SAND2019-2937C

Large index modulation effects using epsilon-near-zero materials

Ting S. (Willie) Luk
Sandia National Laboratories
&
CINT

Presented to Joint CINT, OSE and CHTM Seminar
Mar 21, 2019

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

CINT (Center for Integrated Nanotechnologies) is a DOE Office of Science National User Facility

The DOE/SC nanoscience centers:

- help you understand, create and characterize nanomaterials
- defined by a scientific field, not specific instrumentation.
- NSRC staff support user projects and conduct original research.
- Capabilities involve expertise, hardware and software.
- Users access Synthesis, Fabrication, Characterization and Theory capabilities.

"A DOE/SC user facility has unique world-class research capabilities and technologies which are available broadly to science community worldwide from universities, industry, private laboratories, and other Federal laboratories for work that will be published in the open literature."

CINT is a LANL/SNL partnership to create a National resource for nanomaterials integration

Core Facility

Gateway Facility

Visit http://cint.lanl.gov/ for details

Large index modulation effects using epsilon-near-zero materials

Outline

What is epsilon-near-zero material?
As a Perfect absorption (in ITO)
As an Absorption modulator (GHz Si waveguide modulator)
As a Transient tunable absorber and polarization switching
As a nonlinear medium for third harmonic and higher radiation source

Maxwell’s equations

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

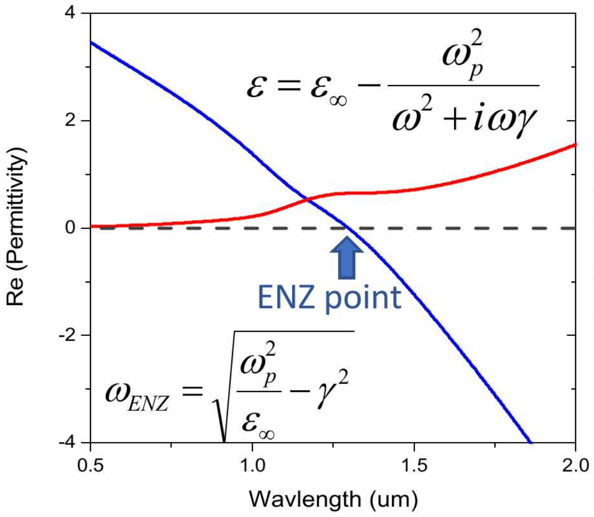
Natural materials: crystals, metals, doped semiconductors, graphene, etc.

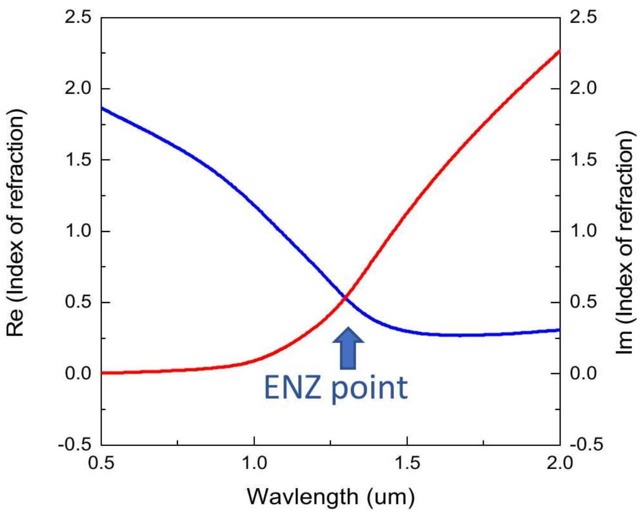
Almost free charges (plasmons): $\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega\gamma}$ $\qquad \omega_{ENZ} = \sqrt{\frac{\omega_p^2}{\varepsilon_\infty} - \gamma^2}$

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

Artificial materials (metamaterials):

Effective zero permittivity - engineered plasmonic and dielectric resonances



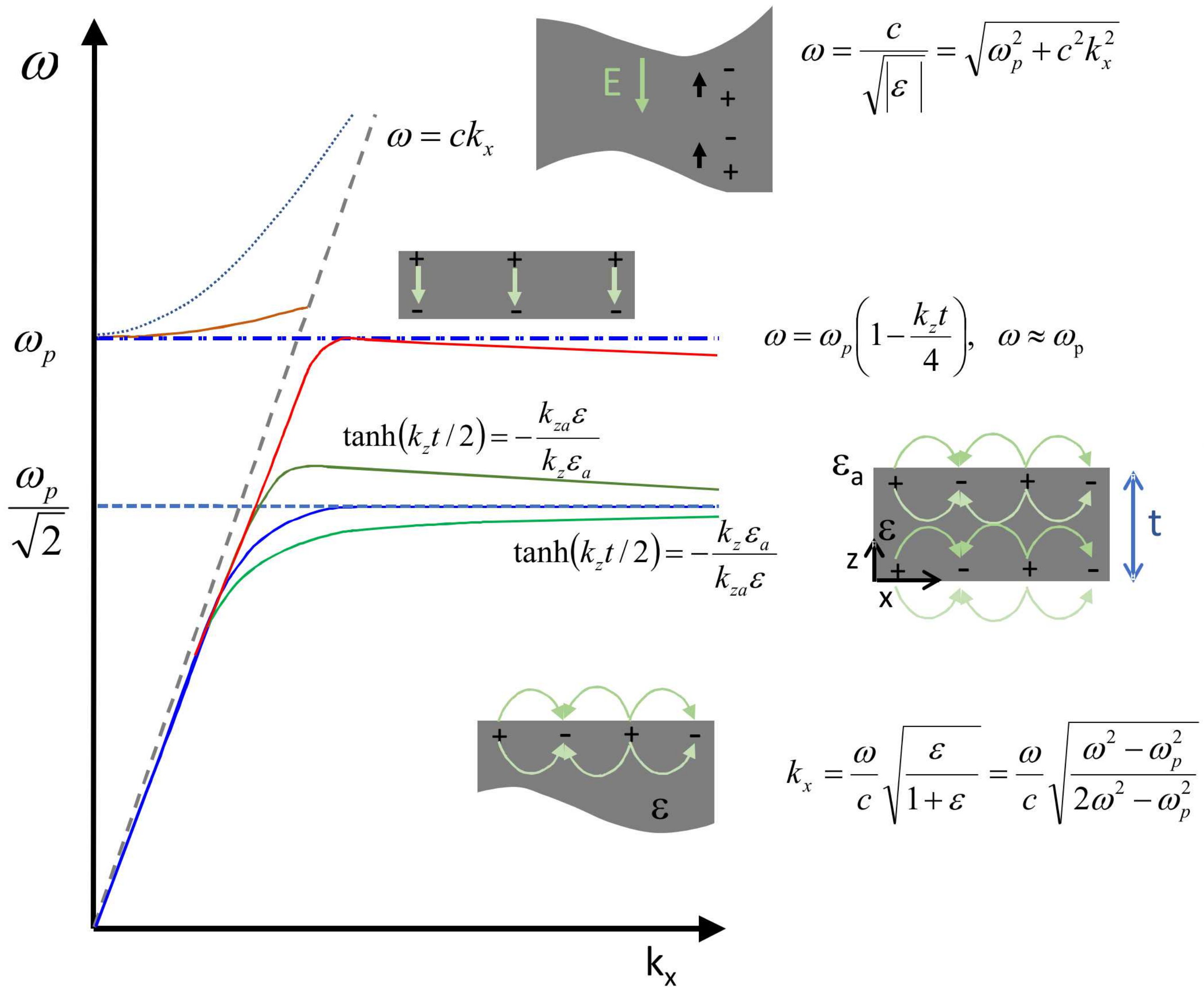


Key properties of ENZ materials are:

- Decoupling of space and time
- Highly dispersive
- Low index of refraction
- Unavoidable loss

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)

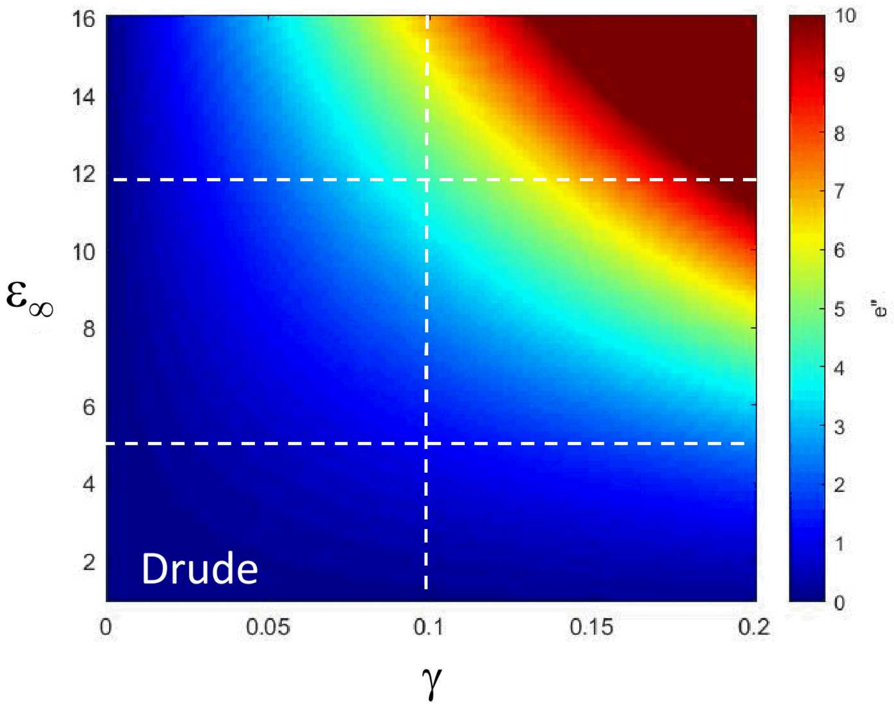
ENZ materials always have loss

Drude model

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

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

ε″ vs gamma @ ENZ frequency

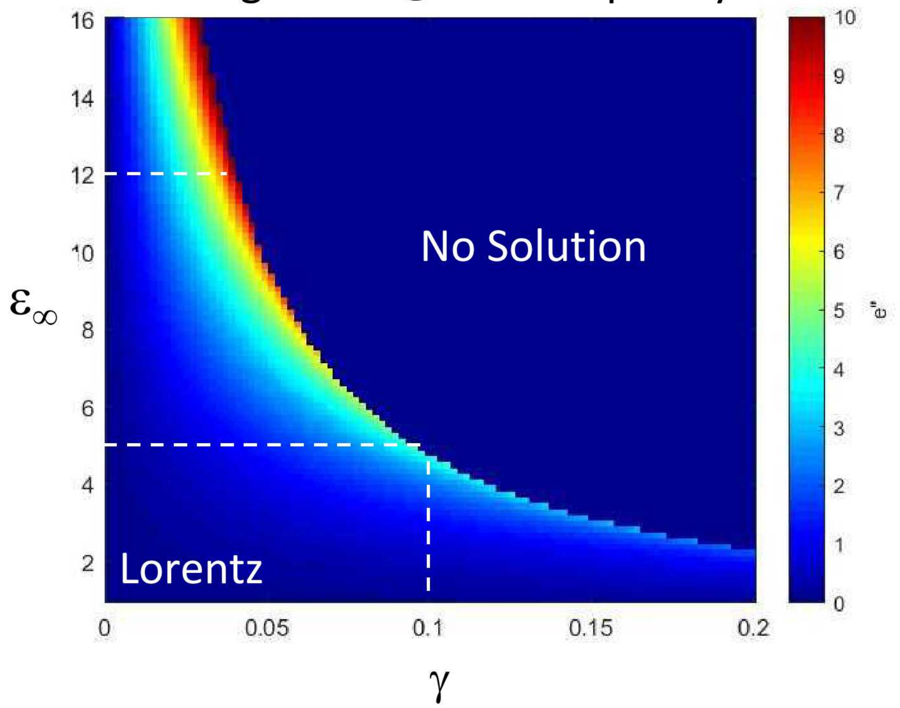


Lorentz model

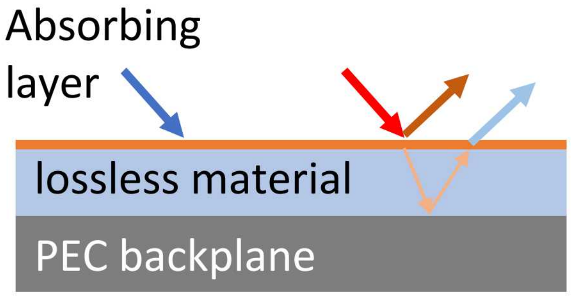
$$\varepsilon = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 - \omega_0^2 + i\omega\gamma}$$

$$\omega_{ENZ} = \frac{1}{\sqrt{2}}\sqrt{\left(\frac{\omega_p^2}{\varepsilon_\infty} + 2\omega_0^2 - \gamma^2\right) \pm \sqrt{\frac{\omega_p^4}{\varepsilon_\infty^2} - 2\frac{\gamma^2\omega_p^2}{\varepsilon_\infty} + \gamma^4 - 4\gamma^2\omega_0^2}}$$

ε″ vs gamma @ ENZ frequency



ENZ material as an ultrathin perfect absorber



- Requires zero transmission
- Requires zero reflection
- Requires a lossy material

Earlier attempts:

Salisbury screen - anti-reflection of planar structures

- $\lambda/4$ lossless spacer material
- Thin top layer of absorbing material

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

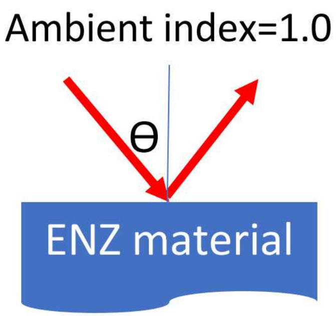
Mid-IR Z. Wang et. al. APL (2015)
Vis J. Guo et. al. Optical Materials Express (2016)

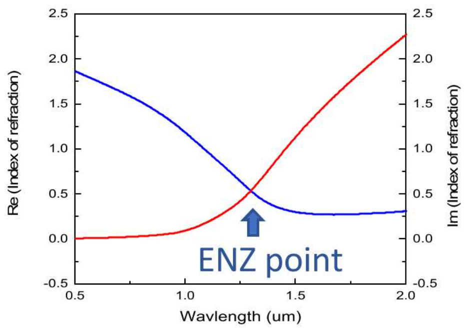
Drawback of this approach

- Requires ¼ wave thickness, a significant thickness for microwaves or radio waves.

ENZ materials can break the quarter wave requirement

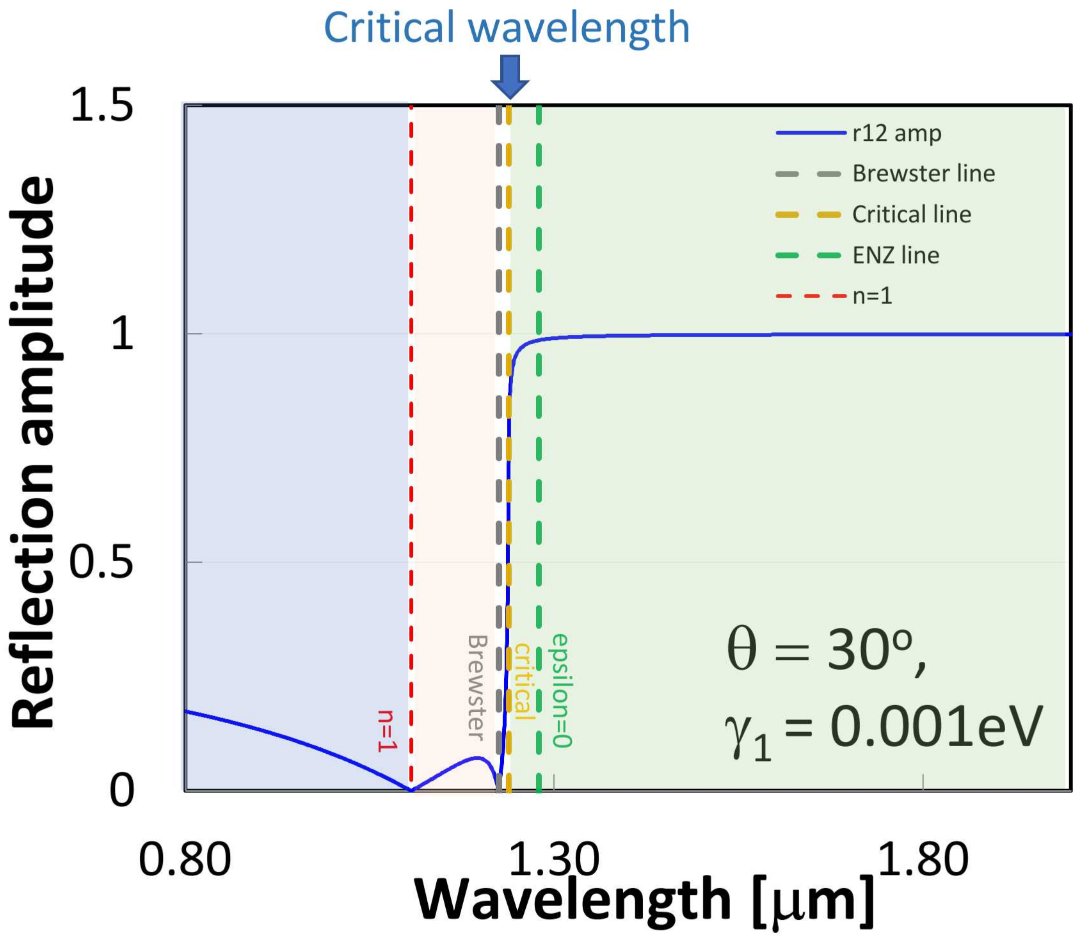
Exploiting the loss of ENZ materials

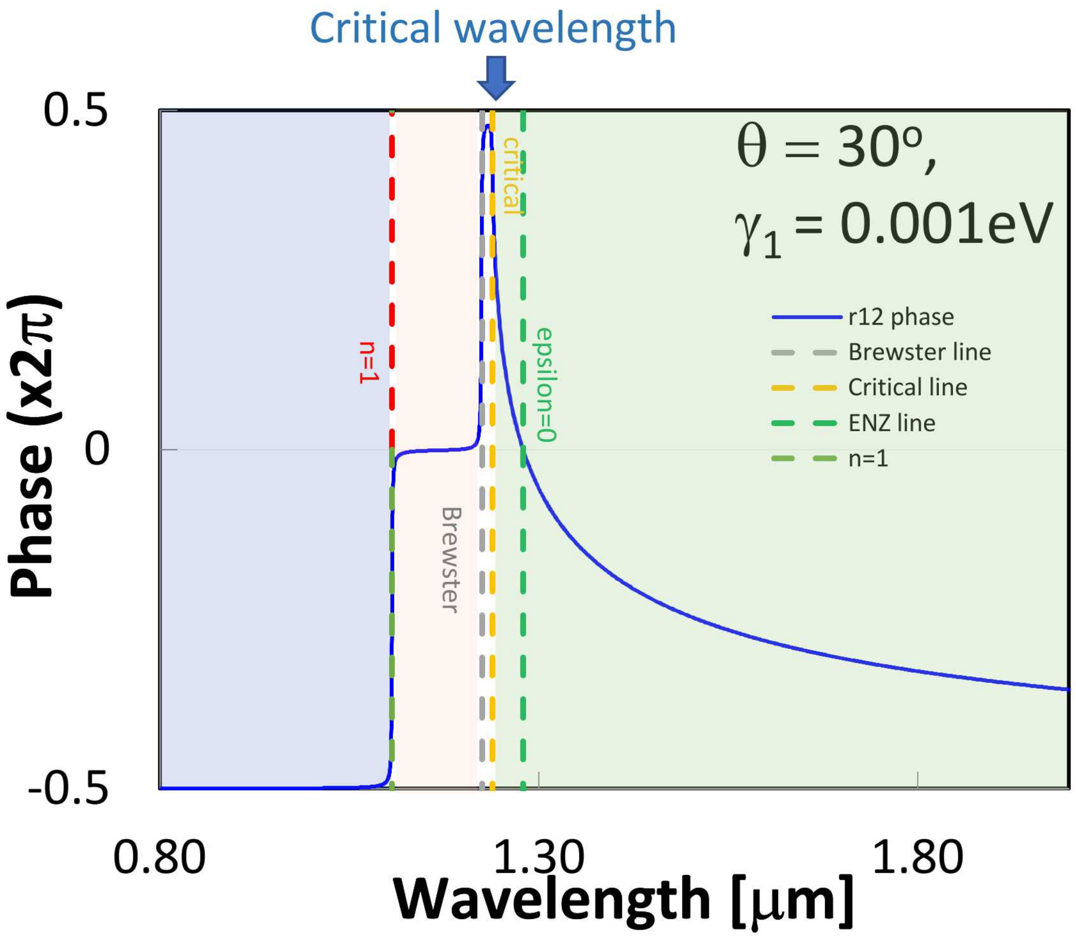




Realizing the needed pi phase shift through total internal reflection

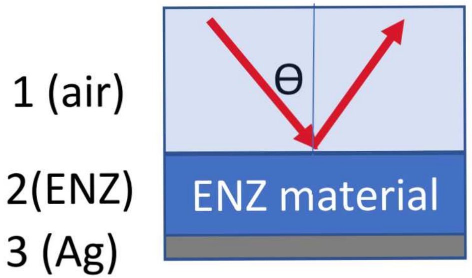
Small loss case





Engineering ENZ perfect absorber using ITO

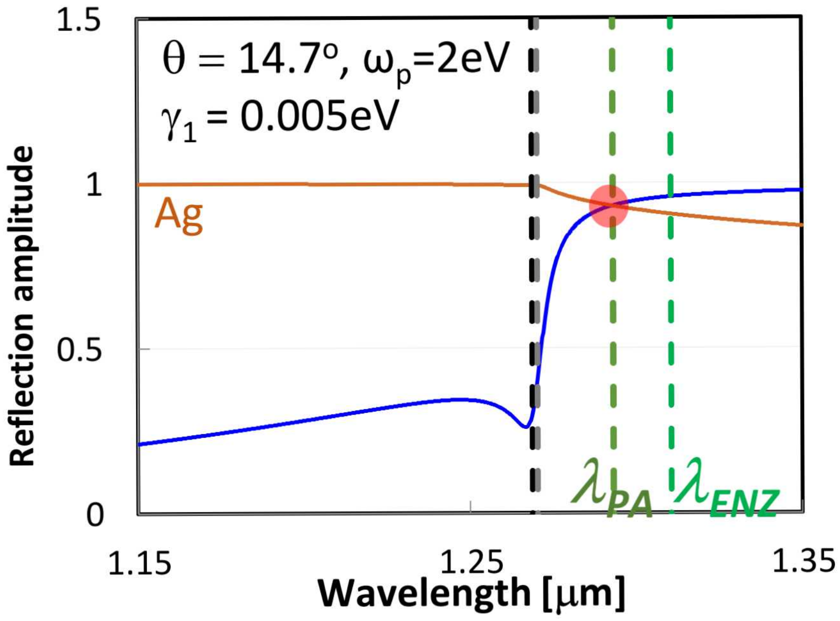
20nm thick ITO, superstrate index 1.5

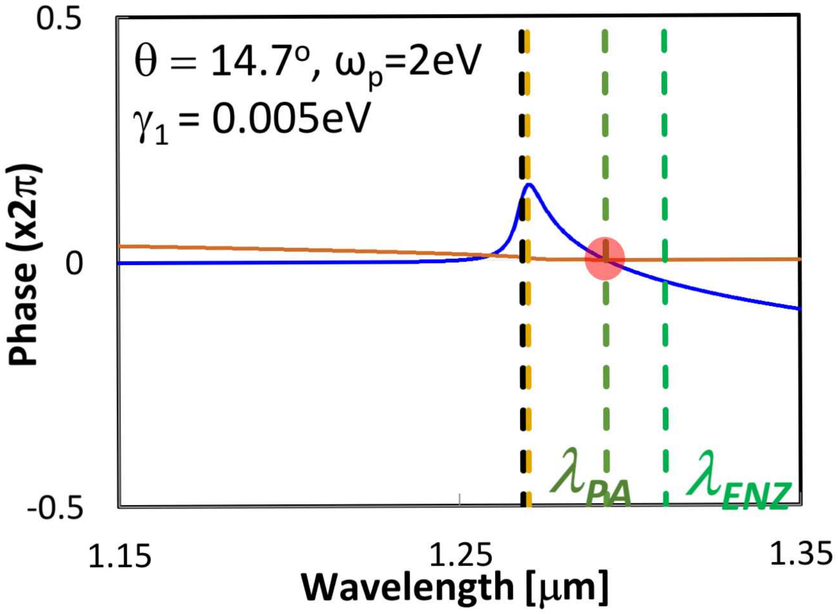


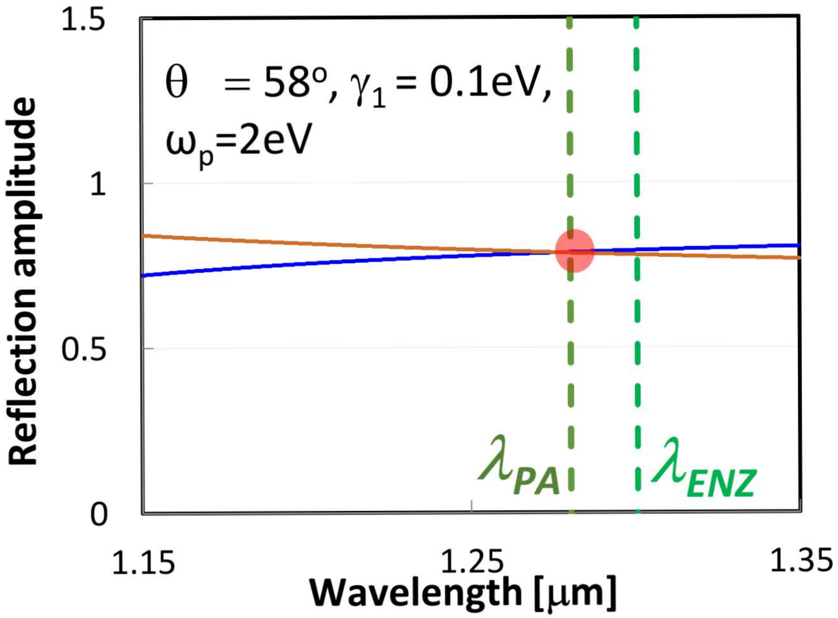
$$\Gamma = \frac{r_{12} + r_{23} e^{2i\phi}}{1 + r_{12} r_{23} e^{2i\phi}}$$

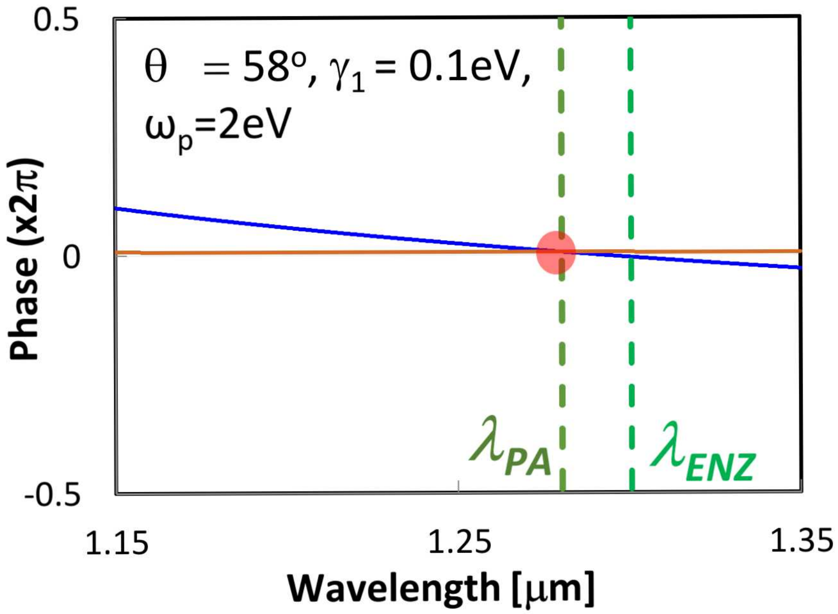
Zero reflection condition

$$r_{12} = -r_{23} e^{2i\phi}$$

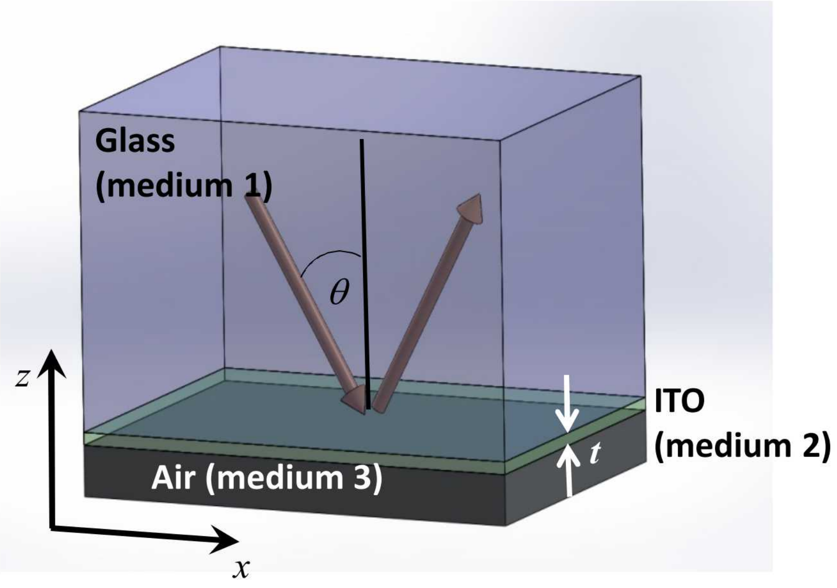








Perfect absorption: critical coupling condition

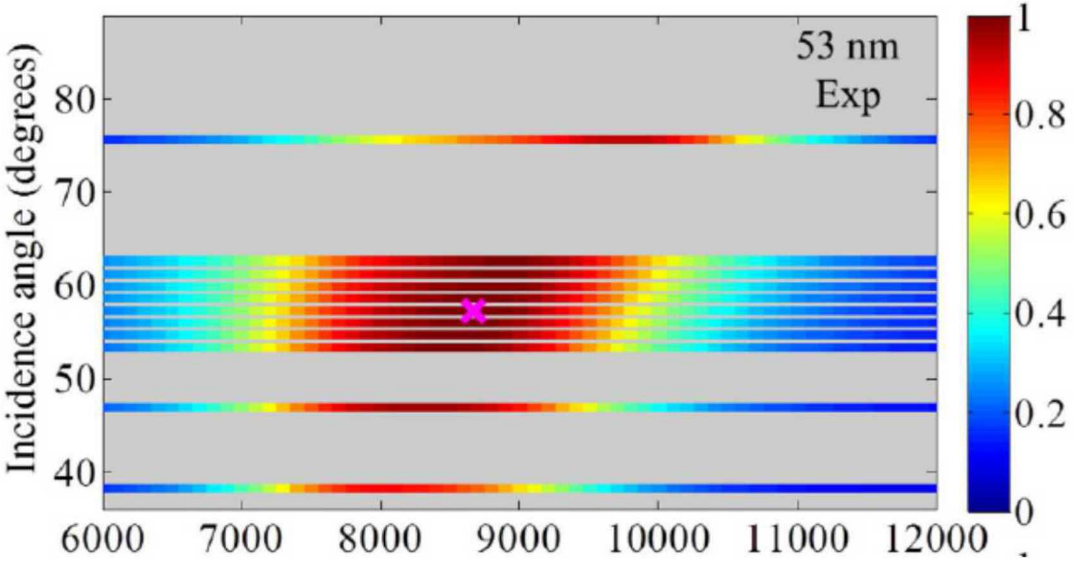


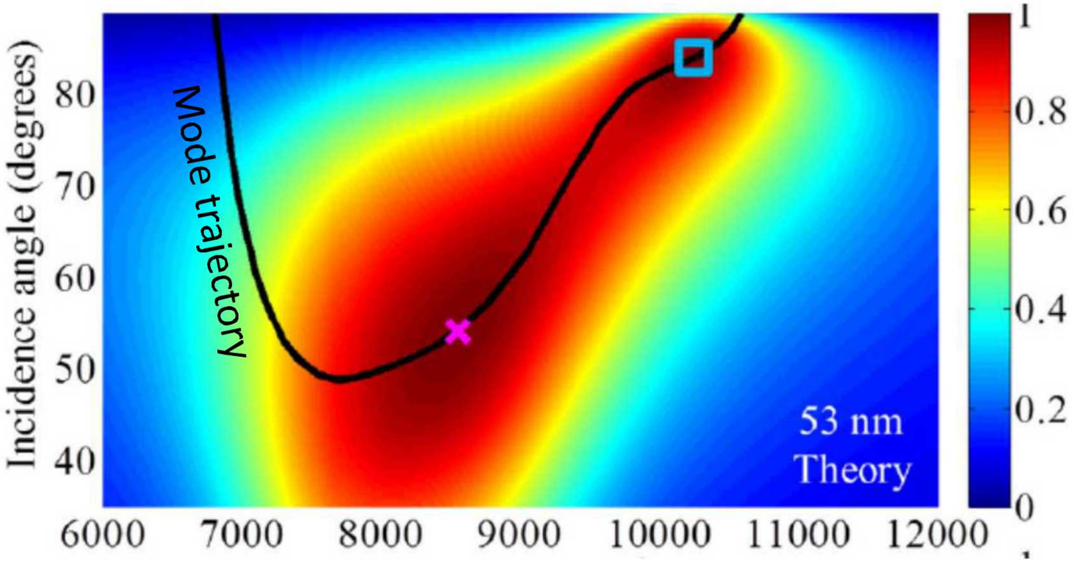
Airy Formula $$\Gamma = \frac{r_{12} + r_{23} e^{2i\phi}}{1 + r_{12} r_{23} e^{2i\phi}} \quad \phi = k_{2\perp} t$$

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)

POLOLOGY of ENZ mode: complex k mode dispersion

$$\Gamma = \frac{r_{12} + r_{23}e^{2i\phi}}{1 + r_{12}r_{23}e^{2i\phi}}$$

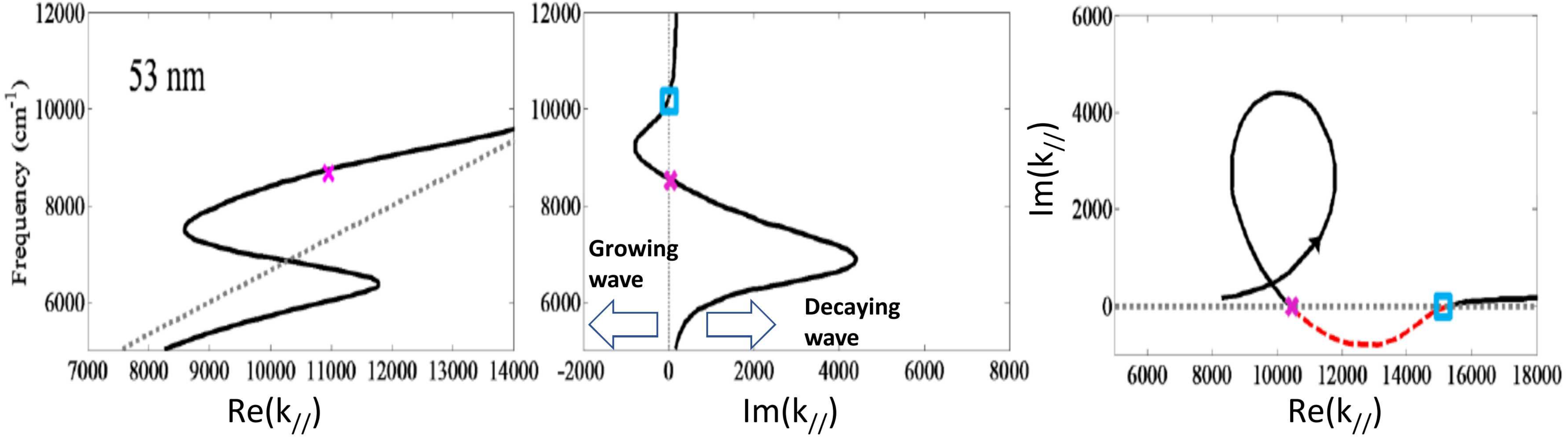
Mode dispersion: $r_{12}r_{23}e^{2i\phi} = -1$

$$r_{ij} = \frac{\dfrac{-k_{i\perp}}{\varepsilon_i} + \dfrac{k_{j\perp}}{\varepsilon_j}}{\dfrac{k_{i\perp}}{\varepsilon_i} + \dfrac{k_{j\perp}}{\varepsilon_j}}$$

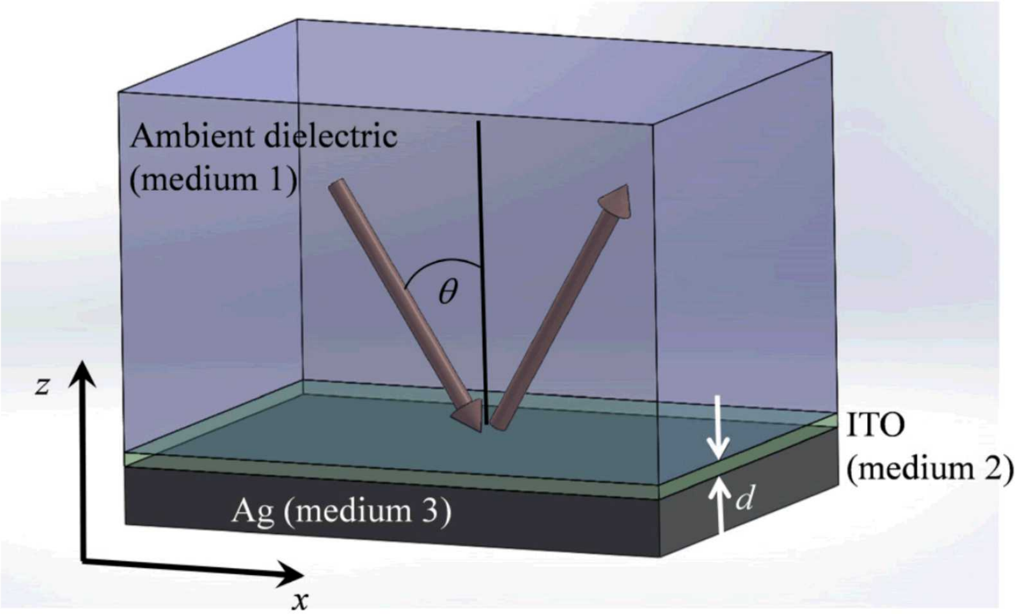
$$k_{i\perp} = \pm\sqrt{\varepsilon_i k_0^2 - k_{i//}^2} \qquad \phi = k_{2\perp}t$$

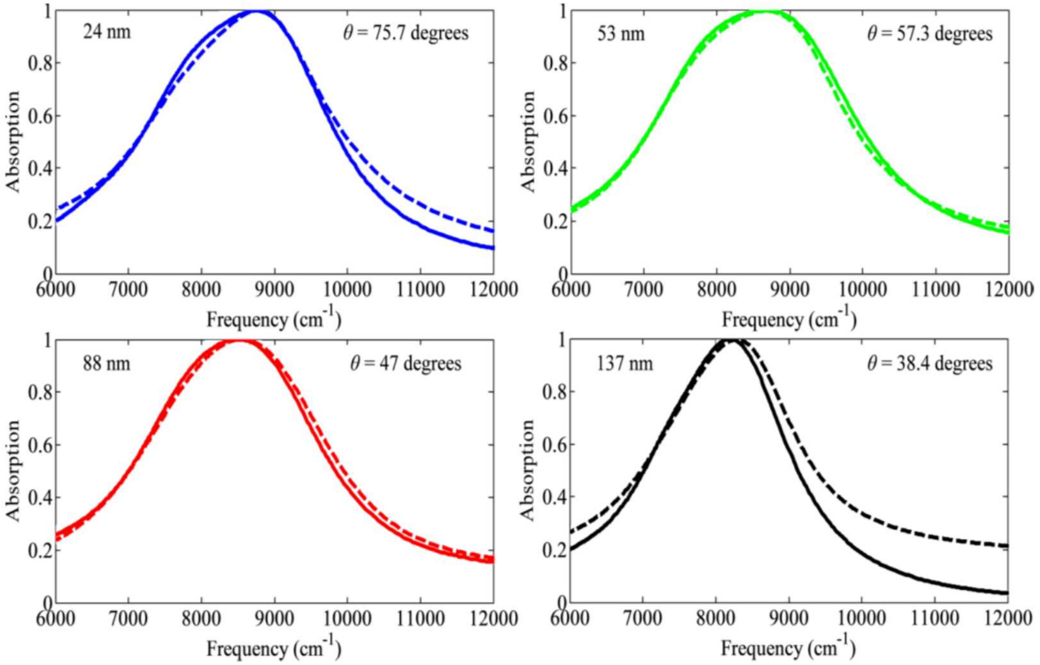
Everything is complex except k_0, ω, and ε_1

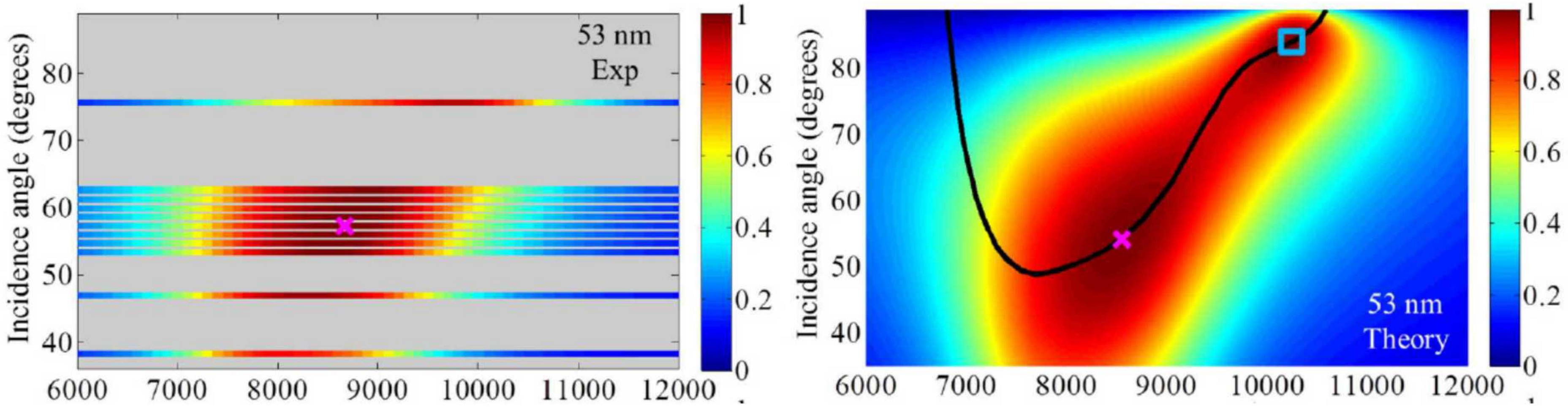
Complex $k_{//}$ solutions



ITO Perfect absorber: experiment



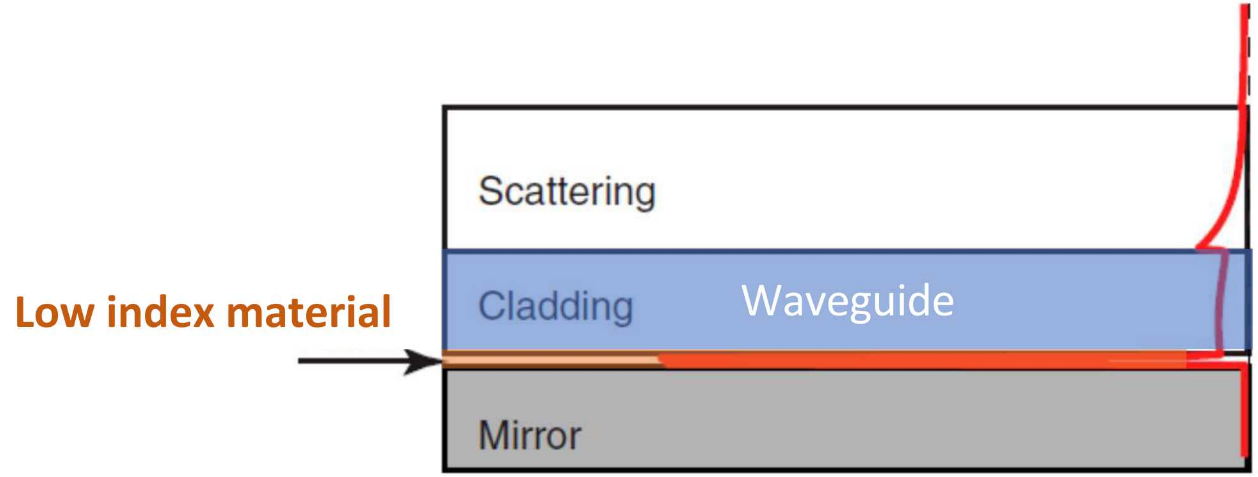




Thinnest perfect absorber we tried is 24nm, $\lambda/50$

20x enhancement in absorption, strongly exceeds the $4n^2/\sin(\theta)$ limit

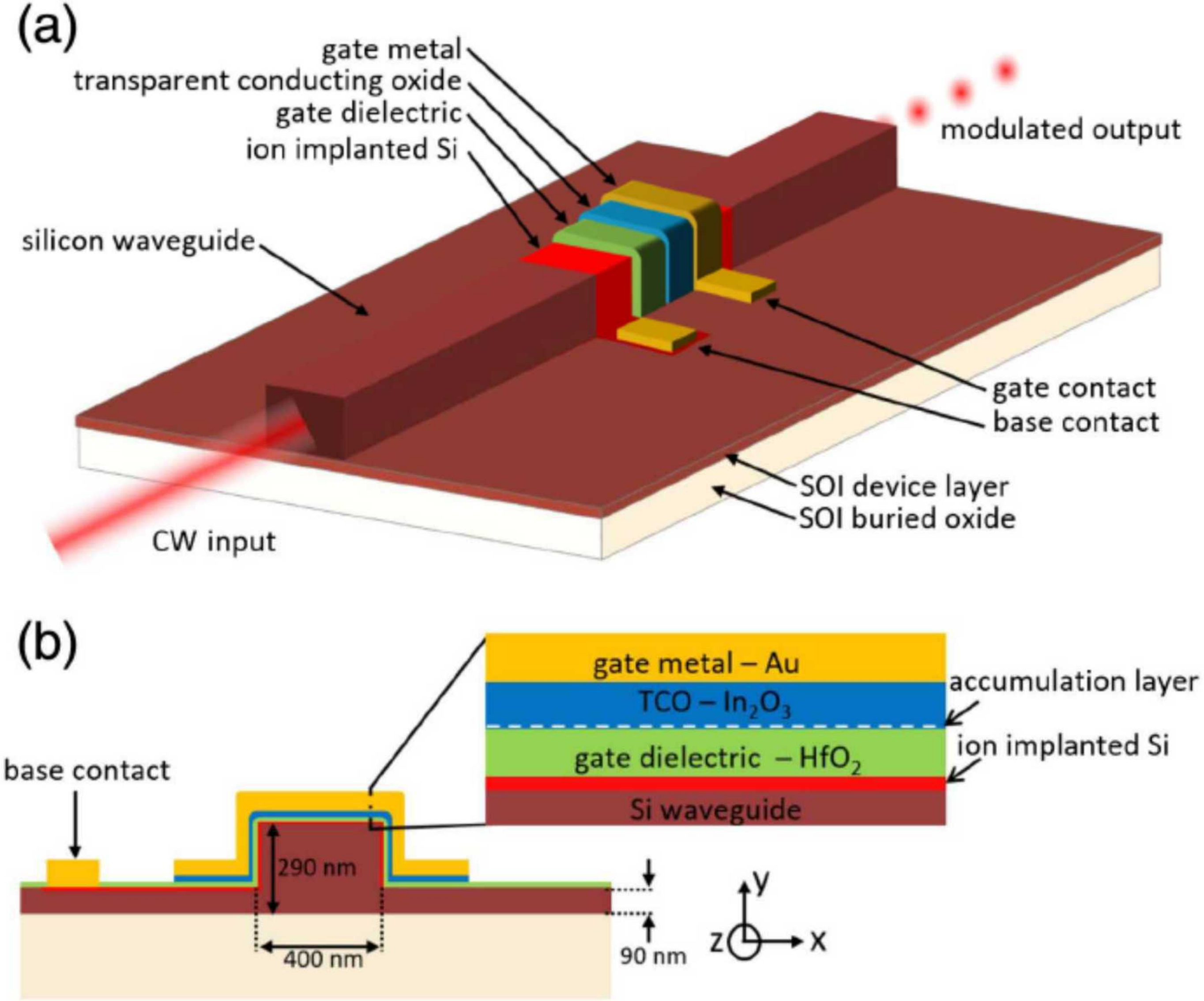
Tuning loss by carrier injection



- 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

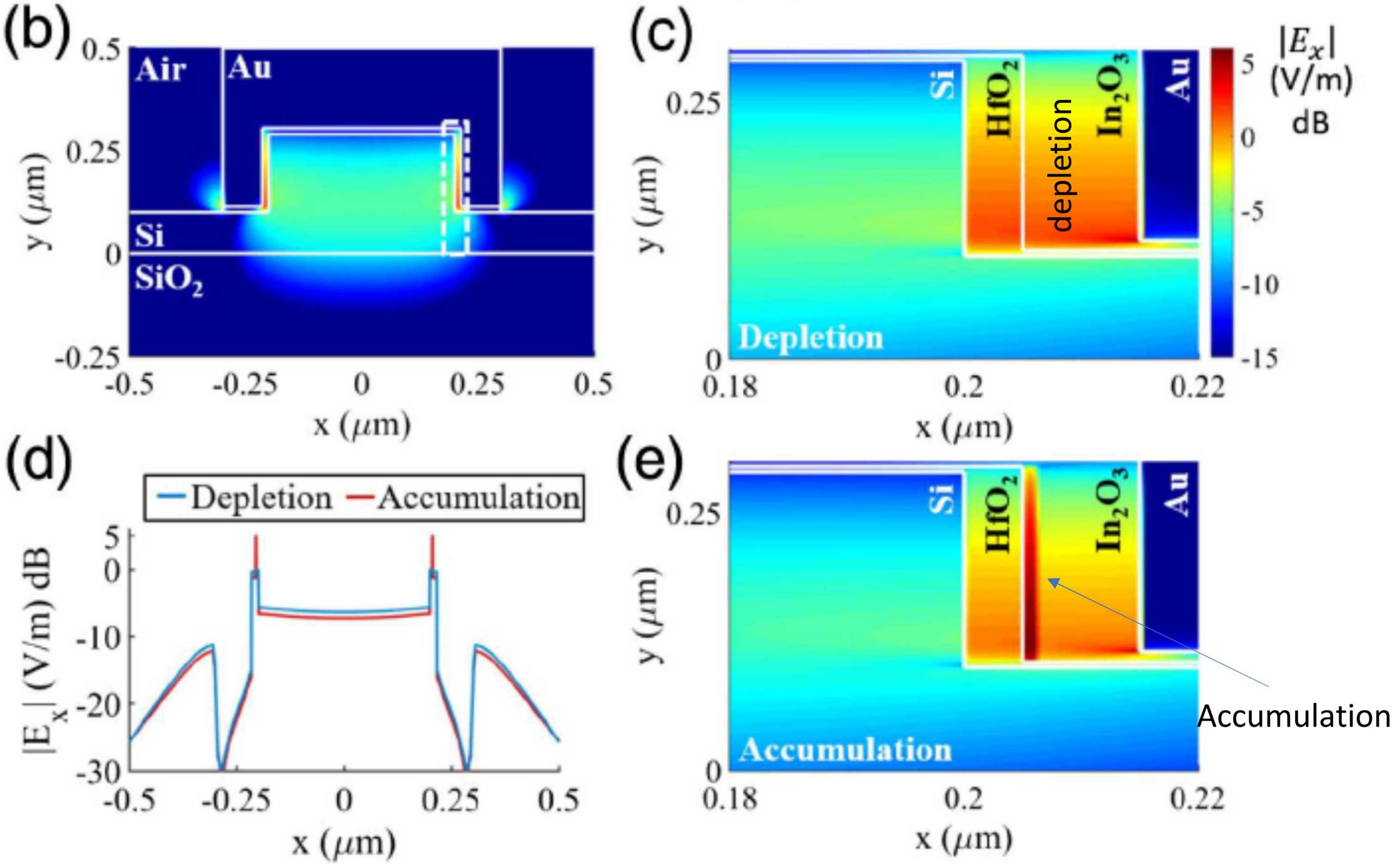
Z. Yu, A. Raman, and S. Fan, "Fundamental limit of nanophotonic light trapping in solar cells," PNAS (2010)

Gigahertz epsilon-near-zero optical modulators



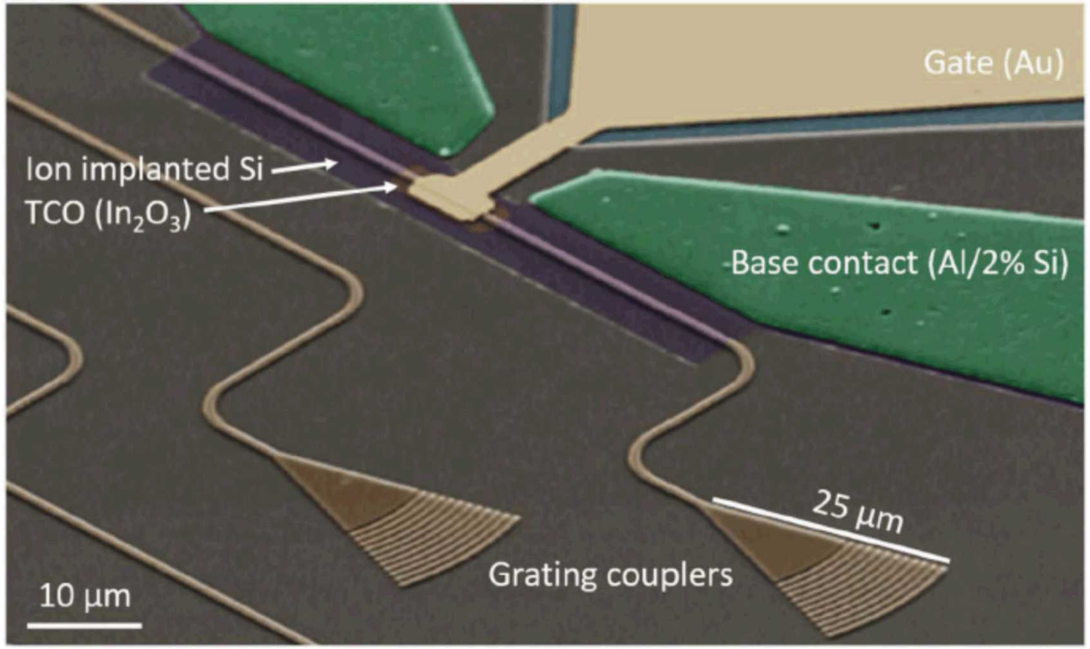
M. G. Wood, S. Campione, S. Parameswaran, T. S. Luk, J. R. Wendt, D. K. Serkland, and G. A. Keeler, "Gigahertz speed operation of epsilon-near-zero silicon photonic modulators," Optica **5**, 233-236 (2018).

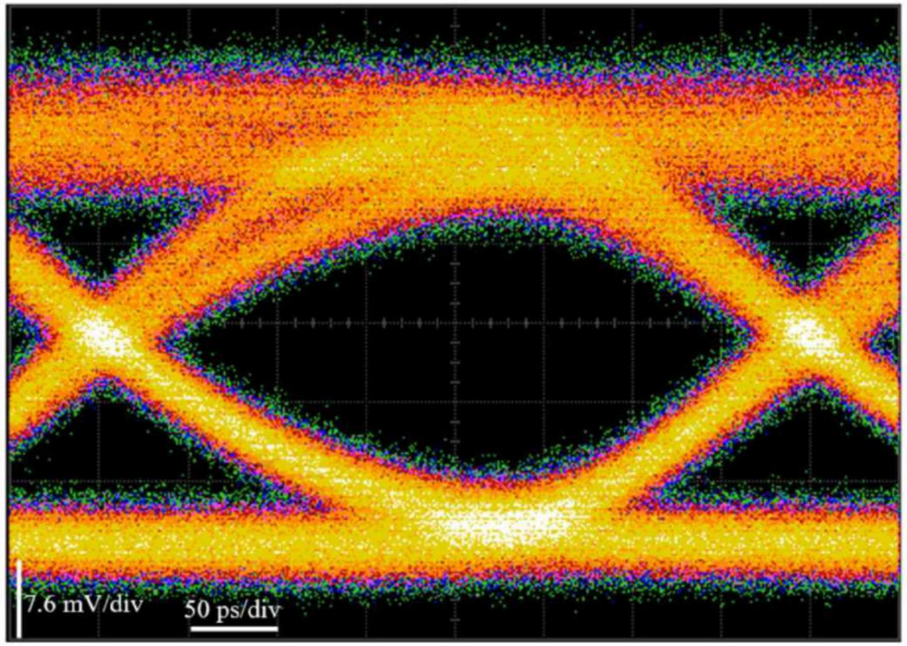
Gigahertz epsilon-near-zero optical modulators



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



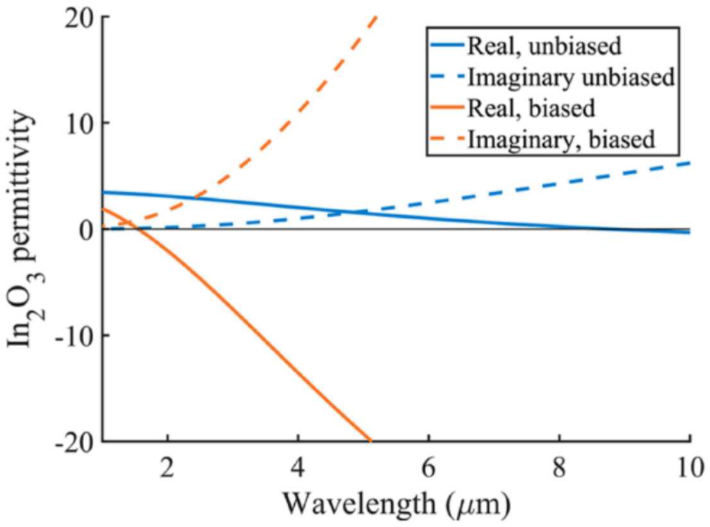


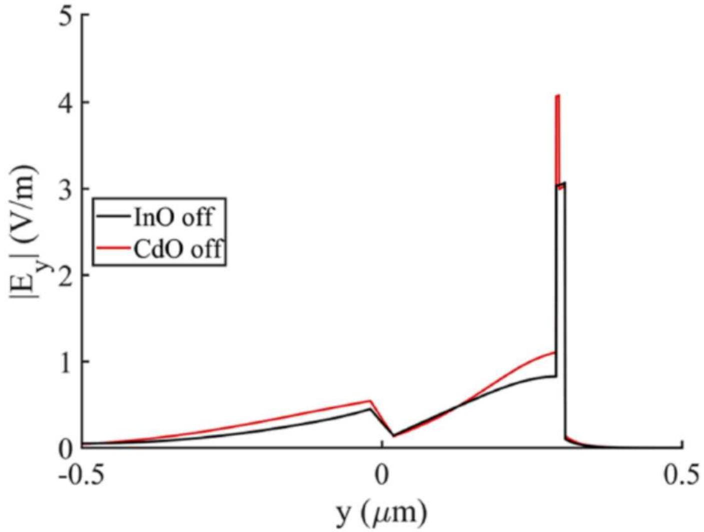
CMOS compatible
Free space coupling through grating
Silicon waveguide: 290x400nm
TCO: In_2O_3 10nm
Gate oxide: Hafnia 5nm
Length: 4um

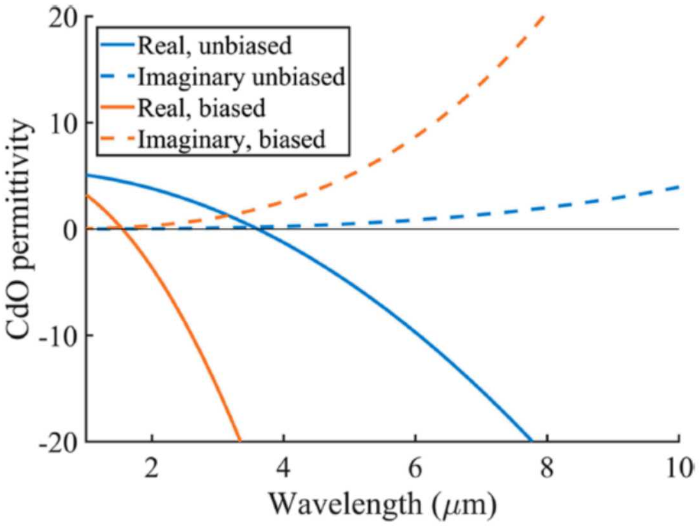
Bandwidth 1530-1590nm
Speed: 2.5 Gb/s, RC limited
Extinction ratio: 6.5 dB
Drive voltage 2 V_{pp}

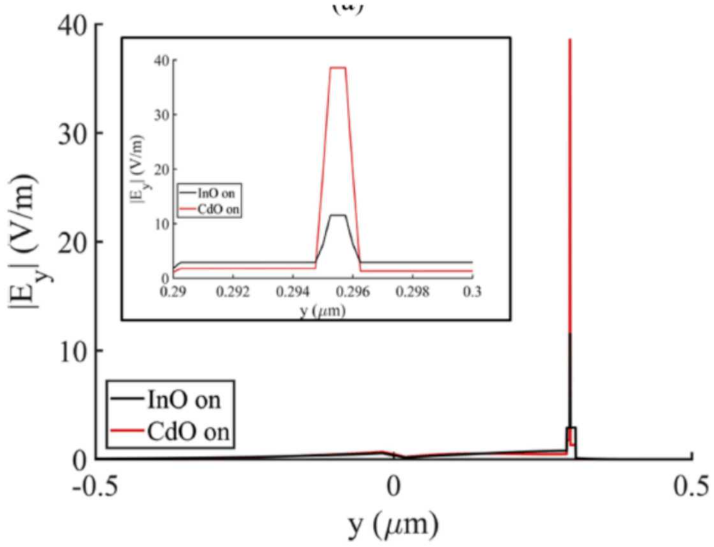
M. G. Wood, S. Campione, S. Parameswaran, T. S. Luk, J. R. Wendt, D. K. Serkland, and G. A. Keeler,
"Gigahertz speed operation of epsilon-near-zero silicon photonic modulators," Optica **5**, 233-236 (2018).

Absorption modulator: In_2O_3 vs CdO

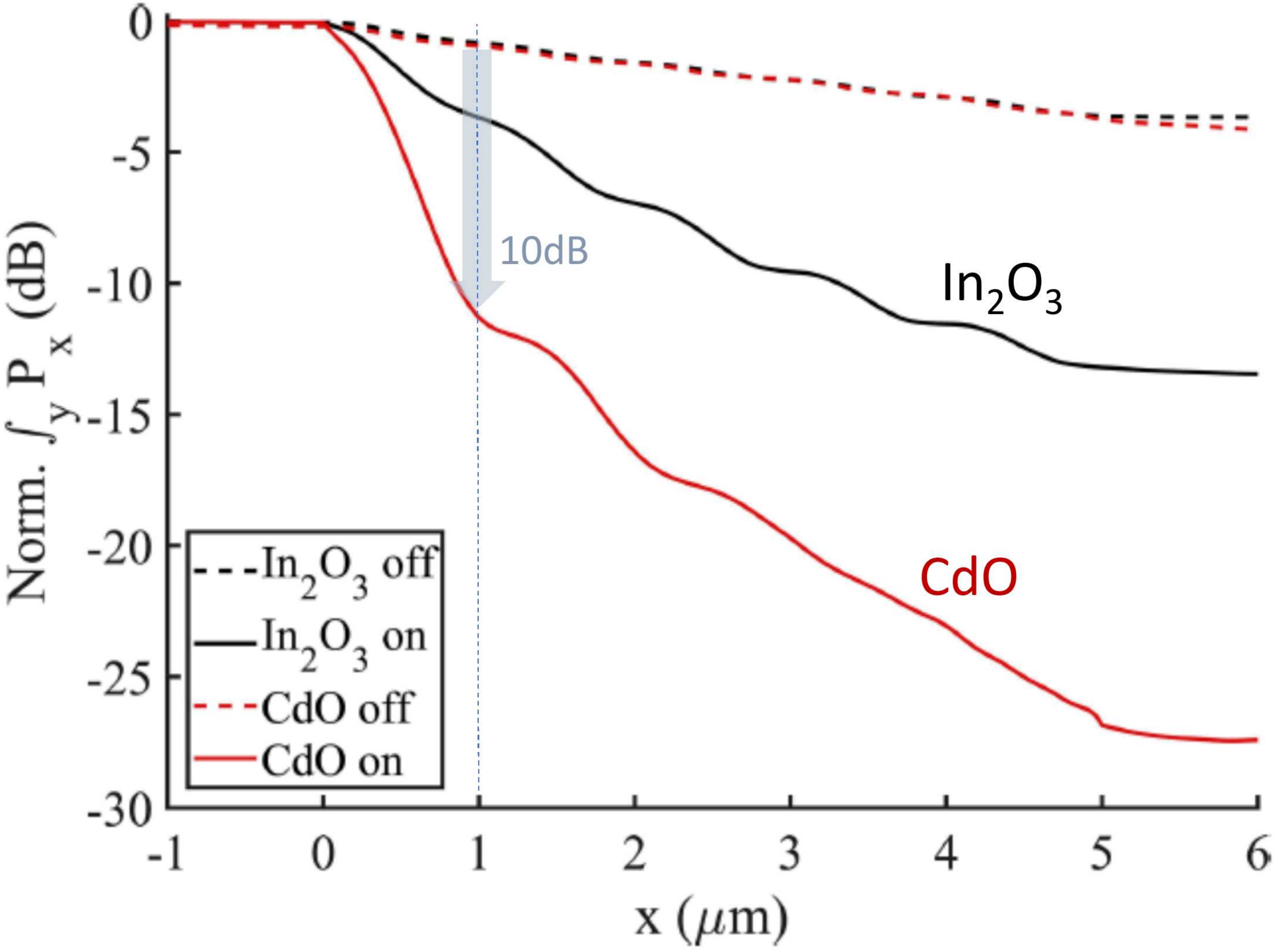




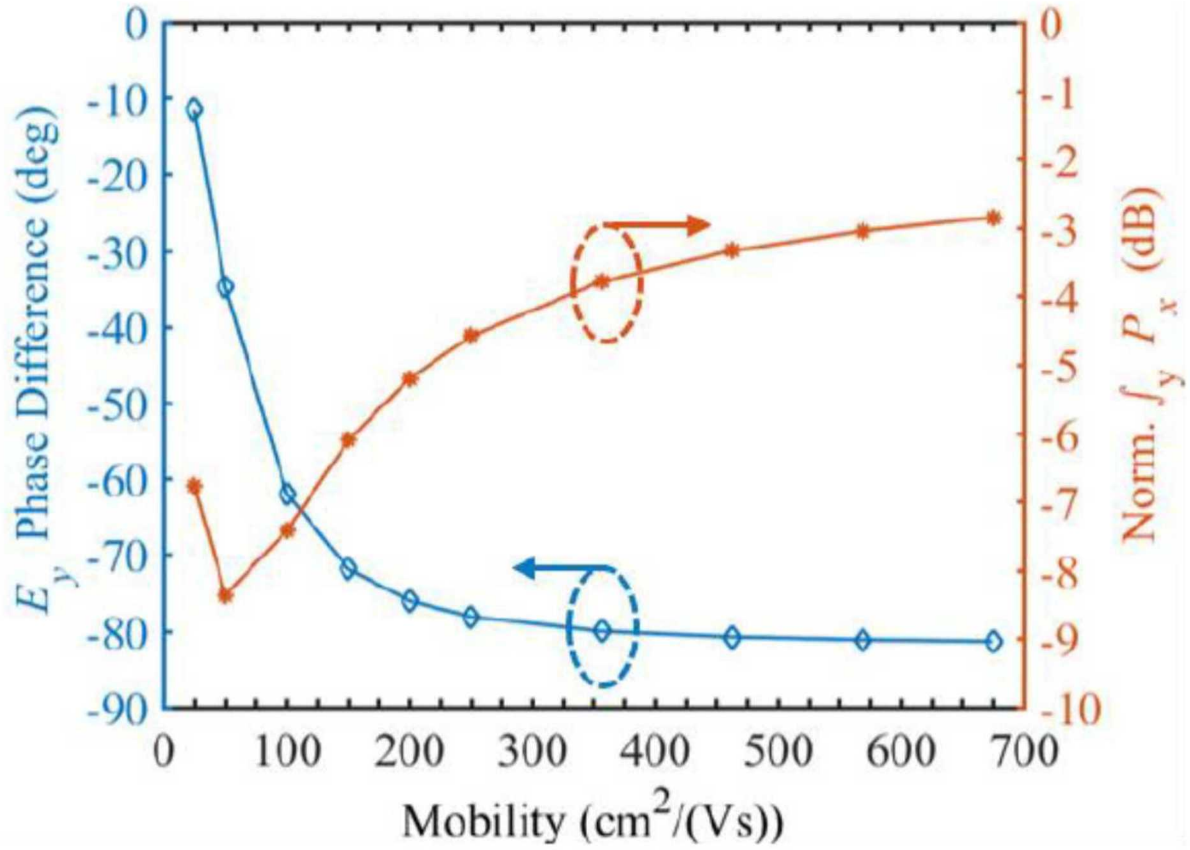


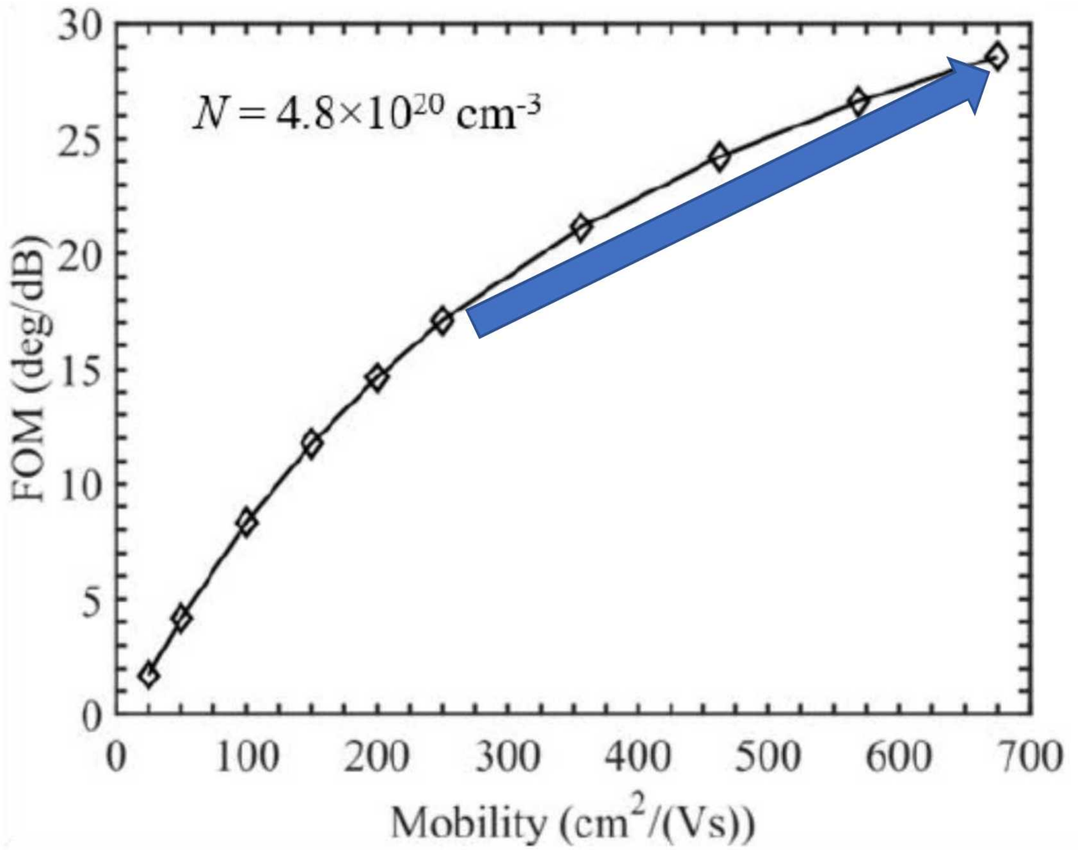


Absorption modulator: In_2O_3 and CdO

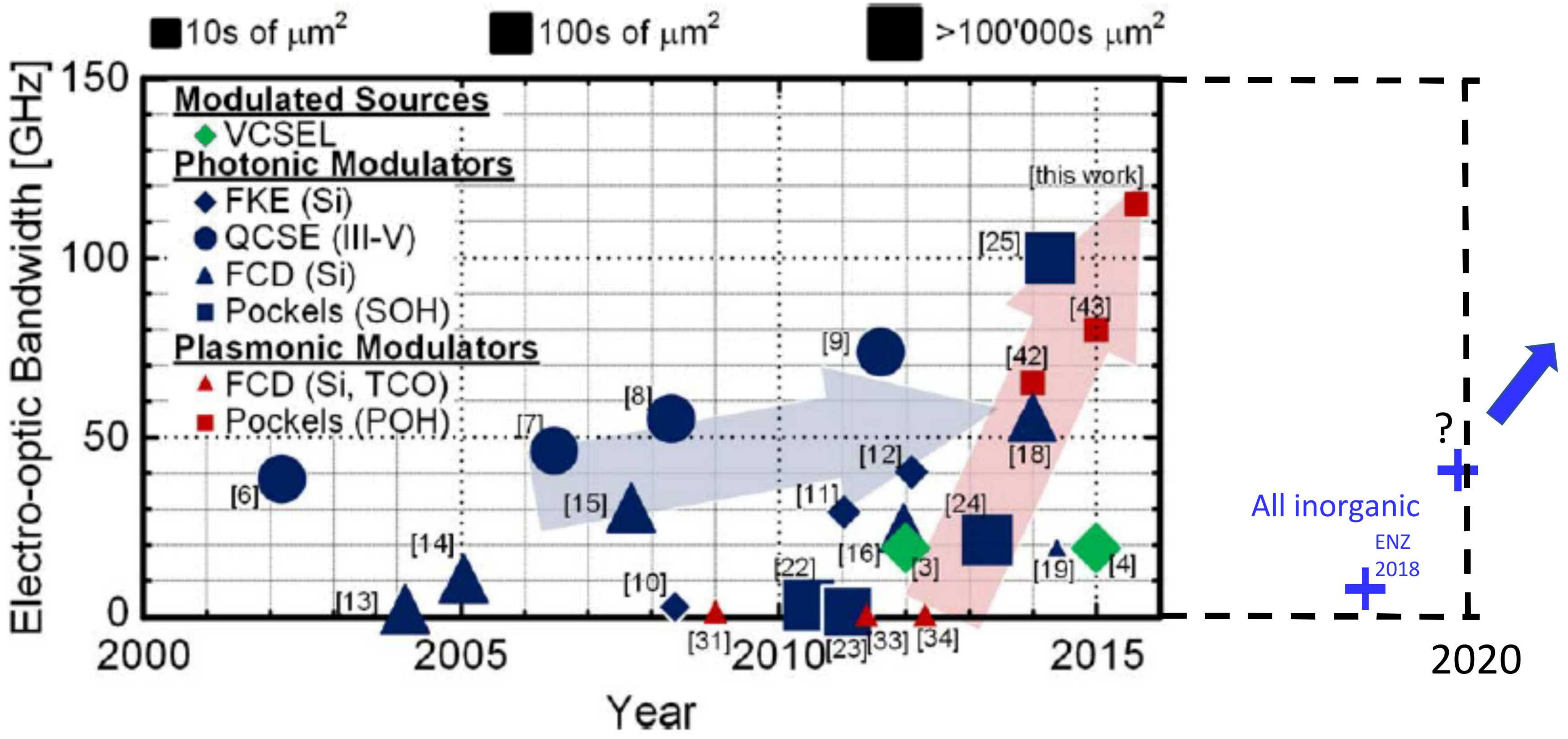


Impact of high mobility conductive oxide





Photonic modulator landscape

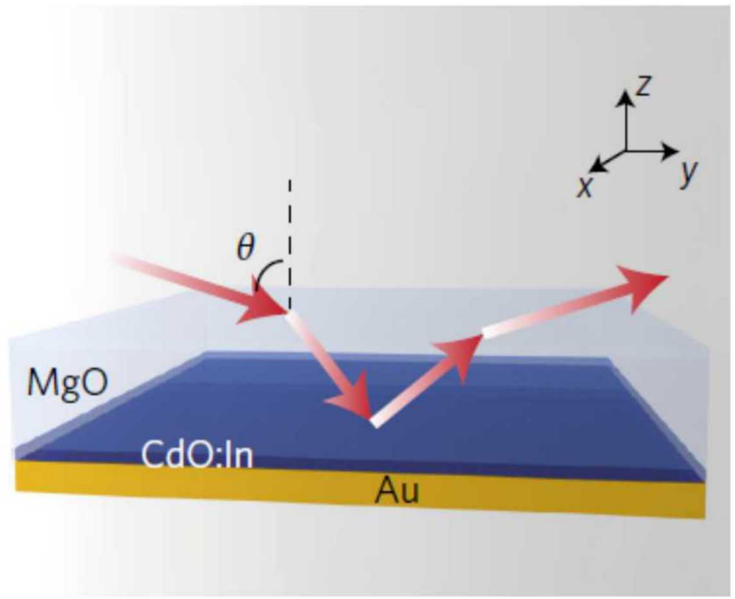


C. Haffner, … , and J. Leuthold, "Plasmonic Organic Hybrid Modulators—Scaling Highest Speed Photonics to the Microscale," Proceedings of the Ieee **104**, 2362-2379 (2016).

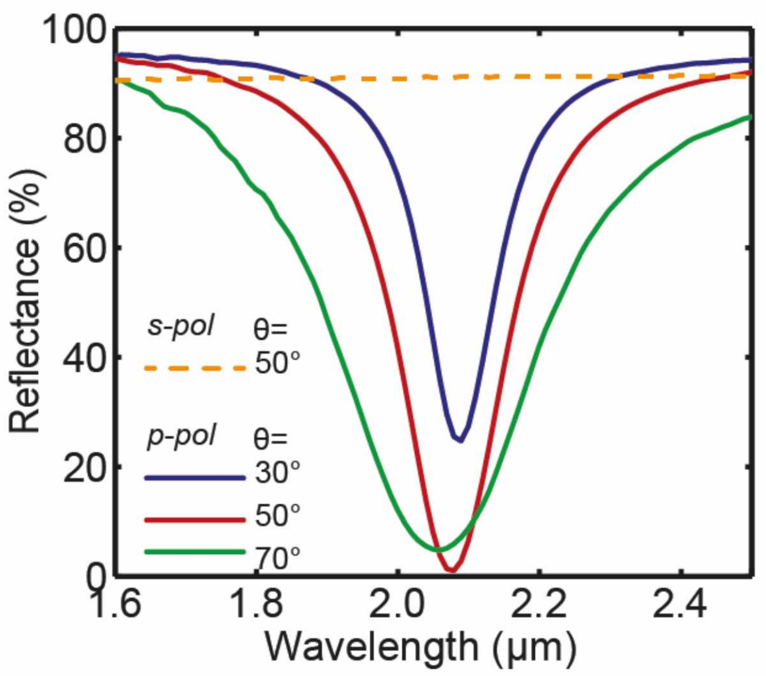
Impact of high mobility conductive oxide

High mobility doped semiconductor CdO

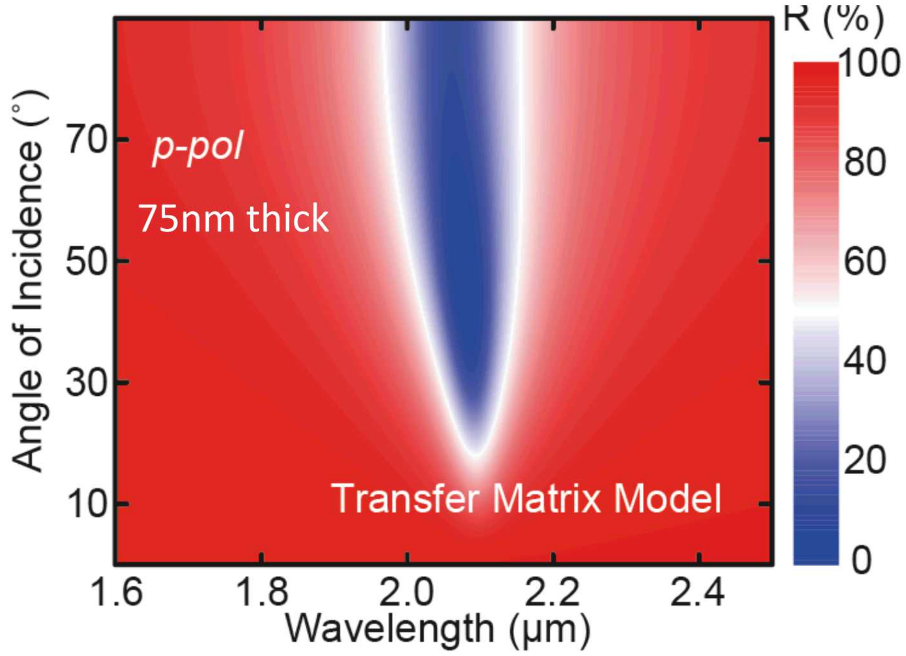
Y:CdO has more than 10x higher mobility than ITO



Measured reflectivity



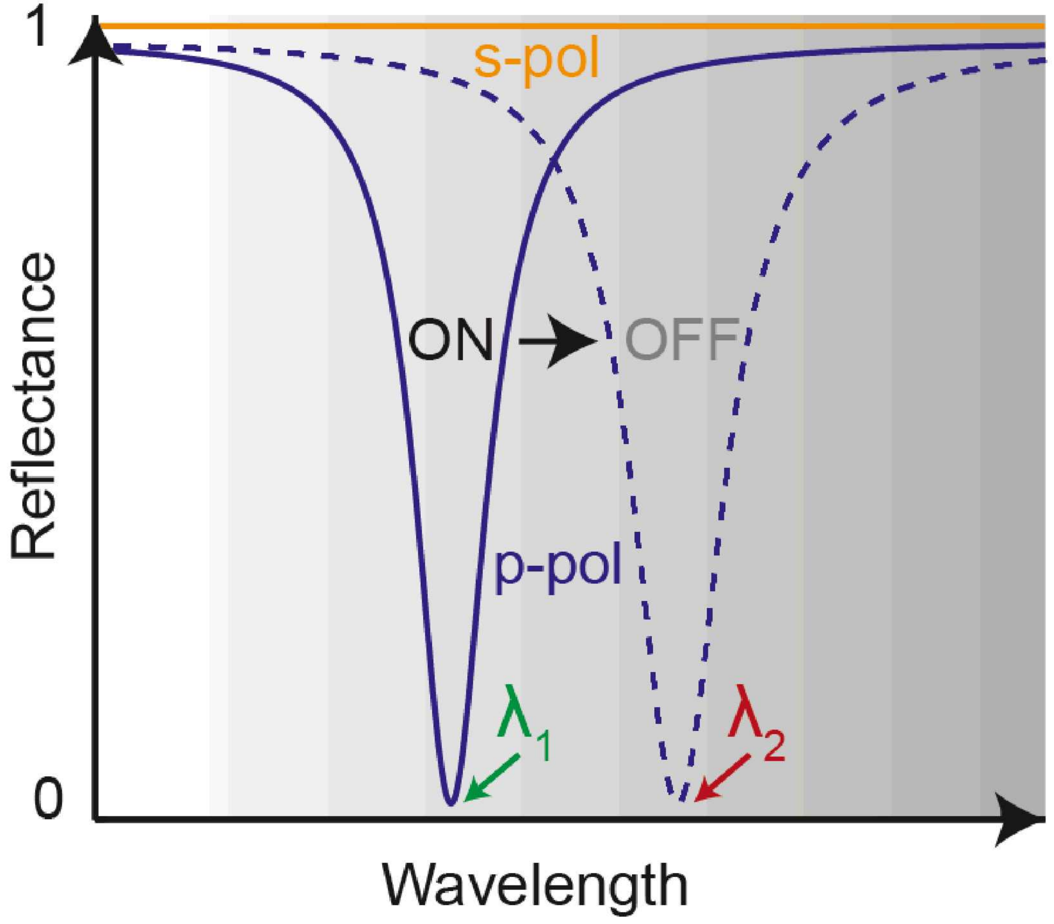
Transfer Matrix simulation



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)

Effective mass tuning of ENZ material

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

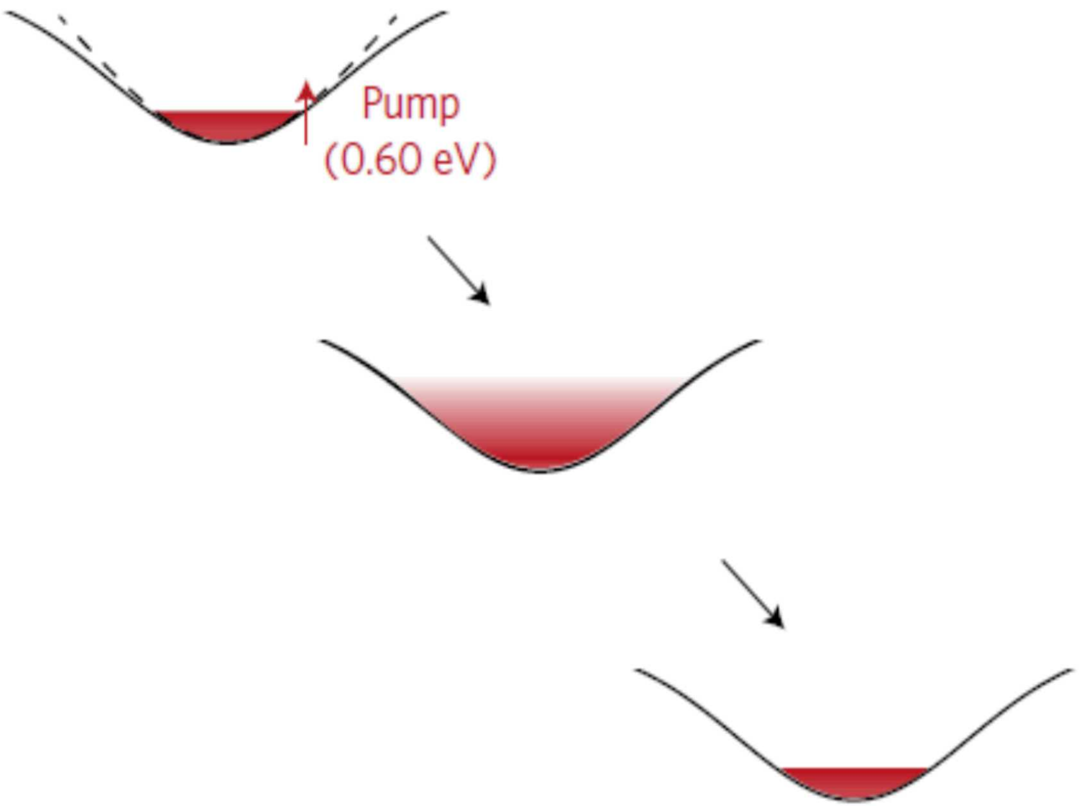


Y. Yang, K. Kelley, E. Sachet, S. Campione, T. S. Luk, J.-P. Maria, M. B. Sinclair, and I. Brener, "Femtosecond optical polarization switching using a cadmium oxide-based perfect absorber," Nature Photonics **11**, 390 (2017)

Tuning effective mass by heating

Electron heating mechanism

Speed limitations: Hot carrier lifetime



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

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

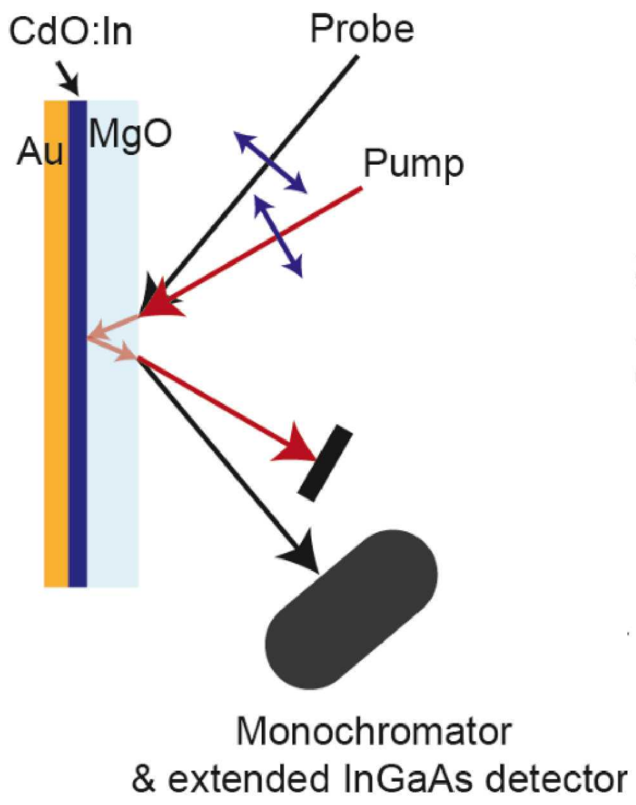
T_{final} (19nm sample)=2.5x10^{4}K

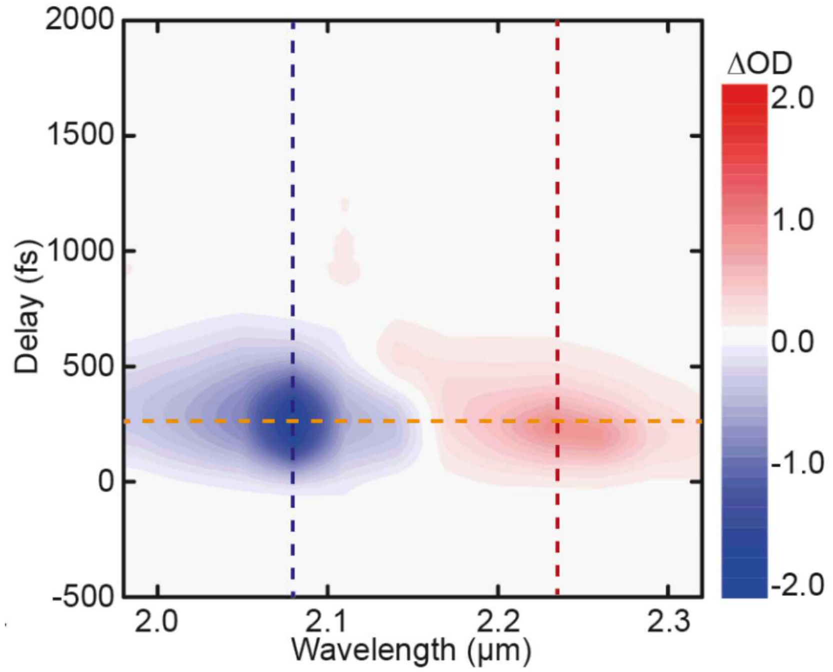
$$m^* = \frac{\hbar^2 \int f(E,T)\,dk}{\int f(E,T)\left(d^2E/dk^2\right)dk}$$

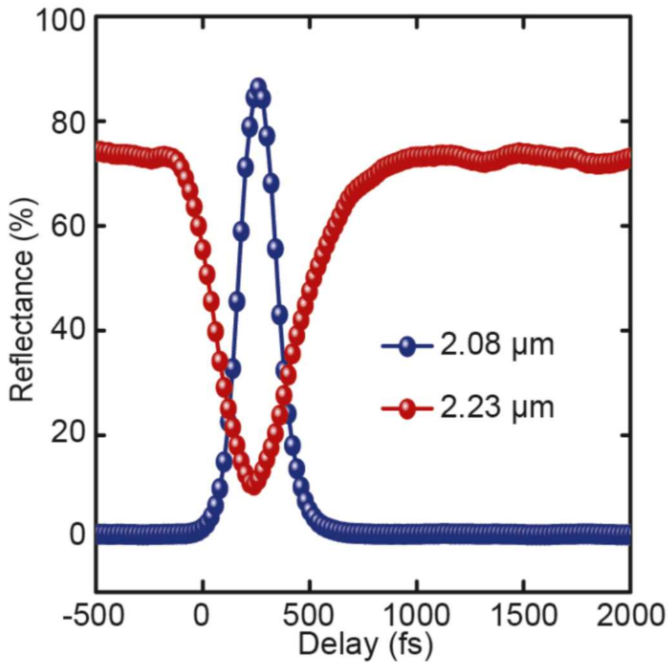
Plasma frequency:

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

Ultrafast AMPLITUDE SWITCHING of the Perfect Absorber



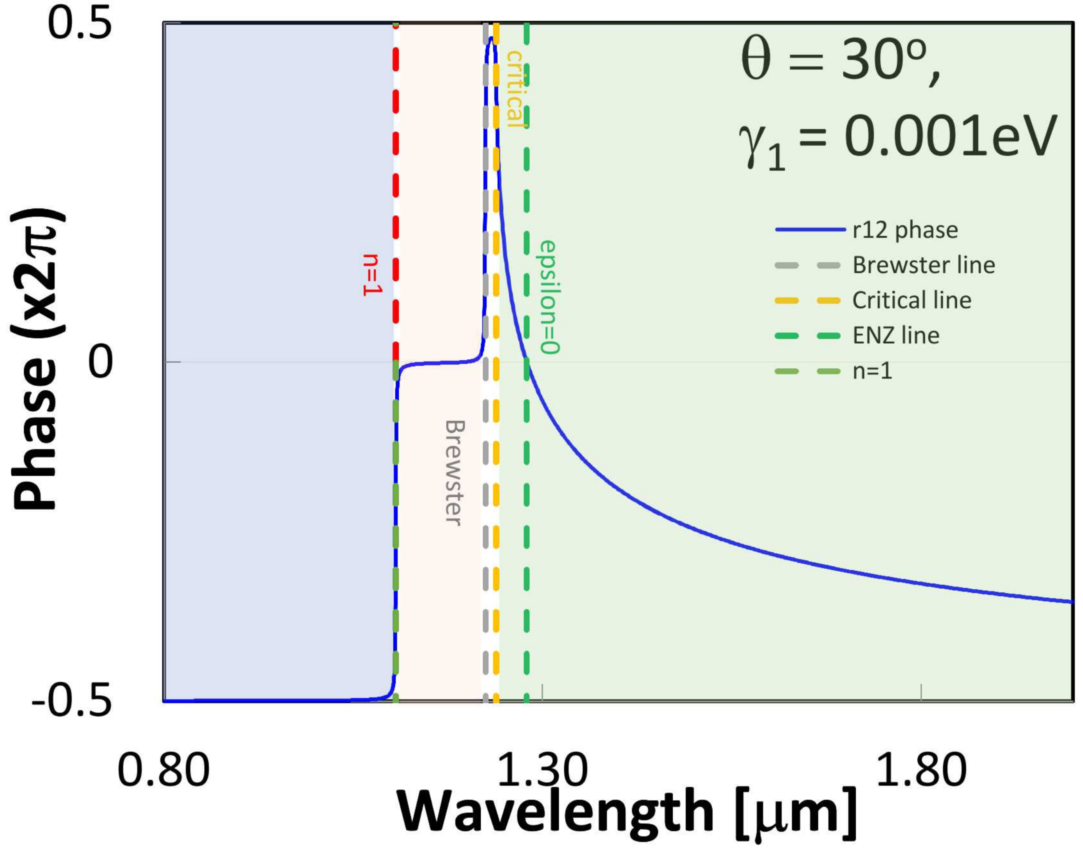




- ***Absolute reflectance* modulation from *1% to 86%!***
- ***Pump energy density: 339 μJ/cm²***
- ***Effective mass change of about 10%***

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)

Recall the phase shift effect for P-polarized light



Intensity dependent index of refraction and permittivity

$$\epsilon(I) = \varepsilon(0) + 3\chi^{(3)}E^2 = \varepsilon(0) + \frac{3\cdot 377}{2}\chi^{(3)}\frac{I}{Re\big(n(I)\big)} = \varepsilon(0) + a\frac{I}{Re\big(n(I)\big)} = \big(n_0 + \Delta n(I)\big)^2$$

$$E^2 = \frac{I}{2Re\big(n(I)\big)}\sqrt{\frac{\mu_0}{\varepsilon_0}} = \frac{377}{2}\frac{I}{Re\big(n(I)\big)}$$

$$a = \frac{3\cdot 377}{2}\chi^{(3)} \qquad n(I) = n_0 + \Delta n$$

$$\Delta\varepsilon = \epsilon(I) - \varepsilon(0) = a\frac{I}{Re\big(n(I)\big)} \cong a\frac{I}{n_0 + \Delta n} = \Delta n^2 + 2n_0\,\Delta n$$

$$\Delta n^3 + 3n_0\Delta n^2 + 2n_0^2\Delta n - aI = 0$$

If ignore the cubic and quadratic term $\Delta n(I) = \frac{3\cdot 377\chi^{(3)}}{4n_0^2}I$

If ignore the cubic term $\Delta n(I) = -\frac{1}{3}n_0 \pm \frac{1}{3\sqrt{n_0}}\sqrt{n_0^3 + \frac{3\cdot 377}{2}\chi^{(3)}I}$

The solution with the minus sign is not physical because $\lim_{I\to 0}\Delta n(I) \neq 0$

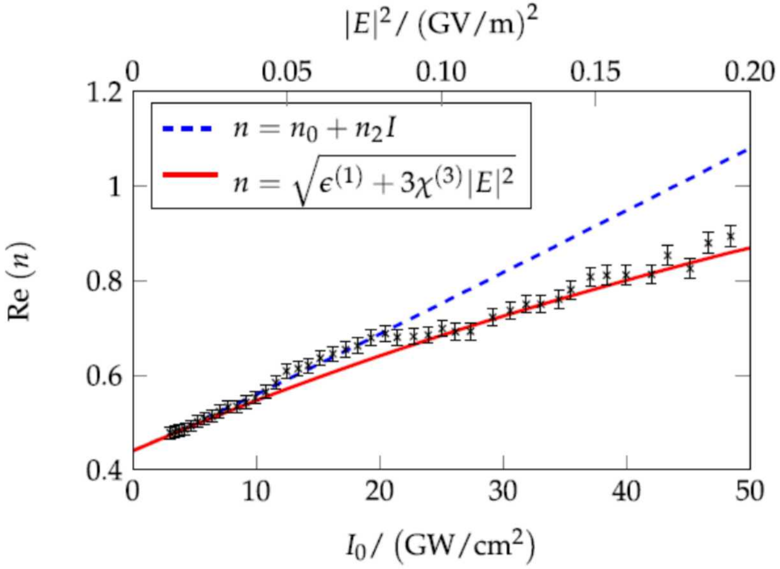
Nonlinear index: ENZ effect

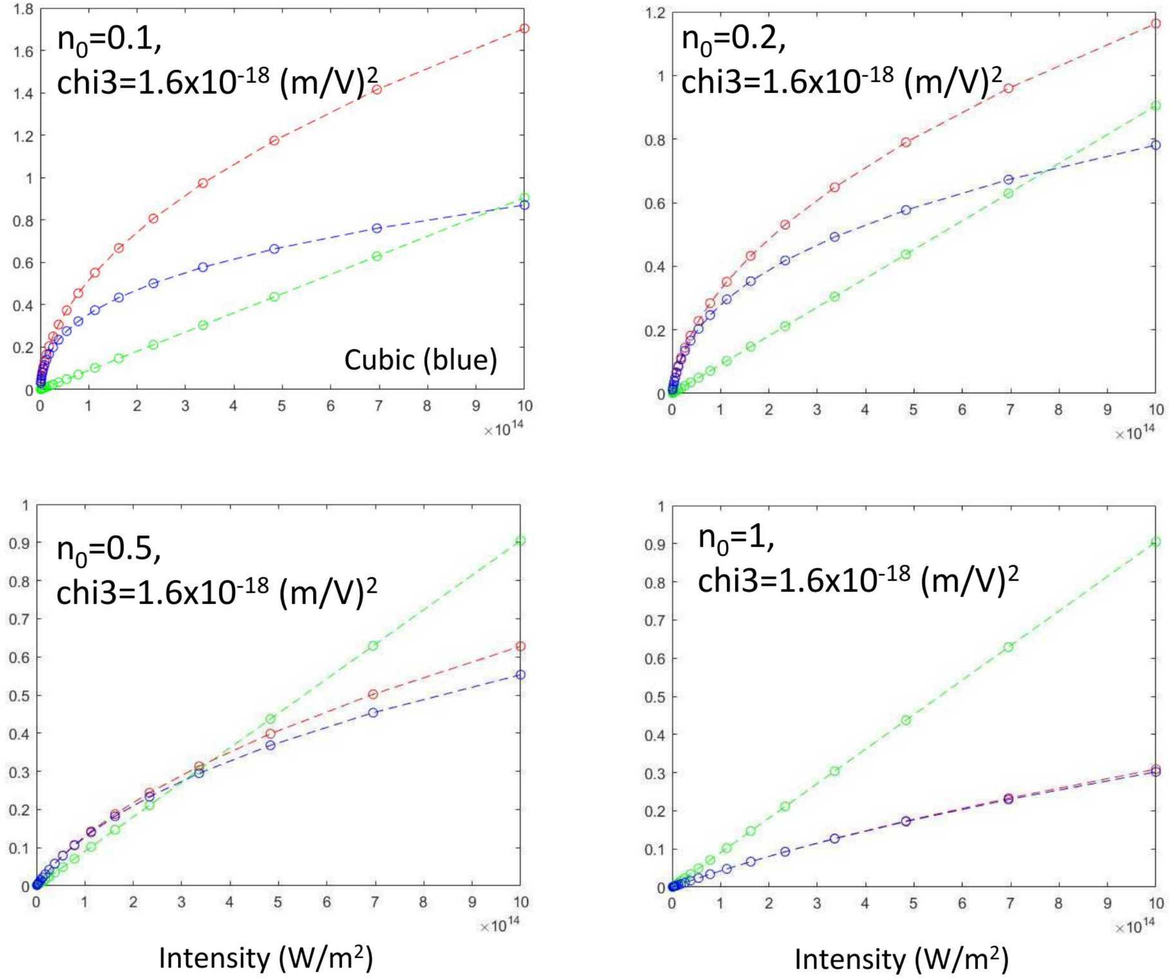
Linear approximation(green)

$$\Delta n(I) = \frac{3 \cdot 377 \chi^{(3)}}{4 n_0^2} I$$

Quadratic approximation(red)

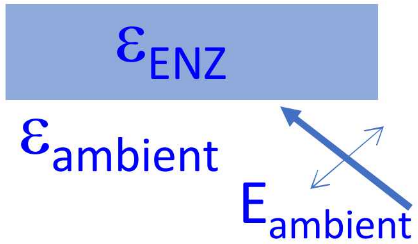
$$\Delta n(I) = -\frac{1}{3} n_0 + \frac{1}{3\sqrt{n_0}} \sqrt{n_0^3 + \frac{3 \cdot 377}{2} \chi^{(3)} I}$$



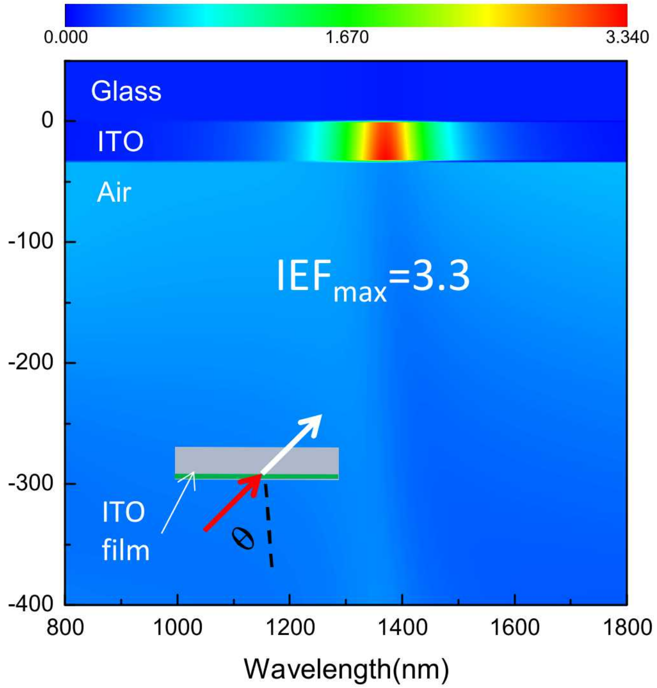


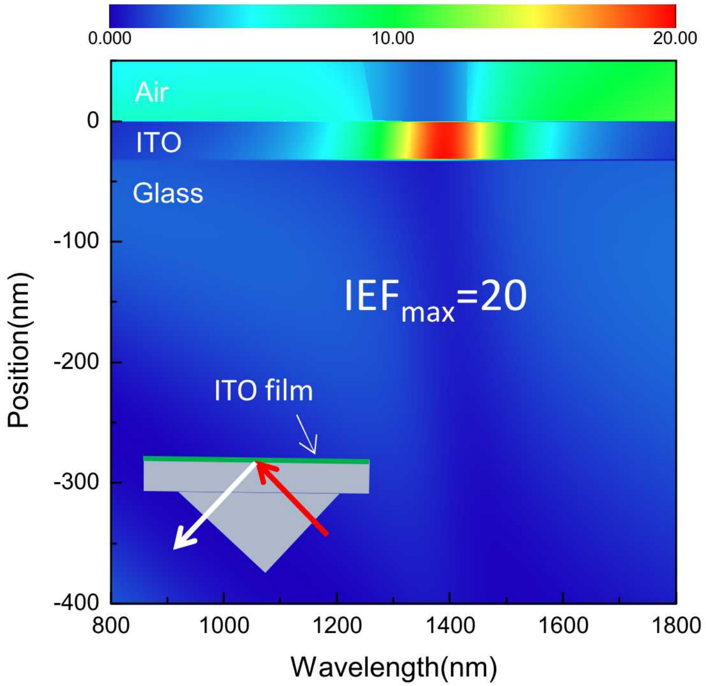
O. Reshef, E. Giese, M. Zahirul Alam, I. De Leon, J. Upham, and R. W. Boyd, "Beyond the perturbative description of the nonlinear optical response of low-index materials," Optics Letters **42**, 3225-3228 (2017)

Enhancing field with ENZ material



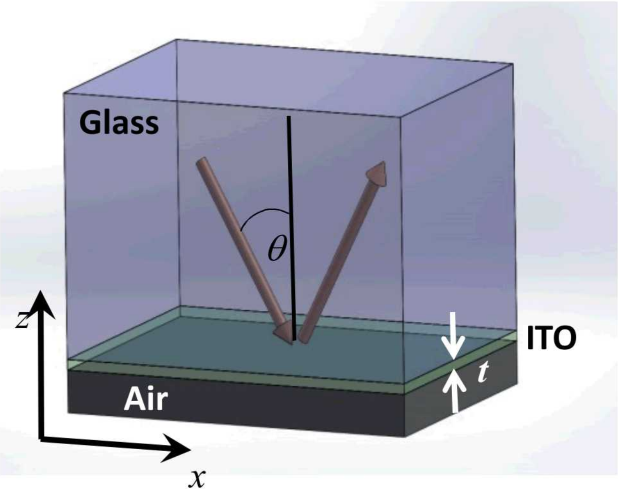
$$E_{ENZ} = D/\varepsilon_{ENZ} = E_{ambient}\varepsilon_{ambient}/\varepsilon_{ENZ}$$





T. S. Luk et. al, "Enhanced third harmonic generation from epsilon-near-zero modes of ultrathin films" *APL, 151103(*2015)

Third harmonic generation using ENZ mode of ITO



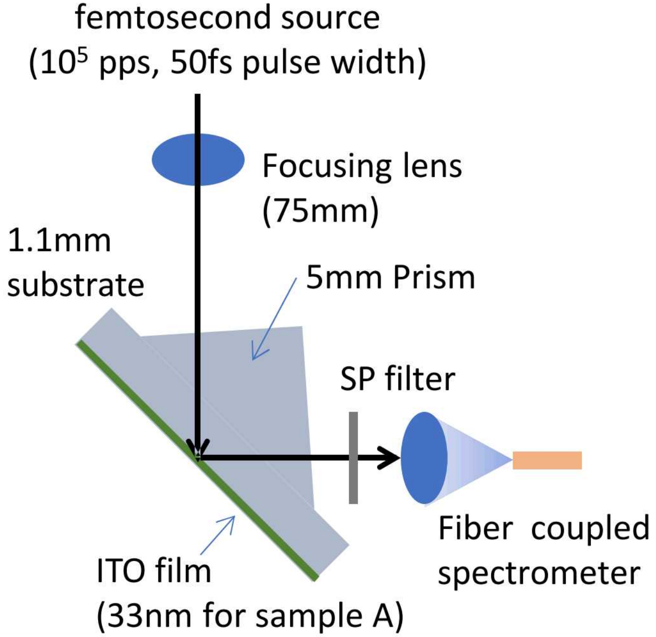
Mechanics of observing TH -

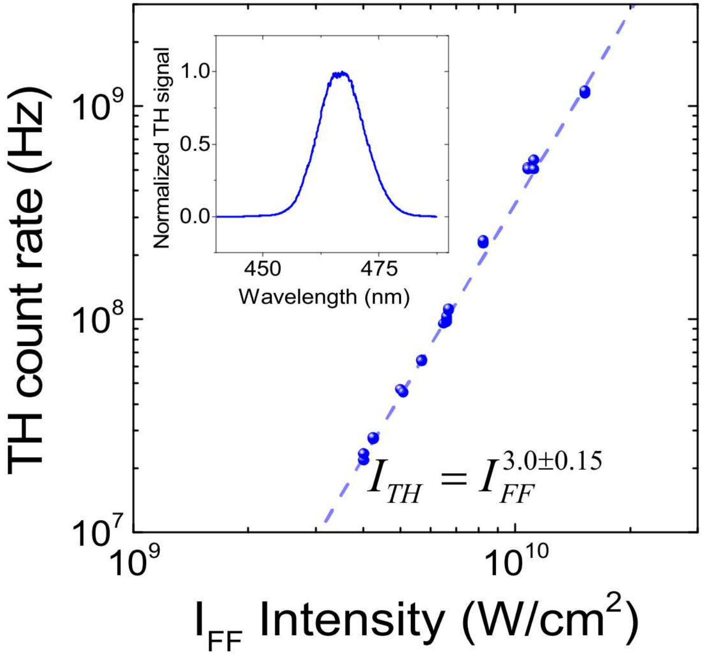
Momentum conservation: $3\vec{k}_{FF} = \vec{k}_{TH}$

Continuity condition: $3k_{FF//} = k_{TH//}$

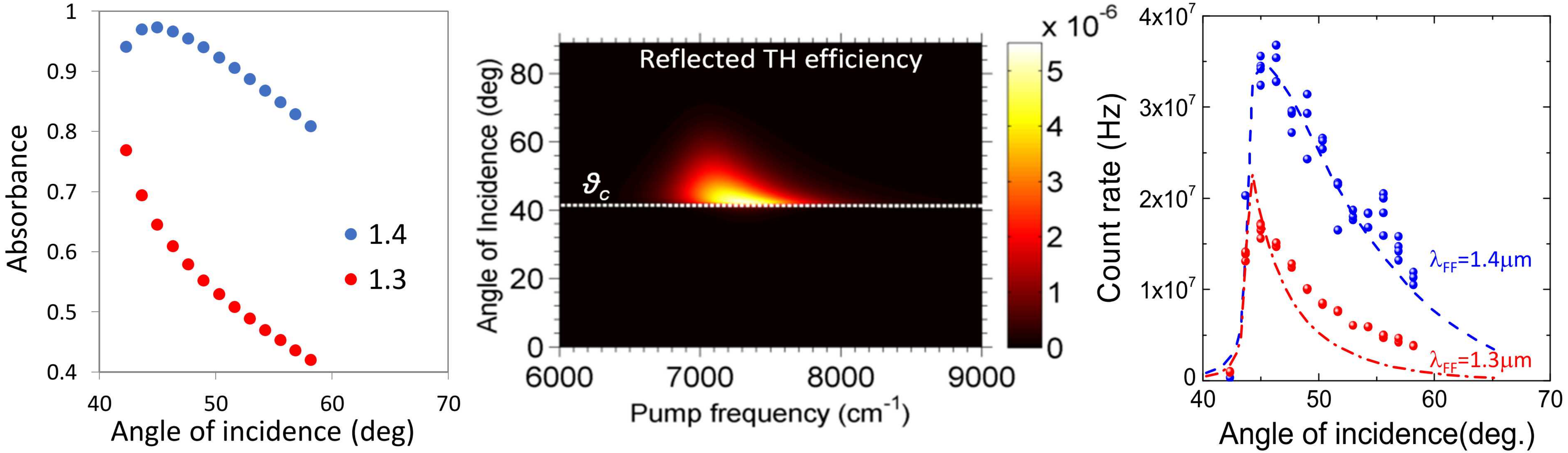
TH emerges in specular direction: $\frac{k_{FF//}}{k_{FF\perp}} = \frac{k_{TH//}}{k_{TH\perp}}$

assume $\varepsilon_{ambient}(\omega) = \varepsilon_{ambient}(3\omega)$





Absorption is not always bad



Use measured conversion efficiency at $2\text{x}10^{10}$ W/cm^2 and obtained ~$3.3\text{x}10^{-6}$

$\chi_{zzzz}^{(3)}$ = $3\text{x}10^{-21}$ m^2/V^2

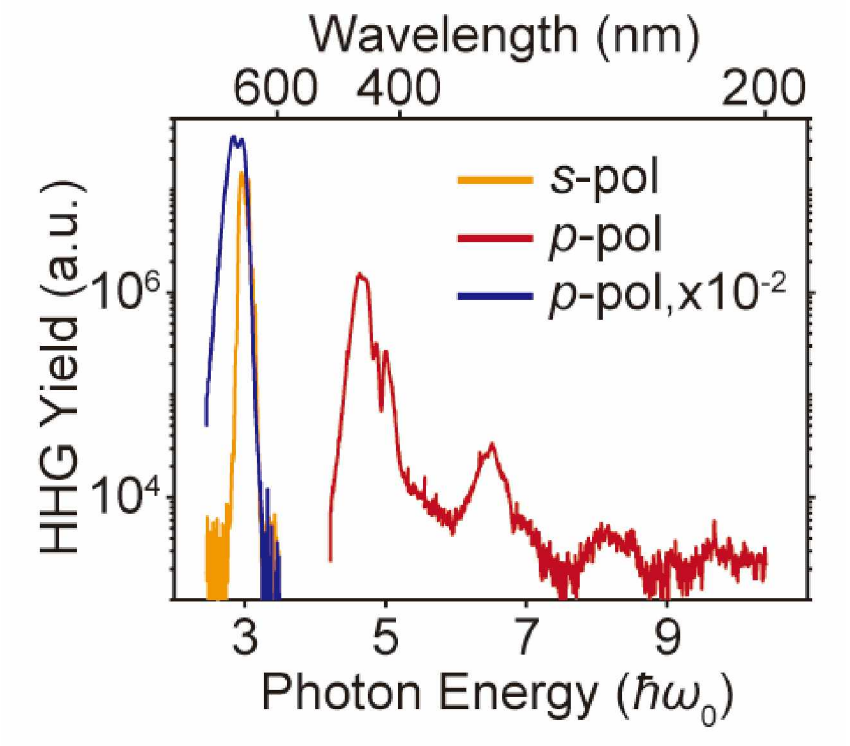
N. Ueda, et. al., APL (1991) – ITO film - $2.8\text{x}10^{-21}$ m^2/V^2

O. Reshef, R. W. Boyd, et. al., and, Optics Letters (2017). ITO film - $1.6\text{x}110^{-18}$ m^2/V^2.

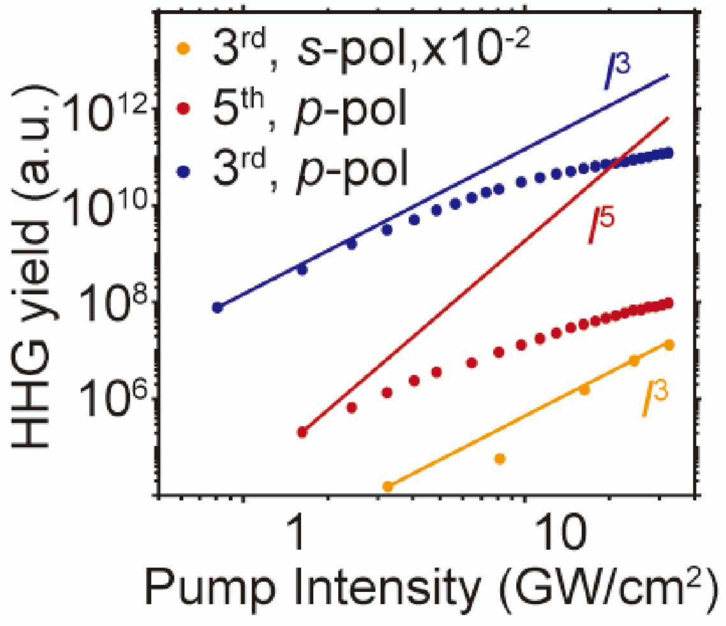
G. Yang, Z. Chen, et. al., Optical Materials (2004), Ag film – $3.5\text{x}10^{-16}$ m^2/V^2

T. S. Luk et. al, “Enhanced third harmonic generation from epsilon-near-zero modes of ultrathin films” *APL, 151103(*2015)

HHG Spectrum



HHG from 3rd to 9th order



Nonperturbative HHG

Introduction ➤ Ultrafast Switch ➤ High-harmonic Generation

Summary

- Free electrons are very polarizable and dispersive
- Exploited these properties to:
  enhanced absorber
  amplitude modulator
  Transient reflectance and polarization modulator
  harmonic generation

Acknowledgements

Salvatore Campione *(SNL)*
Michael B. Sinclair *(SNL)*
Michael Wood *(SNL)*
Gordon Keeler *(ex-SNL)*
Igal Brener *(CINT, SNL)*
Yuanmu Yang *(ex-SNL)*
Iltai Kim *(ex-SNL)*
Y. C. Jun *(ex-SNL)*
Shanhui Fan *(Stanford)*
S. Feng *(ex-Navy Research Lab)*
Domenico de Ceglia *(ex-National Research Council-AMRDEC)*
Maria A. Vincenti *(ex-National Research Council-AMRDEC)*
Michael Scalora (Charles M. Bowden Research Laboratory, AMRDEC)
Kyle Kelley *(North Carolina State University)*
Jon-Paul Maria *(North Carolina State University)*

ITO perfect absorption, Polarization tuning, THG (Salvatore Campione, Michael B. Sinclair)

Si waveguide modulator (Michael Wood, Gordon Keeler)

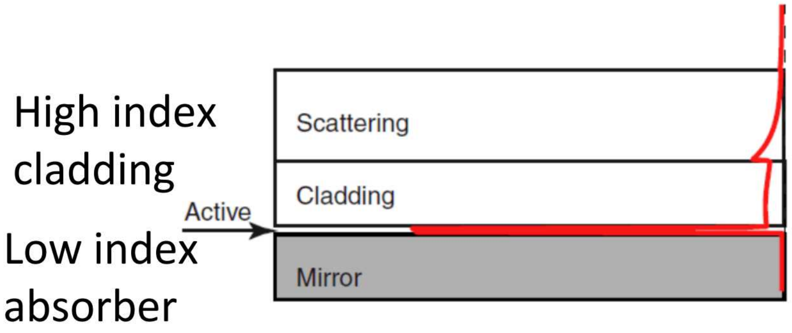
Polarization tuning using CdO (Igal Brener, Yuanmu Yang)

ITO perfect absorption (Iltai Kim, Y. C. Jun, Shanhui Fan, S. Feng)

THG theory (Domenico de Ceglia, Maria A. Vincenti, Michael Scalora)

CdO samples (Michael Scalora, Kyle Kelley, Jon-Paul Maria)

Mode coupling to subwavelength layer of low index material



Total absorption coefficient A_T

$$A_T = \frac{2\pi\gamma_i}{\Delta\omega} \frac{\#\text{ of modes available in the absorber}}{\#\text{ of input channels}}$$

$$\#\text{ of modes available} = 2\left(\frac{2\pi n_{wg}^2 \omega\delta\omega}{c^2}\right)\left(\frac{L}{2\pi}\right)^2$$

$$\#\text{ of input channels} = 2\left(\frac{\pi\omega^2}{c^2}\right)\left(\frac{L}{2\pi}\right)^2$$

Absorption enhancement $F = \left(\frac{A_T}{\alpha_0 d}\right) = 2n_{wg}\left(\frac{\lambda}{d}\right)\left(\frac{\alpha_{wg}}{\alpha_0}\right)$

For ITO case: n_{wg} = ~7, $\left(\frac{\lambda}{d}\right) \cong 50$ $F \cong 350\frac{\alpha_{wg}}{\alpha_0}$

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

Free electrons equation of motion (hydrodynamic model)

Convective: ***SHG and THG***

Coulomb: ***SHG and THG***

$$\ddot{\mathbf{P}}_f + \gamma_f \dot{\mathbf{P}}_f = \frac{n_{0f} e^2}{m_0^*}\mathbf{E} - \frac{1}{n_{0f} e}\left[\left(\nabla\cdot\dot{\mathbf{P}}_f\right)\dot{\mathbf{P}}_f + \left(\dot{\mathbf{P}}_f\cdot\nabla\right)\dot{\mathbf{P}}_f\right] - \frac{e}{m_0^*}\mathbf{E}\left(\nabla\cdot\mathbf{P}_f\right)$$

$$+\frac{\mu_0 e}{m_0^*}\dot{\mathbf{P}}_f\times\mathbf{H} + \frac{5}{3}\frac{E_F}{m_0^*}\nabla\left(\nabla\cdot\mathbf{P}_f\right) - \frac{10}{9}\frac{E_F}{m_0^*}\frac{1}{n_{0f} e}\left(\nabla\cdot\mathbf{P}_f\right)\nabla\left(\nabla\cdot\mathbf{P}_f\right)$$

Drude oscillator

Usually negligible

Lorentz: ***SHG and THG***

Nonlocal gas pressure

Backup slide 1: Susceptibility, Polarization and Dielectric functions

Polarization in real space:

$$P_i(\vec{r}\,', t') = \int \underbrace{\chi_{ij}(\vec{r}, \vec{r}\,', t, t')}_{\text{Susceptibility tensor}} E_j(\vec{r}, t)\, d\vec{r}\, dt$$

Polarization in momentum space:

$$P_i(q,\omega) = \varepsilon_0 \chi_{ij}(q,\omega) E_j(q,\omega)$$

Dielectric tensor:

$$\varepsilon_{ij}(q,\omega) = 1 + \chi_{ij}(q,\omega)$$

(SI units)

Dielectric function is a real function of space and time: $\varepsilon(-q,-\omega) = \varepsilon^*(q,\omega)$

Onsager relations: $\varepsilon_{ij}(q,\omega) = \varepsilon_{ji}(-q,\omega)$

Prospect of using ENZ material for nonlinear optical processes

$$\varepsilon_{NL}(E) = \varepsilon + 3\chi^{(3)}E^2 + \ldots$$

For ENZ material, real part is zero

This becomes the dominant term

$$n_{NL}(E) = \sqrt{\varepsilon + 3\chi^{(3)}E^2}$$

Origin of $\chi^{(3)}$… - Free electrons equation of motion (hydrodynamic model)

*Convective: **SHG and THG***

*Coulomb: **SHG and THG***

$$\ddot{\mathbf{P}}_f + \gamma_f \dot{\mathbf{P}}_f = \frac{n_{0f}e^2}{m_0^*}\mathbf{E} - \frac{1}{n_{0f}e}\left[\left(\nabla\cdot\dot{\mathbf{P}}_f\right)\dot{\mathbf{P}}_f + \left(\dot{\mathbf{P}}_f\cdot\nabla\right)\dot{\mathbf{P}}_f\right] - \frac{e}{m_0^*}\mathbf{E}\left(\nabla\cdot\mathbf{P}_f\right)$$

$$+ \frac{\mu_0 e}{m_0^*}\dot{\mathbf{P}}_f\times\mathbf{H} + \frac{5}{3}\frac{E_F}{m_0^*}\nabla\left(\nabla\cdot\mathbf{P}_f\right) - \frac{10}{9}\frac{E_F}{m_0^*}\frac{1}{n_{0f}e}\left(\nabla\cdot\mathbf{P}_f\right)\nabla\left(\nabla\cdot\mathbf{P}_f\right)$$

Drude oscillator

Usually negligible

*Lorentz: **SHG and THG***

Nonlocal gas pressure

N. Bloembergen, and Y. R. Shen, "Optical Nonlinearities of a Plasma," Physical Review (1966)

J. E. Sipe, V. C. Y. So, M. Fukui, and G. I. Stegeman, "Analysis of second-harmonic generation at metal surfaces," PRB (1980).

M. Scalora, M. A. Vincenti, D. de Ceglia, … and J. W. Haus, "Dynamical model of harmonic generation in centrosymmetric semiconductors at visible and UV wavelengths," PRA (2012).