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Low-Loss Infrared Metamaterials: New Architectures and Unconventional Materials

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Outline

- Metamaterials: Design rules and functionality
- A lithographic approach to 3-D structures
- Unconventional materials for IR metamaterials
  - Polaritonic materials
  - Structure-property relations in MgO thin films
- Strategies for low loss metamaterials
  - Dielectric resonators

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Concept of Metamaterials

Electromagnetic Properties

ε – permittivity – measure of electric polarizability – electric charge separation

μ – permeability – measure of the magnetic polarizability – charge circulation

Index of refraction $n = \sqrt{\varepsilon\mu}$

Natural Material

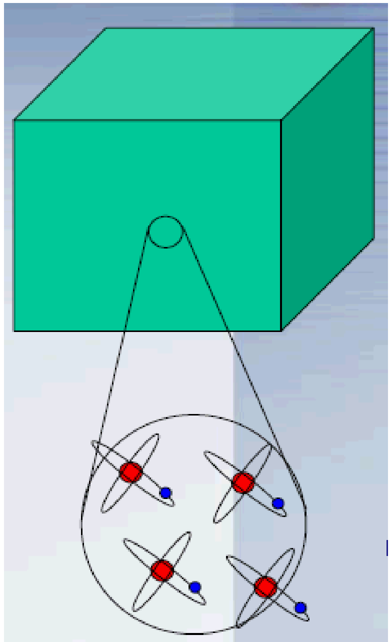
ε – polarizability

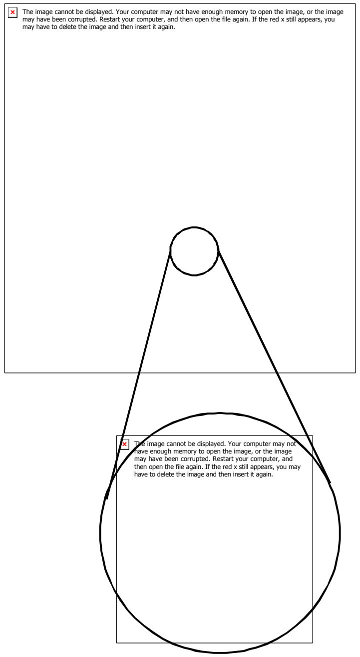
μ – QM spin

Metamaterial

ε – constituent material ε

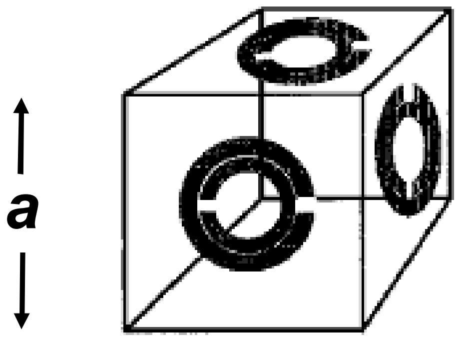
μ – induced charge circulation





3D Metamaterials

- 3D is “easy” in the RF
- Would like to make the same structures in the IR



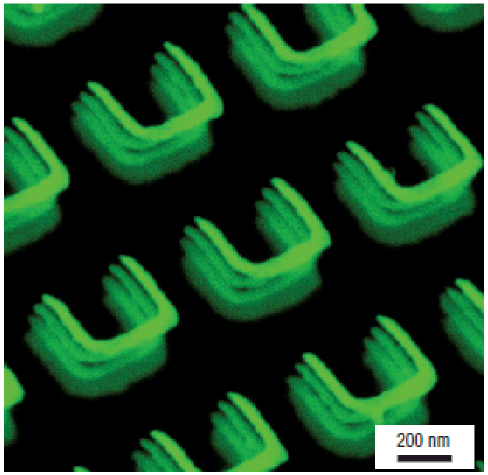
Pendry, IEEE Trans on Microwave Theory and Techniques **47**, #11, 2075 (1999)

Schultz, Science **292**, 77, (2001)

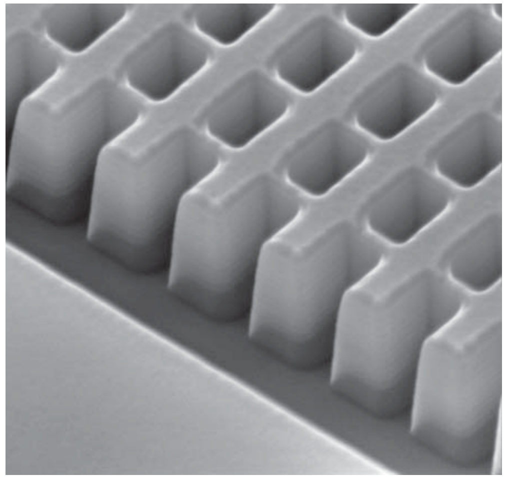
- Geometrically isotropic cubic unit cell.
- Unit Cell Dimensions ~ cm
- Trace Linewidths ~ mm
- SRRs oriented along each coordinate axis

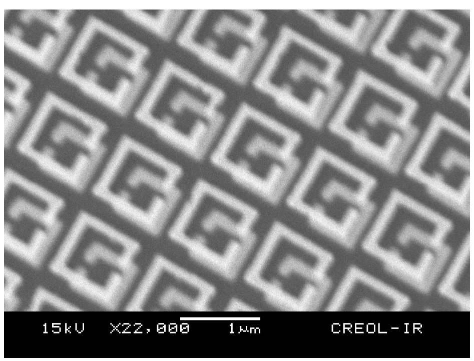
Current State of the Art in IR 3D MM fabrication

Stacked Planar



Giessen- Nat. Mater. **7**, 31, (2008)


Zhang, Nature **455**, 376 (2008)



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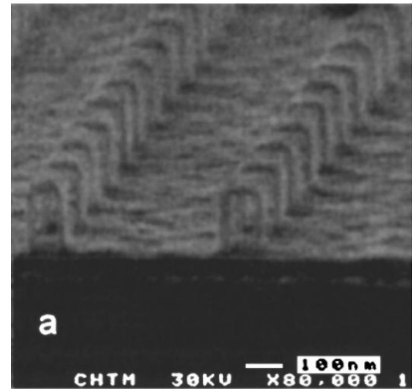
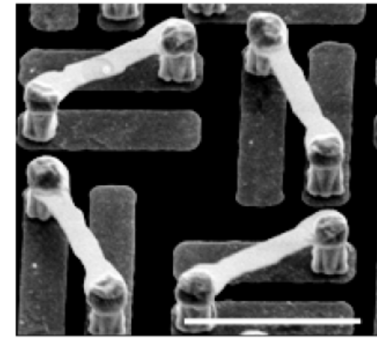
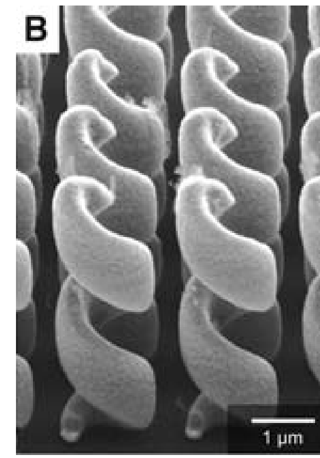
Out-of-Plane Current



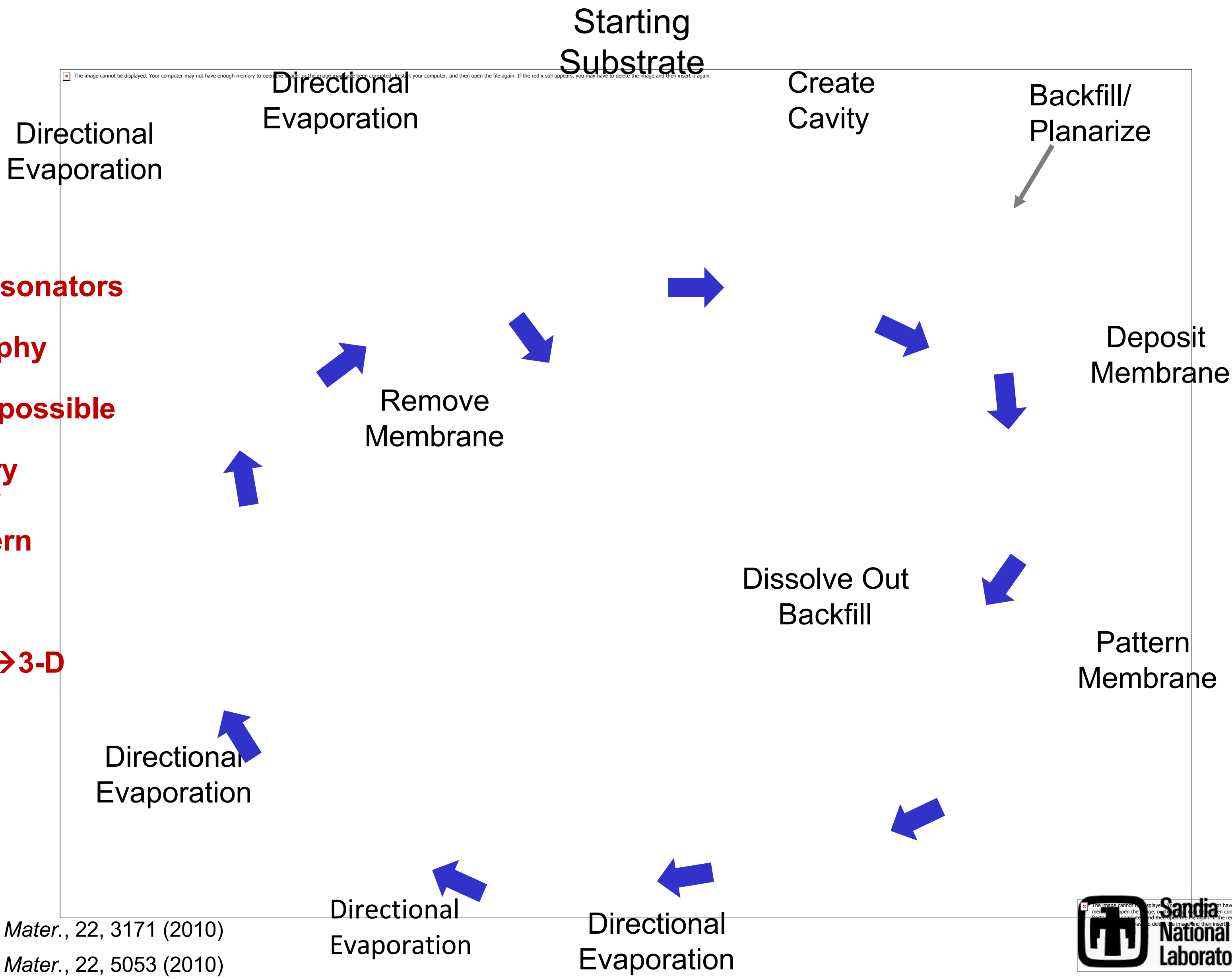
Brueck - JOSA B **23**, 3 434, (2004)


Zhang, PRL **102**, 023901, (2009)



Wegener, Science **325**, 1513-1515, (2009)

Membrane Projection Lithography: MPL



- **Out-of-plane resonators**
- **Planar lithography**
- **Many patterns possible**
- **Cavity geometry independent of resonator pattern**
- **Scalable**
- **Layer-by-layer→3-D**

D.B. Burckel, *et al.*, *Adv. Mater.*, 22, 3171 (2010)

D.B. Burckel, *et al.*, *Adv. Mater.*, 22, 5053 (2010)

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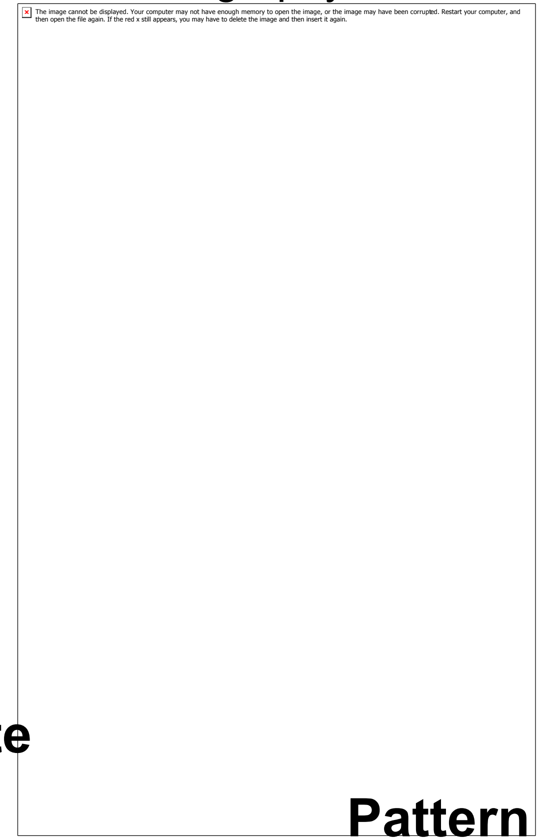
3D Membrane Projection Lithography (3D MPL)

Cavity formation
Optical lithography
Lithography + etch

Evaporations
One to Five
(Normal + 4 Angled)

Cavity shape
Cube
Cylinder
Grating

Lattice material
SU-8
Polyethylene



Pattern
SRRs
Any non-closed loop, structurally stable design

Backfill
Polyimide
SU-8
Oxide

Membrane
PMMA
Positive/Negative resist
hardmask (oxide, nitride)

Substrate
Silicon
Quartz
GaAs
BaF

Pattern Lithography
E-beam (JBX-9300FS)
Optical
Interferometric

D.B. Burckel, *et al.*, *Adv. Mater.*, 22, 3171 (2010)
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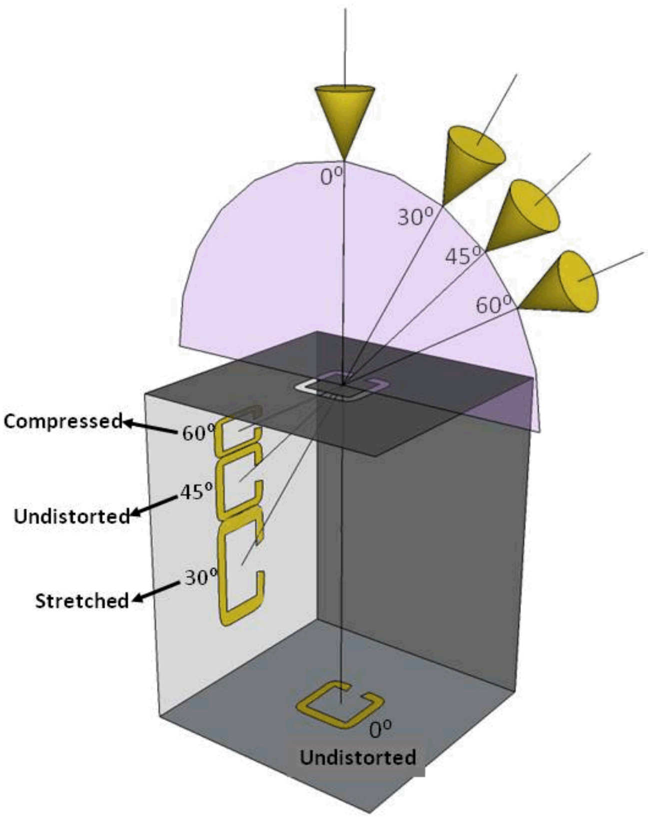
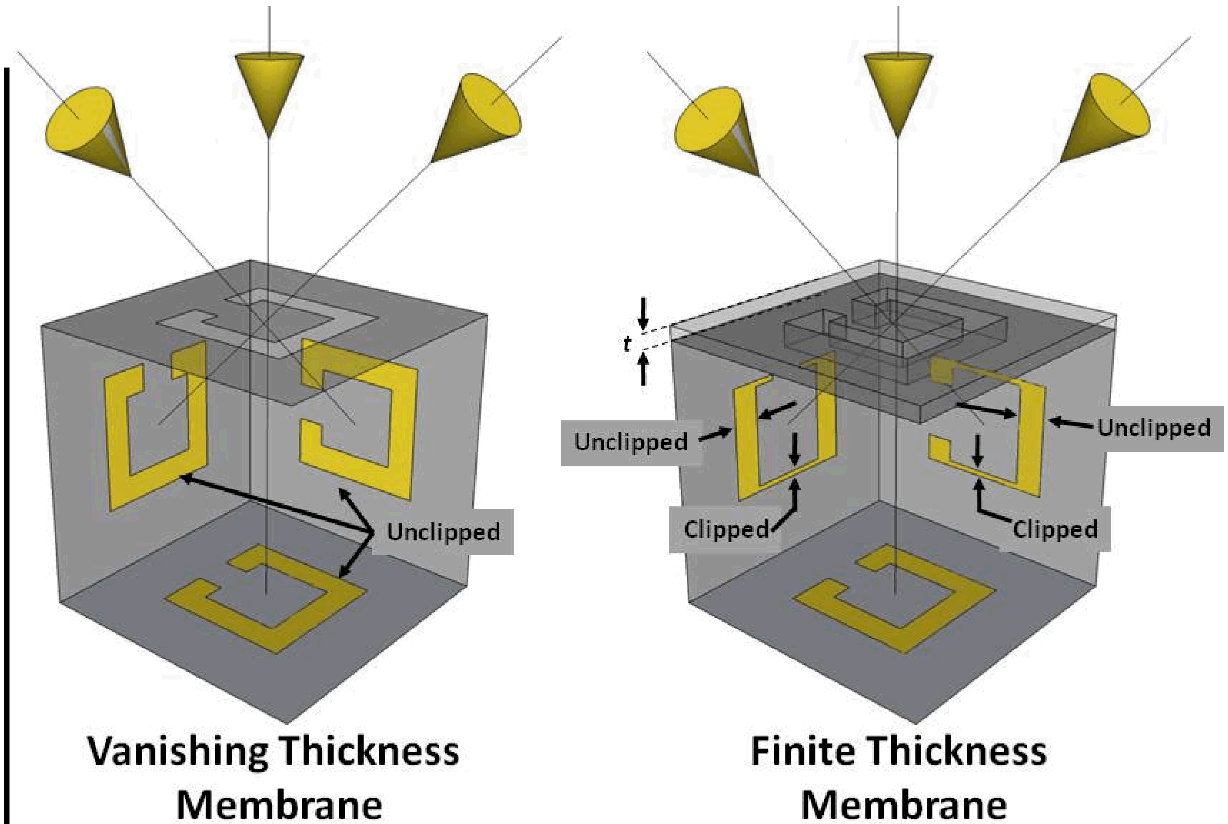
Sources of Pattern Distortion

Projection at 45^0 Preserves Pattern Shape in Cubic Geometries



Real Membranes result in Linewidth Clipping



- Least distortion with thin membranes
- Can pre-compensate membrane pattern

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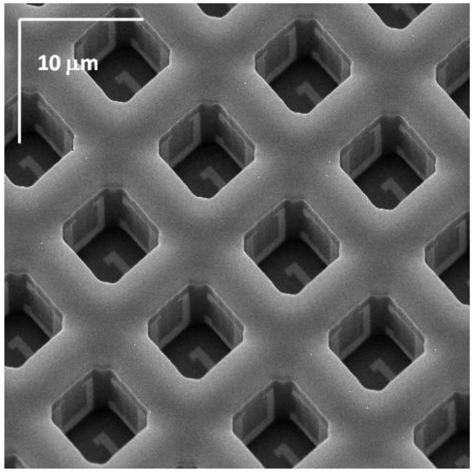
Cubic 3D Metamaterials via MPL



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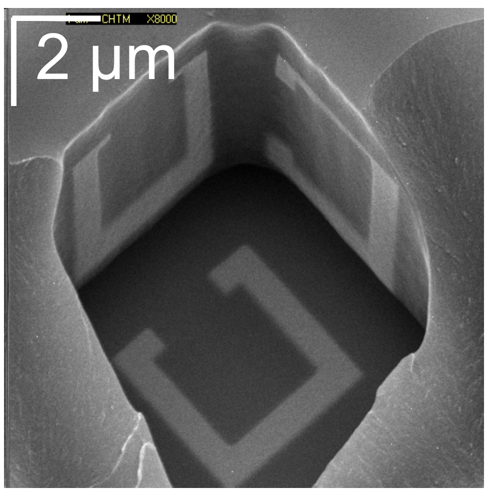

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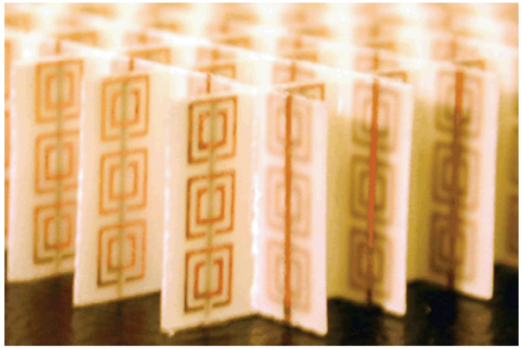
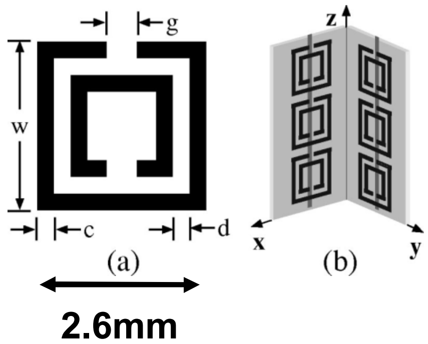
- **Unit Cell Dimension = 14 μm pitch**
- **SRR Dimension = 8 μm**
- **Resonance wavelength ~ 50 μm**

- **Unit Cell Dimension = 6 μm pitch**
- **SRR Dimension = 3 μm**
- **Resonance wavelength ~ 22 μm**

For Reference

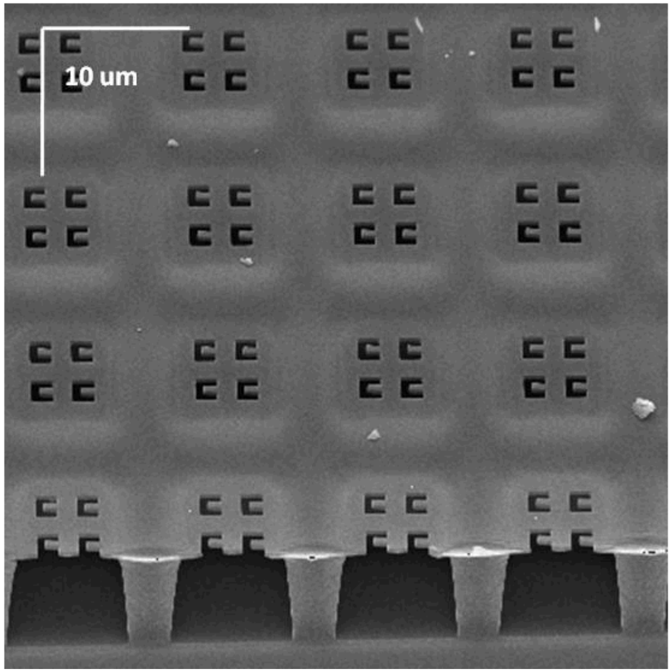


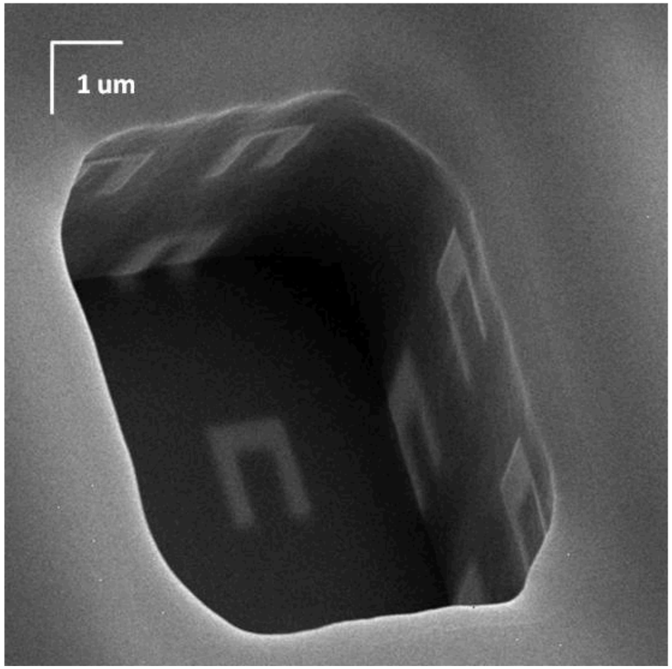
Schultz, Science **292**, 77, (2001)



- **Unit Cell Dimension = 5mm**
- **SRR Dimension = 2.6mm**
- **Operating Wavelength = 3cm**

More 3D MPL Fabrication Results





Patterned PMMA membrane suspended over and aligned to boxes formed in SU-8.

- Gold SRR patterns deposited onto the five box surfaces after liftoff.
- Predictable distortions of the patterns on the vertical walls.

D.B. Burckel, *et al.*,
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D.B. Burckel, *et al.*,
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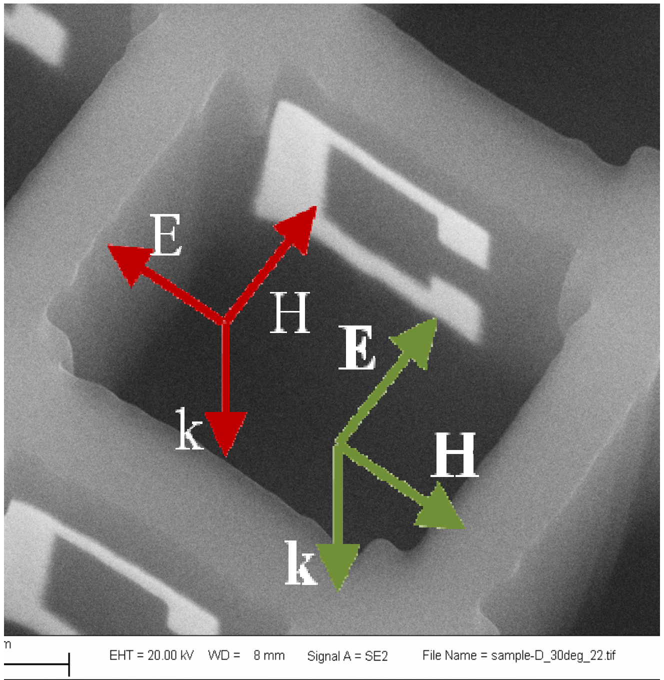
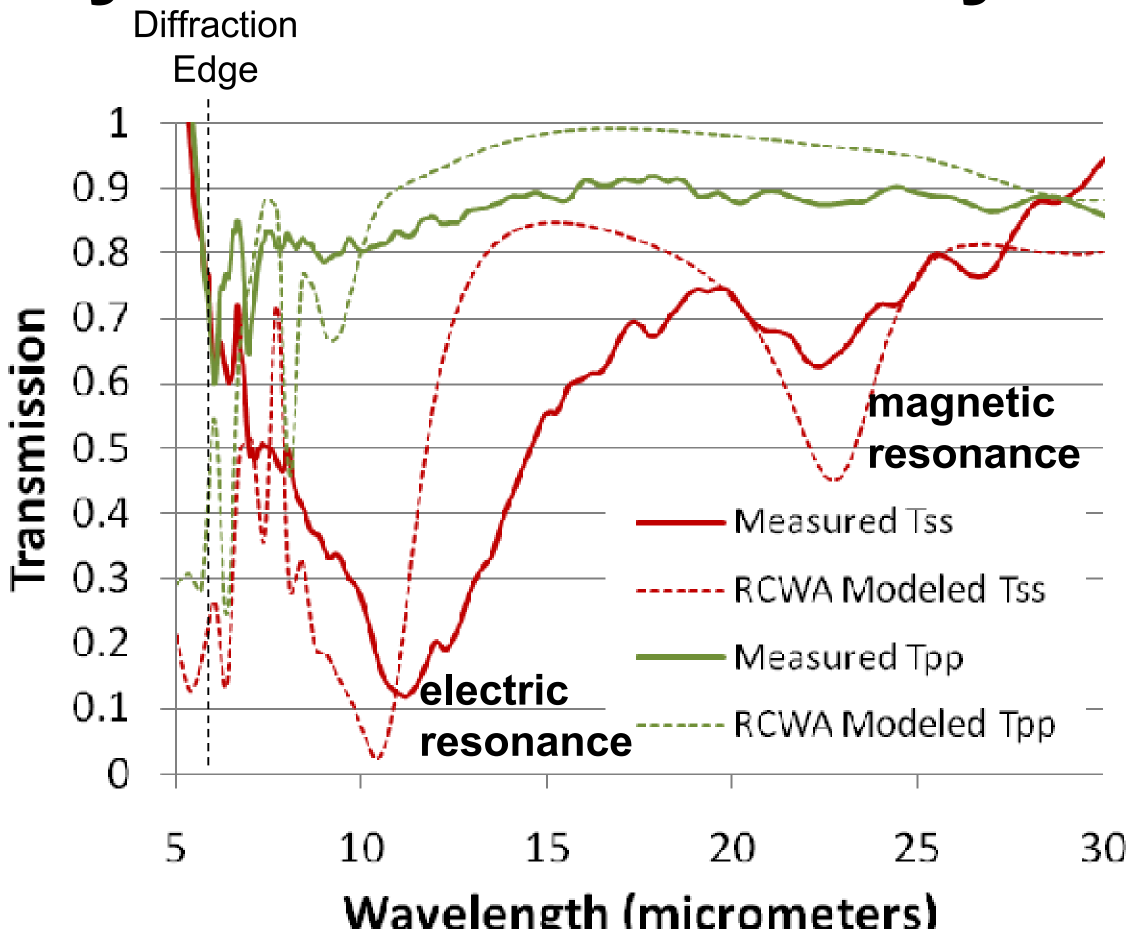
Polarized Transmission of Magnetically Excited SRR Array





S-polarization

- *B*-field excites lowest SRR resonance --- magnetic excitation
- *E*-field excites second order resonance --- electric excitation

P-polarization

- can't couple to any SRR resonances

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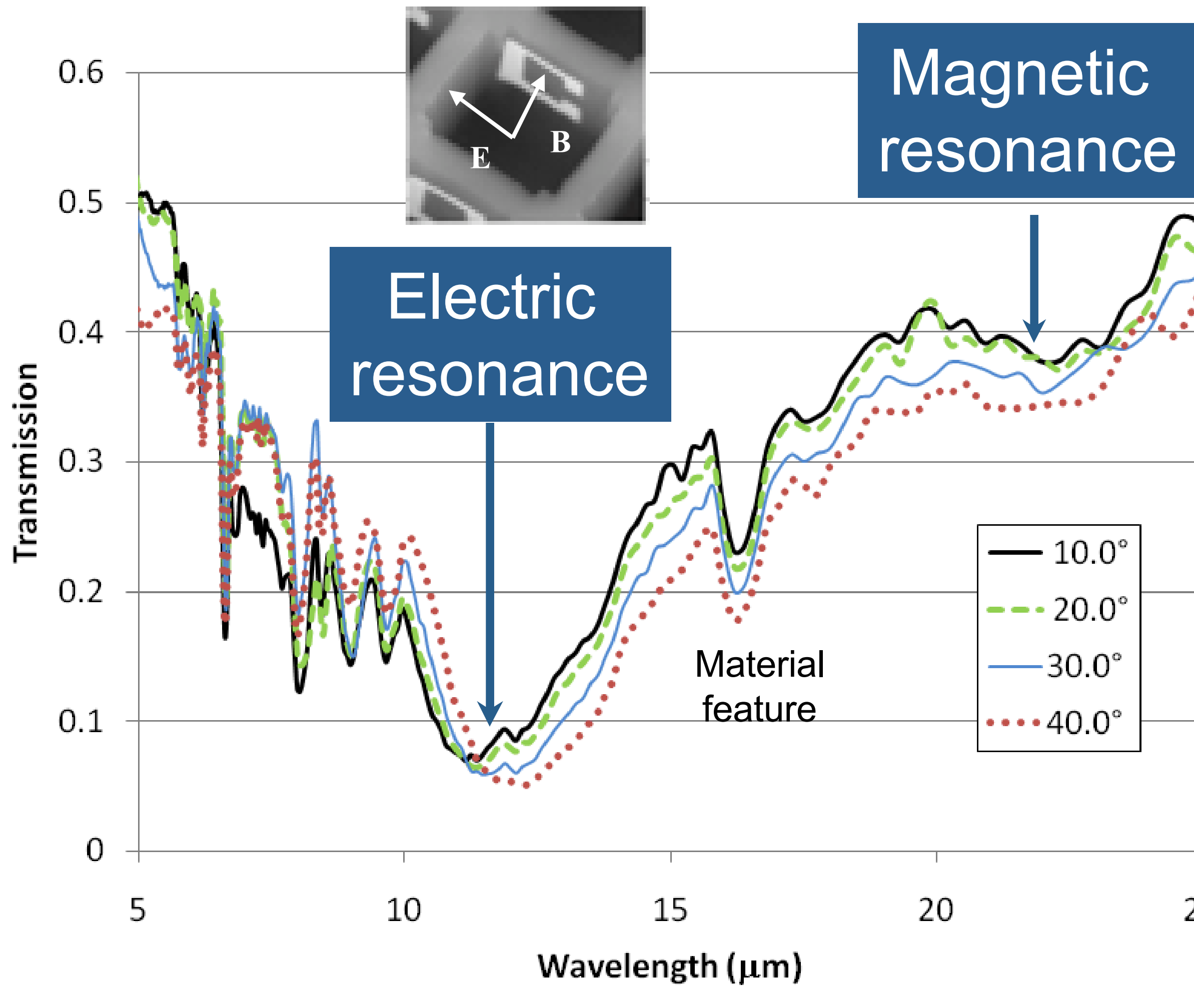
Angle Dependence of Spectral Response

Characterization Techniques

- Hemispherical
- Directional
- Transmission
- FTIR
- Variable Angle Spectroscopic Ellipsometer



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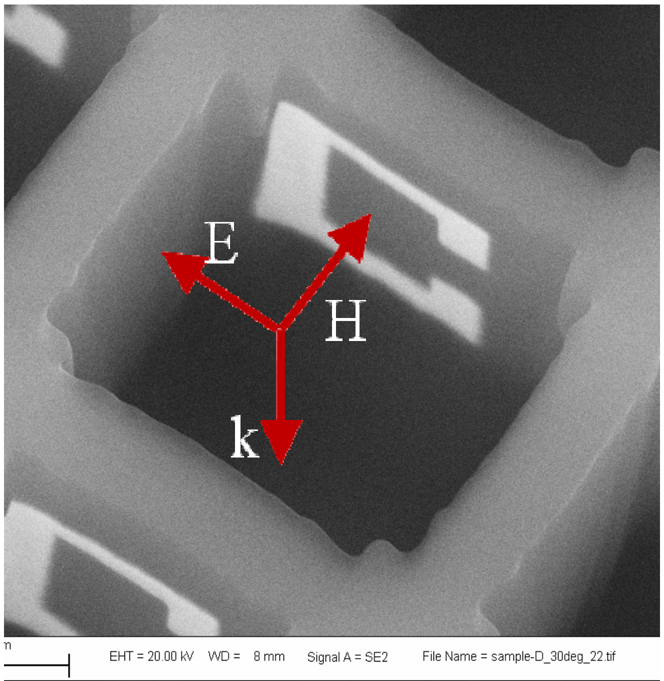
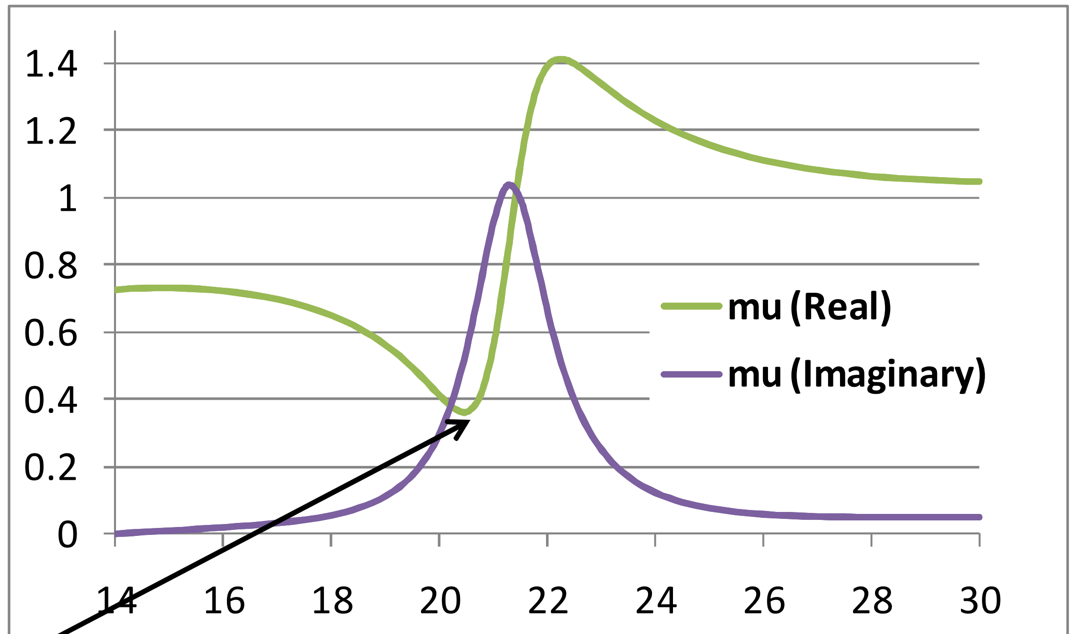
Extracted permeability for modeled array with SRRs on single wall





$\mu = 0.37 + i\,0.56$

Optimized Q-factor for the resonator along with increased packing fraction is expected to improve the strength of the resonance and increase the range of permeability tuning

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Polarized Transmission of 5-SRR Unit Cell

Composite SRRs

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T_{pp}

T_{ss}

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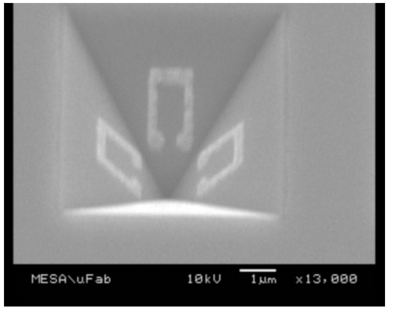
Summary & Future Directions

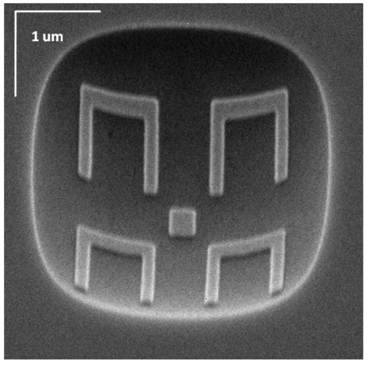
Membrane Projection Lithography (MPL) enables 3D metamaterials

- large area
- out of plane resonators
- can choose orientation & placement of resonators for desired excitation
- demonstrated magnetic response at normal incidence at λ = 22 µm

Currently:

- optimizing EM design for maximal response: higher Q therefore negative μ
- Begin exploring inclusion designs aimed at tuning permittivity (ε).
- currently on track to demonstrate multi-layer MPL and LWIR structures.





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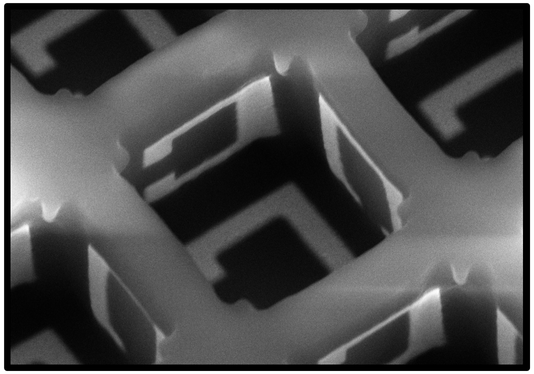
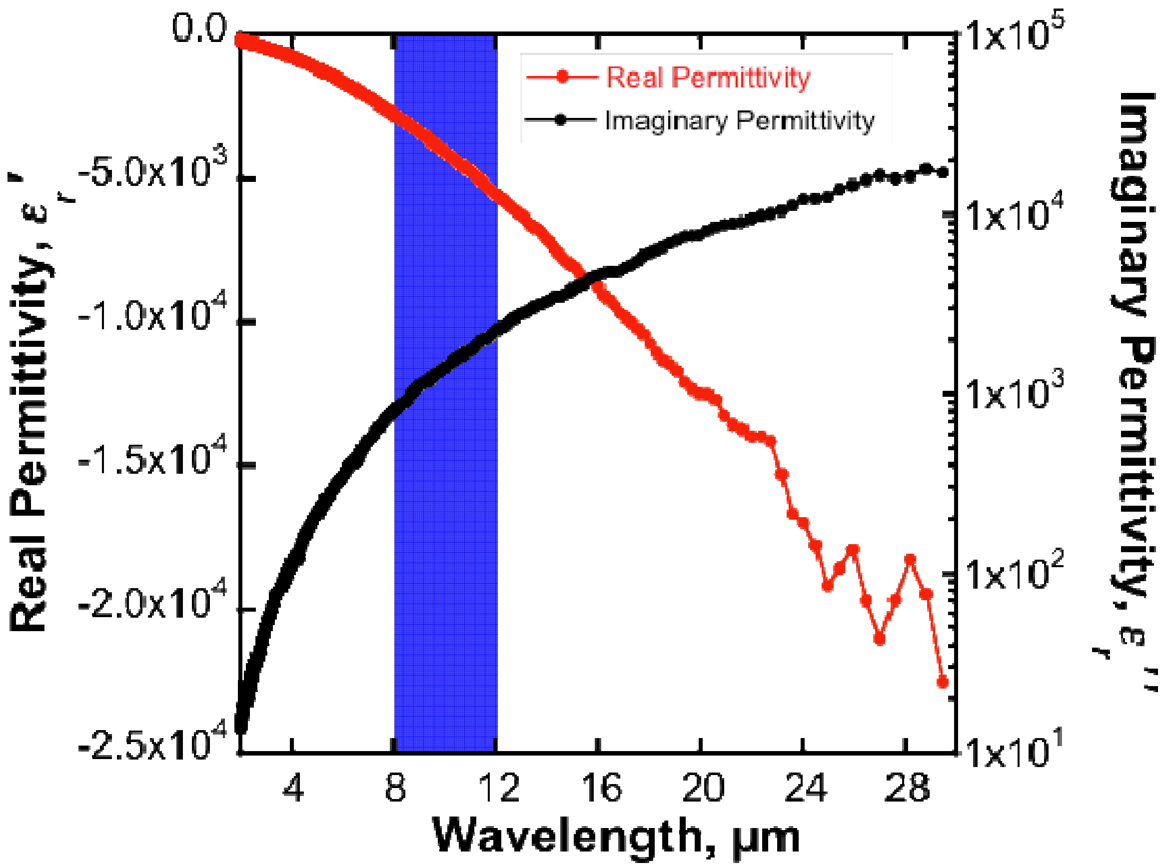
Traditional Metamaterials:



IR Optical Response of Gold

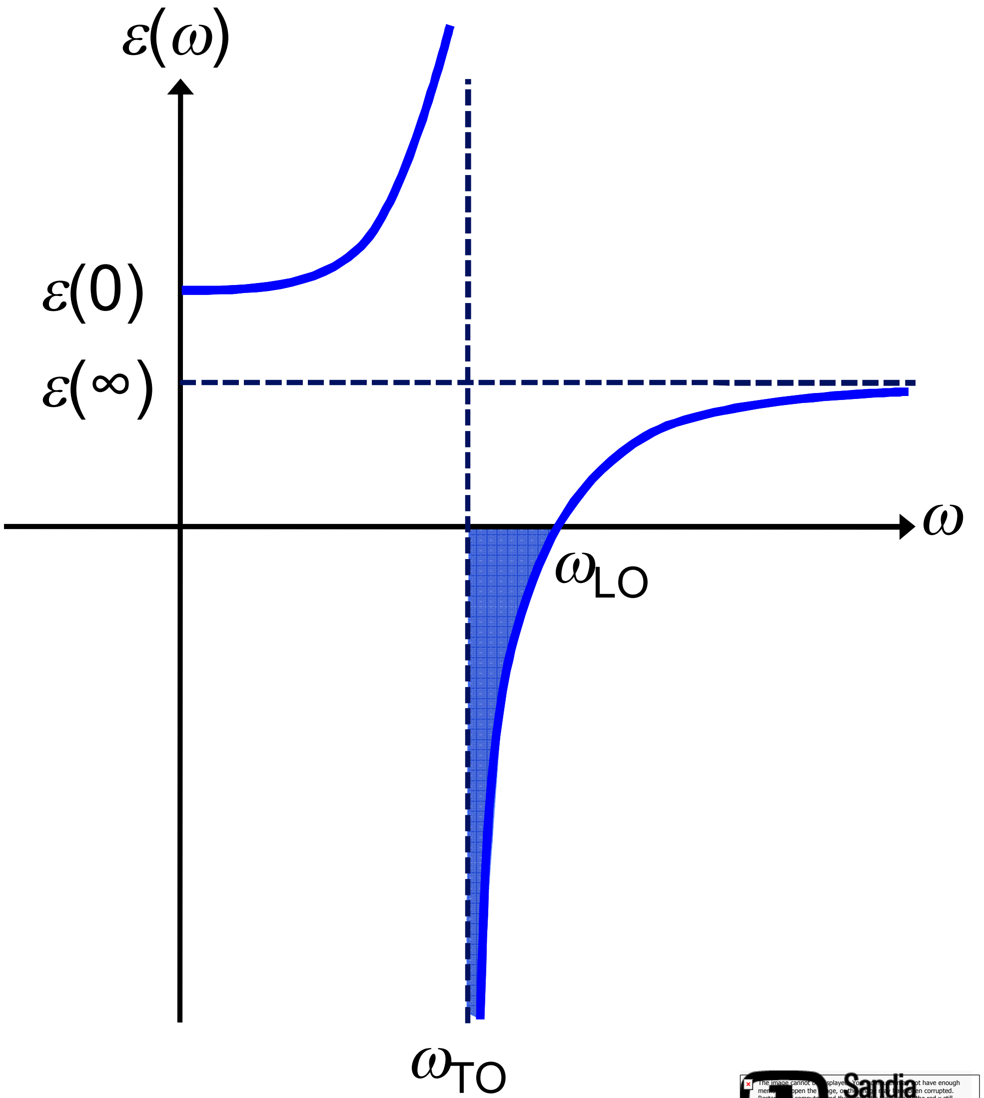


- Traditional metamaterials designed with metallic elements
- Metal conductivity decreases substantially approaching optical frequencies
- Losses high in optical frequencies (Ohmic)

Alternative Low-Loss Materials

Polaritonic (*Reststrahlen*)Materials

- All ionic or partially ionic materials
- Strong phonon-photon coupling in IR
- Negative permittivity between *TO* and *LO* modes
- Can tune frequency through atomic mass or bonding



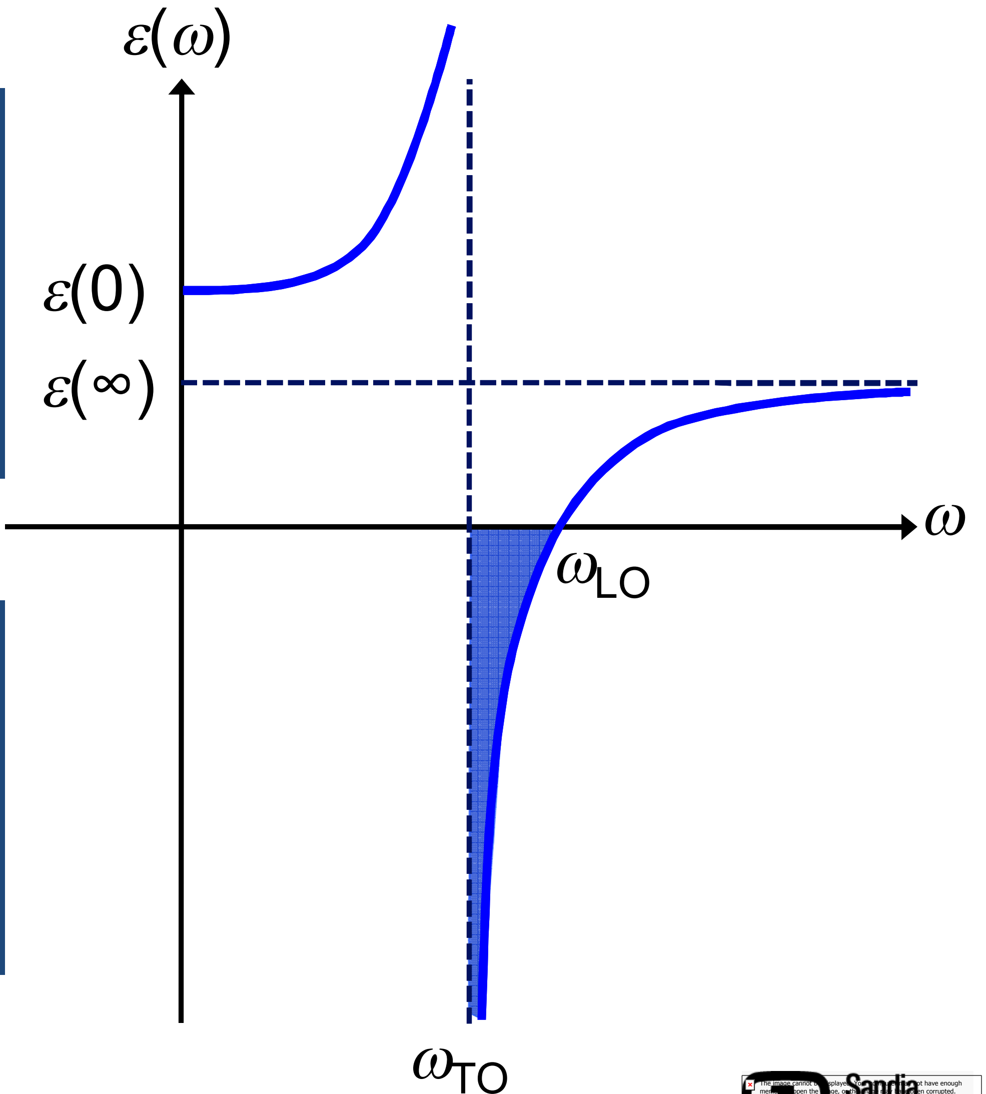
Alternative Low-Loss Materials

Desired Properties

- Strong interaction with electromagnetic waves
- Low dissipation
- Compatible with lithography

Potential Alternative Materials

- Many materials with *Reststrahlen* bands that span the MIR
- *Structure-property relations?*



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Structural Effects on IR Response

Model System: MgO

- *Reststrahlen* Band 15-25 µm
- Simple structure and two ion basis
- Simple phonon dispersion
- Multiple methods to control crystallinity

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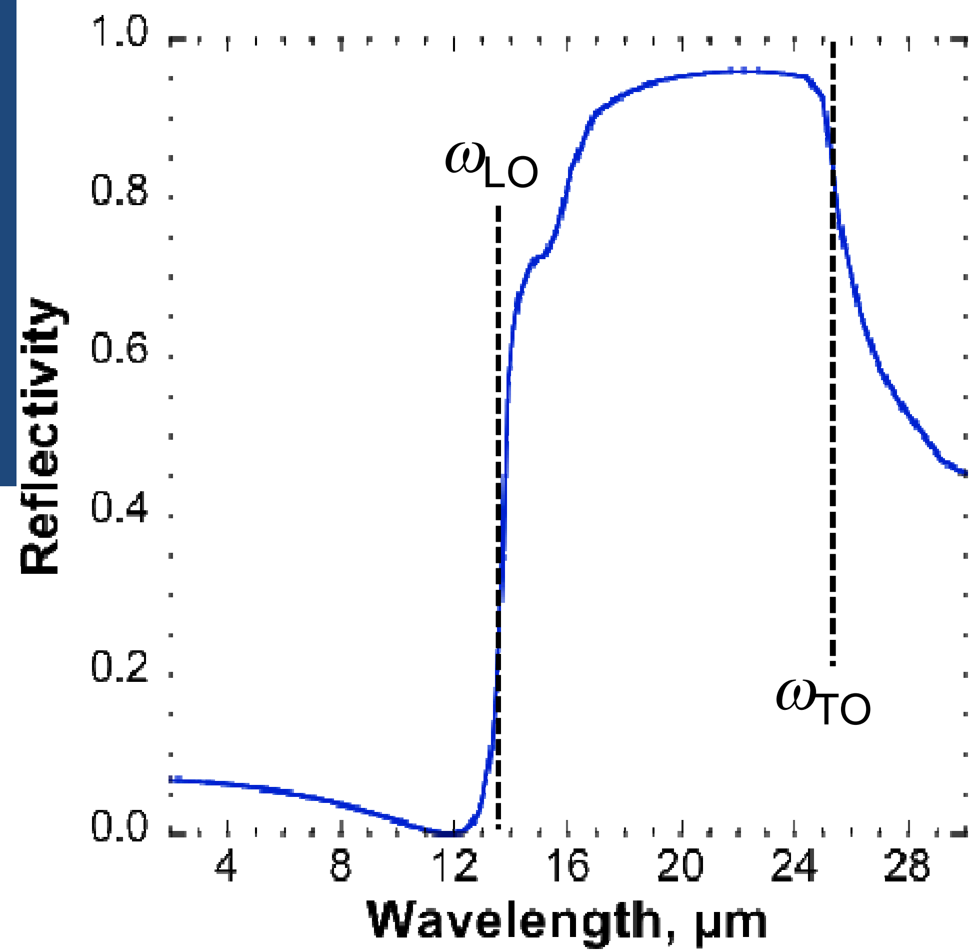
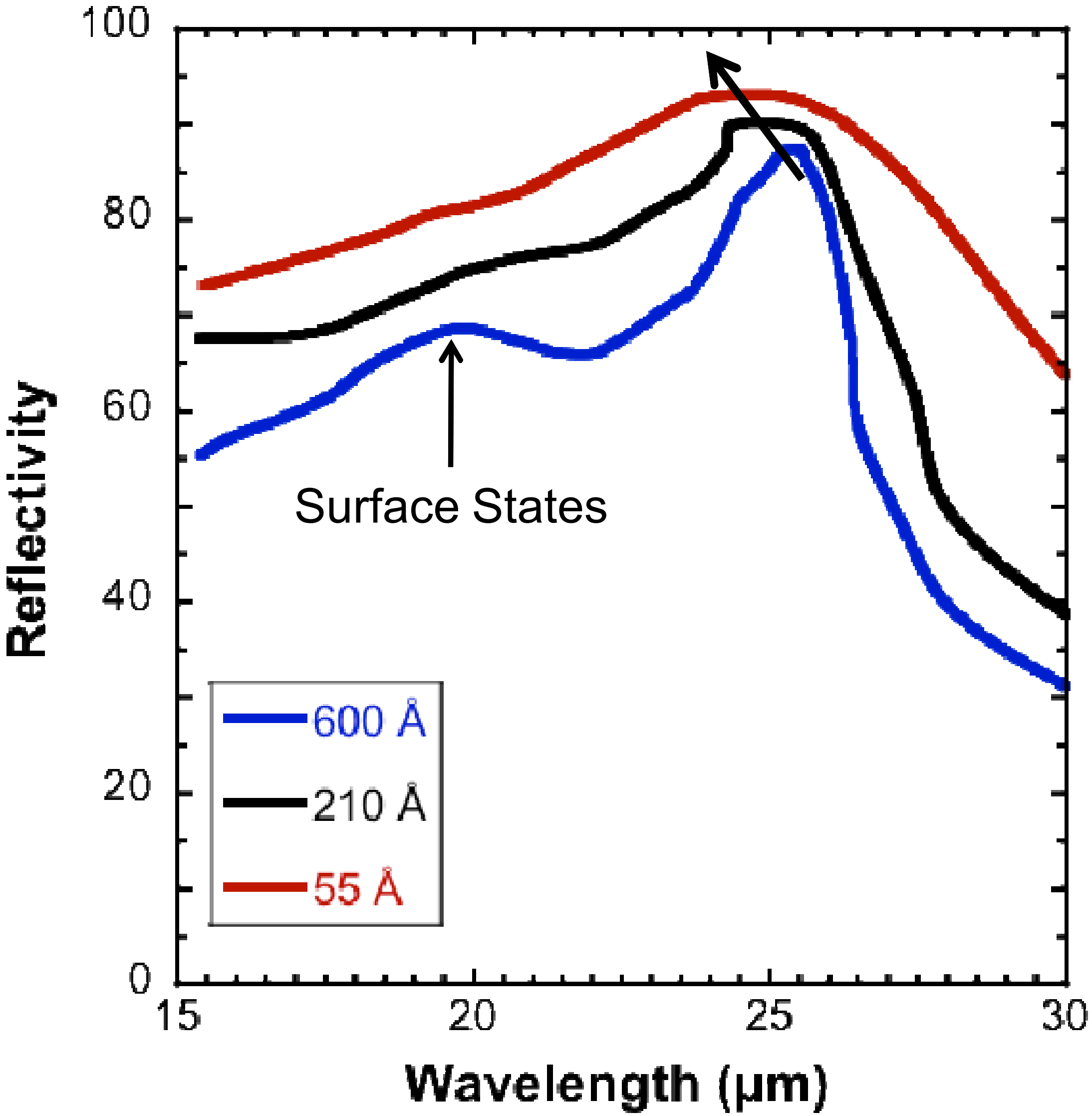


Particle Size Effects on IR Response of MgO



Particle Size Effects

- Reduced particle size broadens *Reststrahlen* response
- Apparent shift in peak reflectivity (ω_{TO})
- Surface modes present

Adapted From: Luxon *et al. Phys. Rev.* **188** (3) 1345 (1969)

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MgO Crystallinity Modification

- 200 nm thick films deposited by RF sputtering on 001-silicon
  - 30° off-axis geometry
  - Argon atmosphere
  - Room temperature substrate
- Thermal anneals used to modify grain/crystal size
  - 200-800°C in air × 1 hour

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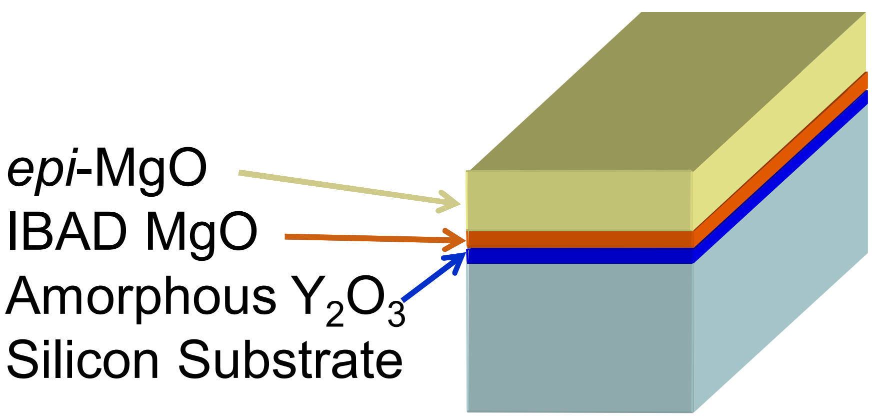
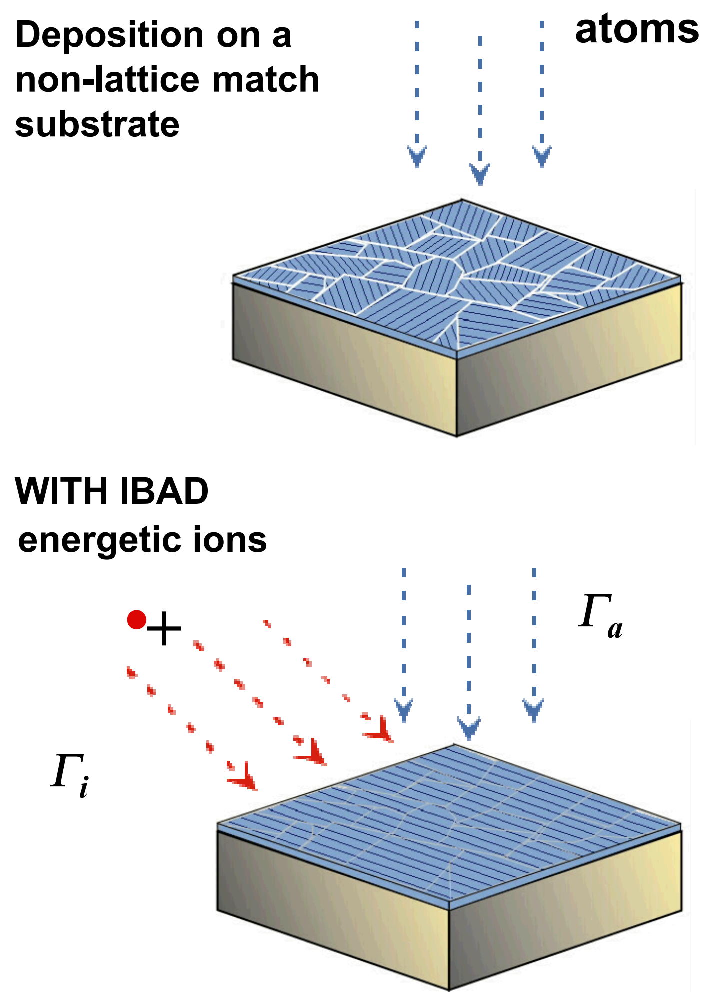
MgO Crystallinity Modification

- Ion-beam assisted deposition
  - (001)-textured material

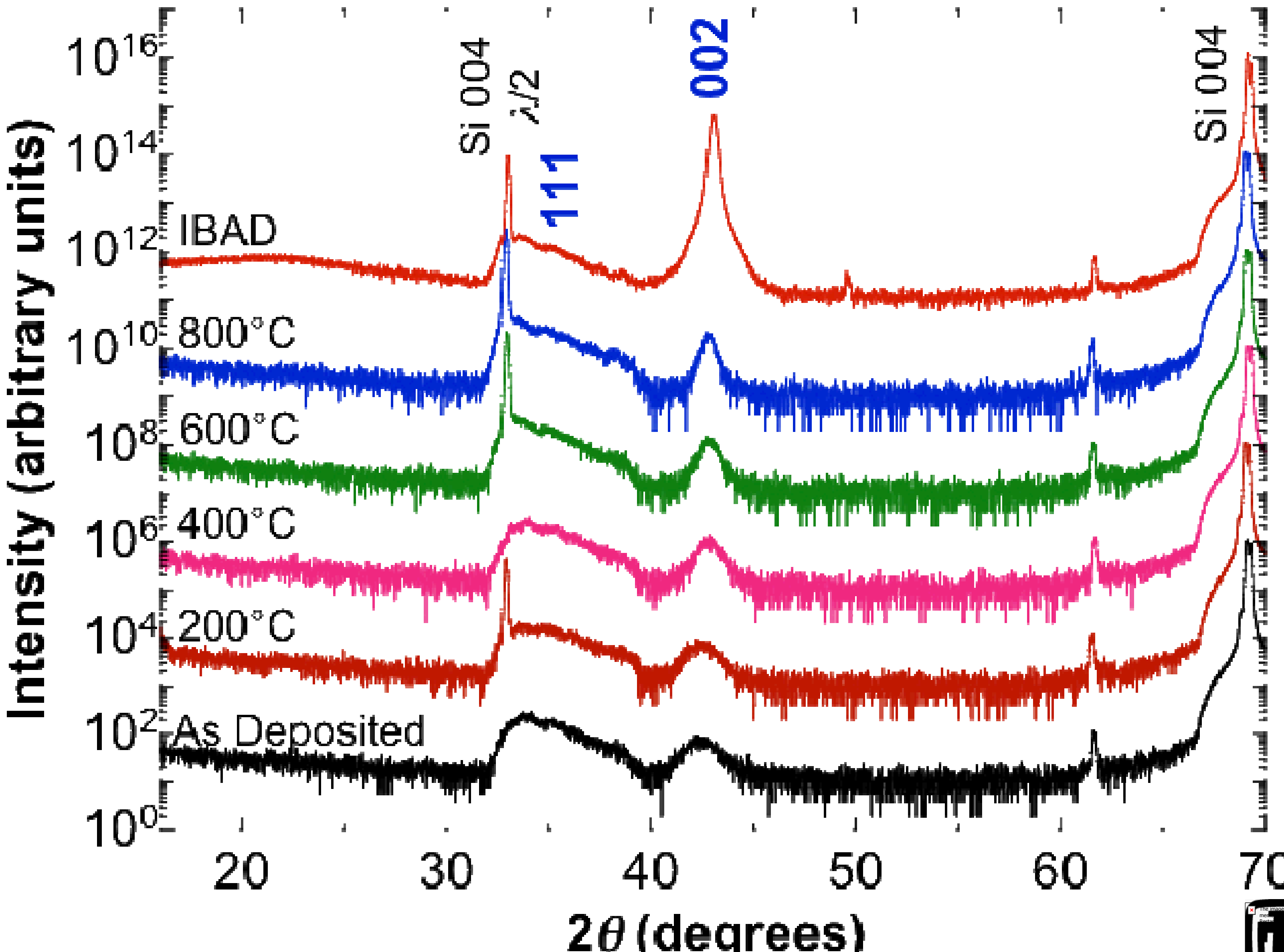


- $\Delta\omega_{002}$ ~ 1.2°
- $\Delta\phi_{220}$ ~ 4.7°



*Vlad Matias, Los Alamos National Laboratory

MgO Structural Properties



J.F. Ihlefeld, *et al.*, *Appl. Phys. Lett.*, 97, 191913 (2010)

Microstructure Characterization

As-Deposited

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As-Deposited

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400°C

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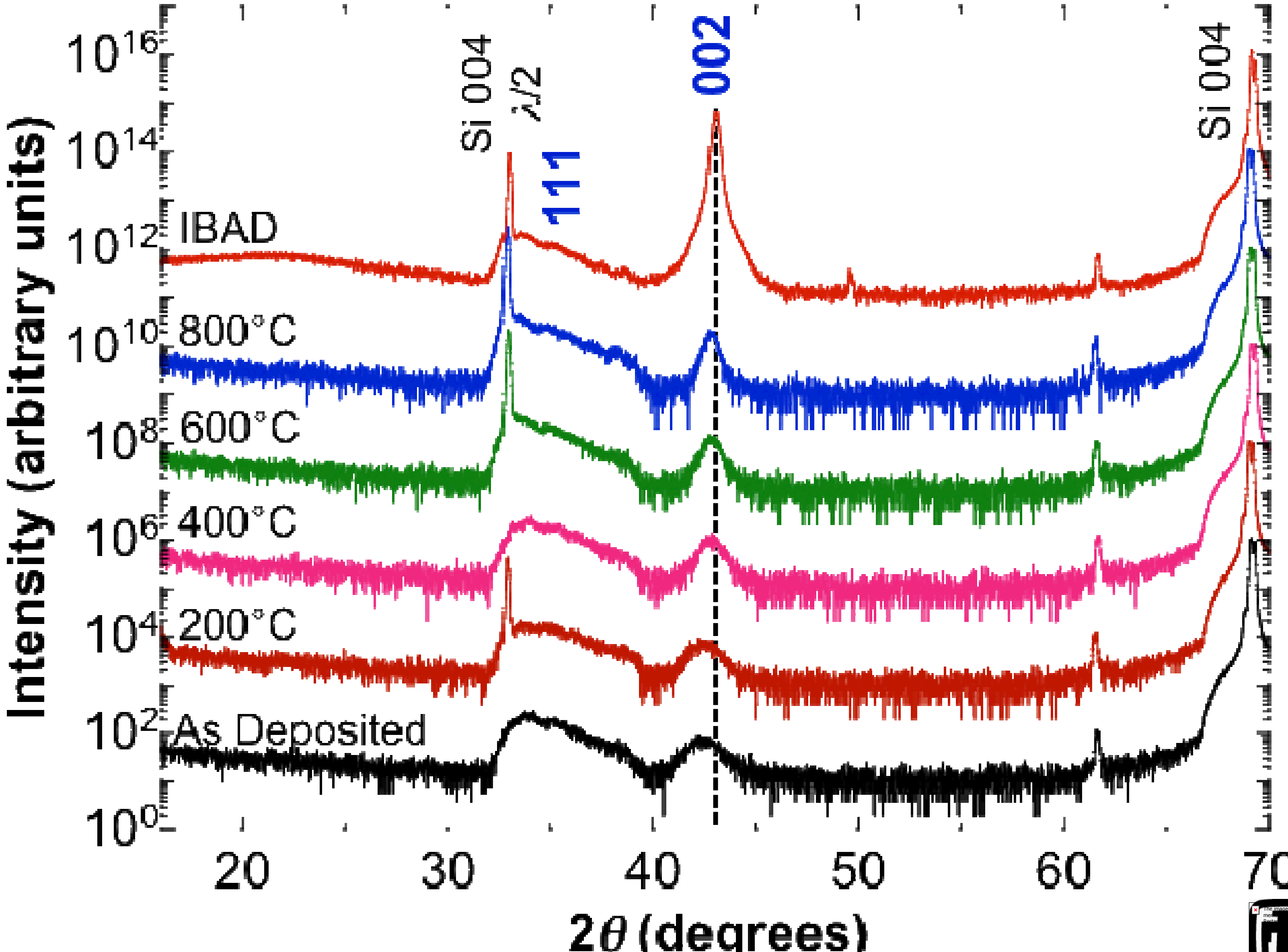
800°C

Particle Size Effects

- Dense microstructure
- Columnar morphology
- No change in apparent grain size with process temperature
  - ~75 nm
  - 6.8 ± 0.3 nm roughness

MgO Structural Properties



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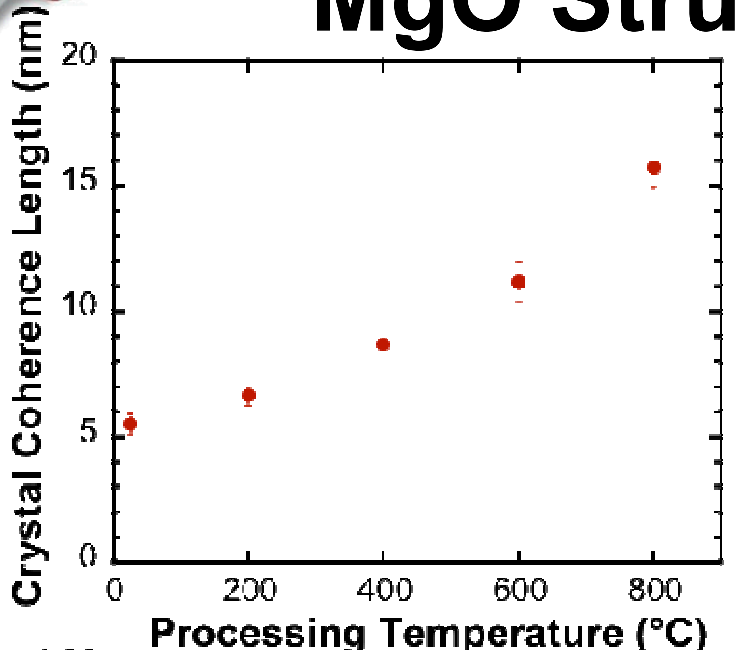
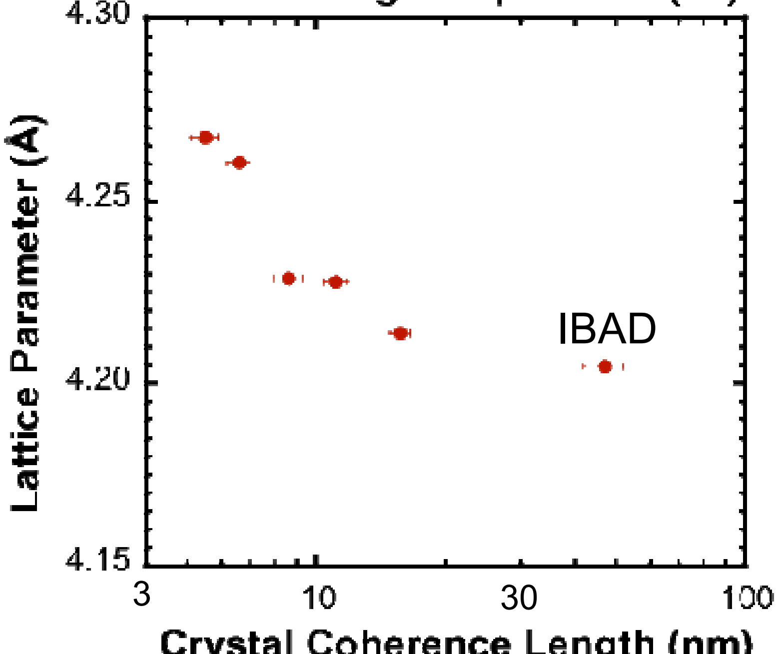
MgO Structural Properties





- Strong scattering dependence on processing temperature
- Lattice parameter decrease with crystallinity*
  - Increased grain/subgrain boundary area
  - High degree of disorder

*$\cos^2\theta$ / $\sin\theta$ height error correction

P. Scherrer, *Nachr. Ges. Wiss. Goettingen, Math. Phys.*, **2**, 96 (1918)
J.F. Ihlefeld, *et al.*, *Appl. Phys. Lett.*, 97, 191913 (2010)

Dark-Field and High-Res TEM

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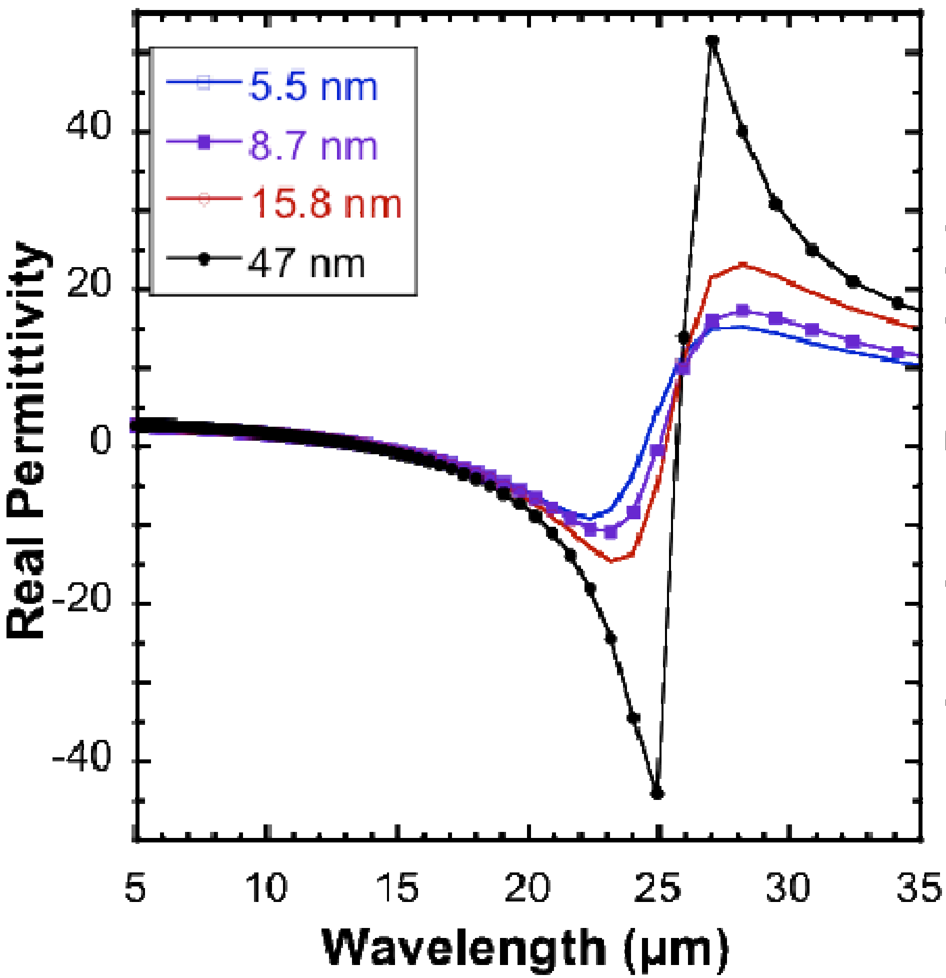
As-Deposited

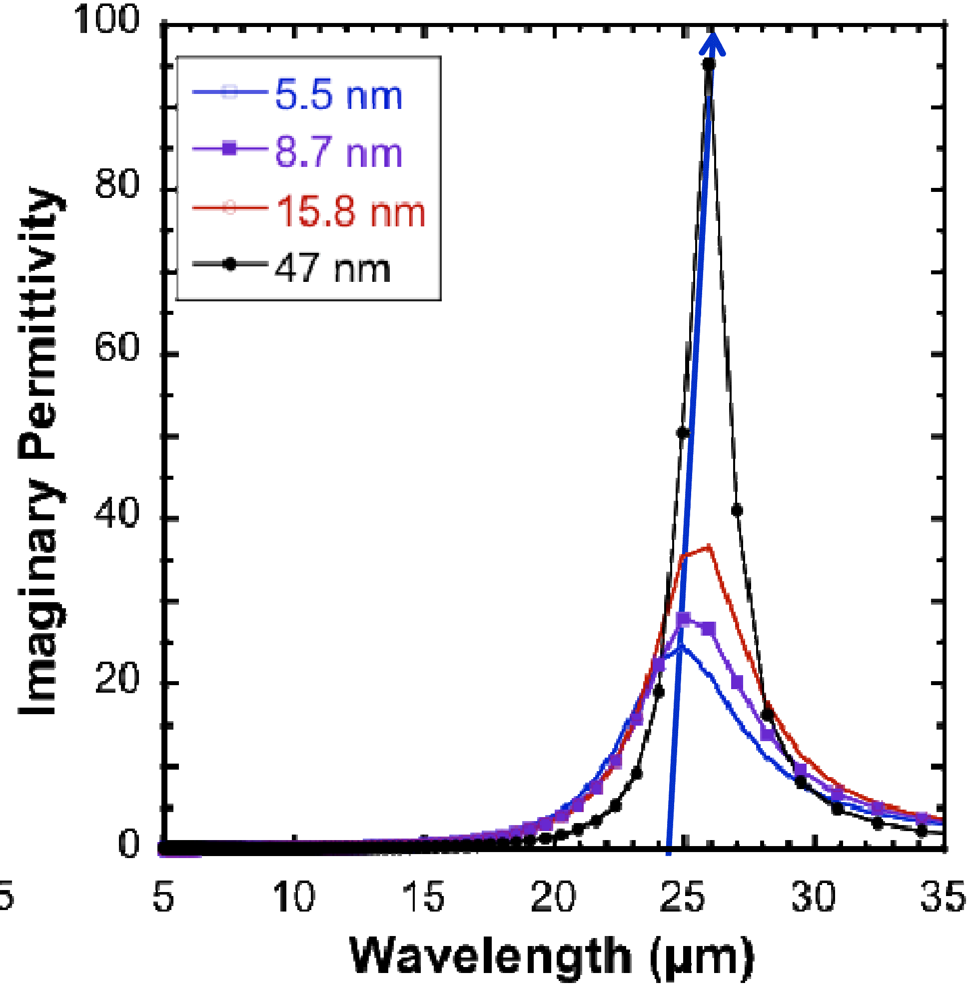
400°C

800°C

As-Deposited

MgO Optical Properties





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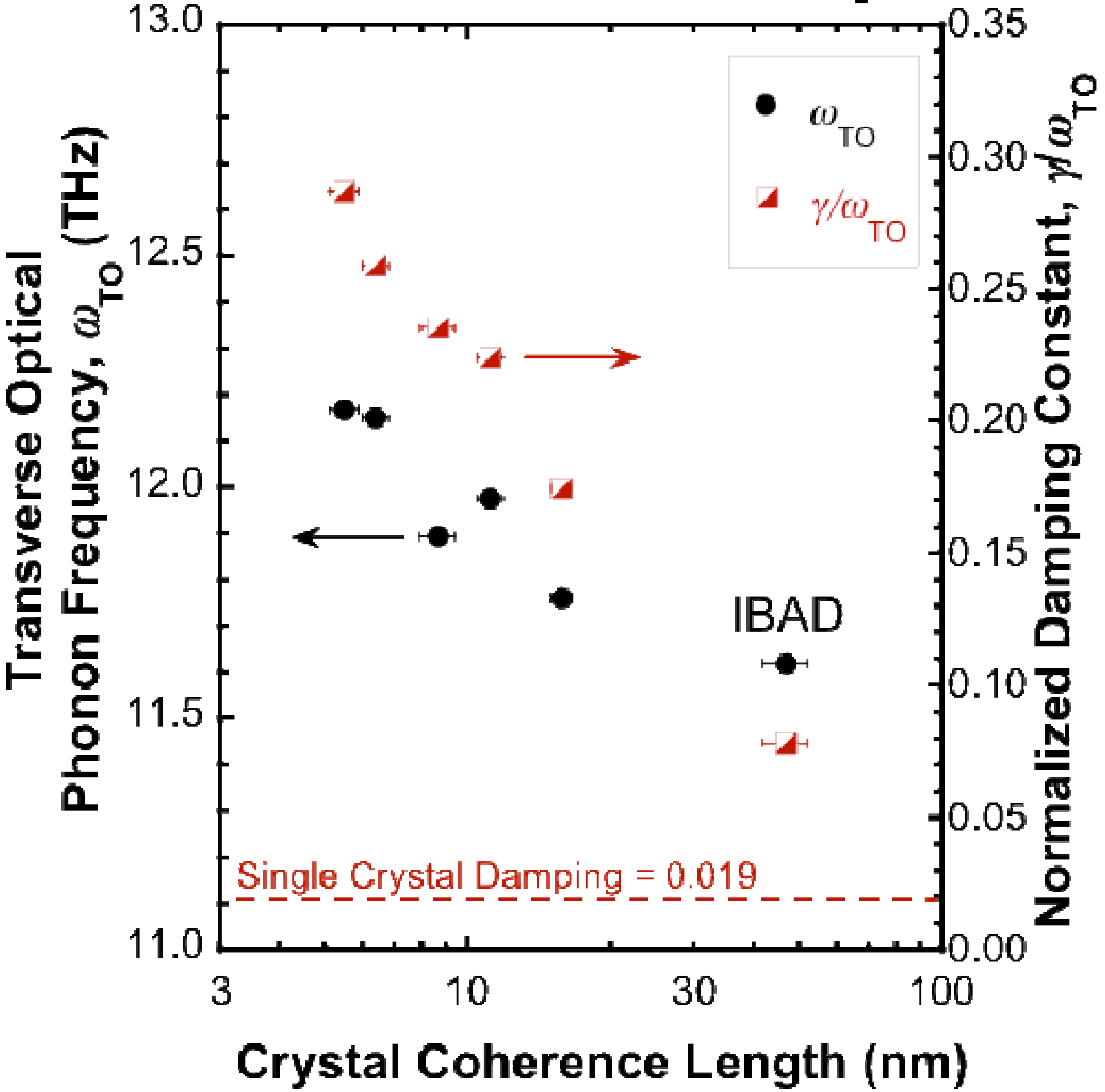
Crystallinity Effects on IR Response



- Response fit with modified classic oscillator model:

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\left[1 + \left(\frac{\omega^2}{\omega_{TO}^2}\right)\right] - i\left(\frac{\omega\gamma}{\omega_{TO}^2}\right)}$$

- Crystal coherence has strong effect on phonon damping constant
- Shift in ω_{TO} concomitant with lattice parameter

J. Jasperse, *et al.*, *Phys. Rev.*, 146:2, 526 (1966) J.F. Ihlefeld, *et al.*,*Appl. Phys. Lett.*, 97, 191913 (2010)

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Summary

- Crystallinity modified by thermal treatments
- Optical response strongly dependent on crystal coherence
- Increased damping with increased disorder
- Transverse optical phonon mode frequency correlates with lattice parameter
- May have broad implications for IR metamaterial design
  - Epsilon-near-zero materials
  - Reduced dimensions of resonators ($\lambda/10$)

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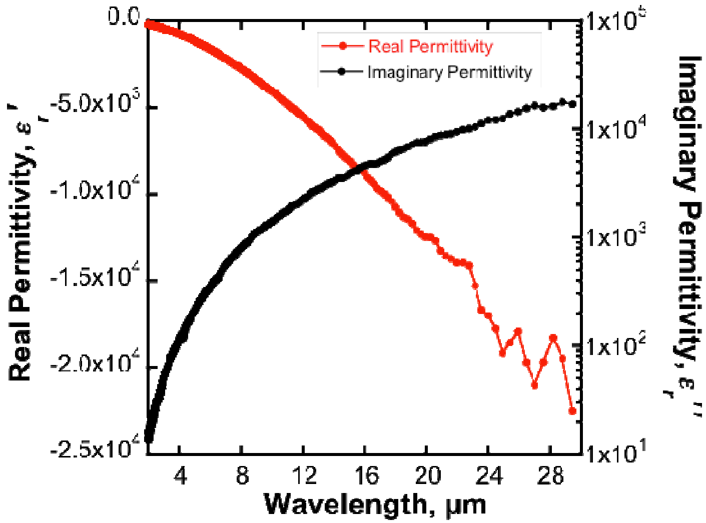
Outline

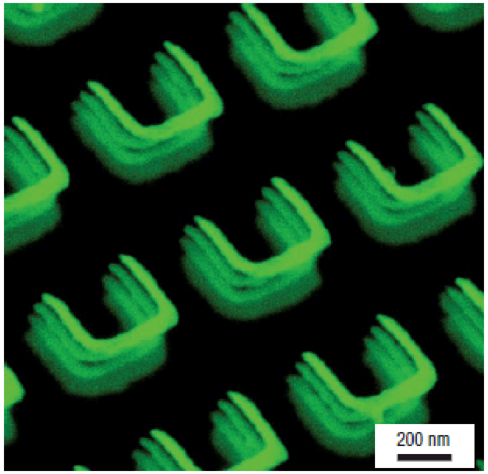
- Metamaterials: Design rules and functionality
- A lithographic approach to 3-D structures
- Unconventional materials for IR metamaterials
  - Polaritonic materials
  - Structure-property relations in MgO thin films
- **Strategies for low loss metamaterials**
  - **Dielectric resonators**

Three Primary Optical MM Limitations

1. Metallic Resonators
   – Ohmic losses
   – Broad and weak resonances
   – Less interaction with light approaching visible frequencies
2. Anisotropic Response of Metallic Resonators
   – Magnetic responses require off-axis excitation
3. Fine scale features
   – Requires high-end e-beam lithography
   – Limitations for large size samples

Need a design approach that mitigates each of these three issues





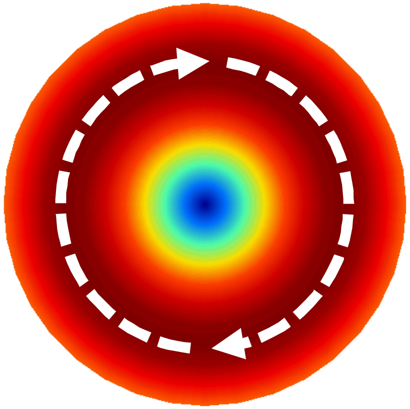
Giessen- Nat. Mater. **7**, 31, (2008)

Spherical Dielectric Resonators

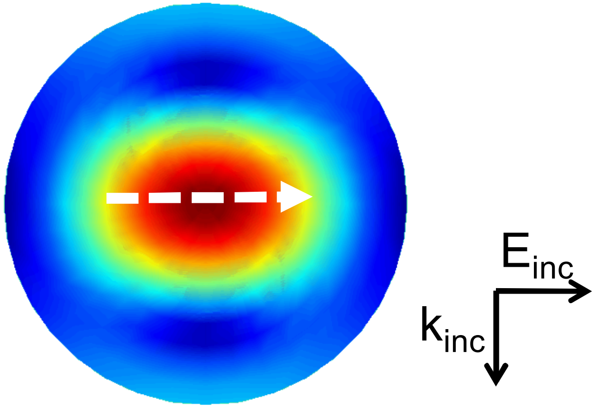
Alternative Means of Achieving Response

- All dielectric materials
  - No electron currents
  - Displacement current
    - Electronic polarization currents
- Spherical (Mie) resonators can provide the desired response
  - Isotropic response
  - Supports both magnetic and electric resonances (dependent upon composition and geometry)

Magnetic mode



Electric mode



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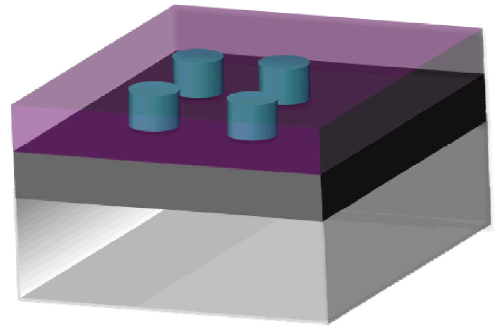
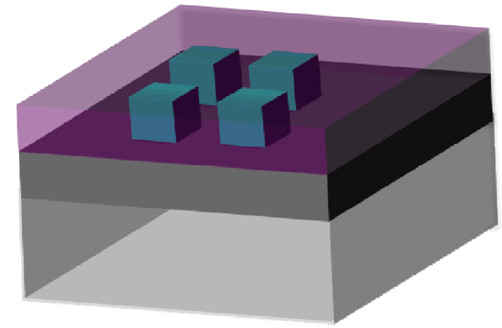
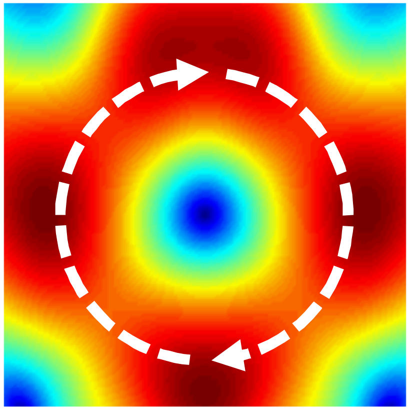
Cubic Dielectric Resonators

- Spherical components not compatible with standard lithography
- Cubic resonators maintain optical isotropy (*m3m* symmetry)
  - No ohmic loss
  - Can be easily integrated into multi-layer composites (through planarization)
  - Satisfies issues with metallic elements

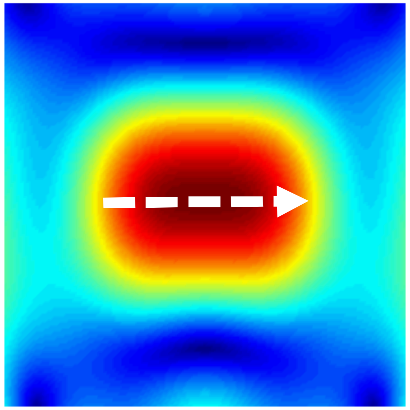


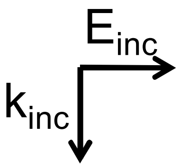


Magnetic mode



Electric mode





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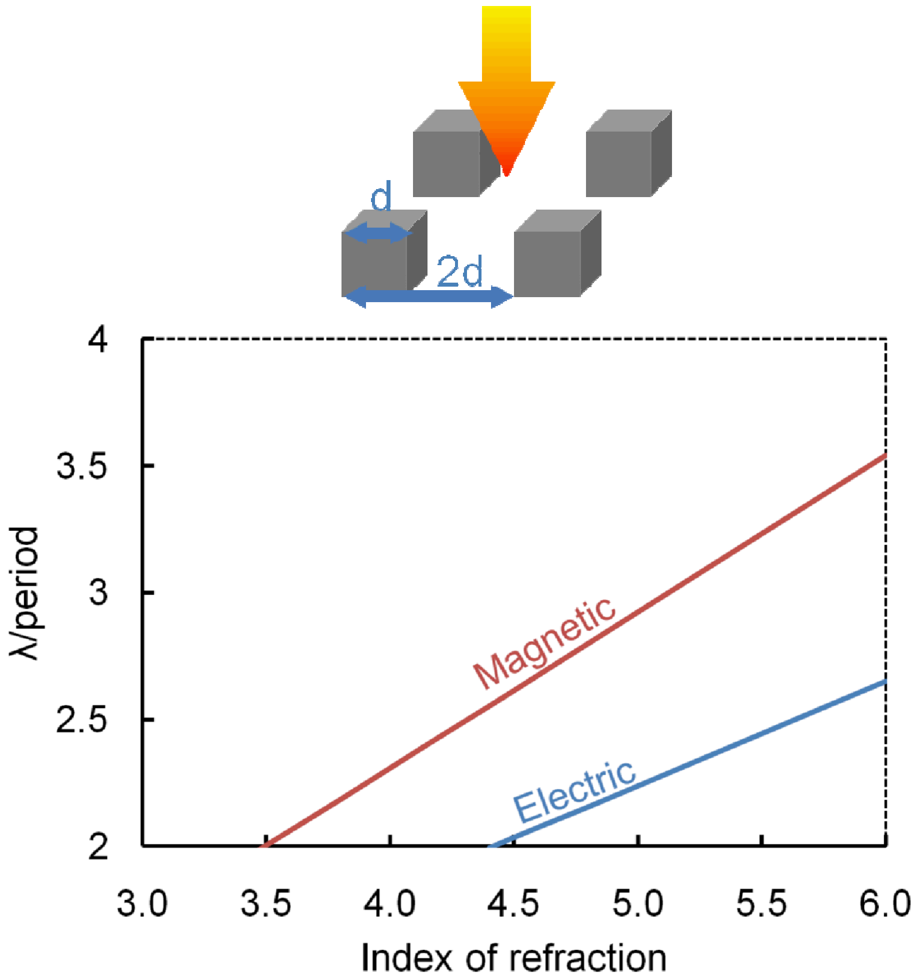
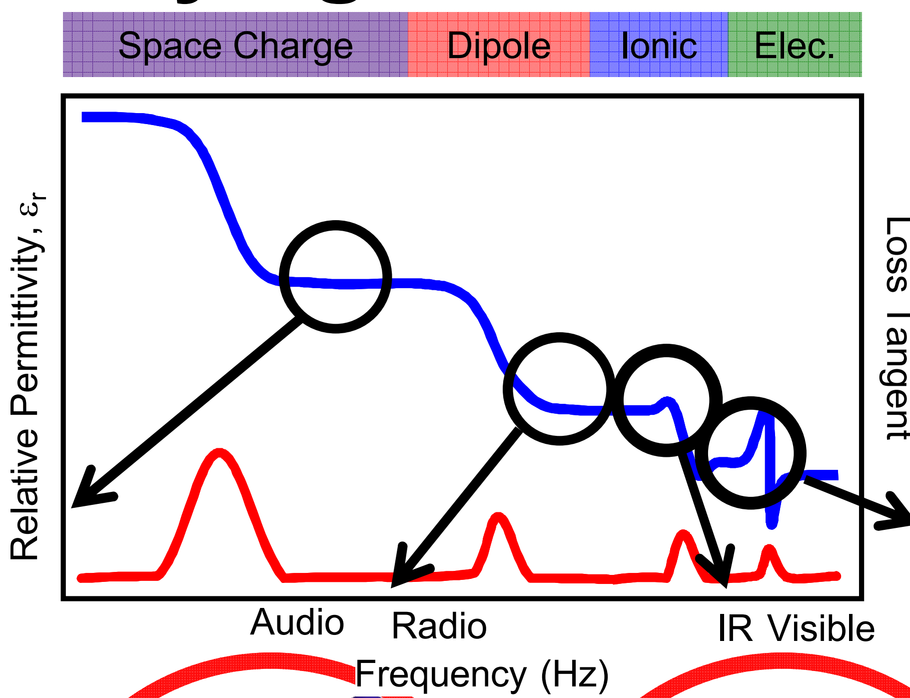
Resonance Behavior

- Mie theory not directly applicable to cubic resonators
  - Modeled via rigorous coupled wave algorithm package (GDCALC)
- Distinct resonances
  - Lowest frequency is magnetic (TM)
  - Second resonance is electric (TE)
  - Resonances scale with the dimensions and refractive index

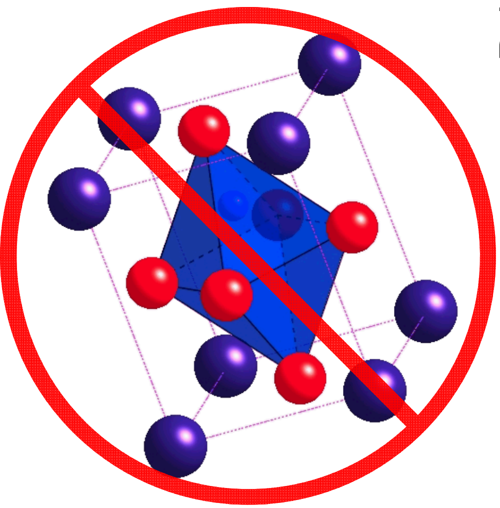


J.C. Ginn, *et al.*, *Nature Photonics, Under Review*

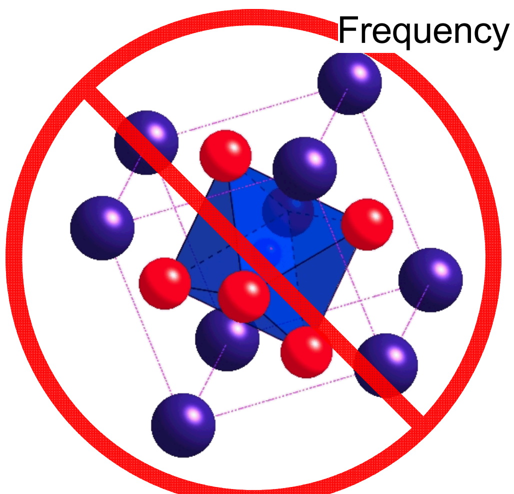
Identifying Ideal Materials



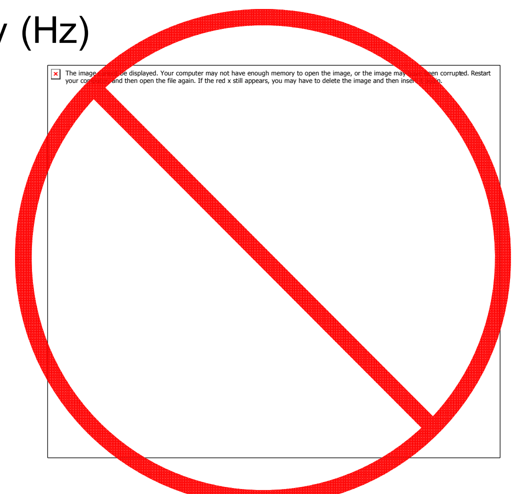
Semiconductors



Ferroelectrics



µW Dielectrics



Polaritonics

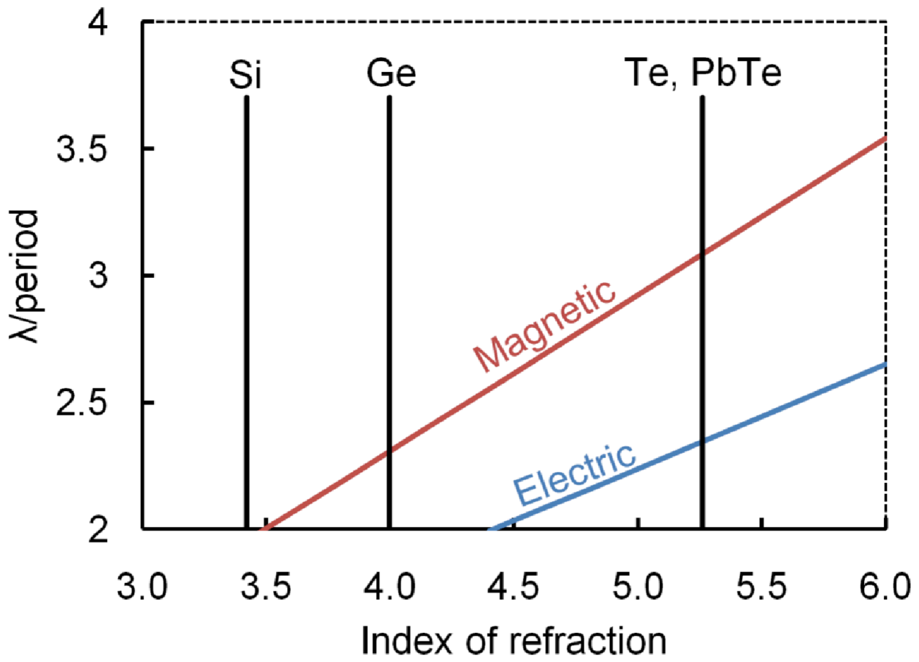
Identifying Materials

Desired Properties

- High refractive index
- Low loss
- Compatible with lithography

Potential Alternative Materials

- Narrow band-gap materials (Ge, Te, PbTe, etc.)
- High-index
- Low-loss below the bandgap



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Optimized IR Resonators

- Tellurium grown via electron-beam evaporation
- Room temperature growth
- Single Crystal BaF_2 substrate

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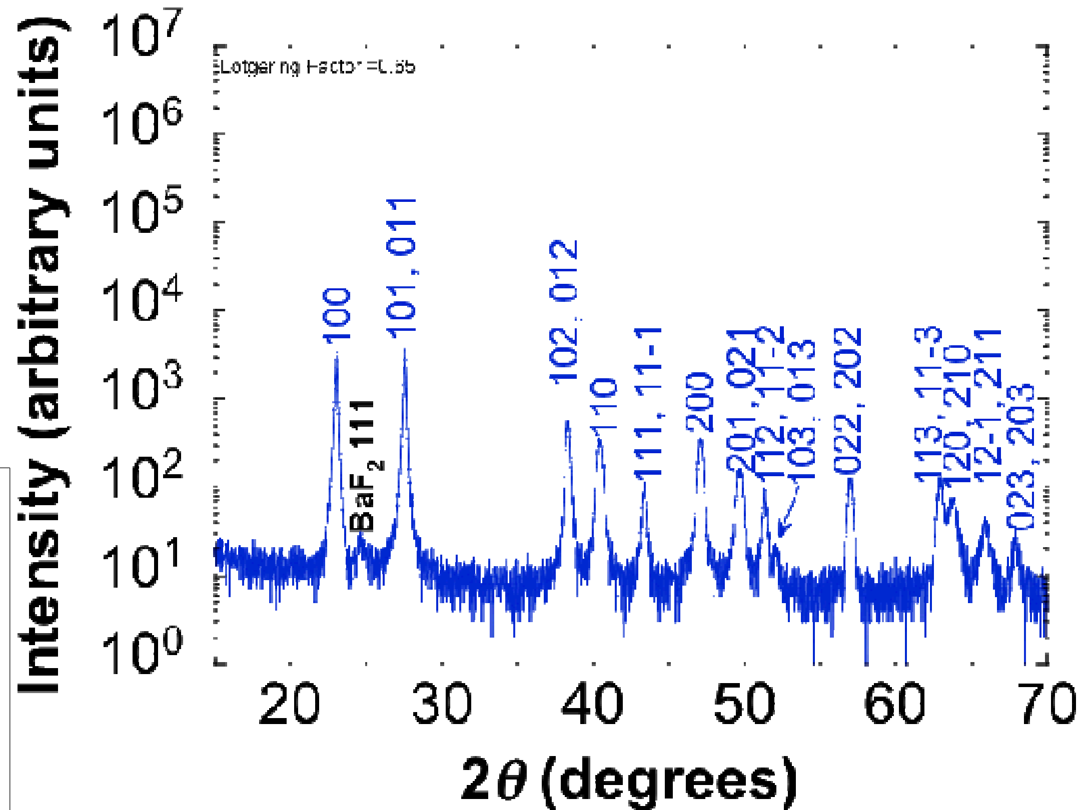

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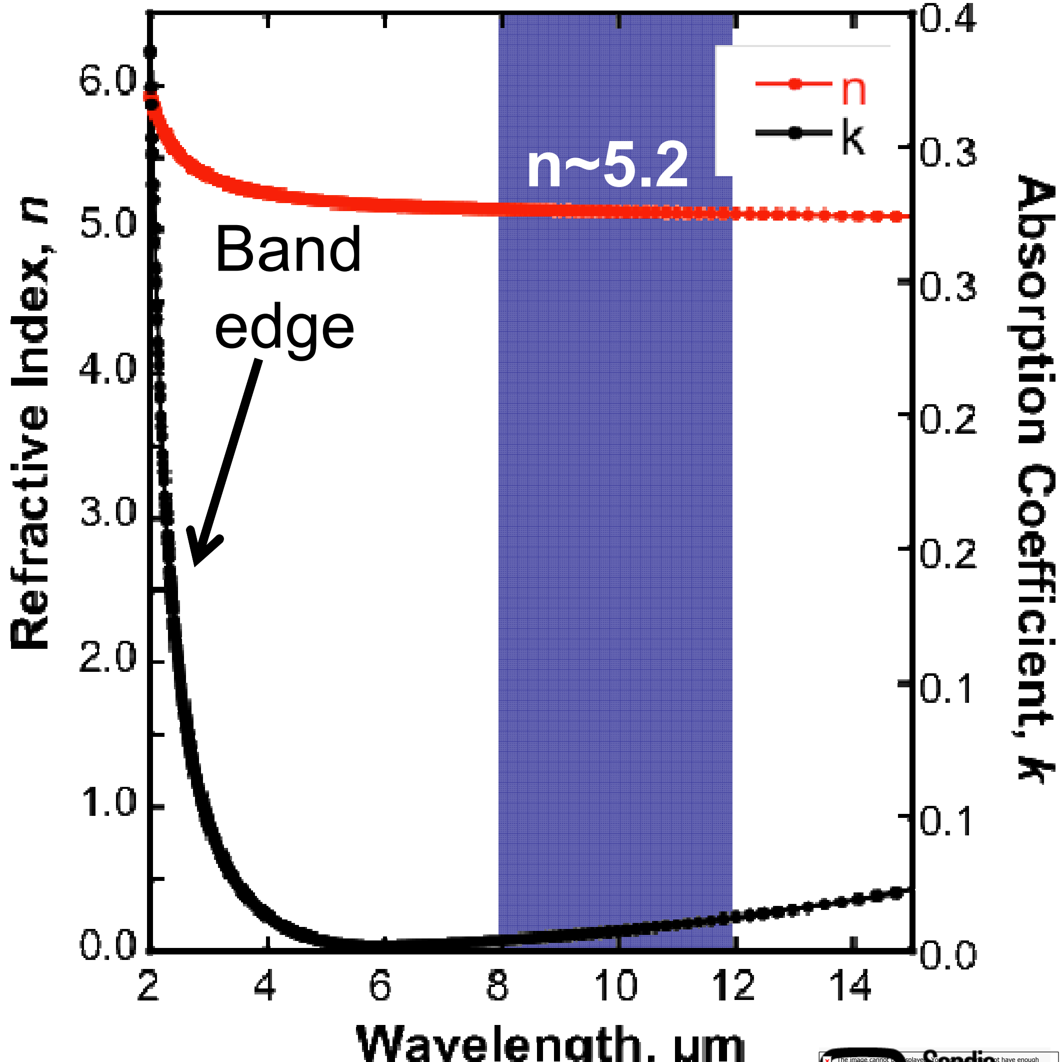
Optimized IR CDR

- Te has $P3_121$ symmetry
  - Polycrystalline required for isotropy
- Measured n = 5.02 + 0.04j @ 10 µm
  - Polycrystalline average = 5.3
- BaF_2 substrate
  - Transparent to 10 µm
- 1.7 µm Te cube will have magnetic resonance at 10 µm



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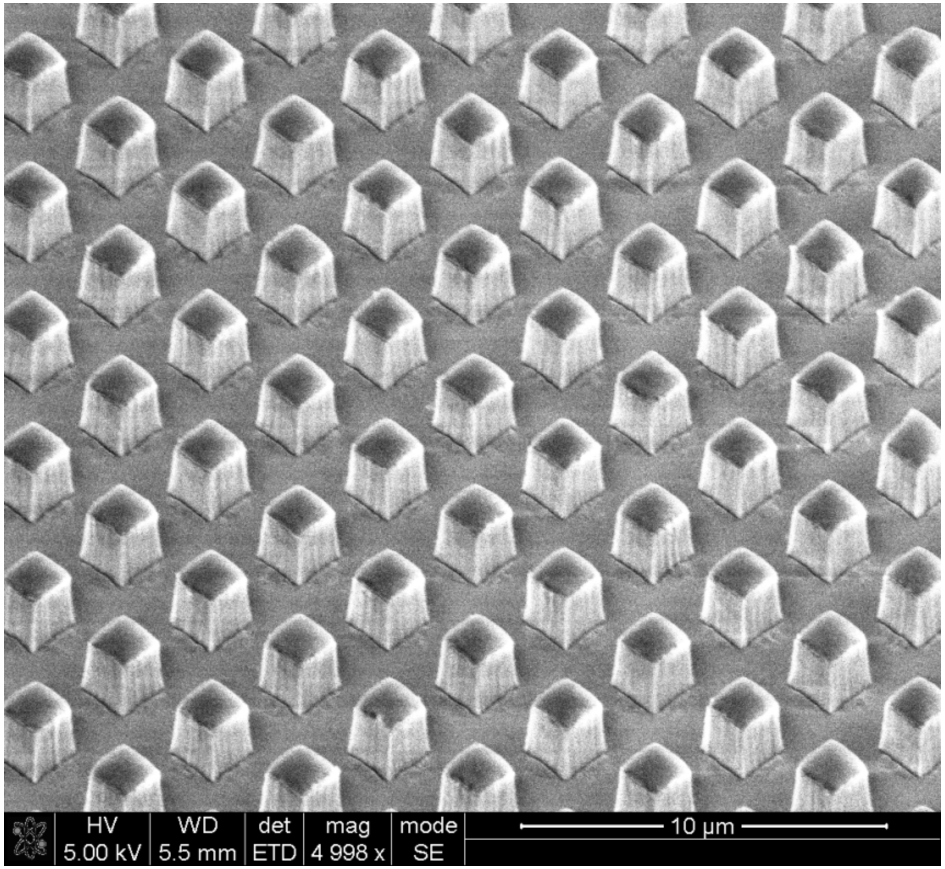
Fabrication of the CDR

- 1.7 μm Tellurium film deposited onto 1” BaF_2 wafer
- Patterned via e-beam lithography
  - Patterned area = 1 cm^2
- Cubes etched using a reactive ion etching process
  - Excellent uniformity over the array
  - Some over etching
    - Base of 1.53 μm
    - 10° sidewall slope
  - Minor polymer redeposition



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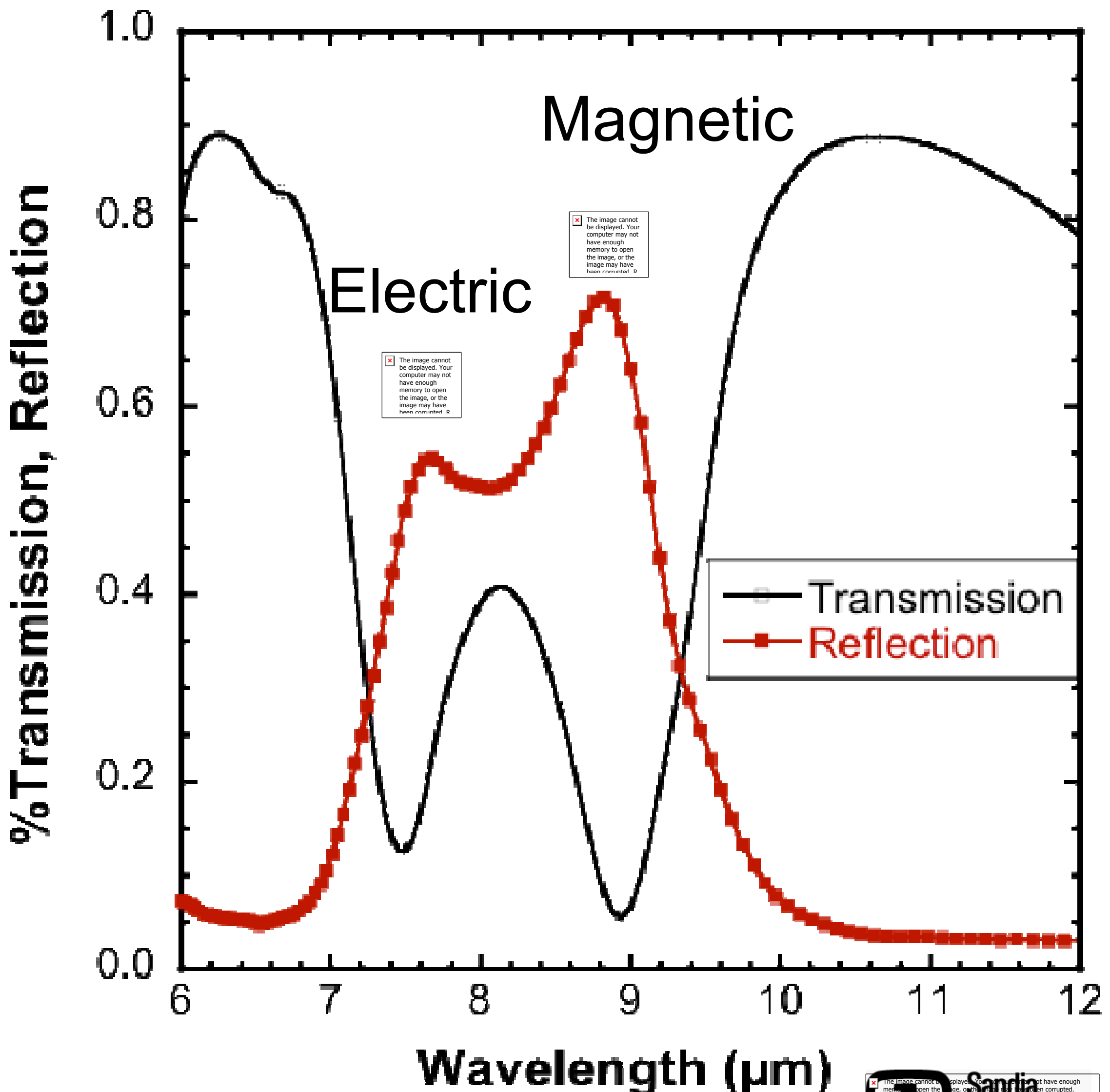
Optical Response

- Measured using a hemispherical directional reflectometer
- Both resonances are well defined and are present above the diffraction cut-off
- Loss (1-R-T) less than 8% between the resonances
- Data is not normalized
- ***First demonstration of an all dielectric optical metamaterial***



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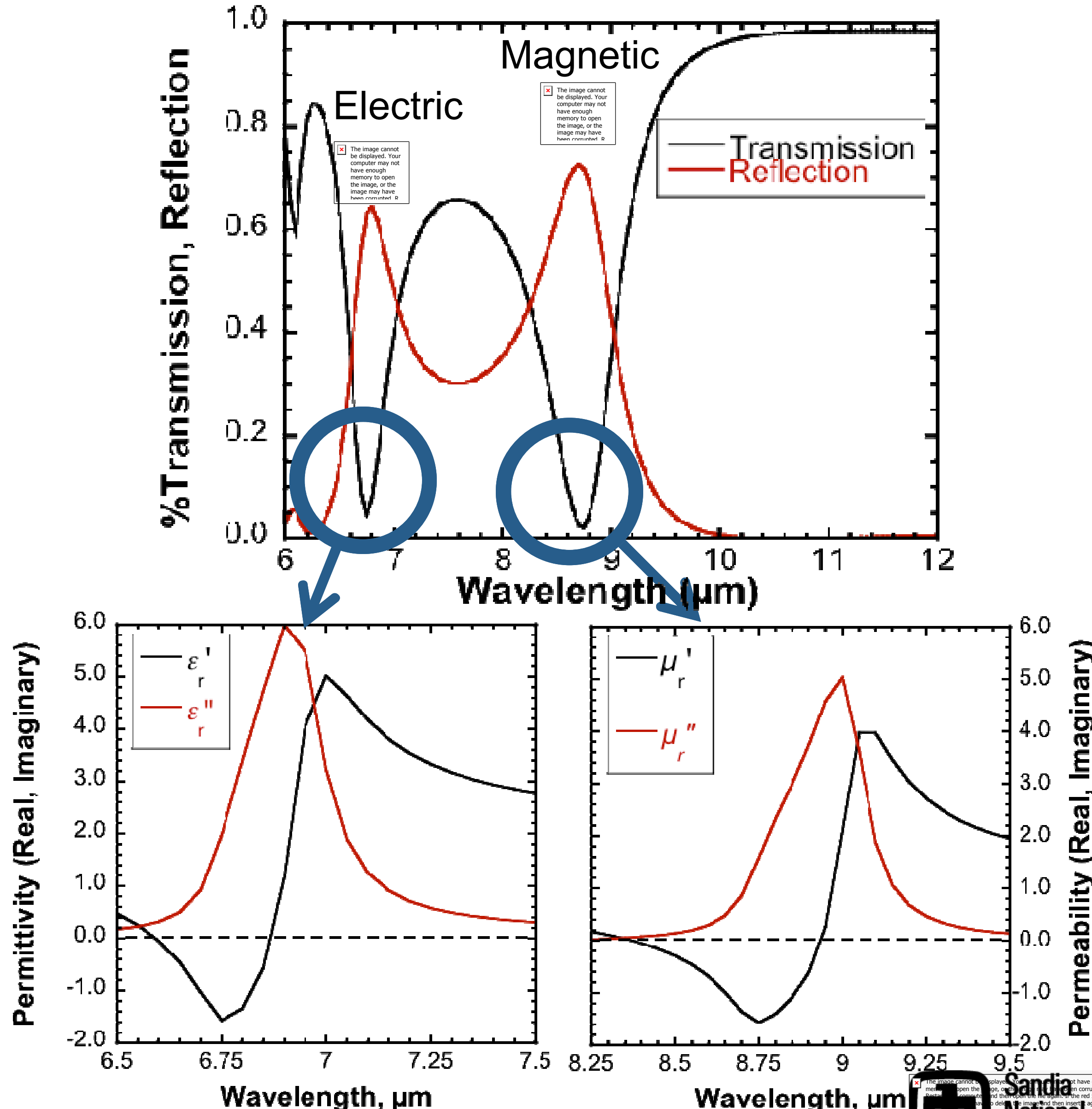
Extracted Optical Properties

- Properties computationally derived averaging real cube dimensions
  - Reasonable agreement
- Permittivity and permeability extracted
  - Both values less than -1
  - Permeability loss is less than 0.5 when permeability is -1
- ***World's First known IR magnetic mirror***



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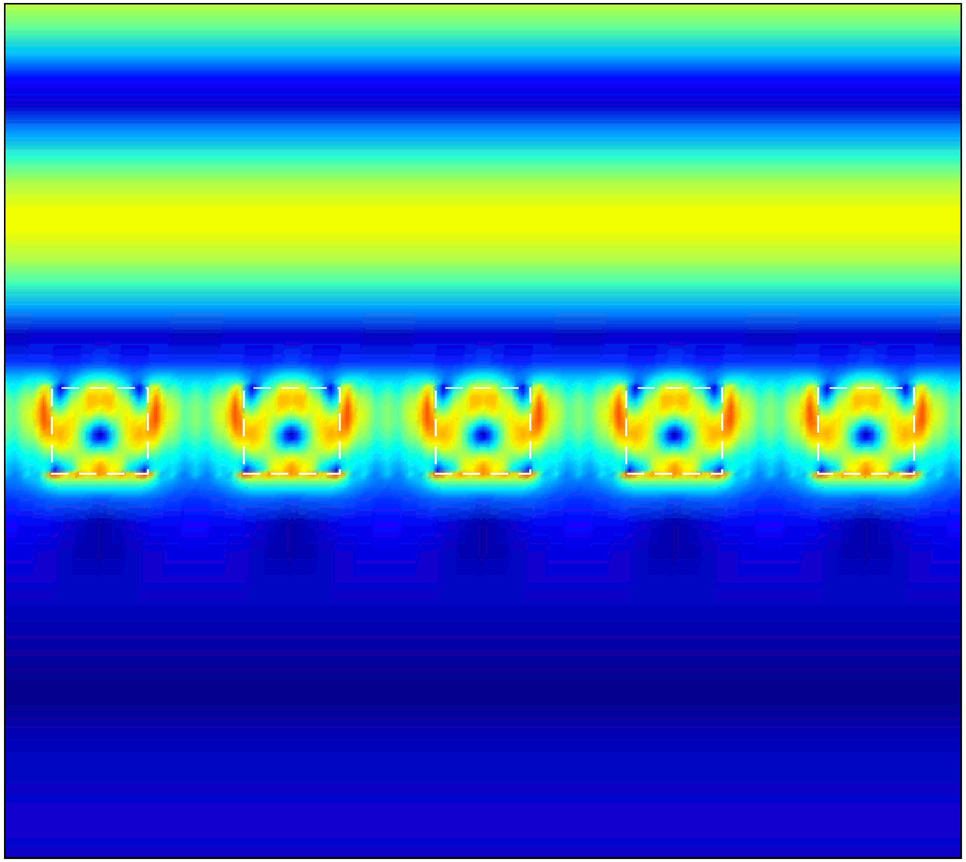
Dielectric Resonator Summary

- Cubic resonator designed with magnetic and electric resonances in the IR for the first time
  - Low loss
  - Isotropic response
- Present efforts focus on improving quality of Te films and etching process
- Integrate into negative index constructs
  - Two intermixed resonators



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Summary

- Multiple 3D IR metamaterial embodiments investigated
- An unconventional materials toolbox is being developed
  - Epsilon near zero materials
  - Dielectric resonators using narrow bandgap semiconductors
- Enabling low loss metamaterial preparation