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All-Dielectric Polaritonic Metasurfaces : From Strong Coupling to Extreme Nonlinearites

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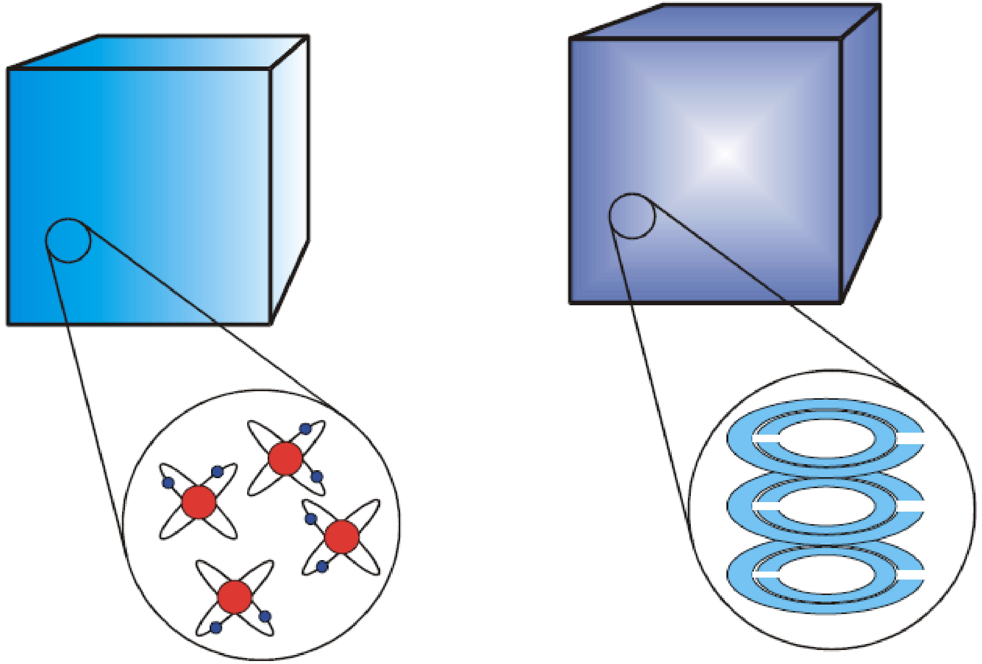
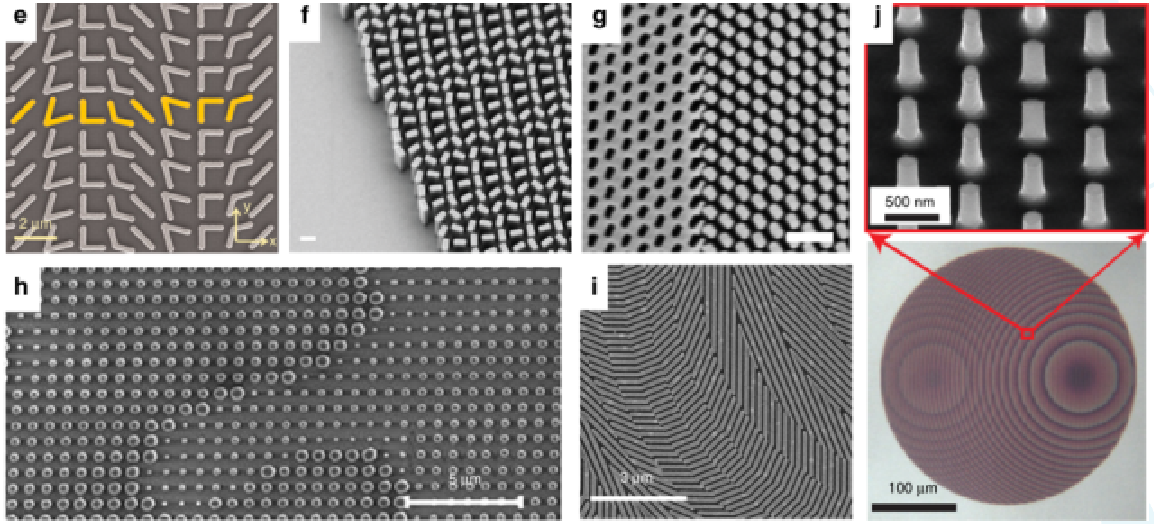
Metamaterials and Metasurfaces

Man-made "atoms" : Metamaterials



Metasurfaces



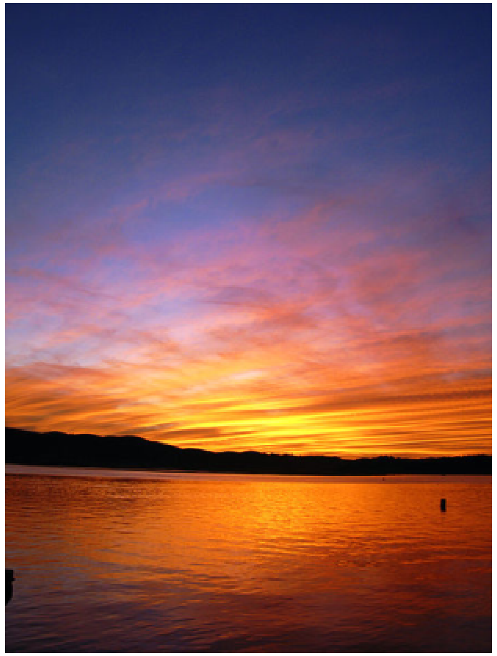
Ref. : Neshev & Aharonovich, Light : Science & Applications 7 (58), 2018.

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

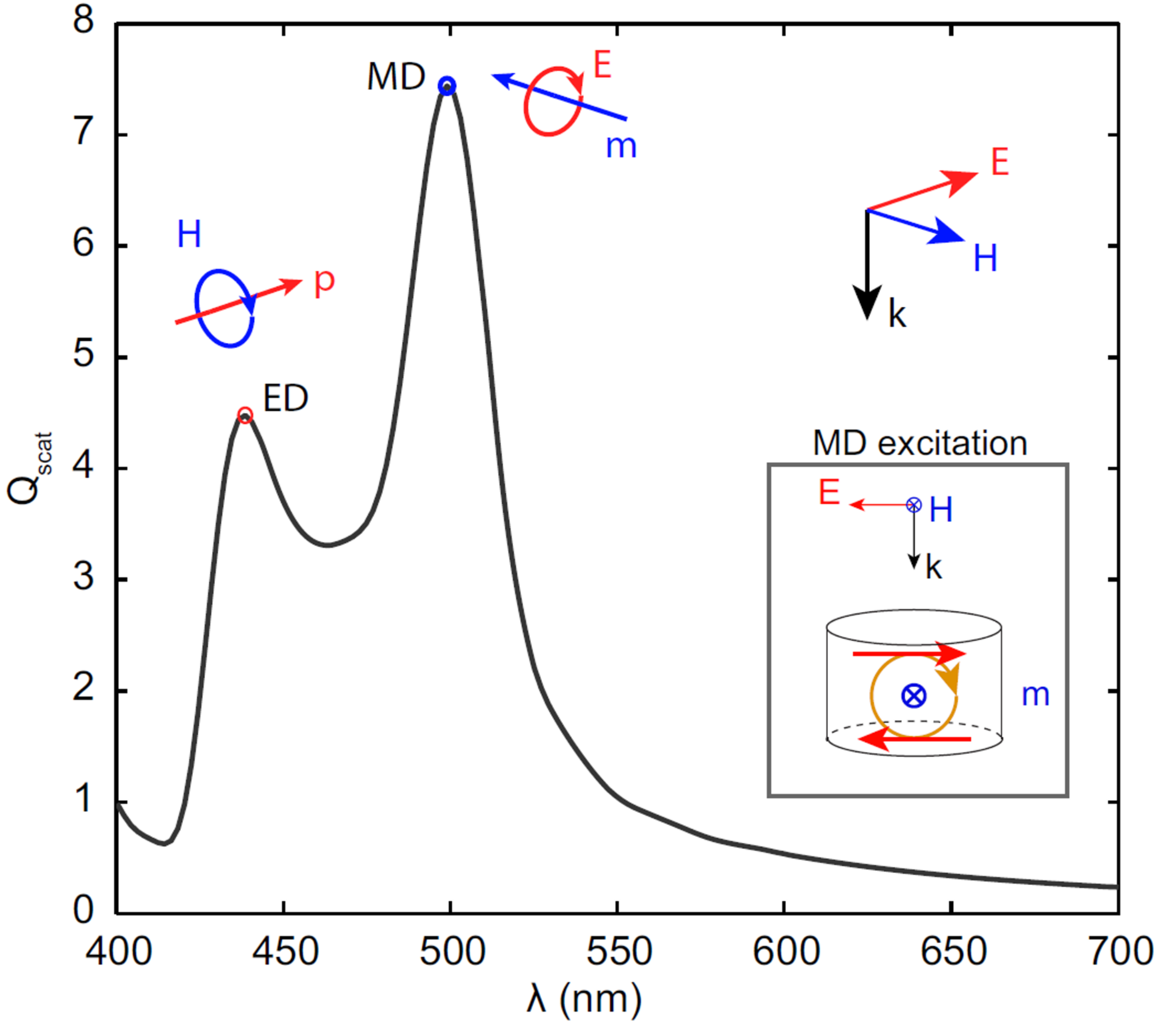
Dielectric particles much smaller than wavelength λ/n



Wikipedia

Rayleigh << λ/n

Mie modes in dielectric particles comparable ~ λ/n



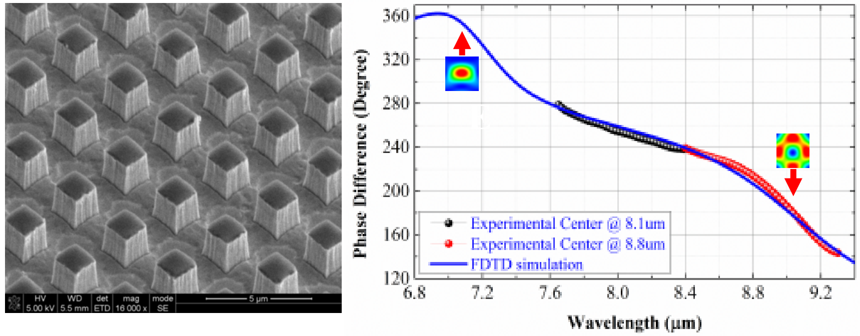
For dielectric particles, the polarizabilities of the electric and magnetic dipole resonances are <u>comparable at optical frequencies</u>

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

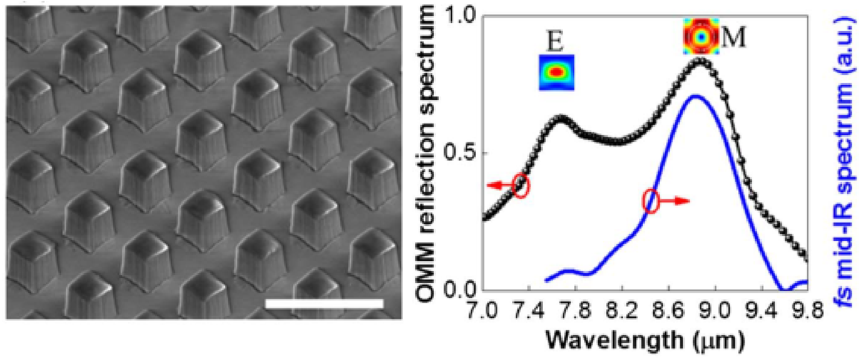
Ref. : Optics Express 21, 26285 (2013)

Applications of Mie Modes in Dielectric Metasurfaces

Magnetic Response

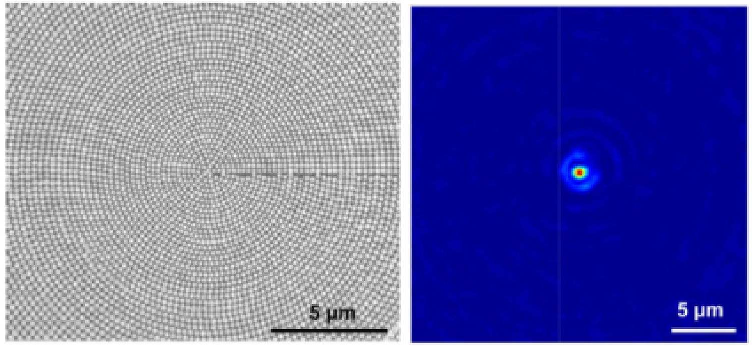


PRL. 108, 097402 (2012)

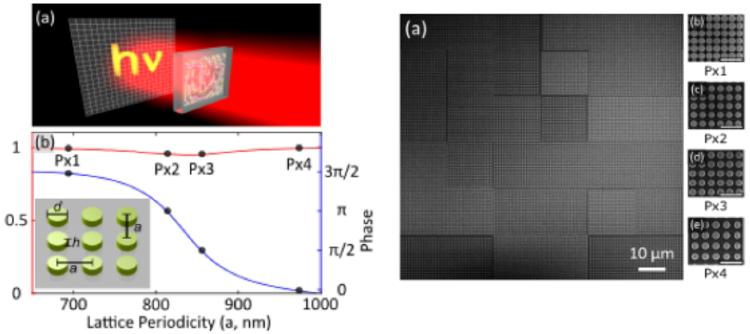


Optica 1, 250 (2014)

Tailoring Linear Transmission

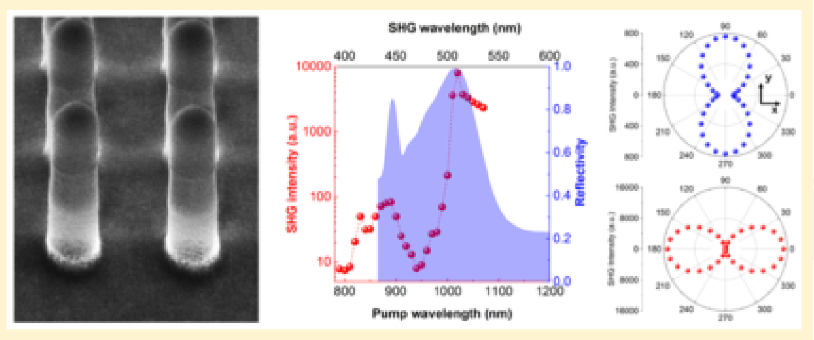


Argonne National Labs

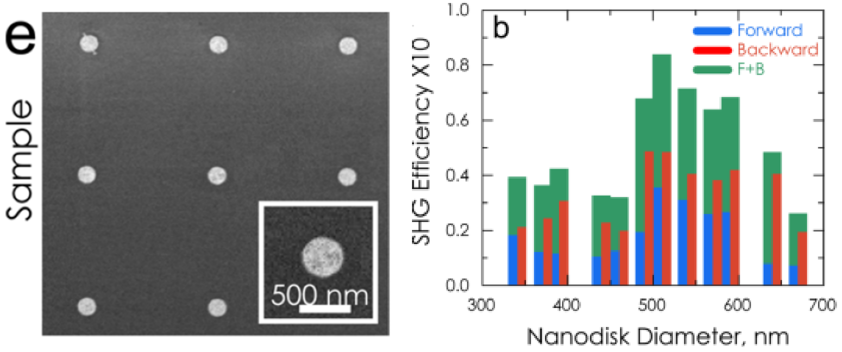


ACS Photonics 3, 514 (2016)

Nonlinear Optics



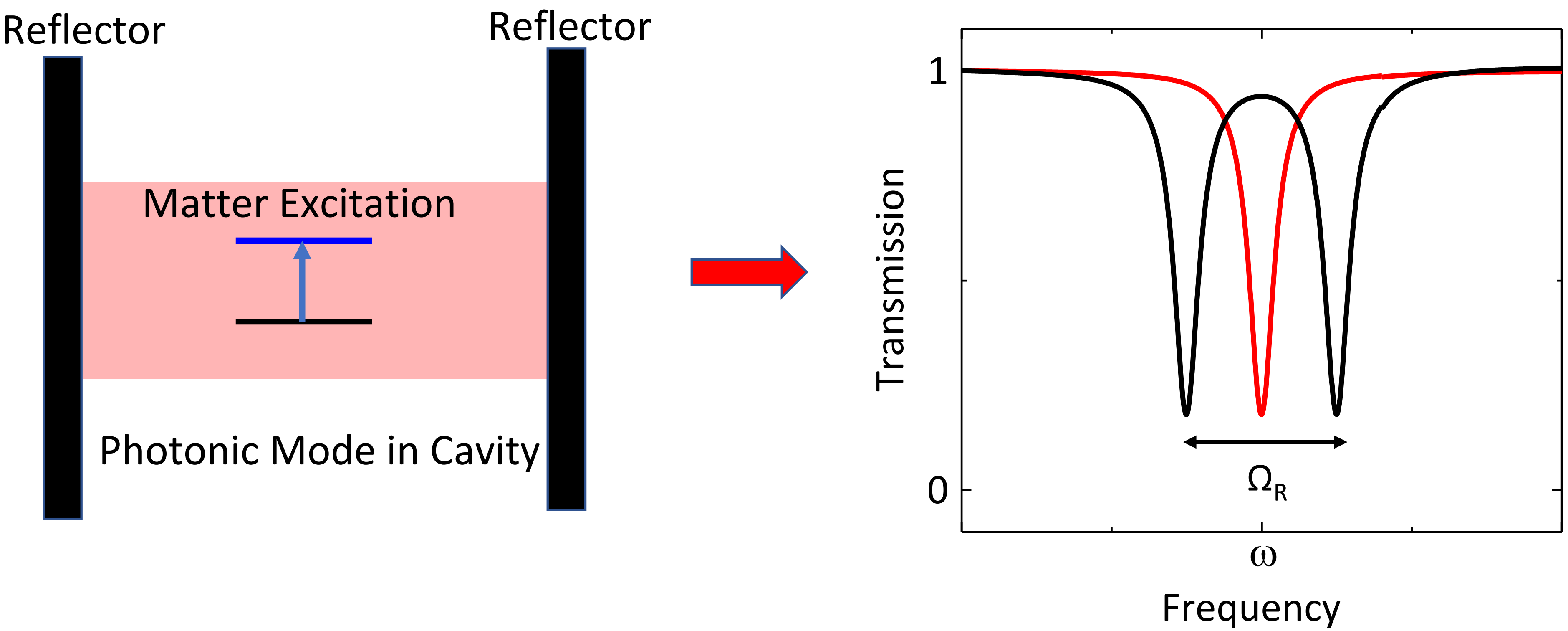
Nano Lett. 16, 5426 (2016)



Nano Lett. 16, 7191 (2016)

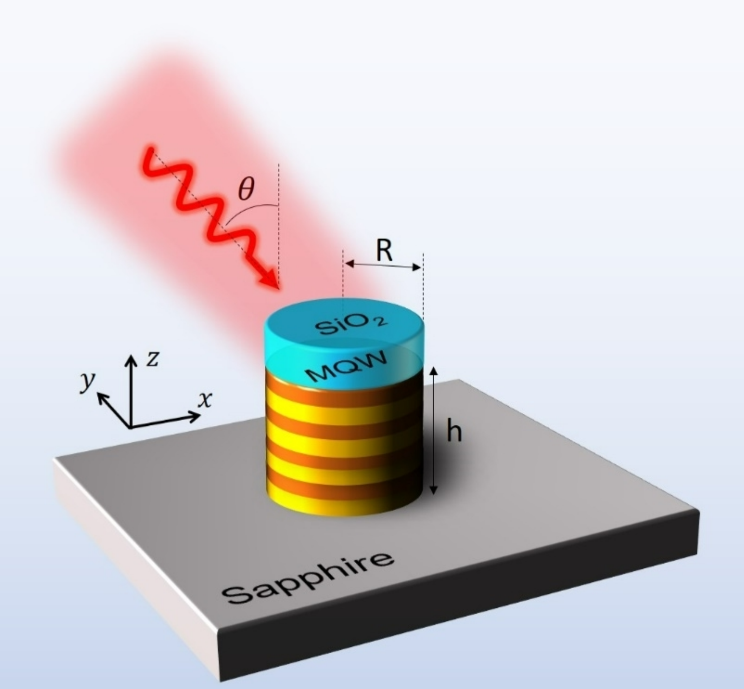
Advantages of Nonlinear Mie Metasurfaces : **1. Ultrathin (relaxed phase matching) 2. Low loss and high damage thresholds 3. Large mode volume (enhanced light-matter interaction) 4. Ease of fabrication**

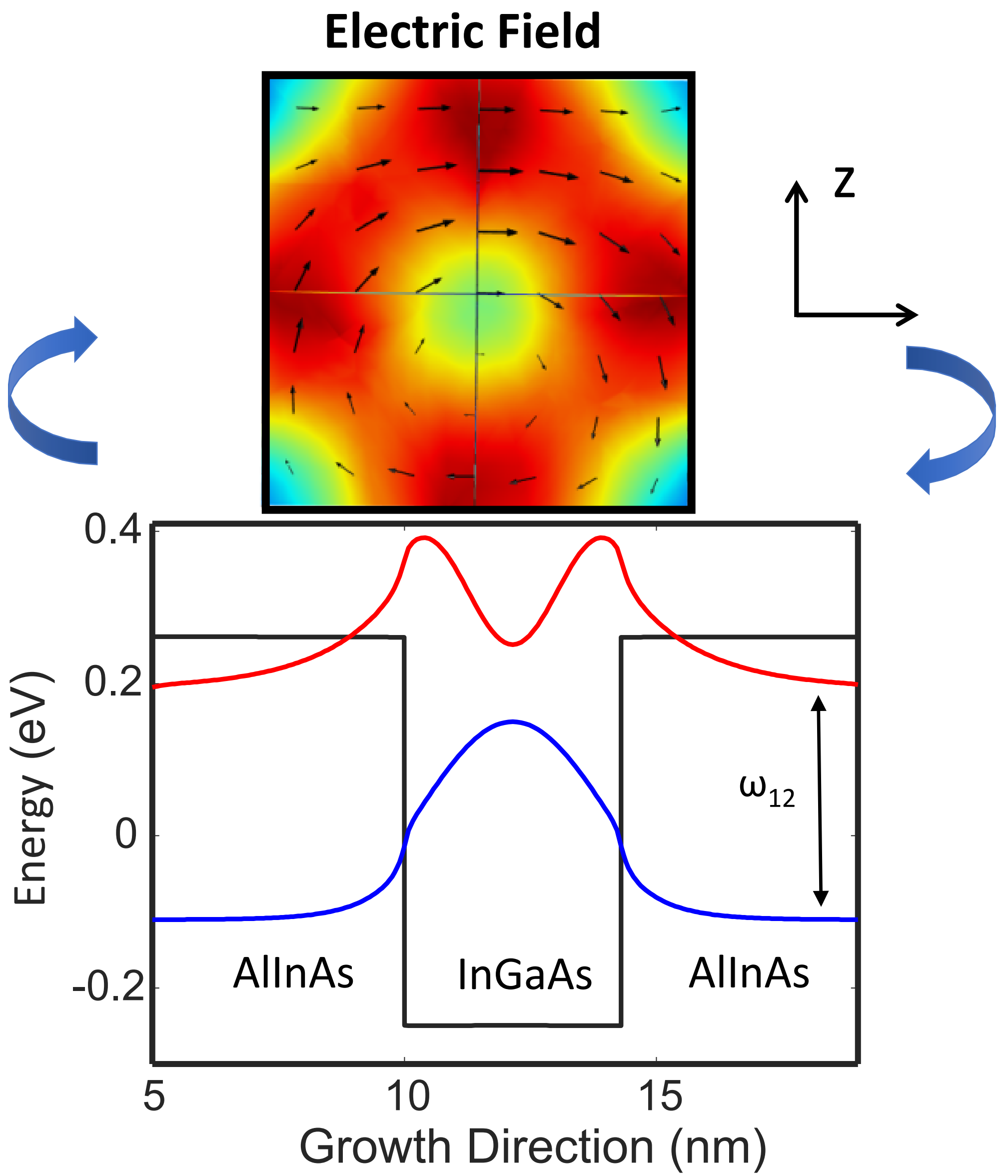
Strong Light Matter Interaction : Polaritons and Rabi Splitting



- **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

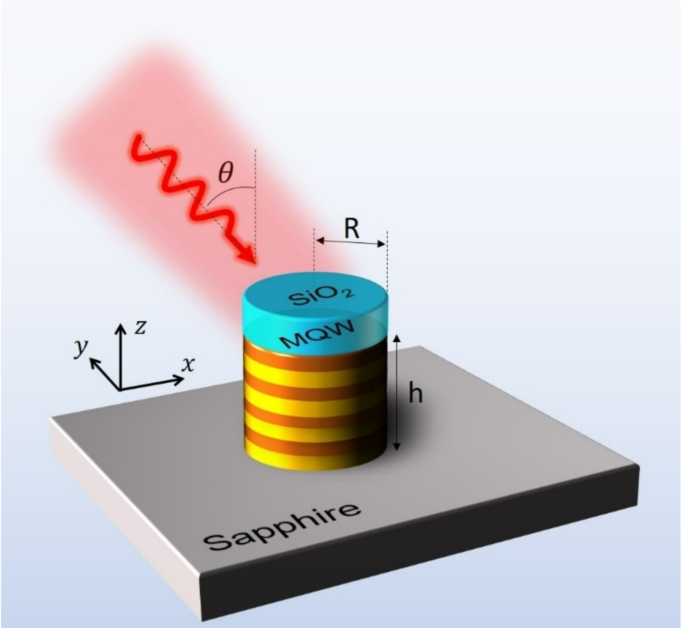


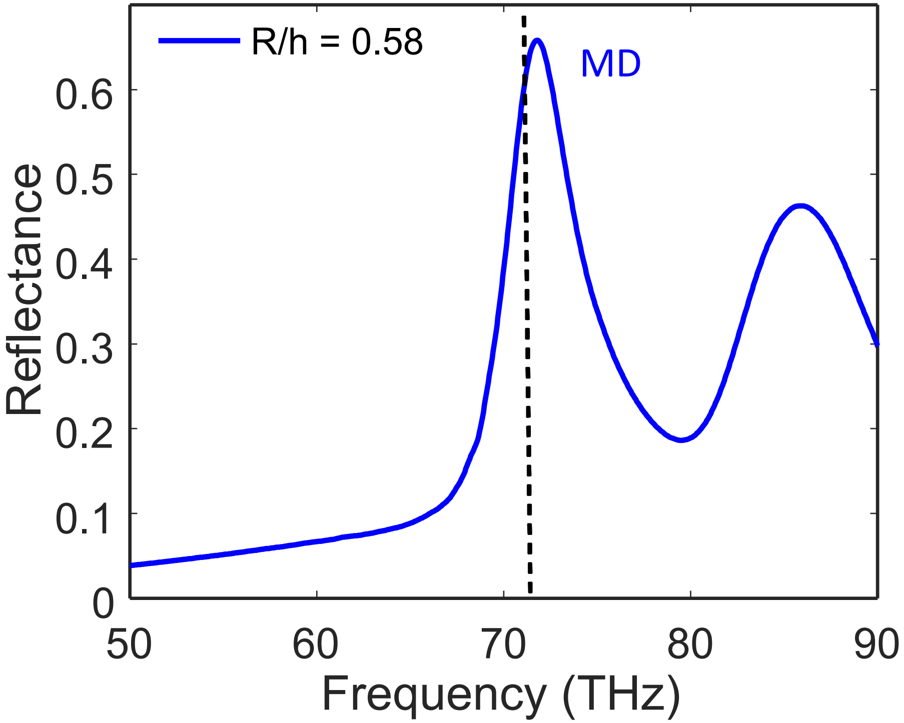


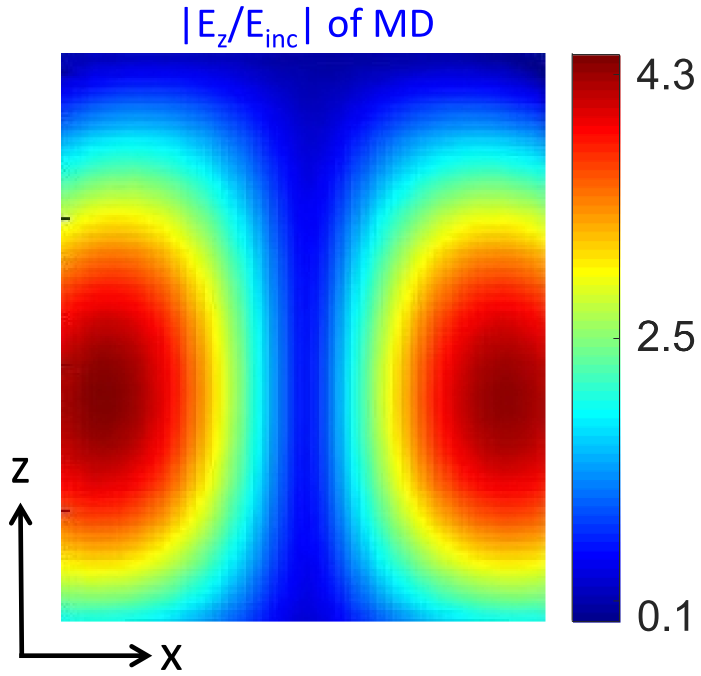
Ω_R depends on :

- **Matter :** Transition dipole moment of intersubband transitions and number of electrons ($N_e = N_d x N_{QW}$) 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

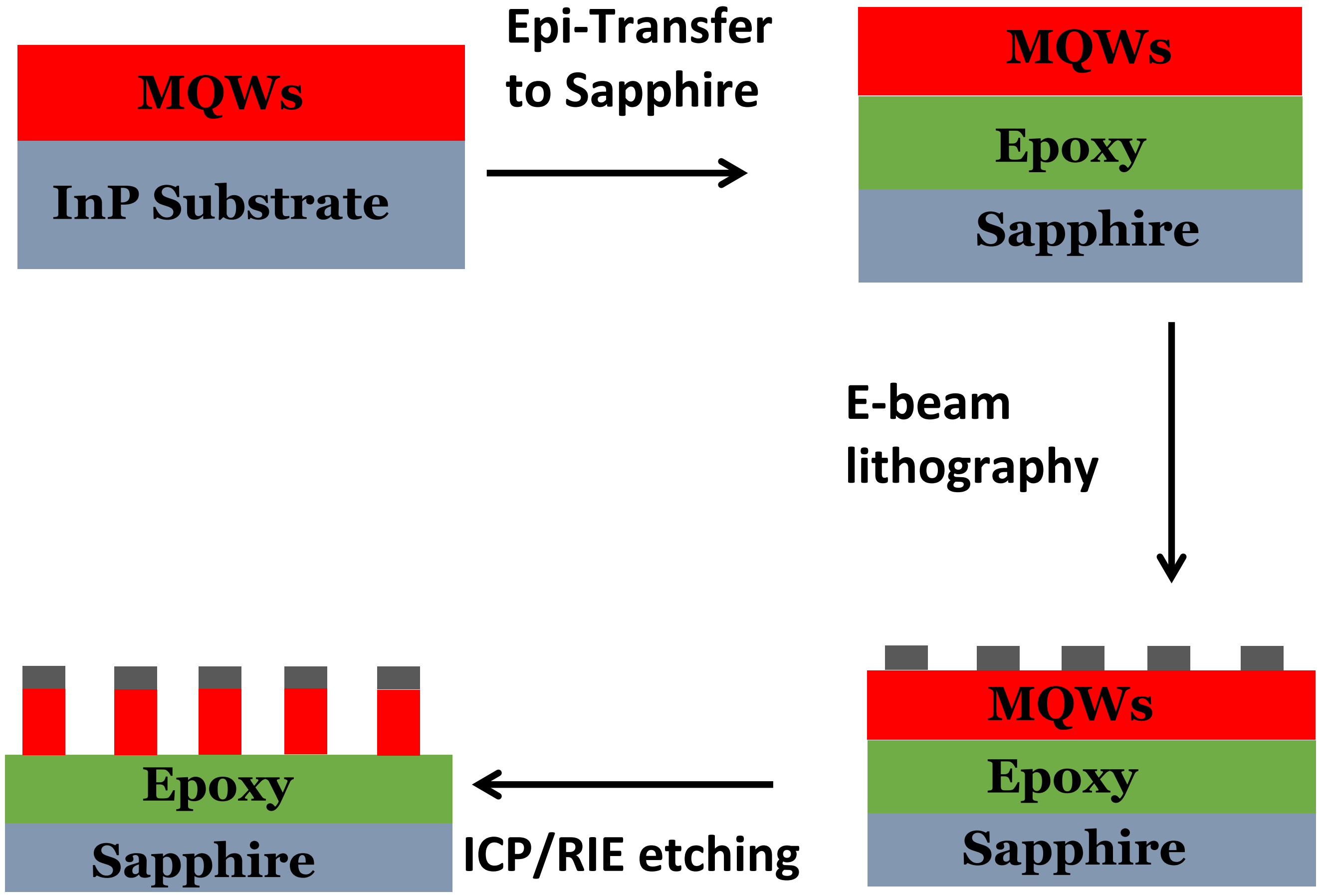




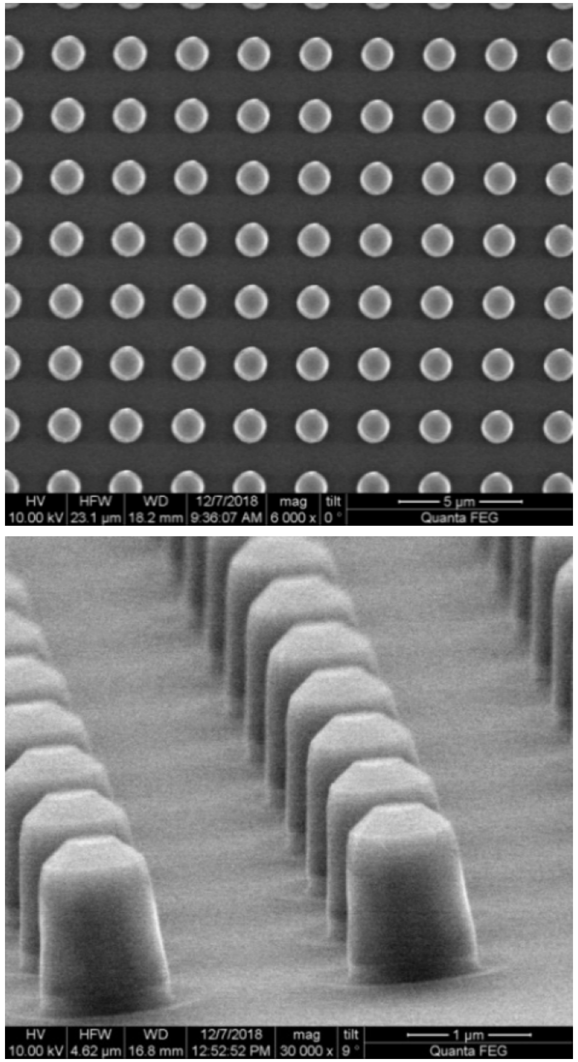


- R/h is tuned to spectrally align the MD mode to the ISB transition
- MD mode has strong z electric field components, allows for normal incidence, and smallest size of the resonator.

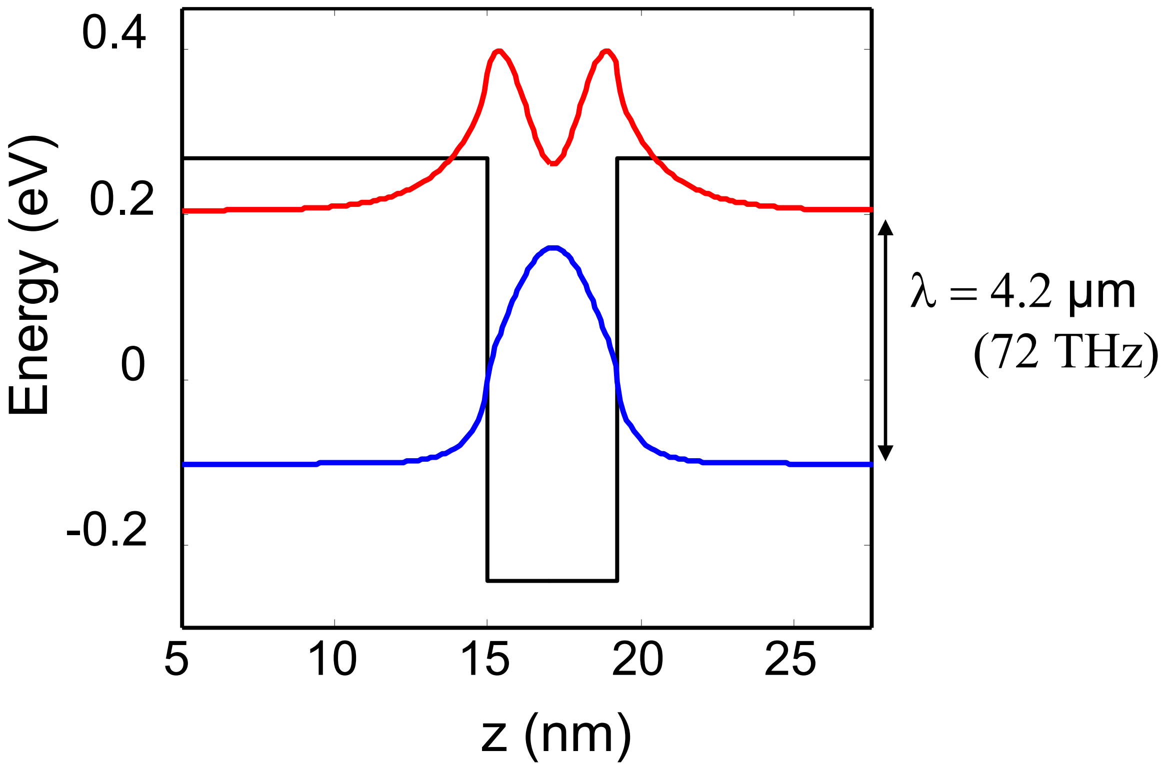
Fabrication of a Polaritonic Metasurface

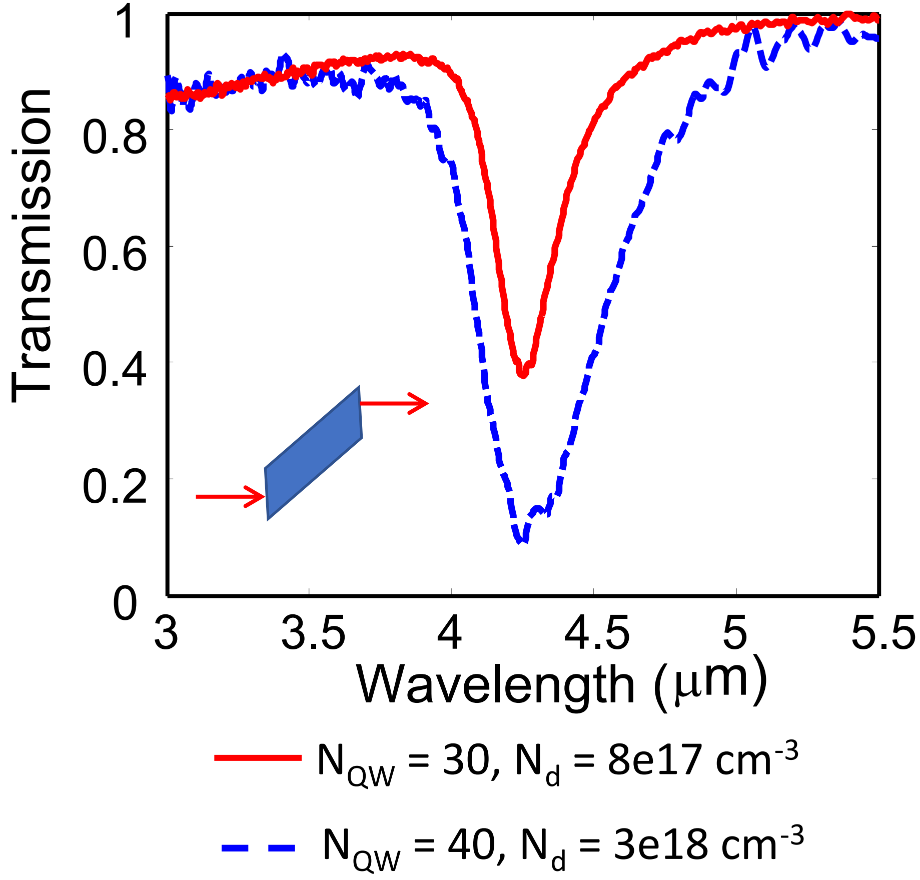


Scanning Electron Micrograph



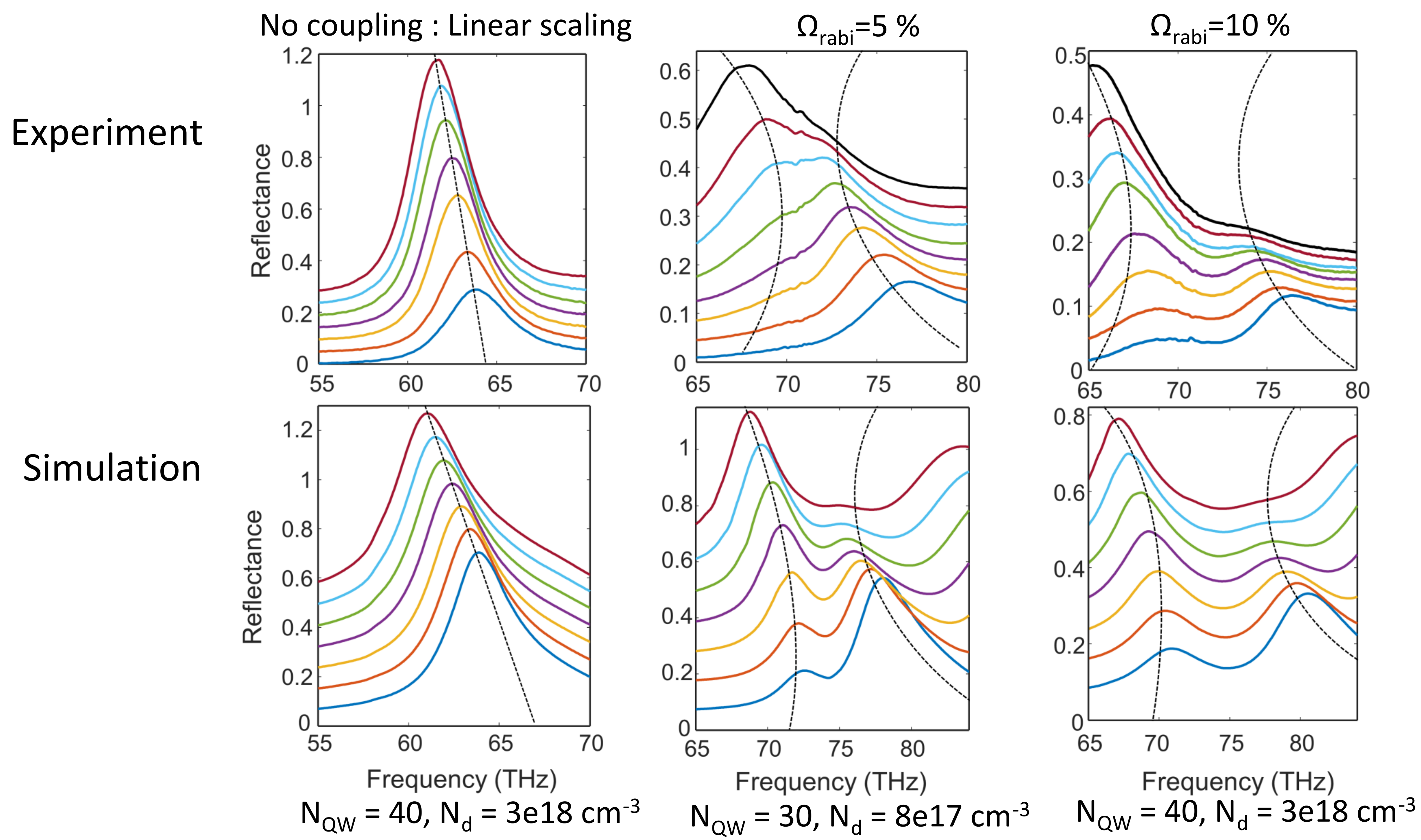
Controlling Light-Matter Interaction by Engineering Heterostructure



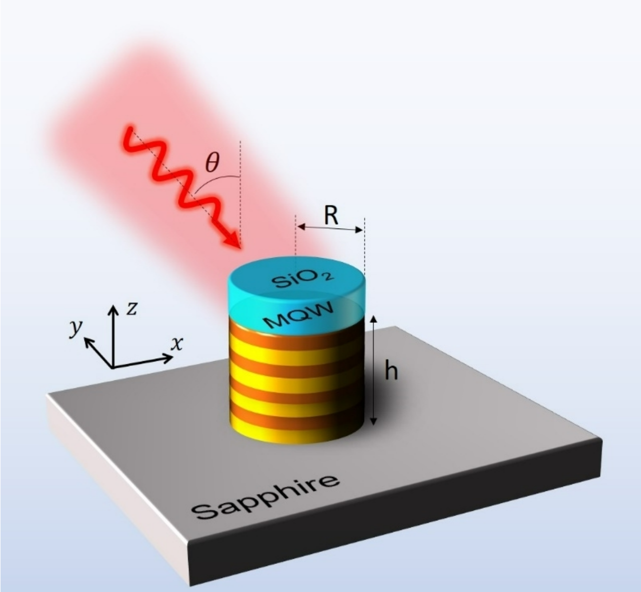


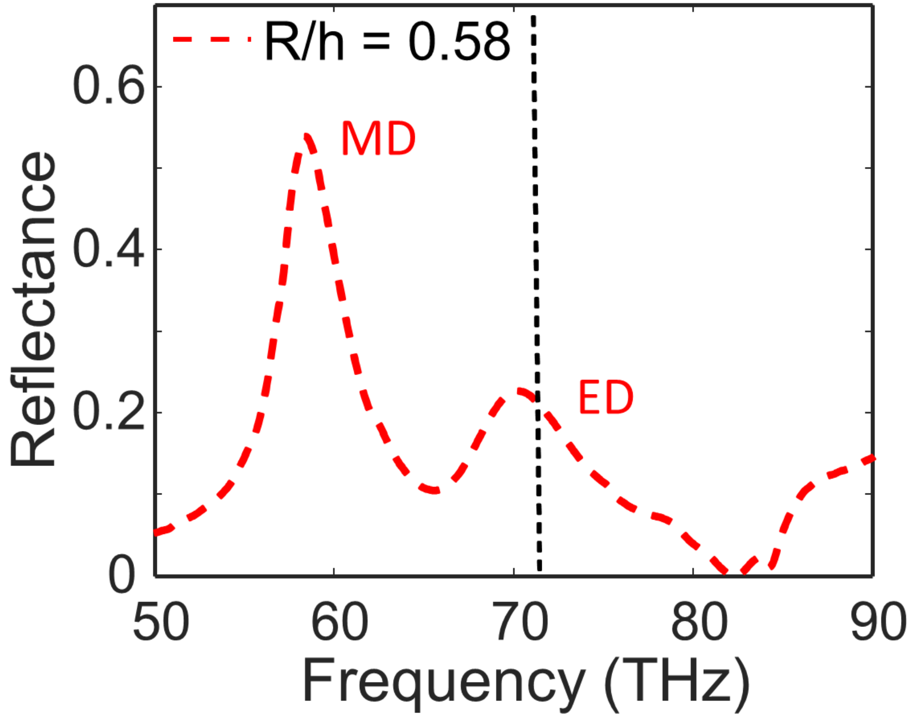
- The strength of light-matter interaction is proportional to number of electrons ($N_e = N_d x N_{QW}$) in the cavity.
- We modify the number of electrons by modifying the doping and number of the quantum wells.

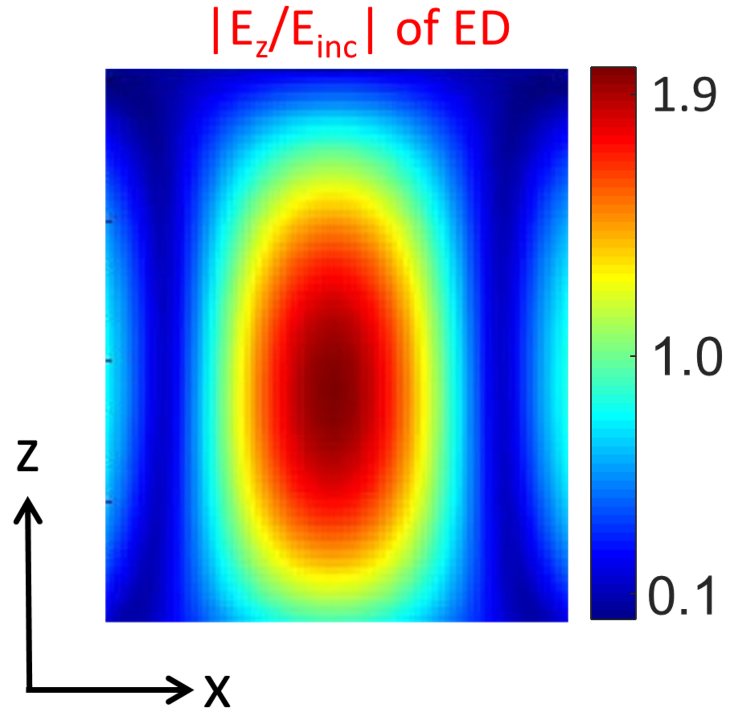
Controlling Light-Matter Interaction by Engineering Heterostructure



Controlling Light-Matter Interaction by Coupling to Different Photonic Modes



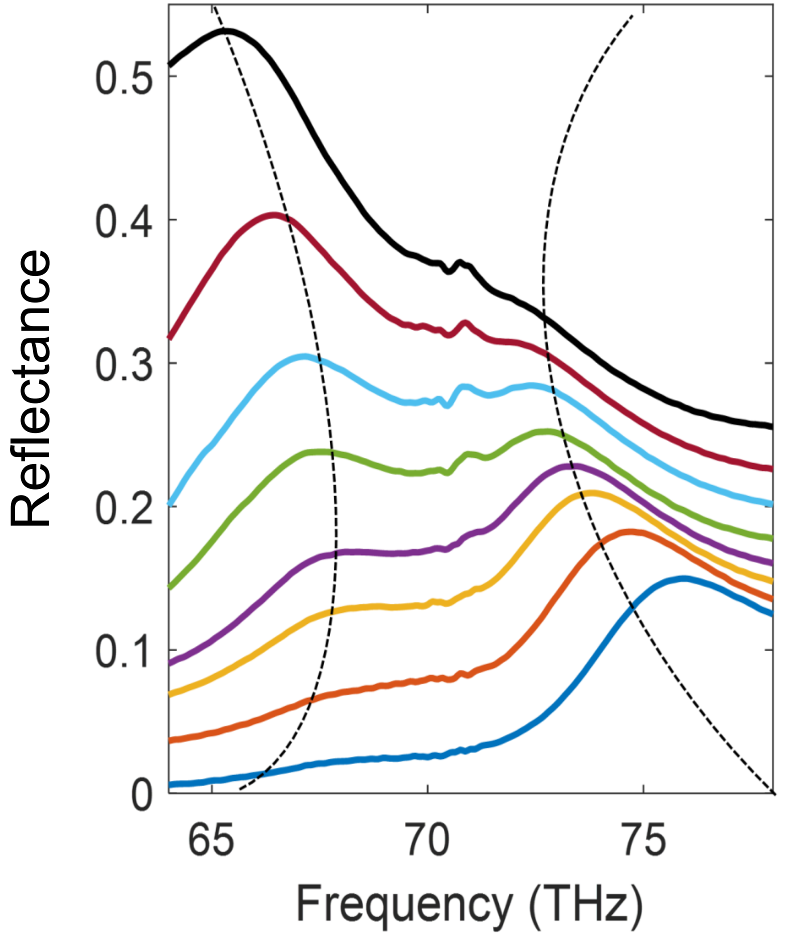




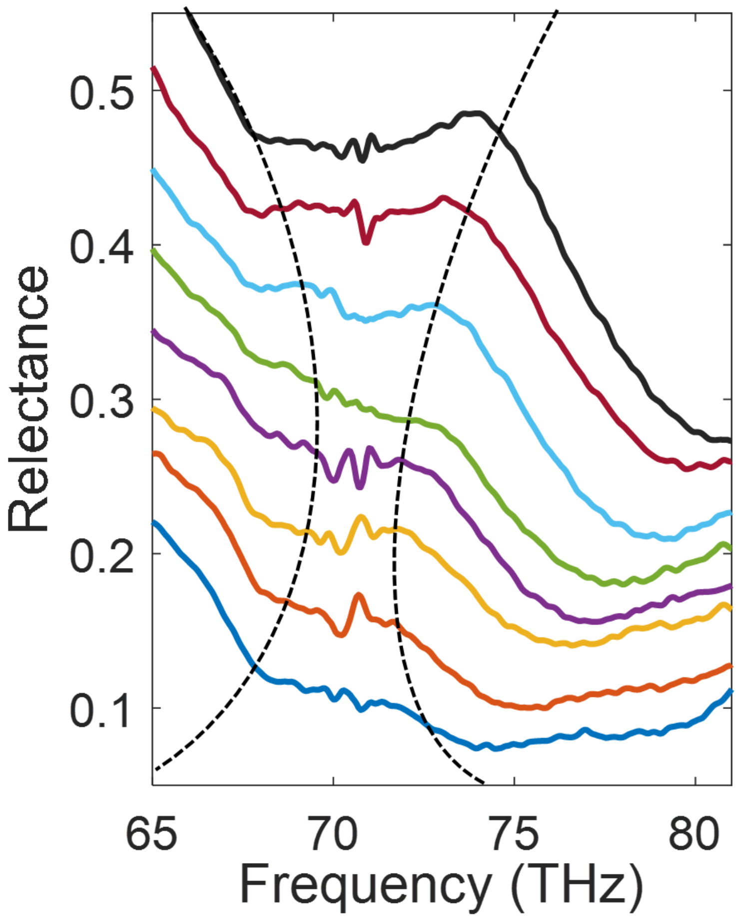
Since different Mie modes have different mode volumes and field enhancements, we can modify light-matter interaction by coupling the IST to different photonic Mie modes (without modifying the heterostructure).

Controlling Light-Matter Interaction by Coupling to Different Photonic Modes

Magnetic Dipole (N_{QW} = 30, N_d = 3e18 cm^{-3})

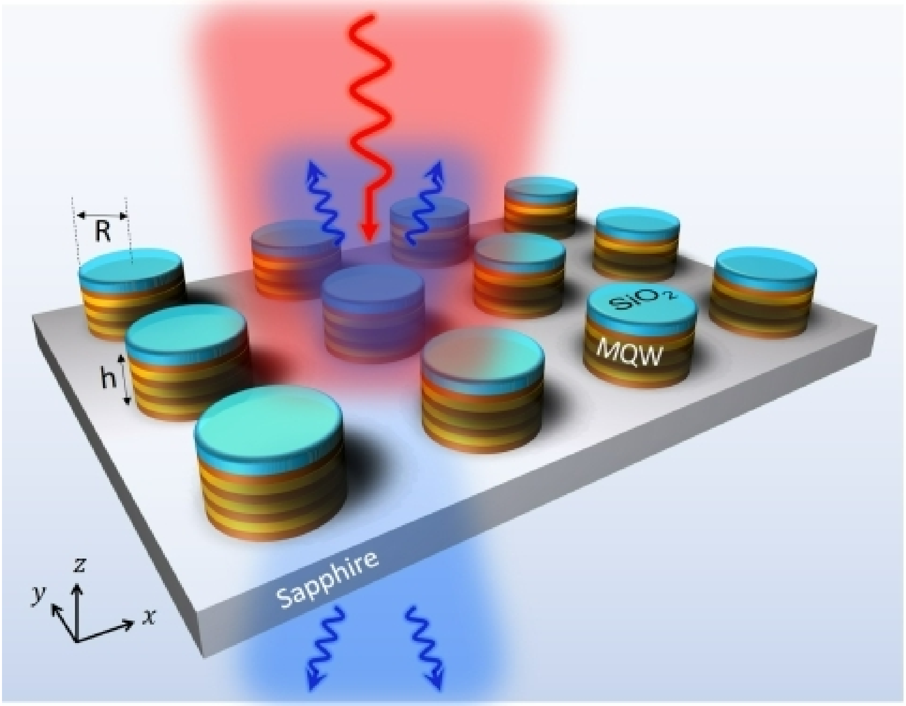


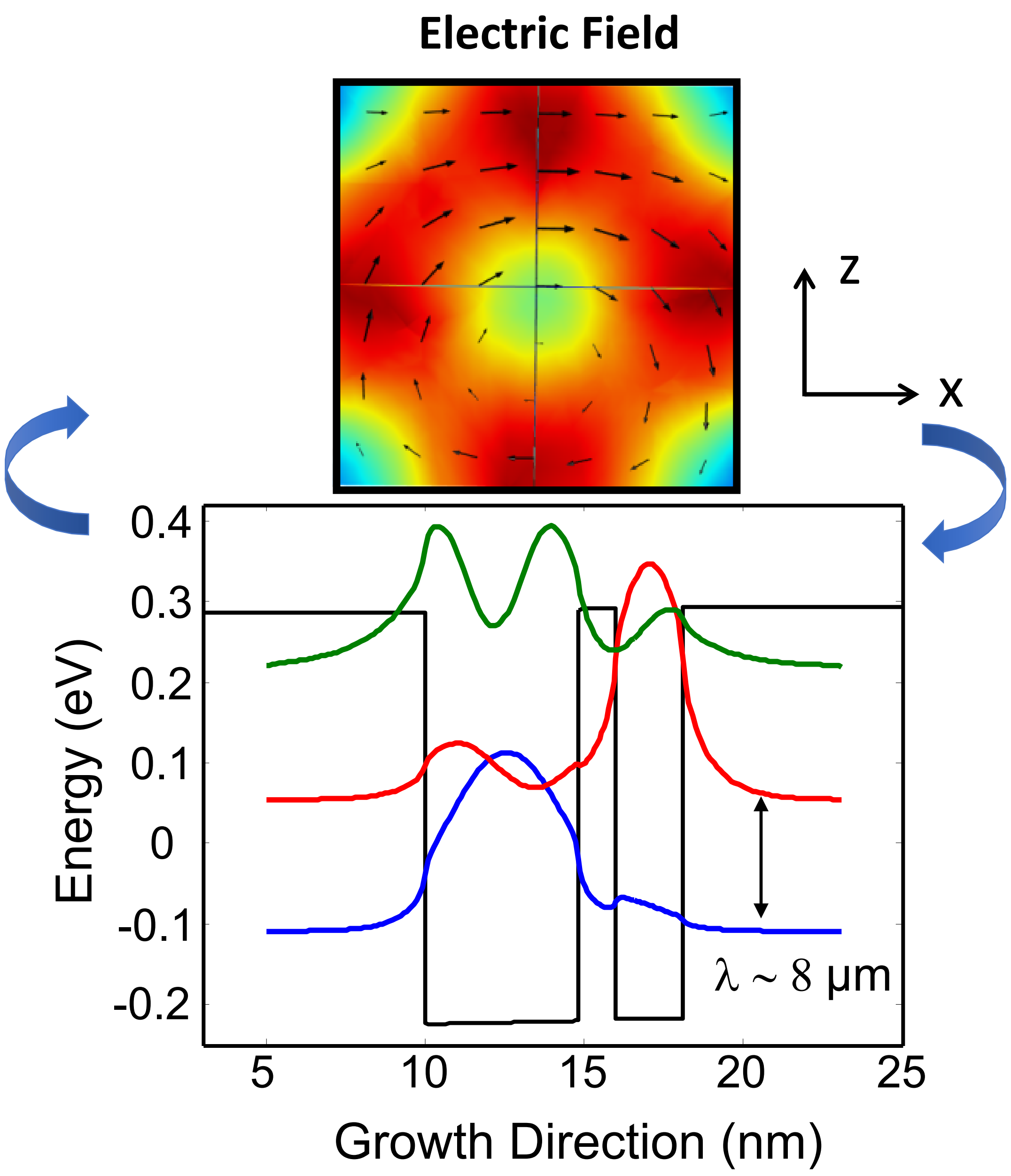
Electric Dipole (N_{QW} = 30, N_d = 3e18 cm^{-3})



The magnetic dipole modes show stronger light-matter interaction !

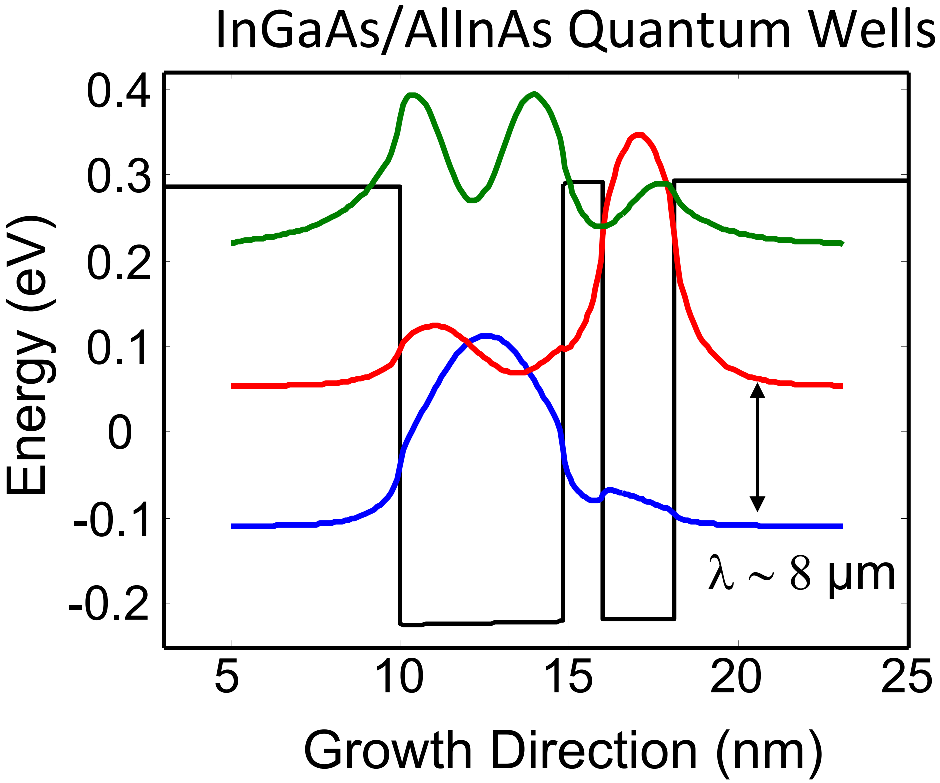
Polaritonic Metasurface : Extreme Nonlinearities

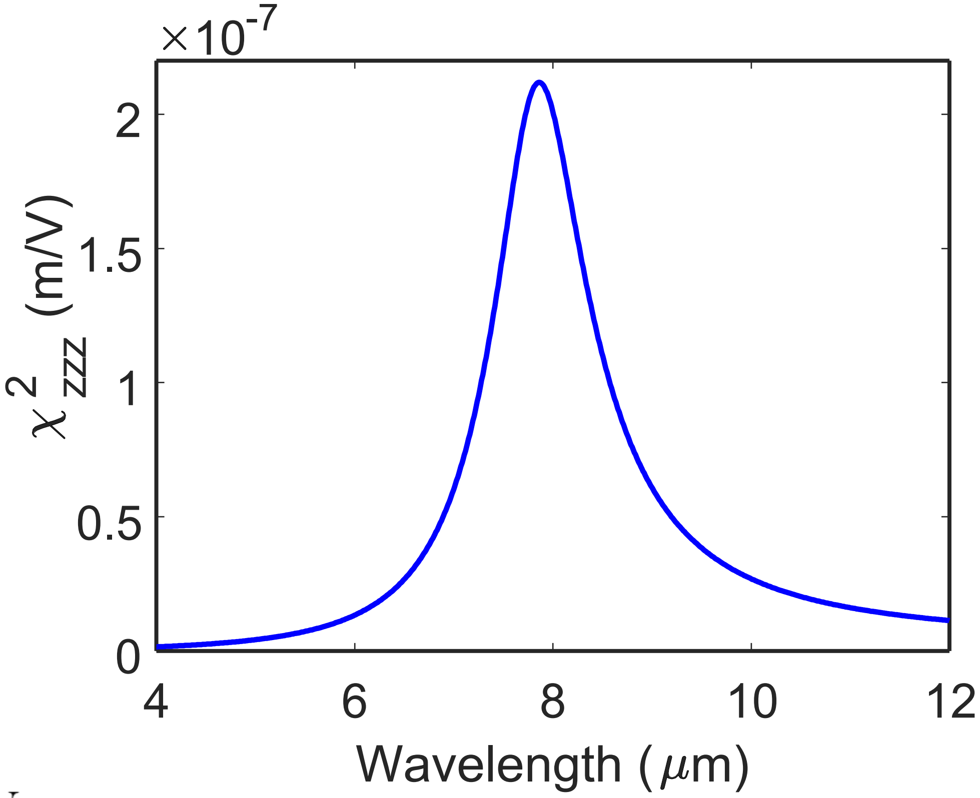




- Light-matter coupling between a MD Mie mode and intersubband electronic excitations.
- MD mode has strong z electric field components, allows for normal incidence, and smallest size of the resonator.

Resonant χ$^{(2)}$ using Intersubband Transitions





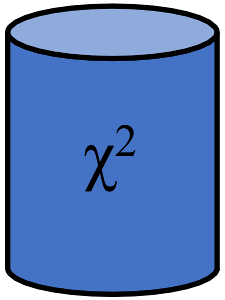
$$\chi^{(2)}(\omega) \propto \frac{N \cdot z_{12} z_{23} z_{13}}{(\omega - \omega_{12} - i\Gamma_{12})(2\omega - \omega_{13} - i\Gamma_{13})}$$

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

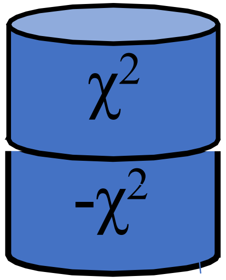
IEEE J. Quantum Electr. 30, 1313 (1994)

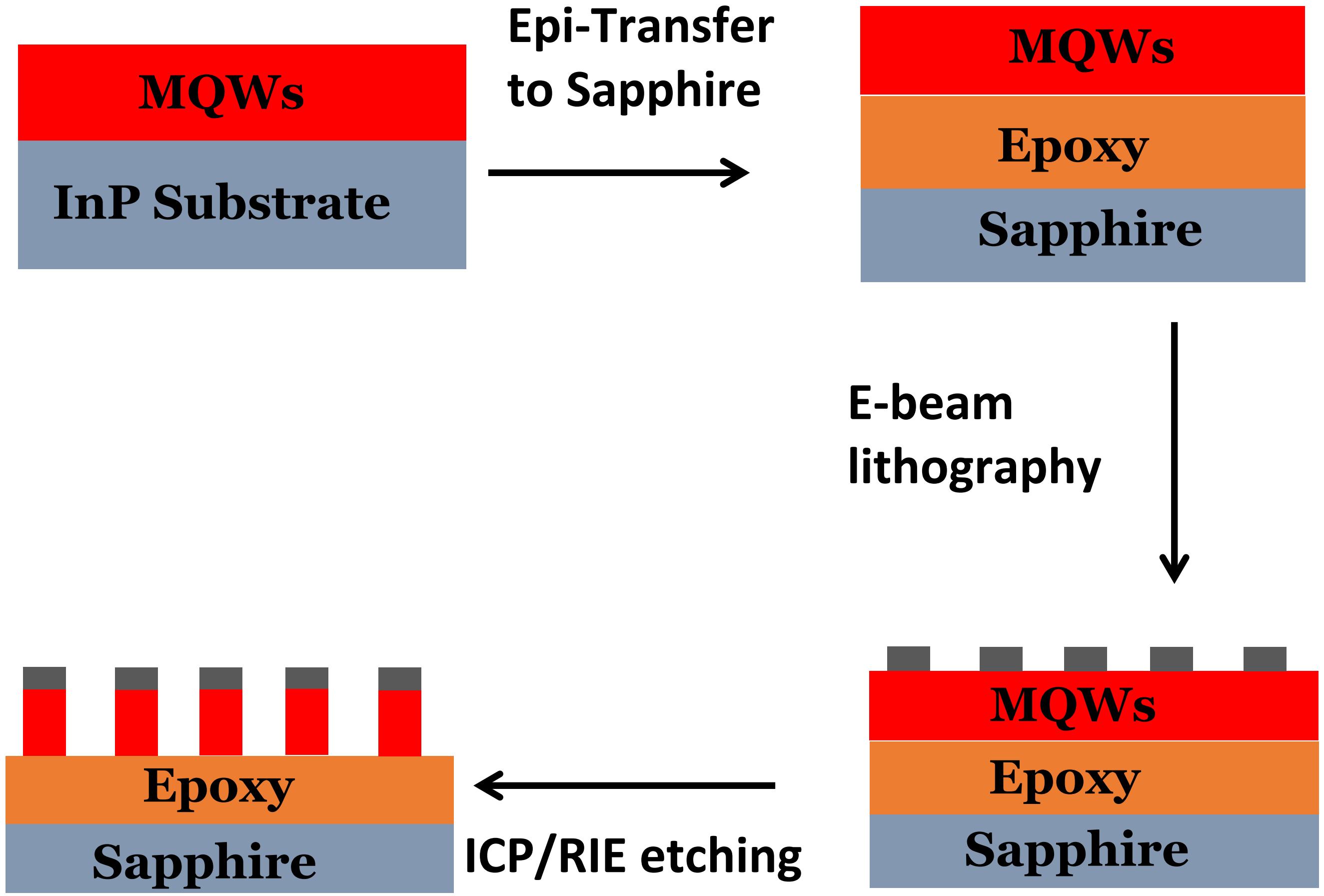
Fabrication of the All-Dielectric Nonlinear Metasurface

(a)



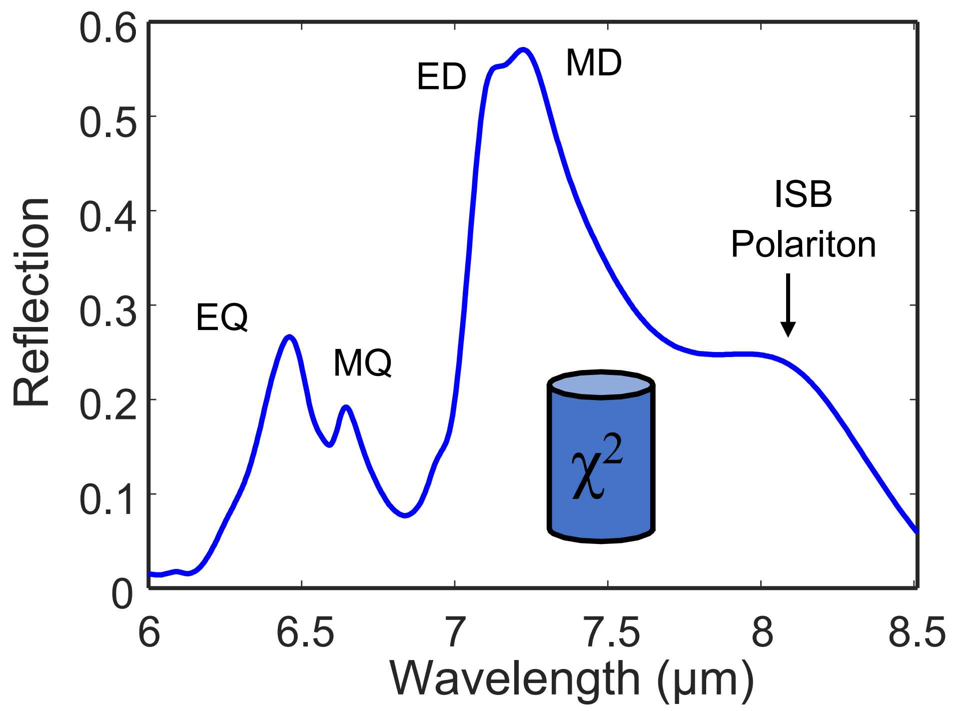
(b)



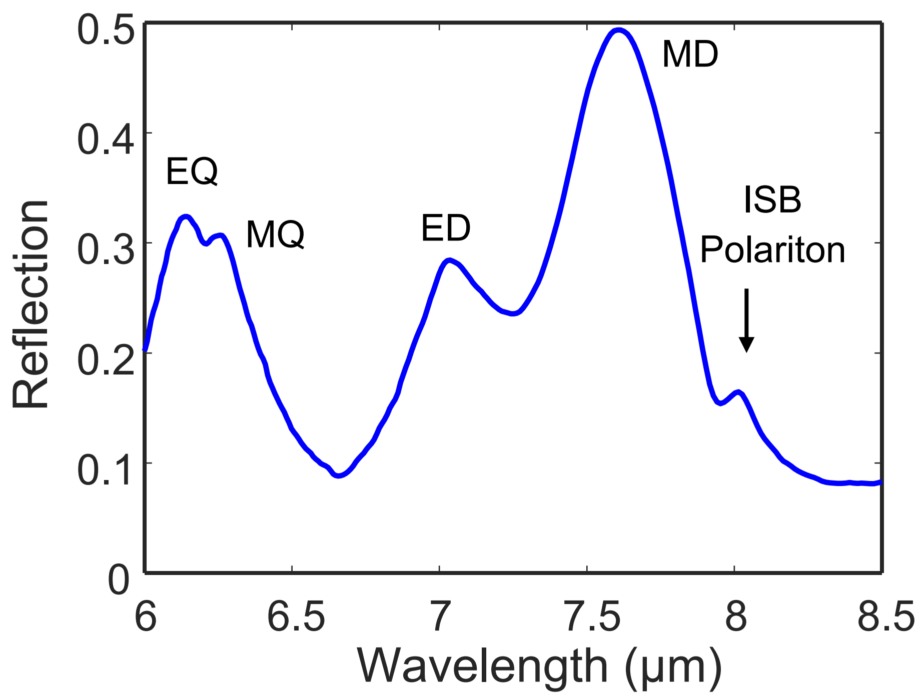


Linear Reflectance Spectra : Simulation and Experiment

Simulation



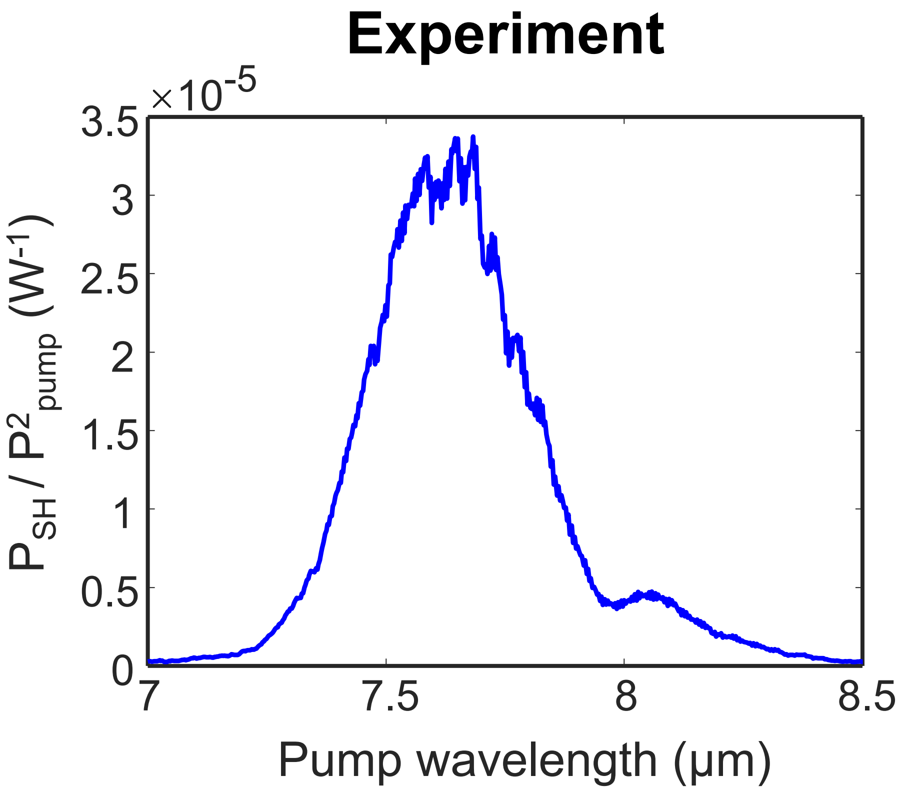
Experiment

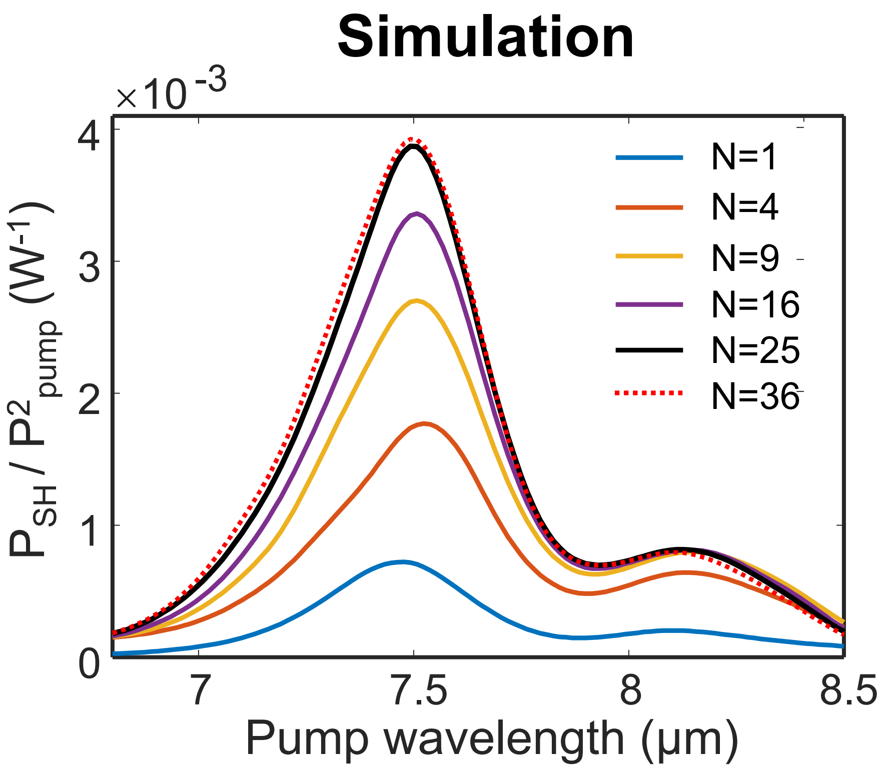


MD

Splitting of Photonic Resonance due to Strong Coupling !

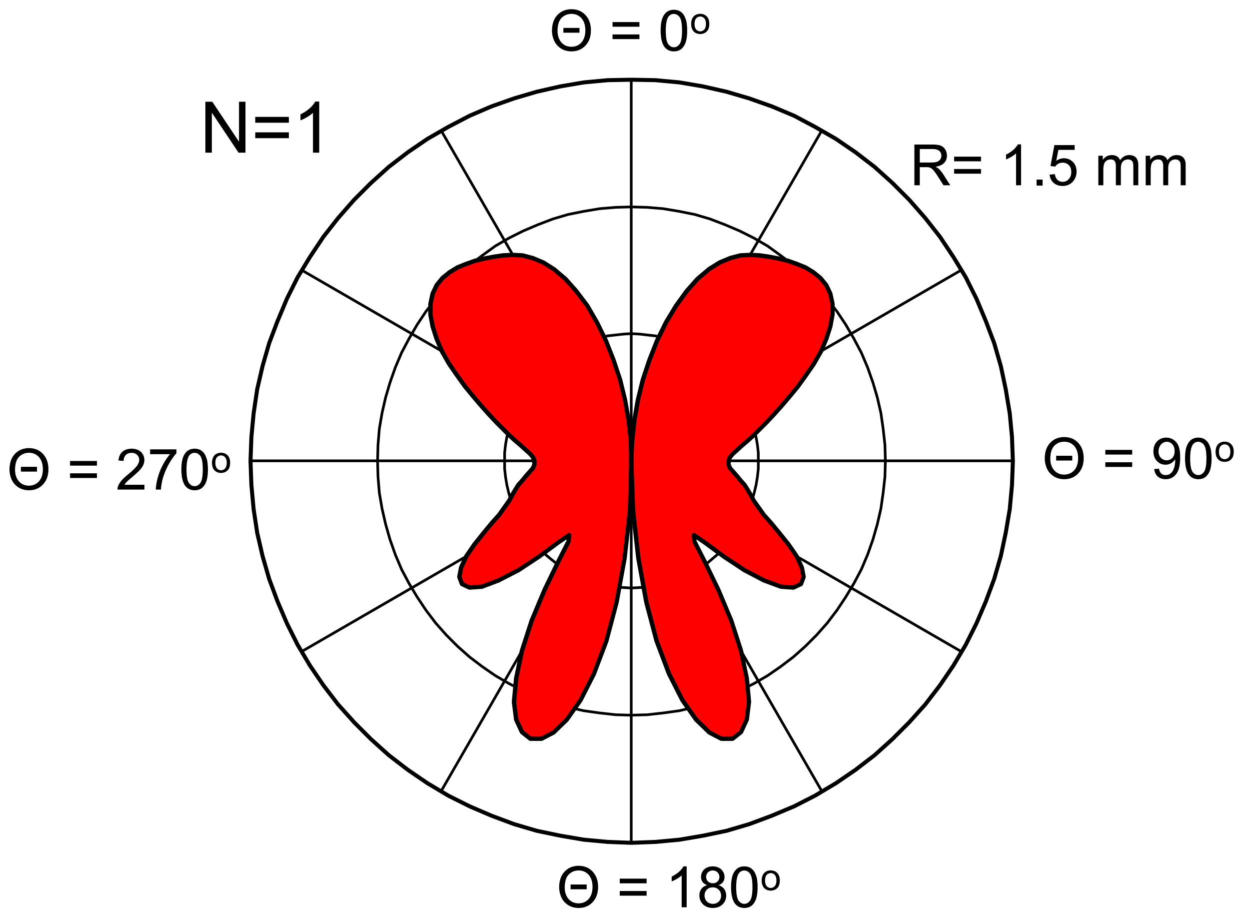
Second-Harmonic Generation : Experiment and Simulations

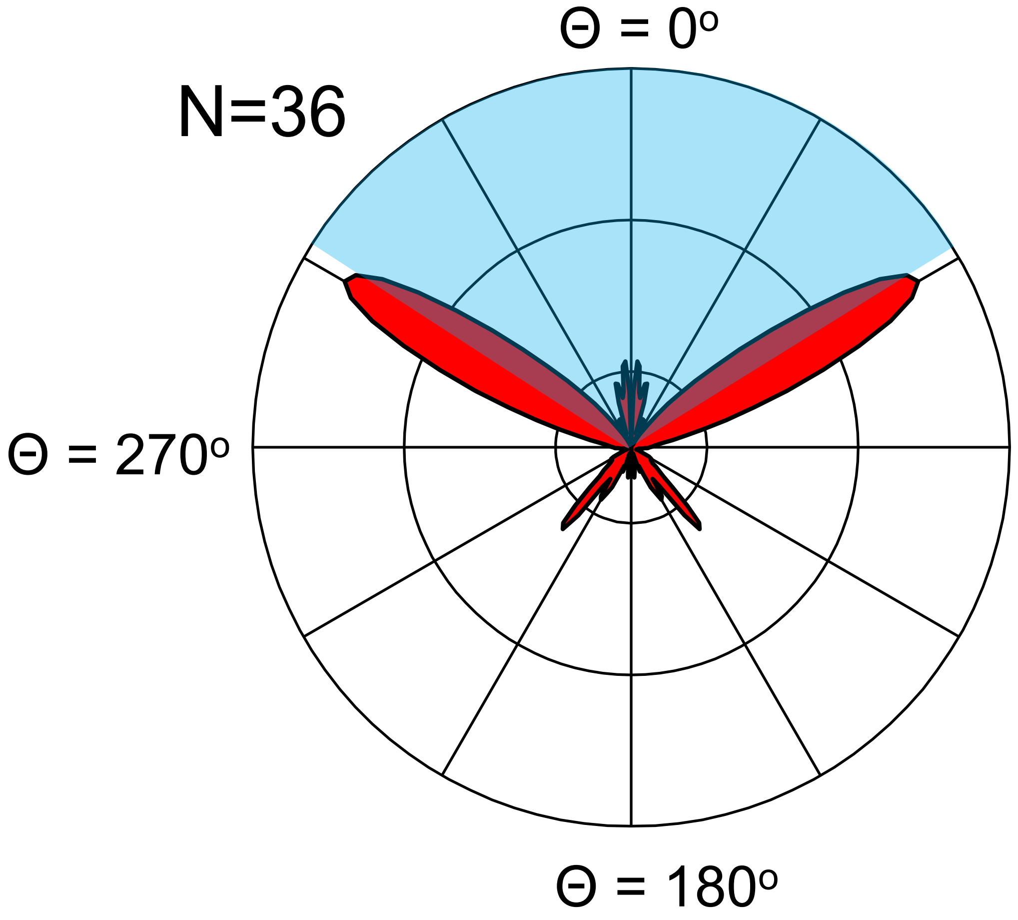




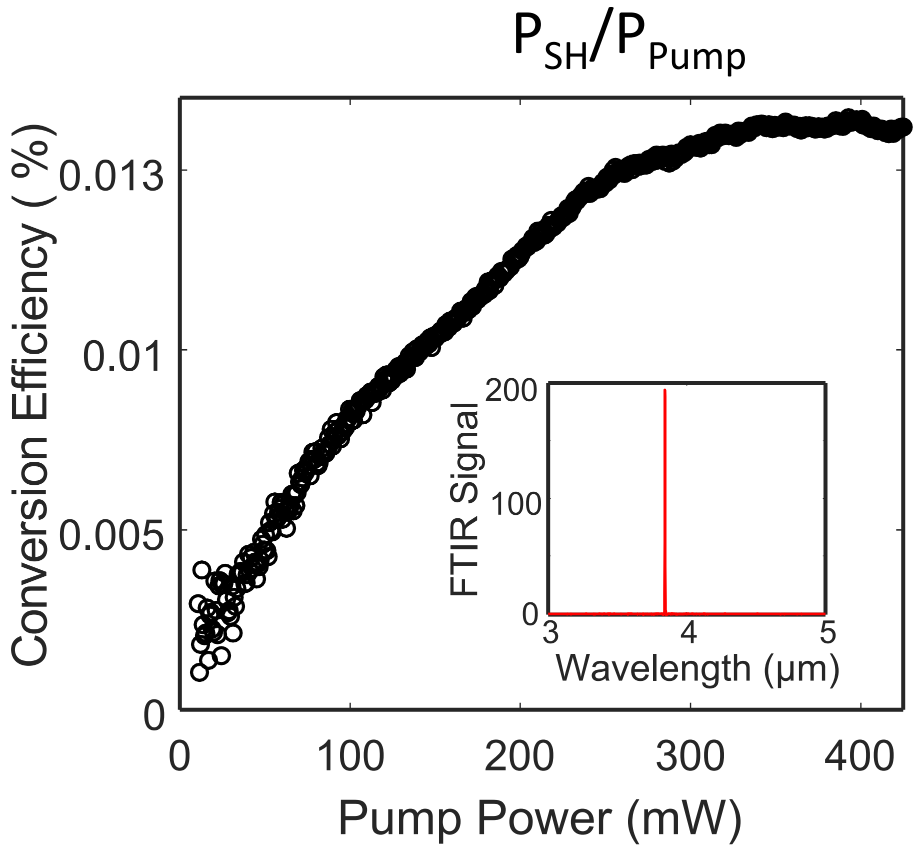
Excellent qualitative areement between experiments and simulations can be seen !

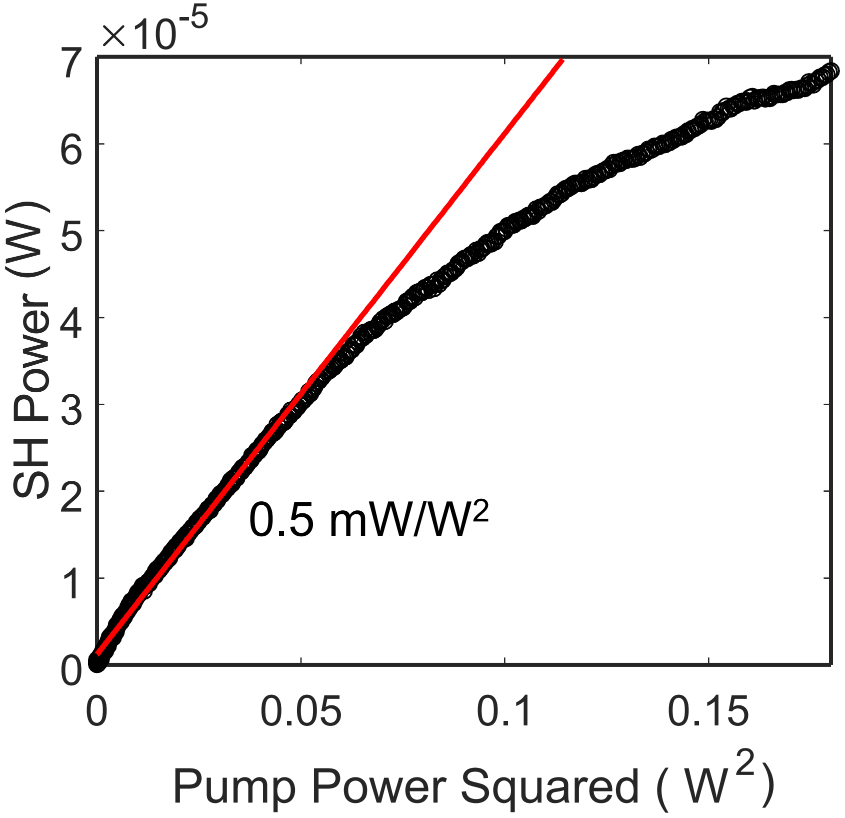
Limited Collection Efficiency due to Diffraction





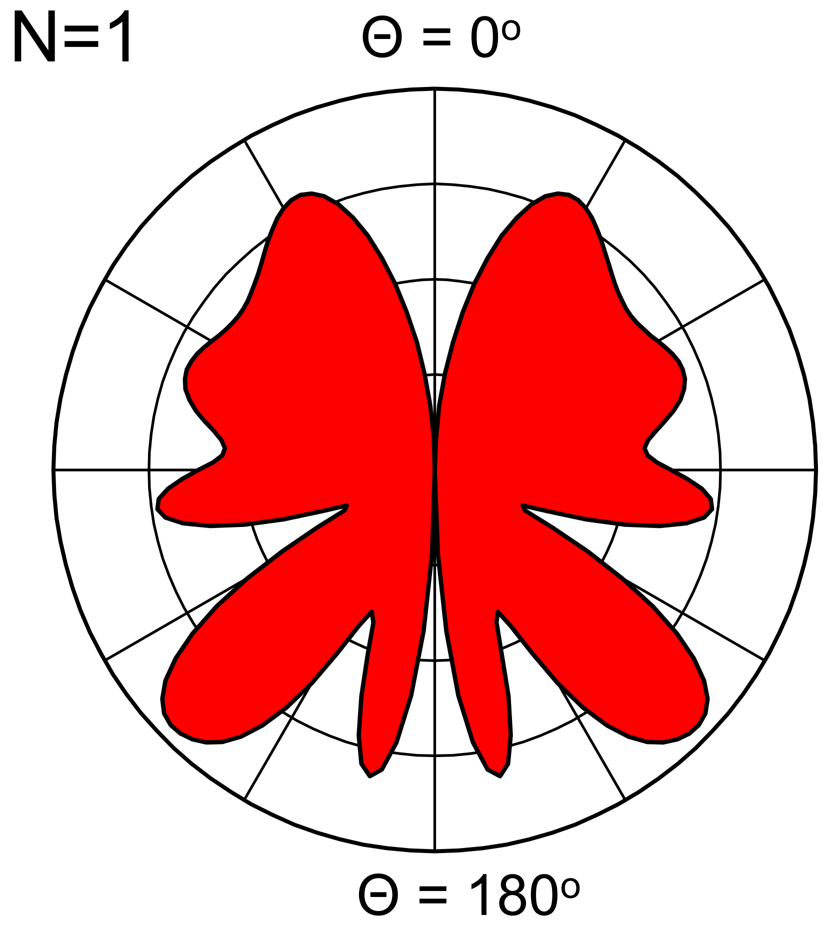
Experimentally Measured SHG Efficiencies

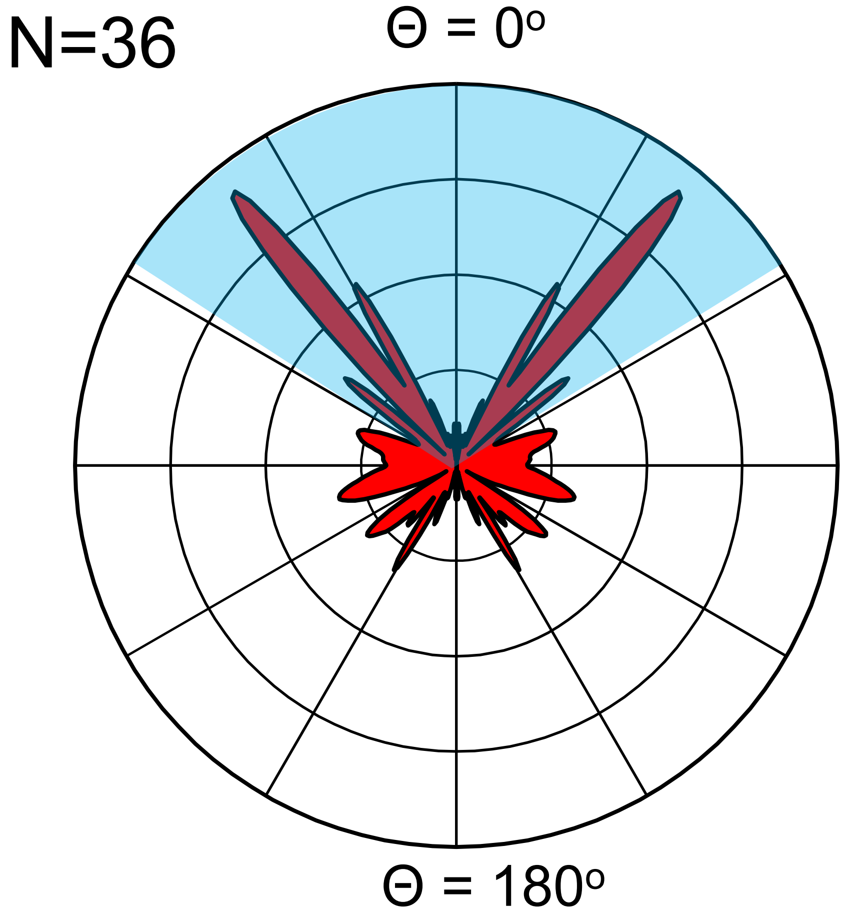




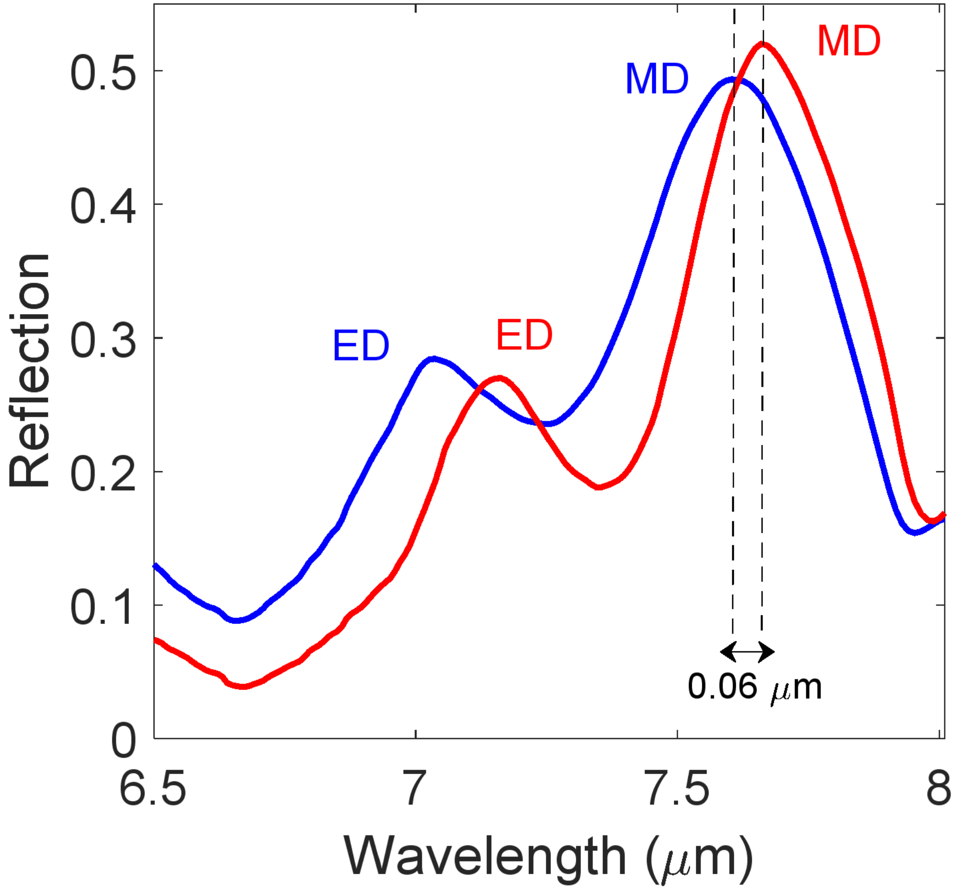
Large magnitude of $\chi^{(2)}$ gives SHG efficiency ~ 0.013 % at 11 kW/cm^2 and conversion factor ~ 0.5 mW/W^2.

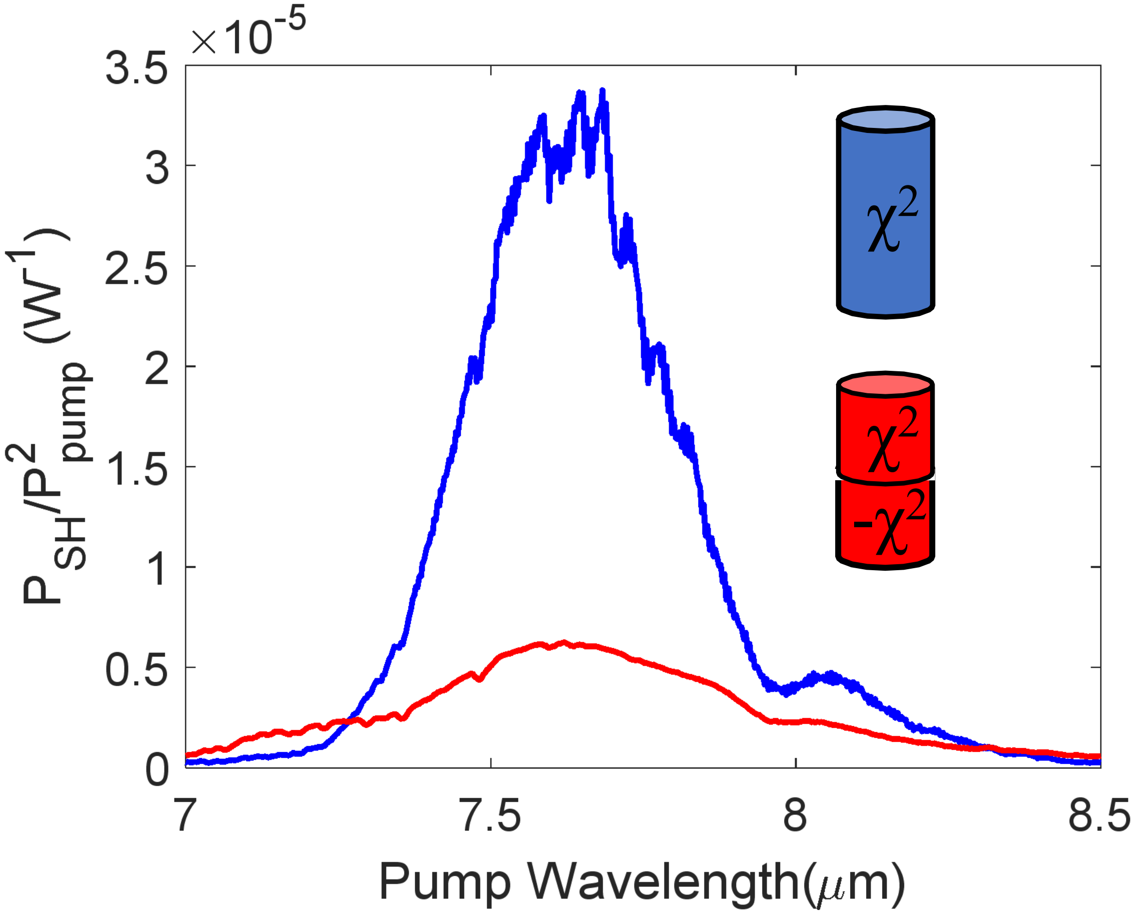
Enhancing Collection Efficiency by Varying Radius & Period





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).