

All-Dielectric Metasurfaces and Waveguided Systems : From Nonlinear Optics to All-Optical Processing

Raktim Sarma¹, Jiaming Xu², Domenico de Ceglia³, Luca Carletti³, Salvatore Campione¹, John Klem¹, Michael Sinclair¹, Mikhail Belkin², Igal Brener¹

¹Sandia National Laboratories, Albuquerque, NM, USA

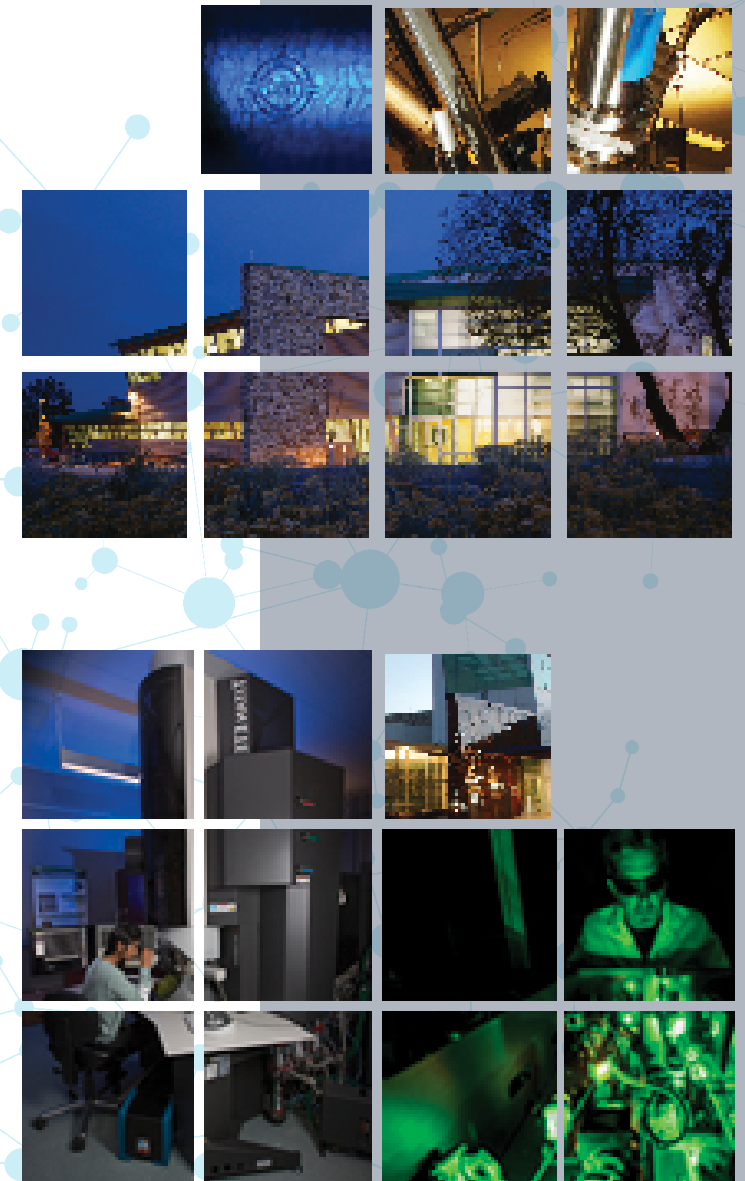
²University of Texas at Austin, Austin, TX, USA

³University of Padova, Padua, Italy



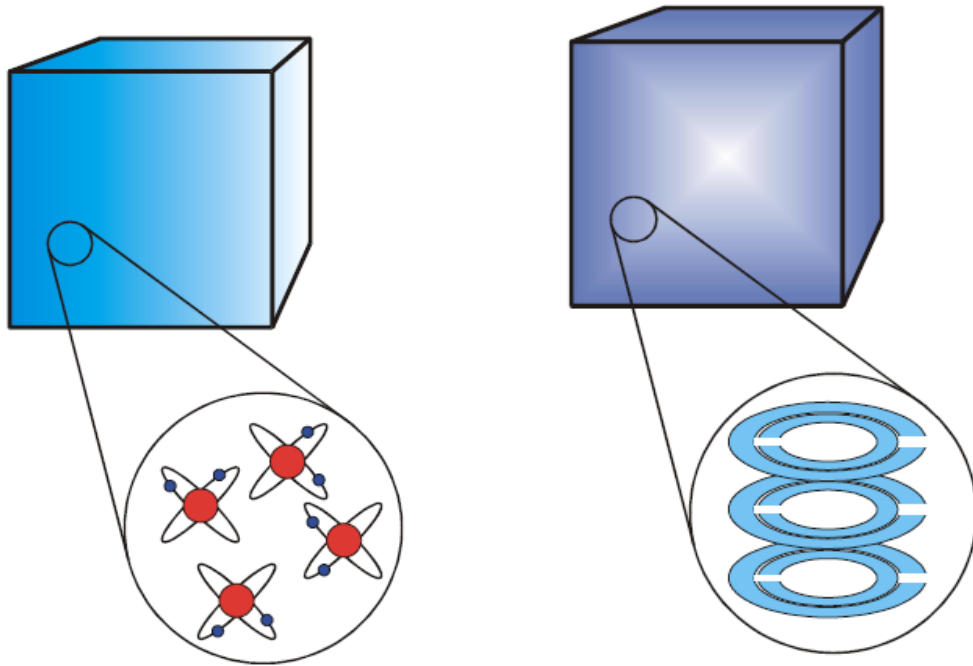
This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering and performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. This presentation describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

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

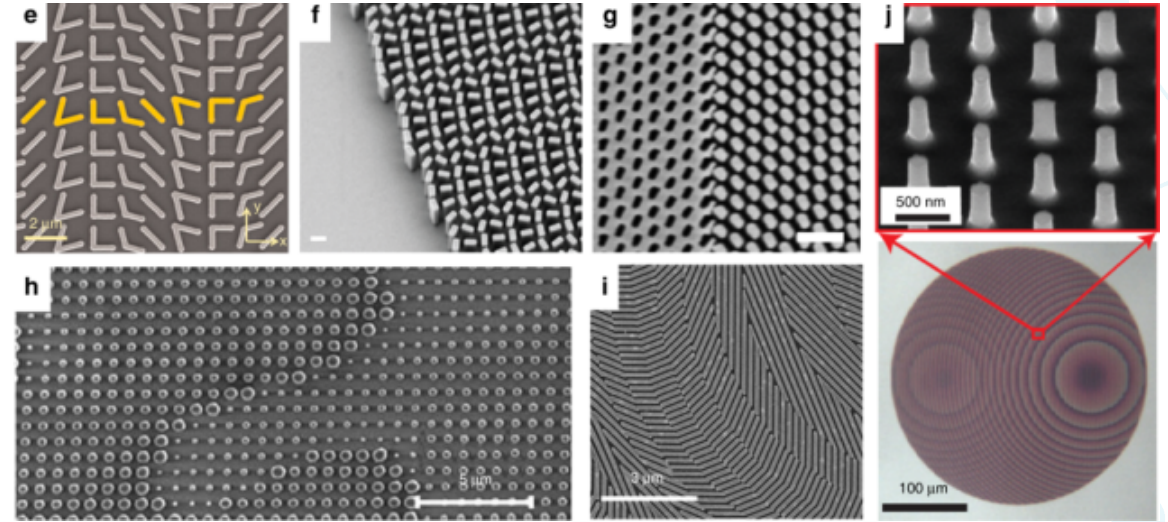


Metamaterials and Metasurfaces

Man-made “atoms” : Metamaterials



Metasurfaces



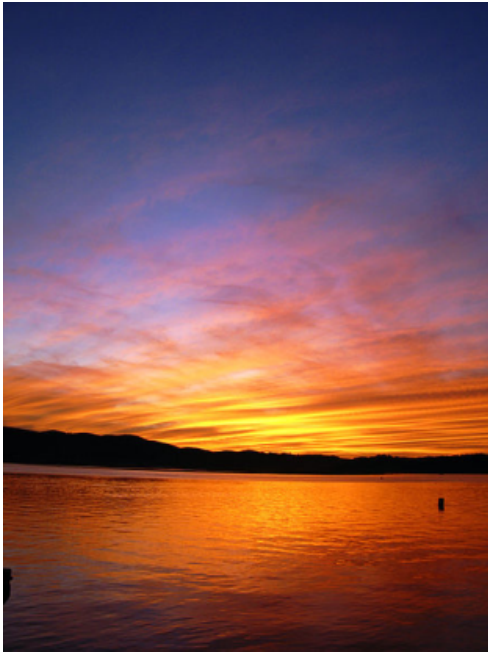
Ref. : Neshev & Aharonovich, Light : Science & Applications 7 (58), 2018.

In metamaterials, optical properties are determined by configuration and properties of meta-atoms.

Metasurfaces are planar (2D) equivalents of metamaterials.

The All-Dielectric Approach: Mie modes

Dielectric particles much smaller than wavelength λ/n

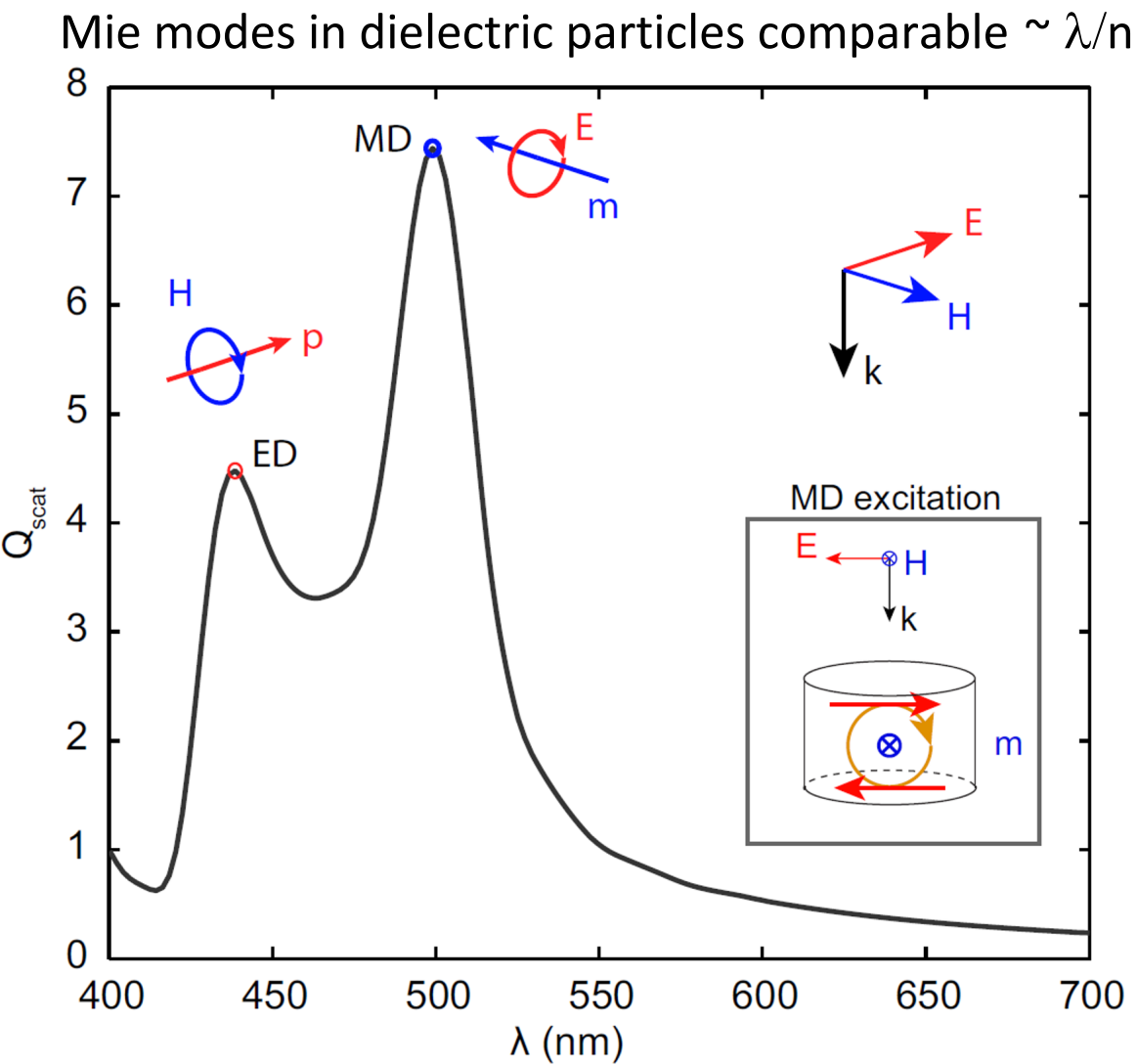


Wikipedia

Rayleigh $\ll \lambda/n$

For dielectric particles, the polarizabilities of the electric and magnetic dipole resonances are comparable at optical frequencies

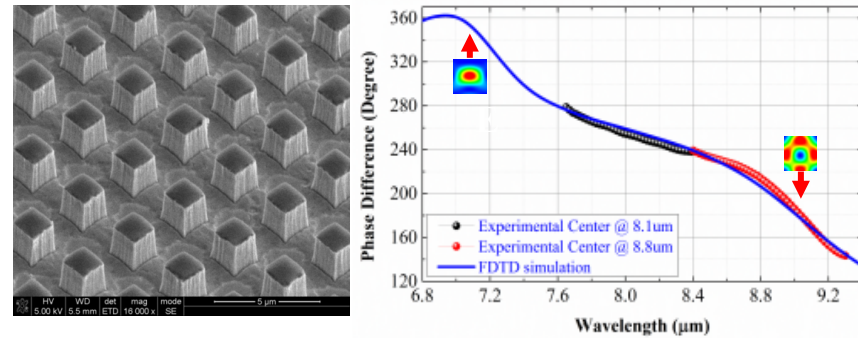
This is not the case for metallic resonators at optical frequencies because of metal losses



Ref. : Optics Express 21, 26285 (2013)

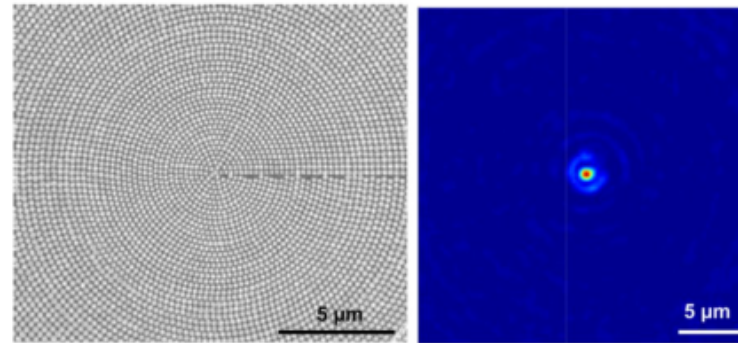
Applications of Mie Modes in Dielectric Metasurfaces

Magnetic Response



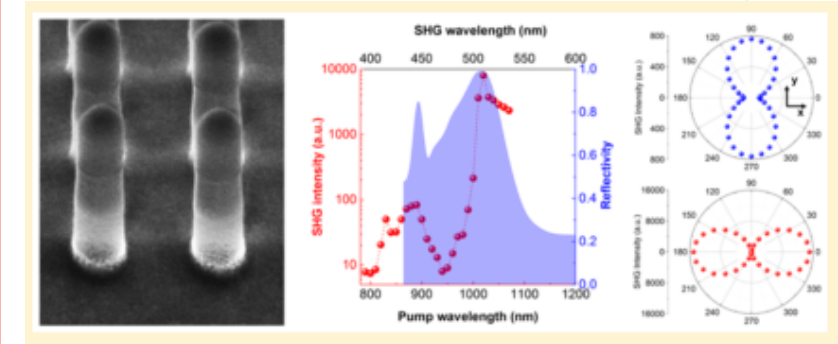
PRL. 108, 097402 (2012)

Tailoring Linear Transmission

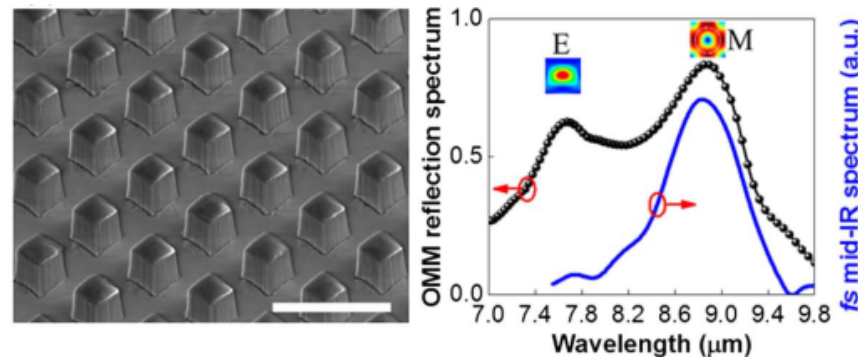


Argonne National Labs

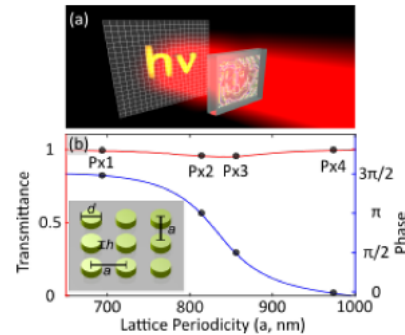
Nonlinear Optics



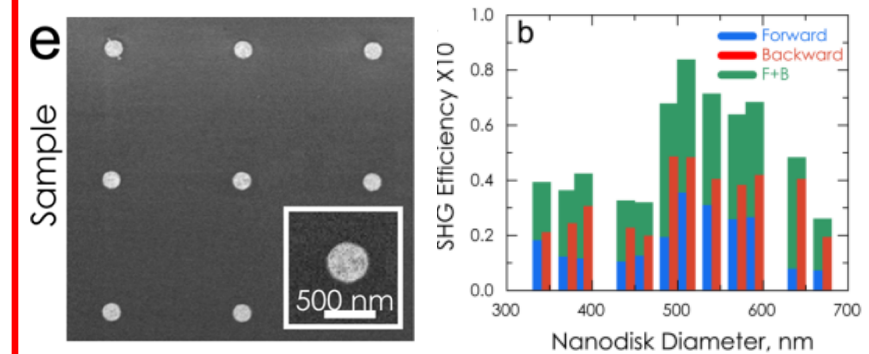
Nano Lett. 16, 5426 (2016)



Optica 1, 250 (2014)



ACS Photonics 3, 514 (2016)

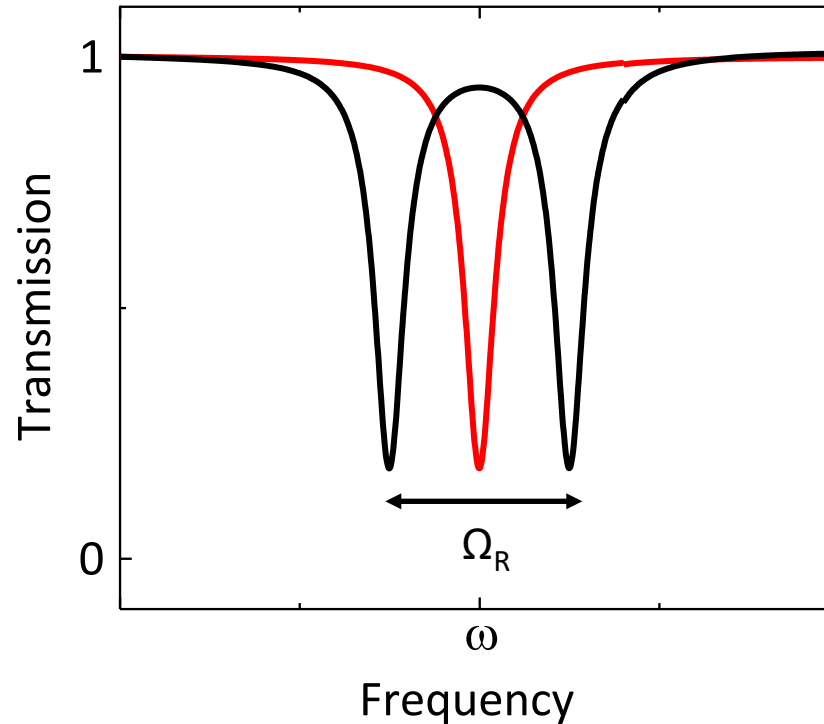
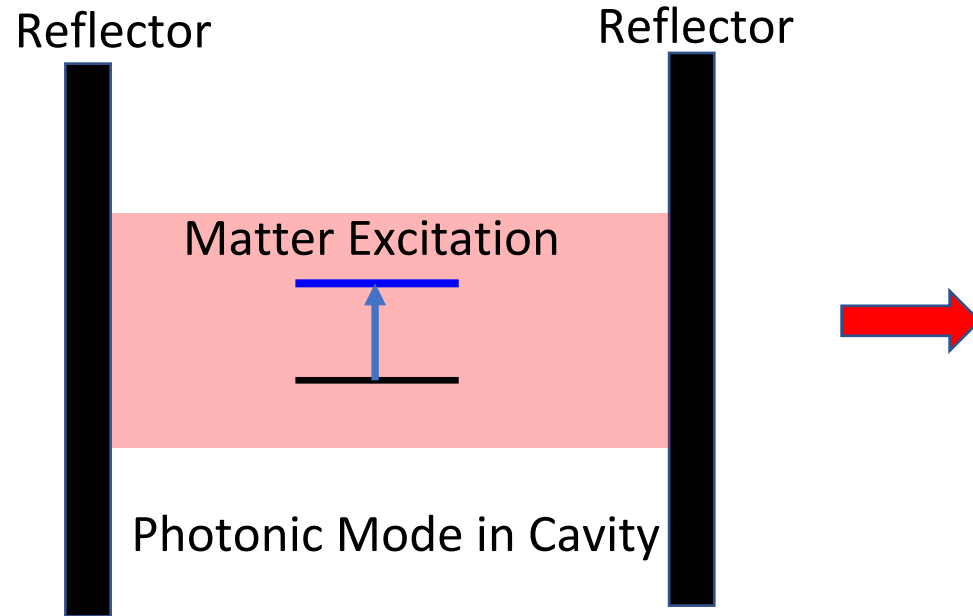


Nano Lett. 16, 7191 (2016)

Advantages of Nonlinear Mie Metasurfaces :

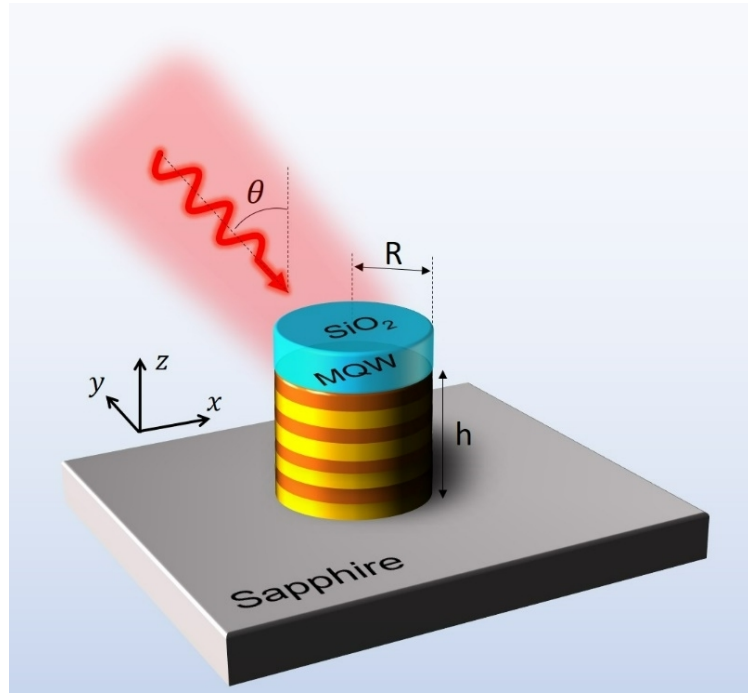
1. Ultrathin (relaxed phase matching)
2. Low loss and high damage thresholds
3. Large mode volume (enhanced light-matter interaction)
4. Ease of fabrication

Strong Light Matter Interaction : Polaritons and Rabi Splitting



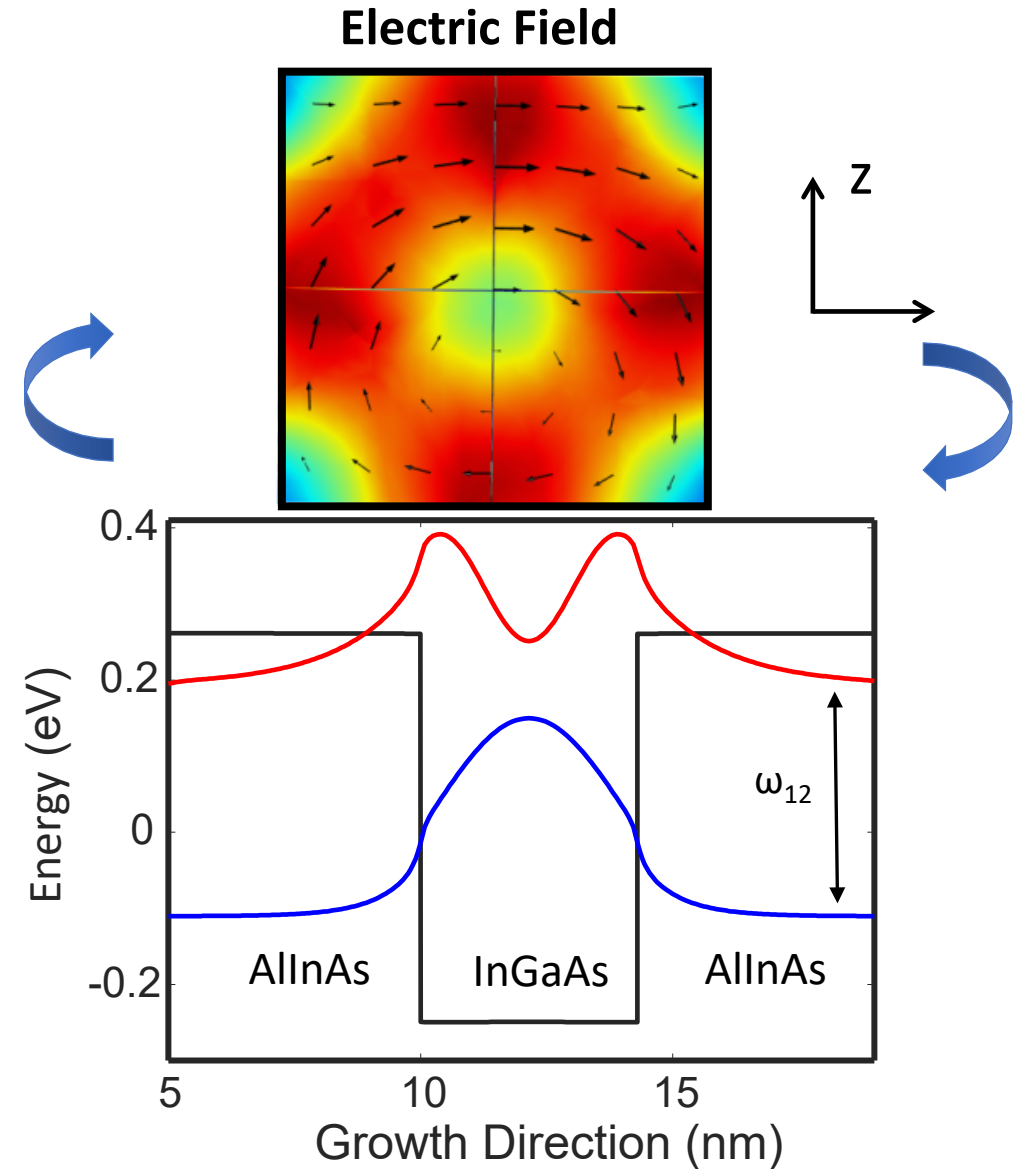
- In strong coupling, light –matter coupling strength > Losses of the system
- Formation of new hybrid states (polaritons) which are superposition of photonic mode and matter excitation and separated by frequency gap Ω_R , called Rabi splitting.

An Intersubband Polaritonic Metasurface : Light-Matter Coupling

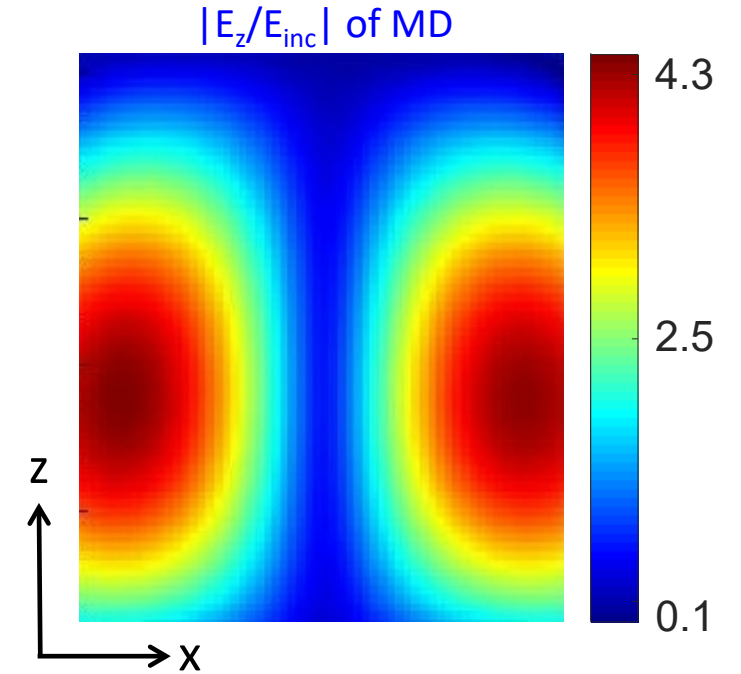
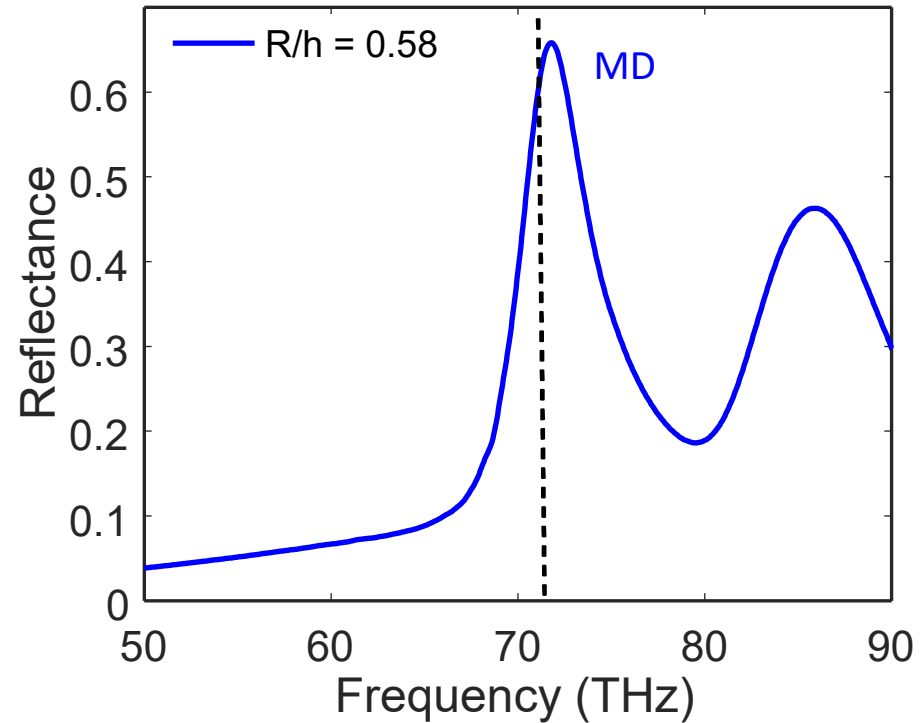
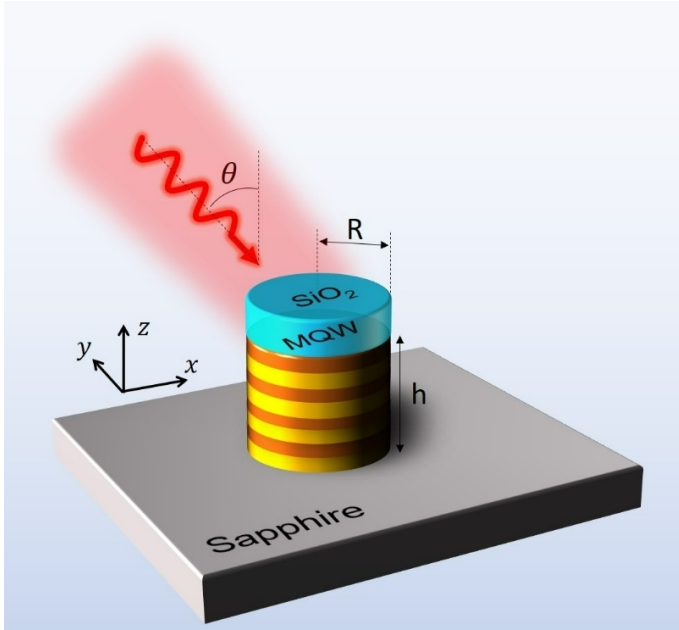


Ω_R depends on :

- **Matter** : Transition dipole moment of intersubband transitions and number of electrons ($N_e = N_d \times N_{QW}$) inside the cavity.
- **Light** : Distribution and enhancement of the *z-directed* electric field components of the photonic mode

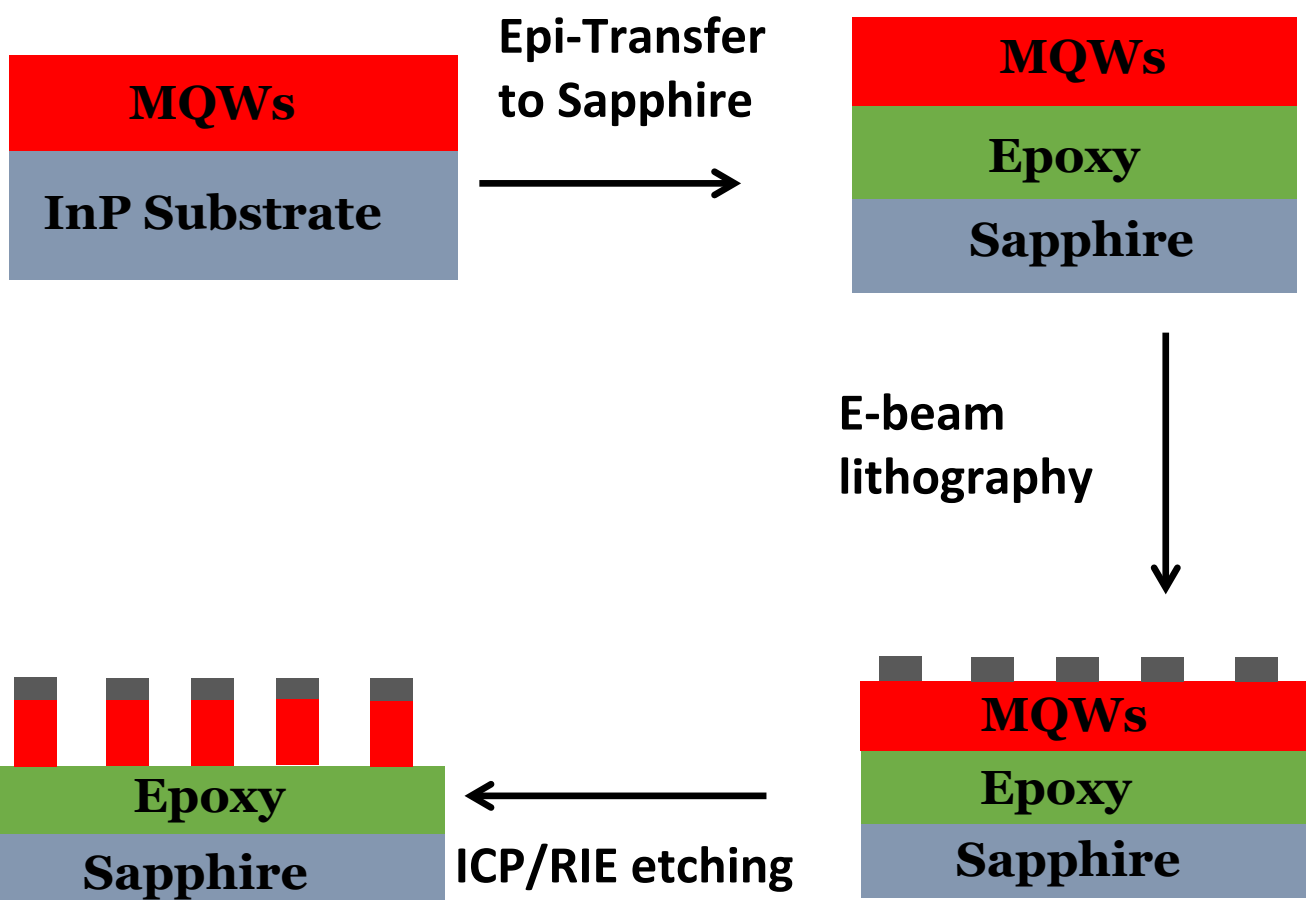


Coupling of Magnetic Dipole Mode to Intersubband Transitions

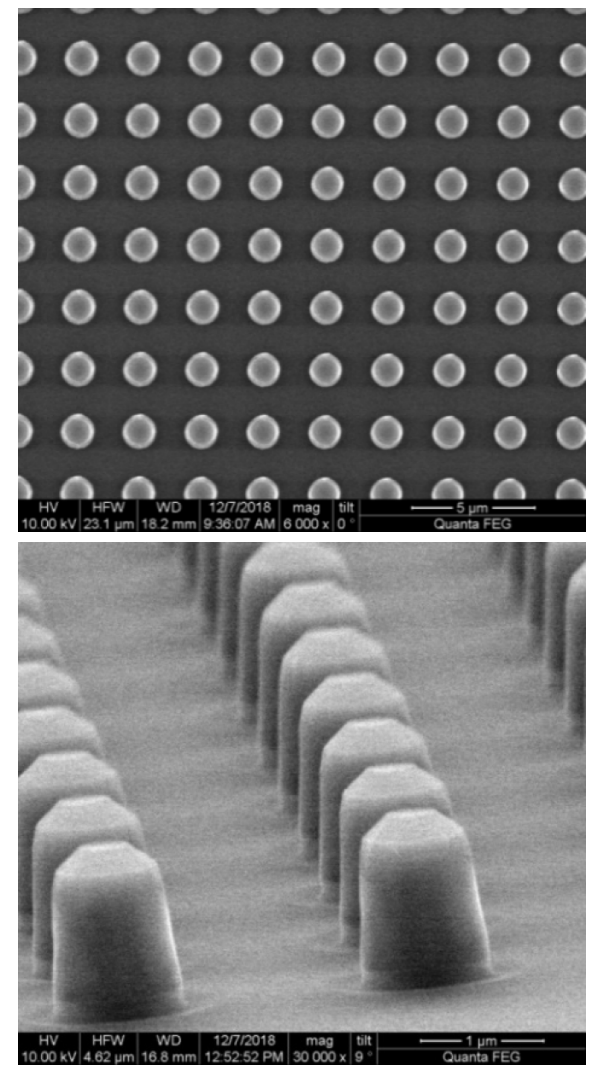


- R/h is tuned to spectrally align the MD mode to the ISB transition
- MD mode has strong z electric field components, allows for normal incidence, and smallest size of the resonator.

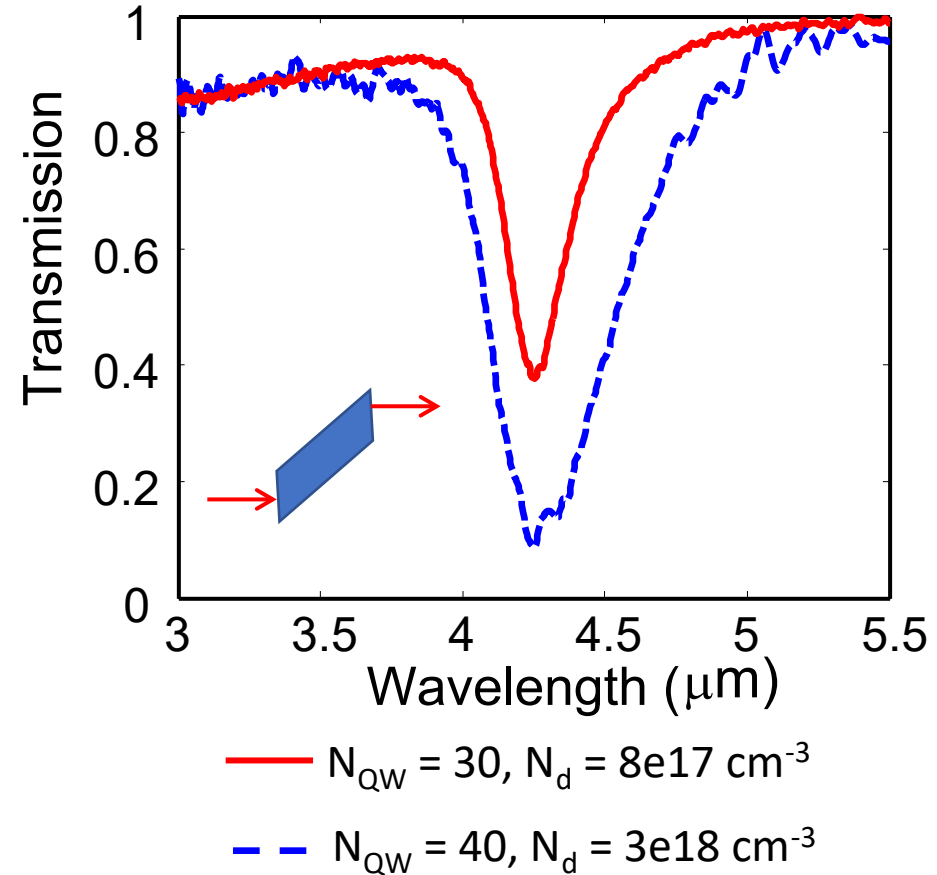
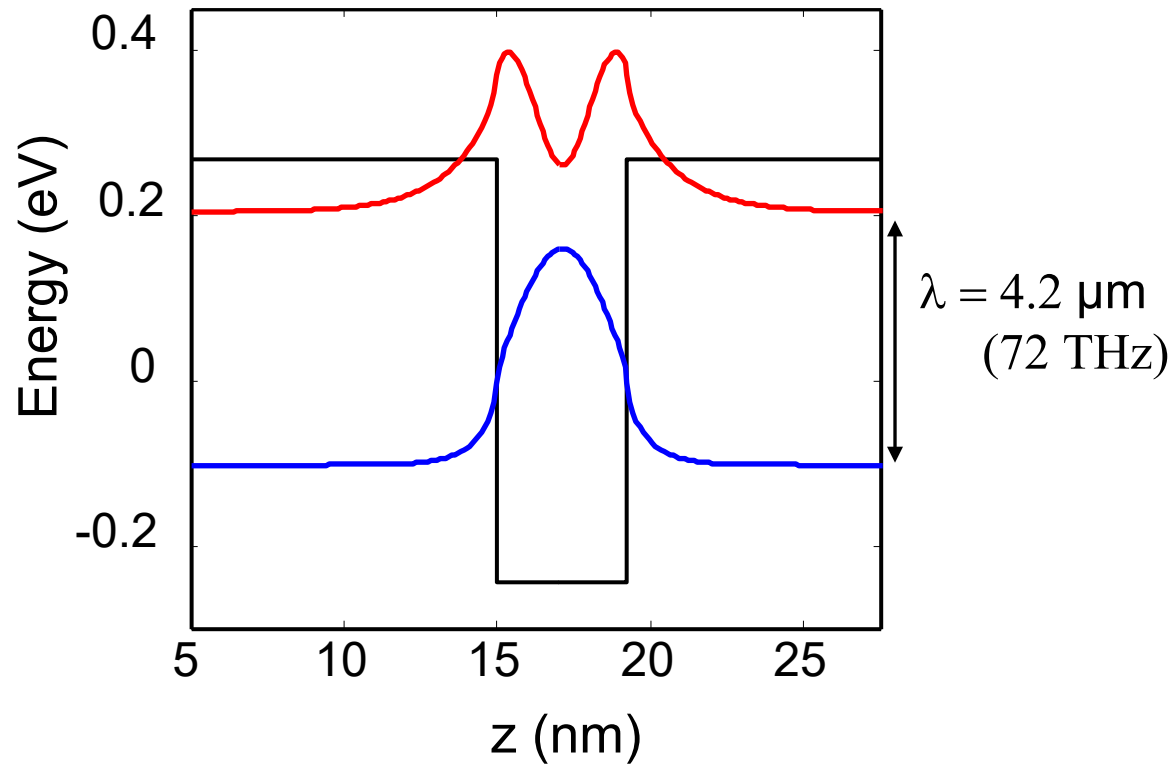
Fabrication of a Polaritonic Metasurface



Scanning Electron Micrograph



Controlling Light-Matter Interaction by Engineering Heterostructure

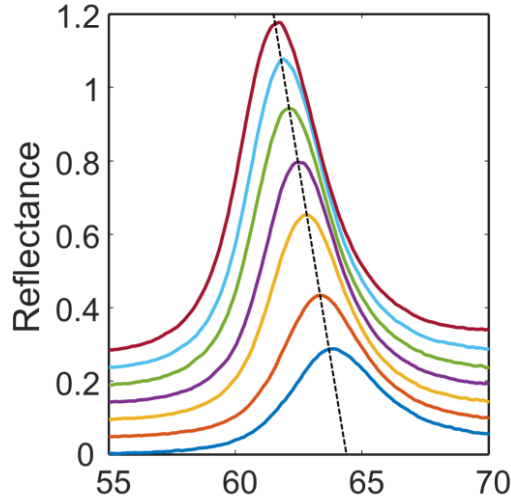


- The strength of light-matter interaction is proportional to number of electrons ($N_{\text{e}} = N_{\text{d}} \times N_{\text{QW}}$) in the cavity.
- We modify the number of electrons by modifying the doping and number of the quantum wells.

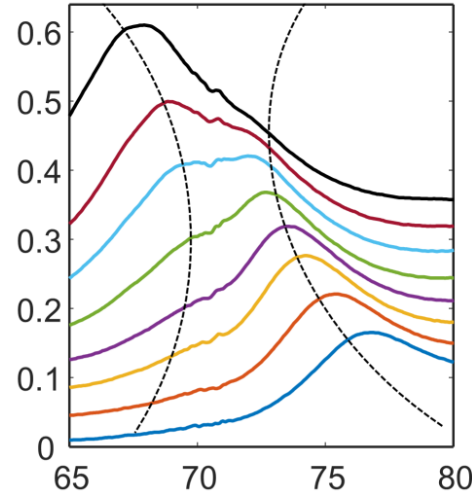
Controlling Light-Matter Interaction by Engineering Heterostructure

Experiment

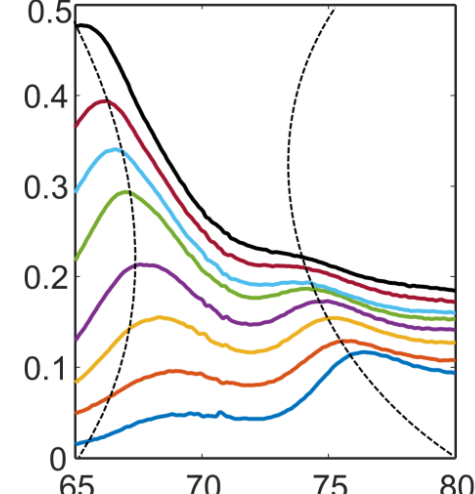
No coupling : Linear scaling



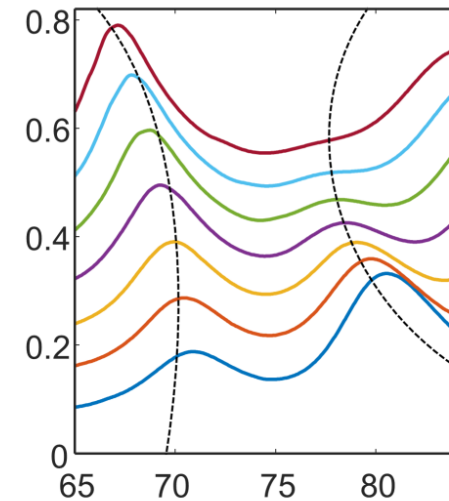
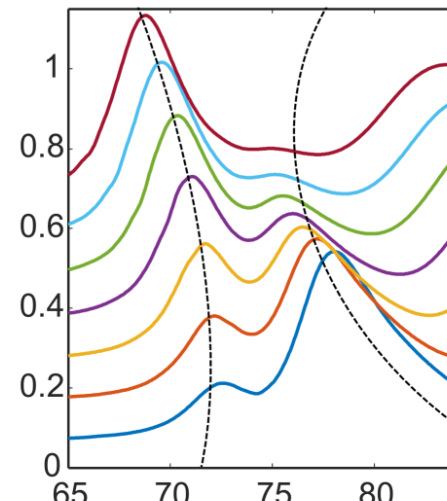
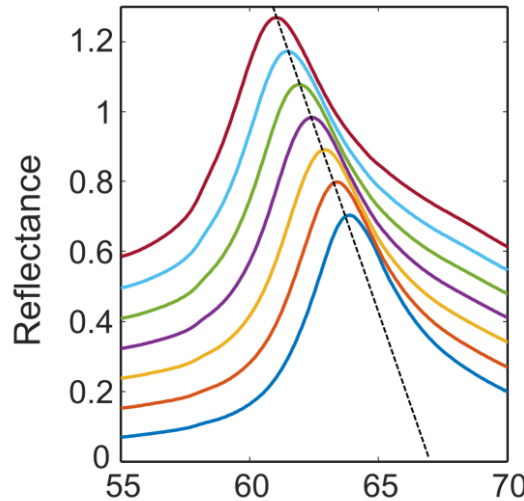
$\Omega_{\text{rabi}}=5\%$



$\Omega_{\text{rabi}}=10\%$



Simulation

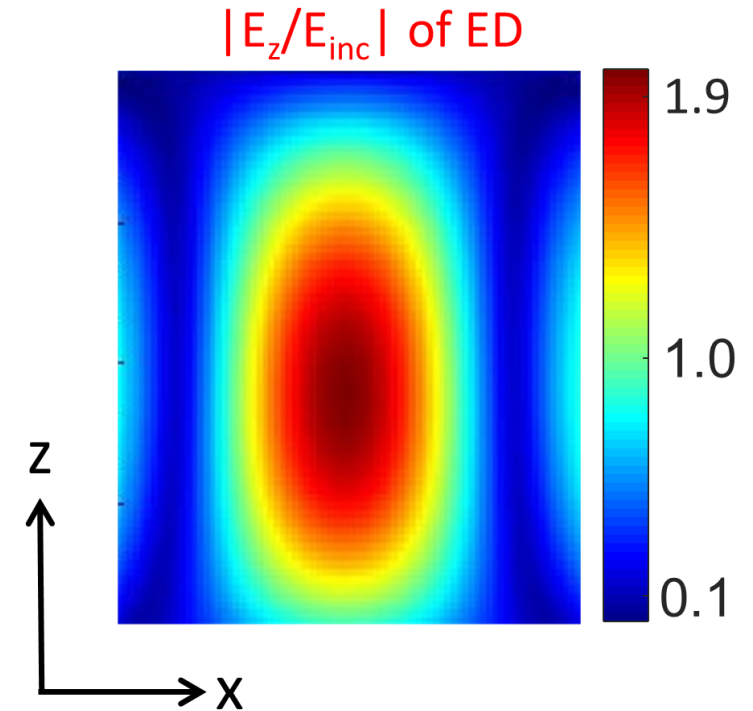
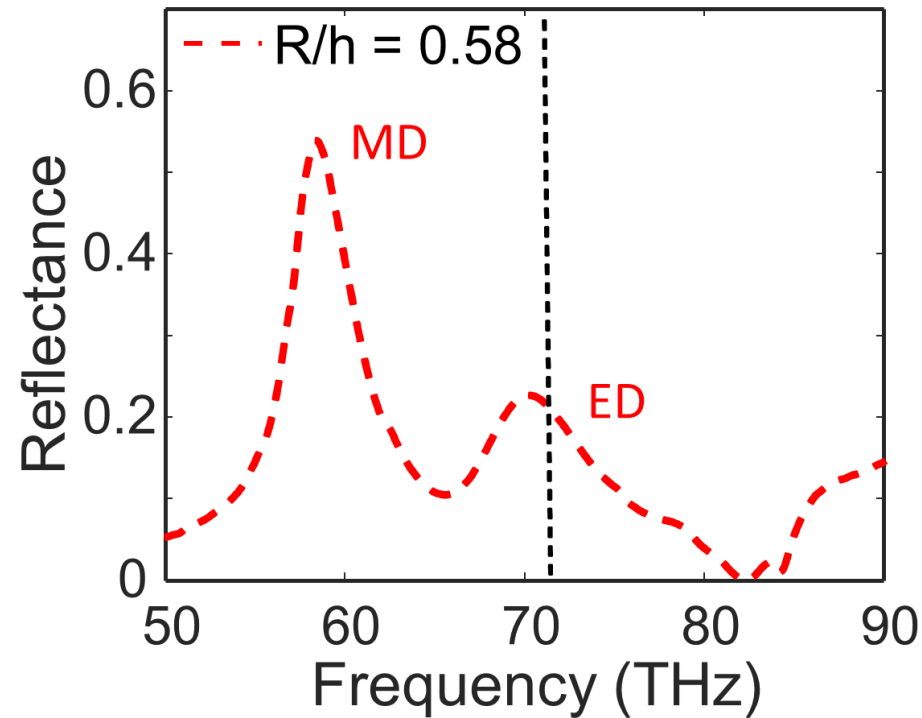
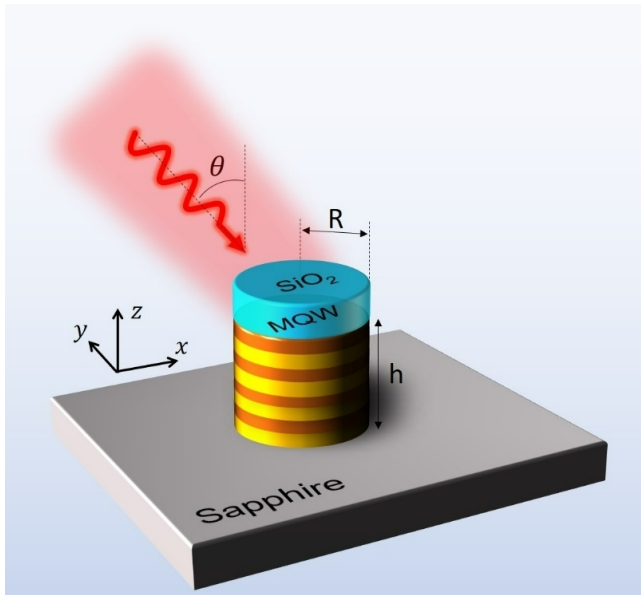


Frequency (THz)
 $N_{\text{QW}} = 40, N_{\text{d}} = 3\text{e}18 \text{ cm}^{-3}$

Frequency (THz)
 $N_{\text{QW}} = 30, N_{\text{d}} = 8\text{e}17 \text{ cm}^{-3}$

Frequency (THz)
 $N_{\text{QW}} = 40, N_{\text{d}} = 3\text{e}18 \text{ cm}^{-3}$

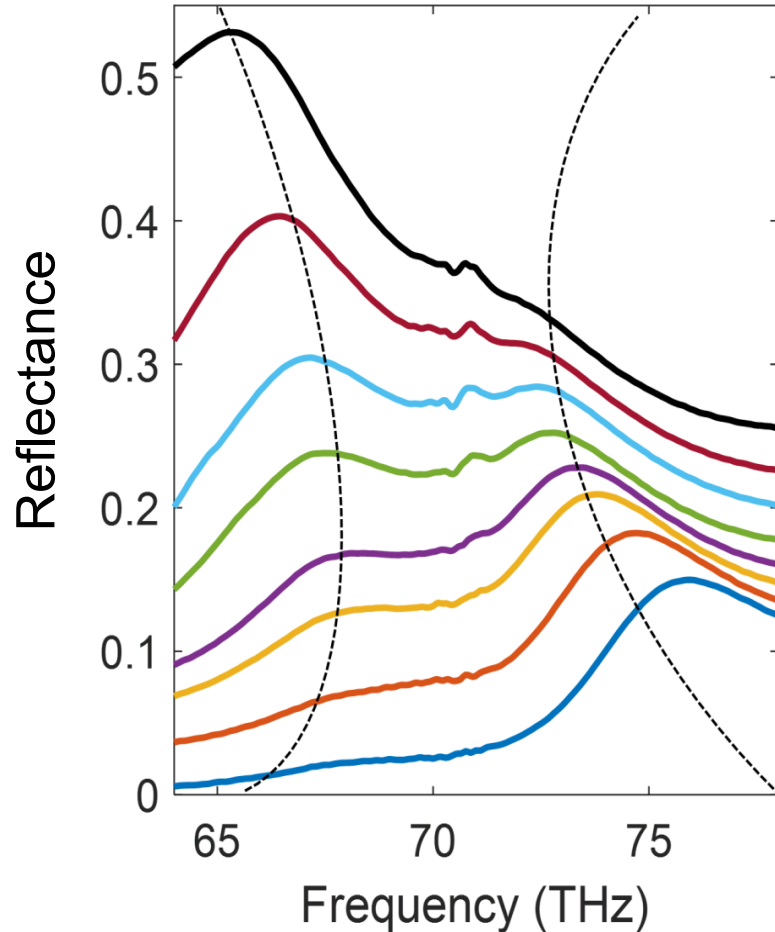
Controlling Light-Matter Interaction by Coupling to Different Photonic Modes



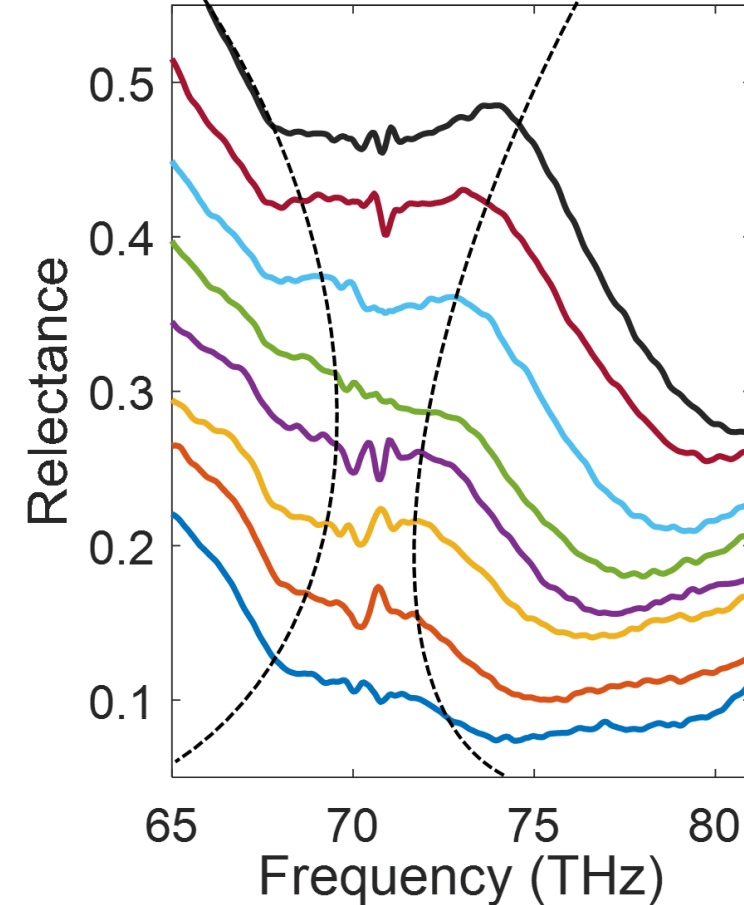
Since different Mie modes have different mode volumes and field enhancements, we can modify light-matter interaction by coupling the IST to different photonic Mie modes (without modifying the heterostructure).

Controlling Light-Matter Interaction by Coupling to Different Photonic Modes

Magnetic Dipole ($N_{\text{QW}} = 30$, $N_{\text{d}} = 3 \times 10^{18} \text{ cm}^{-3}$)

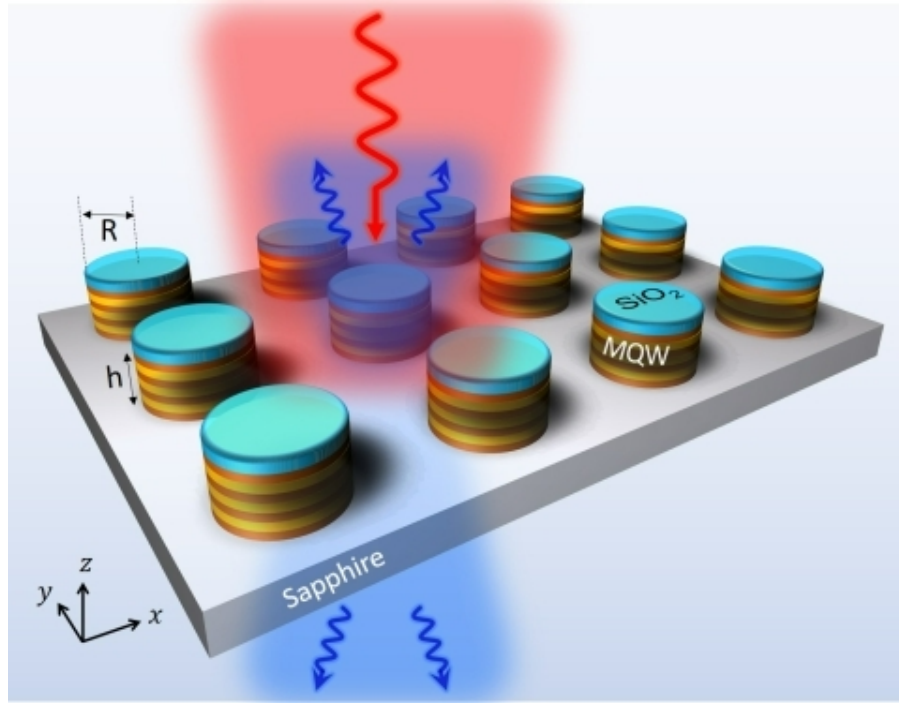


Electric Dipole ($N_{\text{QW}} = 30$, $N_{\text{d}} = 3 \times 10^{18} \text{ cm}^{-3}$)

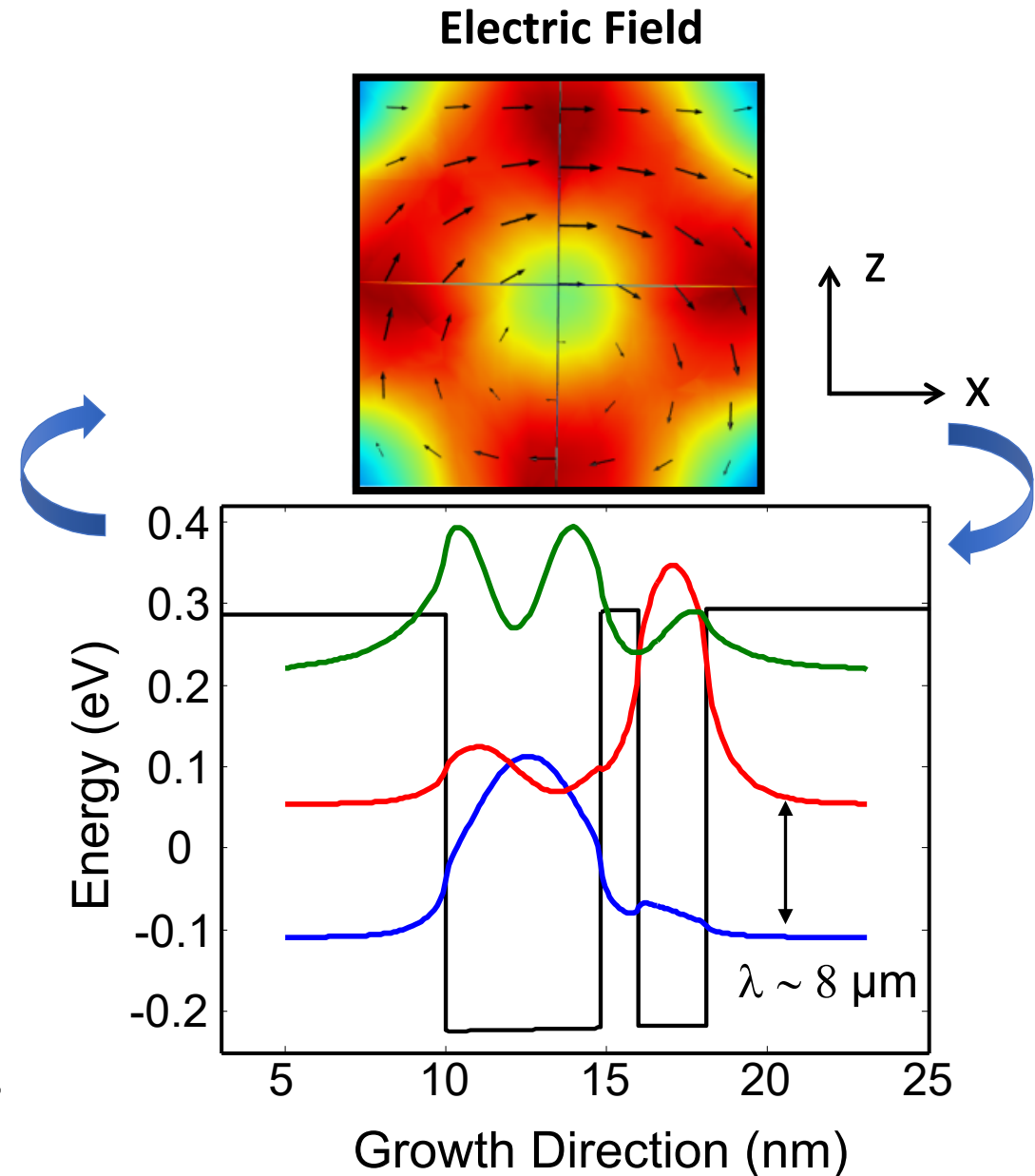


The magnetic dipole modes show stronger light-matter interaction !

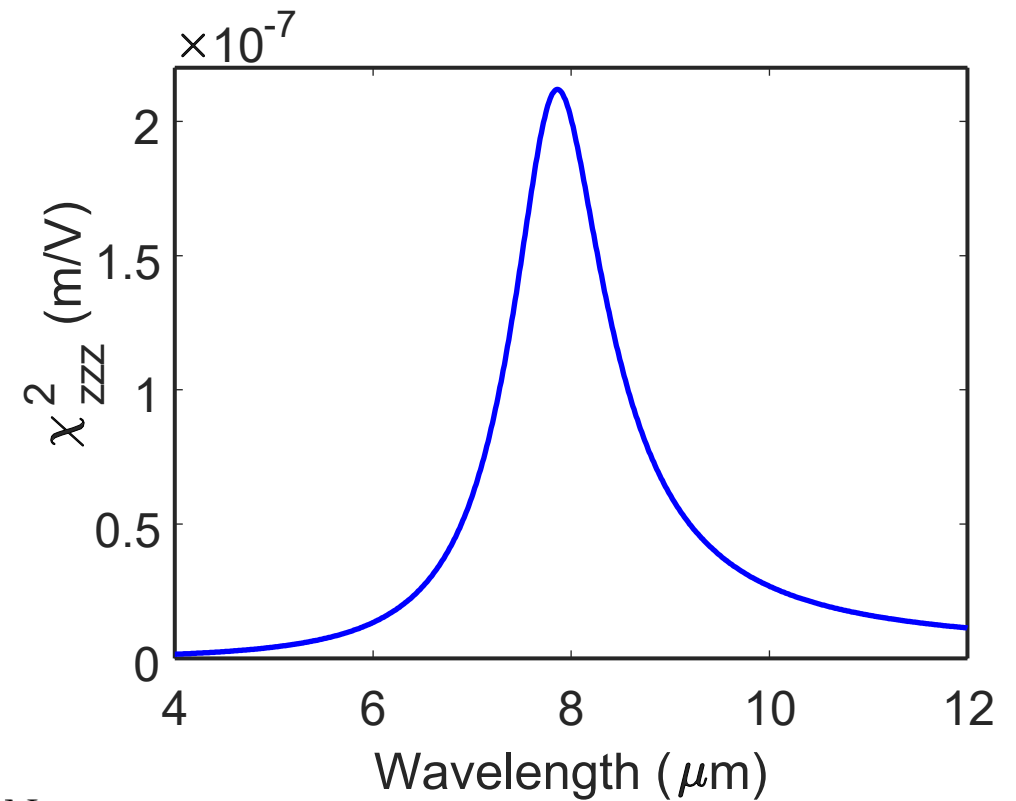
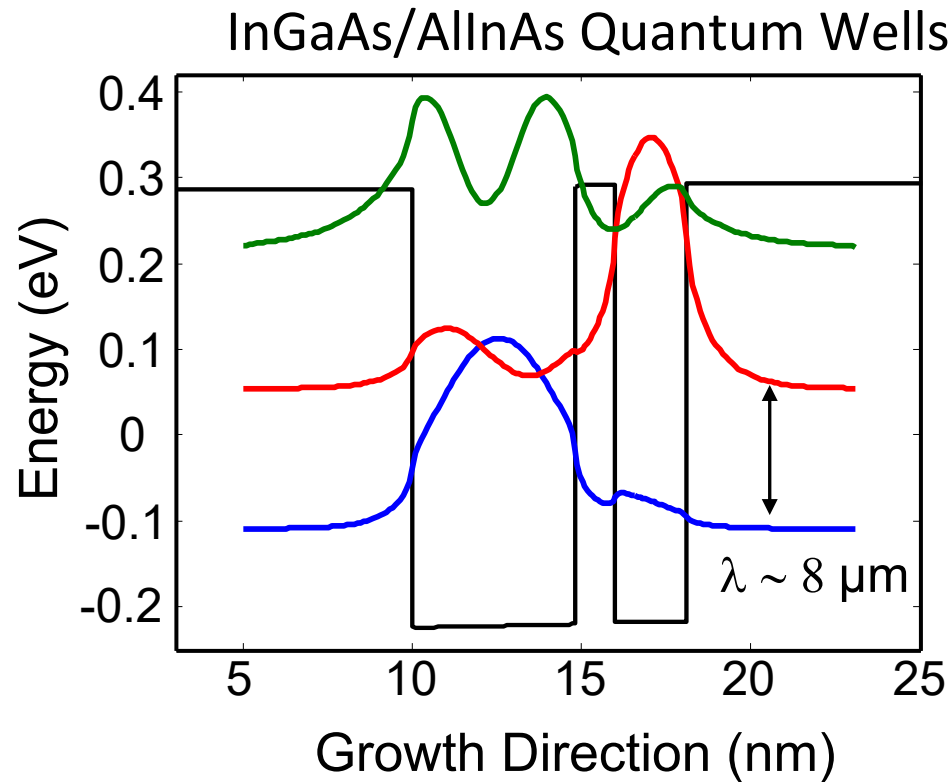
Polaritonic Metasurface : Extreme Nonlinearities



- Light-matter coupling between a MD Mie mode and intersubband electronic excitations.
- MD mode has strong z electric field components, allows for normal incidence, and smallest size of the resonator.



Resonant $\chi^{(2)}$ using Intersubband Transitions

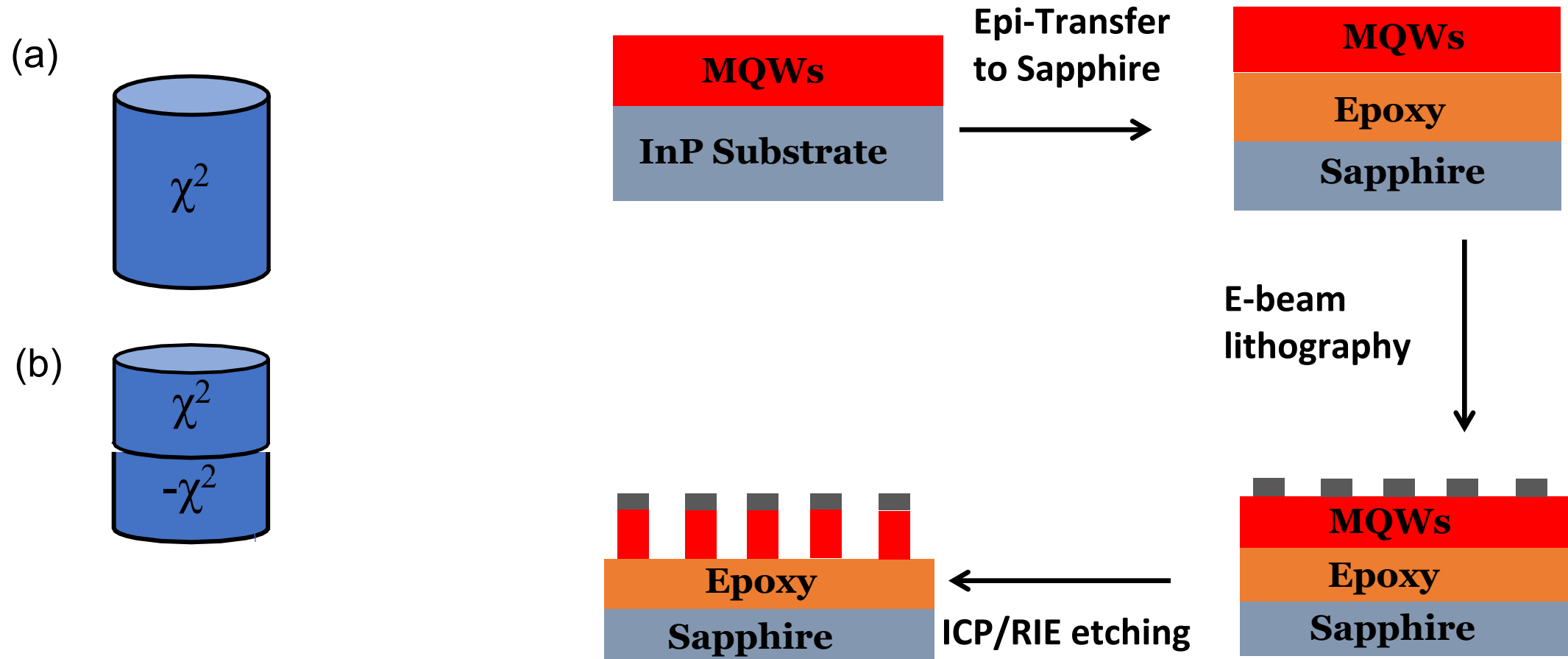


$$\chi^{(2)}(\omega) \propto \frac{N \cdot z_{12} z_{23} z_{13}}{(\omega - \omega_{12} - i\Gamma_{12})(2\omega - \omega_{13} - i\Gamma_{13})}$$

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

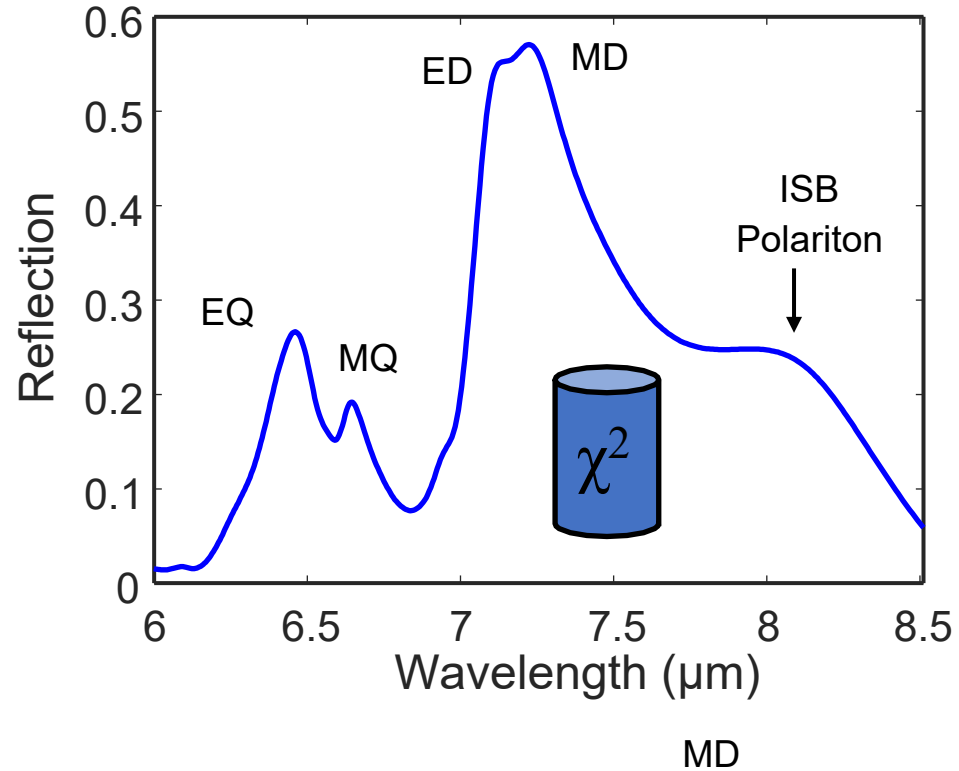
IEEE J. Quantum Electr. 30, 1313 (1994)

Fabrication of the All-Dielectric Nonlinear Metasurface

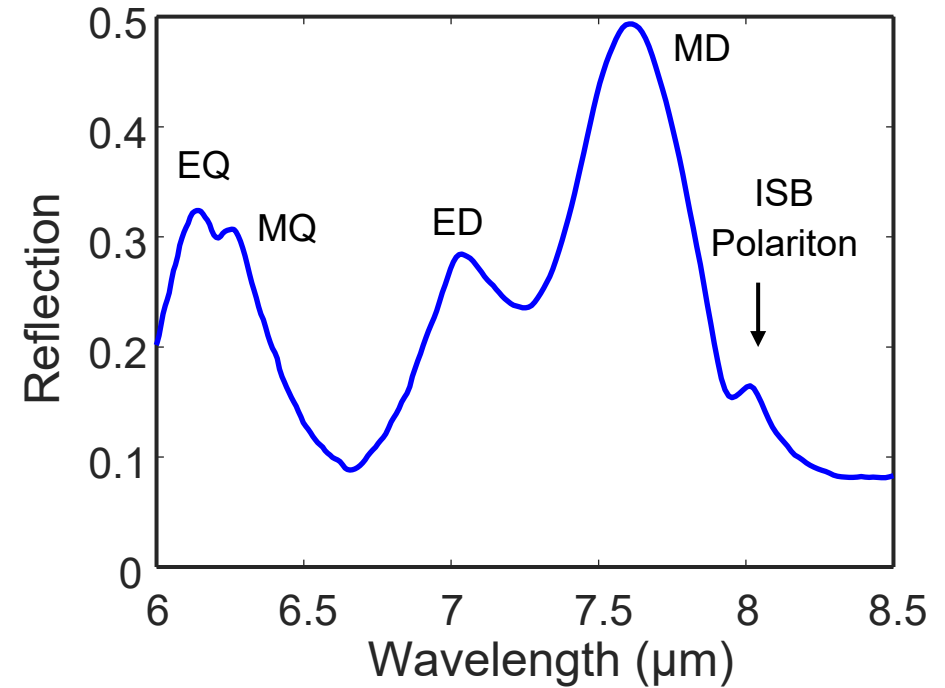


Linear Reflectance Spectra : Simulation and Experiment

Simulation

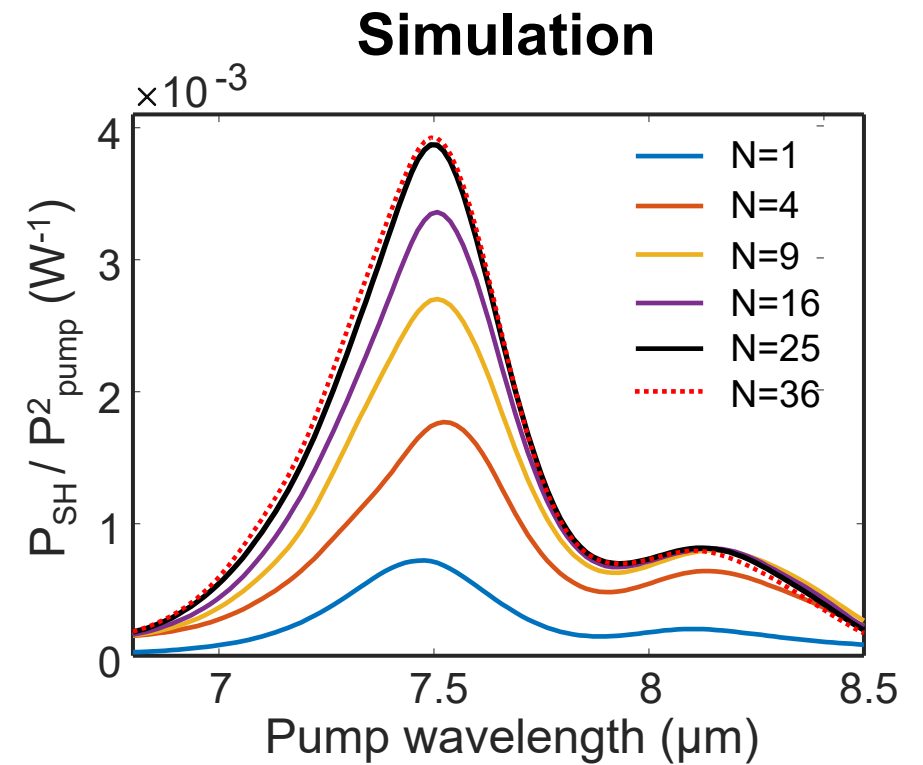
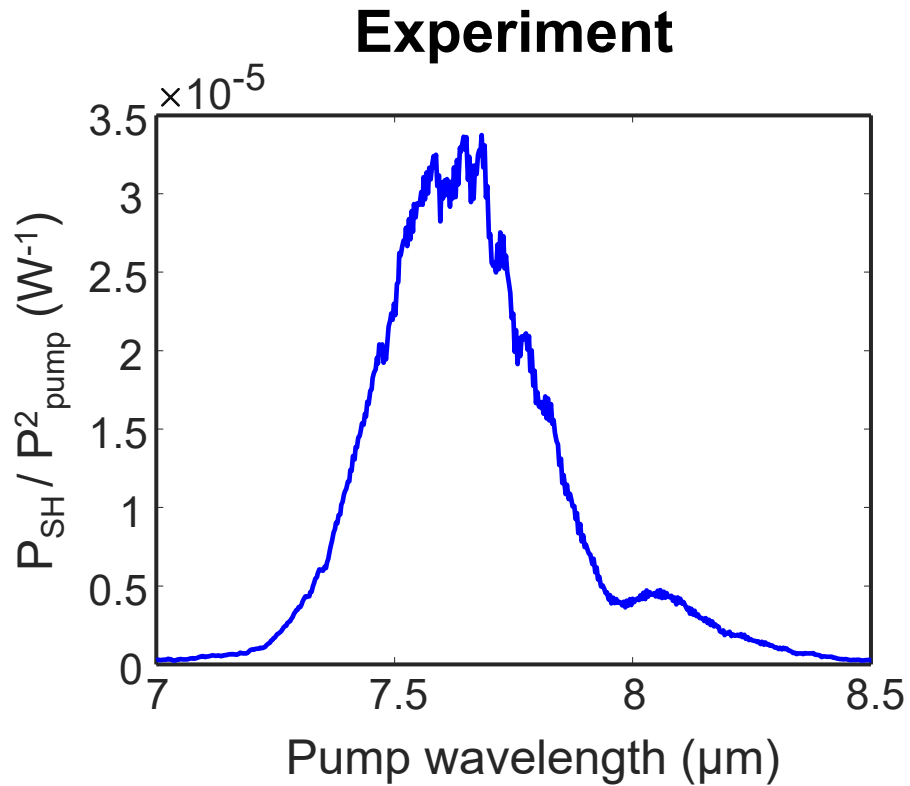


Experiment



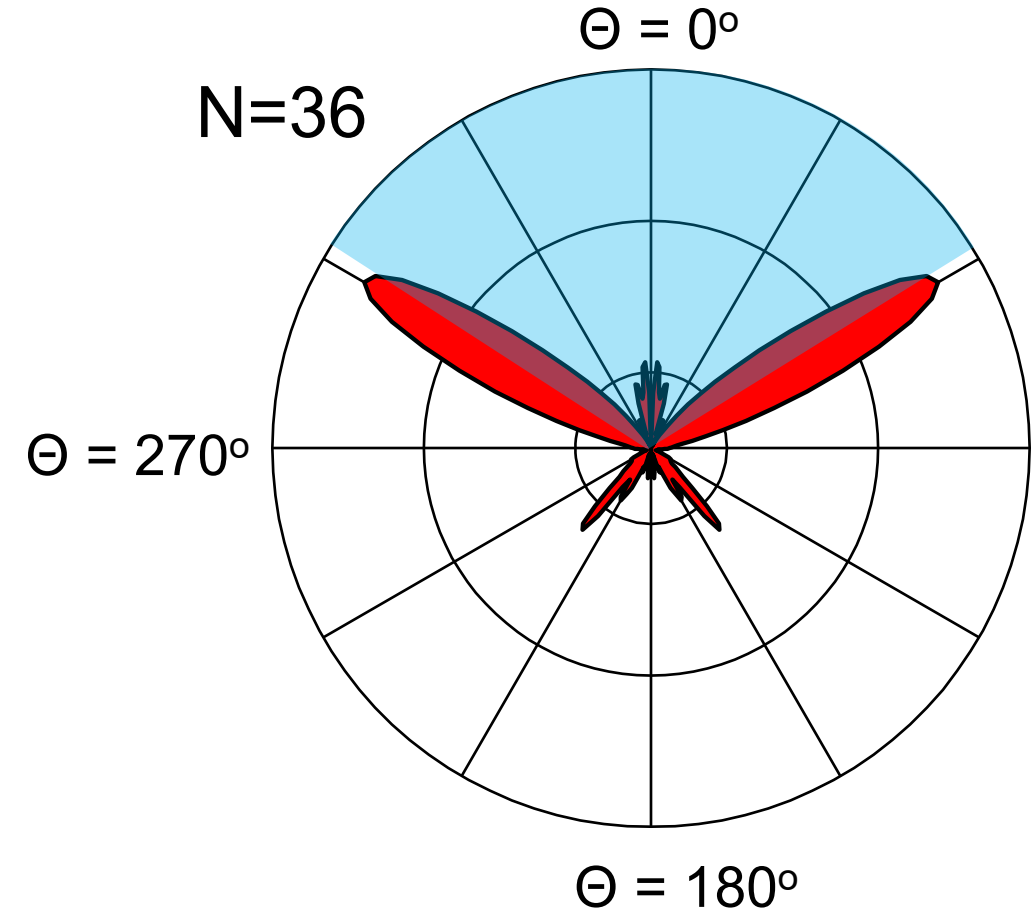
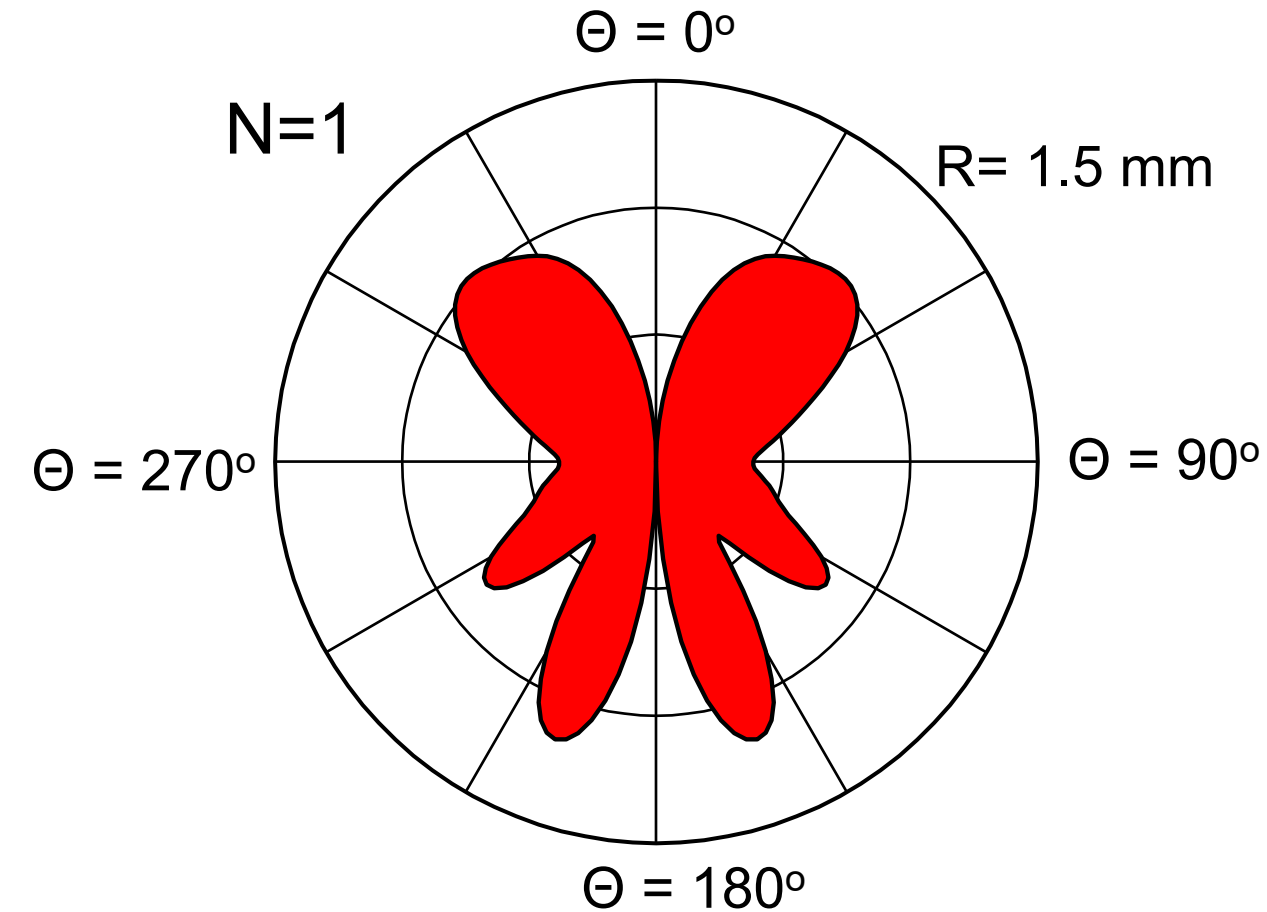
Splitting of Photonic Resonance due to Strong Coupling !

Second-Harmonic Generation : Experiment and Simulations

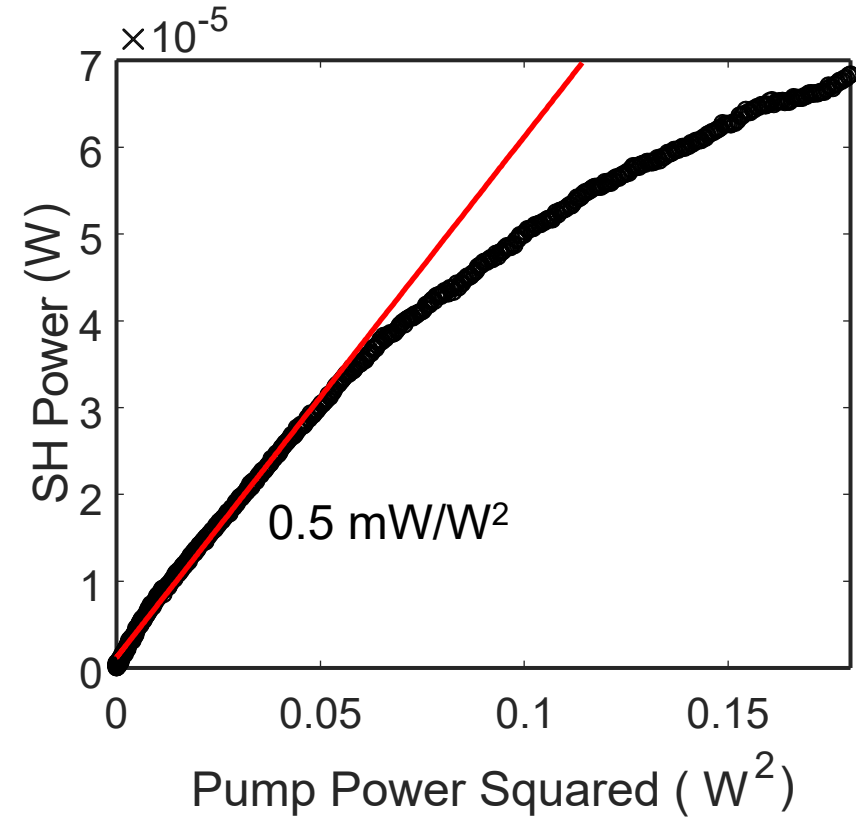
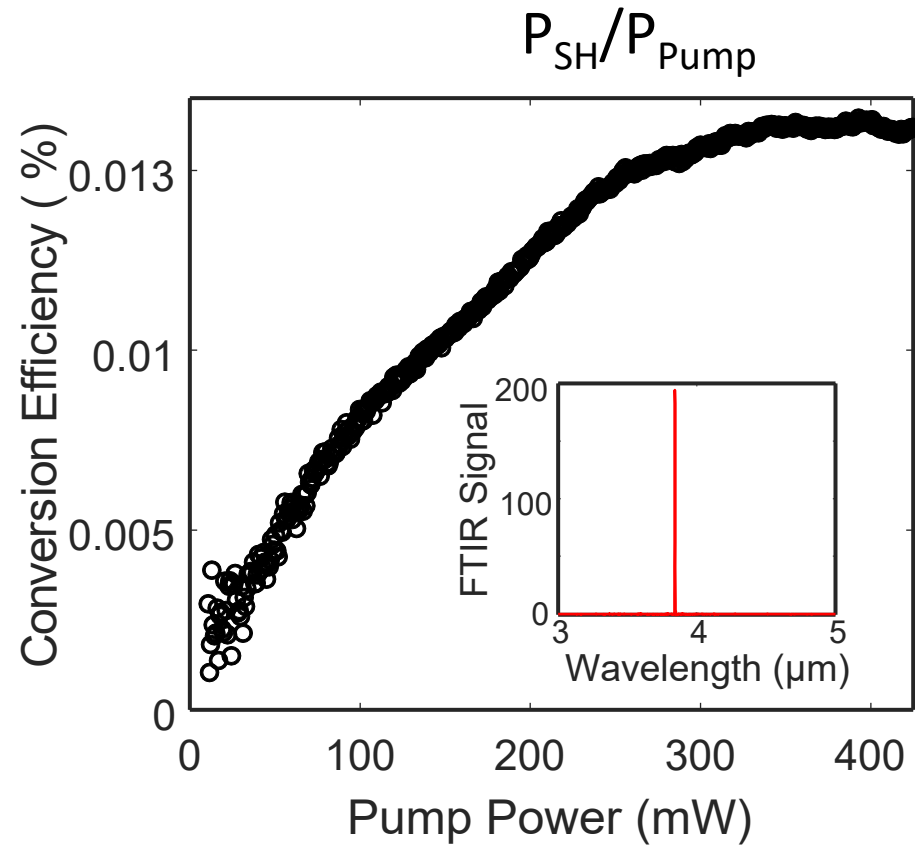


Excellent qualitative agreement between experiments and simulations can be seen !

Limited Collection Efficiency due to Diffraction



Experimentally Measured SHG Efficiencies

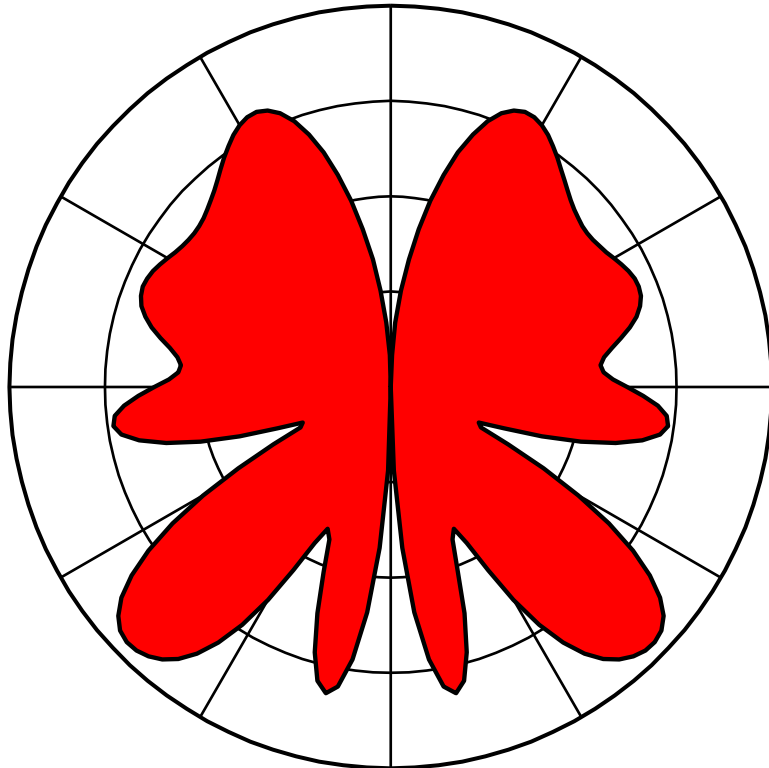


Large magnitude of $\chi^{(2)}$ gives SHG efficiency $\sim 0.013\%$ at $11\text{ kW}/\text{cm}^2$ and conversion factor $\sim 0.5\text{ mW}/\text{W}^2$.

Enhancing Collection Efficiency by Varying Radius & Period

N=1

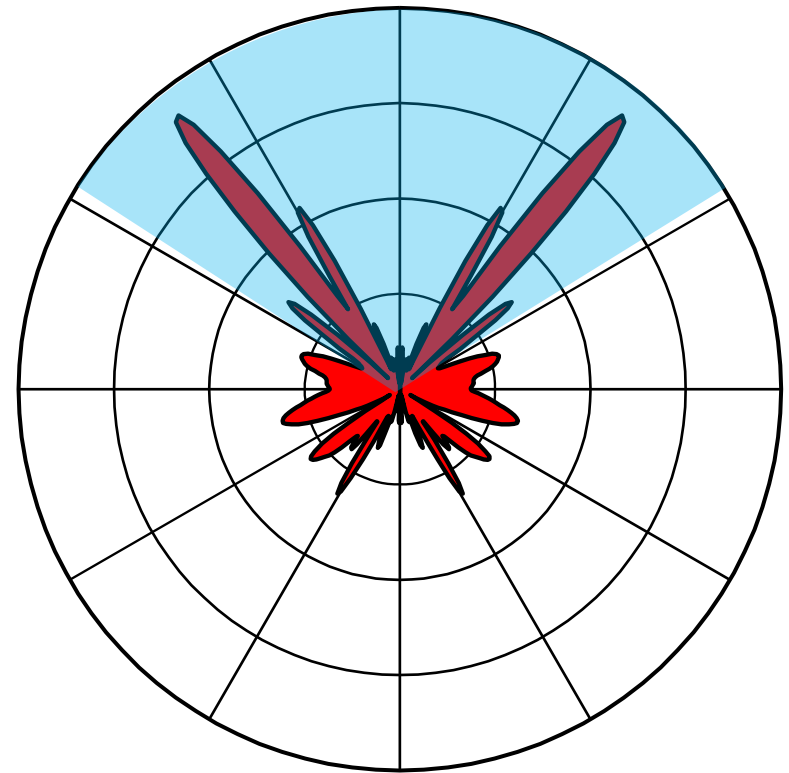
$\Theta = 0^\circ$



$\Theta = 180^\circ$

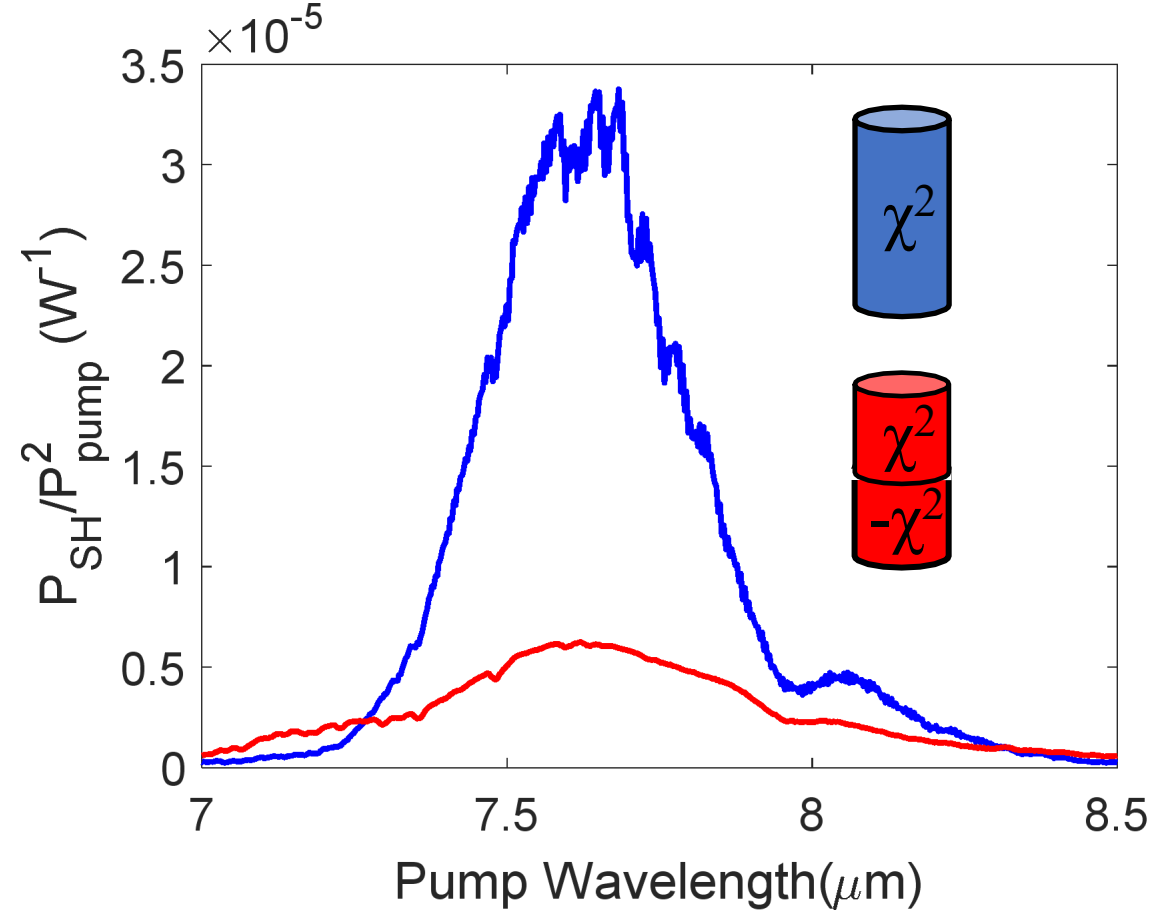
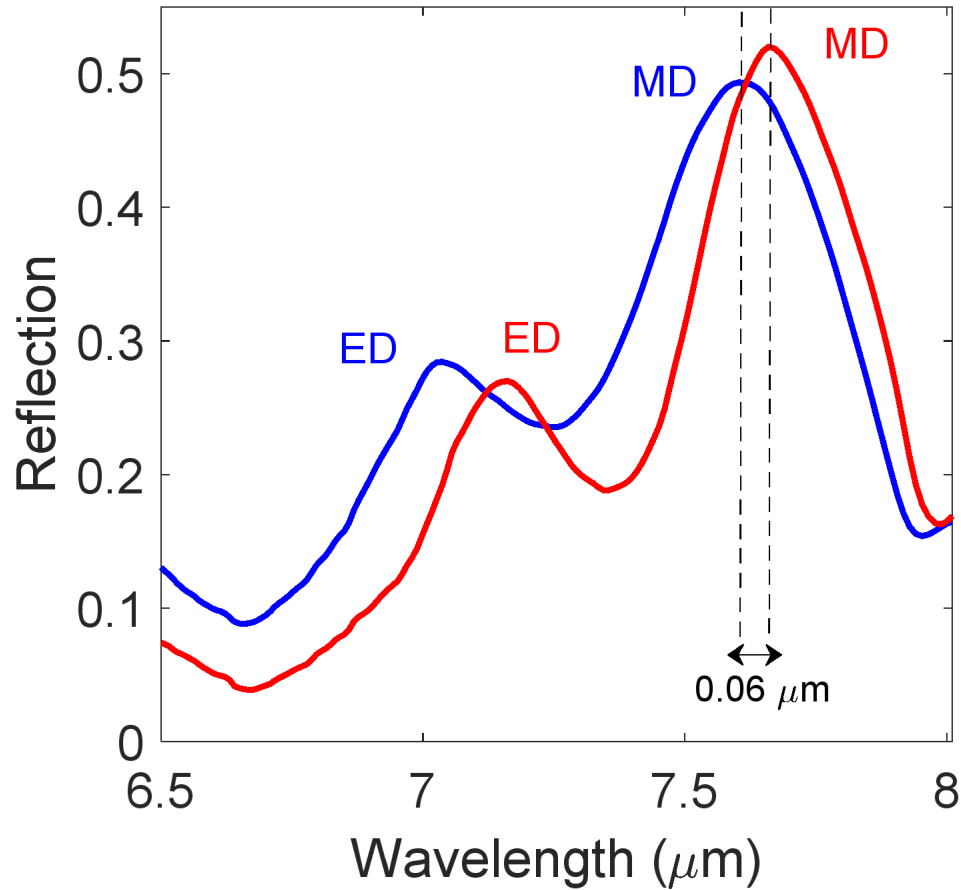
N=36

$\Theta = 0^\circ$



$\Theta = 180^\circ$

Polarity Switching of $\chi^{(2)}$



The SHG efficiency can be controlled by **controlling the sign of $\chi^{(2)}$** inside the Mie resonator

Summary

- We demonstrate giant second-order nonlinearities in polaritonic all-dielectric metasurfaces which can be controlled via microscopic control of magnitude and sign of the material nonlinearity.
- Our results are proof-of-concept and the efficiencies can be improved by optimizing the heterostructure, field overlaps, and interplay between field enhancement and nonlinearity.
- Our approach although demonstrated for a particular wavelength, in principle, can be scaled to other wavelengths from visible to near-IR.

Ref. : [1] R. Sarma et al., Nano Letters 21(1), 367-374 (2021).

[2] R. Sarma et al., Nano Letters 22(3), 896-903 (2022).