



Metasurfaces and Epsilon-Near-Zero Modes in Semiconductors

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Outline

- **What are Epsilon Near Zero modes, what can we do with them?**
 - Fundamentals
 - ENZ modes: coupling to metamaterial resonators
 - Berreman modes: Thermal emission
- **Other examples and uses of Epsilon Near Zero modes**
 - ENZ modes coupled to Intersubband transitions and Metamaterials
 - Enhanced Harmonic Generation

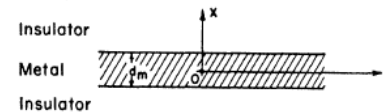


Surface Plasmons in Thin Films

E. N. ECONOMOU*†

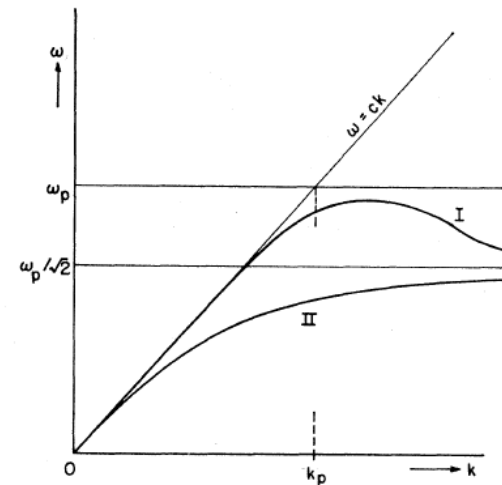
The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

(Received 15 January 1969)



Economou, 1969
Burke et al, 1986
etc

“Thin” here: $\sim 20\text{nm}$ for $\sim 750\text{THz}$
plasma frequency

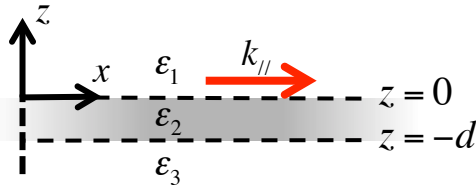


Long range SP

Short range SP

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).

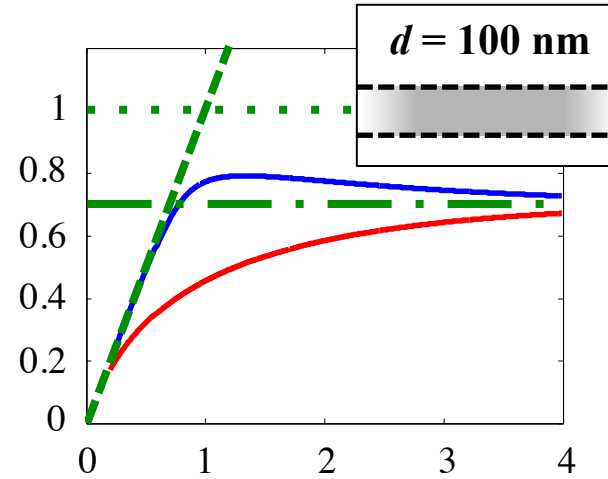
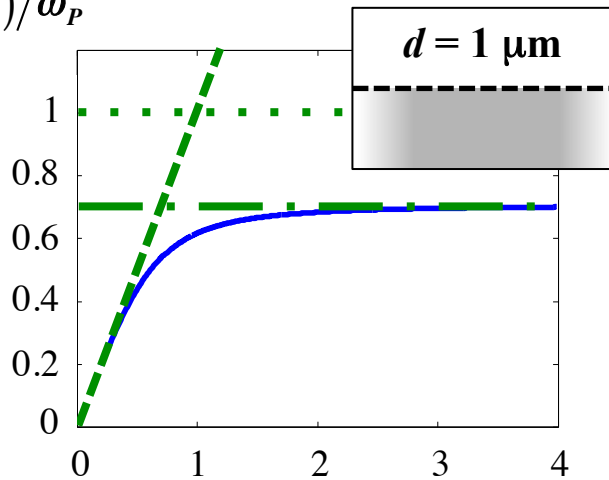
**What happens when the layer becomes much thinner?
($\ll$ skin depth)**



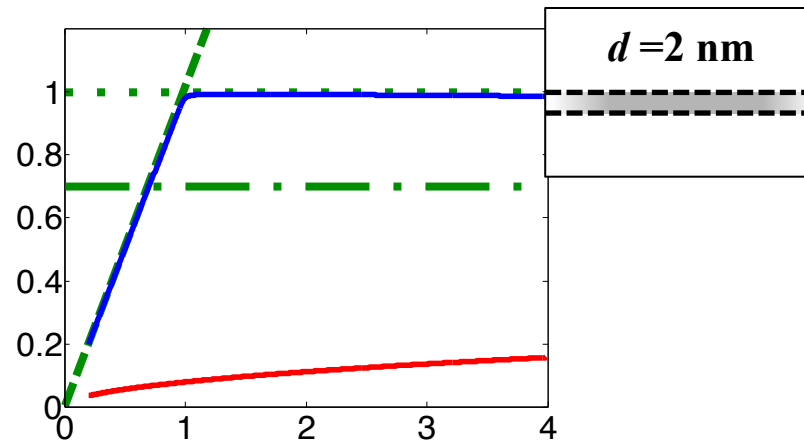
$$1 + \frac{\varepsilon_1 k_{z3}}{\varepsilon_3 k_{z1}} = i \tan(k_{z2} d) \left(\frac{\varepsilon_2 k_{z3}}{\varepsilon_3 k_{z2}} + \frac{\varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1}} \right)$$

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

$\text{Re}(\omega)/\omega_p$



$k_{||} \times c/\omega_p$



“ENZ Mode”

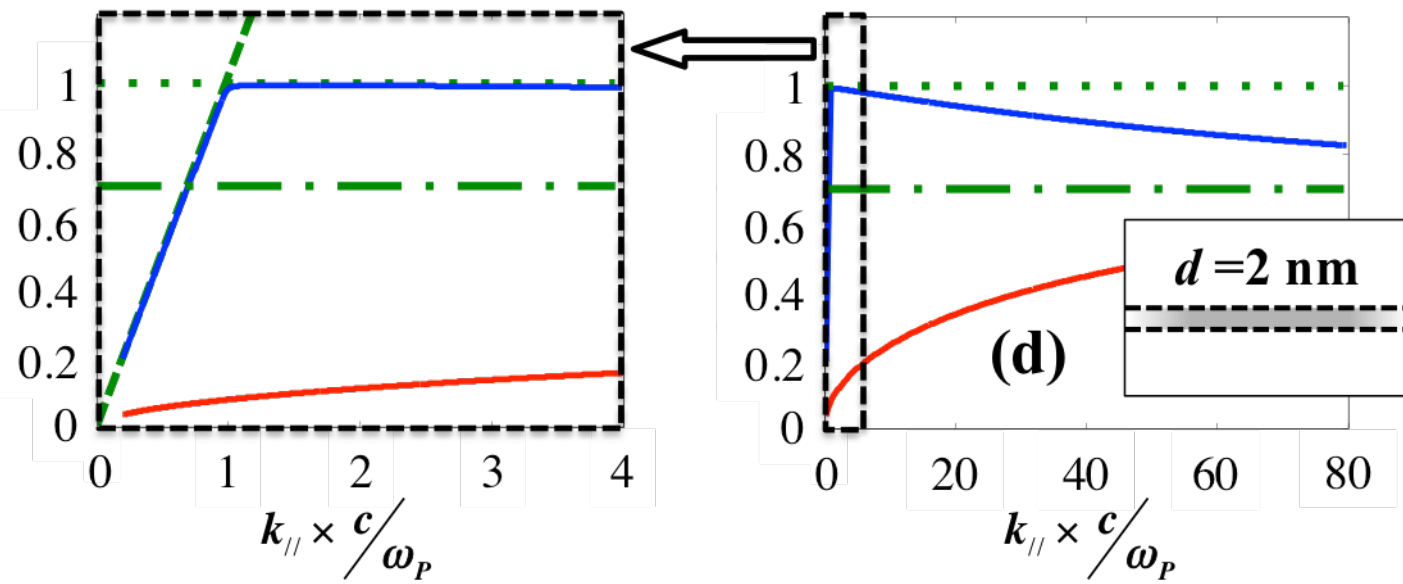
S. Vassant et al., Phys. Rev. Lett.
109, 237401 (2012)
S. Vassant et al., Opt. Express
20, 23971 (2012)

Flat dispersion?



ENZ Dispersion

Flat?...





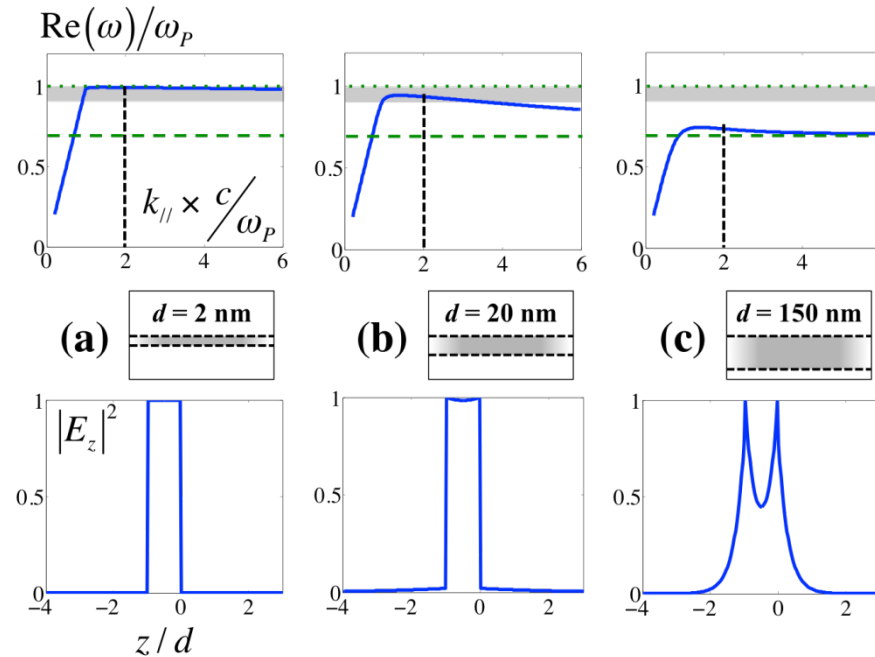
ENZ Modes: E_z is Constant and Large



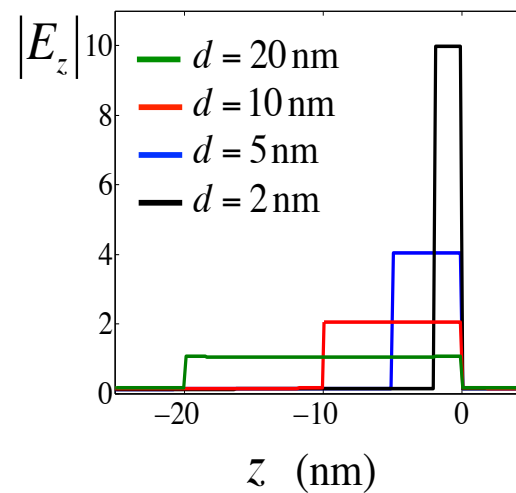
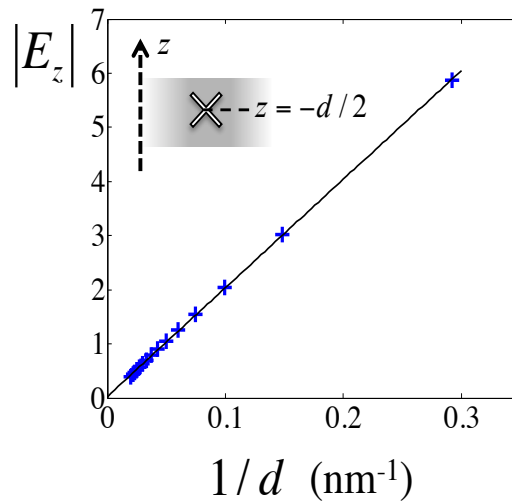
Salvo Campione



Prof. Francois Marquier



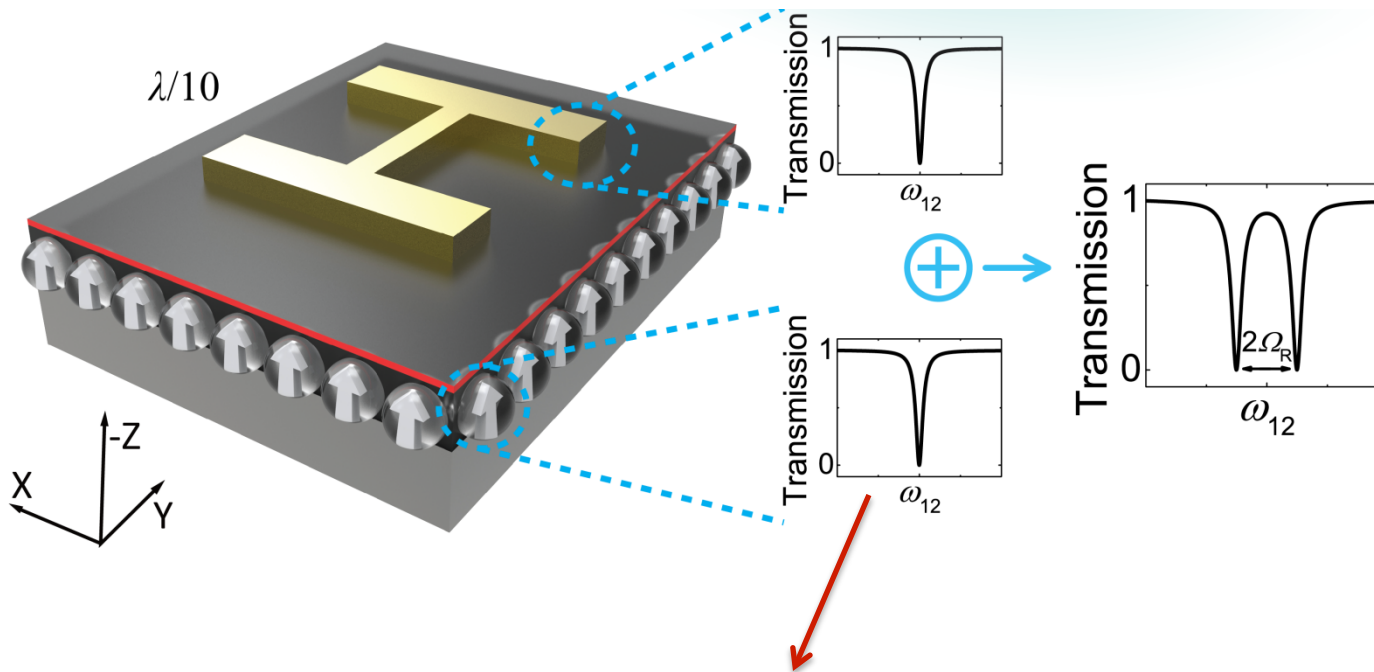
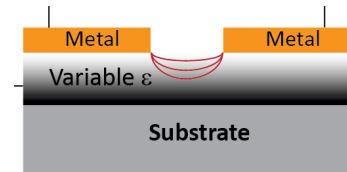
$$E_z \propto \frac{1}{d}$$





What Can We Do With ENZ Modes: Coupling to Planar Metamaterials

MM resonators create strong optical fields that lead to strong coupling

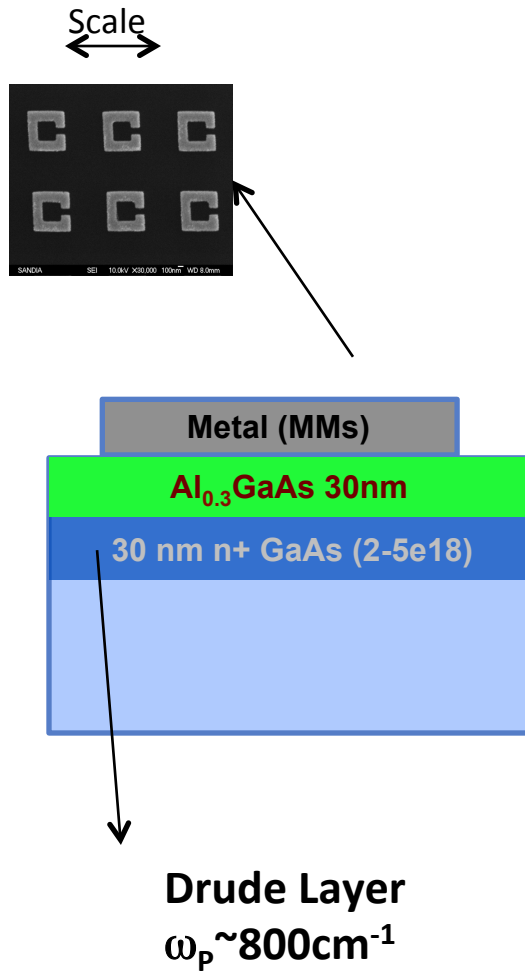


Optical Phonons: Nano Letters 11, 2104 (2011)
Intersubband Transitions: Nature Communications 4, (2013)
Epsilon Near Zero modes: Nano Letters 13, 5391 (2013)

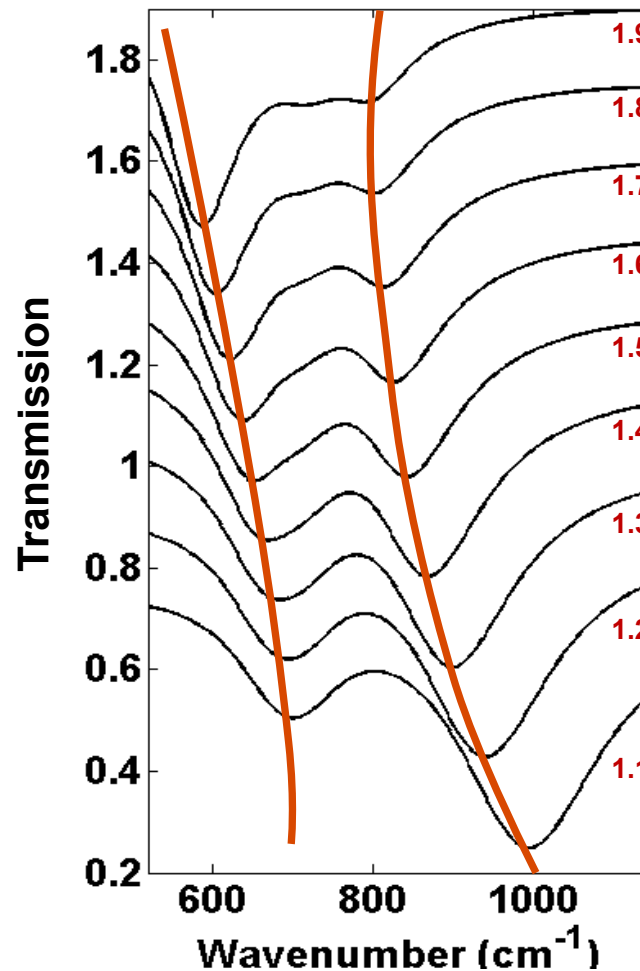


Young Chul Jun

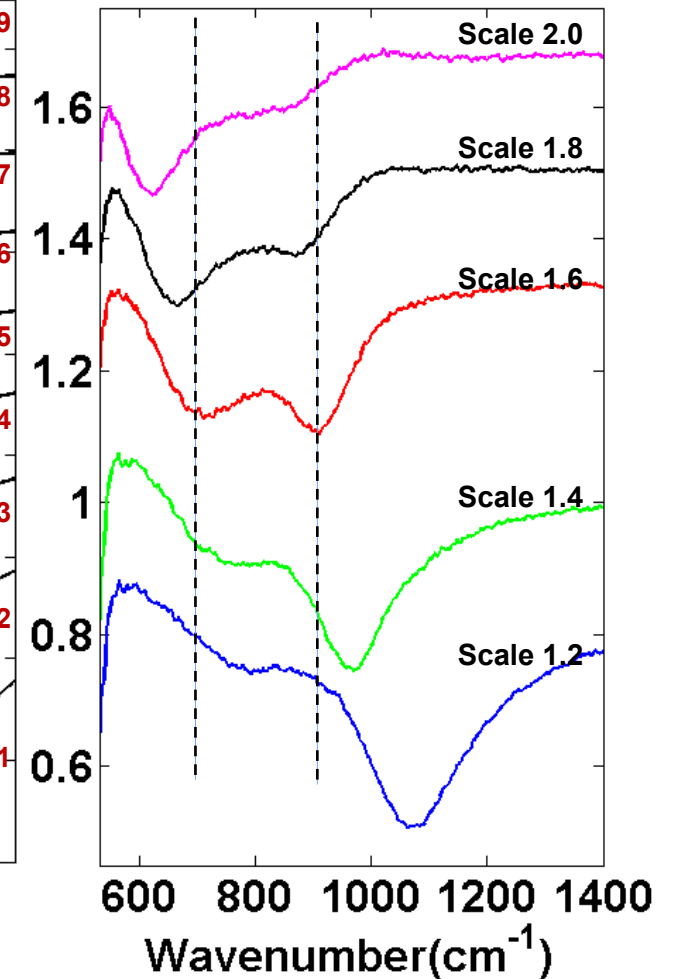
Strong Coupling to ENZ Modes



Numerical simulation (FDTD)

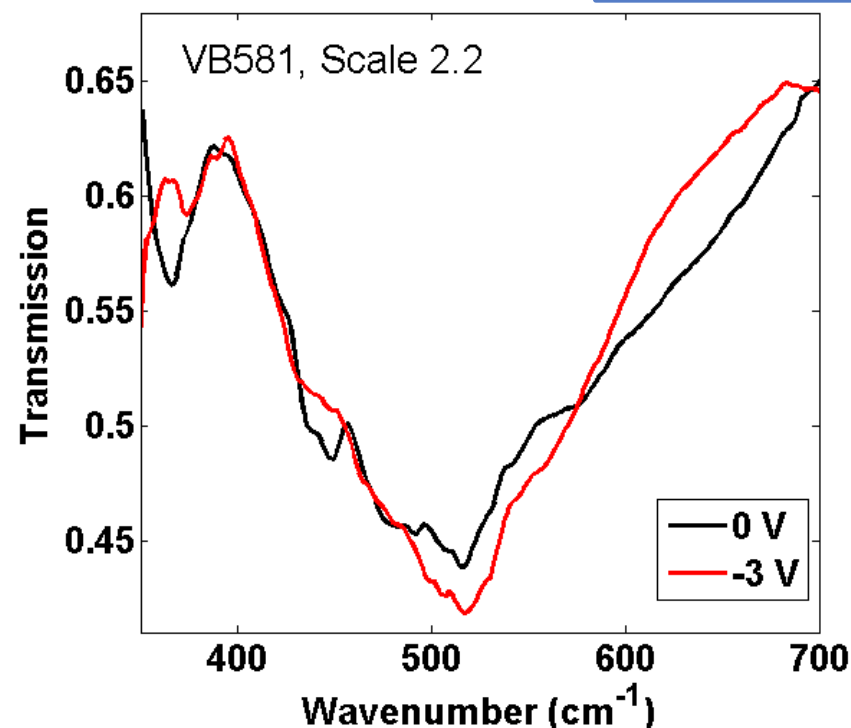
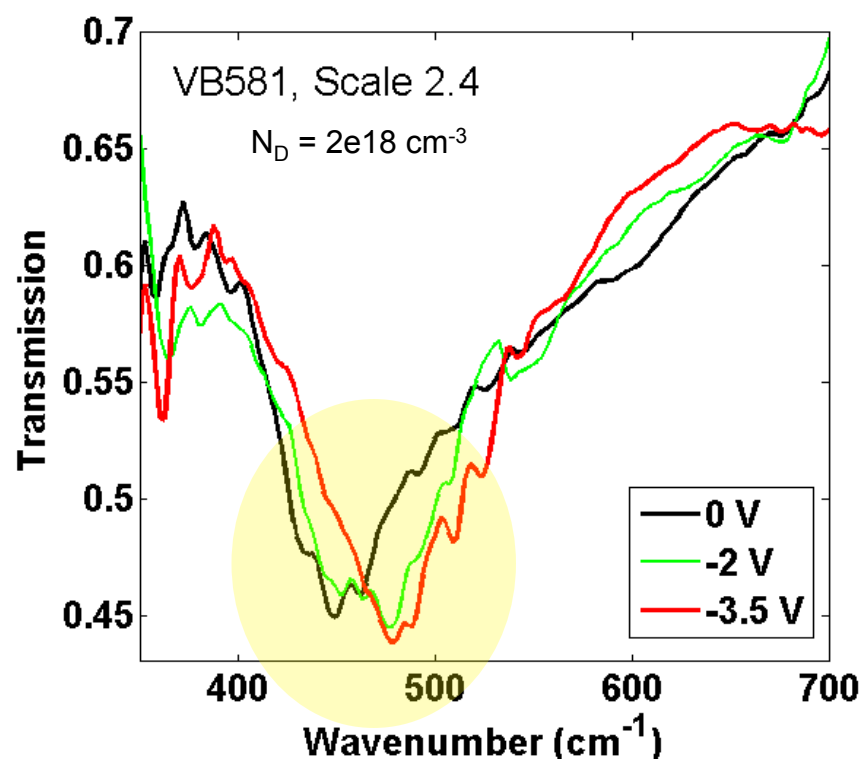
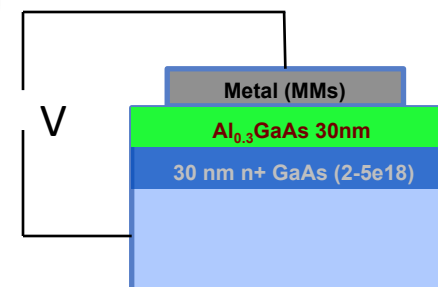


FTIR transmission measurement





Electrically Tuning the Coupling to the ENZ Mode



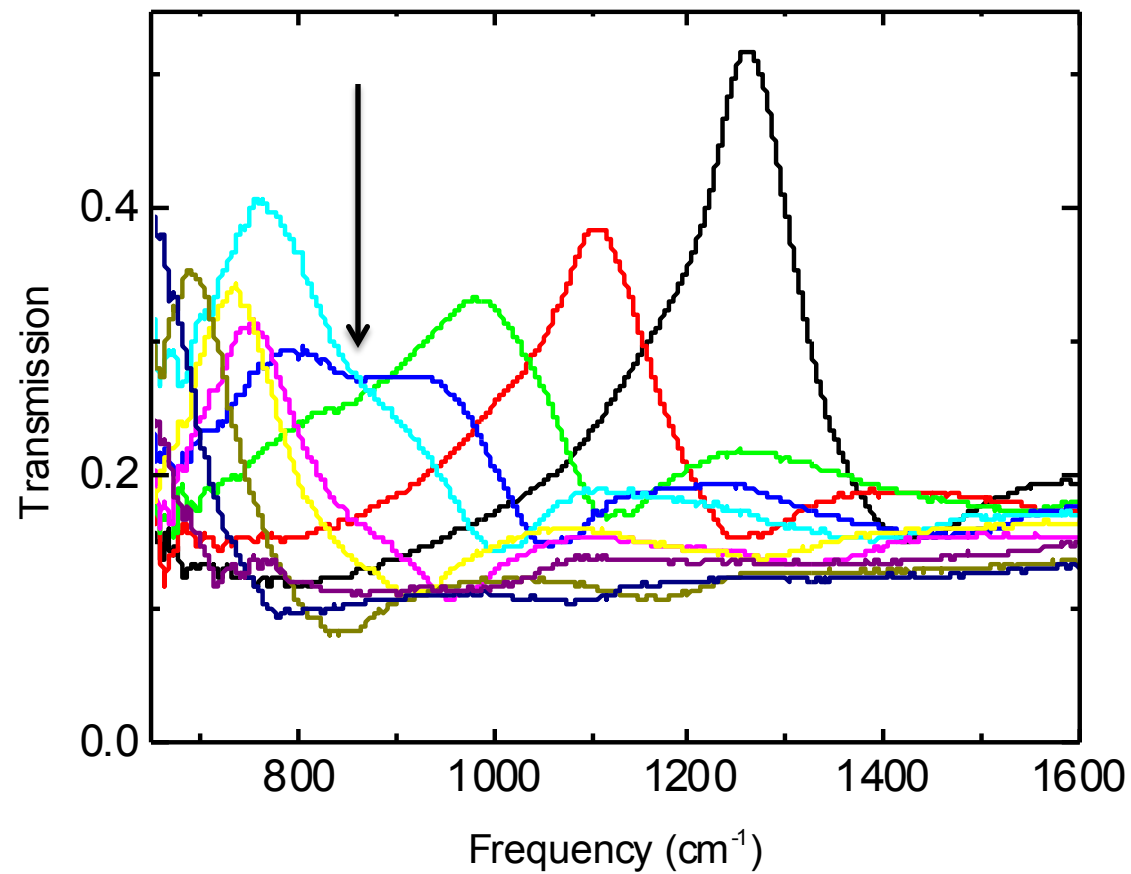
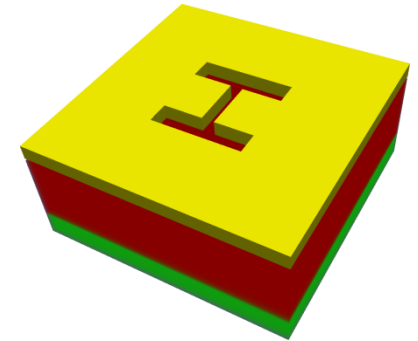
***Fundamentally different than tuning just by changing a local permittivity!
Removal of carriers -> removal of ENZ mode***



Alex Benz

Inverted Dogbone Resonator + ENZ Mode

Tunable Passband Filter

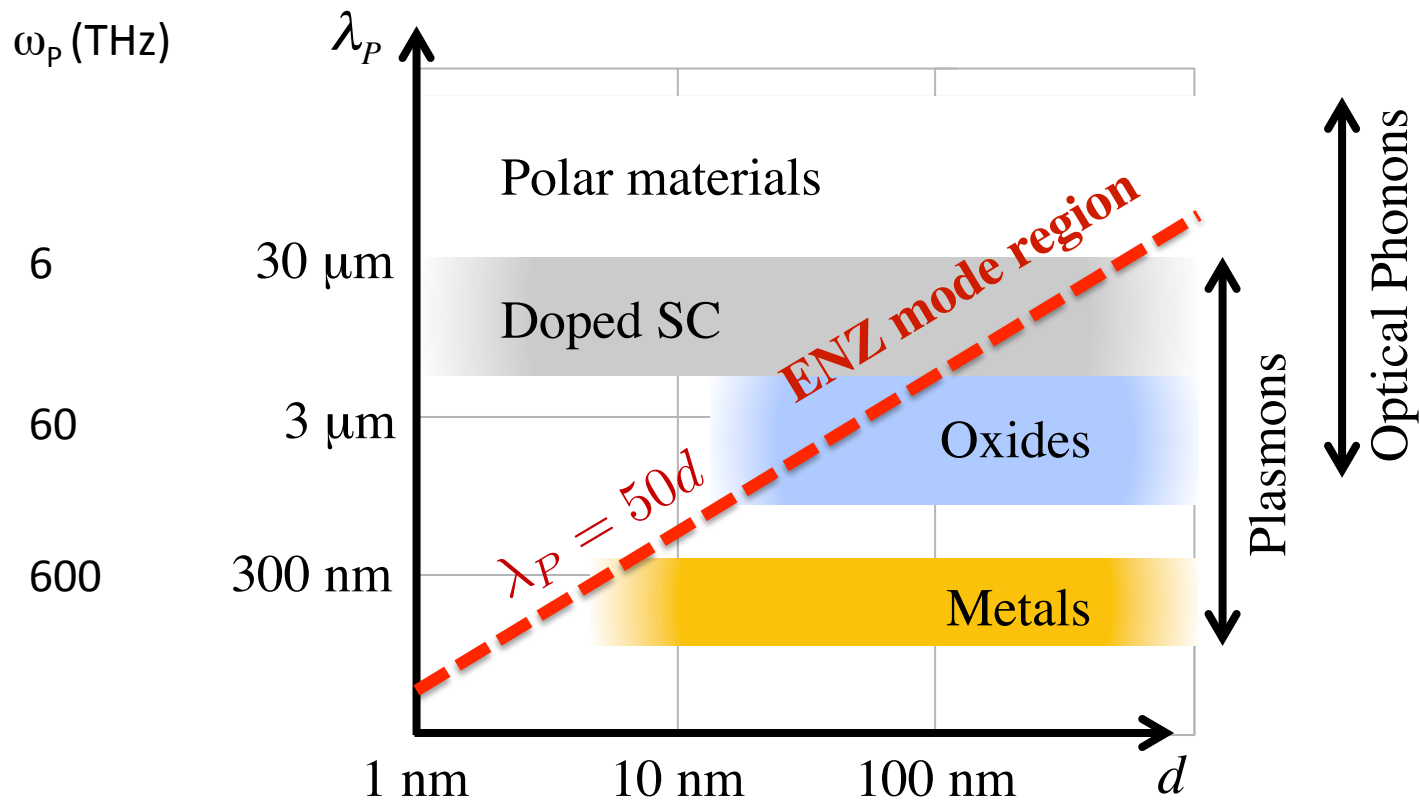




Where Can We Find ENZ Modes?

Close to $\omega \approx \omega_P$ $\omega \approx \omega_P \left[1 - \frac{k_{||} d}{4} \right] - i \frac{\gamma}{2}$

Condition for ENZ modes $\frac{\omega_P}{c} < k_{||} < \frac{4}{d}$



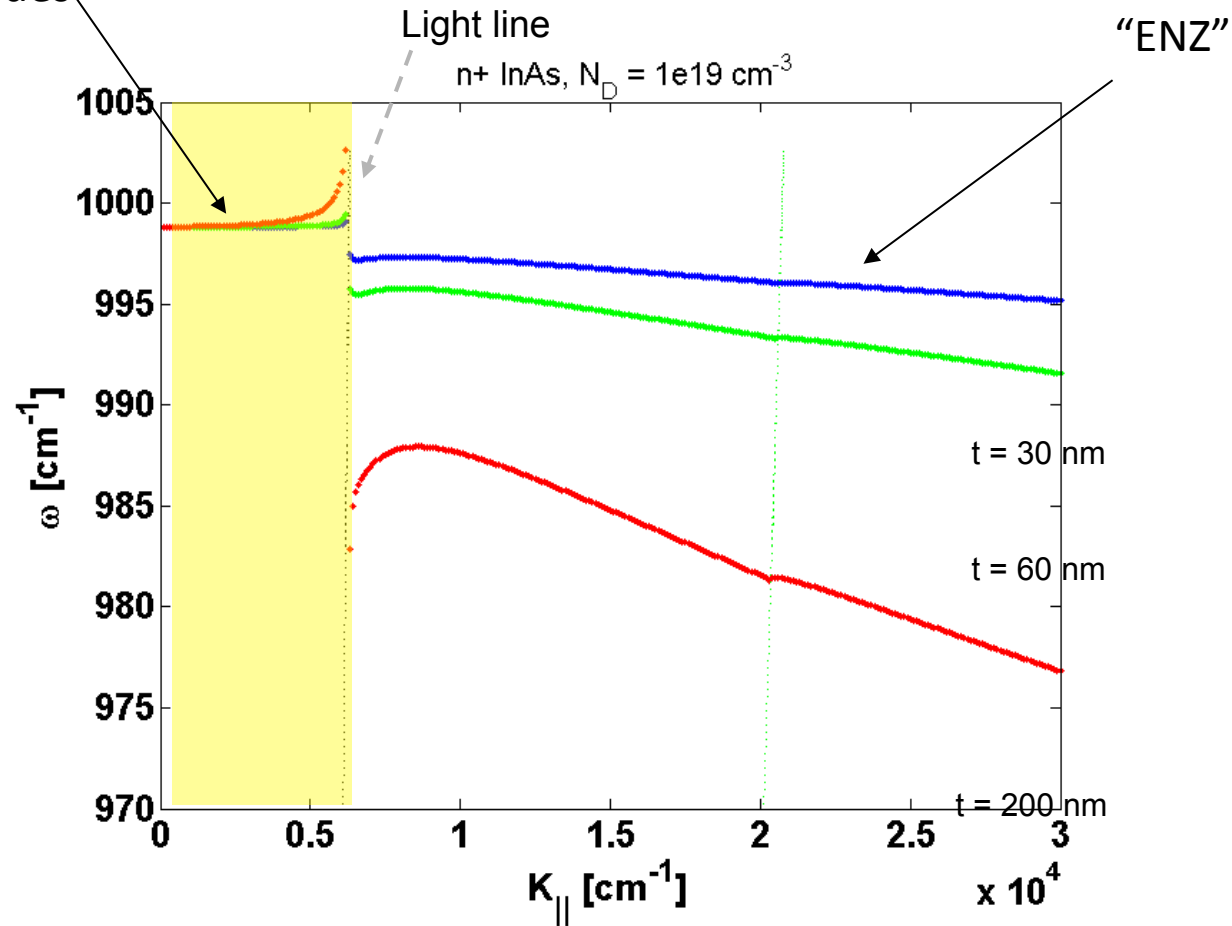


Young Chul Jun

Related to ENZ: Berreman Modes

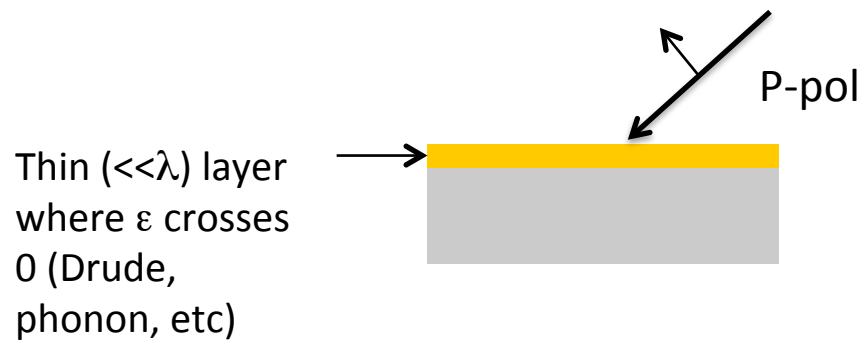
Leaky modes: can couple from free space

“Berreman
Modes”

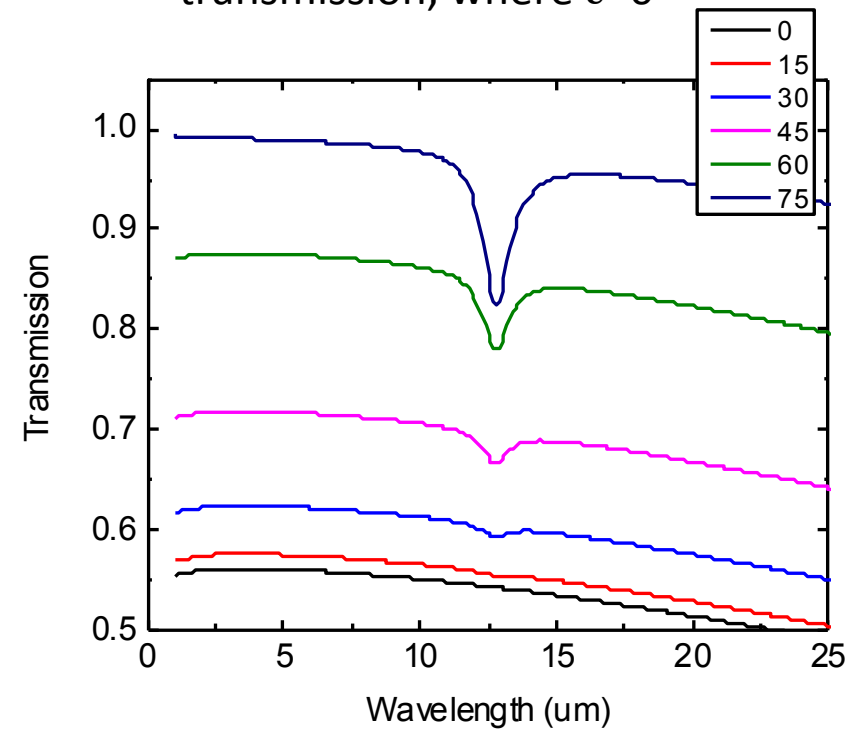




Coupling to Berreman Modes From Free Space



Transmission vs angle (p-pol):
A sharp dip is observed in transmission, where $\epsilon \sim 0$

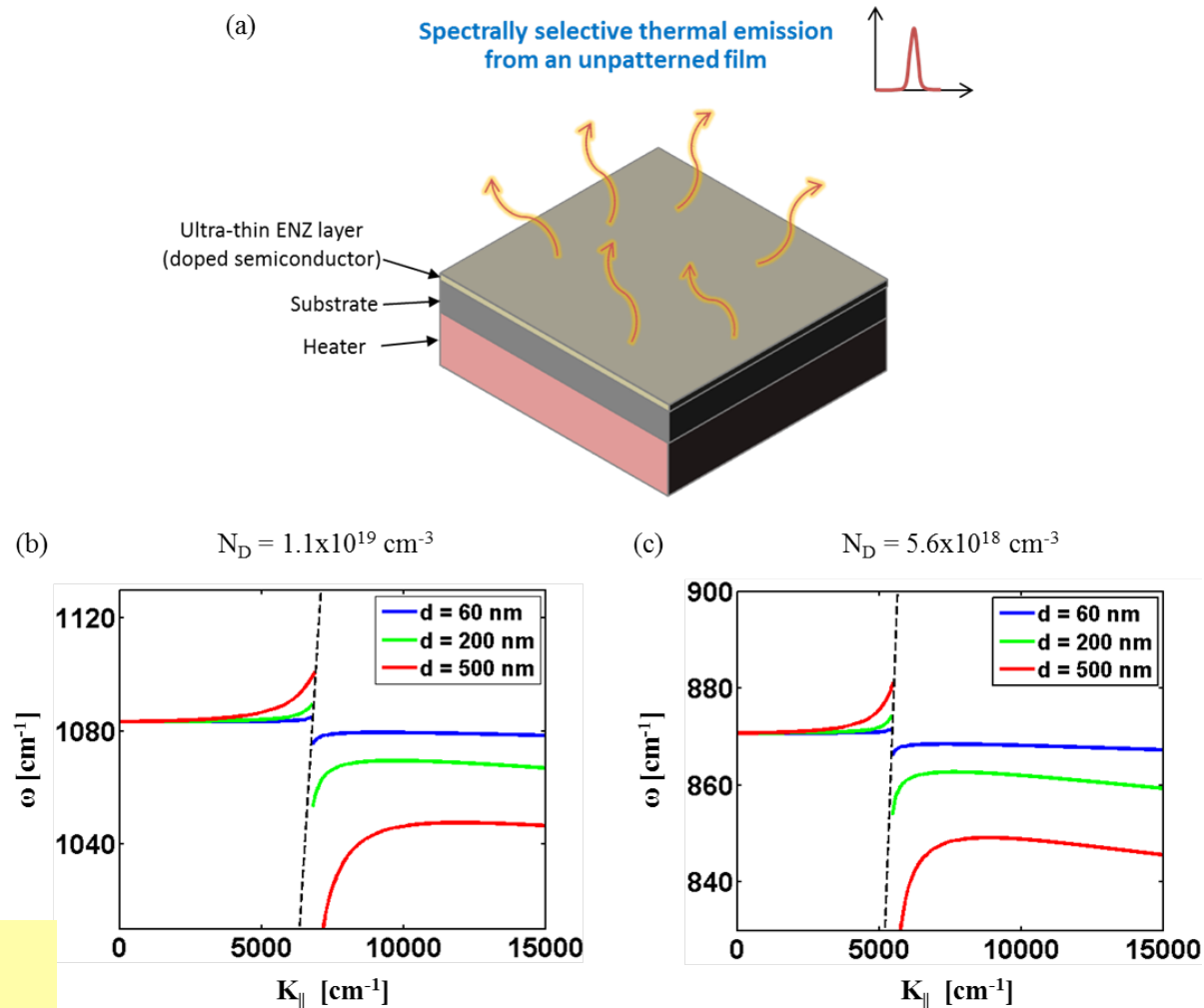


("Berreman" dip)



Thermal Emission: Berreman Modes

Kirchhoff's law of thermal radiation: the absorptivity of an object should be equal to its emissivity ($A = \epsilon$)

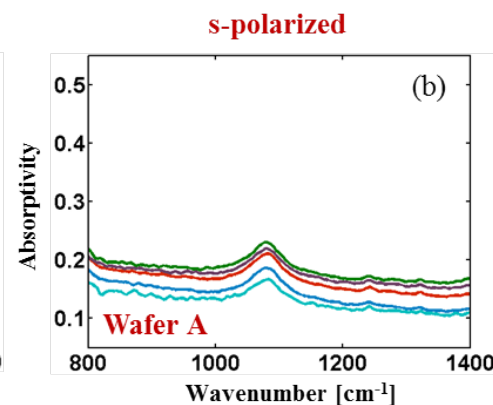
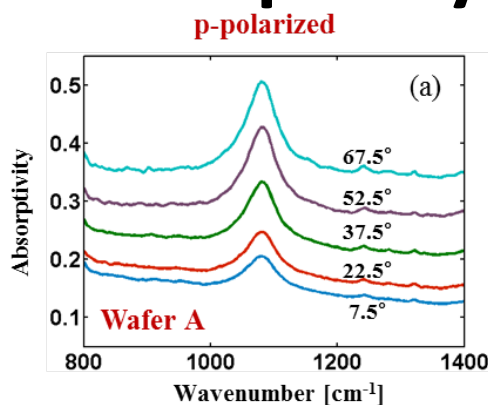


Appl. Phys. Lett. **105**,
131109 (2014)

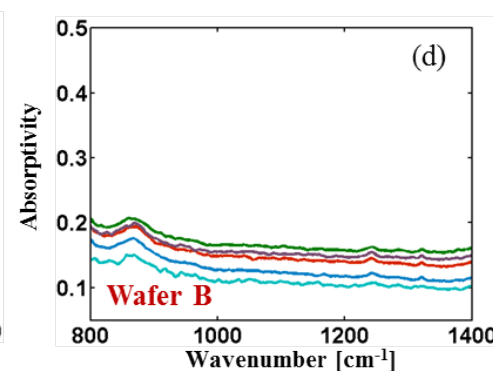
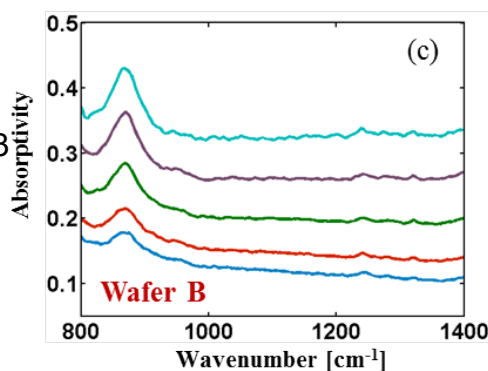


Angle-resolved, polarization-dependent absorptivity measurements

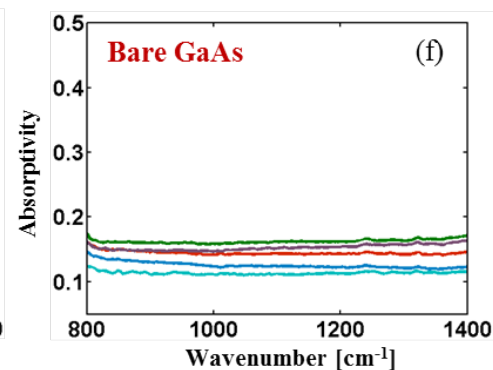
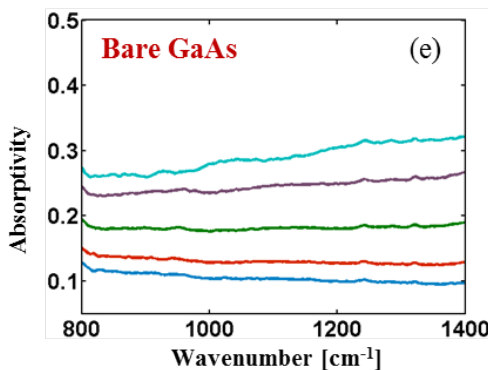
$$N_D = 1.1 \times 10^{19} \text{ cm}^{-3}$$



$$N_D = 5.6 \times 10^{18} \text{ cm}^{-3}$$



Bare GaAs



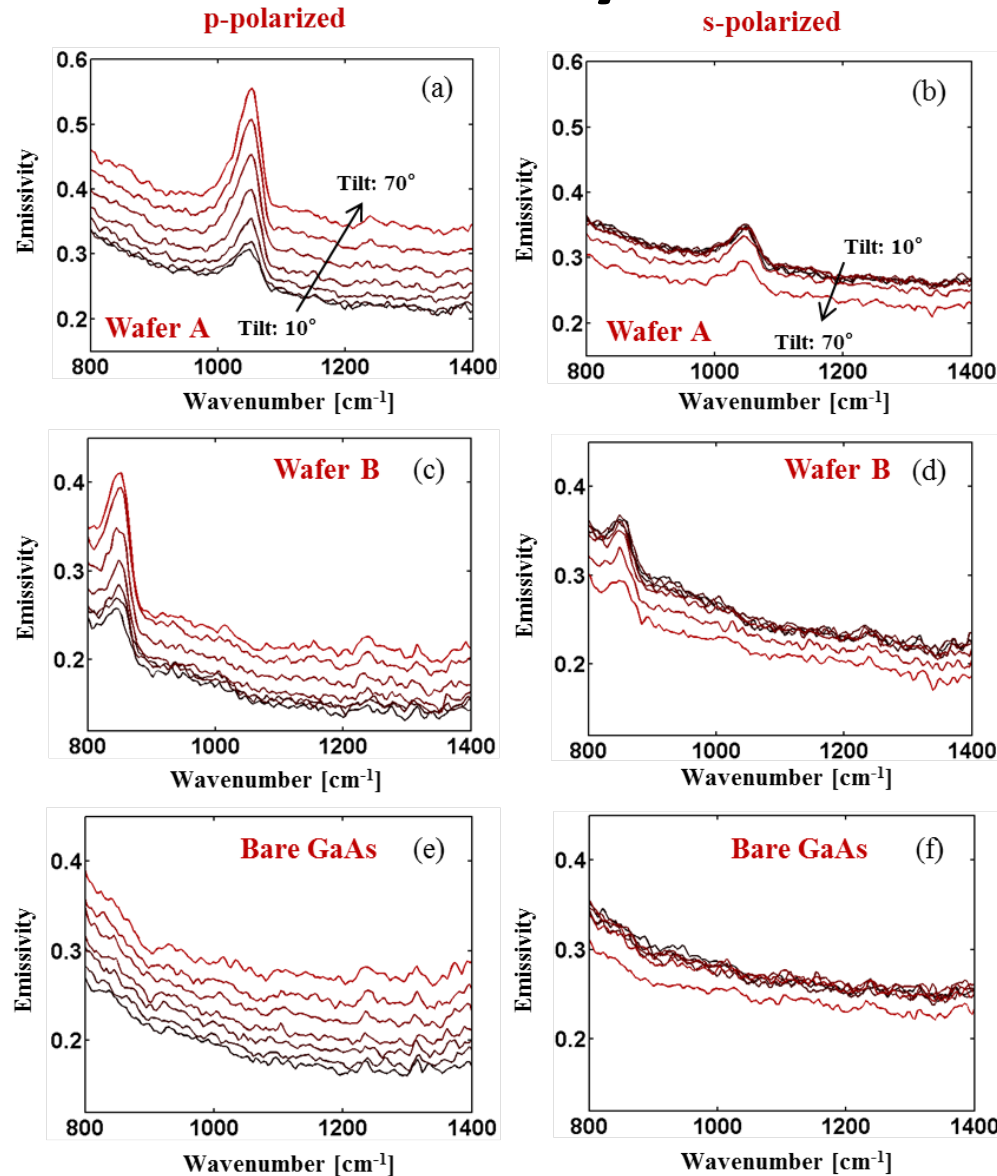
60nm InAs

Hyperhemispherical
Directional
Reflectometer

$$A = 1 - R - T$$



Angle-resolved, polarization-dependent emissivity measurements (140°C)



- Unpatterned semiconductor thin films generate spectrally selective thermal emission near the ENZ frequencies.
- Wavelength determined purely by doping densities
- An ENZ film works as a “leaky wave antenna”



Outline

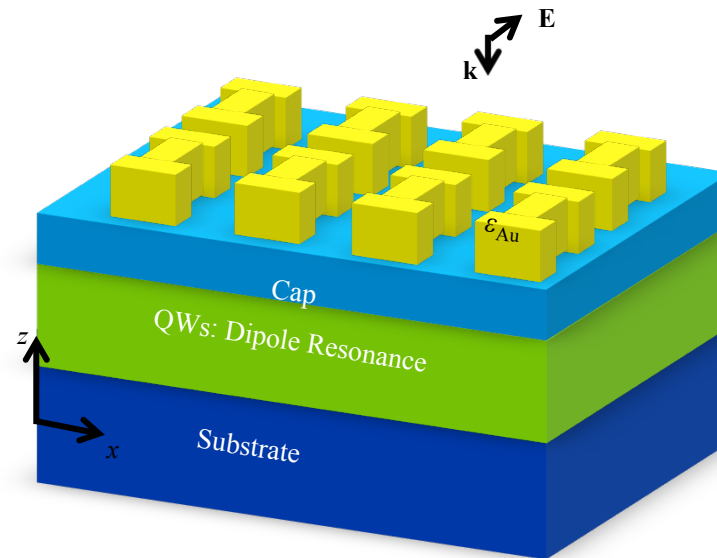
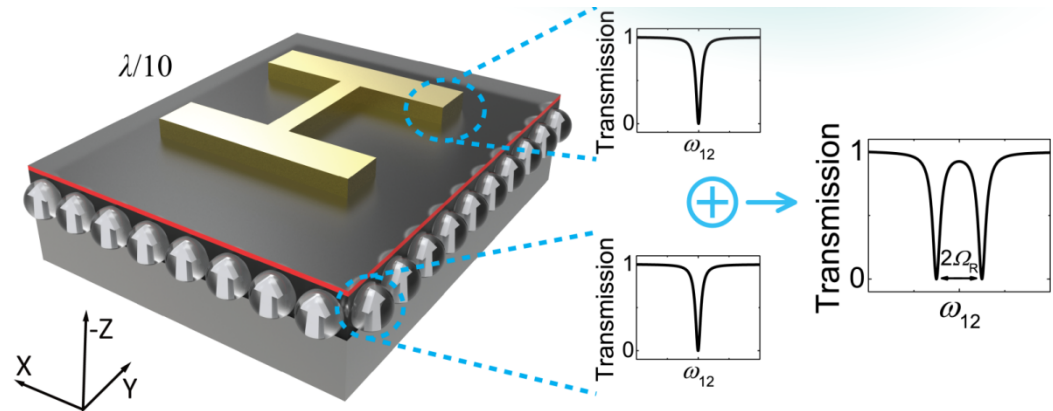
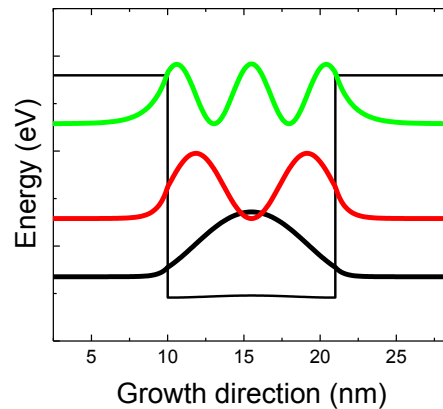
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 - **Enhanced Harmonic Generation**



Strong Coupling of Metamaterials to Inter-subband Transitions in Quantum Wells



Alex Benz

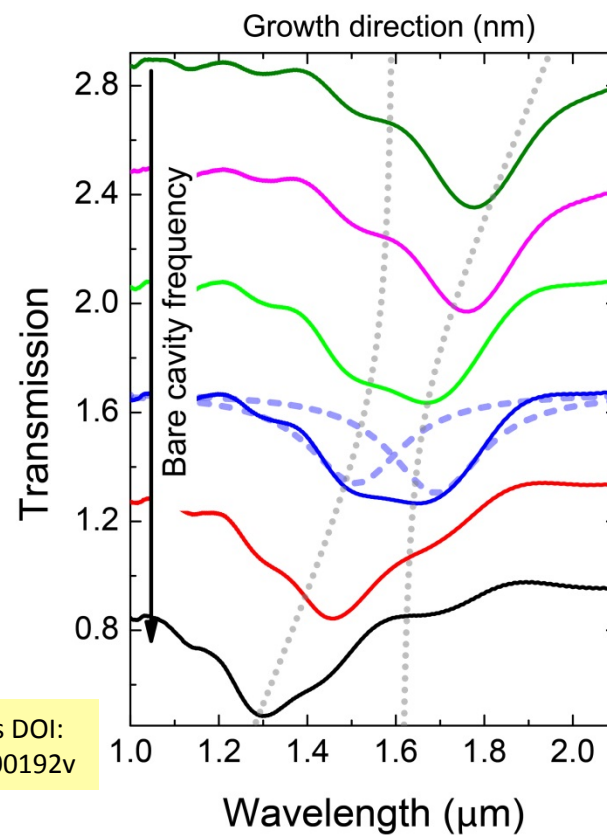
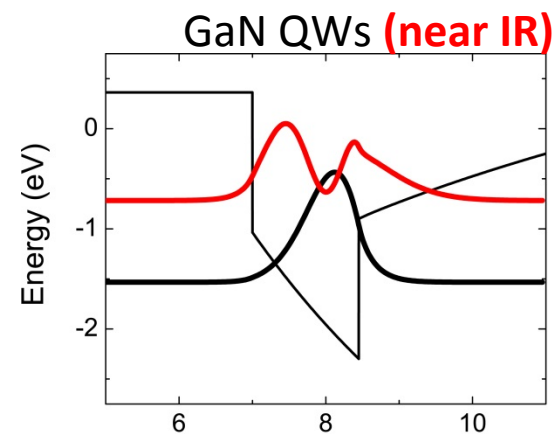
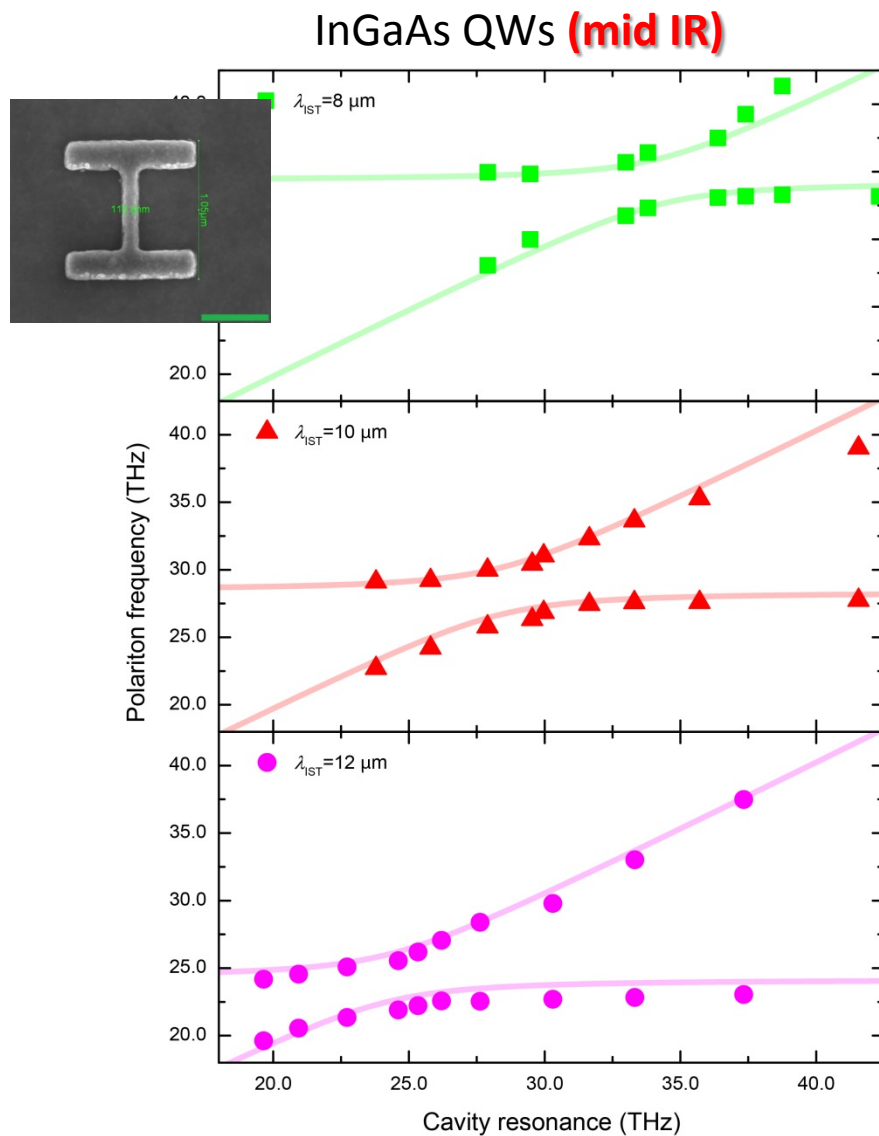


$$\epsilon_{xx} = \epsilon_{yy} \neq \epsilon_{zz}$$

Opt. Express 20, 6584 (2012),
APL 98, 203103 (2011)
Nature Communications 4,
(2013)



Strong Coupling: From Mid-IR to Near IR

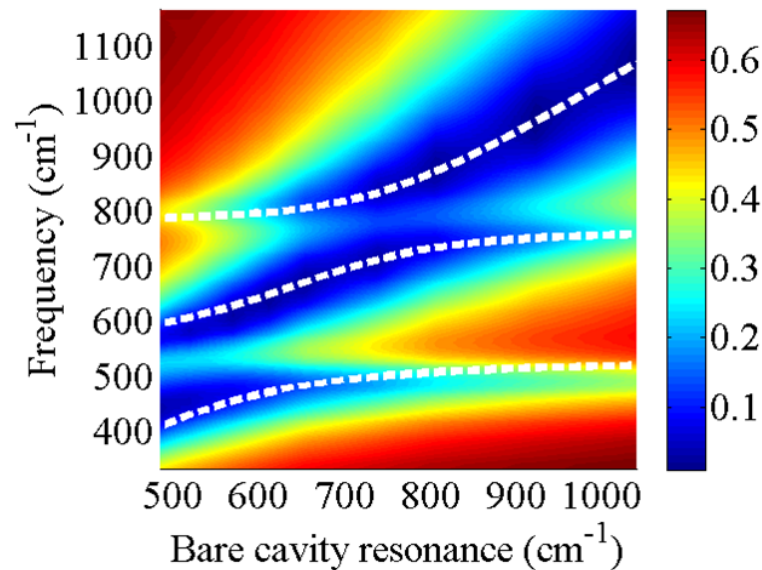
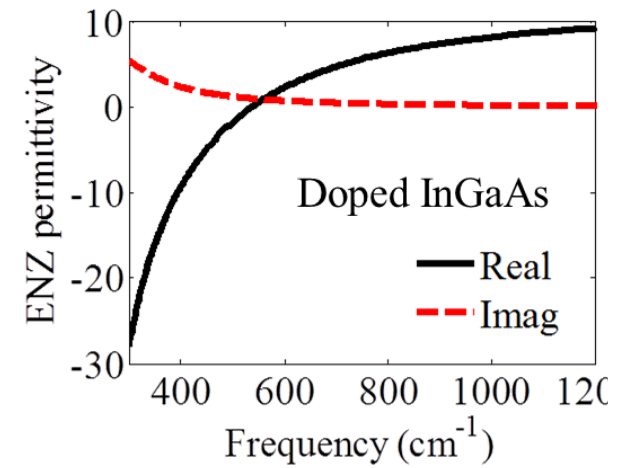
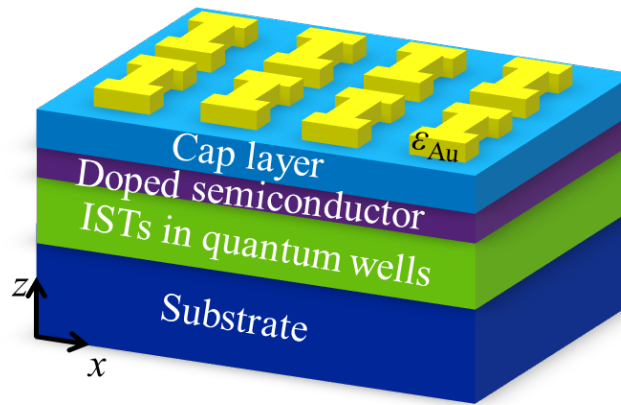
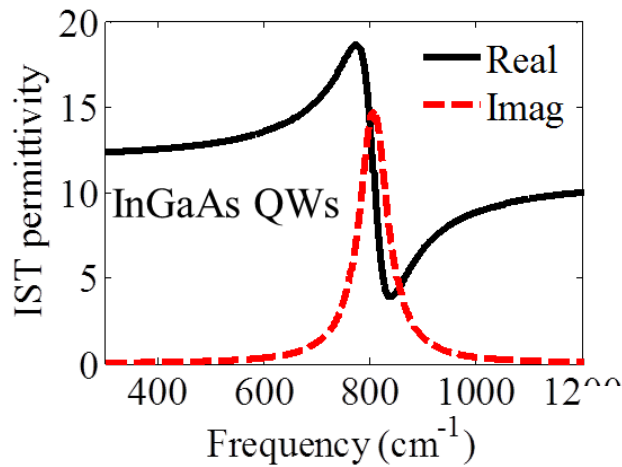


ACS Photonics DOI:
10.1021/ph500192v



Addition of ENZ layer to QWs

$\omega_p \neq$ IST transition frequency

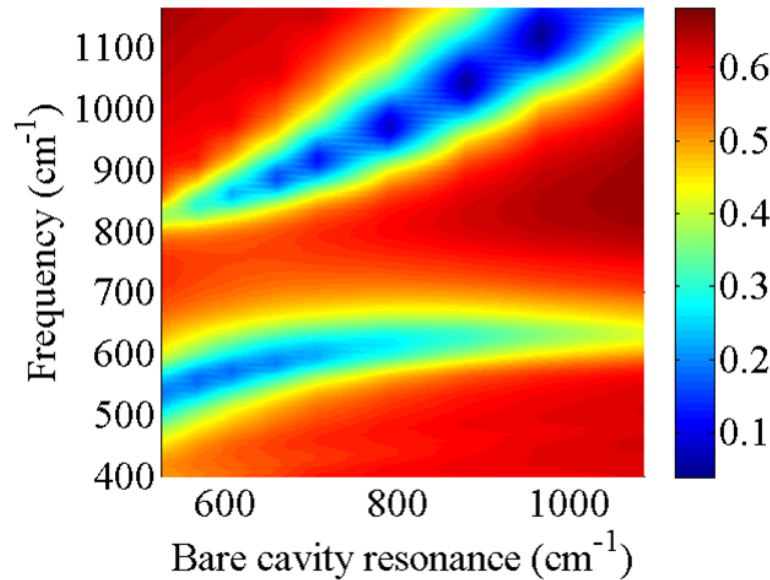
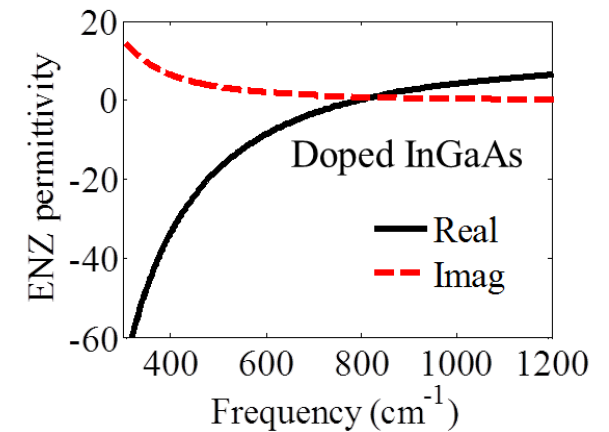
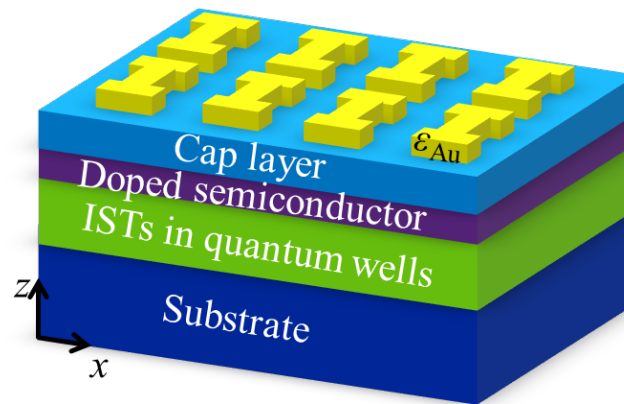
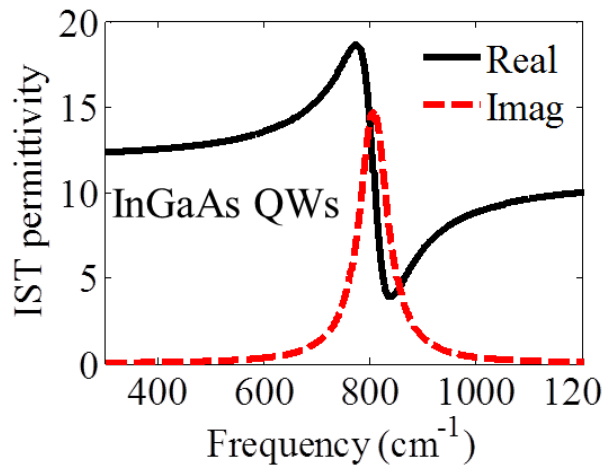


2 Polariton Branches



Addition of ENZ layer to QWs

$\omega_p = \text{IST transition frequency}$



1 Polariton Branch!
Bigger Rabi Splitting

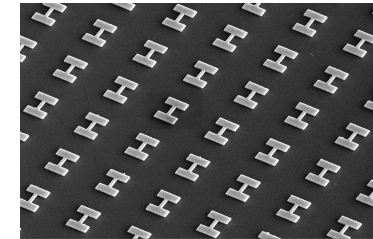
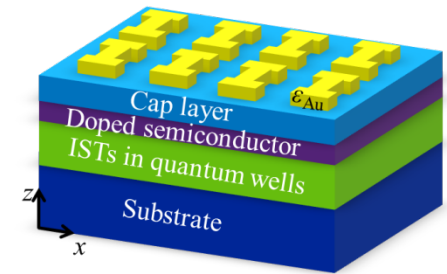
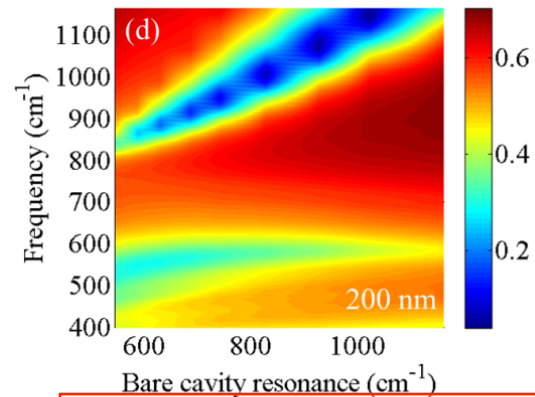
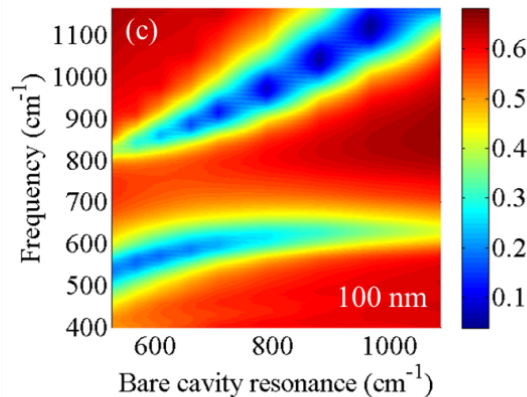
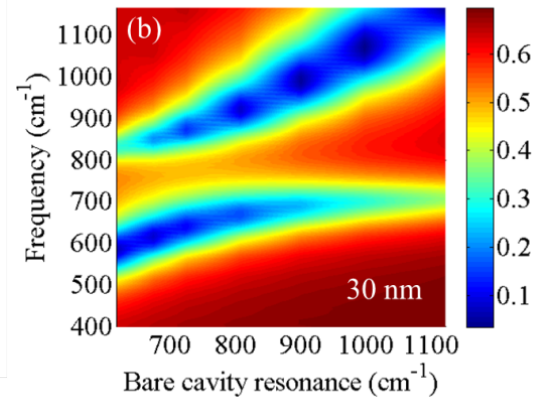
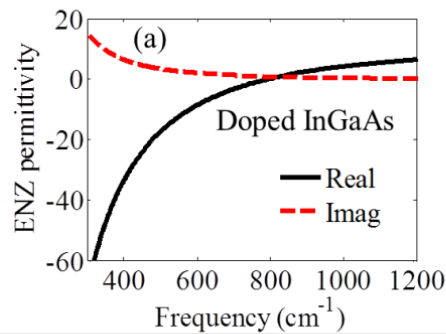


Using ENZ: Larger Rabi Splitting

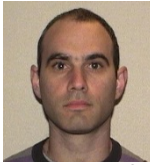


Salvo
Campione

Simulations

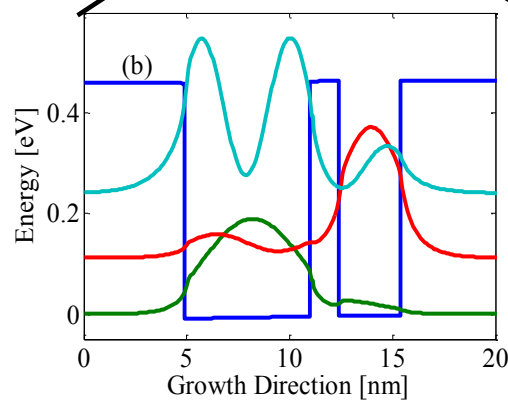
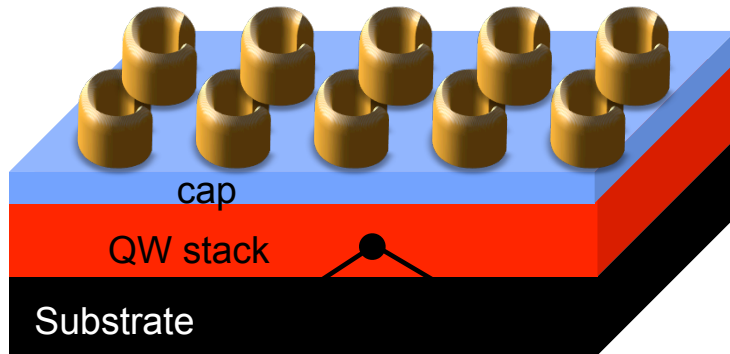


(e)	IST only	ENZ only 30 nm	ENZ only 100 nm	ENZ only 200 nm	ENZ 30 nm and 651 nm IST	ENZ 100 nm and 651 nm IST	ENZ 200 nm and 651 nm IST
Theory	150	177	250	280	240	300	315
Exp	120	180	NA	NA	NA	320	340



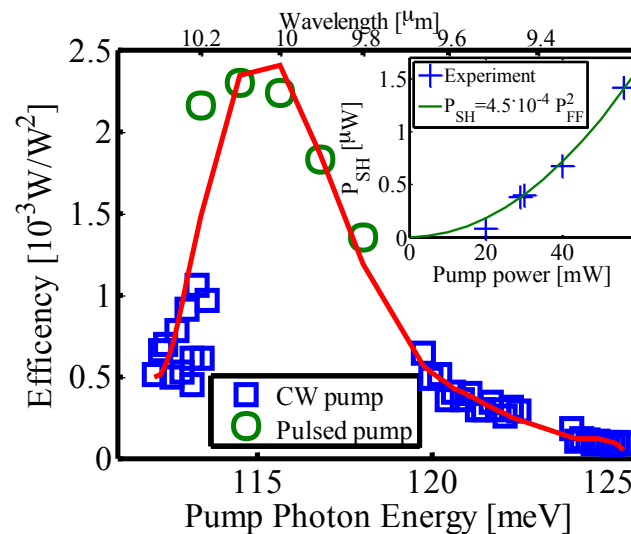
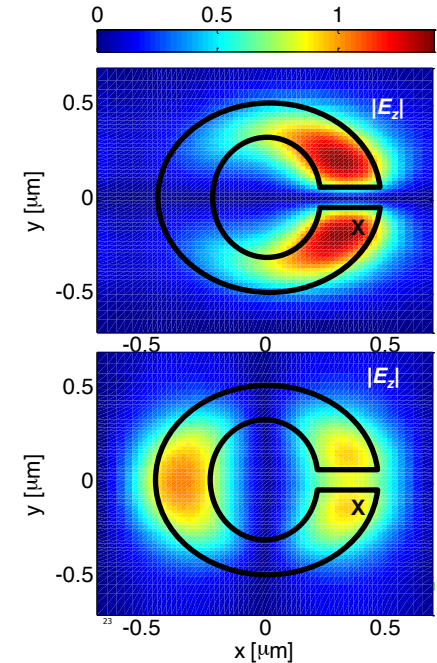
Omri Wolf

SHG From Strongly Coupled MMs and Intersubband Transitions



3 levels are designed to create a $\chi(2)$

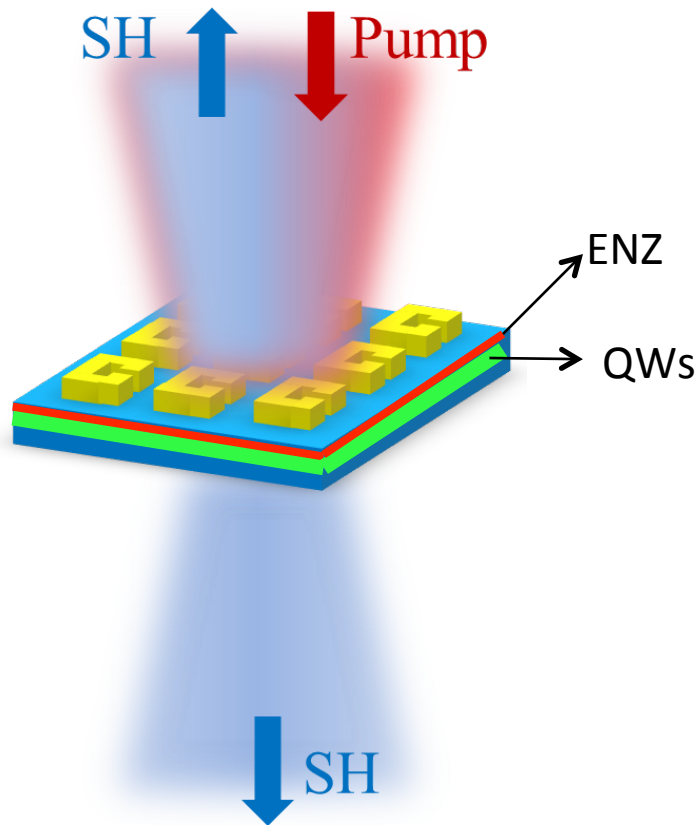
Resonators are designed to have resonances at 30 & 60 THz



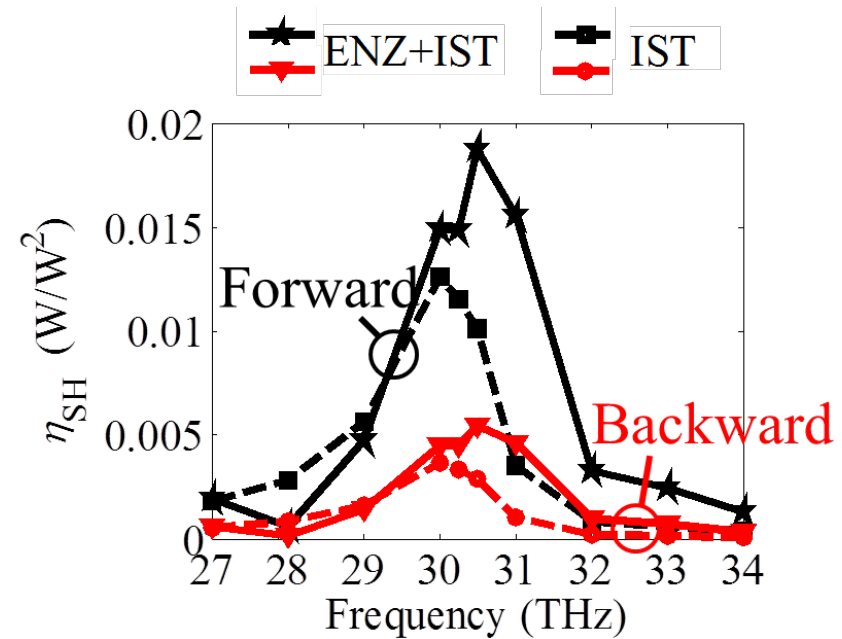
High Efficiency with $\sim 700\text{nm}$ thickness!



Addition of ENZ Layer Also Enhances SHG



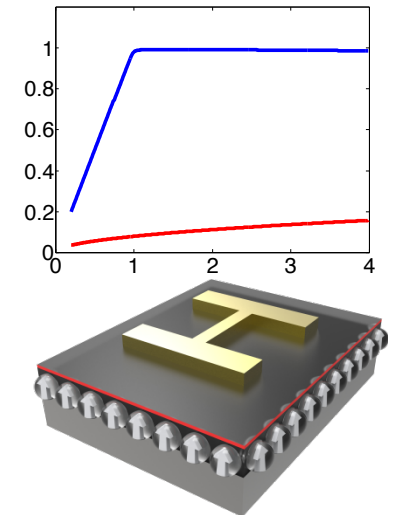
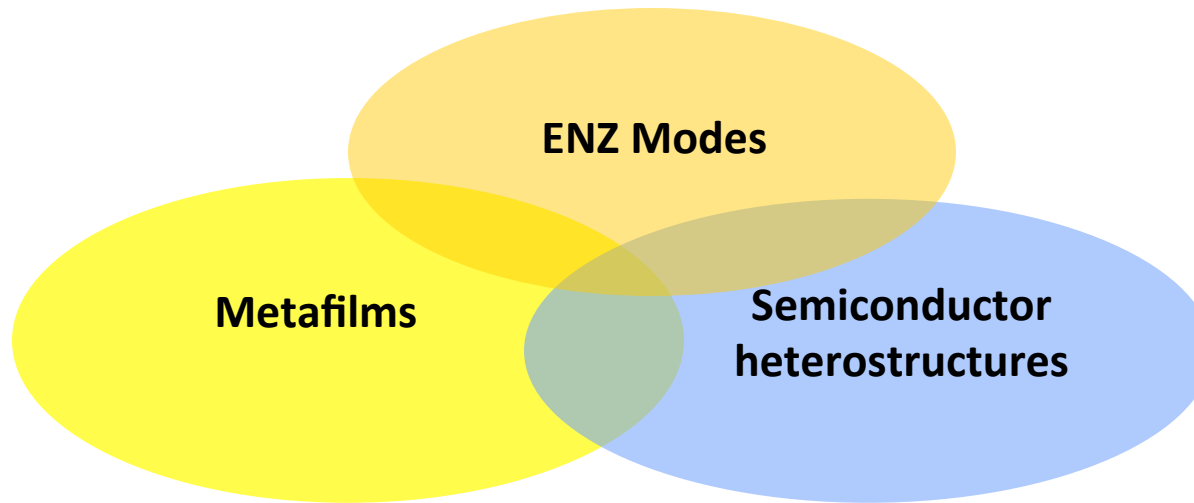
Conversion efficiency



The addition of an ENZ layer enhances SHG efficiency too



Summary



- **Metamaterial resonators + semiconductor heterostructures + ENZ modes :** platform for tunable spectral behavior and variable coupling
- **Addition of ENZ layer to metafilms enhances coupling to other dipole resonances (useful for spectral tuning, enhanced nonlinearities, etc)**
- **Berremian modes (close-relative of ENZ modes) can be used for spectrally selective thermal emission**



Acknowledgments

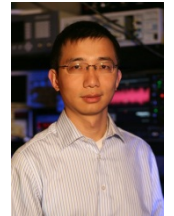
Postdocs: Young Chul Jun (n+, ENZ),
(Phonons),



Alex Benz (QWs),



Sheng Liu



Salvo Campione (ENZ, Sims),

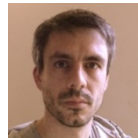


, Omri Wolf (SHG)



CINT + Sandia: John Reno, Mike Sinclair, Ines Montano, John Klem (InGaAs), Andy Allerman (GaN)

UC Irvine: Filippo Capolino



France: J.J. Greffet, F. Marquier

(ENZ)



SPARES

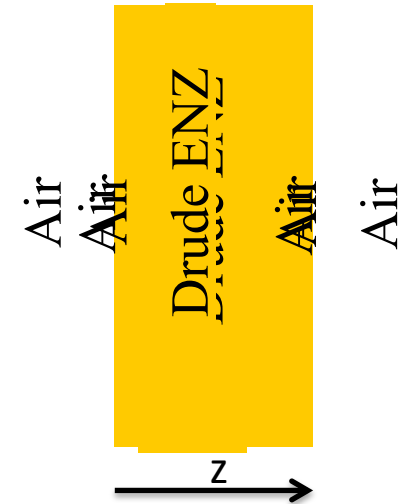
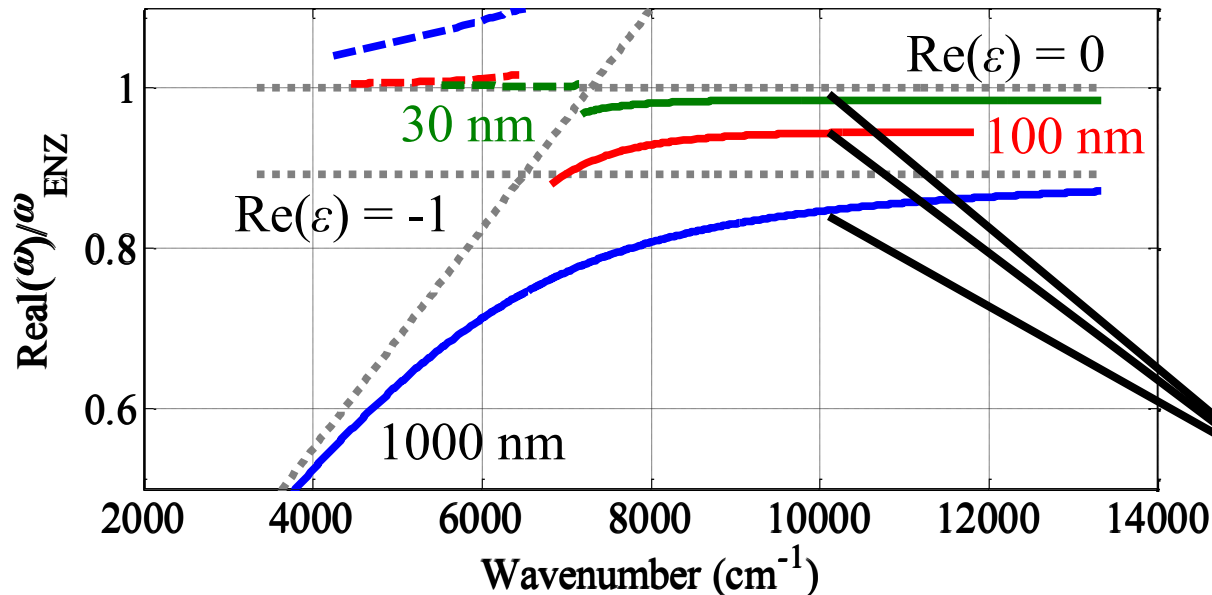


Plasmon Modes of a Thin Film: “ENZ mode”

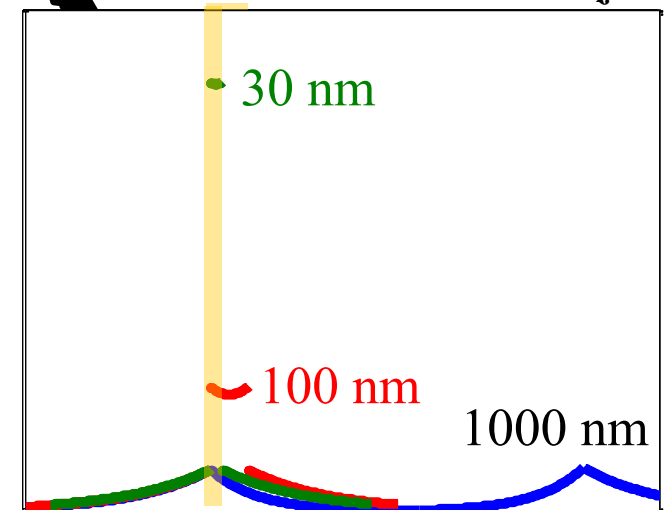


Salvo

“Thin”: ~20nm for ~30THz plasma frequency



Mode profile, E_z



The smaller the thickness, the larger E_z at the surface plasmon polariton

$$\left(1 + \frac{\epsilon_1 k_{z,1}}{\epsilon_3 k_{z,3}}\right) = i \tan(k_{z,2}d) \left(\frac{\epsilon_2 k_{z,3}}{\epsilon_3 k_{z,2}} + \frac{\epsilon_1 k_{z,2}}{\epsilon_2 k_{z,1}}\right)$$

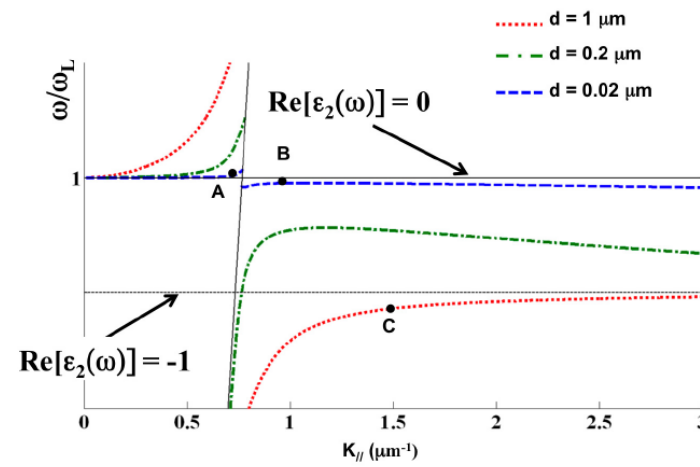
Numerical solution in complex domain
(assuming complex ω , real k)



Berreman mode and epsilon near zero mode

Simon Vassant,* Jean-Paul Hugonin, Francois Marquier, and
Jean-Jacques Greffet

*Laboratoire Charles Fabry, Institut d'Optique, Univ Paris Sud, CNRS, 2 av. Fresnel, 91127
Palaiseau, France*

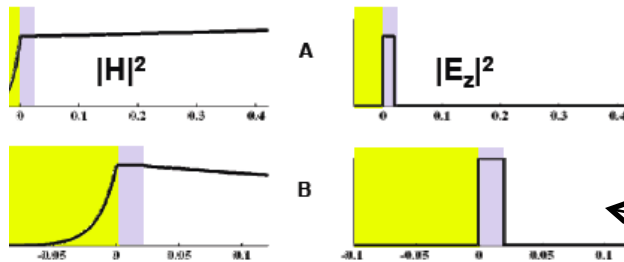
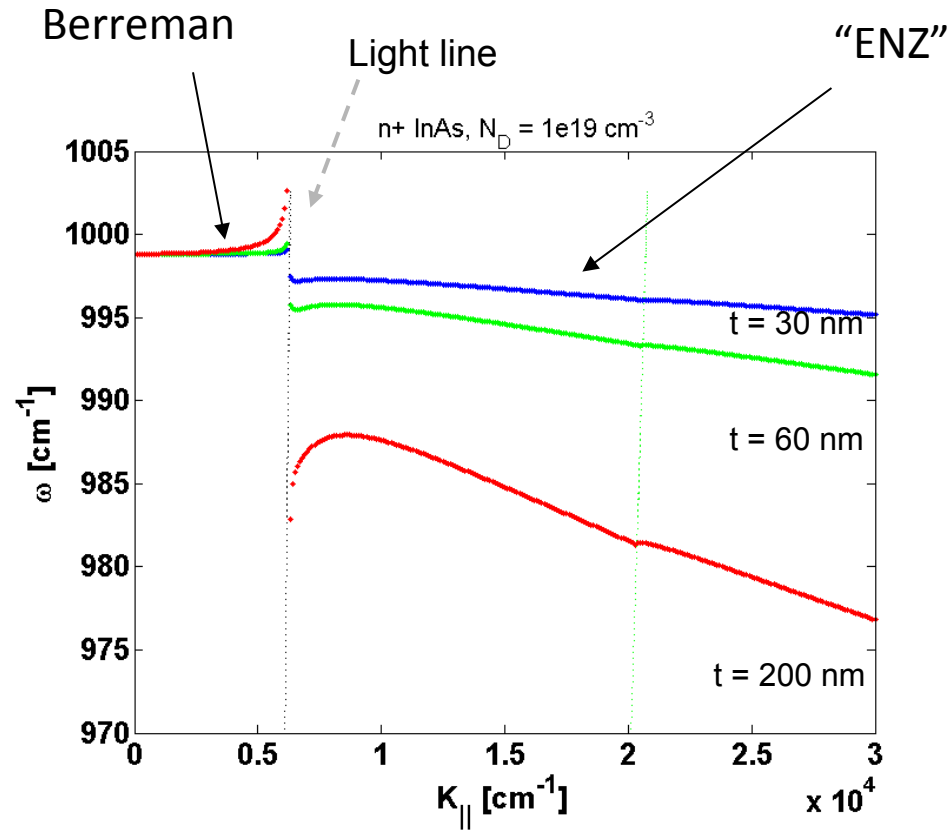




"Epsilon Near Zero" vs Berreman Modes



Young Chul Jun

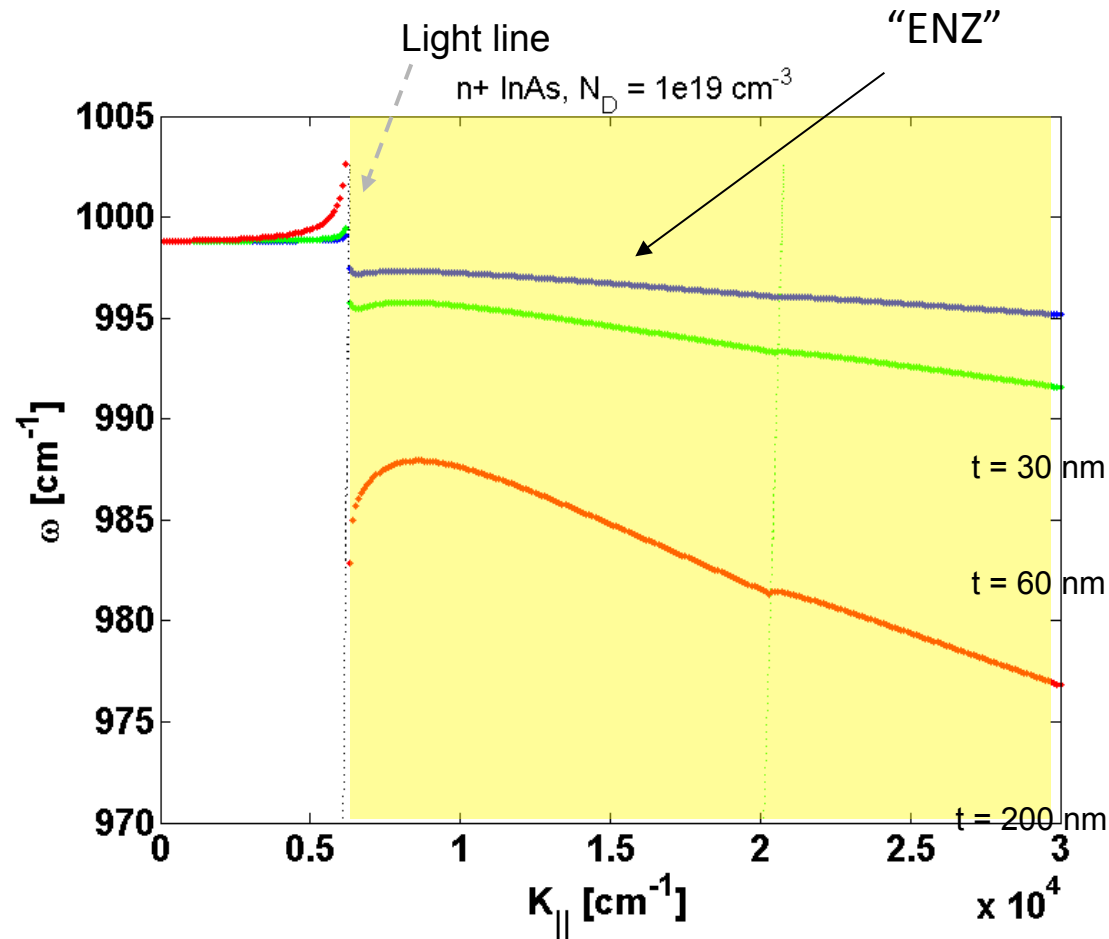


- Berreman mode (leaky mode),
- ENZ mode (bound mode)
- ENZ mode is a special type of a plasmon polariton



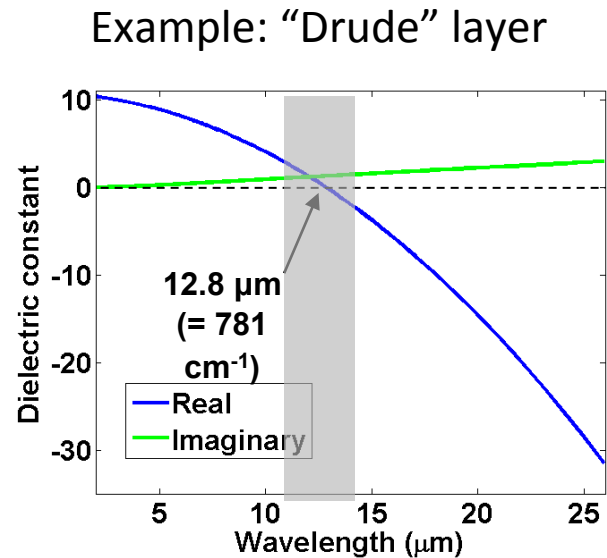
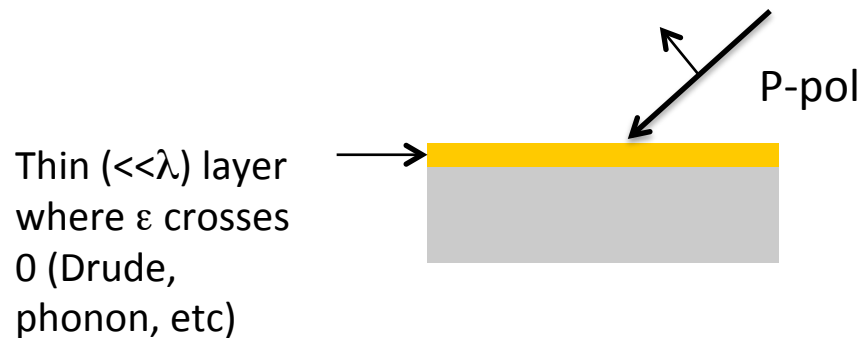
ENZ Modes

To the right hand side of the light line: cannot couple from free space

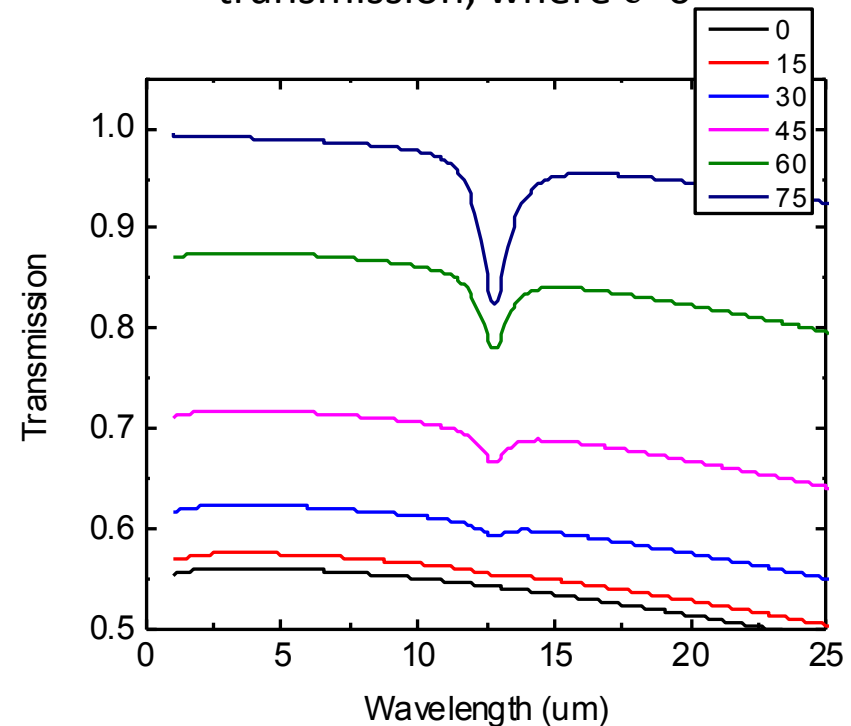




Coupling to Berreman Modes From Free Space



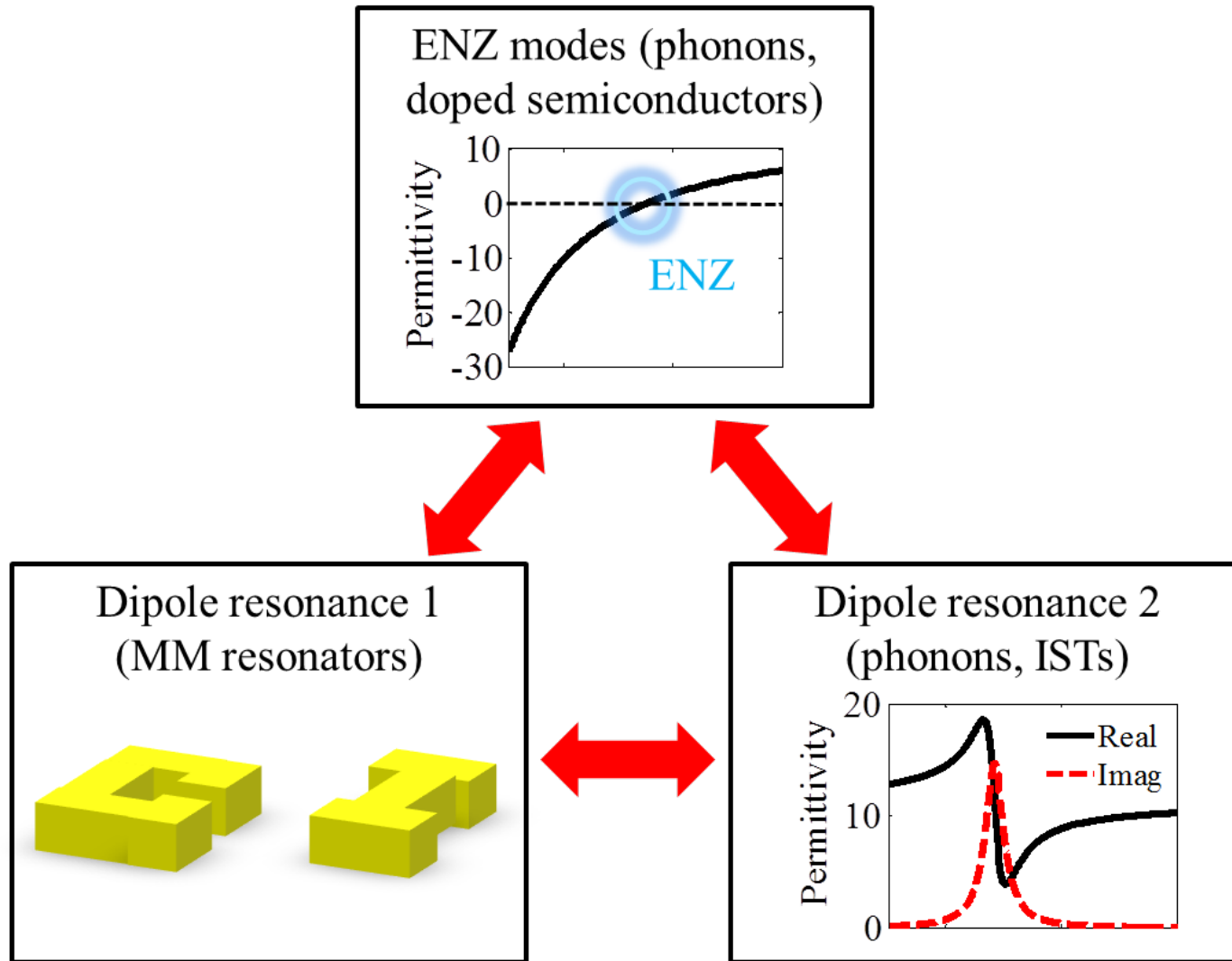
Transmission vs angle (p-pol):
A sharp dip is observed in transmission, where $\epsilon \sim 0$



("Berreman" dip)



Coupling of ENZ Modes to Other Resonances

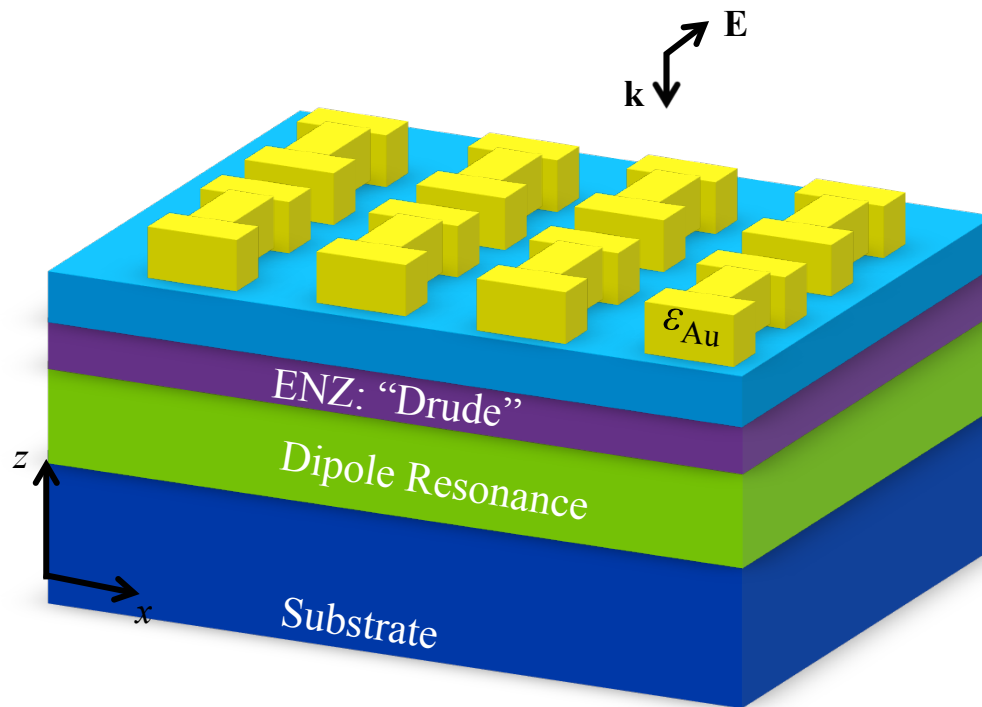
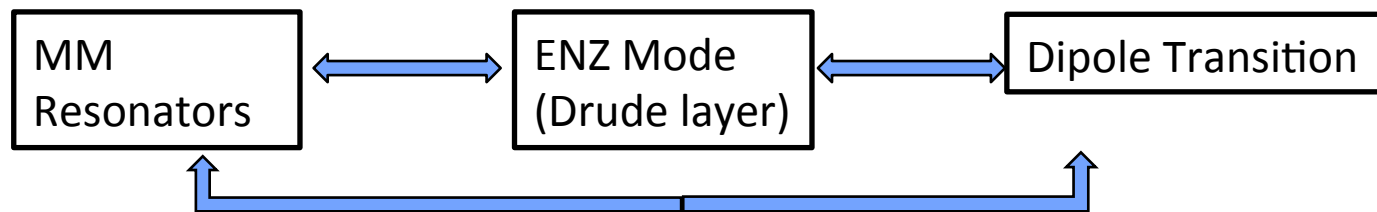


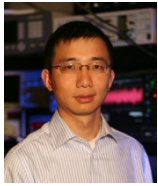


Salvo

Let's Separate the ENZ Layer and the Dipole Resonance Layer

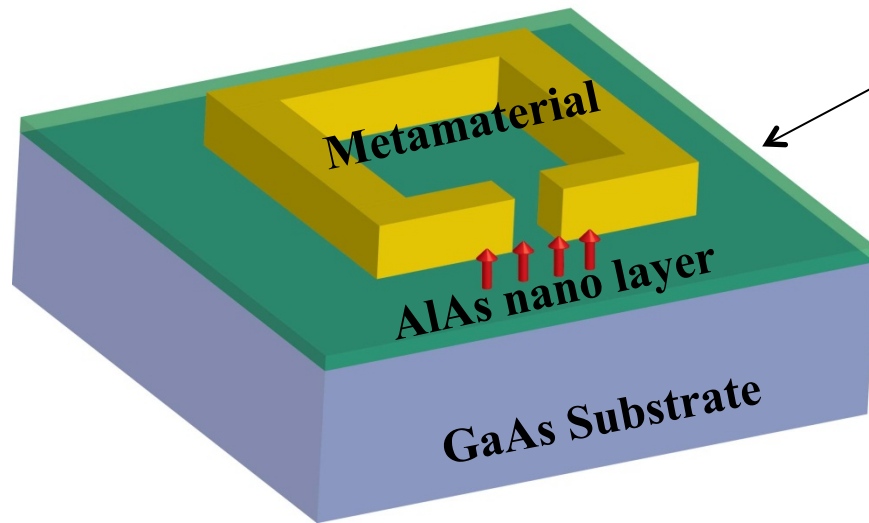
3 Interacting Systems



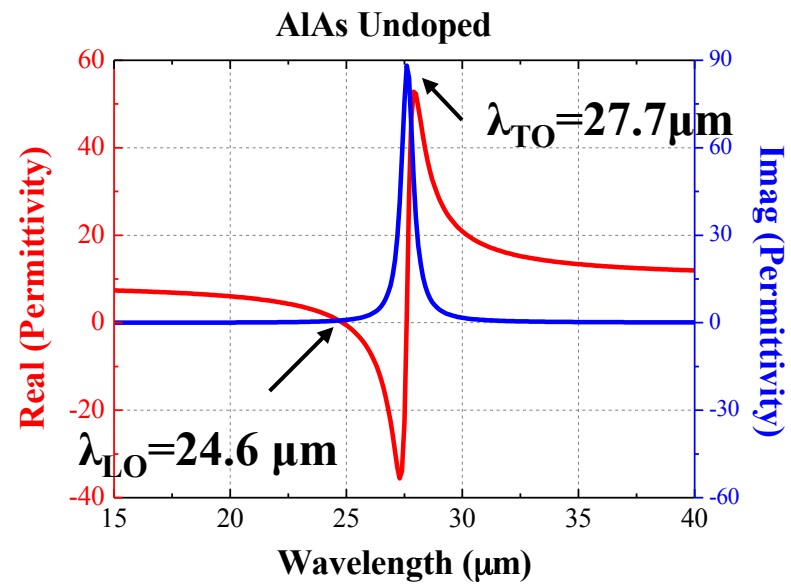


Sheng

ENZ Layer With an Optical Phonon Resonance



Optical Phonon



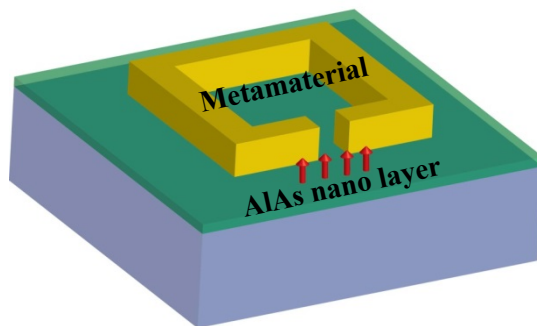
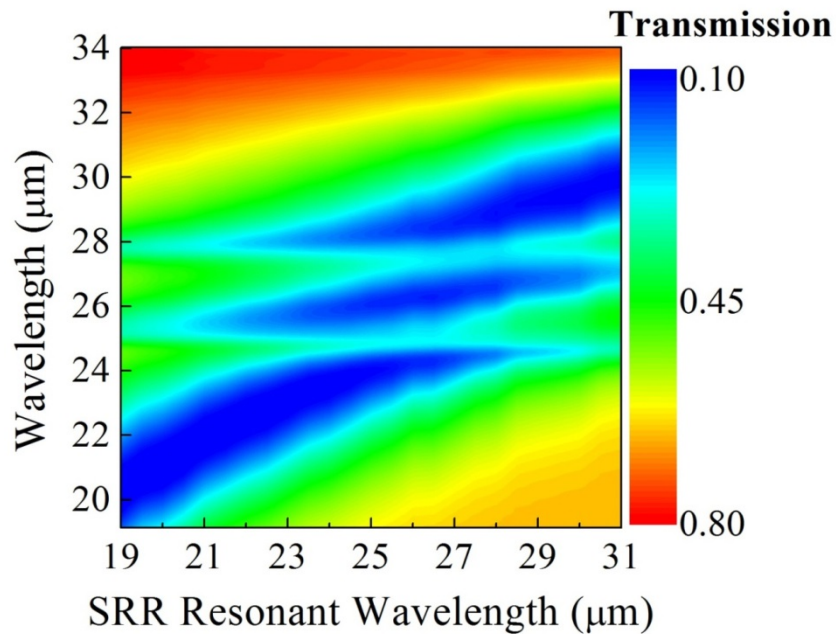
One thin layer contains:

- $\epsilon=0$ crossing -> **ENZ mode**
- TO Phonon -> **“dipole” transition**



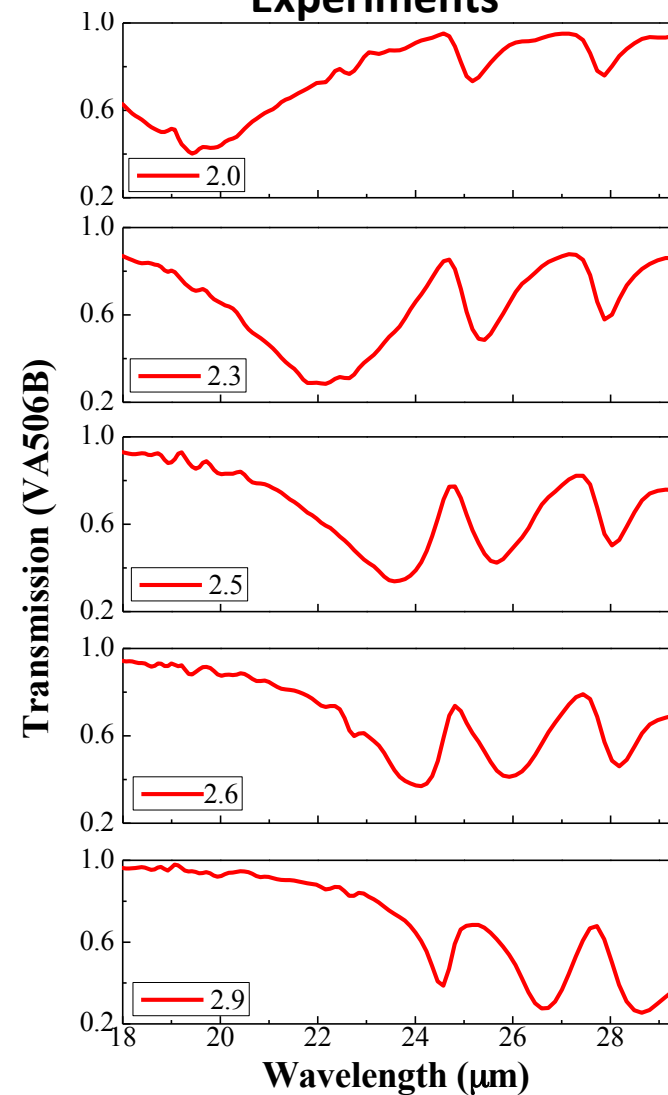
Strong Coupling: 2 Polariton Branches

Simulations



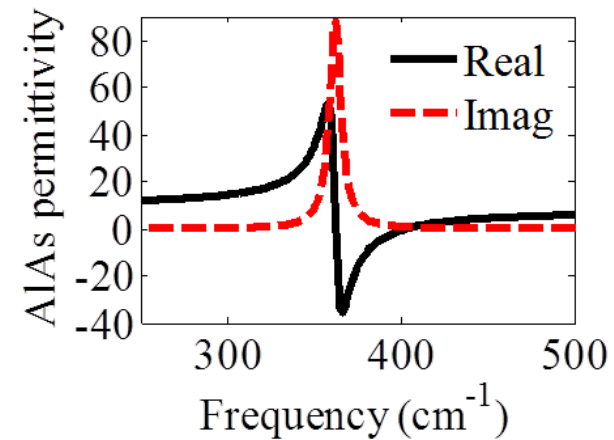
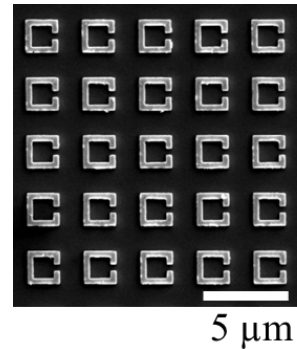
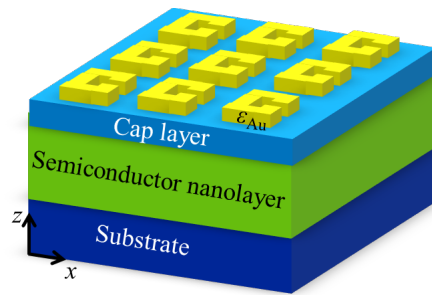
Increasing SRR size

Experiments

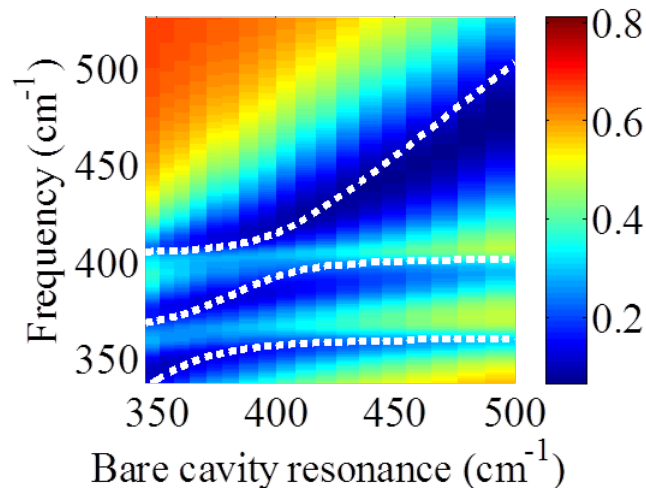




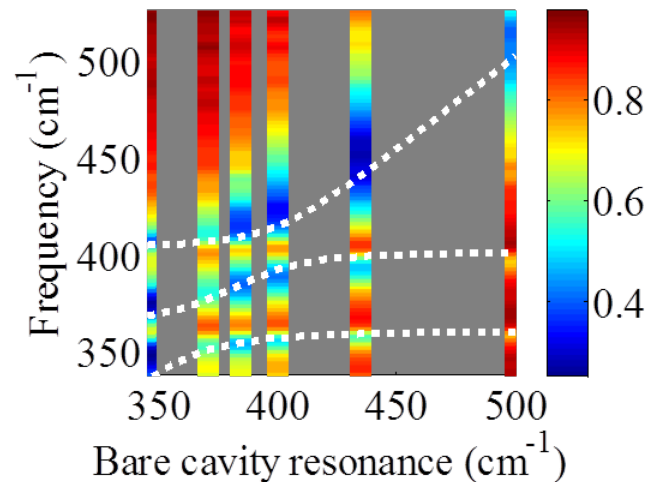
ENZ Layer With an Optical Phonon Resonance



Full-wave simulations



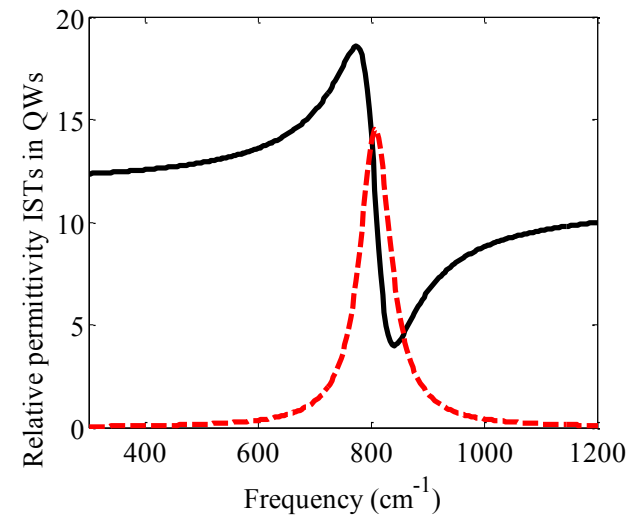
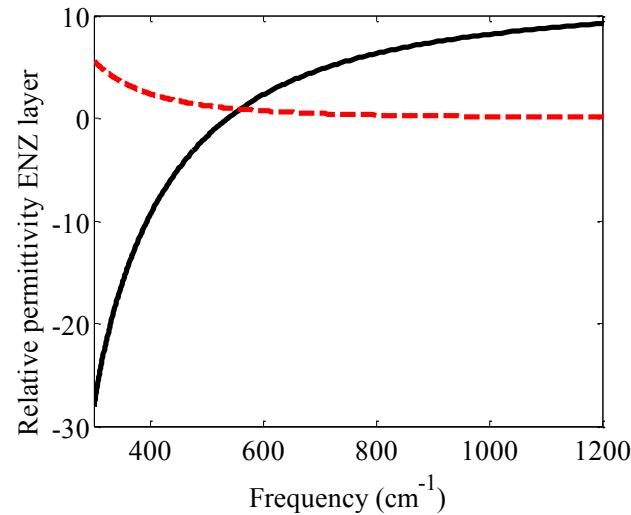
Experiments



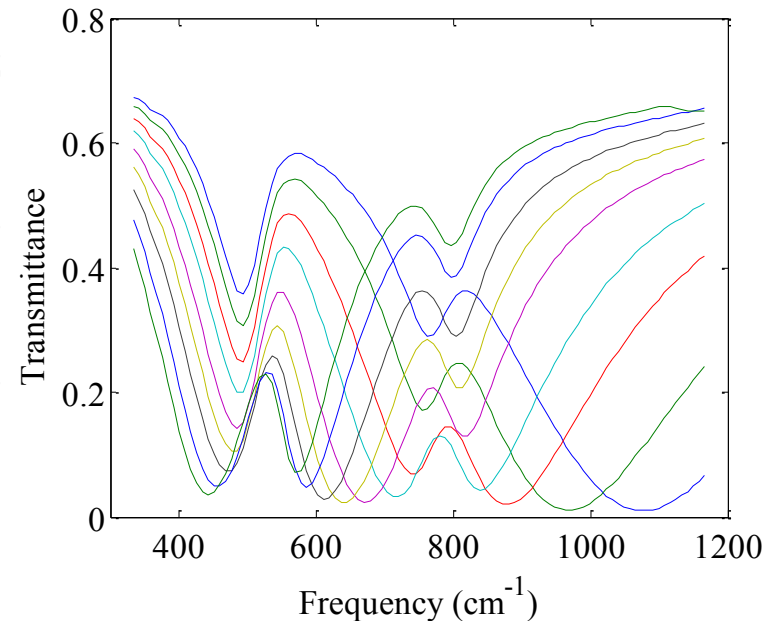
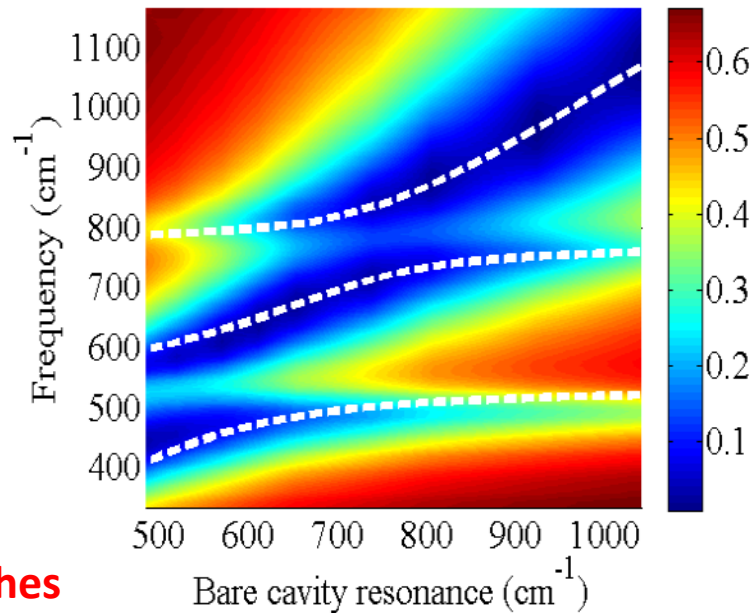
- Formation of three polariton branches
- This implementation is limited by nature: as optical phonons are defined by material properties and can hardly be modified



Case 1: Different Frequencies for Dipole Transition and ENZ



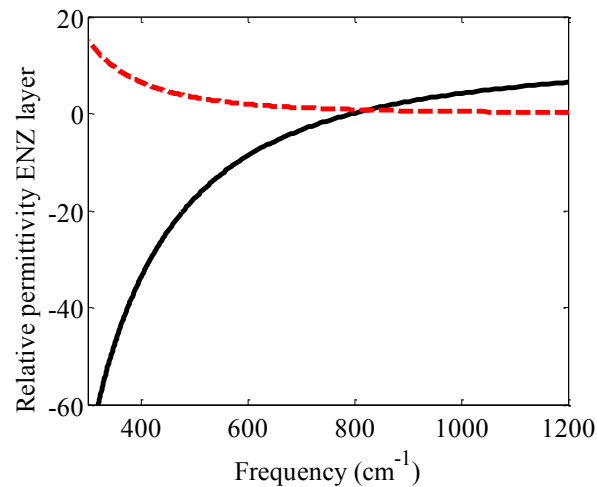
(simulations)



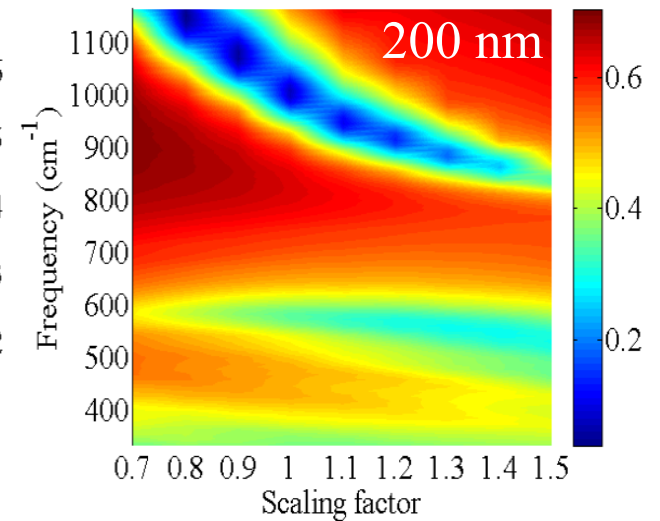
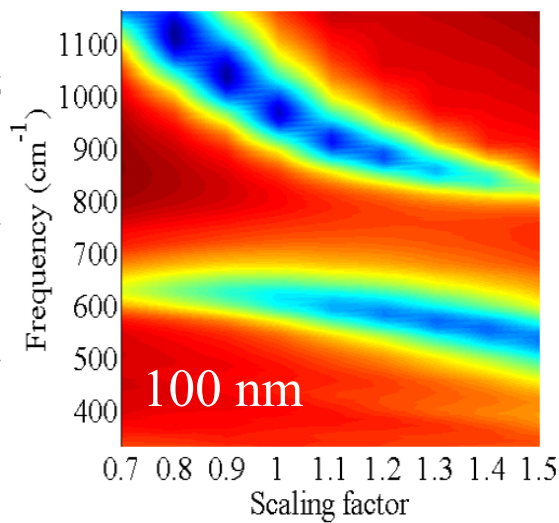
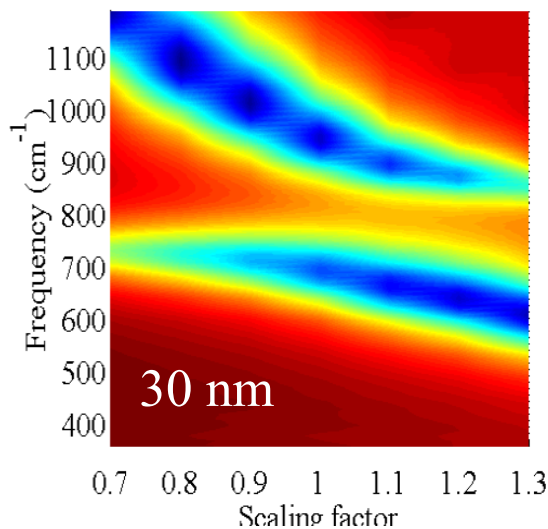
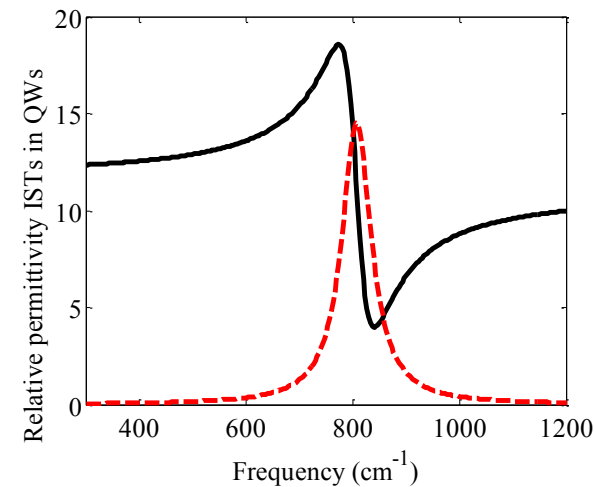
**2 Different
Polariton Branches**



Case 2: Same Frequencies for Dipole Transition and ENZ



(simulations)

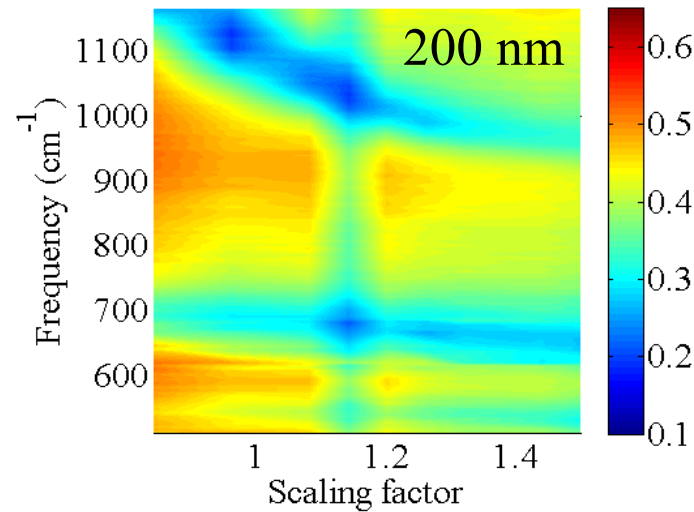
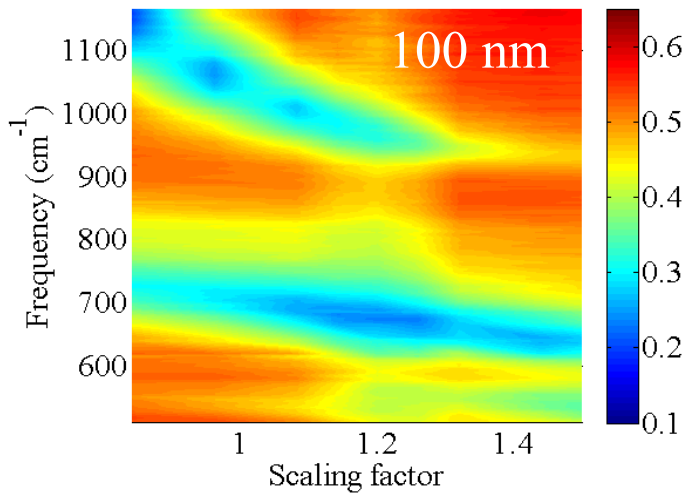
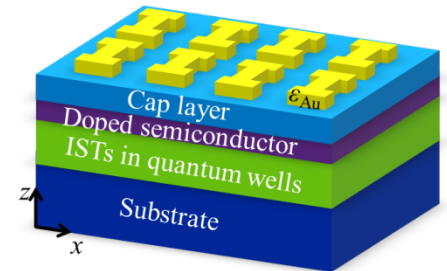
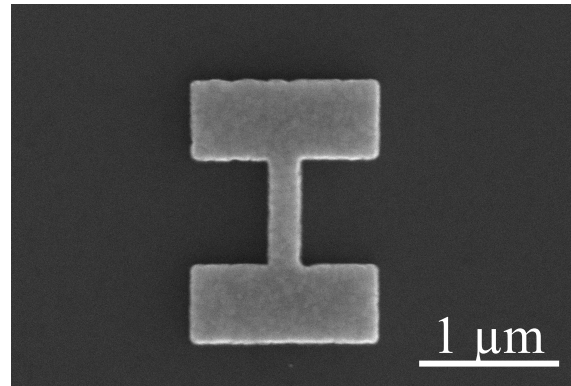
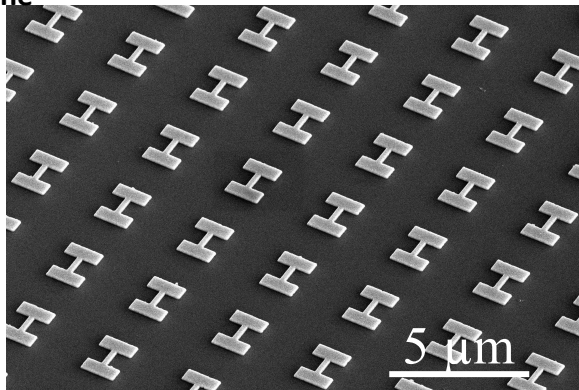


Only 1 Polariton, larger Rabi Splitting!



Salvo
Campione

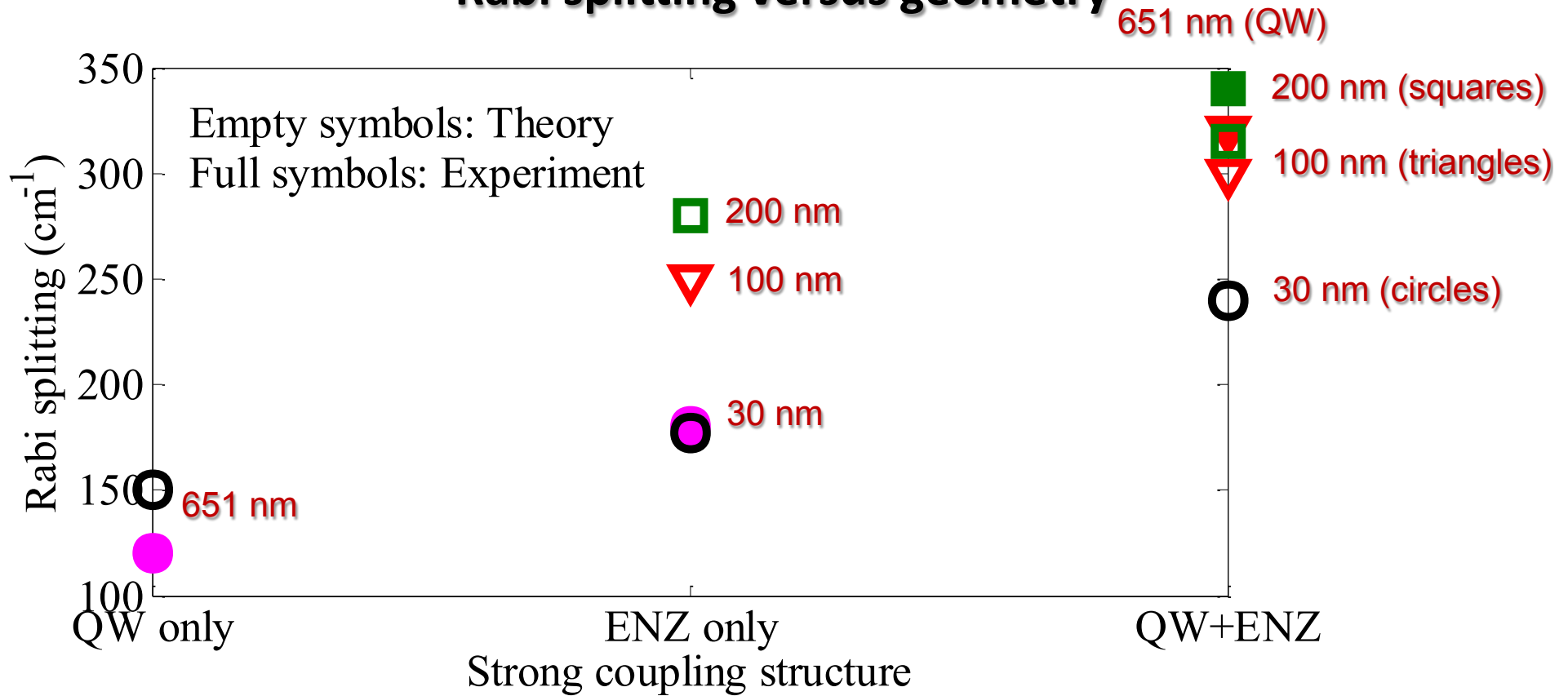
Experimental Verification of Combined Strong Coupling: (ENZ layer + dipole transition in QWs)





ENZ layer + dipole transition in QWs

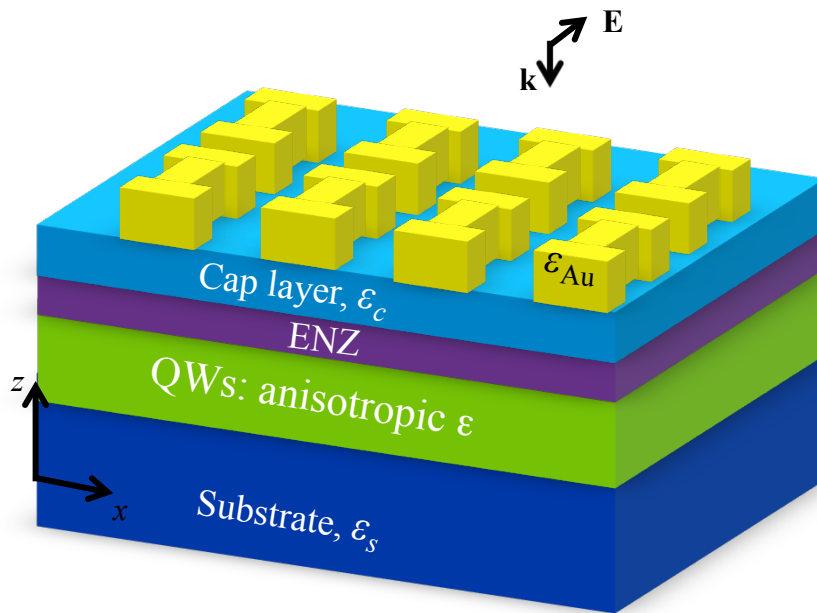
Rabi splitting versus geometry



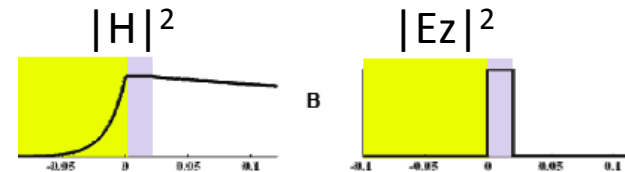
The structures comprising QW+ENZ exhibit the largest Rabi splittings for a given ENZ thickness



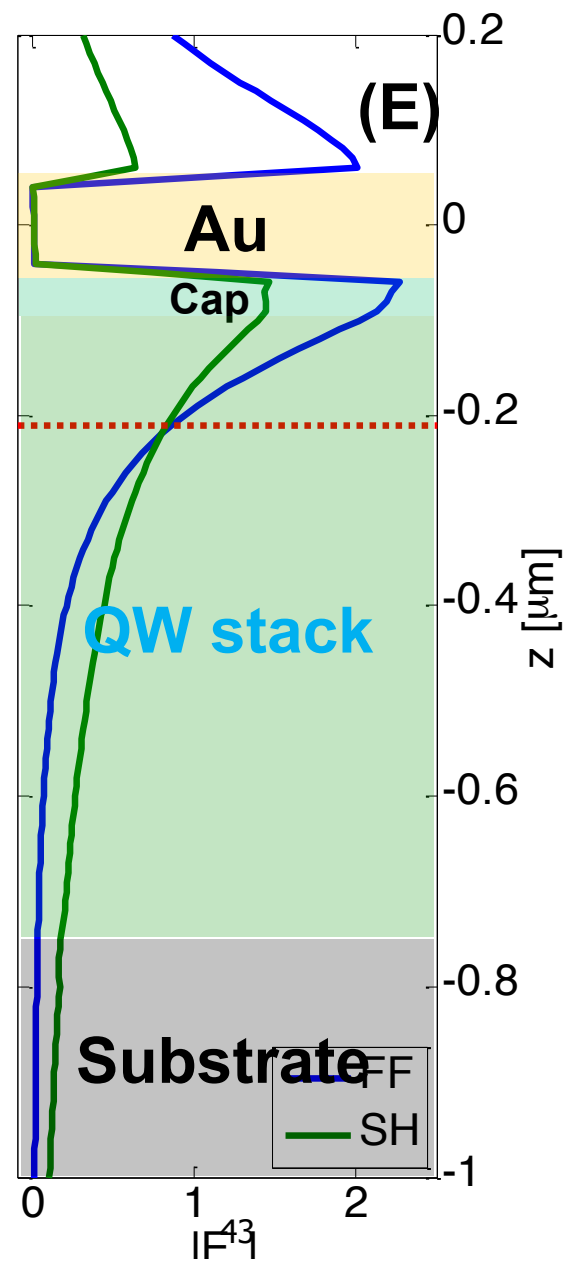
Combining ENZ Modes and Intersubband Transitions for Stronger Coupling



- Field decays exponentially with distance from MM resonators
- An ENZ thin layer acts as a “transducer” and potential “amplifier” of E_z field



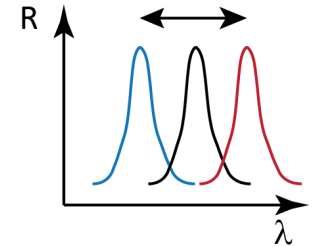
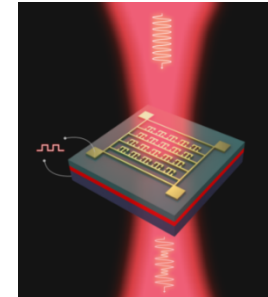
The addition of an ENZ layer enhances the coupling
-> larger Rabi splitting
-> larger SHG efficiency





Tunable Planar Metamaterials: Why?

A voltage tunable IR spectral filter would be a very useful device:

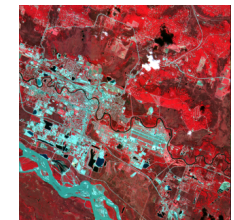
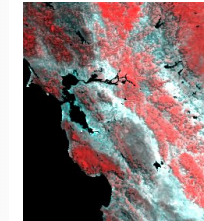


1. Application example: IR hyperspectral imaging



(Cedip)

(NASA)



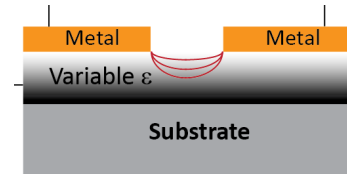
Has mechanical filters!

2. Fundamentals: (ultra?) strongly coupled systems are interesting for control of emission, quantum optics, cavity QED, etc



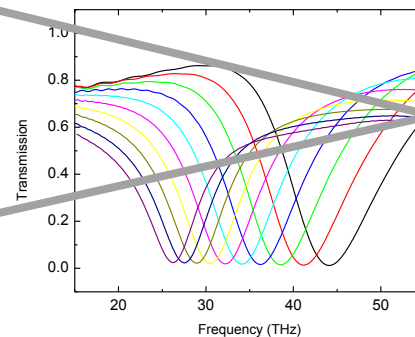
Planar Metamaterials: Strong Coupling & Tuning

MM resonators interact strongly with thin layers underneath
(see papers by Chen, Giessen, Zheludev, us, etc)

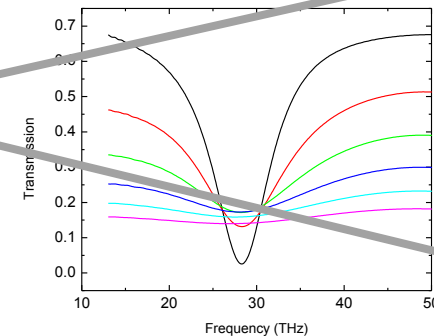


- Nonresonant

Change ϵ_1 (n)

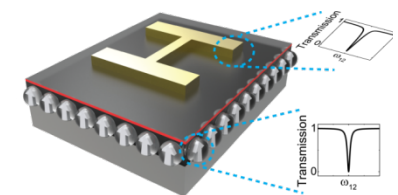
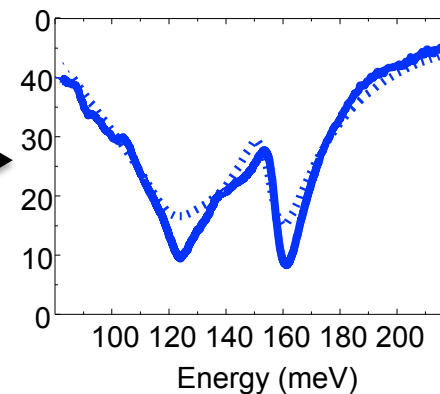
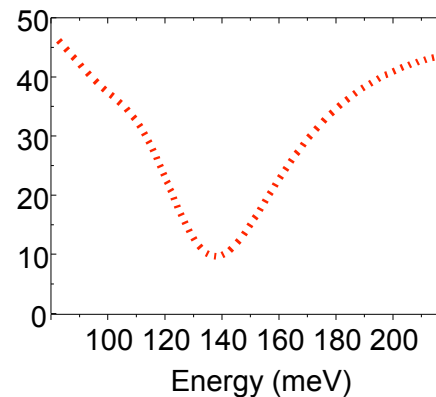


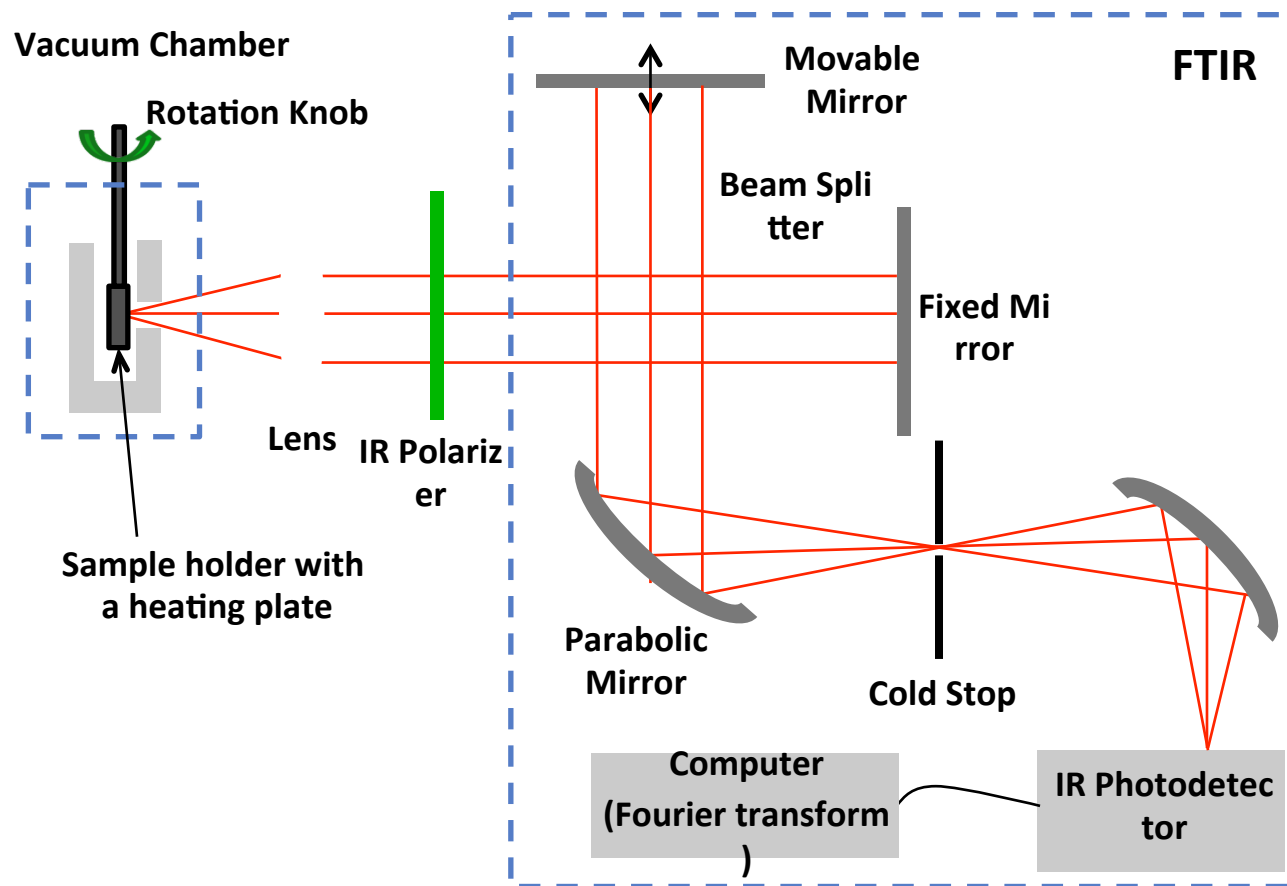
Change ϵ_2 (n)



- **Resonant**
(Use a dipolar resonance, example: phonon)

Nano Letters 11, 2104 (2011)



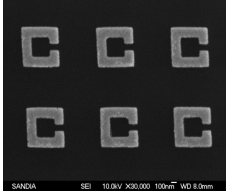
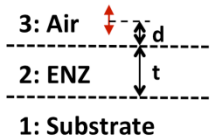




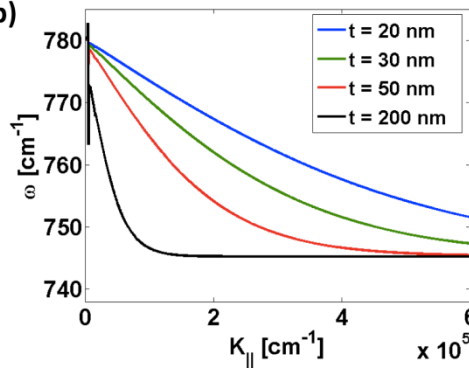
MM Resonators Provide k-vectors to Excite ENZ Modes

ENZ dispersion

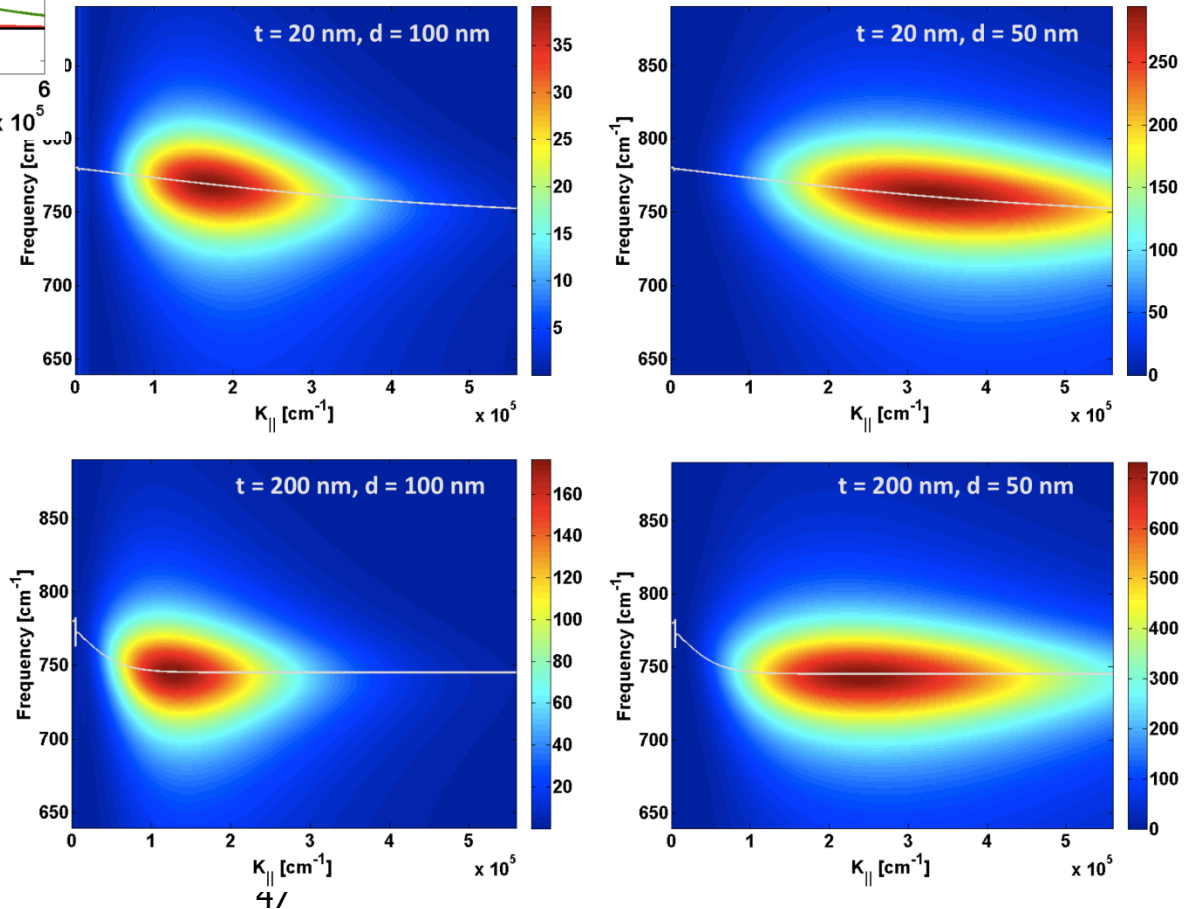
(a)



(b)



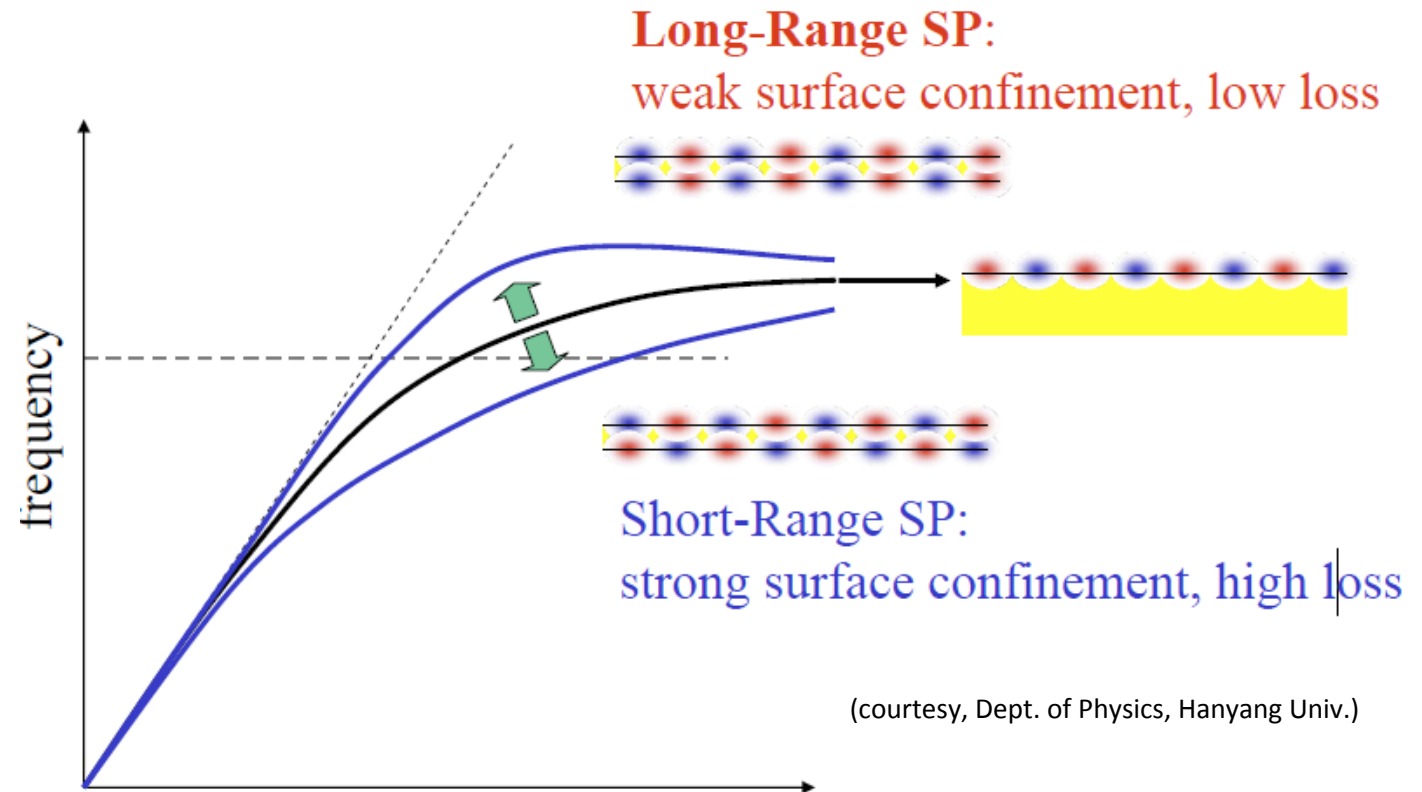
Superimpose dipole emission spectra on
the ENZ dispersion





Plasmon Modes of a Thin Film

“Thin”: ~20nm for ~750THz plasma frequency



$$\left(1 + \frac{\epsilon_1 k_{z,1}}{\epsilon_3 k_{z,3}}\right) = i \tan(k_{z,2}d) \left(\frac{\epsilon_2 k_{z,3}}{\epsilon_3 k_{z,2}} + \frac{\epsilon_1 k_{z,2}}{\epsilon_2 k_{z,1}}\right)$$

assuming real ω , complex k

Economou, 1969
Burke 1986
etc



“Epsilon Near Zero” vs Berreman Modes

Air

Drude

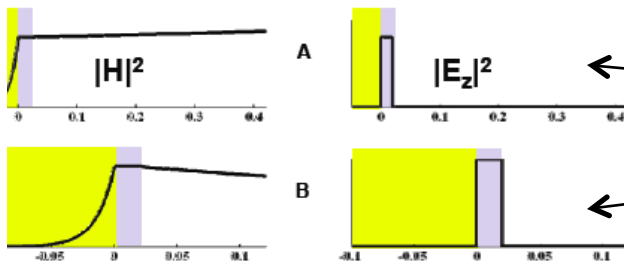
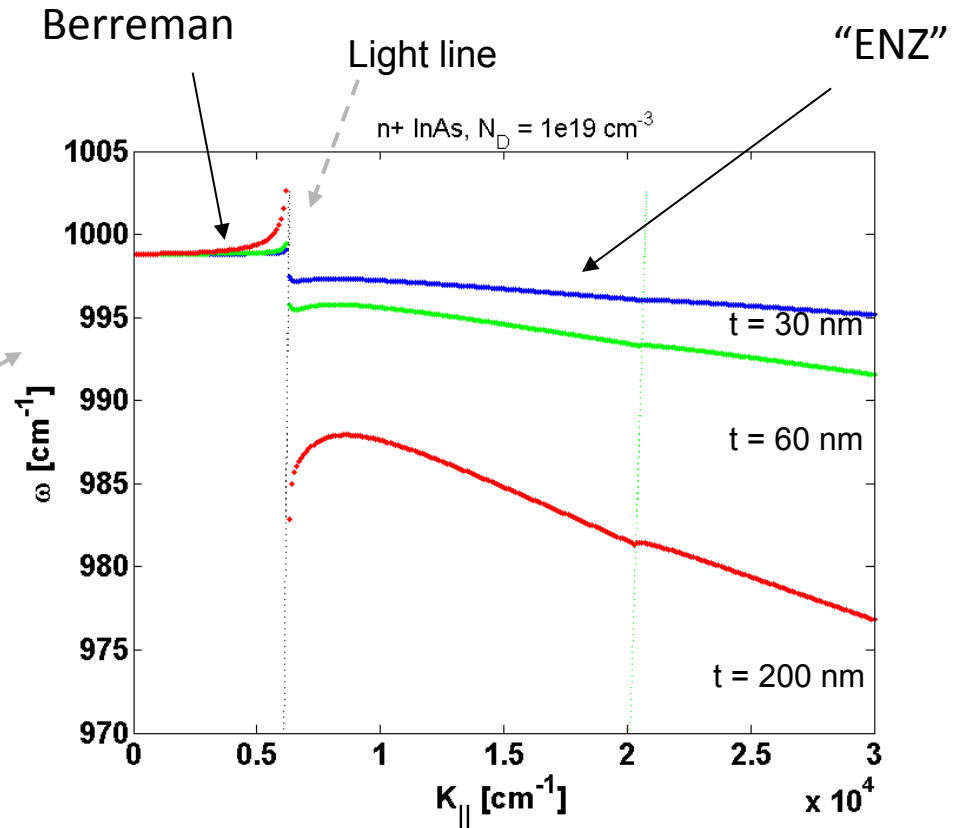
Substr.



Dispersion relation of the TM mode in a 3-layer

$$\left(1 + \frac{\epsilon_1 k_{z,1}}{\epsilon_3 k_{z,3}}\right) = i \tan(k_{z,2}d) \left(\frac{\epsilon_2 k_{z,3}}{\epsilon_3 k_{z,2}} + \frac{\epsilon_1 k_{z,2}}{\epsilon_2 k_{z,1}}\right)$$

Numerical solution in complex domain
(assuming complex freq, real k)

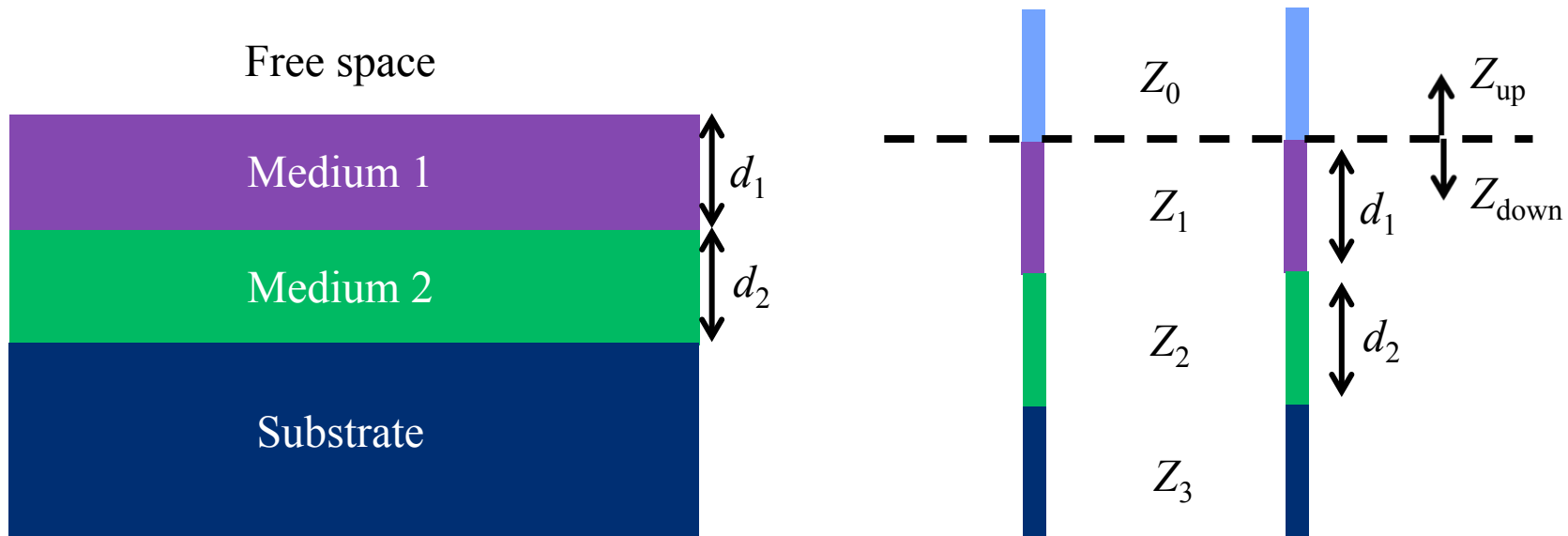


- Berreman mode (leaky mode),
- ENZ mode (bound mode)
- ENZ mode is a special type of a plasmon polariton



Modal analysis

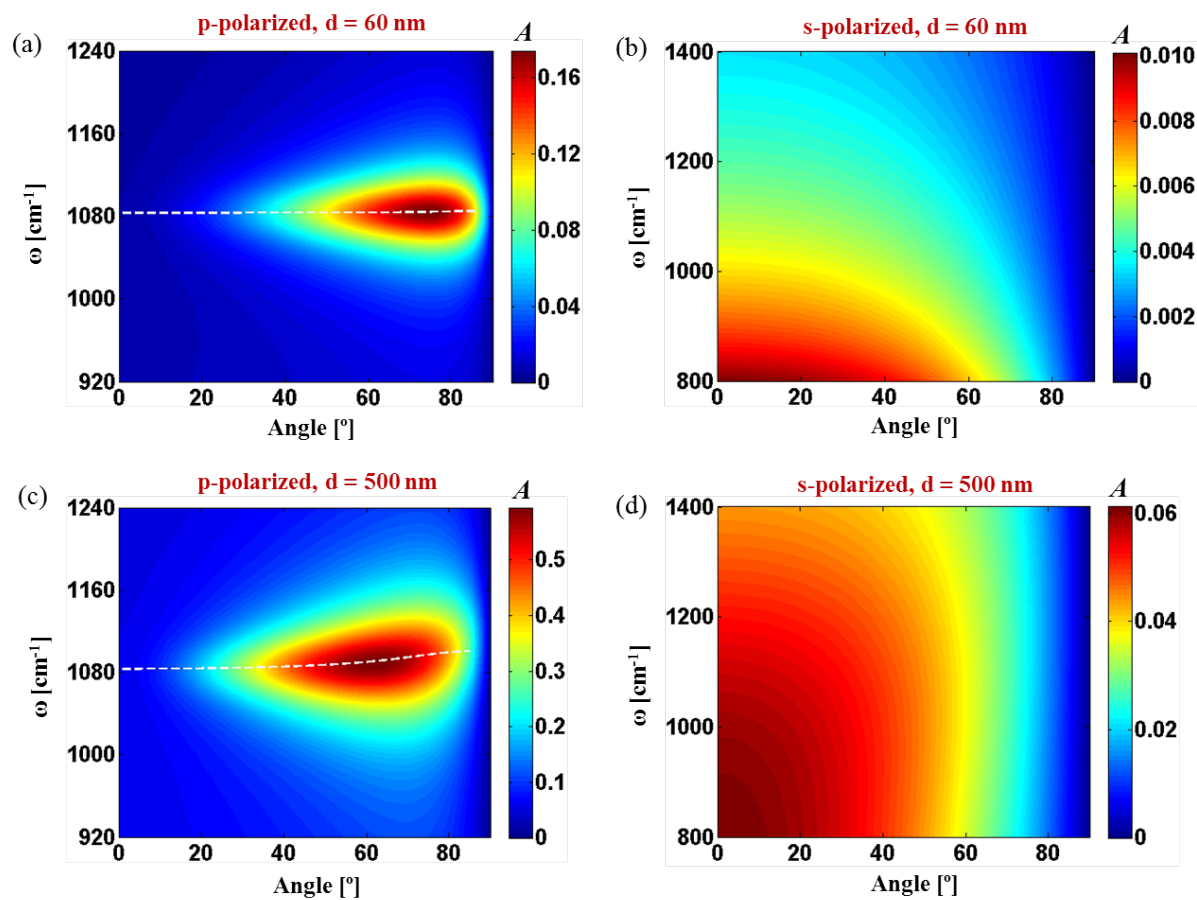
Can be calculated in a different way: The layered medium can be modeled as a set of cascaded transmission lines



Select a reference plane, then compute the impedances looking up and looking down accounting for the dispersion in each medium

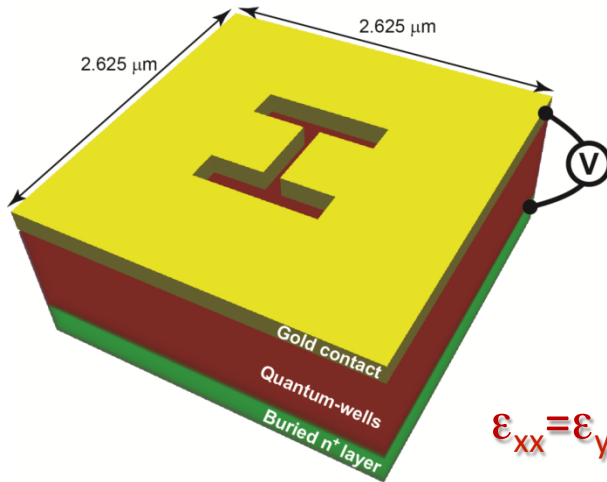
Compute the (complex) modes as the zeroes of

$$Z_{\text{up}} + Z_{\text{down}} = 0$$

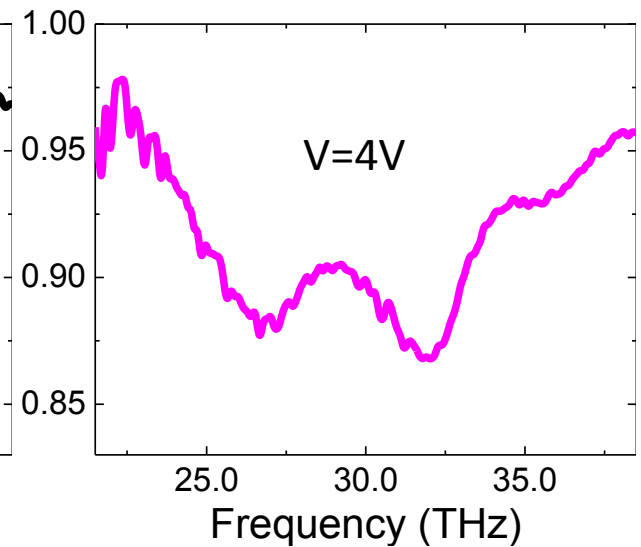
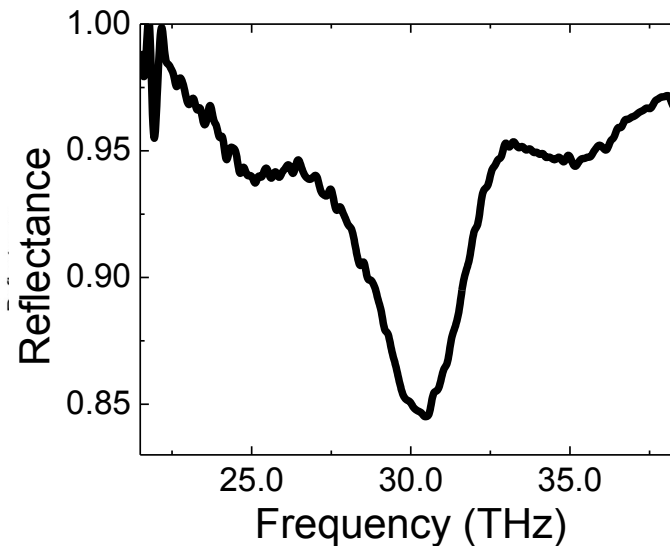
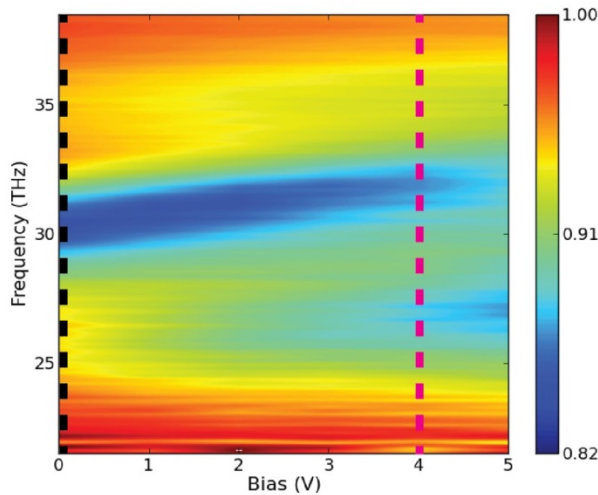
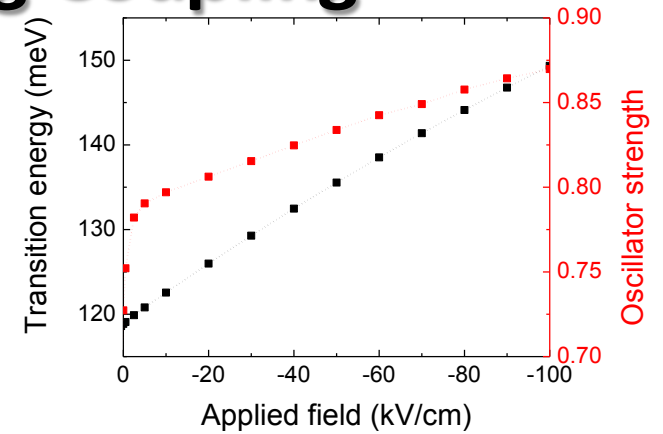
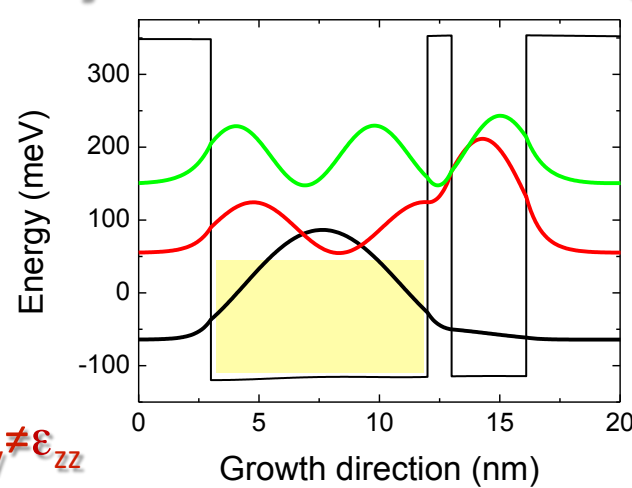




Electrically Tunable Strong Coupling



$$\epsilon_{xx} = \epsilon_{yy} \neq \epsilon_{zz}$$

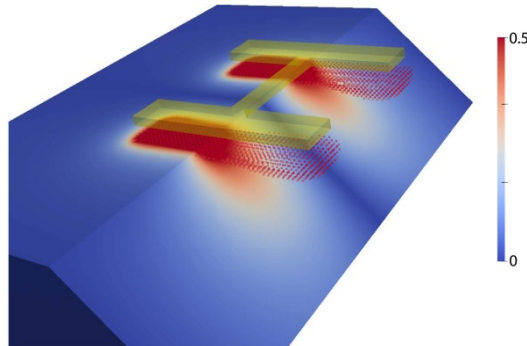
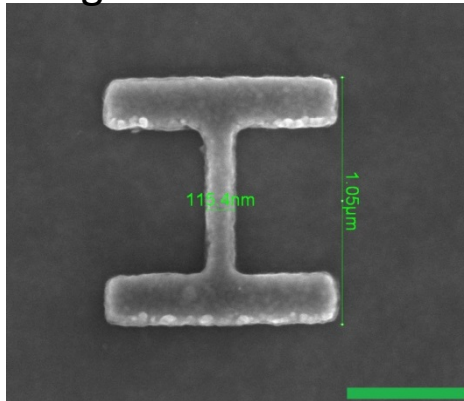


Upper polariton shifted by 2.5 THz (~8% of center frequency)



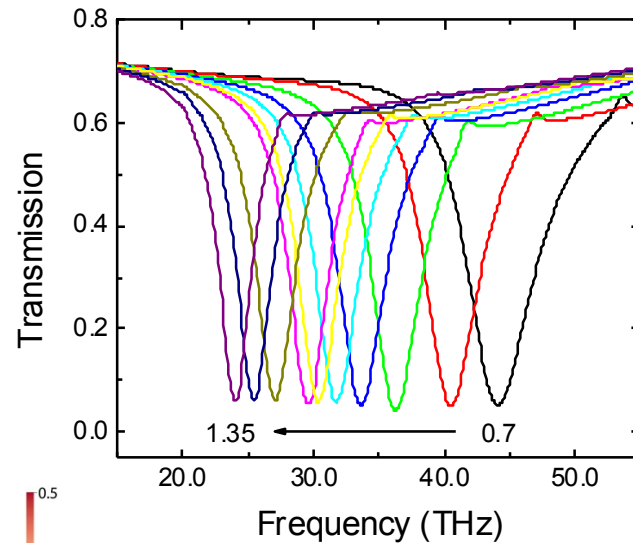
Strong Coupling Theory vs. Experiment

“Dogbone” resonators



Rabi frequency depends on simple parameters

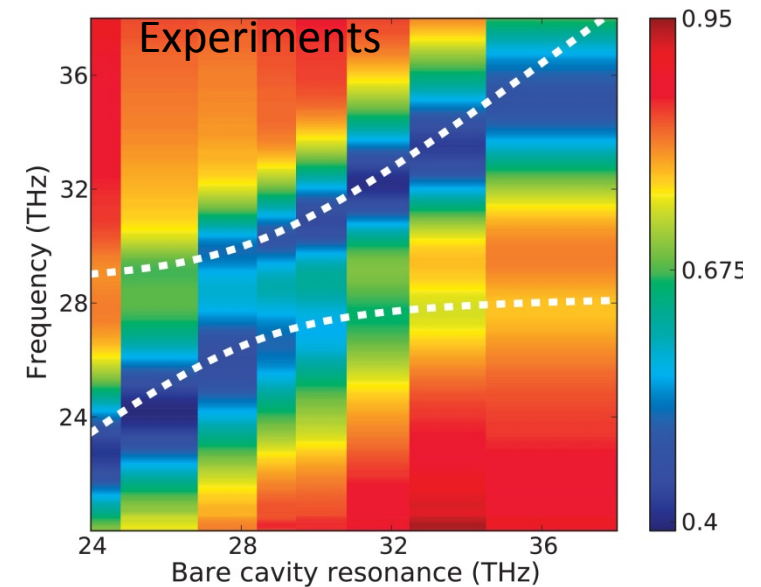
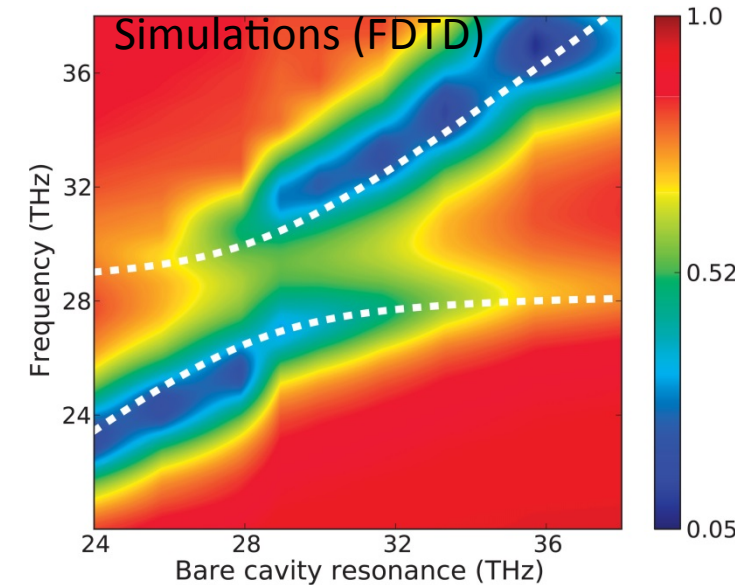
Nature Communications 4, (2013)
Phys. Rev. B 89, 165133



$$\Omega \downarrow R = \sqrt{f \downarrow W} \quad \omega \downarrow p / 2$$

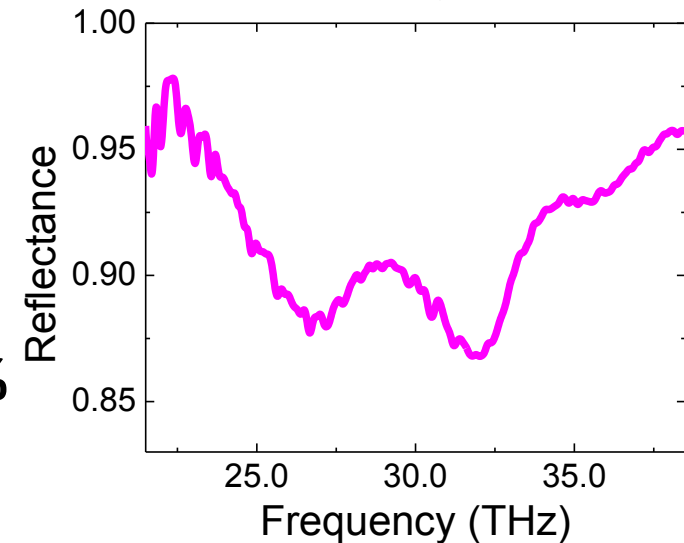
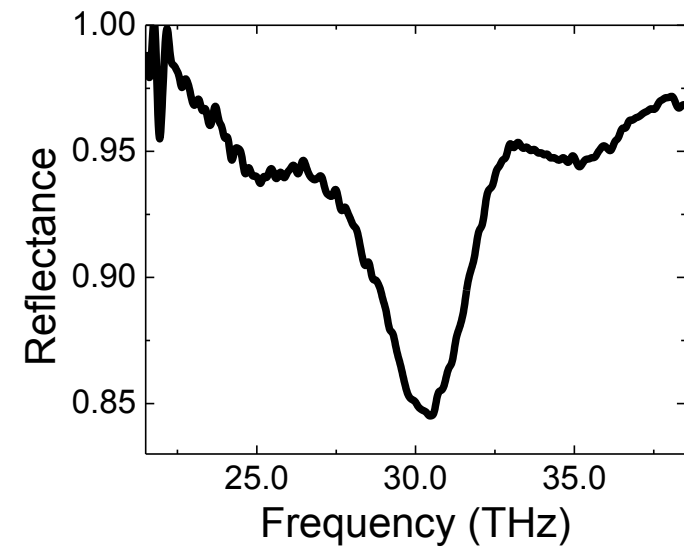
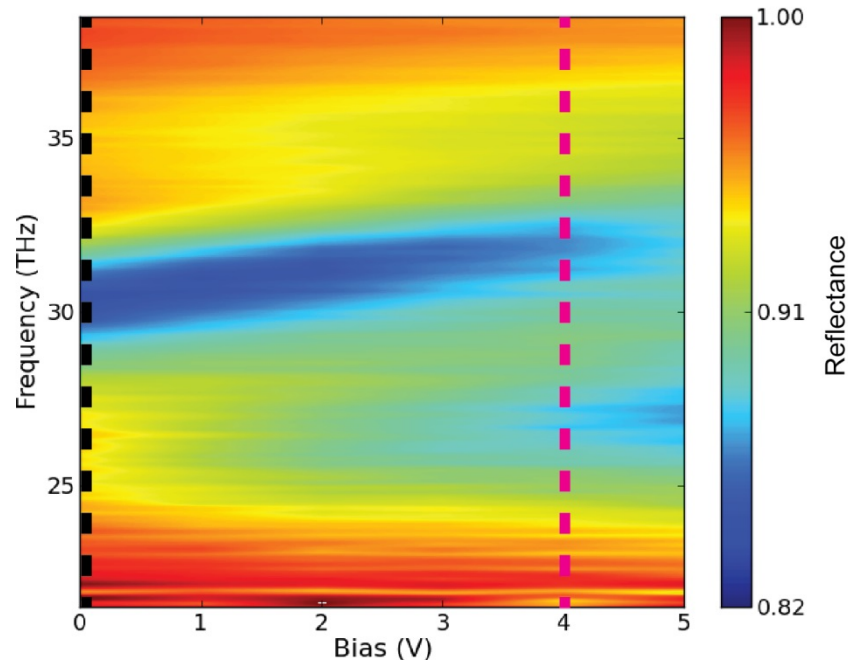
Plasma frequency
Geometry factor

53





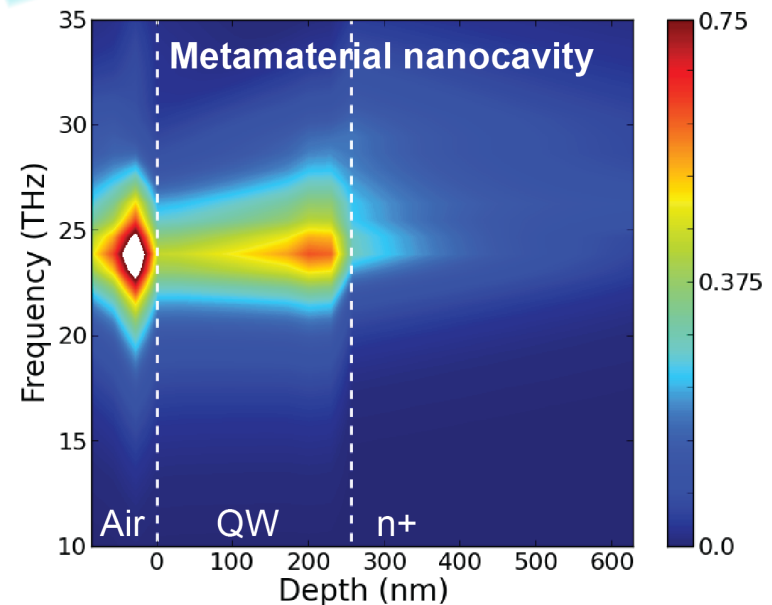
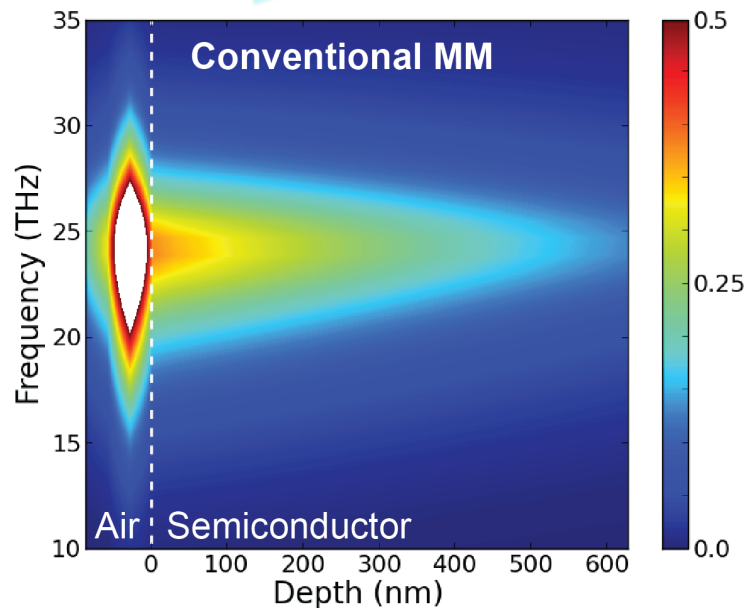
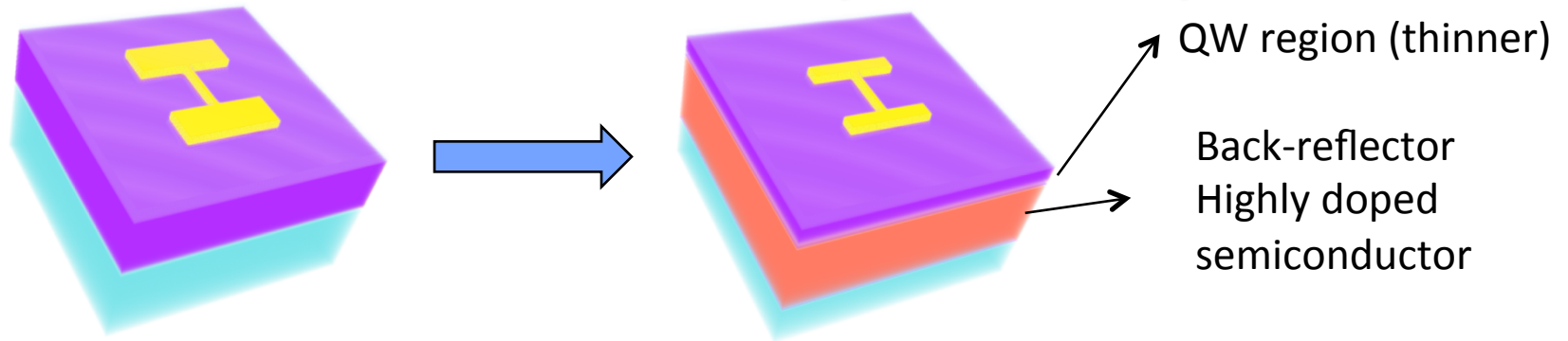
Electrically Tunable Strong Coupling



- Metamaterial spectral response tuned by voltage
- Upper polariton shifted by 2.5 THz (=8% of center frequency or one full intersubband linewidth)



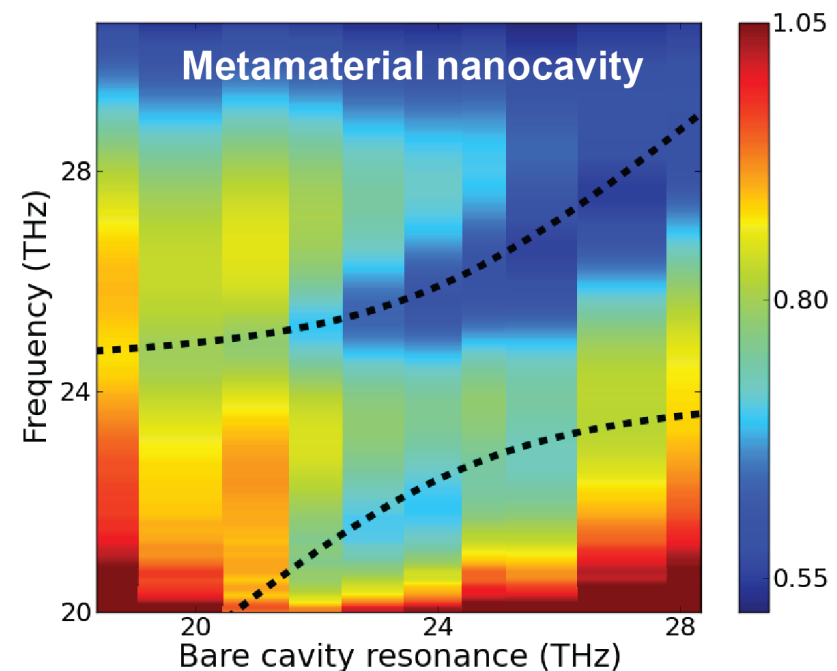
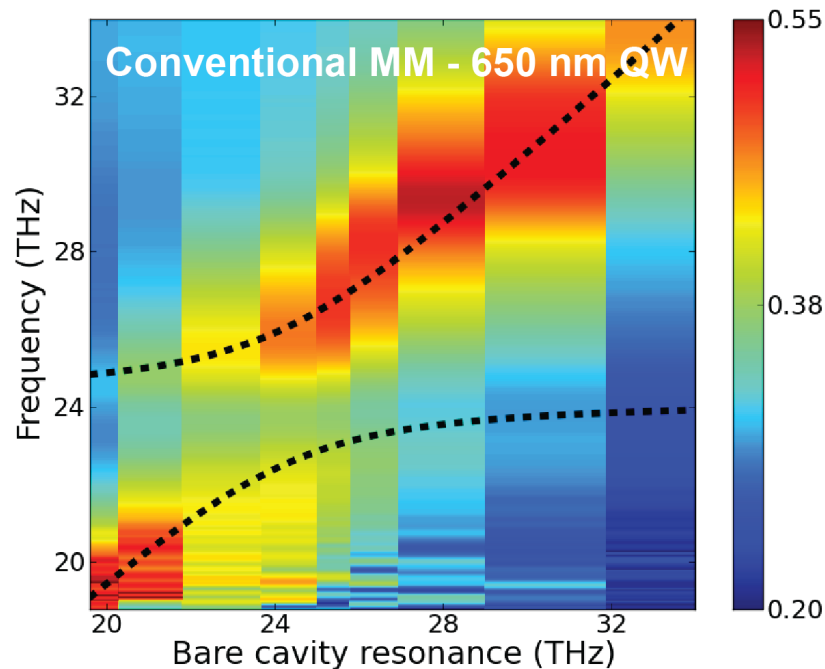
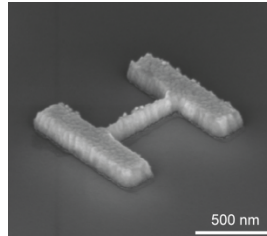
How to Increase the Rabi Frequency: Metamaterial Nanocavity + Ground-plane



- Ground plane limits field penetration into the semiconductor
- Mode volume reduced from: $2.49 \times 10^{-3} (\lambda/n)^3$ to $1.34 \times 10^{-3} (\lambda/n)^3$



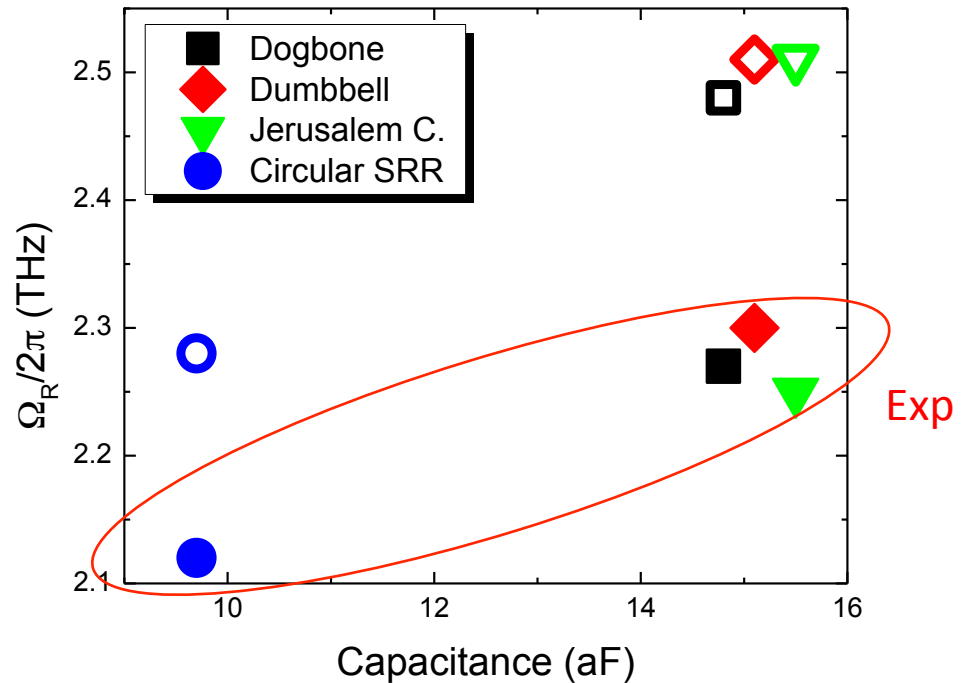
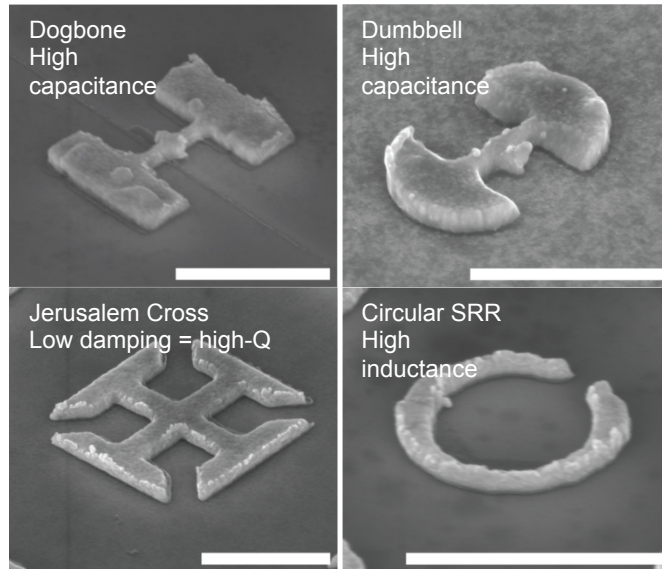
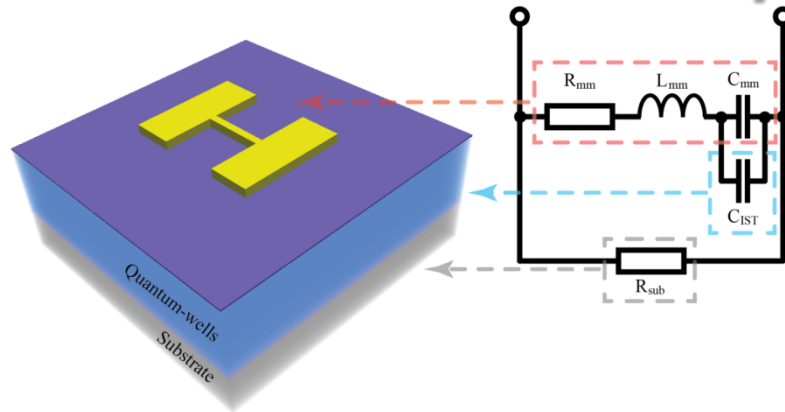
Reduced Mode Volume: Experiments



- Rabi frequency remains almost identical despite reduced QW-stack thickness (factor 3.3) or reduced mode volume (factor 1.9)
- Conventional sample: 2.1 THz vs. Nanocavity: 2.5 THz



Rabi Frequency vs. Geometry



Physical Review **B89**, 165133 (2014)

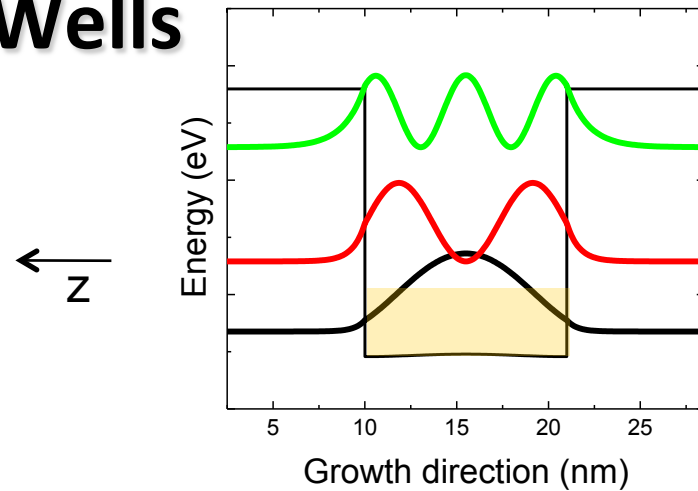
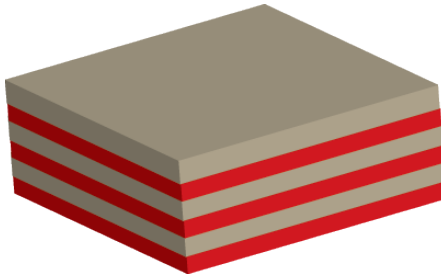
- Larger capacitance leads to larger Rabi splitting



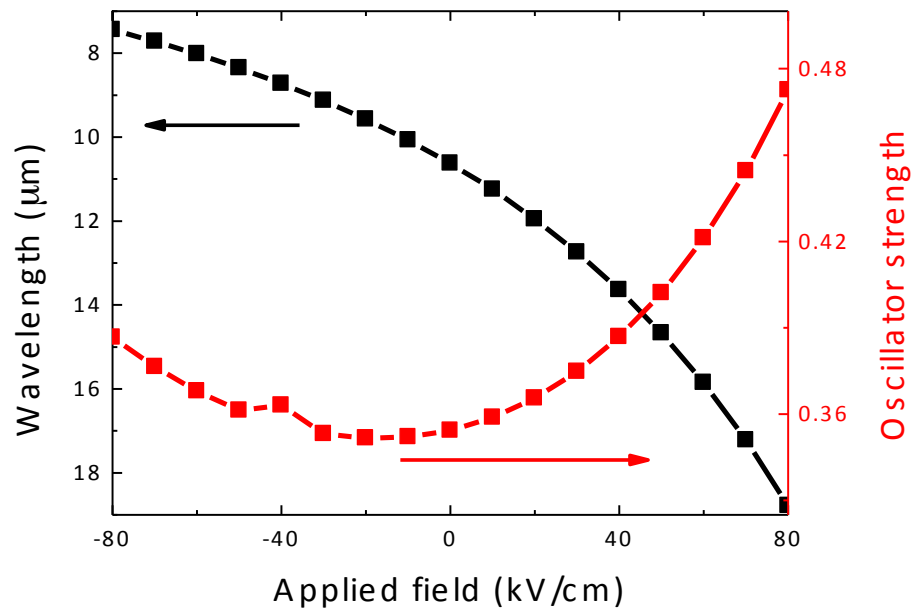
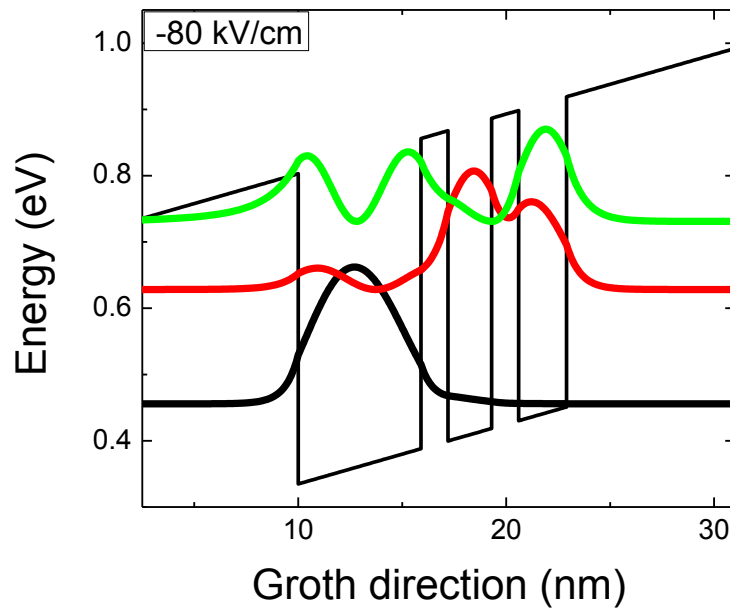
Strong Coupling to Inter-subband Transitions in Quantum Wells

- Scalable (far IR to near IR), Mature, Versatile

Opt. Express 20, 6584 (2012),
APL 98, 203103 (2011)



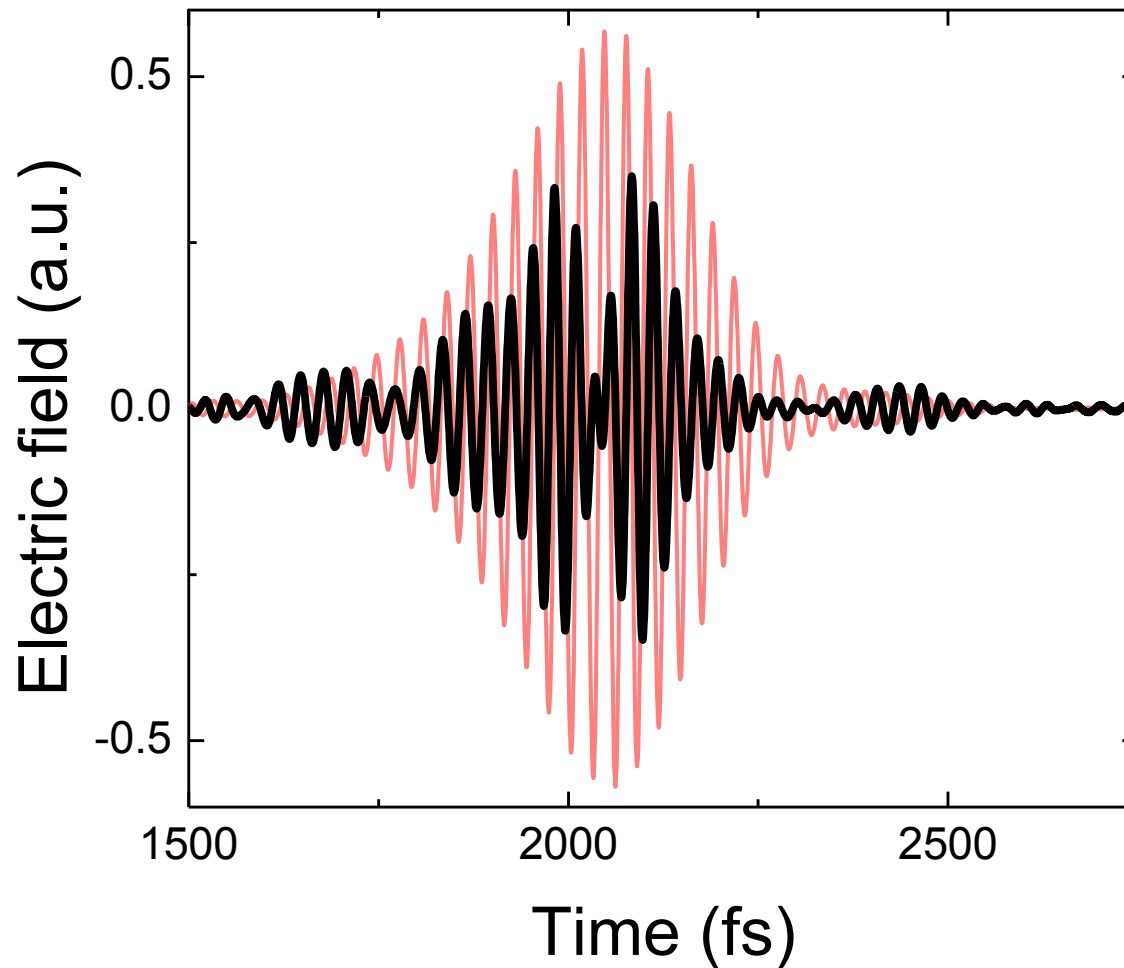
Change levels using an electric field





Experimental Rabi Oscillations

(using a mid-IR Time-domain fsec system)

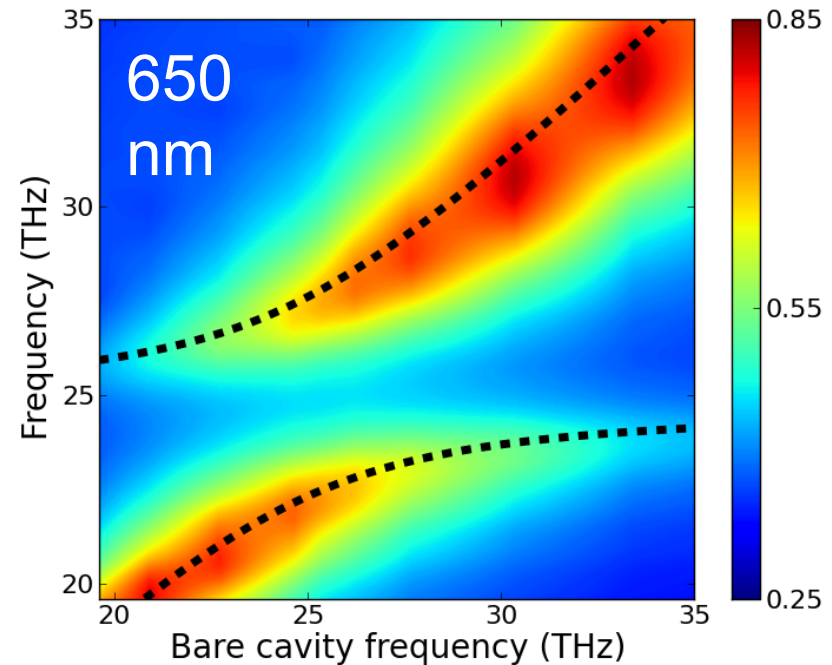
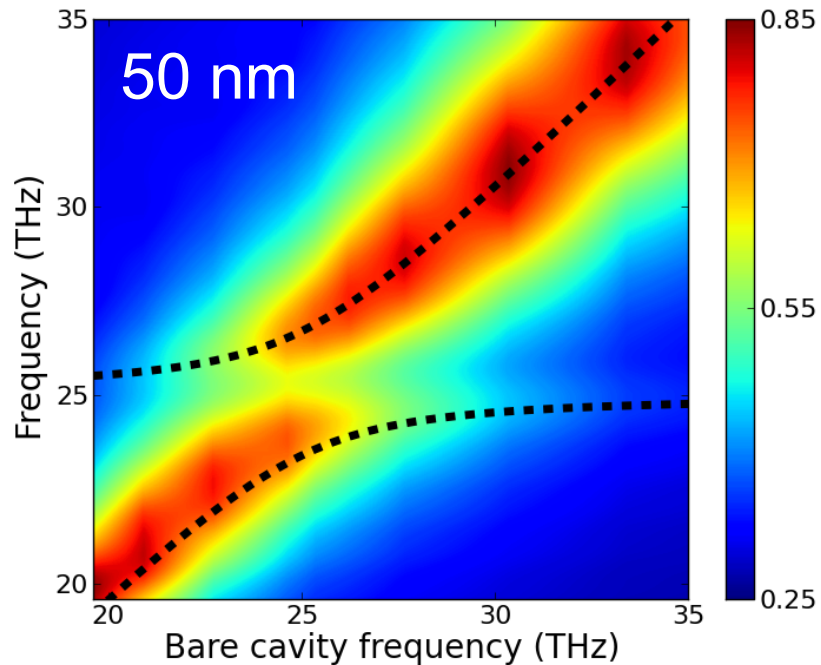


- Energy exchange probed in time
 - 33 fs oscillation
 - 480 fs beating
- System strongly coupled
- Splitting of 4.2 THz measured
 - 15 % of ω_{12}



How to Increase the Rabi Frequency

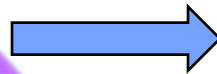
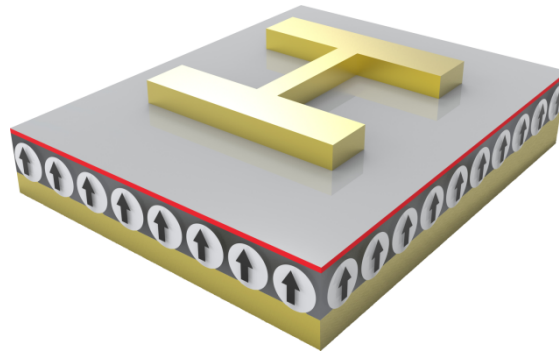
Effect of QW-stack thickness



- Thickness of the QW-stack influences the Rabi frequency
- Increased from 1.65 to 2.64 THz

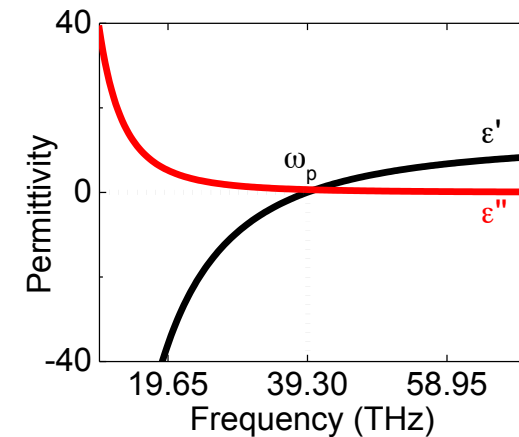


How to Increase the Rabi Frequency: Metamaterial Nanocavity + Ground-plane



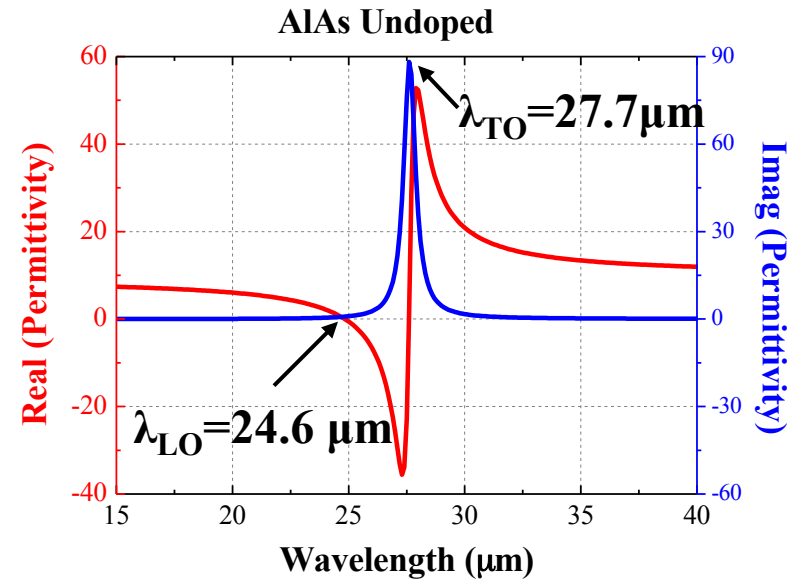
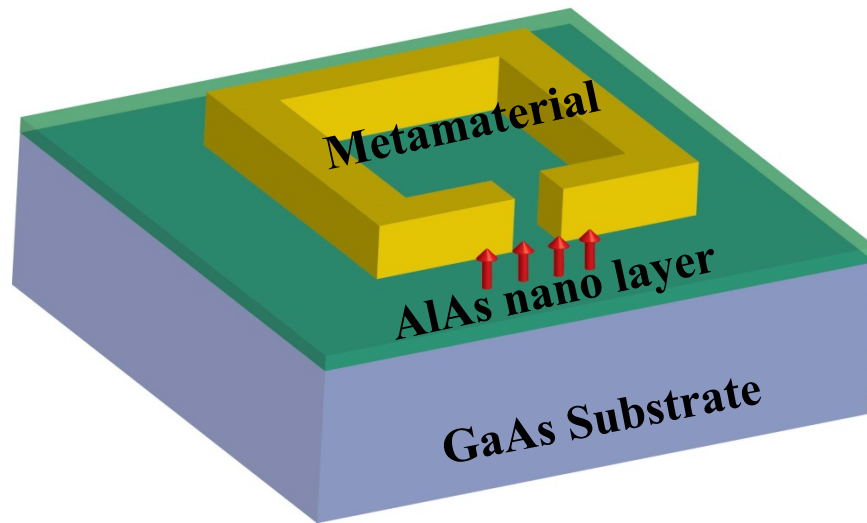
QW region (thinner)

Back-reflector
Highly doped
semiconductor





Coupling of Metamaterial Resonators to Dipole Resonances + ENZ modes



Lorentz model

$$\epsilon = \epsilon_{\infty} \left(1 + \frac{\omega_L^2 - \omega_T^2}{\omega_T^2 - \omega^2 + i\omega\Gamma} - \frac{\omega_p^2}{\omega(\omega - i\gamma)} \right) = (\eta - i\kappa)^2$$

Γ : Phonon damping constant
 ω_p : Plasma frequency
 γ : Free carrier damping constant

One thin layer contains:

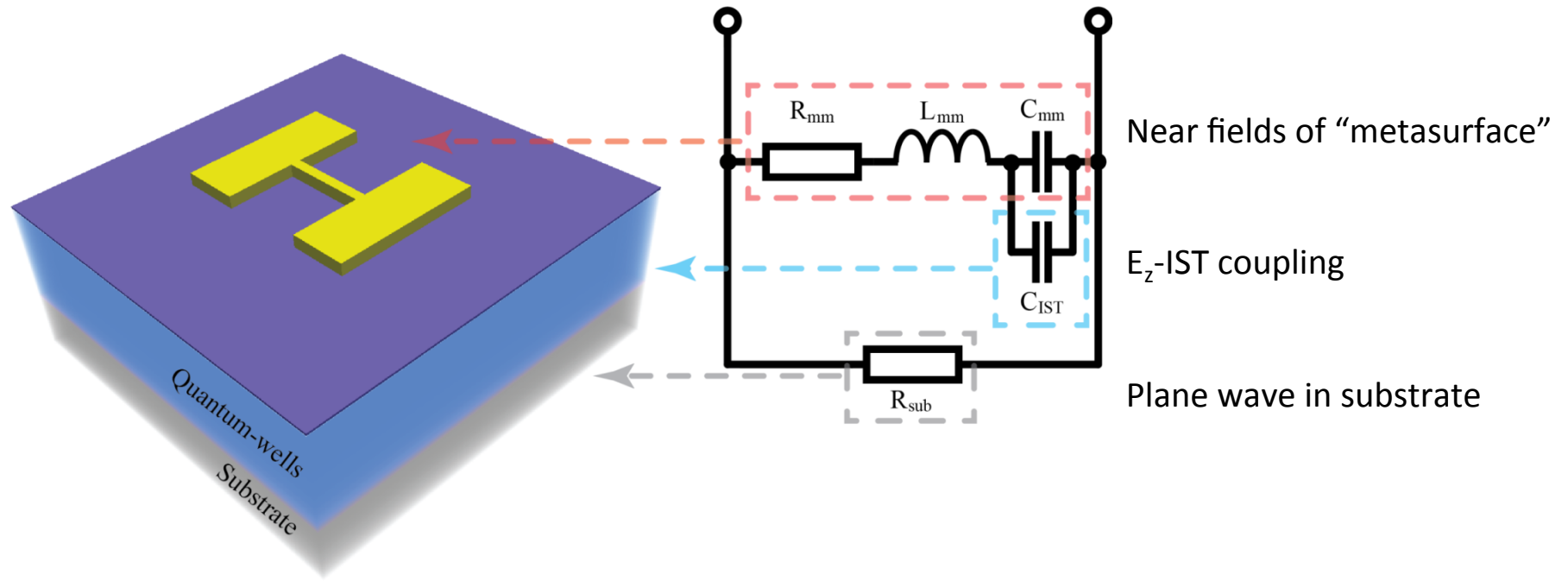
- $\epsilon \sim 0$ crossing $\rightarrow$ ENZ mode
- TO Phonon: “dipole” transition

J. Appl. Phys., **48**, 212 (1976).

Solid State Communications, **62**, 645-647, (1987).



Circuit Model for Strong Coupling



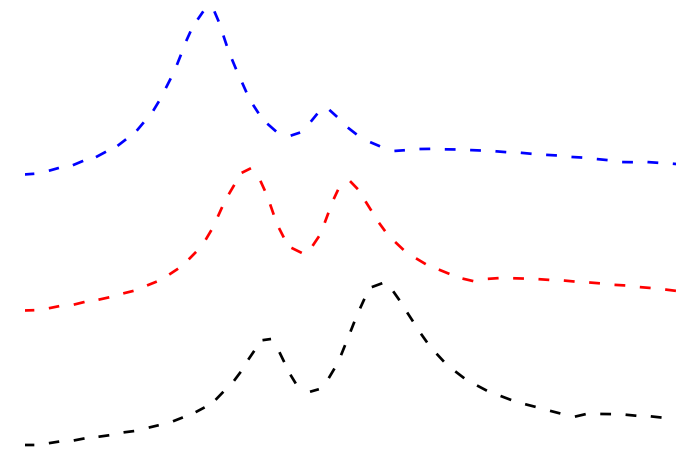
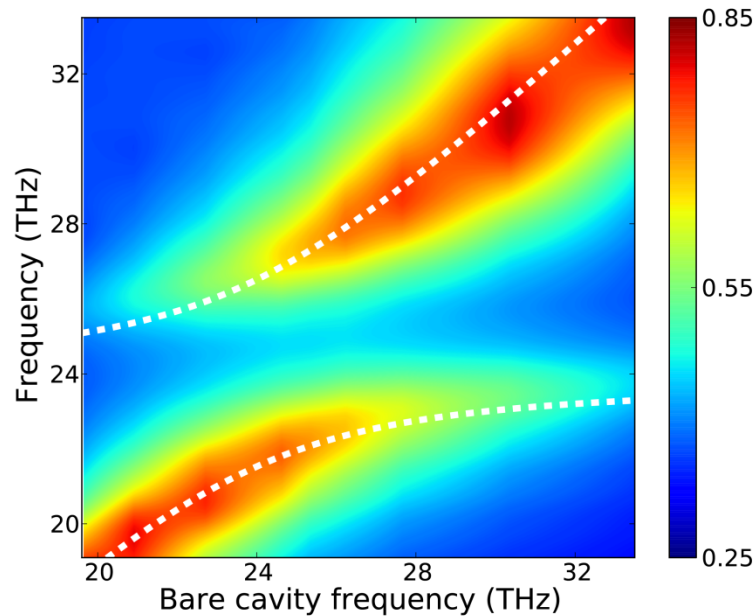
- Metamaterial modelled as simple RLC harmonic oscillator
- Strong coupling represented by dispersive capacitor
- No free fitting parameters for dispersive capacitor

$$\epsilon_{\downarrow IST}(\omega) = \epsilon_{\downarrow back} + \Delta N_{\downarrow 12} e^{i\phi} / \epsilon_{\downarrow 0} m^* f_{\downarrow 12} /$$

$$C_{\downarrow IST}(\omega) = \sqrt{\epsilon_{\downarrow back} \epsilon_{\downarrow IST}(\omega) - \epsilon_{\downarrow back}} / \epsilon$$



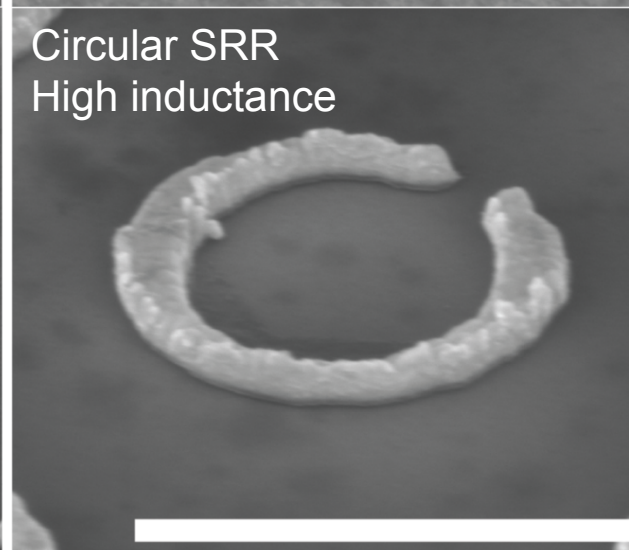
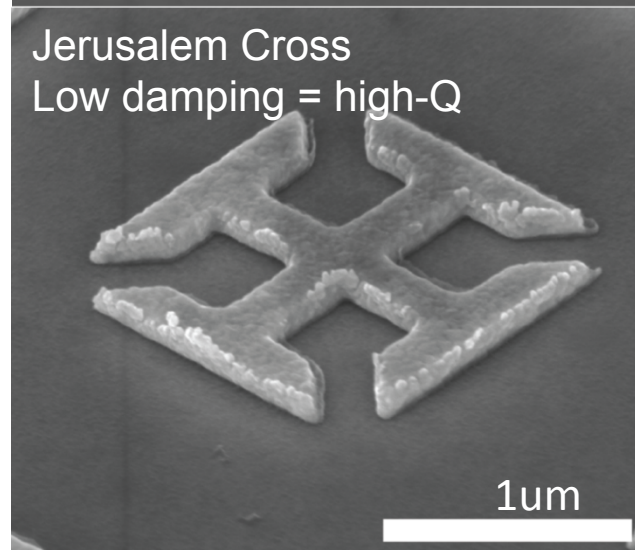
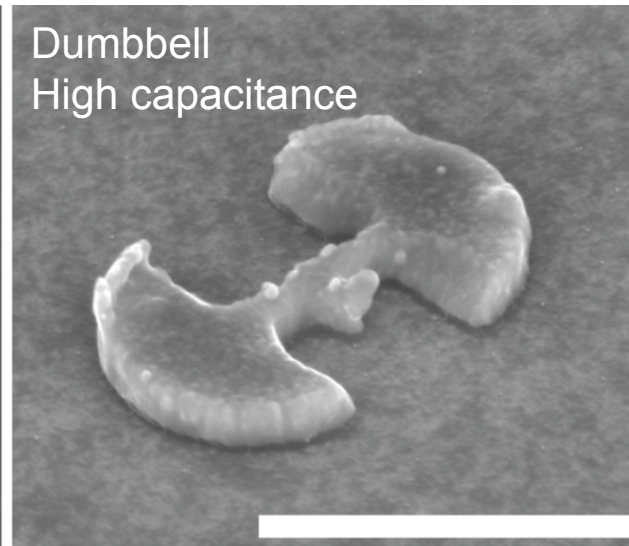
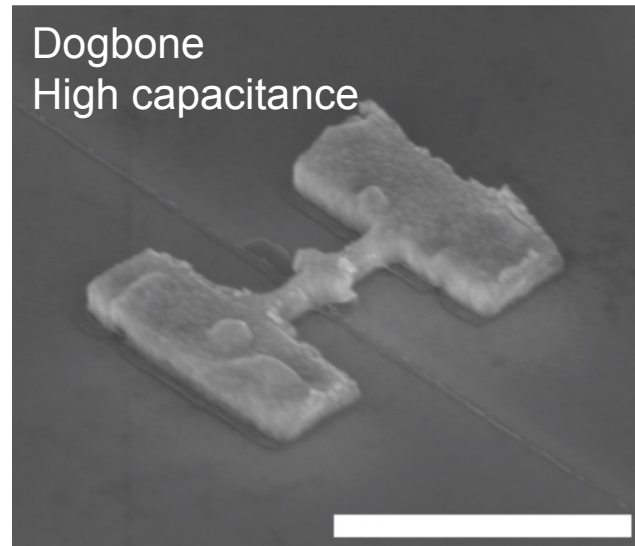
Accuracy of circuit model



- FDTD simulations show clear anti-crossing as a function bare cavity frequency
- Circuit model can recover full lineshape of reflectance (or transmittance) spectra



Resonator Geometries Analyzed

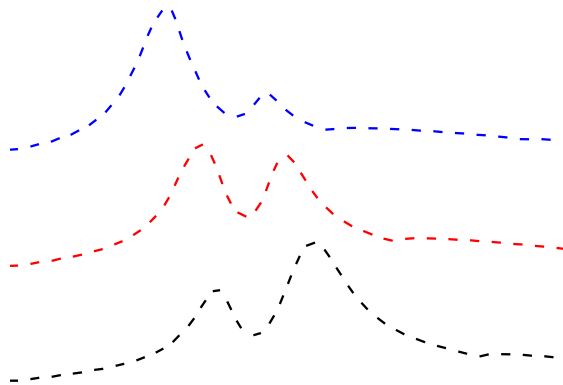


- All metamaterial have identical bare cavity resonances but different RLC values
- Metamaterial defined by electron-beam lithography
- Same QW-stack used for all samples
- Study only geometry effect

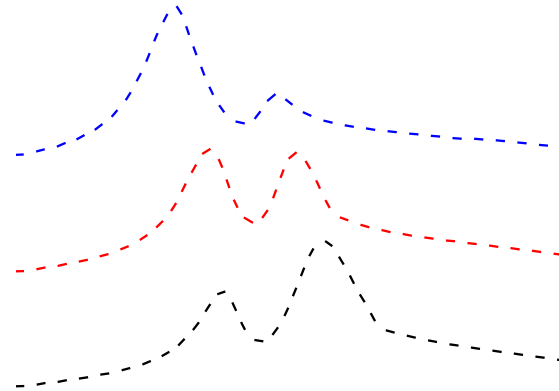


Circuit model accuracy

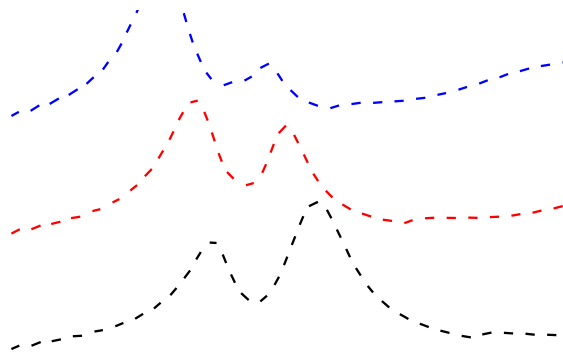
Dogbone



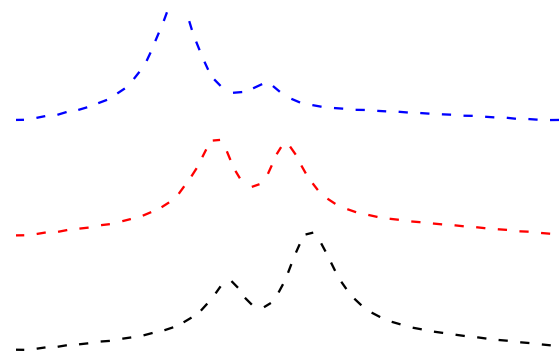
Dumbbell



Jerusalem
Cross



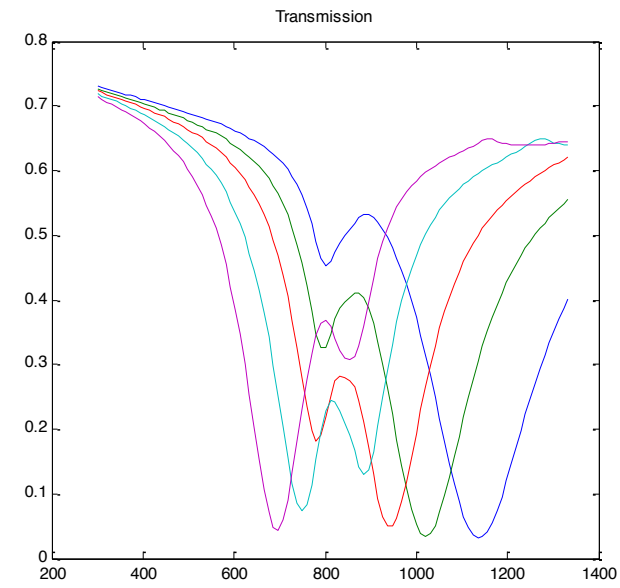
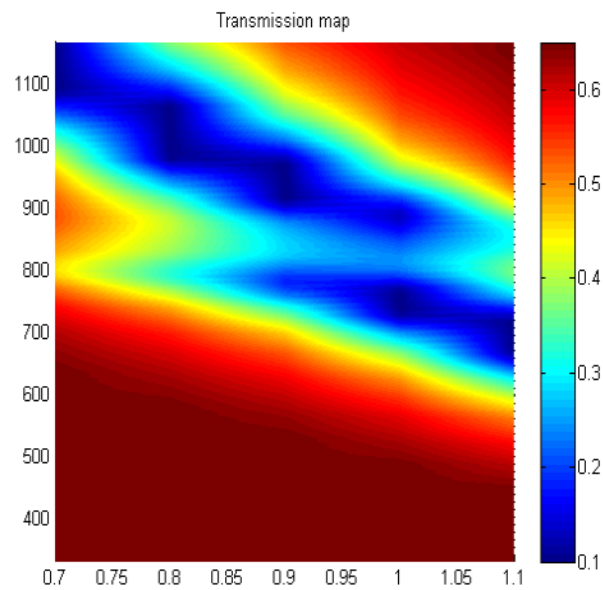
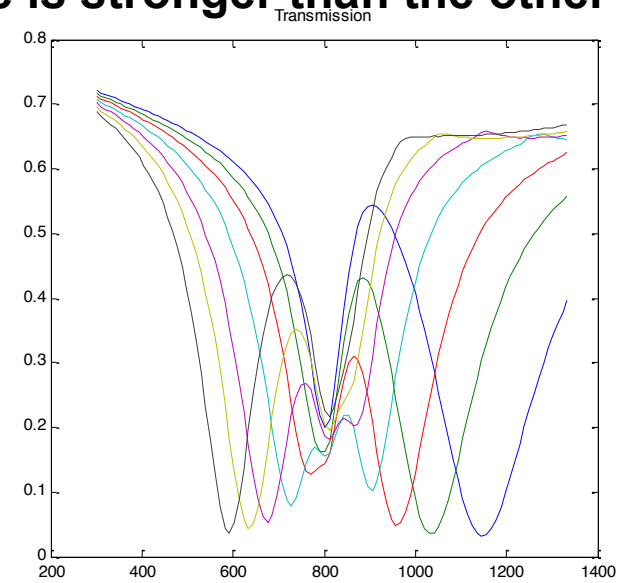
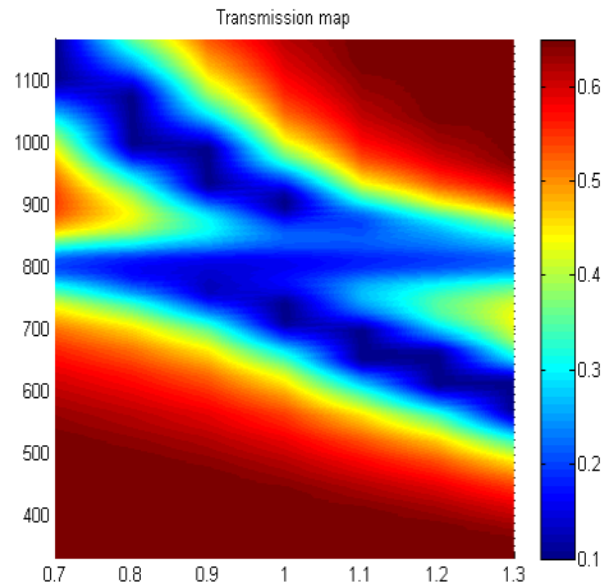
Circular SRR



- RLC model describes strong coupling accurately despite the exact geometry
- Only cold cavity requires fitting



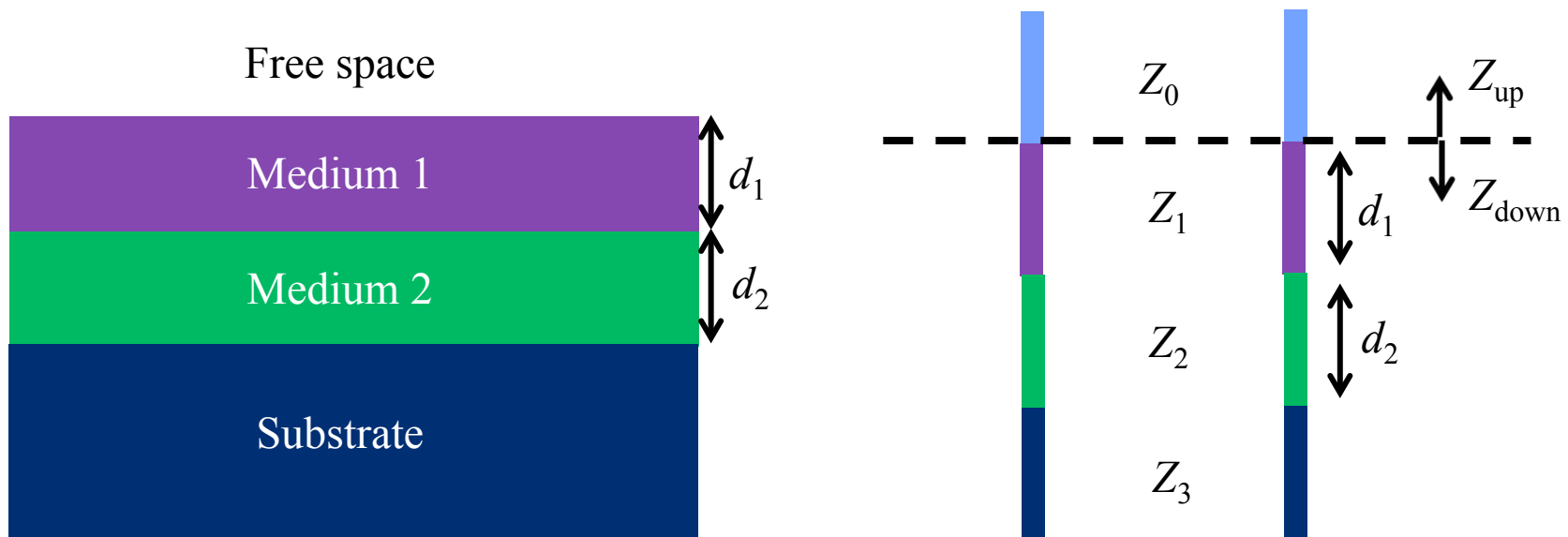
Top: Two isotropic Lorentzians; one is stronger than the other



Bottom: Two anisotropic Lorentzians; one is stronger than the other

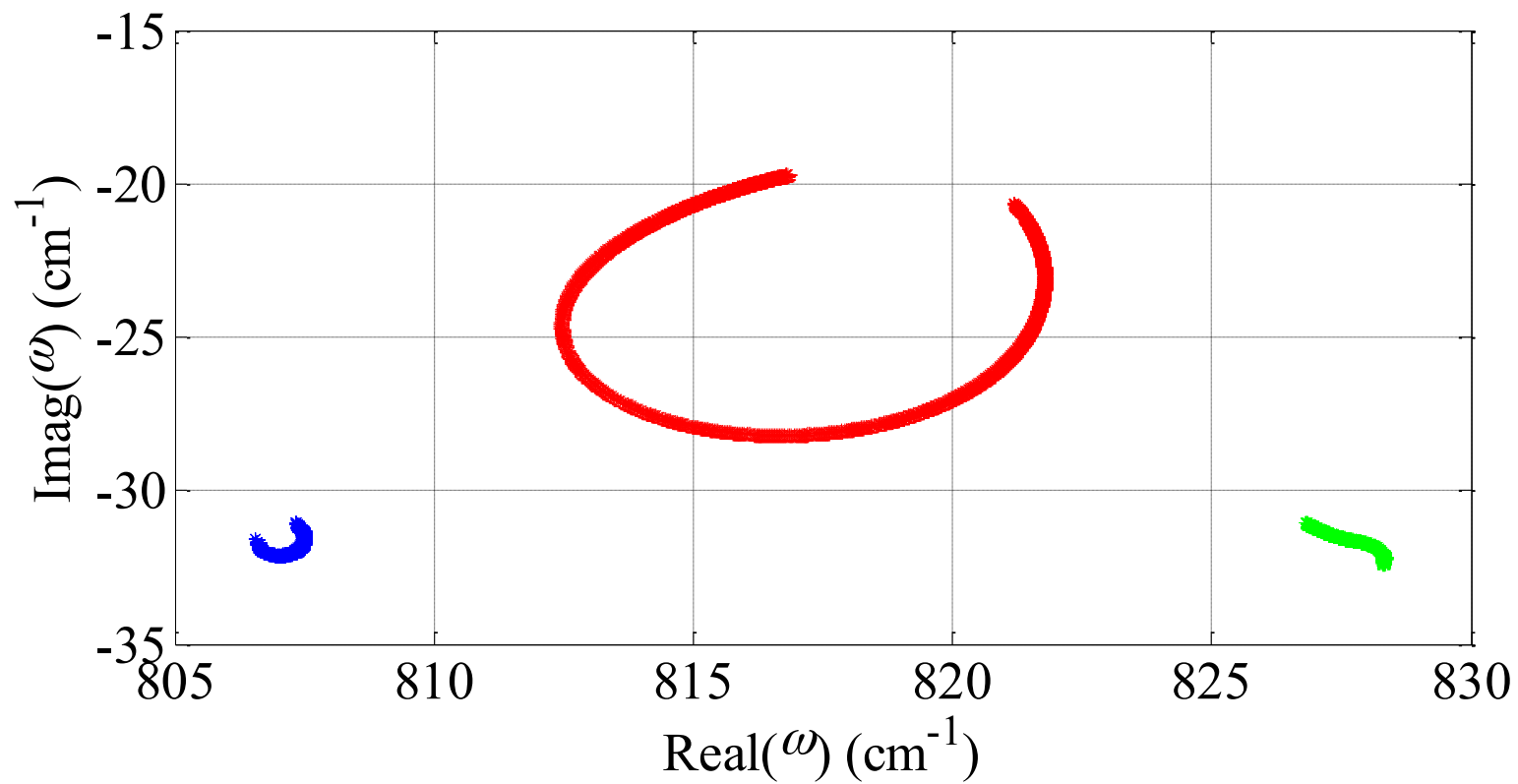
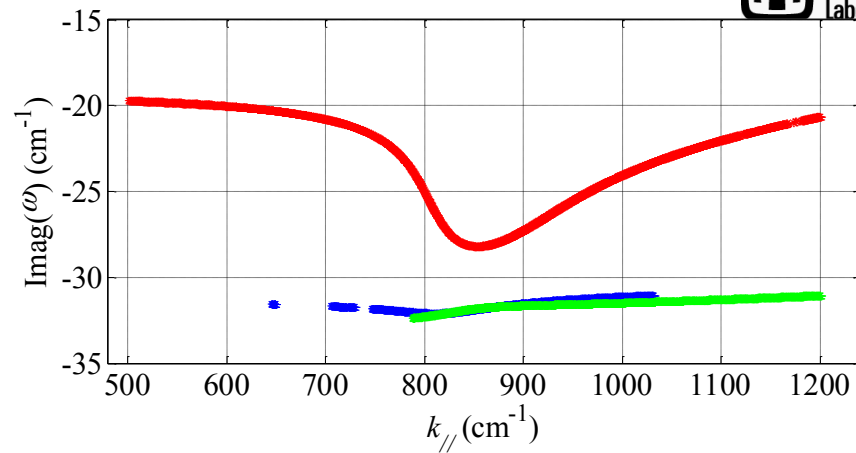
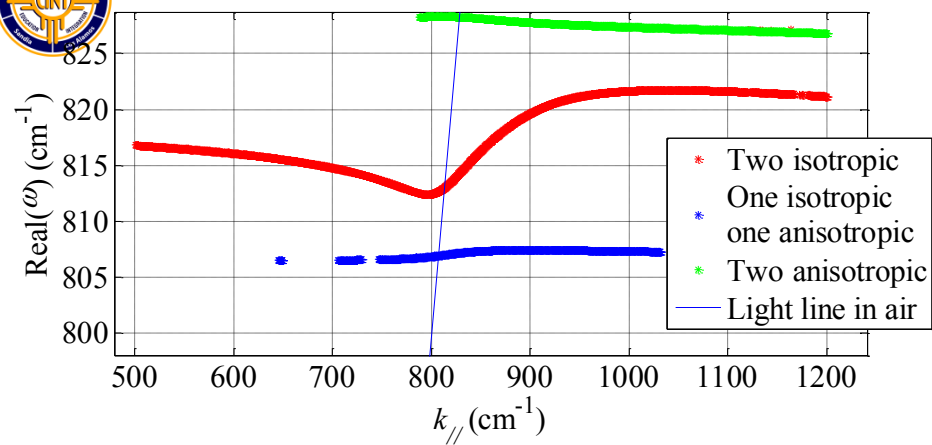
Modal analysis

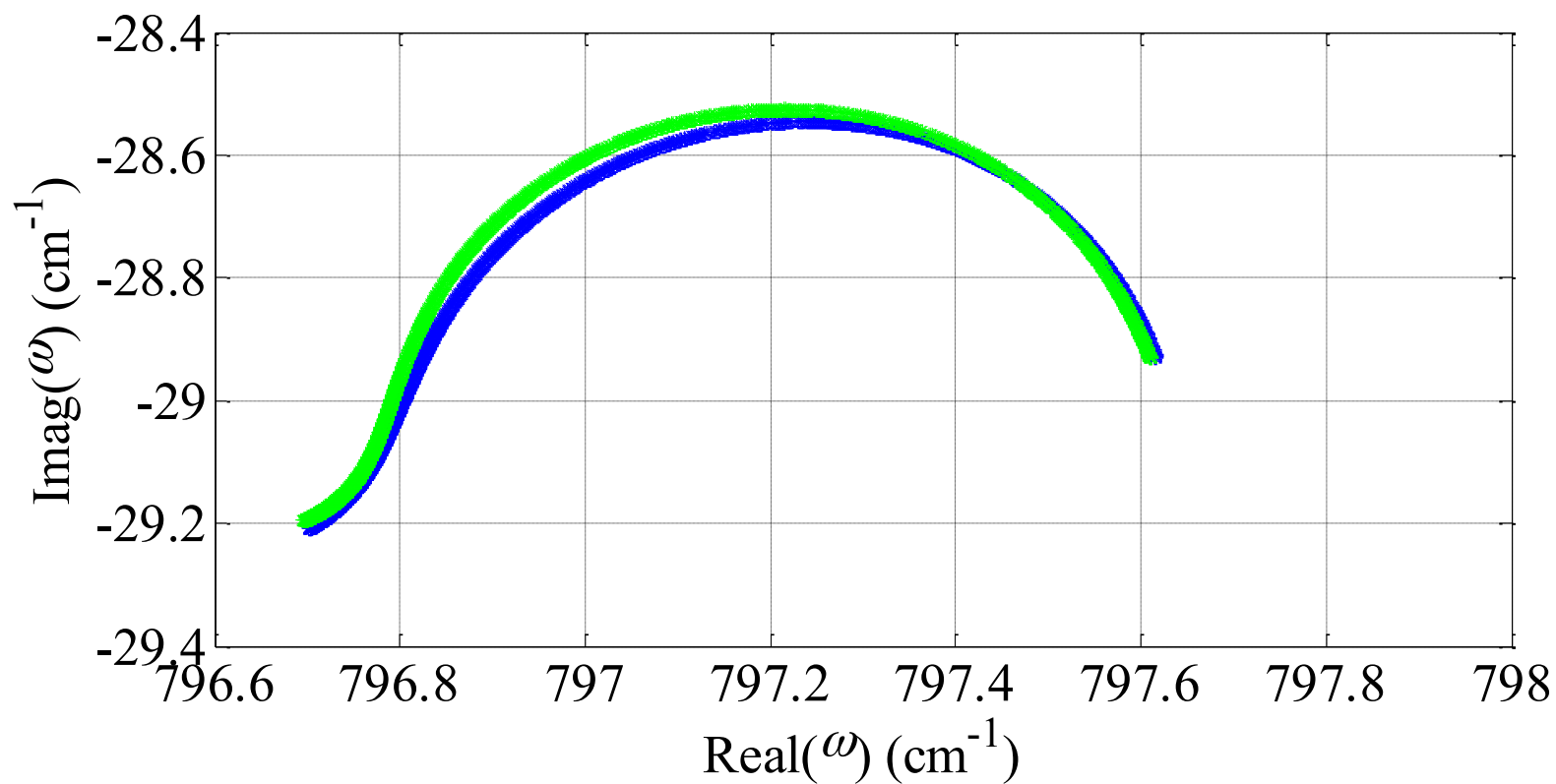
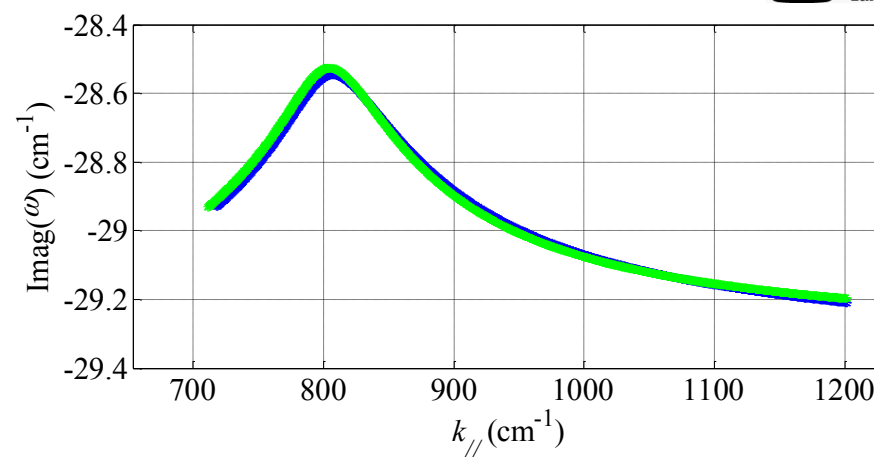
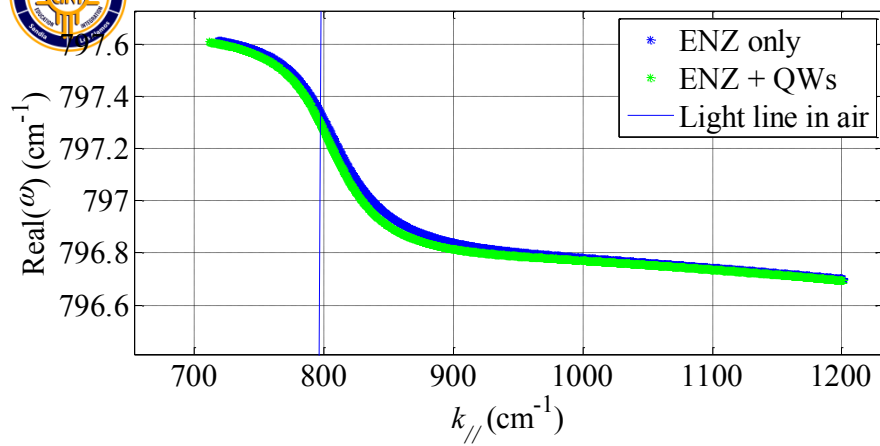
The layered medium can be modeled as a set of cascaded transmission lines



Select a reference plane, then compute the impedances looking up and looking down accounting for the dispersion in each medium

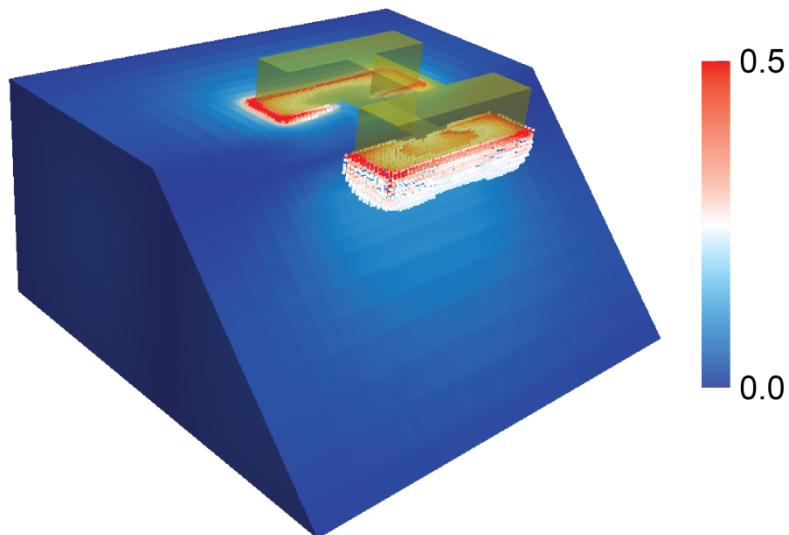
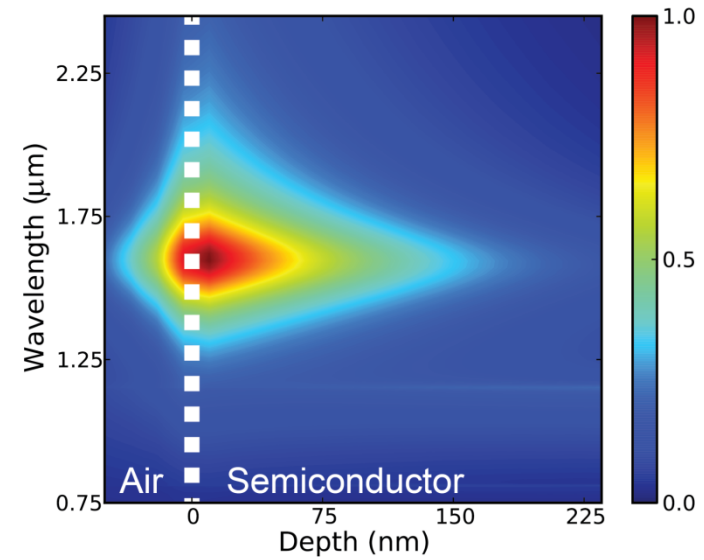
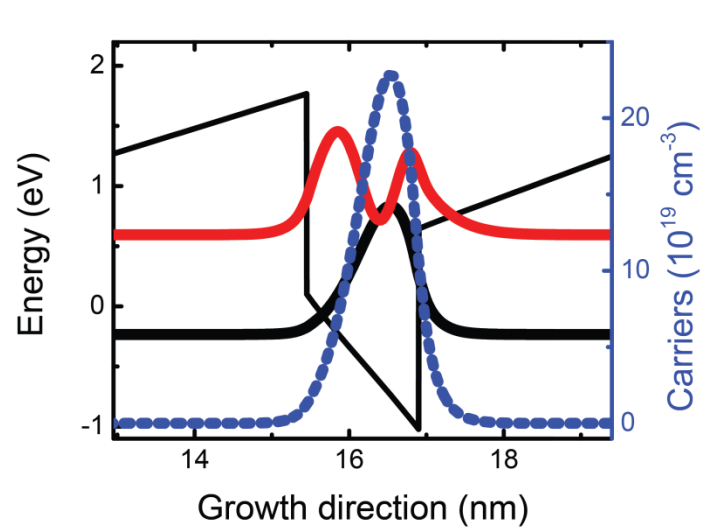
Compute the (complex) modes as the zeroes of $Z_{up} + Z_{down} = 0$





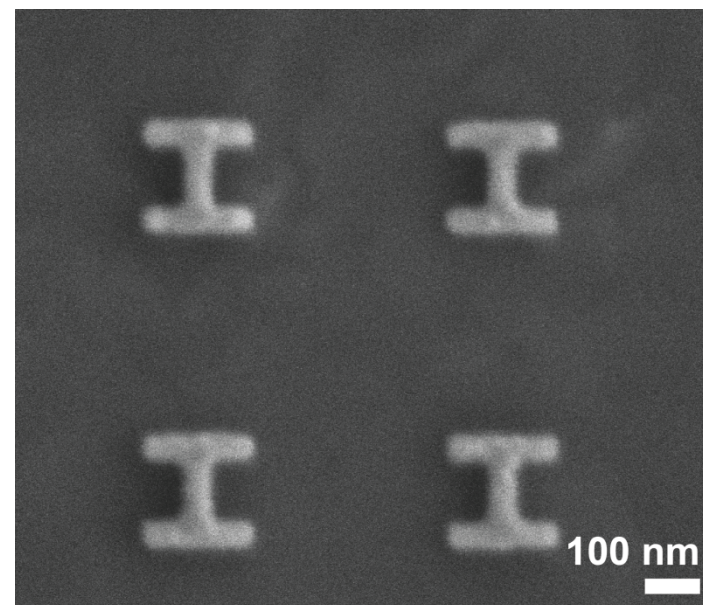
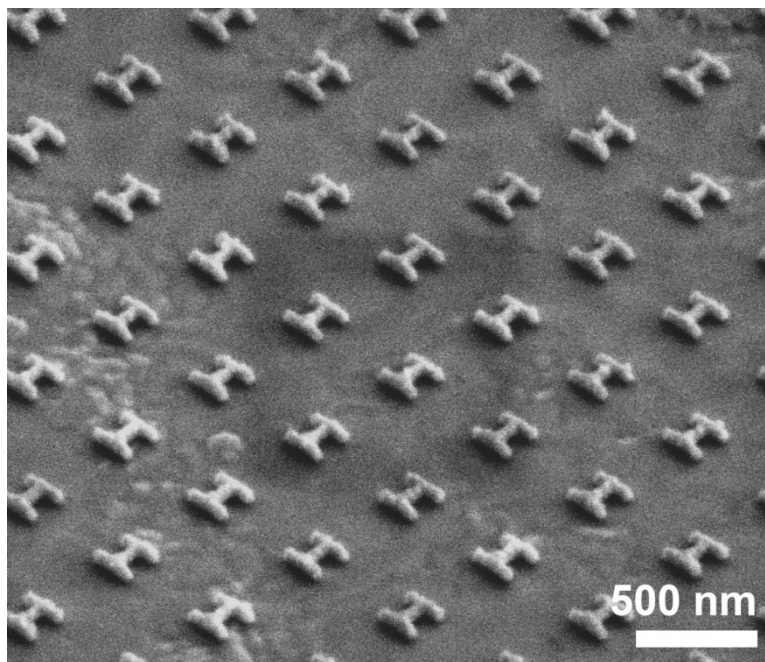


Strong Coupling, Near IR



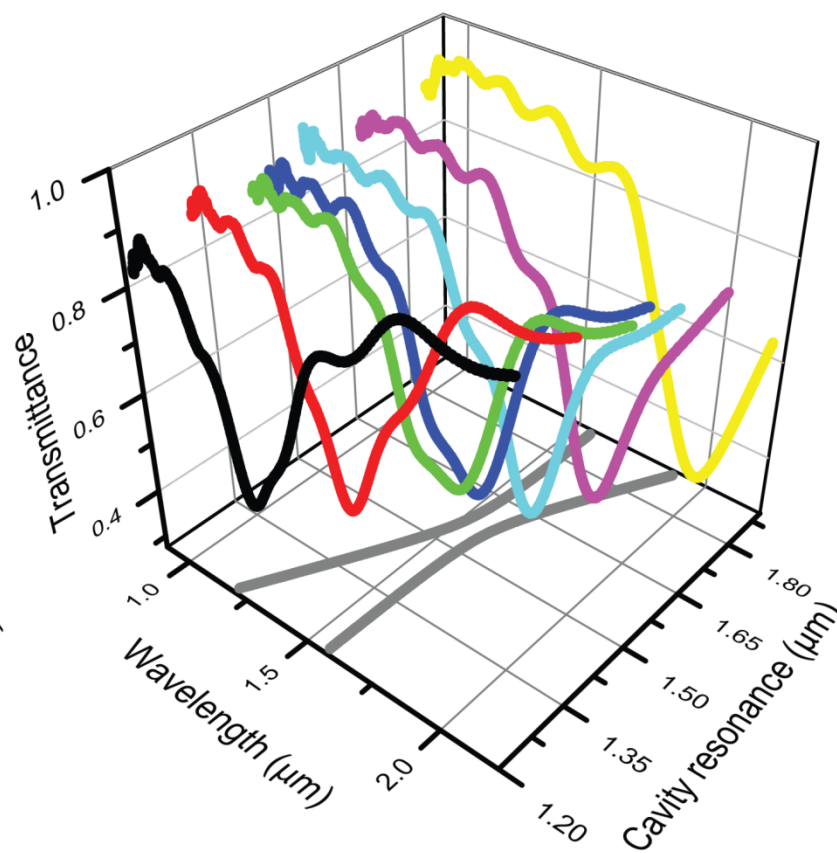
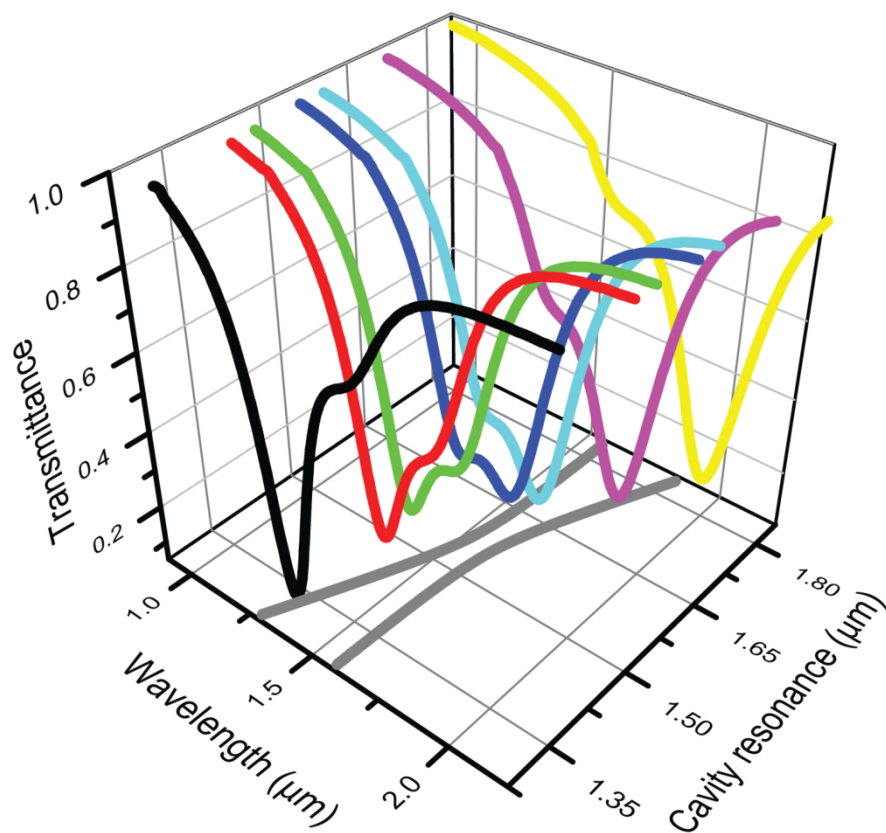


Dogbone Resonators, Near IR



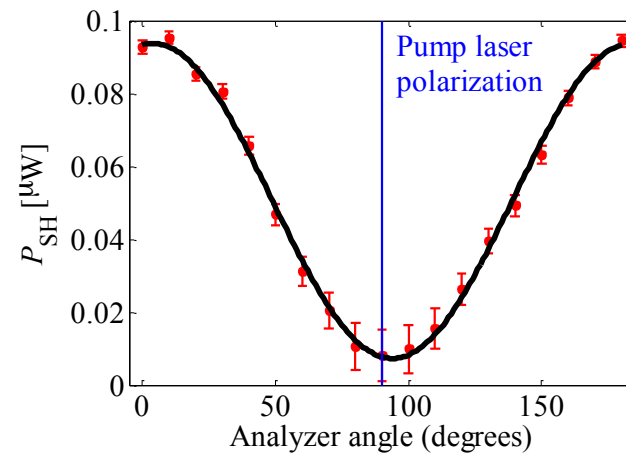
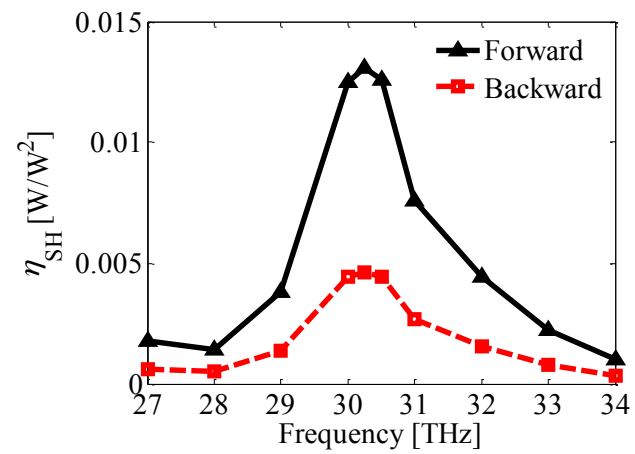
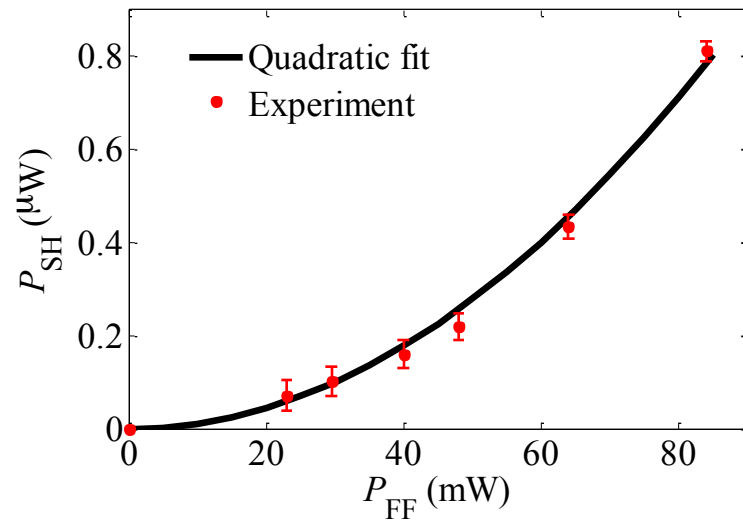


Strong Coupling, Near IR





SHG



Appl. Phys. Lett. **104**, 131104 (2014)