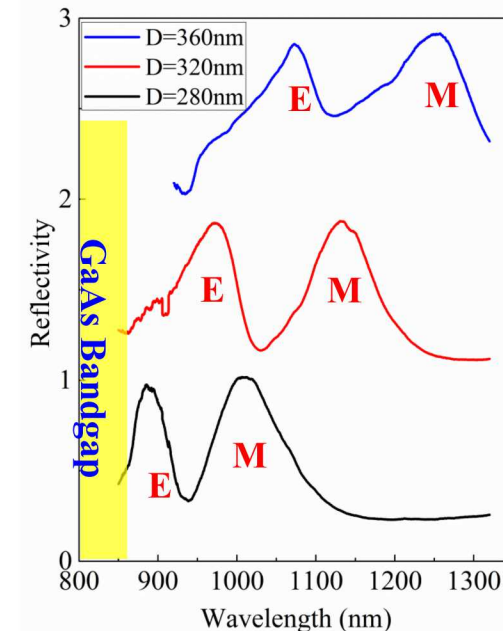
GaAs: ~200pm/V

Material	d _{eff} (pm/V)
MgO:PPLN	14pm/V (typical)
KTP	3.4pm/V
BBO	2.5pm/V
LBO	0.85pm/V

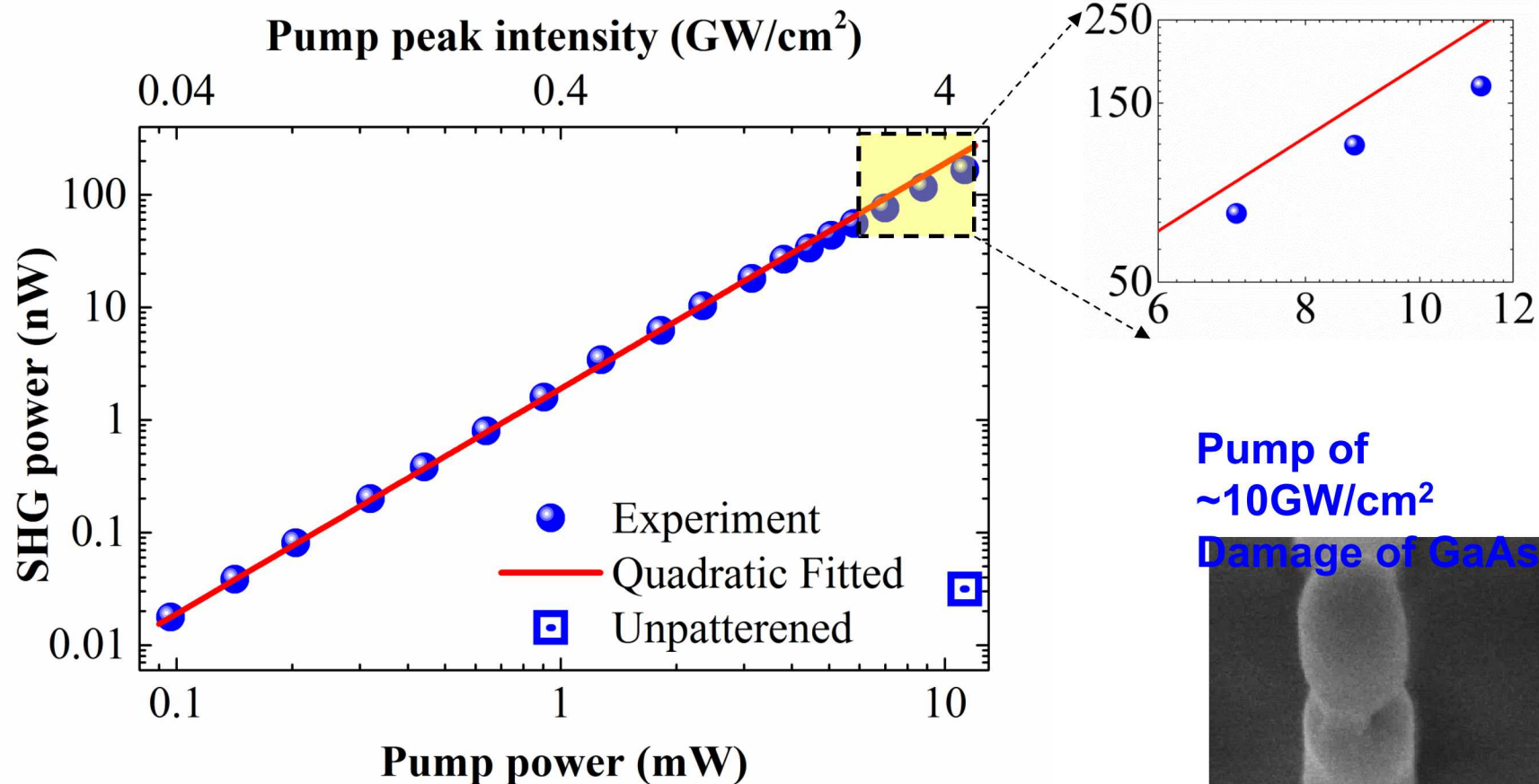


How to Increase SHG Efficiency from all-Dielectric Metasurfaces

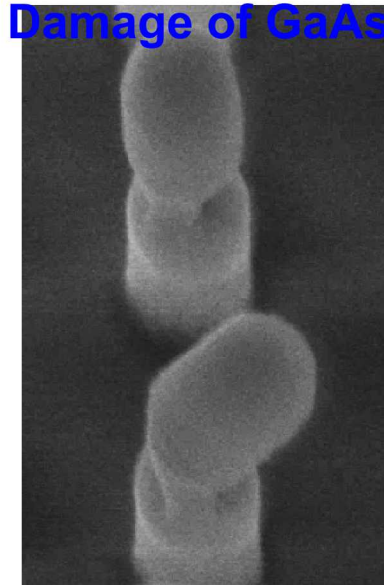
- Find another high index material with higher $\chi^{(2)}$ 😞
 - GaAs already has $\chi^{(2)} > 200 \text{ pm/V}$
- Find combination of modes and symmetry that maximize nonlinear polarization $P_{NL,x}^{2\omega} \propto 2\chi_{xyz}^{(2)} E_y^\omega E_z^\omega, \dots$ 😞
- Increase Q of resonances ($E_{inside} \propto Q$) 😊



SHG power vs pump power



Pump of
~10GW/cm²
Damage of GaAs

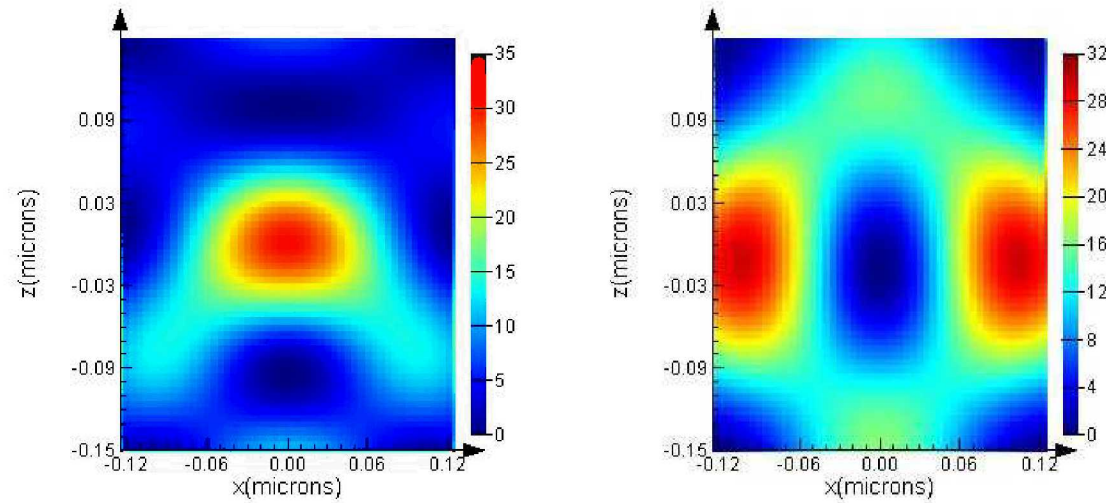


- Quadratic relationship sustained at lower pump power.
- Irreversible damage of GaAs occurred > ~1.5 GW/cm².
- Two-photon-absorption of GaAs followed by thermal damage due increased free carrier absorption

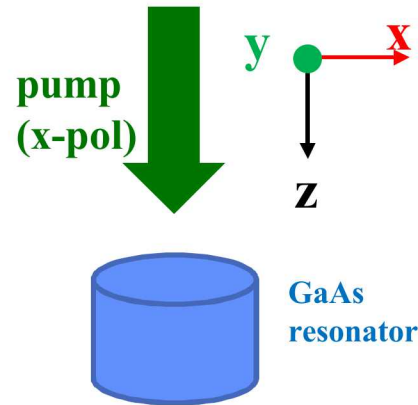
Polarization, Selection Rules

$|E|^2$ enhancement for both electric and magnetic dipole resonance.

($I_{2\omega}$ proportional to I_{ω}^2)

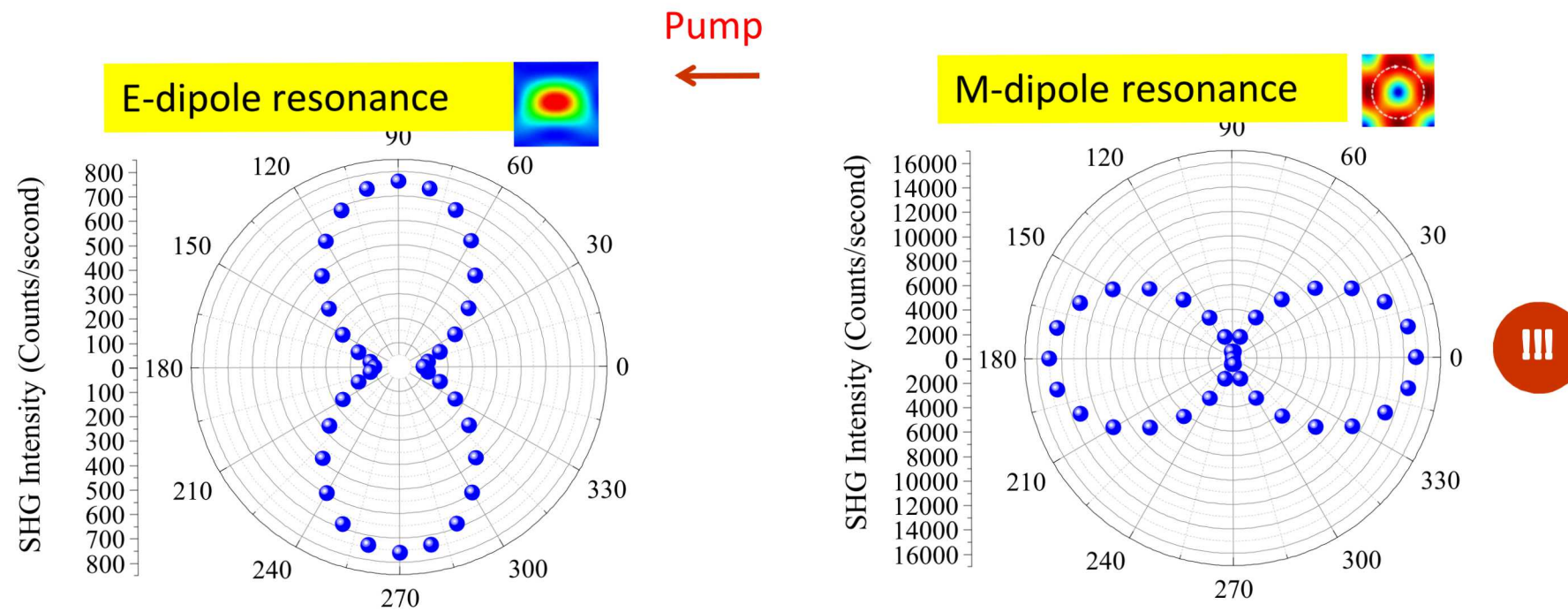


There are only E_x and E_z component inside the GaAs resonators.



$$E_y^{2\omega} = 2\chi_{xxy}^{(2)} E_x^{\omega} E_z^{\omega}$$

Orthogonal SHG polarization compared to the pump is expected



This is not consistent with bulk $\chi^{(2)}$

What happens at the M-dipole resonance?

1. Surface nonlinearity changes symmetry to $mm2$

$$E_x^{2\omega} = 2\chi_{xxz}^{(2)} E_x^\omega E_z^\omega$$

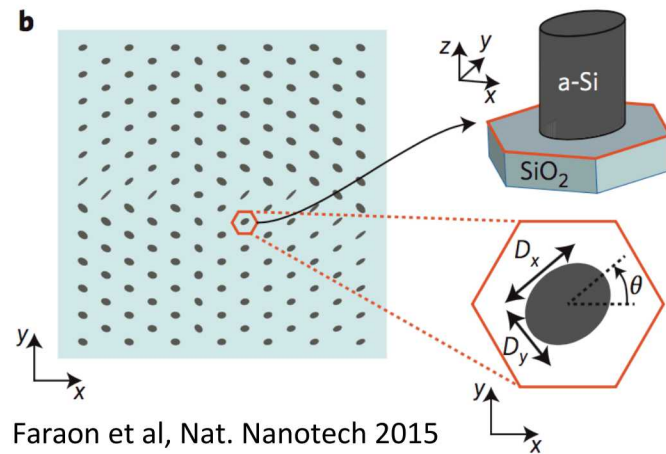
can be large, considering the large field enhancement at the surface for this mode.

2. SHG is above bandgap of GaAs, absorption favors SHG generated at the surface.

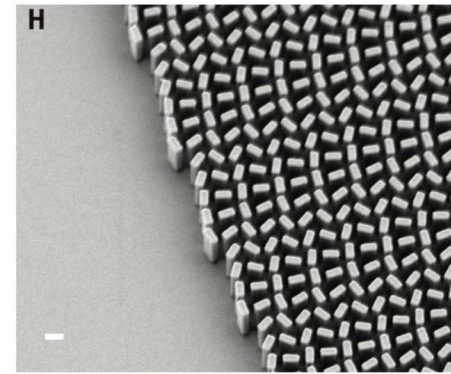
Not predicted: Carletti *et al.*, "Enhanced second-harmonic generation from magnetic resonance in AlGaAs nanoantennas", *Optics Express*, 23, 26544 (2015)

School #2: High Aspect Ratio Dielectric Metasurfaces for Flat Optics

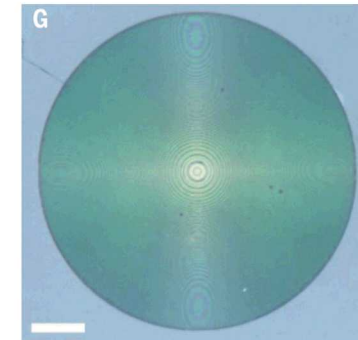
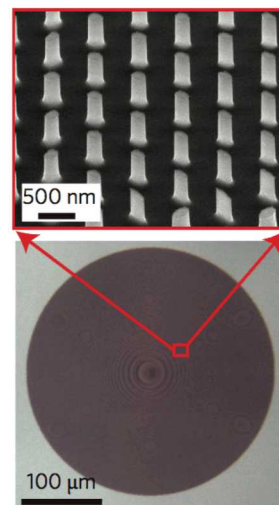
Use “tiny waveguides”, birefringent because of asymmetrical cross section.
Phase given by orientation (Faraon, Hasman, Capasso)



Faraon et al, Nat. Nanotech 2015

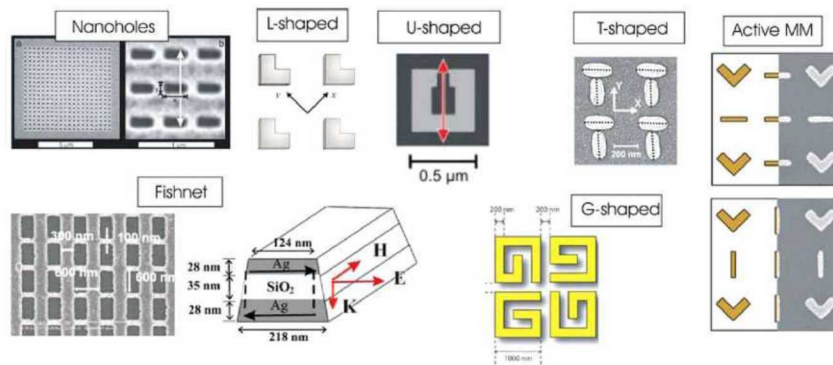


Capasso et al, Science 2016



Nonlinear Optics from Plasmonic Metasurfaces

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

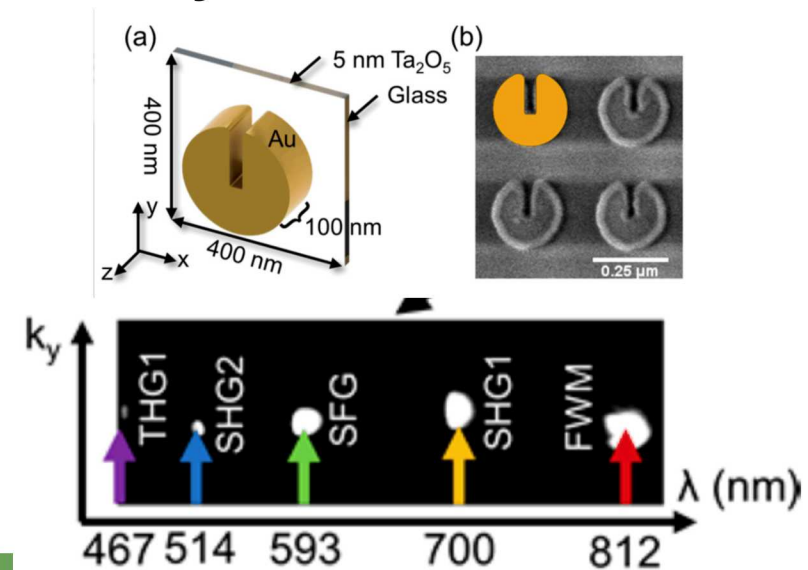


Y. Kivshar et al, Laser Photonics Review, **9**, 195 (2015)

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

Nonlinear plasmonics

Martti Kauranen¹ and Anatoly V. Zayats²

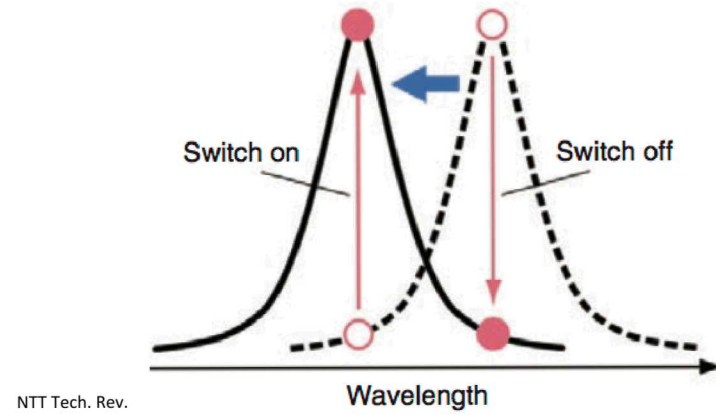


Sartorello, et al. *ACS Photonics* 3.8 (2016): 1517-1522.

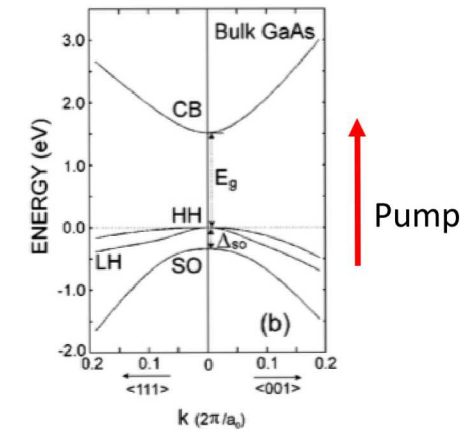
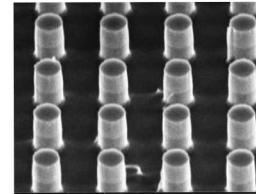
SHG efficiency $\sim 10^{-9}$

**Plasmonic structure: small mode volume
(usually only uses surface nonlinearities)**

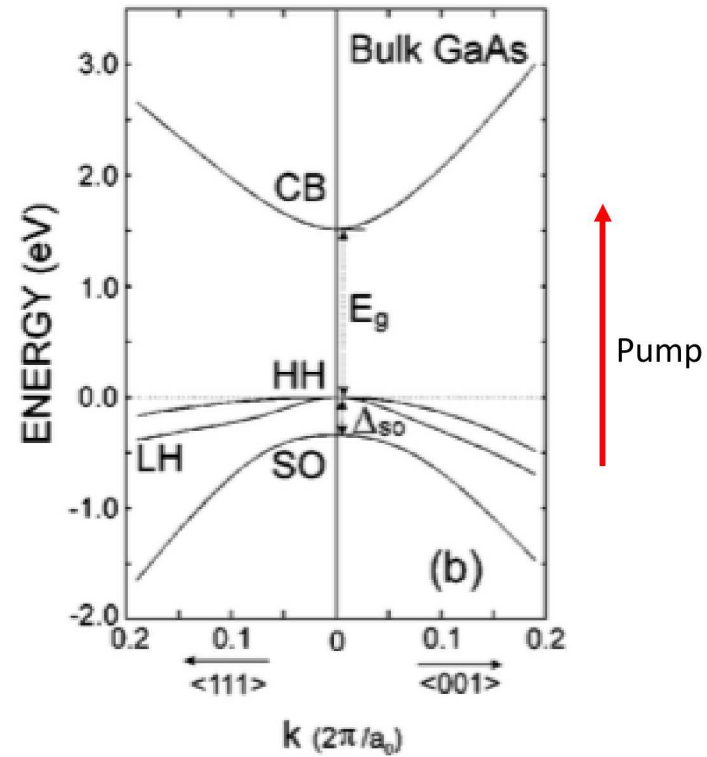
Ultrafast Switching Using Metasurfaces



• Dielectric Metasurfaces

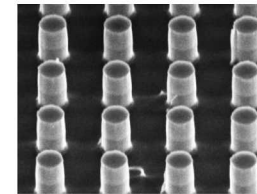


Vurgaftmana
JAPN (2001)

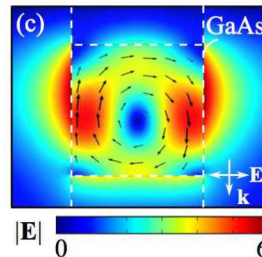
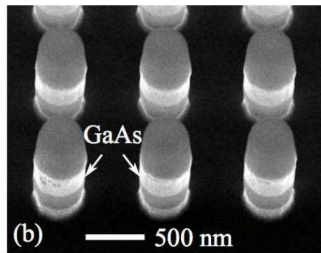
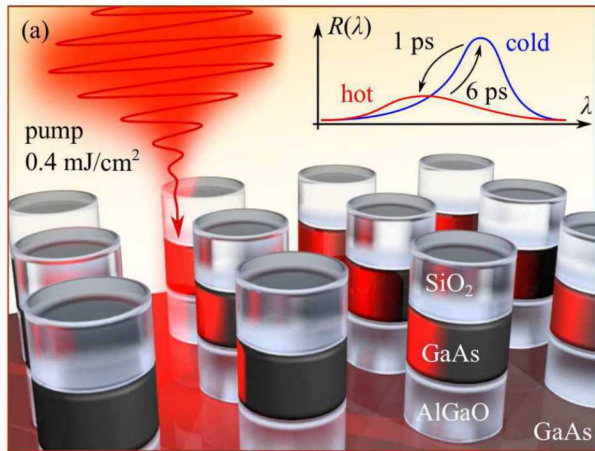


To first order, the laser beam gets absorbed, generates electrons and holes, and these cause a refractive index change.

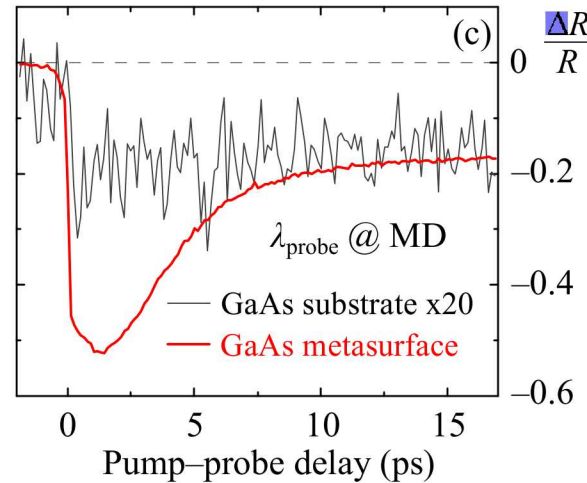
Then the Mie resonances will change (spectrally) since resonances occur at $\sim \lambda/n$



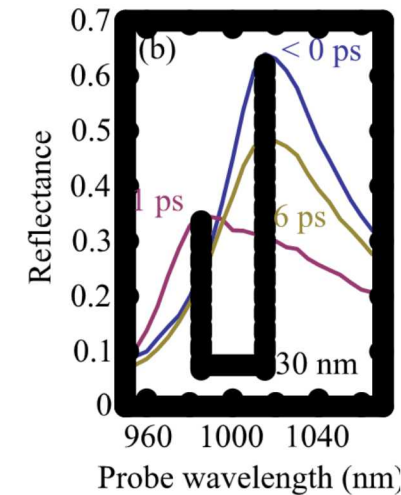
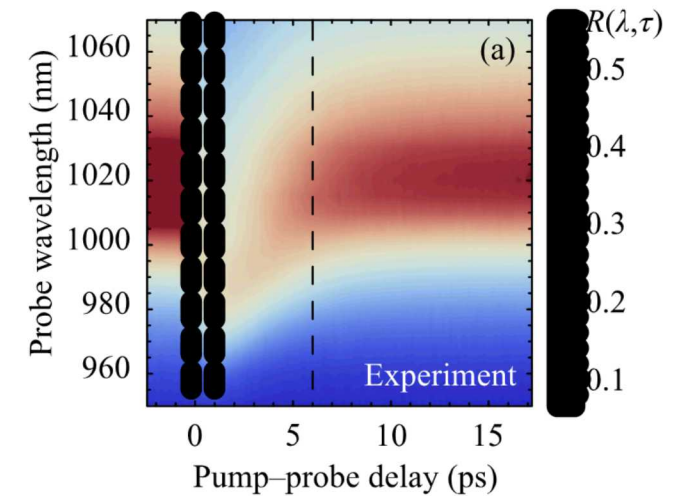
Dielectric Metasurface: Ultrafast Tuning of Magnetic Dipole Mode



Modulation 50x of substrate



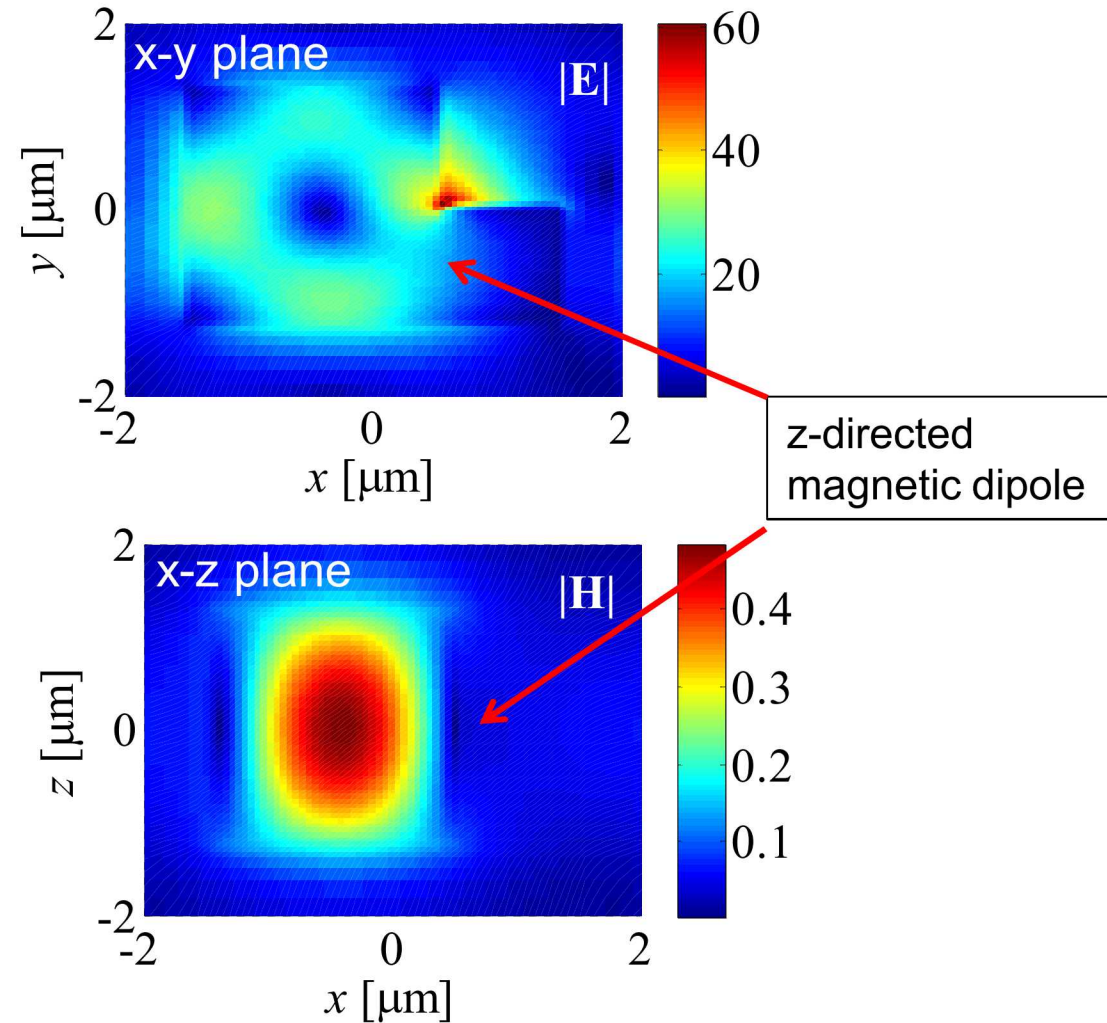
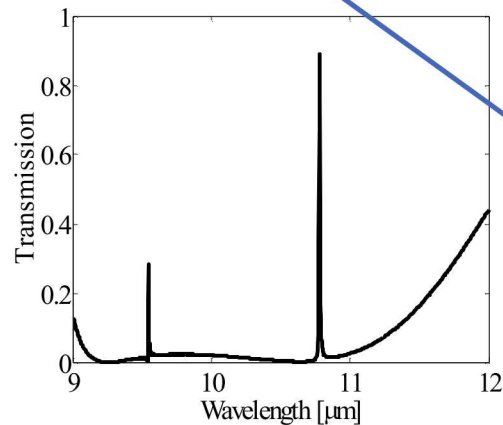
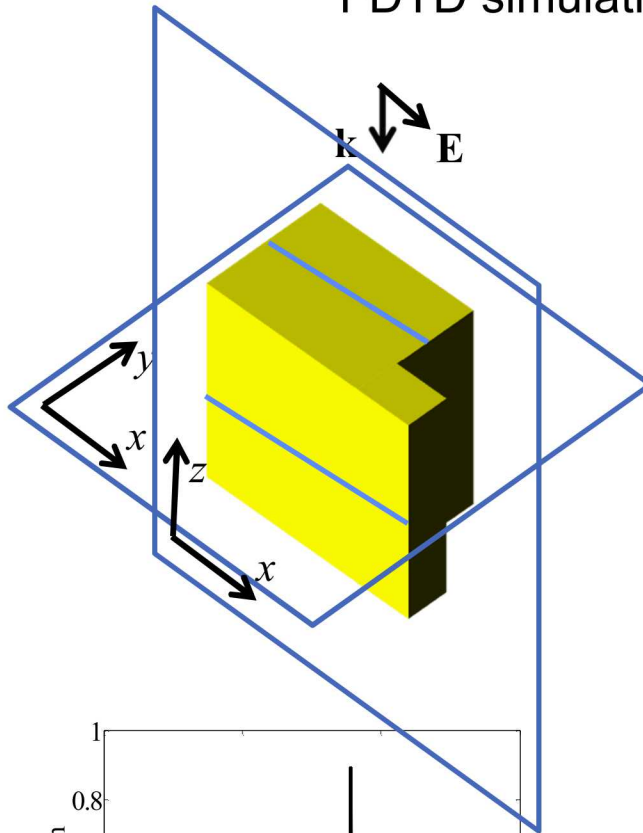
Resonance tuning by 30 nm



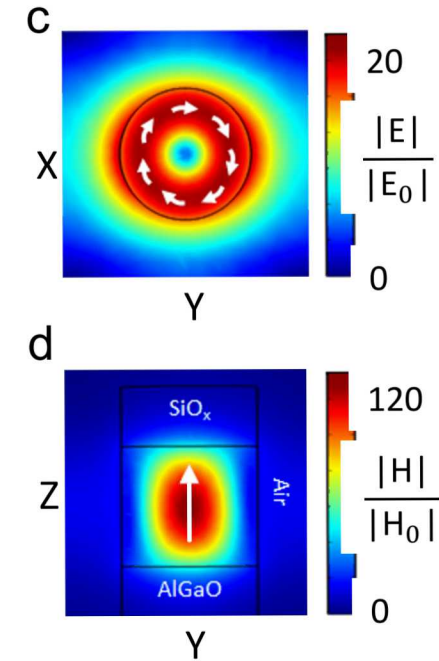
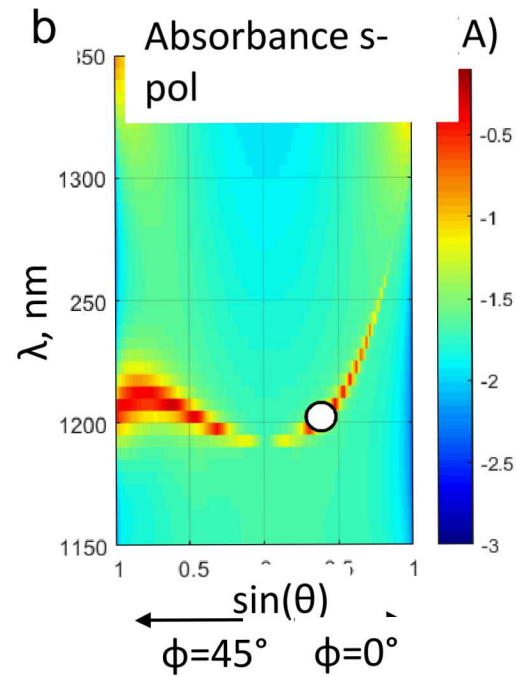
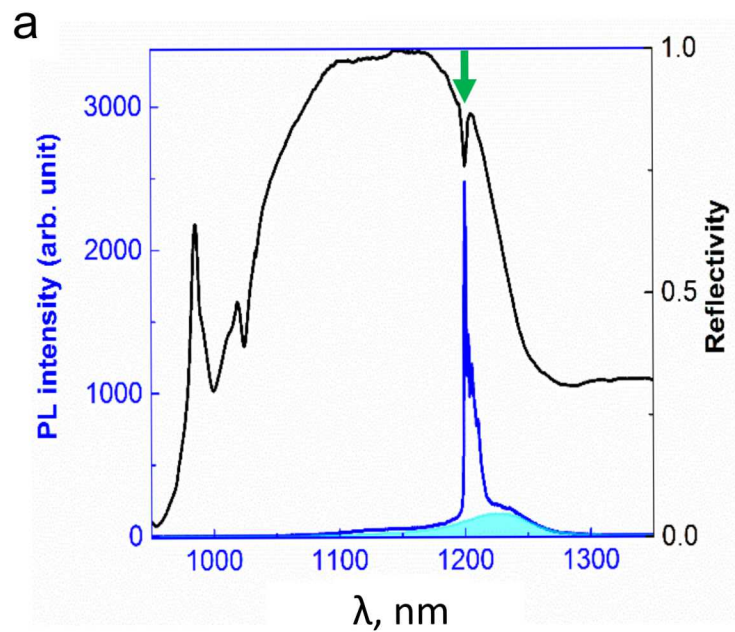
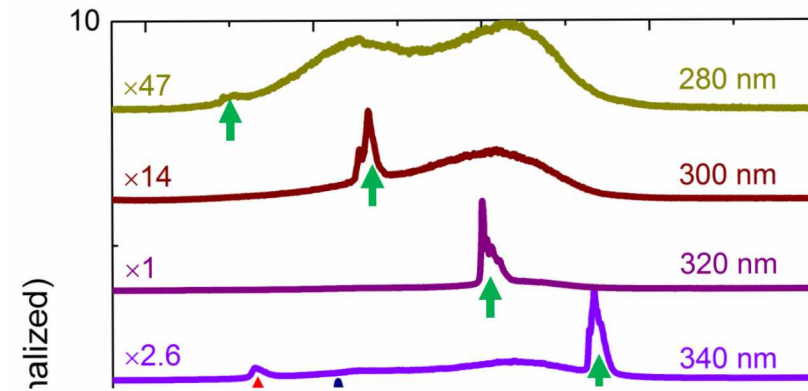
- Fast recovery due to surface recombination (~ 2.5 ps from low power expts)
- Maximum index modulation is -0.14 due to Drude dispersion and band filling effects

Resonant Mode Field Profiles

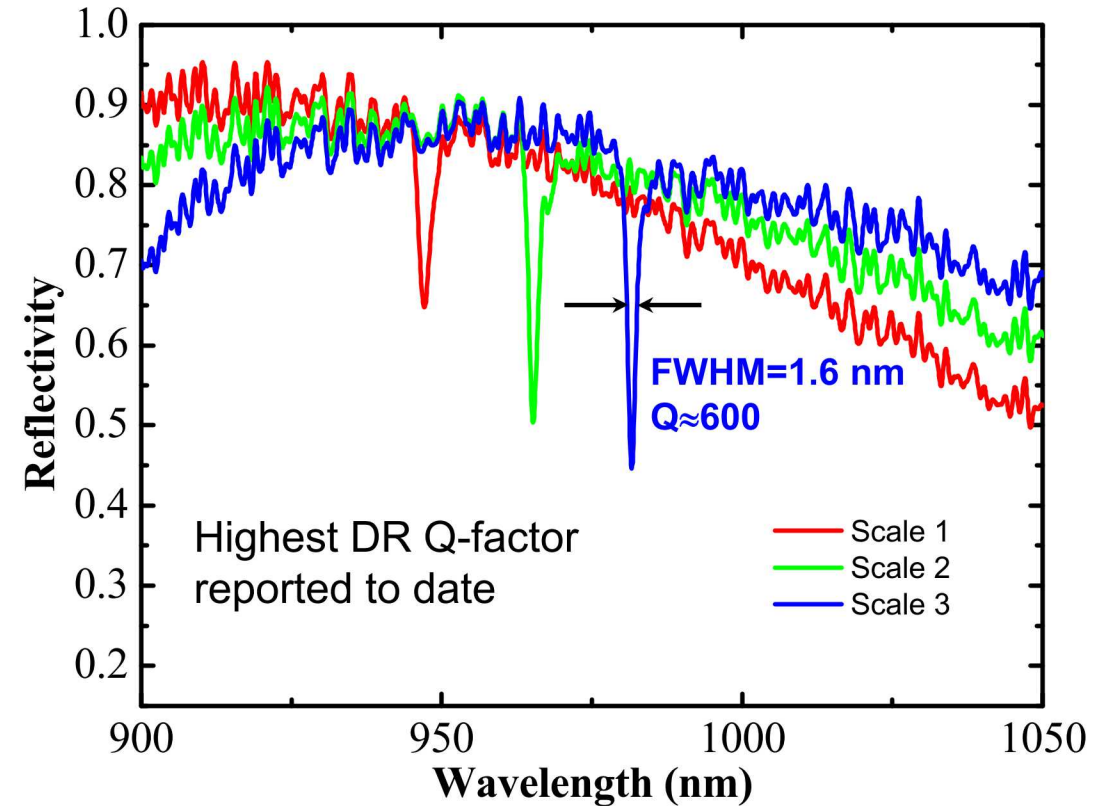
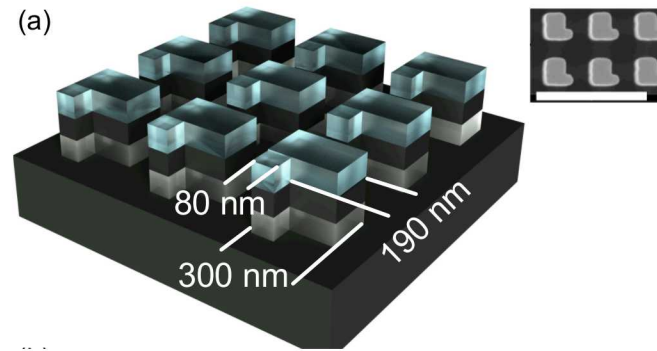
FDTD simulations at the $\lambda = 10.8 \mu\text{m}$ resonance



Photoluminescence



GaAs Fano Resonators: $Q \approx 600$!



- GaAs is direct bandgap $\rightarrow$ lower absorption losses
- GaAs has a large $\chi^{(2)}$ $\rightarrow$ nonlinear devices (SHG, down-conversion, etc.)
- Can incorporate InGaAs quantum wells for gain and photon detection