

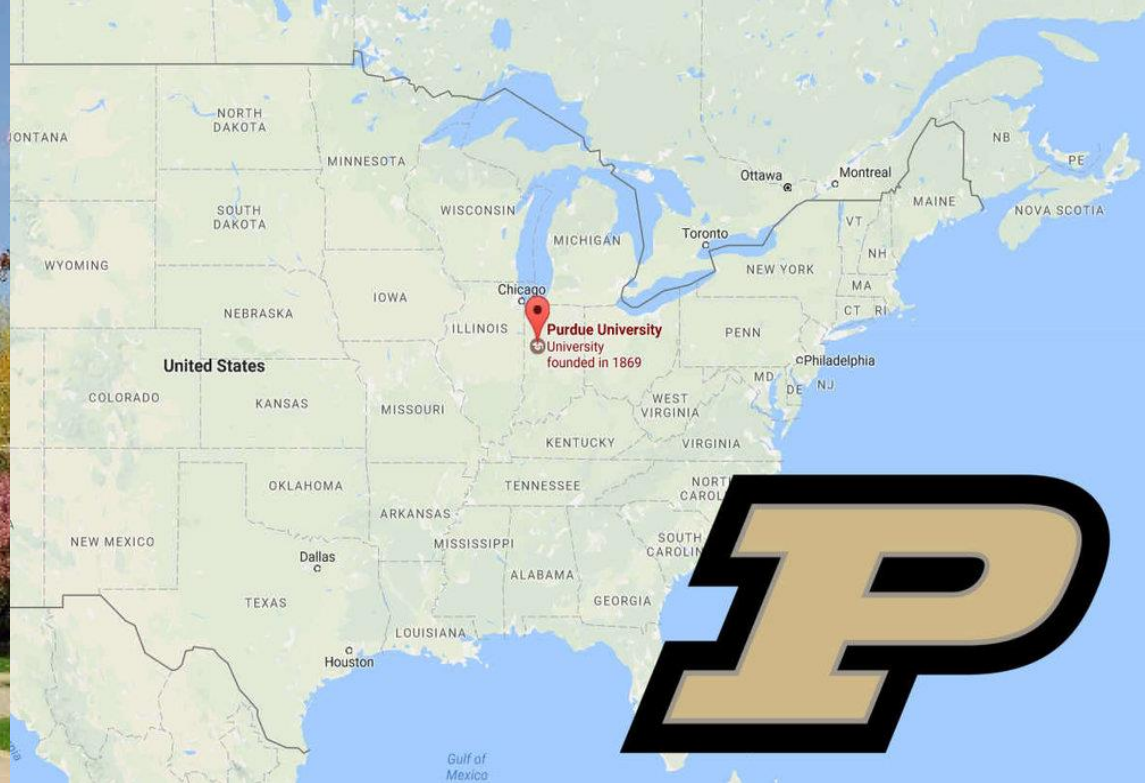
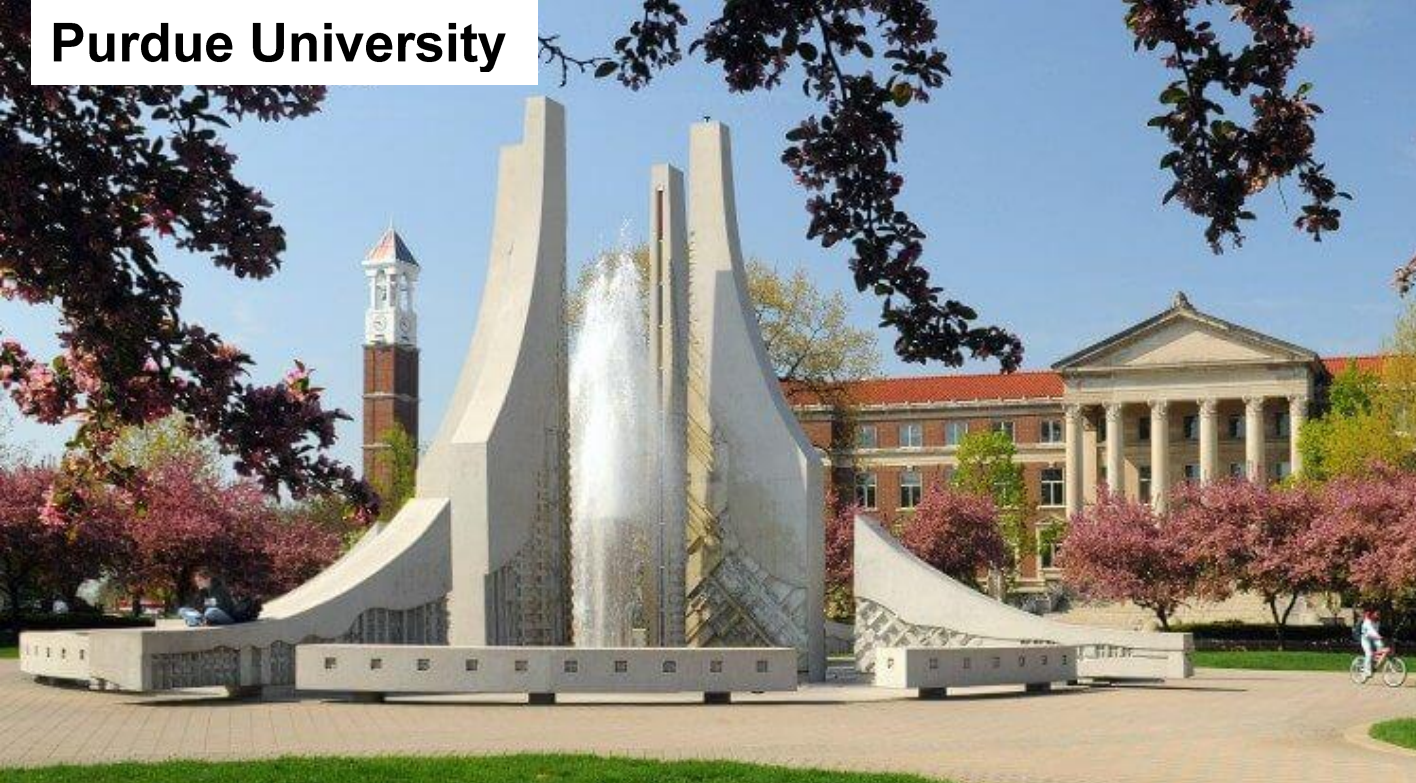
# Innovations in Tunable Optical Switches: Permittivity Control in Metals, Semiconductors and Dielectrics

**Soham Saha**

Ph.D., Electrical Engineering,  
*Purdue University (2021)*



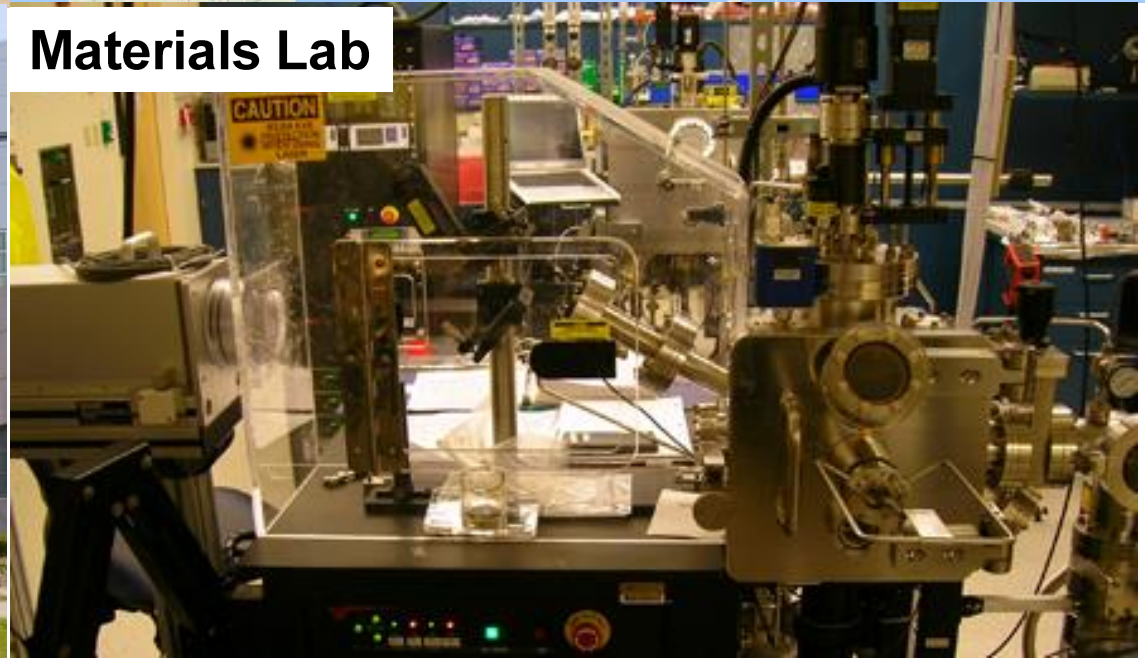
# Purdue University



# Birck Nanotechnology Center

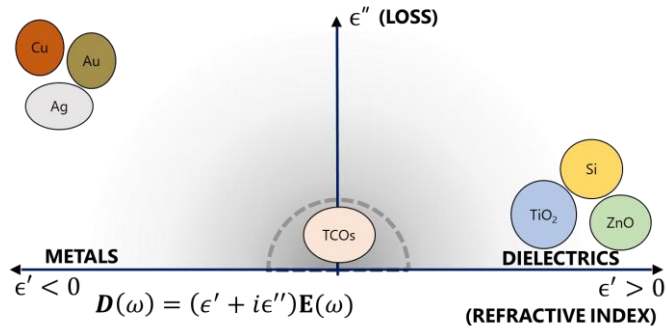


# Materials Lab

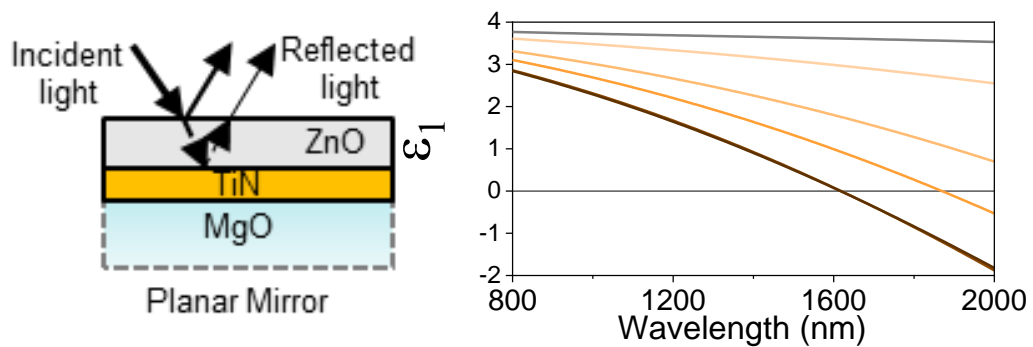




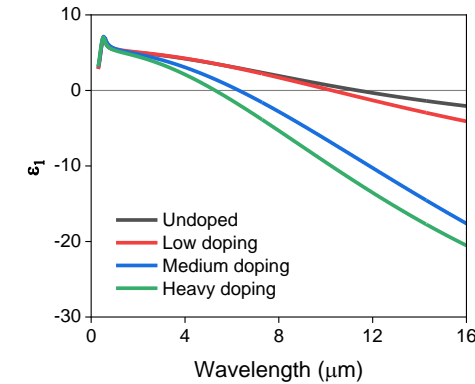
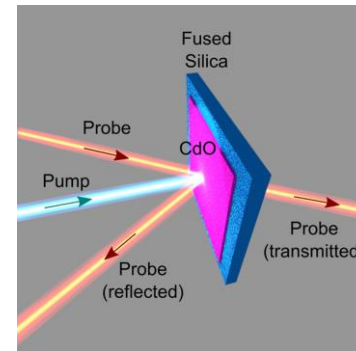
# Outline



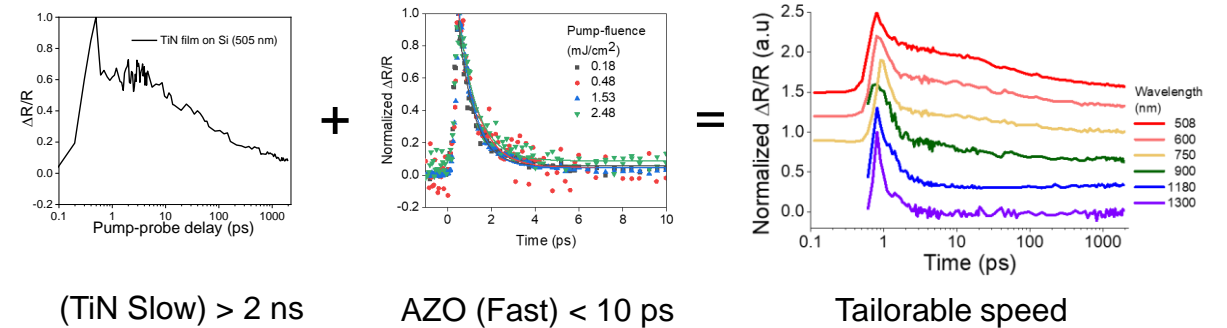
Permittivity and the Epsilon near Zero Point:  
 How to **tune** or **tailor** them



How much can we **tune** the permittivity of zinc oxide **without** adding dopants?



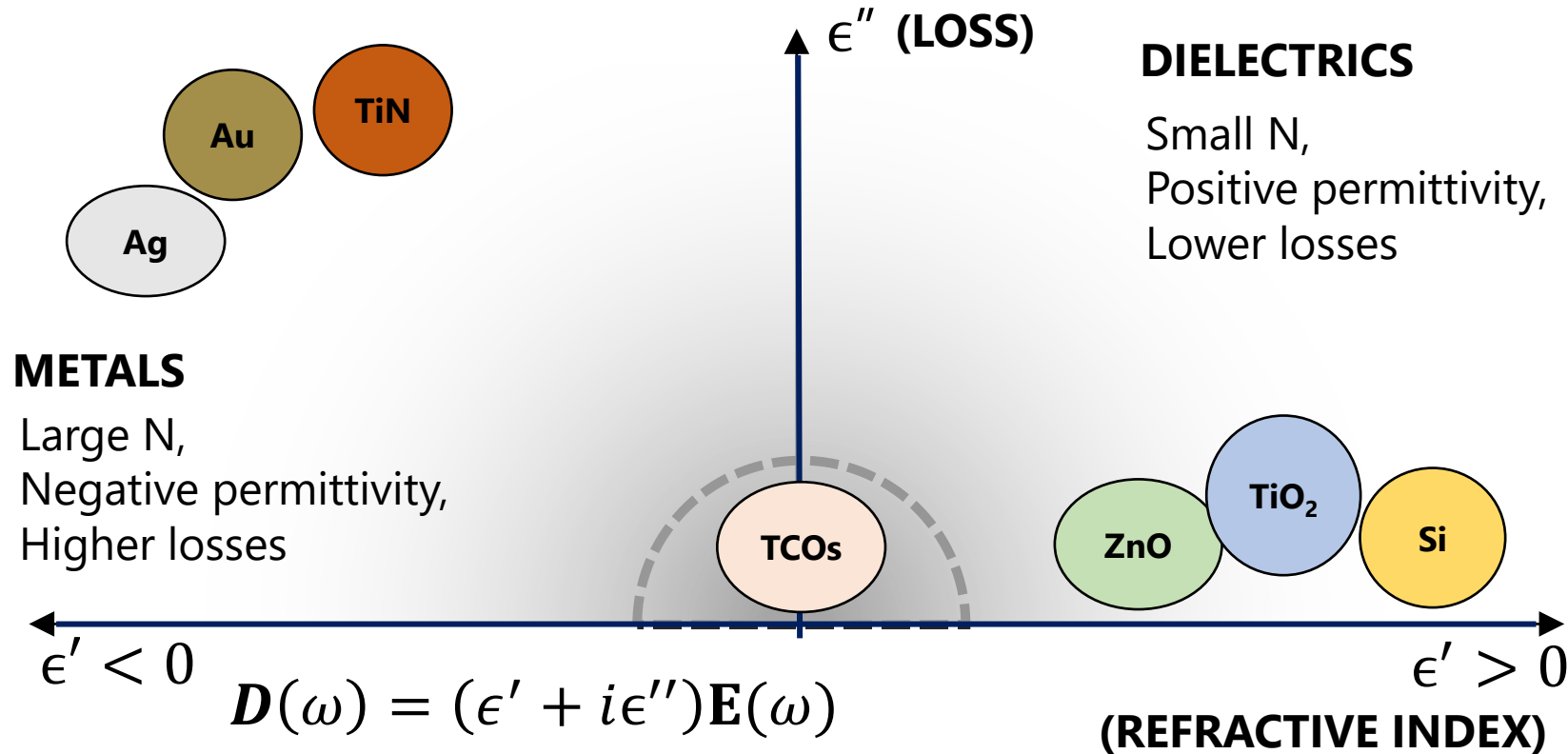
How much can we **tailor** the permittivity and relaxation-time of cadmium oxide by adding dopants?



Can we **control** the overall **response time** of a metasurface?

# Effect of Free-Electrons on Permittivity

$$\mathbf{D}(\omega) = (\epsilon' + i\epsilon'')\mathbf{E}(\omega)$$



## Drude Lorentz Model

$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^*}$$

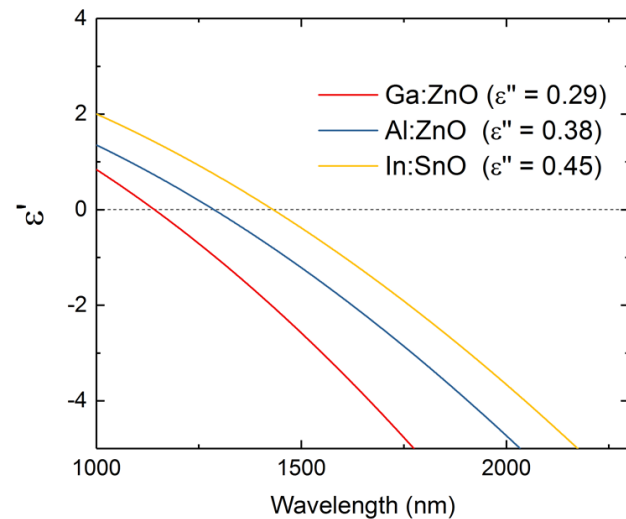
$$\epsilon' = \epsilon_B - \frac{\omega_p^2}{\omega^2 + \Gamma_d^2}$$

$$\epsilon'' = \frac{\omega_p^2 \Gamma_d}{\omega^3 + \Gamma_d^2 \omega}$$



# Epsilon-Near-Zero (ENZ) Effects

ENZ regime is the wavelength range where the permittivity changes sign  
Happens near telecom freq. in TCOs



Reflectance modulation enhanced near ENZ

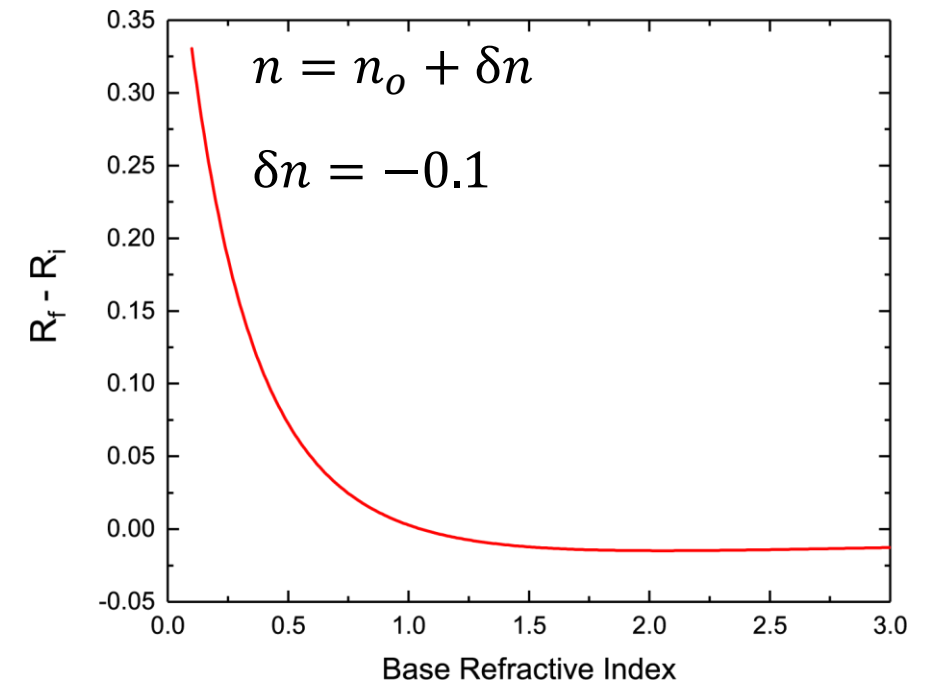
$$n = \sqrt{\epsilon}$$

$$\delta n = \frac{\delta \epsilon}{(2\sqrt{\epsilon})}$$

$$\delta R = \frac{1}{2\sqrt{\epsilon}} \frac{dR}{dn} \delta \epsilon$$

$$\epsilon \rightarrow 0$$

$$\delta R \rightarrow \infty$$



Large Field Enhancements  $\epsilon_1 \mathbf{E}_1 = \epsilon_2 \mathbf{E}_2$

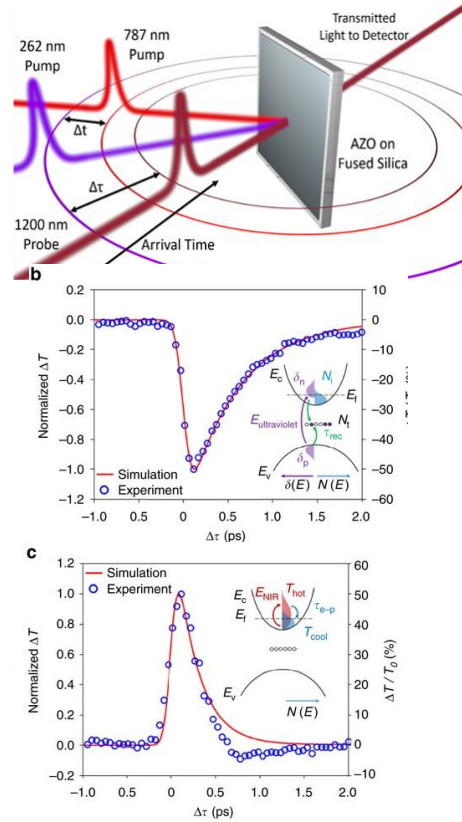
**Slow light effects** => Enhanced light-matter interaction

$$v_g = c \sqrt{\epsilon'(\omega)} \left( 1 + \frac{2}{\pi} \int_0^\infty \frac{\epsilon''(\omega_1)}{(\omega_1^2 - \omega^2)^2} \omega_1^3 d\omega_1 \right)^{-1}. \quad \text{Khurgin et al. } \textit{Optica} \text{ (2020)}$$

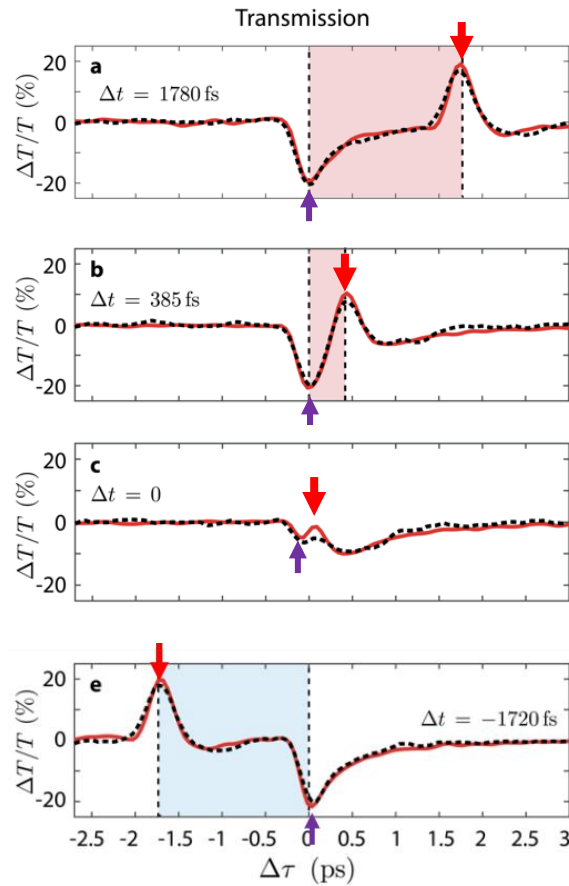
Kinsey et al. *Optica* (2015)

# Application of Large ENZ-Enhanced Reflectance Modulation

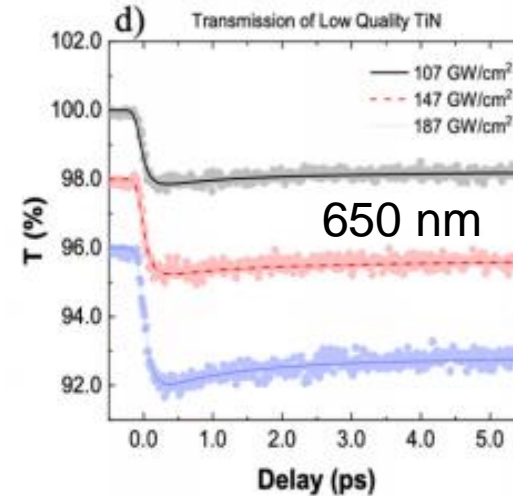
All optical addition of signals at femtosecond timescales



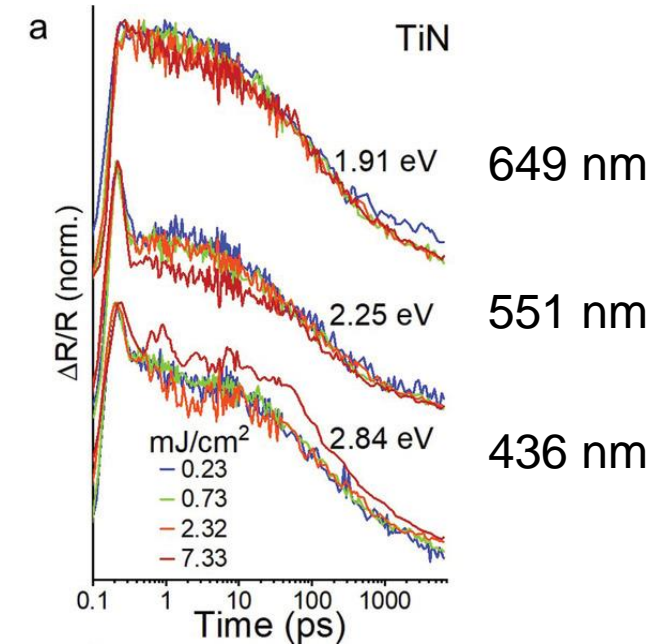
Clerici et al. *Nat. Comm.* (2016)



Extraction of hidden dynamics of hot-electrons



H. George et al. *Opt. Mat. Exp.* (2019)



B. Diroll et al. *Adv. Opt. Mat.* (2020)

# Nonlinearities Enhanced by ENZ

## REPORT Large optical nonlinearity of indium tin oxide in its epsilon-near-zero region

M. Zahirul Alam<sup>1</sup>, Israel De Leon<sup>1,3,\*</sup>, Robert W. Boyd<sup>1,2</sup>  
+ See all authors and affiliations

Science 13 May 2016:  
Vol. 352, Issue 6287, pp. 795-797  
DOI: 10.1126/science.aae0330



## Negative Refraction in Time-Varying Strongly Coupled Plasmonic-Antenna-Epsilon-Near-Zero Systems

V. Bruno, C. DeVault, S. Vezzoli, Z. Kudyshev, T. Huq, S. Mignuzzi, A. Jacassi, S. Saha, Y. D. Shah, S. A. Maier, D. R. S. Cumming, A. Boltasseva, M. Ferrera, M. Clerici, D. Faccio, R. Sapienza, and V. M. Shalaev  
Phys. Rev. Lett. **124**, 043902 – Published 30 January 2020

PhysiCS See Synopsis: [Plasmonic Metamaterials Bend Light Backwards](#)

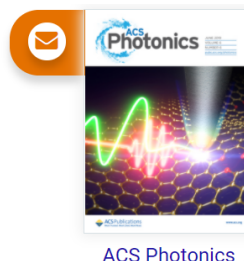
## RETURN TO ISSUE | < PREV LETTER NEXT > Second Harmonic Generation from Phononic Epsilon-Near-Zero Berreman Modes in Ultrathin Polar Crystal Films

Nikolai Christian Passler, I. Rzdolski, D. Scott Katzer, D. F. Storm, Joshua D. Caldwell, Martin Wolf, and Alexander Paarmann\*

Cite this: *ACS Photonics* 2019, 6, 6, 1365–1371  
Publication Date: June 3, 2019  
<https://doi.org/10.1021/acsp Photonics.9b00290>  
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ACS Photonics

## Published: 01 May 2017 Femtosecond optical polarization switching using a cadmium oxide-based perfect absorber

Yuanmu Yang , Kyle Kelley, Edward Sachet, Salvatore Campione, Ting S. Luk, Jon-Paul Maria, Michael B. Sinclair & Igal Brener

*Nature Photonics* **11**, 390–395(2017) | [Cite this article](#)  
**2406** Accesses | **108** Citations | **60** Altmetric | [Metrics](#)

## Letter | Published: 15 July 2019 High-harmonic generation from an epsilon-near-zero material

Yuanmu Yang , Jian Lu, Alejandro Manjavacas, Ting S. Luk, Hanzhe Liu, Kyle Kelley, Jon-Paul Maria, Evan L. Runnerstrom, Michael B. Sinclair, Shambhu Ghimire & Igal Brener

*Nature Physics* **15**, 1022–1026(2019) | [Cite this article](#)  
**6416** Accesses | **28** Citations | **15** Altmetric | [Metrics](#)

Review Article | Published: 26 September 2019

## Near-zero-index materials for photonics

Nathaniel Kinsey , Clayton DeVault, Alexandra Boltasseva & Vladimir M. Shalaev

*Nature Reviews Materials* **4**, 742–760(2019) | [Cite this article](#)  
**4468** Accesses | **41** Citations | **14** Altmetric | [Metrics](#)

## Article | Open Access | Published: 01 May 2020 Broadband frequency translation through time refraction in an epsilon-near-zero material

Yiyu Zhou , M. Zahirul Alam, Mohammad Karimi, Jeremy Upham, Orad Reshef, Cong Liu, Alan E. Willner & Robert W. Boyd

## RETURN TO ISSUE | < PREV ARTICLE NEXT > Field-Effect Tunable and Broadband Epsilon-Near-Zero Perfect Absorbers with Deep Subwavelength Thickness

Aleksei Anopchenko\*, Long Tao, Catherine Arndt, and Ho Wai Howard Lee\*

Cite this: *ACS Photonics* 2018, 5, 7, 2631–2637  
Publication Date: April 23, 2018  
<https://doi.org/10.1021/acsp Photonics.7b01373>  
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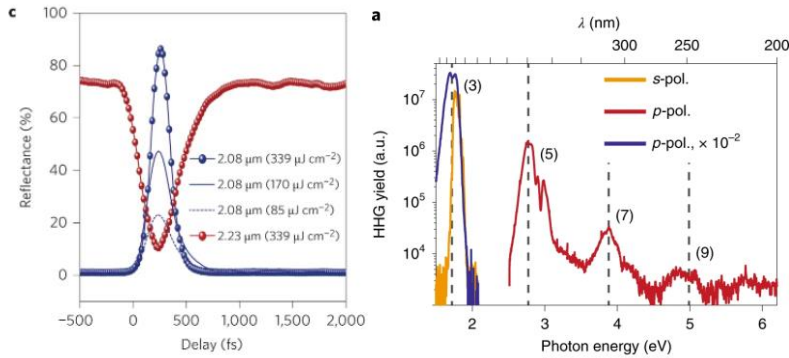
Can we **extend Epsilon Near Zero** effects to any wavelength of our choice?

How much can we **control** the permittivity in **real time**?

Can we **control** the **response-time** of an all-optical switch?

# Materials Explored in This Work

## Cadmium Oxide (CdO)

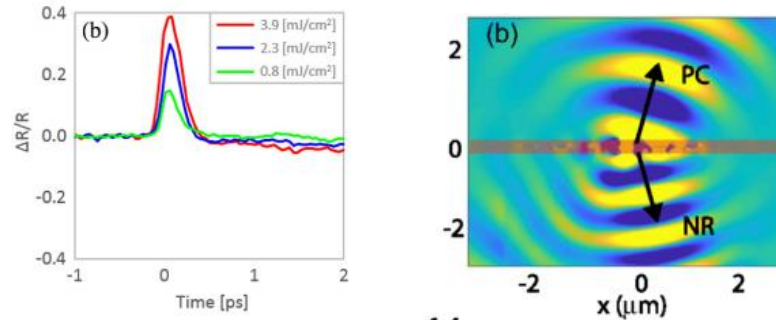


Femtosecond switching:  
Nat. Phot. (2017)

High harmonic generation:  
Nat. Phys (2019)

- **Low-loss metal** for MID IR Plasmonics
- **High mobility** for electroabsorption modulators
- **ENZ Applications:** ultrafast switches, high harmonic Generation

## Zinc Oxide (ZnO) and Al:ZnO

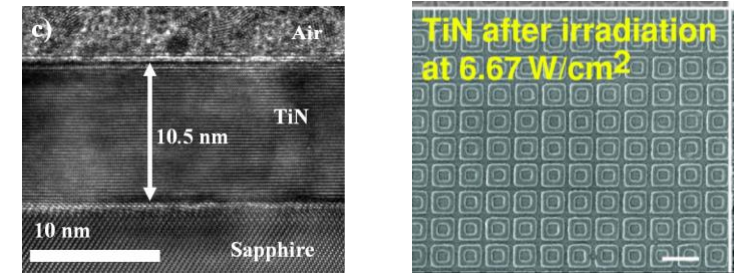


Kinsey et. al. Optica (2015)

V. Bruno et. al. PRL. (2020)

- ZnO: laser tolerant, used in optoelectronic applications from **single photon emitters** to **solar cells**
- AZO Demonstrated in various ENZ applications like **reflectance enhancement**, **negative refraction**, etc.

## Titanium Nitride (TiN)



Shah et. al. AOM. (2017)

Guler et. al. Adv. Mat. (2014)

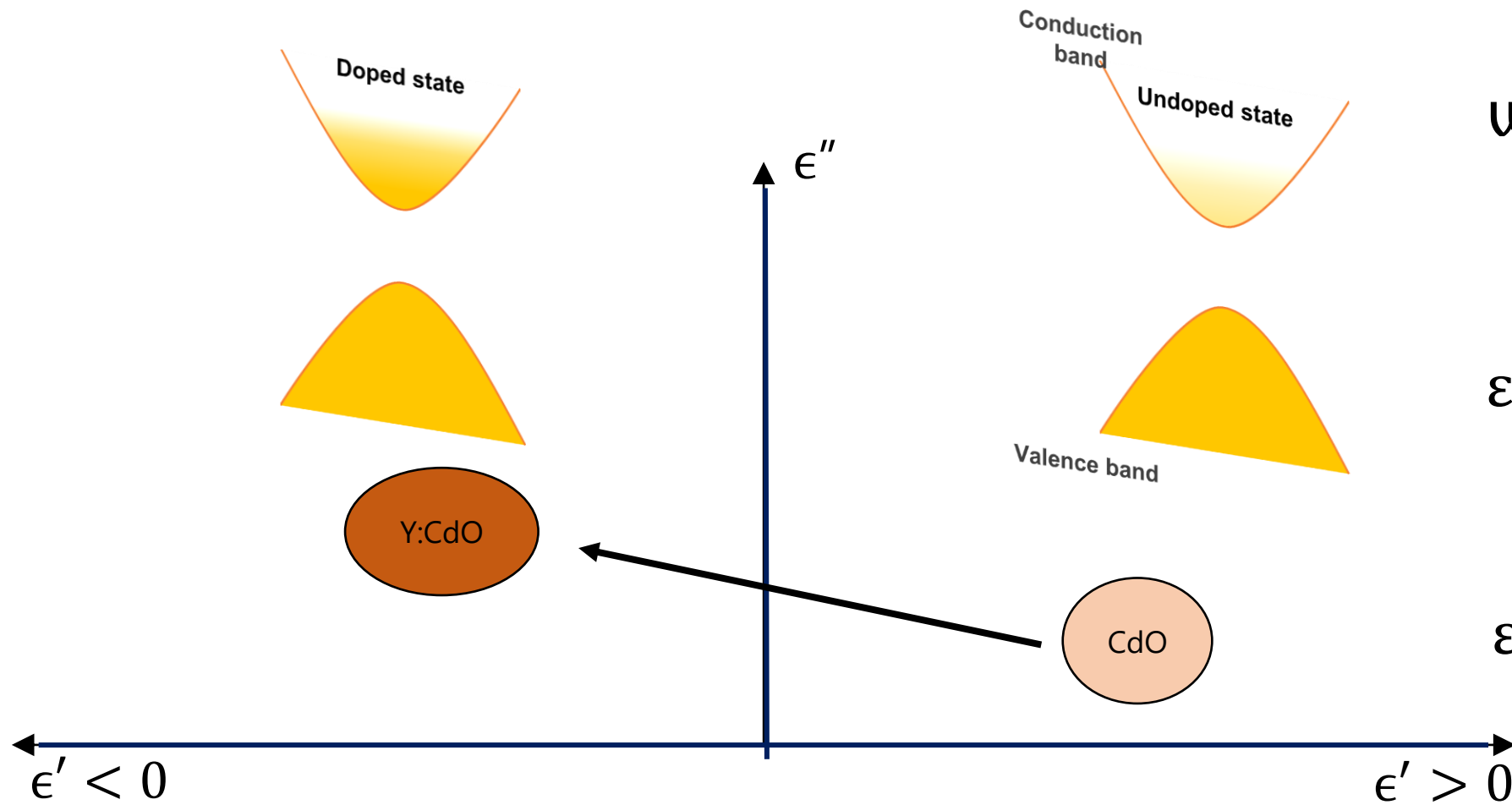
- **Gold-like** metallic properties
- Large **laser tolerance**
- Interesting dynamics with a **nanosecond** response time



How much can we tailor the **permittivity** and **relaxation time** of cadmium oxide **by adding dopants**?



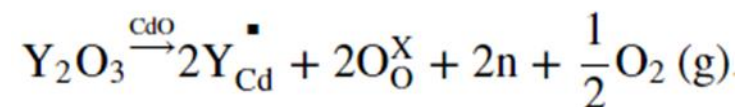
# How do We Tailor the Permittivity ?



$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^*}$$

$$\epsilon' = \epsilon_B - \frac{\omega_p^2}{\omega^2 + \Gamma_d^2}$$

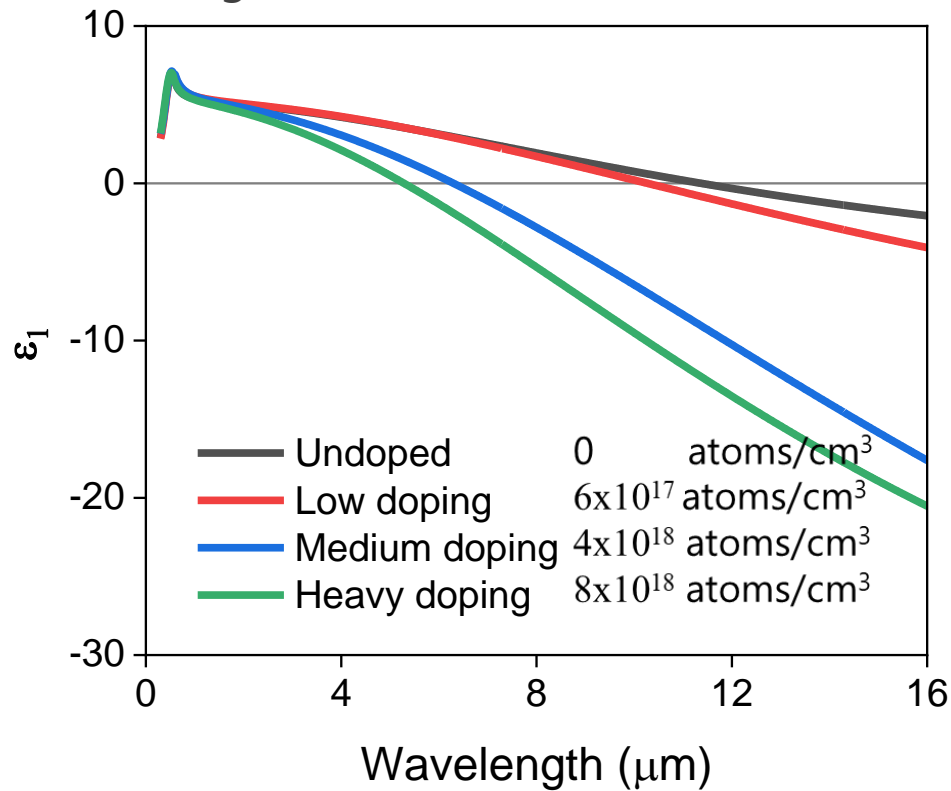
$$\epsilon'' = \frac{\omega_p^2 \Gamma_d}{\omega^3 + \Gamma_d^2 \omega}$$



# Yttrium Doping of Cadmium Oxide to Tailor the Permittivity

**Material: CdO ~500 nm films grown on fused silica by co-sputtering CdO and Yttrium**

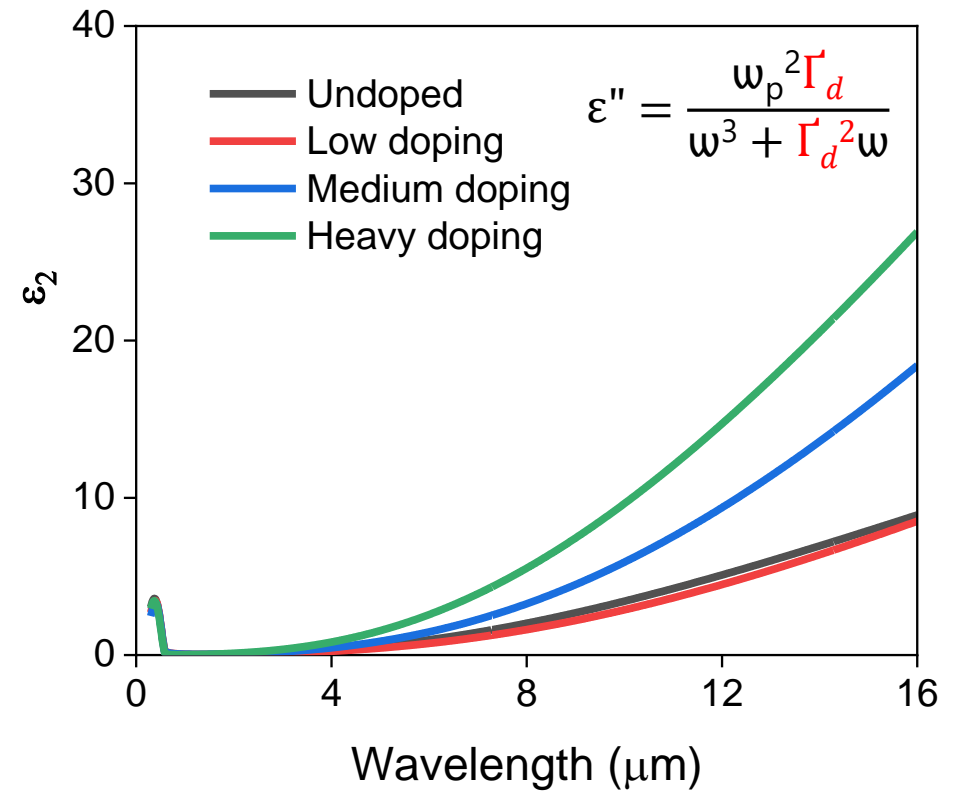
Adding more Y dopant increases electrons and blue shifts the ENZ point from 11 to 5 microns, making the films more metallic



$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^*}$$

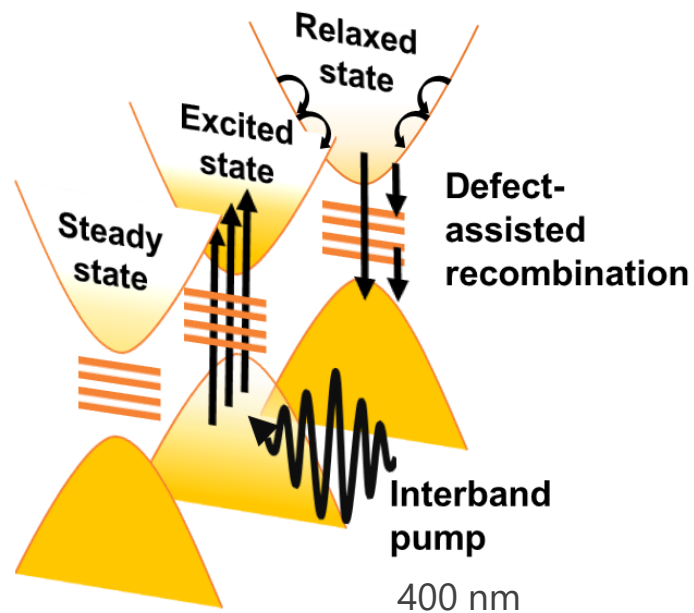
$$\epsilon' = \epsilon_B - \frac{\omega_p^2}{\omega^2 + \Gamma_d^2}$$

Increased electrons and defects increases the losses with doping

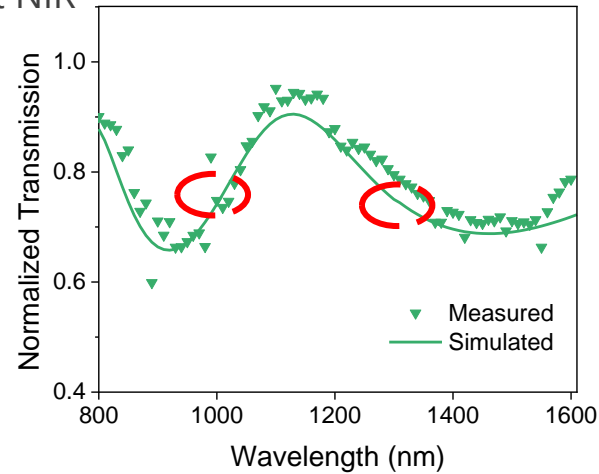


$$\epsilon'' = \frac{\omega_p^2 \Gamma_d}{\omega^3 + \Gamma_d^2 \omega}$$

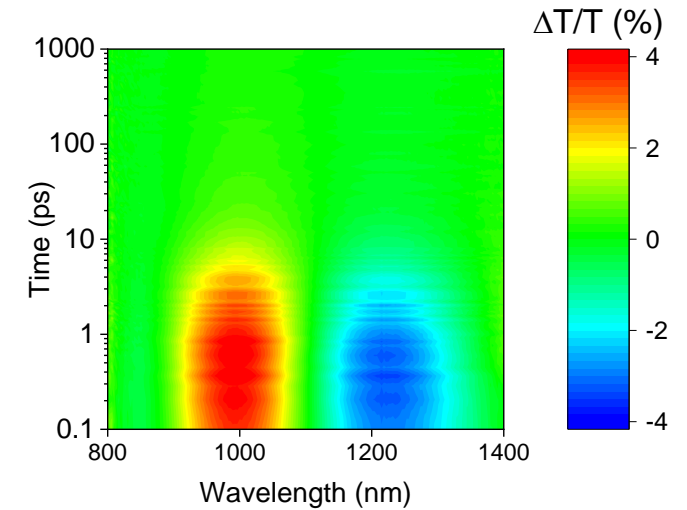
# ENZ Enhanced All-optical Switching in Mid-IR



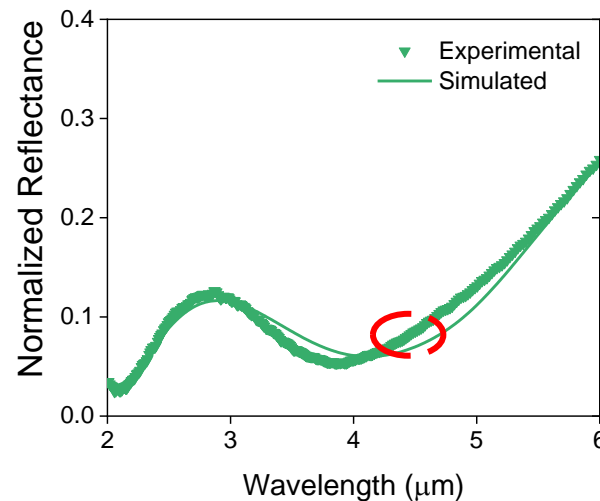
Steady state transmittance vs wavelength at NIR



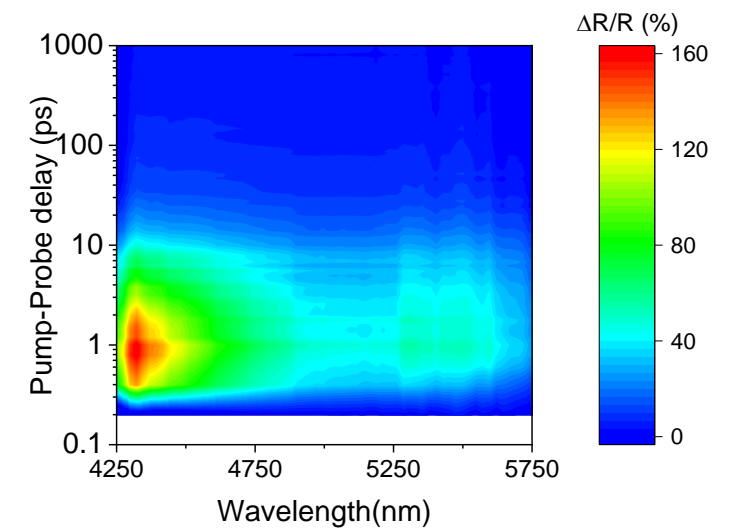
Moderate modulation near transmittance dips (4%)



Steady state reflectance vs wavelength at MIR

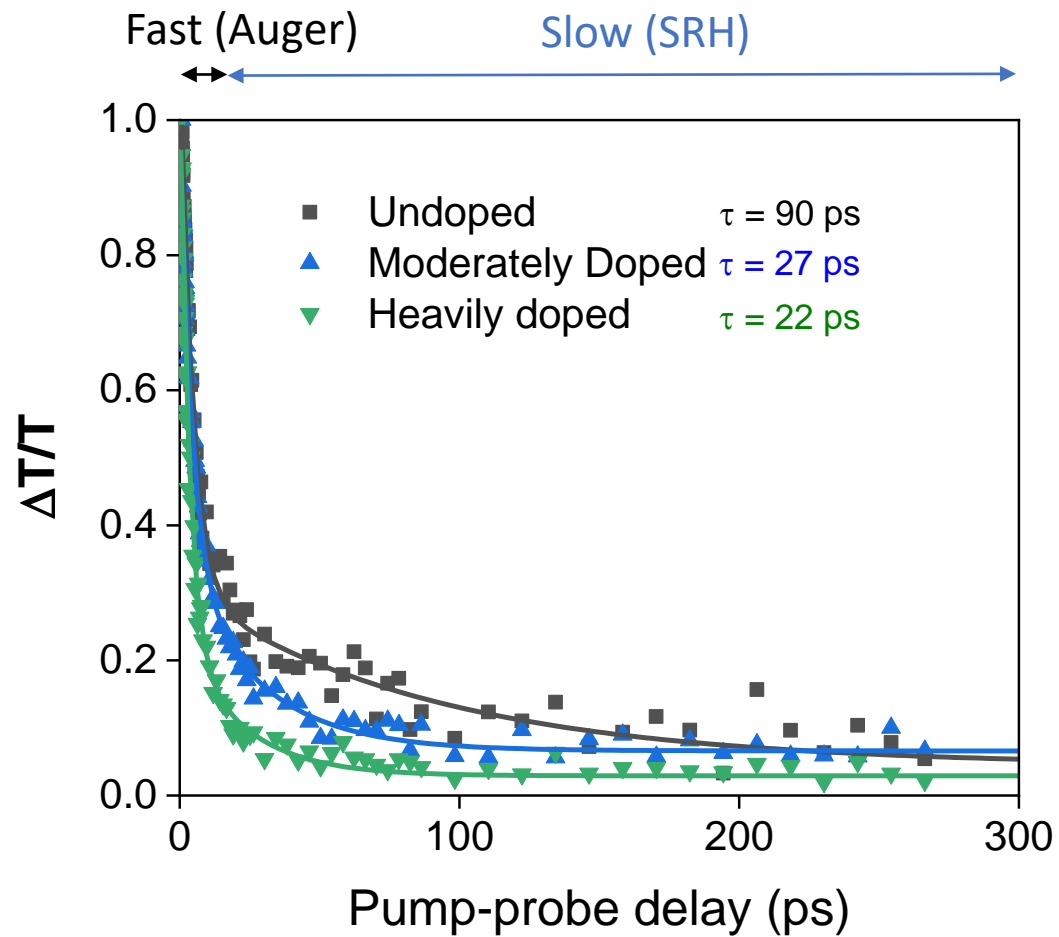


Further enhanced near the ENZ point (160%)

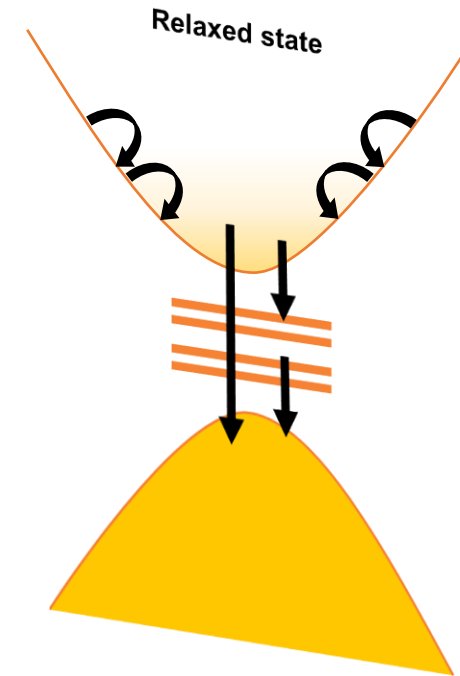




# The Overall Response Time can be Engineered by Doping



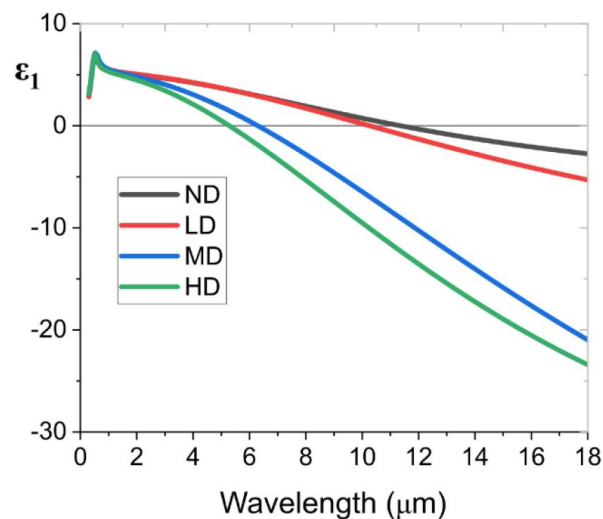
## Defect-assisted recombination



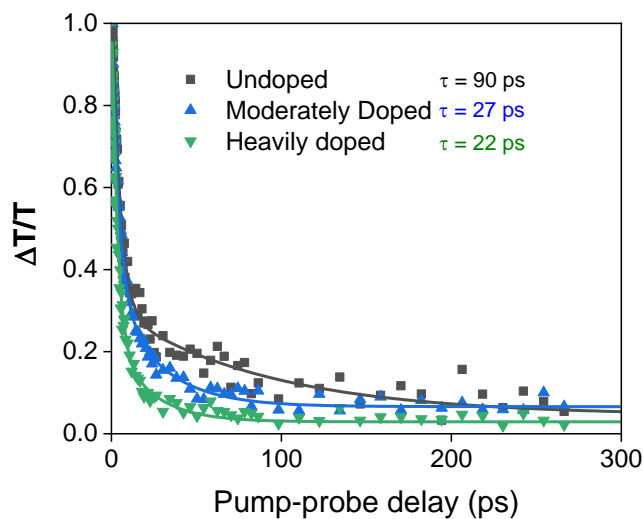
More defects, faster recombination

## Application: All optical switching in the MIR for satellite communications

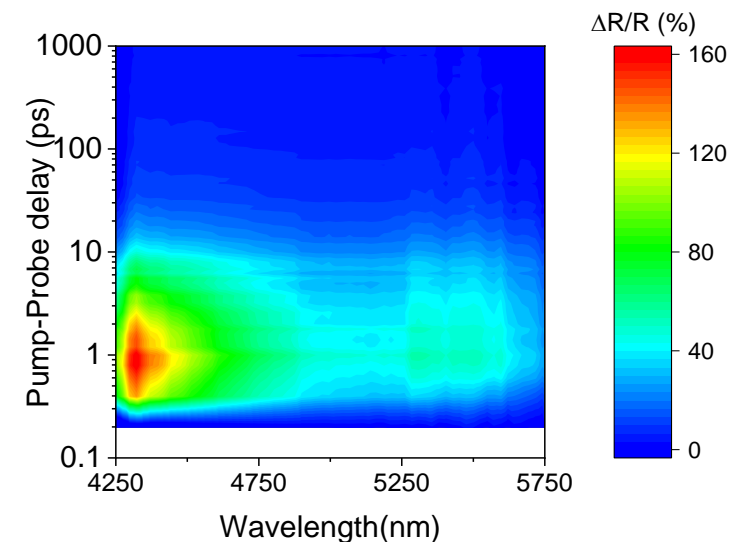
### Application 2: ENZ enhanced phenomena across a broad MIR range



ENZ can be tailored from 5 to 11 microns by adding dopants



Doping reduces the relaxation time



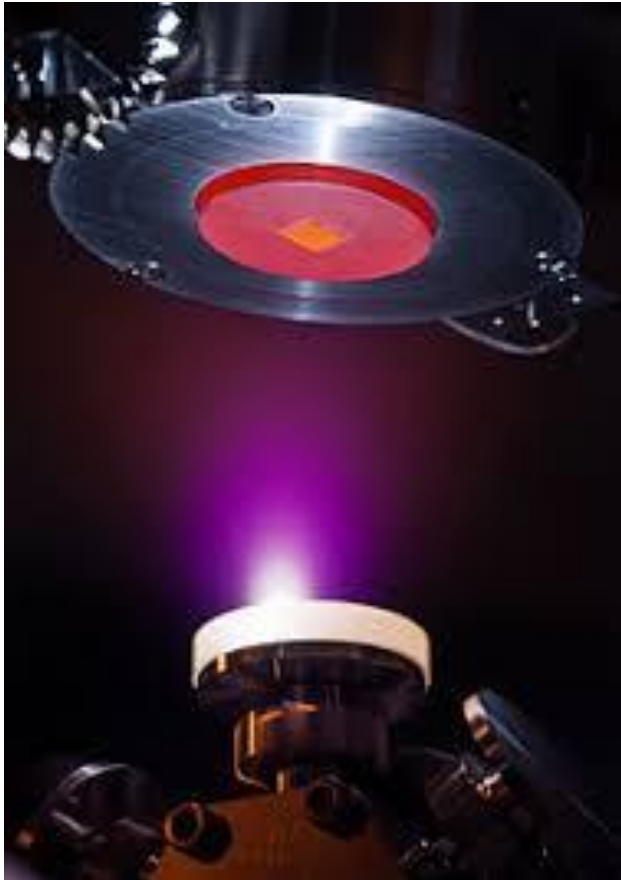
Modulation of 160% near ENZ at  $1.3 \text{ mJ/cm}^2$  pump power with 50ps relaxation time

**Doping introduces losses in the oxide**  
**Large on-state losses**  
**The process is irreversible**

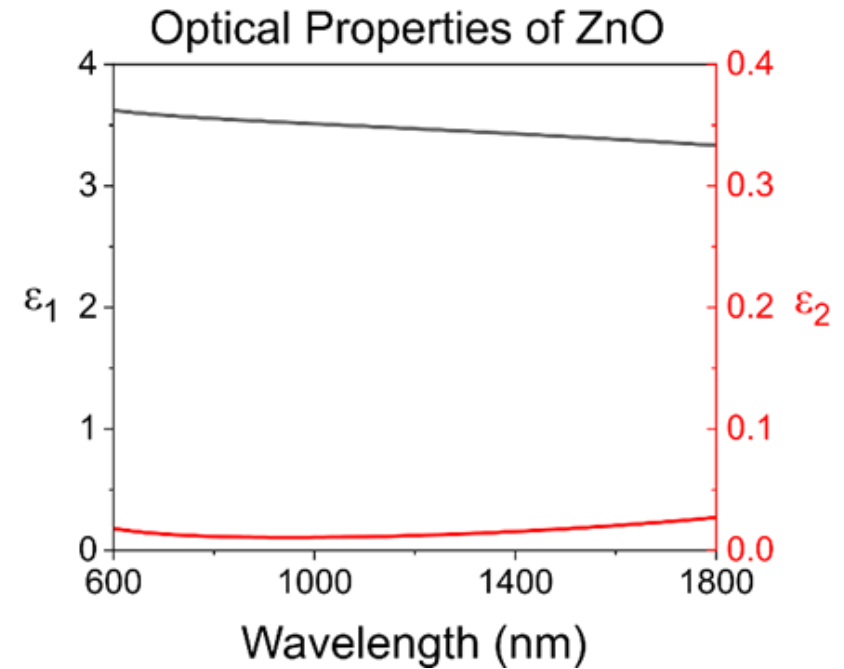
How much can we tune the **permittivity** of zinc oxide **in real time, without** adding dopants?

# Material of Choice: Zinc Oxide (Undoped)

ZnO films grown by  
Pulsed Laser Deposition

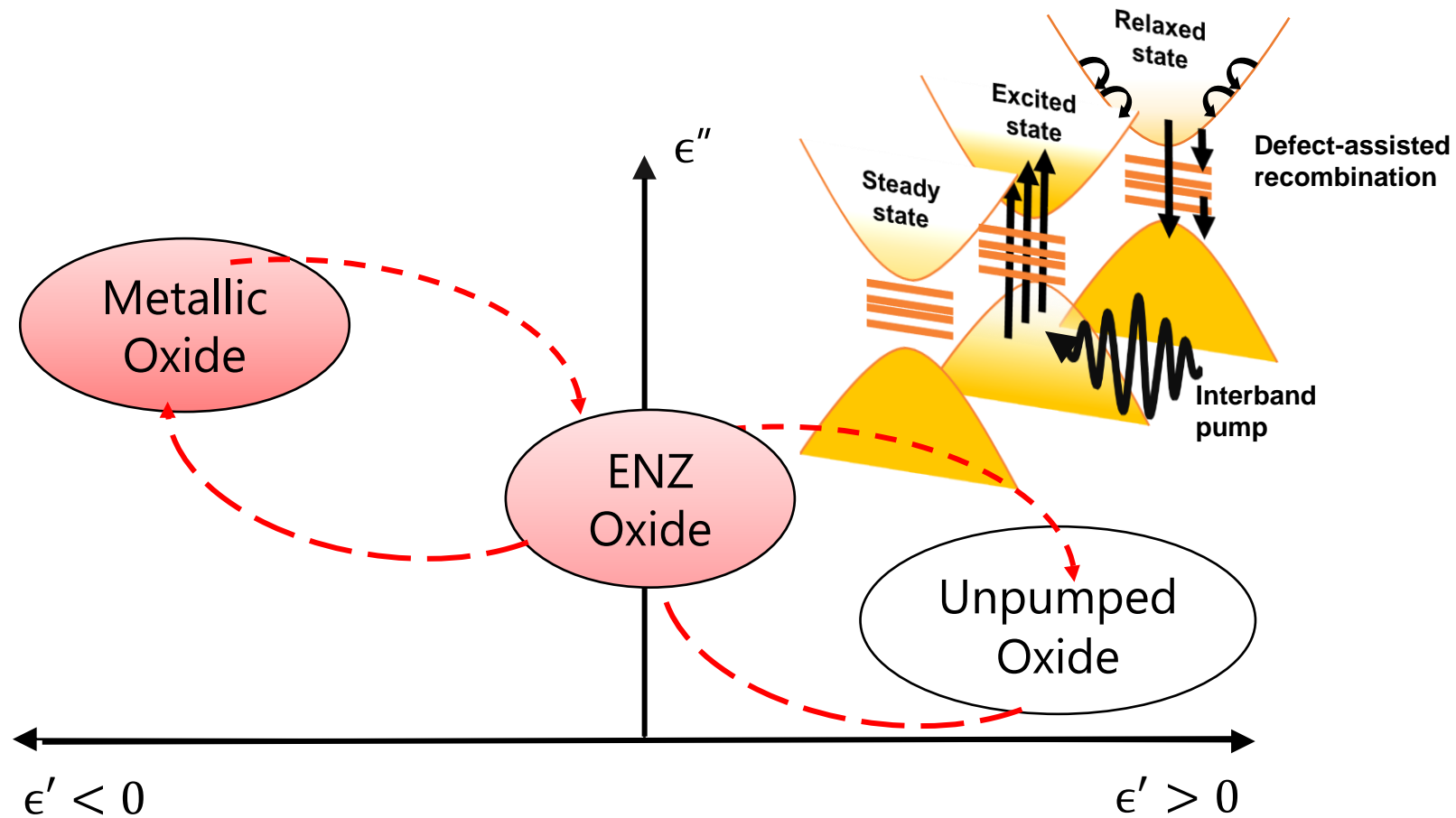


Dielectric with low optical losses



$$\varepsilon = \varepsilon_{\infty} + \frac{A_1}{E_1^2 - (\hbar\omega)^2 - iB_1\hbar\omega} - \frac{A_0}{(\hbar\omega)^2 + iB_0\hbar\omega}$$

# How do We Tune the Permittivity ?



$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^*}$$

$$\epsilon' = \epsilon_B - \frac{\omega_p^2}{\omega^2 + \Gamma_d^2}$$

$$\epsilon'' = \frac{\omega_p^2 \Gamma_d}{\omega^3 + \Gamma_d^2 \omega}$$

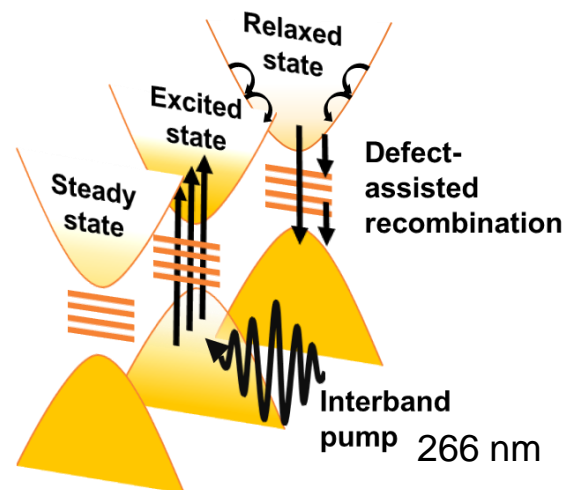
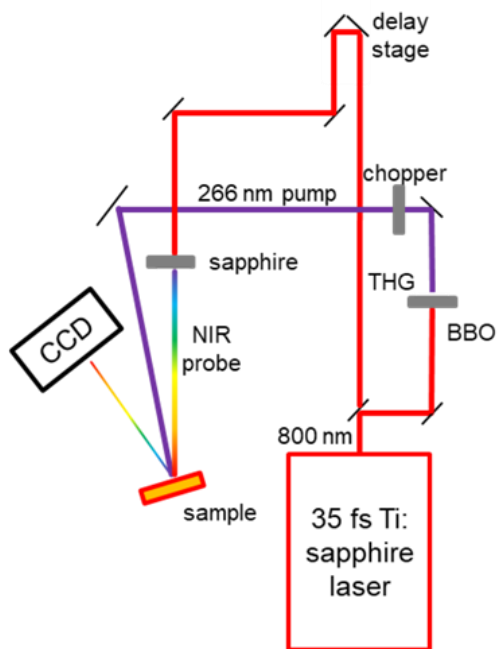
Optically pumping with an interband pump, generating electron hole pairs

Tuning is a dynamic method: Instantaneous, reversible results

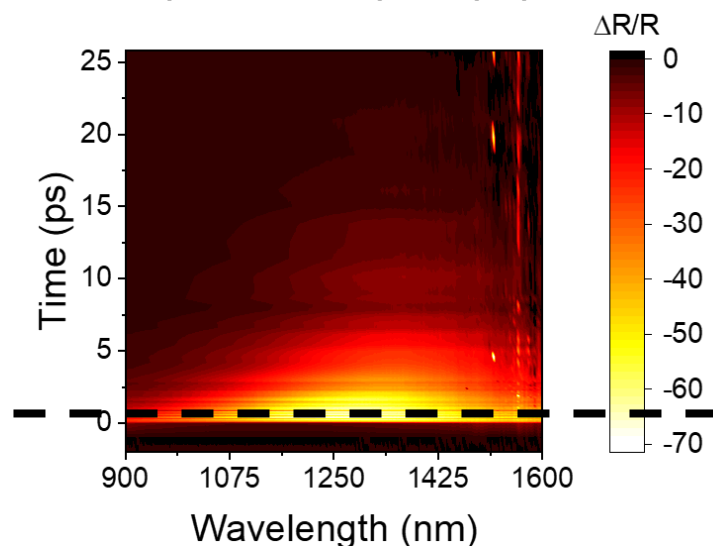


# Interband Pump – Near Infrared Probe

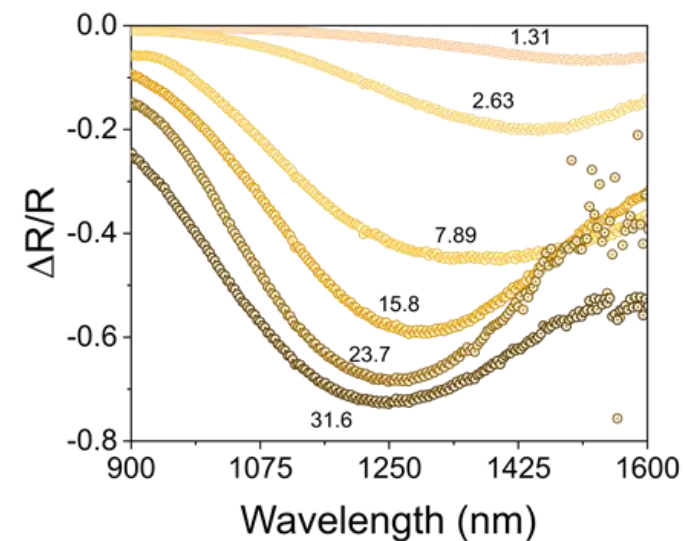
Pump-probe schematic



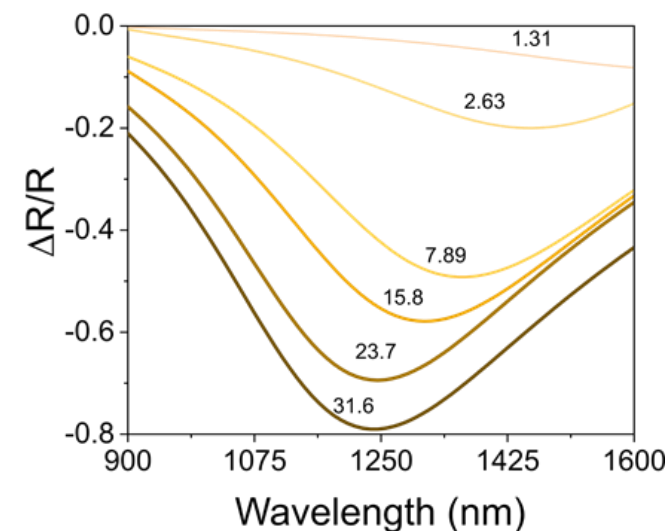
$\Delta R/R$  captured vs pump-probe delay



Reflectance extracted

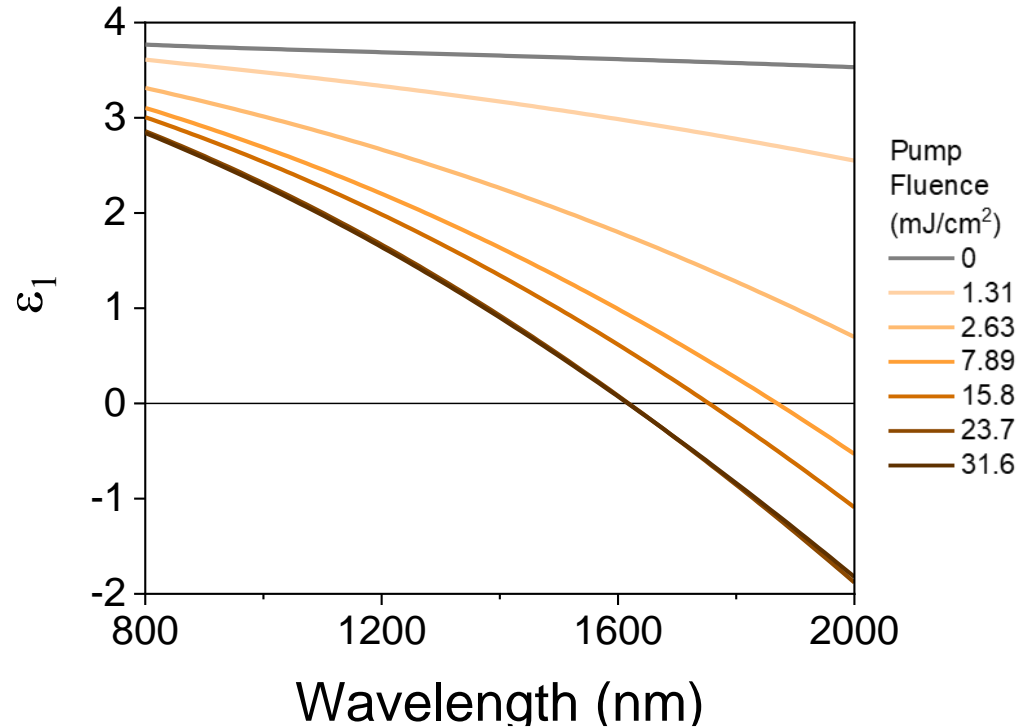


Drude Parameters extracted from fits



# Unity Order Permittivity Changes at Telecom Wavelengths

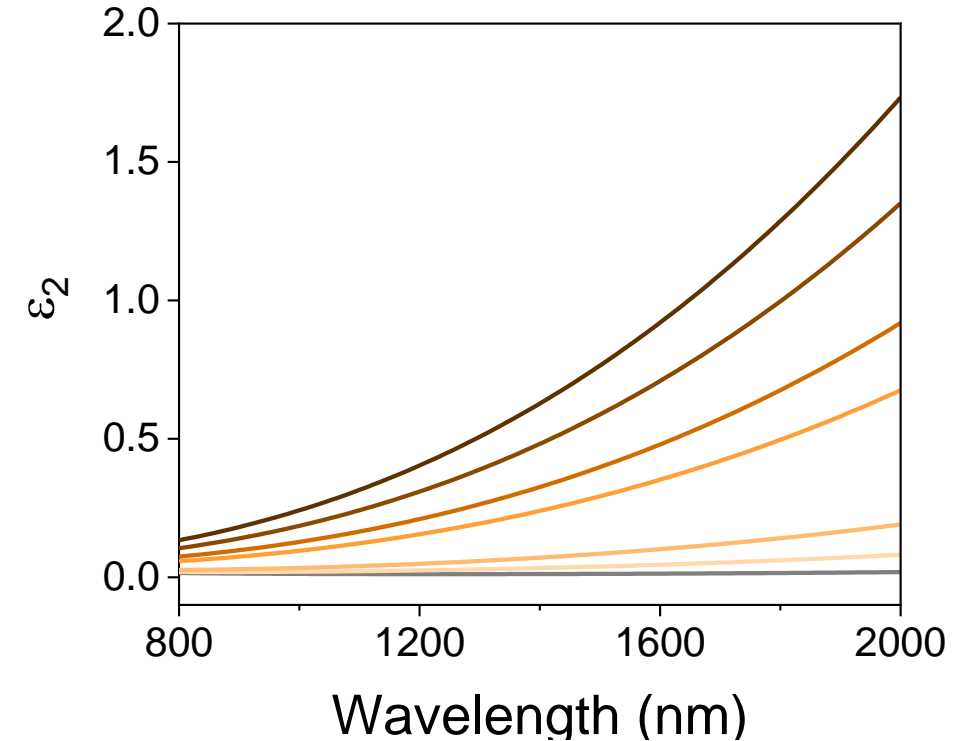
Extraordinary permittivity modulation  
(-4 at 1600 nm) saturates at 23 mJ/cm<sup>2</sup>



N saturates due to saturable absorption  
As N rises, m\* rises due to non parabolic band

$$\omega_p^2 = \frac{Ne^2}{\epsilon_0 m^*} \quad \epsilon' = \epsilon_B - \frac{\omega_p^2}{\omega^2 + \Gamma_d^2}$$

With increased power, the absorption in the films increase

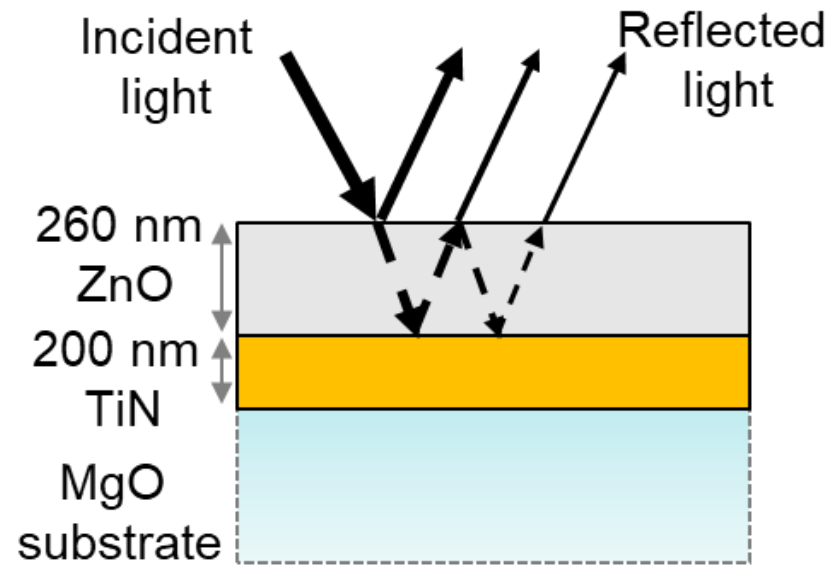


Increased free carrier concentration and heating of the lattice increases the scattering  $\Gamma$

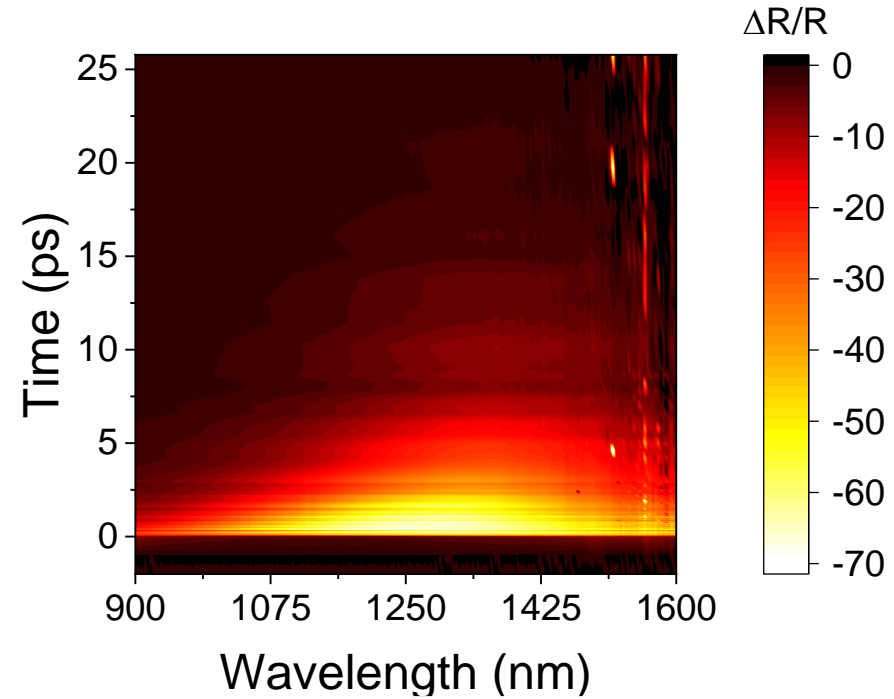
$$\epsilon'' = \frac{\omega_p^2 \Gamma_d}{\omega^3 + \Gamma_d^2 \omega}$$

# Large, Broadband Modulation in Planar Mirrors Without Lithography

Lithography-free mirror



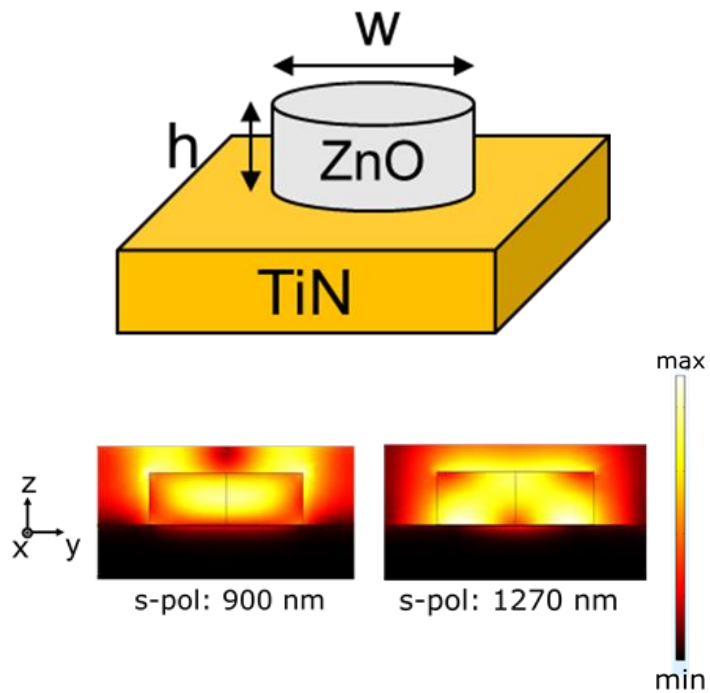
Broadband modulation of 70% at telecom  
Without Doping or patterning



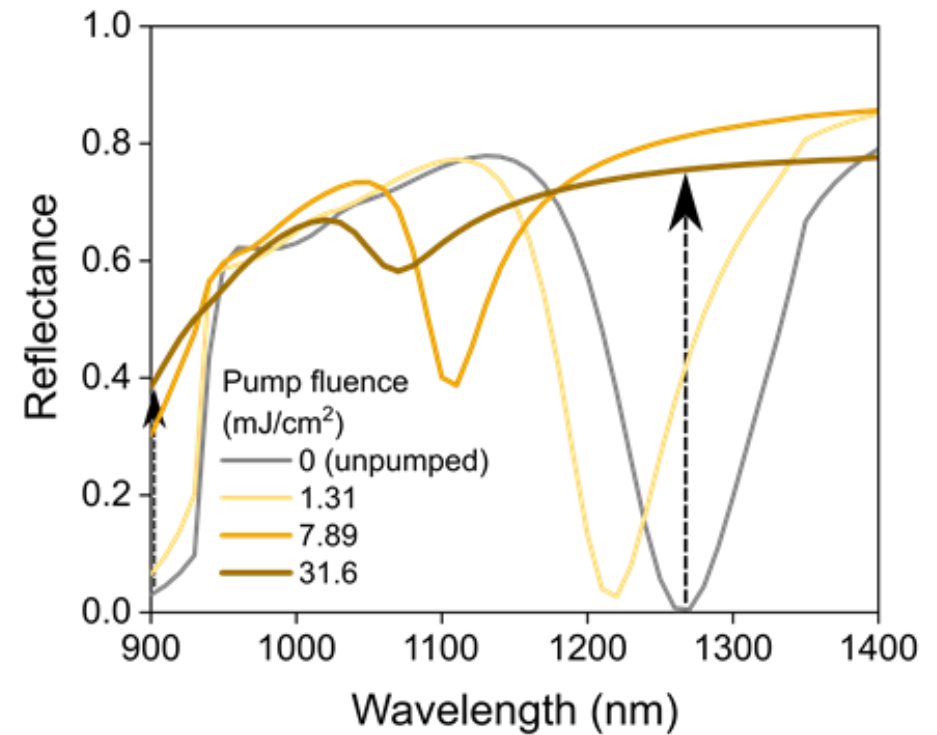
20ps response time!

# Selective Enhancement at Specific Wavelengths: Design

## Hybrid Plasmonic Resonators

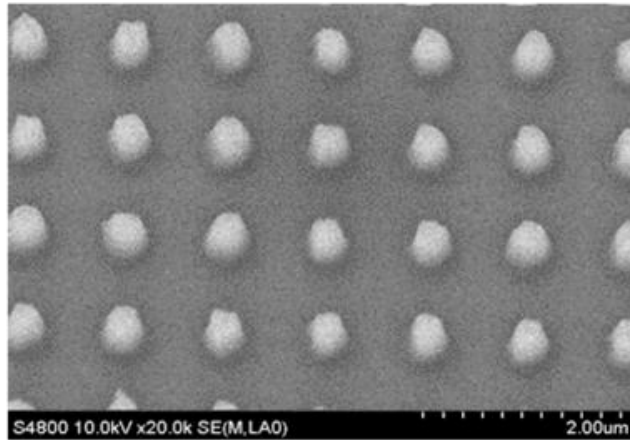


## Resonance shifts under a pump

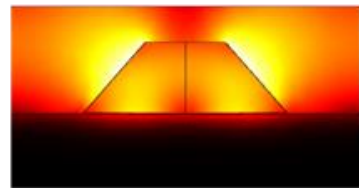
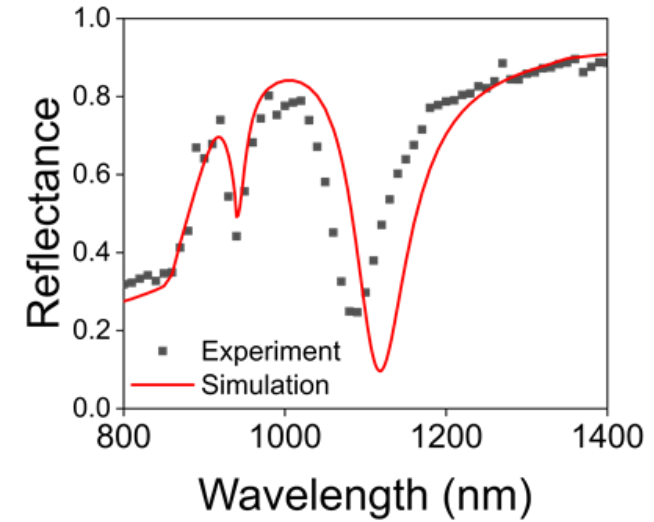


# Selective Enhancement at Specific Wavelengths

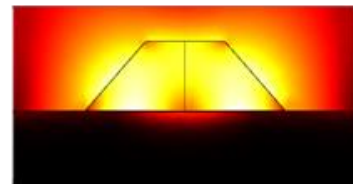
ZnO nanodisks on TiN



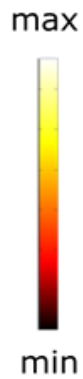
Steady-state resonance shows two dips



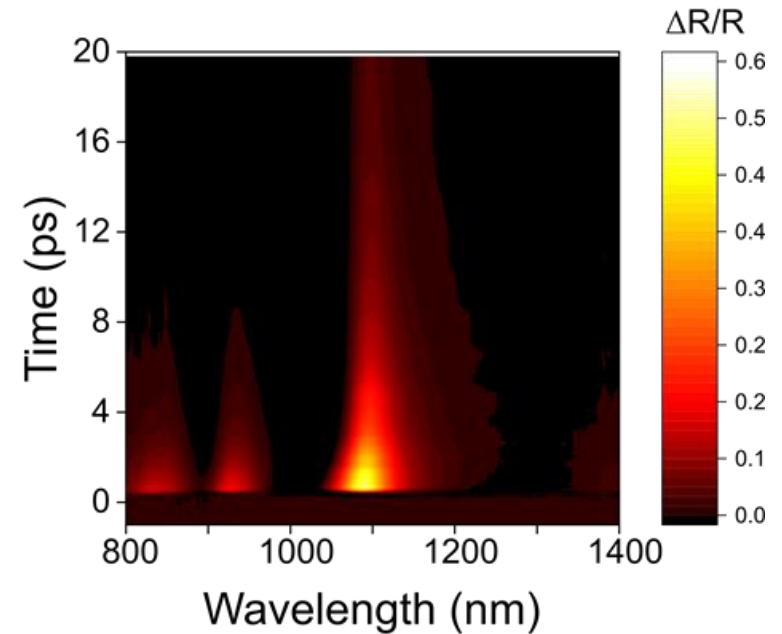
940 nm



1120 nm



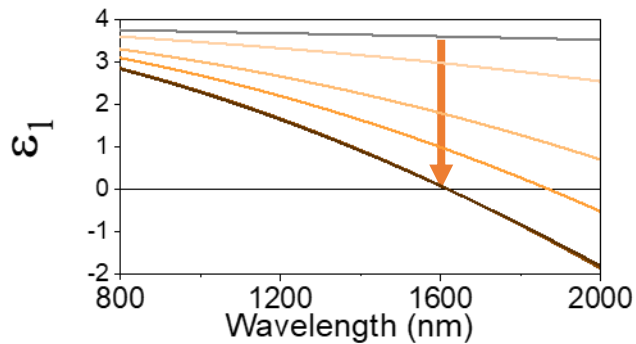
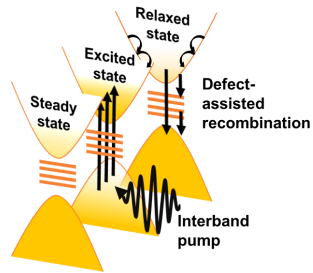
Maximum change of 55% at 7.8 mJ/cm<sup>2</sup> with 20 ps Response time





# Summary

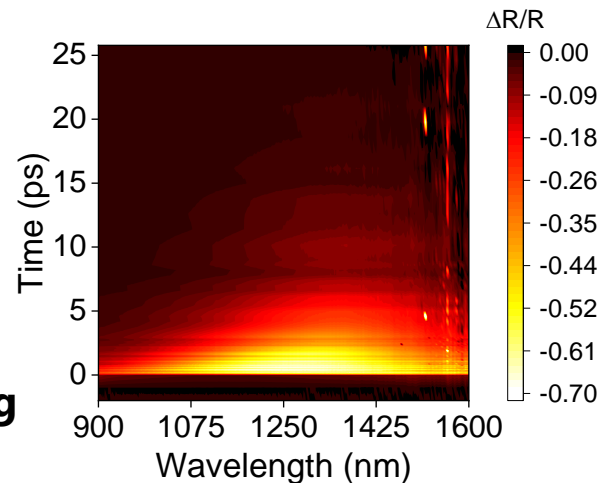
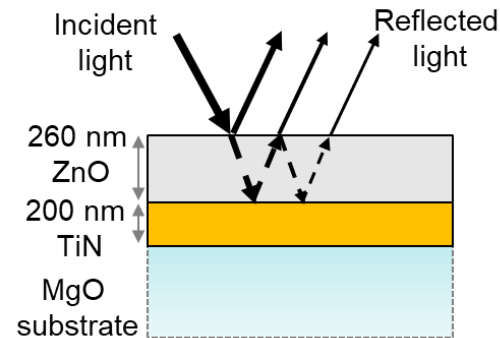
Large optical changes are demonstrated in UNDOPED ZnO by interband pumping



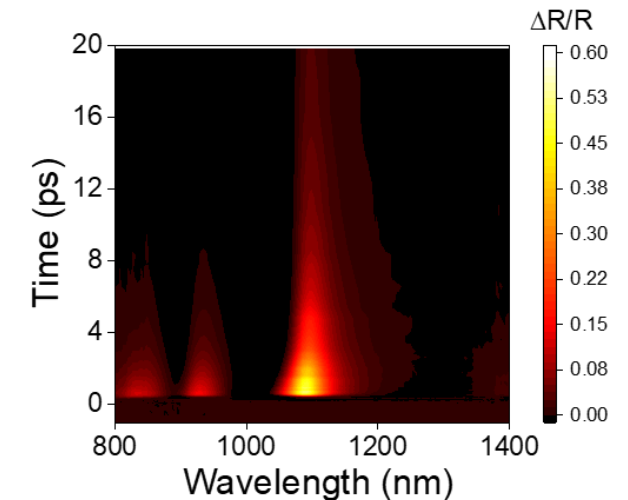
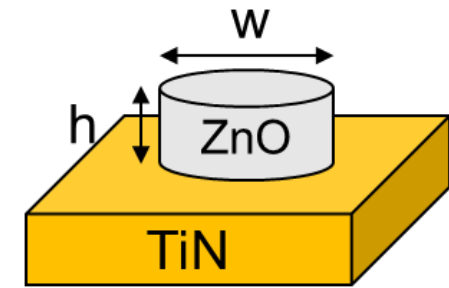
**Application: All optical switching**

**Transient permittivity data useful for designing dynamic polarization switches, HMMs, etc**

Planar metal-backed ZnO mirror can be used to demonstrate broadband modulation with ps response time

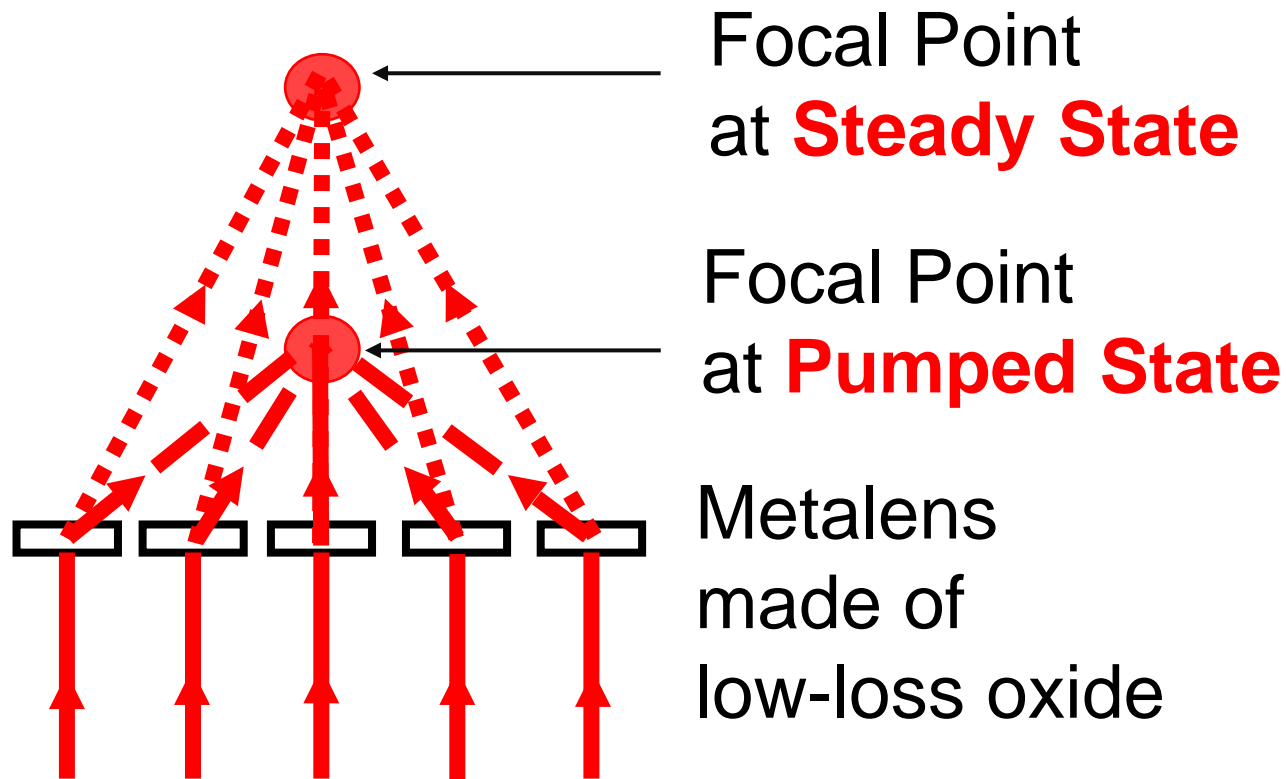


The resonances can be selectively enhanced at specific wavelengths with simple resonant structures



# Optically Tunable Lens

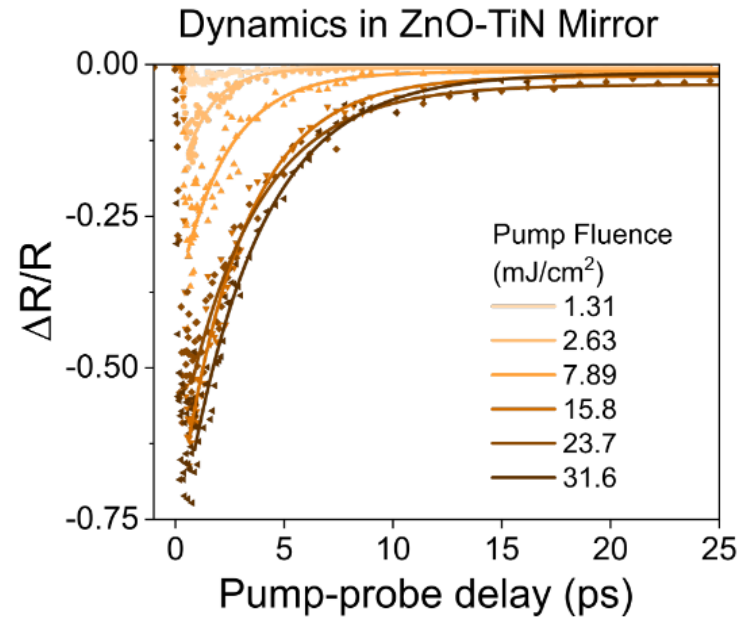
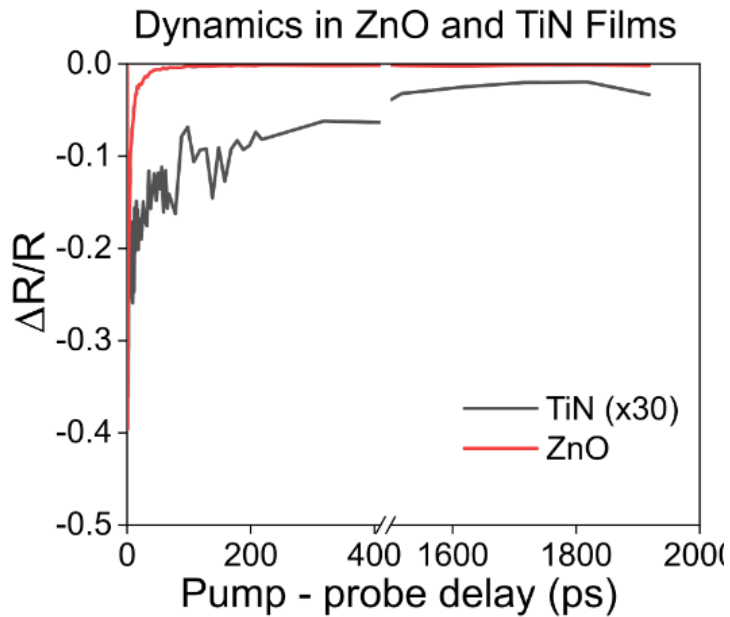
Tunable focusing, polarization rotators, filters, etc



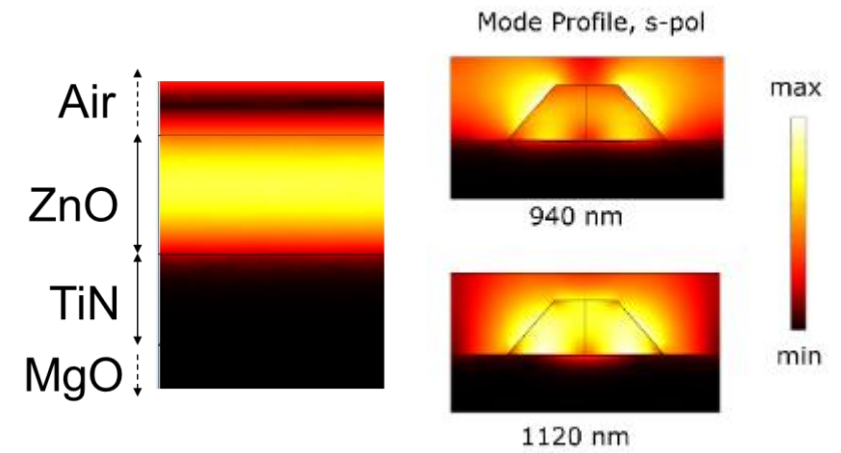
## Impact

- Focusing high power beams used to burn off tissue
- All-optical scanning

# The Switches are Fast, Despite the Slower Response of TiN



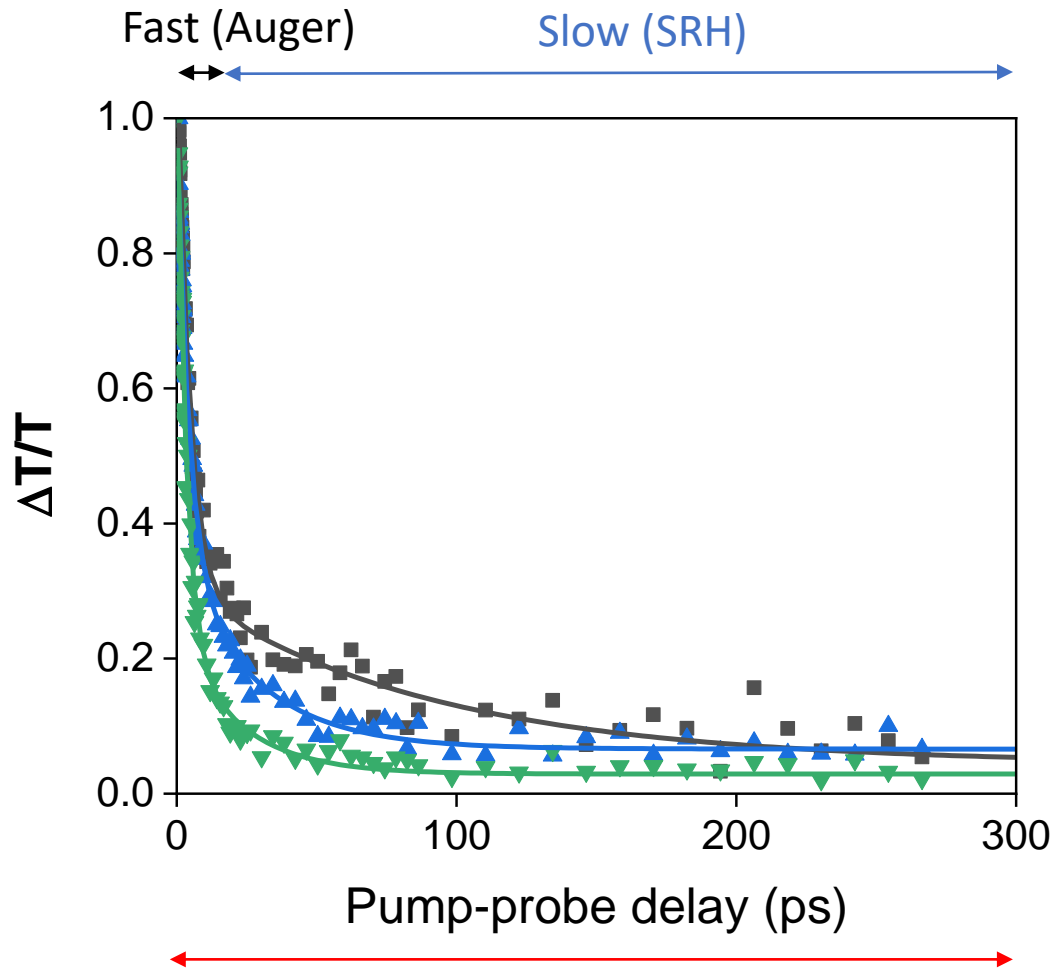
Most of the light matter interaction happens in the ZnO



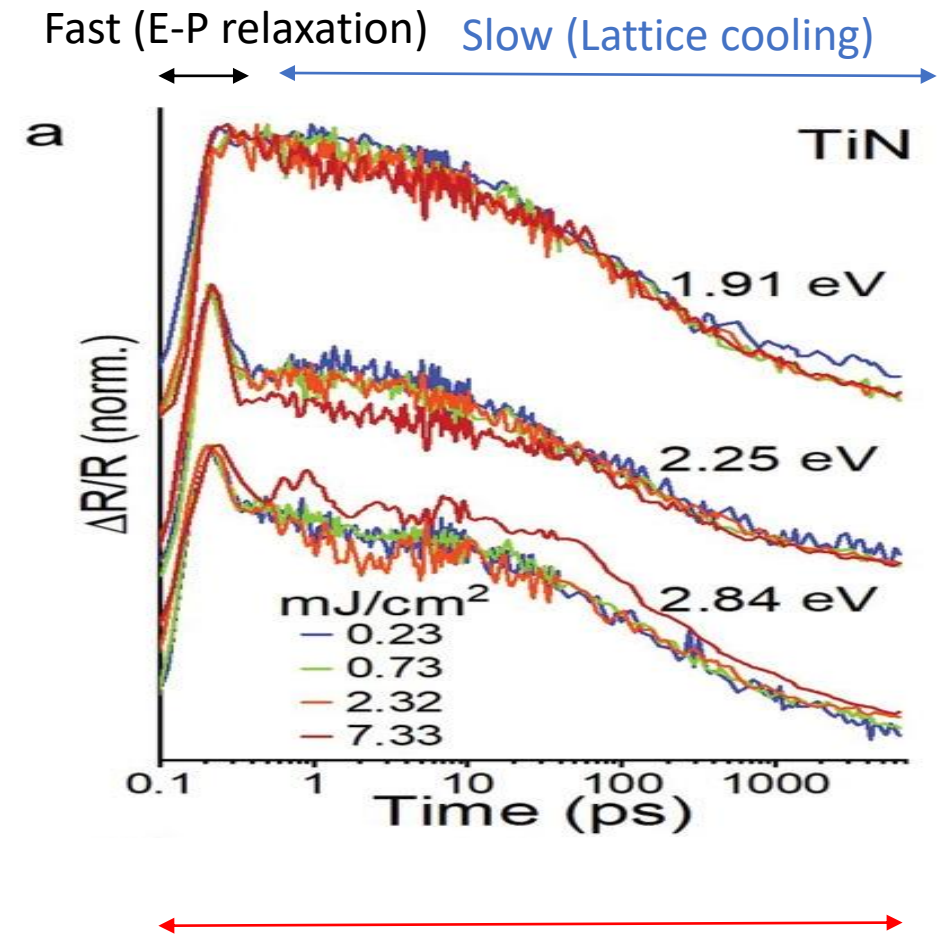
As a result, ZnO dictates the modulation dynamics, even when TiN is excited

Can we **control** the **speed** of an all-optical switch?

# In Materials, Response Time is Dictated by the Slowest Dynamics



Saha et. al, Adv. Func. Mat. (2019)

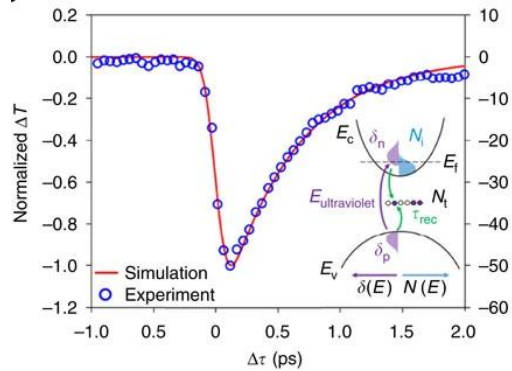


B. Diroll, S. Saha et. al, Adv. Opt. Mat. (2020)

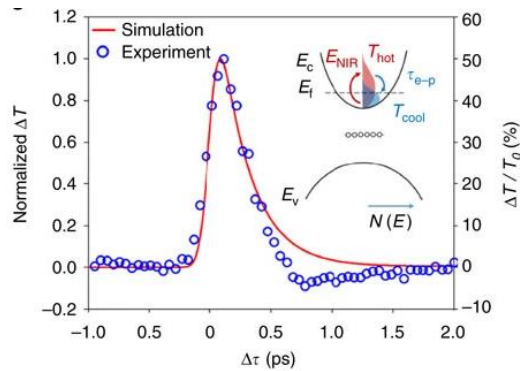


# Pump Induced Control of Switching Dynamics

Utilizing interband vs intraband  
Relaxation times

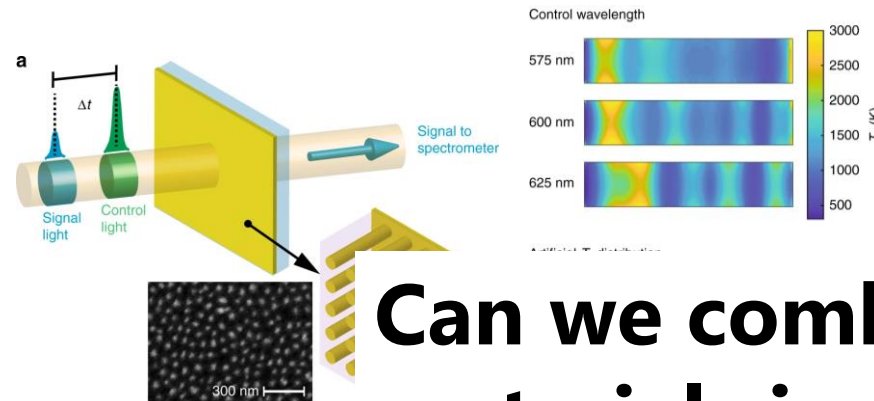


Interband – picoseconds

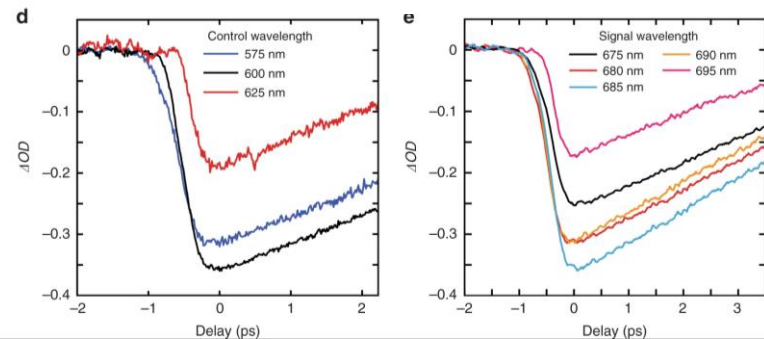


Intraband – 100s of fs

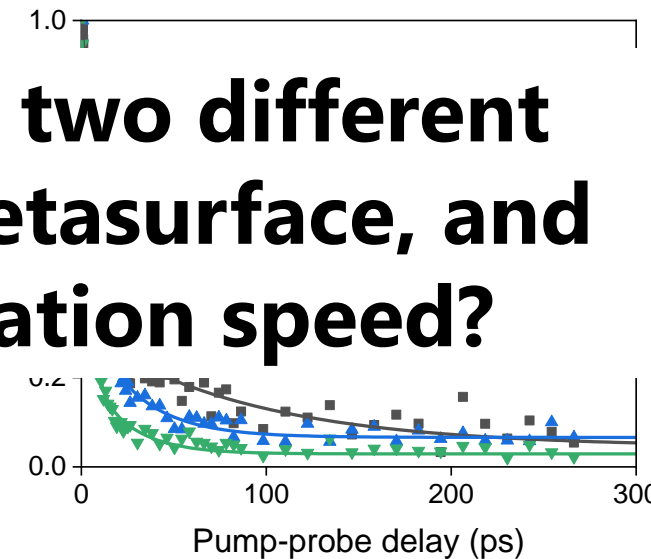
Controlling temperature distribution in nanorods controls the photon dynamics to an extent



**Can we combine two different materials in a metasurface, and control the operation speed?**



Controlling doping to control relaxation times



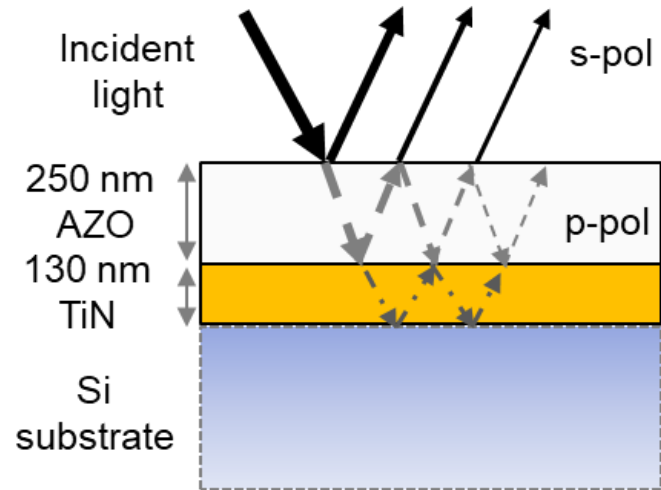
Clerici et al., *Nat Comm.* 2016

Zayats et al., *Nat Comm.* 2019

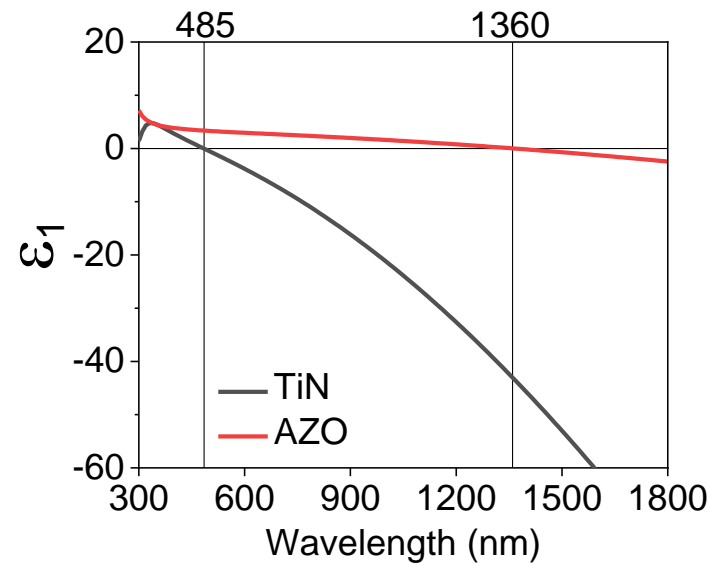
Saha et al., *Adv. Opt. Mat.* 2020

# Berremian Metasurface Strongly Absorbs p-polarized Light Near ENZ

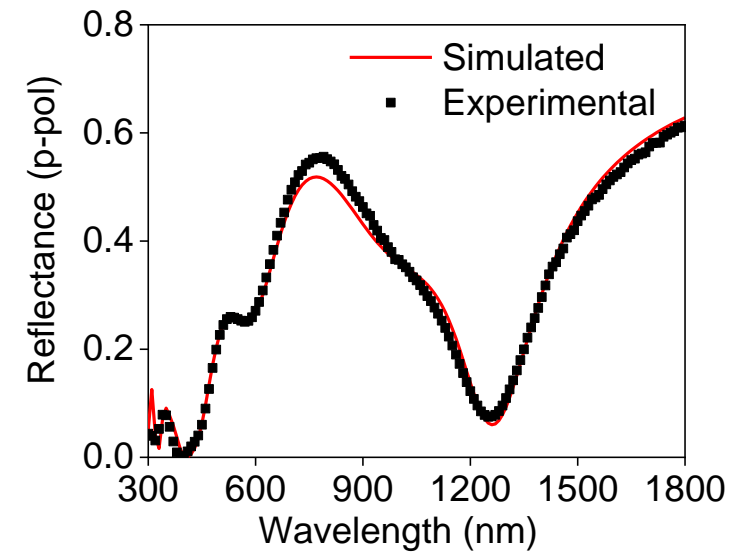
## AZO film on TiN



## ENZs of TiN and AZO

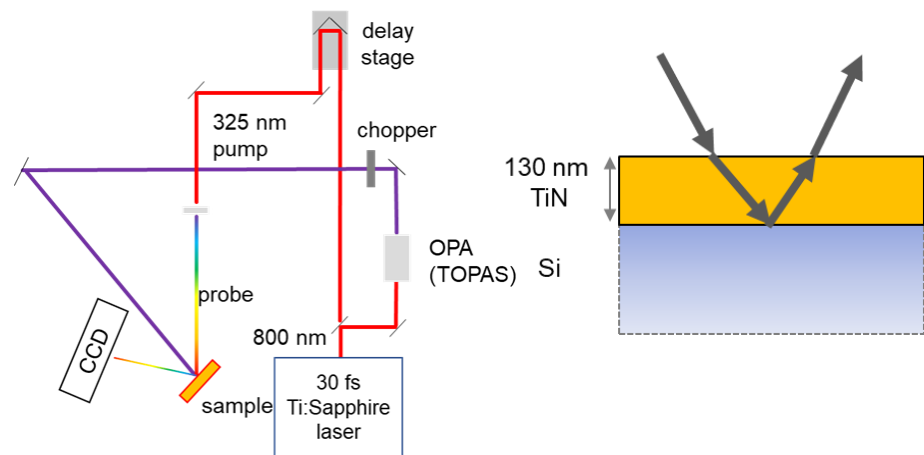


## Near perfect absorption near ENZ

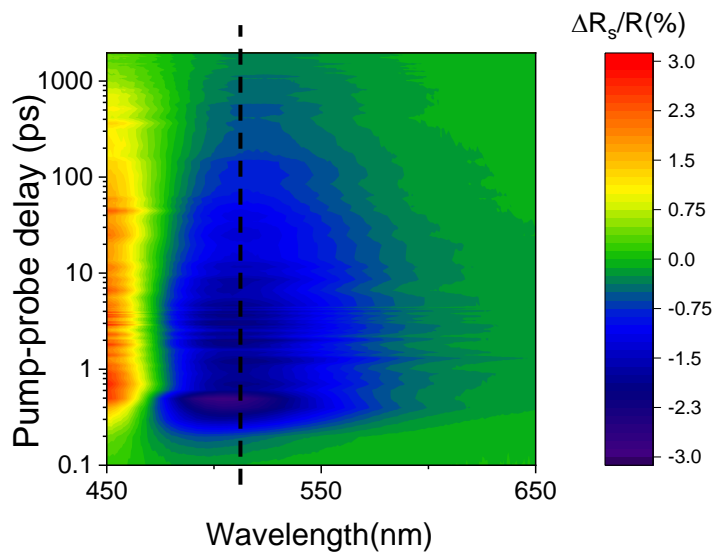
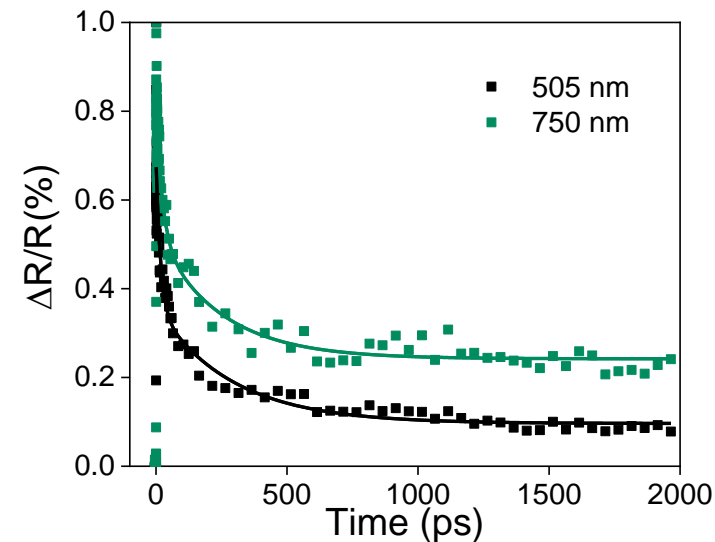
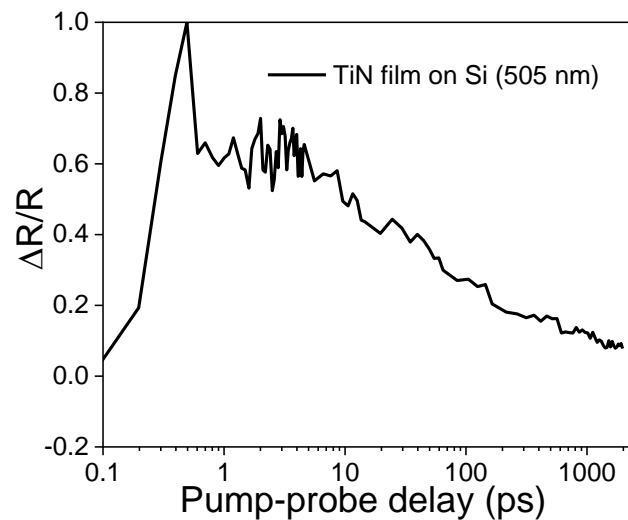


# TiN is Slow (325 nm pump, Vis probes, 1.3 mJ/cm<sup>2</sup>)

## Experimental Setup



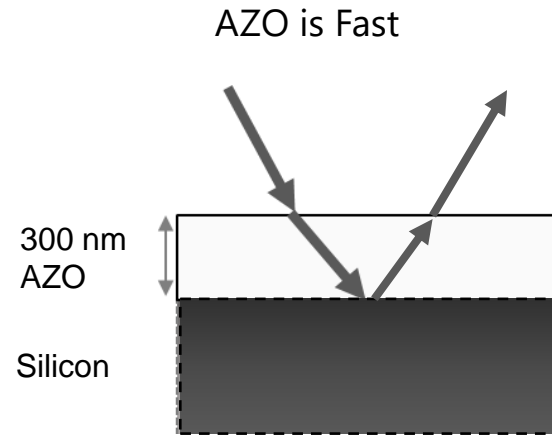
## TiN Response time > 2 ns



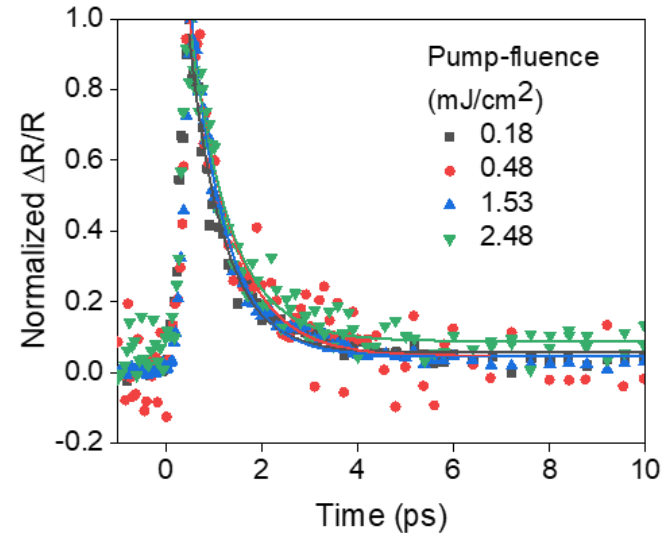
$$\left(\frac{\Delta R}{R}\right)(t) = A e^{\frac{-t}{\tau_1}} + B e^{\frac{-t}{\tau_2}} + C$$

Wavelength (nm)	A	B	C	$\tau_1$ (ps)	$\tau_2$ (ps)
505	0.33	0.26	0.096	17.3	287
750	0.29	0.28	0.24	24.3	242

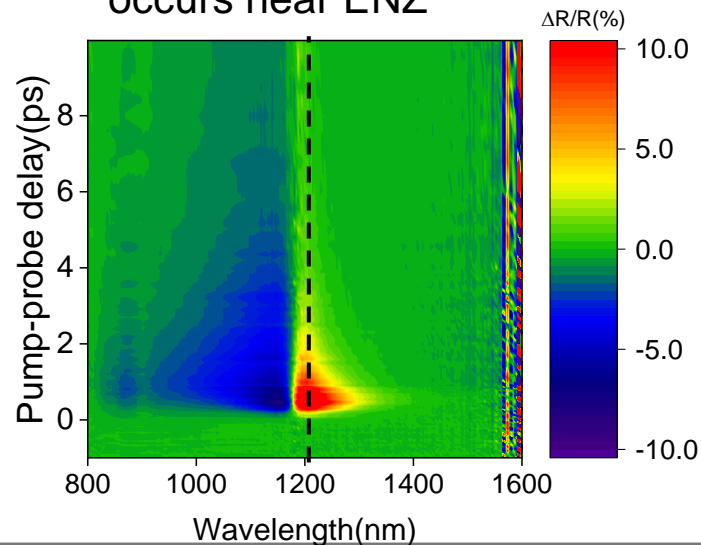
# AZO is Fast (325 nm pump, NIR probes, 1.3 mJ/cm<sup>2</sup>)



Sub 10 ps response time



Maximum modulation occurs near ENZ

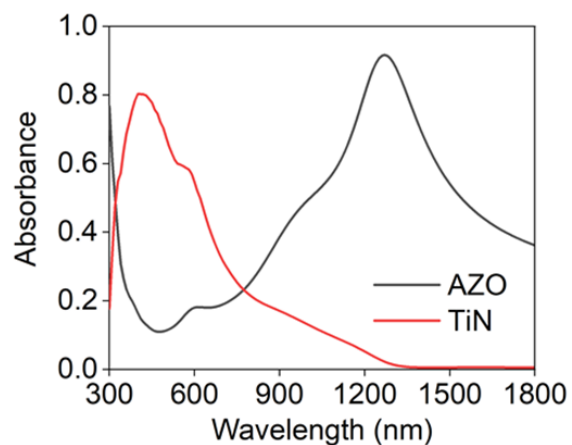
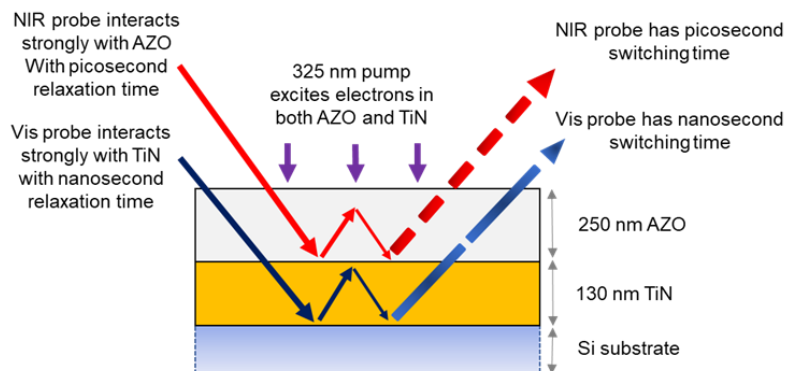


$$\left(\frac{\Delta R}{R}\right)(t) = D e^{\frac{-t}{\tau}} + E$$

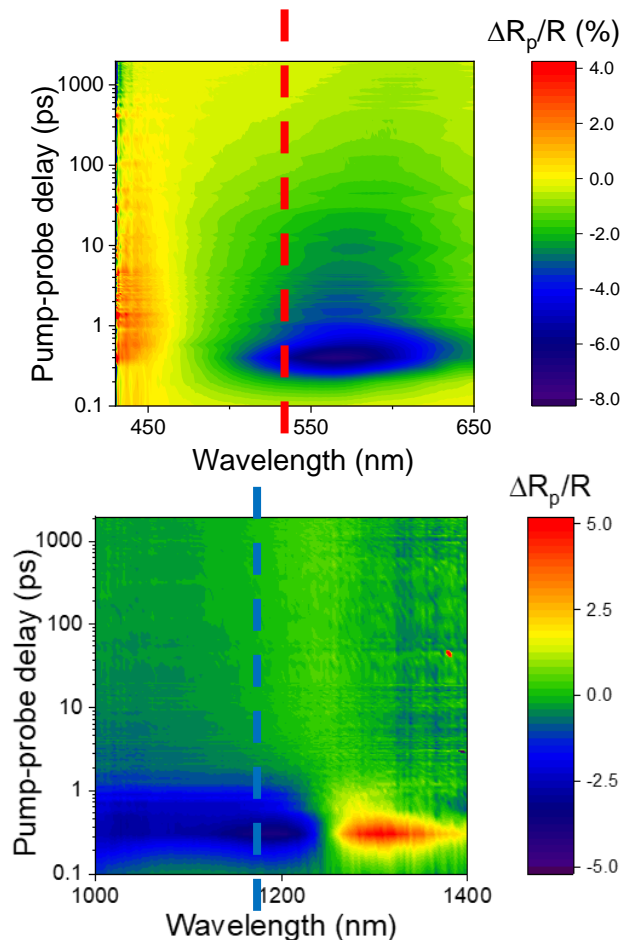
Pump Fluence (mJ/cm <sup>2</sup> )	D	E	$\tau$ (ps)
0.18	1.76	0.056	0.715
0.49	1.69	0.046	0.873
1.53	1.98	0.045	0.723
2.48	1.65	0.088	0.834

# Metasurface Can Operate Both Fast and Slow

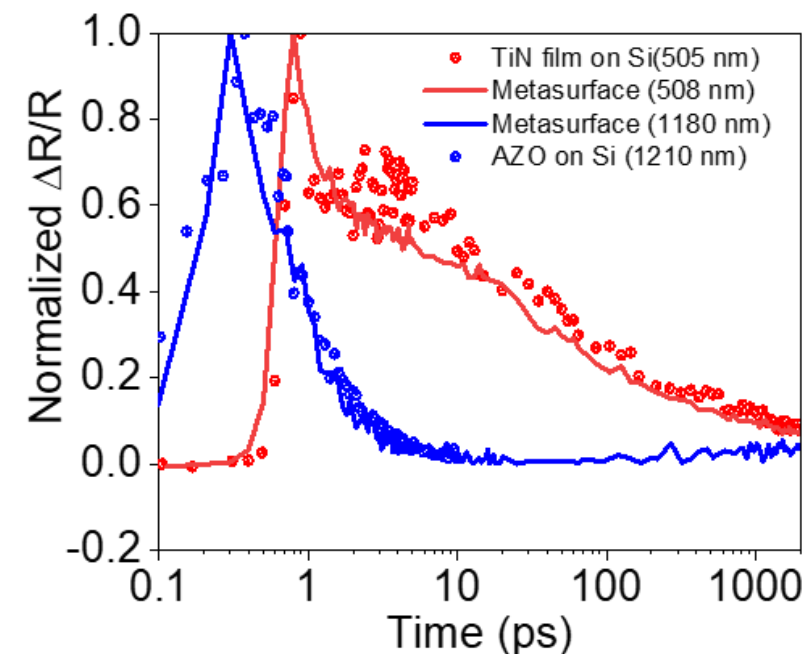
In the metasurface, light is strongly absorbed by the individual material close to the ENZ point



The metasurface reacts slowly near 500 nm, and fast near 1300 nm.



The metasurface switching responses (solid lines) near ENZ match well with that of the individual films (dots)

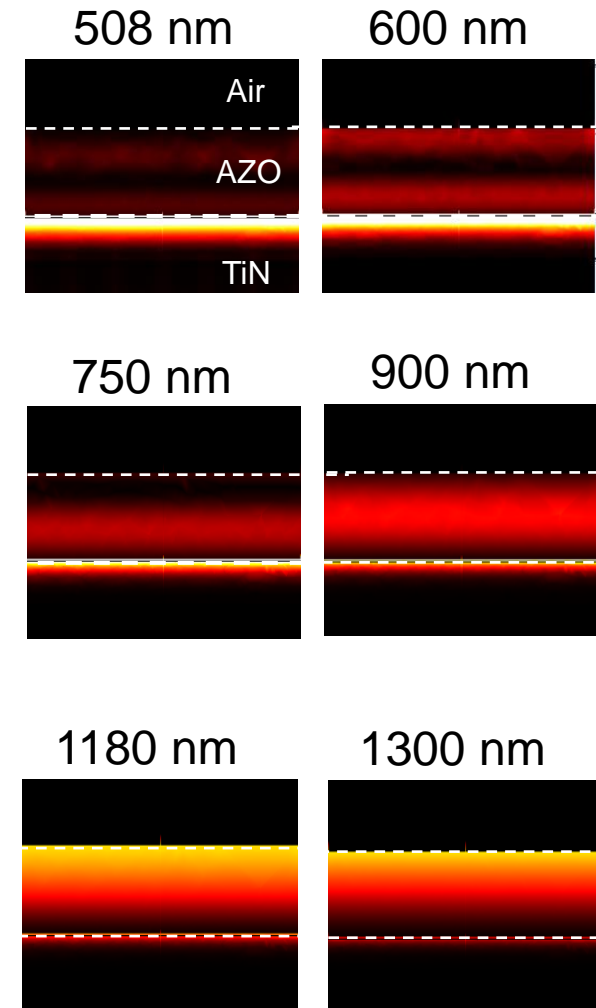
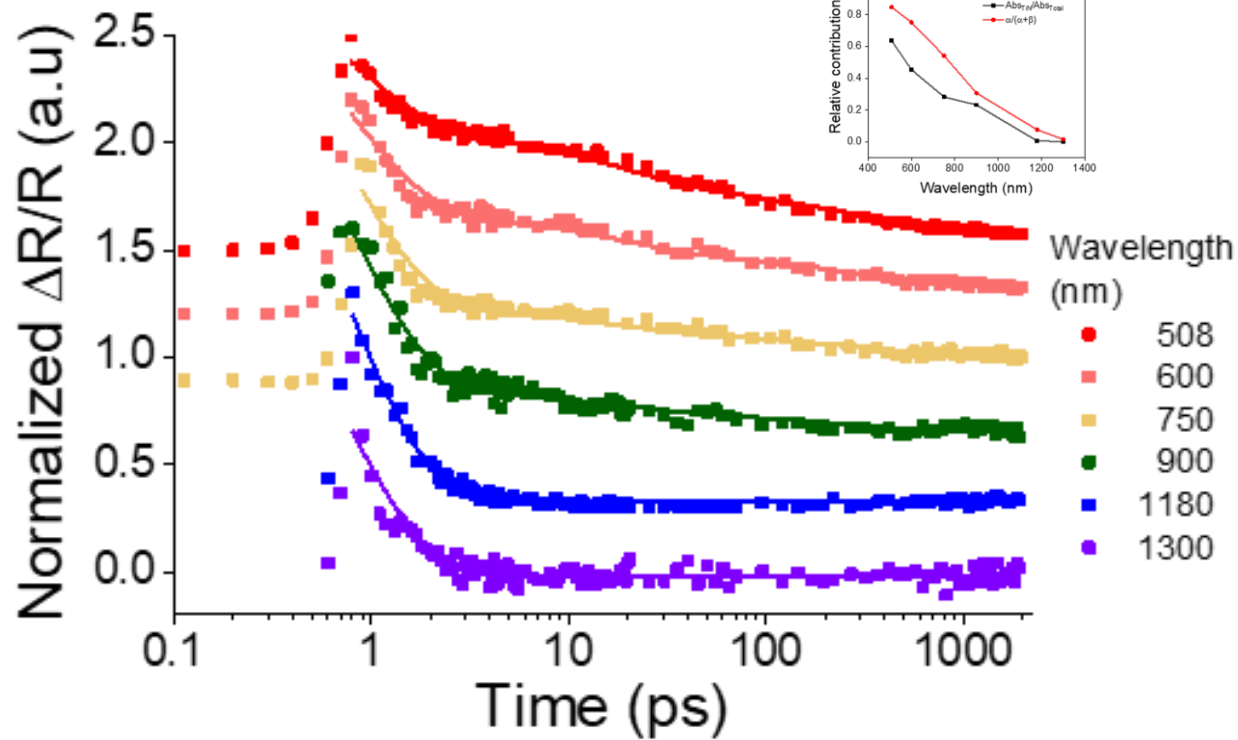




# Designer Dynamics Determined by the Light Matter Interaction

$$\left(\frac{\Delta R}{R}\right)(t) = \underbrace{\alpha(Ae^{\frac{-t}{17.3}} + Be^{\frac{-t}{287}})}_{\text{TiN response}} + \underbrace{\beta e^{\frac{-t}{0.72}}}_{\text{AZO response}} + \Gamma$$

The decay as a weighted sum of the TiN and AZO dynamics

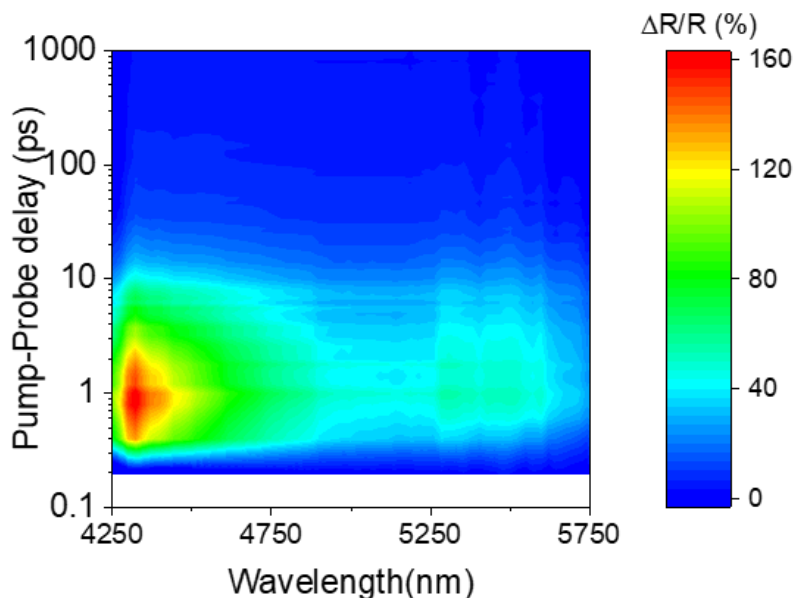


# Final Summary

Understanding tuning and tailoring can help us understand the steady state and temporal dynamics of TCOs  
A deeper understanding of the materials will help us develop new devices

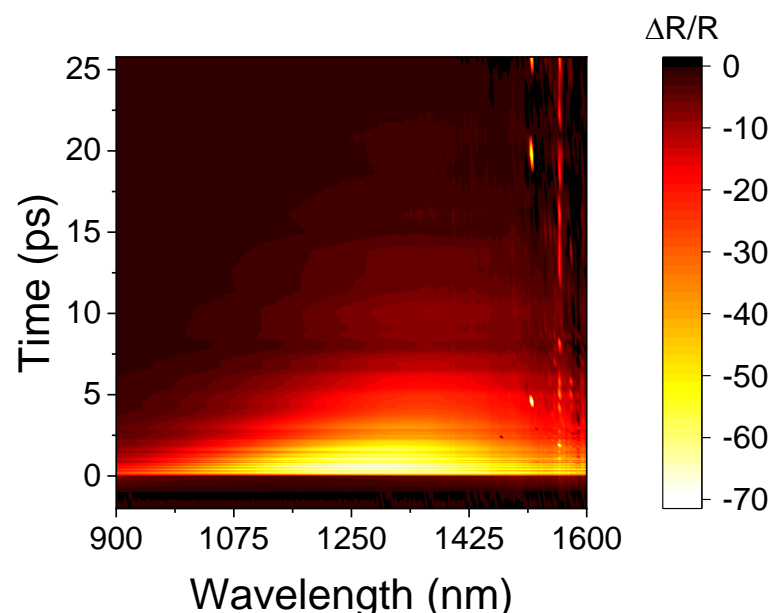
## Y:CdO

- **ENZ Tailoring** with different doping
- Controlling **decay time** with dopants
- **Large modulation** in the MIR



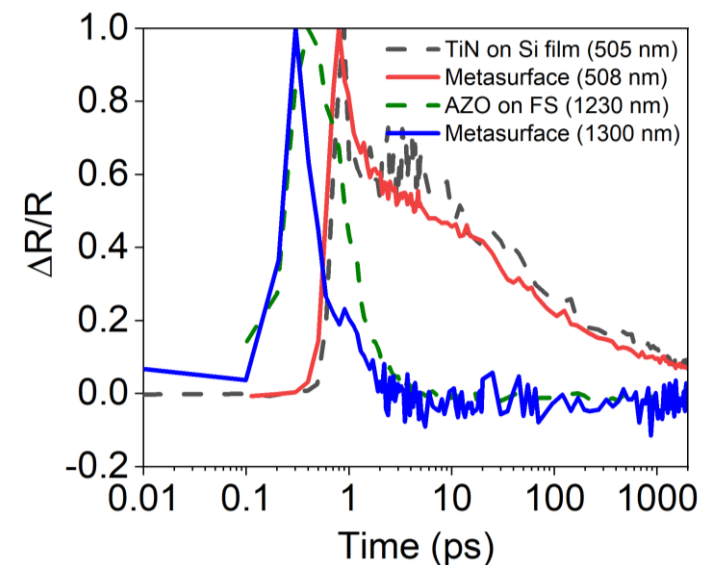
## ZnO

- **Permittivity Tuning** with increased **pump fluence**
- Large modulation in the NIR



## TiN-AZO

- **Fast** and **Slow** switching in the same Berreman metasurface
- **<10 ps** to **>3 ns** switching time by controlling the probe



# Acknowledgement

Work on Y:CdO  
published in  
Advanced Functional  
Materials



Work on ZnO modulators  
preprint in arxiv



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