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# High harmonic generation from a degenerate plasma

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&
CINT

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# Acknowledgements

## Contributors to the HHG work

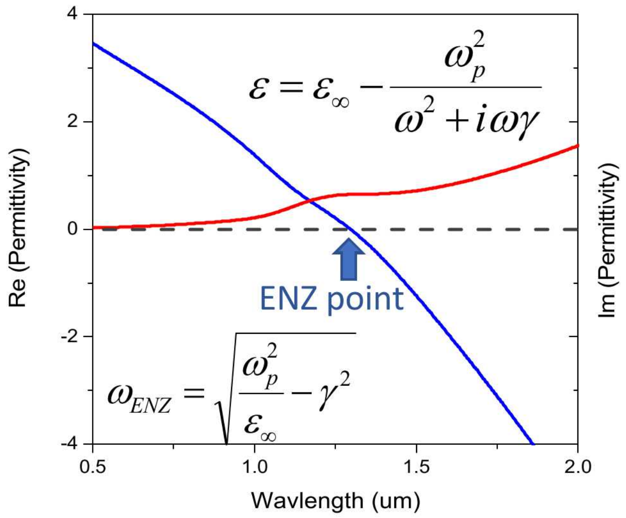
Igal Brener (Sandia) – project lead
Yuanmu Yang (Tsinghua University) – principal investigator
Hanzhe Liu (Tsinghua University) - analysis
Jian Lu (Stanford PULSE Institute) – experiment
Shambhu Ghimire (Stanford PULSE Institute) – experiment
Alejandro Macjavacas (UNM) - theory
Kyle Kelley (Oak Ridge National Lab) - samples
Jon-Paul Maria (Penn State U) – samples
Evan Runnerstrom (Army Research Office, NC) - samples

# What are epsilon-near-zero (ENZ) materials?

**Natural effective medium:** crystals, metals, doped semiconductors, graphene, etc.

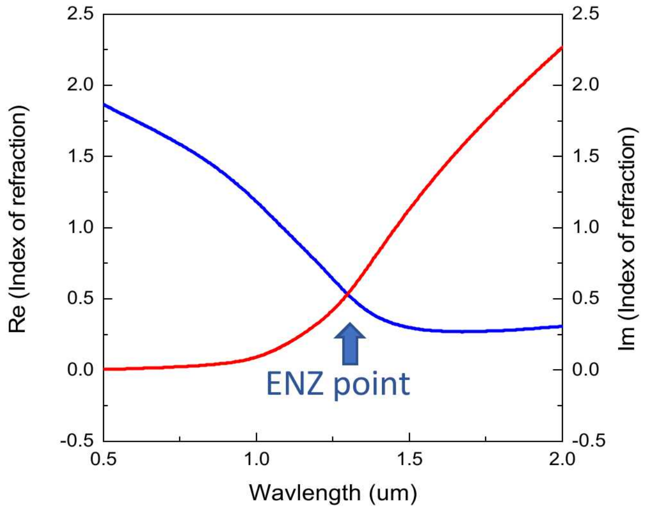
Almost free charges (plasmons): $$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\gamma}$$ $$\omega_{ENZ} = \sqrt{\frac{\omega_p^2}{\varepsilon_\infty} - \gamma^2}$$

Bound charges (phonons): $$\varepsilon = \varepsilon_\infty + \frac{\Omega_p^2}{\omega_{TO}^2 - \omega^2 + i\omega\gamma}$$ $$\omega_{ENZ} = \omega_{LO} = \sqrt{\omega_{TO}^2 + \frac{\Omega_p^2}{\varepsilon_\infty}}$$




$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\gamma}$$

$$\omega_{ENZ} = \sqrt{\frac{\omega_p^2}{\varepsilon_\infty} - \gamma^2}$$



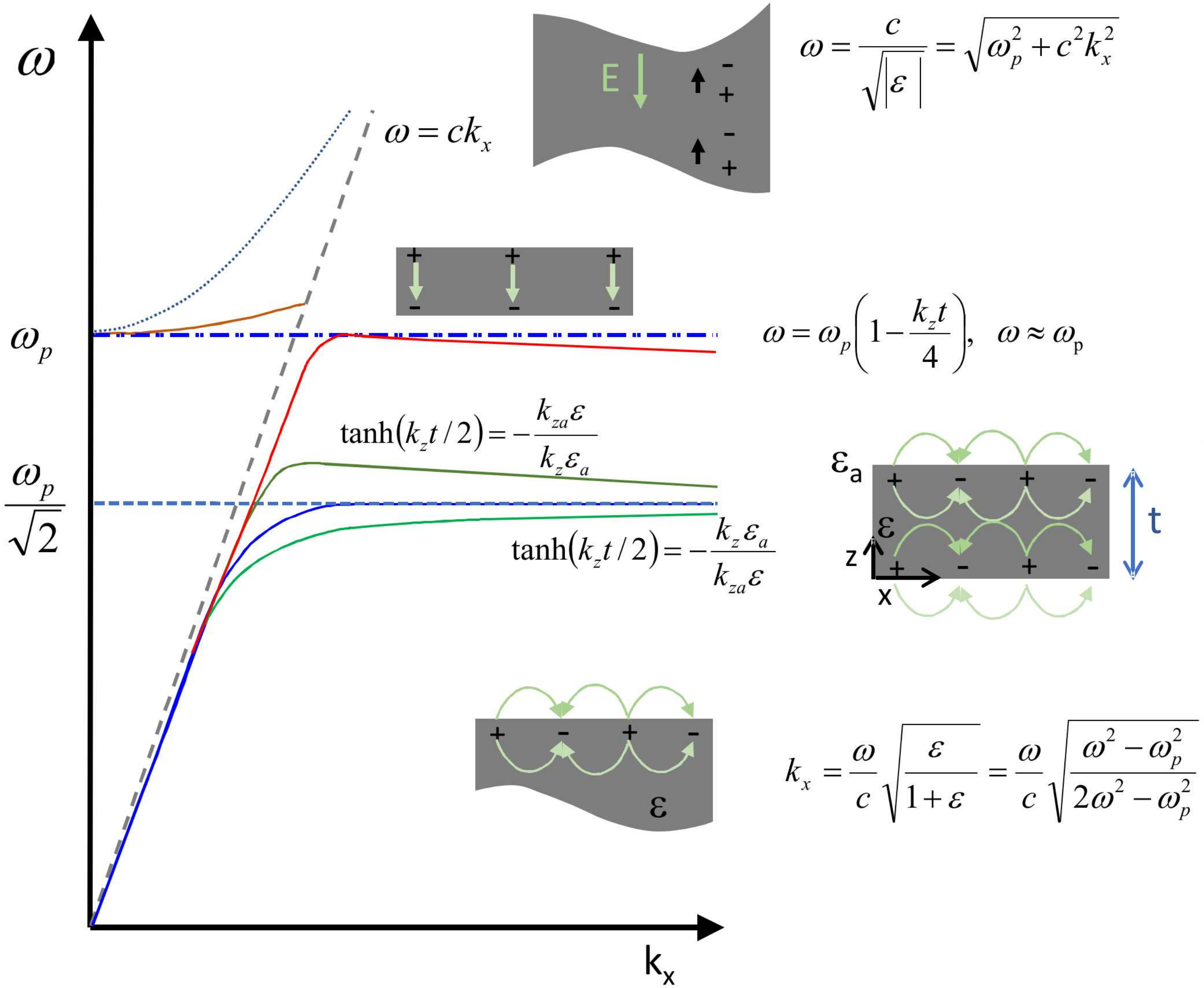

Key properties of ENZ materials are:

- Highly dispersive
- Low index of refraction
- Unavoidable loss

**Artificial effective medium materials (metamaterials):** engineered plasmonic (hyperbolic metamaterials); cutoff waveguides; Coupled dielectric resonances (photonic crystals)

# What are epsilon-near-zero modes?

$$\varepsilon \cong 1-\frac{\omega_p^2}{\omega^2}+i\frac{\gamma\omega_p^2}{\omega^3},\ \omega >> \gamma$$




**Homogeneous metal:** no propagating mode; above the plasma frequency, longitudinal wave with parabolic dispersion.

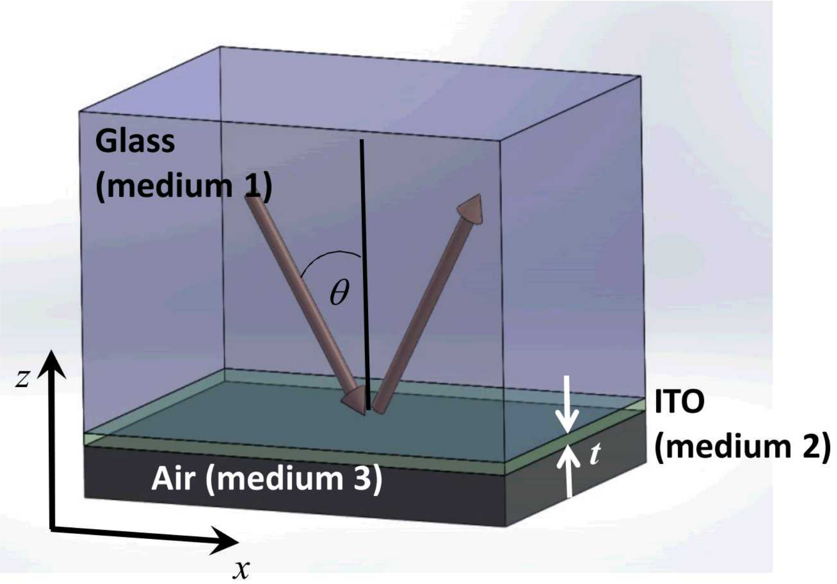
**Two interfaces, subwavelength thickness:** epsilon-near-zero mode and Berreman mode.

**Two interfaces:** short-range and long-range SPP.

**One interface:** Surface propagating wave called surface plasmon polariton (SPP).

Vassant, Marquier, Greffet et al., Opt. Express **20**, 23971 (2012)
Campione, Brener, Marquier, Phys. Rev. B **91**, 121408 (2015)
Campione et al., Opt. Express **24**, 18782 (2016)

# Critical coupling of light to absorbing materials: Perfect absorption

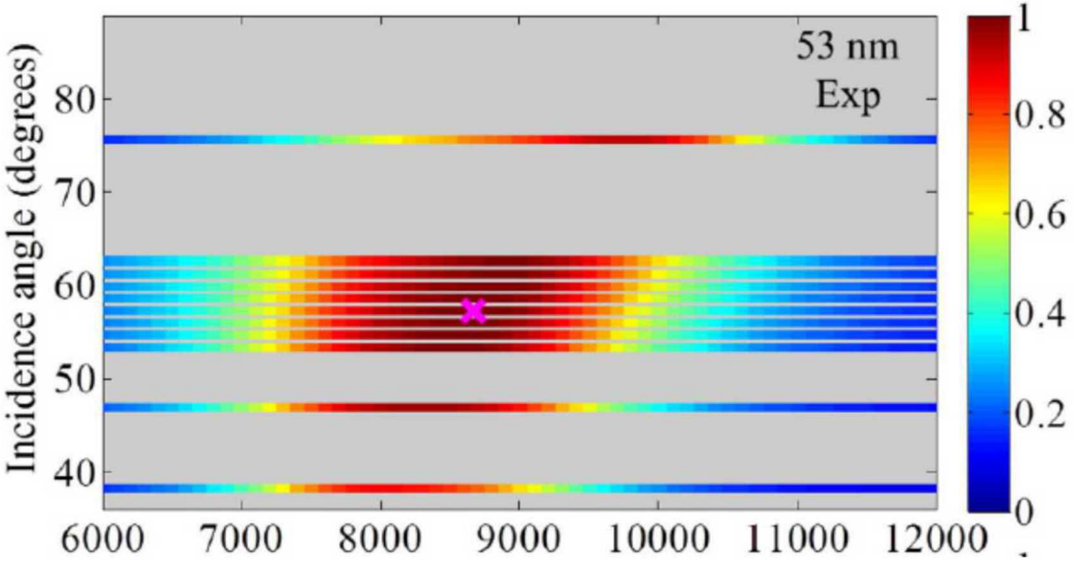


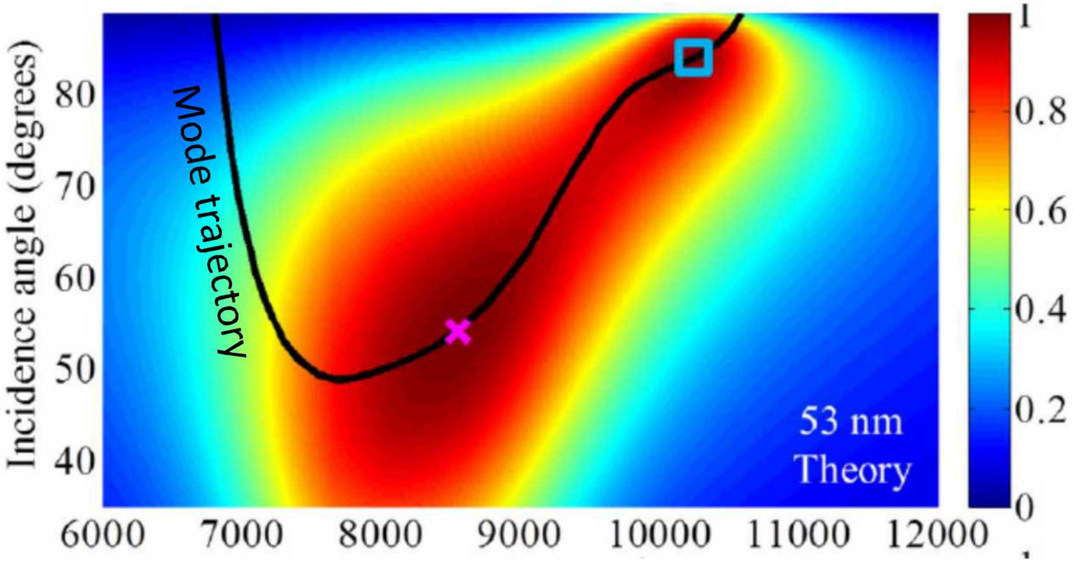

Airy Formula $$\Gamma = \frac{r_{12} + r_{23}e^{2i\phi}}{1 + r_{12}r_{23}e^{2i\phi}} \quad \phi = k_{2\perp}t$$

Critical coupling condition: assume $r_{23} = -1$

$$r_{12} = e^{2i\phi}$$

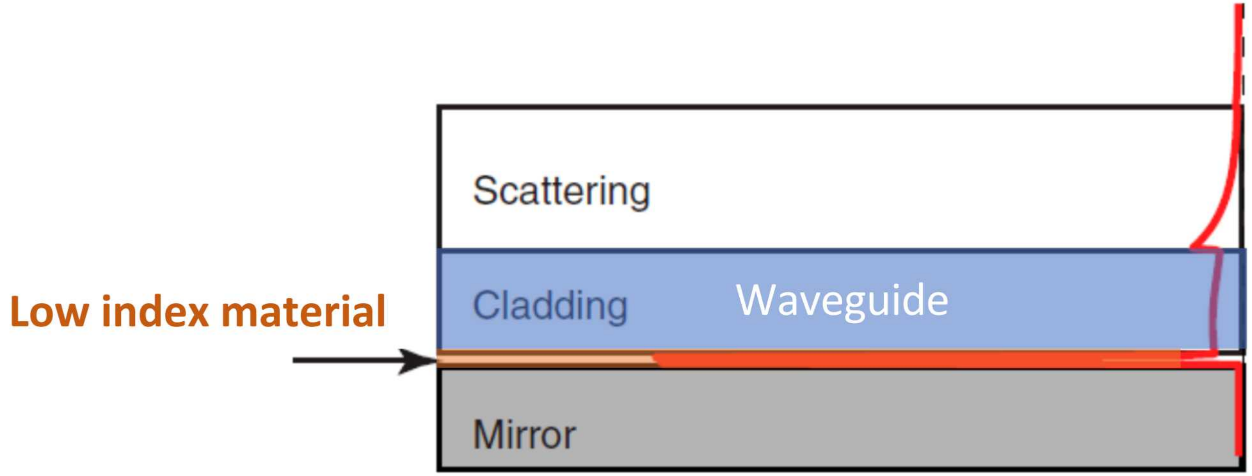
Solution: $$\frac{2\pi d_{PA}}{\lambda_{PA}} = \left[\frac{\varepsilon_2'^2 + \varepsilon_2''^2}{\varepsilon_1^{3/2}\varepsilon_2''}\right]\frac{1}{\tan\theta_{PA}\sin\theta_{PA}}$$




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

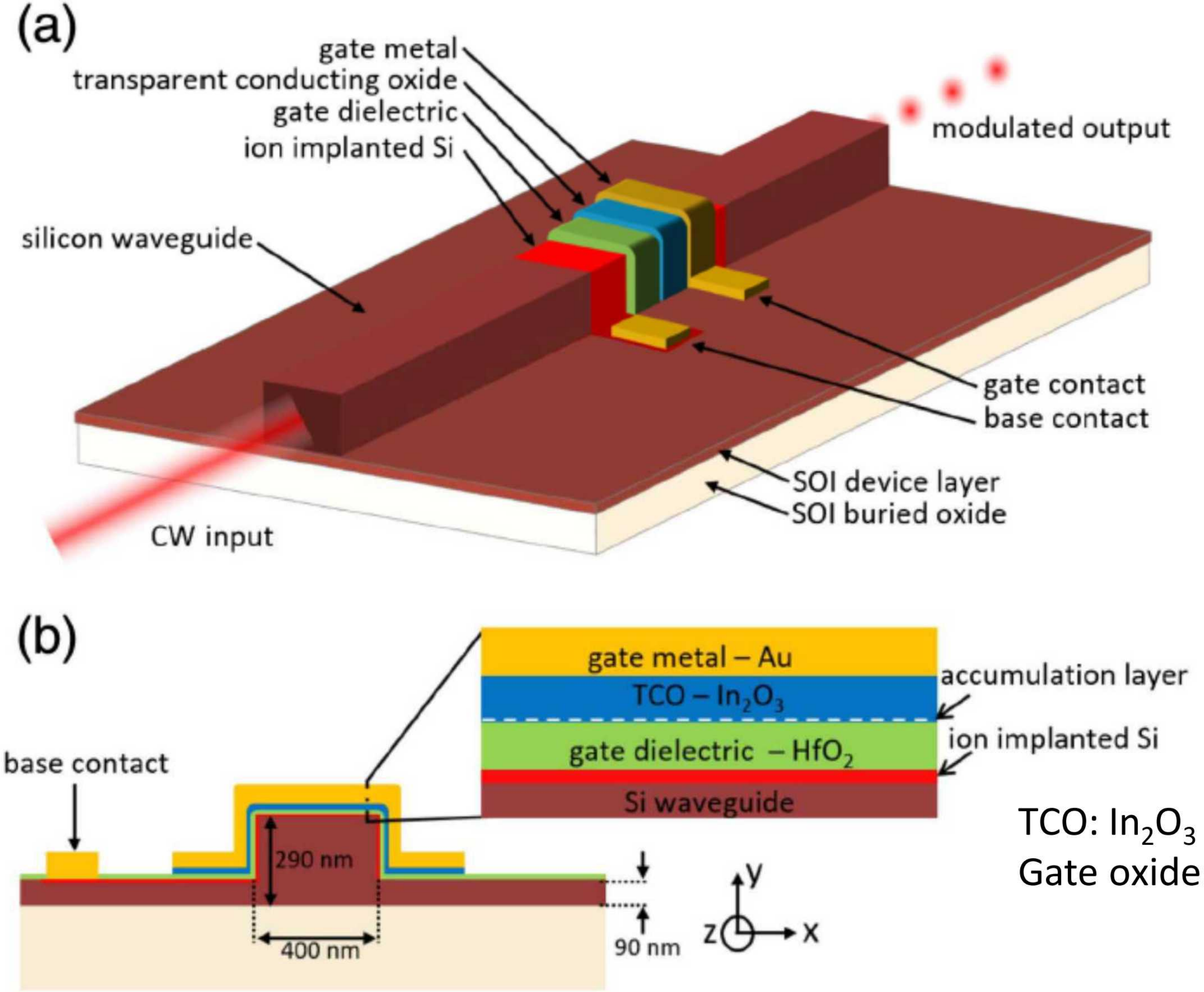
# Tuning the light-ENZ material interaction




- Modes in the waveguide get sucked into the low index material
- Tune the low index layer by carrier injection, making a lossy layer
- Waveguide loss is modulated

Z. Yu, A. Raman, and S. Fan, "Fundamental limit of nanophotonic light trapping in solar cells," PNAS (2010)

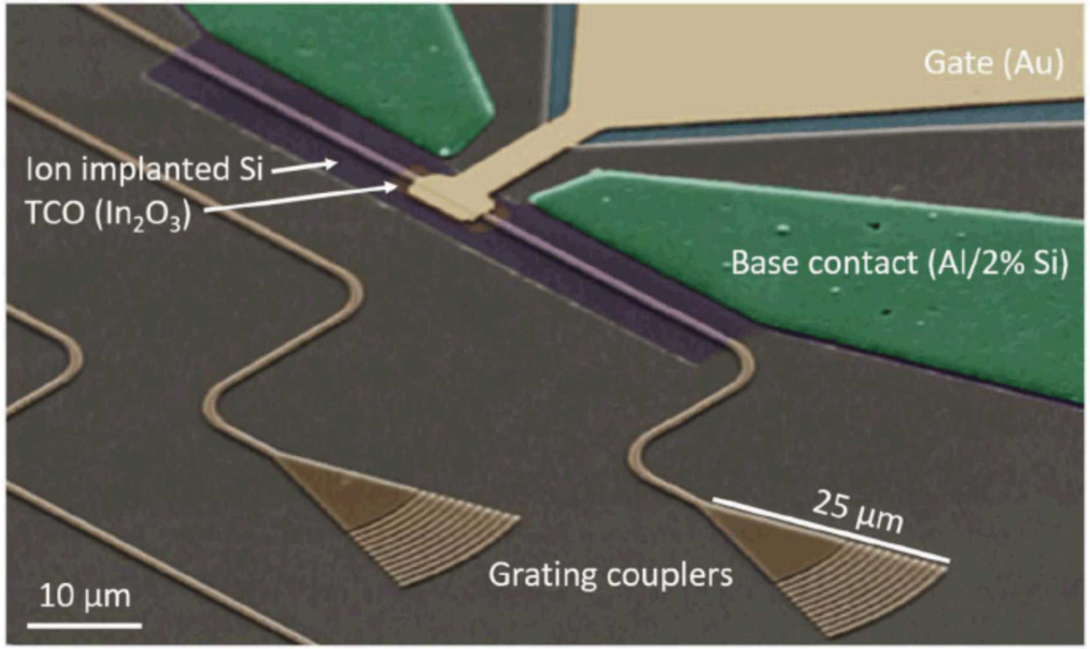
# Application: epsilon-near-zero optical modulators

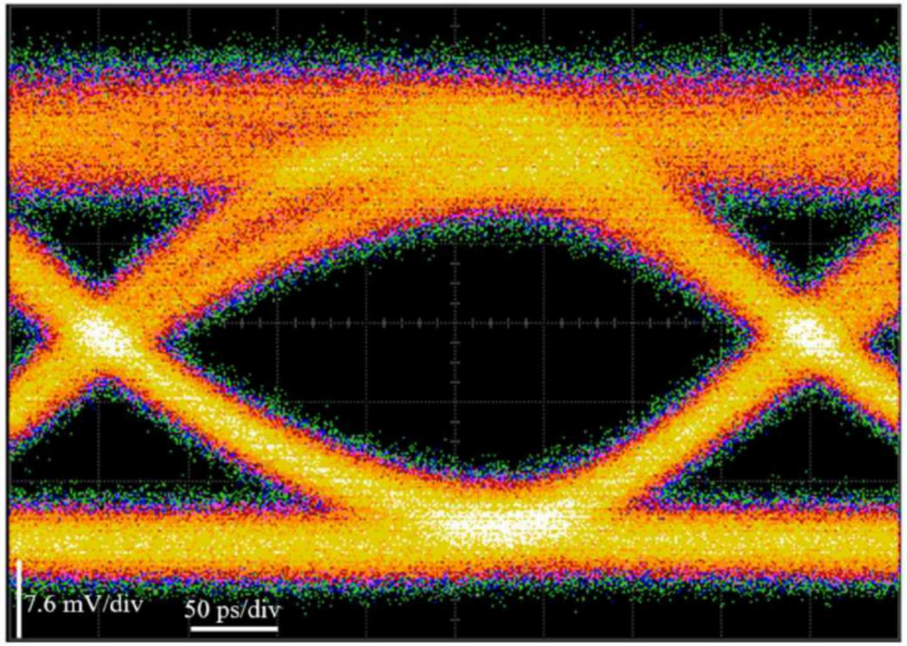



TCO: $In_2O_3$ 10nm
Gate oxide: Hafnia 5nm

M. G. Wood, S. Campione, S. Parameswaran, T. S. Luk, J. R. Wendt, D. K. Serkland, and G. A. Keeler, "Gigahertz speed operation of epsilon-near-zero silicon photonic modulators," Optica **5**, 233-236 (2018).

# Gigahertz epsilon-near-zero optical modulators




CMOS compatible
Free space coupling through grating
Silicon waveguide: 290x400nm
TCO: $In_2O_3$ 10nm
Gate oxide: Hafnia 5nm
Length: 4um

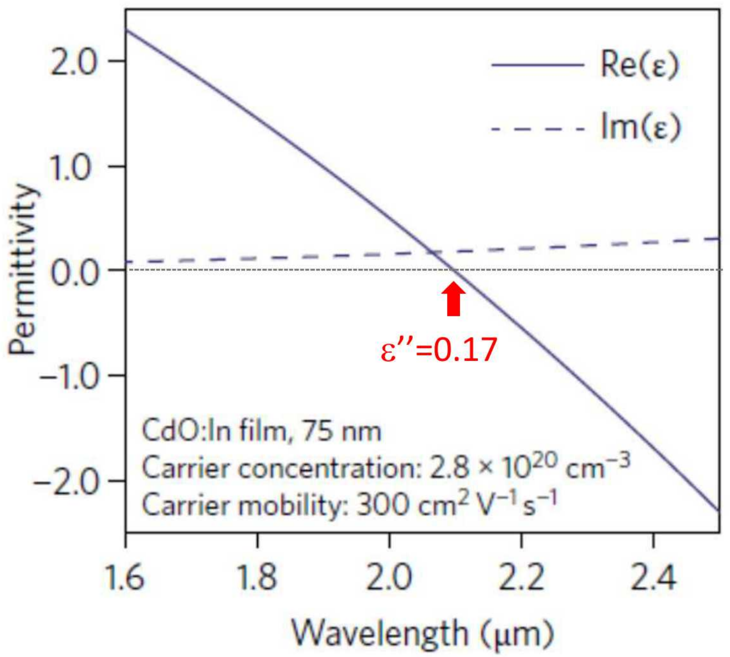
Bandwidth 1530-1590nm
Speed: 2.5 Gb/s, RC limited
Extinction ratio: 6.5 dB
Drive voltage 2 $V_{pp}$


M. G. Wood, S. Campione, S. Parameswaran, T. S. Luk, J. R. Wendt, D. K. Serkland, and G. A. Keeler,
"Gigahertz speed operation of epsilon-near-zero silicon photonic modulators," Optica **5**, 233-236 (2018).

# High quality ENZ material: High mobility CdO

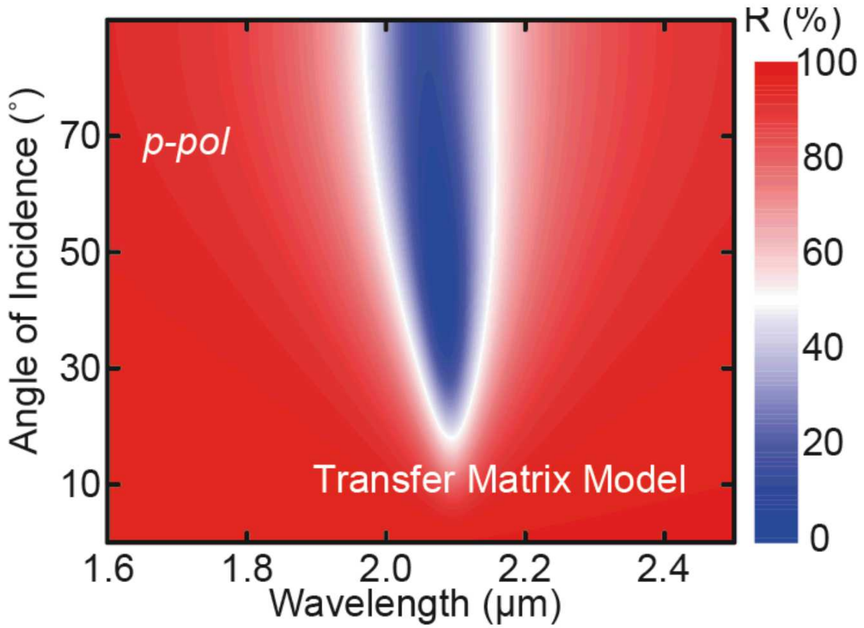
***Low Loss***

Y:CdO has more than 10x higher mobility than ITO

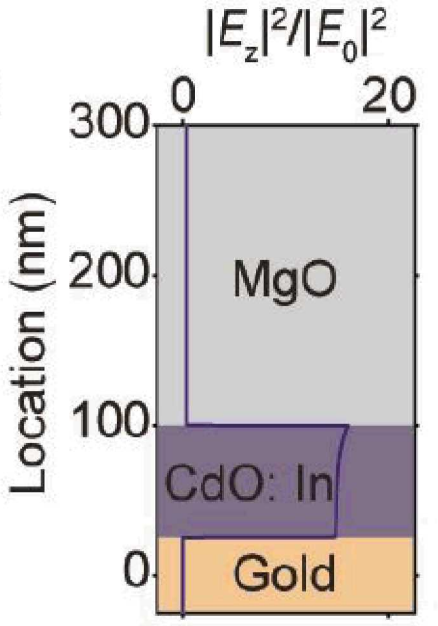



***Perfect absorber with no prism***

Transfer Matrix simulation

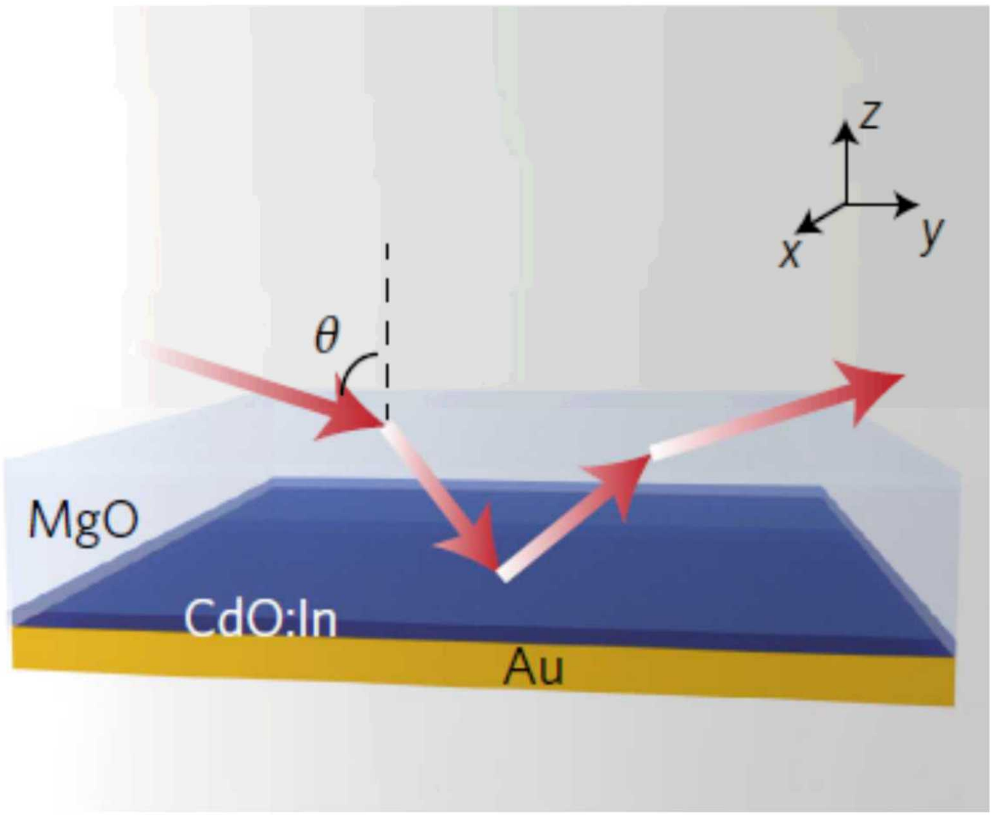


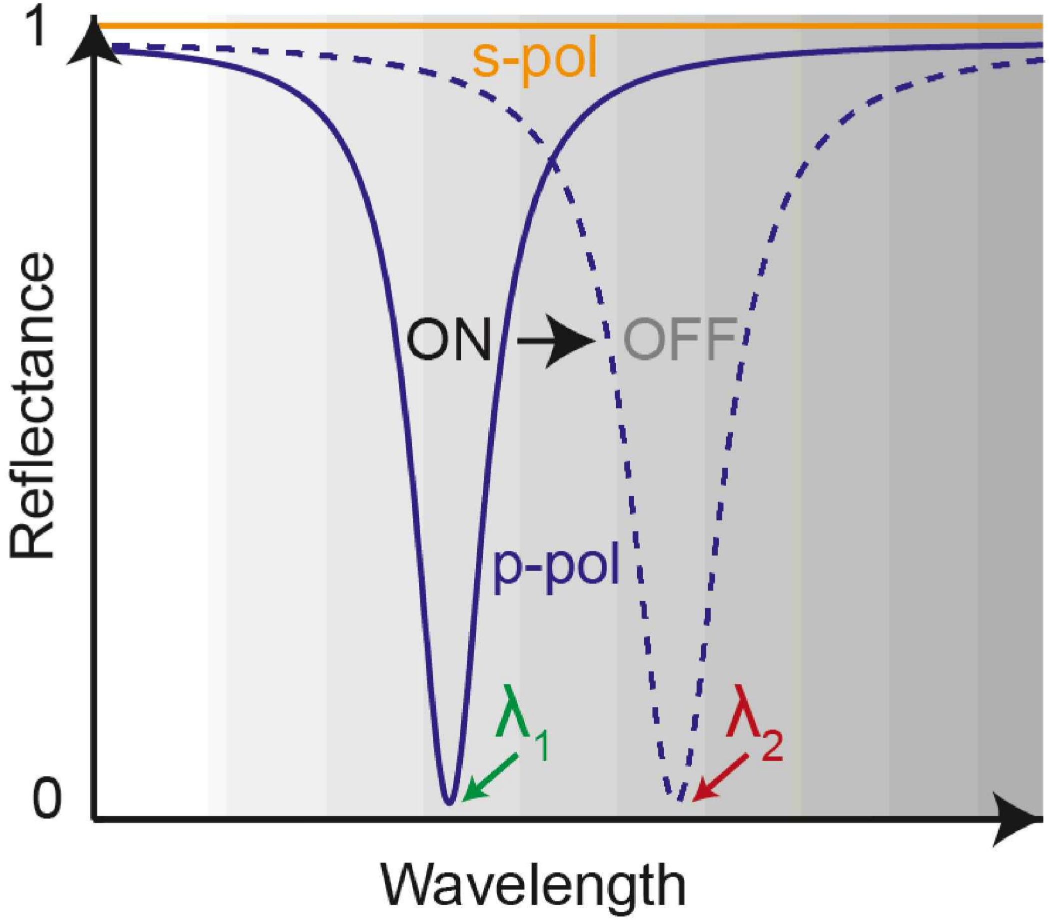

***Large field enhancement***



# Absorption modulation by effective mass tuning

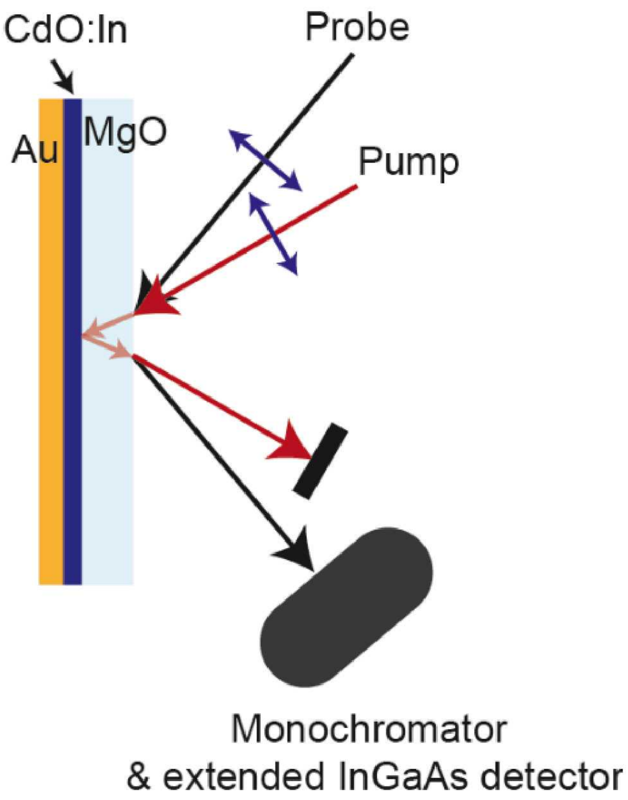
$$\omega_p = \sqrt{ne^2/\varepsilon_0 m^*}$$

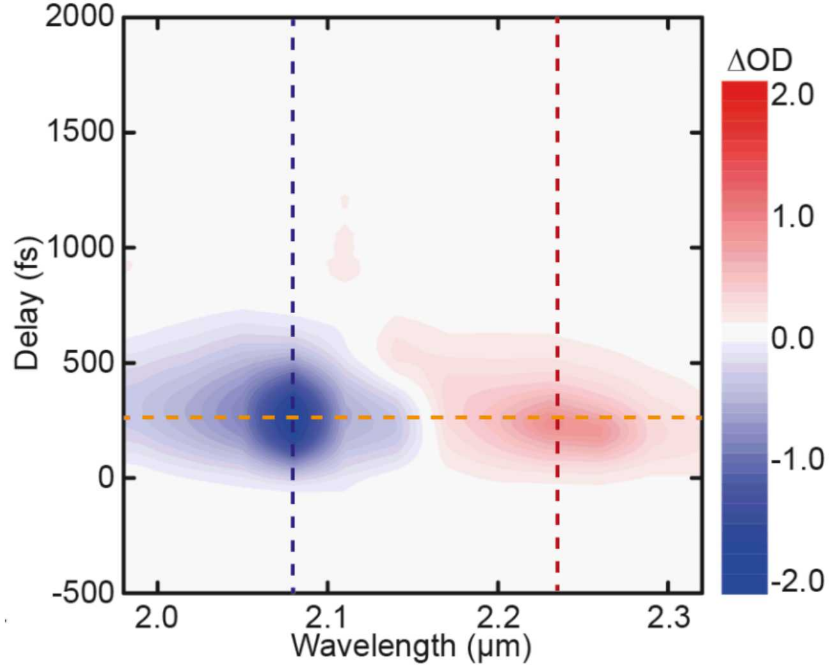


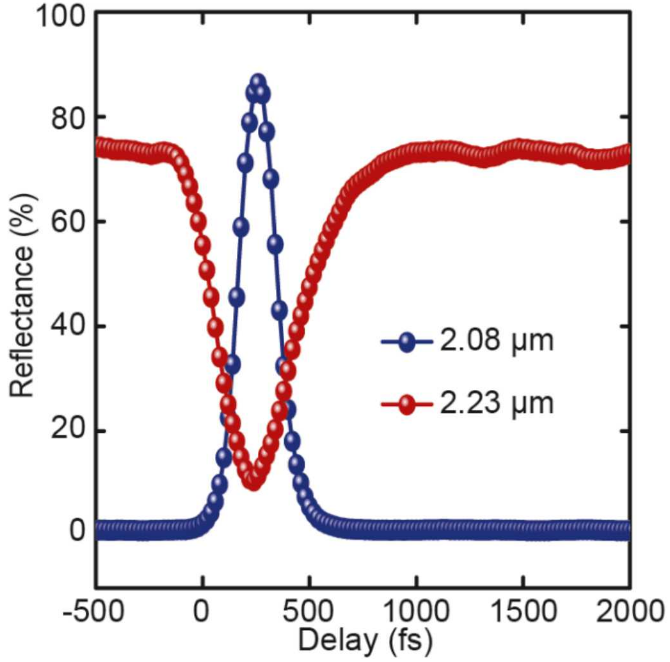




Y. Yang, K. Kelley, E. Sachet, S. Campione, T. S. Luk, J.-P. Maria, M. B. Sinclair, and I. Brener,
"Femtosecond optical polarization switching using a cadmium oxide-based perfect absorber," Nature Photonics **11**, 390 (2017)

# Ultrafast high contrast reflectance modulator



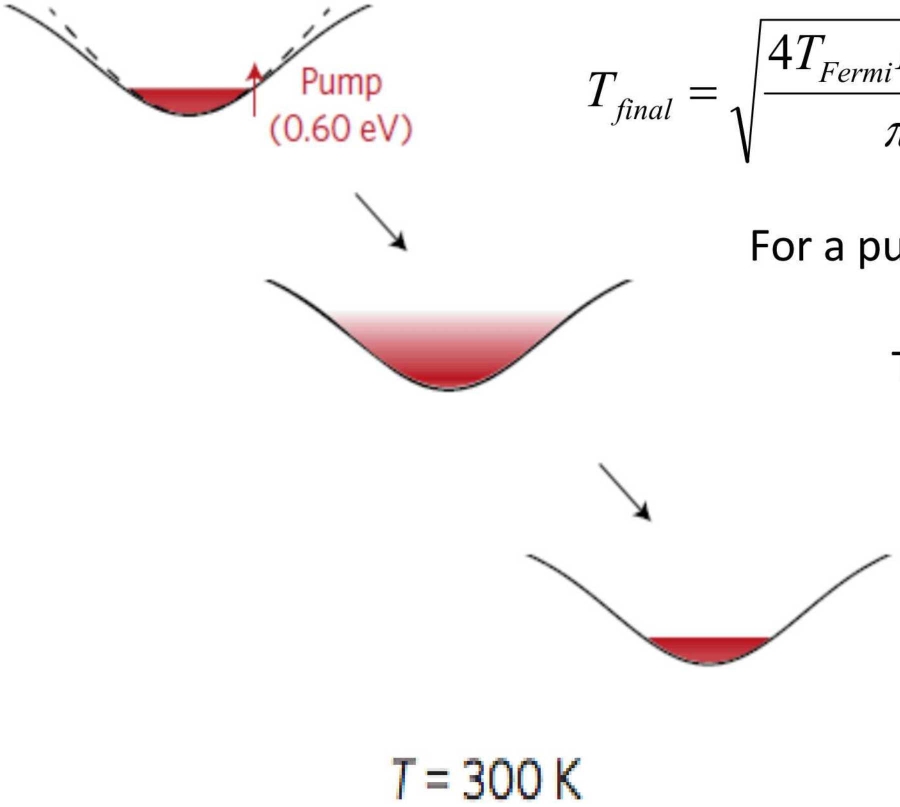

- ***Absolute reflectance* modulation from *1% to 86%!***
- ***Pump energy density: 339 μJ/cm²***

Y. Yang, K. Kelley, E. Sachet, S. Campione, T. S. Luk, J.-P. Maria, M. B. Sinclair, and I. Brener,
"Femtosecond optical polarization switching using a cadmium oxide-based perfect absorber," Nature Photonics **11**, 390 (2017)

# Reflectance modulation by effective mass tuning via heating

**Electron heating mechanism**

Speed limitations: Hot carrier lifetime




Electron temperature estimate:

$$T_{final} = \sqrt{\frac{4T_{Fermi}E_{per\_electron}}{\pi^2 k_B}}$$

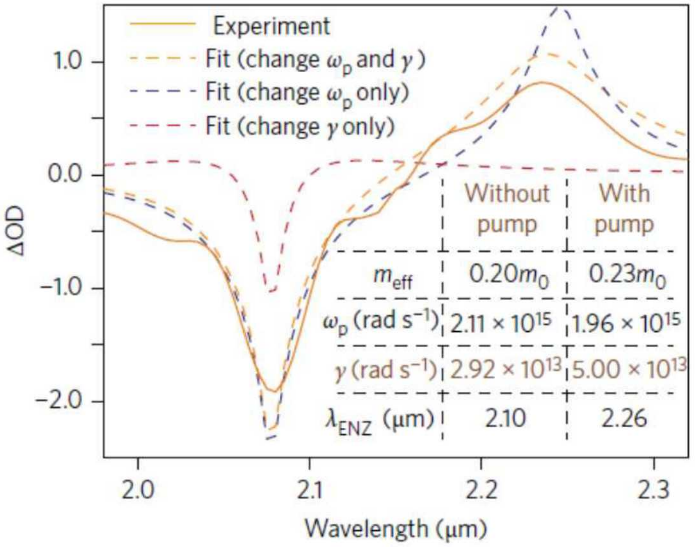
For a pump fluence of 300uJ/cm$^2$

$T_{final}$ (75nm sample)=6.7x10$^3$K

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

Plasma frequency:

$$\omega_p = \sqrt{ne^2/\varepsilon_0 m^*}$$

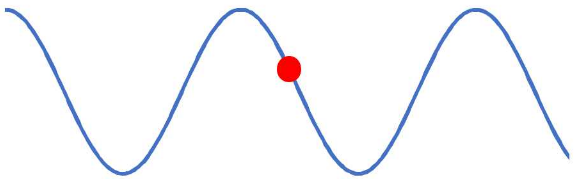



- ***Effective mass change of about 10%, ~7% shift in plasma frequency***

Guo et. al., Nat. Photon, (2016)

# Types of nonlinear electronic response

## Sinusoidal field driven free electron (ignore Lorentz force)



$$\frac{dp}{dt} = -eE_0 \sin\omega t$$

⇩

$$j(t) = -e\frac{\partial \varepsilon}{\partial p} = -\frac{eE_0}{m^*\omega}\cos\omega t$$

Since $m^*(\varepsilon) = \frac{\hbar^2}{\frac{\partial^2 \varepsilon(k)}{\partial k^2}}$

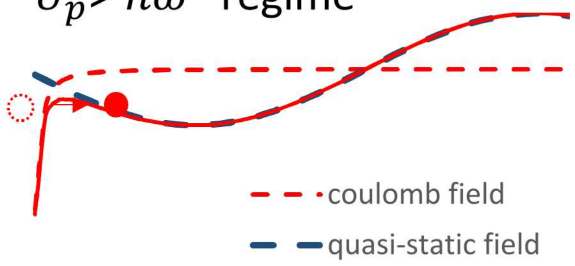
For parabolic band,

$m^*$ is constant since $\frac{\partial^2 \varepsilon(k)}{\partial k^2}$ = constant

**Therefore $j(t)$ has no higher harmonic**

## Quasi-free electron in a sinusoidal field and an ion core field

$U_p > \hbar\omega$ regime




$$\max(n)\hbar\omega \le I_p + 3U_p$$

$I_p$ is the ionization energy $\quad U_p = \frac{e^2E_0^2}{4m\omega^2}$

$$p = -\frac{eE_0}{\omega}(\cos\omega t - \cos\omega t_0)$$

$$<\frac{p^2}{2m}> = U_p(1 + 2\cos^2\omega t_0)$$

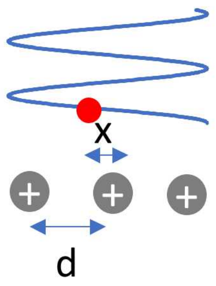
$t_0$ is the time of birth of a quasi-free electron

T. F. Gallagher, "Above-Threshold Ionization in Low-Frequency Limit," PRL(1988).

P. B. Corkum, "Plasma perspective on strong field multiphoton ionization," Physical Review Letters **71**, 1994-1997 (1993)

# Types of nonlinear electronic response

## Sinusoidal driven electron in a lattice fields




$$x_{max} = \frac{eE_0}{m^* \omega^2}$$

For CdO d – 0.47nm

When E is so large that , $x_{max} > d$,
additional modulation comes from lattice field (Bloch oscillation)

Bloch oscillation frequency $\omega_B = \frac{eE_0 d}{\hbar}$

The nth harmonic current

$$j_n(t) = c_n E_0 \frac{d^2 m^*}{\hbar^2} \frac{\omega_p^2}{\omega} \left[ J_{2n-2}\left(\frac{\omega_B}{\omega}\right) + J_{2n}\left(\frac{\omega_B}{\omega}\right) \right]$$

D. Golde, T. Meier, and S. W. Koch, "High harmonics generated in semiconductor nanostructures by the coupled dynamics of optical inter- and intraband excitations," PRB (2008)

S. Ghimire, A. D. DiChiara, E. Sistrunk, P. Agostini, L. F. DiMauro, and D. A. Reis, "Observation of high-order harmonic generation in a bulk crystal," Nat Phys **7**, 138-141 (2011)

## Sinusoidal driven electron in an non-parabolic potential

$$j(t) = -e\frac{\partial \varepsilon}{\partial p} = -\frac{eE_0}{m^* \omega} \cos \omega t$$

For non-parabolic dispersion $\frac{p^2}{2m^*} = \varepsilon + \frac{\varepsilon^2}{\varepsilon_g}$

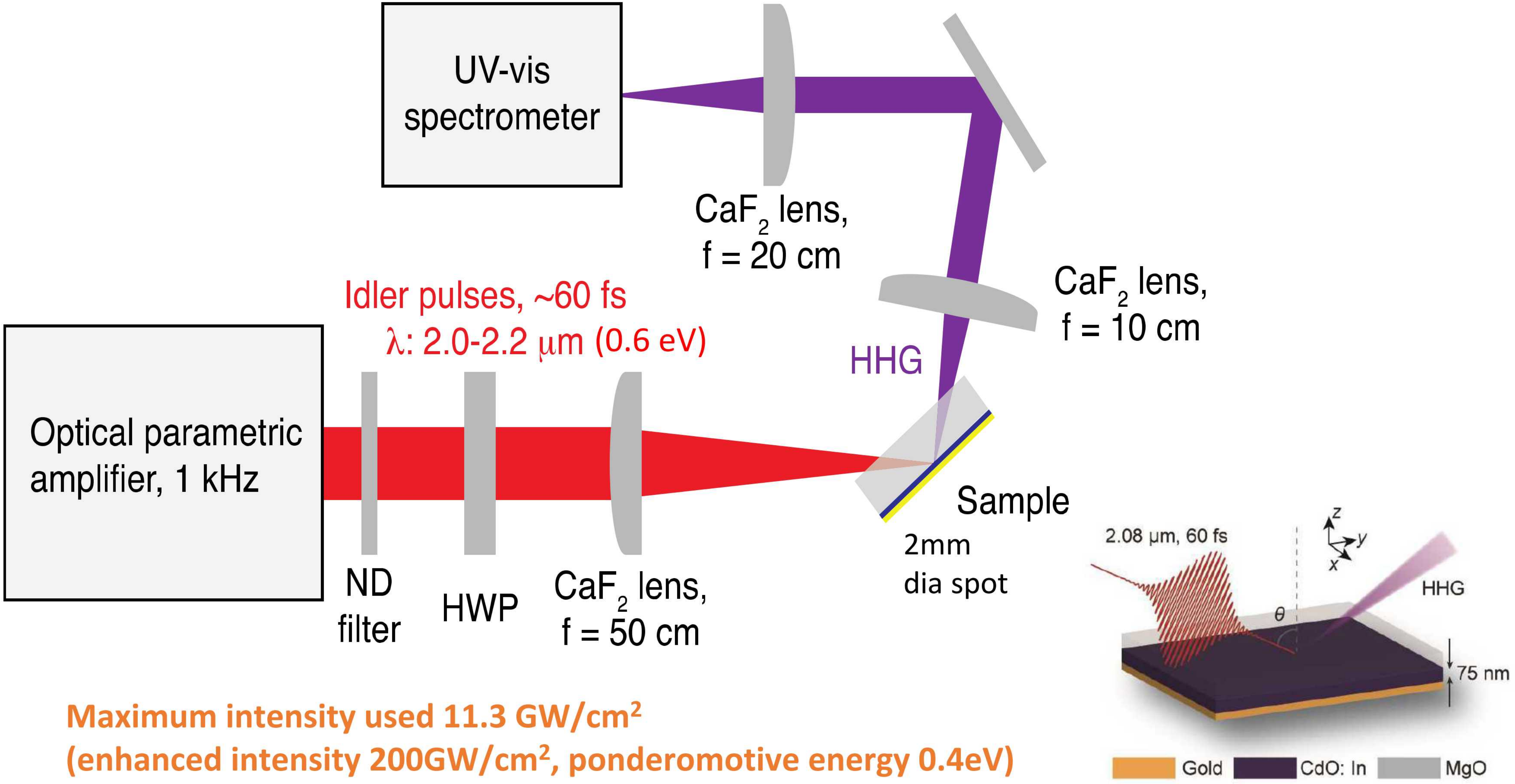
$\varepsilon_g$ is non-parabolicity parameter

$$\Rightarrow \begin{cases} \varepsilon = -\frac{\varepsilon_g}{2} + \sqrt{\frac{\varepsilon_g^2}{4} + \frac{\varepsilon_g p^2}{2m^*}} \\ \frac{\partial \varepsilon}{\partial p} = \frac{1}{\left(1 + \frac{2\varepsilon}{\varepsilon_g}\right)} \frac{p}{m^*} \end{cases}$$

$$j(t) = -\frac{e^2 E_0 \cos \omega t}{m^* \omega \left(1 + \frac{2e^2 E_0^2 cos^2 \omega t}{m^* \omega^2 \varepsilon_g}\right)}$$

P. Guo, et. al., "Ultrafast switching of tunable infrared plasmons in indium tin oxide nanorod arrays with large absolute amplitude," Nat Photon (2016)

# Experimental setup of high harmonic generation from CdO



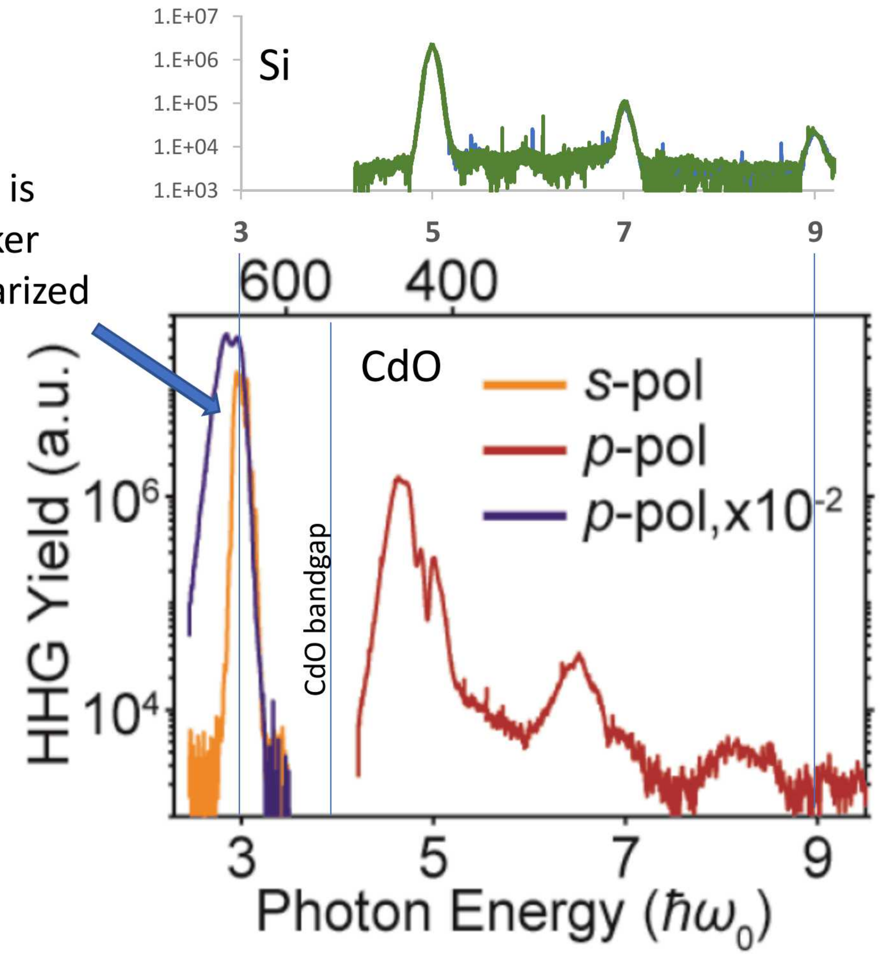

**Maximum intensity used 11.3 GW/cm$^2$**
**(enhanced intensity 200GW/cm$^2$, ponderomotive energy 0.4eV)**

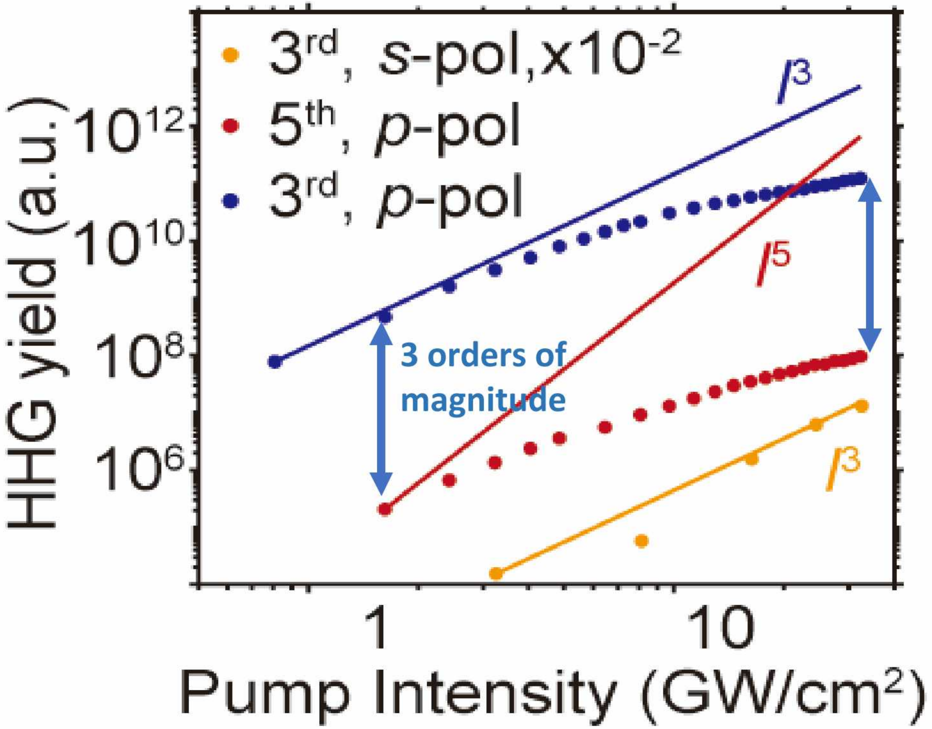
# Highly nonlinear response

## *HHG from 3rd to 9th order*

- **Incident intensity 11.3GW/cm$^2$**
- **Internal intensity ~200GW/cm$^2$**
  - **(12MV/cm internal field)**

S-polarized is much weaker than p-polarized

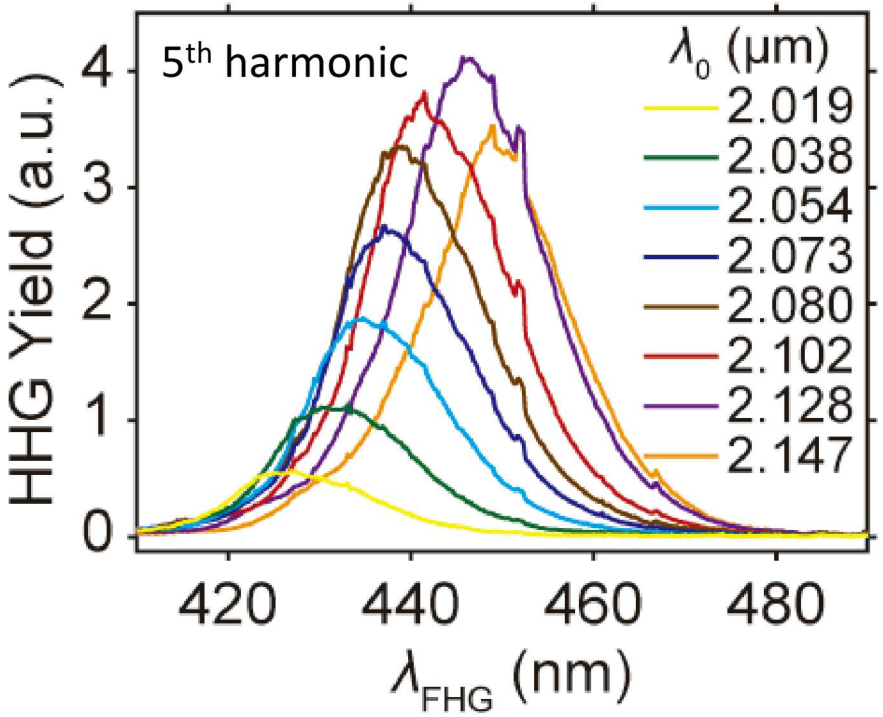


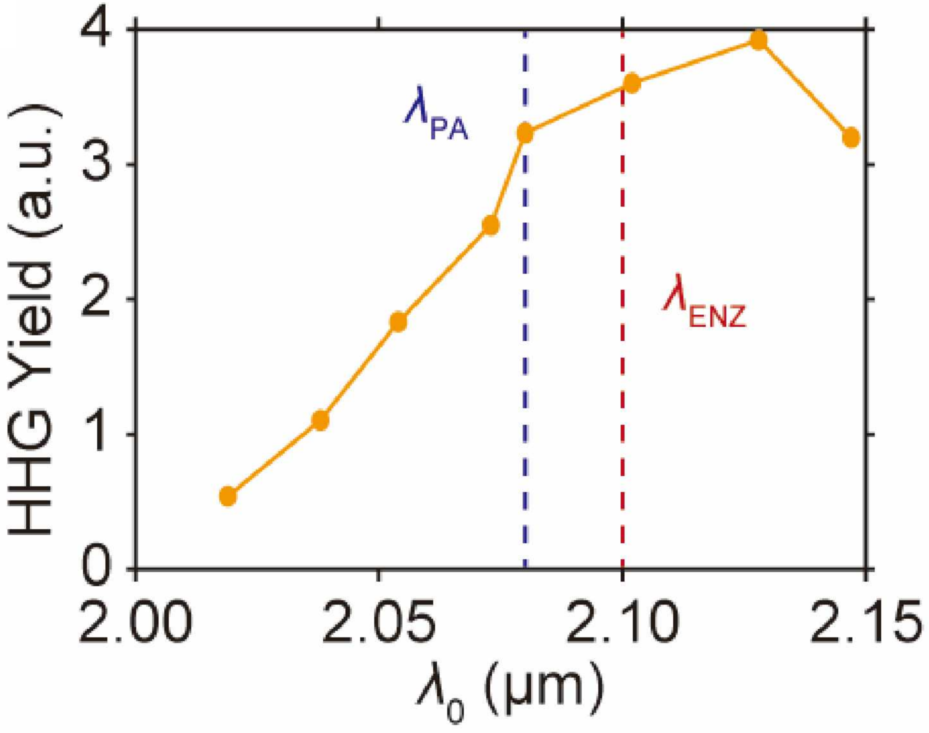




Y. Yang, et. al., to be published in Nat. Phys. (2019)

# Nonlinear response dominated by electrons in conduction band

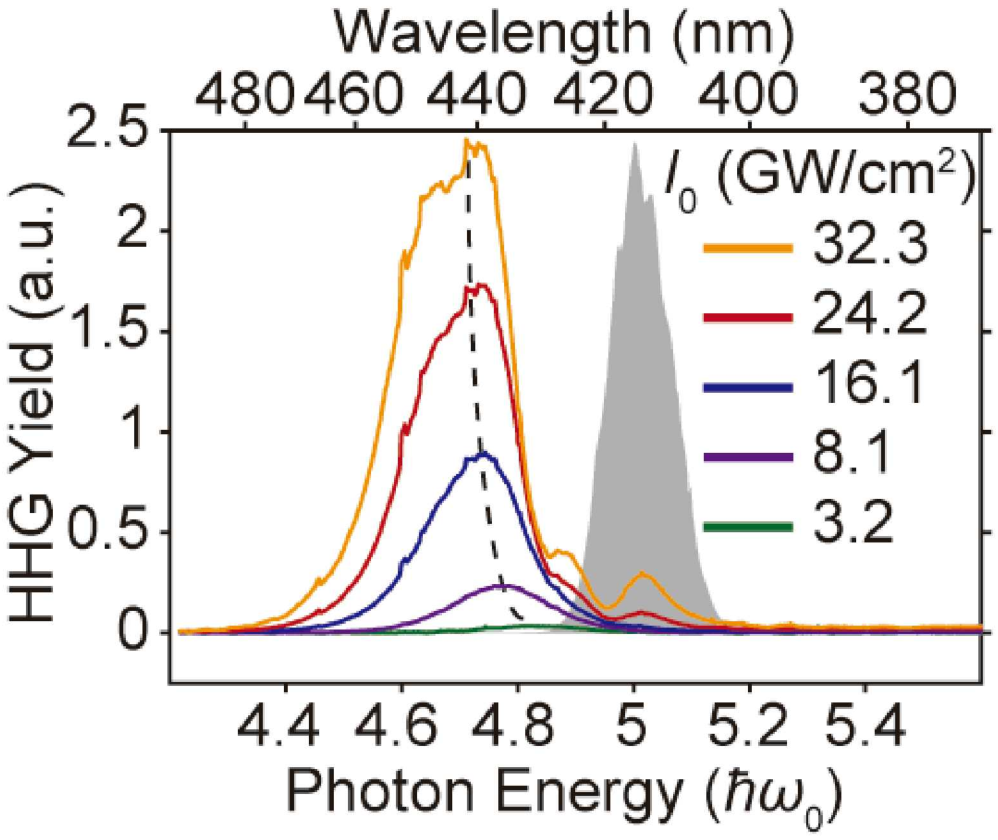
Laser intensity ~5.6 GW/cm$^2$
(8MV/cm internal enhanced field)



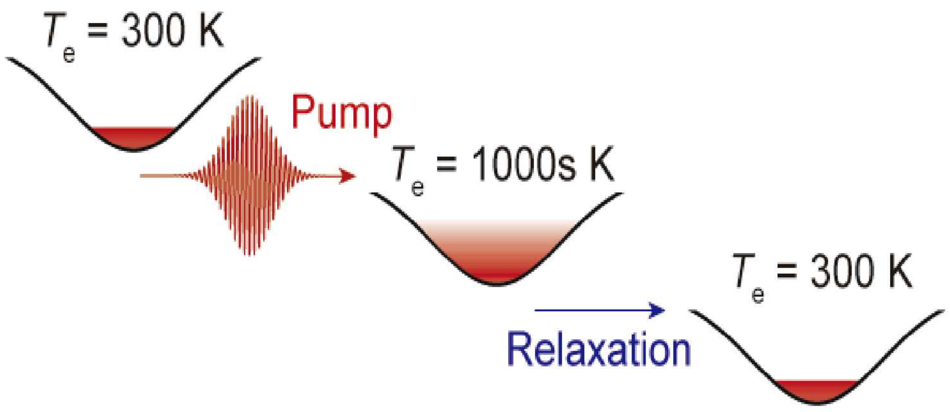




**Harmonic yield decreases when detuned from the ENZ frequency**

# Shift and broadening of the harmonic fields



# Plasma frequency shift from heating




Optical power absorbed

$$P_{\rm abs} = \frac{\omega}{8\pi} {\rm Im}\{\varepsilon_{\rm CdO}\} \left|E_{\rm CdO}\right|^2$$

$$U_{\rm abs} = P_{\rm abs}\Delta t = \frac{\omega}{c} {\rm Im}\{\varepsilon_{\rm CdO}\} \left|g(\omega,\theta)\right|^2 \cos\theta,$$

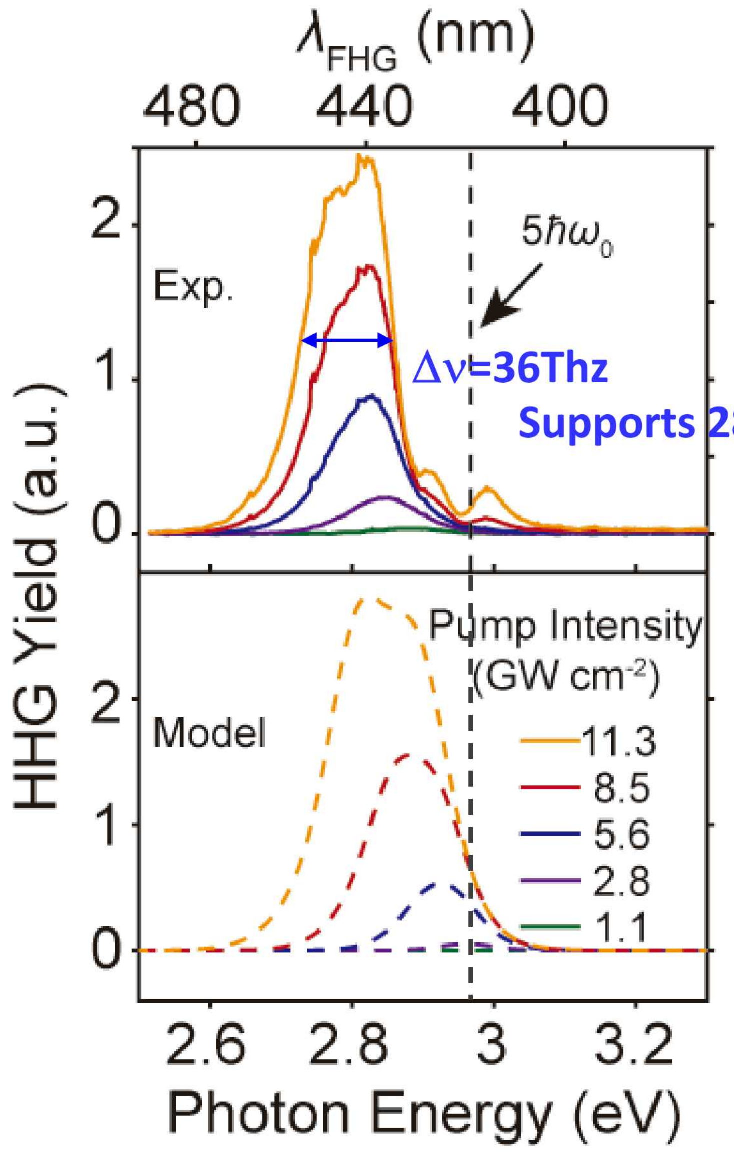
Solve for the electron temperature

$$U(T) = \frac{\sqrt{2}}{\pi^{3/2}} \frac{m^{3/2}}{\hbar^3} \int_0^\infty dE E \left(1 + \frac{2E}{E_g}\right) \sqrt{E + \frac{E^2}{E_g}} f_0(T,\mu),$$

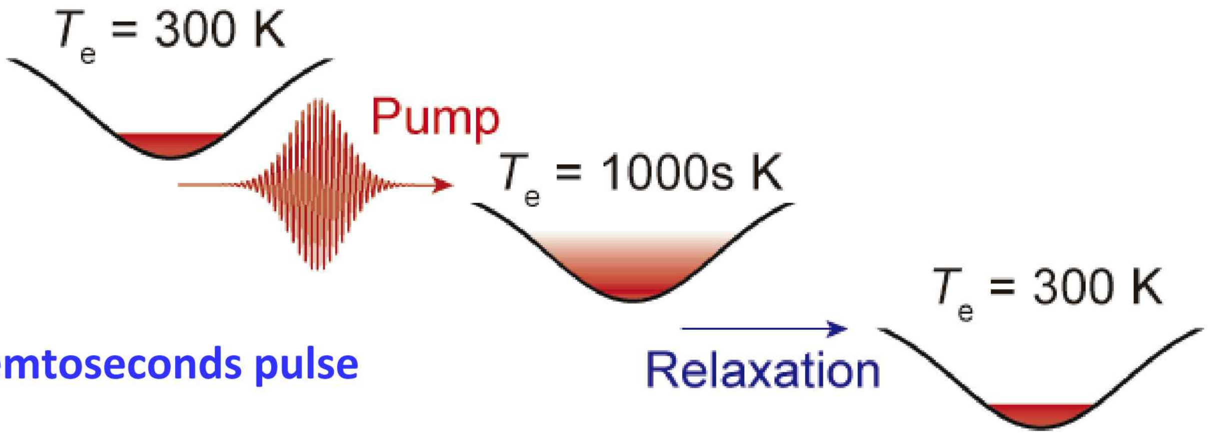
Plasma frequency at temperature T

$$\omega_p(\mu,T)^2 = \frac{e^2}{3\pi^2}\int_0^\infty dE \frac{m}{\hbar^2}\left(1+2E/E_g\right)\left(\frac{2m}{\hbar^2}\left(E+E^2/E_g\right)\right)^{\frac{1}{2}} \frac{\hbar^2 \frac{2m}{\hbar^2}\left(E+E^2/E_g\right)}{m^2\left(1+2E/E_g\right)^2}\left(-\frac{\partial f_0(\mu,T)}{\partial E}\right)$$

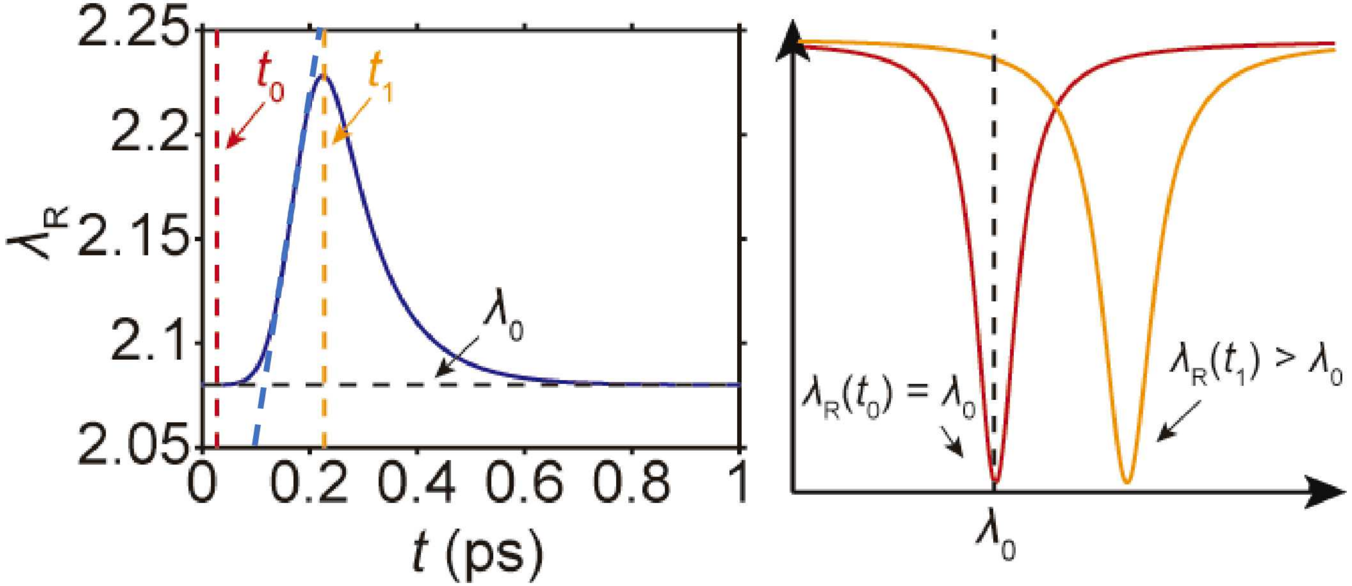
# Adiabatic time dependence model




Supports 28 femtoseconds pulse




Linear chirp region 0.6Tera-rad/s




Y. Yang, et. al., to be published in Nat. Phys. (2019)

# Summary

Quasi-free electrons confined in nano-structures
enables strong light-matter interactions

- We exploited these properties to show:
  enhanced absorber
  amplitude modulator
  Transient reflectance and polarization modulator

- High harmonic generation from high mobility degenerate electron plasma

- New nonlinear mechanisms (non-parabolicity and heating) to explain the nonlinear harmonic behavior

# More acknowledgements


| | |
| --- | --- |
| Salvatore Campione *(SNL)*<br>Michael B. Sinclair *(SNL)* | ITO perfect absorption |
| Michael Wood *(SNL)*<br>Gordon Keeler *(ex-SNL)* | Si waveguide modulator |
| Igal Brener *(CINT, SNL)*<br>Yuanmu Yang *(ex-SNL)* | Polarization tuning using CdO |
| Iltai Kim *(ex-SNL)*<br>Y. C. Jun *(ex-SNL)*<br>Shanhui Fan *(Stanford)*<br>S. Feng *(ex-Navy Research Lab)* | ITO perfect absorption |

**Thank you for your attention**