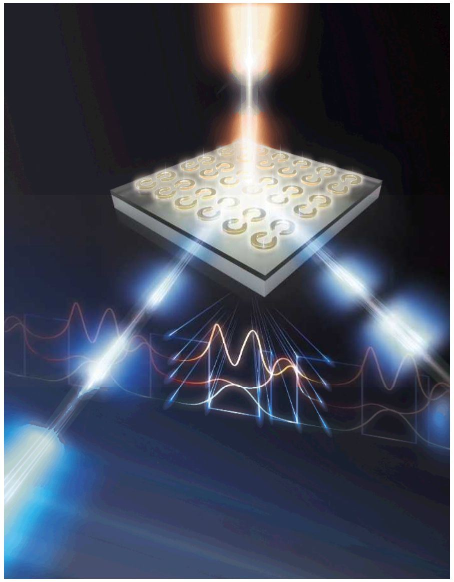
SAND2015-10658C

Nonlinear Optics with Metasurfaces



Igal Brener

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Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company,
for the United States Department of Energy's National Nuclear Security Administration
under contract DE-AC04-94AL85000.

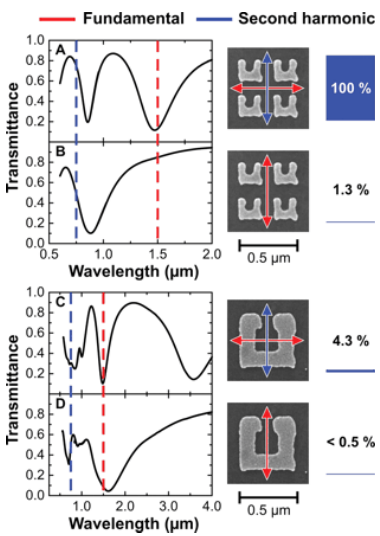
Outline

- Harmonic generation from metallic metasurfaces coupled to inter-subband transitions in semiconductor quantum wells
- Harmonic generation from all-dielectric metasurfaces

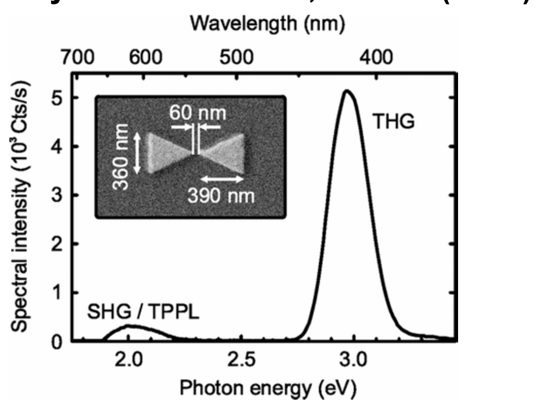
Nonloinear Metasurfaces: Harmonic Generation from Metallic Resonators

- **Harmonic generation has been observed from planar metamaterials ("metasurfaces") and plasmonic arrays.**

***First reports*: Science 502, 313 (2006)**



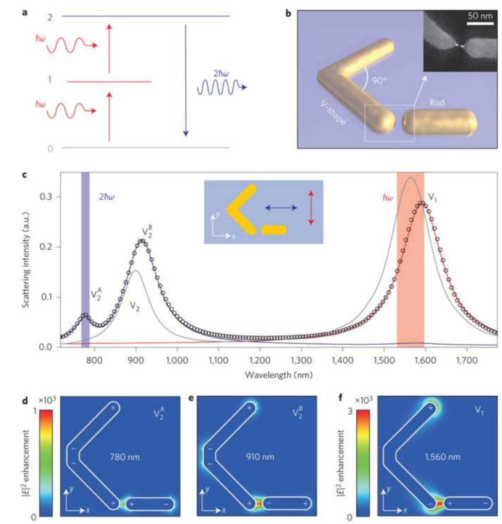
Other geometries:
Phys. Rev. Lett. 103, 257404 (2009)



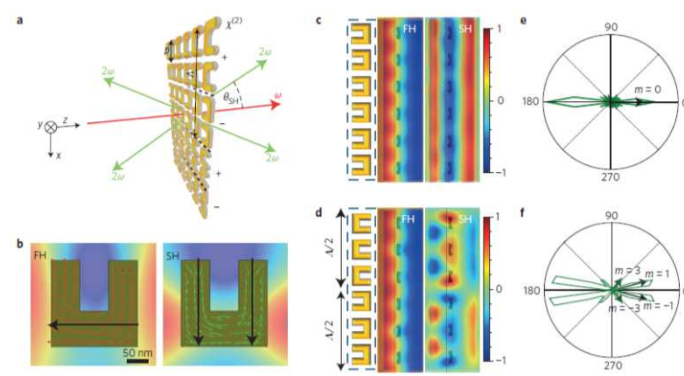
And many more….

Efficiencies have been very, <u>very</u> low!

Mode Matching:
Nature Nanotechnology 10, 412 (2015)



Metamaterials Photonic Crystals:
Nature Photonics 9, 180 (2015)



How Metasurfaces on Semiconductors Can Be Used for Enhanced Optical Nonlinearities

Conventional SHG, perfect phase-matching

$I_{2\omega} \sim (I_\omega \chi^{(2)} L)^2$

$\chi^{(2)}$

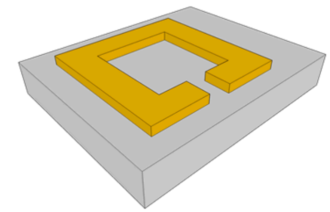
L

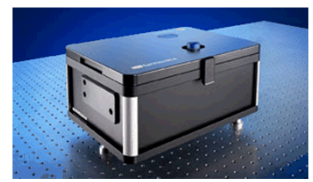
Increase I or L

Cavity-enhanced

$I_{2\omega} \sim Q^2$

$\chi^{(2)}$





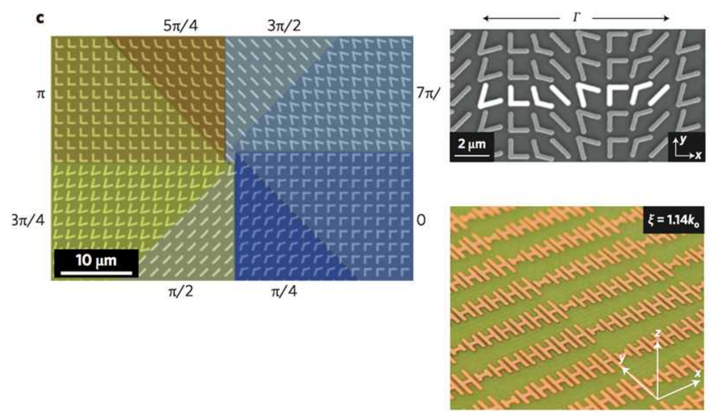
Coherent Inc.

Increase $\chi^{(2)}$

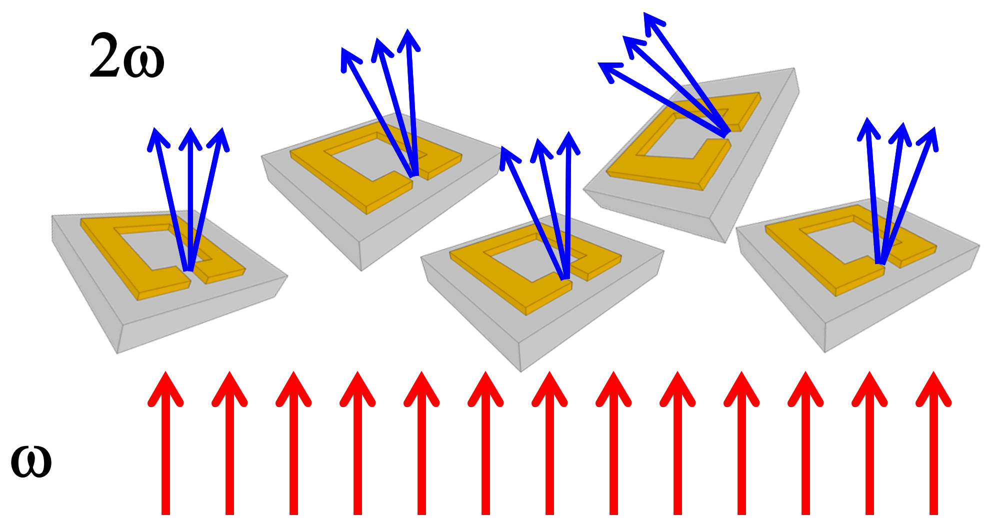
Use resonant $\chi^{(2)}$ with semiconductor heterostructures ~ 10^4 enhancement in $\chi^{(2)}$

Metasurfaces Provide New Functionality for Harmonic Generation

Metallic Metasurfaces have been used extensively for beam/phasefront/polarization manipulation: ***this is all degenerate:***

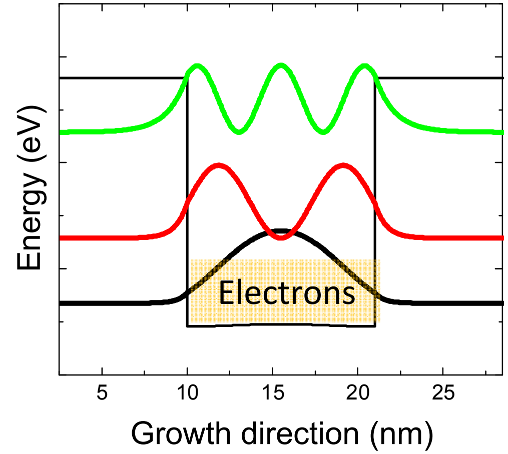


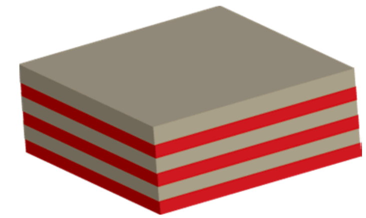
(Shalaev, Capasso, Zhou,etc..)



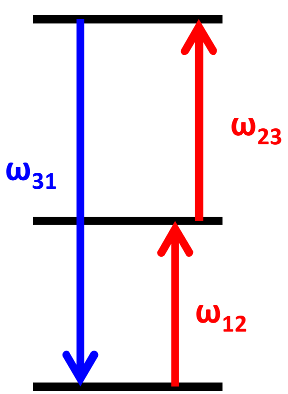
Resonant Optical Nonlinearities: Intersubband Transitions in Quantum Wells

Electronic levels and wavefunctions can be engineered with semiconductor heterostructures





Resonant **optical nonlinearities can be engineered too:**

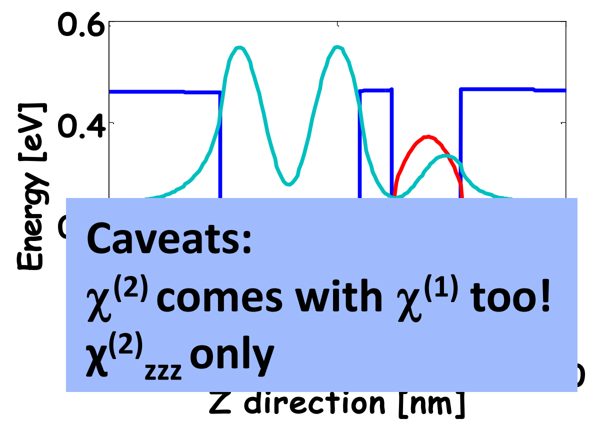


$$\chi^{(2)}(\omega) \propto \frac{N \cdot z_{12} z_{23} z_{13}}{(\omega - \omega_{12} - i\Gamma_{12})(2\omega - \omega_{13} - i\Gamma_{13})}$$

$$z_{ij} \propto \langle \Psi_i | \vec{R} | \Psi_j \rangle$$

~250 nm/V - QWs

10s of pm/V - $LiNbO_3$

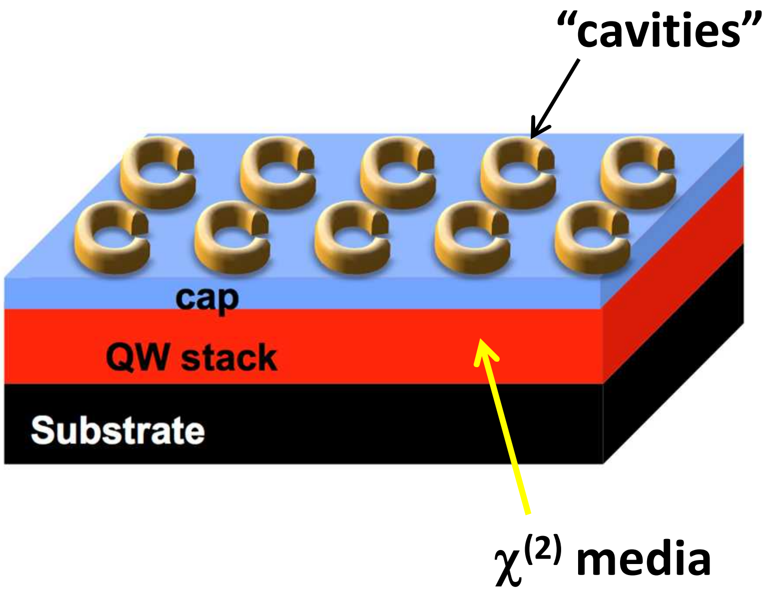


Caveats:
$\chi^{(2)}$ comes with $\chi^{(1)}$ too!
$\chi^{(2)}_{zzz}$ only

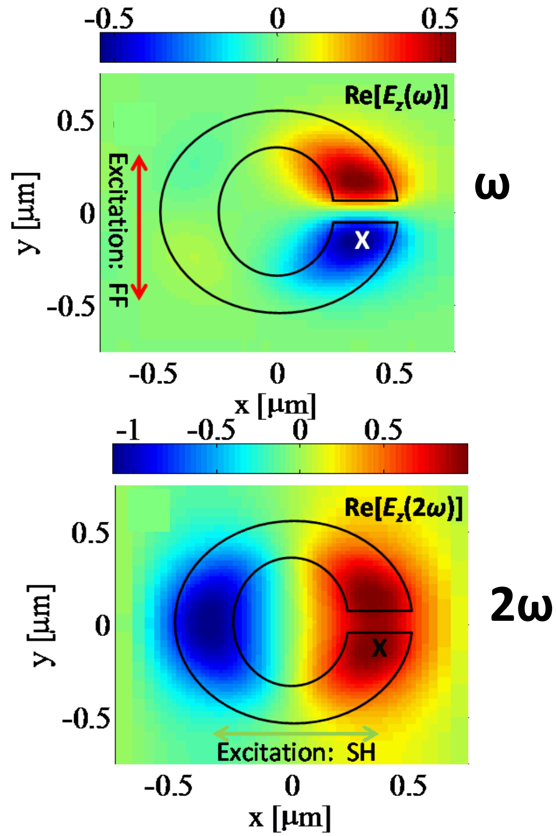
F. Capasso, C. Sirtori, and A.Y. Cho, IEEE J. Quantum Electron. 30, 1313 (1994)

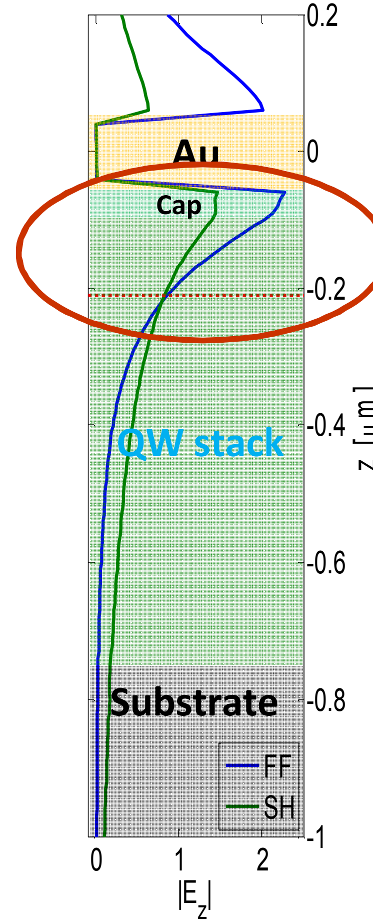
Salvo Campione

Metasurfaces Coupled to Resonant $\chi^{(2)}$



Resonators are designed to have resonances at 30 & 60 THz (5&10um)

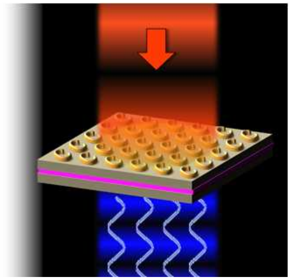


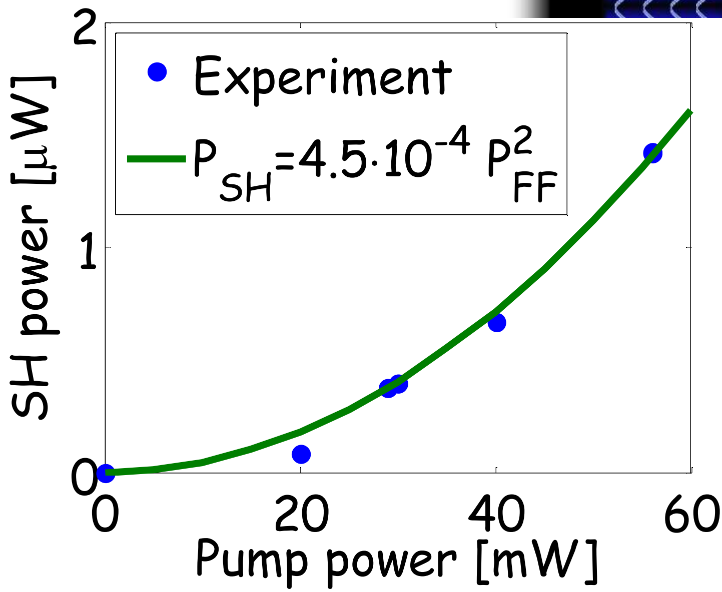


APL 104, 131104 (2014)

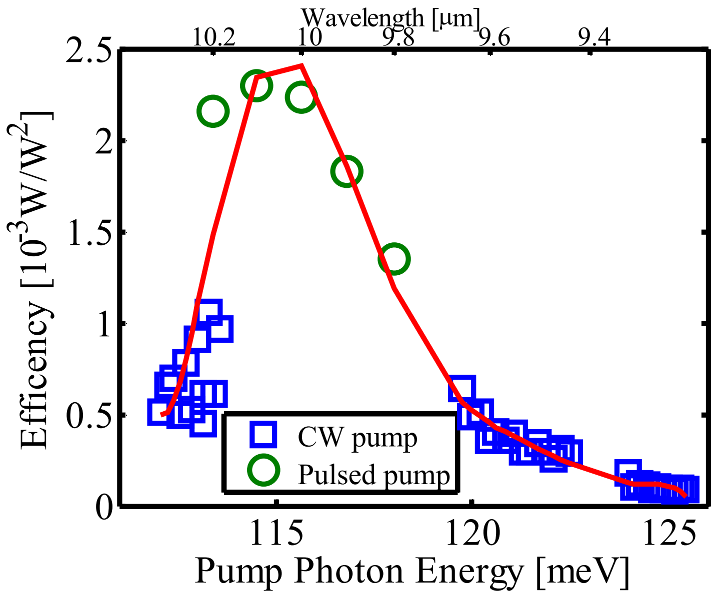
SHG: Power and Frequency Dependence

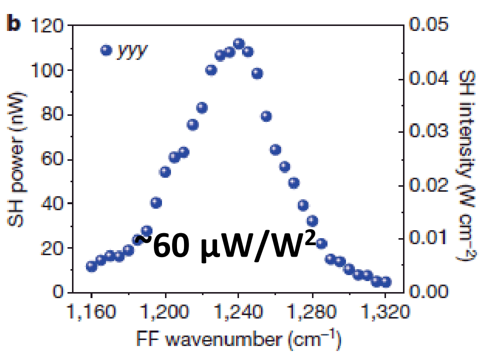
Pump: CW CO_2 laser





max efficiency: ~2.3 mW/W²



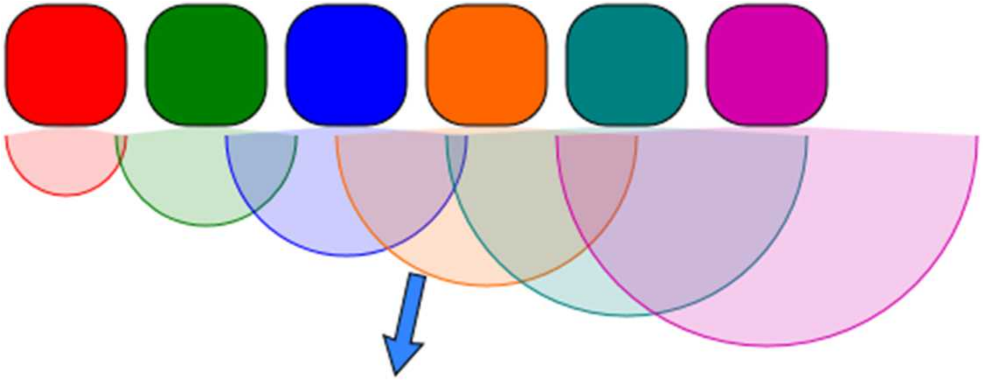


Lee, Belkin, Alu, et al. *Nature* 511, 65-69 (2014).

- **Saturation is an issue**
- **Max. conversion efficiencies of few % are possible**
- <u>**Added functionality is the advantage**</u>

Nature Communications 6, 7667 (2015)

Phased Arrays



Identical point sources

+

Radiating with controllable phase difference

==

Full control over beam direction and shape

Image from Wikipedia

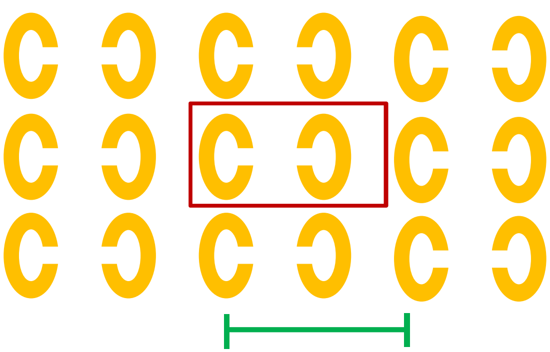
SHG Beam Manipulation


Omri


Salvo

"edge-fed dipole"

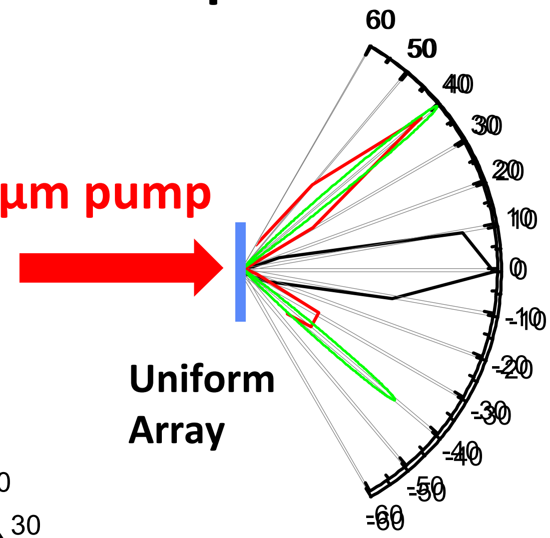
flipping induces π phase shift



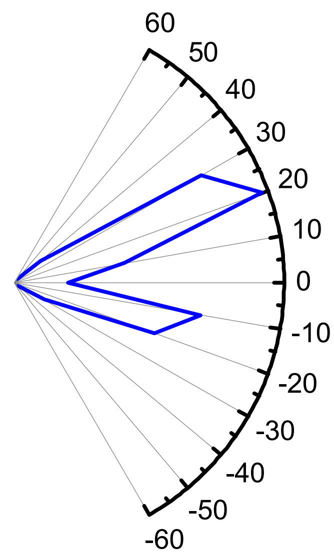
Period determines angular separation

10µm pump

5µm beam



Uniform Array



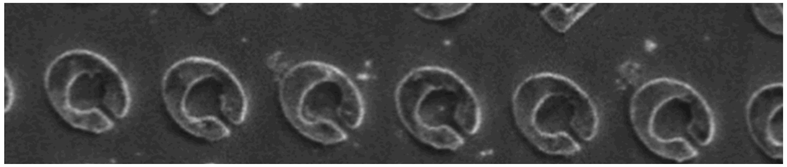
2-in-1:

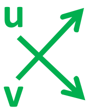
Source +beam splitter

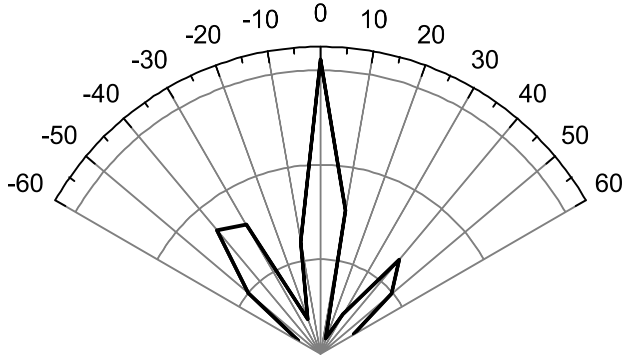
Nature Communications 6, 7667 (2015)

SHG Beam Manipulation II

- **Cavities radiate polarized light**





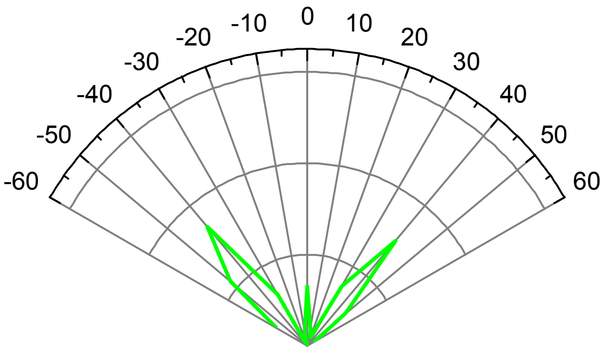


unpolarized

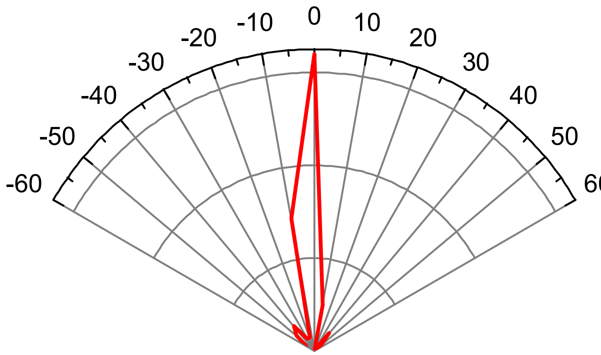
Pump polarization

3-in-1:

Source + Polarizer + Beam Splitter

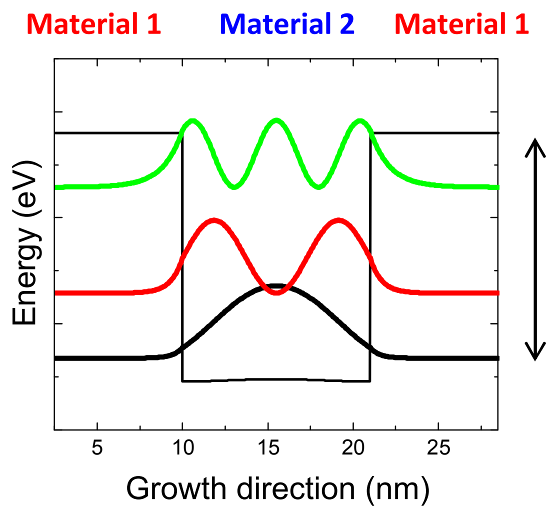


v polarized



u polarized

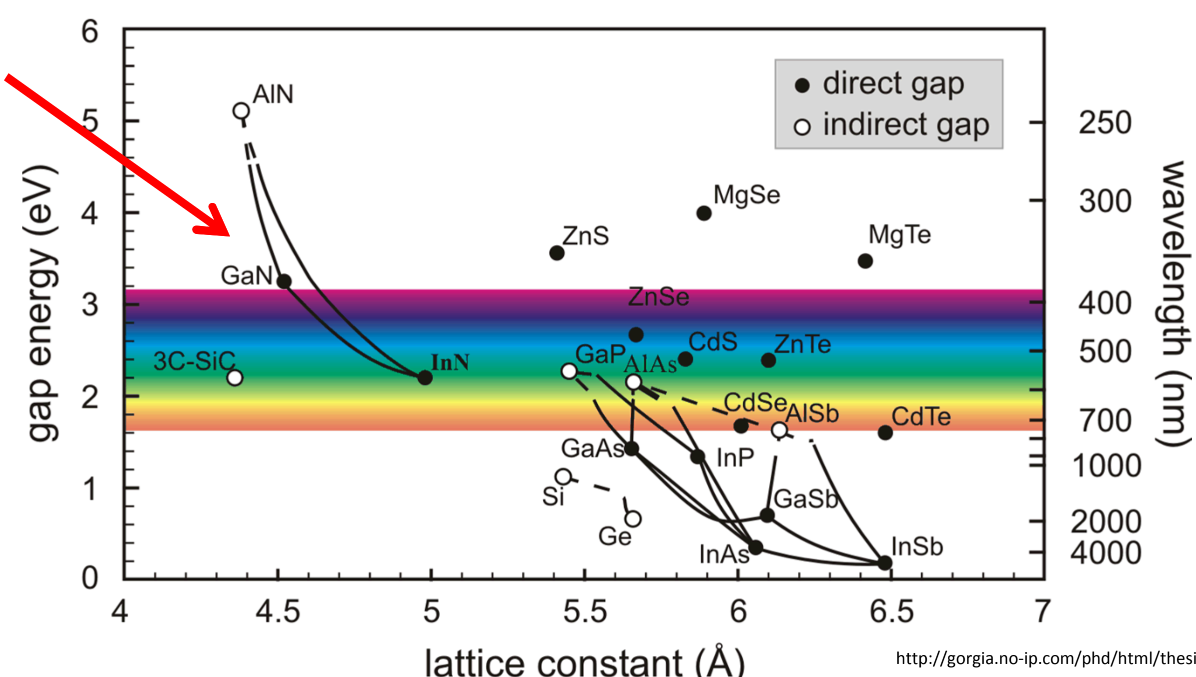
Scaling This Approach to Shorter Wavelengths



10um to 5um: AlInAs barriers
InGaAs wells
2ω ~ 0.24eV

How about 1.55um to 0.75um?
This requires a conduction band offset of ~1.6eV!!

III-Nitrides

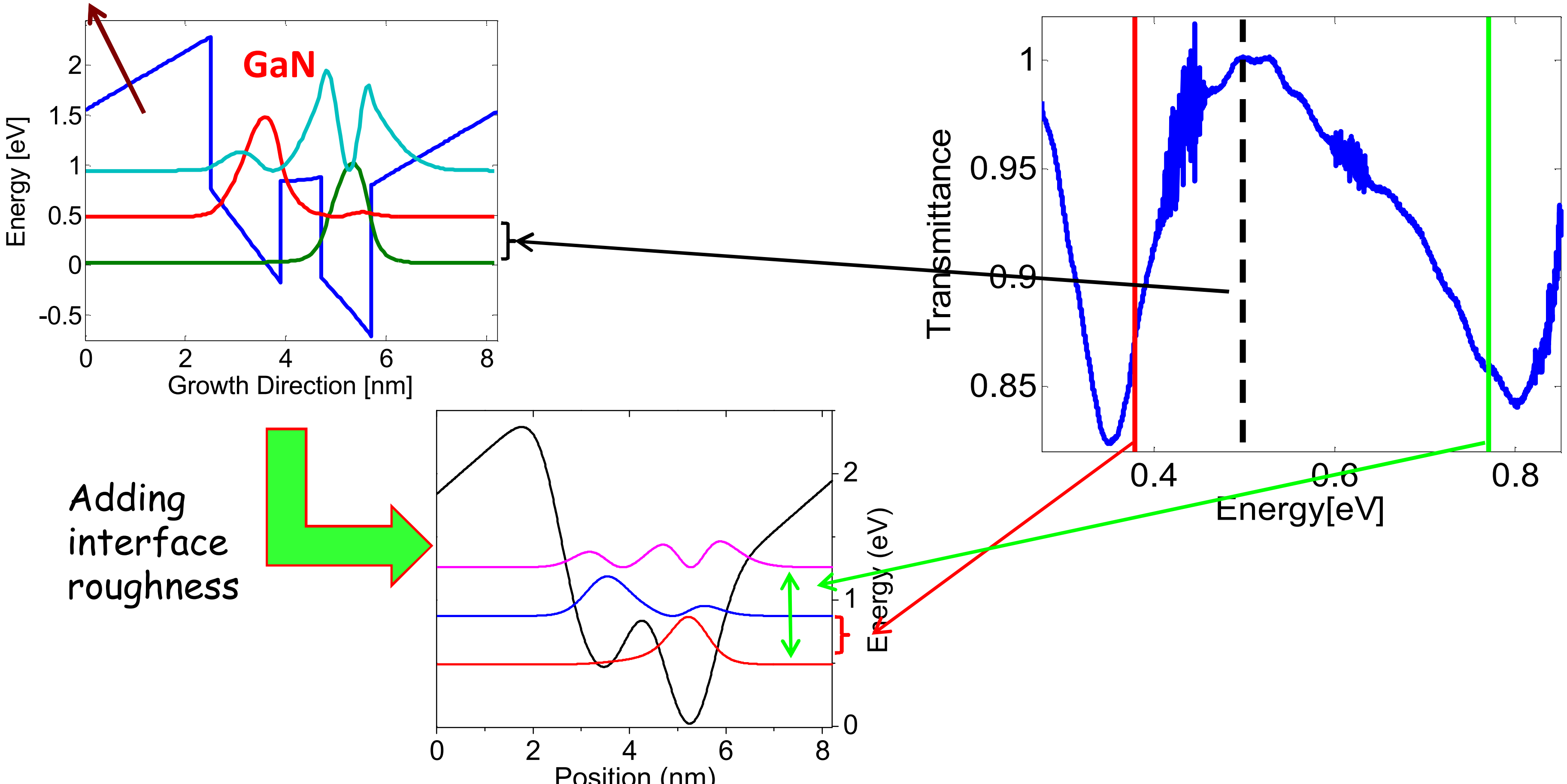


http://gorgia.no-ip.com/phd/html/thesis/phd_html/simone-jbook.html

Omri Wolf

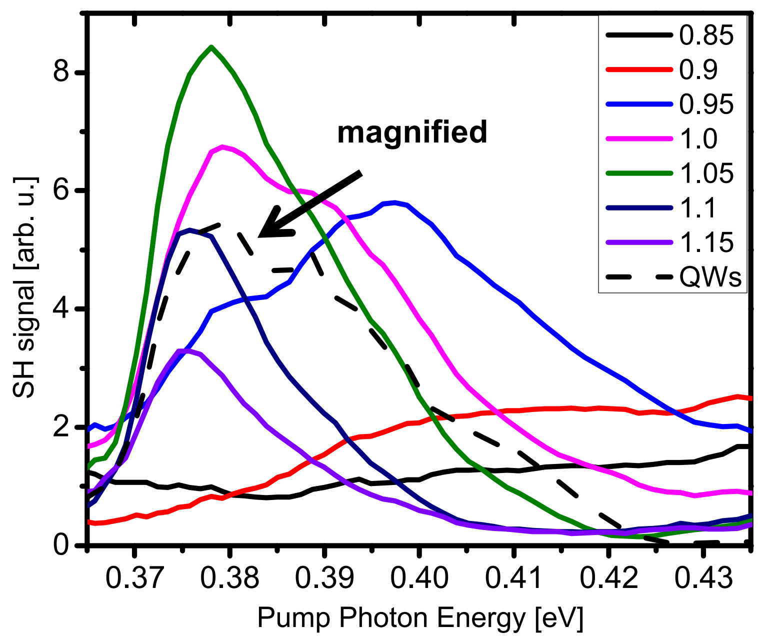
SHG (3.2μm>1.6μm) Using AlGaN/GaN QWs

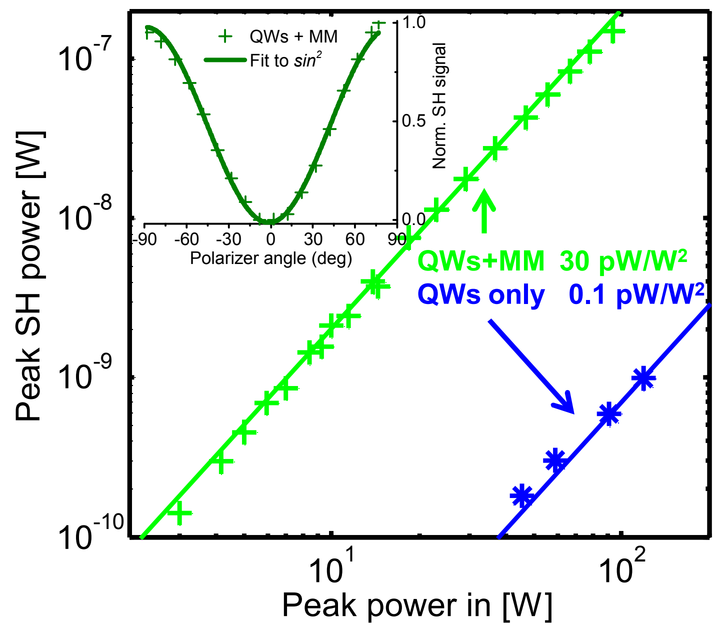
- **Design: SHG 3 μm → 1.5 μm (0.4 eV → 0.8 eV)**
- **Traditional band structure calculation <u>not enough</u>**
  - **Over estimation of IST**

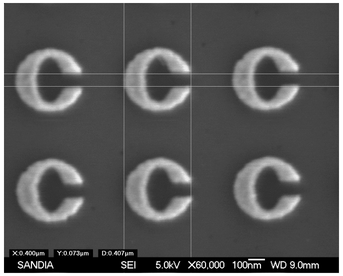


O. Wolf et al. Under review

SHG (3.2μm>1.6μm) Using AlGaN/GaN QWs

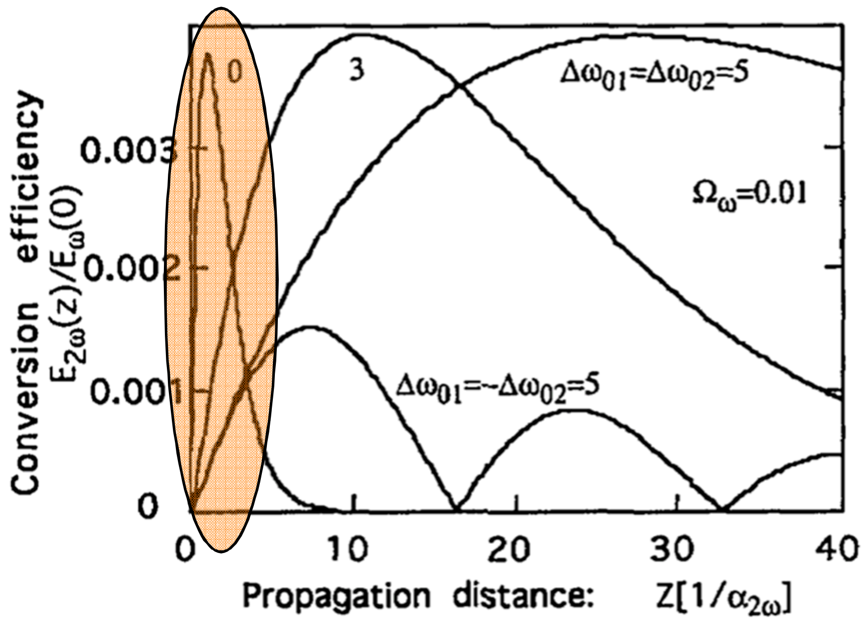






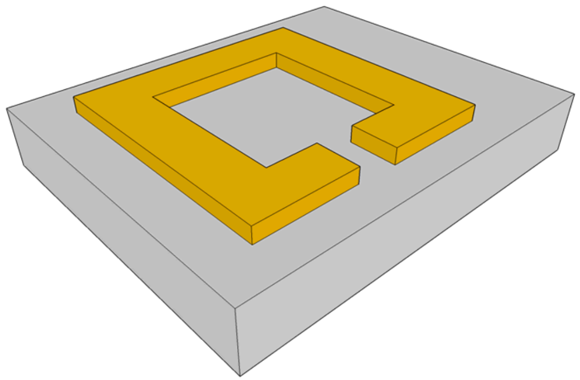
In a III-N heterostructure, metamaterials increase efficiency by over 2 orders of magnitude

Effect of Absorption on SHG (unavoidable when using resonant $\chi^{(2)}$)



Almogy&Yariv, *Optics Letters* (1994)

Dealt first by Boyd, Ashkin, Dziedzic, & Kleinman, (1965). *Physical Review, 137*(4A), A1305.



For α~1000cm^{-1} (1/α~10um), "effective length" (*QxL*) needs to be <few microns

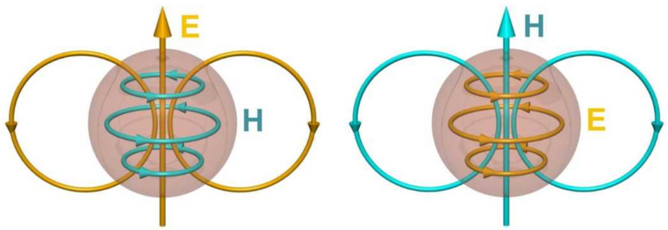
"Cavity" with Q~10
Field decay length (*L*) ~200nm
***QxL*~2um…. >> OK,**
We cannot gain too much more by increasing Q!

Outline

- Harmonic generation from metallic metasurfaces coupled to inter-subband transitions in semiconductor quantum wells
- Harmonic generation from all-dielectric metasurfaces

Dielectric Resonators for Metamaterials

Magnetic dipole resonance: tailor μ
Electric dipole resonance: tailor ε



Images: A. Miroshnichenko

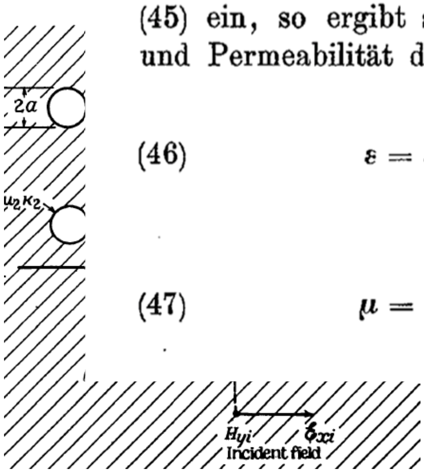
3. *Zur Optik kolloidaler Metallösungen;* *von R. Gans und H. Happel.*

Annalen der Physik, 1909

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

(46) $$\varepsilon = \varepsilon_0 \frac{1 + \frac{4\pi N}{3}\left(\frac{\lambda'}{2\pi}\right)^3 (a_1 - a_2)}{1 - \frac{2\pi N}{3}\left(\frac{\lambda'}{2\pi}\right)^3 (a_1 - a_2)},$$

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



$$\epsilon'_{re} = \epsilon_{r1}\left(1 + \frac{3v_f}{\frac{F(\theta)+2b_e}{F(\theta)-b_e} - v_f}\right)$$

(Ack: Ed Kuester, CU Boulder)

MATERIAL LOADED WITH
ICLES*

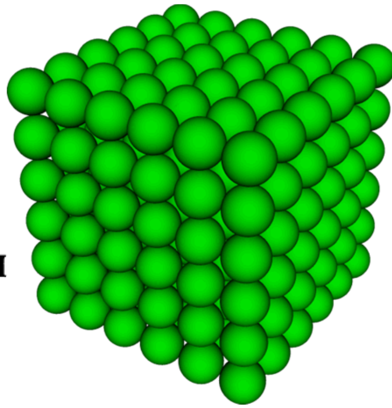
†

ed form 27th September, 1946.)

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

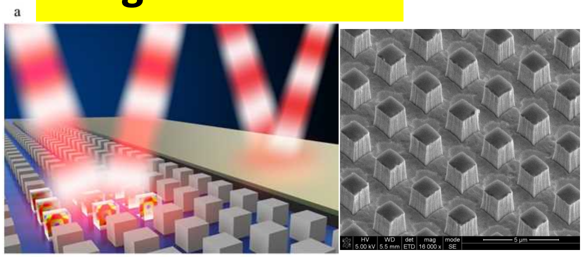
$$\theta = k_0 a \sqrt{\epsilon'_{r2}\mu'_{r2}}$$

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



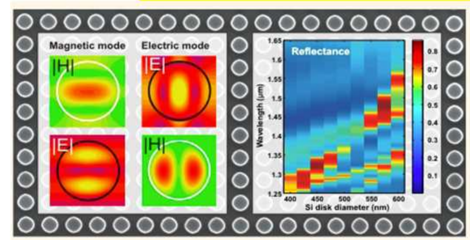
Some Recent Results with Dielectric Metasurfaces (Linear)

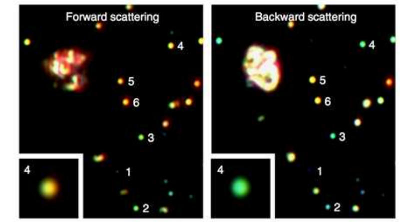
Magnetic mirror



Optica 2015

Tailoring scattering

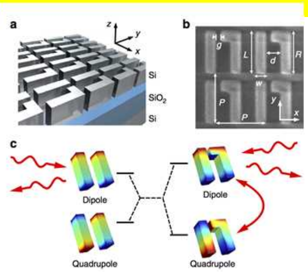


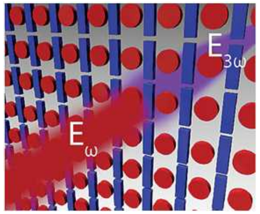


ACS Nano 7*, 7824 (2013)*

Nat. Comm. 4*, 1527 (2013)*

Fano resonances

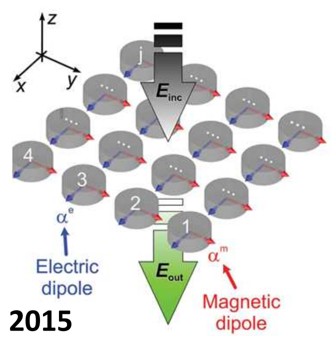




Nature Comm*. 2014.*

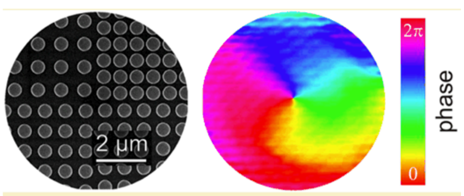
Nano Lett. 15, 7388 (2015)

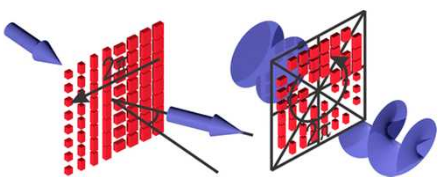
Huygens Metasurfaces

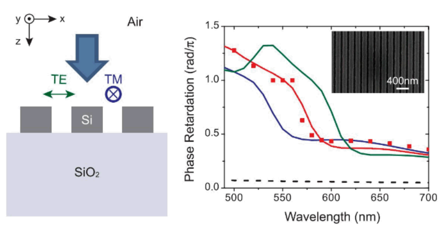


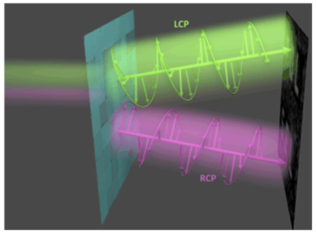
Adv. Optical Mater. **2015**

Phase-front manipulation









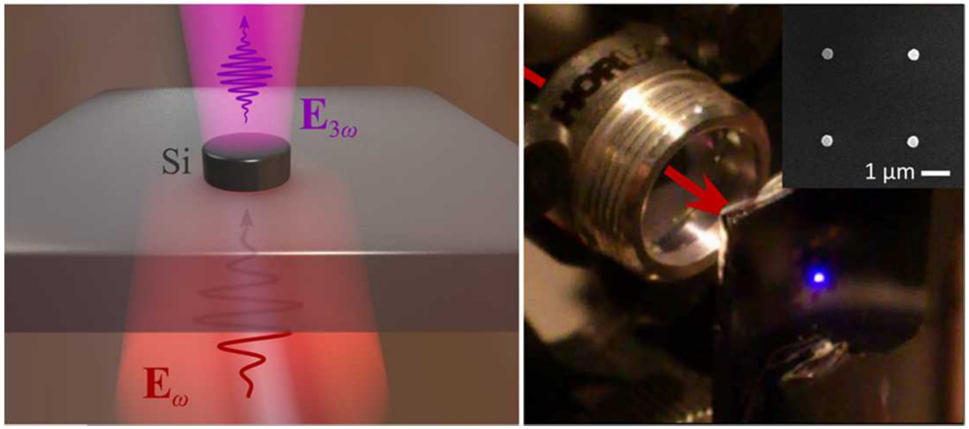
Nano Lett. 15, 5369 (2015)

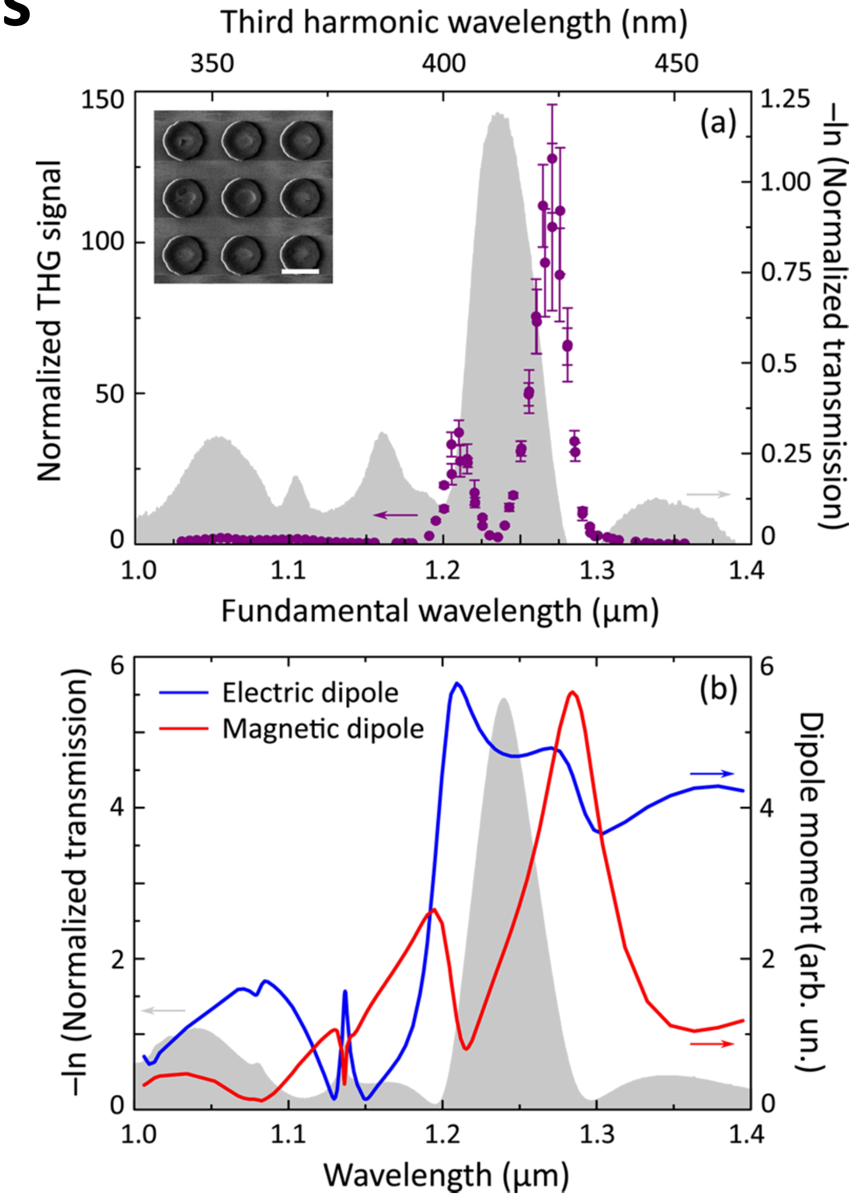
Nano Lett.15, 6261 (2015)

Science 345*, b*

Opt. Express 23*, 22611 (2015)*

Optical Nonlinearities (THG) in Si Dielectric Metasurfaces





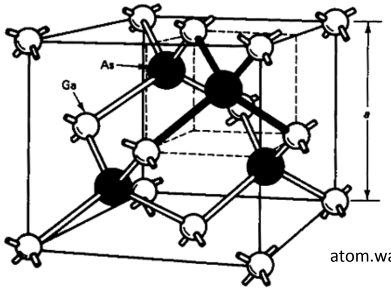
Strong enhancement near magnetic dipole resonance

(With ANU & Moscow State)

Nano Letters (2015)

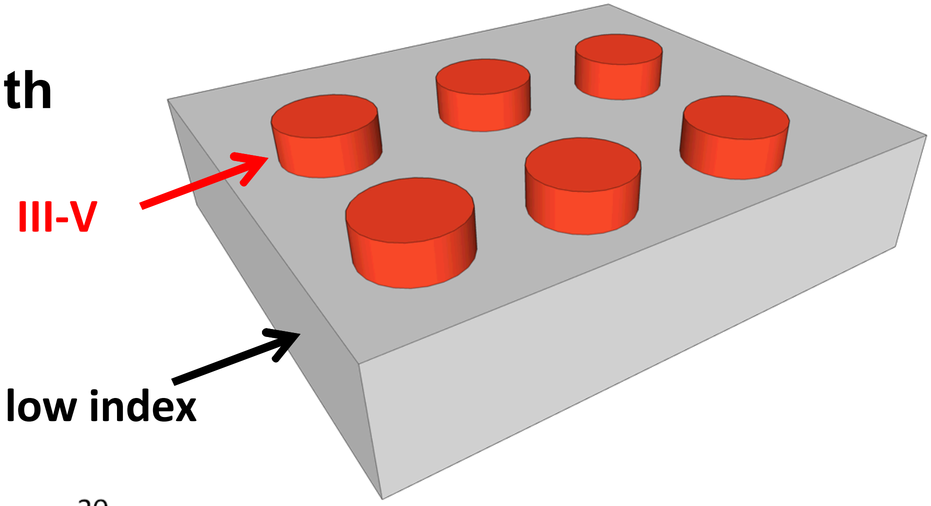
Silicon is "limited"for Optical Nonlinearities

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

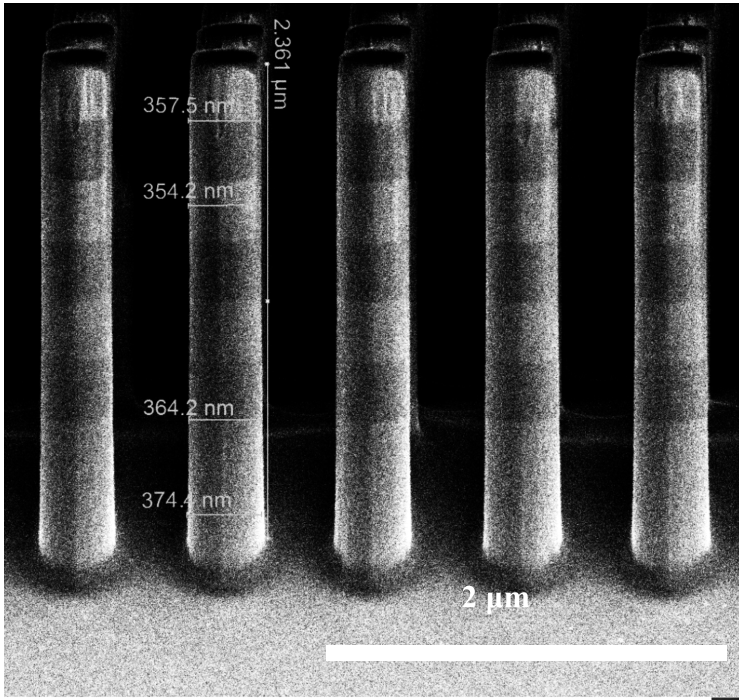


atom.waterloo.ca

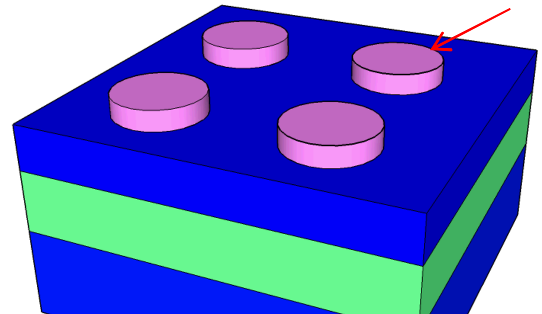
How to obtain high index dielectric nanoresonators with III-V semiconductors??

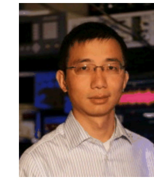


Fabrication of Al(In)GaAs Based Dielectric MMs

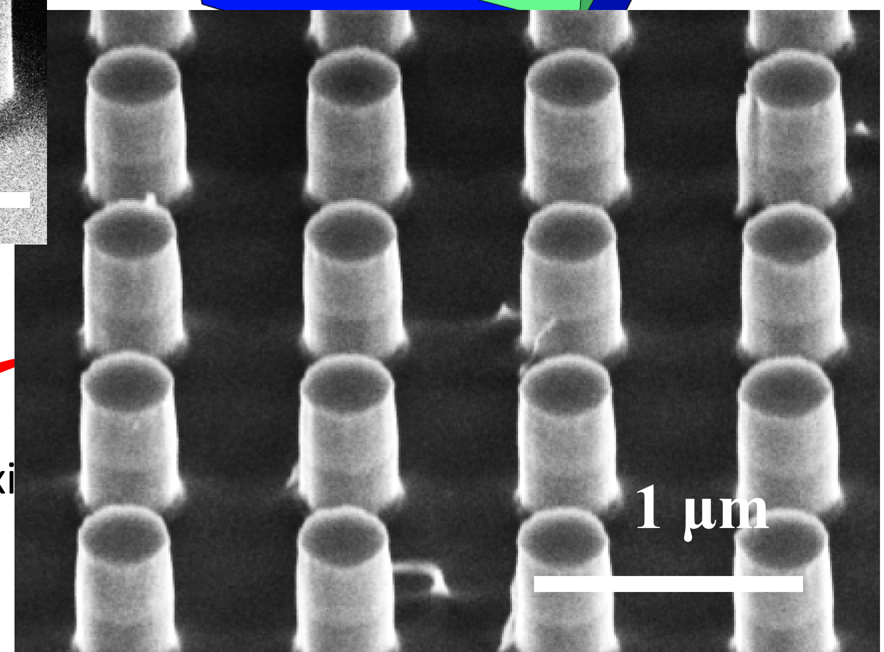


D





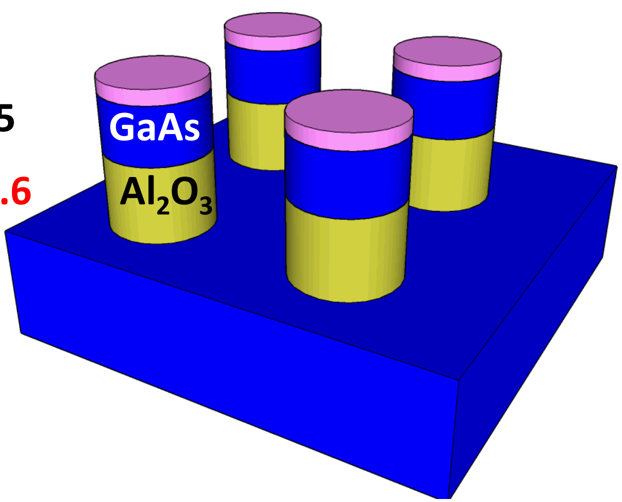
Sheng Liu



Oxi

n~3.5

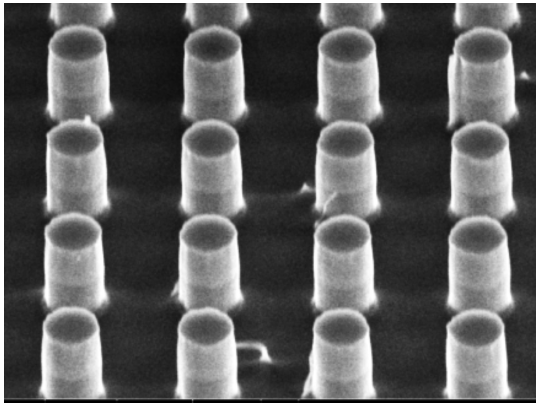
n~1.6



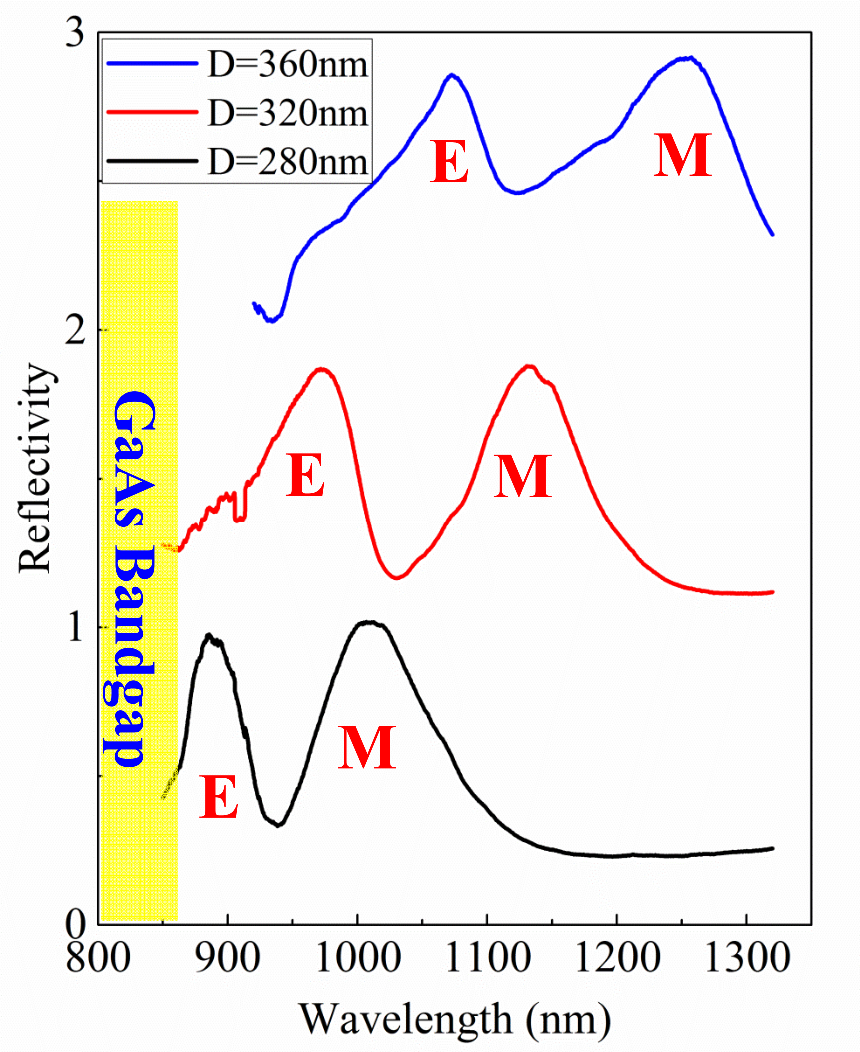
GaAs Dielectric Resonators (1 layer)

GaAs disk height ~300nm

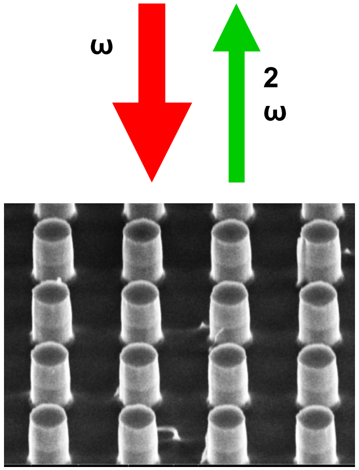
Different diameters

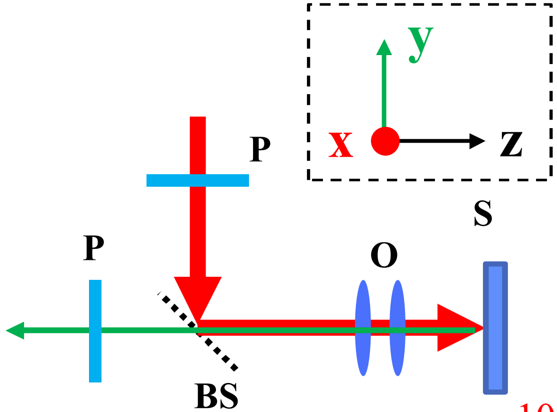


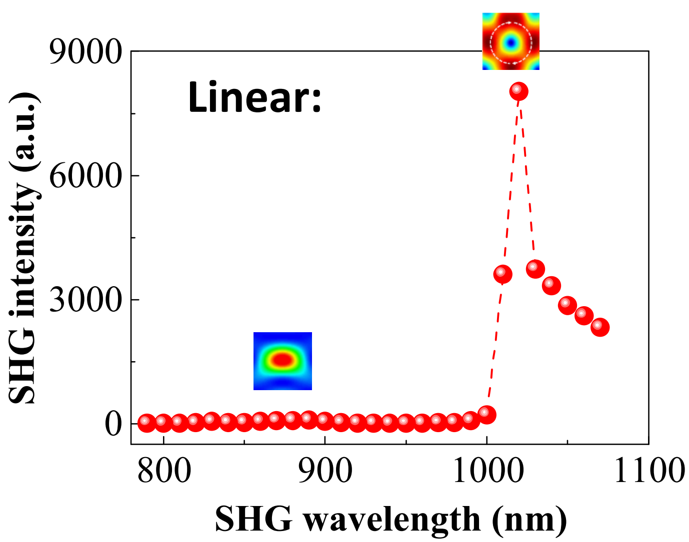
- **Extremely low loss below bandgap**
- **Crystalline**

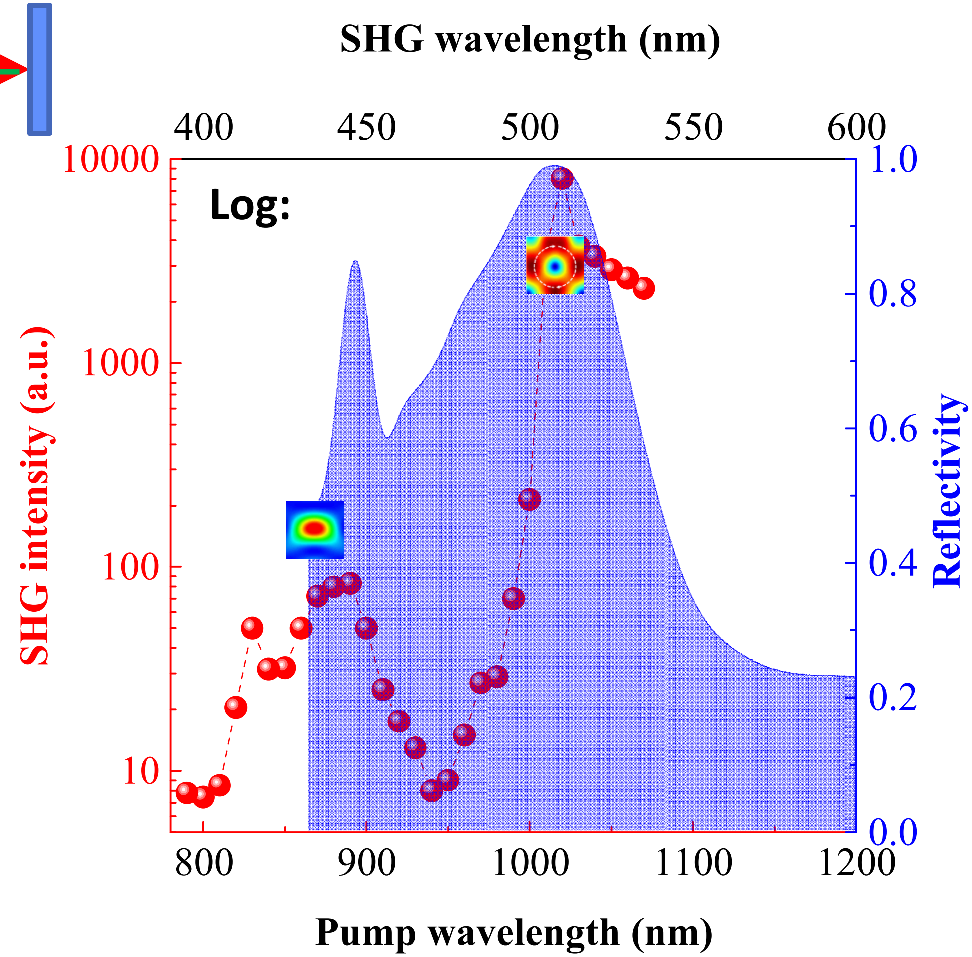


Second Harmonic spectral dependence

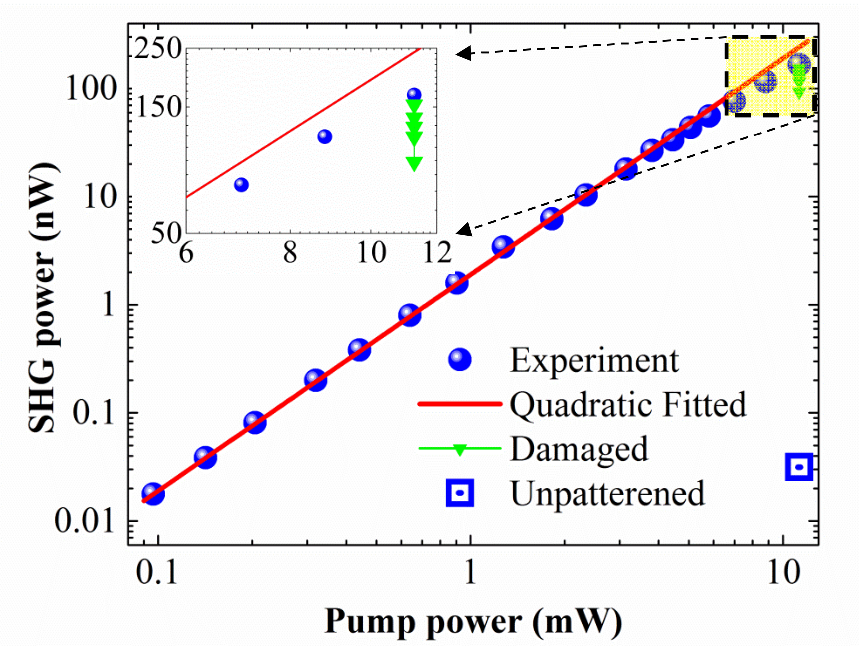


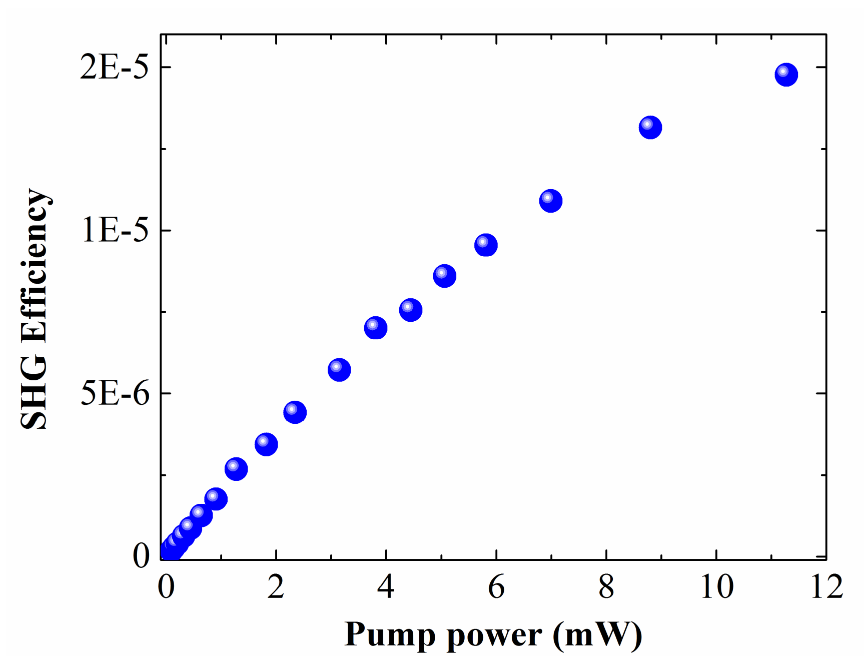






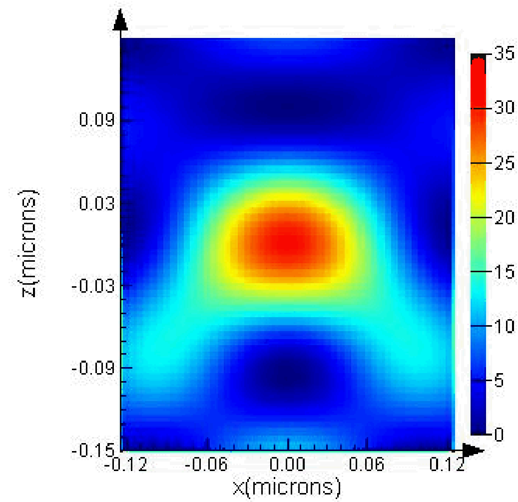
Power Dependence

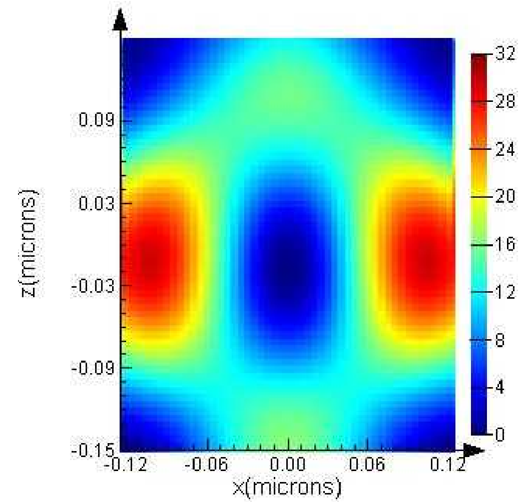




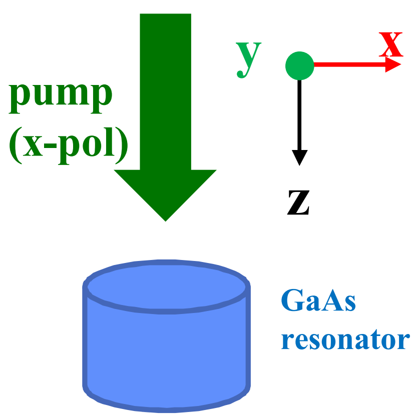
Polarization, Selection Rules

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





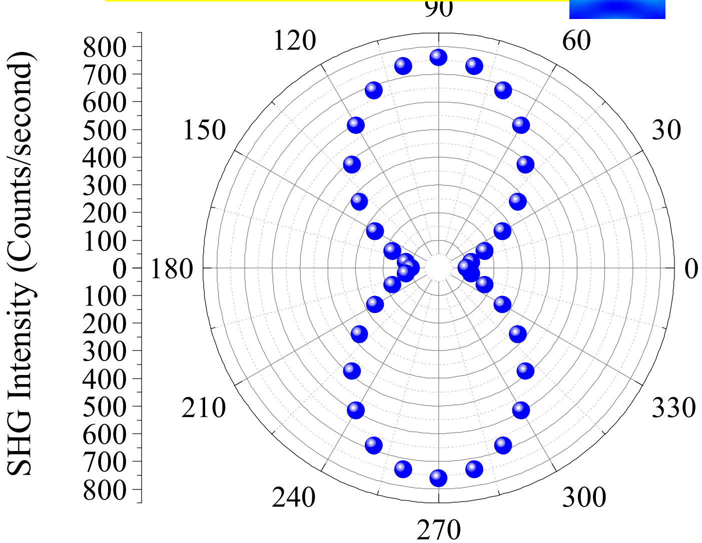
There are only E_x and E_z component inside the GaAs resonators.



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

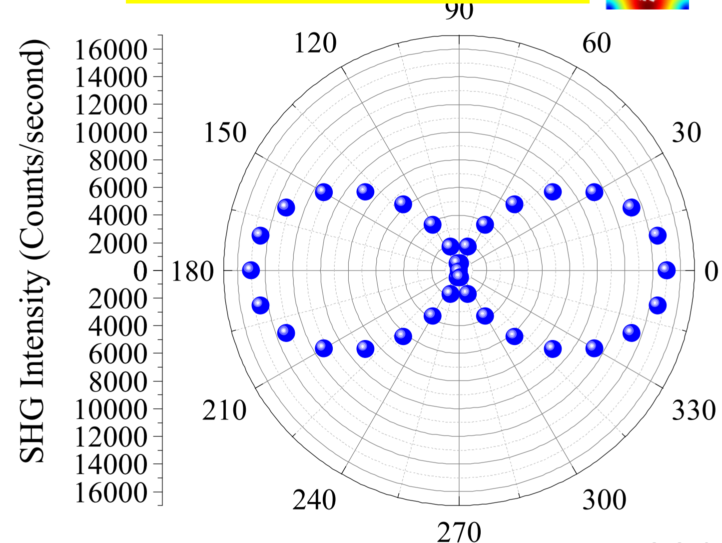
Orthogonal SHG polarization compared to the pump is expected

E-dipole resonance



Pump

M-dipole resonance



oops!

What happens at the M-dipole resonance?

1. Surface nonlinearity changes symmetry to mm2

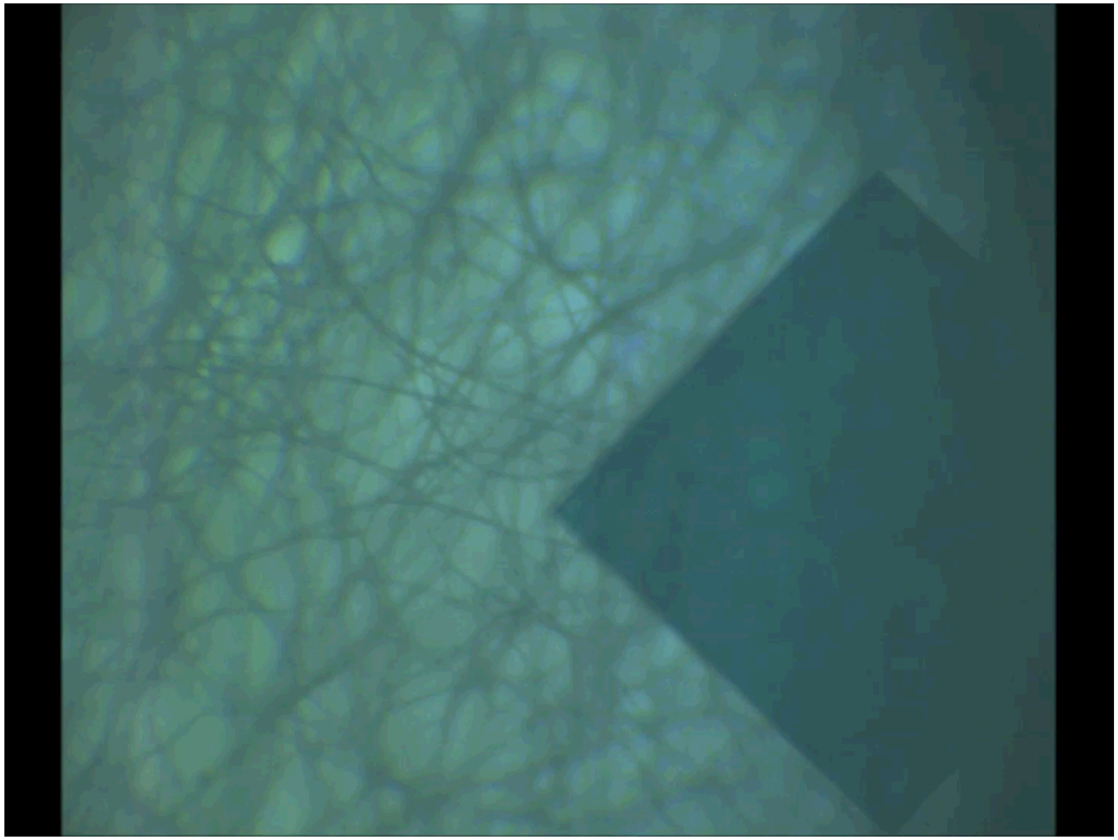
$$E_x^{2\omega} = 2\chi_{xxz}^{(2)} E_x^{\omega} E_z^{\omega}$$

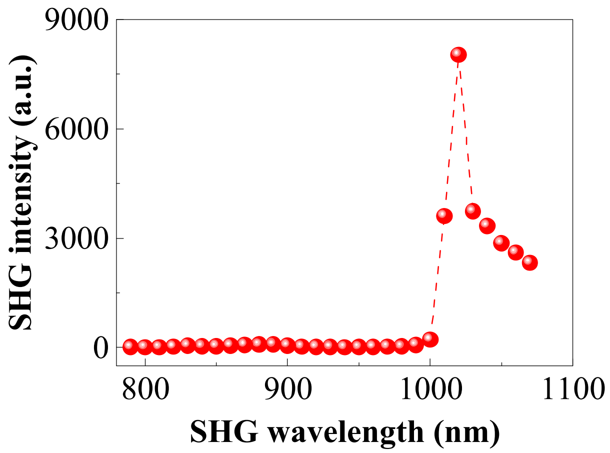
can be large, considering the large field enhancement at the surface for this mode.

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)

SHG & THG….





9000
6000
3000
0
800
900
1000
1100
SHG intensity (a.u.)
SHG wavelength (nm)

Acknowledgments

Postdocs: Salvo Campione, Omri Wolf, Sheng Liu

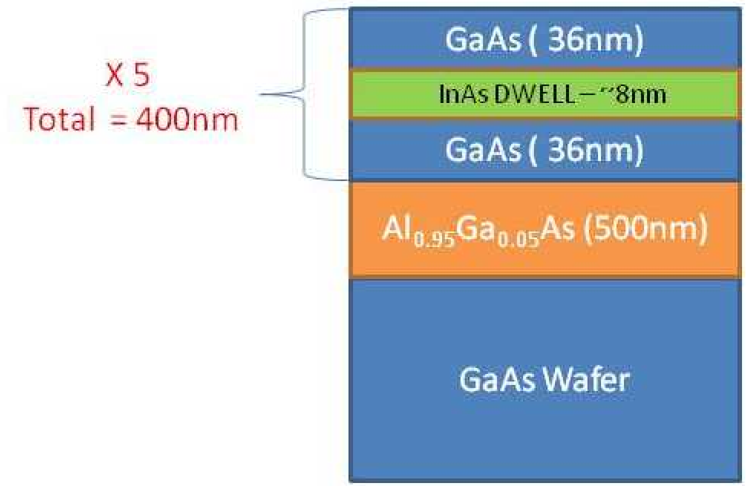
InGaAs

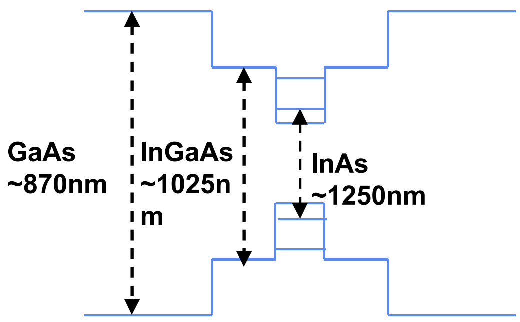
CINT + Sandia: John Reno, Mike Sinclair, Ines Montano, John Klem, Eric Shaner, Mike Wanke, Joel Wendt, Paul Clem, Jon Ihlefeld, Bruce Burckel, Jason Domingues, Andy Allerman, Sally Samora, Jon Wierer, ……

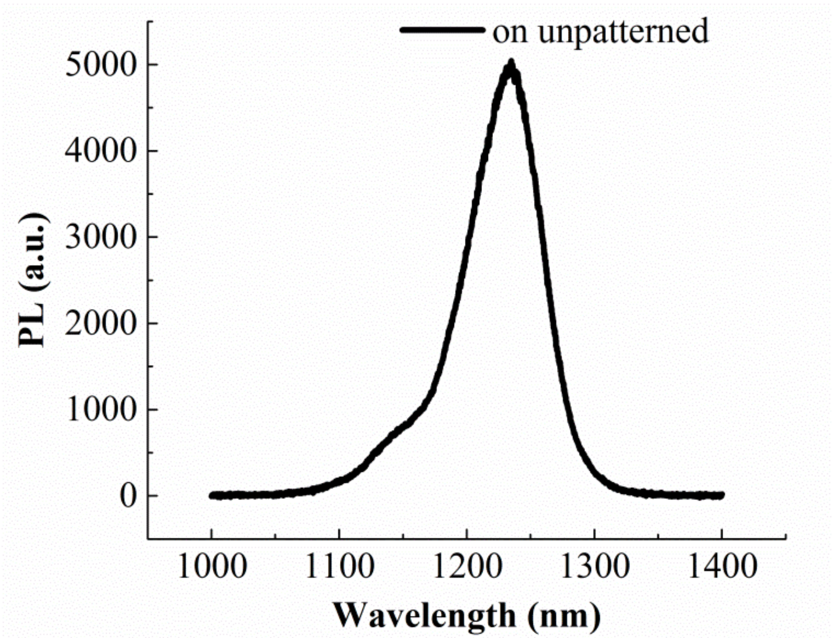
GaN

U.S. DEPARTMENT OF ENERGY | Office of Science

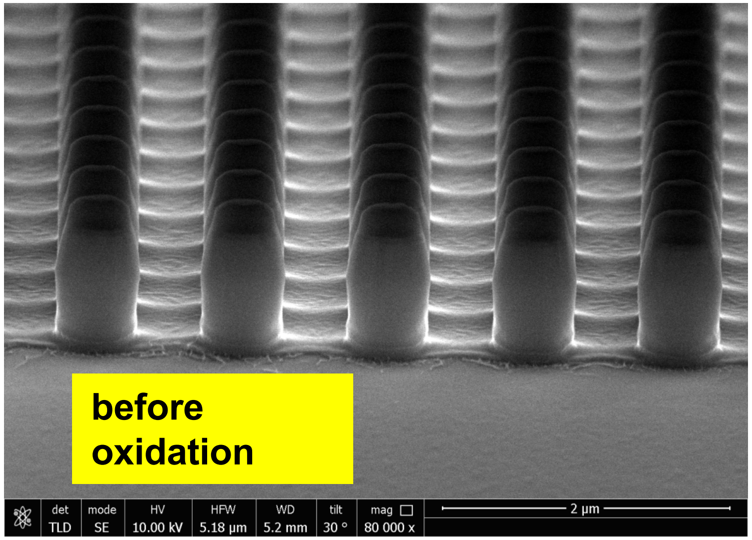
Active III-V Dielectric Resonators

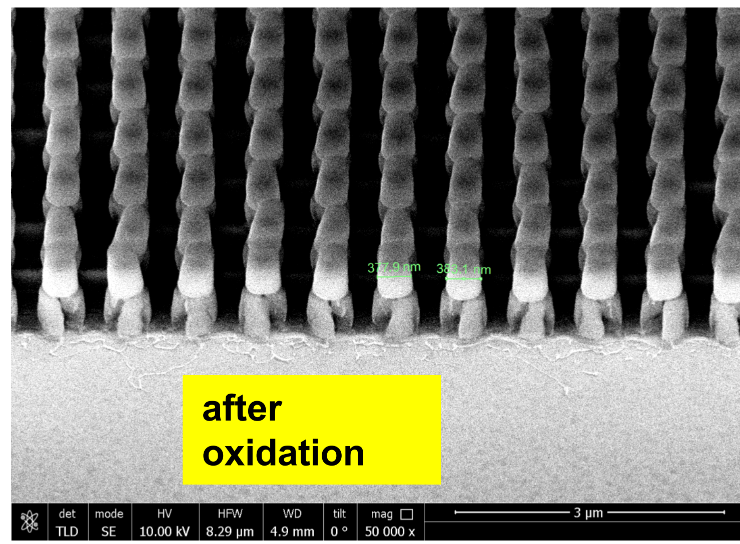


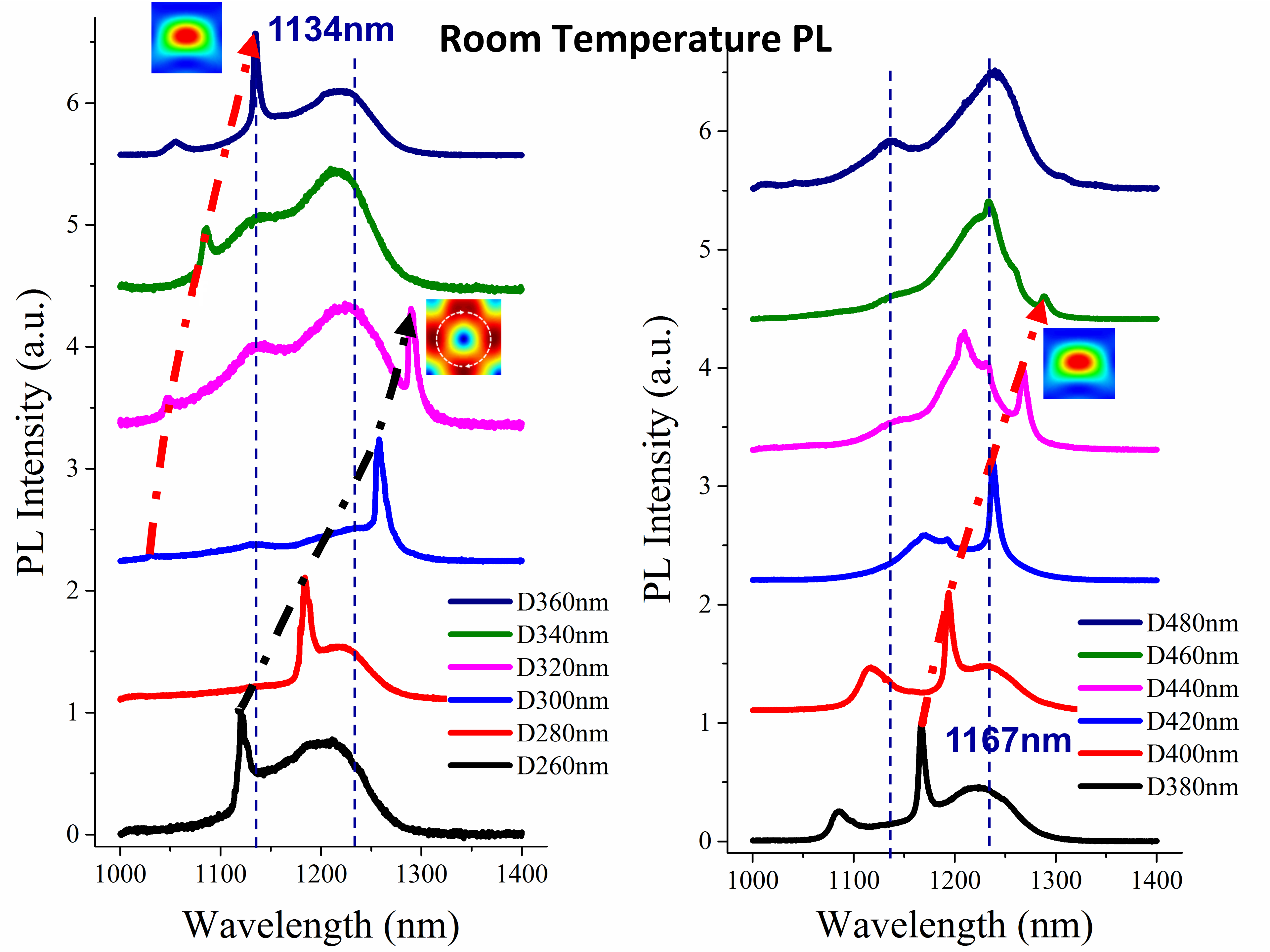




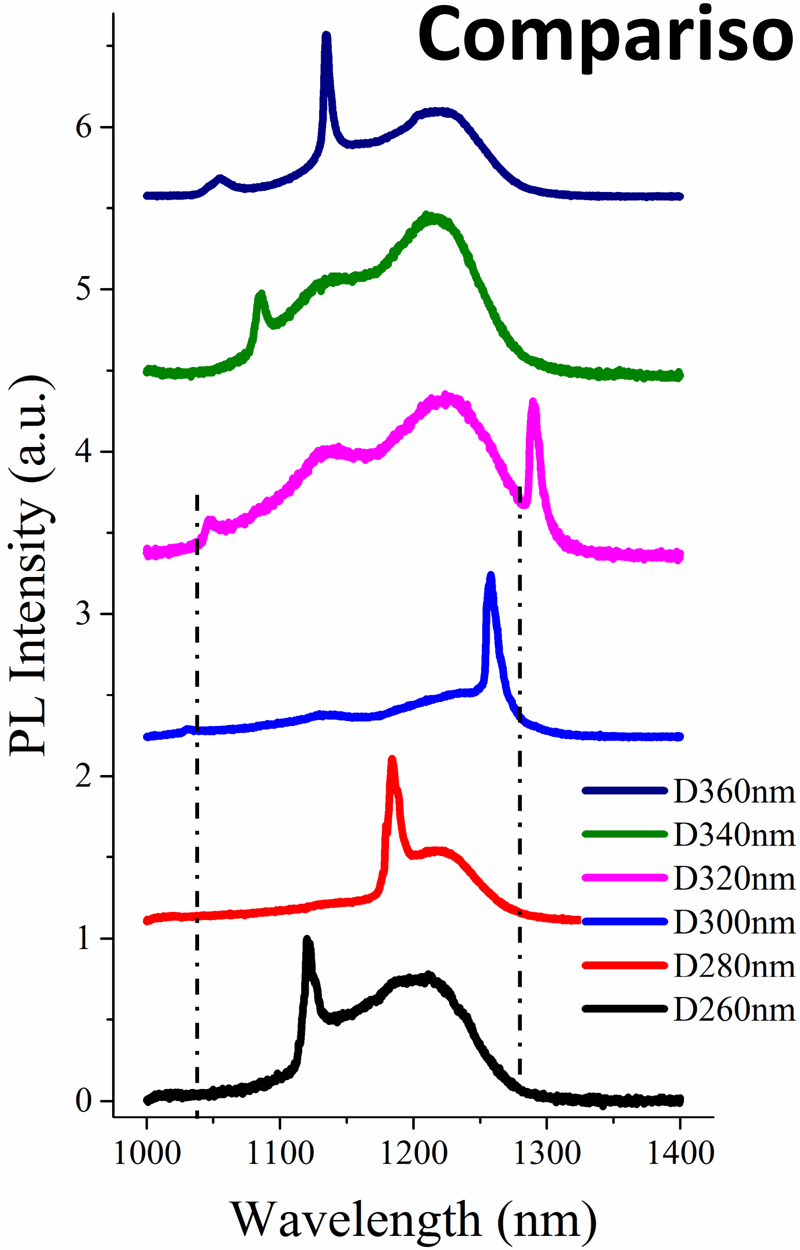
Fabrication

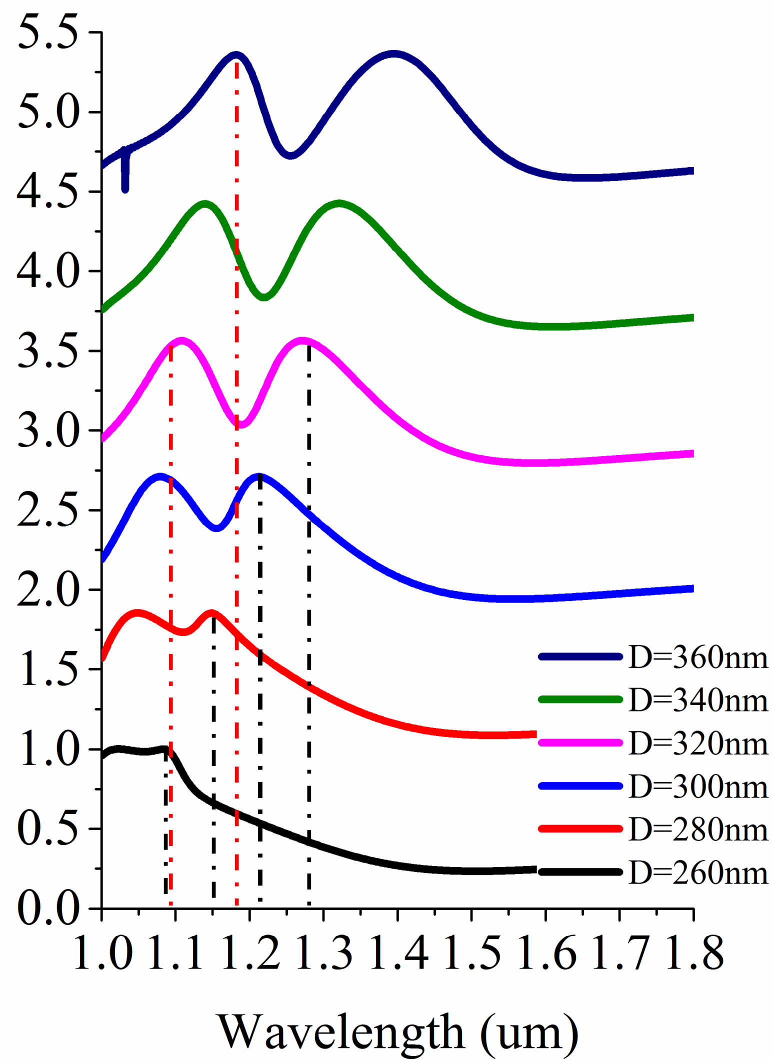


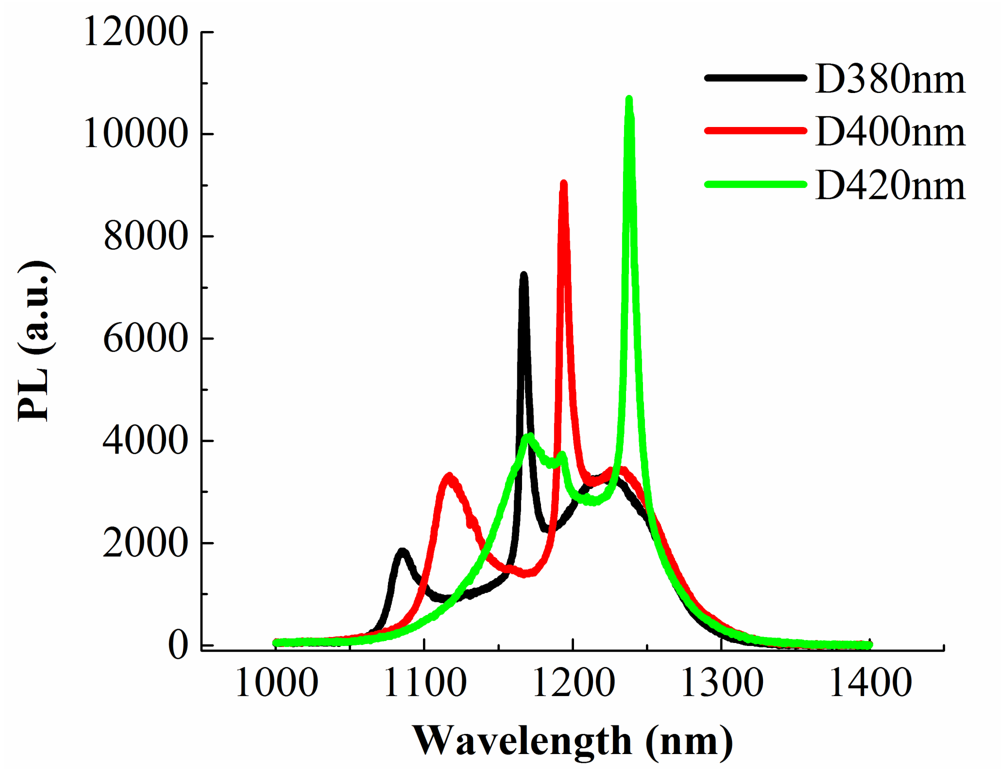



Room Temperature PL
1134nm
1167nm
PL Intensity (a.u.)
Wavelength (nm)
D360nm
D340nm
D320nm
D300nm
D280nm
D260nm
D480nm
D460nm
D440nm
D420nm
D400nm
D380nm
0
1
2
3
4
5
6
1000
1100
1200
1300
1400

Comparison to FDTD Modeling

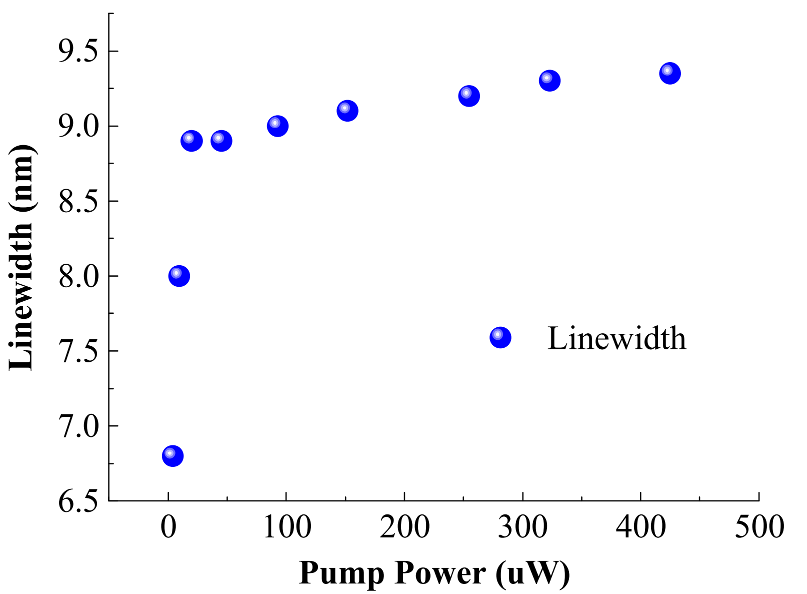


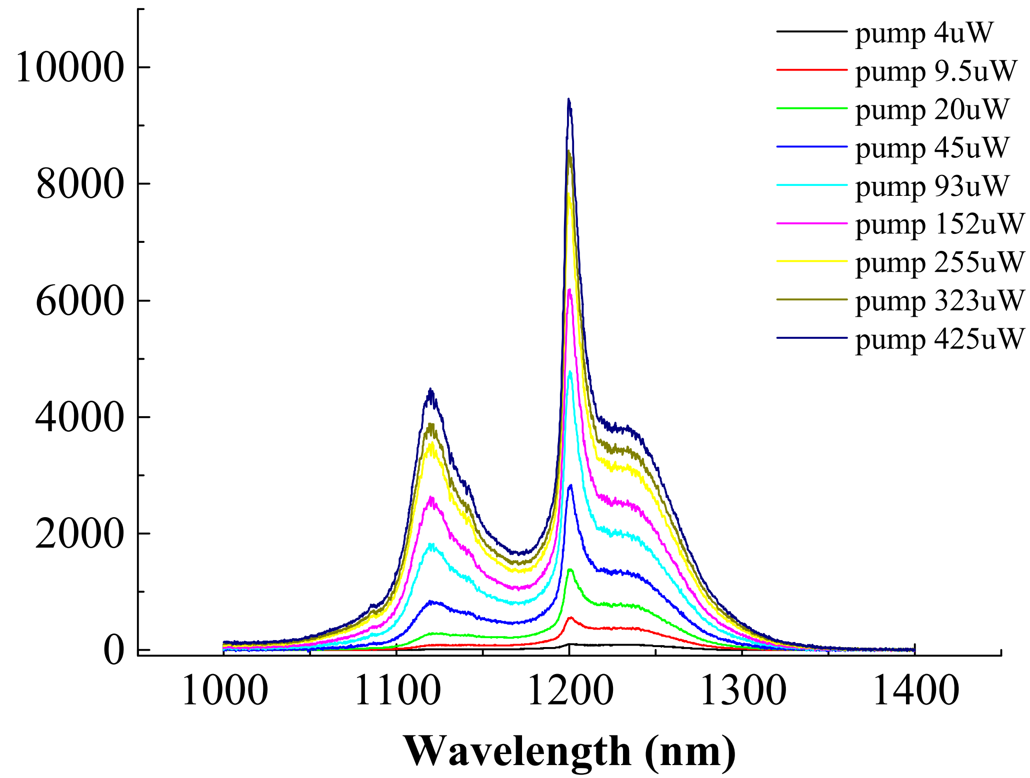




The PL intensity enhancement is larger at electric dipole resonance than at the magnetic dipole resonance.

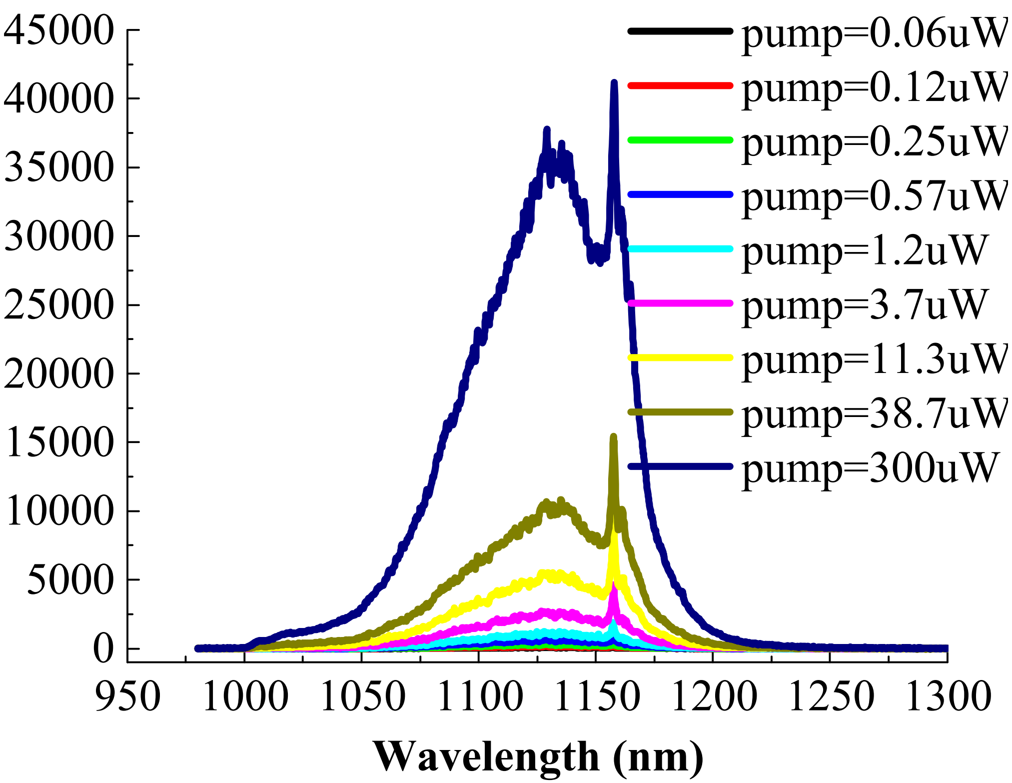
This is not Lasing!

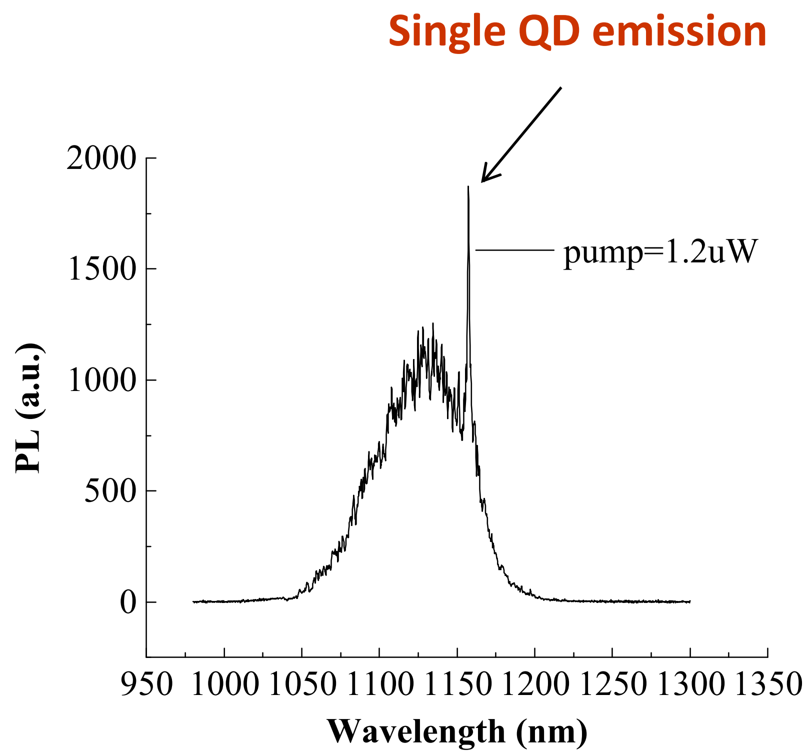




T=8K Results

At low powers it is quite easy to see *very* narrow peaks





Effect of Absorption on SHG

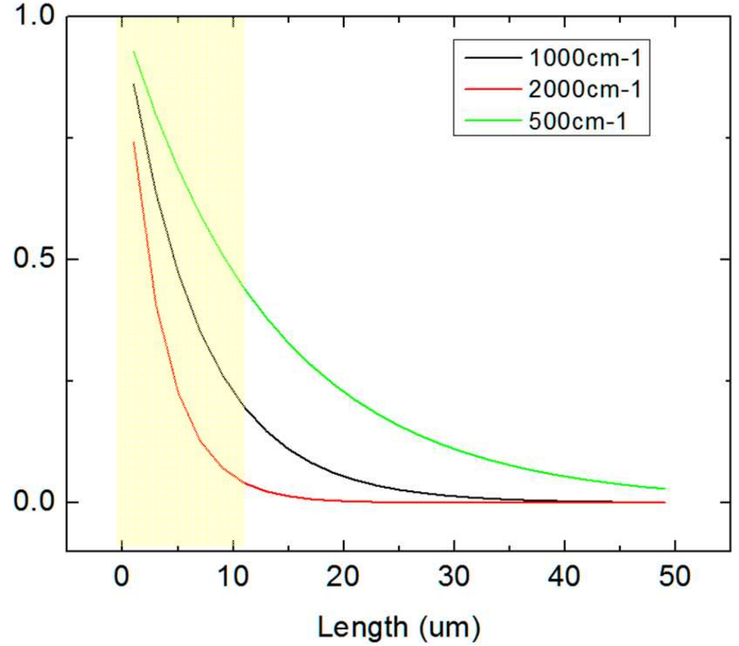
$$E(2\omega) \propto e^{\frac{-\alpha_{2\omega} l}{2}} \int_0^l e^{\alpha' z} \quad , \quad \alpha' = \alpha_\omega - \frac{\alpha_{2\omega}}{2}$$

$$I_{2\omega}(l) \propto e^{-\alpha_{2\omega} l} \frac{\left(e^{-\alpha' l} - 1\right)^2}{\alpha'^2}$$

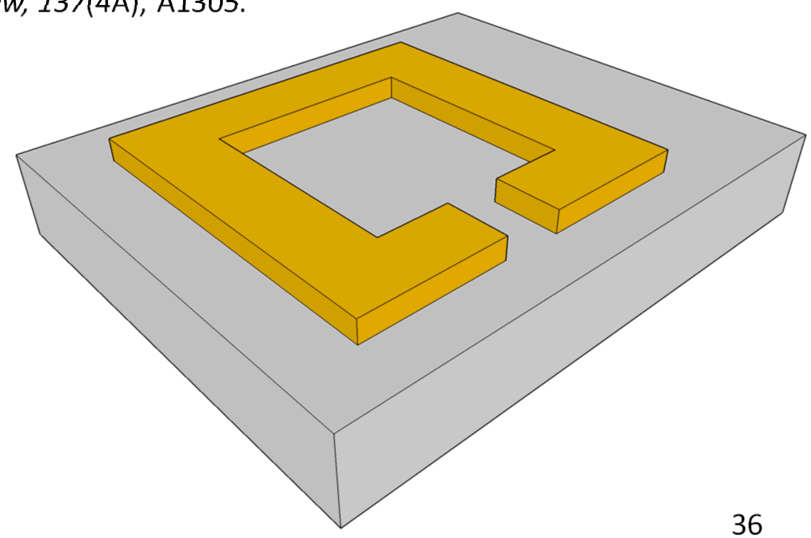
$$\alpha_\omega \approx \alpha_{2\omega} \approx \alpha$$

$$I_{2\omega}(l) \propto e^{-\alpha l} \frac{\left(e^{\frac{-\alpha l}{2}} - 1\right)^2}{\left(\frac{-\alpha l}{2}\right)^2}$$

Boyd, Ashkin, Dziedzic, & Kleinman, (1965). *Physical Review, 137*(4A), A1305.



For α~1000cm^{-1}, "effective length" (*QxL*) needs to be <few microns

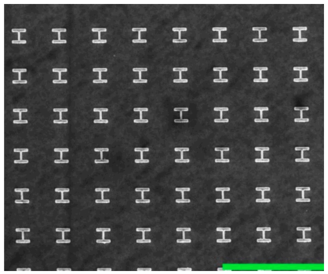


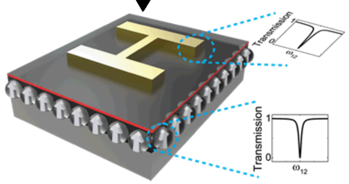
Q~10
Field decay length (*L*) ~200nm
QxL~2um....

Outline
Metasurfaces

Metallic

Planar resonators on III-V substrate

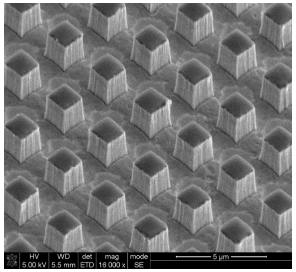


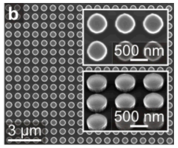


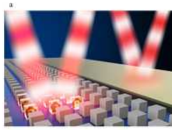
- Strong coupling to phonons, plasmons, intersubband transitions
- Voltage tuning, tunable filters
- Nonlinear optics

Dielectric

Tellurium cubes on BaF2





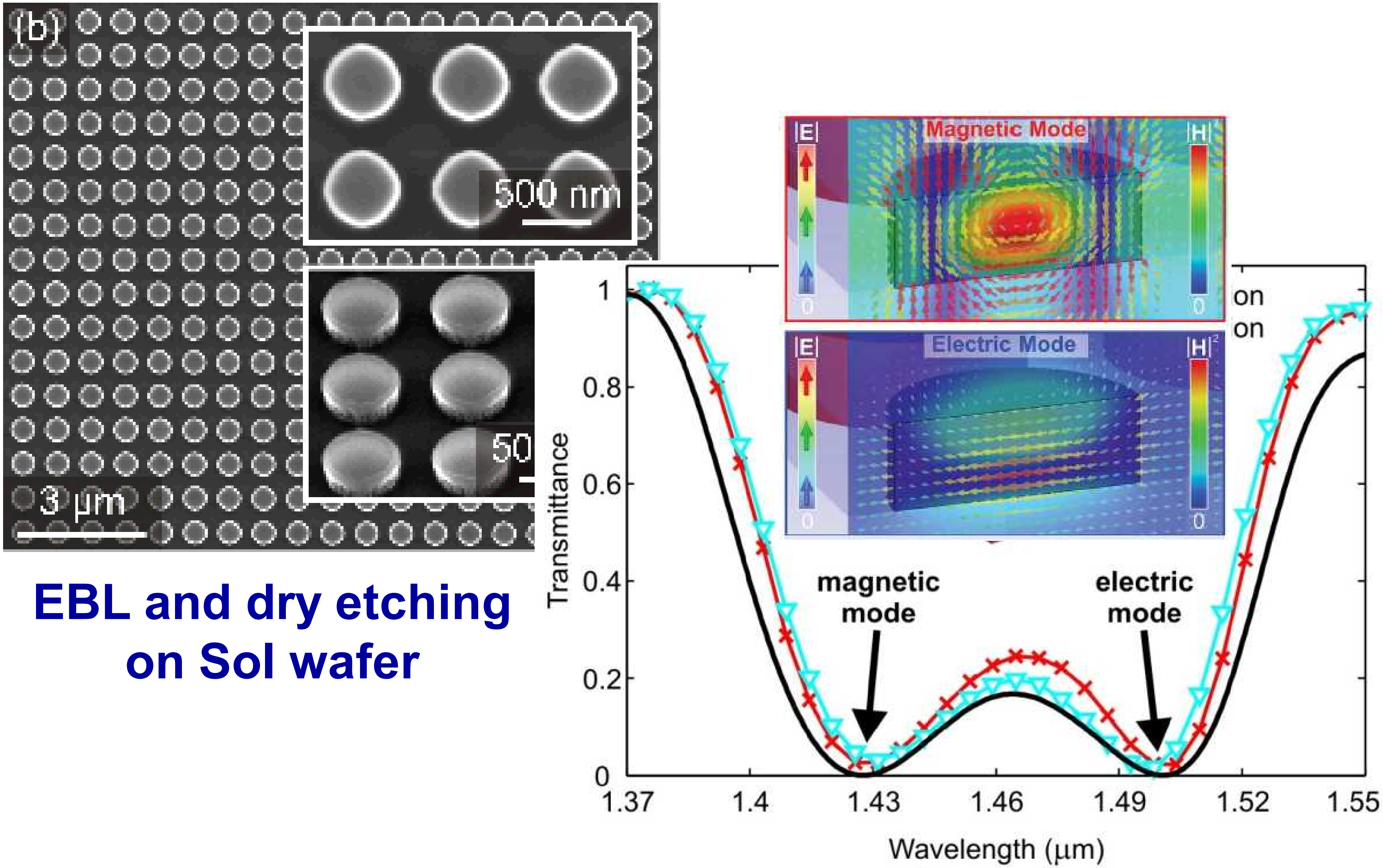


- Optical magnetism
- Unidirectional scattering, Huygens metasurfaces
- Nonlinear optics

Metasurfaces

& Semiconductors

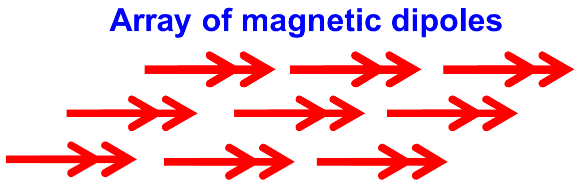
LC tunable dielectric MSs



EBL and dry etching on SoI wafer

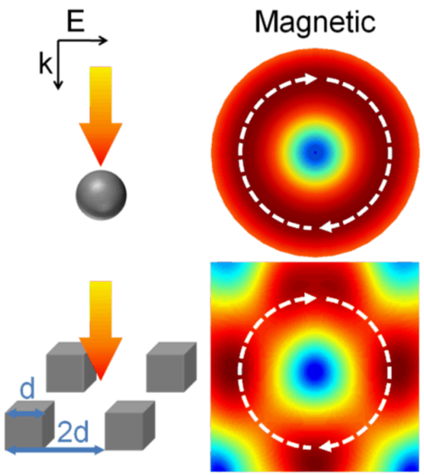
Creating a Magnetic Mirror from a Collection of Magnetic Dipoles

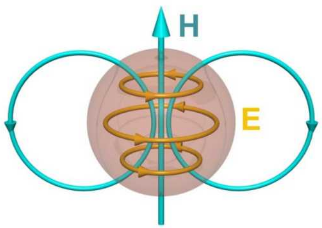
A perfect magnetic conductor does not exist in nature



Because the magnetic dipole responds in phase with the electric field, this represents an artificial magnetic conductor

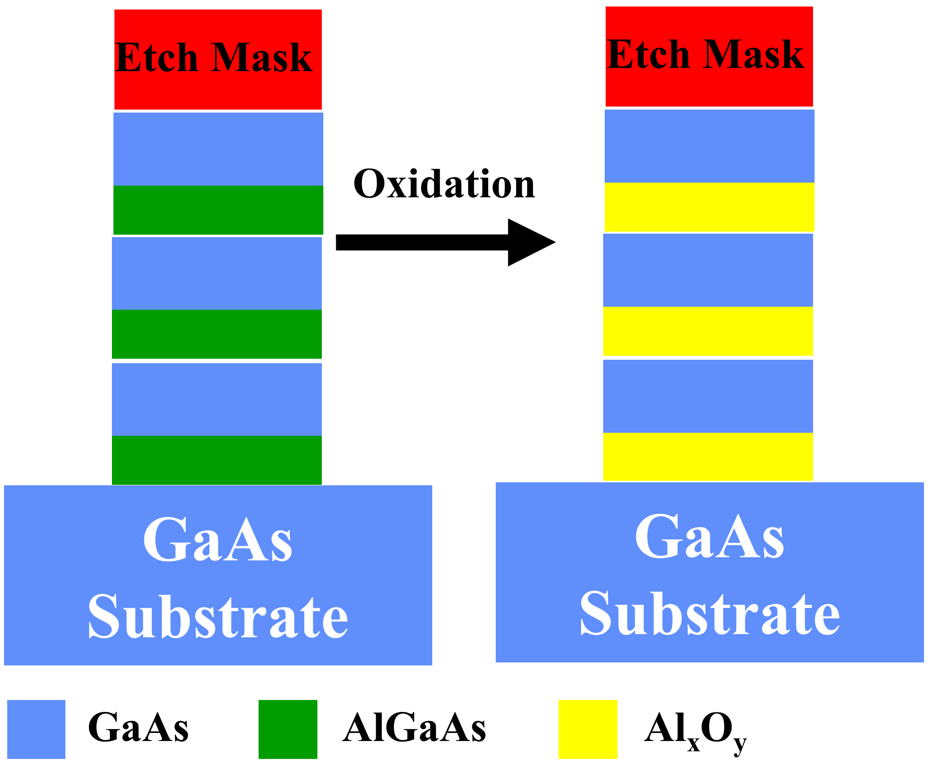
We can create magnetic dipoles with Dielectric Resonators

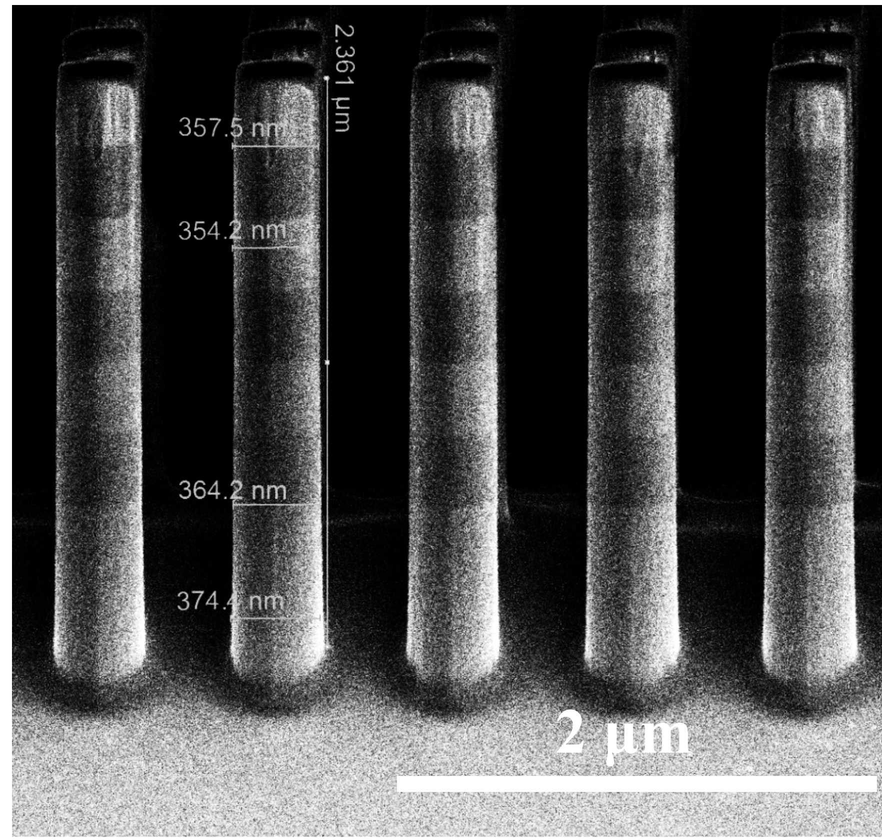




Multilayer III-V Dielectric Metamaterials

Epitaxially grown **multi-layer GaAs/AlGaAs**

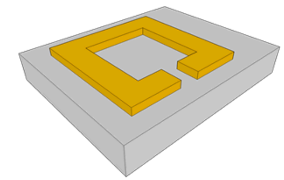




Cavity-enhanced

$I_{2\omega} \sim L_{eff}^2 \sim (QL)^2$

$\chi^{(2)}$



Conventional SHG, perfect phase-matching

$I_{2\omega} \sim (I_\omega \chi^{(2)} L)^2$

$\chi^{(2)}$

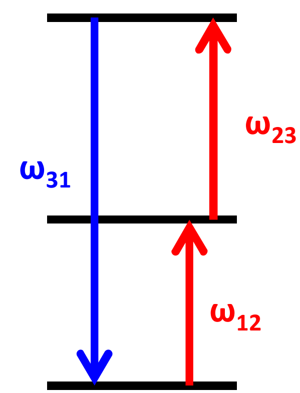
L

I, L

$\chi^{(2)}$

Non-resonant $\chi^{(2)}$, conventional materials , ~10's pm/V

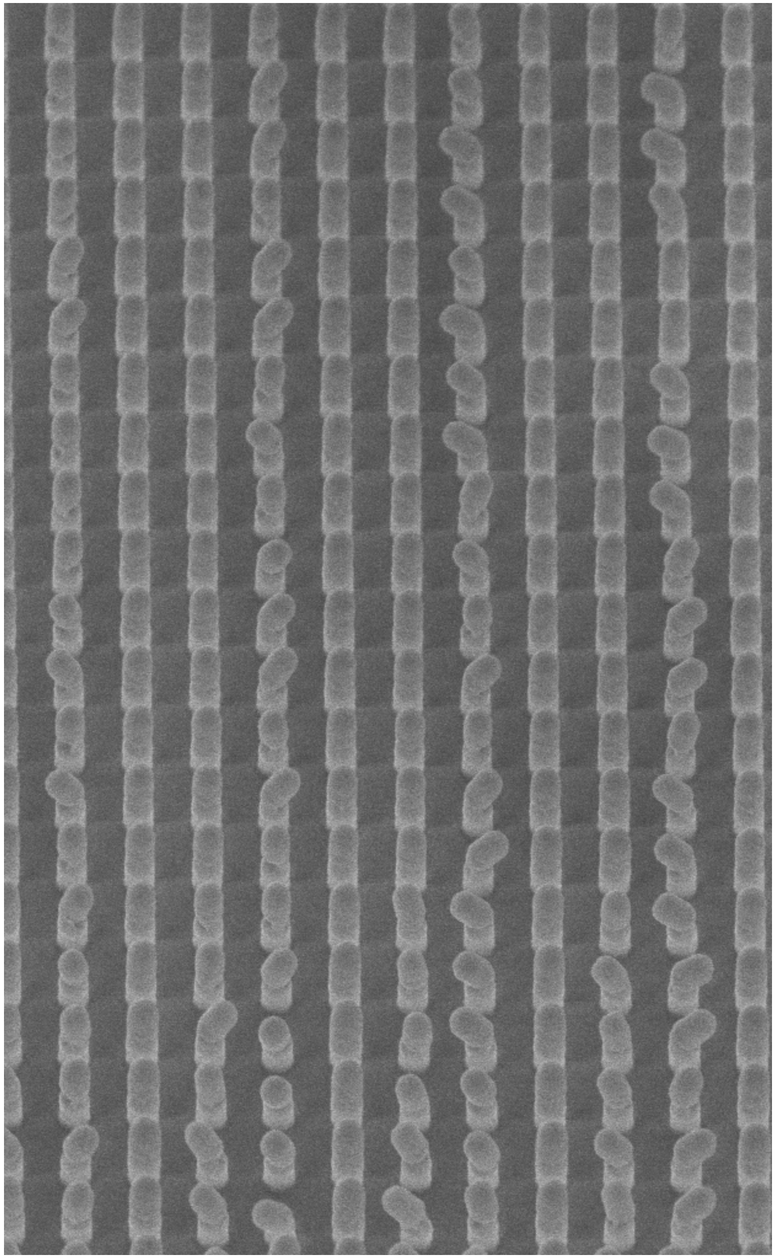
Resonant $\chi^{(2)}$ ~ 250 nm/V (QWs)

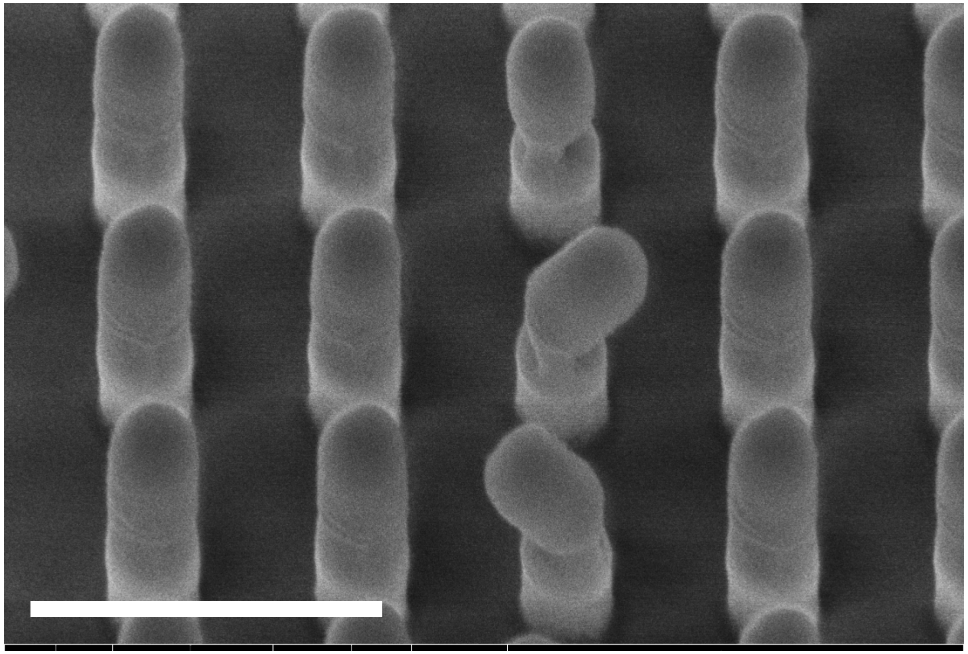


chi(1)

But this comes with absorption!

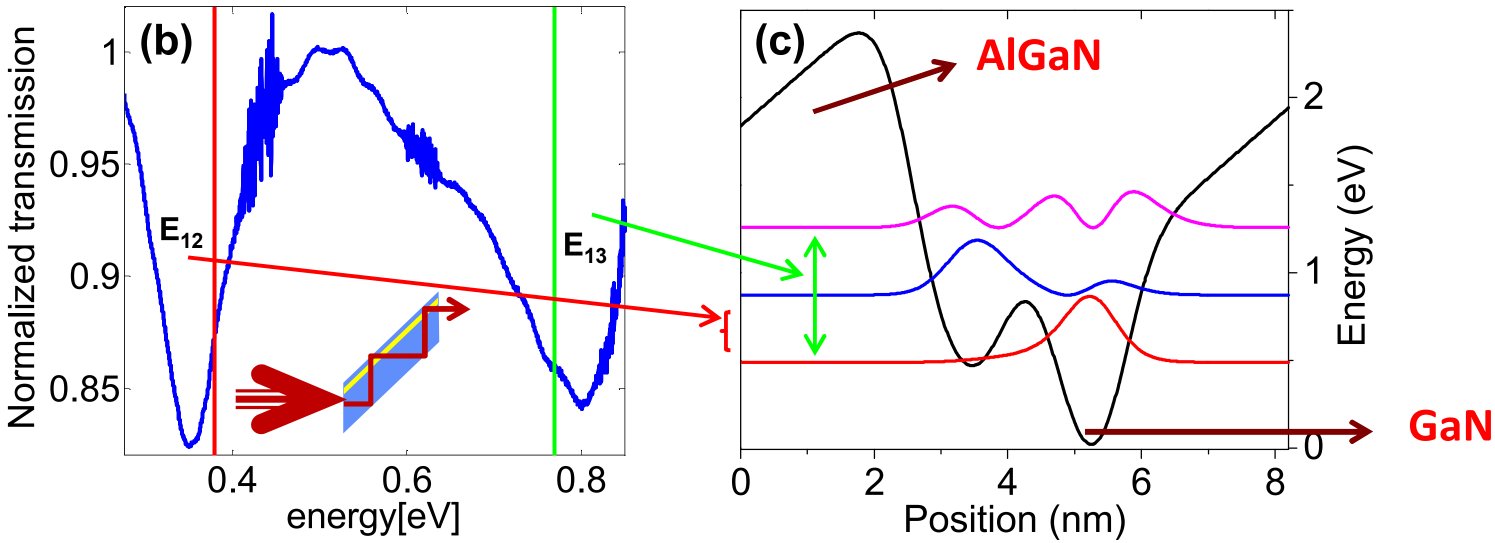
Sample damaged by strong pump and SEM images showing damage.





Low damage threshold, maybe due to etching induced defect on the surface.

SHG (3.2μm>1.6μm) Using AlGaN/GaN QWs



In a III-N heterostructure, metamaterials increase efficiency by over 2 orders of magnitude