Winter 1-1-2008

Lecture 11, Part 3: Glass dielectrics for microelectronics and energy applications - Glass & glass ceramics as high energy materials

Mike Lanagan
Penn State University

Dielectric Properties and Metamaterials

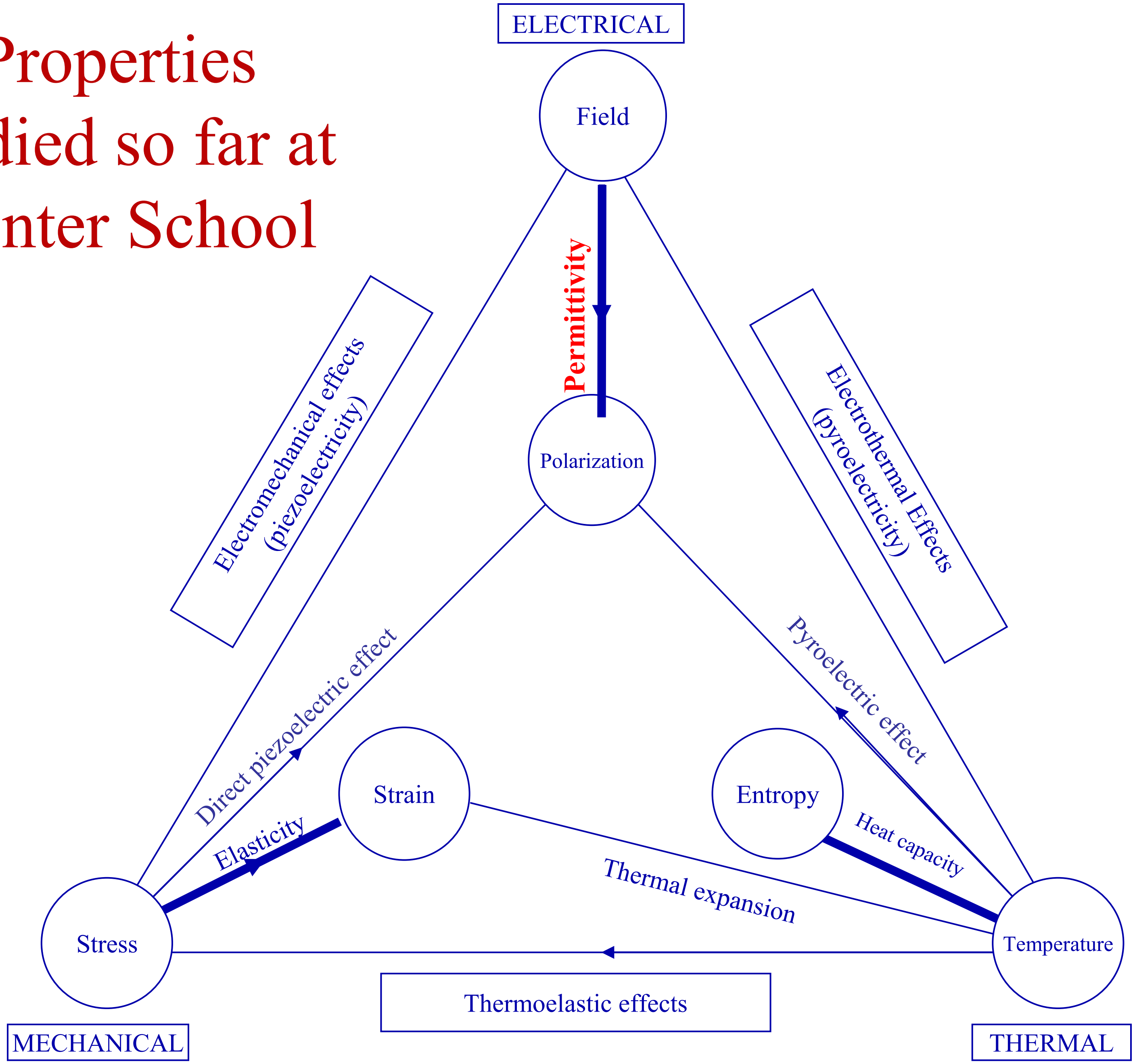
Mike Lanagan
Materials Research Institute
Penn State University

US-Japan Winter School on New Functionality in Glass
January 15, 2008
Kyoto Japan

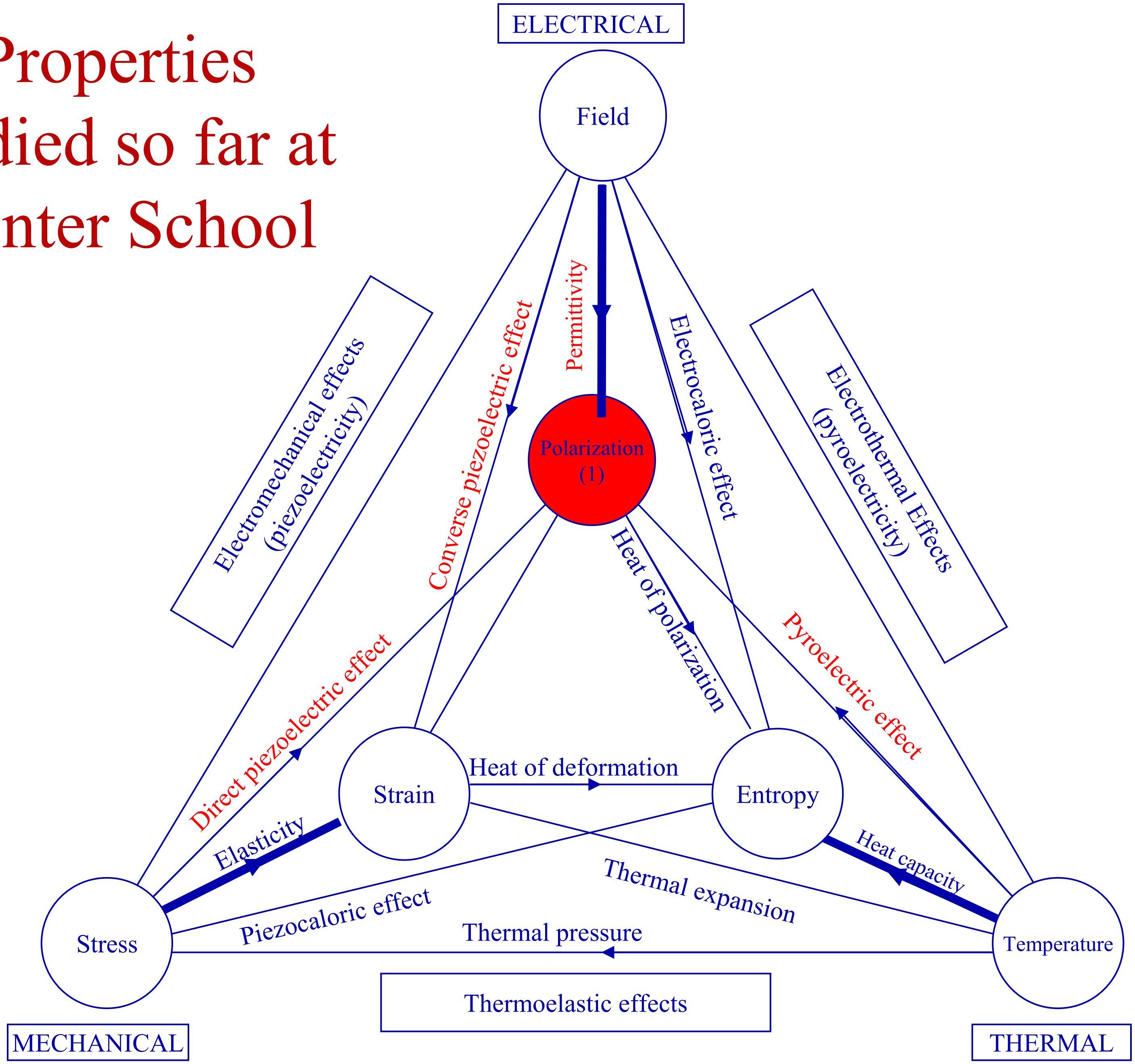
Dielectric Properties and Metamaterials

- Dielectric Properties (i.e Permittivity)
  - Fundamental frequency dependence
- Metamaterials
  - Negative permittivity and refractive index
  - Based on resonance response
  - Discussion on the potential of glass as a meta-material

Properties studied so far at Winter School



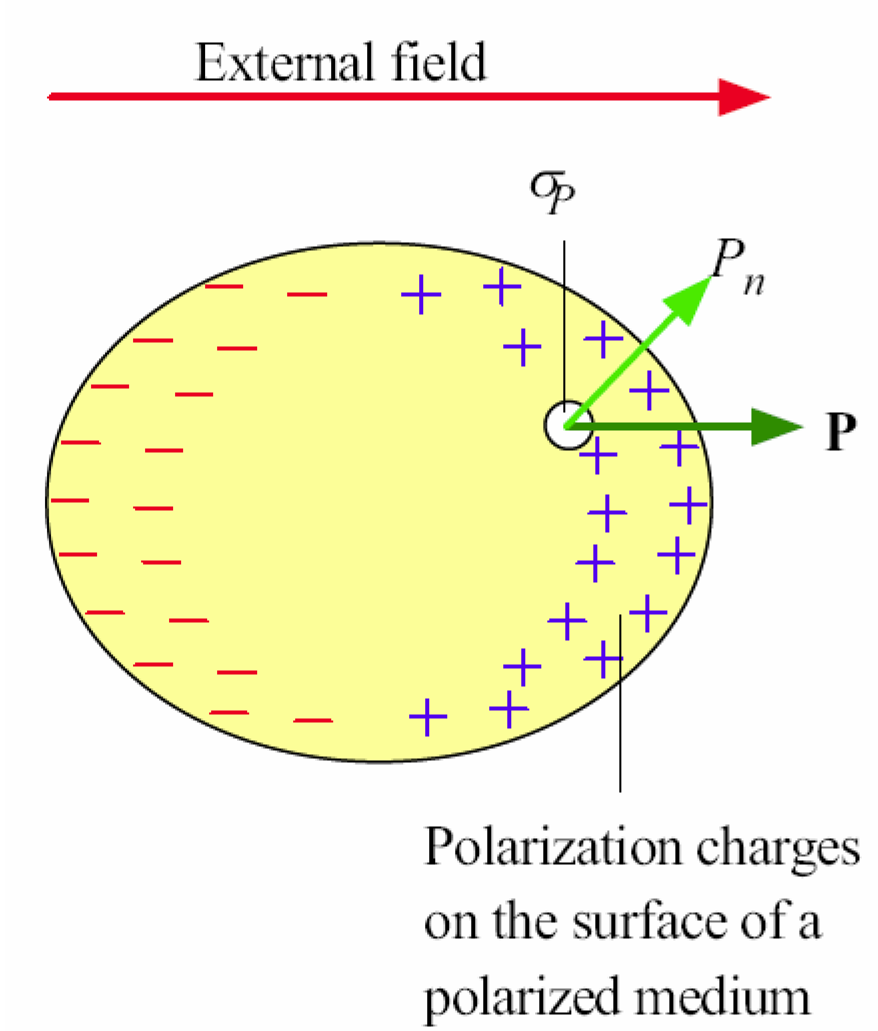
Properties studied so far at Winter School



Dielectric Polarization

- Contributes to Permittivity
- 4 basic mechanisms
  - Electronic
  - Ionic
  - Rotational or Dipolar
  - Space charge

External Electric Field Polarizes a Material



Polarization charge density on the surface of a polarized medium is related to the normal component of the polarization vector.

Fig 7.6

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Relative Permittivity and Polarizability

$$\varepsilon_r = 1 + \frac{N\alpha_e}{\varepsilon_o}$$

ε_r = relative permittivity

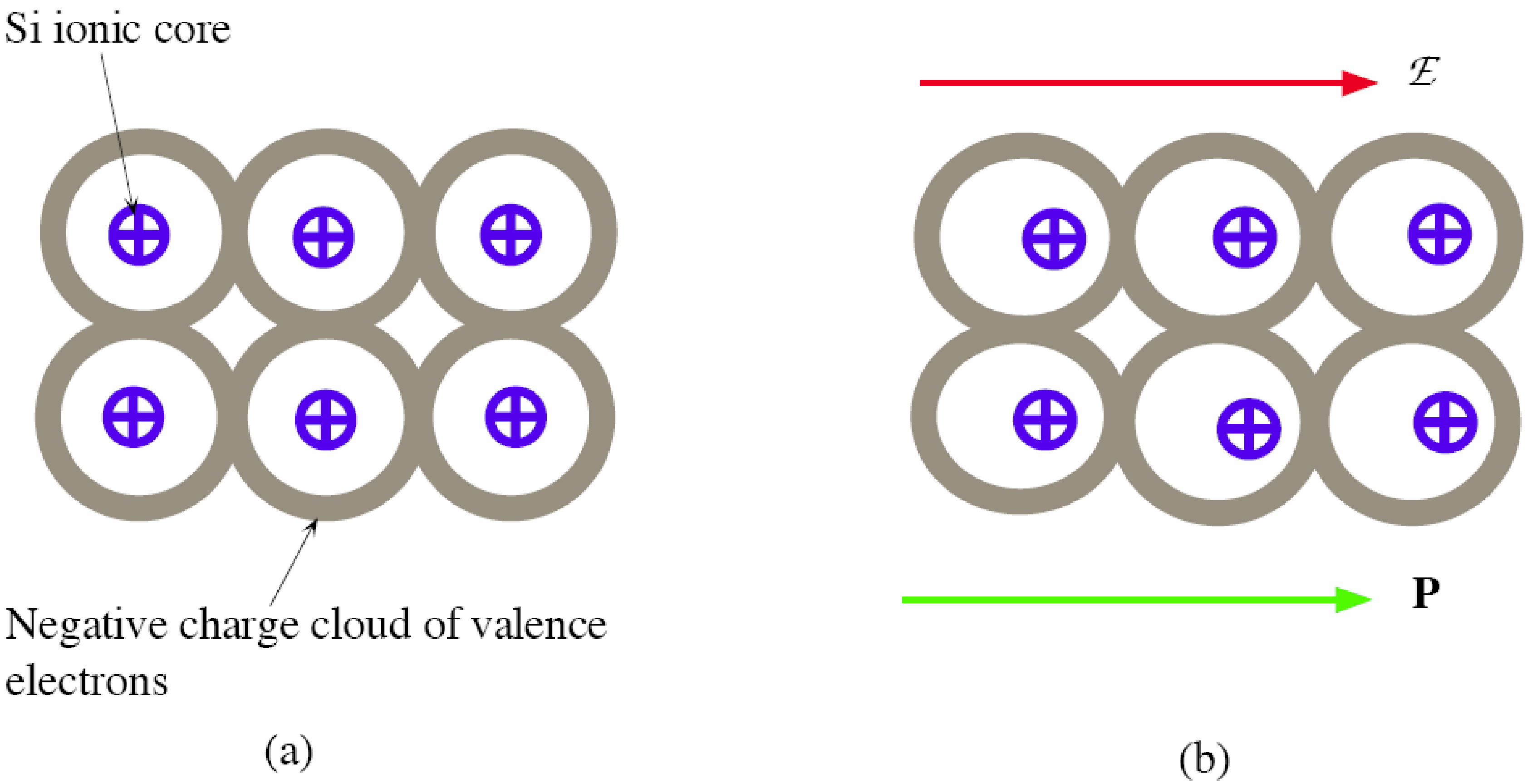
N = number of molecules per unit volume

α_e = electronic polarizability

ε_o = permittivity of free space

Assumption: Only electronic polarization is present

Electronic Polarization



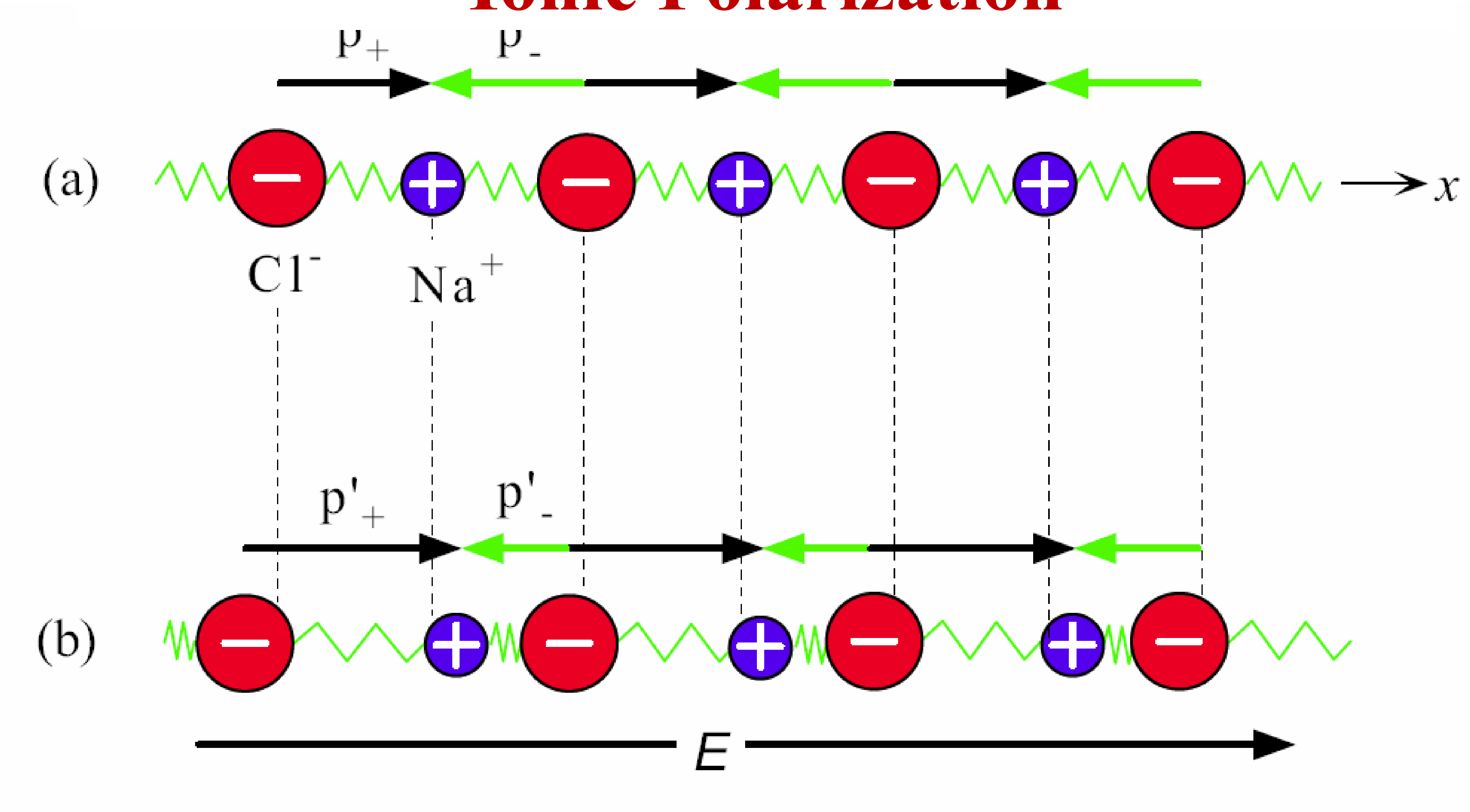
(a) Valence electrons in covalent bonds in the absence of an applied field.

(b) When an electric field is applied to a covalent solid, the valence electrons in the covalent bonds are shifted very easily with respect to the positive ionic cores. The whole solid becomes polarized due to the collective shift in the negative charge distribution of the valence electrons.

Fig 7.8

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Ionic Polarization



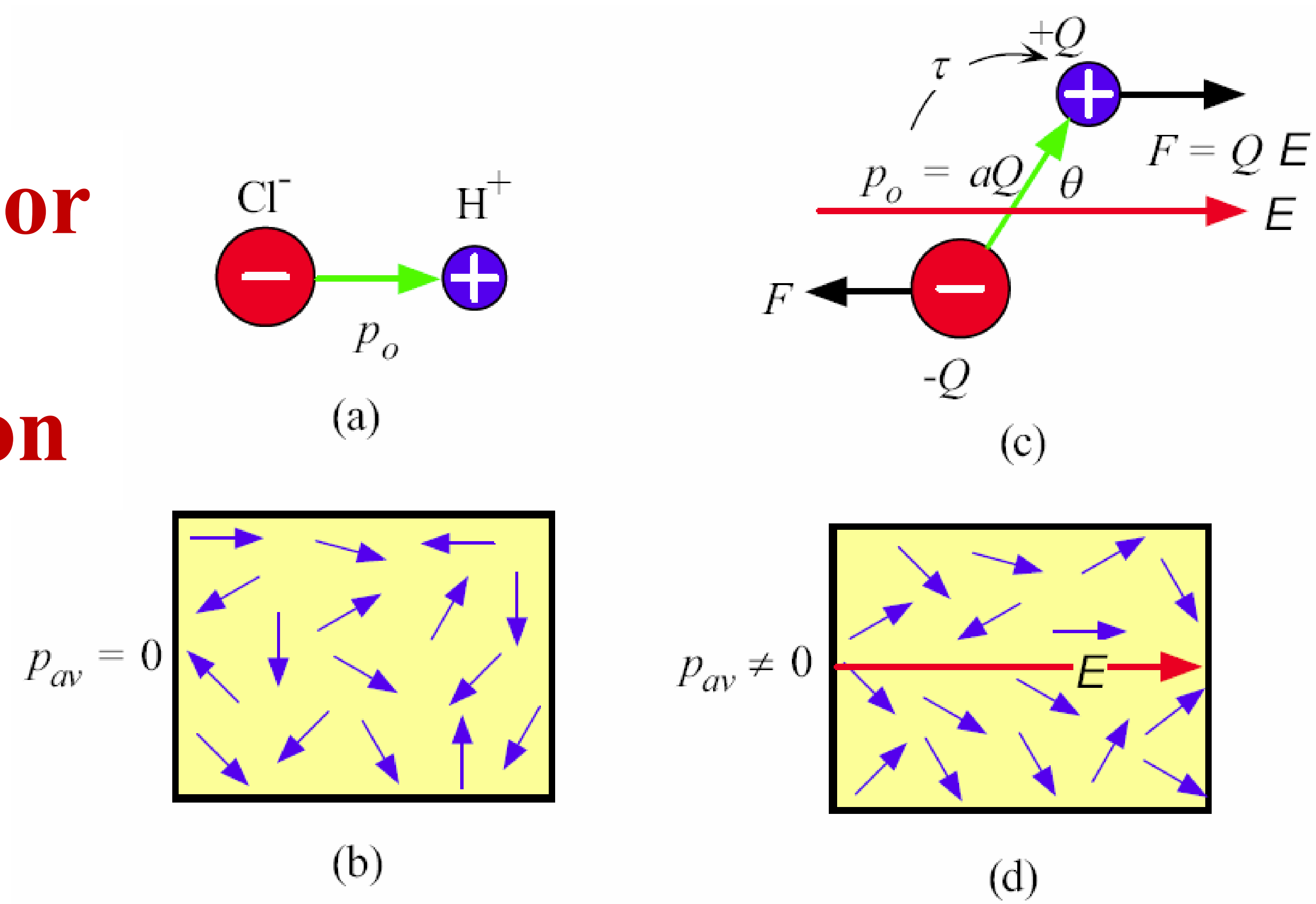
moment per ion is zero.

(b) In the presence of an applied field the ions become slightly displaced which leads to a net average dipole moment per ion.

Fig 7.9

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Rotational or Dipolar Polarization



(a) A HCl molecule possesses a permanent dipole moment p_0.
(b) In the absence of a field, thermal agitation of the molecules results in zero net average dipole moment per molecule.
(c) A dipole such as HCl placed in a field experiences a torque that tries to rotate it to align p_0 with the field E.
(d) In the presence of an applied field, the dipoles try to rotate to align with the field against thermal agitation. There is now a net average dipole moment per molecule along the field.

Fig 7.10 From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Complex Relative Permittivity (related to time response)

$$\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$$

ε_r = dielectric constant

ε'_r = real part of the complex dielectric constant

ε''_r = imaginary part of the complex dielectric constant

j = imaginary constant $\sqrt{(-1)}$

Dielectric Loss Factor

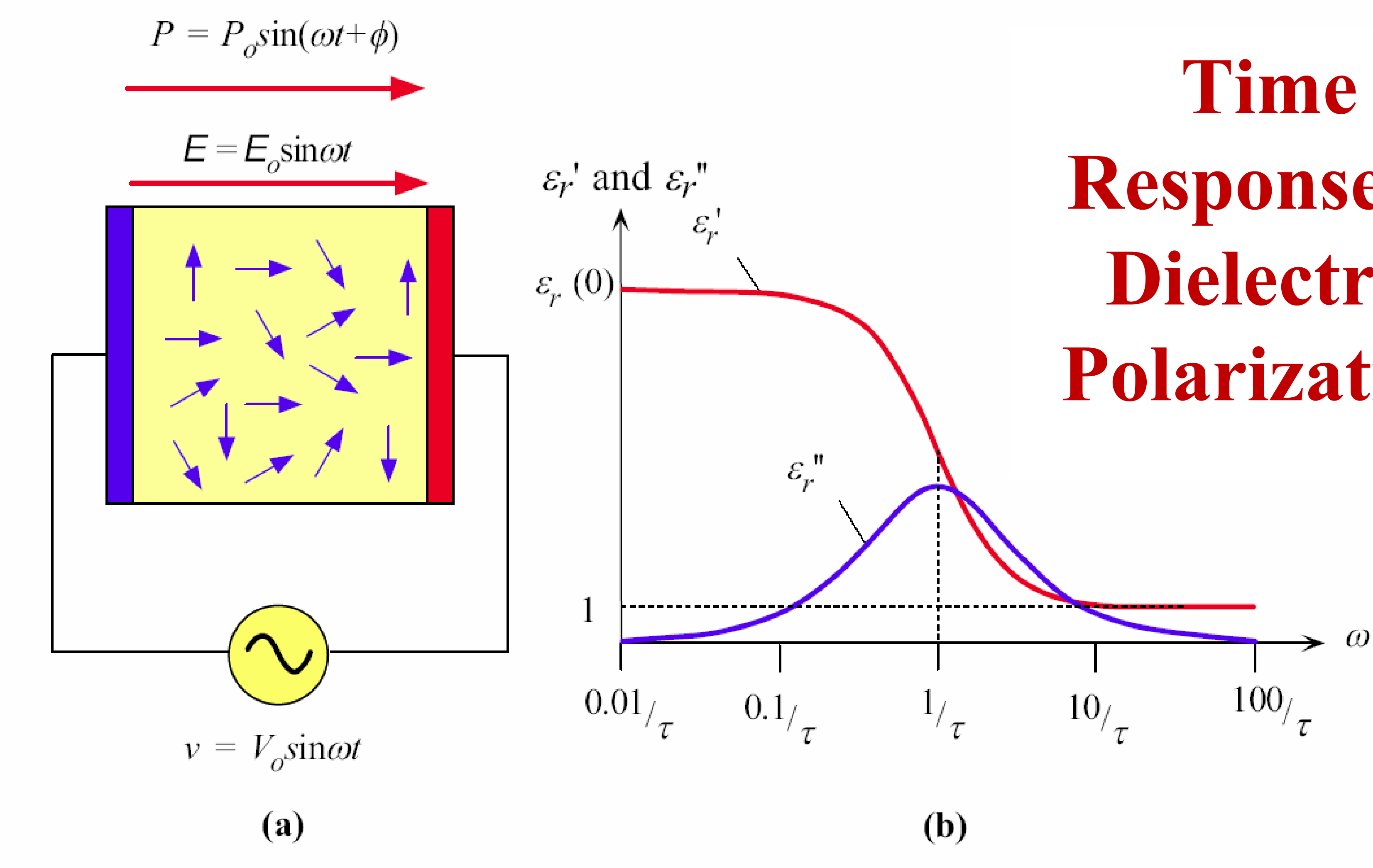
Loss Tangent (related to energy loss)

$$\tan\delta = \frac{\varepsilon_r''}{\varepsilon_r'}$$

$\tan\delta$ = loss tangent or loss factor, ε'_r = real part of the complex dielectric constant, ε''_r = imaginary part of the complex dielectric constant

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Time Response of Dielectric Polarization

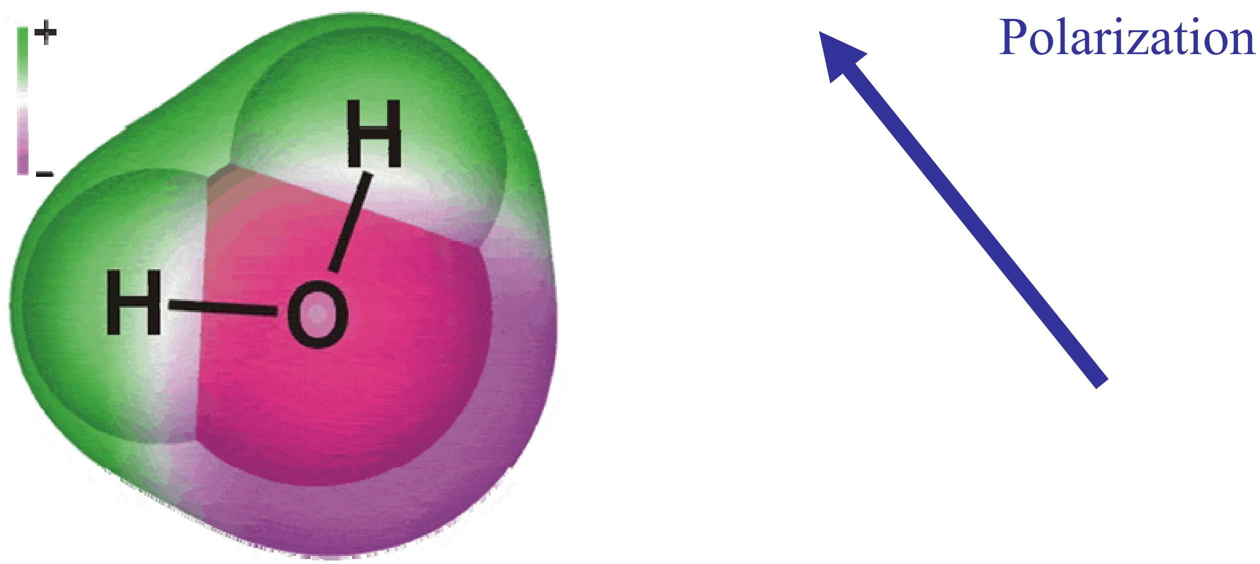


(a) An ac field is applied to a dipolar medium. The polarization P ($P = Np$) is out of phase with the ac field.
(b) The relative permittivity is a complex number with real (ε_r') and imaginary (ε_r'') parts that exhibit frequency dependence.

Fig 7.13

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

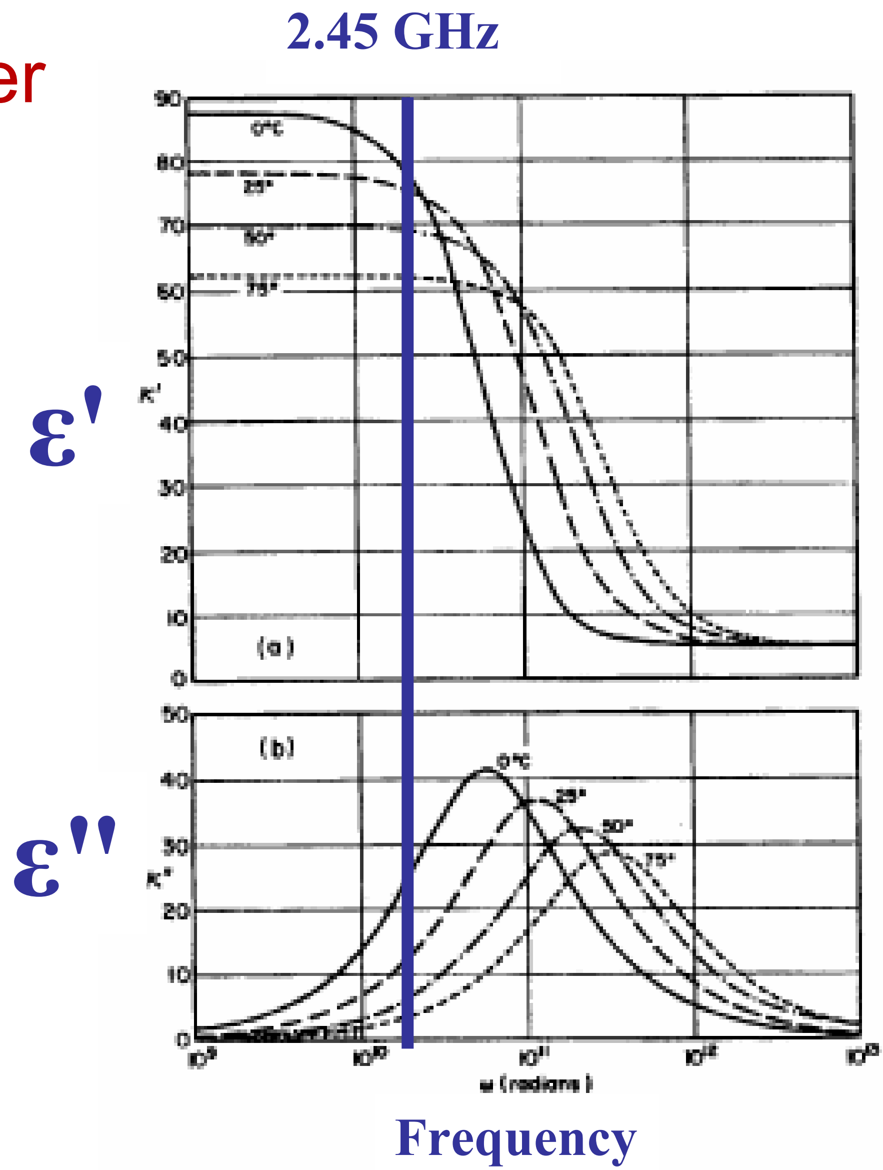
Example: Water Molecule



www.lsbu.ac.uk/water/molecule.html

Microwave Dielectric Relaxation of Liquid Water

- Dielectric relaxation indicated by:
  - Decrease in real permittivity
  - Peak in imaginary permittivity
- Maximum ε” corresponds to maximum conversion from EM energy to thermal energy.
- High ε”
  - good for microwave oven
  - Bad for device



Relaxation spectrum of water as function of frequency and temperature: (a) dispersion; (b) absorption.

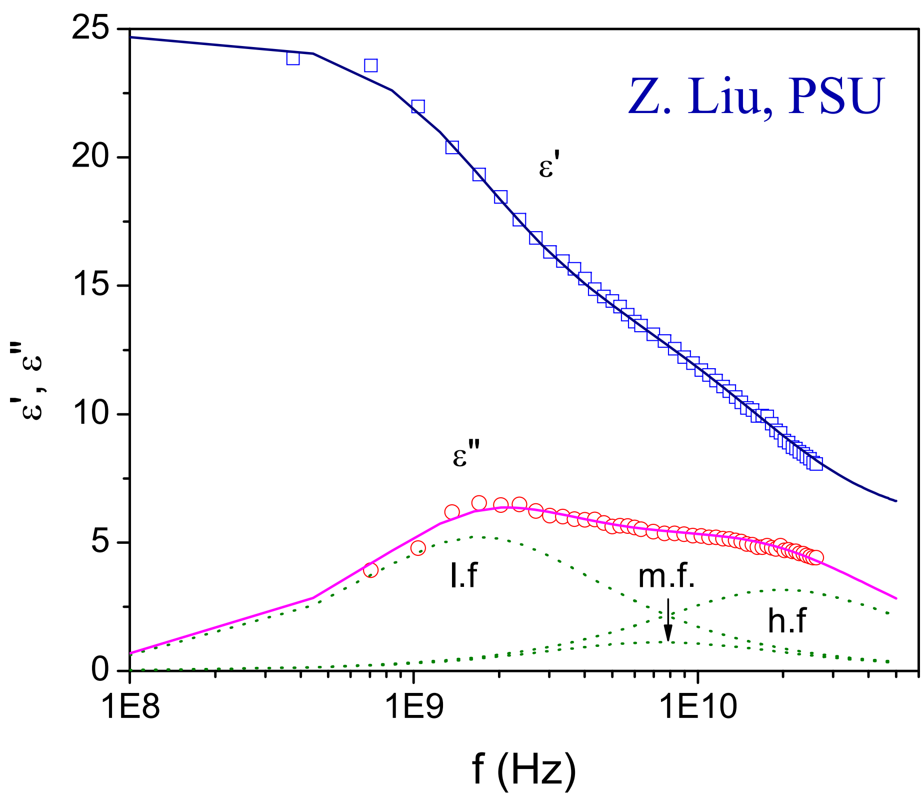
A. Von Hipple, IEEE Trans. Insul, 1988

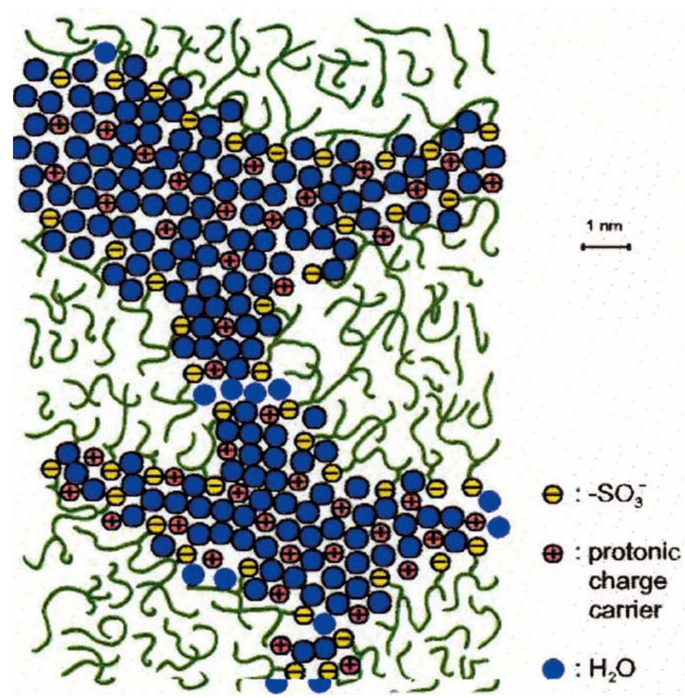
Why not 20 GHz operation for a microwave oven?

- Open bands
  - 915 MHz (not all countries)
  - 2.45 GHz
  - 5.8 GHz
  - 24.1 GHz
- Cost constraints

Water in a proton exchange membrane

- ***Current PEM fuel cells are based on PSA membranes, e.g. Nafion. The essential feature of Nafion is the nano-separation of hydrophilic/hydrophobic domains***

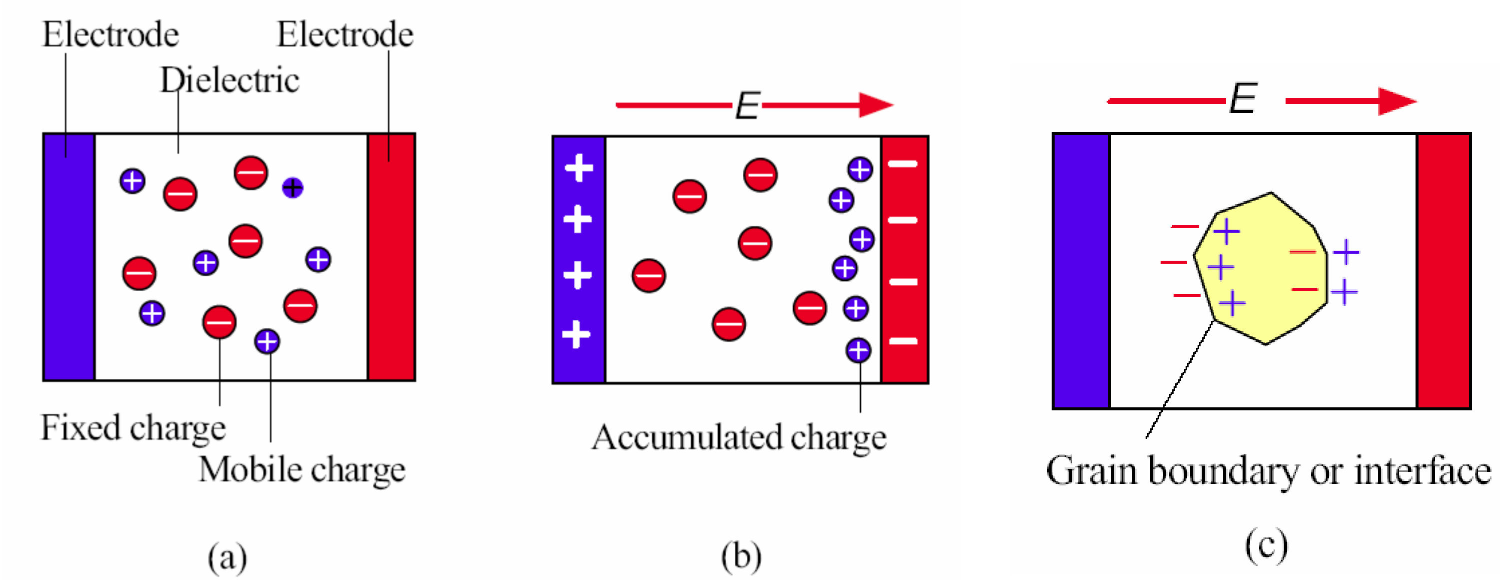




K.D. Kreuer, *J. Membr. Sci.*, **185**, 29 (2001)

Why not characterize water in porous glass in this way?

Space Charge Polarizability



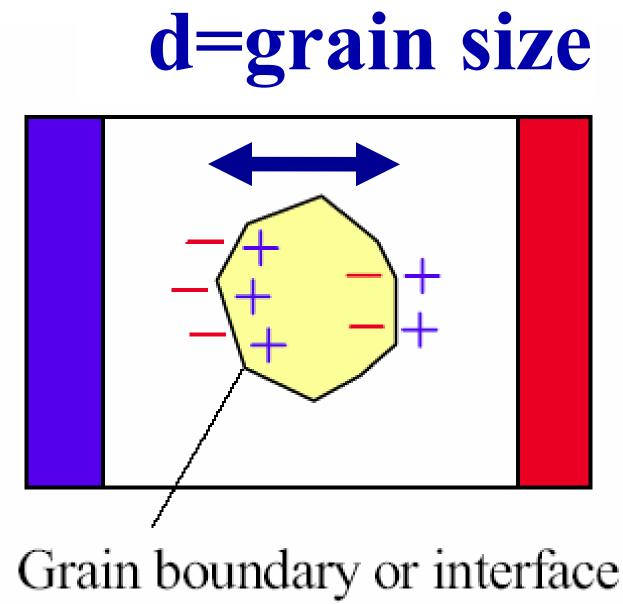
(a) A crystal with equal number of mobile positive ions and fixed negative ions. In the absence of a field, there is no net separation between all the positive charges and all the negative charges.
(b) In the presence of an applied field, the mobile positive ions migrate toward the negative charges and positive charges in the dielectric. The dielectric therefore exhibits interfacial polarization.
(c) Grain boundaries and interfaces between different materials frequently give rise to Interfacial polarization.

Fig 7.11

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Space Charge in Ceramic Capacitors with Glass

- Need long enough time for charge to move to boundary



Relaxation Time $\tau \propto \dfrac{d}{\sigma}$ Grain Size / Grain Conductivity

Role of Glasses in MLCC

SAMSUNG ELECTRO-MECHANICS

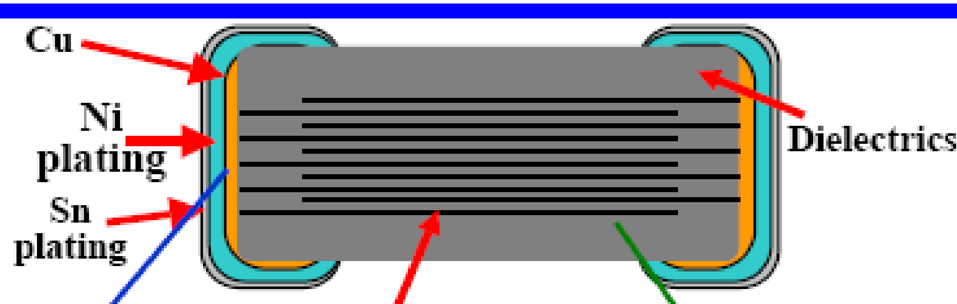
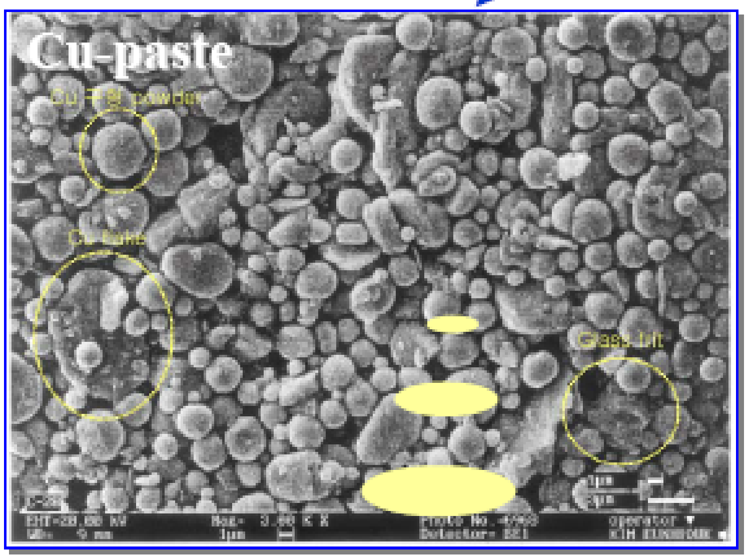
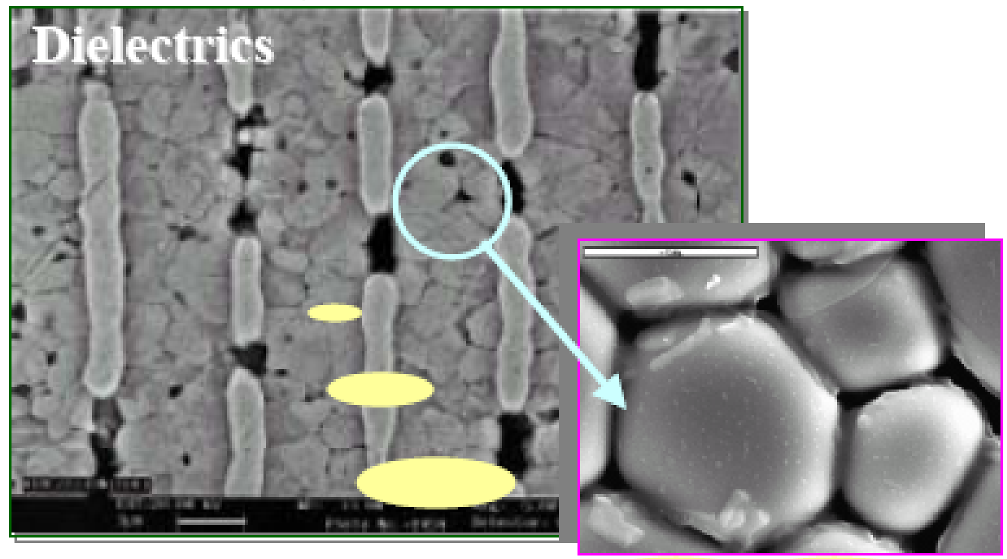


Fig. Cut-away view of MLCC





Glass is of major importance as an bond between ceramic and metal, and a filler.

Glass is of major importance as an additive to ceramics in order to promote sintering at low temperature.

Presented by Dr. Song Moon Song, Center for Dielectric Studies Fall Meeting

BaTiO$_3$ Ceramics with Glass boundaries

Relaxation time = 0.0001 s

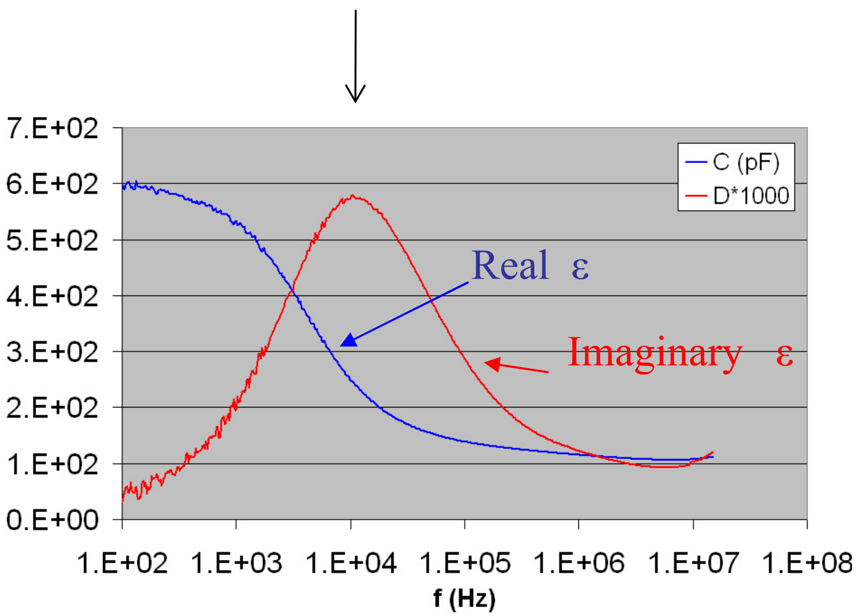
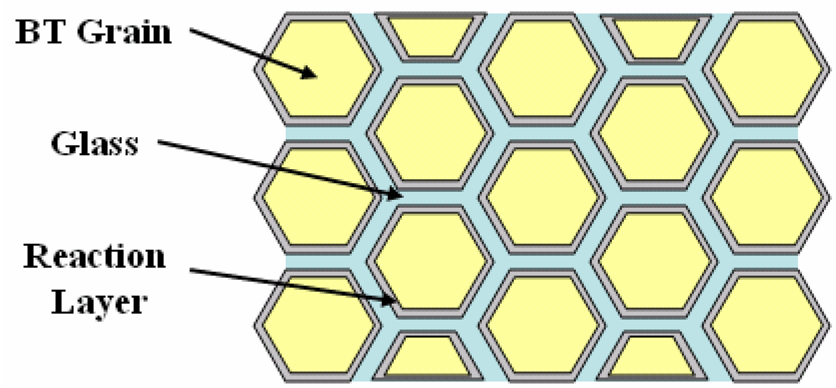


Figure shows dielectric response of a BaTiO3 – Glass composite

Microstructure Schematic



- Conductive grain and insulating grain boundary
- Maxwell-Wagner relaxation

From the relaxation time and microstructure, we can determine the grain conductivity

Janosik Thesis

Frequency Response of Dielectric Polarization

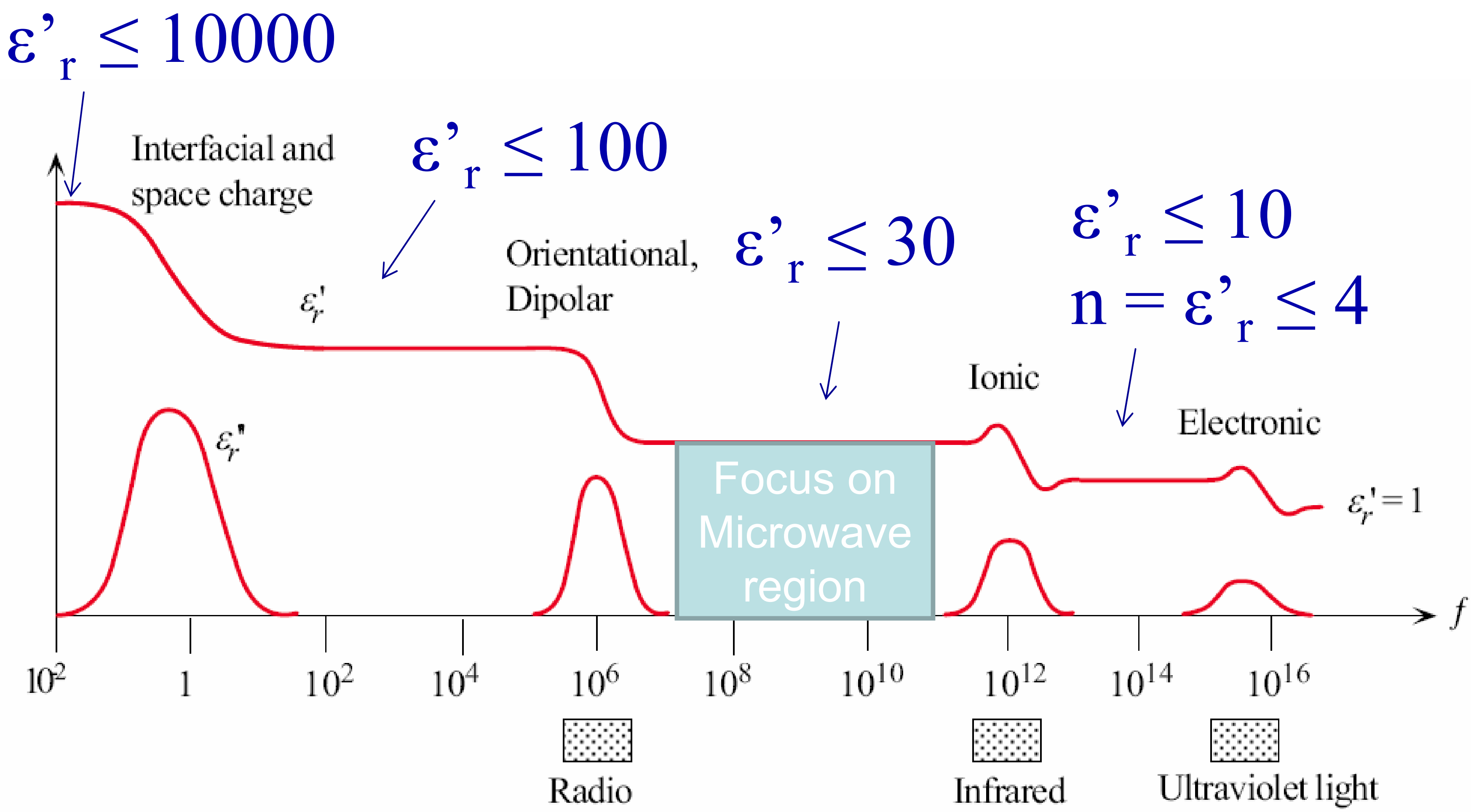


Fig 7.15

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Table 7.2 Typical examples of polarization mechanisms

Example	Polarization	Static ε_r	Comment
Ar gas	Electronic	1.0005	Small N in gases: $\varepsilon_r \approx 1$
Ar liquid (T < 87.3 K)	Electronic	1.53	van der Waals bonding
Si crystal	Electronic polarization due to valence electrons	11.9	Covalent solid; bond polarization
NaCl crystal	Ionic	5.90	Ionic crystalline solid
CsCl crystal	Ionic	7.20	Ionic crystalline solid
Water	Orientational	80	Dipolar liquid
Nitromethane (27 °C)	Orientational	34	Dipolar liquid
PVC (polyvinyl chloride)	Orientational	7	Dipole orientations partly hindered in the solid

BaTiO3 permittivity (dielectric constant) = 1,000

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Permittivity of Amorphous Materials

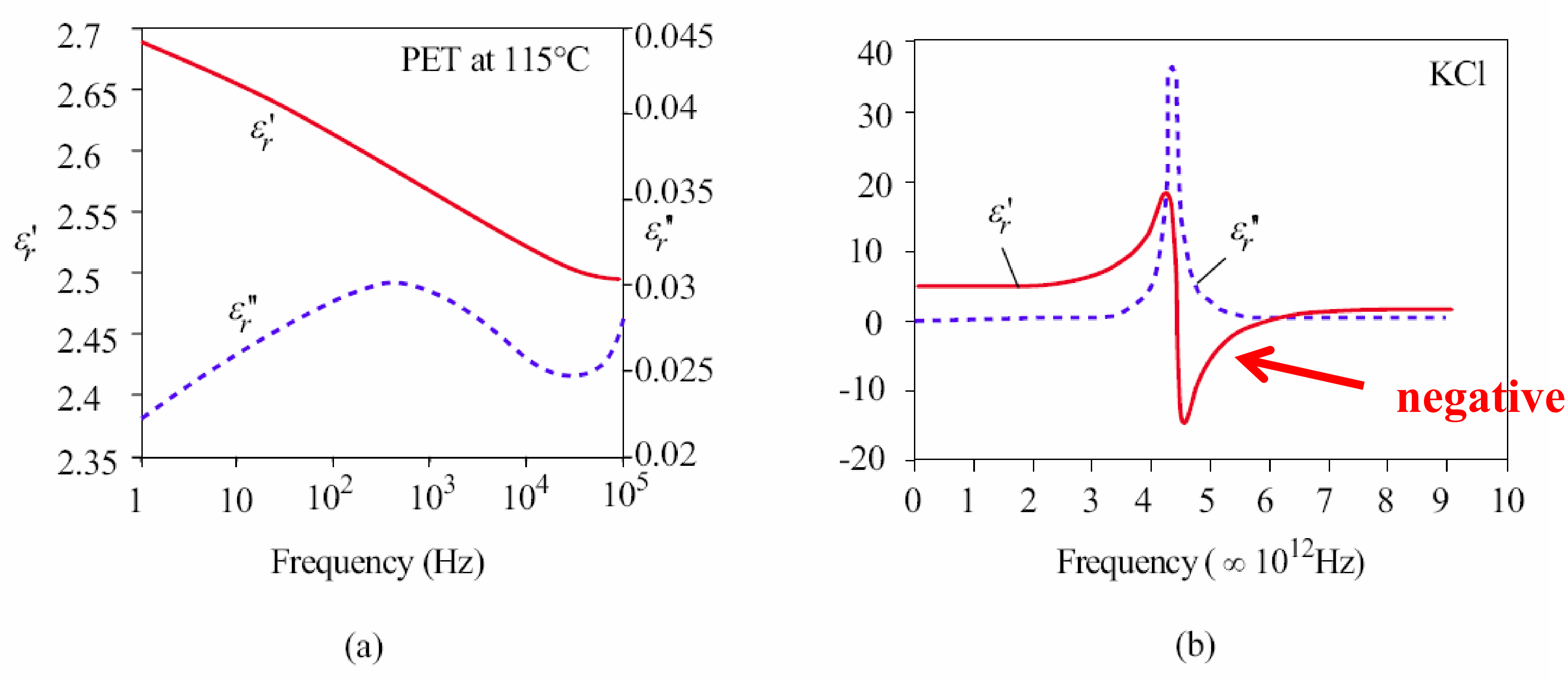
Permittivity values are related to the electron density an ionic charge

- $Si0_2$ ε_r=4
- Commercial flat panel Ba-Si-O ε_r= 8
- 40%Ba-20%Ti-40%Si-0 ε_r= 15
- Ta_2O_5 ε_r= 25
- Nb_2O_5 ε_r= 40
- Mainly electronic and ionic contributions

Frequency (or time) Response

- Relaxation Response
  - Based on diffusion mechanisms
  - Significant damping in oscillations
  - Describes Dipolar and Space charge mechanisms
- Resonance Response
  - High frequency response
  - Not as much damping as relaxation response

Relaxation vs Resonant Response



(a) Real and imaginary part is of the dielectric constant, ε_r' and ε_r'' versus frequency for (a) a polymer, PET, at 115 °C and (b) an ionic crystal, KCl, at room temperature.

both exhibit relaxation peaks but for different reasons.

SOURCE:

(b) from C. Smart, G.R. Wilkinson, A. M. Karo, and J.R. Hardy, International Conference on lattice Dynamics, Copenhagen, 1963, as quoted by D. G. Martin, "The Study of the Vibration of Crystal Lattices by Far Infra-Red Spectroscopy," *Advances in Physics*, 14, no. 53-56, 1965, pp. 39-100.

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Summary of Dielectric Response

- 4 basic mechanisms with each mechanism having a characteristic frequency response
- Glasses potentially have electronic, ionic and space charge contributions
- Highest permittivity for a glass is less than 20
- Discussion point – is rotational polarization possible in glass?

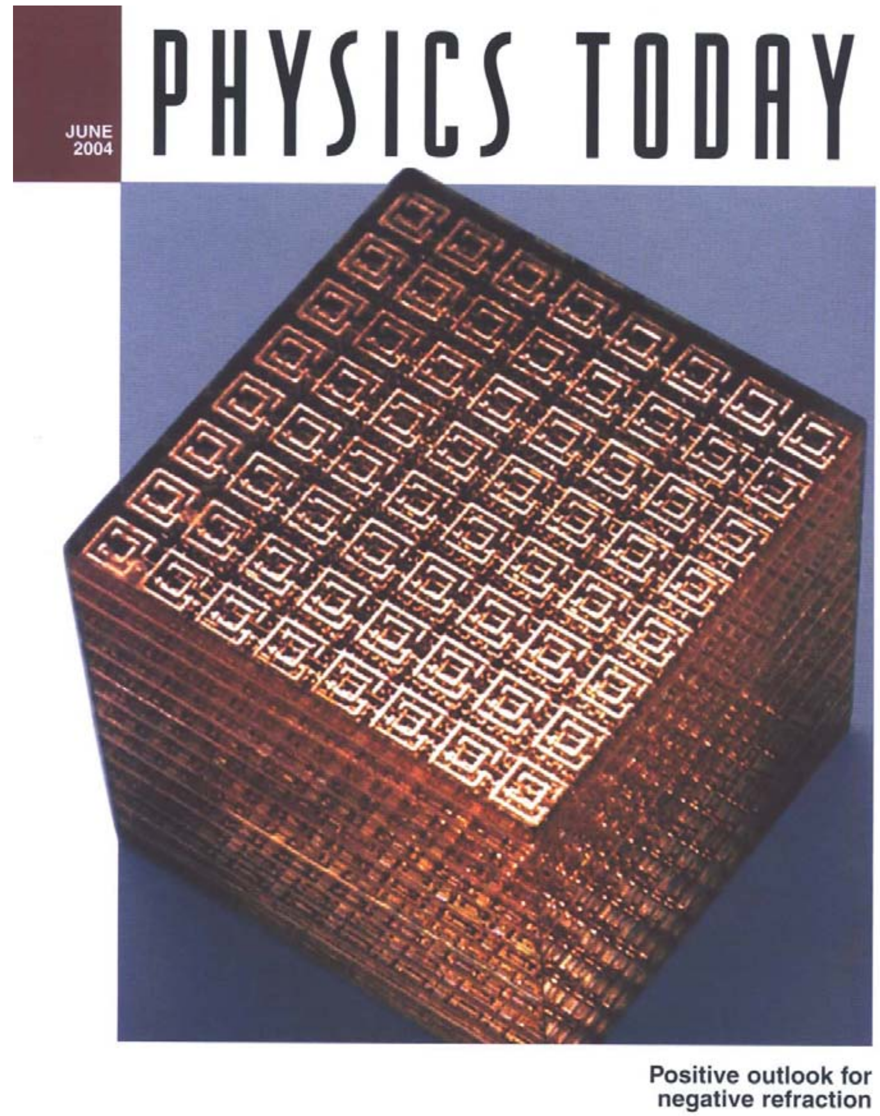
Metamaterials

Mike Lanagan, Khalid Rajab, Masato Iwasaki, Doug Werner and Elena Semouchkina

Materials Research Institute

Penn State University

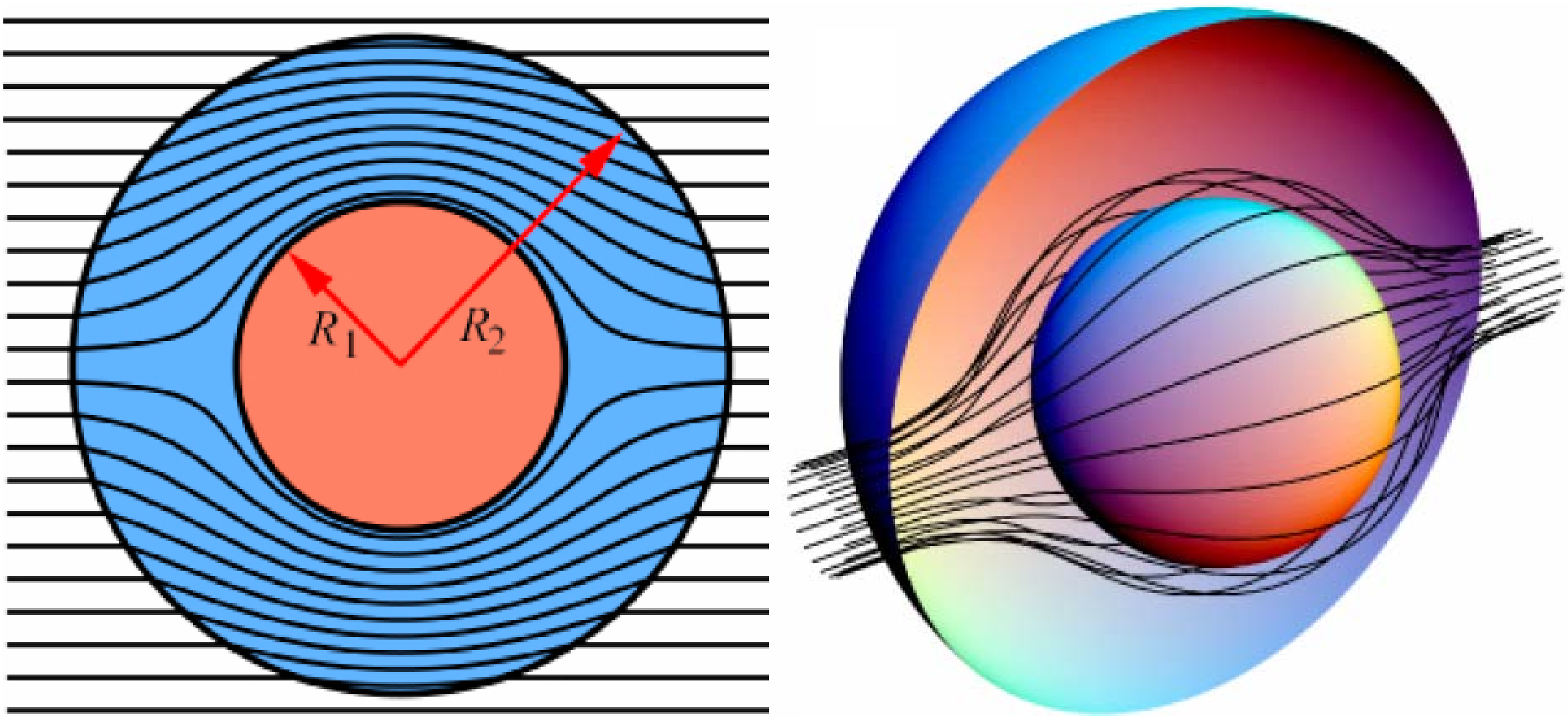
Metamaterials
Reading assignment



Metamaterials
(Based on negative permittivity)

- Description and Definition of Metamaterials
- Discovery and Application
- Creating materials with a resonant response
  - Plasmonic resonances for optics (not covered here)
  - Dielectric Resonators (interesting for Microwave and THz)
- Why Glass is an Interesting Medium for Metamaterials
  - Low dielectric loss
  - Easy to create spheres and periodic structures
  - Particular interest for mm-wave and THz frequencies

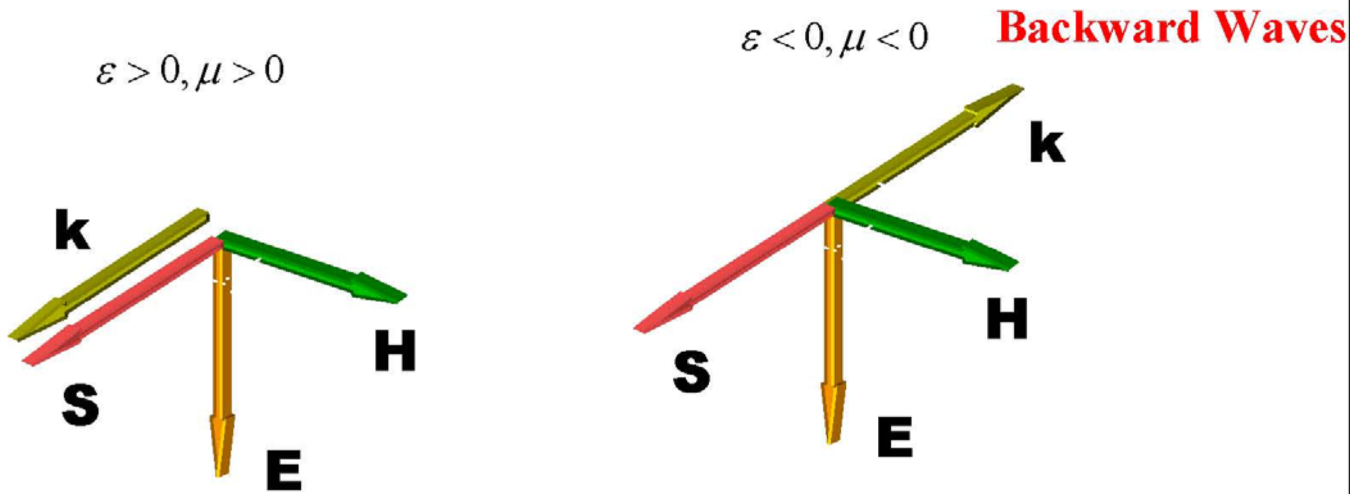
Electromagnetic Cloaking Using Metamaterials



*J.B. Pendry *et al.*, *Science* **312**, 1780 (2006).

LEFT-HANDED ε<0 AND μ<0 METAMATERIALS

Veselago, 1960s



Regular Materials

(right-handed)

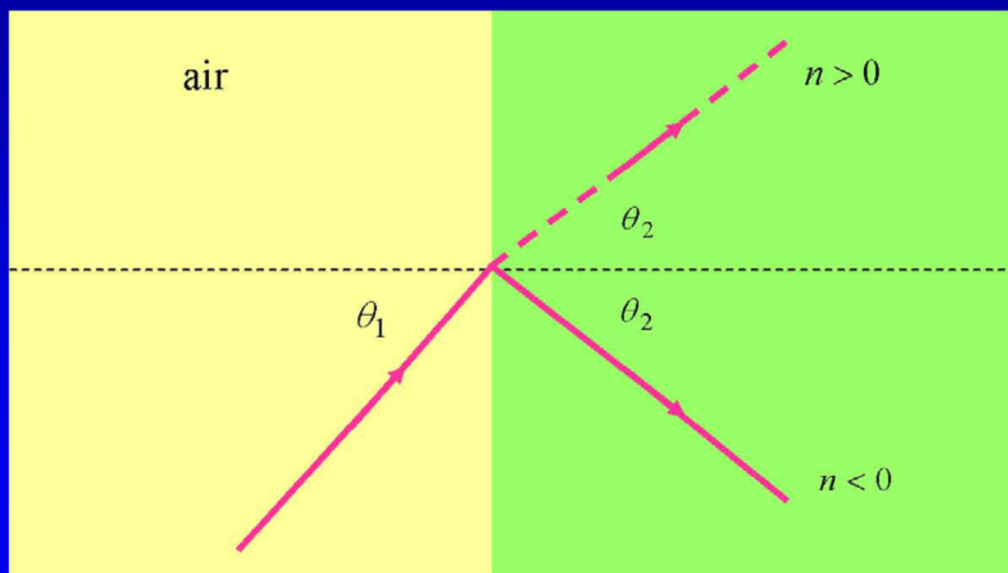
Left-handed Materials

$n = -\sqrt{\varepsilon\mu}$

Negative-Refractive-Index (NRI) Materials

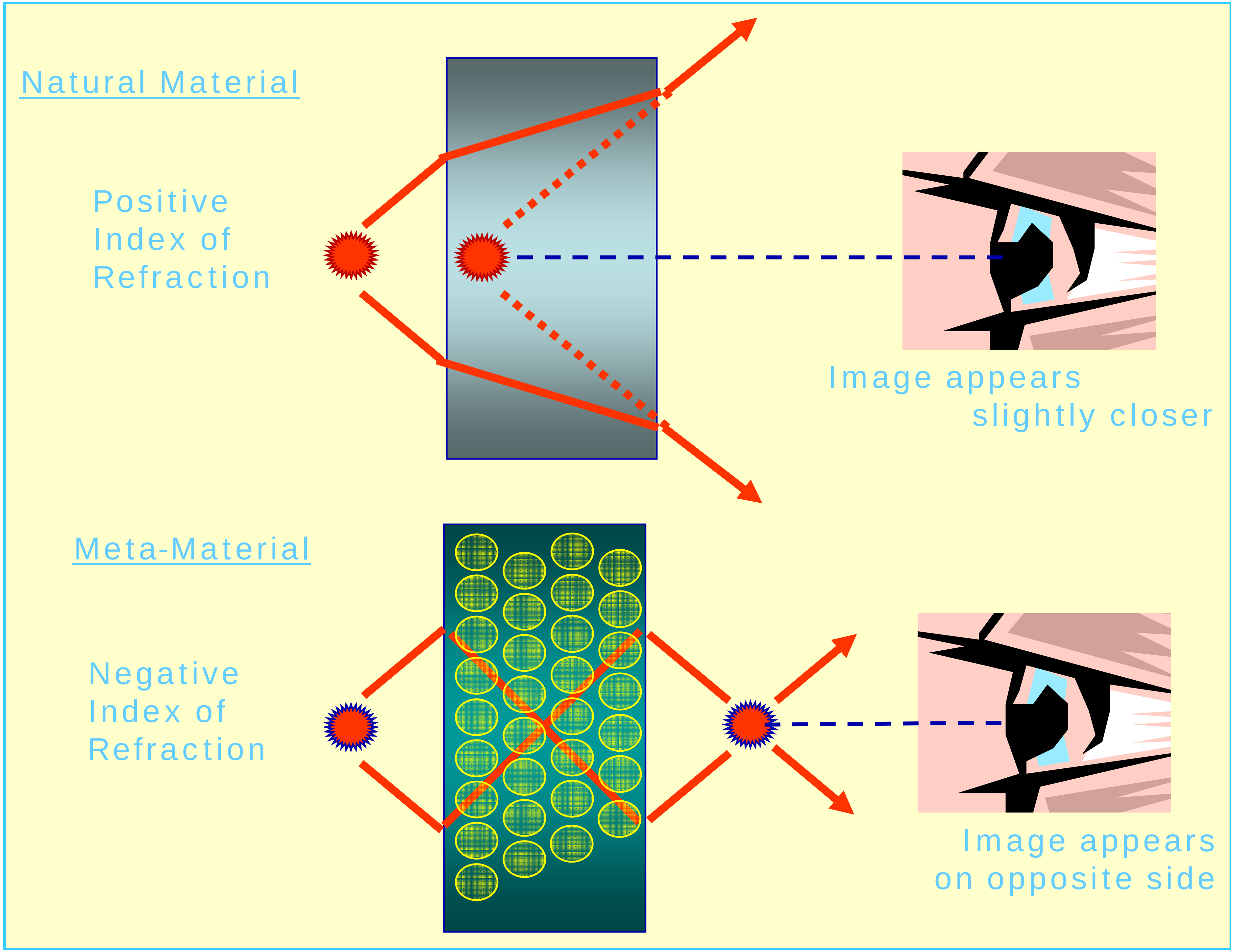
George V. Eleftheriades//University of Toronto

NEGATIVE REFRACTION



$$\frac{\sin\theta_1}{\sin\theta_2} = n$$

Negative-Refractive-Index (NRI) Media


Natural Material
Positive
Index of
Refraction
Image appears
slightly closer
Meta-Material
Negative
Index of
Refraction
Image appears
on opposite side

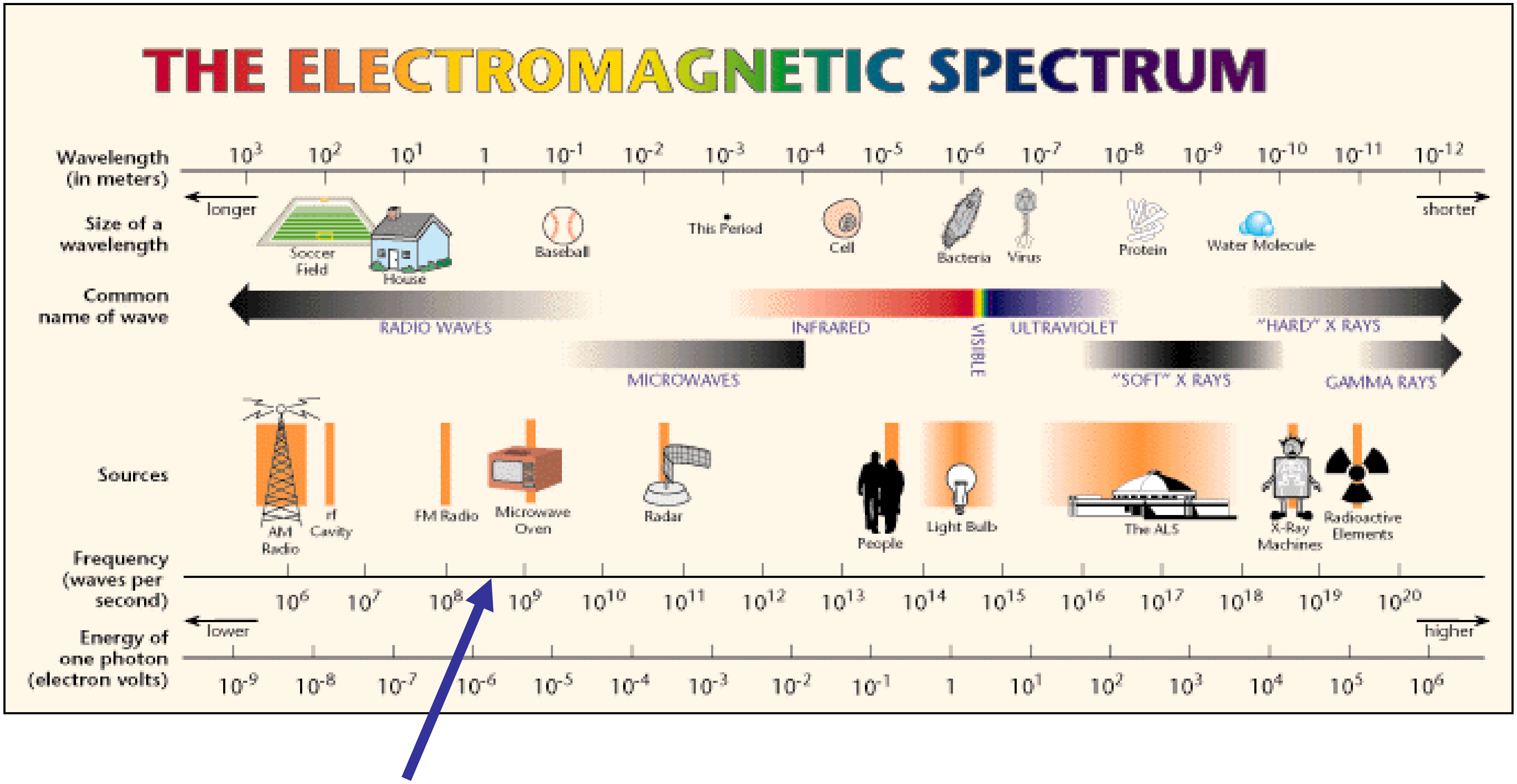
Discovery of Metamaterials

- Predicted by Veselago in 1960s
- First Experiments at UC San Diego in 2000
- Significant interest for applications
  - Magnetic resonant imaging
  - THz imaging
  - Cloaking

How can one make metamaterials?

- Think of resonance
  - Result of standing waves
  - Function of the wavelength and structure size
- We will use ring resonators as a example

Source: Wikipedia



Microwave: Resonator Size should be in centimeters

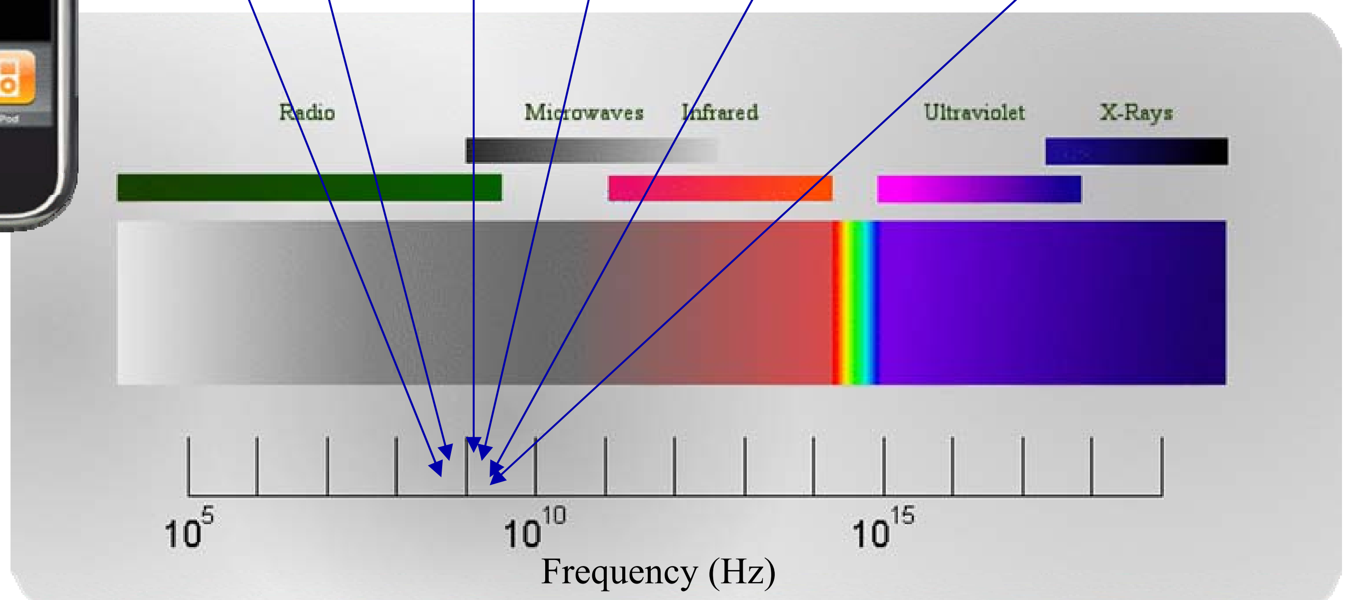
Resonant frequency

$$f_r \propto \frac{1}{d\varepsilon_r}$$

d=Resonator size

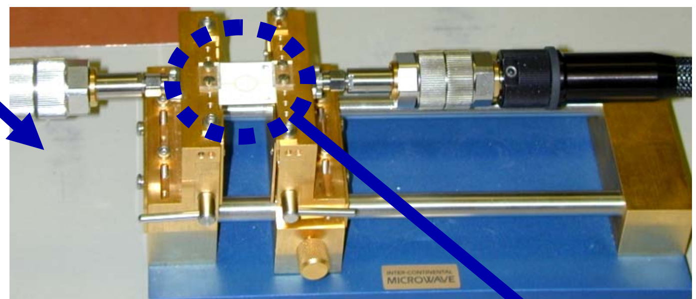
Apple iPhone and Microwaves

- Quad-band GSM
  850 MHz 900 MHz 1800 MHz 1900 MHz
- Bluetooth
  2400 MHz
- 802.11 WiFi
  2400 MHz

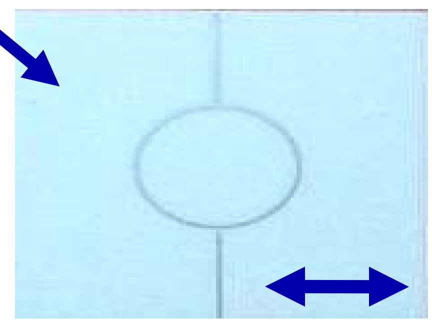


Ring Resonator Measurements

HP8510T Network Analyzer
45 MHz to 26 GHz



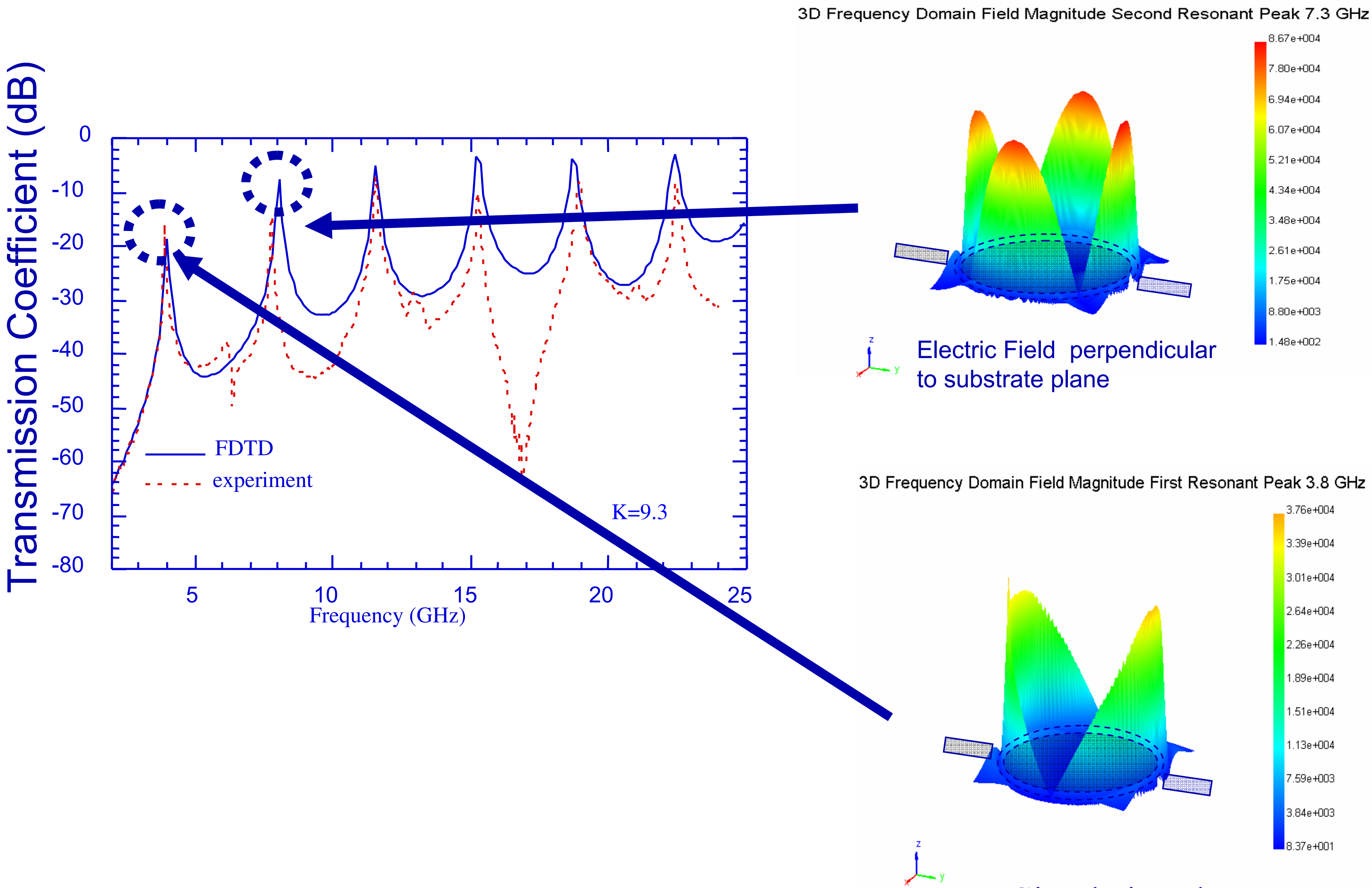
Intercontinental Microwave
Fixture



1 cm

Ring
Resonator

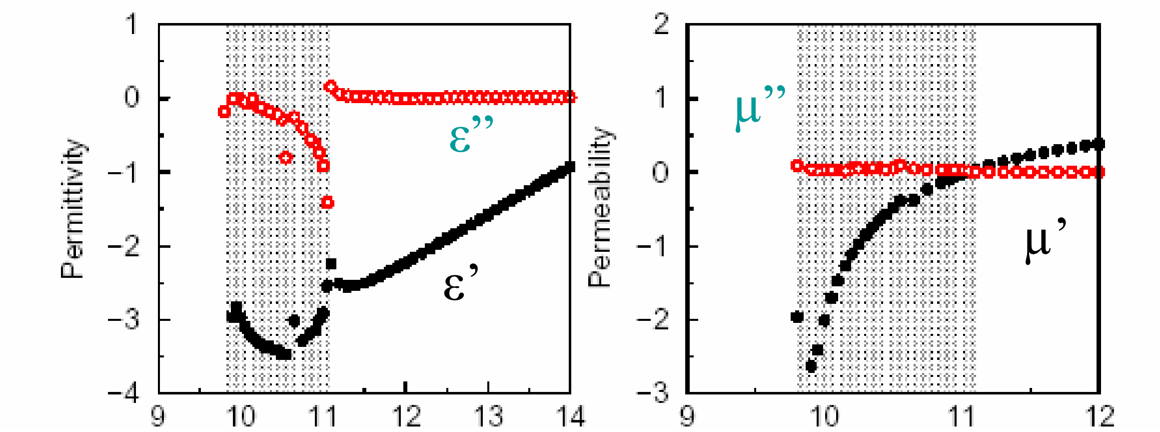
Resonant Behavior in Ring Resonators

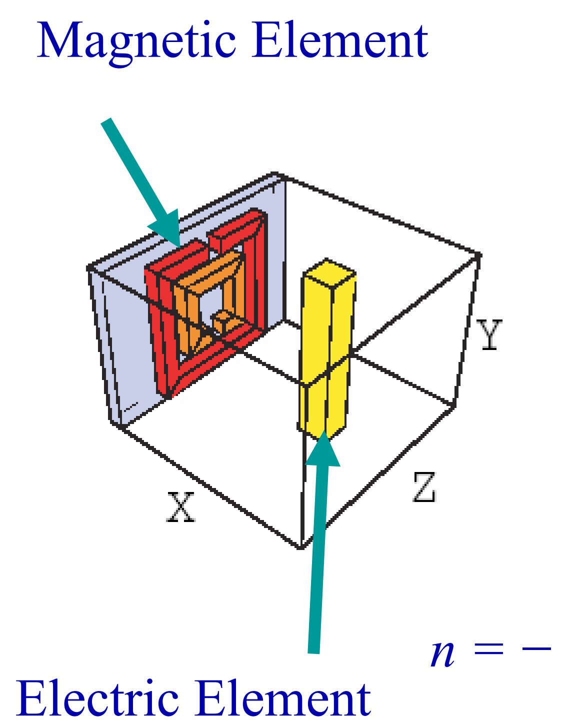


Simulations by L. Haney

Double Negative Materials*

Frequency in GHz →



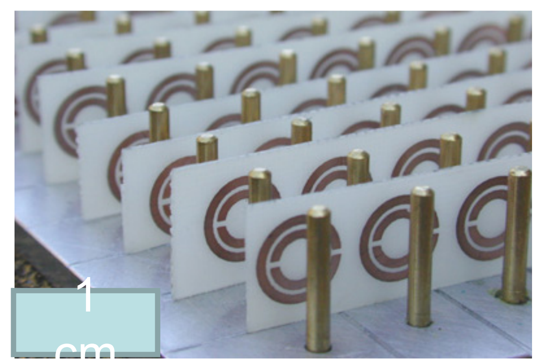


$n = -(\varepsilon\mu$

Negative permeability results from the resonating magnetic element

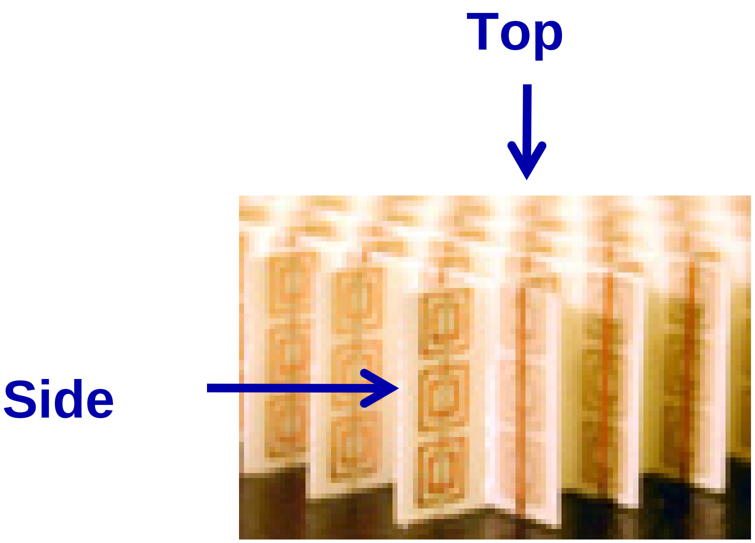
Negative permeability results from the resonating electric element

Recall $n = -(\varepsilon\mu)^{1/2}$

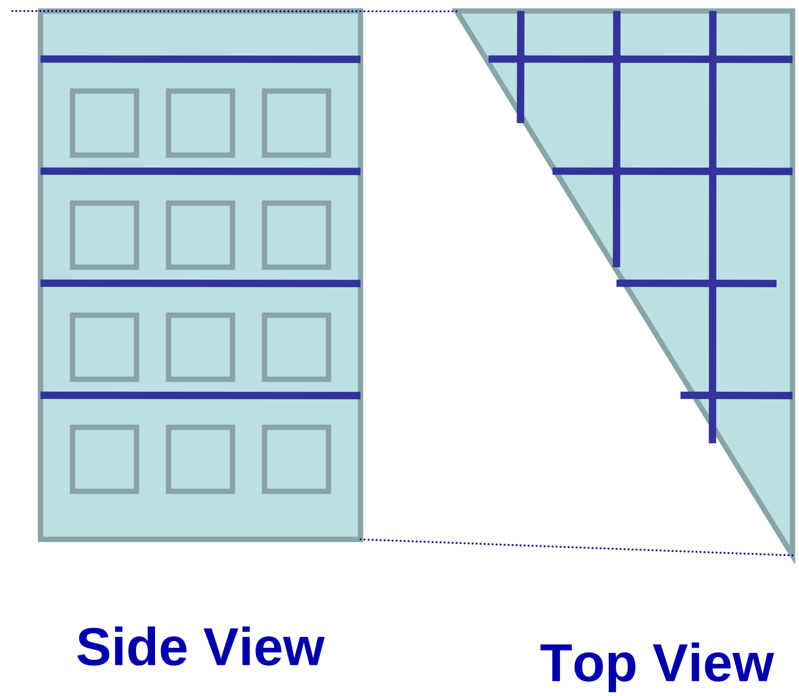


P.M. Markos and C. M. Soukoulis, Opt. Exp., p. 649 (2003)

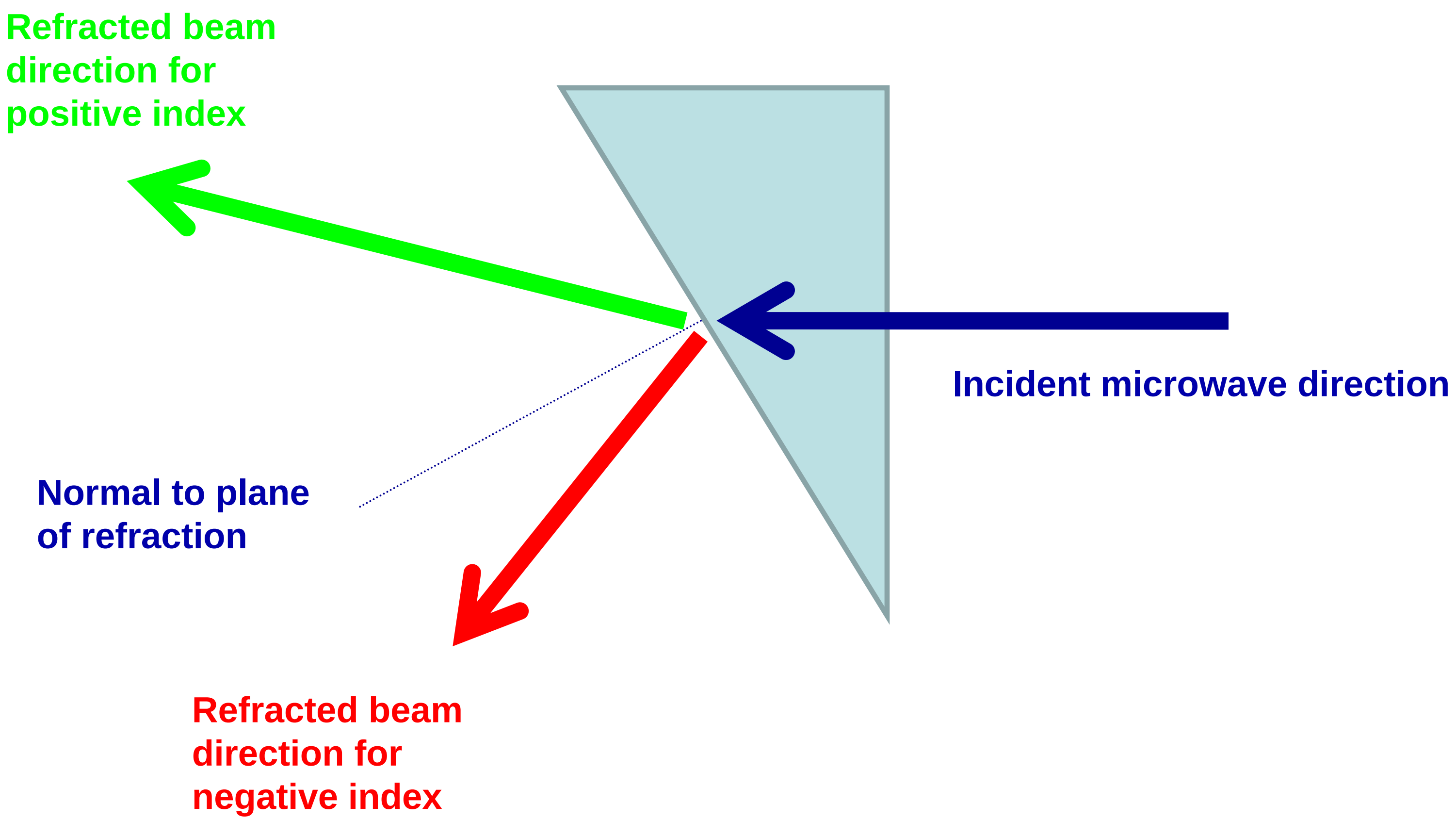
Critical Experiment for Metamaterial



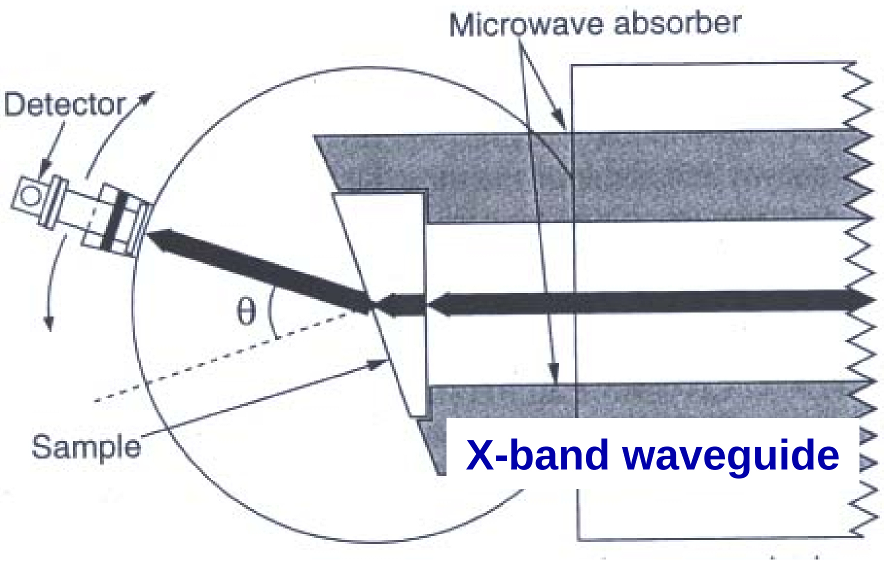
First make a metamaterial prism

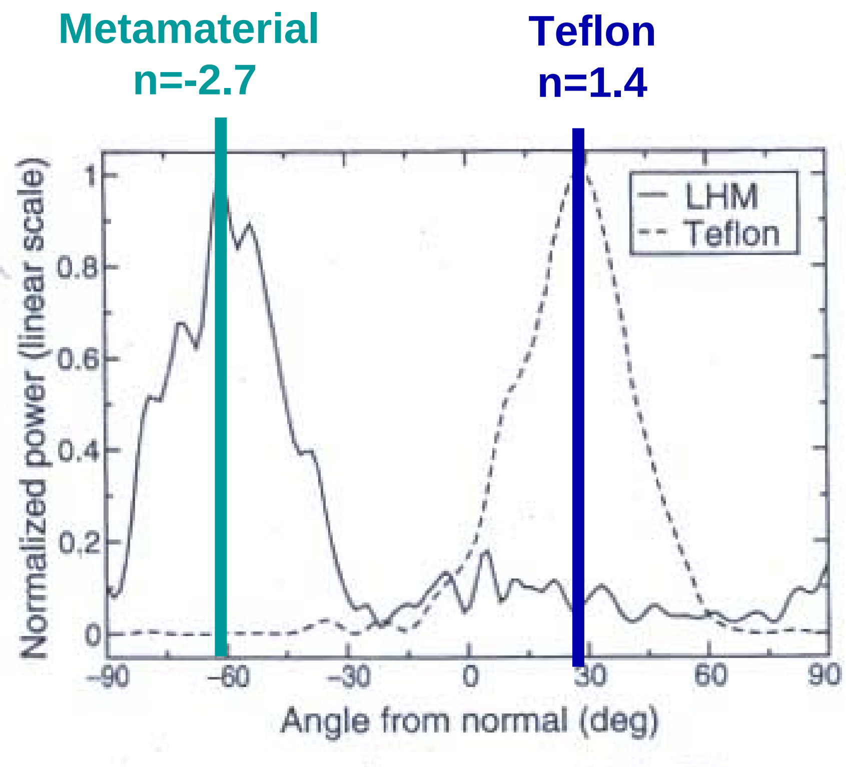


Critical Experiment for Metamaterial



Experimental Confirmation of a Meta-material*





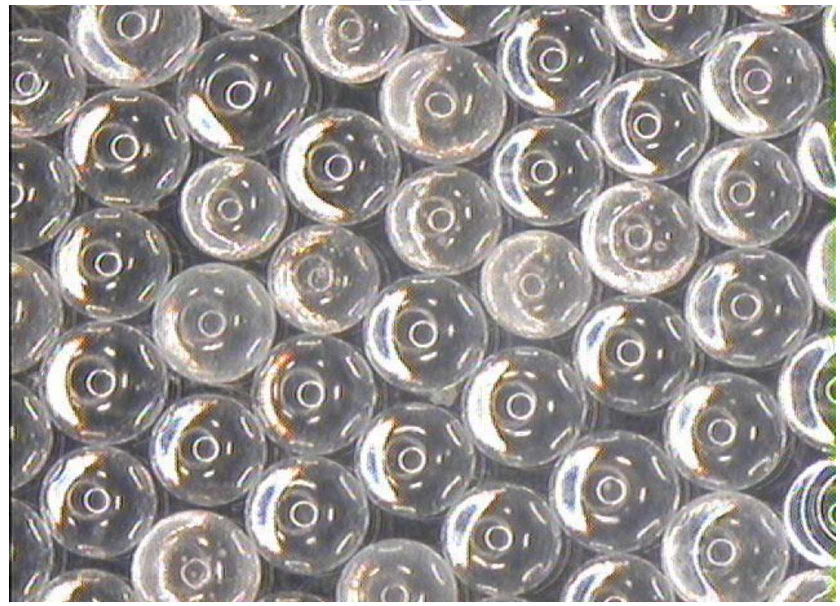
R. A. Shelby et al., Science, pg. 77 (2001)

Why not other resonant structures for Metamaterials?

Ceramic Cylinders

Microwave ceramic resonators made by Murata

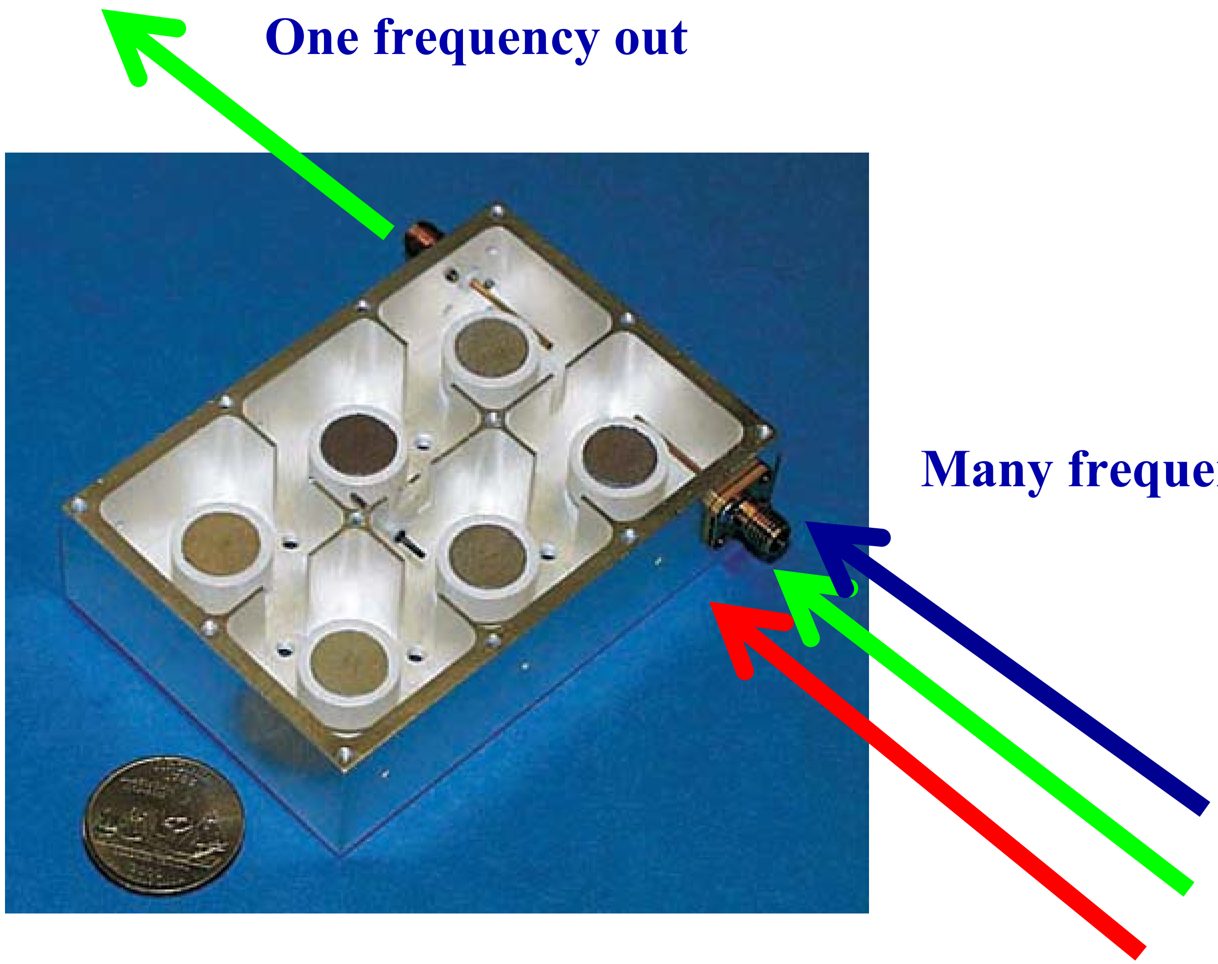
Glass Spheres



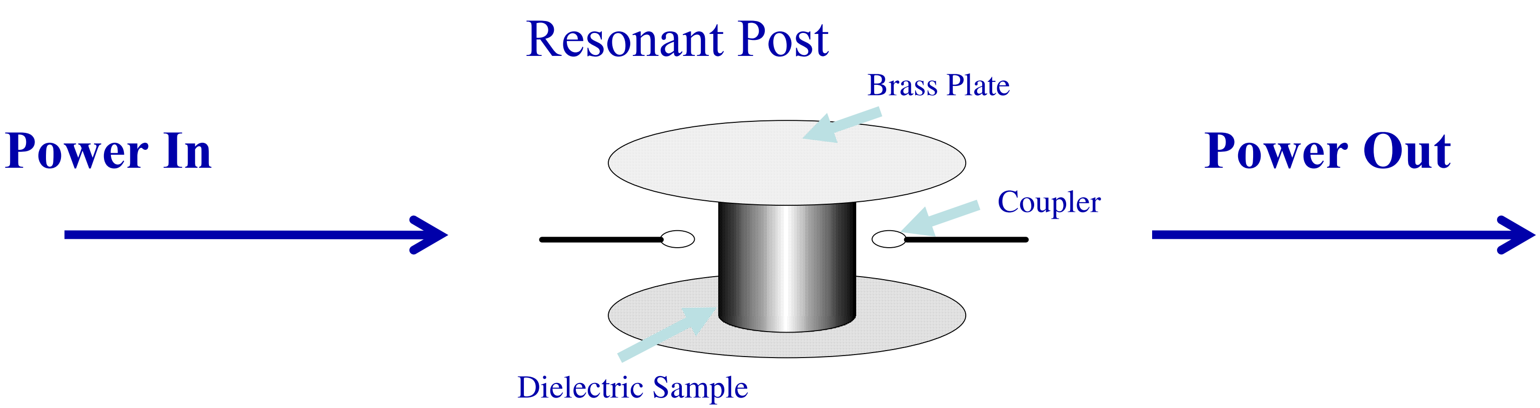
1 mm diameter silica spheres. Fabricated by Amanda Baker

Dielectric properties and geometry are key factors for resonators

Microwave Filter for Cell Phone Base Station: Commercial Application of Ceramic Dielectric Resonators

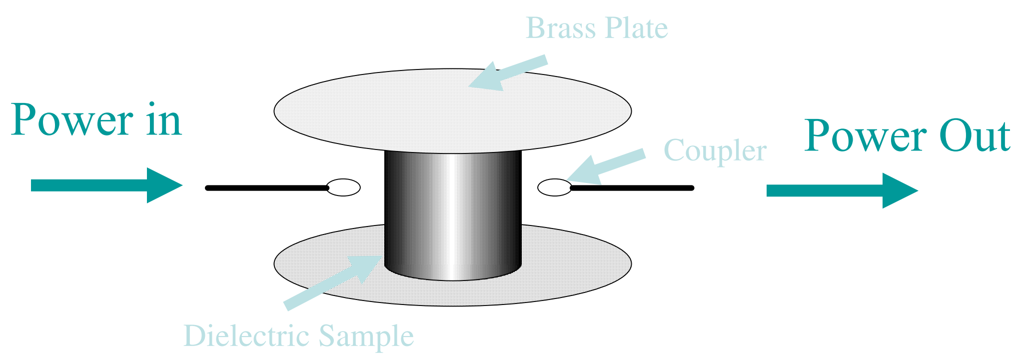


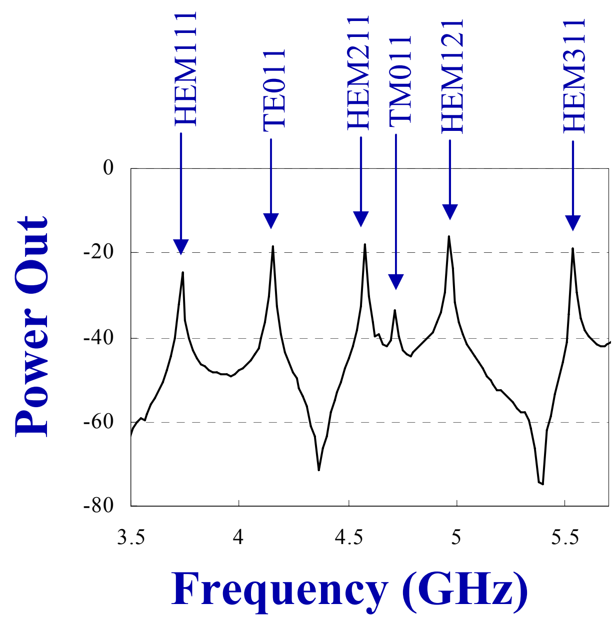
Microwave Characterization



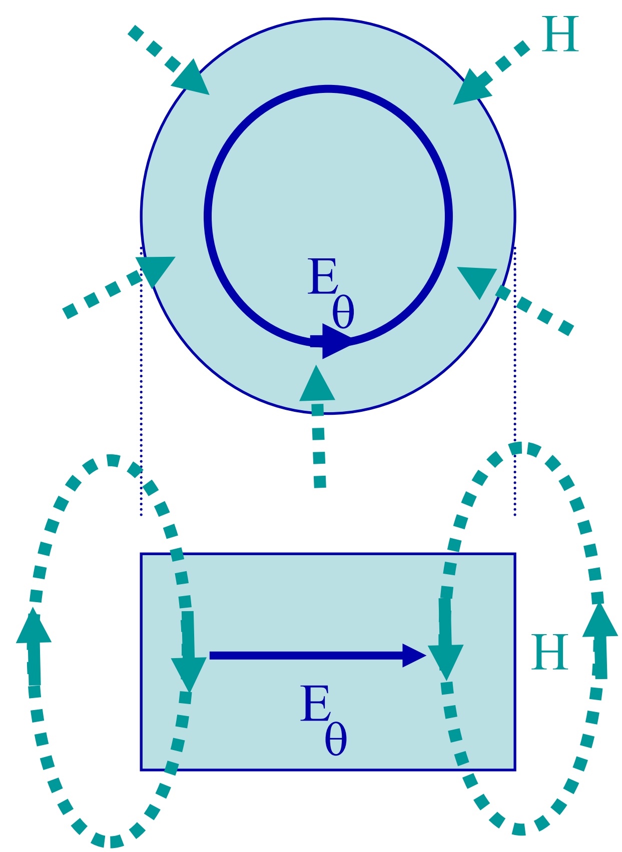
Resonant Post Method*

Resonant Post Method



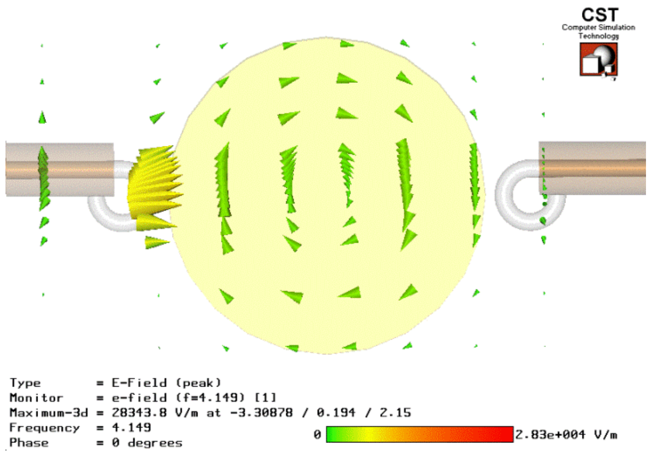
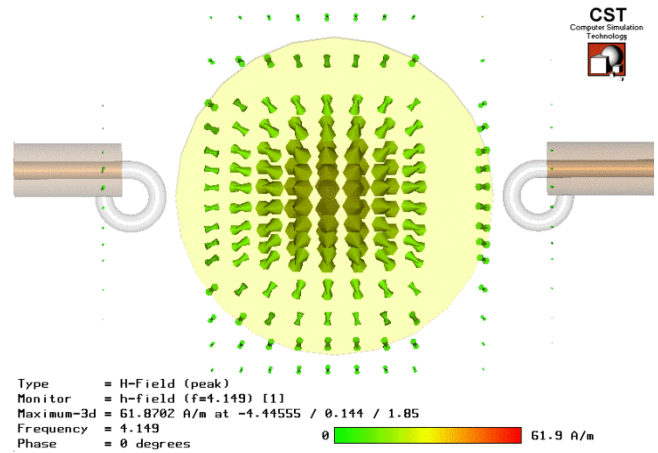
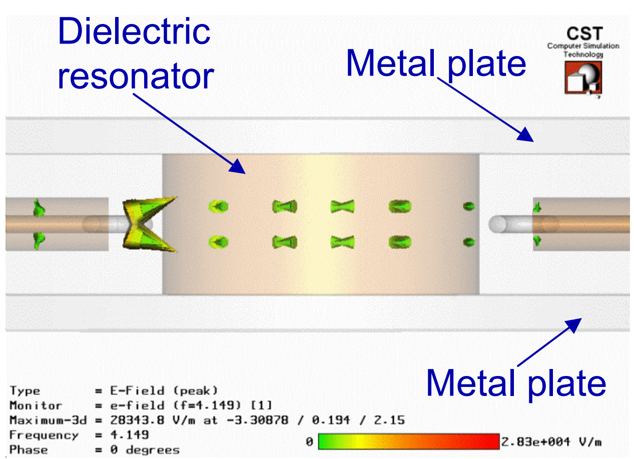
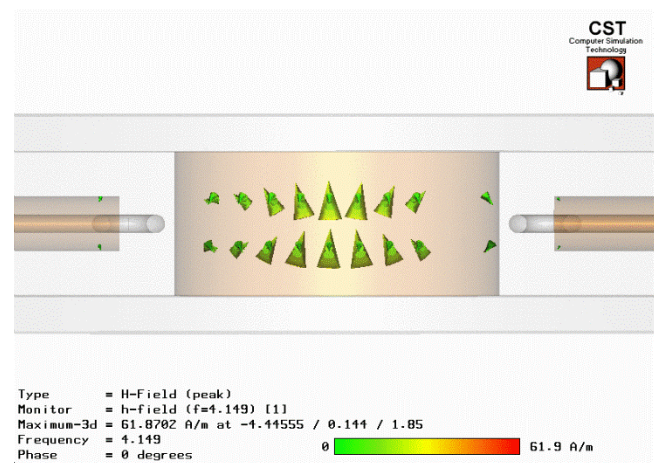


TE_{011} Mode



*Hakke and Coleman, IEEE (1960)

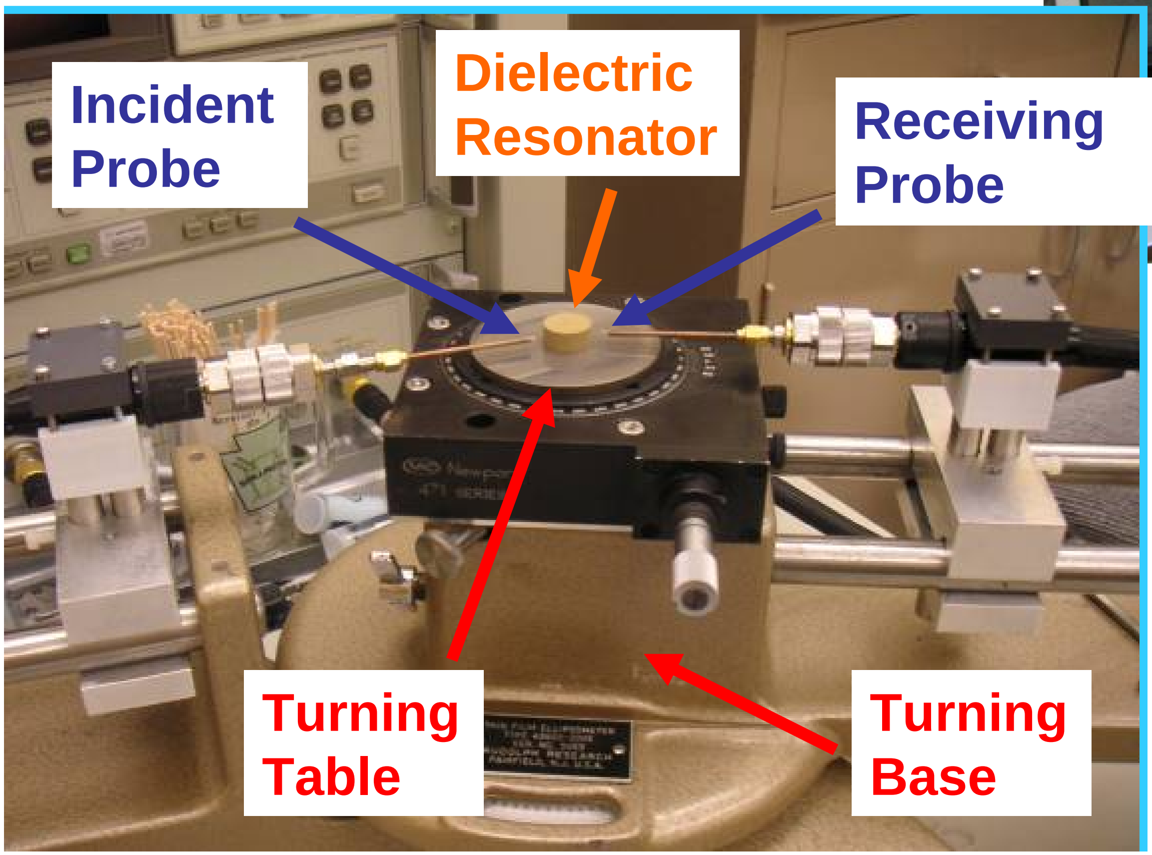
Field Distribution of TE_{011} Mode from FDTD Simulation

TE011	E-field	H-field
Top-view		
Side-view		

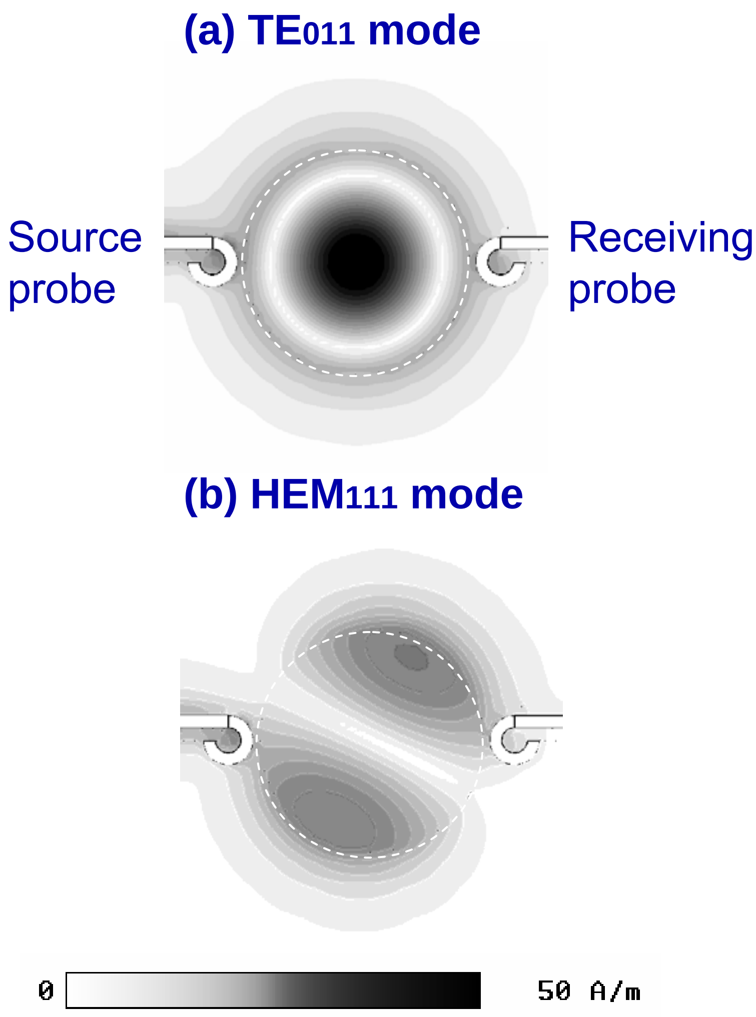
M. Iwasaki

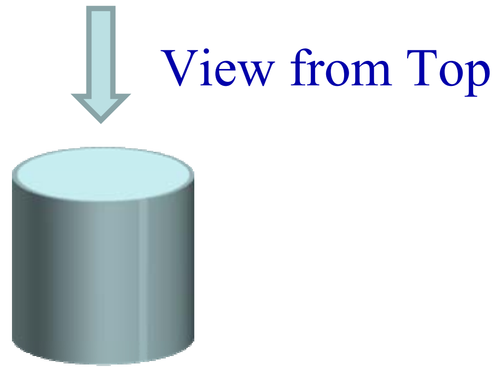
Masato "Mat" Iwasaki, Visiting Scientist
NGK Spark Plug

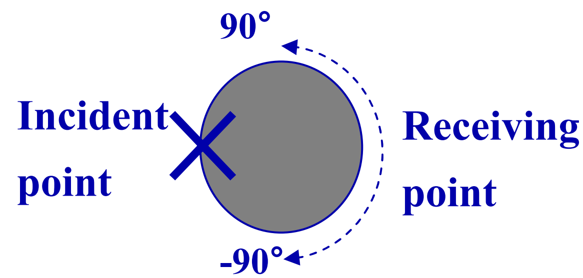
- **Meta-materials**
- **Electromagnetic Simulation**

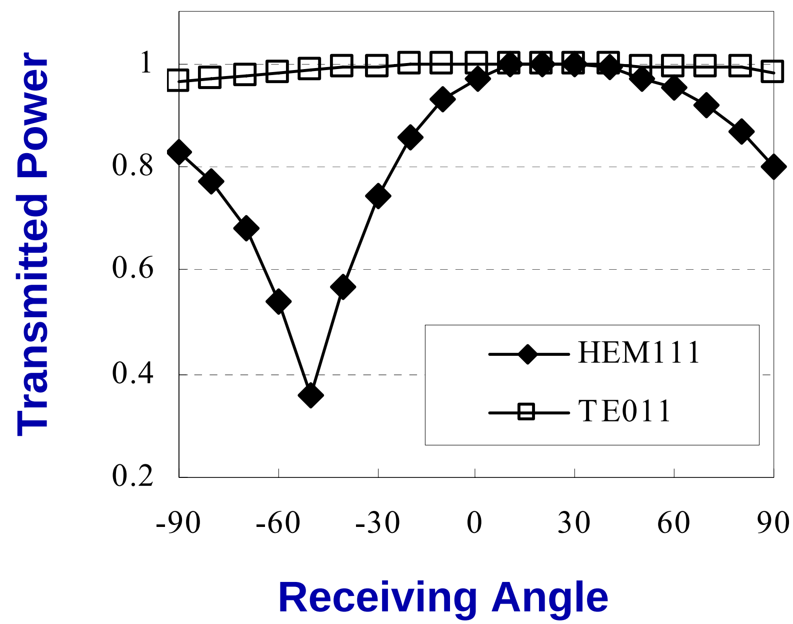


Simulation and Experimental Results





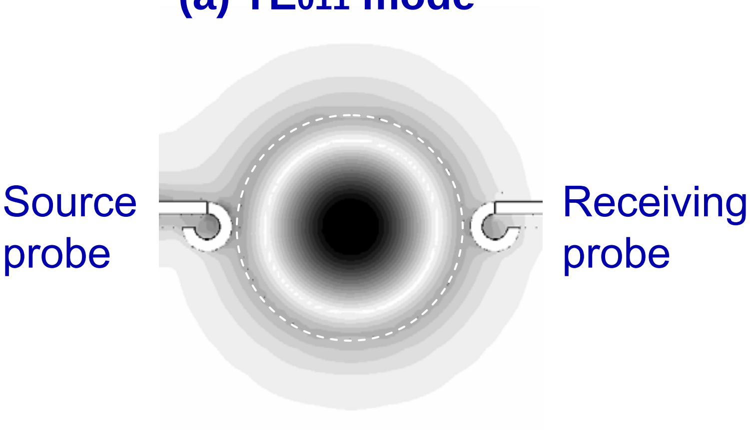




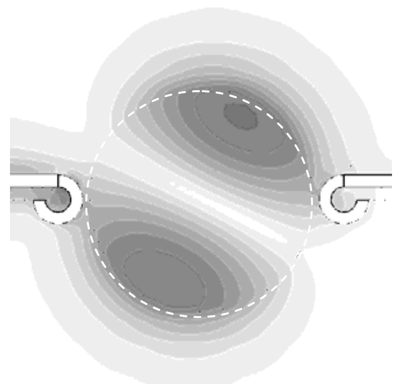
Electric field distribution of single DR

-By simulation results, magnetic field distributions were drawn in longitudinal direction at the half height of DR.

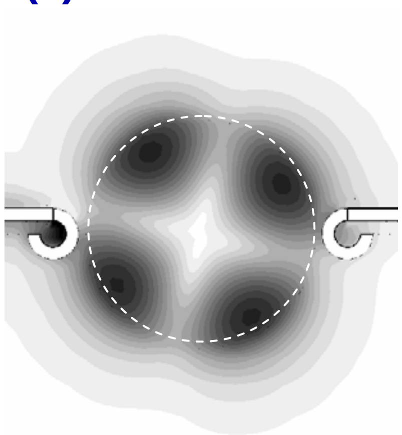
(a) TE_{011} mode



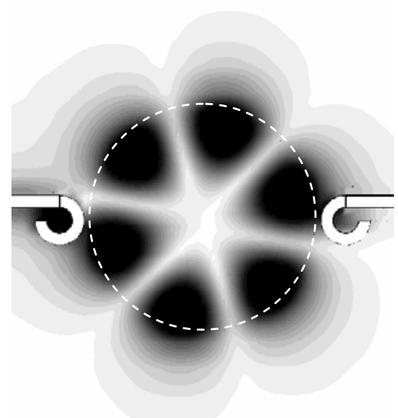
(b) HEM_{111} mode



(c) HEM_{211} mode



(d) HEM_{311} mode



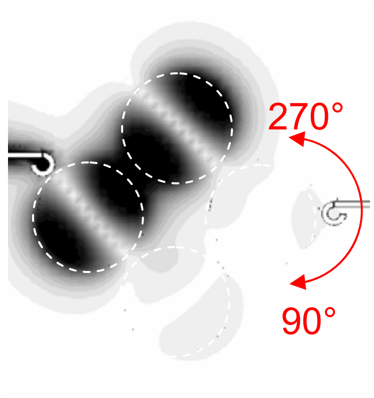
M. Iwasaki

Field Distribution of Square DR Cluster

-Magnetic field distributions in longitudinal direction at the half height of DR were drawn.

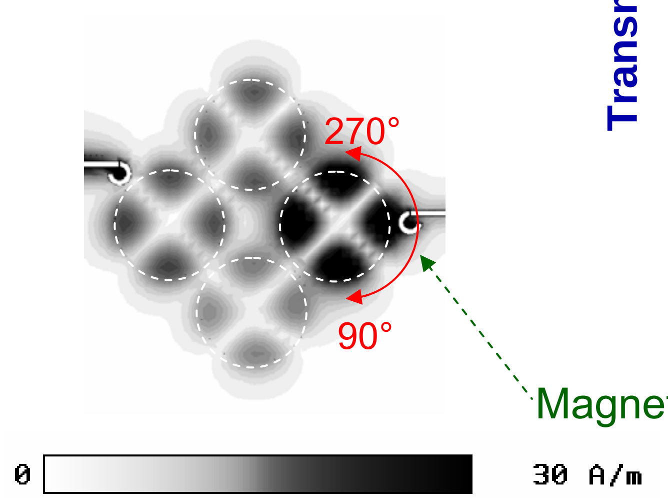
HEM111 mode

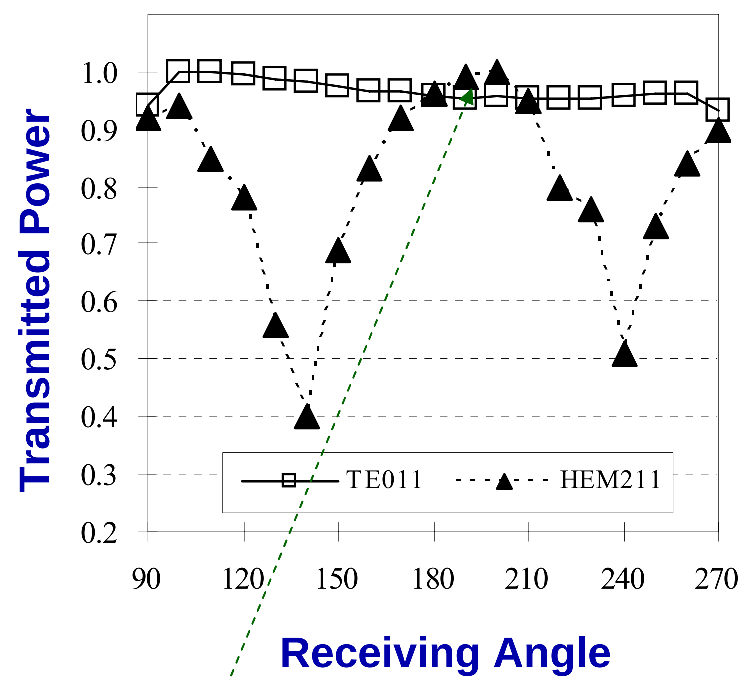
No HEM 111 mode propagation for square symmetry



HEM211 mode

HEM 211 mode propagation for square symmetry is consistent with lattice symmetry



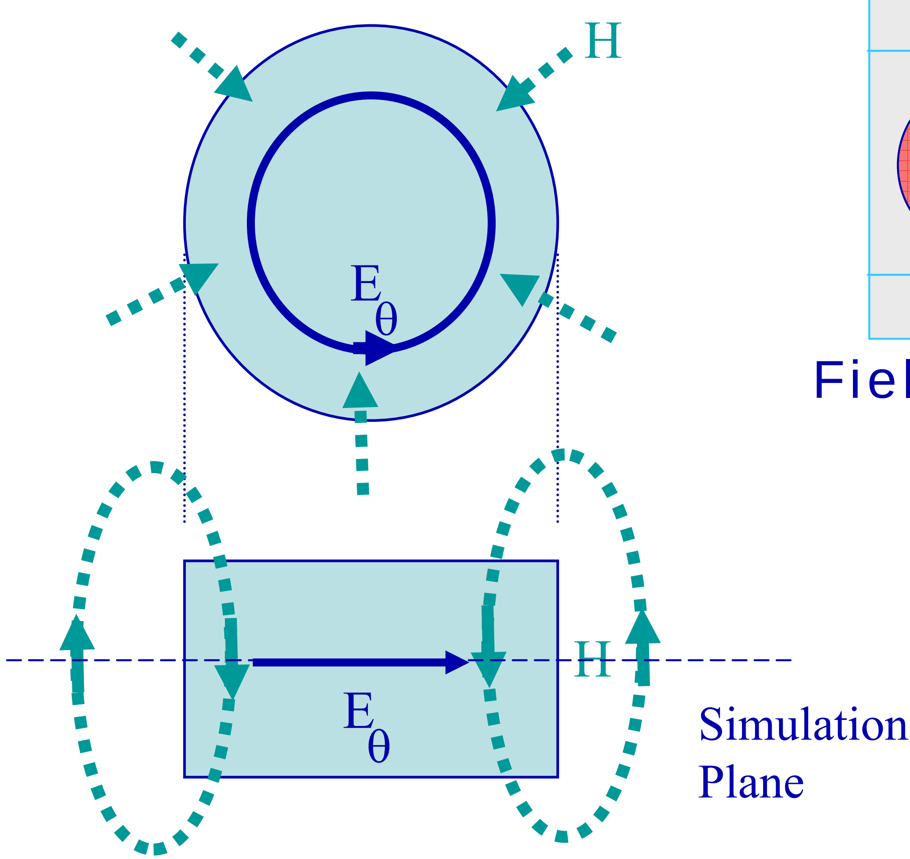


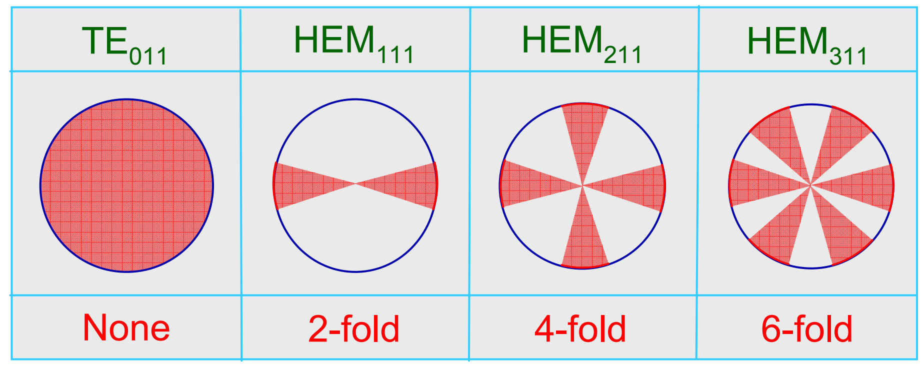
Magnetic field maximum

M. Iwasaki

Magnetic Field Symmetry in Dielectric Resonator Modes

TE_{011} Mode





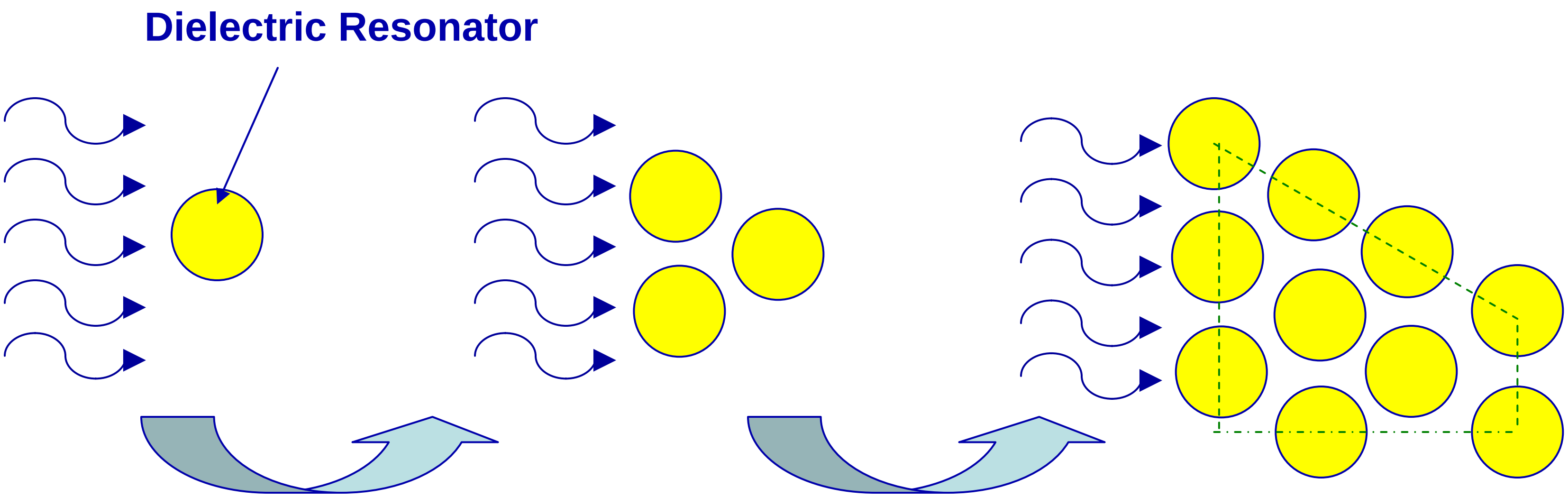
Field Distribution of Each Resonant Mode

M. Iwasaki, E.A. Semouchkina, G.B. Semouchkin, K.Z. Rajab*, C.A. Randall, and M.T. Lanagan, "Symmetry Matching of Hybrid Modes for Dielectric Metamaterials," *Japanese Journal of Applied Physics* , (2006)

Moving from individual resonators to clusters to arrays

-For characterizing the refracted waves through wedge-shaped DR arrays,
Simulations and measurements starting from one DR will be performed.

-**Excitation with large area** should be employed.

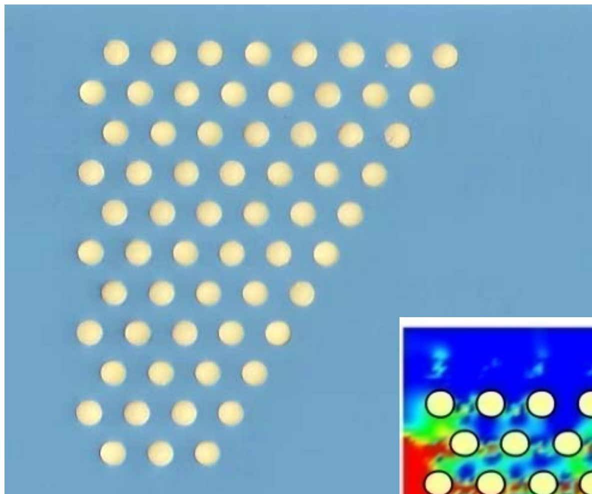


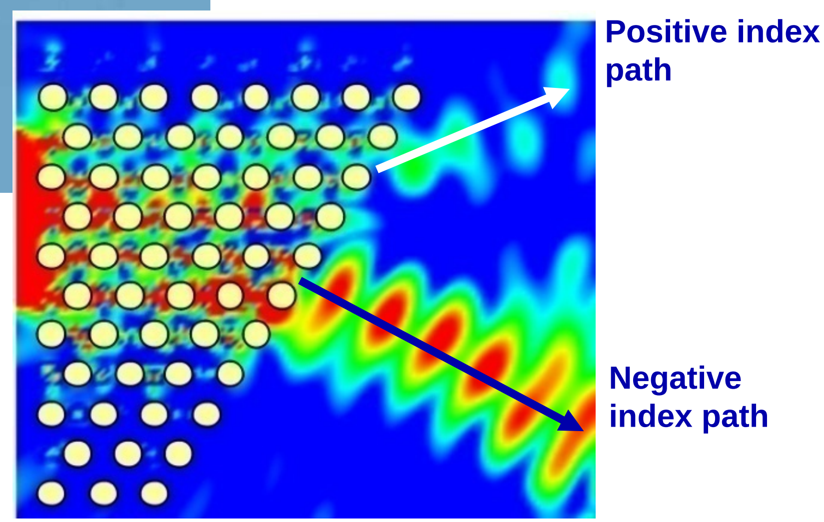
Increasing the number of DRs to form wedge shape

: Incident waves

M. Iwasaki

Ceramic Dielectric Resonator Arrays





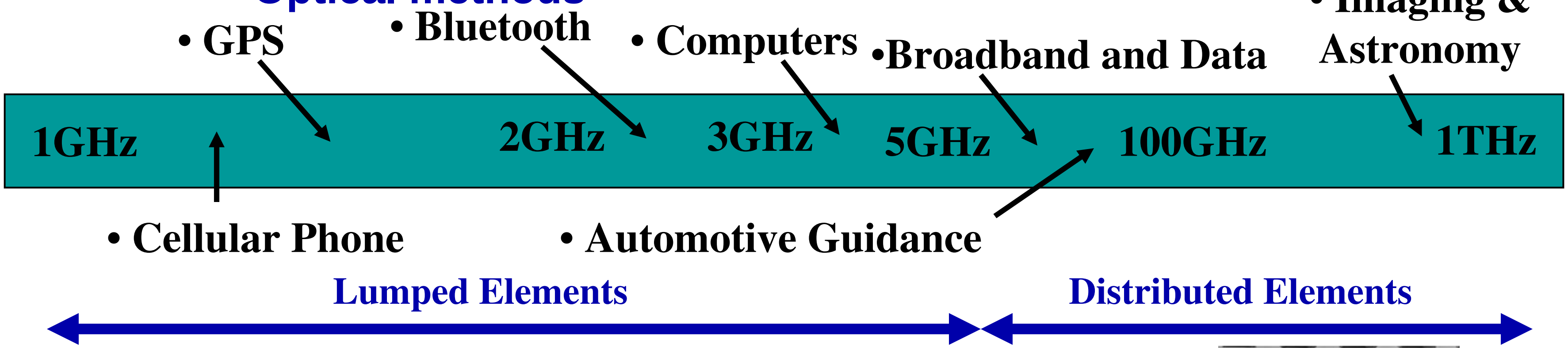
Top View
Resonator array
in a low permittivity matrix

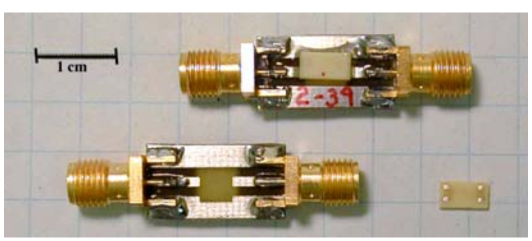
Simulation by Elena Semouchkina

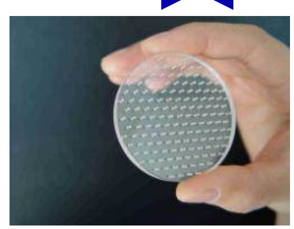
Operating
Frequency 15 GHz

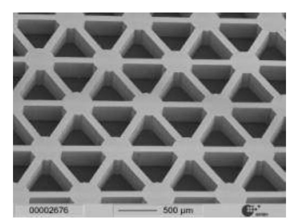
Moving Beyond Microwaves

- **Materials Trends**
  - **Higher application frequencies (both communications and computing)**
  - **Lower permittivity (dielectric constant) and lower loss (higher Q)**
  - **All dielectric (no metal?) structures**
- **Design and Process Implications**
  - **More compact designs**
  - **Dimensional control becomes more critical**
- **Measurement Implications**
  - **Optical methods**



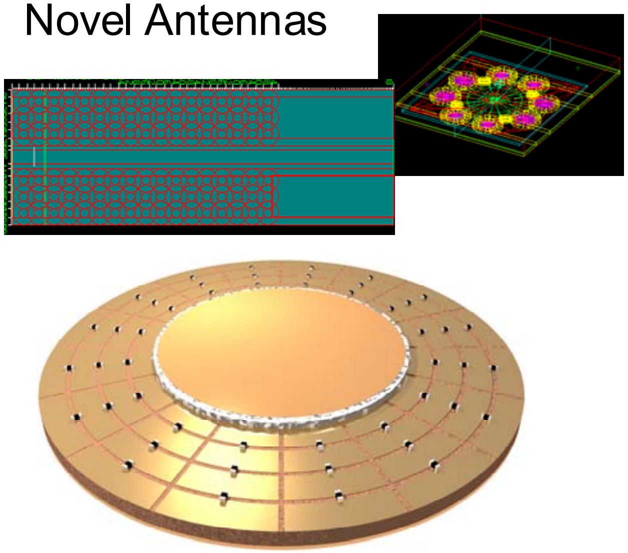




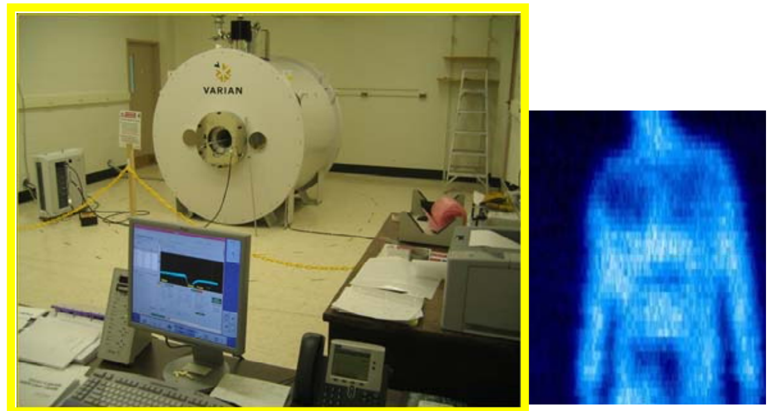


*http://www.fz-juelich.de/isg/isg2/isg2-sh/ebg_materials.htm

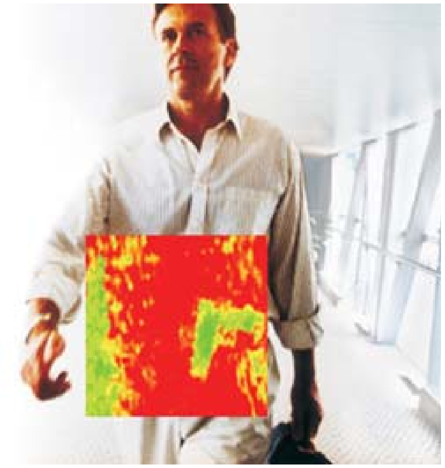
Novel Antennas



MRI Systems

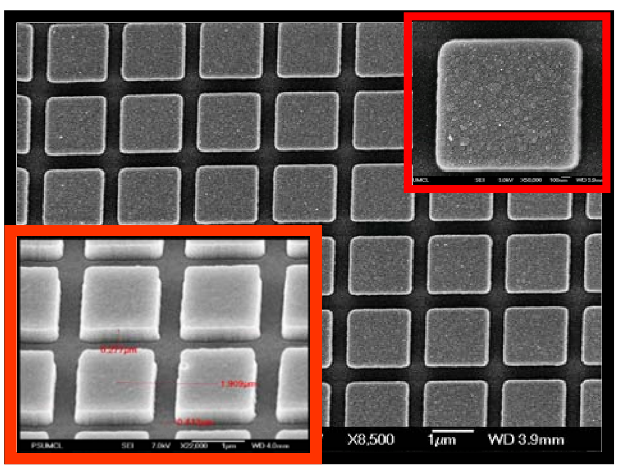


THz Imaging



Systems Level: *Innovative devices for precision measurement, shielding, imaging, telecommunications, energy, and biomedicine*

Metamaterials for IR Devices

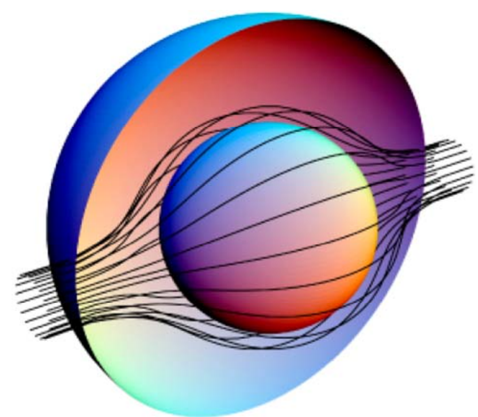


Superlens-based Nanopatterning

Fang *et al*, Science, 2005

D. Werner

EM Cloaking

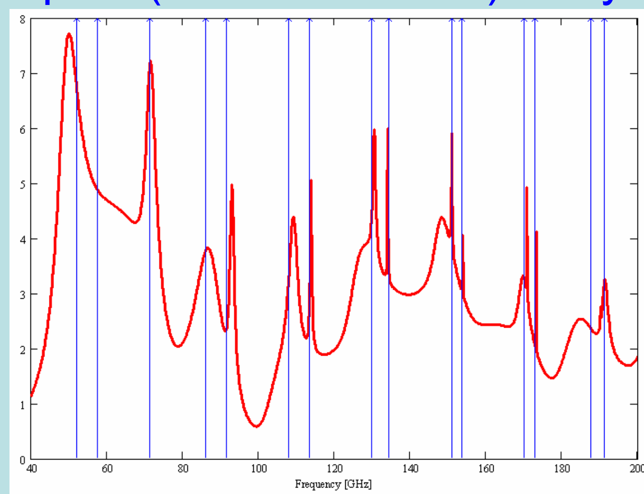


Pendry *et al.*, *Science*, 2006

THz Characterization of Arrays

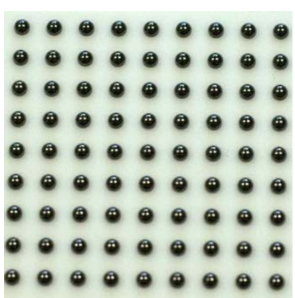
- Materials:
  - Silicon Nitride, Si_3N_4 $\varepsilon_r \approx 8.9$
  - Brass
- Lattices:
  - Square
  - Hexagonal
- Unit cells:
  - 4mm
  - 3mm
  - 2mm

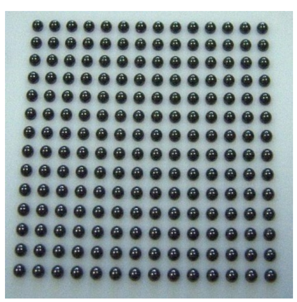
Mie theory (single sphere) and loosely coupled (unit cell = 4mm) array

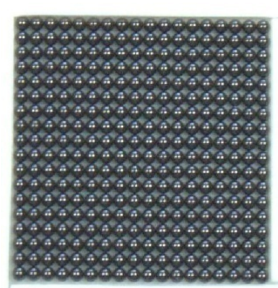


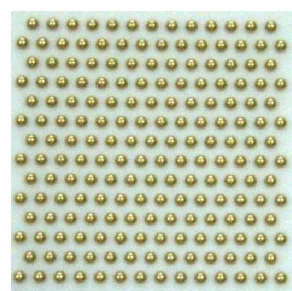
Blue – Measured resonant frequencies

Red – Scattering cross-section (Mie)





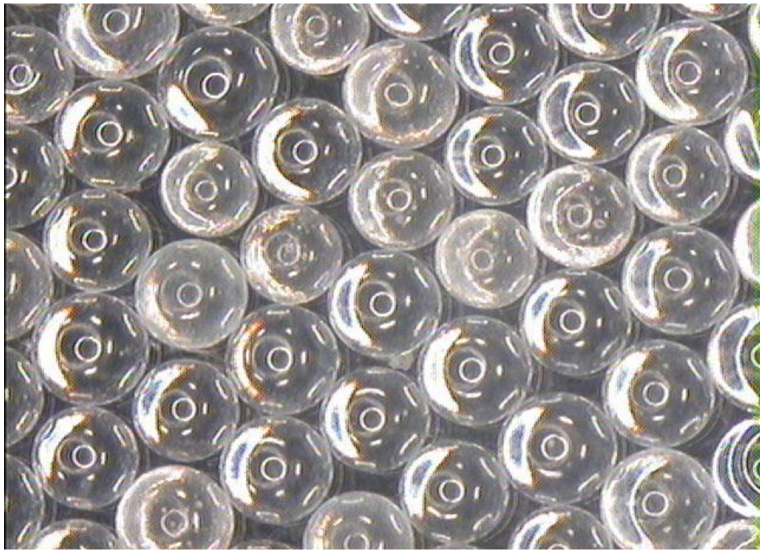




Khalid Rajab

What's Next for Ceramic Dielectric Materials and Structures?

- Higher Frequencies pushing into the THz range
- What size resonators do we need?
- What types of dielectrics (glass?) do we need?



1 mm diameter silica spheres.
Fabricated by Amanda Baker

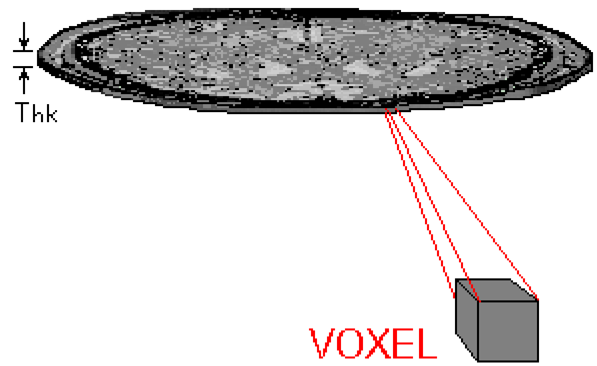
Quiz

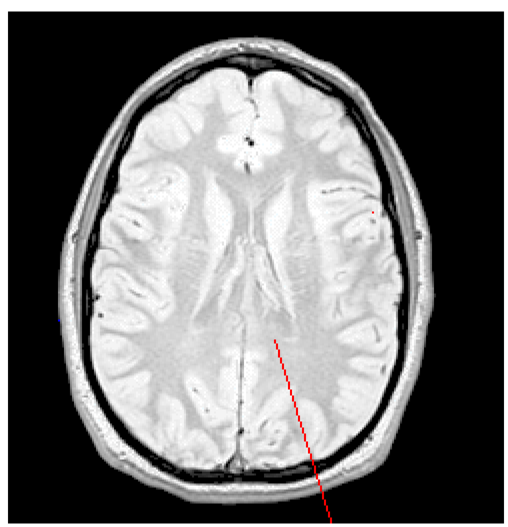
- What material property affects the resonator frequency?
- What other parameter affects resonance?
- Why would we NOT want to make the resonator too small?
- What functionalities of glass are potentially important for metamaterials?

Metamaterials in Magnetic Resonance Imaging?

- Overview of how MRI works
- Use of resonators in MRI (not metamaterials yet)
- Case Study: Glass Metamaterials for MRI

Background on Magnetic Resonance Imaging (MRI)





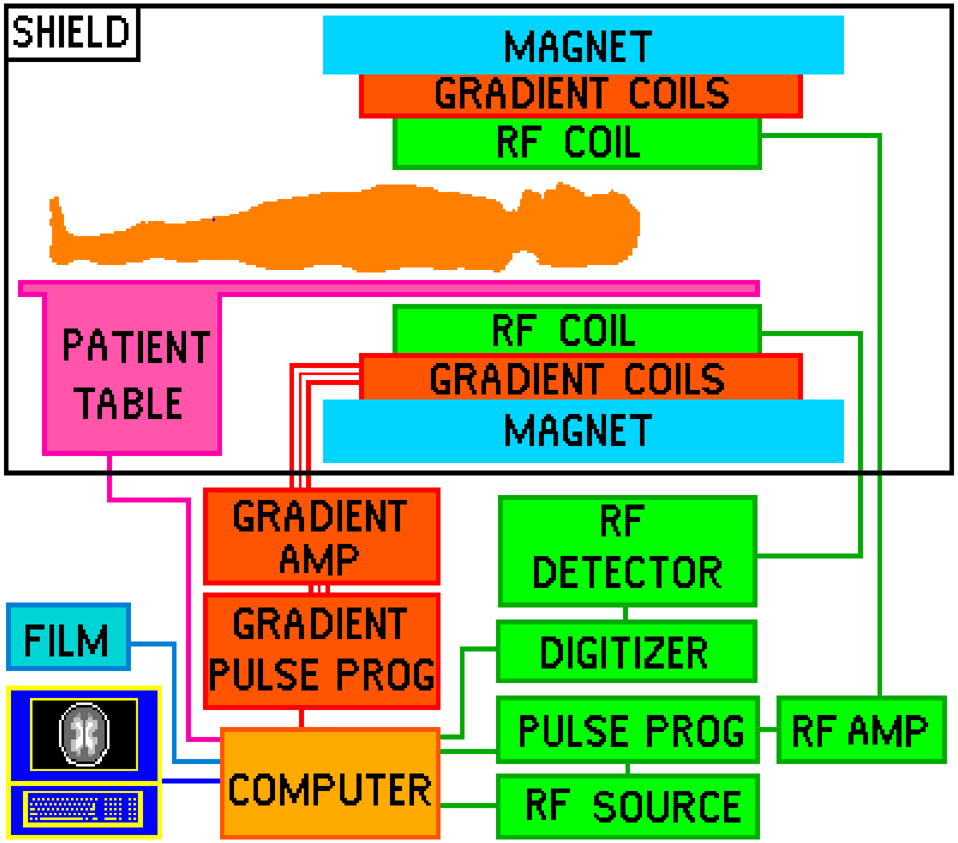
Magnetic resonance imaging is based on the absorption and emission of energy in the radio frequency range of the electromagnetic spectrum. Radio Frequency (RF) Coils are used to transmit and receive energy from the samples.

(Adapted from Reference: http://www.cis.rit.edu/htbooks/mri)

Background on MRI contd.

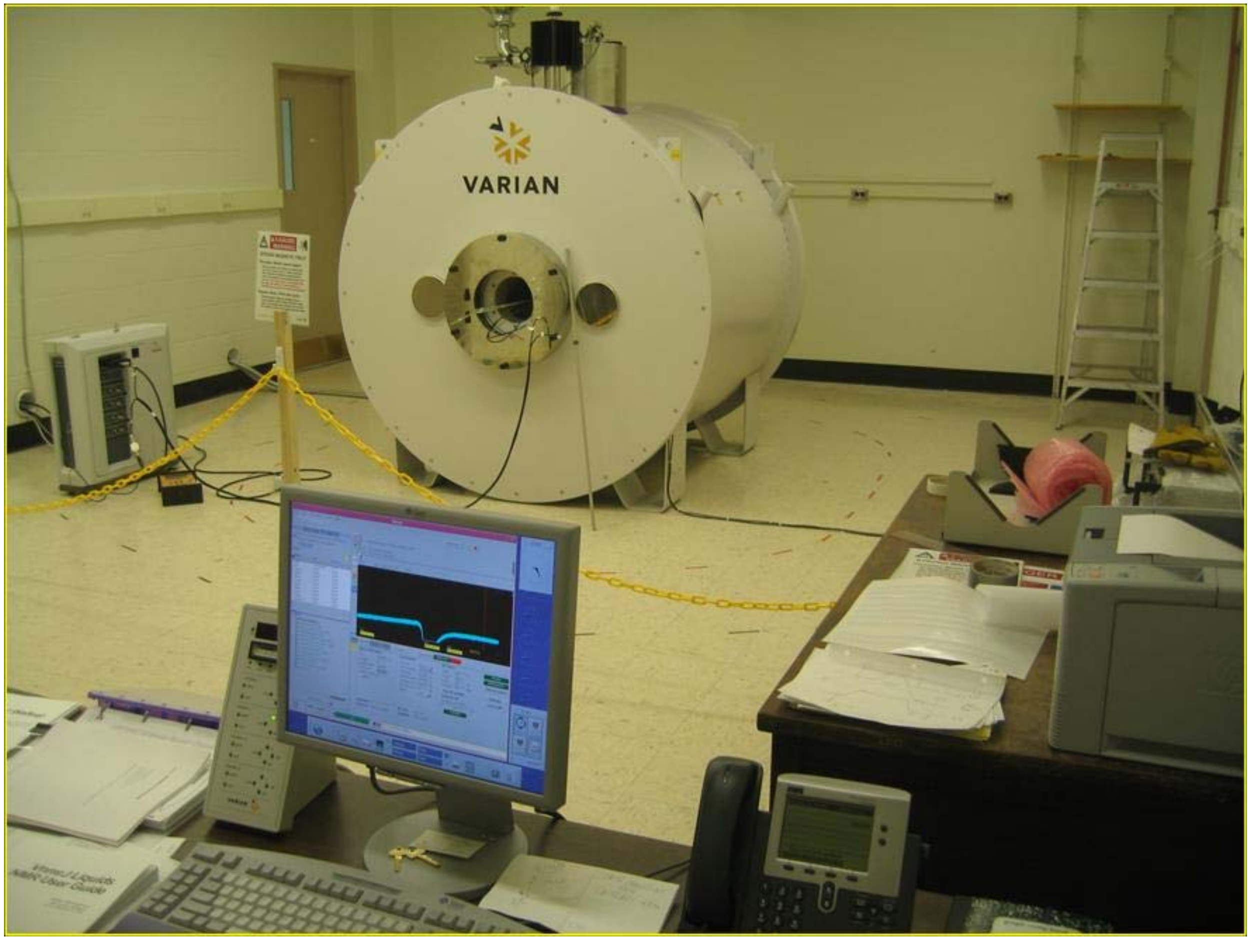
- MRI is based on spatial variations in the phase and frequency of the radio frequency energy being absorbed and emitted by the imaged object.
- Important microscopic property responsible for MRI is the spin property within hydrogen nuclei
- The human body is primarily fat and water. Fat and water have many hydrogen atoms which make the human body approximately 63% hydrogen atoms

Block diagram of MRI Equipment



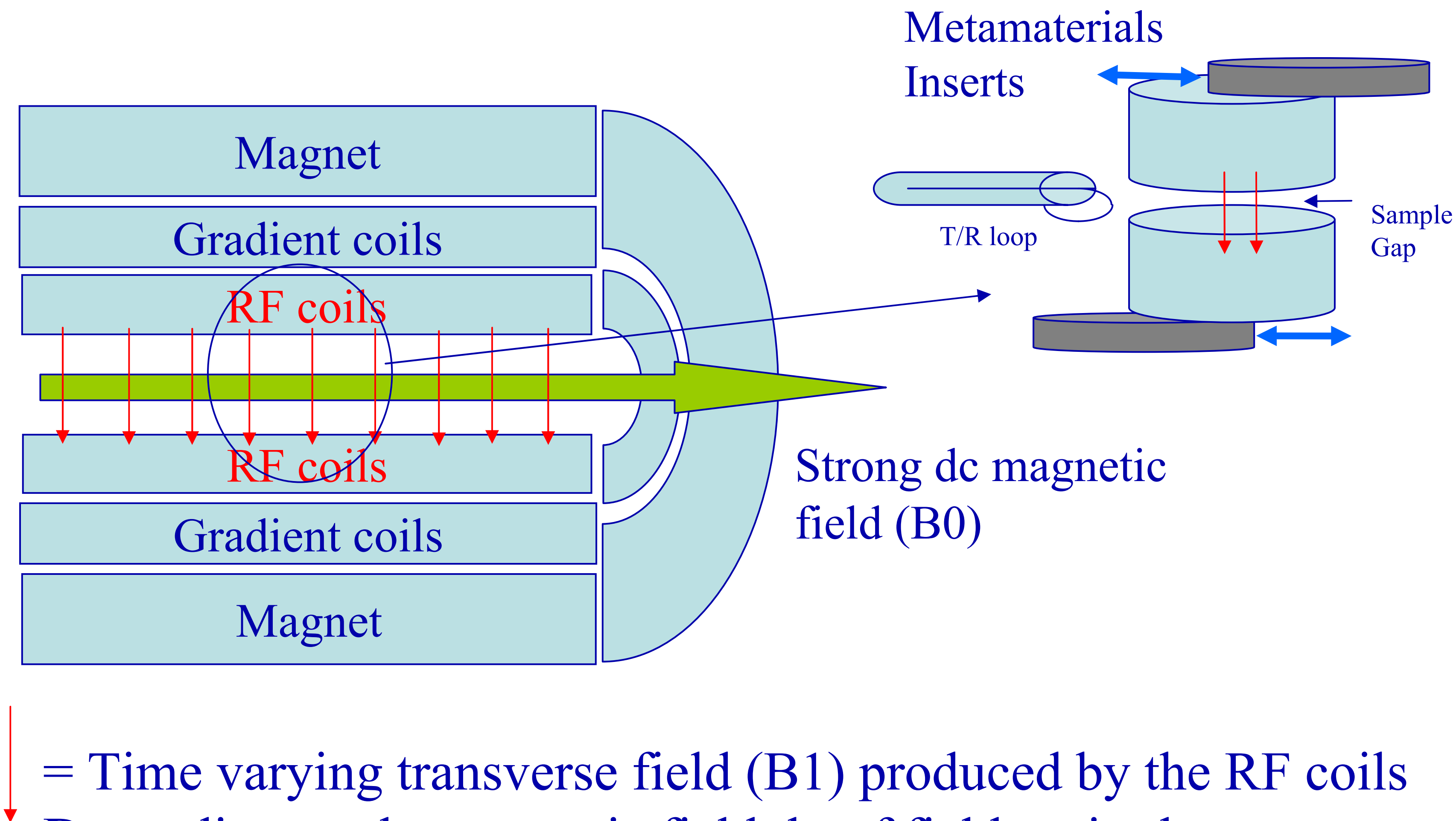
(Adapted from Reference: http://www.cis.rit.edu/htbooks/mri)

7 Tesla MRI device at the NMR spectroscopy lab



Andrew Webb Penn State University

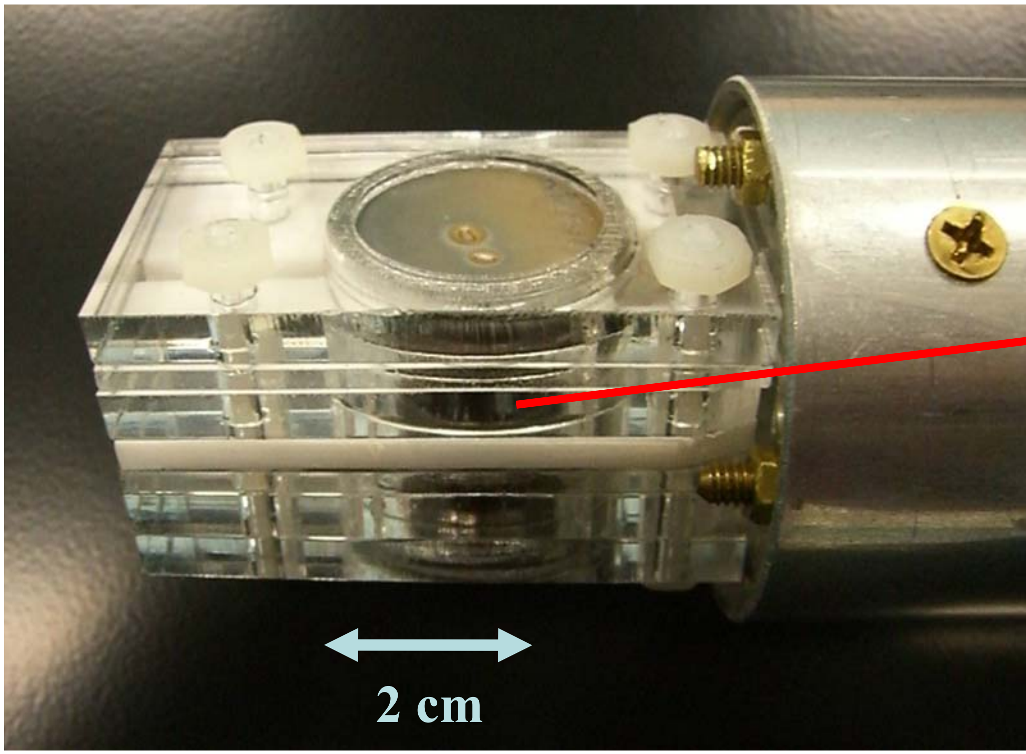
Schematic MRI System



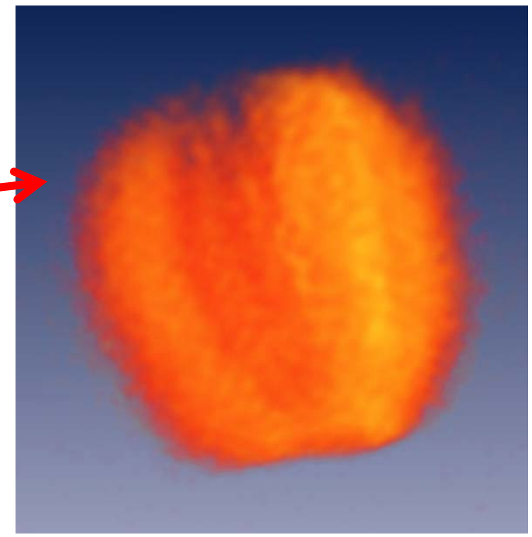
= Time varying transverse field (B1) produced by the RF coils
Depending on the magnetic field the rf field varies between 100 and 1,000

Imaging a Canola Seed

Ceramic Cylinder as an MRI insert



Canola Seed Image



2.5 mm

Elena Semouchkina, Varun Tyagi, Michael Lanagan, Amanda Baker, Andrew Webb, Thomas Neuberger

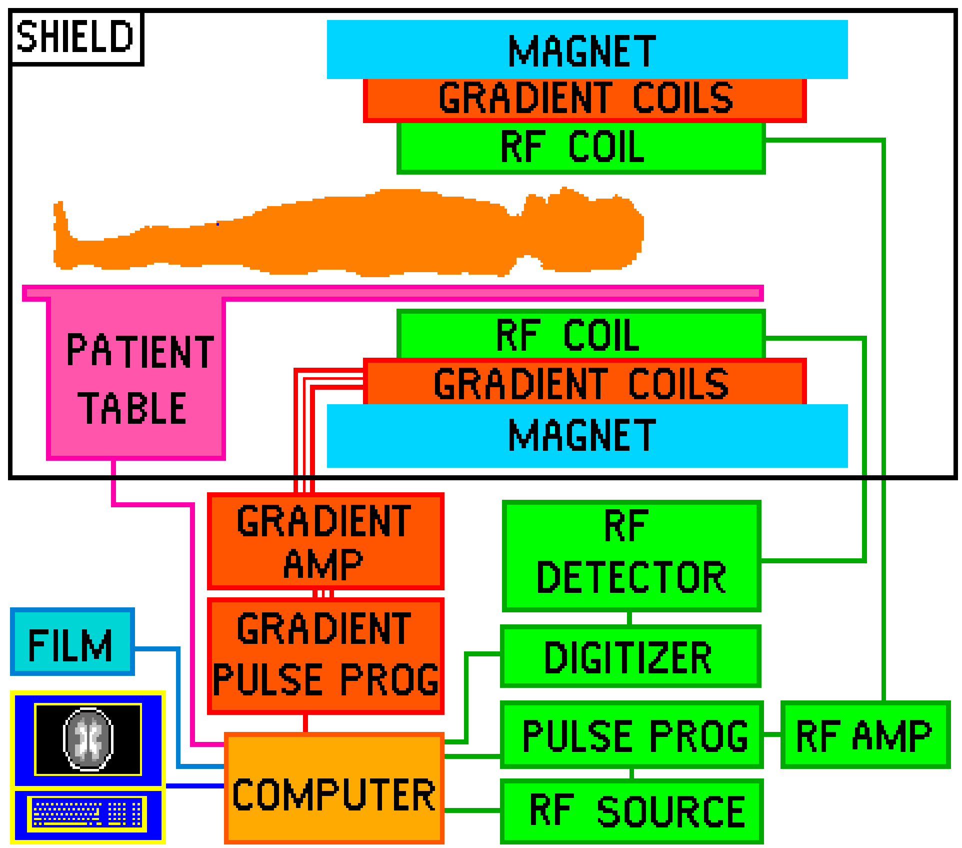
Case Study

- Design a resonator for a 3Tesla MRI
- Frequency = 300 MHz

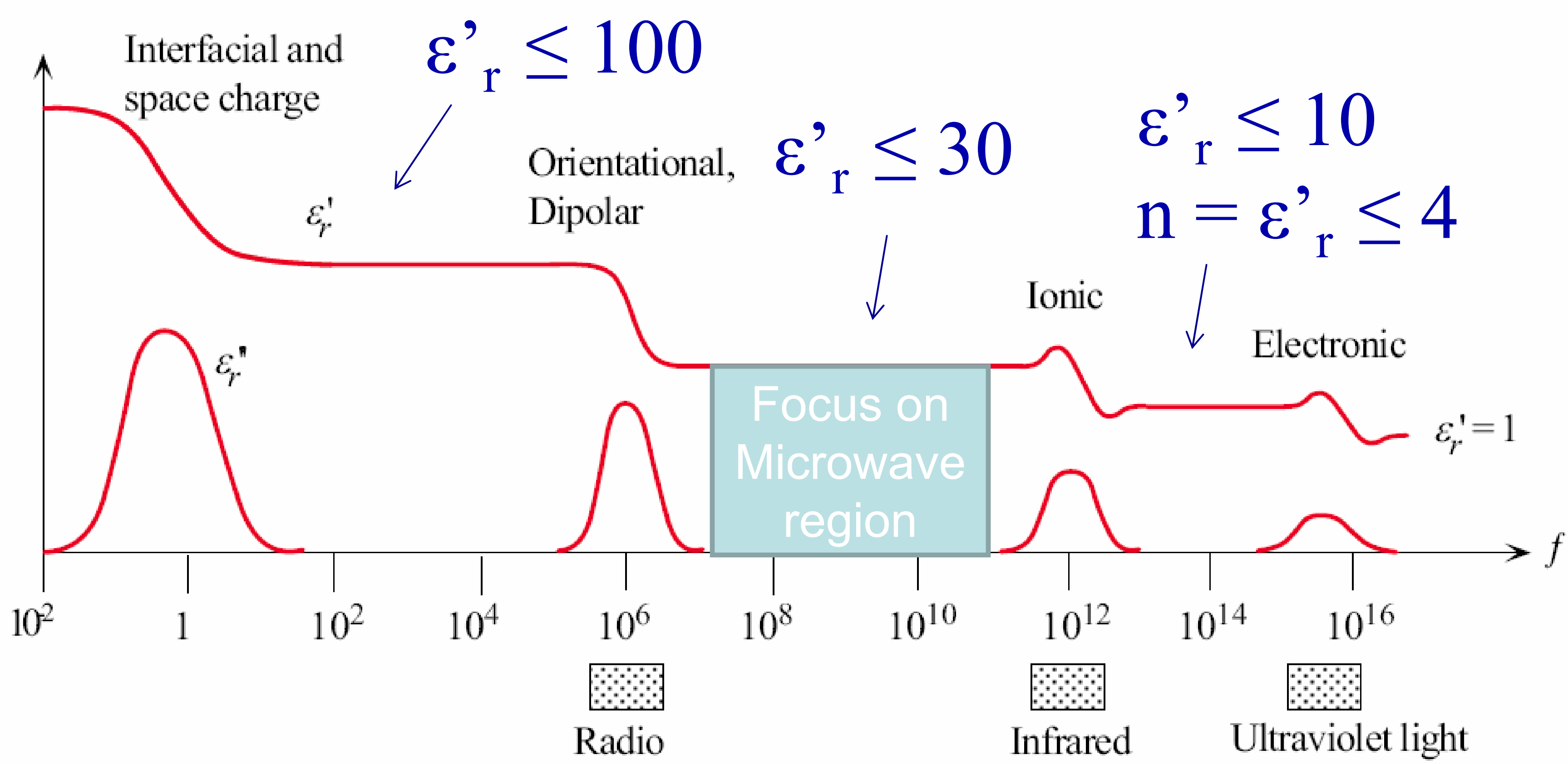
Case Study

- First think of the wavelength for the resonator replacing RF coil insert
- What will be size of the glass resonator
  - How do we shrink the size
  - Do you think that loss is important?

Block diagram of MRI Equipment



Frequency Response of Dielectric Polarization



The frequency dependence of the real and imaginary parts of the dielectric constant in the presence of interfacial, orientational, ionic, and, electronic polarization mechanisms.

Fig 7.15

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

How do we make a high permittivity glass?

$$\varepsilon_r = 1 + \frac{N\alpha_e}{\varepsilon_o}$$

ε_r = relative permittivity

N = number of molecules per unit volume

α_e = electronic polarizability

ε_o = permittivity of free space

Assumption: Only ionic and electronic polarization is present

Summary of glass as a dielectric

- Dielectric response for glass occurs over a wide frequency range
- New applications for dielectrics could involve glass
- Functionality of glass
  - Related to dielectric properties (permittivity and loss
  - Formability and cost

Dielectric Loss and Q Factor

Loss Tangent

$$\tan\delta = \frac{\varepsilon_r''}{\varepsilon_r'}$$

Q Factor

$$Q = \frac{1}{\tan\delta} = \frac{energy\_stored}{energy\_dissipated}$$

ε'_r = real part of the complex dielectric constant, ε''_r = imaginary part of the complex dielectric constant

From *Principles of Electronic Materials and Devices, Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)