

# Nonlinear optics of epsilon near zero material using CdO

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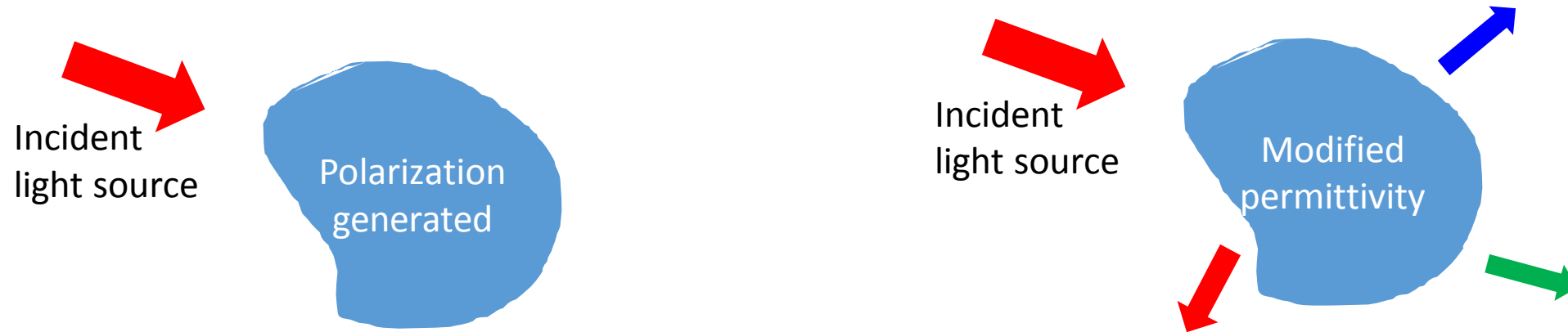
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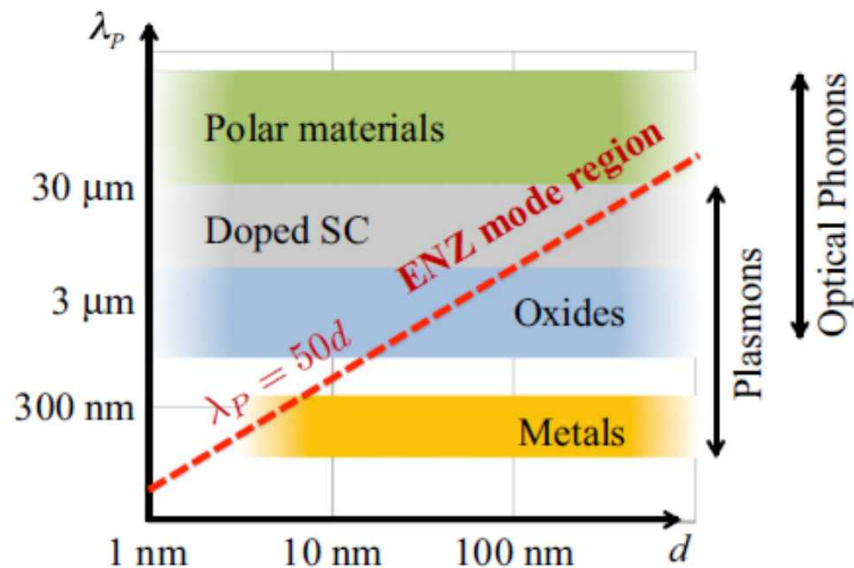
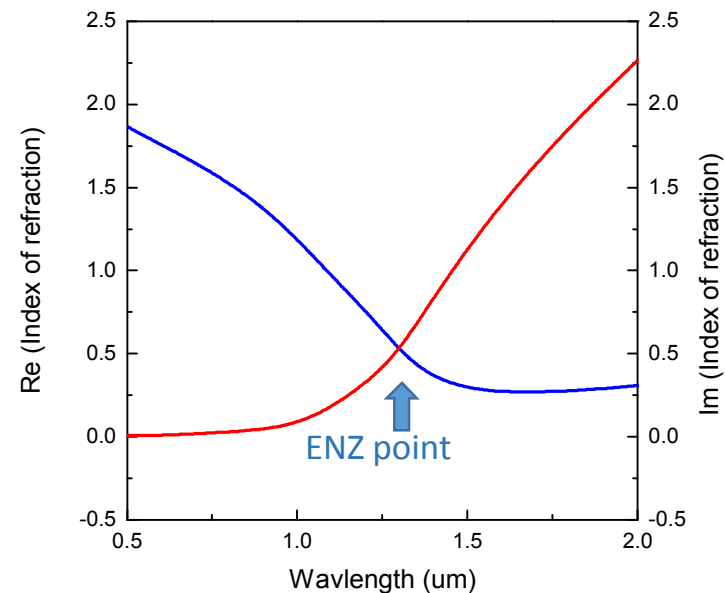
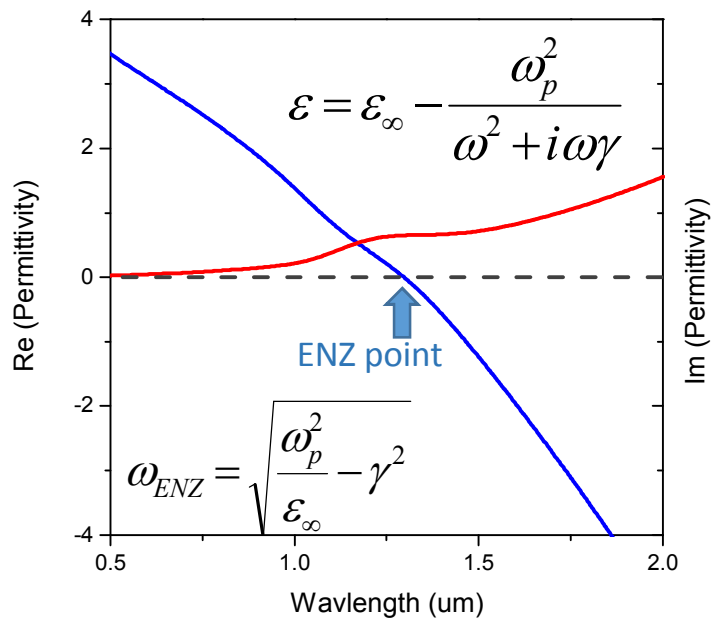
Gordon Keeler (*SNL*)

# Strong Light matter interaction starts with absorption



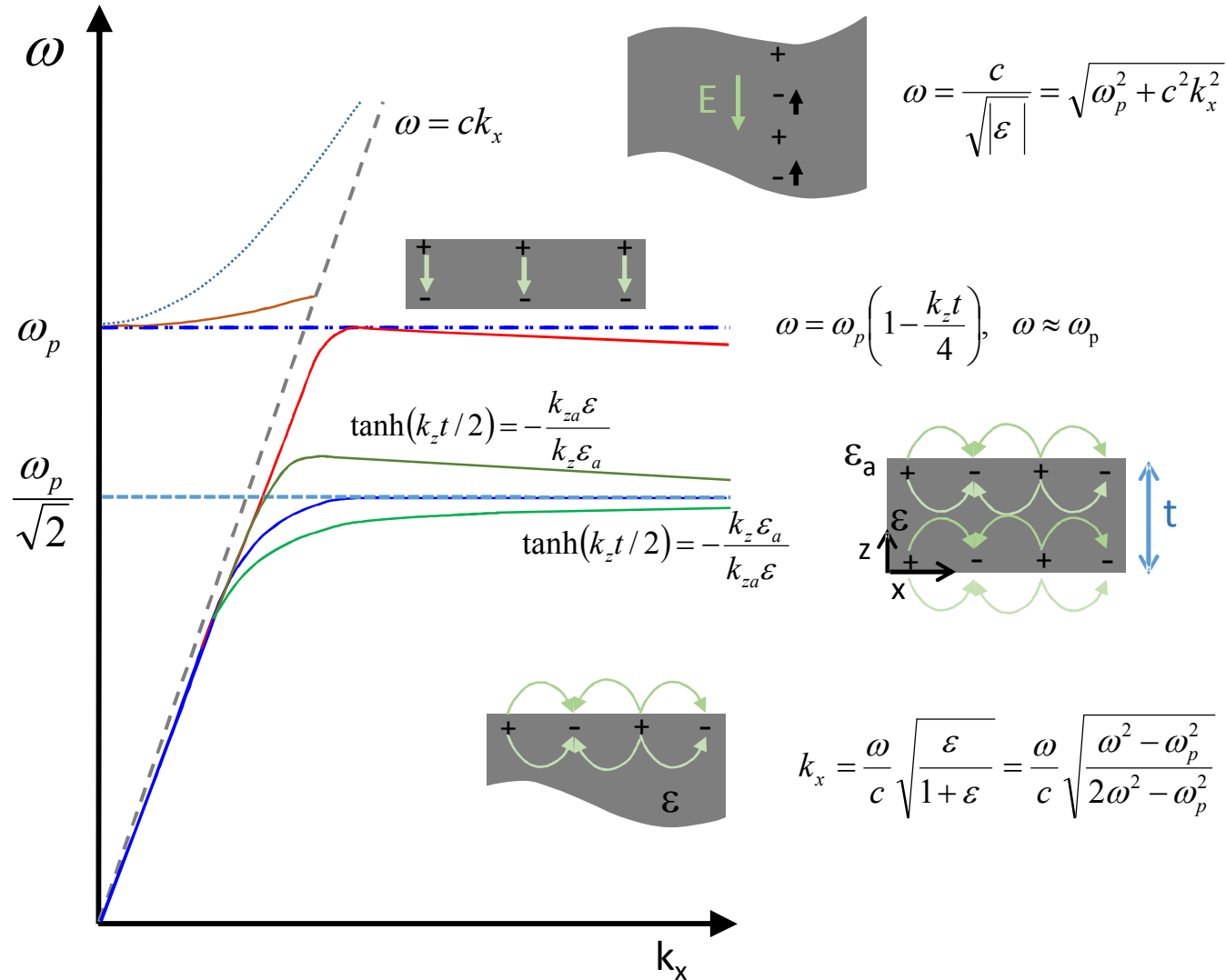
- Maximum absorption produces maximum polarization
- Free electrons are very polarizable:  $\text{Re}(\epsilon)=0$  at plasma frequency
- ENZ modes enable complete absorption
- Optical interaction can induce large change in permittivity
- Polarization decay slow enough for time scale of applications

# Epsilon-near-zero is not so exotic

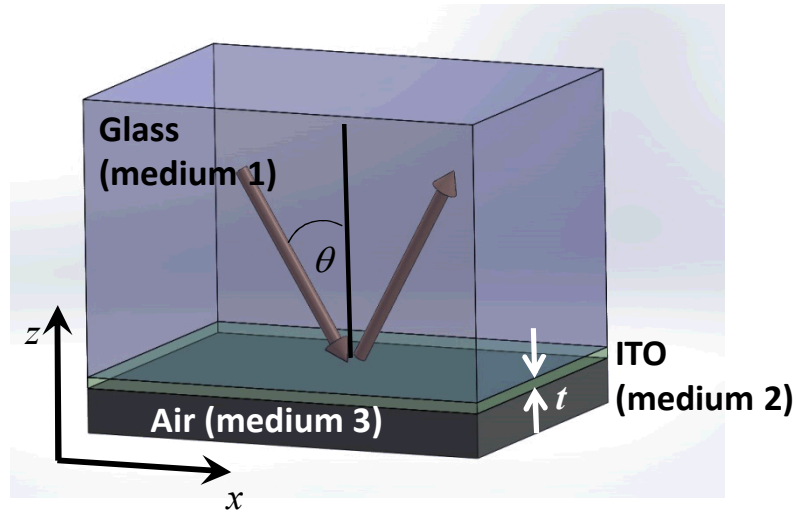


# Epsilon-near-zero (ENZ) modes and dispersion

$$\epsilon \cong 1 - \frac{\omega_p^2}{\omega^2} + i \frac{\gamma \omega_p^2}{\omega^3}, \quad \omega \gg \gamma$$



# ENZ mode dispersion and critical coupling



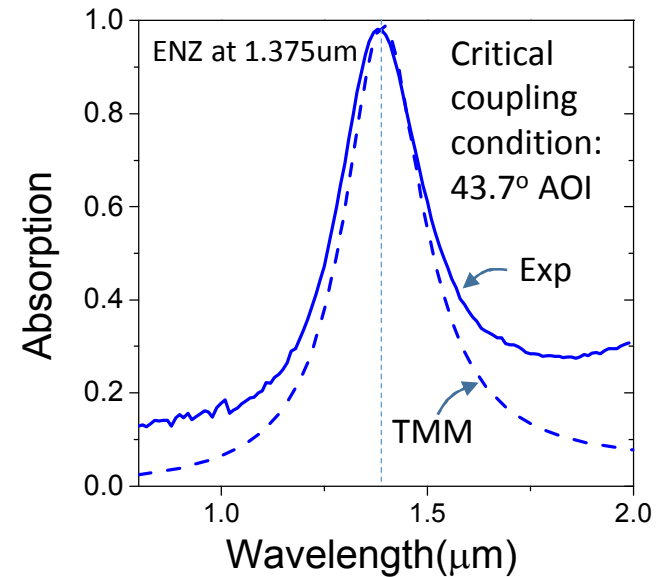
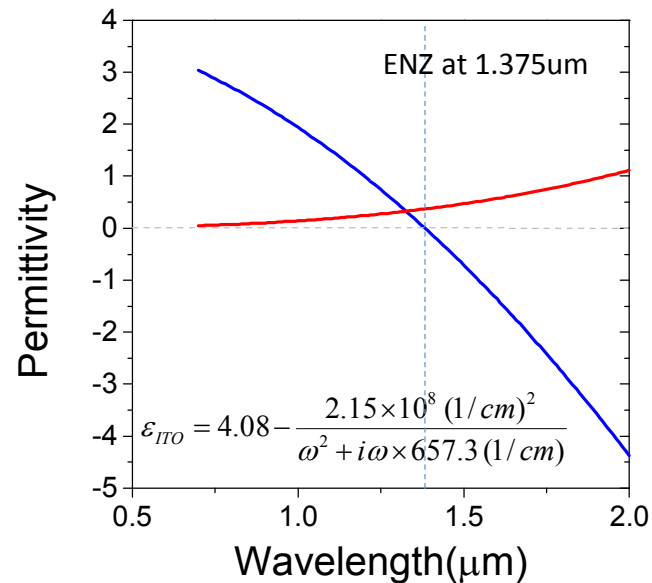
Airy Formula 
$$\Gamma = \frac{r_{12} + r_{23}e^{2i\phi}}{1 + r_{12}r_{23}e^{2i\phi}} \quad \phi = k_{2\perp}t$$

Mode dispersion: 
$$r_{12}r_{23}e^{2i\phi} = -1$$

Critical coupling condition: 
$$r_{12} = -r_{23}e^{2i\phi}$$

T. S. Luk et. al, " Directional perfect absorption using deep subwavelength low-permittivity films" *PRB*, 085411(2014)

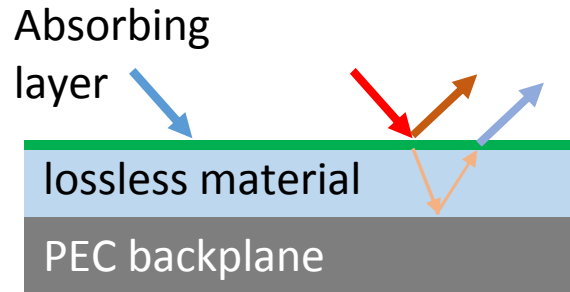
33nm ITO film (sample A)



# Other perfect absorption schemes

## Salisbury screen: anti-reflection of planar structures

[US 2599944 \(A\)](#) "Absorbent body for electromagnetic waves". Salisbury W. W. June 10, 1952

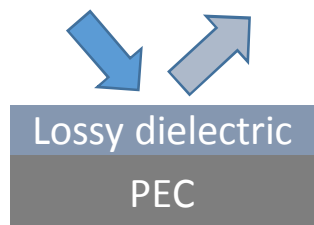


- Requires a metal backplane
- $\lambda/4$  lossless spacer material
- Thin top layer of absorbing material

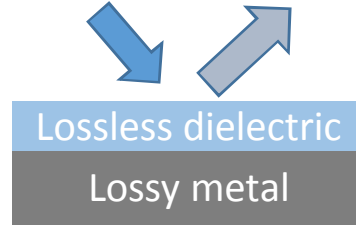
Mid-IR Z. Wang et. al. APL (2015)

Vis J. Guo et. al. Optical Materials Express (2016)

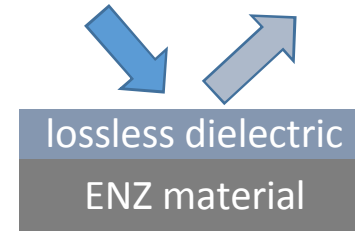
## Other Impedance matching variations (2 interfaces)



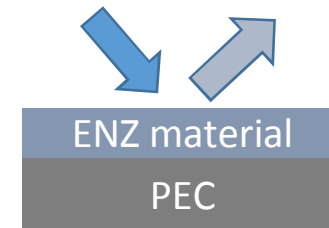
Kats, Nat. Mater. (2013)



Driessen, APL (2009)

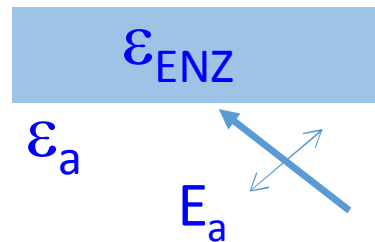


S. Campione, T. S. Luk OE (2016)

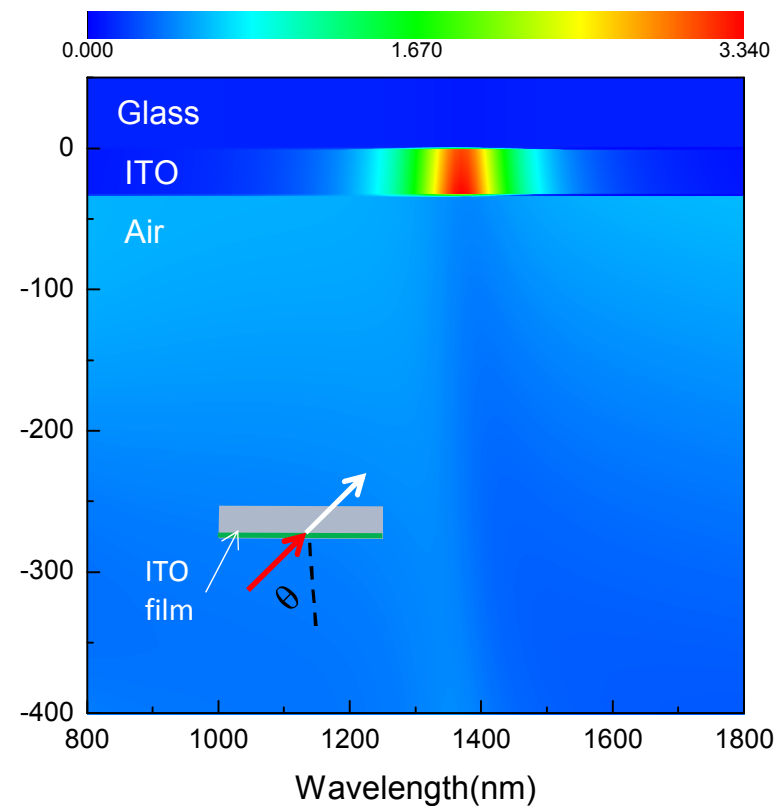
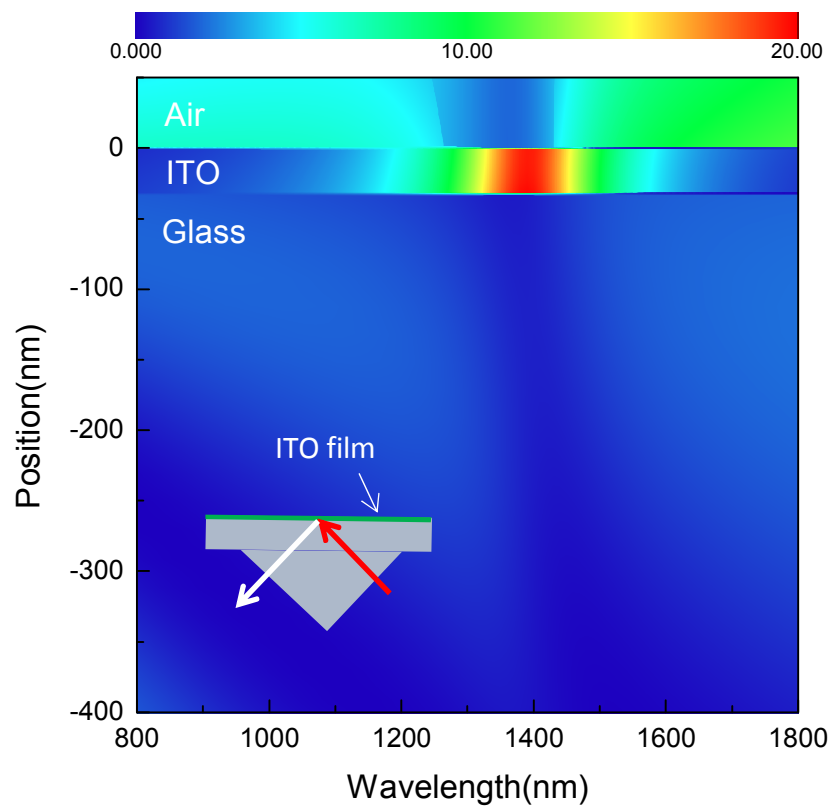


T. S. Luk et. al., PRB (2014)

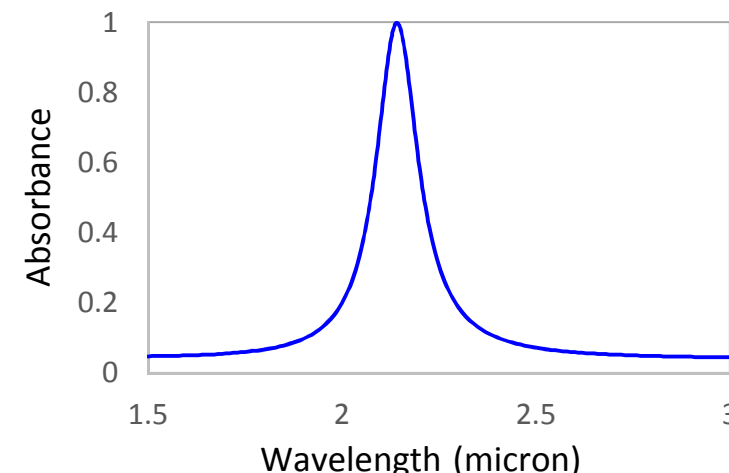
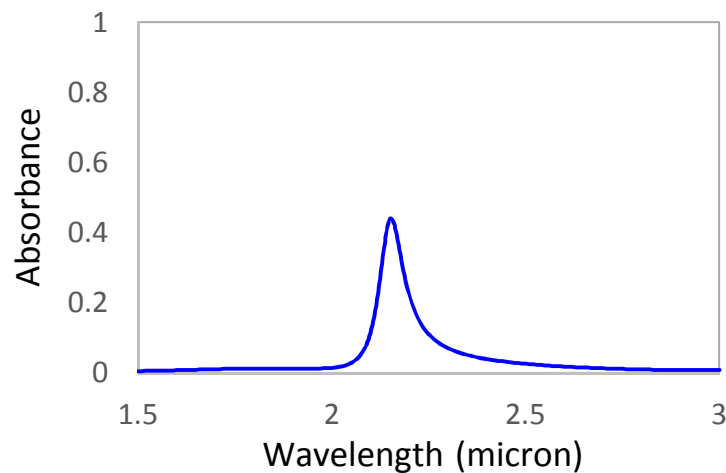
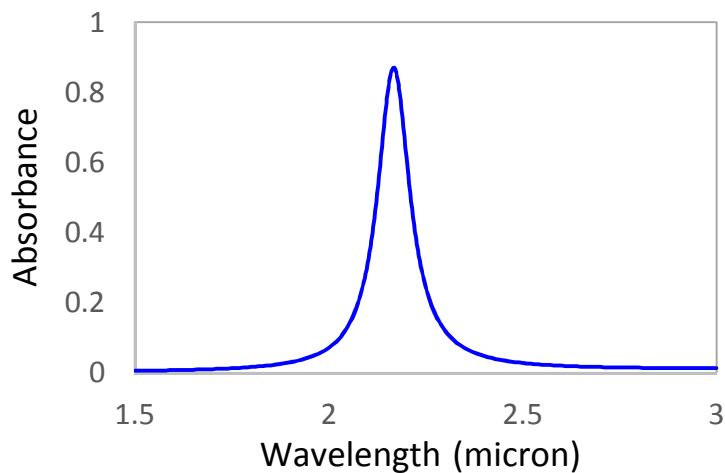
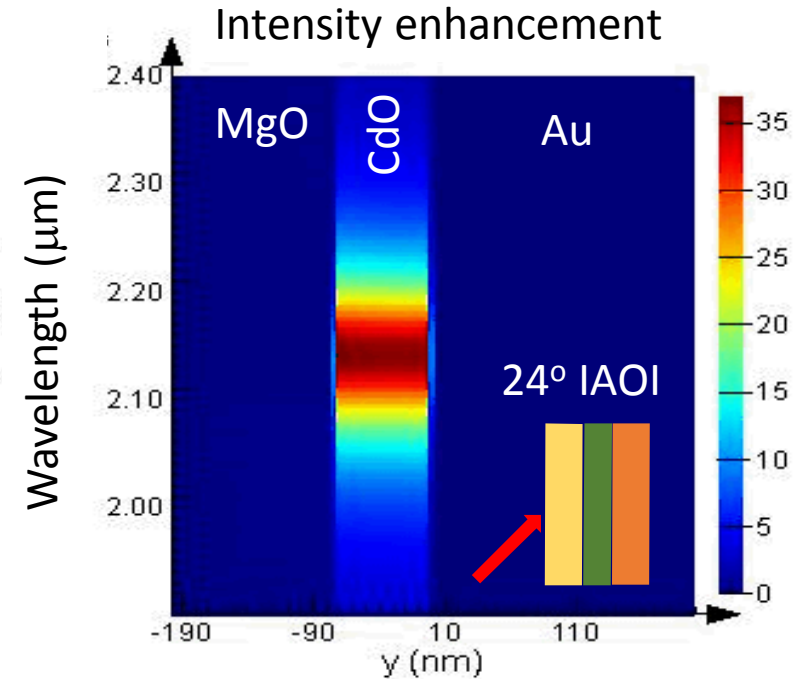
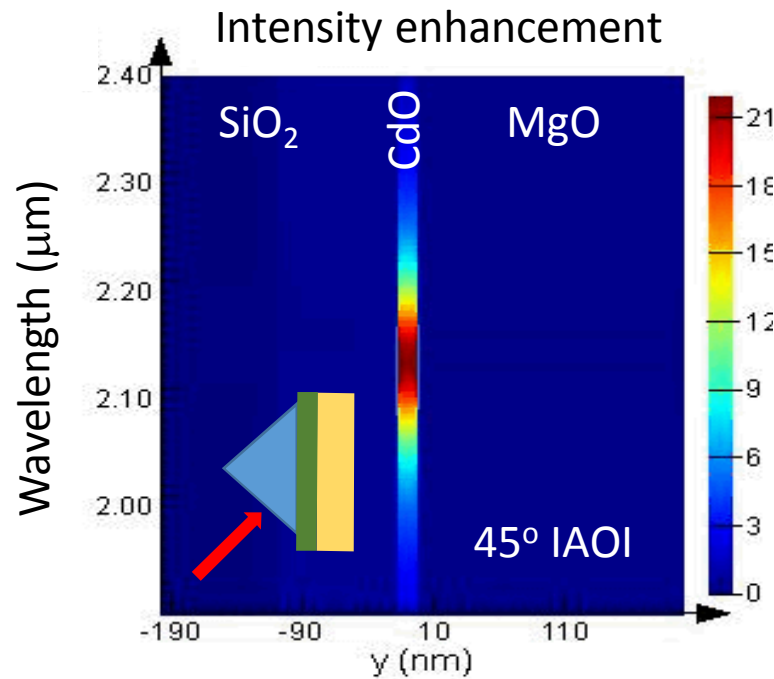
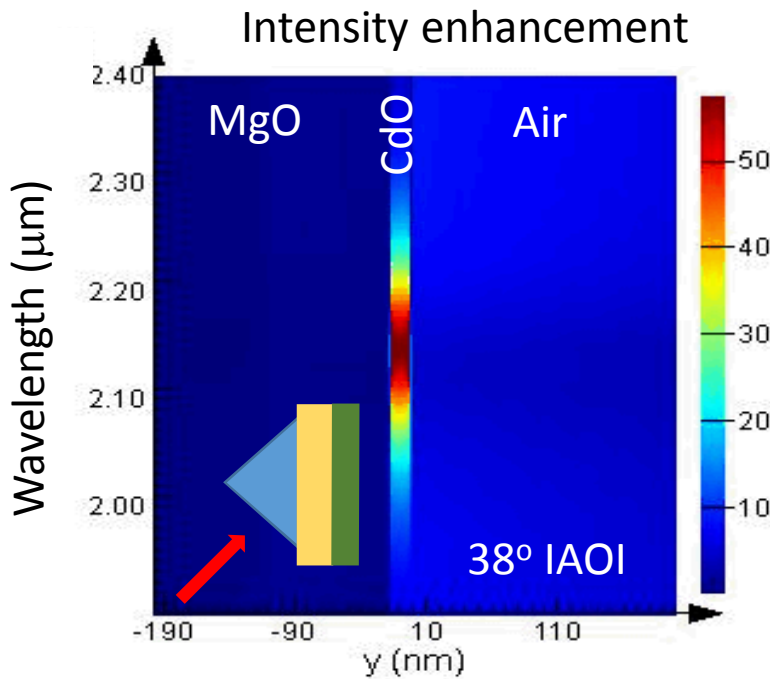
# Intensity enhancement: Berreman vs ENZ mode



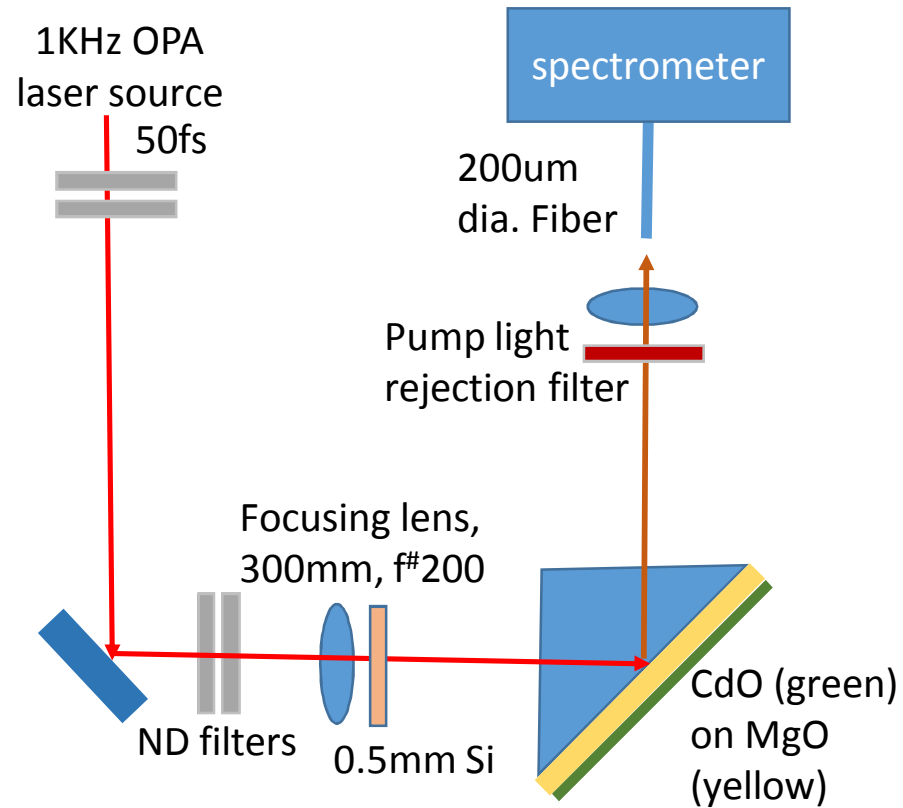
$$E_{ENZ} = D/\epsilon_{ENZ} = E_a \epsilon_a / \epsilon_{ENZ}$$



# Field enhancement of CdO ENZ material



# Third harmonic generation using ENZ mode

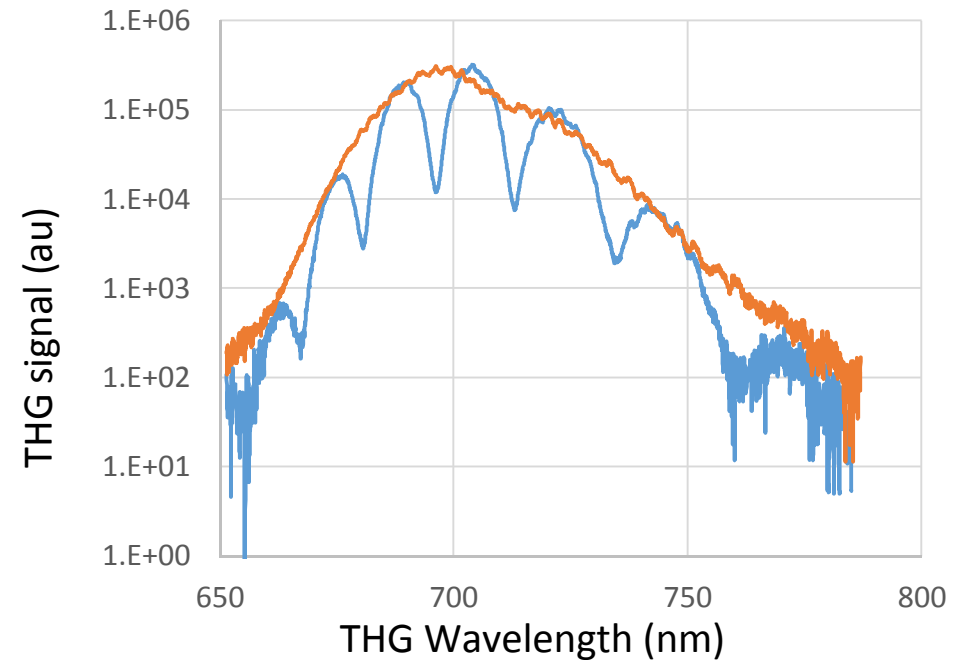


Momentum conservation:  $3\vec{k}_{FF} = \vec{k}_{TH}$

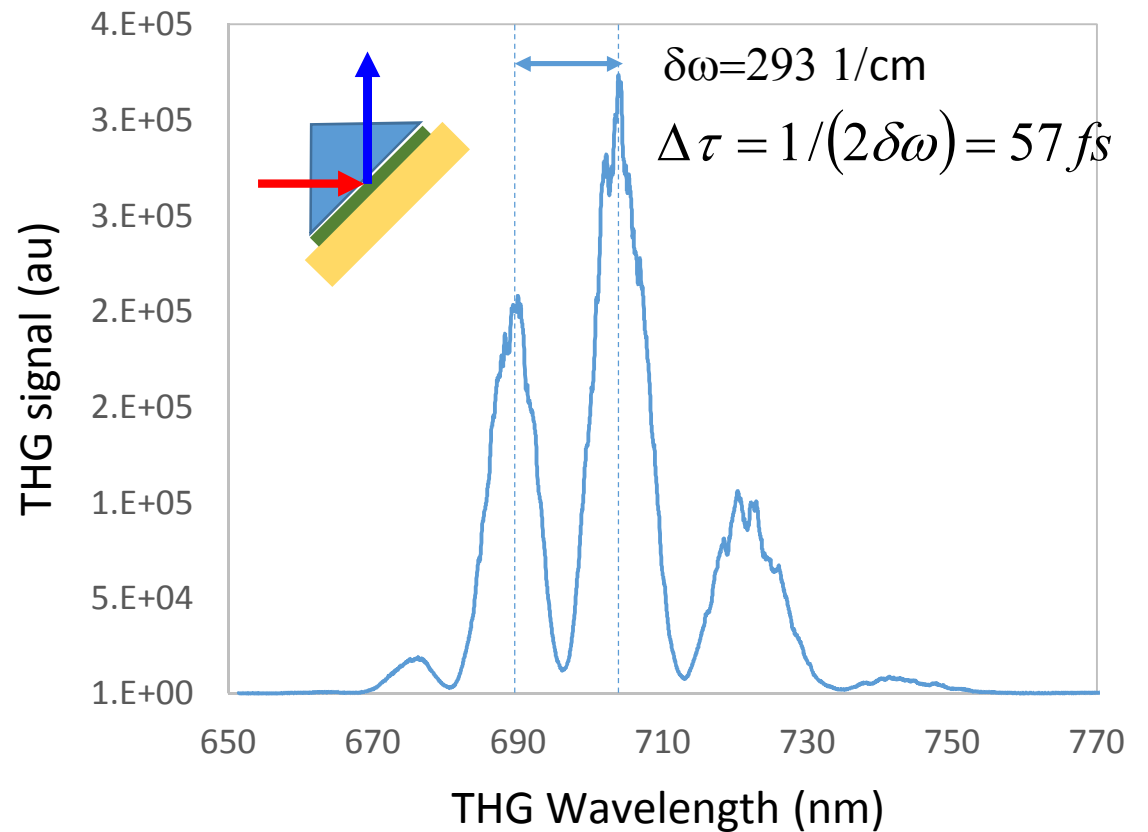
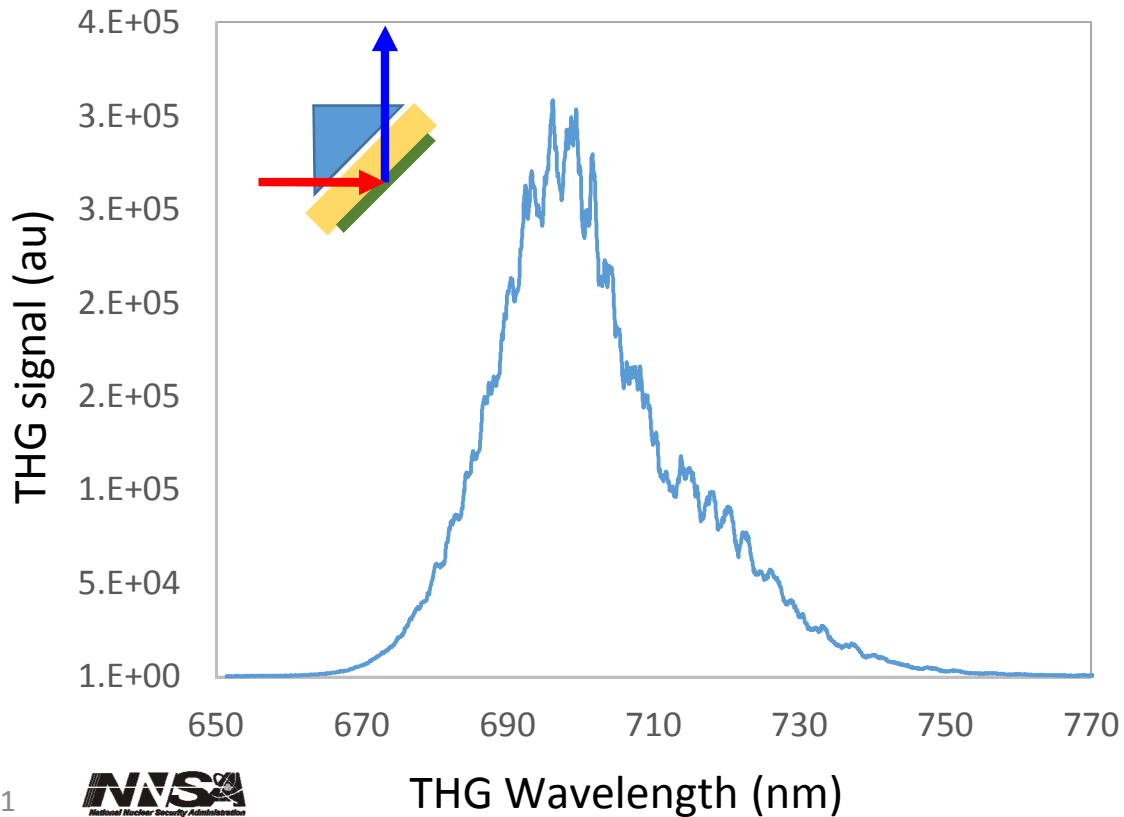
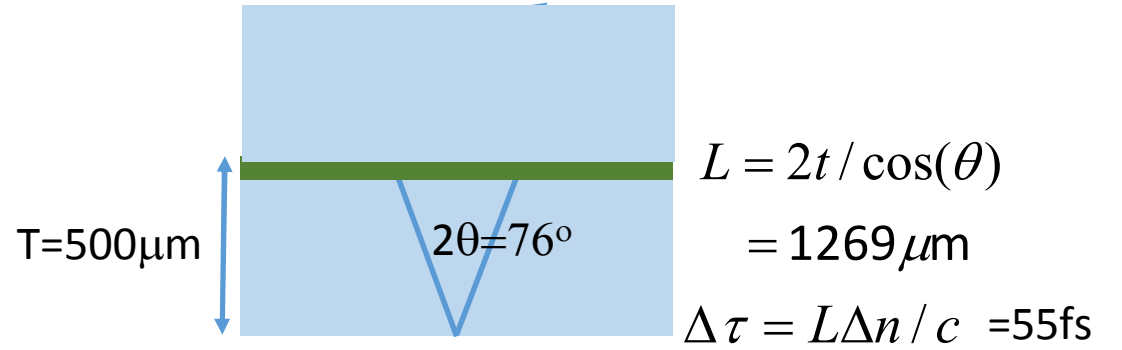
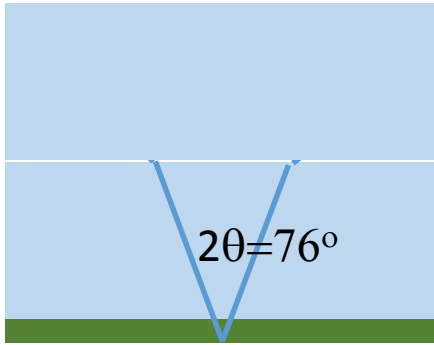
Continuity condition:  $3k_{FF\parallel} = k_{TH\parallel}$

TH emerges in specular direction:  $\frac{k_{FF\parallel}}{k_{FF\perp}} = \frac{k_{TH\parallel}}{k_{TH\perp}}$

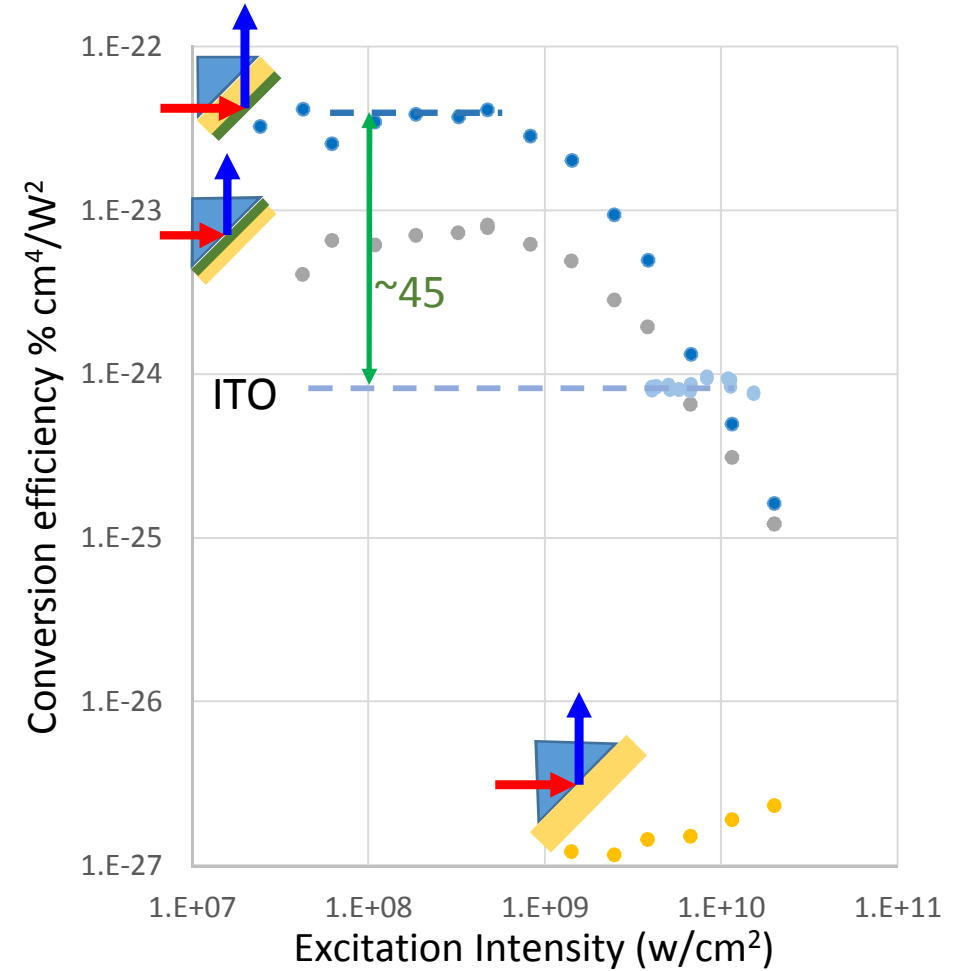
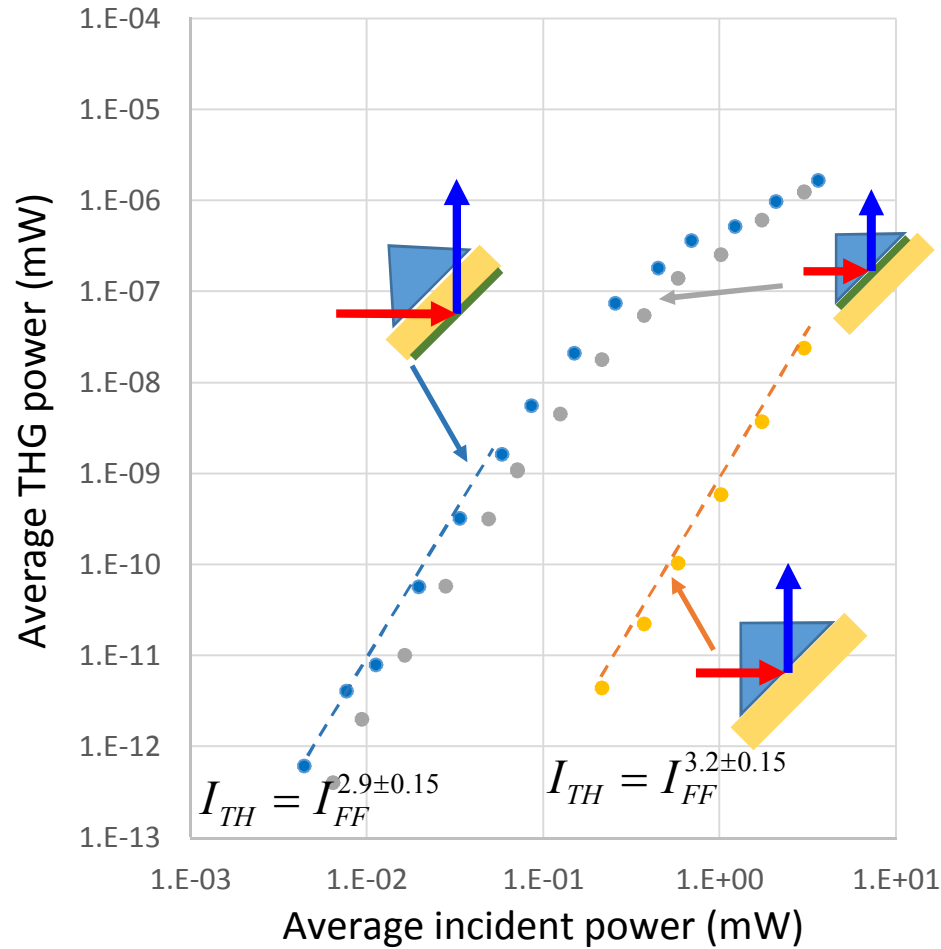
Provided  $\epsilon_a(\omega) = \epsilon_a(3\omega)$



# Origin of spectral modulation



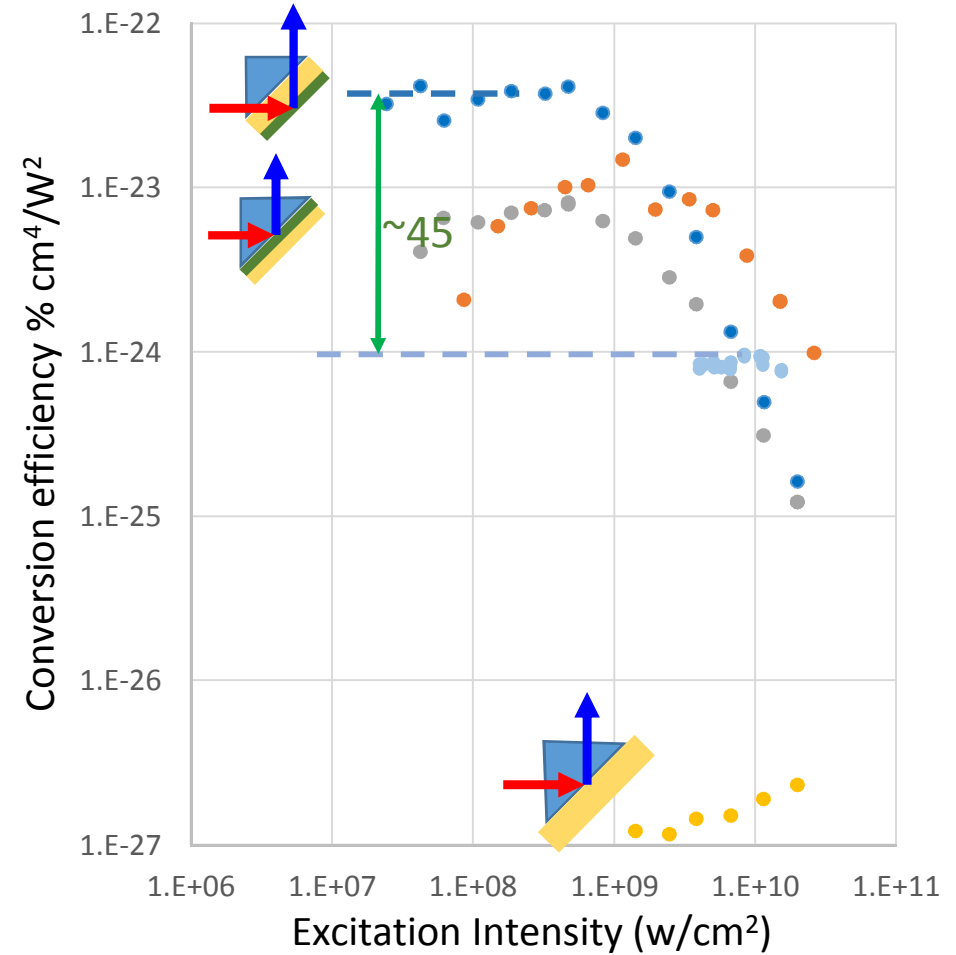
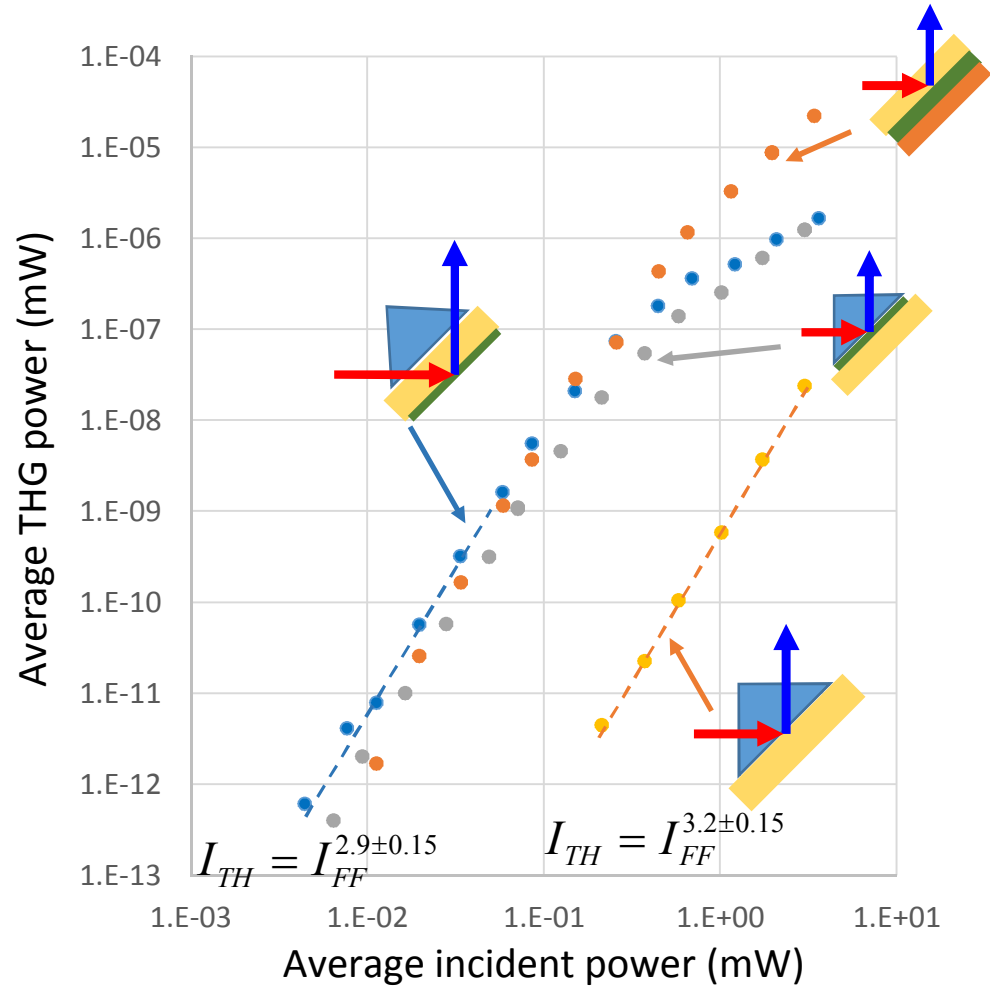
# Third harmonic generation from plasmonic CdO



## Comparison of THG efficiency in the metric of % $\text{cm}^4/\text{W}^2$

CdO-19nm	$3.5 \times 10^{-23}$ ( $\chi^{(3)} = 2 \times 10^{-20} \text{ m}^2/\text{V}^2$ ?? from Scalora*)
Chalcogenide glass	$2 \times 10^{-19} \text{ m}^2/\text{V}^2$
ITO <sup>1</sup>	$8 \times 10^{-25}$ ( $\chi^{(3)} = 3 \times 10^{-21} \text{ m}^2/\text{V}^2$ )
Anapole <sup>2</sup>	$2.22 \times 10^{-22}$

# Third harmonic generation from plasmonic CdO

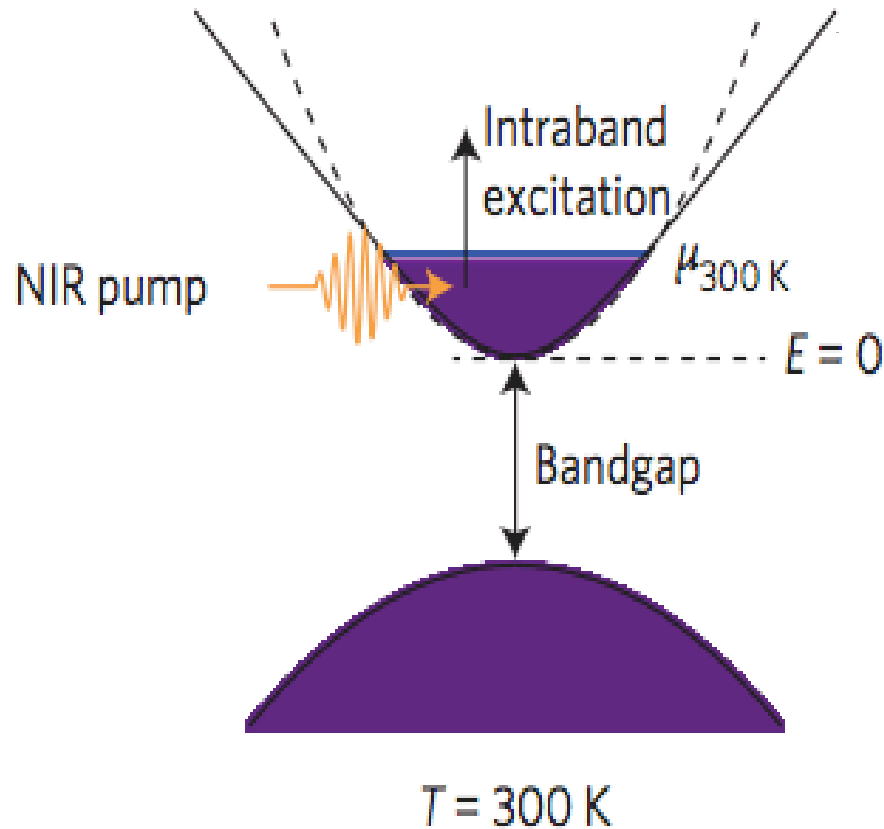


# Summary of THG from plasmonic CdO

- CdO is a superior plasmonic material in the mid-IR region.
- Field enhancement from ENZ mode enables strong nonlinear effects.
- About 45 times better than ITO and comparable to nano-patterned dielectric resonators.
- Significant heating is probably the root cause of saturation behavior.

# Heating modifies optical dielectric function and for switching applications

Photon-“perturb” electrons:



$$m^* = \frac{\hbar^2 \int f(E, T) dk}{\int f(E, T) (d^2E/dk^2) dk}$$

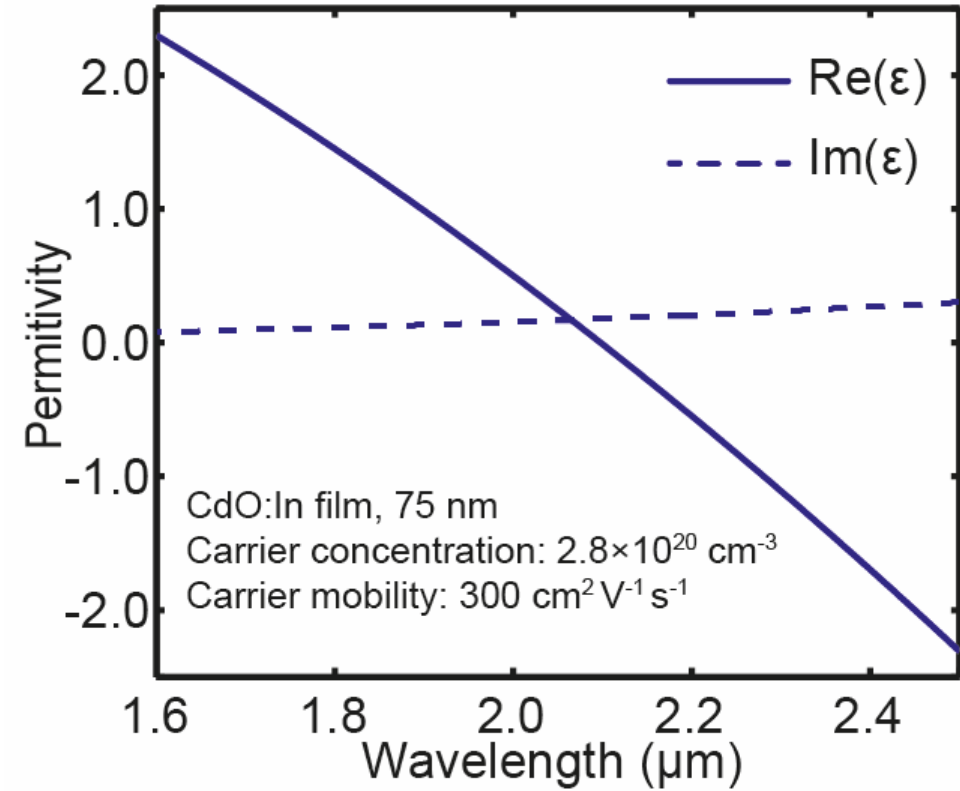
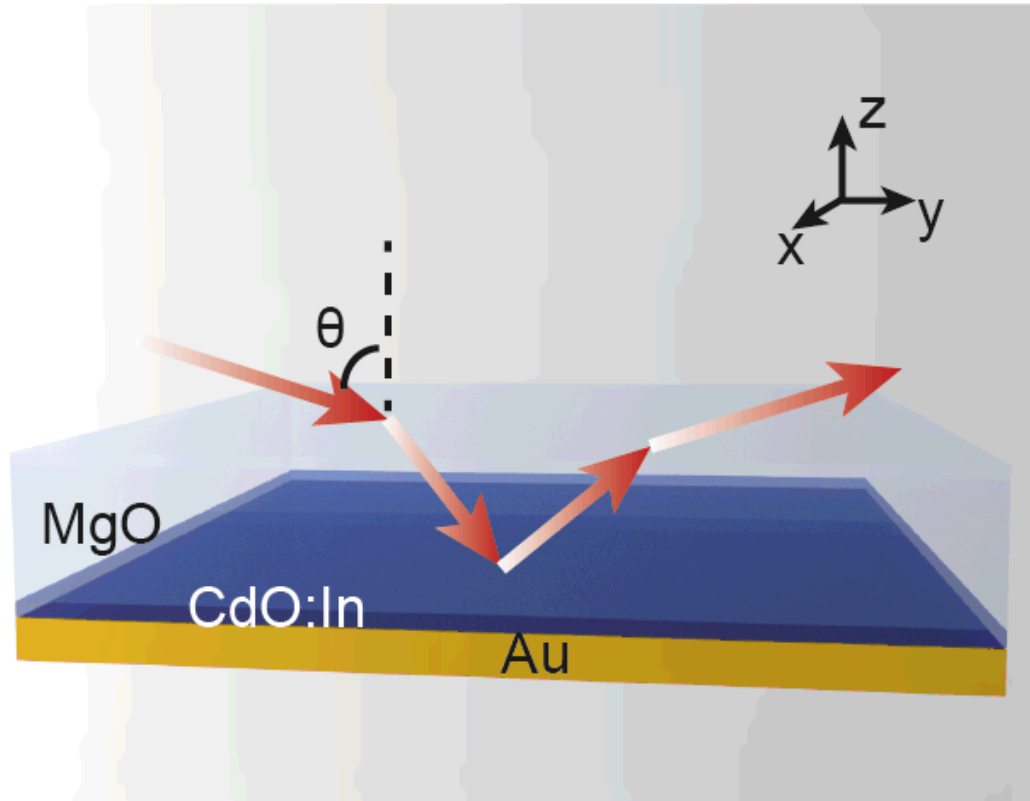
Plasma frequency:

$$\omega_p = \sqrt{ne^2 / \epsilon_0 m^*}$$

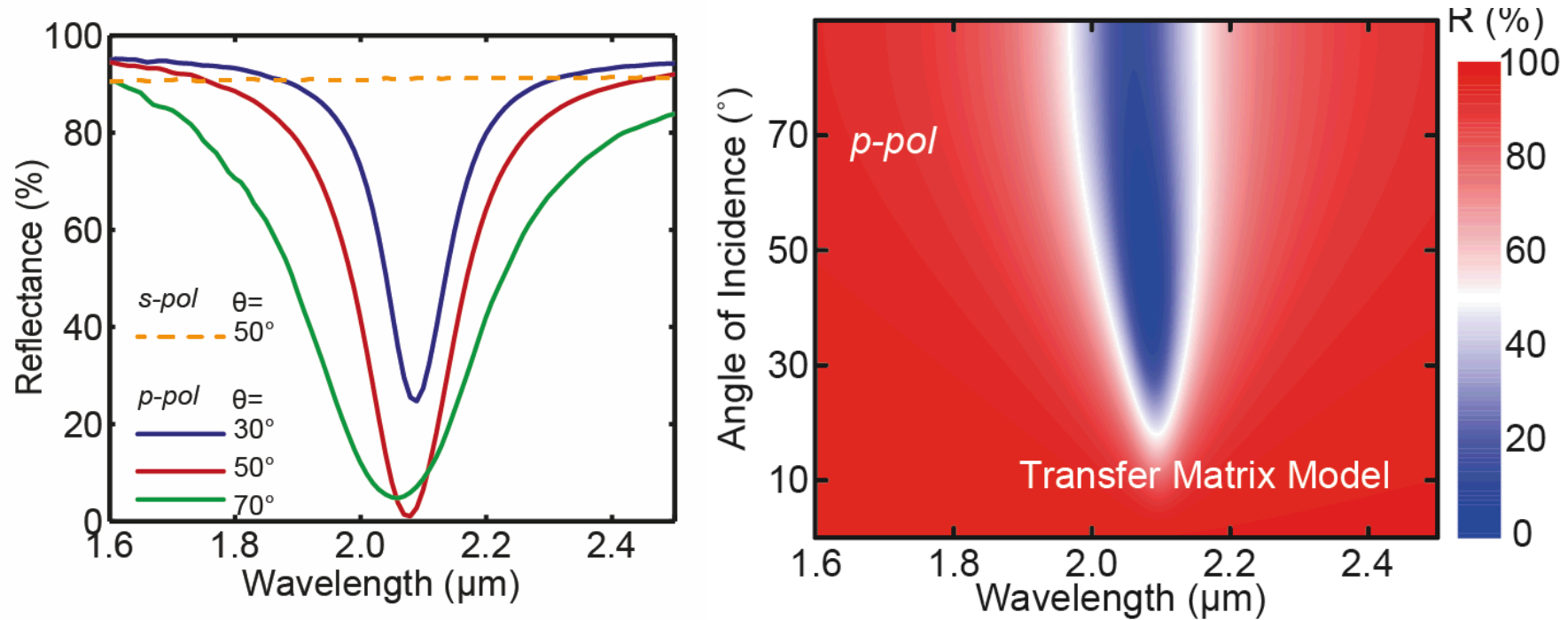
Speed limitations:

- Hot carrier lifetime

# CdO-based Berreman-mode Perfect Absorber

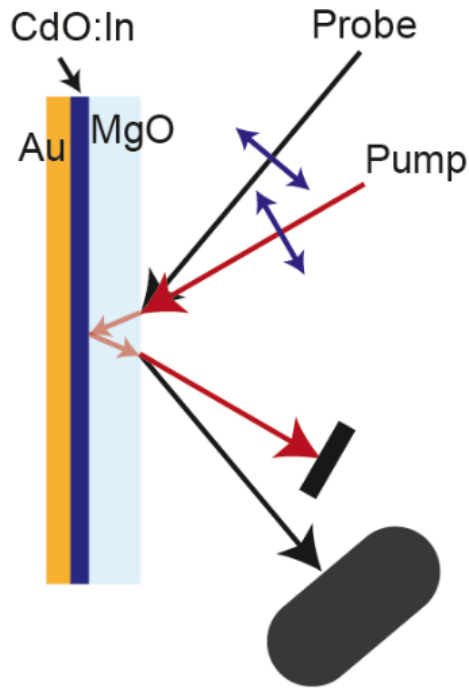


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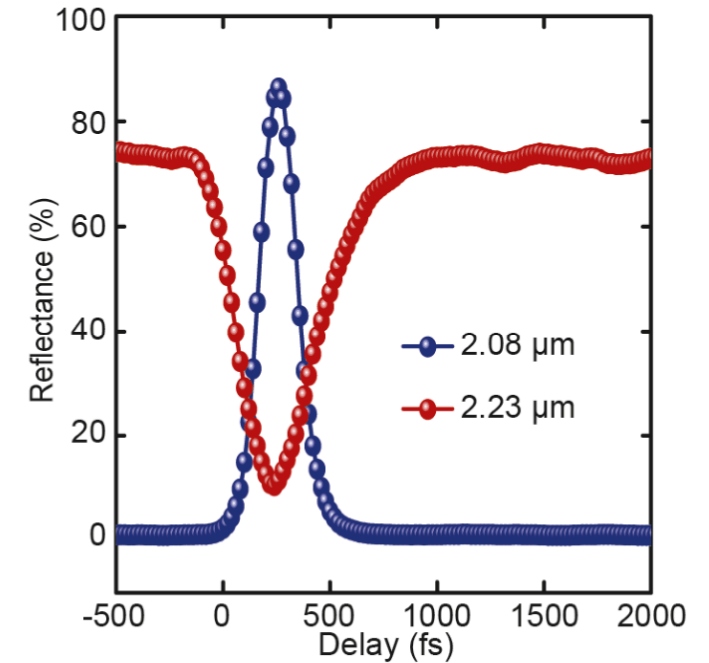
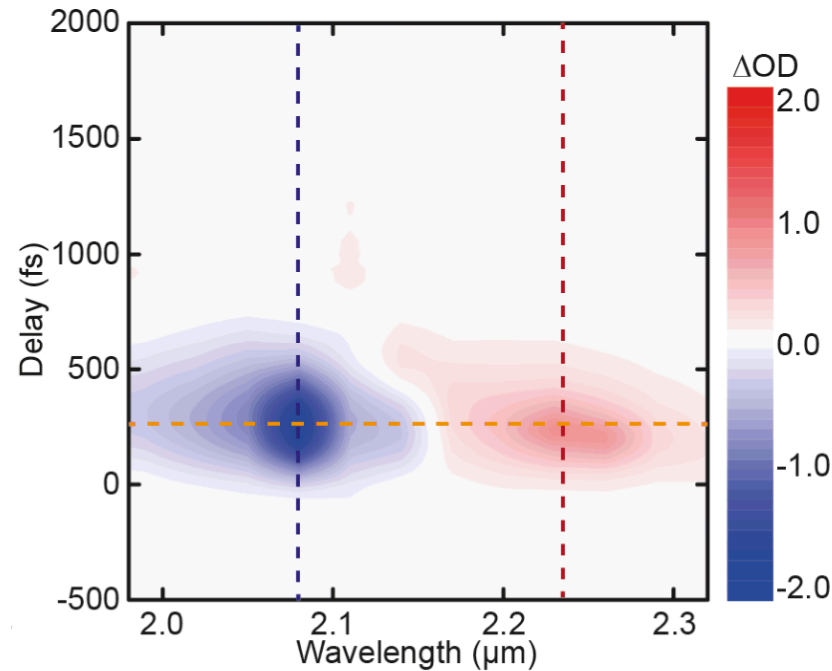


# Ultrafast AMPLITUDE SWITCHING of the Perfect Absorber

- **Absolute reflectance modulation from 1% to 86%!**
- **Pump energy density:  $339 \mu\text{J}/\text{cm}^2$**

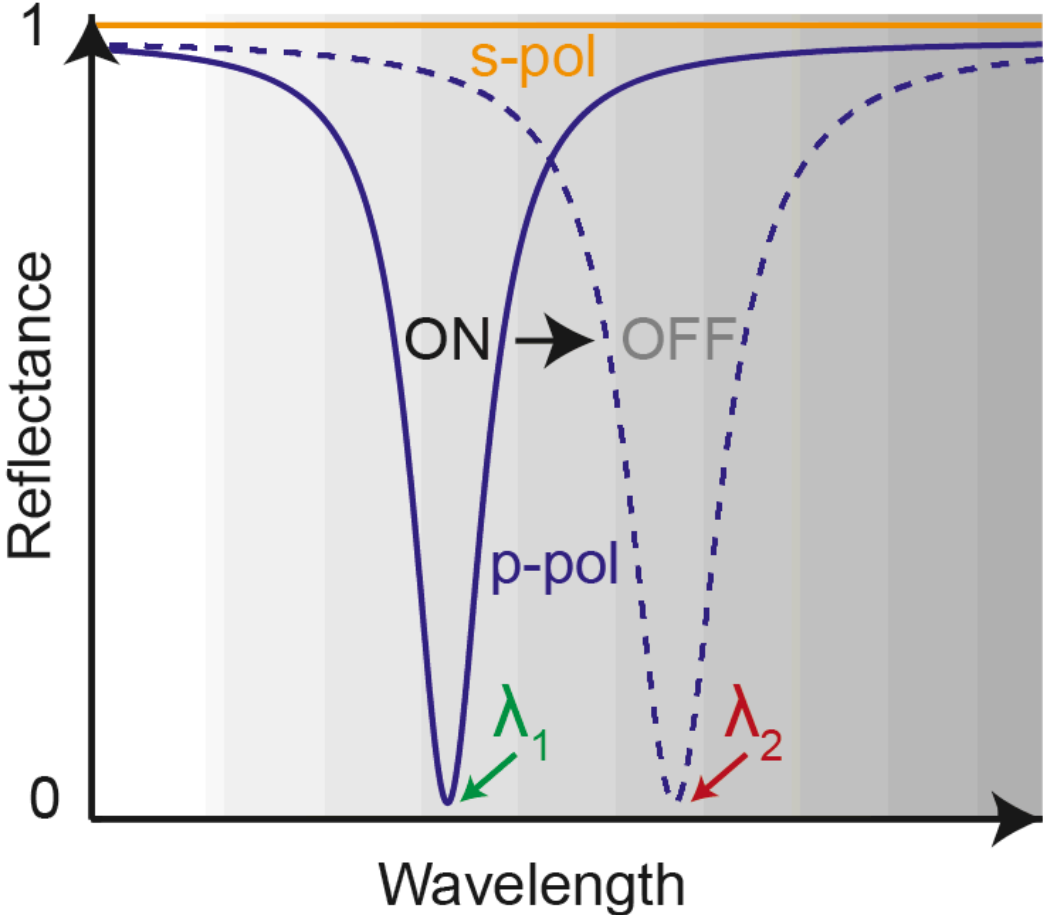


Monochromator  
& extended InGaAs detector

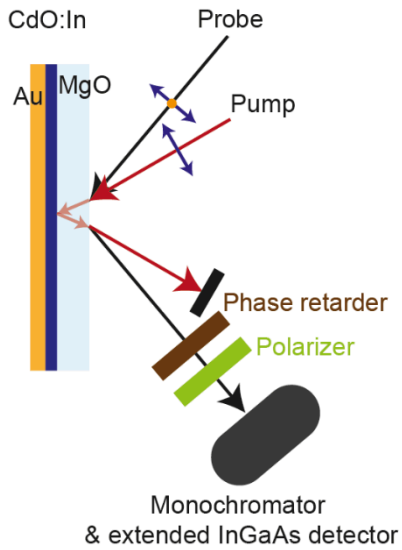


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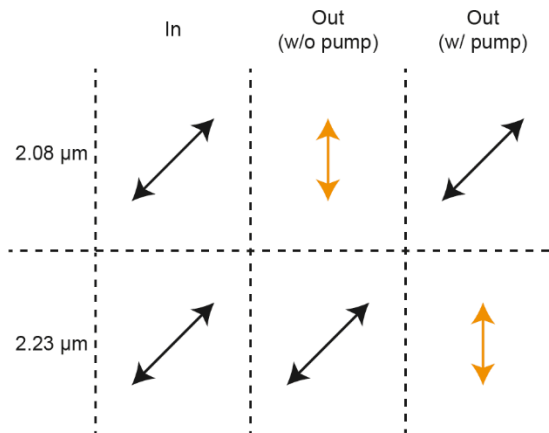
# Ultrafast POLARIZATION SWITCHING



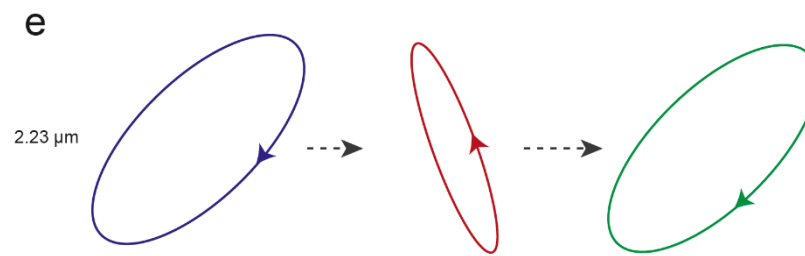
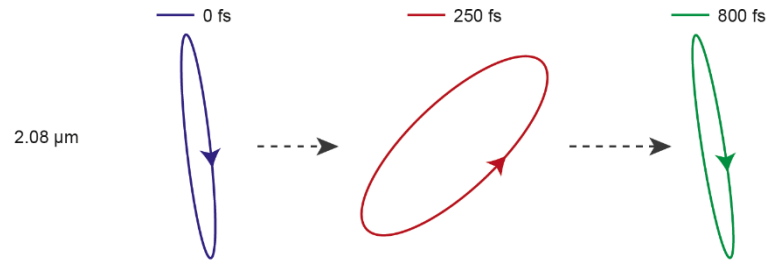
# Ultrafast Polarization Switching



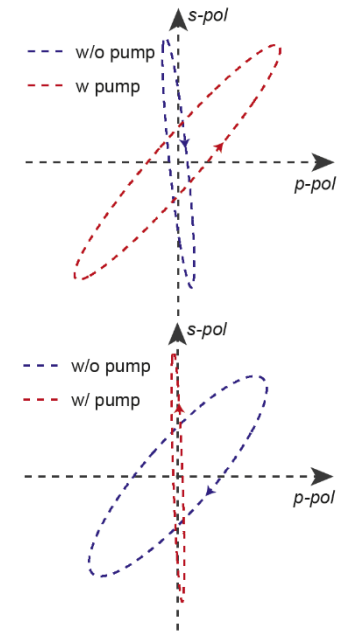
Predicted polarization input/output:



**Experiments**



**Simulations**

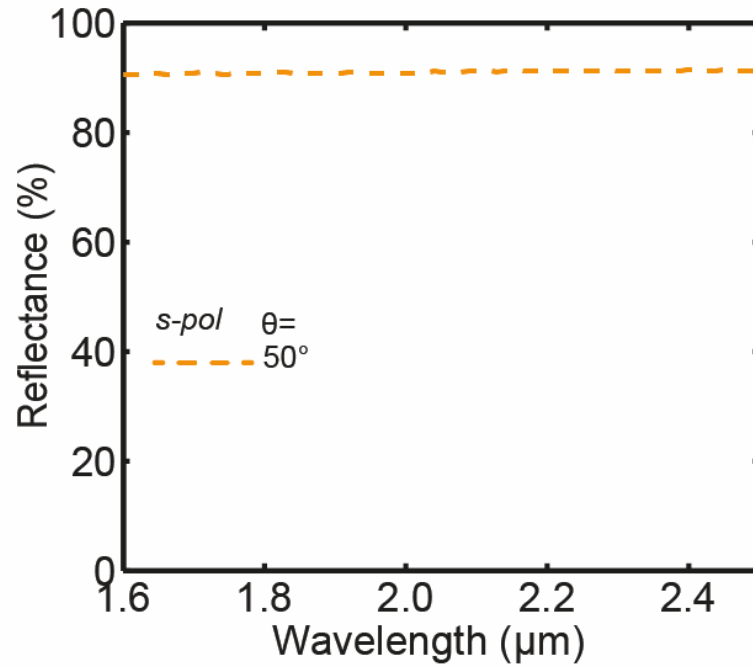


# Summary

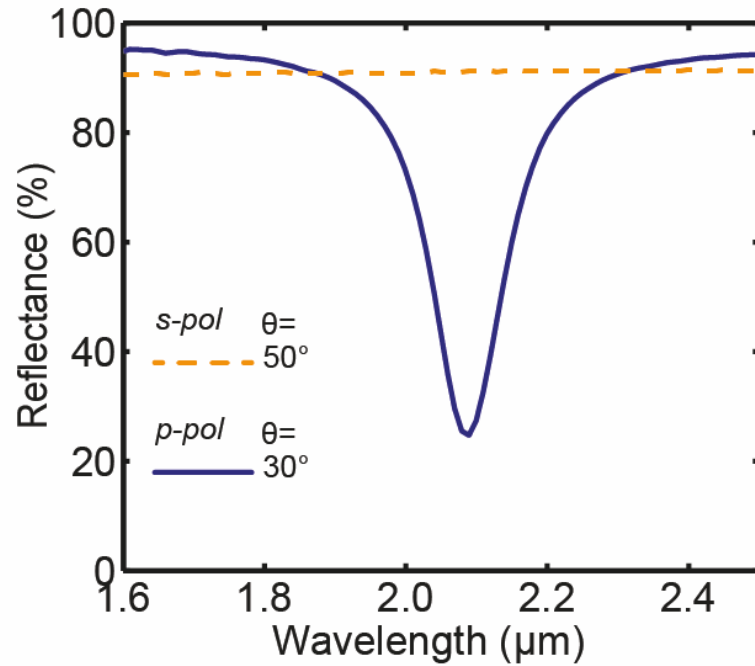
- A great deal can be accomplished with a simple thin film structure
- Field enhancement from ENZ enables efficient TH generation
- Thicker film can reduce saturation effect
- Heating effect enables ultrafast reflectivity or polarization switching

Thank you for your attention

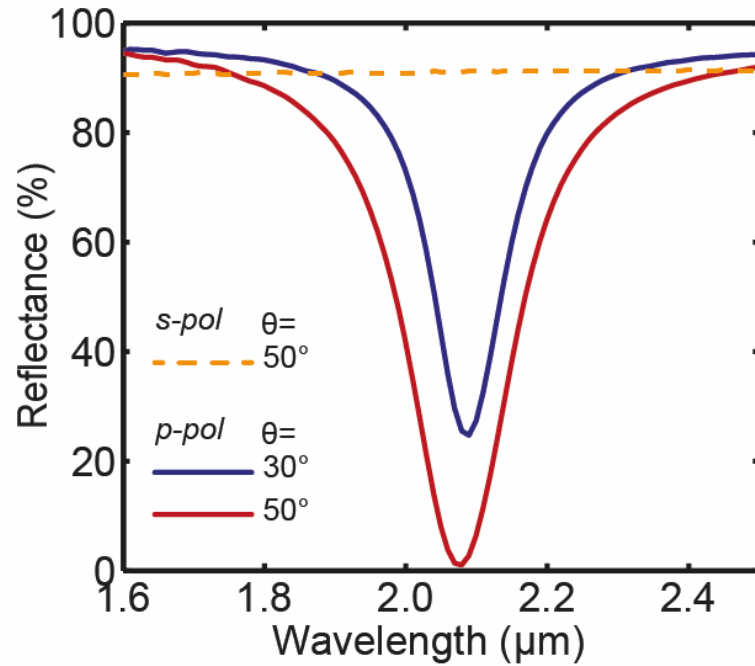
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