SAND2019-10893C

RECENT PROGRESS IN NONLINEAR OPTICS FROM METASURFACES AND EPSILON NEAR ZERO MATERIALS

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Sandia National Labs and Center for Integrated Nanotechnologies

ICQNN, Sofia, 2019

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LDRD LABORATORY DIRECTED RESEARCH & DEVELOPMENT
U.S. DEPARTMENT OF ENERGY Office of Science

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Revisiting Nonlinear Optics

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

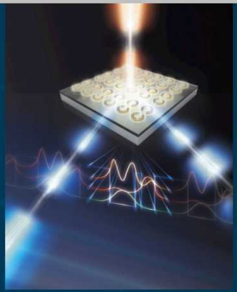
High nonlinearity

High fields

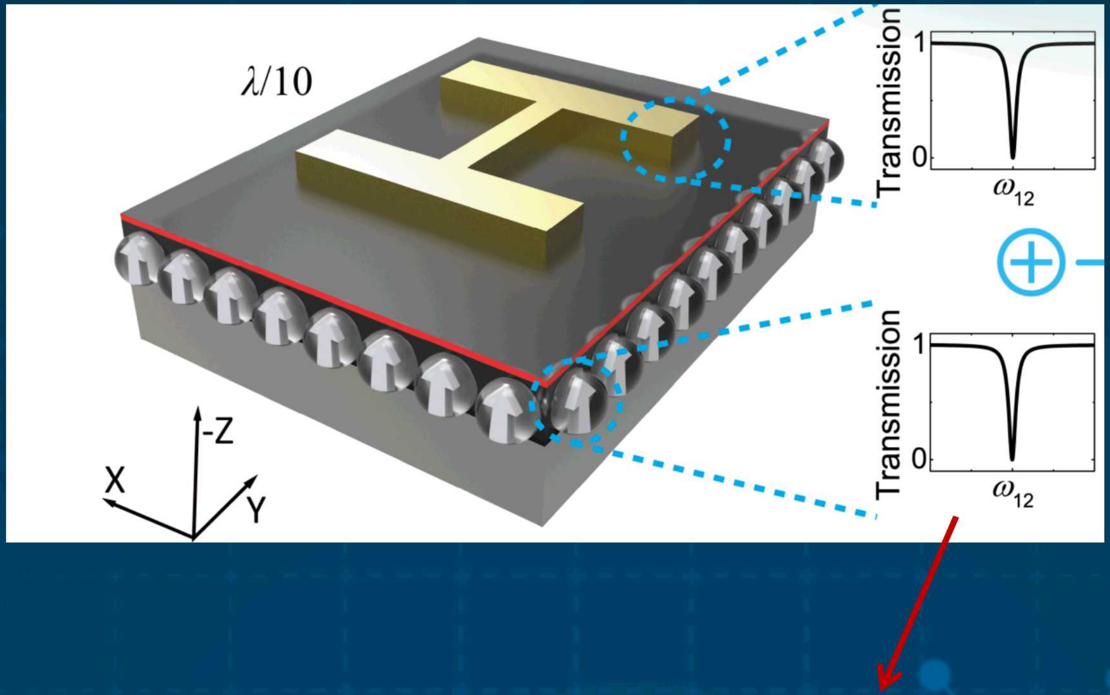
Energy and momentum conservation: phase matching

Paths to Nonlinear Metasurfaces

- Metallic/Plasmonic over "engineered" nonlinearity



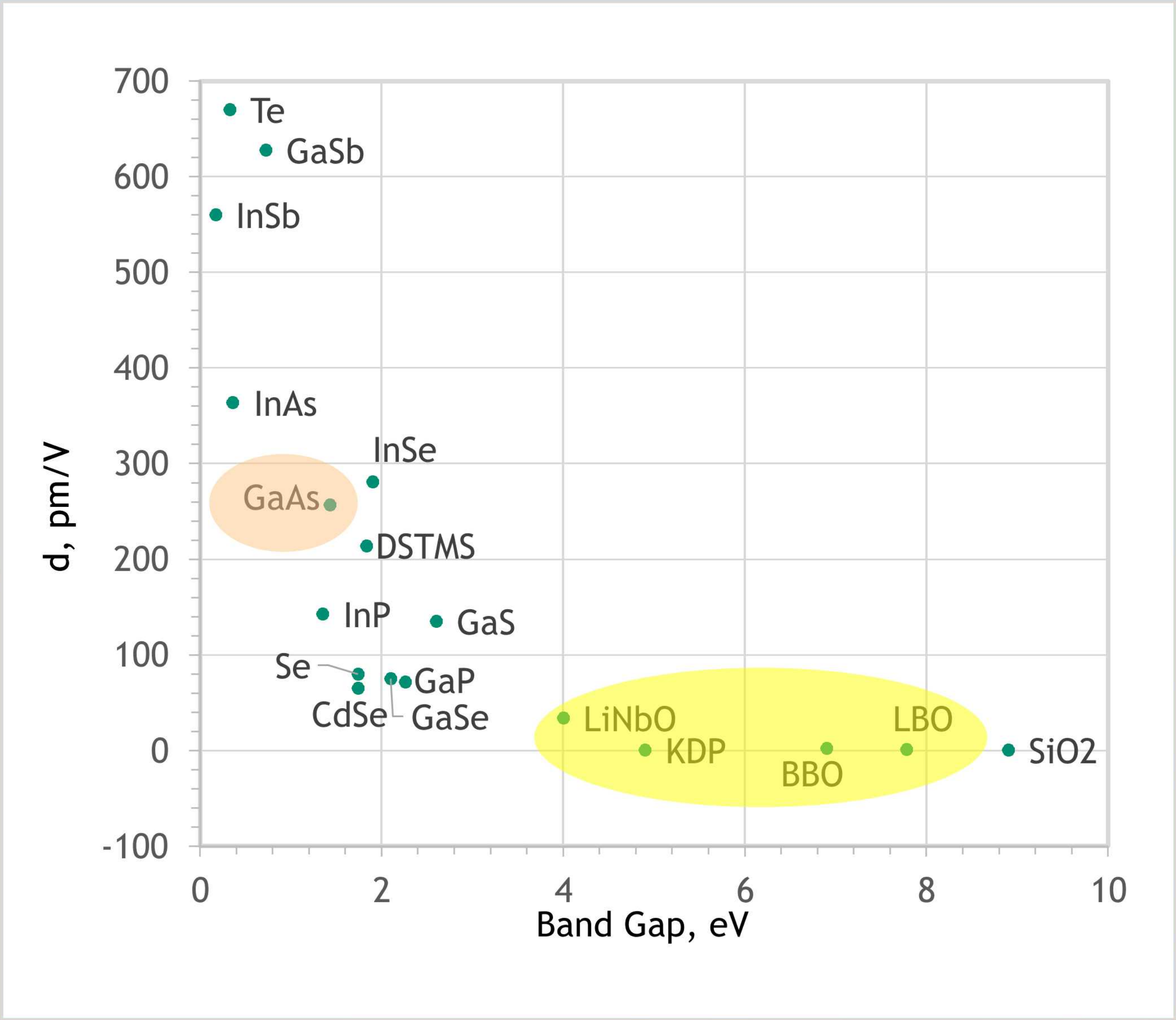
1. Nonlinear Metasurfaces: Metallic Resonators + Engineered Nonlinearity



For linear phenomena:

Optical Phonons: Nano Letters 11, 2104 (2011)
Epsilon Near Zero modes: Nano Letters 13, 5391 (2013)
Intersubband Transitions: Nature Communications 4, (2013)

Materials for NLO (i.e., SHG)



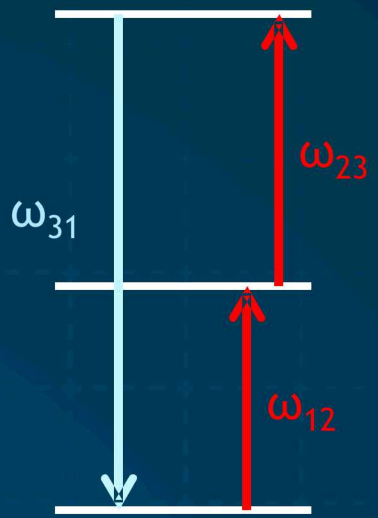
Intersubband transitions ~x1000

Resonant $\chi^{(2)}$ in a Three-Level System

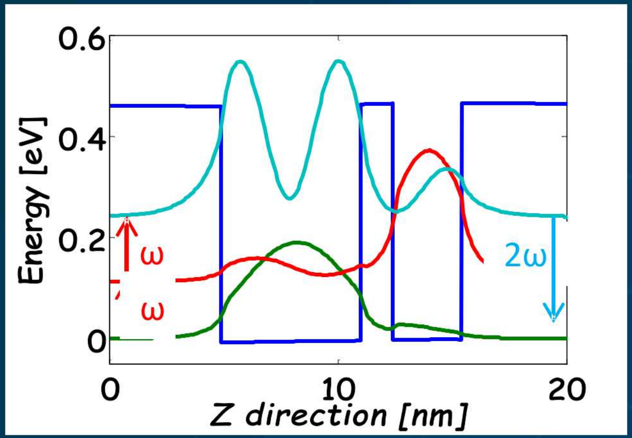
Using perturbation theory:

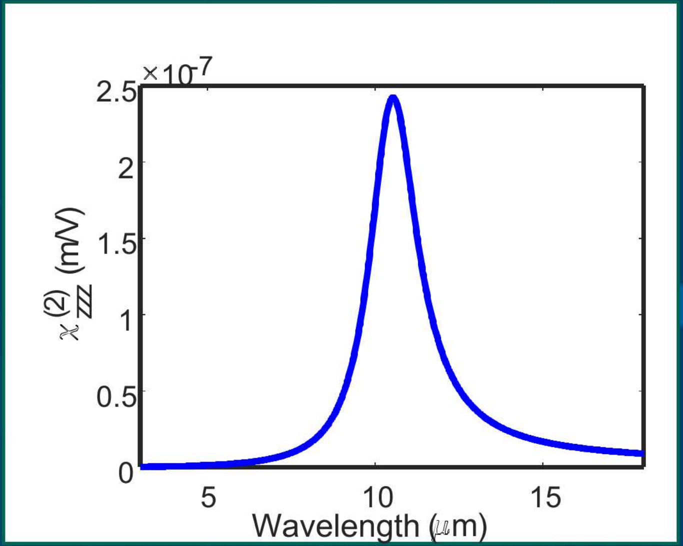
$$\chi^{(2)}(\omega) \propto \frac{N \cdot z_{12} z_{23} z_{13}}{(\omega - \omega_{12} - i\Gamma_{12})(2\omega - \omega_{13} - i\Gamma_{13})}$$

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



refs





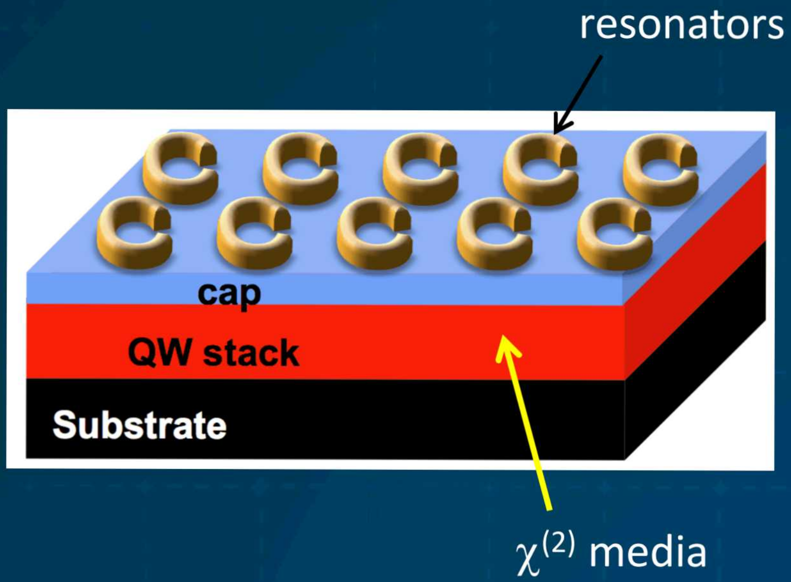
~250 nm/V - QWs

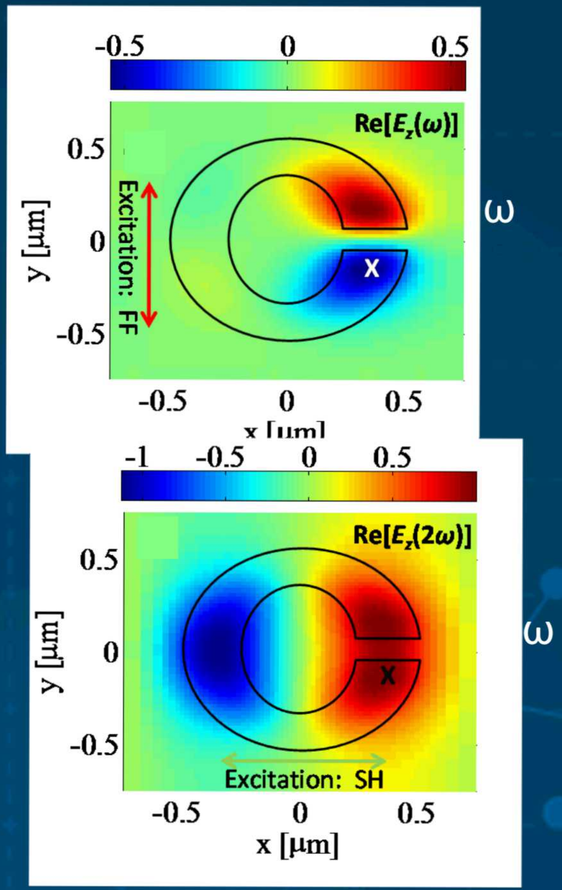
~15 of pm/V - $LiNbO_3$

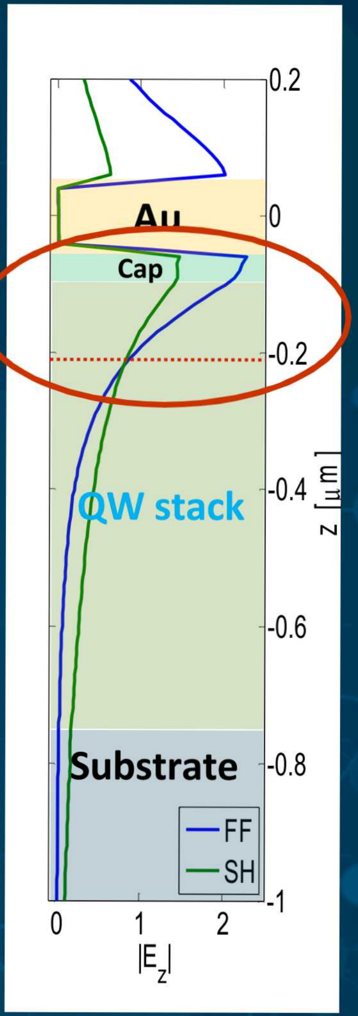
IEEE J. Quantum Electron. 30, 1313 (1994)

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

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



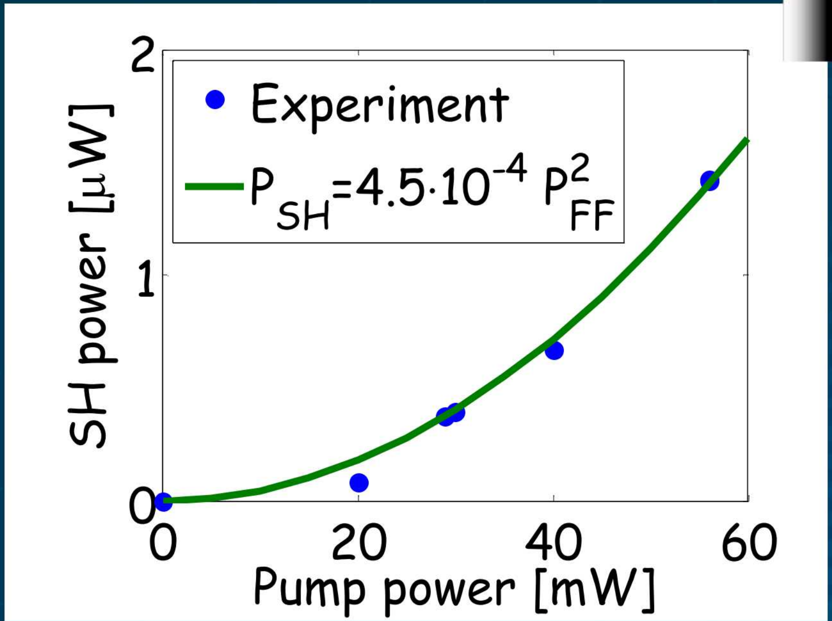


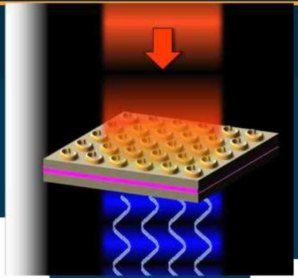


APL **104**, 131104 (2014)

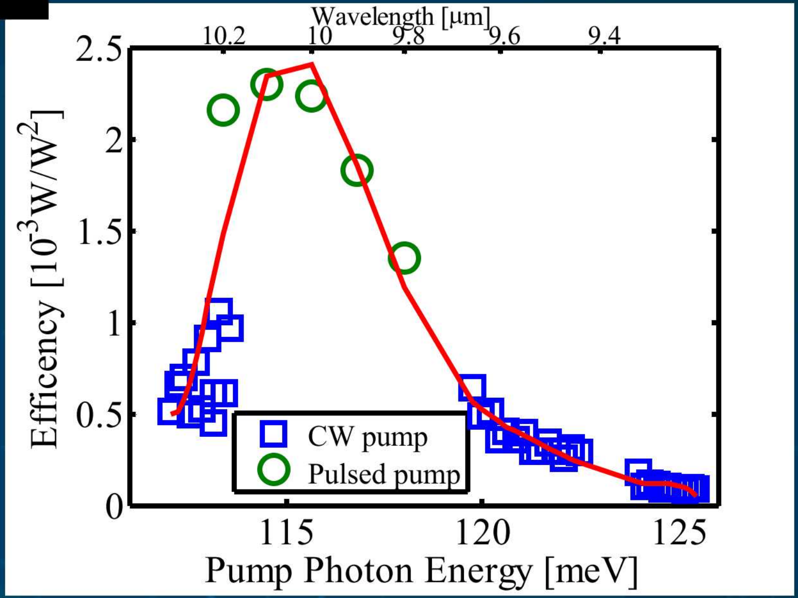
SHG: Power and Frequency Dependence

Pump: CW CO_2 laser





max efficiency: ~2.3 mW/W^2



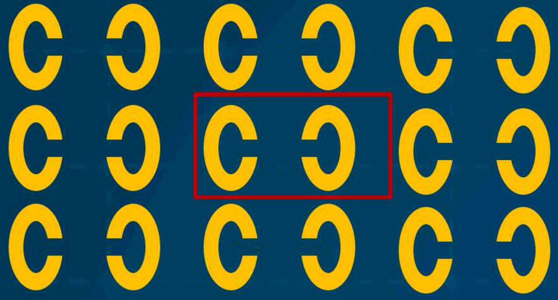
- Saturation and damage are issues
- Max. conversion efficiencies of few % are possible
- Added functionality is the advantage

Nature Communications 6, 7667 (2015)
See also: Lee, Belkin, Alu, et al. Nature 511, 65-69 (2014).

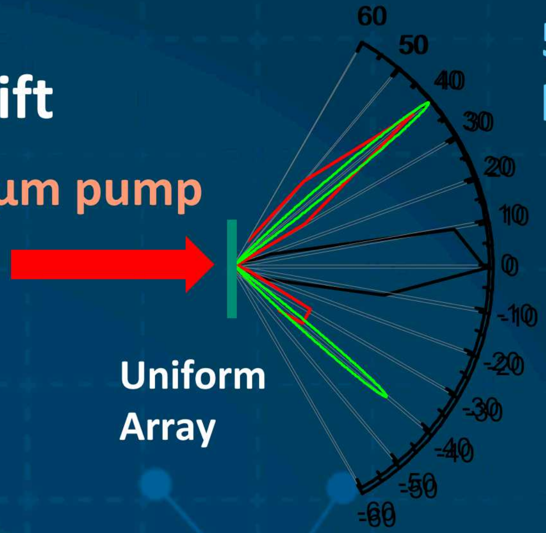
SHG Beam Manipulation

“edge-fed dipole”

flipping resonators induces π phase shift



10µm pump



Uniform Array

5µm beam

2-in-1:

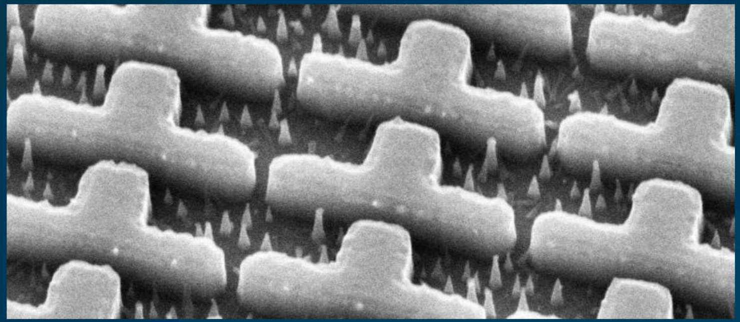
Source +beam splitter

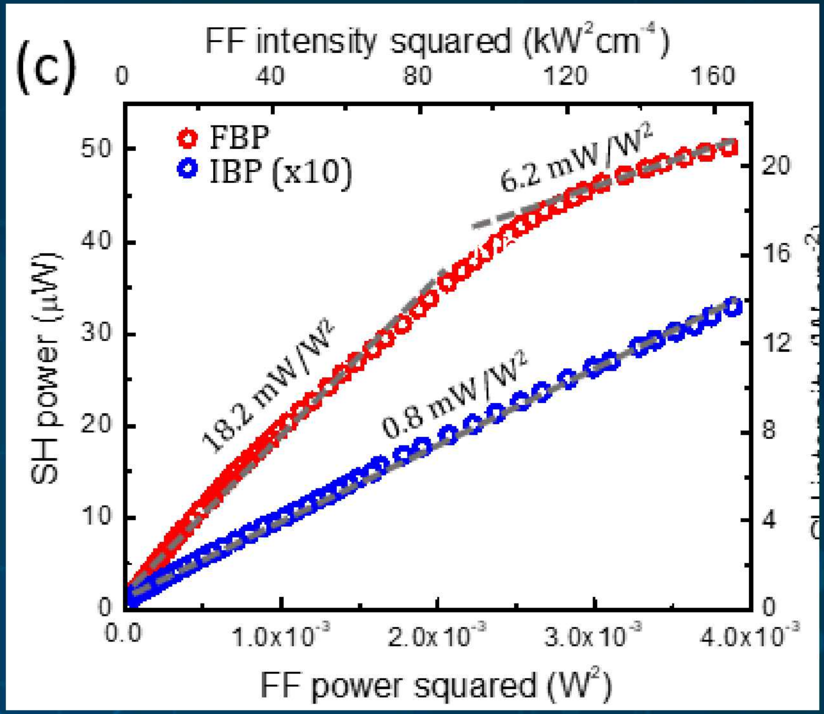
More complex manipulation is possible, depending on metasurface arrangement
See also work by Alu&Belkin, and Ellenbogen

Nature Communications 6, 7667 (2015)

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

Local field enhancement (100 nm beneath top Au layer)





Max efficiency:

18.2 mW/W² (Belkin & Brener 2018)

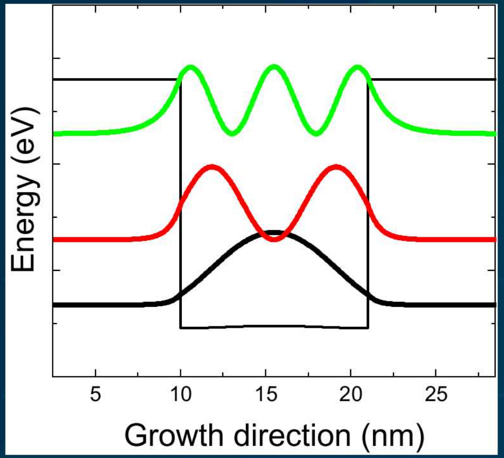
$\chi^{(2)}_{eff}$~ $1.31\times10^{6}\ pmV^{-1}$

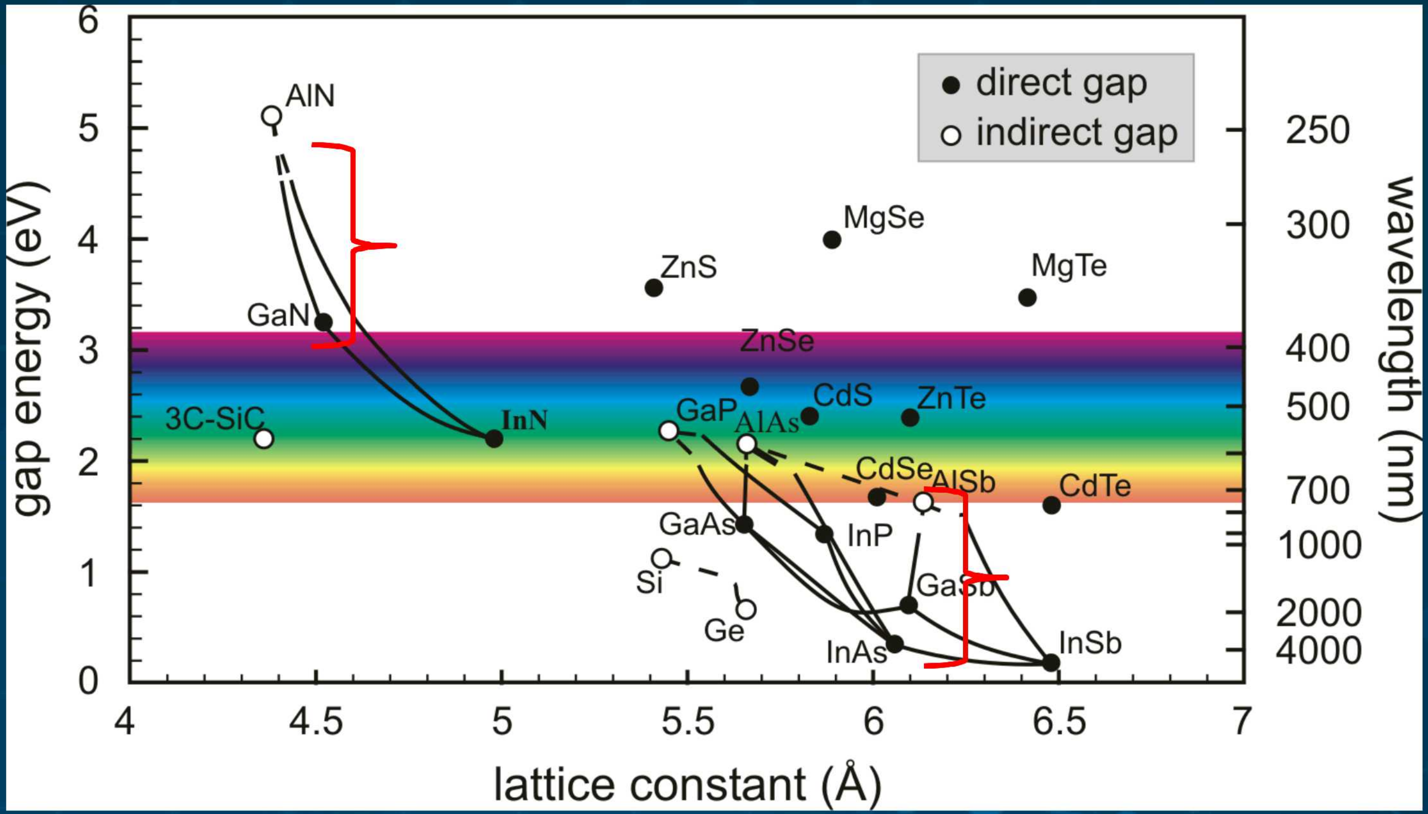
(With Belkin's group)

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

Scaling to Shorter Wavelengths

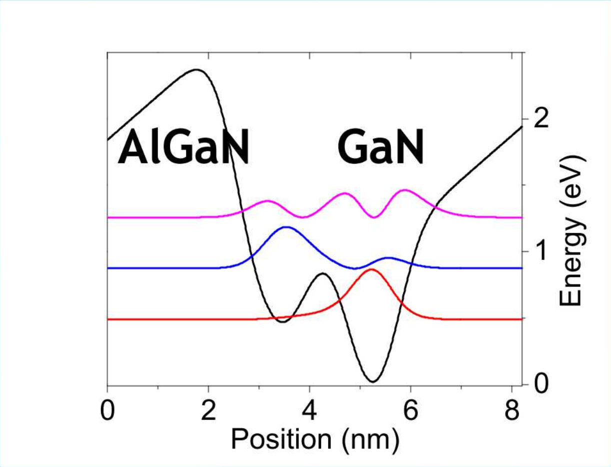
Material 1 Material 2 Material 1

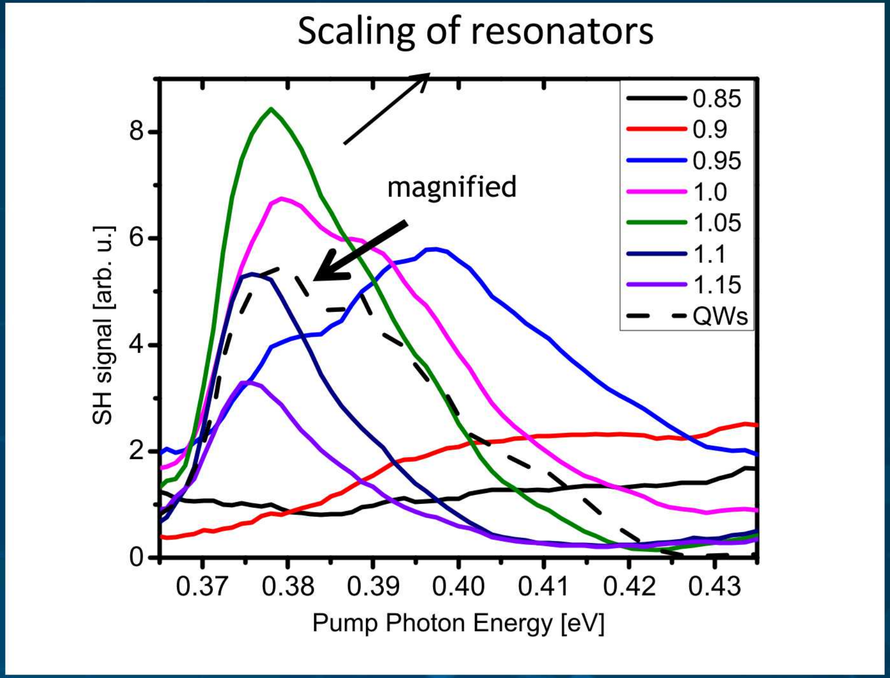




http://gorgia.no-ip.com/phd/html/thesis/phd_html/simone-jbook.html

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

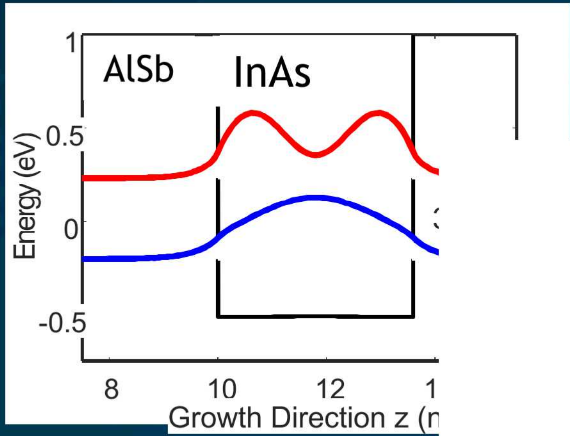


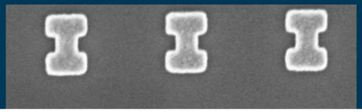


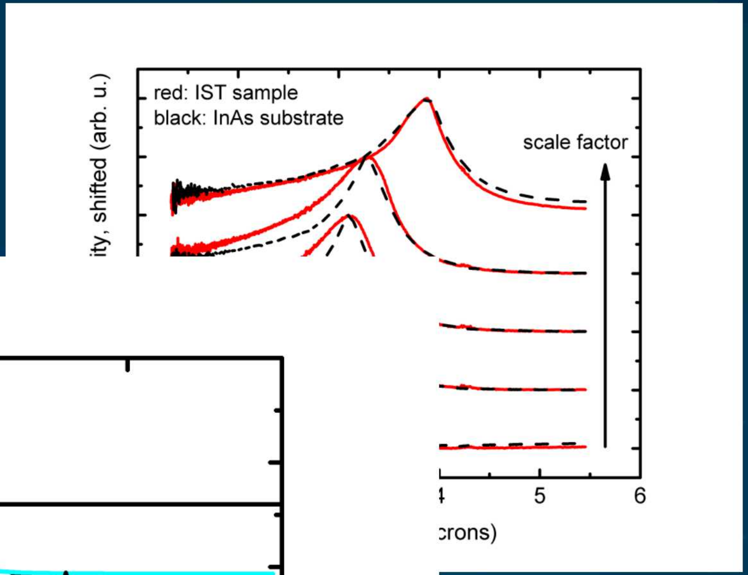
APL107, 151108 (2015)

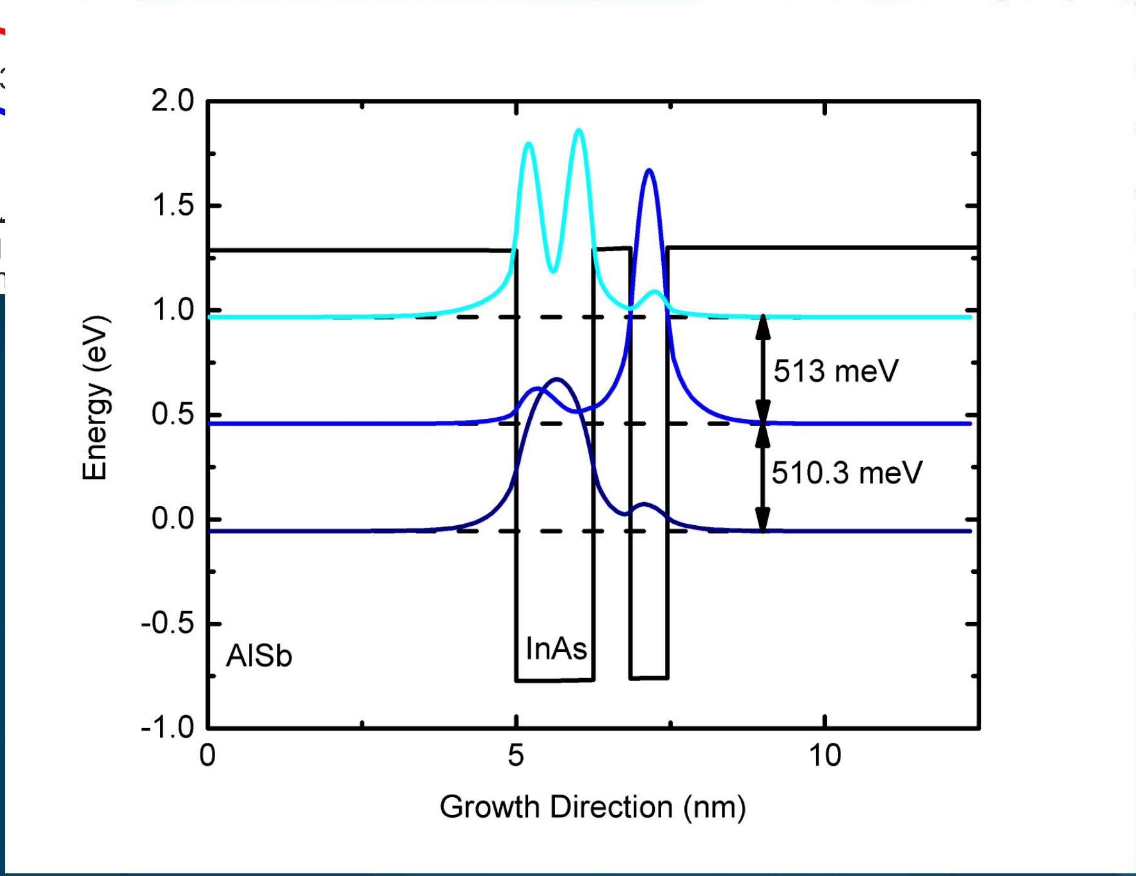
Near IR SHG (SHG @~1.2μm) Using AlSb\InAs QWs

The Conduction Band Offset of 2.1 eV





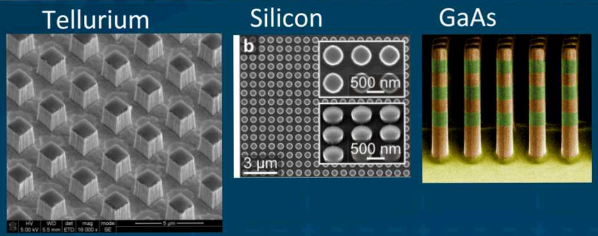




Design for SHG

Paths to Nonlinear Metasurfaces

- All dielectric

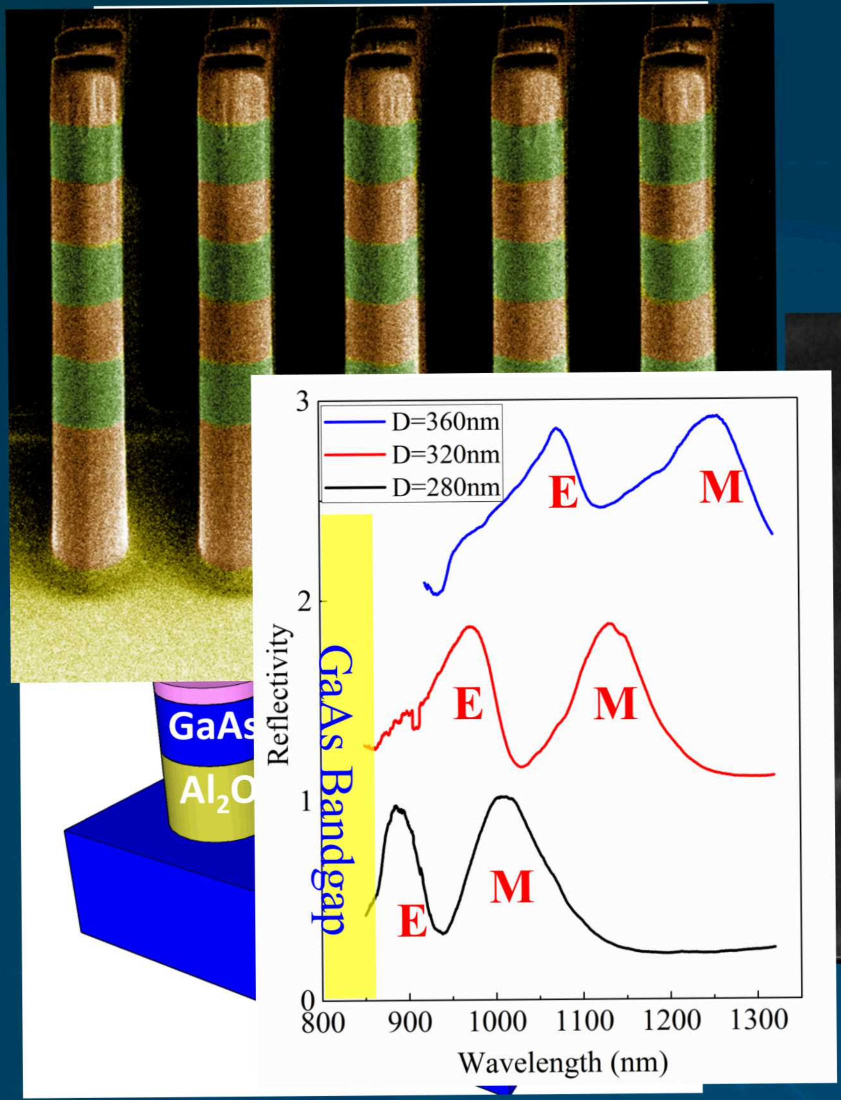


I. Nonlinear All-Dielectric III-V Metasurfaces

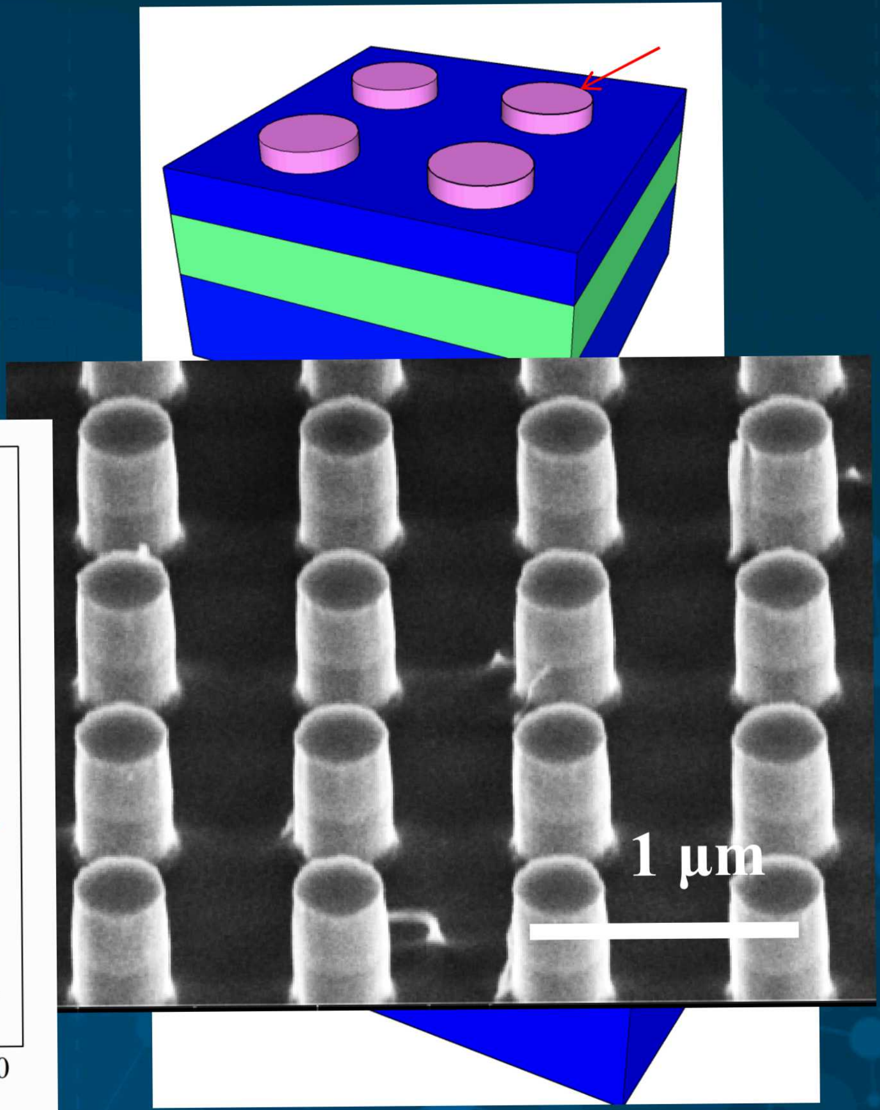
Epitaxially grown: MBE, MOCVD

n~3.5

n~1.6

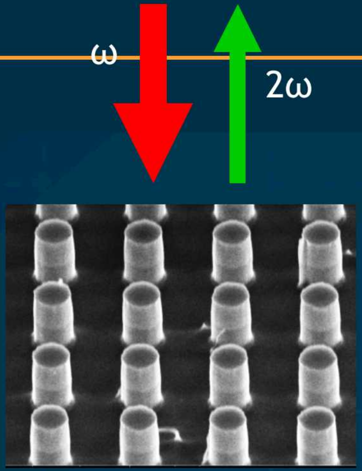


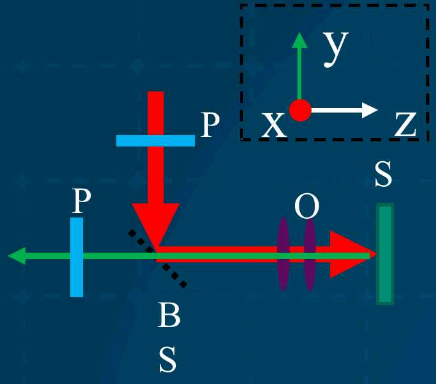
Etch mask

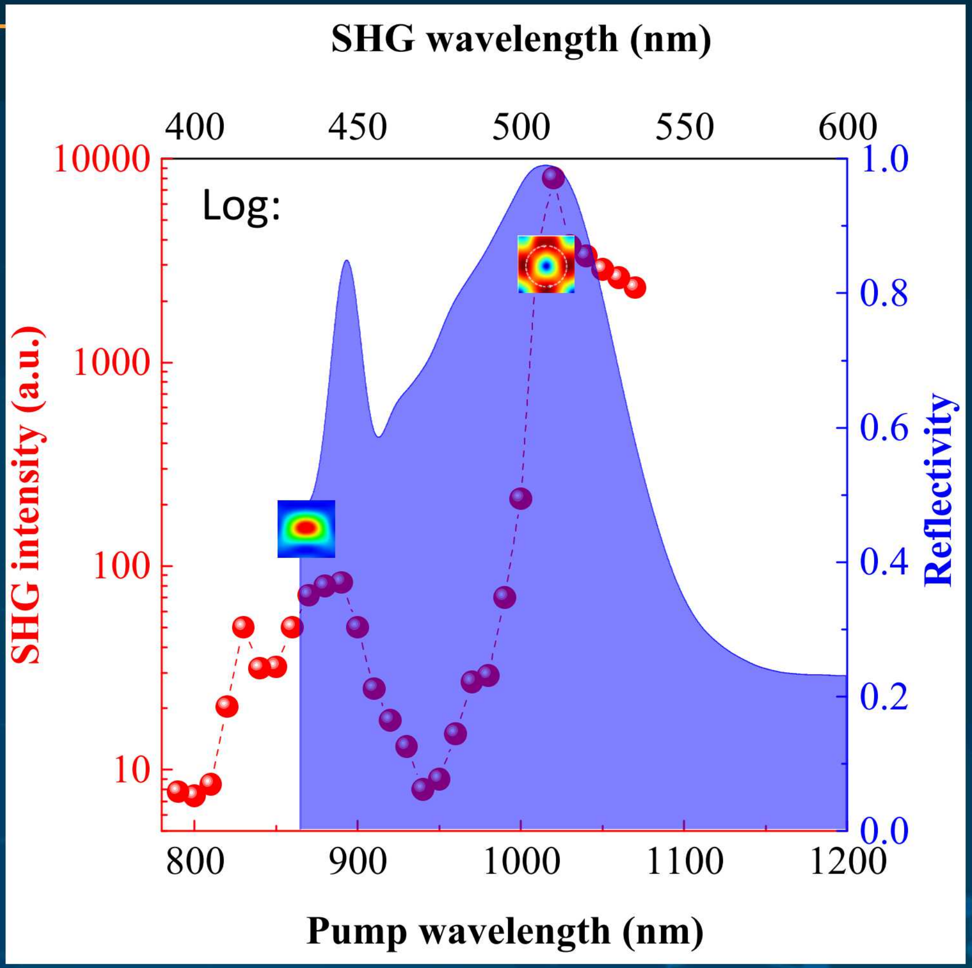


Advanced Opt. Mat 2016

I. Nonlinear All-Dielectric III-V Metasurfaces: SHG





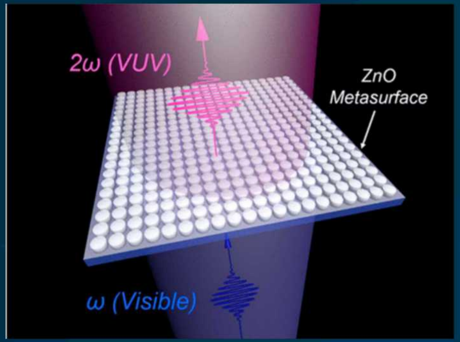


ACS Photonics 2018, 5, 1786–1793

Nano Letters 2016
Also ANU, Costa (Italy)

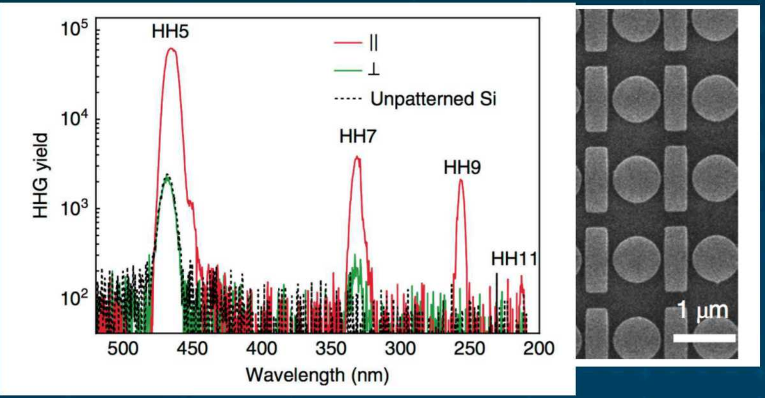
Nonlinear Optics with All-dielectric Metasurfaces

Vacuum Ultraviolet Light Generation



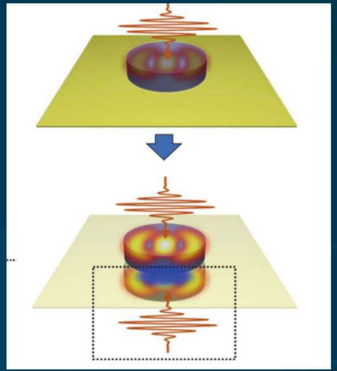
Semmlinger, M. et al. *Nano Lett.*, Article ASAP
DOI: 10.1021/acs.nanolett.8b02346

High-harmonic generation



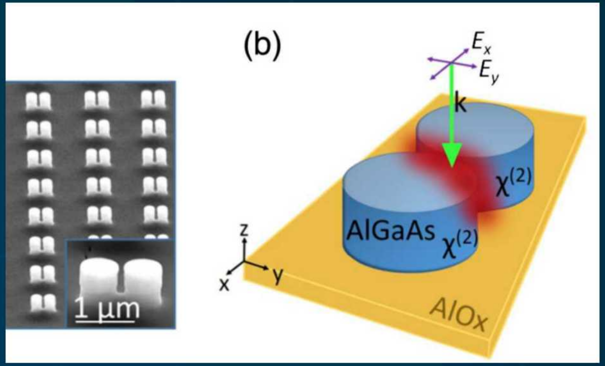
Liu, H., et al. *Nature Physics* (2018): 1.

THG in a mirror-enhanced anapole resonator



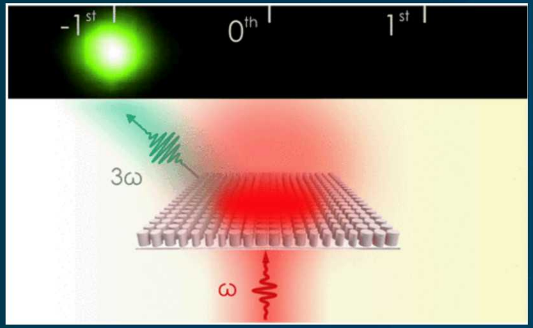
Xu, Lei, et al. *Light: Science and Applications* 7 (2018): 1-8.

Tuning the SHG in AlGaAs nanodimers



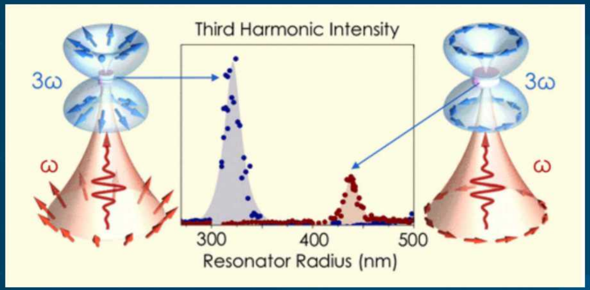
Rocco, D., et al. *Photonics Research* 6.5 (2018): B6-B12.

Nonlinear wavefront control



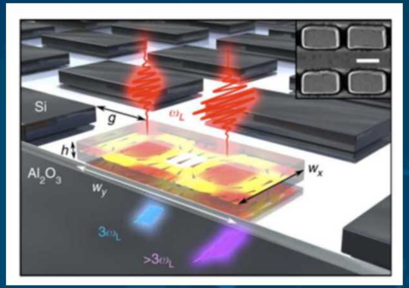
Wang, L., et al. *Nano letters* *18* (6) (2018): 3978–3984.

Selective Third-Harmonic Generation



Melik-Gaykazyan, E. V., et al.
ACS Photonics 5.3 (2018): 728–733.

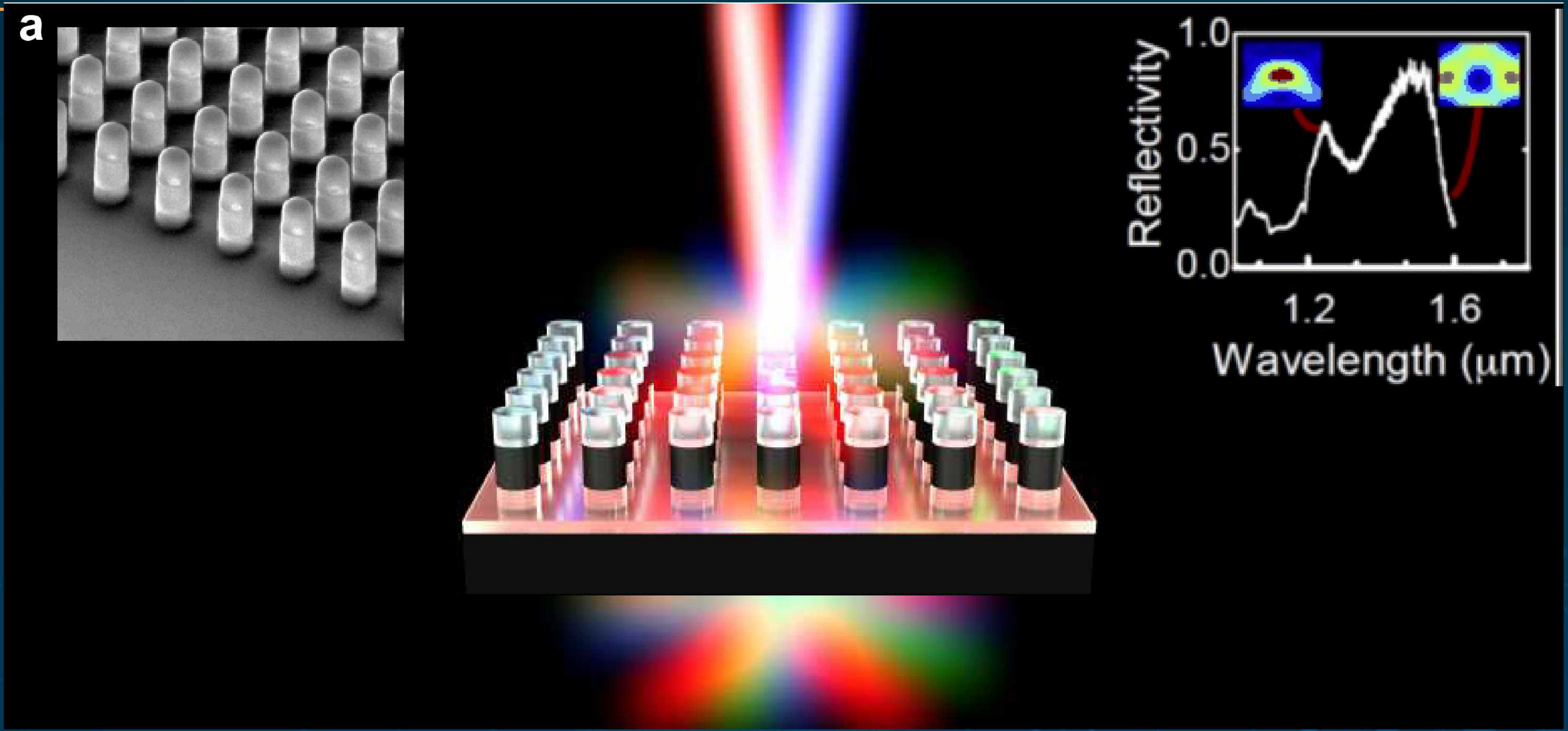
Tunable THG



Shcherbakov, Shvets et al, Nature Comm. 2019

And a lot of good work by other groups: Kivshar, Neshev, Staude, Valentine, Maier, de Angelis, etc

Higher Harmonics: Mixing Several Beams

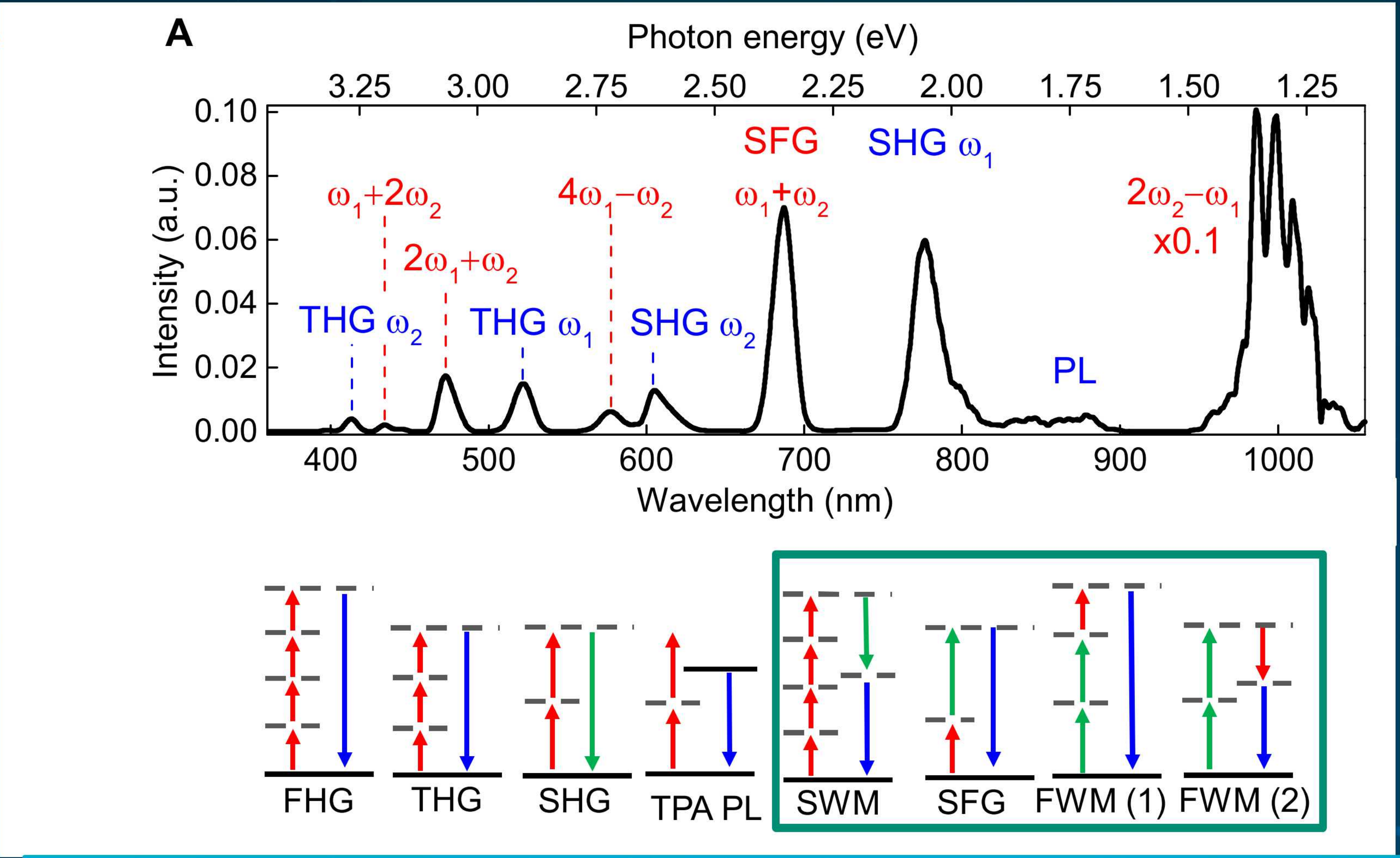


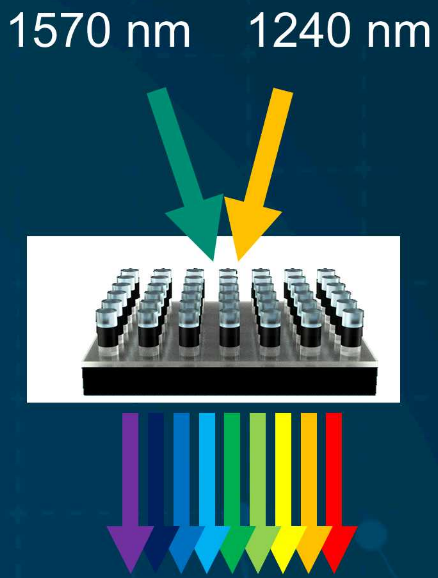
The closest thing to an electronic mixer for optics

Nature Comm. 2018

Polina Vabishchevich

Frequency mixing spectra

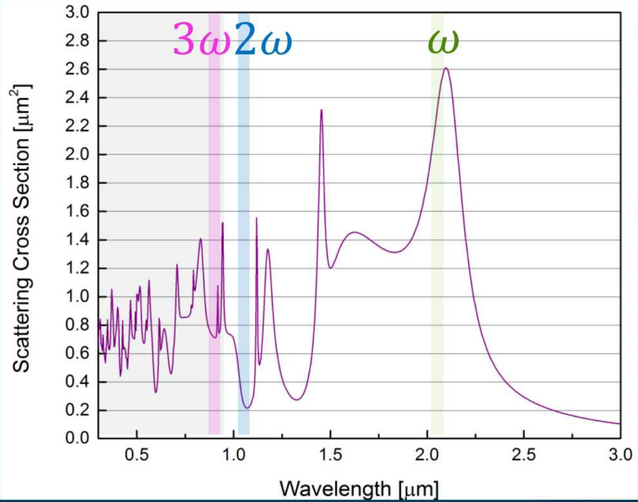




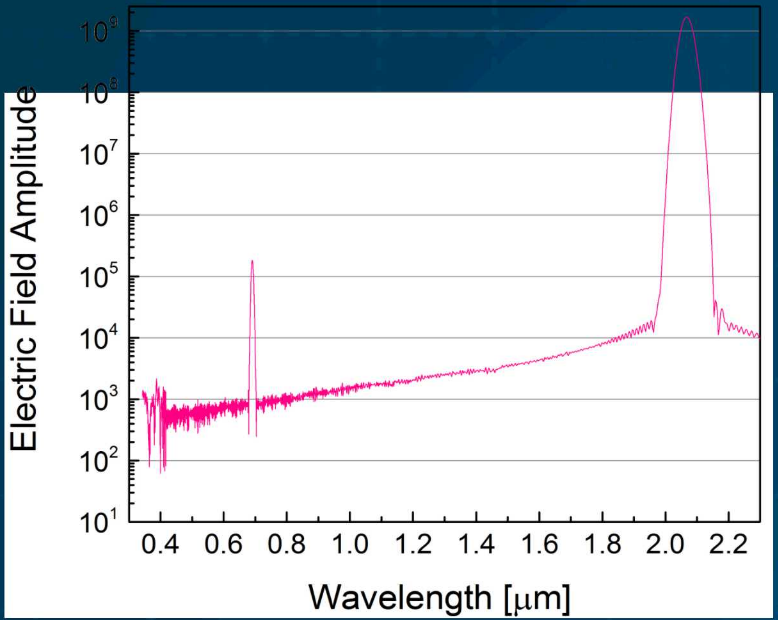
Nature Comm. 2018

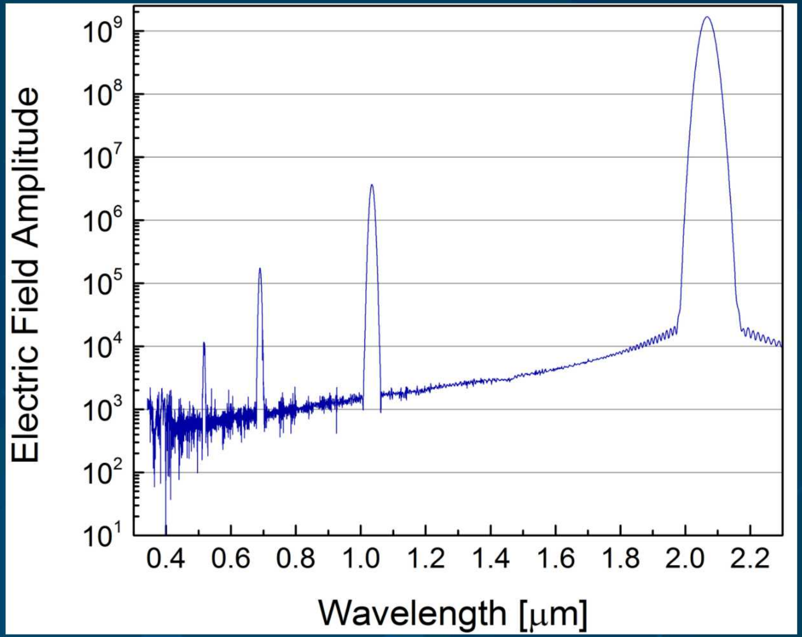
7 different nonlinear processes: 2nd, 3rd, 4th harmonics, sum frequency generation, 4 wave-mixing, six-wave mixing:
simultaneous high order processes with no phase matching and comparable efficiency

$\chi^{(3)}$, $\chi^{(4)}$, $\chi^{(5)}$,…. or Not?



Cascaded nonlinearities?





FOM:

$$\frac{\chi^{(2)}(2\omega;\omega,\omega)\chi^{(2)}(3\omega;2\omega,\omega)}{\epsilon_r(2\omega)\chi^{(3)}(3\omega;\omega,\omega,\omega)}$$

$\chi^{(3)}(3\omega;\omega,\omega,\omega) \propto 10^{-16}\ m^2\backslash V^2$

$\chi^{(2)}(2\omega;\omega,\omega) \propto 10^{-8}\ m\backslash V$

Back to the future….

IEEE PHOTONICS TECHNOLOGY LETTERS, VOL. 11, NO. 6, JUNE 1999

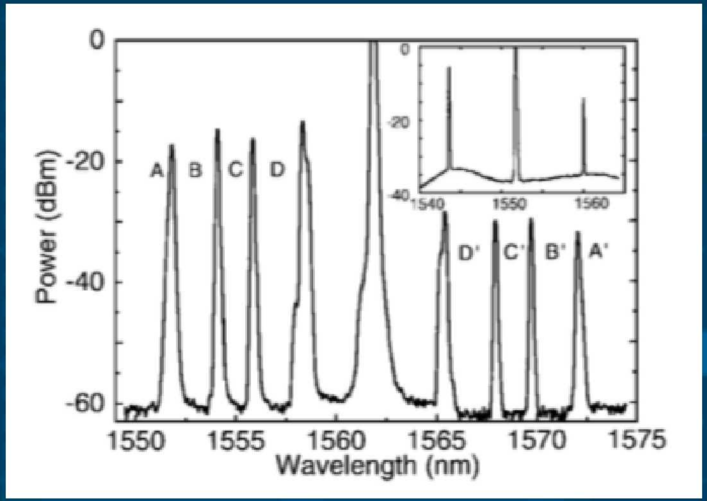
1.5-μm-Band Wavelength Conversion Based on Cascaded Second-Order Nonlinearity in $LiNbO_3$ Waveguides

M. H. Chou, I. Brener, M. M. Fejer, E. E. Chaban, and S. B. Christman

82 IEEE PHOTONICS TECHNOLOGY LETTERS, VOL. 12, NO. 1, JANUARY 2000

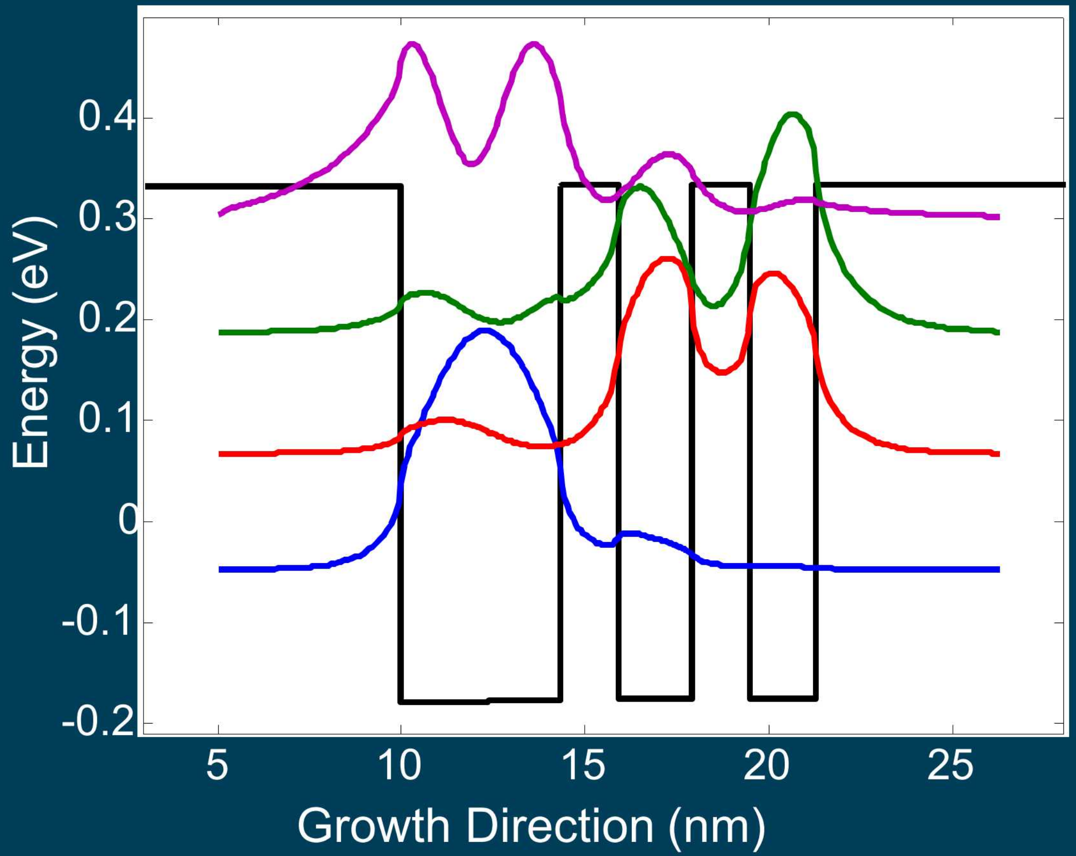
Efficient Wide-Band and Tunable Midspan Spectral Inverter Using Cascaded Nonlinearities in $LiNbO_3$ Waveguides

M. H. Chou, *Member, IEEE*, I. Brener, *Member, IEEE*, G. Lenz, R. Scotti, *Member, IEEE*, E. E. Chaban, J. Shmulovich, D. Philen, S. Kosinski, K. R. Parameswaran, *Member, IEEE*, and M. M. Fejer, *Member, IEEE*



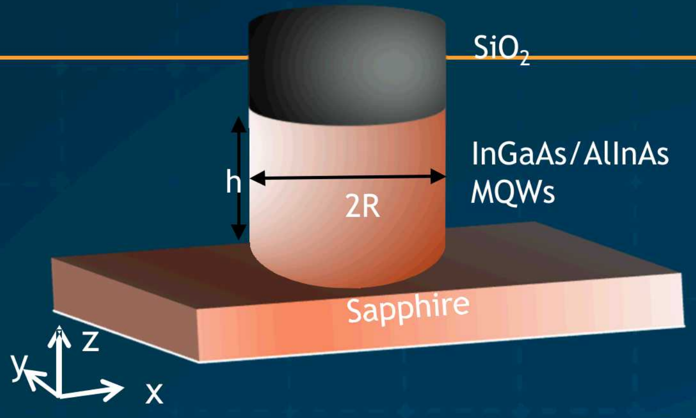
The Best of Both Worlds: Mie Metsurfaces and IST's!

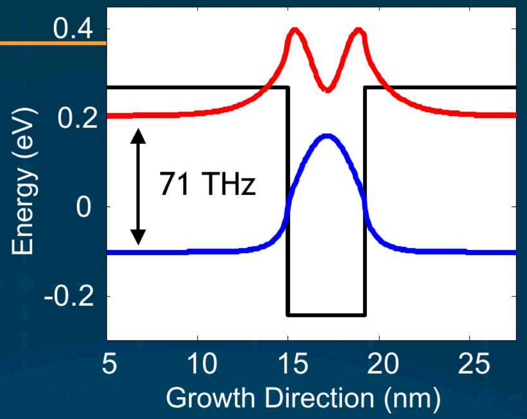
4 levels -> $\chi(3)$



- Height (h) determines the number of repetitions of QWs we can embed into the resonator.
- MD resonance can be spectrally scaled by changing R_{cyl} (h = 1.25 µm for our case)

Precursor to Nonlinear Experiments: Strong Coupling

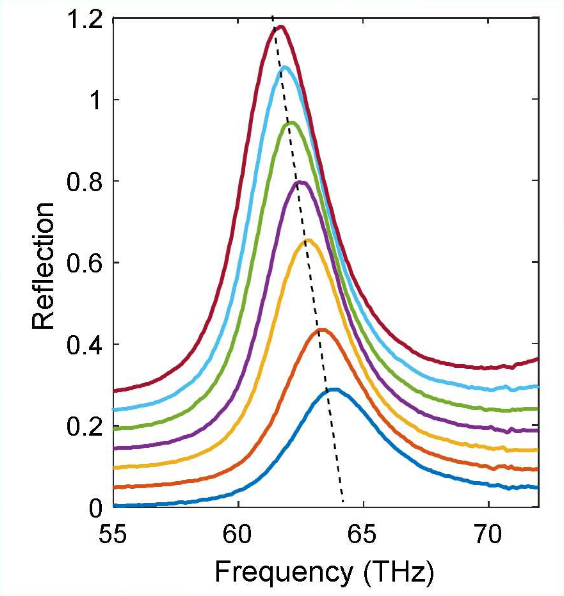




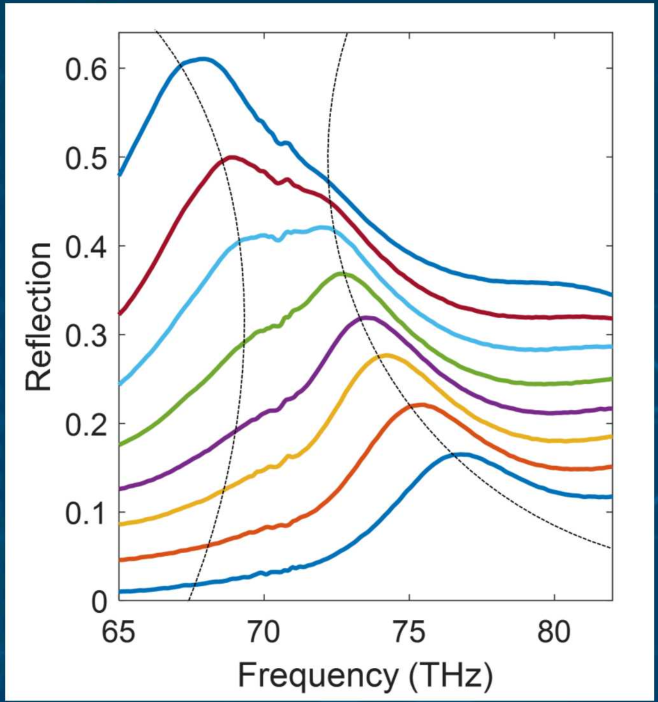
$$\Omega_{rabi} \propto \sqrt{N}$$

N~ number of carriers ~number of wells for fixed doping

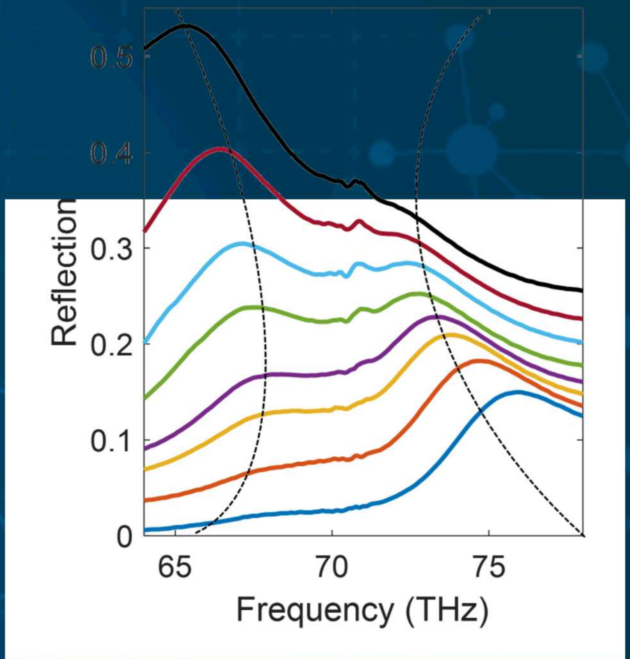
No coupling : Linear scaling



Ω_{rabi}=5 % (N-)



Ω_{rabi}=10 % (N++)



Comparison of SHG (normalized by spot size)

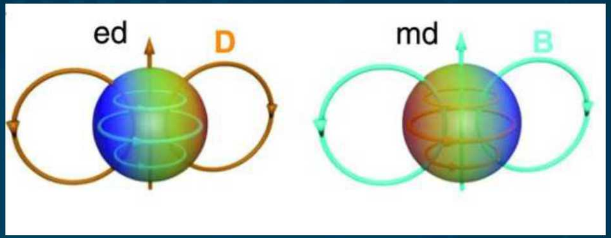
Nonlinear Metasurface Approaches	Maximum Normalized Conversion Efficiency ($W/W^2/cm^2$)
IST + Plasmonic Resonator + Metal Backplane + Etching of QWs	1746 (Experimental)
IST+ Leaky Mode Resonance	101 (Experimental)
IST + Plasmonic Resonator	29 (Experimental)
GaAs/AlGaAs nonlinear Mie metasurfaces	0.05 (Experimental)
IST + Mie Resonator (Not optimized !)	**3250 (14 micron spot size)**

Nonlinear Optics in Metasurfaces: How to Optimize?

Traditional nonlinear optics with crystals

$$I_{2\omega} \propto \frac{(\chi^2)^2}{n_\omega n_{2\omega}{}^2}$$

Nonlinear optics with metasurfaces



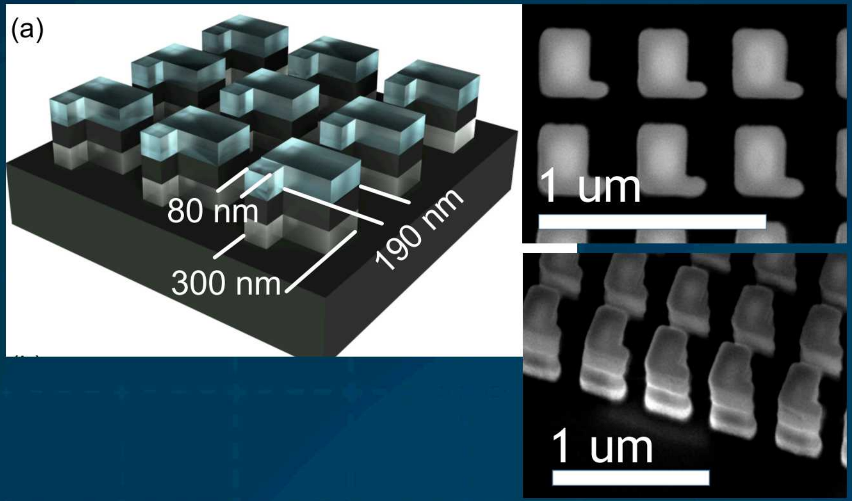
$$I_\omega \propto Q \; to \; Q^2 \propto n^{2-4}$$

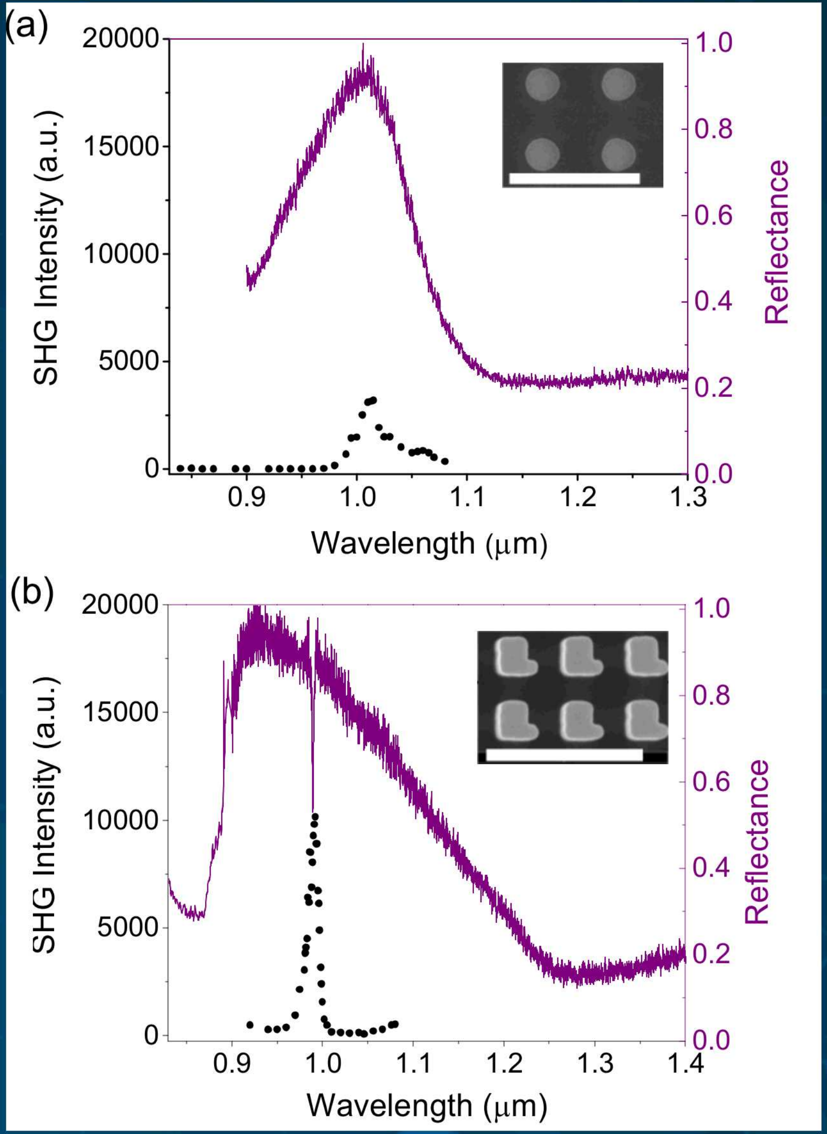
$$I_{2\omega} \propto I_\omega^2 \, (\chi^2)^2 \sim n^?$$

However with an additional resonance at 2ω it could be

$$I_\omega \propto Q(\omega)^? Q(2\omega)^? \propto n^{??}$$

Q: SHG from Broken Symmetry Metasurfaces
(or Bound States in the Continuum)



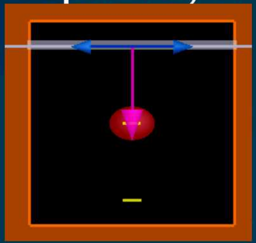


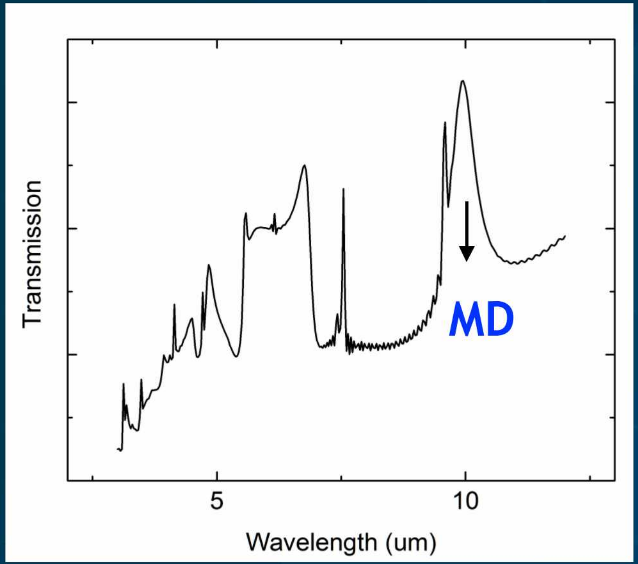
13-fold enhancement of SHG in Broken Symmetry metasurface compared with the nanodisk Mie metasurface

ACS Photonics 2018

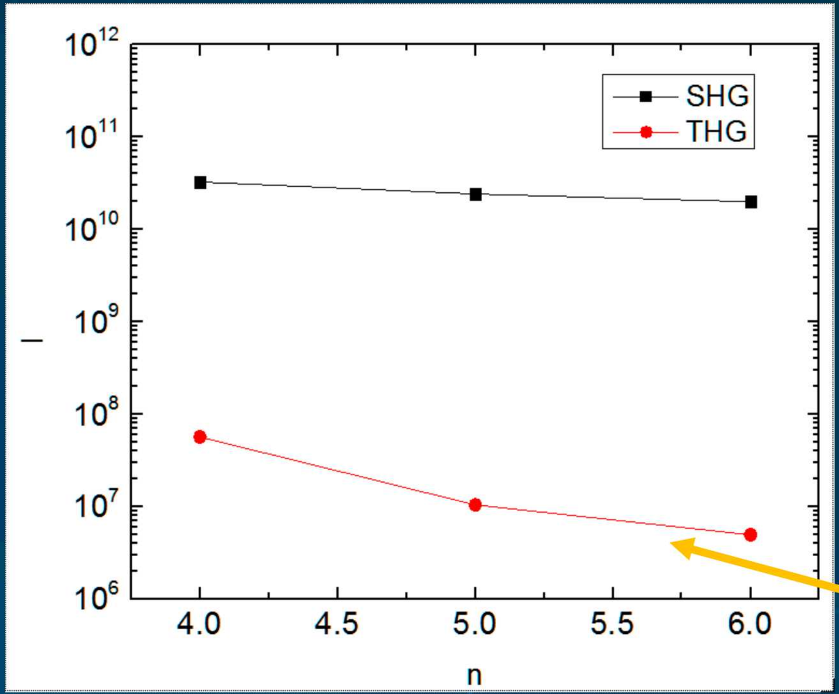
Dependence on n
Spherical Mie Resonator

Single sphere, n=5





$$I_{2\omega} \propto I_{\omega}^{2}\,(\chi^{2})^{2} \not\propto n^{?}$$



Cascaded

Index is not a big knob ☹ (at least for SHG and cascaded NLO)

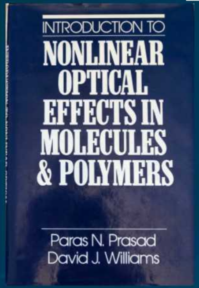
Nonlinear Optics with Metasurfaces: No Traditional Constraints

$$\begin{bmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{bmatrix} = 2\epsilon_0$$

1.9.3 Second Harmonic Generation Coefficients

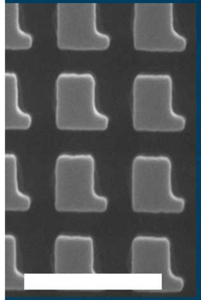
Crystal System—Cubic

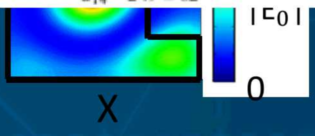
Cubic material	Symmetry class	d_{im} (pm/V)	Wavelength λ (μm)
$NaBrO_3$	23	$d_{14} = 0.19$	0.6943
$NaClO_3$	23	$d_{14} = 0.46$	0.6943
AlSb	-43m	$d_{14} = 49 \pm 36$	1.058
$Bi_4Ge_3O_{12}$	-43m	$d_{14} = 1.28$	1.064
CdTe	-43m	$d_{14} = 167.6 \pm 63$	10.6
		$d_{14} = 59.0 \pm 24$	28.0
CuBr	-43m	$d_{14} = 8.04 \pm 30\%$	10.6
		$d_{14} = -4.38 \pm 20\%$	1.318
		$d_{14} = -6.37 \pm 20\%$	1.064
		$d_{14} = -6.53 \pm 20\%$	0.946
CuCl	-43m	$d_{14} = 6.7 \pm 30\%$	10.6
		$d_{14} = -4.0 \pm 20\%$	1.318
		$d_{14} = -3.97 \pm 20\%$	1.064
		$d_{14} = -3.47 \pm 20\%$	0.946
CuI	-43m	$d_{14} = 8.04 \pm 30\%$	10.6
		$d_{14} = -5.47 \pm 20\%$	1.318
		$d_{14} = -6.08 \pm 20\%$	1.064
		$d_{14} = -6.04 \pm 20\%$	0.946
GaAs	-43m	$d_{14} = 134.1 \pm 41.9$	10.6
		$d_{14} = 209.5 \pm 13.3$	1.058
		$d_{14} = 256.5$	0.694
GaP	-43m	$d_{14} = 71.8 \pm 12.3$	1.058
		$d_{36} = 59.5 \pm 6.0$	1.318
		$d_{14} = +70.6$	3.39
GaSb	-43m	$d_{14} = +628 \pm 6.3$	10.6
InAs	-43m	$d_{14} = 364 \pm 47$	1.058
		$d_{14} = 249 \pm 62$	10.6



An additio… phase-matched… d_{33} is the large… further in Sect… …e to …since …ssed

In metasurfaces, all field components can contribu… nonlinearities





Nonlinear Optics with Dielectric Metasurfaces: the "new rules":

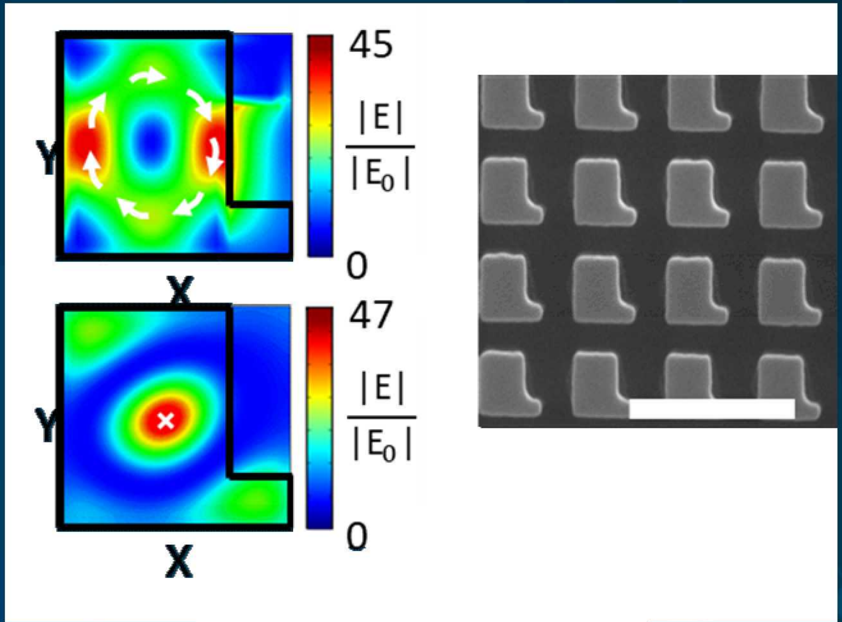
- Fundamental beam induces spatial distribution of *E(ω)* in resonator (usually a resonator mode)
- The vector components of *E(ω)* contract quadratically with the tensor components of $\chi^{(2)}$ to generate *P(2ω)*:

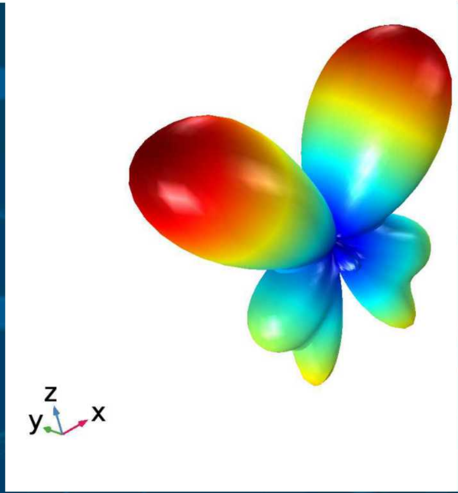
$$P_k(2\omega) = E_i\ E_j\chi^{(2)}_{ijk}$$

- *P(2ω)* excites a distribution of resonator modes at 2ω
- Angular distribution of second harmonic radiation dictated by radiation distribution of the excited modes

Rich physics!
Plenty of opportunity for tuning & optimizing desired behaviors.

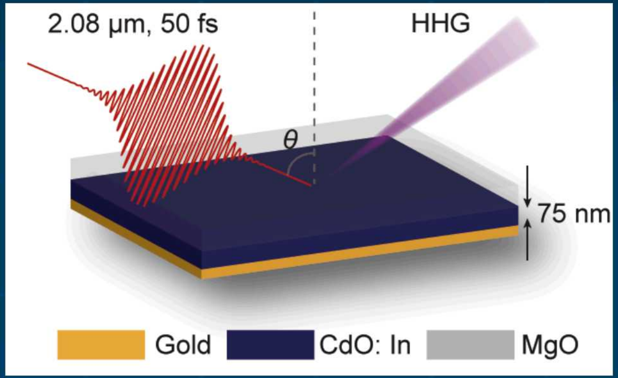
Field distributions





Second harmonic emission

High-harmonic generation in Epsilon Near Zero materials

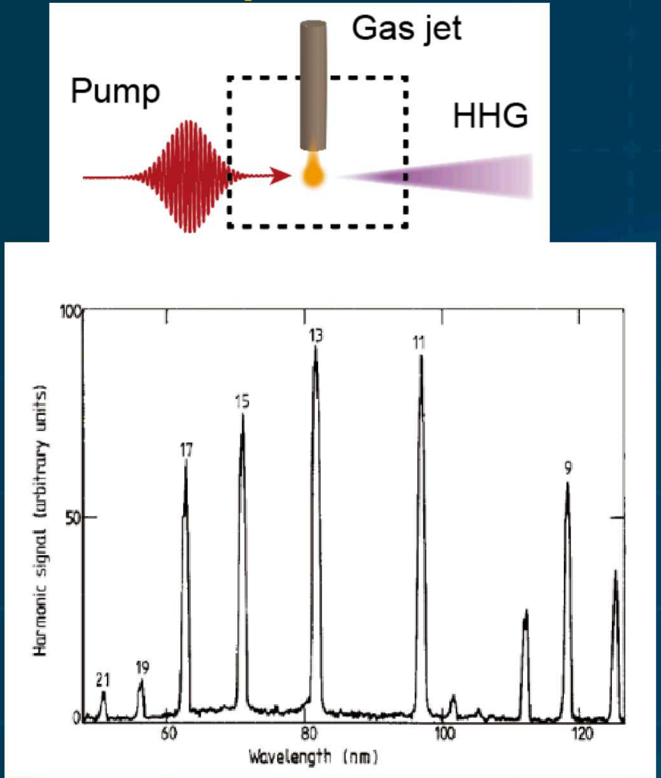


- Physics of ENZ & Berreman modes: Physical Review B **91**, 121408 (2015).
- fsec switching with CdO ENZ films: Nature Photonics 11, 390 (2017)
- Doped CdO as ENZ material: Nature Materials 14, 414 (2015)
- HHG: Nature Physics 2019

High Harmonic Generation

High-harmonics are used to generate attosecond pulses

Gas-phase HHG

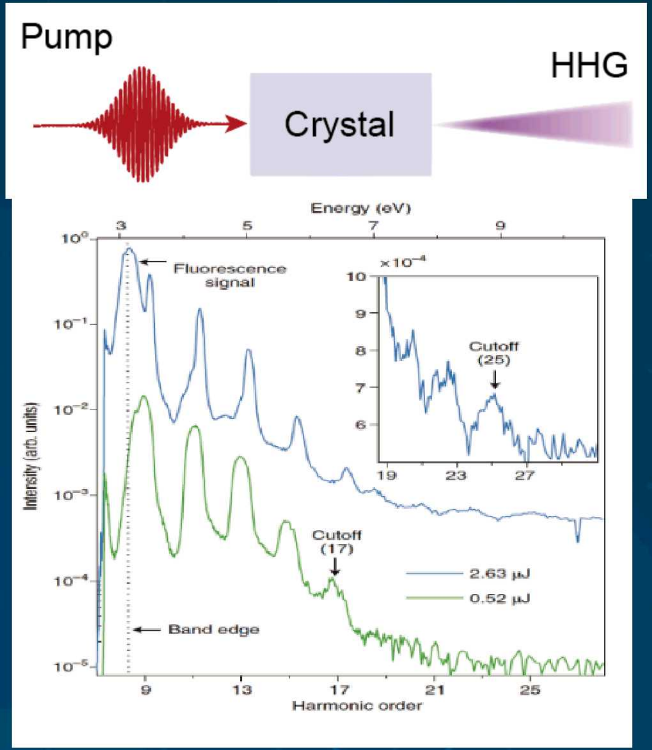


M. Ferray et al, *Journal of Physics B* (1988)

Pros:
- High damage threshold

Cons:
- Cumbersome setup

Solid-phase HHG



S. Ghimire et al, ***Nature Physics*** (2010)

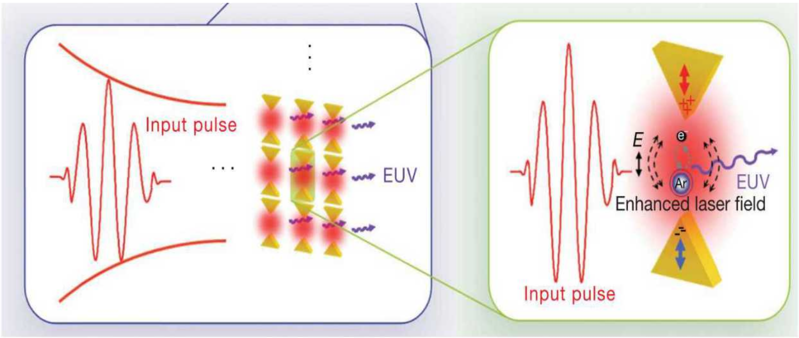
Pros:
- Compact setup

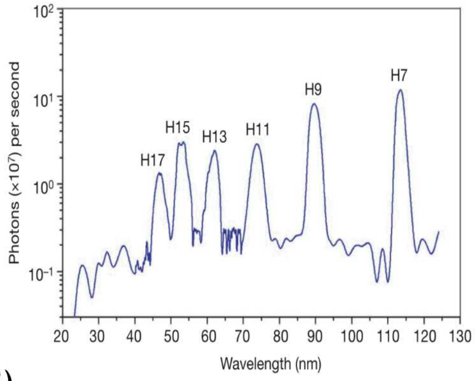
Cons:
- Low damage threshold
- Absorption above bandgap

See review by Ghimire & Reis, Nat. Phys. 2018

HHG enhanced by Plasmonics

Gas-phase HHG enhanced by plasmonics:

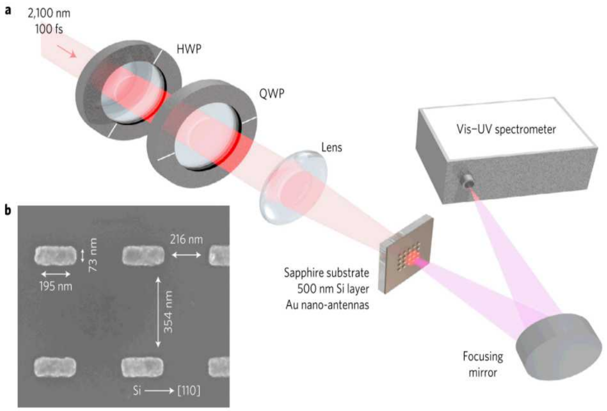


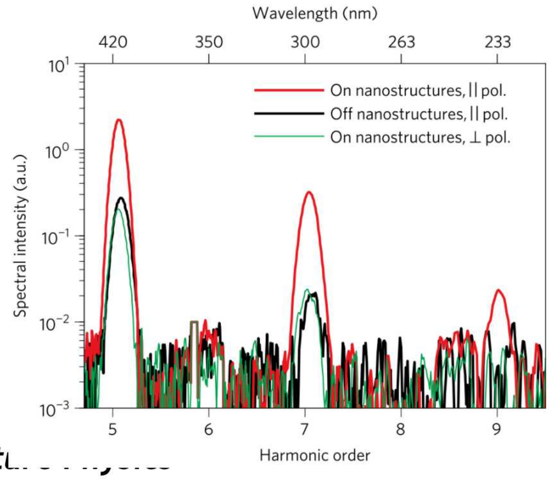


S. Kim et al, ***Nature*** (2008)

Problem:
Low damage threshold!

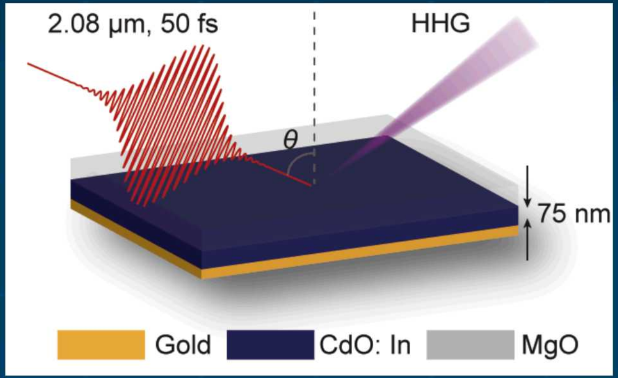
Solid-phase HHG enhanced by plasmonics:





G. Vampa et al, ***Natu***
(2017)

Epsilon Near Zero thin films might provide a new platform for HHG

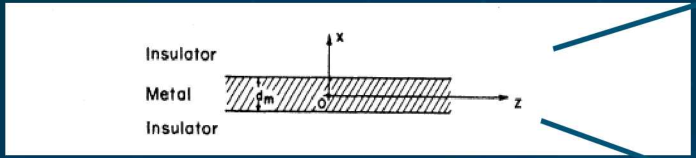


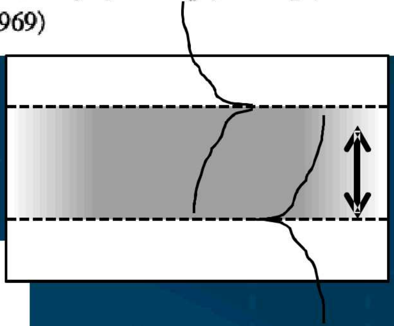
- Lower optical power density
- No patterning or nanostructuring
- No damage

PHYSICAL REVIEW VOLUME 182, NUMBER 2 10 JUNE 1969

Surface Plasmons in Thin Films

E. N. Economou*†
The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637
(Received 15 January 1969)





Skin depth

Econoumou, 1969
Burke et al, 1986
etc

"Thin" here: ~20nm for ~400nm plasma wavelength

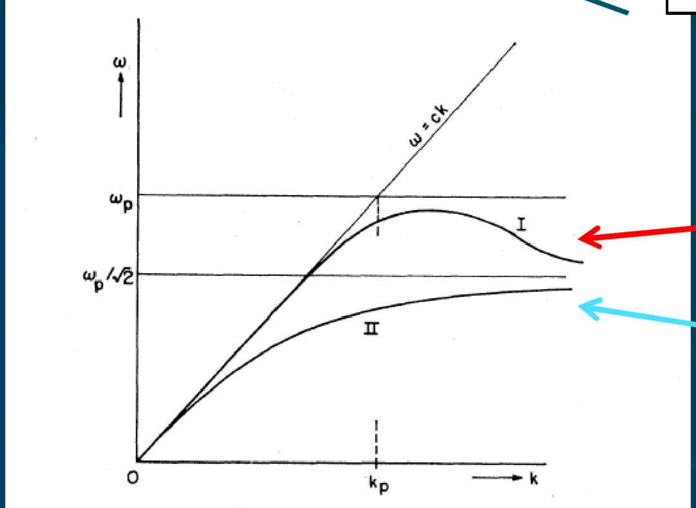


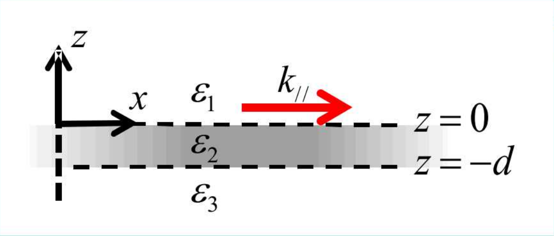
FIG. 4. Geometry and the dispersion relations for SPO of a metal film between two semi-infinite insulators ($\epsilon_m = 1 - \omega_p^2/\omega^2$, $\epsilon_i = 1$). The analytical expressions for $k \gg k_p$ or $k \ll k_p$ for the curves shown schematically here are (3.23b) and (3.18).

Long range surface plasmon

Short range surface plasmon

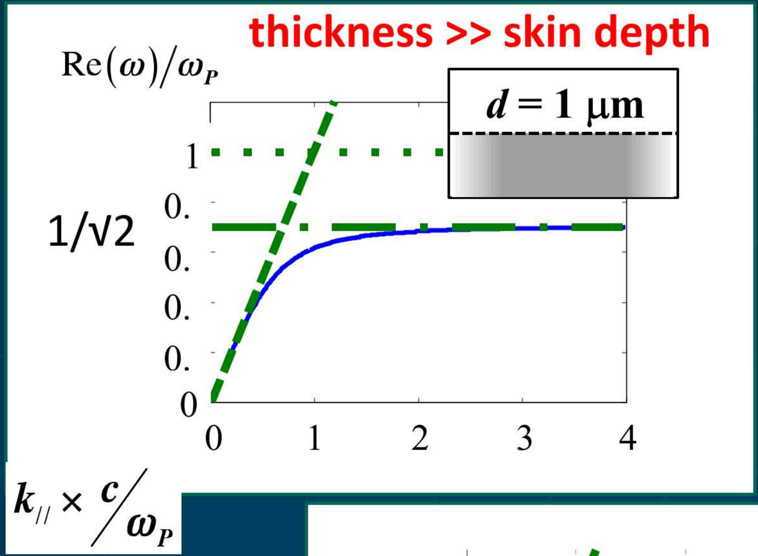
What happens when the layer becomes much thinner? (<< skin depth)

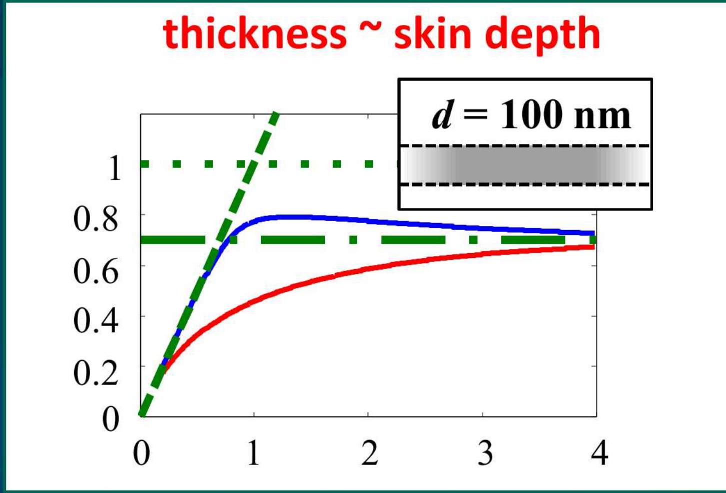
Film thickness dependence of surface plasmons and ENZ mode dispersion

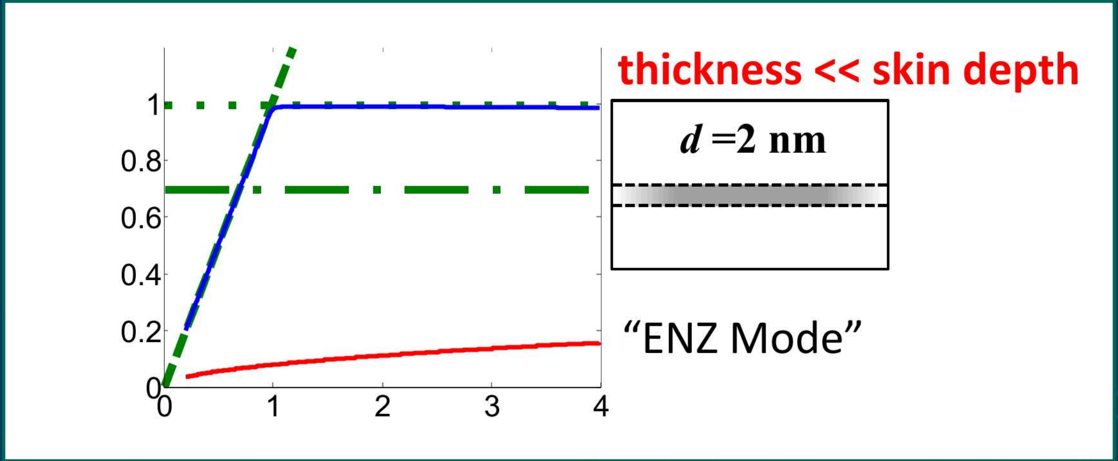


$$1+\frac{\varepsilon_1 k_{z3}}{\varepsilon_3 k_{z1}} = i\tan(k_{z2}d)\left(\frac{\varepsilon_2 k_{z3}}{\varepsilon_3 k_{z2}}+\frac{\varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1}}\right)$$

$k_{zi}^2=\varepsilon_i\frac{\omega^2}{c^2}-k_{//}^2$ is the longitudinal wavenumber in medium i

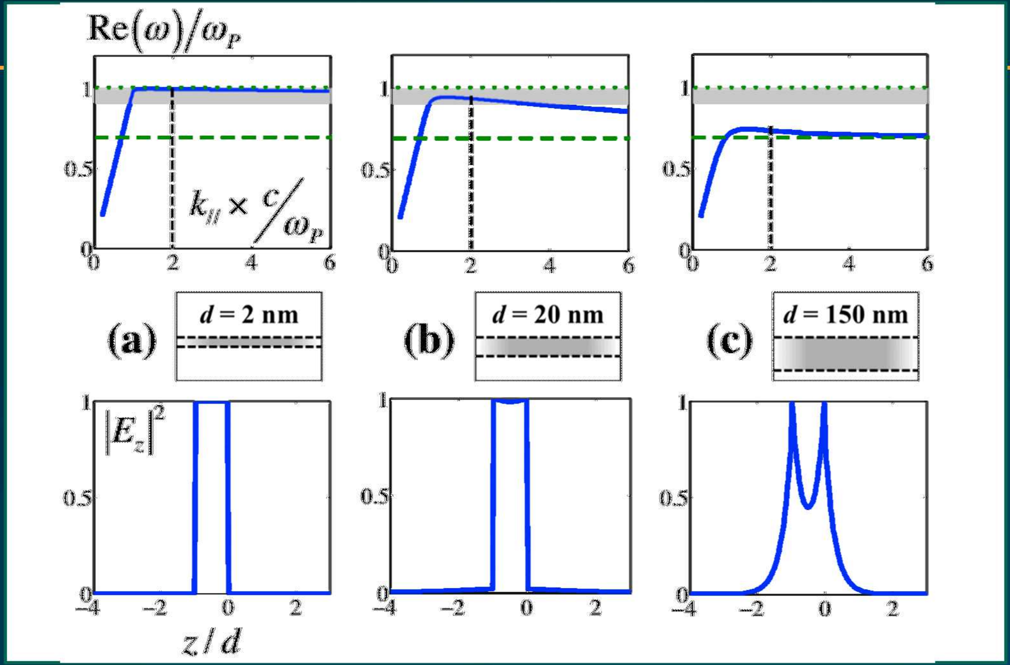






1. Vassant, Marquier, Greffet et al., Phys. Rev. Lett. **109**, 237401 (2012)
2. Vassant, Marquier, Greffet et al., Opt. Express **20**, 23971 (2012)
3. Campione, Brener, Marquier, Phys. Rev. B Rapid Commun. **91**, 121408 (2015)

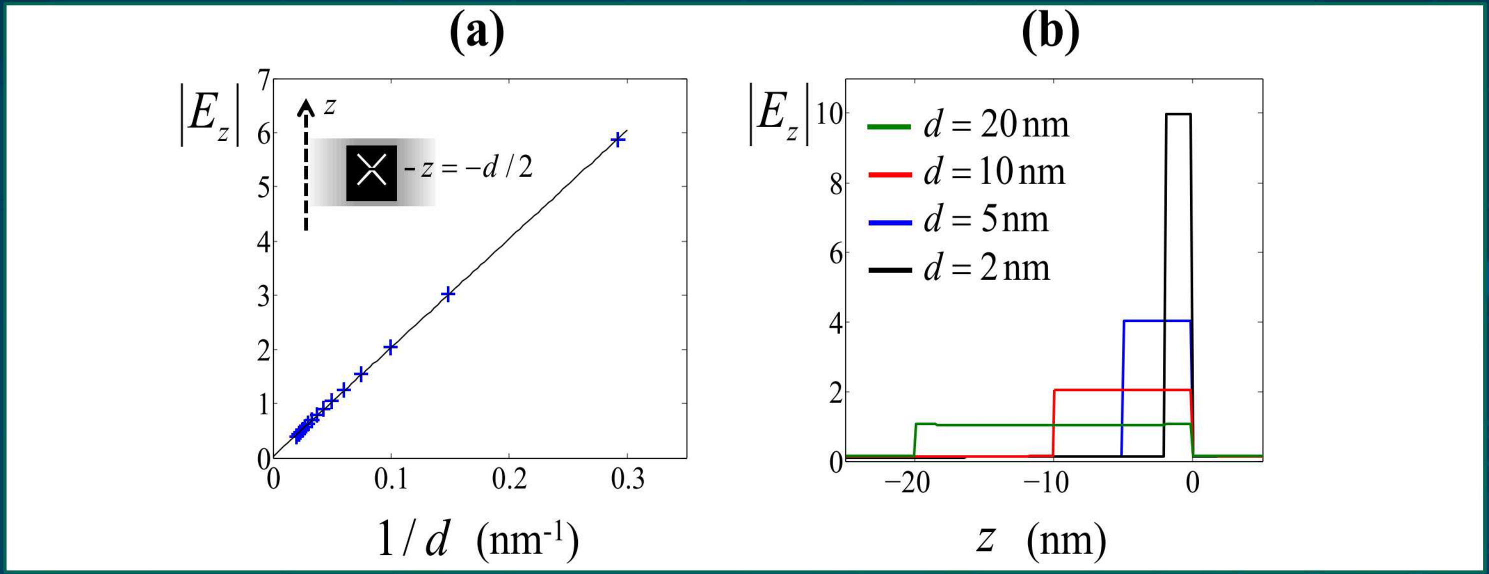
ENZ Modes: E_z is Constant and Large



Phys. Rev. B Rapid Commun. **91**, 121408 (2015)

$$E_z \propto \frac{1}{d}$$

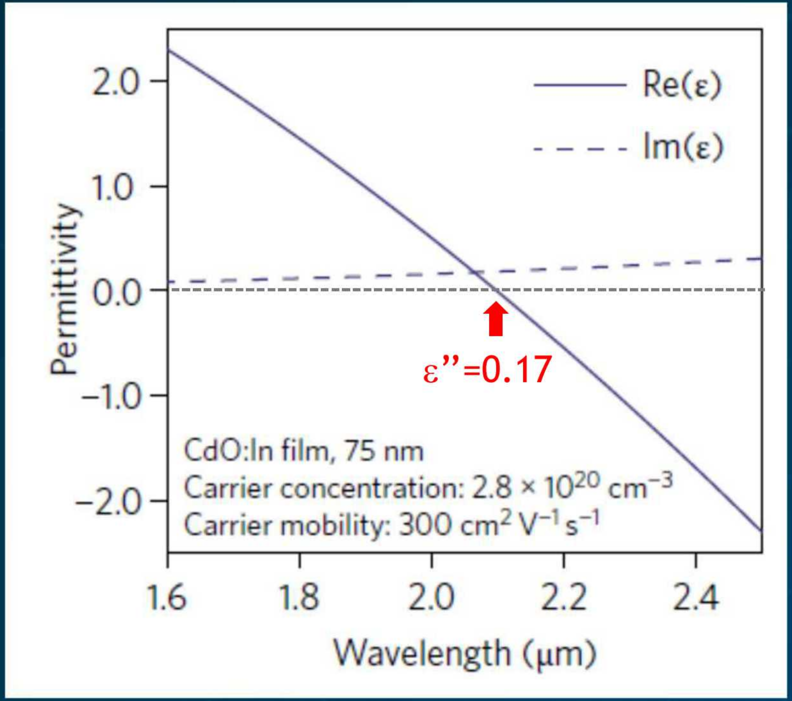
"Thin" is better
But not too thin …..



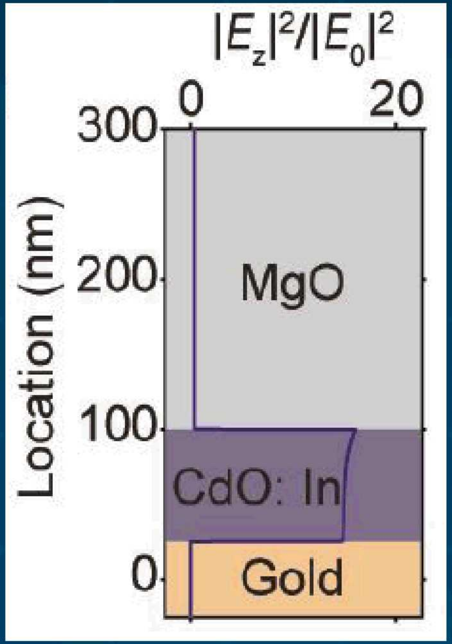
High Quality ENZ Material: High Mobility CdO

Low Loss

Y:CdO has more than 10x higher mobility than ITO

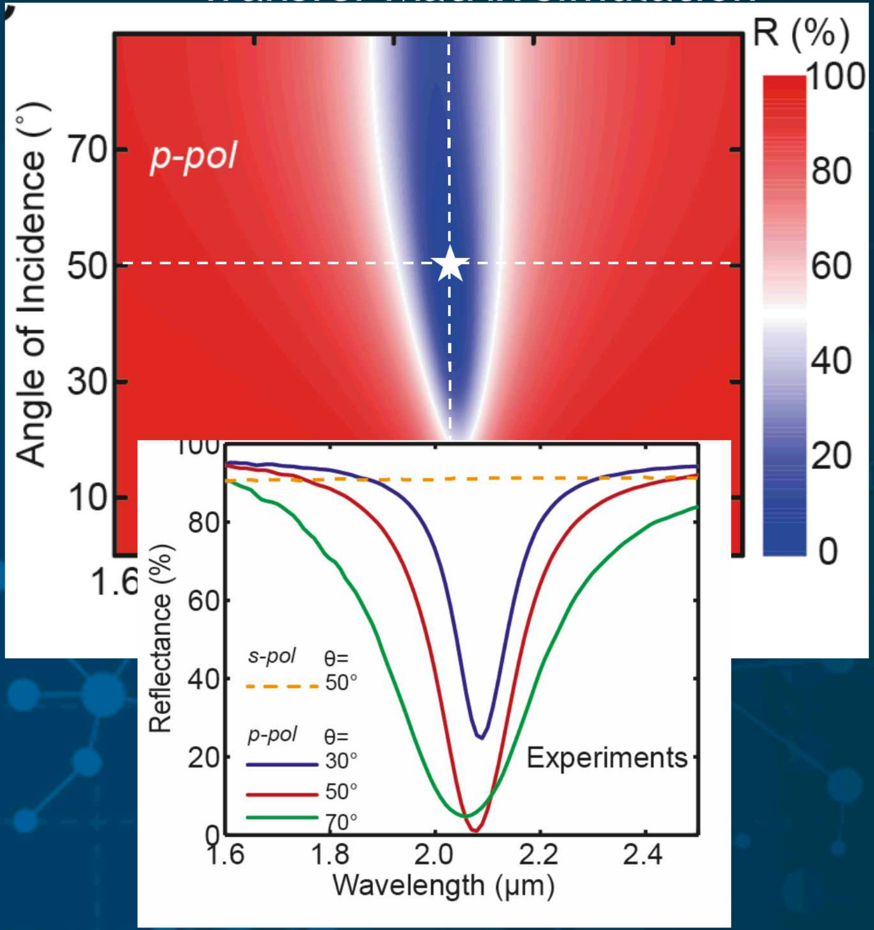


Large field enhancement

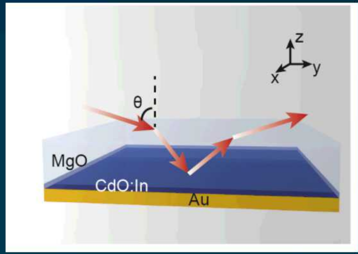


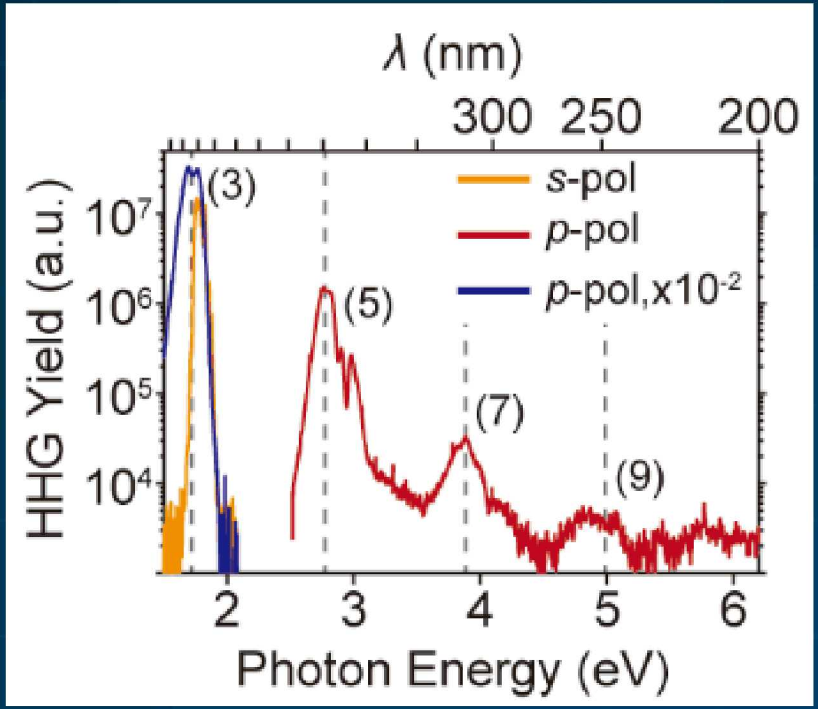
Perfect absorber without prism

Transfer Matrix simulation

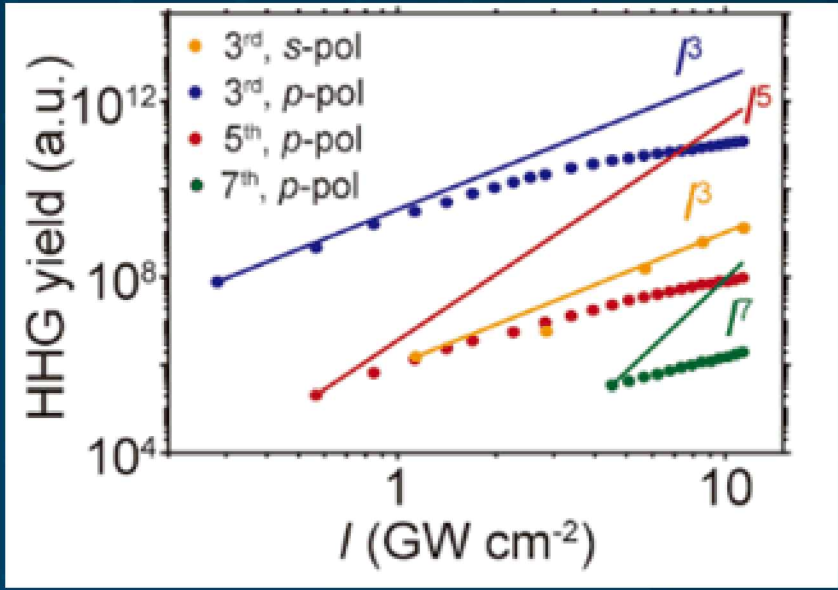


HHG Spectrum





HHG from 3rd to 9th order

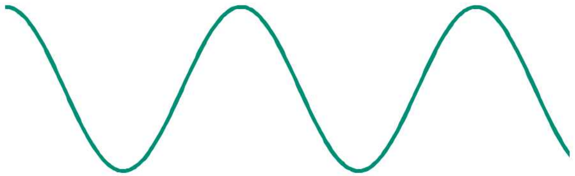


Non-perturbative scaling

10 GW/cm^2 pump intensity (**TW/cm^2** required for typical gas or solid phase HHG)!

Source of Nonlinearity

Sinusoidal field driven free electron



$$\frac{dp}{dt} = -\,eE_0 \sin\omega t$$

$$j(t) = -e\frac{\partial \varepsilon}{\partial p} = -\frac{eE_0}{m^*\omega}\cos\omega t$$

Since $m^*(\varepsilon) = \frac{\hbar^2}{\frac{\partial^2 \varepsilon(k)}{\partial k^2}}$

For parabolic band,

m^* is constant since $\frac{\partial^2 \varepsilon(k)}{\partial k^2}$ = constant

$j(t)$: **no higher harmonic**

Sinusoidal-field driven electron in a non-parabolic potential

$$j(t) = -e\frac{\partial \varepsilon}{\partial p} = -\frac{eE_0}{m^*\omega}\cos\omega t$$

For **non-parabolic** dispersion $\frac{p^2}{2m^*} = \varepsilon + \frac{\varepsilon^2}{\varepsilon_g}$

ε_g is non-parabolicity parameter

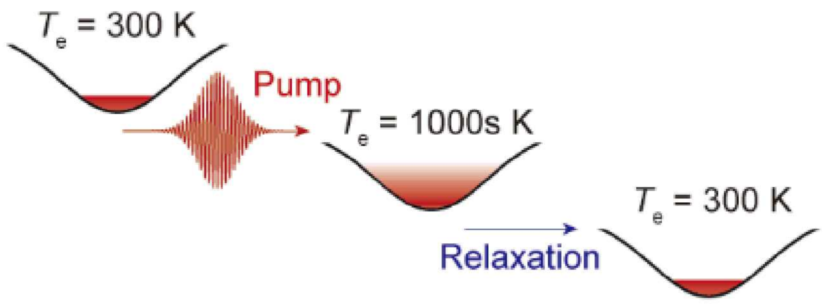
$$\Rightarrow \begin{cases} \varepsilon = -\frac{\varepsilon_g}{2} + \sqrt{\frac{\varepsilon_g^2}{4} + \frac{\varepsilon_g p^2}{2m^*}} \\ \frac{\partial \varepsilon}{\partial p} = \frac{1}{\left(1 + \frac{2\varepsilon}{\varepsilon_g}\right)}\frac{p}{m^*} \end{cases}$$

$$j(t) = -\frac{e^2 E_0 \cos\omega t}{m^*\omega\left(1 + \frac{2e^2 E_0^2 cos^2\omega t}{m^*\omega^2 \varepsilon_g}\right)}$$

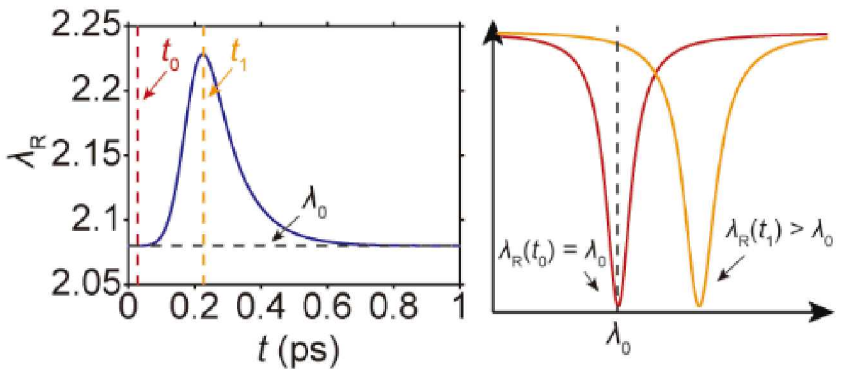
P. Guo, et. al., Nat Photon (2016)

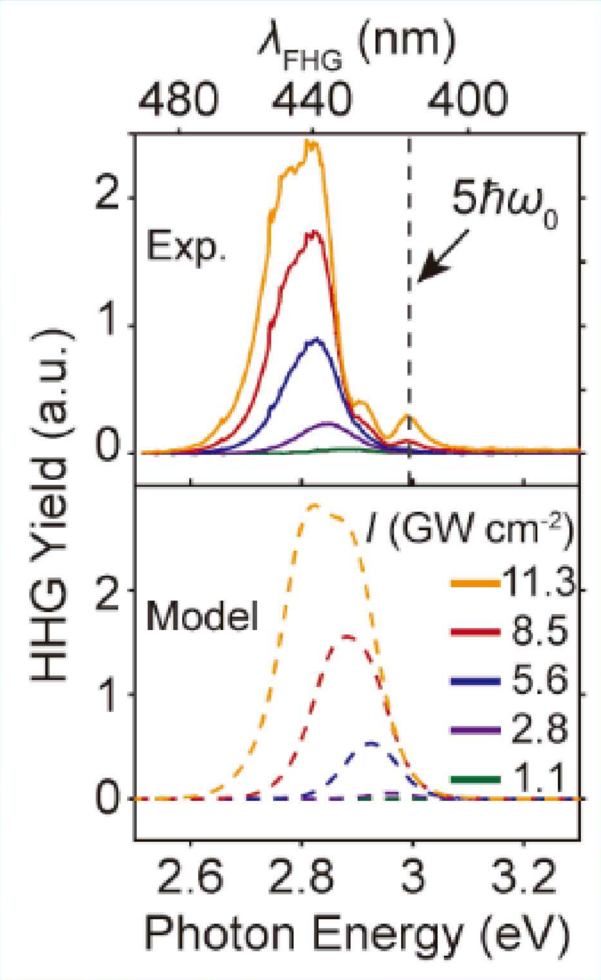
HHG Spectral Shift and Broadening

Hot electron dynamics in CdO:



Time-varying resonant wavelength in CdO:



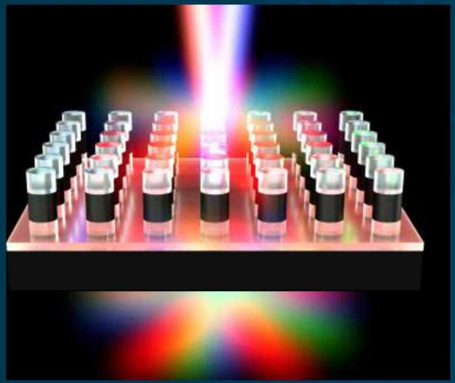


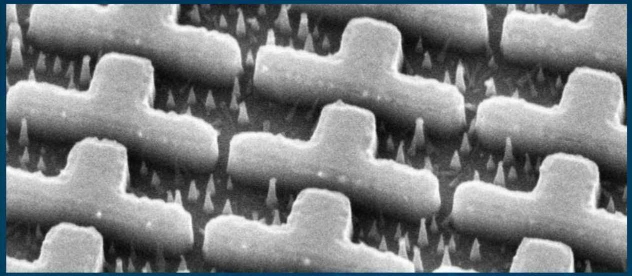
Maybe this broadening can be used to recompress pulses

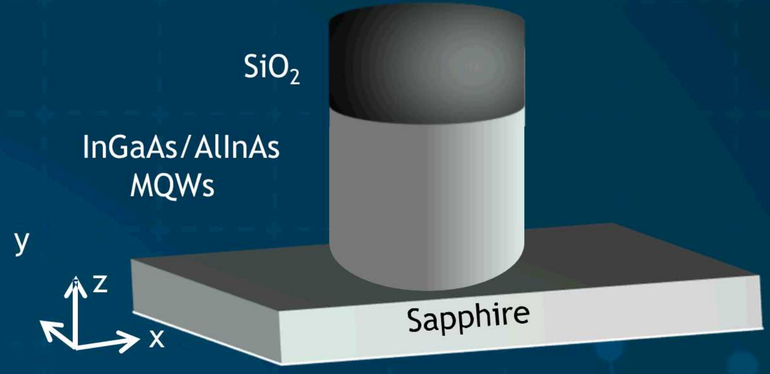
Nature Physics 2019

Summary

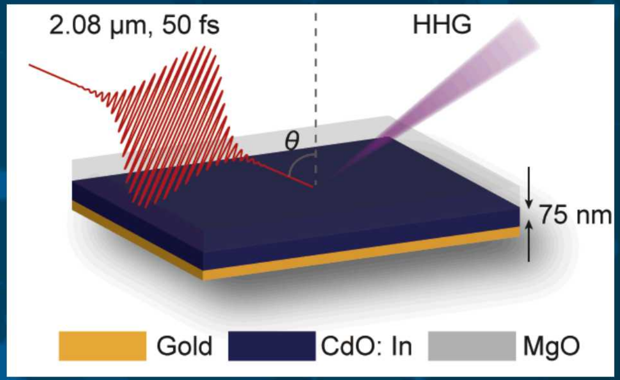
Free of phase matching constrains, nonlinear metasurfaces offer a new playground for nonlinear optics: nonlinear mixing, HHG, etc.







ENZ materials provide a new path for High-harmonic generation with lower powers and interesting physics.



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UT Austin: Nish Nookala, Misha Belkin

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JP Maria: *NC State*
Shambhu Ghimire, Jian Lu: *SLAC*
Alejandro Manjavacas: *UNM*

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