

# Plasmonics and Optical Metamaterials: Looking beyond Gold and Silver

*EXPLORING AND UTILIZING  
ALTERNATIVE  
CONSTITUENT MATERIALS*

***Alexandra Boltasseva***

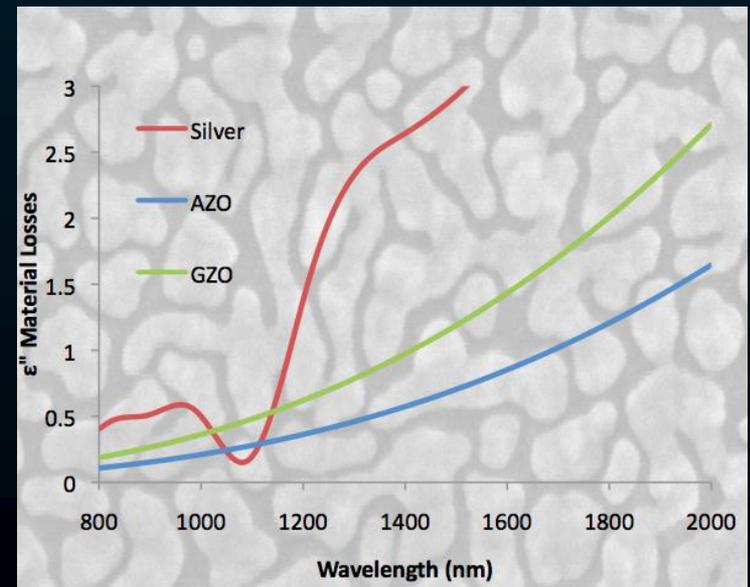
*School of Electrical & Computer Engineering*

*Birck Nanotechnology Center*

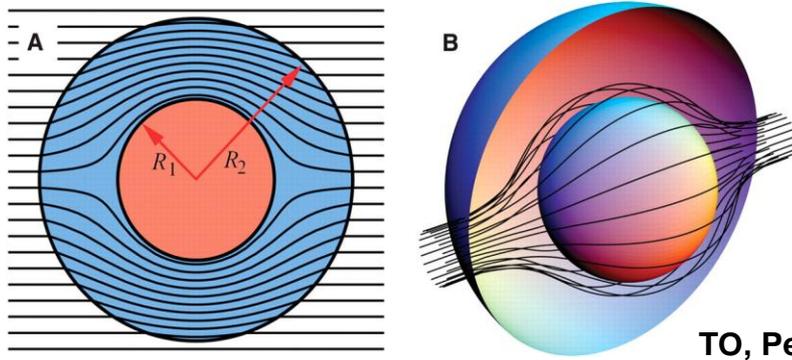
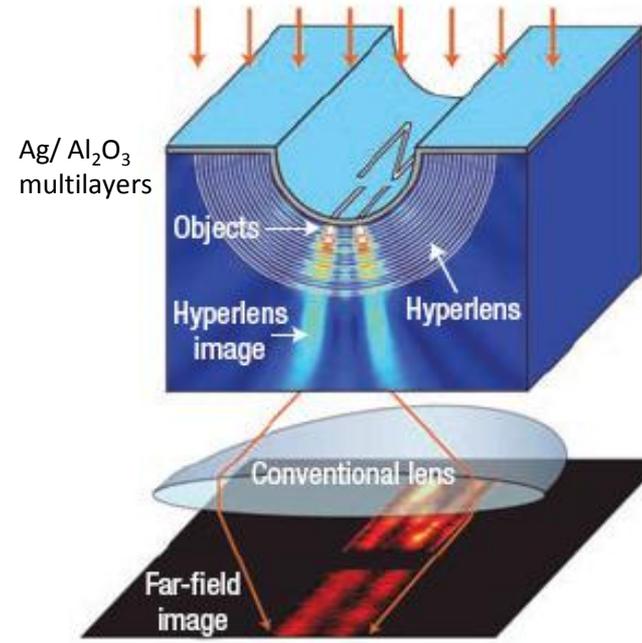
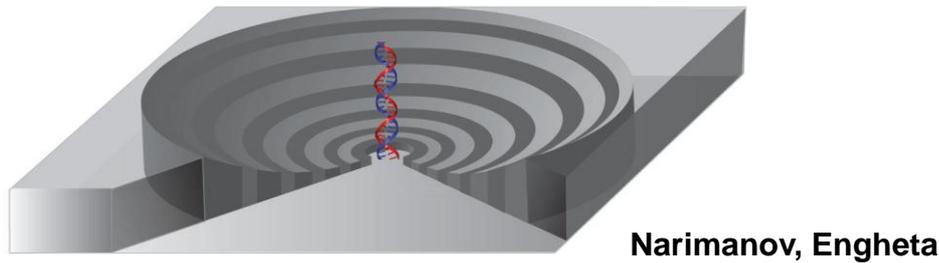
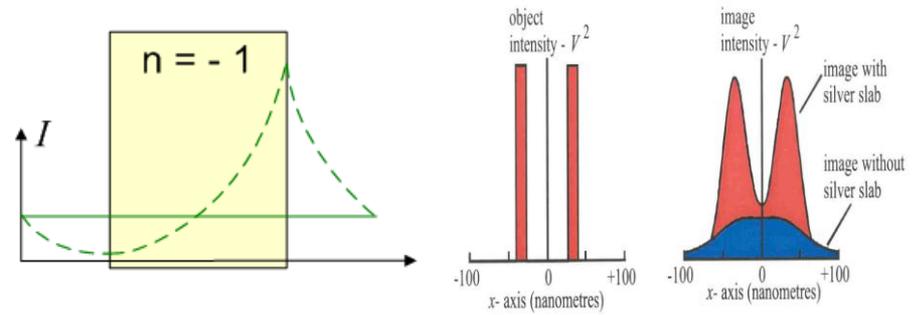
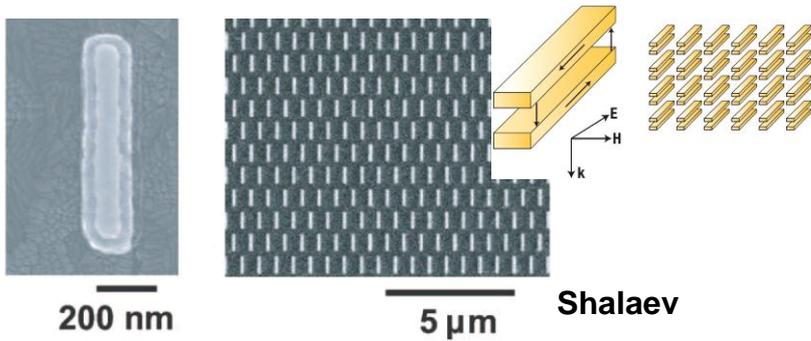
*Purdue University*

# OUTLINE

- Introduction: Challenges with Gold and Silver
- Material Requirements: Plasmonics & Metamaterials
- Alternative Materials?
- Transparent Conducting Oxides
- Transition Metal Nitrides
- Figures of Merit
- What is the Right Choice?
- Outlook



# METAMATERIALS/TRANSFORMATION OPTICS



Z. Liu et. al, Science (2007)

# CHALLENGES

## Conventional plasmonics: Gold and Silver

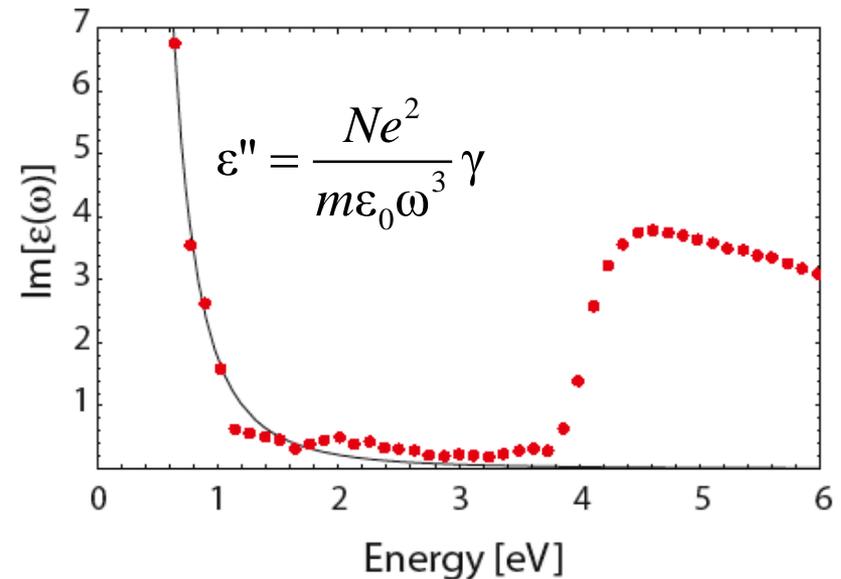
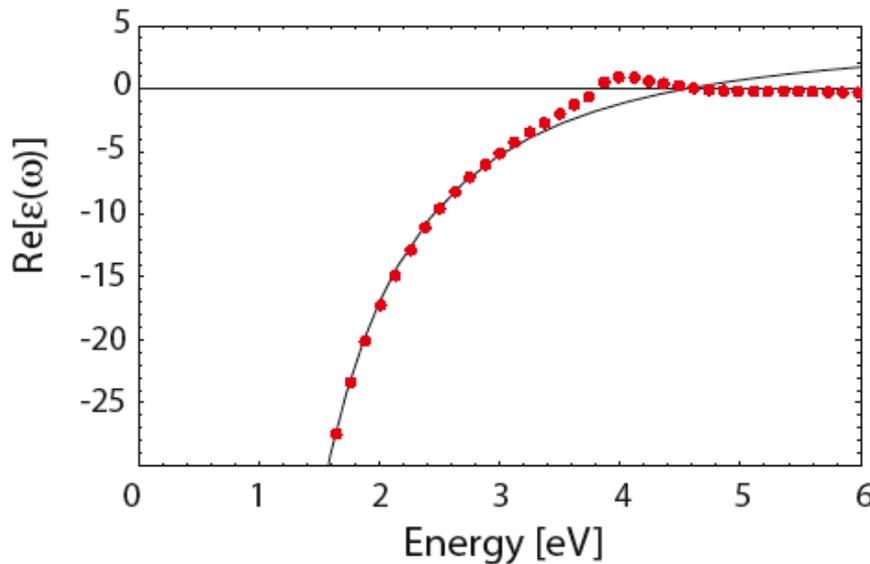
- Large losses in near-IR and visible ranges

Interband transitions

Surface roughness, grain boundaries, etc...

$$\epsilon(\omega) = \epsilon_{ib} - \frac{Ne^2 / m\epsilon_0}{\omega^2 + i\omega\gamma_m(\omega)}$$

Real and Imaginary  $\epsilon(\omega)$  for Silver (Ag)



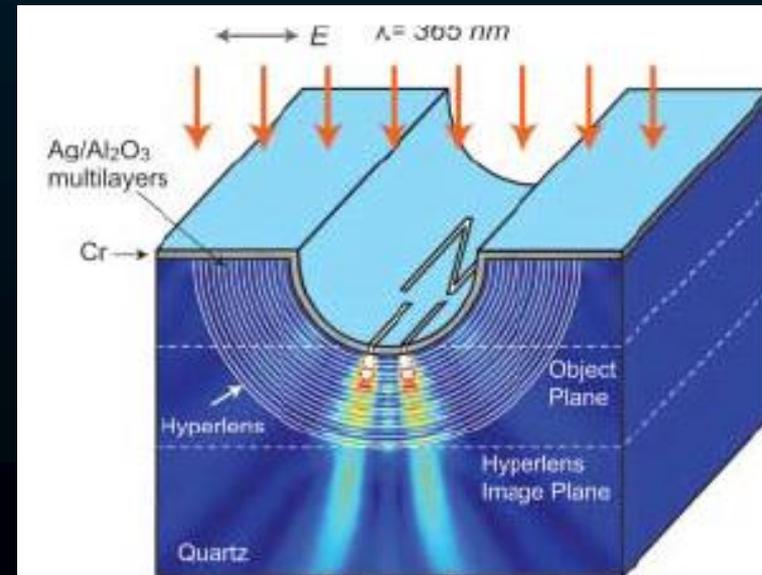
# MATERIAL REQUIREMENTS

Emerging innovative fields such as Transformational Optics require **comparable magnitudes of  $\epsilon'$**  of metal and dielectric and dielectric

- Epsilon-near-zero (ENZ) materials
- Effective permittivity nearly zero: e.g. optical cloaks, hyperlens etc.

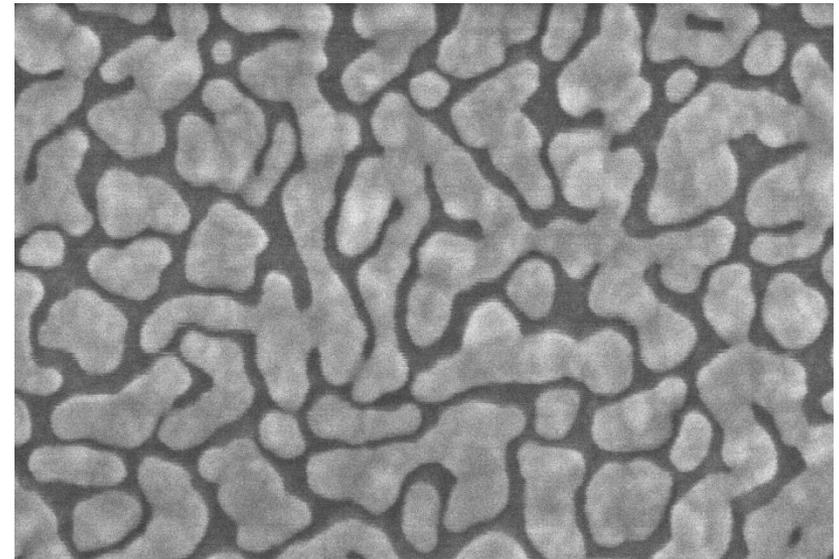
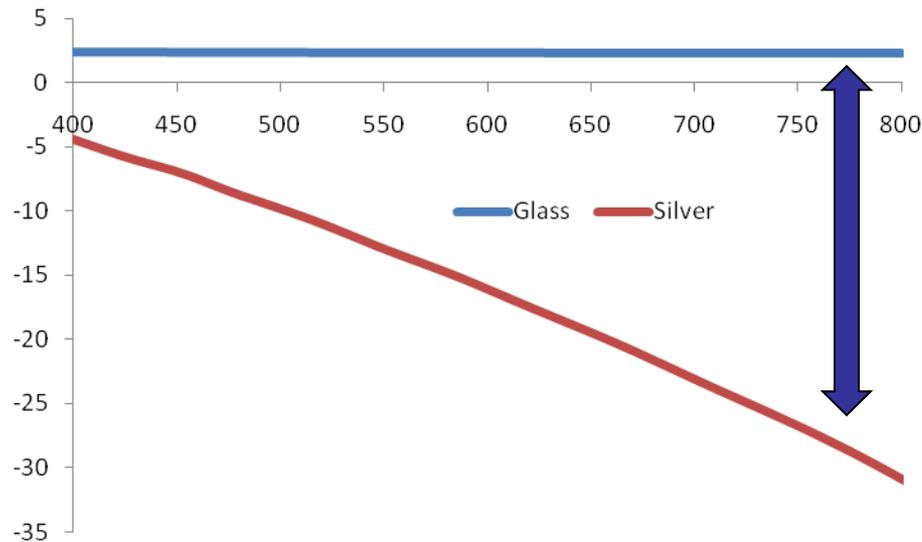
Novel devices would require **tunable  $\epsilon'$**

- Switching and modulation capabilities



# MATERIALS FOR TO / ENZ

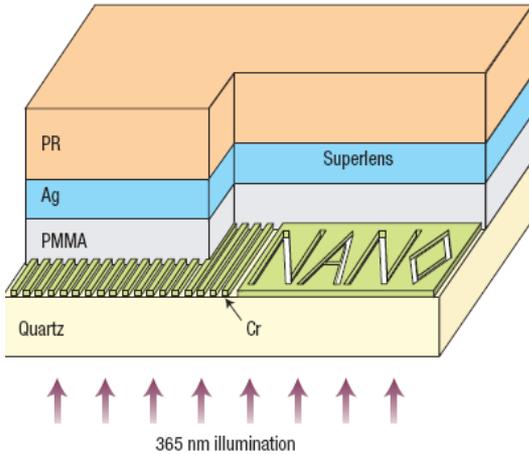
- Effective permittivity nearly zero  $\epsilon_{\text{effective}} \sim 0$ : cloaks, hyperlens etc.
- Real permittivity of metals must be comparable to that of dielectrics (for example,  $\epsilon_{\text{dielectric}} \sim 2$  requires  $\text{Re}(\epsilon_{\text{plasmonic material}}) \sim -2$  while  $\text{Re}(\epsilon_{\text{Ag}}) \ll -2...$ )



Ag: threshold for uniform continuous films is around 12-23 nm

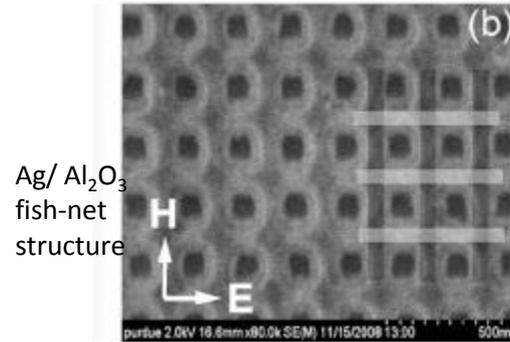
# BEYOND GOLD AND SILVER?

## Superlens



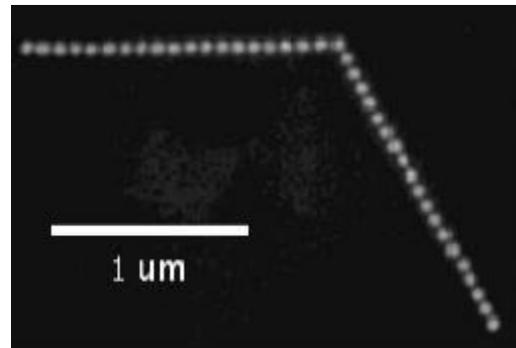
N. Fang et. al, Science (2005)

## 2-D Negative refractive index materials (NIM)



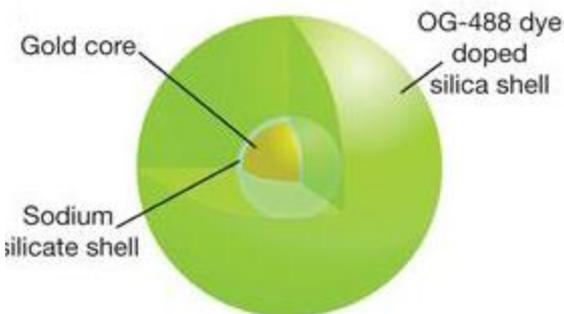
U. K. Chettiar et. al, MRS Bulletin (2008)

## Plasmonic nanoparticle waveguide



M.L. Brongersma, et al., Phys. Rev. B (2000)

## Nanolaser

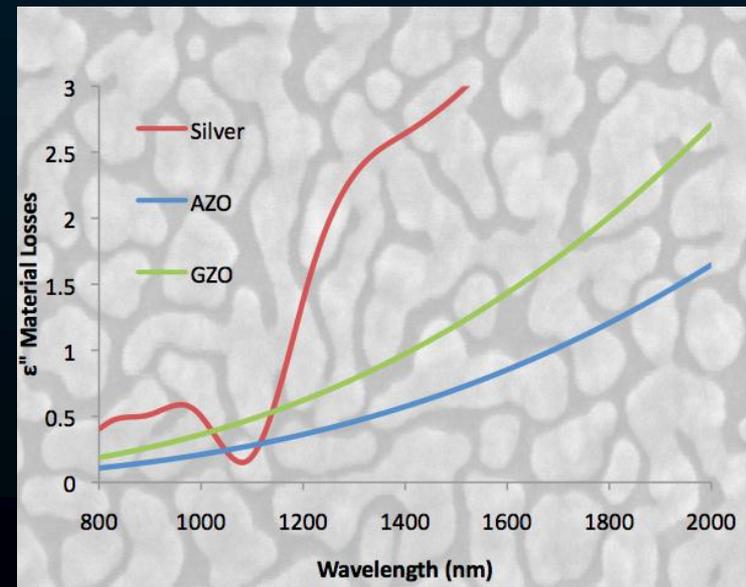


M.A Noginov et. al, Nature (2010)

- Too large magnitude of real permittivity
- Large losses (VIS/NIR)
- Optical properties are not tunable
- Fabrication of very thin films/nanostructures is difficult
- Nanopatterning increases losses, grain boundaries, surface roughness...
- **Not CMOS compatible**

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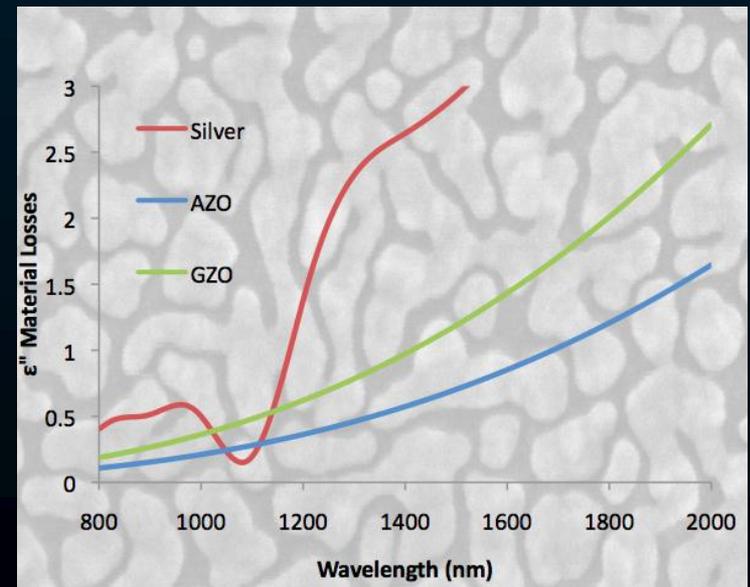


# MATERIAL REQUIREMENTS

- **Low loss** components
  - Dielectrics can be nearly loss-less
  - Metals have large losses
- **Adjustable / Tunable** optical properties  
Some Metamaterial + TO designs require **comparable magnitudes of  $\epsilon'$**  of metal and dielectric
  - Epsilon-near-zero (ENZ) materials
  - Effective permittivity nearly zero: e.g. optical cloaks, hyperlens etc.
- **Switchable** devices
- **SC-compatible** components

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# POSSIBILITIES

- **Metals** (Ag, Au, Cu, Al, Alkali)
- **Alloys** (Noble-Alkali<sup>1</sup>, alloys of noble metals with transition metals like Cadmium and Zinc<sup>2</sup>)
- **Doped Semiconductors:** Highly doped semiconductors<sup>3</sup>, doped conducting oxides (ITO<sup>4</sup>, Al:ZnO and Ga:ZnO<sup>5</sup>)
- **Intermetallics** (nitrides, germanides, ...)

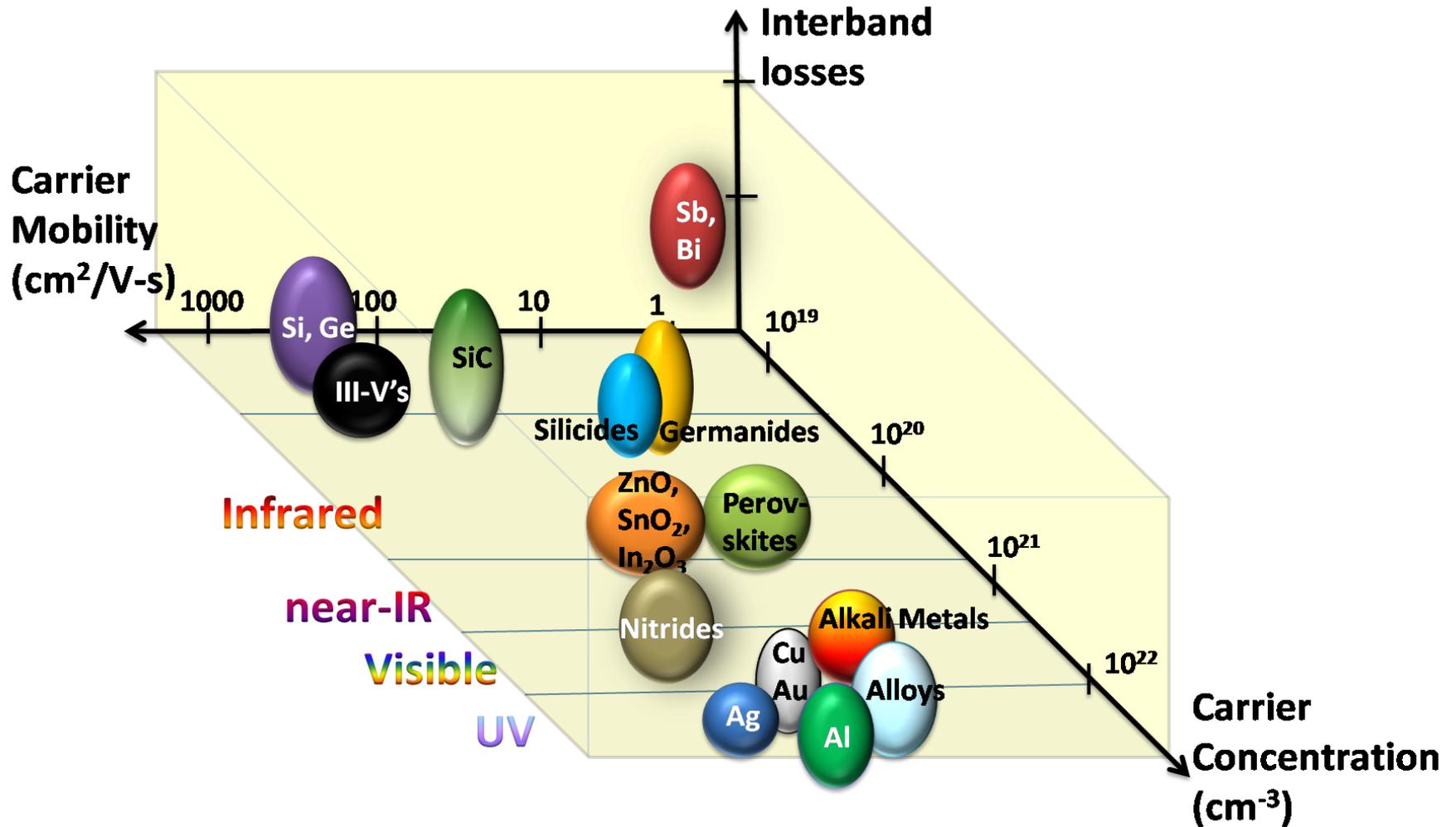
1:M. G. Blaber et al., J.Phys. Cond. Matter, 21, 144211 (2009),

2:D. A. Bobb et al., Appl. Phys. Lett. 95, 151102 (2009); 3:A. J .Hoffman et al., Nature Mater., 6, 946 (2007)

4:C. Rhodes et al., J. Appl. Phys., vol. 100, 54905 (2008)

5: LPR 4, 795 (2010), Phys. Status Solidi RRL 4, 295 (2010), Metamaterials 5, 1 (2011), OMEEx (2011)

# ALTERNATIVE MATERIALS



# METALS

## Ag – Conventional Plasmonics, usual choice

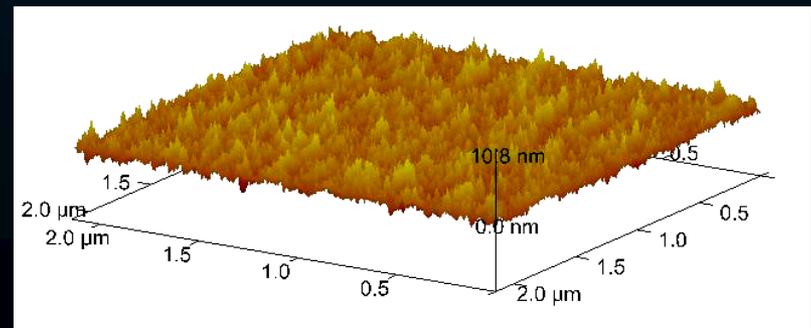
- Low loss
- Standard physical vapor deposition (PVD) methods + chemical methods
- But *degrades* in air

## Au – Second Best for VIS, NIR

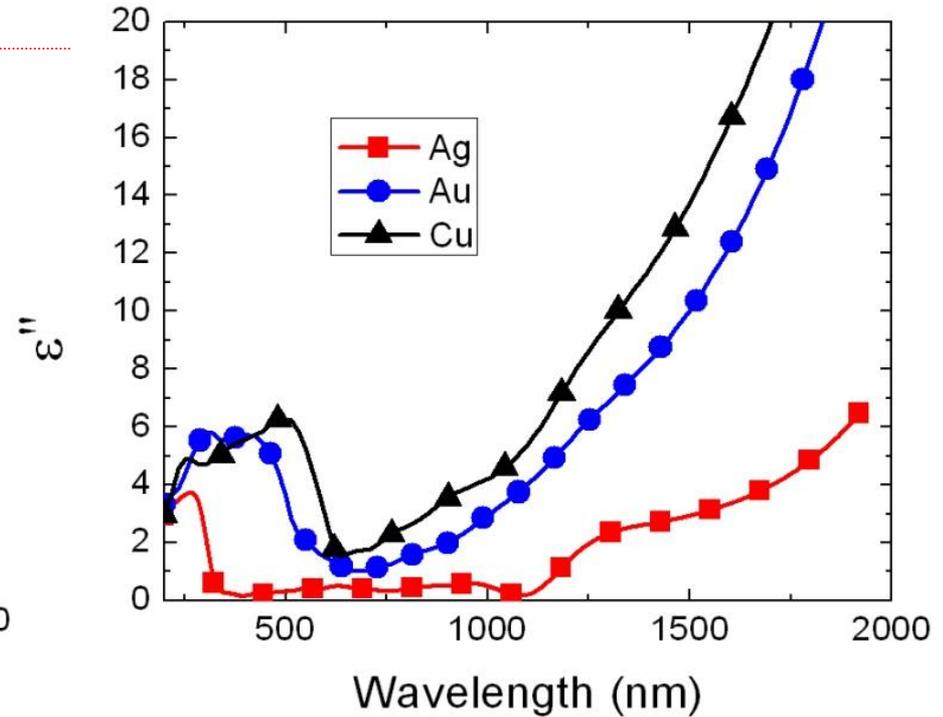
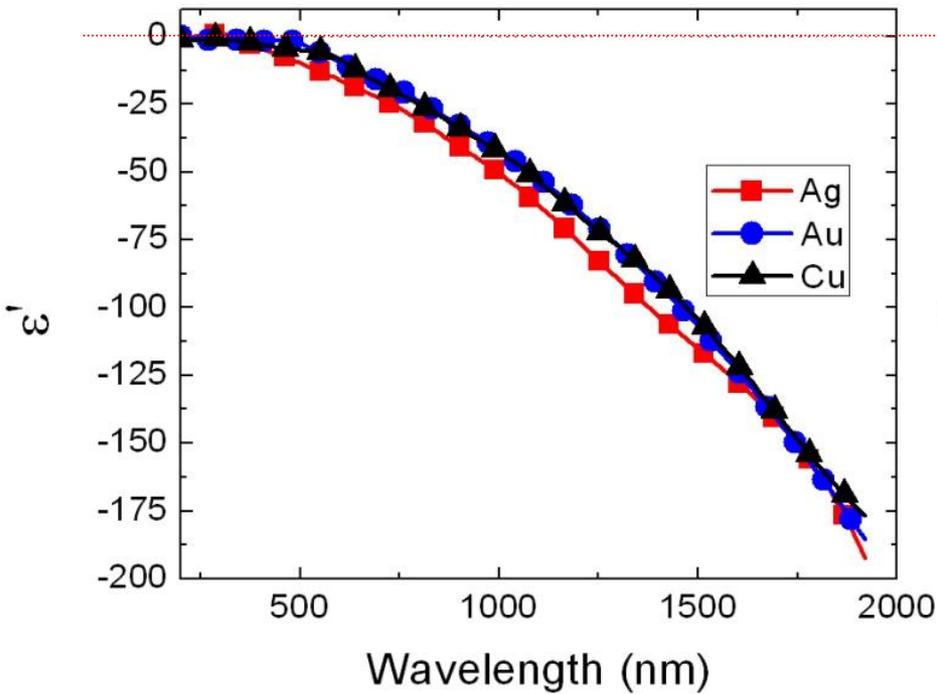
- Acceptable loss but *interband transition* (5d-6p) within VIS range
- Standard PVD methods + chemical methods: Chemically stable
- Continuous film at thickness of 2-7nm

## Cu – Ok for VIS (similar to Au)

- High conductivity + low cost
- But prone to surface *oxidation*



# Ag, Au, Cu

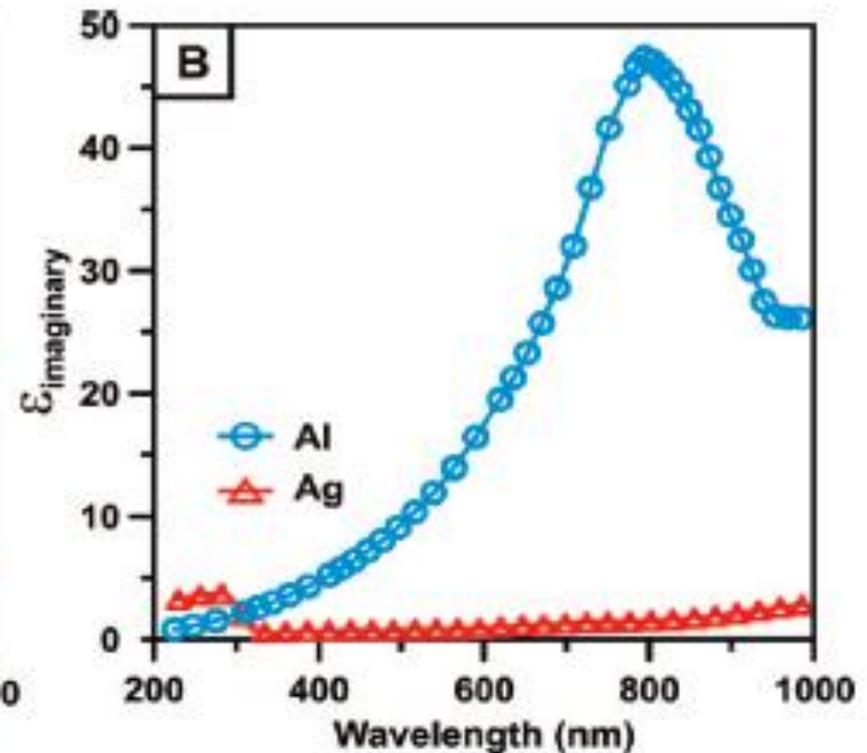
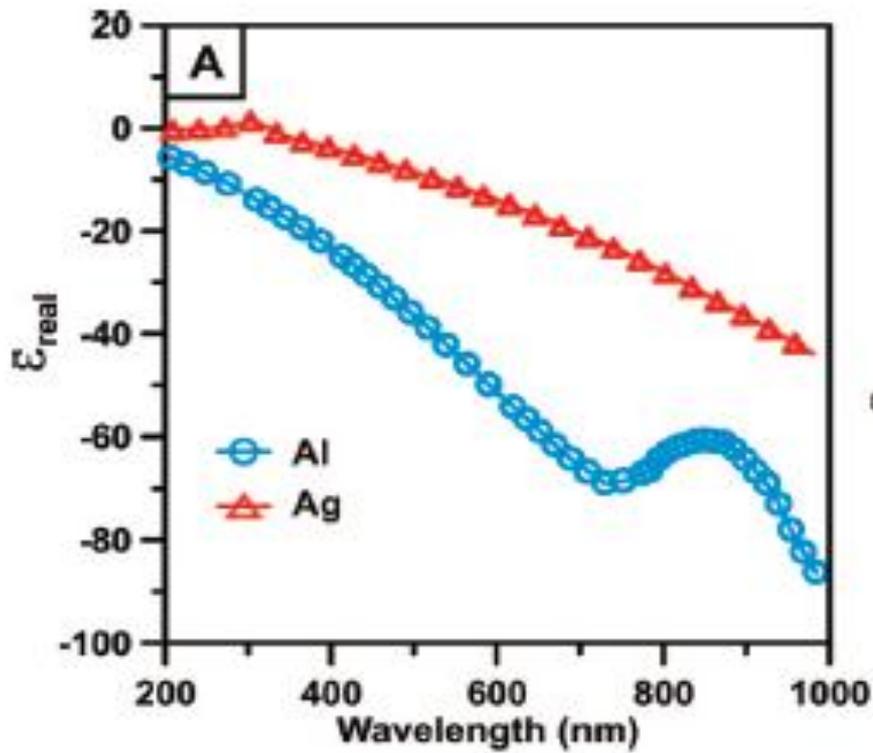


Cu – similar to Au 600-750 nm  
fabrication is challenging (easily oxidizes)

# ALUMINUM

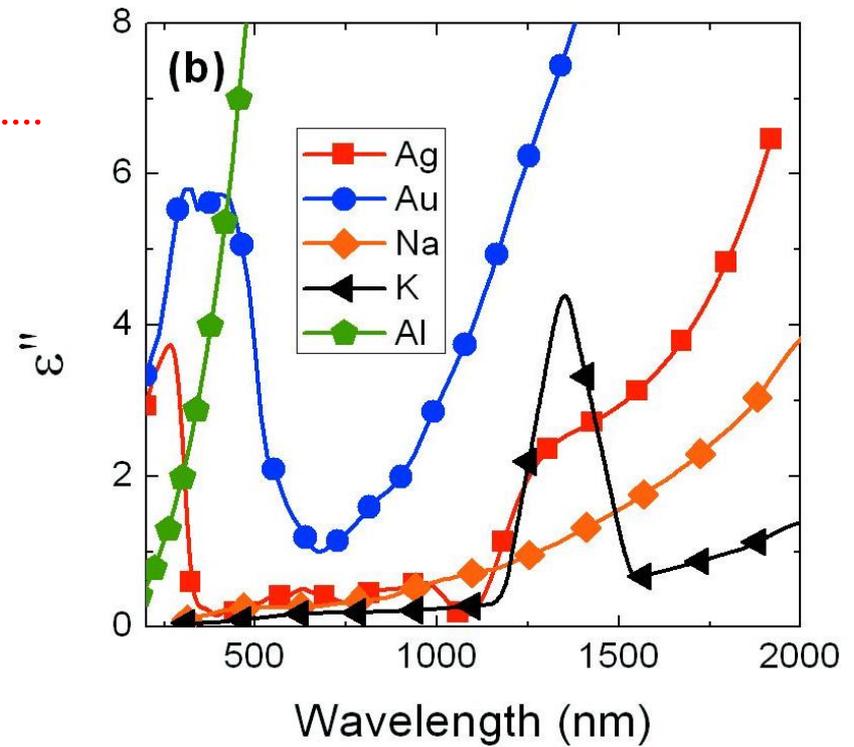
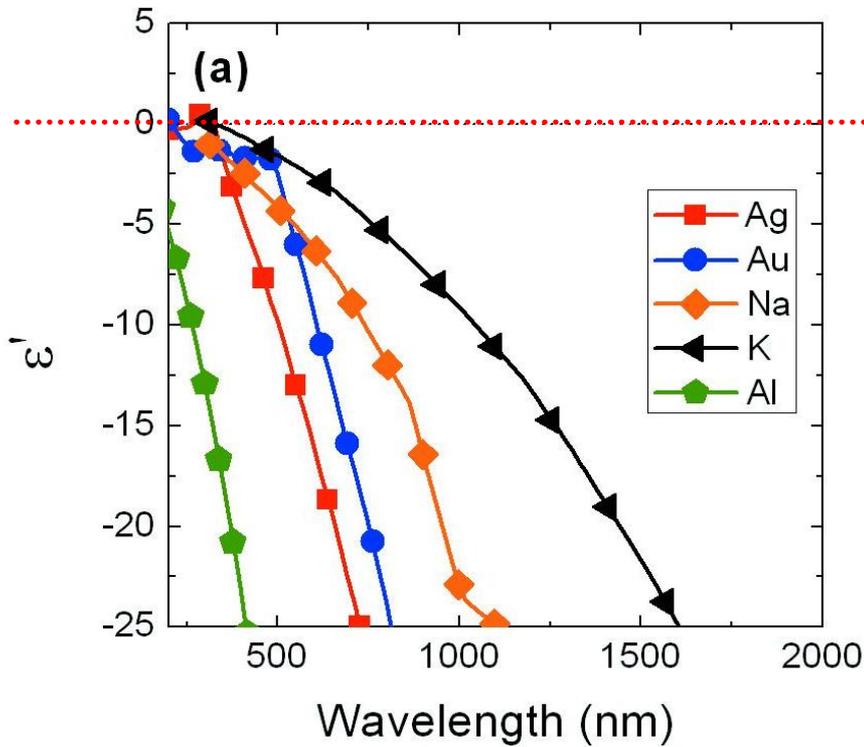
Al – Higher loss in VIS + NIR

- Best for short wavelengths (still plasmonic below 200 nm)
- Prone to surface *oxidation* ( $\text{Al}_2\text{O}_3$  2.5~3nm)



# ALKALI

Alkali (Sodium, Potassium) – Lowest losses, closest to free-electron gas  
 - Very *reactive* (ultra-high vacuum  $10^{-19}$  Torr requirement, passivation)



# ALLOYS: IMPROVING METALS

## Improving Noble Metals:

- To shift interband transitions to another (unimportant) part of the spectrum
- By alloying two or more elements to create unique band structures that can be fine-tuned by adjusting the proportion of each alloyed material

## Noble-Transition Metal Alloys

Bivalent transition metals (Cadmium and Zinc) contribute one extra electron to the free-electron plasma n-type doping  $\Rightarrow$

- Increasing of  $\omega_p$
- Shifting the threshold for interband transitions
- Reducing the absorption at a specific wavelength

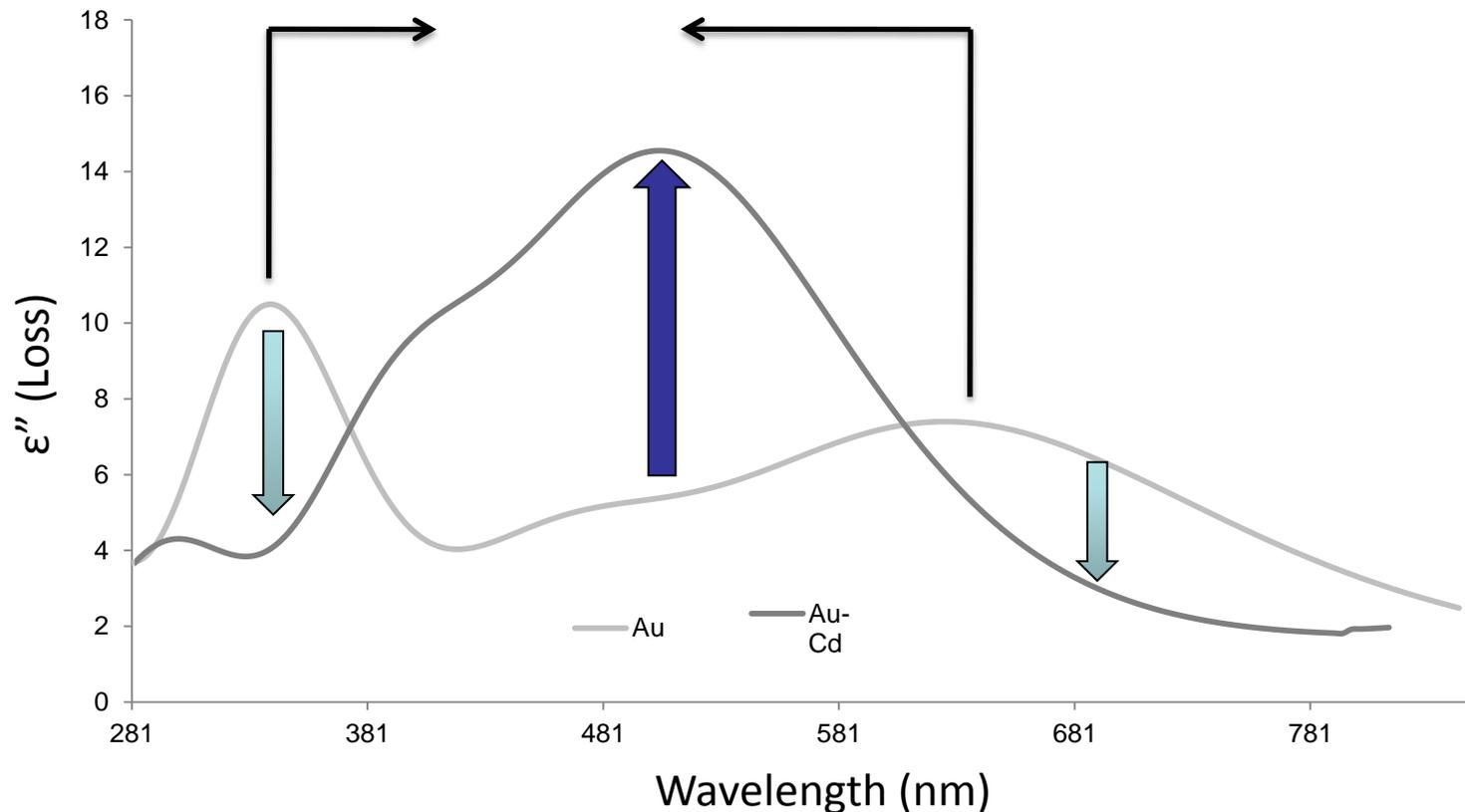
## “Band Engineering”

# NOBLE-TRANSITION ALLOY

Cadmium + Gold  $\Rightarrow$  Additional electron to free electron gas

Shift of the Lorentz resonance peaks  $\Rightarrow$  Tuning of the optical parameters

Optimum – 3.3% Cadmium in Gold

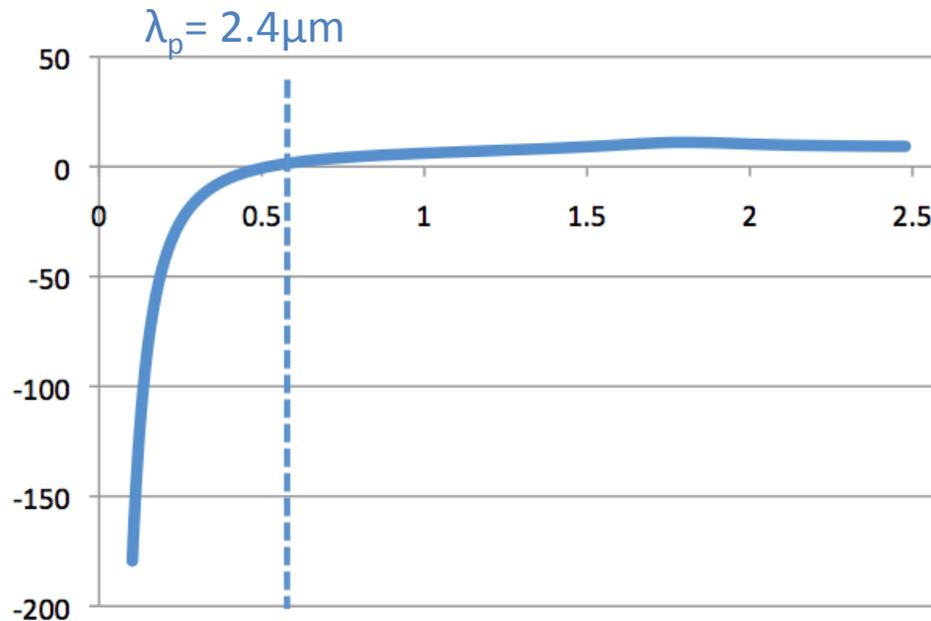


# ALKALI-NOBLE COMPOUNDS

## Alkali-Noble Metal: Intermetallic Compounds

Group I alkali metals: Strongest free-electron-like behavior

Most promise: Potassium Gold (KAu)



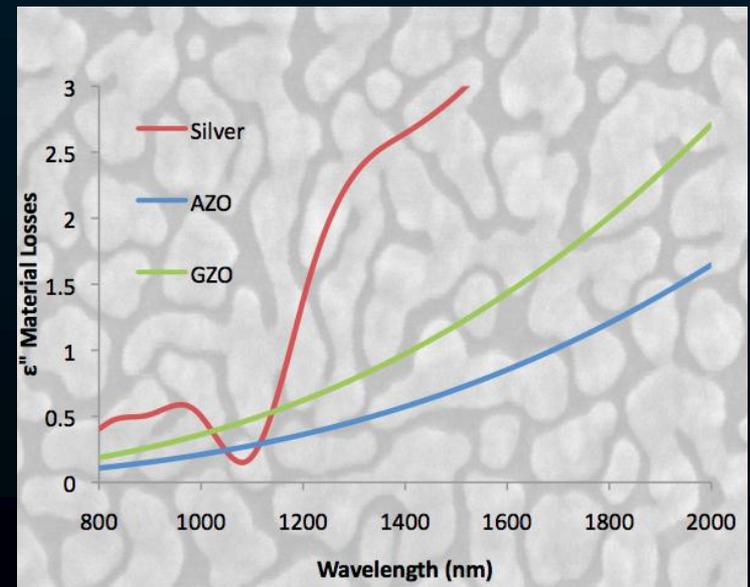
KAu:

Crossover point is 0.5 eV (2.4  $\mu\text{m}$ )

$\Rightarrow$  KAu does not have negative  $\epsilon'$  values in the NIR and VIS ranges

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# SEMICONDUCTORS AS “METALS”

- Metals: Too large carrier concentration
  - Large plasma frequency ( $\omega_p$ )
    - $\omega_p \propto \sqrt{n}$  :  $n \sim 10^{22} \text{ cm}^{-3}$  in metals
  - Large loss ( $\epsilon'' \propto \omega_p^2$ ) + large magnitude of  $\epsilon'$
- Semiconductors: **Doping** can control carrier concentration
  - Conventional semiconductors: too low carrier concentration (dielectrics)
  - Doping density of  $10^{21} \text{ cm}^{-3}$  could produce  $\epsilon' < 0$  in NIR

*METALS ARE TOO METALLIC...*

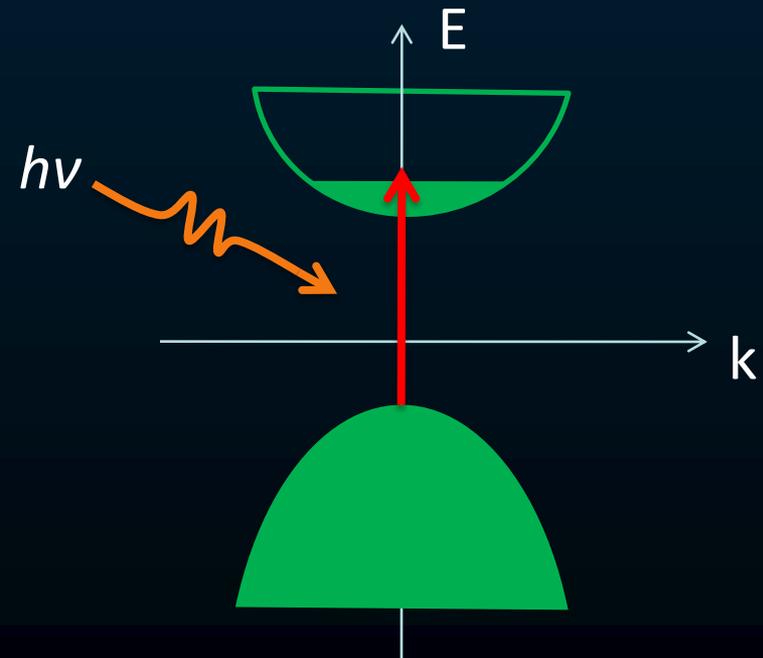
*MAKE SCs MORE METALLIC...*

# METALS AS “LESS-METALS”

- Lower carrier concentration in metals
  - Abstract electrons by non-metal inclusions
  - Non-stoichiometric: controllable properties

# SEMICONDUCTOR-BASED “METALS”

- Make semiconductors more metallic:  
Increase carrier concentration to  $10^{21} \text{ cm}^{-3}$
- Wide Bandgap Semiconductors:  
*Negligible interband transition losses*
- Bandgap should be larger than frequency of interest  
Material Bandgap (eV):  
Si - 1.12, GaAs - 1.42, SiC - 2.36-3.05
- Large carrier mobility:  
*Low damping losses*



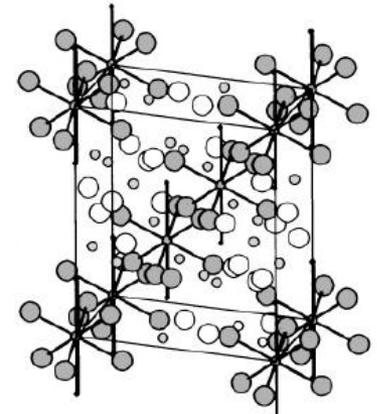
# SEMICONDUCTORS

## ◦ Semiconductors

- A lot of potential candidates like GaAs, GaN, Zn and Cd compounds (Oxides, Sulphides, Selenides, Tellurides),  $\text{In}_2\text{O}_3$ ,  $\text{SnO}_2$ , SiC, etc.
- Bandgap should be larger than frequency of interest (Material Bandgap (eV): Si - 1.12, GaAs - 1.42, SiC - 2.36-3.05)
- Negative  $\epsilon$  at IR: heavy doping or resonance (*Hoffmann et. al: Alternating layers of InGaAs and InAlAs; Doping  $7 \times 10^{18} \text{ cm}^{-3}$  to achieve negative epsilon at  $\sim 10 \mu\text{m}$* )

## ◦ Transparent Conducting Oxides

- Tune plasma frequency by doping
- Great switching opportunities
- **Indium Tin Oxide (ITO), Al doped Zinc Oxide (AZO) and Ga doped Zinc Oxide (GZO)**



Soliman et al.,  
Thin Solid Films, 502, 205 (2006)

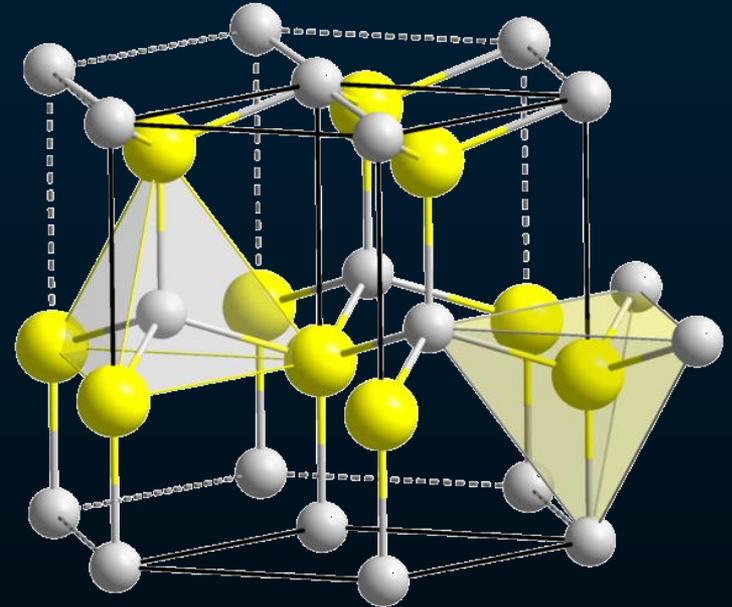
# SEMICONDUCTORS AS “METALS”

## *NIR*: Transparent Conducting Oxides

- **Tune plasma frequency by doping**
- **Great switching opportunities**
- Gallium doped zinc oxide (GZO)
- Aluminum doped zinc oxide (AZO)
- Tin doped indium oxide (ITO)

# ZINC OXIDE

- II-VI semiconductor
- Wide band-gap of 3.37 eV at 300 K
- Applications:
  - Display panels
  - Piezo-electric devices
  - Paints, anti-corrosive coatings
  - Bio-compatible devices



# DOPING ZnO

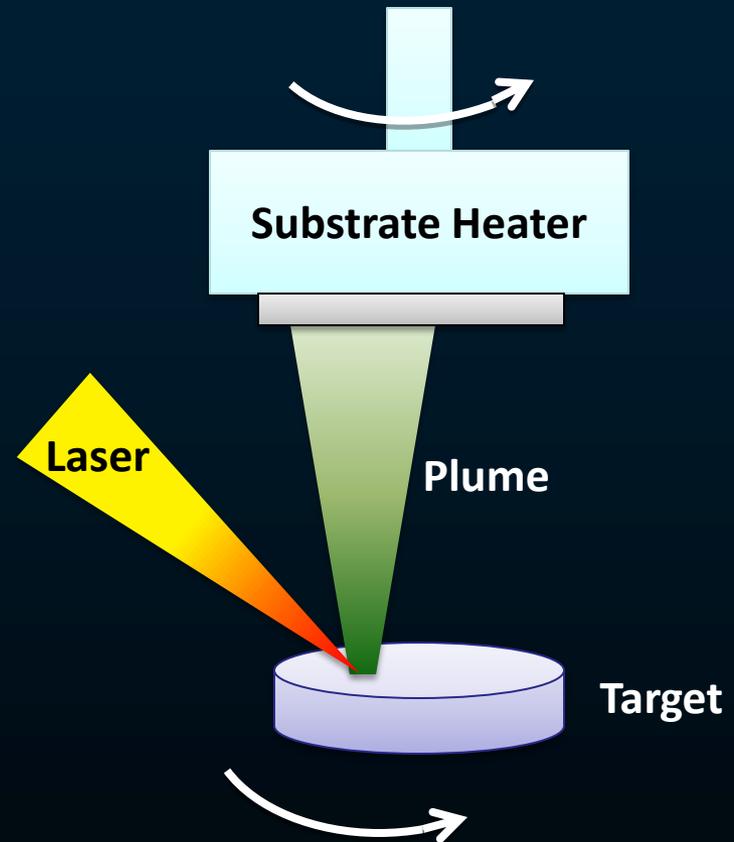
- By substituting a fraction of the  $\text{Zn}^{2+}$  ions by  $\text{Al}^{3+}$  ions, which serve as electron donors, ZnO is turned into a n-type semiconductor

## Ultra-High Doping Challenges

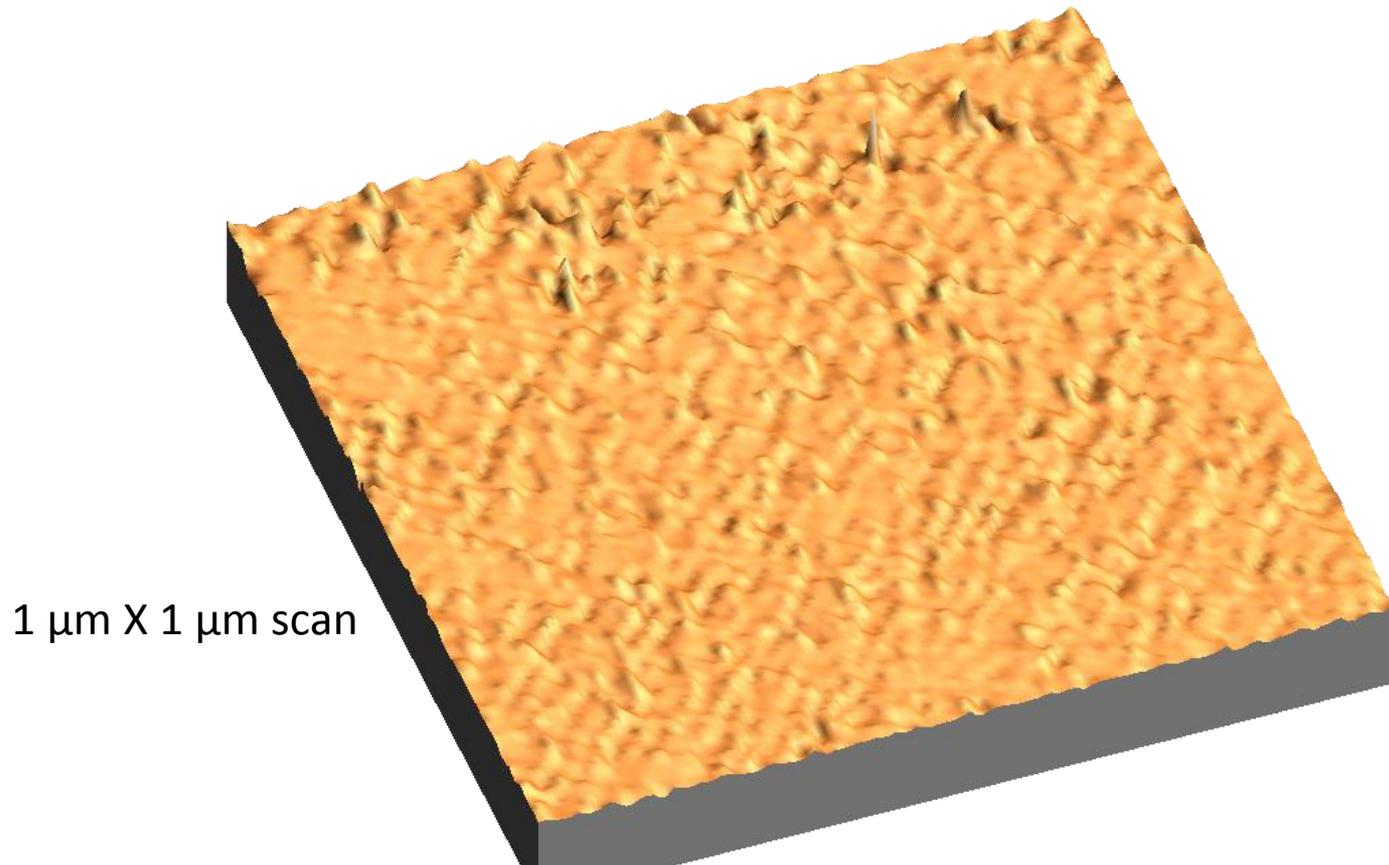
- Solid solubilities (maximum concentrations) of Ga and Al in ZnO are high (2-4 wt%)<sup>1</sup>
- High carrier concentration<sup>1</sup>  $\sim 10^{21} \text{ cm}^{-3}$  (close to solid solubility limit)
- Very heavy doping lowers mobility of carriers due to increased impurity scattering
- Properties depend largely on the deposition method and the deposition conditions

# PULSED LASER DEPOSITION (PLD)

- Pulsed LASER Deposition (PLD)
  - Excimer laser pulses ablate the target of desired material
  - Ablation of more than one targets can deposit material with mixed composition
- Optical Characterization: Ellipsometry
  - Spectroscopic ellipsometer (J.A. Woollam Co.)
  - Drude-Lorentz model to extract the dielectric function



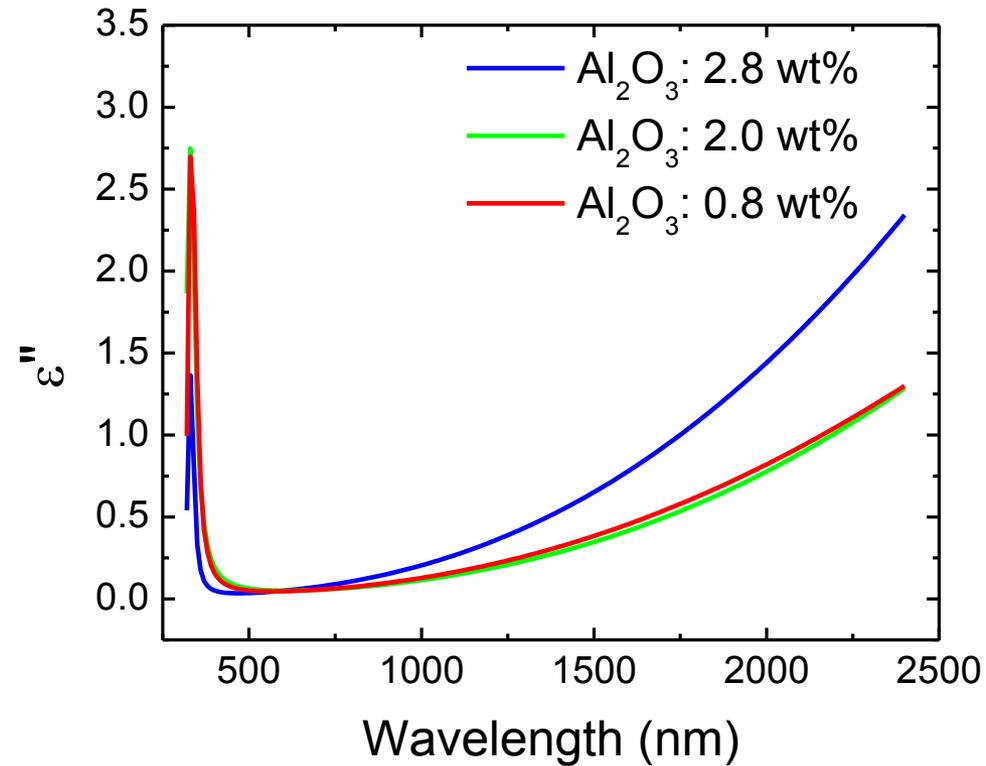
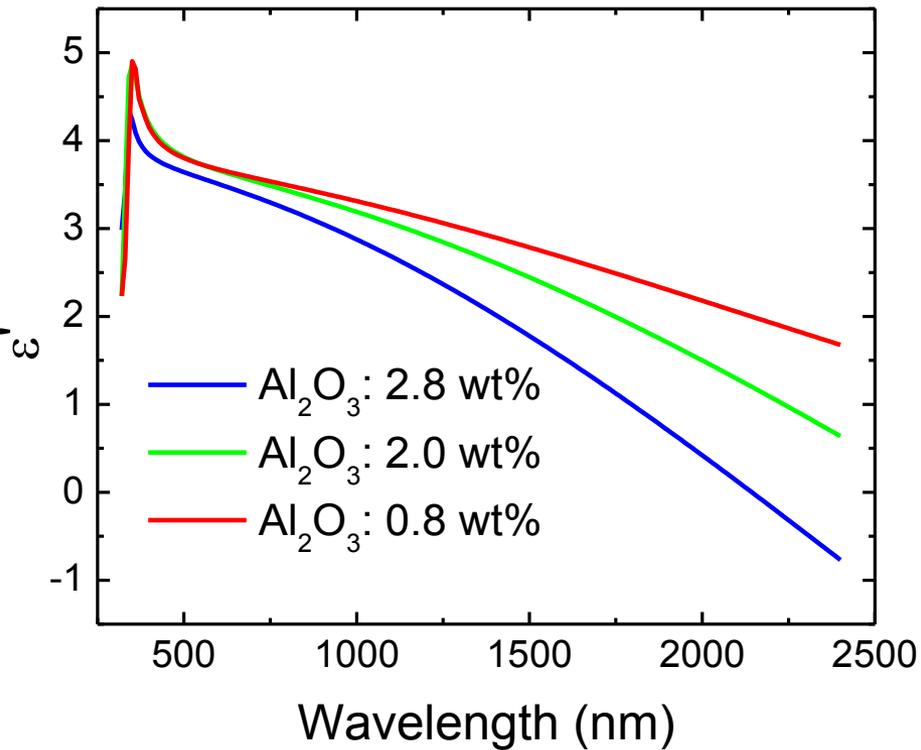
# AFM OF AZO FILMS



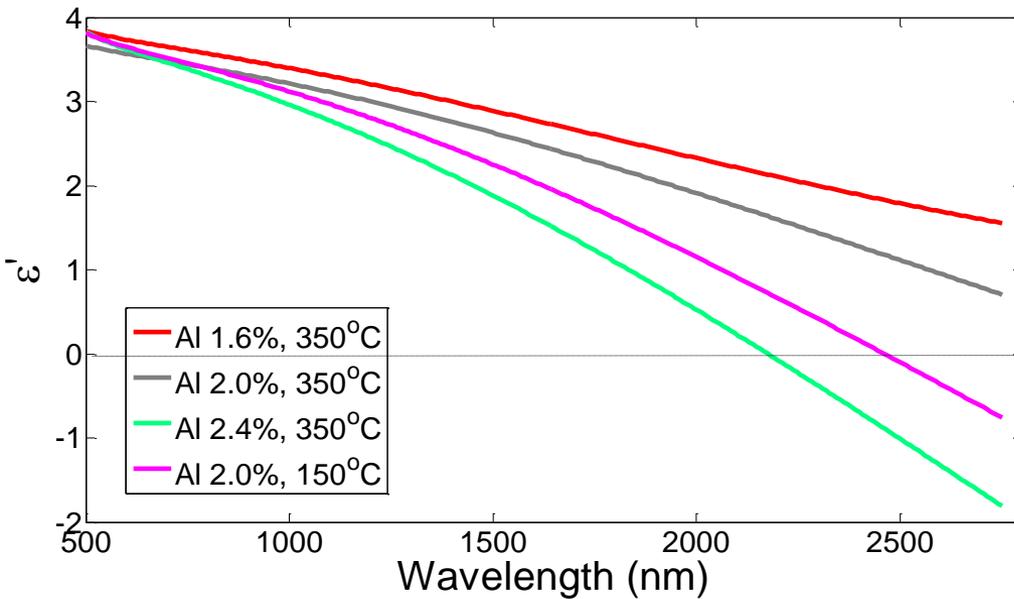
1 μm X 1 μm scan

Mean grain size: 8 nm; Roughness (rms): 1.3 nm; Thickness 62 nm

# AZO THIN FILMS



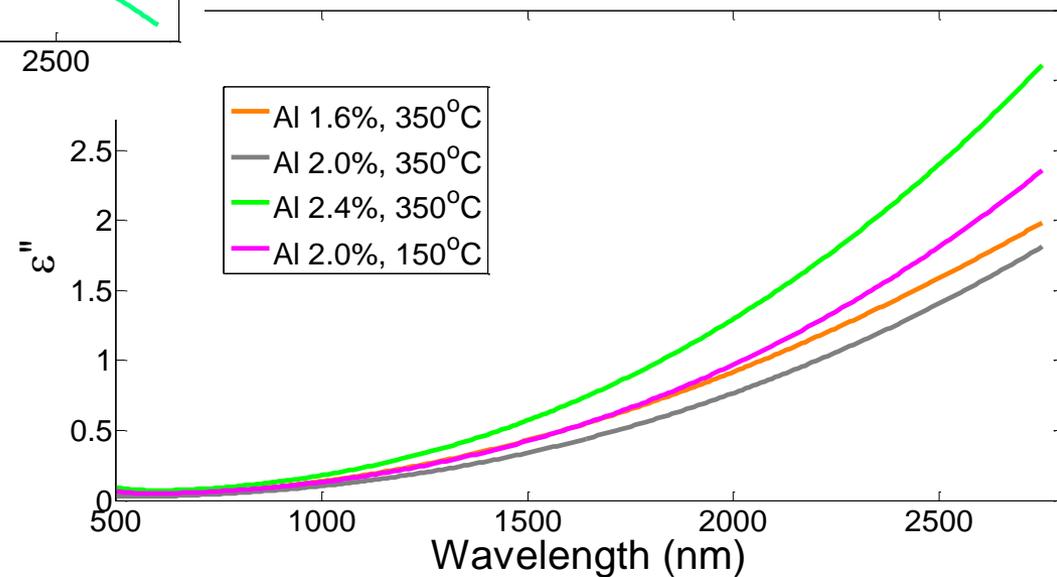
# OPTICAL PROPERTIES



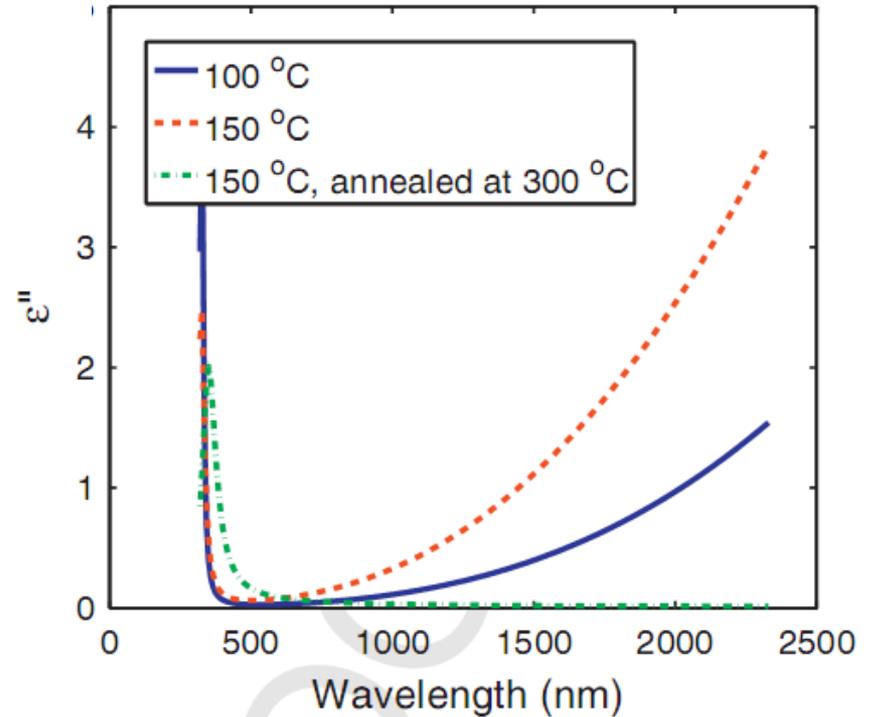
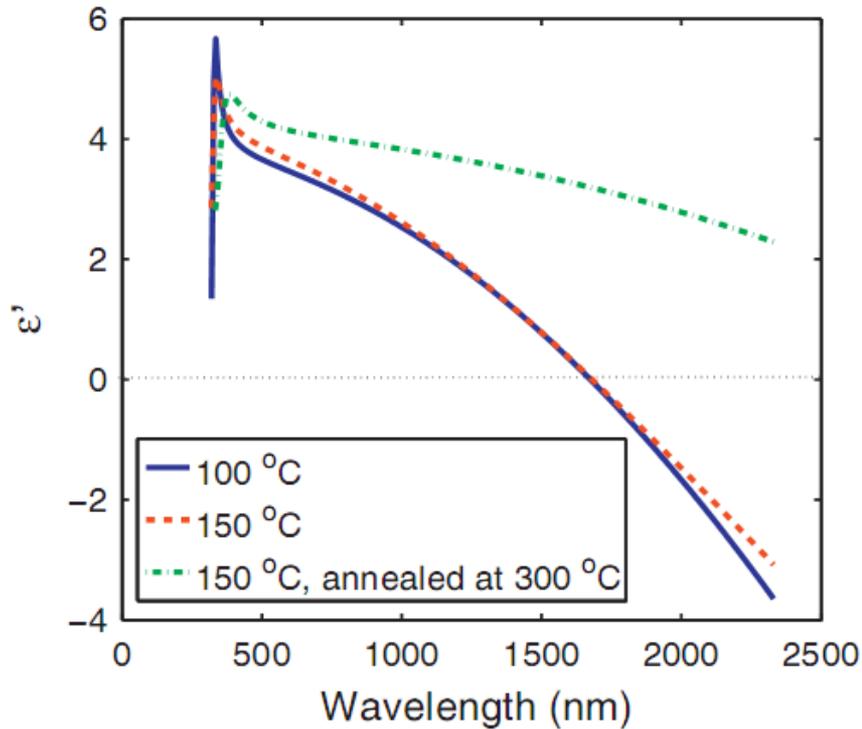
Permittivities for AZO films deposited at 350°C with 2.4, 2, 1.6% Al and AZO film deposited at 150°C with 2% Al

The cross-over frequency increases with larger Al

Film deposited at 150°C has higher cross-over frequency owing to larger efficiency of doping



# OPTICAL PROPERTIES

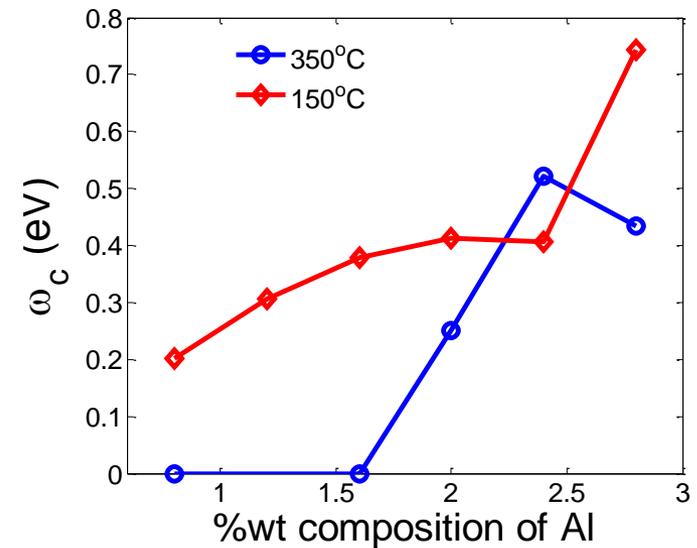


AZO films on c-sapphire substrates: Al doping 3.0 wt%

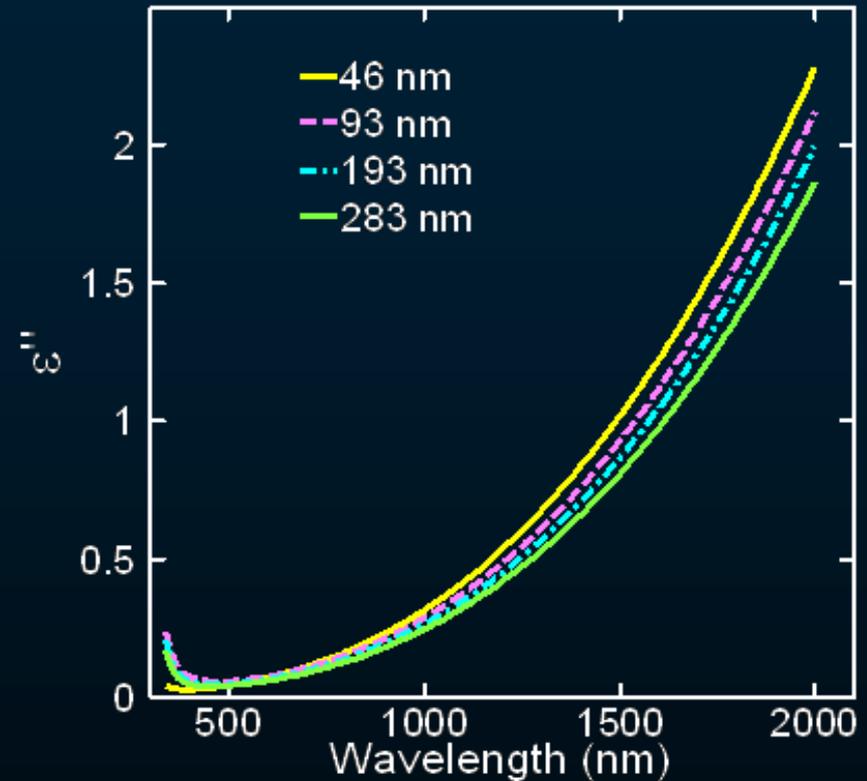
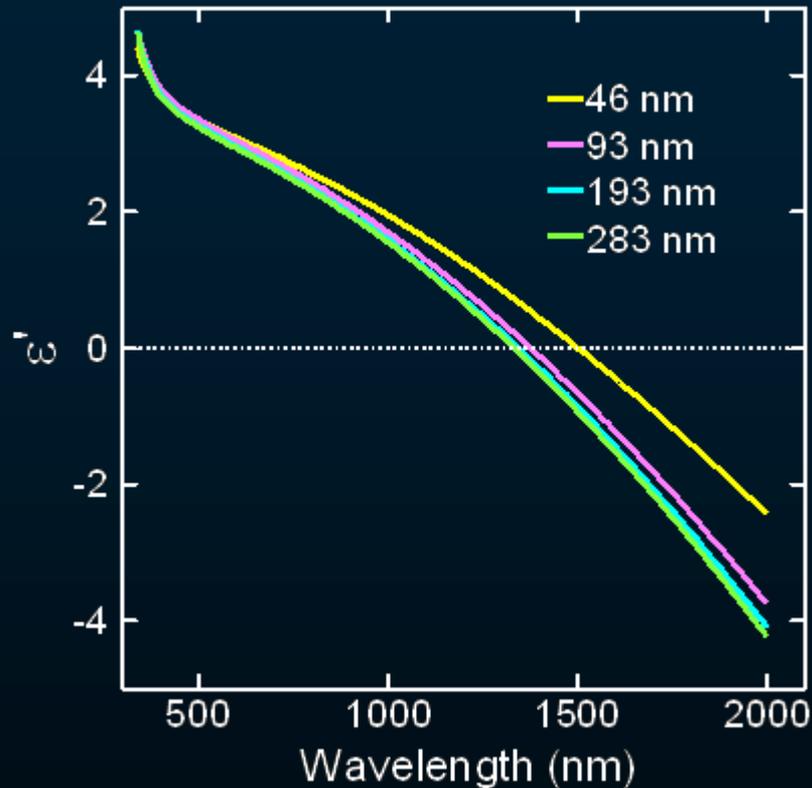
One of the films deposited at 150 °C was subjected to forming gas anneal at 300 °C for 2 h.

# ZnO DOPING

- Larger doping does not necessarily mean larger cross-over frequency
- Trend shows that lower temperature deposition can produce larger cross-over frequency with larger doping

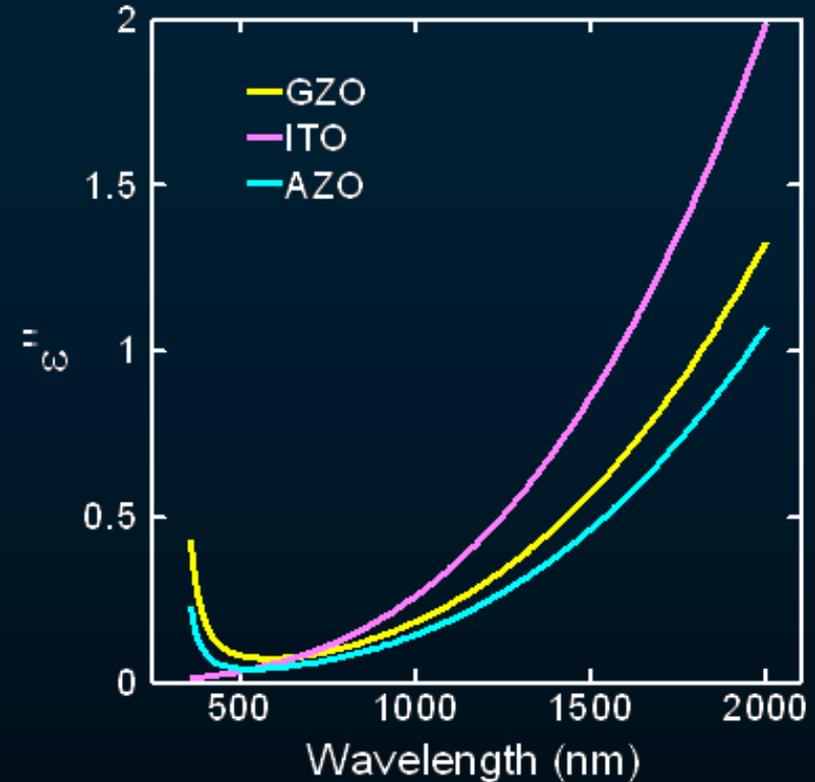
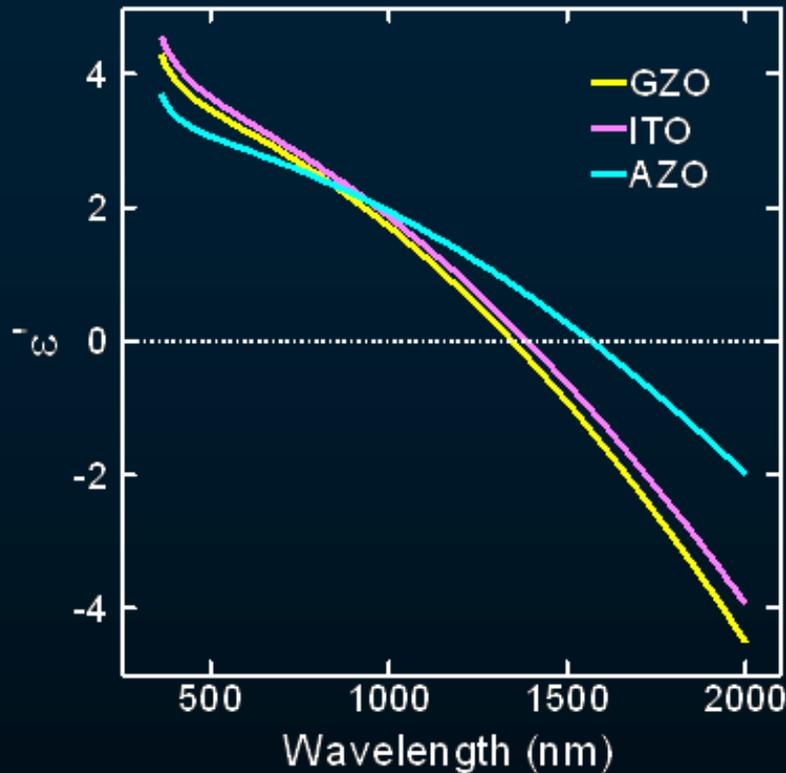


# THICKNESS-DEPENDENT PROPERTIES



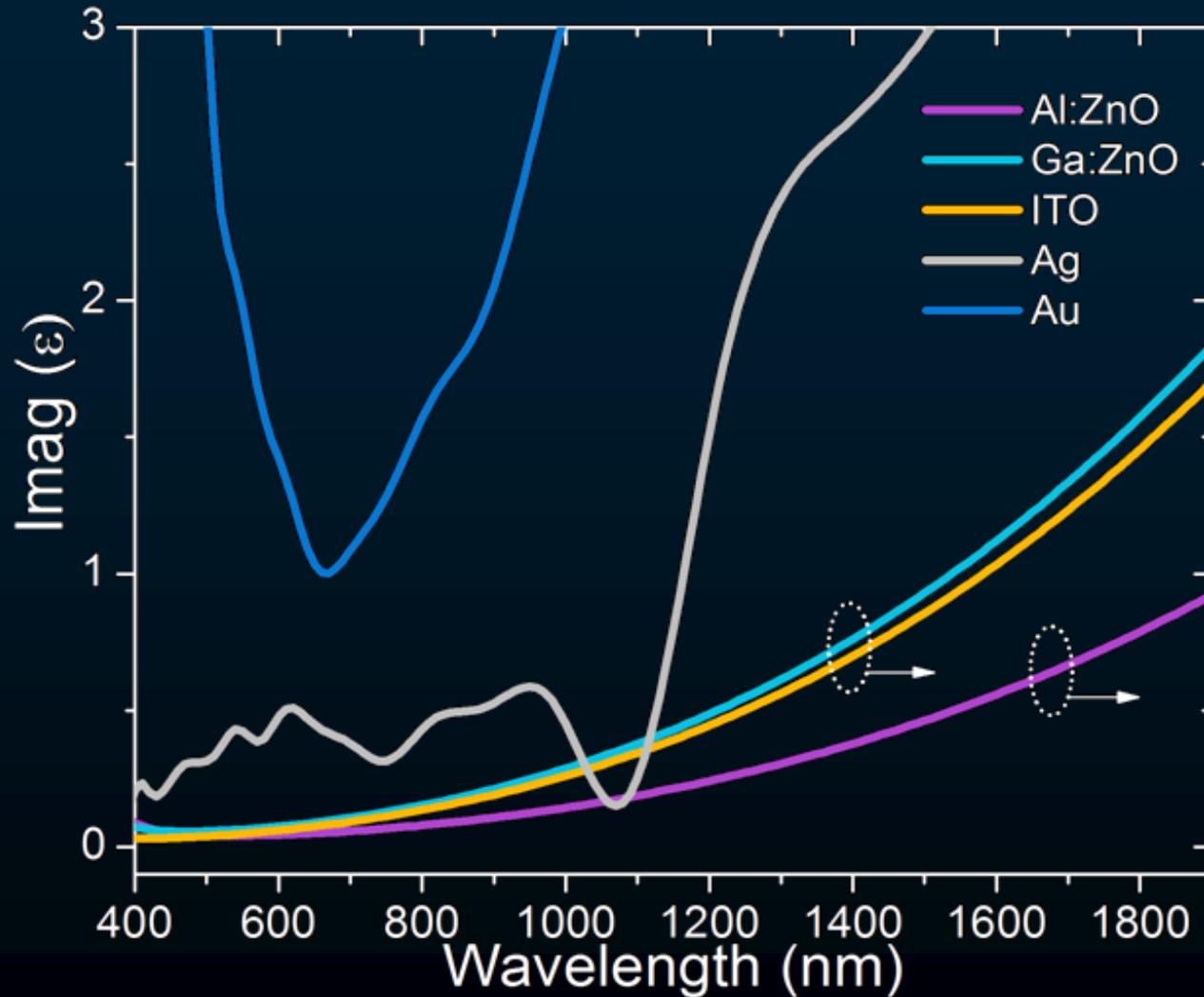
- GZO films deposited on glass under identical condition except film thickness
- Understanding thickness-dependent properties is essential for device fabrication

# OPTICAL PROPERTIES: TCOs

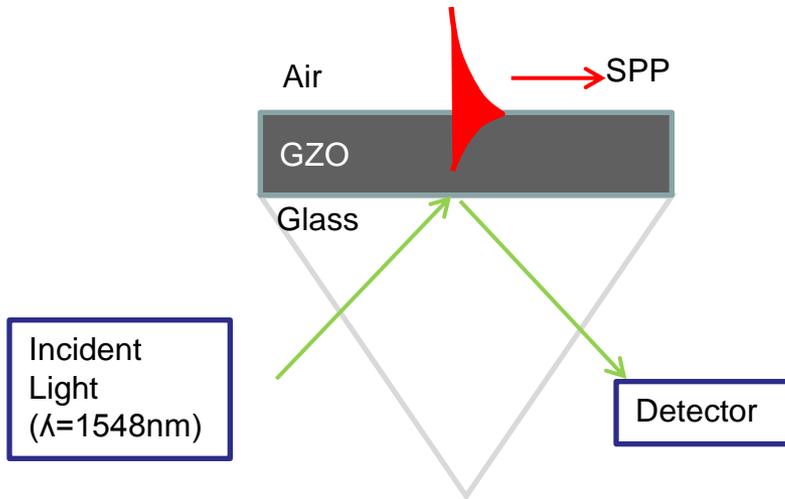


- TCOs films deposited on glass substrate
- AZO: Lowest Drude damping, Longest cross-over wavelength
- GZO, ITO: Cross-over wavelength as low as 1.2  $\mu\text{m}$
- Drude damping in GZO is slightly higher than that in AZO and lower than that in ITO

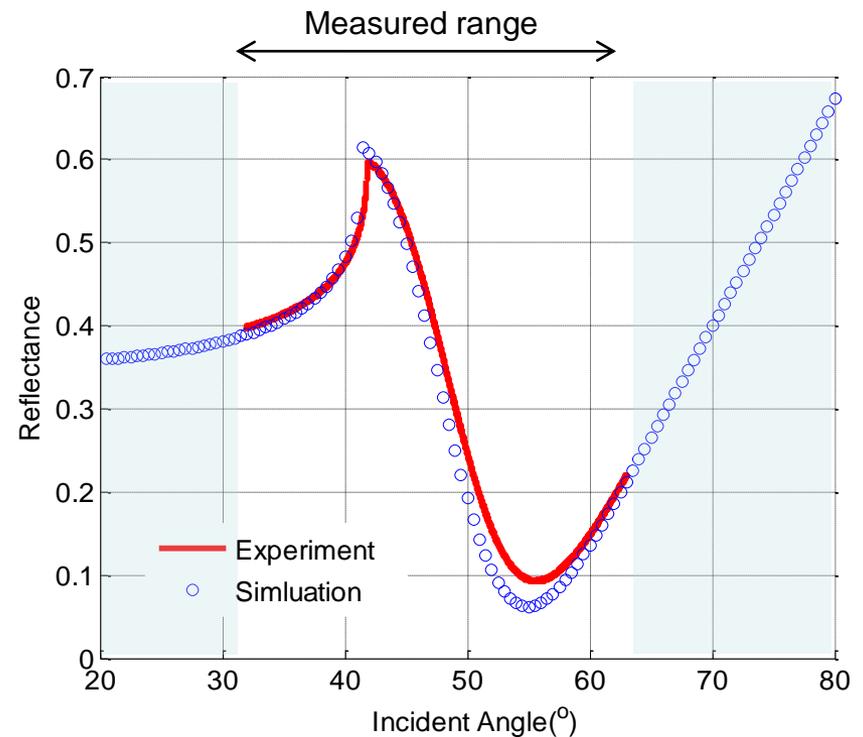
# TCOs COMPARED TO GOLD/SILVER



# SPPs ON GZO FILMS at 1.55um



- GZO is deposited onto glass prism.
- $\epsilon_{\text{GZO}} = -2.12 + 1.2i$  (at 1.55um)
- Angular reflectance shows dip at angles corresponding to excitation of SPPs



# NEXT STEP: PATTERNING



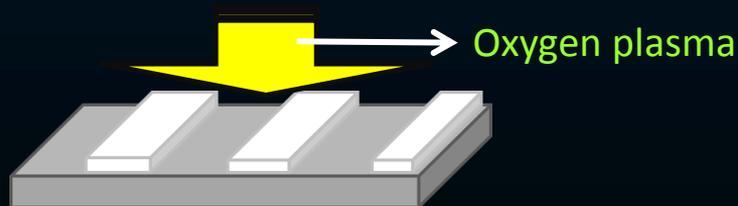
Patterning ZEP 520A using E-beam lithography



Depositing GZO thin film onto patterned ZEP 520A

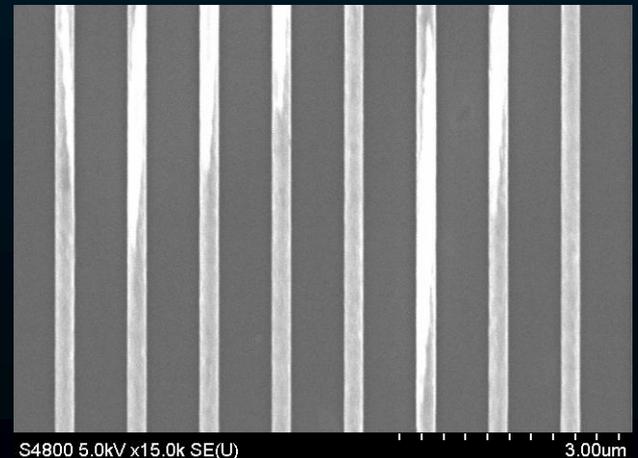
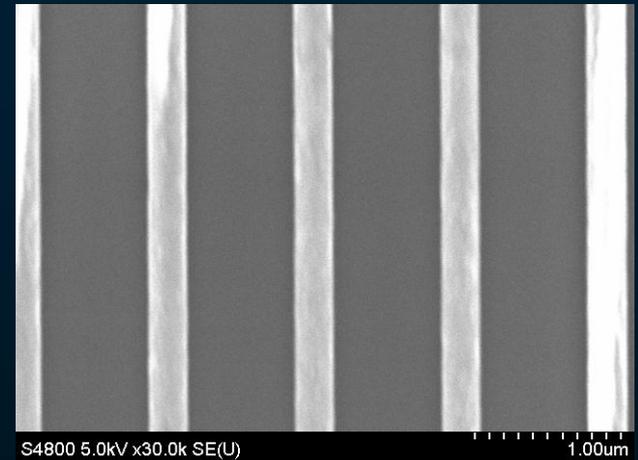


Lift-off ZEP 520A by soaking in ZDMAC

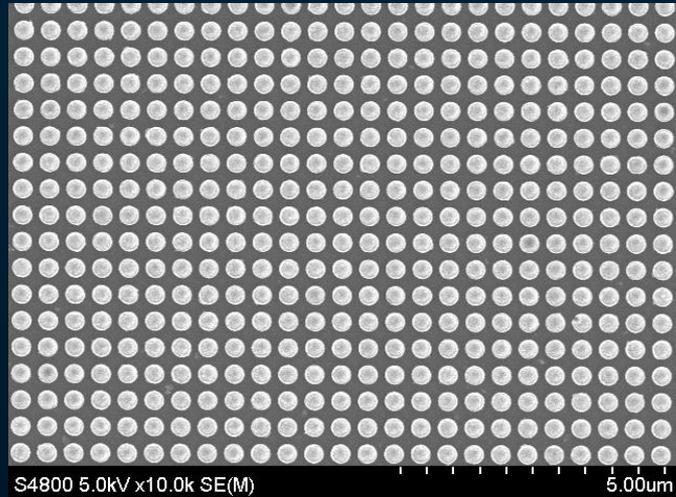
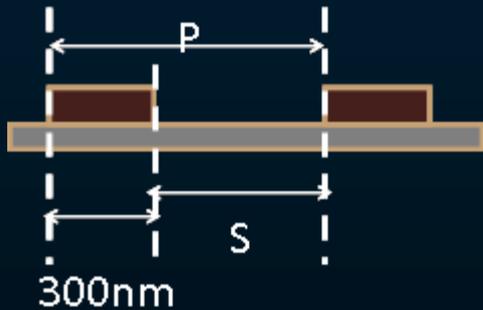


Removing residual ZEP 520A and cleaning a surface with oxygen plasma

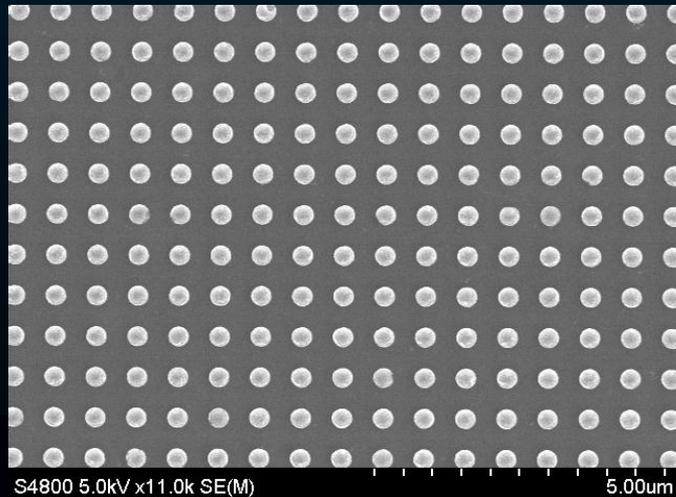
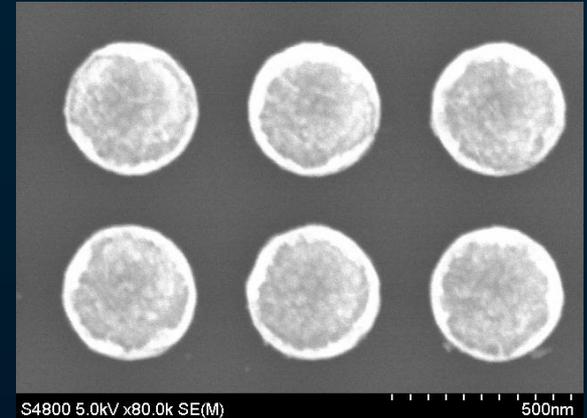
200nm  
↔



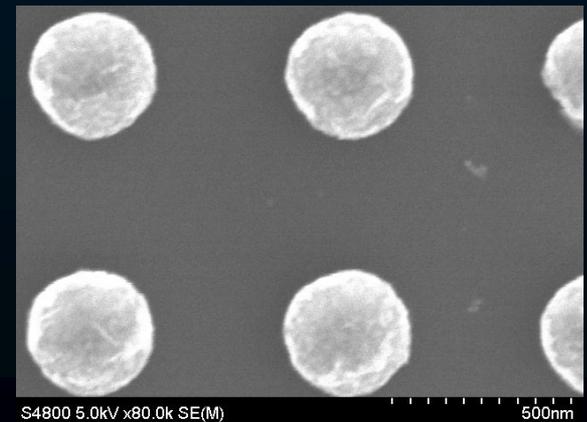
# TCO LSPR STRUCTURES FABRICATION



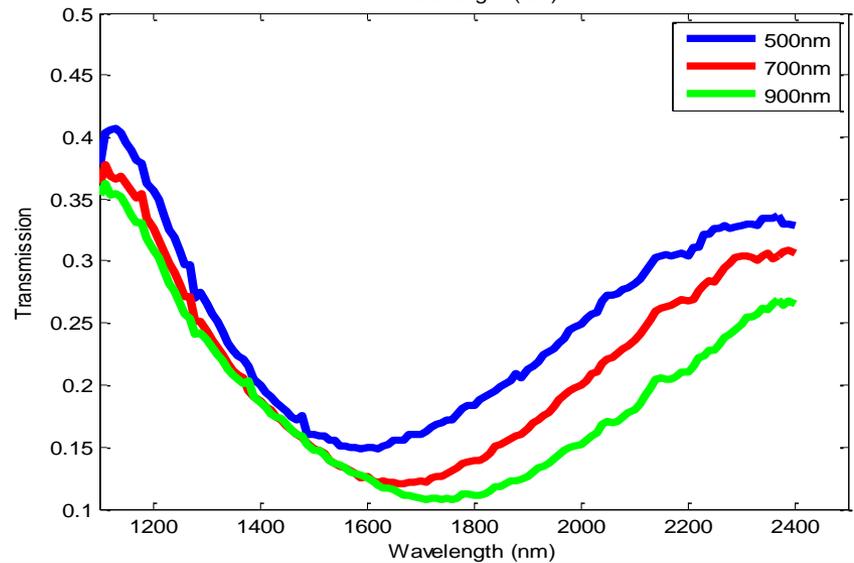
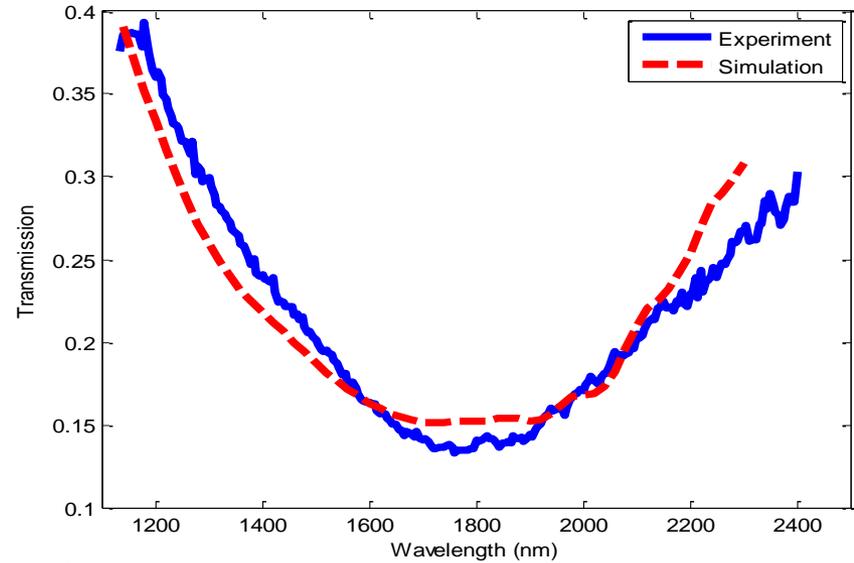
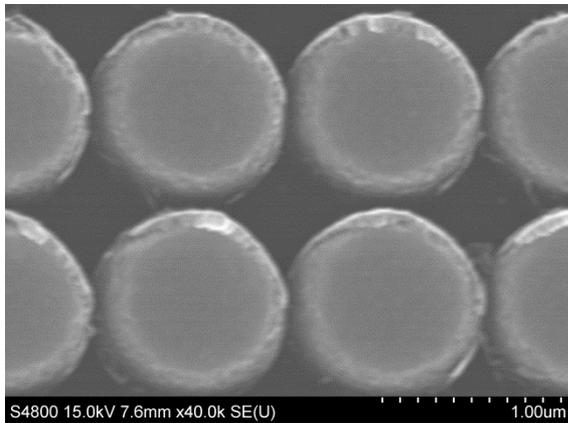
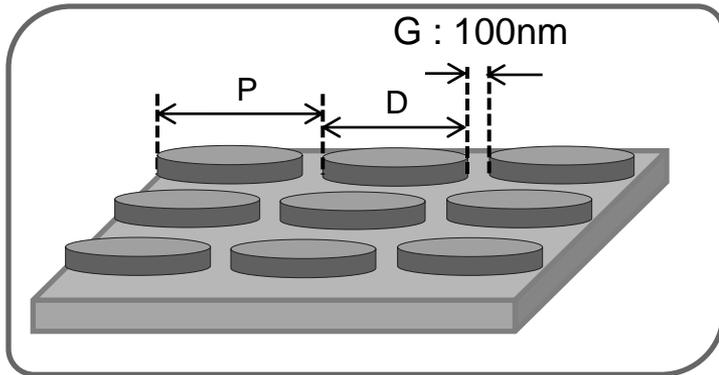
$P : 500\text{nm}, S : 200\text{nm}$



$P : 700\text{nm}, S : 400\text{nm}$

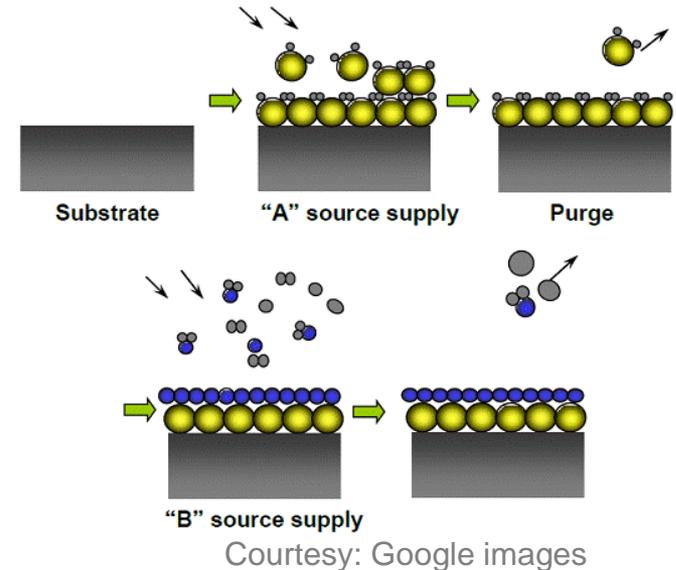


# TCO LSPR STRUCTURES

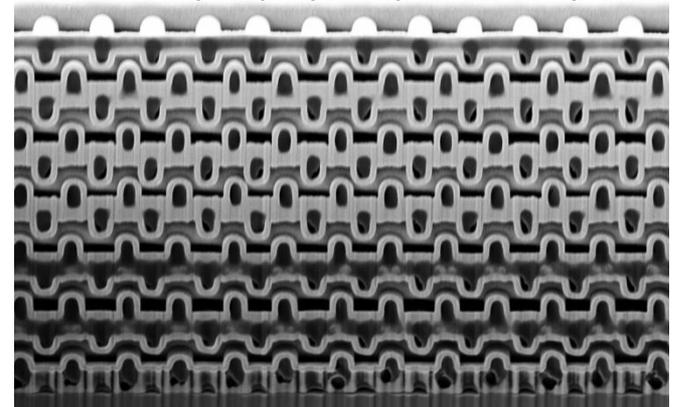


# ATOMIC-LAYER-DEPOSITION OF AZO

- Diethyl zinc and oxygen as precursors to deposit ZnO
- Al or Ti dopants are introduced during deposition
- Smallest cross-over wavelength:  $1.6 \mu\text{m}$
- Drude-damping loss  $\approx 0.2 \text{ eV}$
- Smooth films: rms roughness  $\sim 0.59 \text{ nm}$

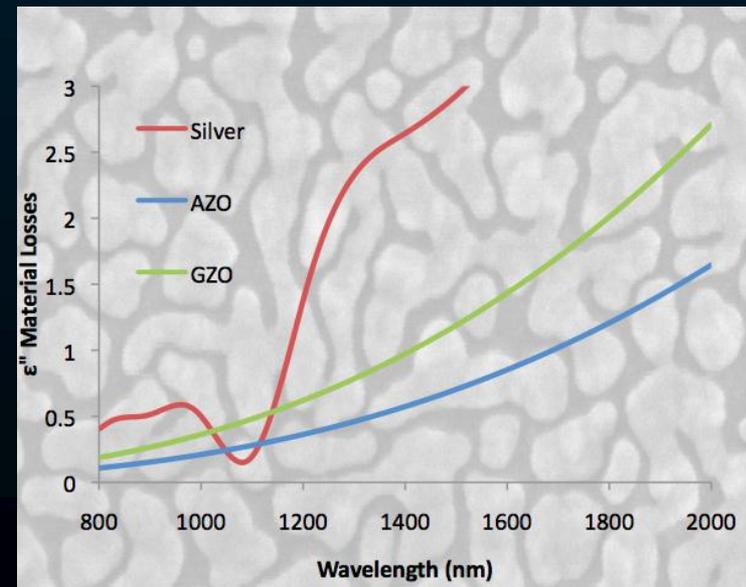


conformal on woodpile polymer photonic crystal



# OUTLINE

- Introduction: Challenges with Gold and Silver
- Material Requirements: Plasmonics & Metamaterials
- Alternative Materials?
- Transparent Conducting Oxides
- **Transition Metal Nitrides**
- Figures of Merit
- What is the Right Choice?
- Outlook



# METALS TO 'LESS-METALS'

- Reduce carrier concentration:  
Mixing them with non-metals  $\Rightarrow$

## Intermetallics

## Ceramics

- Silicides
- Germanides
- Borides
- Nitrides
- Oxides
- Metallic alloys

# TITANIUM NITRIDE

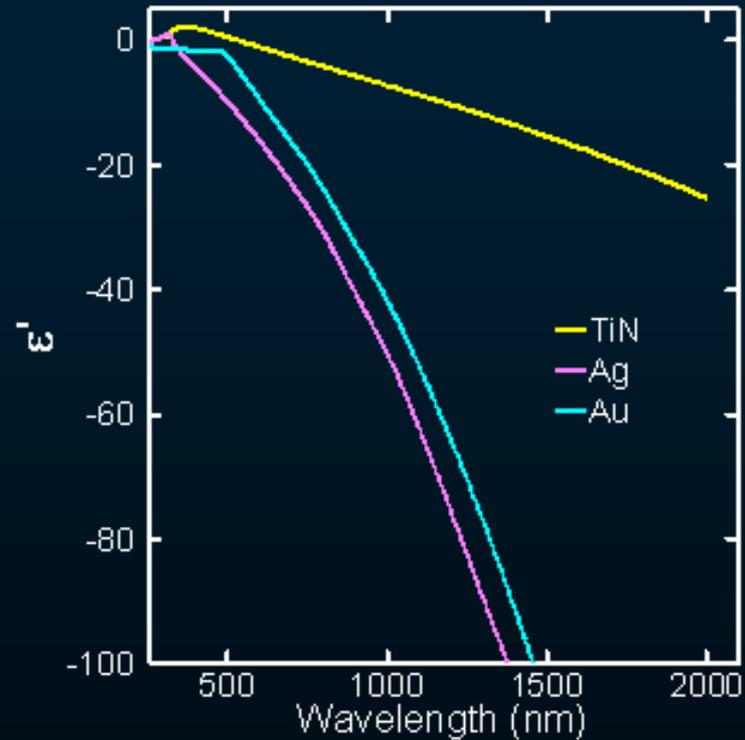
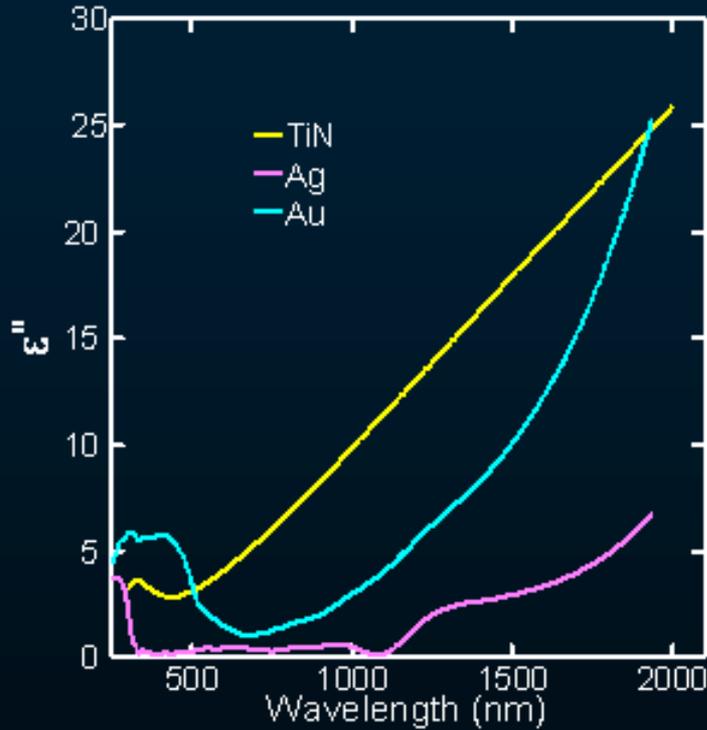
- Metallic: Golden luster
- Hard & tough: high speed drill-bits
- Deposition: CVD, sputtering, evaporation...

**CMOS COMPATIBLE**

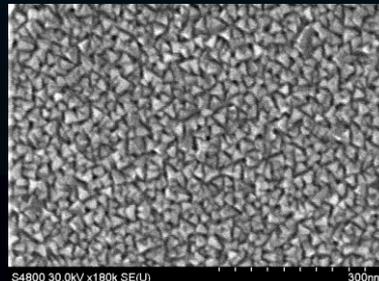


# TITANIUM NITRIDE

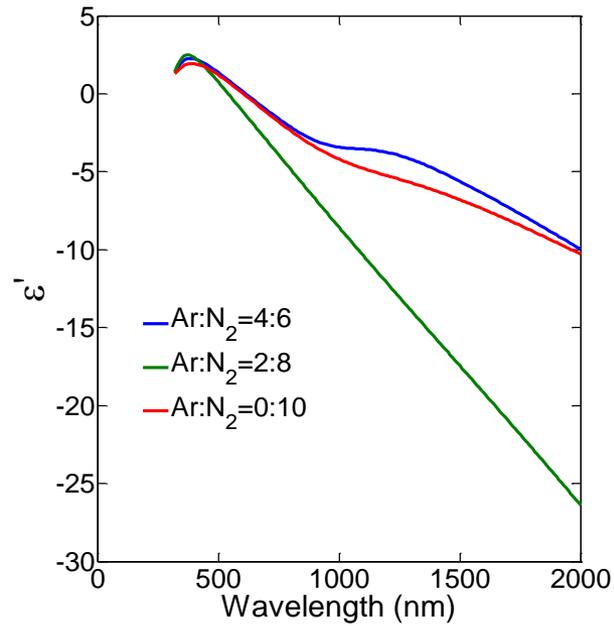
TiN comparison with Ag and Au



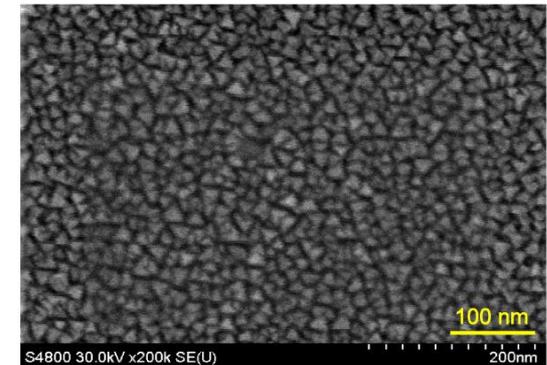
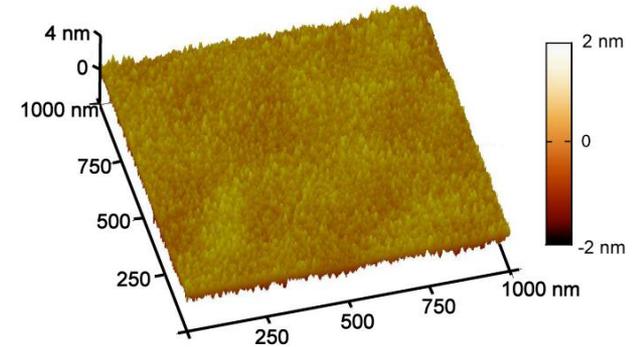
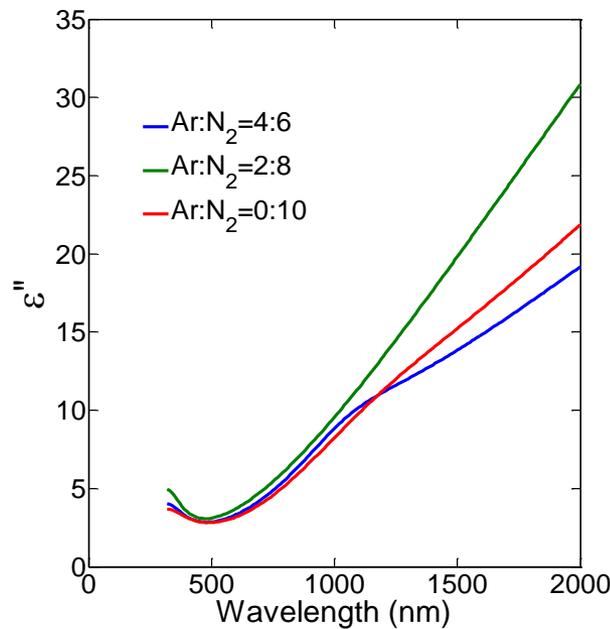
Crystalline TiN on c-Sapphire  
(rms roughness 0.5 nm)



# TITANIUM NITRIDE: OPTIMIZATION

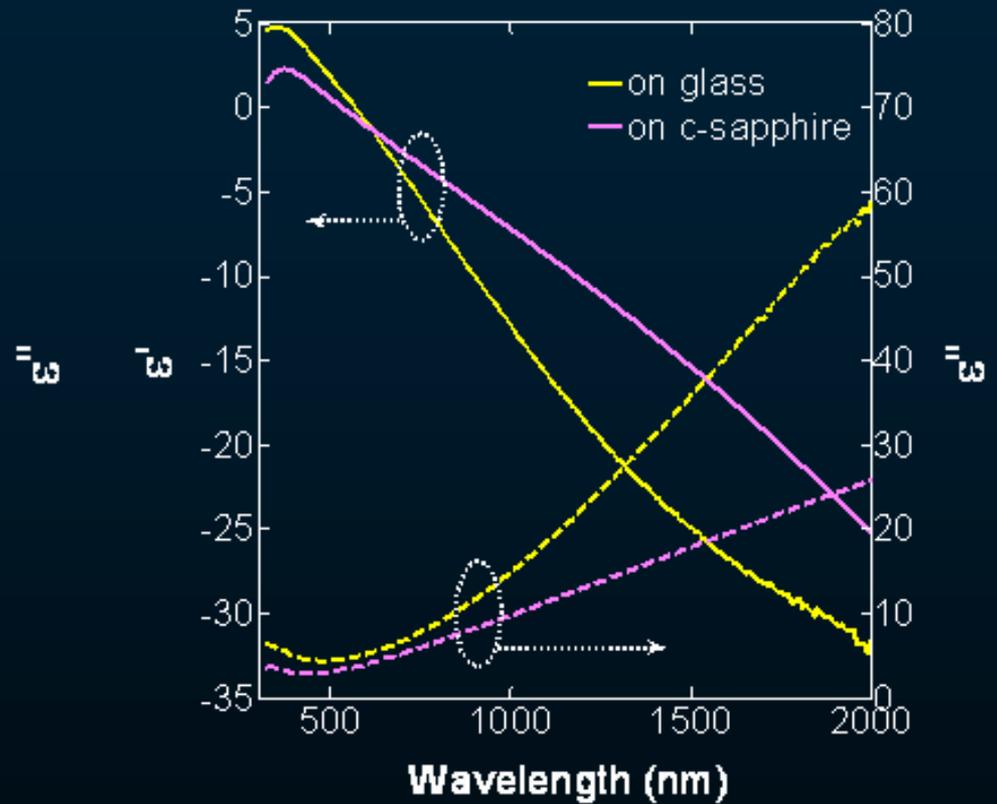
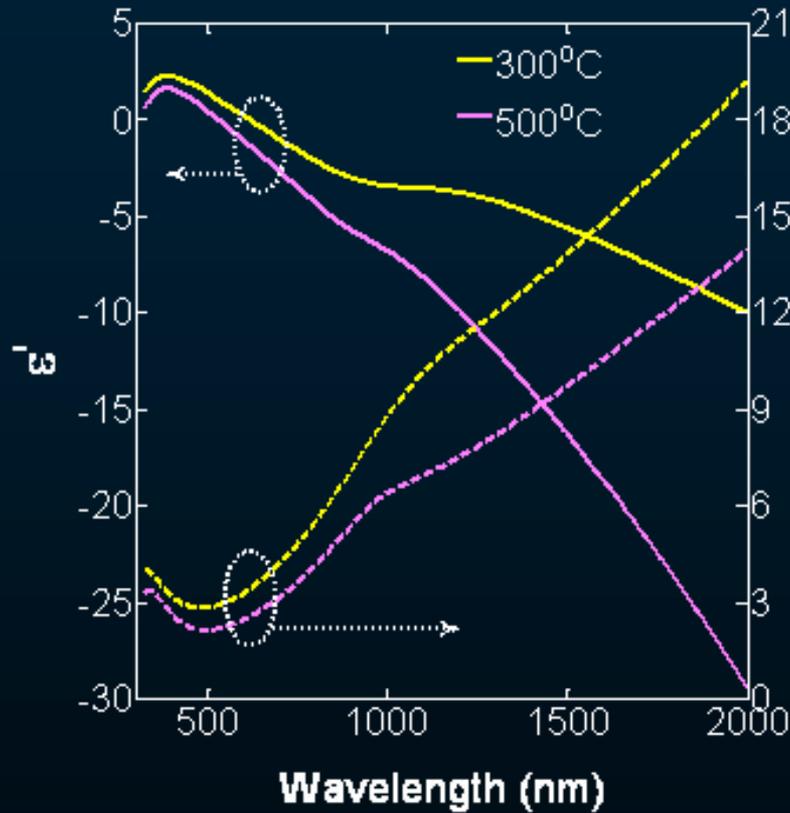


Changing optical properties



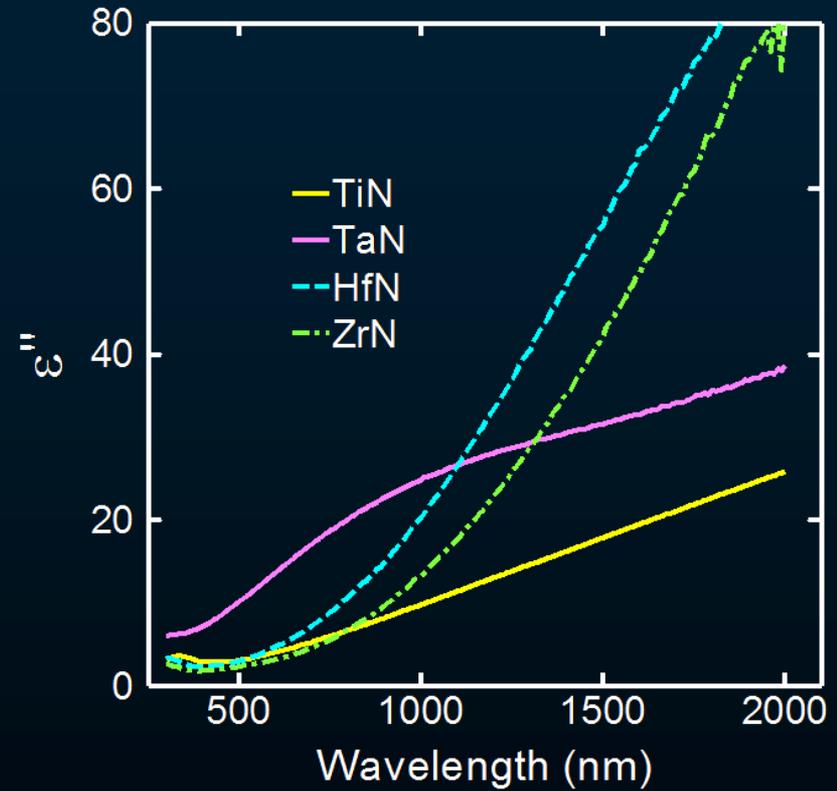
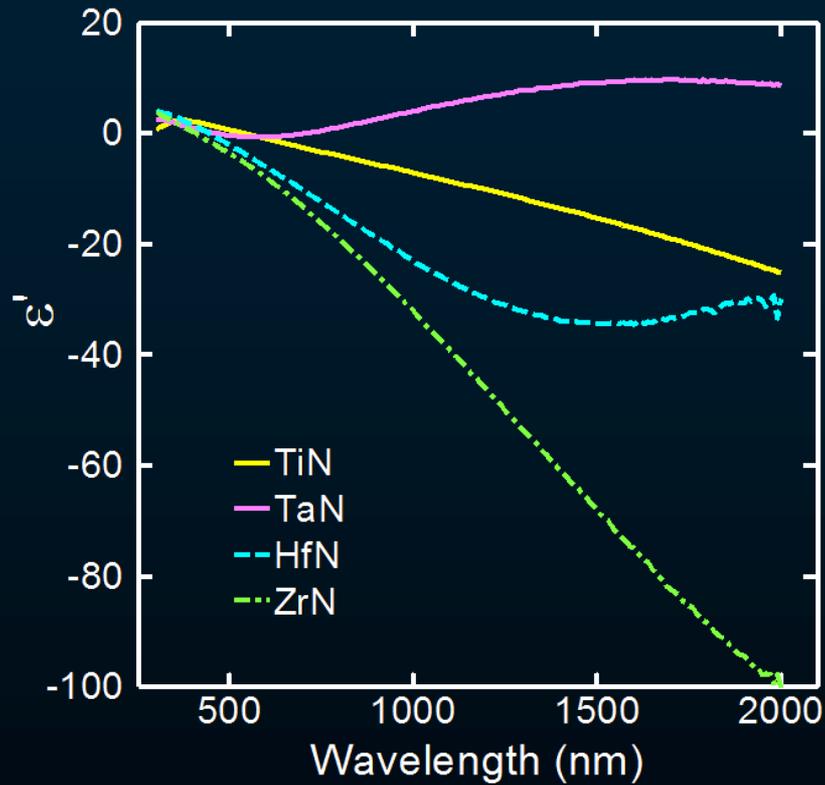
Ultra-thin and smooth films; epitaxial growth

# TITANIUM NITRIDE



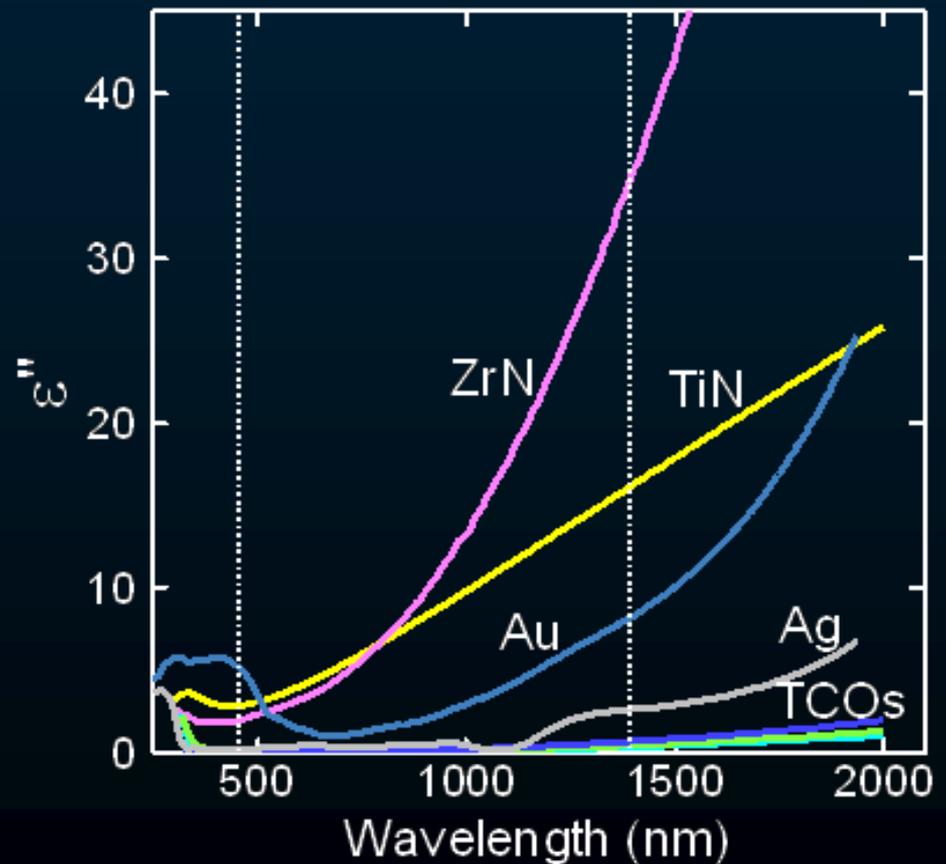
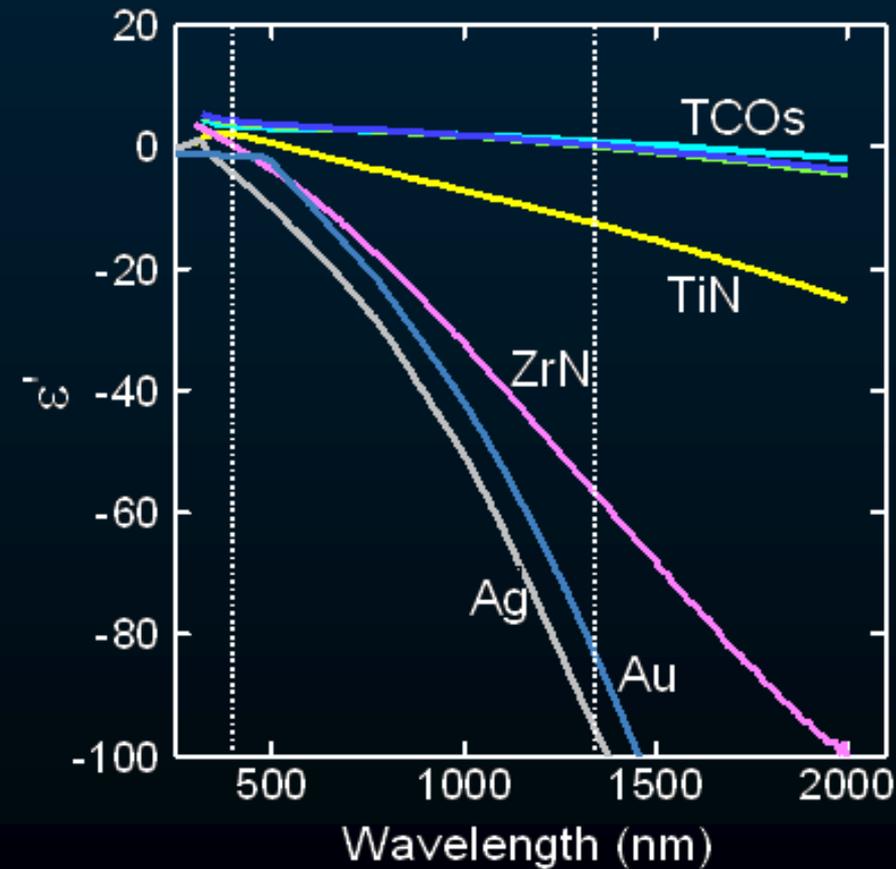
- TiN films deposited at 300°C and 500°C (left)
- TiN films deposited on different substrates (right)
- The films were deposited with the flow ratio of Ar and N set to 4:6

# OTHER METAL NITRIDES



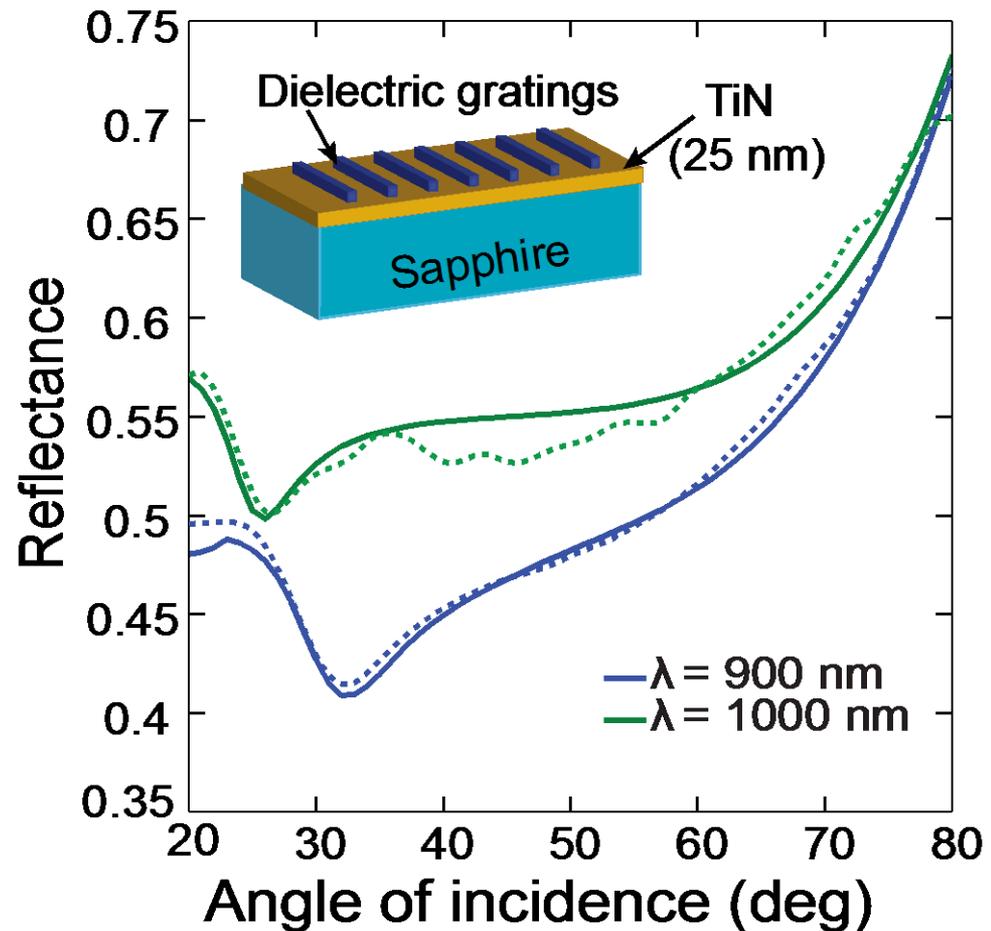
# COMPARISON WITH SILVER/GOLD

TaN, HfN, ZrN also exhibit metallic properties in the visible!

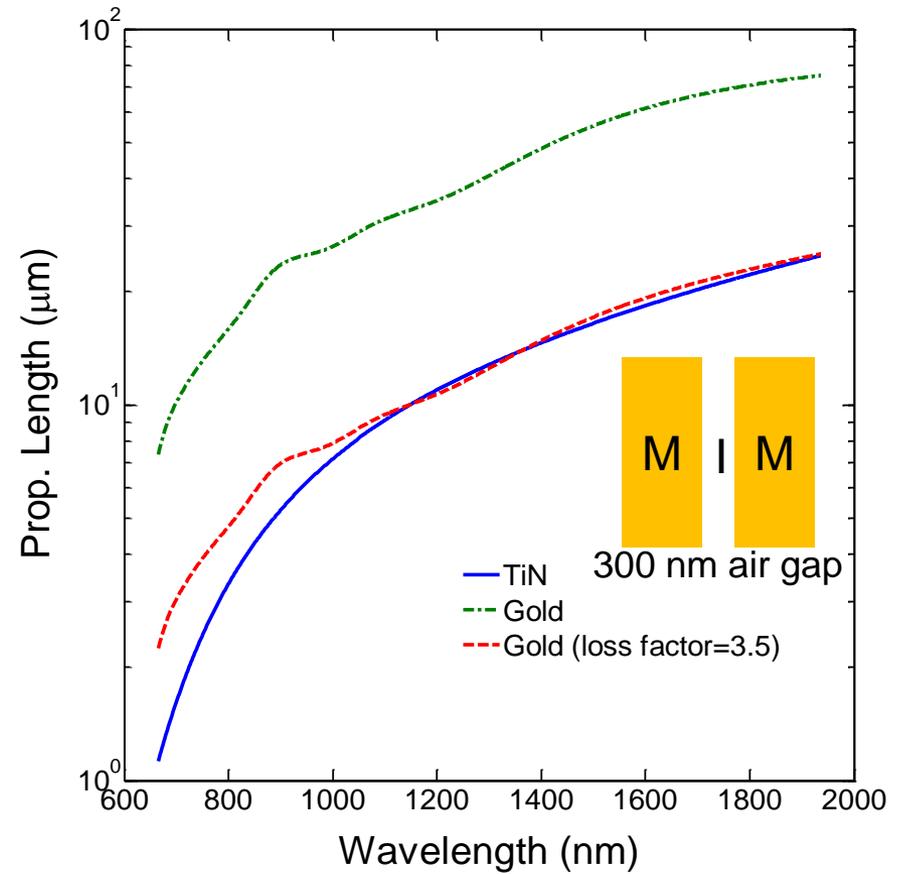
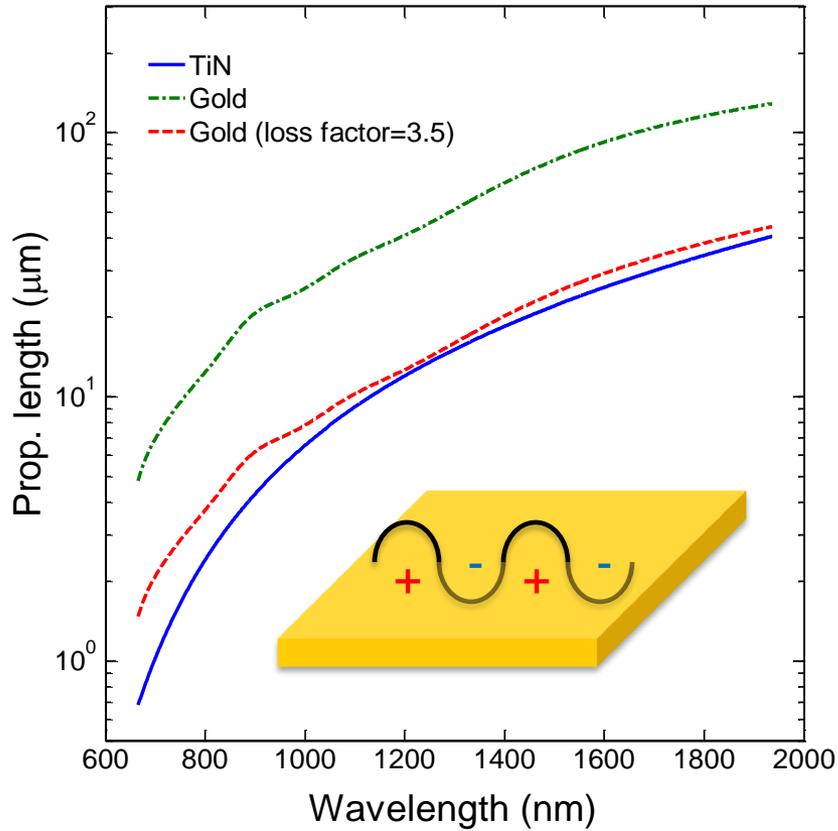


# SPP ON TiN FILM

- Dielectric gratings are used to couple SPPs
- Electron-beam resist is patterned into gratings on top of 25 nm thin TiN film
- Angular reflectance shows dip at angles corresponding to excitation of SPPs

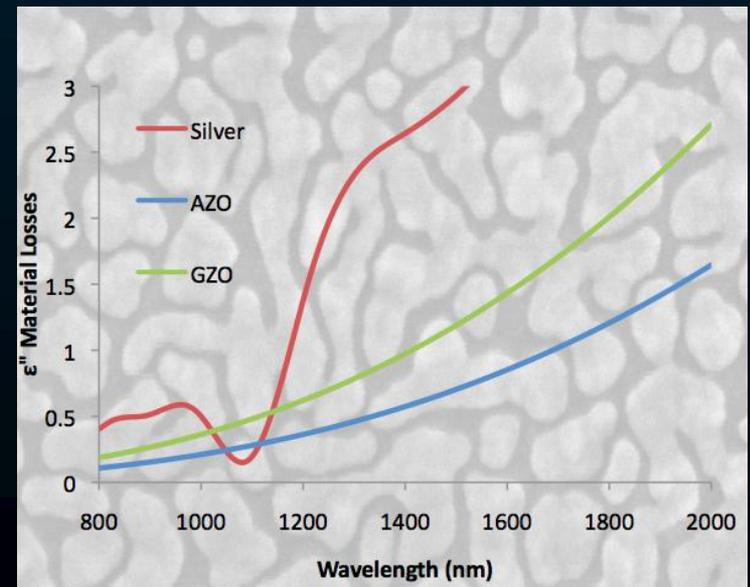


# SPPs ON TiN FILM



# OUTLINE

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# QUALITY FACTORS

- **$\text{Im}(\epsilon) = \epsilon''$**  is a necessary indicator of performance *but*
- **$\text{Re}(\epsilon) = \epsilon'$**  is also important in quantifying the overall material quality in devices

## **Metrics/Figures-of-Merit/Quality Factors**

are to be estimated for each

- Material
- Application
- Wavelength range...

# QUALITY FACTOR FOR SP

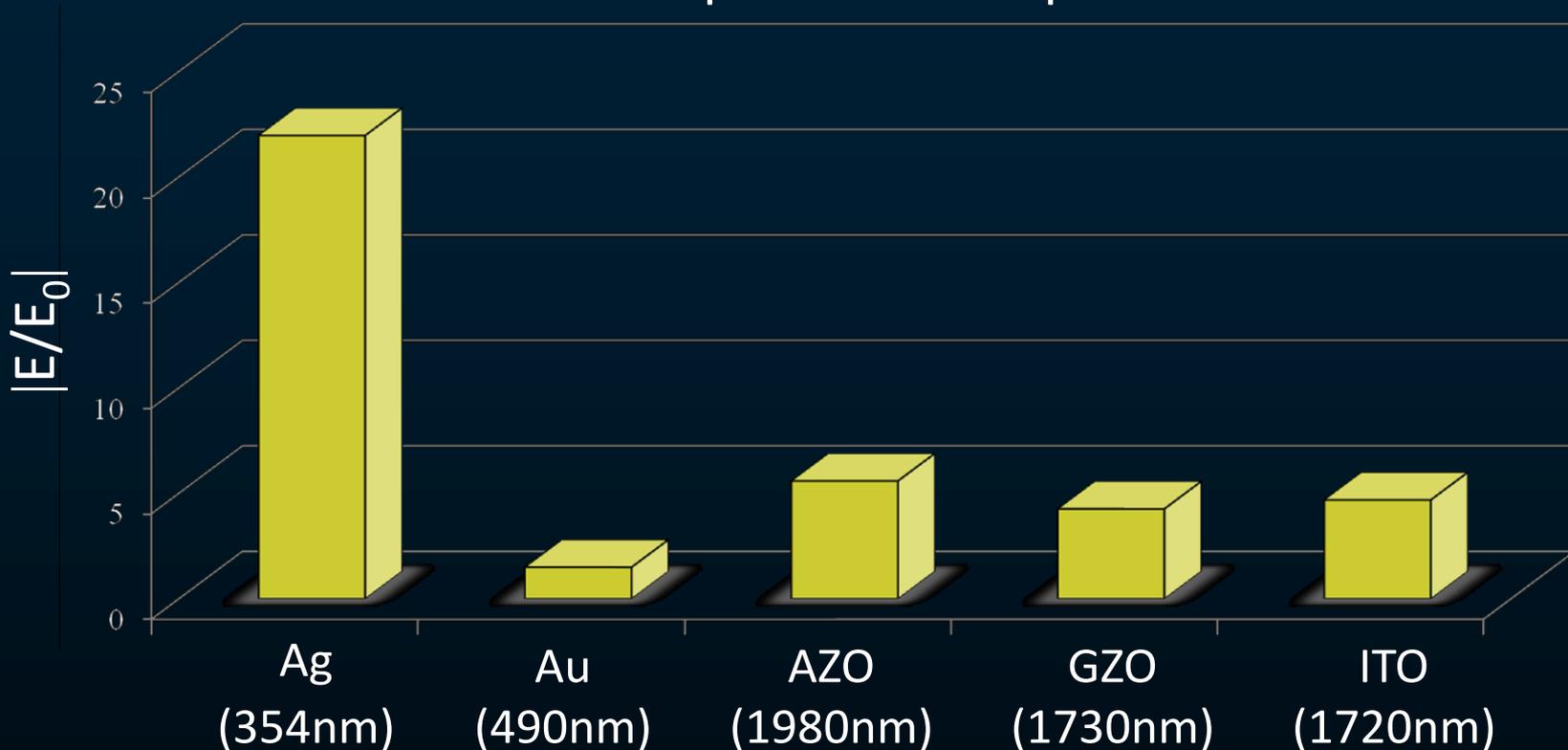
LSPR and SPR systems: Local-field enhancement

$Q_{\text{LSPR}} = (\text{Enhanced local-field}) / (\text{Incident field})$   
*(depends on the shape)*

$$Q_{\text{LSPR}}(\omega) = \frac{-\varepsilon'(\omega)}{\varepsilon''(\omega)} \quad (\text{sphere})$$

# COMPARISON: LSPR

LSPR field enhancement for spherical nanoparticles in air



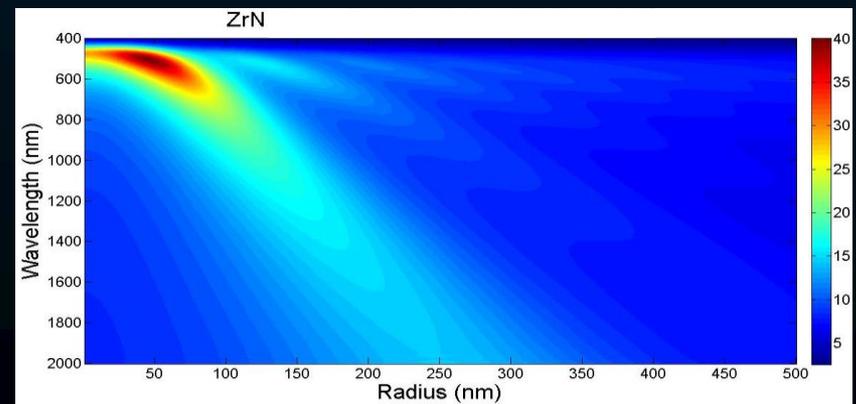
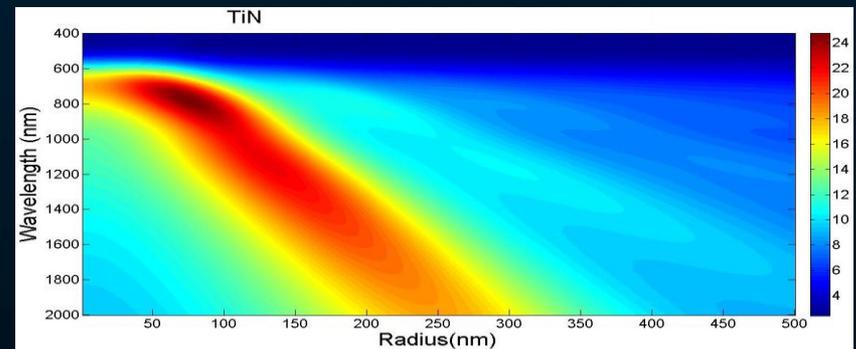
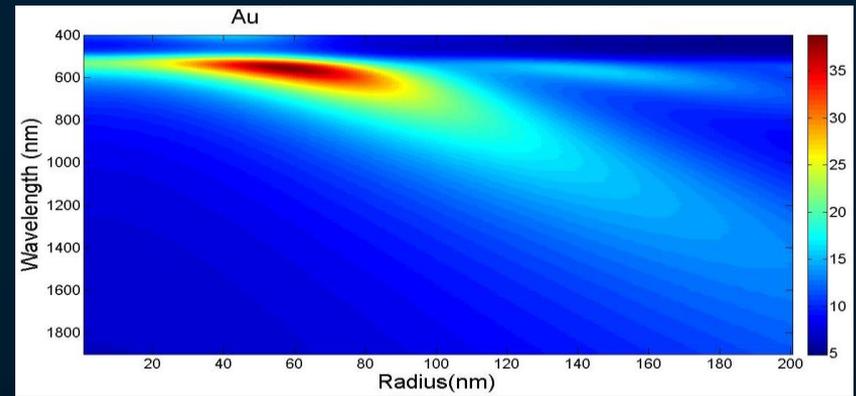
LSPR occurs at  $\epsilon_{\text{metal}}' = -2\epsilon_{\text{dielectric}}$  (dipolar):

- Cannot be satisfied in Ag and Au in NIR
- Need other geometries (core-shells): more challenging to fabricate
- TCOs produce strong LSPR in the near and farther IR

# COMPARISON: LSPR

- Field enhancement of metal nanospheres at the surface calculated using Mie theory
- ZrN and TiN nanospheres: Field enhancement comparable to that of Au
- TiN has good performance over broadband in the visible and near-IR

near field intensity enhancement



# HYPERBOLIC METAMATERIALS

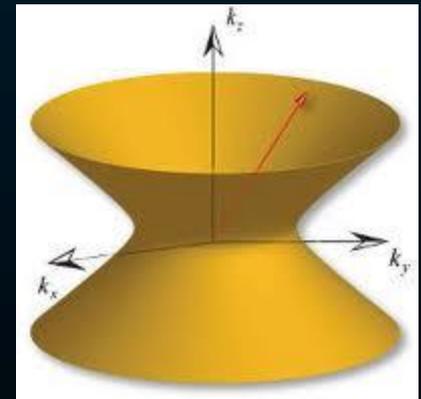
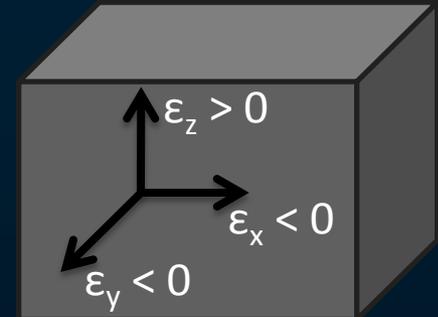
## Extremely anisotropic materials

- Extremely large effective index
- Extremely high photonic density of states

## Applications

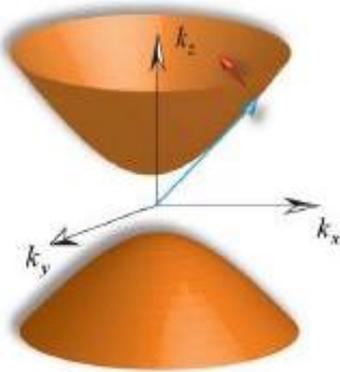
- Quantum optics with metamaterials
- Tailor mechanical, thermal and electromagnetic properties for interesting physics and devices

**Designing a material which is metallic in one direction but dielectric in the another**



# HYPERBOLIC METAMATERIALS

A metamaterial has hyperbolic dispersion relation



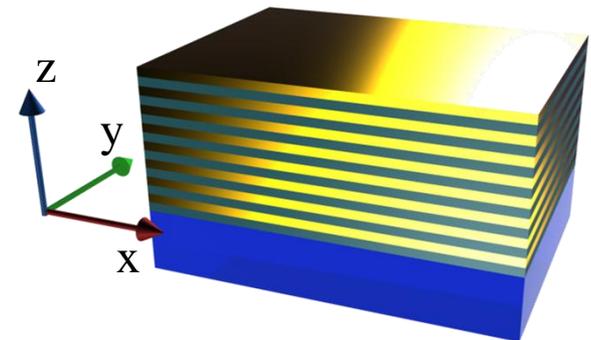
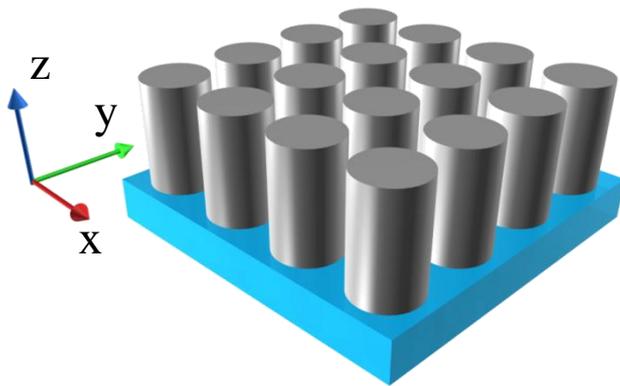
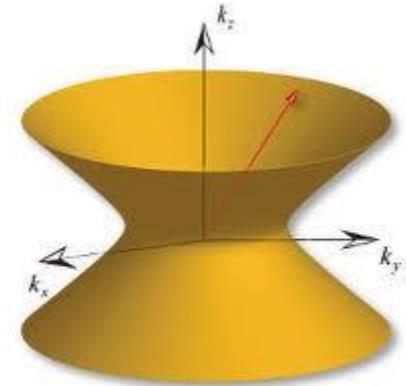
**Hyperbolic dispersion**

$$\epsilon_{x,y} > 0 ; \epsilon_z < 0$$

**Transverse positive**

$$\epsilon_{x,y} < 0 ; \epsilon_z > 0$$

**Transverse negative**

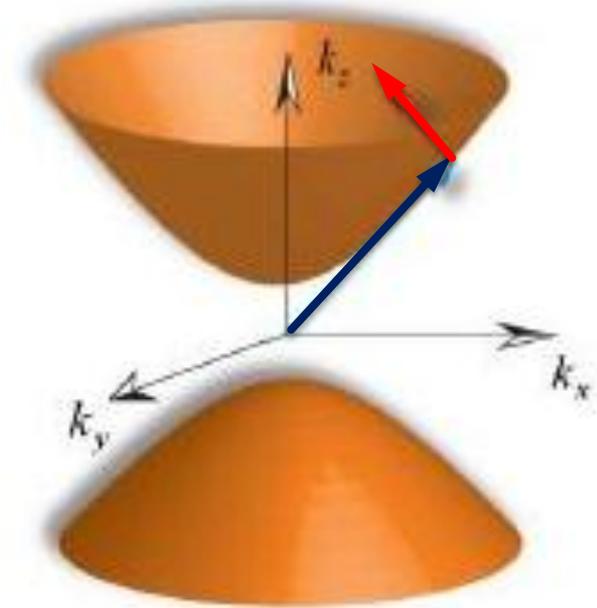


# NEGATIVE REFRACTION IN HMMs

- Only transverse-positive HMM can exhibit negative refraction.
- Only transverse-positive HMM produces measurable transmission.
- Only transverse-positive HMM can produce hyperlensing.
- Figure-of-merit<sup>1</sup>:  $\text{Re}\{\beta_z\} / \text{Im}\{\beta_z\}$

<sup>1</sup>A. Hoffman et. al, Nature Materials **6** (2007);

$\epsilon_{x,y} > 0 ; \epsilon_z < 0$   
**Transverse positive**



Z. Jacob and V.M. Shalaev, Science **334** (2011)

# HMM PERFORMANCE: TCOs / NITRIDES

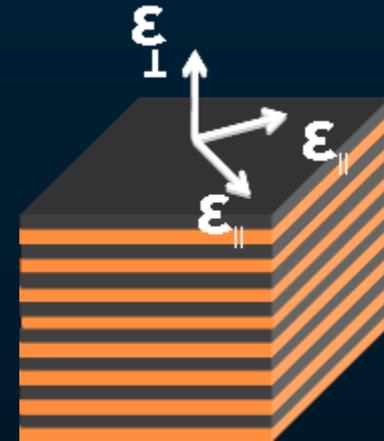
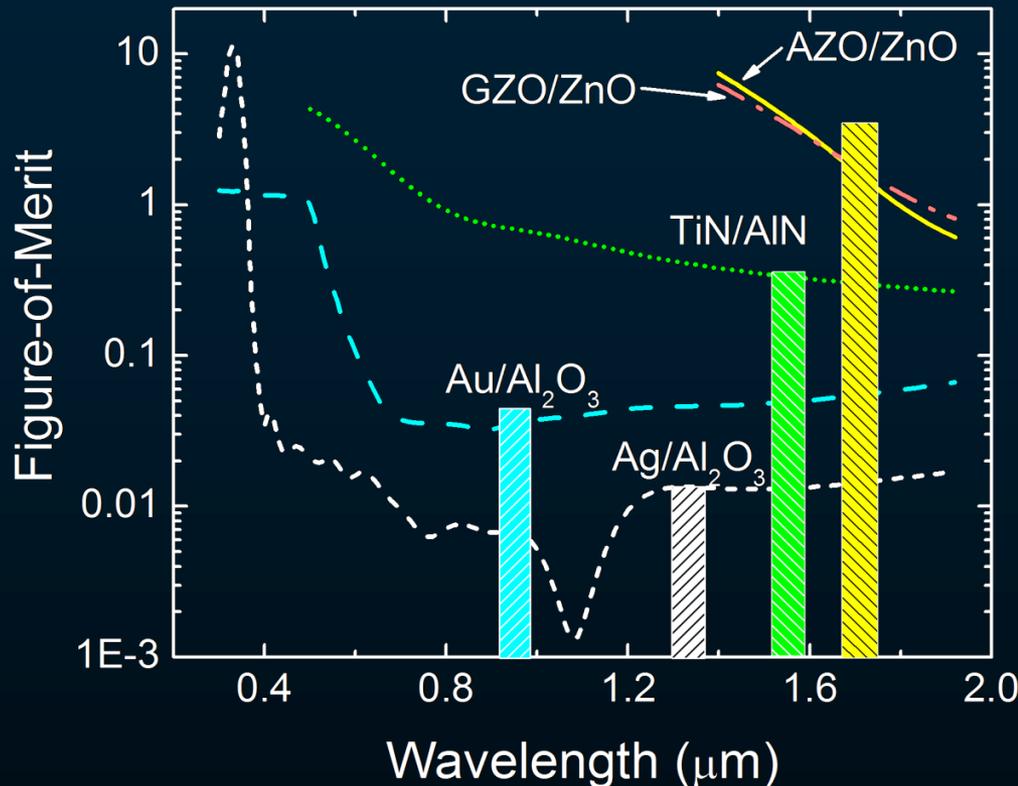


Figure-of-Merit =  $Re\{\beta_{\perp}\} / Im\{\beta_{\perp}\}$

$\beta_{\perp}$  – propagation vector in  $\perp$  - direction

Polycrystalline AZO/ZnO, single crystal AZO/ZnO, Au/Al<sub>2</sub>O<sub>3</sub> and Ag/Al<sub>2</sub>O<sub>3</sub>  
 AZO/ZnO - Figure-of-Merit of **11** at 1.8 μm

# NEGATIVE REFRACTION EXPERIMENT

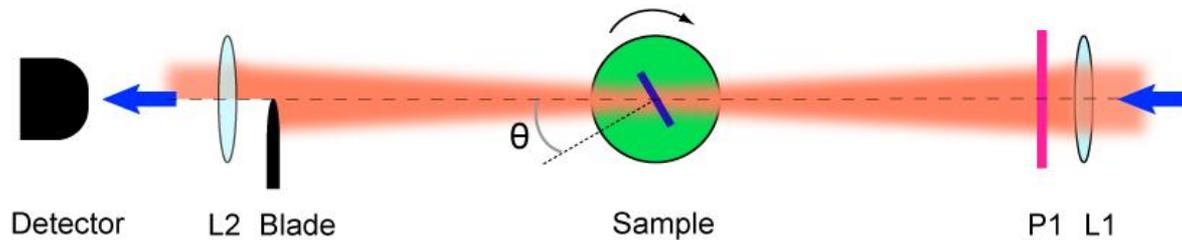
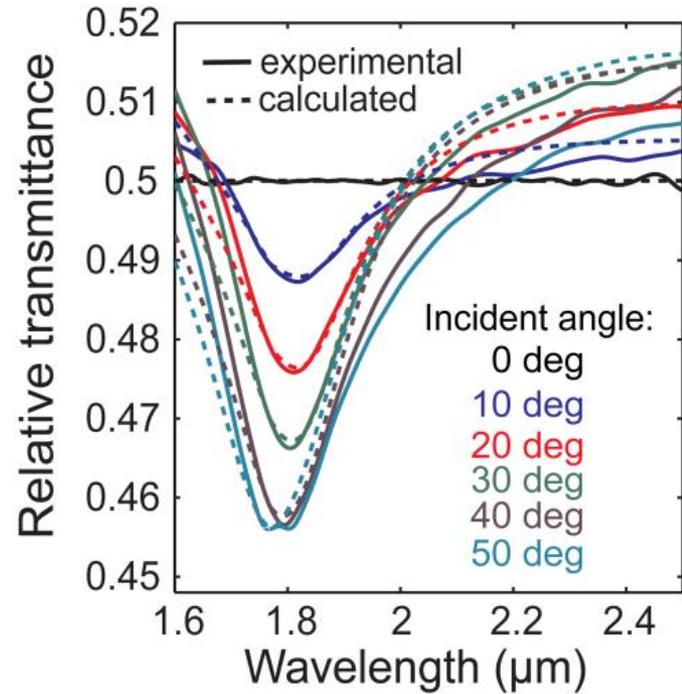
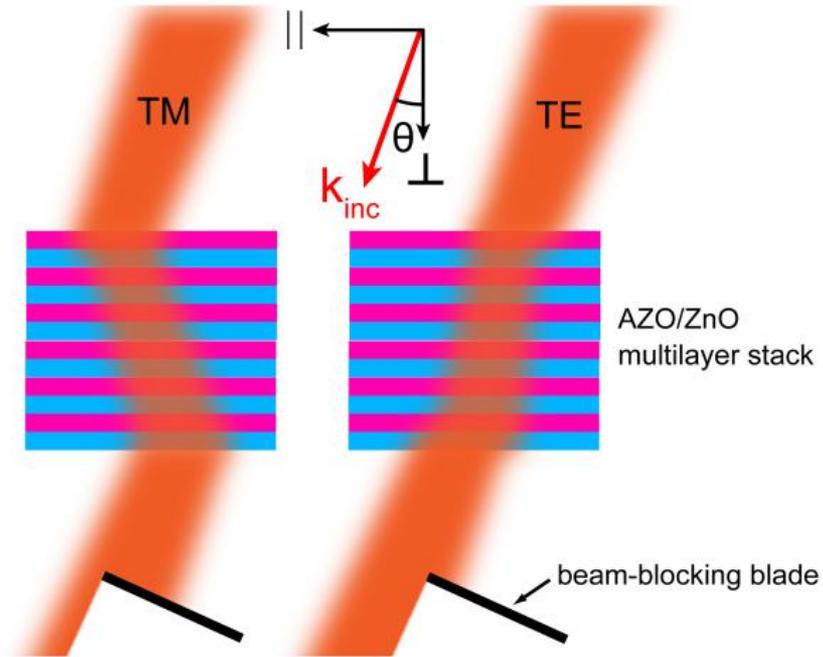
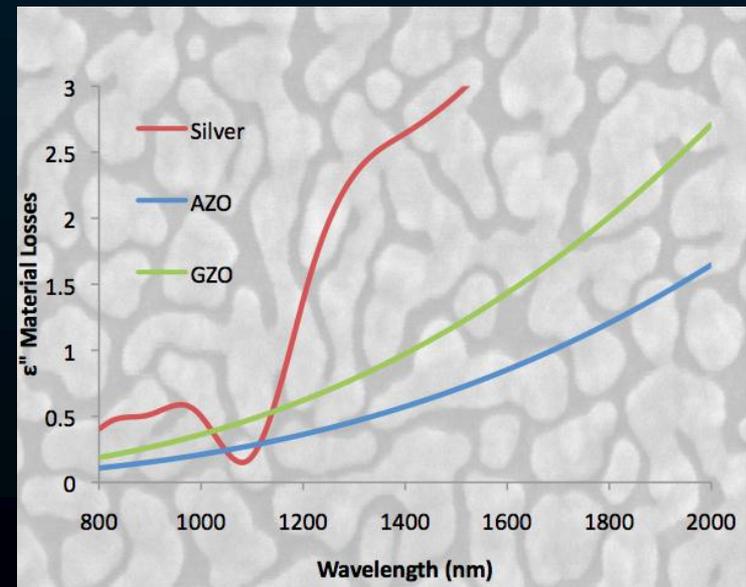


Figure-of-merit (FoM) : 11  
at 1.8  $\mu\text{m}$  wavelength  
Highest FoM demonstrated  
in NIR

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# WHAT IS THE BEST MATERIAL FOR...

## Applications

LSPR, SPPs & Waveguides, NIMs, TO, ENZ...

## Materials

Noble Metals, Metal Nitrides, TCOs, Noble Alkali Alloys, Alkali Metals, Graphene, Other Metals...

# What is the Best Plasmonic Material For...

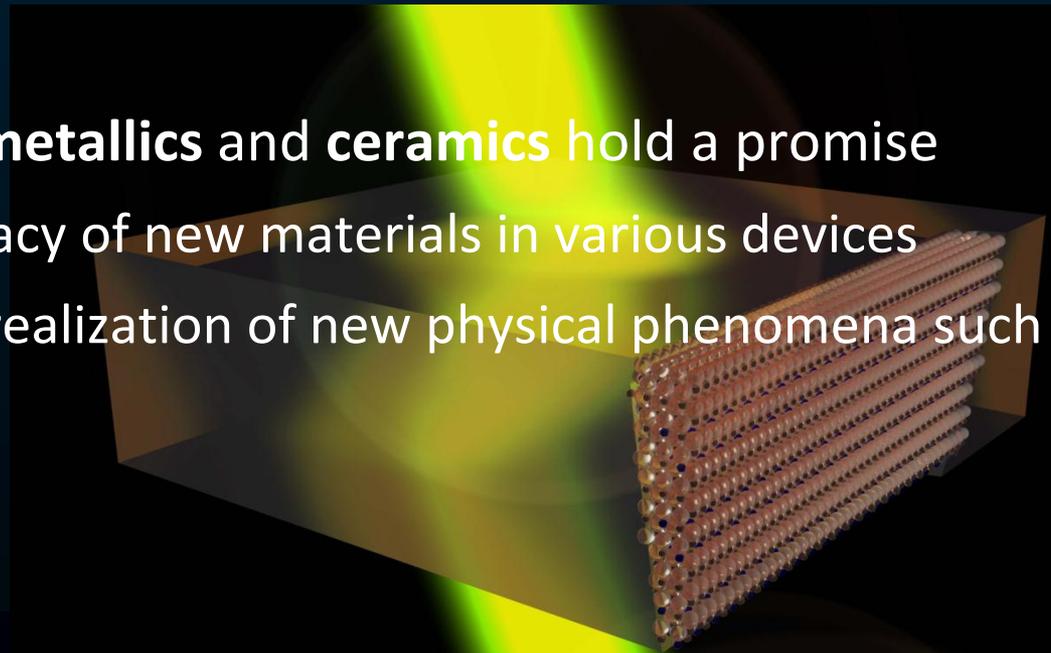
	LSPR	SPPs & Waveguides	NIMs	TO	ENZ	Switchable MMs
Noble Metals	✓	✓	✓	✗	✗	✗
Transition Metal Nitrides	✗	✓	✗	✓	✗	✗
TCOs	✓	✗	✗	✓	✓	✓
Intermetallics	✗	✗	✗	✓	✓	✗
Alkali Metals	✓	✓	✓	✗	✗	✗
Graphene	✗	✗	✗	✗	✗	✓
Other Metals	✗	✗	✗	✗	✗	✗

# MATERIAL CHOICES: OUTLOOK

- **Infrared:**
  - Silicides, Germanides, GaAs, SiC
- **Near-infrared:**
  - TCOs: AZO, GZO, ITO
  - Perovskites: Heavily doped barium tin oxide, strontium titanate, cadmium tellurium oxide, calcium titanate, strontium tin oxide
    - *Tune plasma frequency by doping*
    - *Great switching opportunities*
- **Visible:**
  - Intermetallics: Silicides, Germanides, Nitrides...
  - Titanium nitride, tantalum nitride

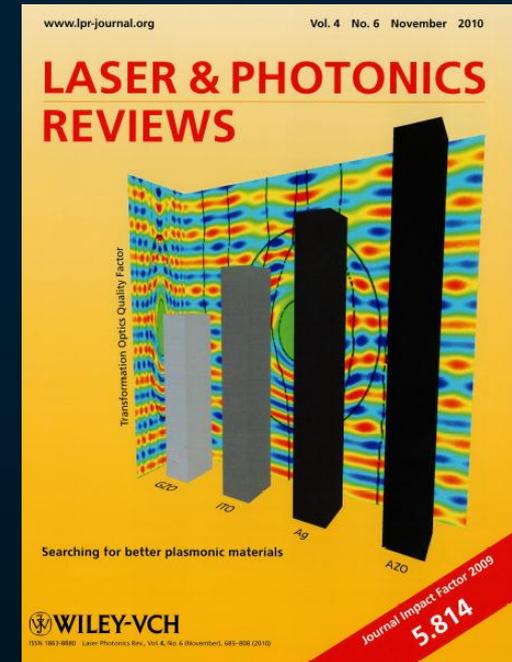
# SUMMARY

- There is not a single choice for the best low-loss plasmonic material for all applications and all wavelength
- **At 1.5- $\mu\text{m}$  TCOs (AZO, ITO, GZO)** may be the best materials for **TO devices** and **NIR applications** providing low loss and not that negative  $\epsilon'$
- At **visible** range **intermetallics** and **ceramics** hold a promise
- Demonstration of efficacy of new materials in various devices
- New materials enable realization of new physical phenomena such as ENZ properties

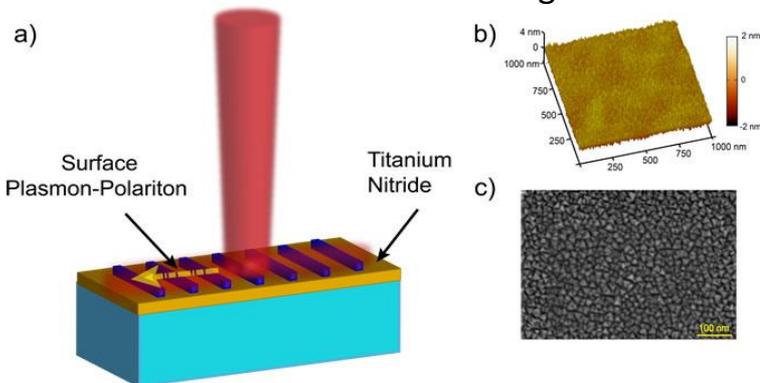


# PLASMONIC MATERIALS RESEARCH

- Laser & Photonics Reviews 4, 795–808 (2010)
- Phys. Status Solidi RRL 4, 295–297 (2010)
- Metamaterials 5, 1–7 (2011)
- Science 331, 290 (2011)
- Optical Materials Express 1 (6), 1090–1099 (2011)  
MM and Plasmonics Focus Issue, **Invited**
- Optical Materials Express 2 (4), 478-489 (2012)  
**Highlighted article**
- Proc. Natl. Acad. Sci. (2012)



Researchers Discover a New Path for Light Through  
Metal: Novel Plasmonic Material May Merge  
Photonic and Electronic Technologies



OSA press release March 27, 2012

*Nature Photonics News&Views Highlight*

news & views

VIEW FROM... NANOMETA 2011

**In search of new materials**

NATURE PHOTONICS | VOL 5 | MARCH 2011

MATERIALS SCIENCE

**Low-Loss Plasmonic Metamaterials**

21 JANUARY 2011 VOL 331 SCIENCE

# TEAM and SUPPORT

## Students

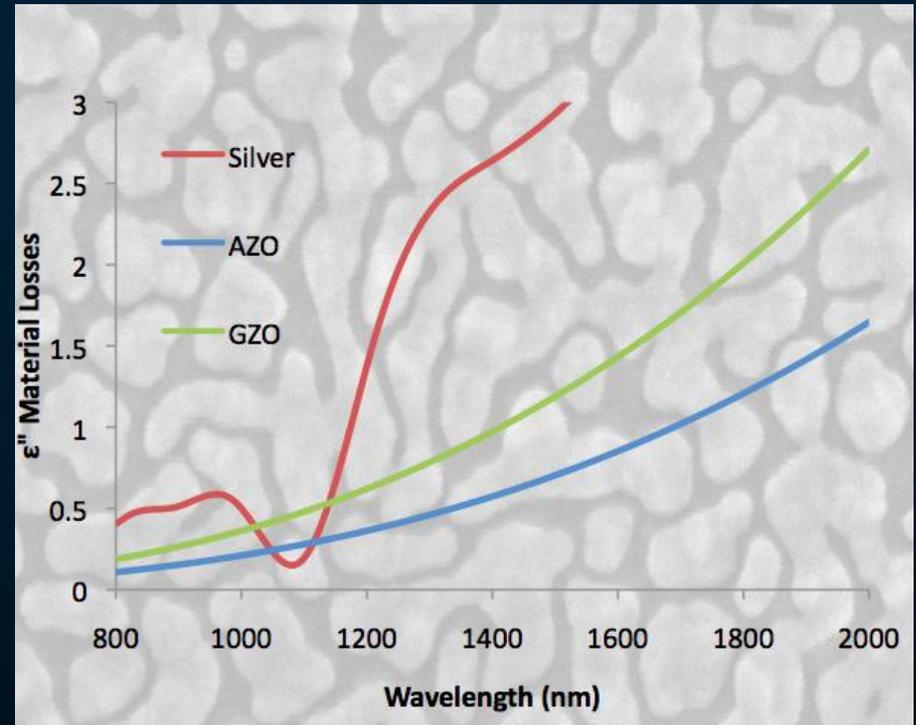
- Gururaj Naik
- Jongbum Kim
- Paul West
- Naresh Emani

## Collaborations

- Prof. T. Sands (Purdue)
- Prof. V. Shalaev (Purdue)
- Dr. A. Kildishev (Purdue)

## Support

- ONR-MURI, ARO



THANK YOU!