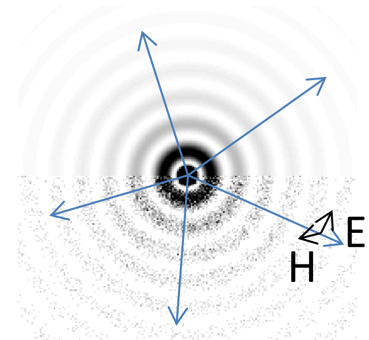
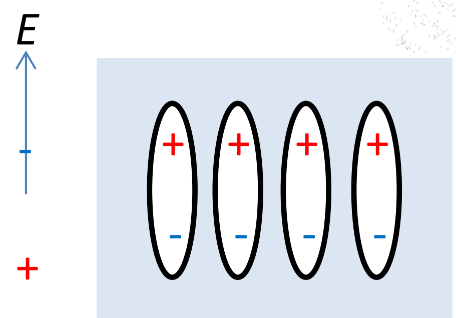
Optical materials

We need to know material properties
to estimate refraction, reflection, absorption.
<u>These are frequency-dependent.</u>
This week we will focus on propagation *in* material



- Macroscopic view
- Microscopic view
- Modelling material properties
- Advanced materials



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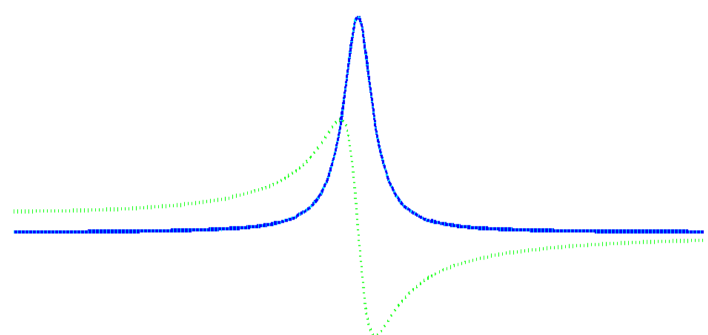


Waves in materials propagate with complex refractive index

- Plane wave solution gives $E \propto \exp(i[kx - \omega t])$
- Usually, we will consider behaviour at a given temporal frequency ω
- **Propagation** is then **determined by the spatial frequency or wavenumber *k***
- We can define the wavenumber as $k = 2\pi/\lambda = (n + i\kappa)2\pi/\lambda_0 = (n + i\kappa)k_0$
    - "0" means in a vacuum, *c* is the speed in a vacuum, λ is wavelength
- $n + i\kappa$ is the **complex refractive index**
- ***n* is the real part of the refractive index**
    - should be familiar (e.g. first year physics)
    - describes how much the wave slows down in a material
    - affects how quickly the wave oscillates *in a spatial sense*
- **$\boldsymbol{\kappa}$ is the imaginary part of the refractive index**
    - affects how quickly the wave decays *in a spatial sense*
- **Generally refractive index varies with frequency**
- We will typically observe *light* in terms of intensity $I \propto |E|^2$

Example optical materials and their EM properties

Material	*n (visible)*	T band μm	$\varepsilon/\varepsilon_0$ (low ω)	μ/μ_0-1
Vacuum	1	all	1	0
Air (STP)	1.0003		1.0006	
Water	1.34	0.3-0.8	80	-0.9×10^{-5}
"Glass" (silica)	>1.45	0.18 - 2	3.9	
MgF_2	1.37	0.13 - 8	5	
C (diamond)	2.38	wide	5.5	-2×10^{-5}
Si	3.4	1-10	11.7	

http://refractiveindex.info

Macroscopic view: effect of n

- **Propagation** in material is affected by **complex refractive index *n+iκ***

How does n affect propagation (ignoring κ)?

$k = nk_0$

then, (for a single wave)

$E \propto \exp(\,ink_0x)$

$I \ \propto |E|^2$ is constant

Reminder

Sign convention:

$$E = E_0 \exp[i\{kr - \omega t\}]$$

$$H = H_0 \exp[i\{kr - \omega t\}]$$

"Physics" $\varepsilon_c = \varepsilon' + i\varepsilon''$

Hence, to see how n affects propagation in a material,
we will generally need interference:
e.g. with equal intensity counter-propagating waves

$$E = \exp(ikx) + \exp(-ikx)$$

… $|E|^2 \propto 2\cos(4\pi nx/\lambda_0) + 1$

For general optical scenarios a more complicated calculation is needed (see next week)

Saleh & Teich Ch5

Macroscopic view: effect of κ

- **Propagation** in material is affected by **complex refractive index *n+iκ***

How does κ affect propagation?

$$k = (n + i\kappa)2\pi/\lambda_0$$

Assuming a single wave (i.e. no reflection)

$$E \propto \exp(\, ikx)$$

Then, converting to intensity

$$\boxed{I \propto |E|^2 \propto \exp(\, - 4\pi\kappa x/\lambda_0)}$$

κ>0: Intensity decays (Beer-Lambert "law")

Reminder

Sign convention:

$$E = E_0 \exp[i\{kr - \omega t\}]$$

$$H = H_0 \exp[i\{kr - \omega t\}]$$

"Physics" $\varepsilon_c = \varepsilon' + i\varepsilon''$

In general, some reflection will occur, so accurate predictions are more complicated (see next week)

Related definitions

- **Skin-depth (1/e)**
- **Absorption coefficient: $4\pi\kappa/\lambda_0$**

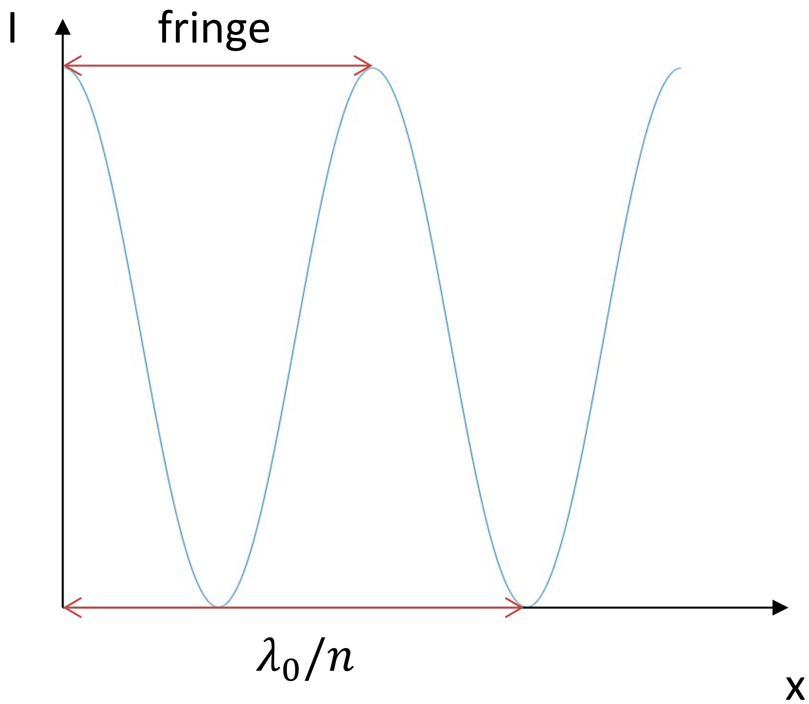
Saleh & Teich Ch5

Exercise

Interference measurements were taken within an optical material.
The fringes were spaced by 111nm @ 4.13eV, and 769nm @ 0.620eV.
What is the refractive index of this material?

$$\lambda_0 = \frac{hc}{U} = \frac{1.24\mu m}{eV}$$

$\Delta I \sim \cos(4\pi n x/\lambda_0)$ [Two waves interfering, assuming no absorption]

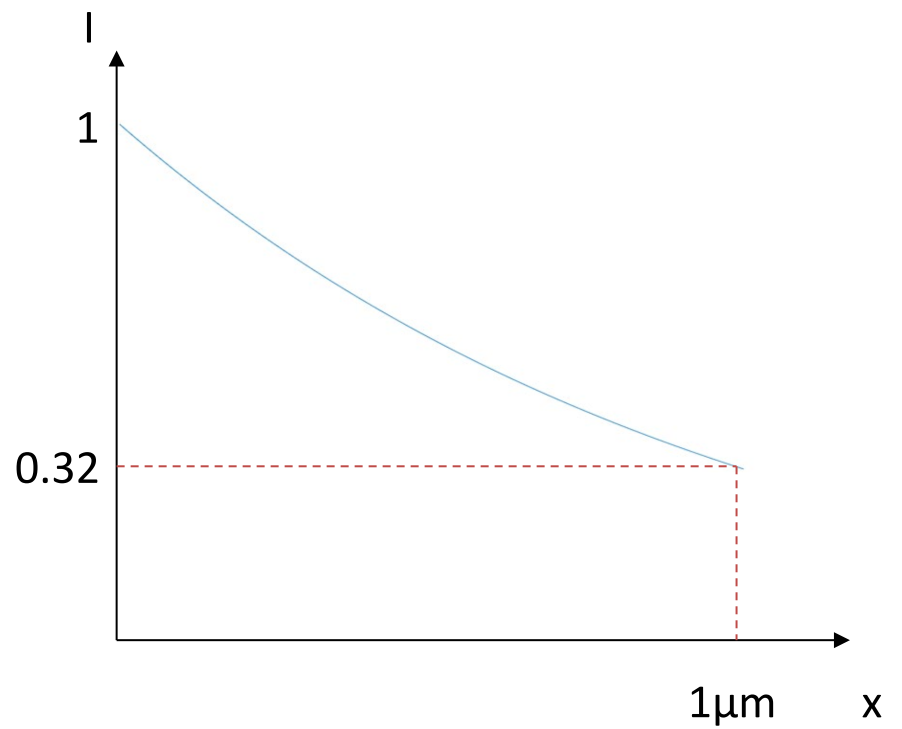


Exercise

1μm thickness of a material internally transmits 32% of light at 0.413eV.
What is the imaginary part of the refractive index at this frequency?

$$\lambda_0 = \frac{hc}{U} = \frac{1.24\mu m}{eV}$$

$I \sim \exp(-4\pi\kappa x/\lambda_0)$ [Single wave, i.e. ignoring reflection]



Relating EM material properties

n related to electric (ε) & magnetic (μ) properties

$$\boxed{n + i\kappa = \sqrt{\frac{\varepsilon\mu}{\varepsilon_0\mu_0}}}$$

Most optical materials $\mu = \mu_0$, i.e. <u>not</u> "magnetic", so **ε most important**:

$$\boxed{\varepsilon / \varepsilon_0 = \varepsilon' + i\varepsilon'' = (n + i\kappa)^2 = (n^2 - \kappa^2) + i(2n\kappa)}$$

Relative impedance $\eta_r = \frac{\eta}{\eta_0} = \sqrt{\mu_r/\epsilon_r} = \mu_r/(n + i\kappa)$

= $1/(n + i\kappa)$ *for non-magnetic*

Relates magnetic and E fields (more next week)

Exercise

The refractive index of a material is 1.26+0.11i at a particular frequency. Assuming a non-magnetic material, what is the relative complex permittivity?

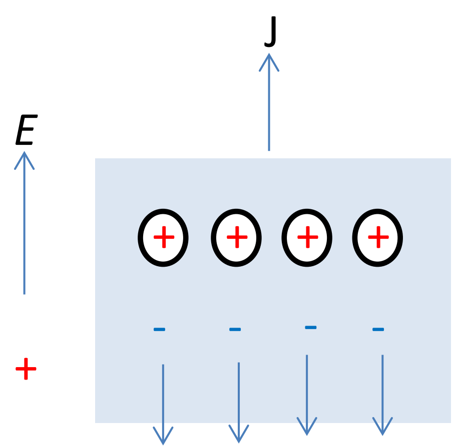
$$\boxed{\varepsilon / \varepsilon_0 = \varepsilon' + i\varepsilon'' = (n + i\kappa)^2 = (n^2 - \kappa^2) + i(2n\kappa)}$$

Where do optical properties come from?

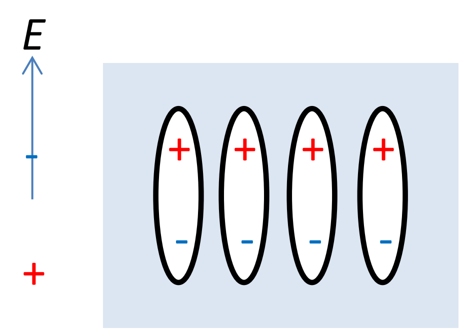
Basic concept: optical properties dominated by <u>electrons, either free or bound</u>.
We need to know how electrons can move to counteract incoming field.

- Why do we care?
  - Understanding of matter
  - Prediction from first-principles
  - Engineering of desirable properties

Mobile charges (e-)
Moving charges = induced J



Bound charges (+ cores & - e)
Charge-displacement = **dipole**



http://en.wikipedia.org/wiki/Permittivity

Charges affect electrical properties: ε, σ

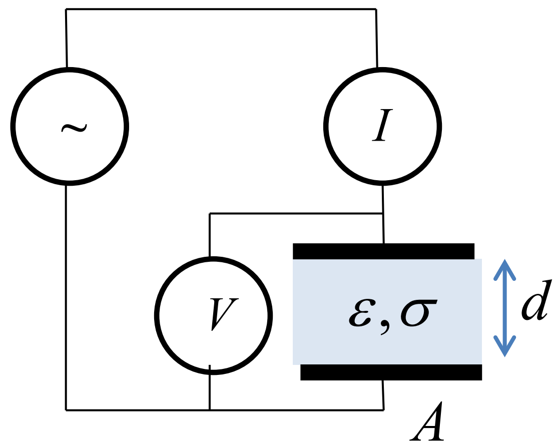
Sample between parallel plate electrodes
AC voltage & current measurements

Resistance

$$R \equiv \frac{V}{I} = \rho \frac{d}{A}$$

Conductance

$$G \equiv \frac{I}{V} = \sigma \frac{A}{d}$$



Capacitance

$$C = \frac{I}{\frac{dV}{dt}} = \varepsilon \frac{A}{d}$$

Maxwell: (free) charge flux

$$\boxed{J = \sigma E}$$

$$J = \frac{I}{A} \quad E = \frac{V}{d}$$

(bound) charge displaced

$$\boxed{D = \varepsilon E}$$

$$E = \frac{V}{d}$$

Permittivity *ε* & conductivity *σ* defined by constitutive relations

<u>At *low* frequency *ω*</u>, are related by $\boxed{\varepsilon_c = \varepsilon + i\sigma / \omega}$

Real = phase
Imag = absorb

“Low” frequency general means Radio. Drude model better (includes radio and optical).

Exercise

A material has a DC conductivity of $6x10^7$ S/m.

What values would the DC conductivity model predict for the imaginary part of the permittivity at 1.24eV and 0.62eV respectively?

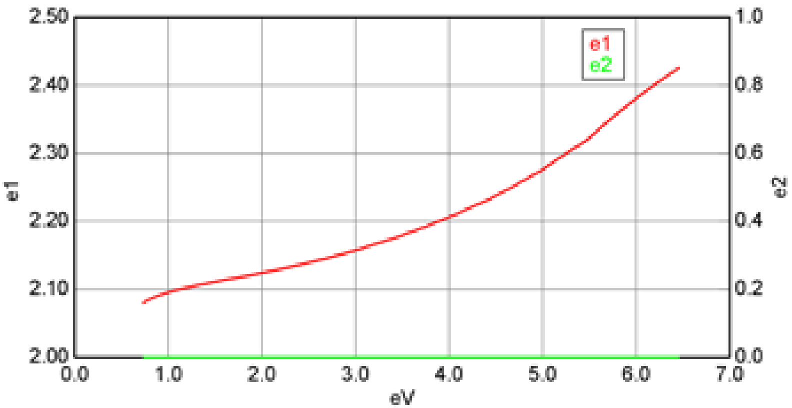
[Model is NOT accurate at these frequencies: check later with Drude]

$$\lambda_0 = \frac{hc}{U} = \frac{1.24\mu m}{eV} \qquad \omega = 2\pi c/\lambda_0$$

$$\varepsilon_c = \varepsilon + i\sigma/\omega$$

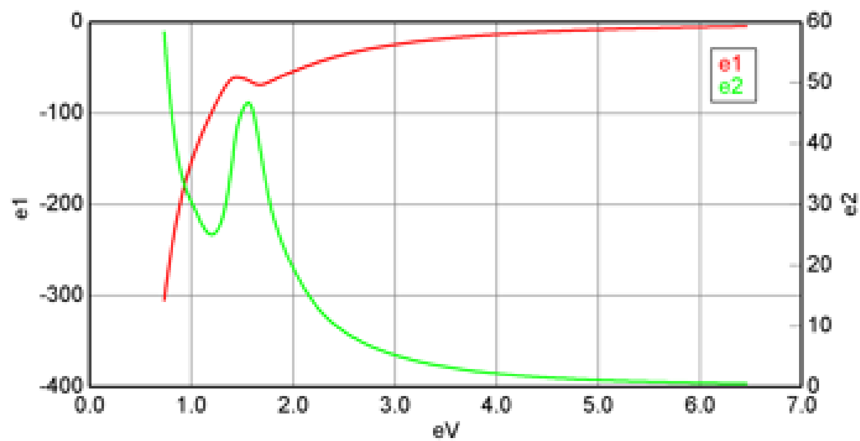
EM properties are frequency dependent

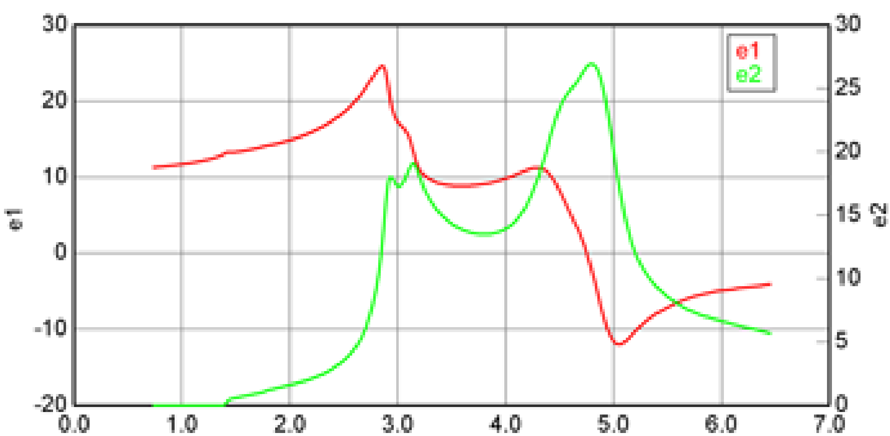
e1(real) = phase
e2(imag) = absorb



Dielectric that is transparent over the entire spectral region. Notice e1 is positive, but e2=0 indicates a transparent material

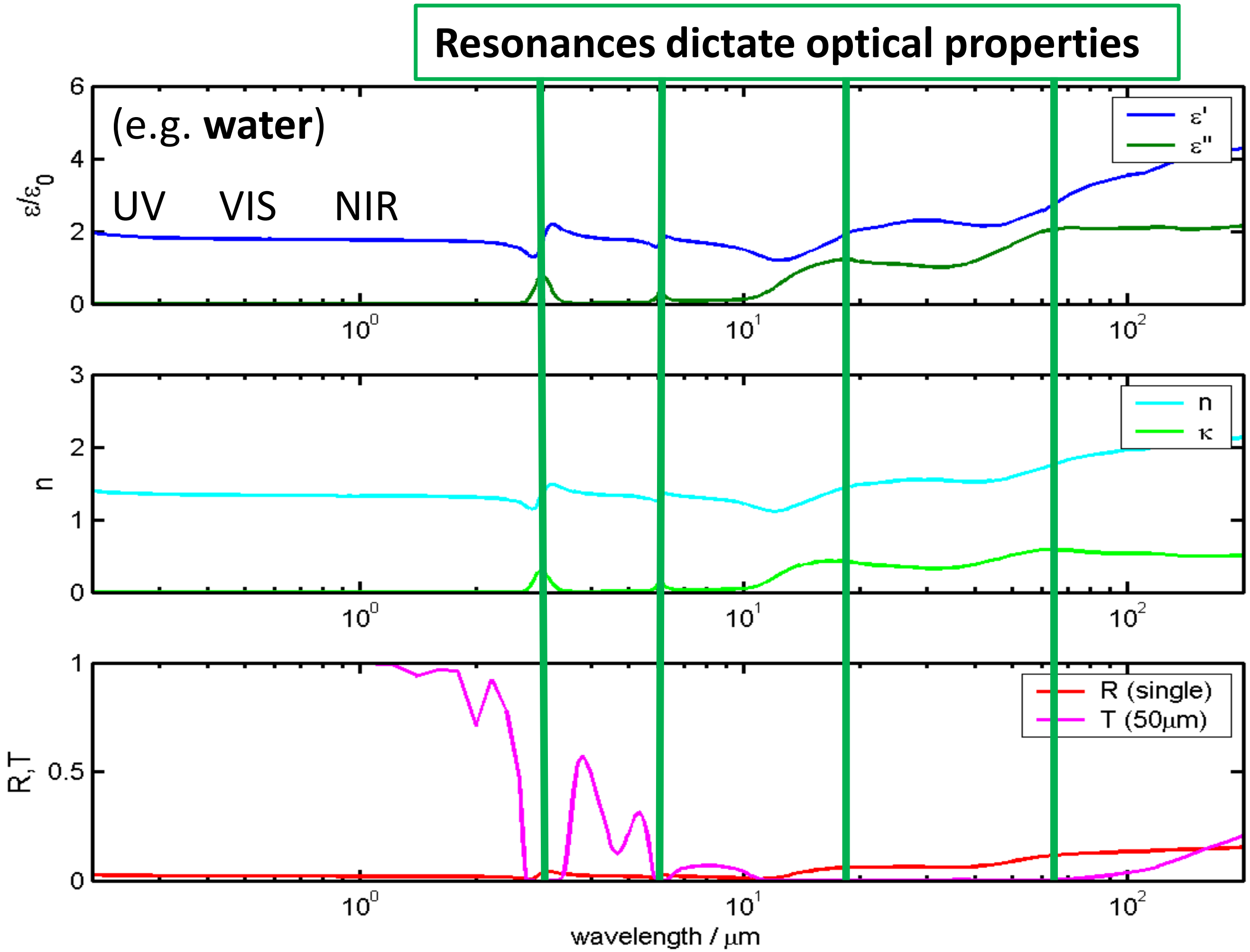
Metal material that has absorption due to free carriers over the entire spectral region, causing e2 to be nonzero. e1 is negative over significant part of spectrum.





Semiconductor material that has a bandgap near 1.42 eV. Note e2 is zero below the bandgap, with absorption (e2>0) above the bandgap.

Why frequency dependent?



μ-wave

Low ω

Vibrational
Dipolar

Electronic resonances

Physical models for optical properties

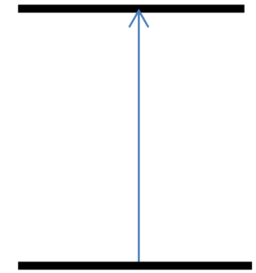
Model optical response of materials:

- **Based on physics of charge carrier environment**
- **Predict frequency dependent permittivity $\epsilon(\omega)$**
- **Imaginary part corresponds to absorption**

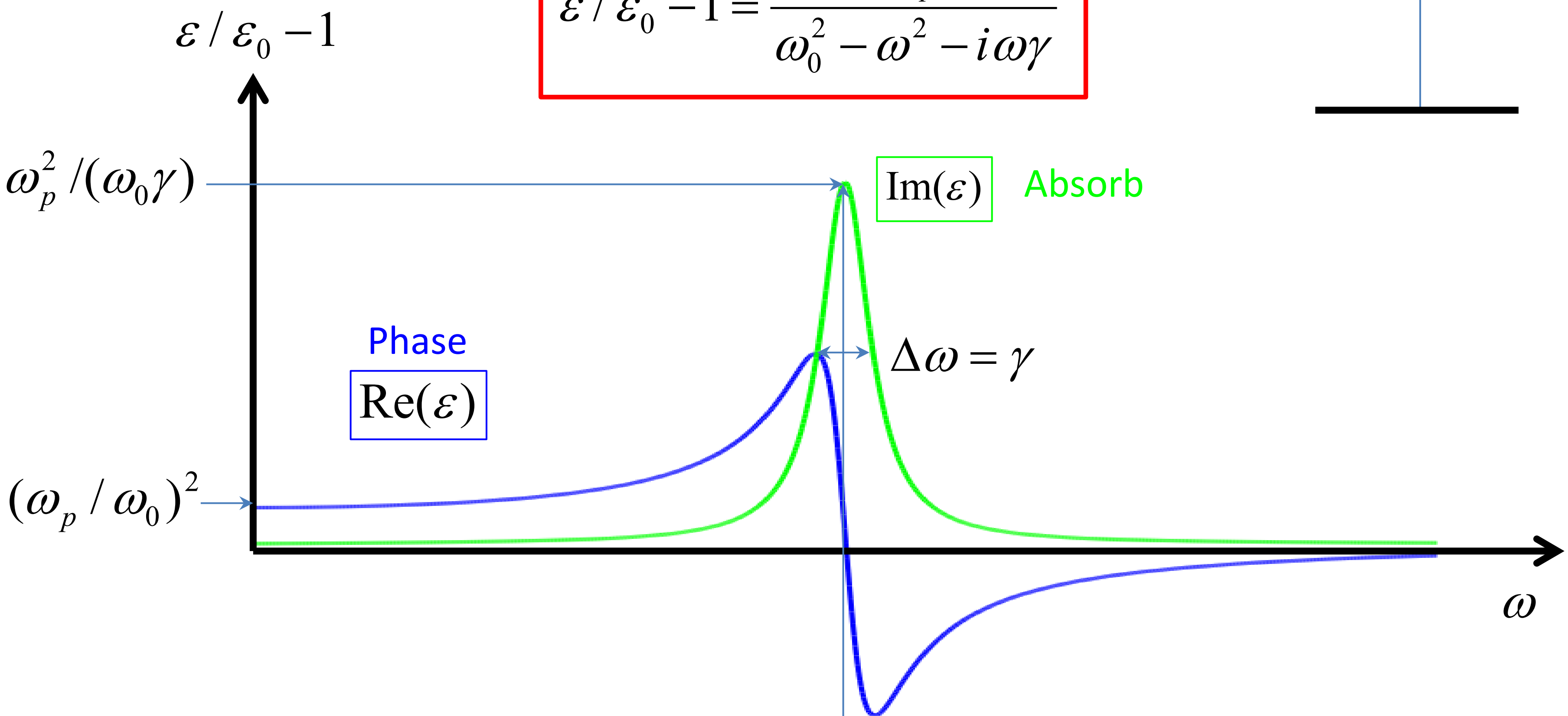
- Lorentz model – bound charges & defects
- Drude model – nearly free charges
- Tauc-Lorentz model – band-gaps
- Debye model – polar fluids

Lorentz oscillator model

e.g. bound electrons transiting between defects



$$\boxed{\varepsilon/\varepsilon_0 - 1 = \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega\gamma}}$$



$$\omega_{peak} \approx \sqrt{\omega_0^2 - (\gamma/2)^2}$$

NOTE: absorption → wobble in real part.
Hence *n* for glasses higher in UV & lower in IR

Electron oscillator model

Lorentz-Drude model

Charge carrier experiences elastic and inelastic forces, with associated frequencies

Equation of motion:

$$\overset{\text{net force}}{m\frac{d^2x}{dt^2}} = \overset{\text{damping}}{-\gamma\frac{dx}{dt}} \overset{\text{elastic}}{-\,\omega_0^2 x} \overset{\text{driving}}{-\,eE}$$

Assuming *x* & *E* sinusoidal ($e^{-i\omega t}$): $(-\omega^2 - i\omega\gamma + \omega_0^2)x = -(e/m)E$

Polarization field: $P = Np = -Nex = \underbrace{\boxed{\dfrac{Ne^2/m}{\omega_0^2 - \omega^2 - i\omega\gamma}}}_{(\varepsilon-\varepsilon_0)} E$

$$P = D - E = (\varepsilon - \varepsilon_0)E$$

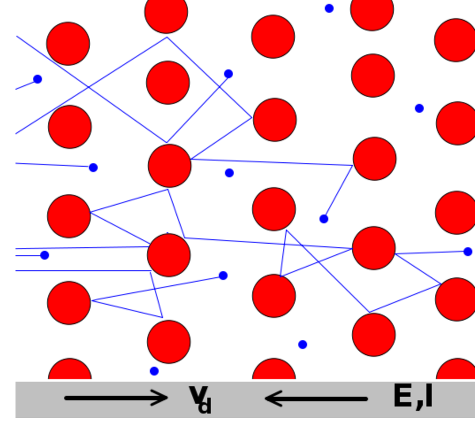
NOTE: complex signs

$e^{-i\omega t} \rightarrow \mathrm{Re}(\varepsilon) + i\,\mathrm{Im}(\varepsilon)$

Really there are additional factors to account for oscillator cross-section

Can add up multiple oscillators

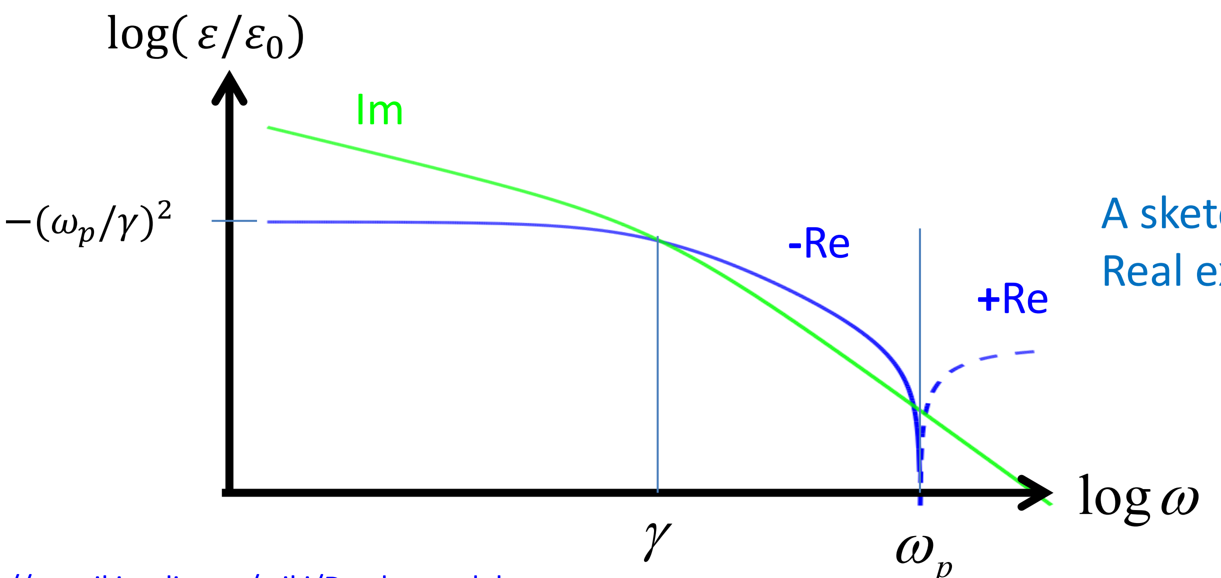
Drude model



Nearly free carriers damped by interactions: metals, plasmas

$$\varepsilon/\varepsilon_0 - 1 = \frac{\omega_p^2}{\cancel{\omega_0^2} - \omega^2 - i\omega\gamma}$$

- Metal = free carriers, zero elastic force
- Special case of Lorentz model, ω_0=0
- Characterized by **plasma frequency** $\omega_p^2 = Ne^2/(m\varepsilon_0)$
  - **Enables experimental estimate of charge concentration *N***
- Damping rate γ experimentally related to DC resistivity $\gamma = \varepsilon_0 \omega_p^2 \rho_{DC}$
  - Arises from charge-phonon coupling



A sketch to summarize special points
Real examples follow

http://en.wikipedia.org/wiki/Drude_model

Exercise: Silver

Estimate ω_p

$$N = n_v \rho_m N_A / A_r$$

Mass density of Ag: ρ_m=10.5 g cm^{-3}

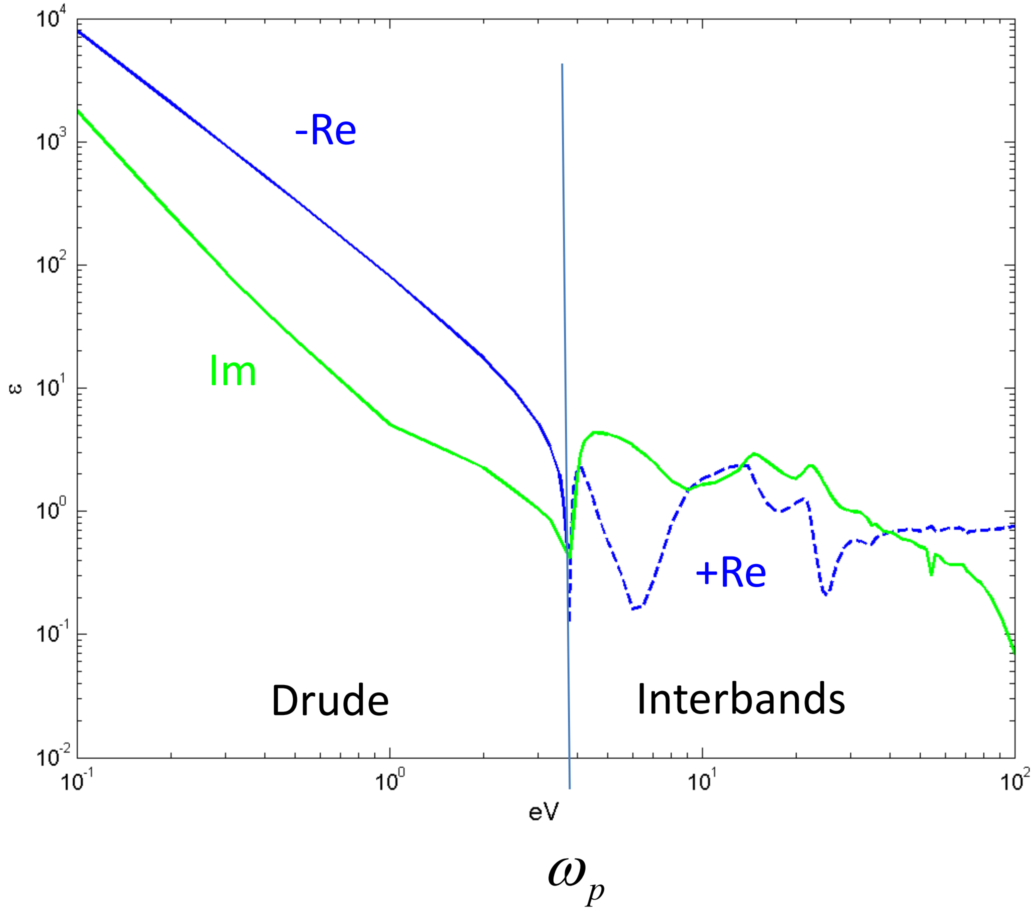
Atomic mass: A_r=108 g mol^{-1}

Valence: n_v=1

N =

$$\omega_p = q_e \sqrt{N/(m_e \varepsilon_0)}$$

Plasma frequency =



- ➢Observed frequency is 4eV due to interbands (however Drude should be adequate for low ω)

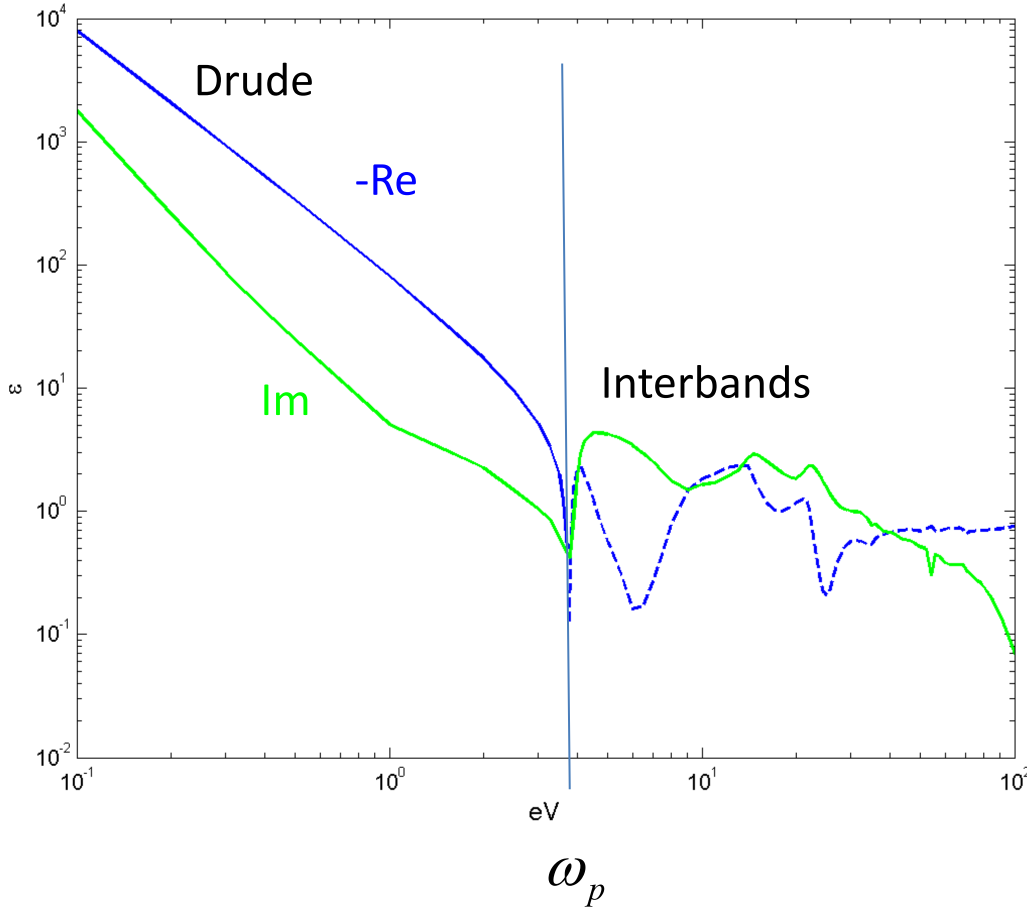
http://hyperphysics.phy-astr.gsu.edu/hbase/tables/fermi.html

Exercise: Silver

Estimate γ

$$\gamma = \varepsilon_0 \omega_p^2 \rho_{DC}$$

σ=1/ρ=6x10^{7} S/m
ε_0~ 9x10^{-12} F/m



http://hyperphysics.phy-astr.gsu.edu/hbase/tables/fermi.html

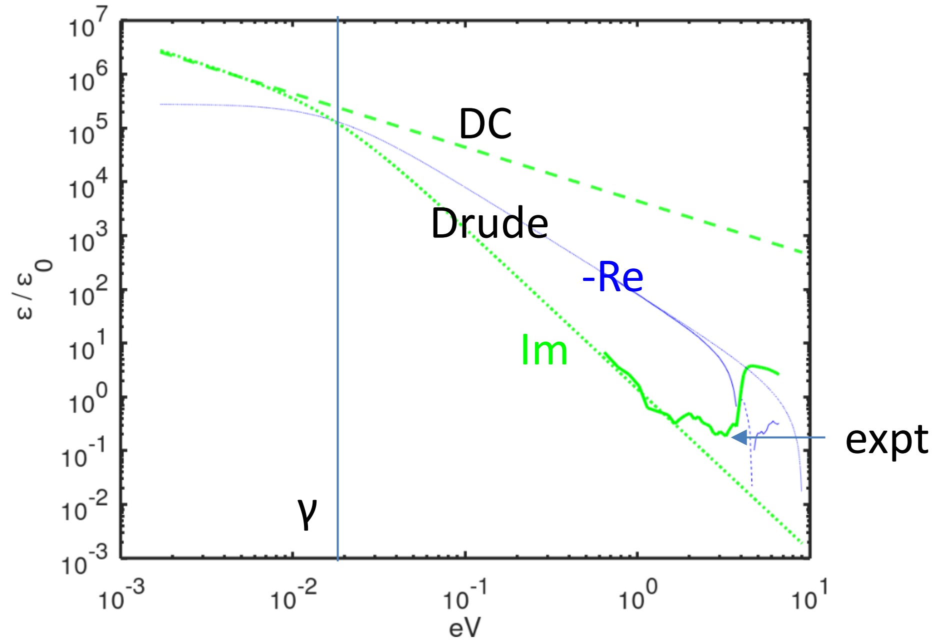
Exercise

A material has a DC conductivity of 6×10^7 S/m.

What values would the *Drude* model predict for the imaginary part of the permittivity at 1.24eV and 0.62eV respectively?

How inaccurate was the previous DC model estimate?

$$\frac{\varepsilon}{\varepsilon_0} - 1 \quad = \quad -\frac{\omega_p^2}{\omega(\omega + i\gamma)}$$



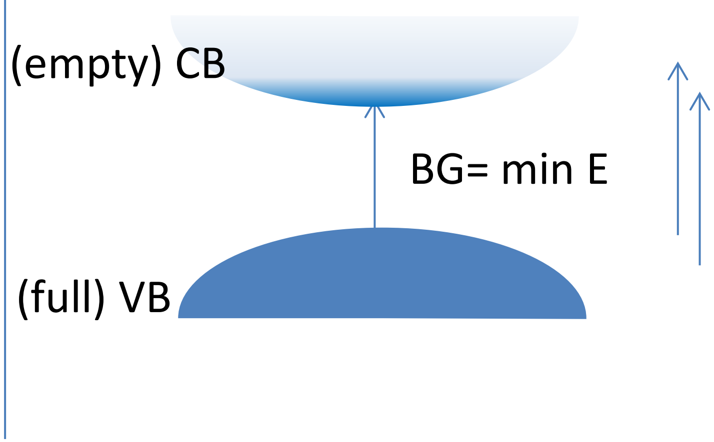
Quantum model

Optical absorption corresponds to electron energy transitions between bands

Absorption (@ E diff) = sum of overlap product of all available transitions

$E = \hbar\omega$

Interband transition (e.g. Semiconductor)

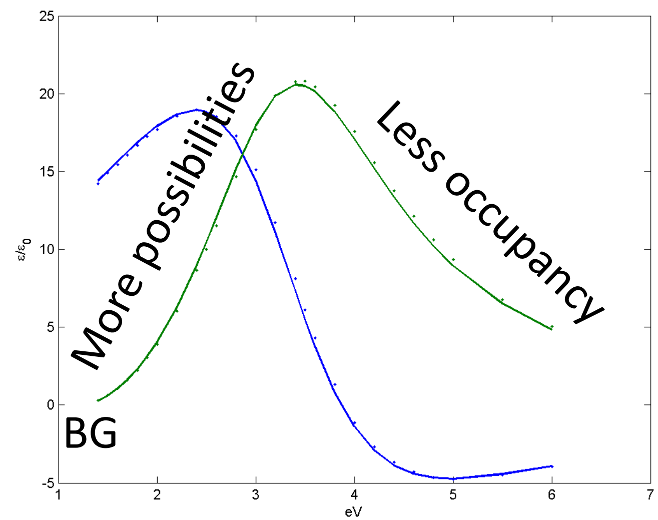


Larger E also allowed:
- More possible transitions, absorption ↑
- Less thermal occupancy away from BG, absorption ↓

e.g. Tauc-Lorentz model (strictly for *amorphous* IB)

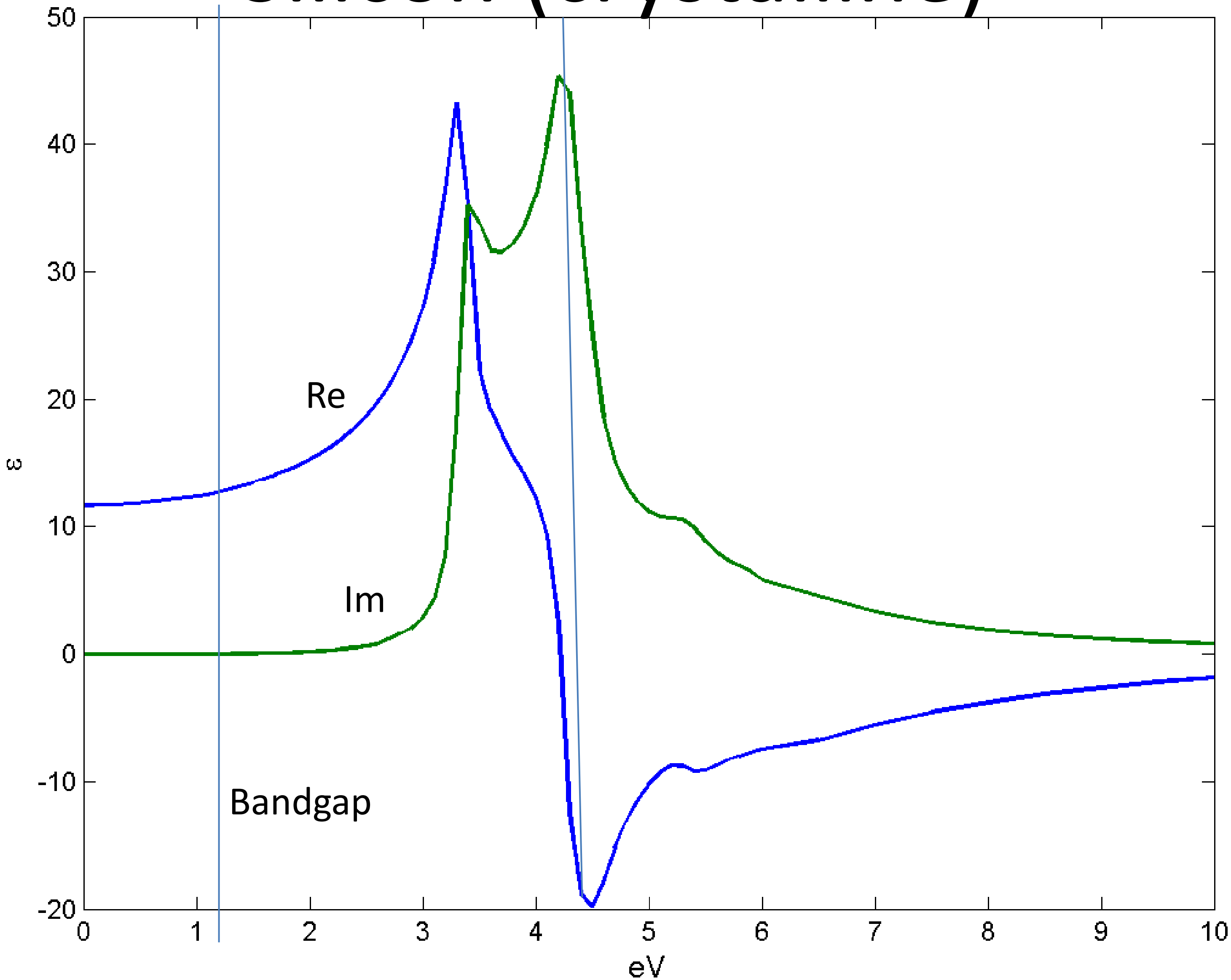
$$\mathrm{Im}[\varepsilon(\omega > \omega_g)] = \frac{A\omega_0 C(\omega - \omega_g)^2}{[(\omega^2 - \omega_0^2)^2 + (C\omega)^2]\omega}$$

e.g. a-Si: A=123, ω_g=1.2eV, ω_0=3.4eV, C=2.5eV



Saleh & Teich Ch15

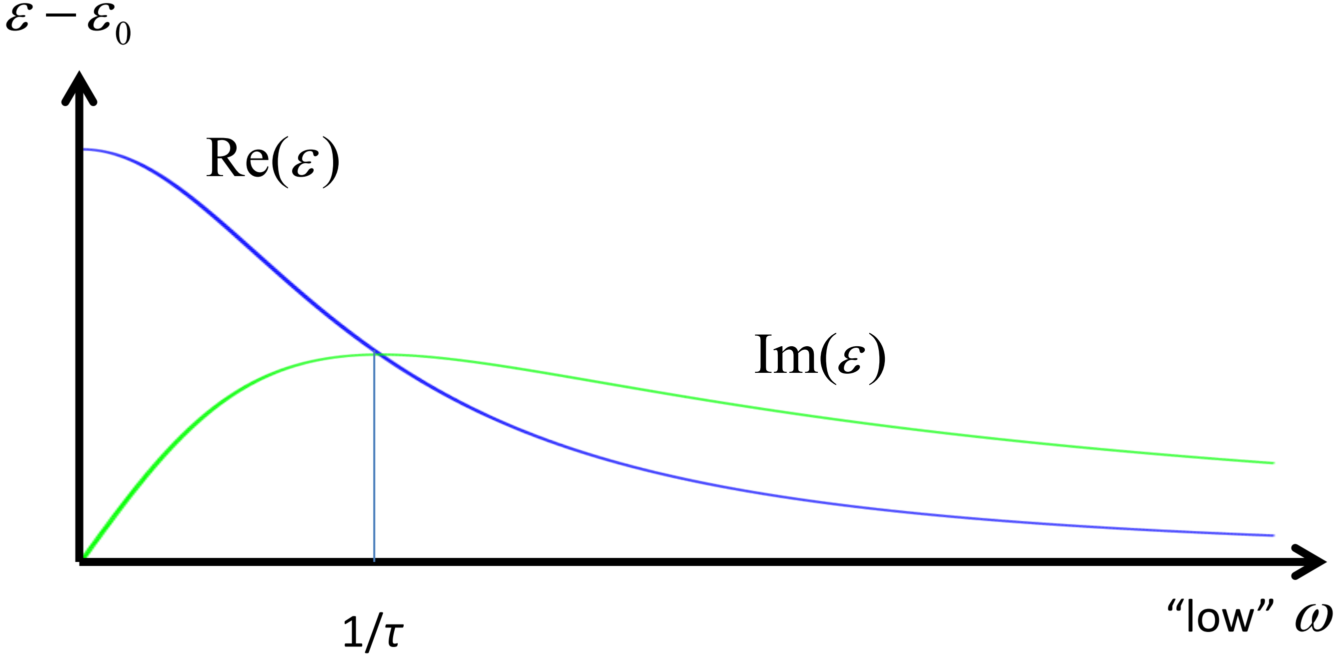
Silicon (crystalline)



Debye model

e.g. polar liquids

$$\varepsilon - \varepsilon_0 \propto \frac{1}{1 - i\omega\tau}$$



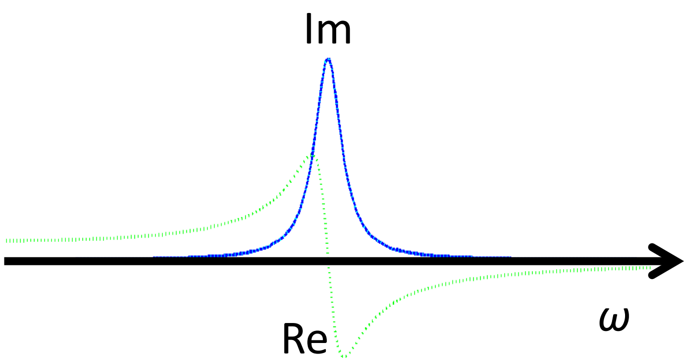
Why does absorption cause a wobble?

Answer: effect (material response) follows cause (light)

➢Not always practical to measure both Re & Im parts
Kramers-Kronig (KK) analysis uses causality to relate them

$$\mathrm{Re}[\varepsilon(\omega)] = 1 + \frac{2}{\pi}\int_0^\infty \frac{\Omega\,\mathrm{Im}[\varepsilon(\Omega)]}{\Omega^2 - \omega^2}\,d\Omega$$

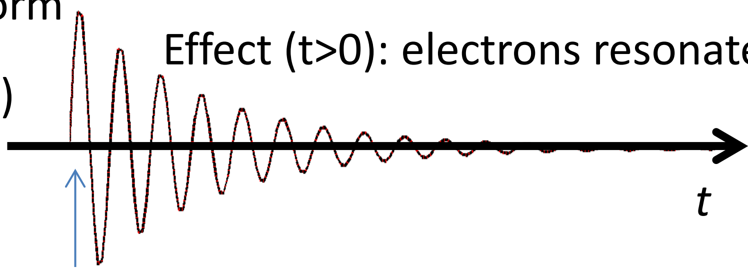
e.g. Im part (absorption expt / quantum calc) → Re part



➢KK is nasty. More speed & insight from Fourier transform

$$2\,\mathrm{Im}\left[\int_0^\infty \mathrm{Im}[\varepsilon(\omega)]e^{i\omega t}\,d\omega\right] \rightarrow \varepsilon(t)$$ Re, causal (***t*>0**)

$$\int_0^\infty \varepsilon(t)e^{-i\omega t}\,dt \rightarrow \varepsilon(\omega)$$ Re + i Im



➢Although in widespread use,
KK-type analysis is inaccurate (requires *all* freq),
fits to models are easier.
Direct measurement of both parts is best practice.

http://en.wikipedia.org/wiki/Kramers-Kronig_relations

How to measure?

- Refraction
- Interference (Optics) [must know *d*]
- Reflection (Next week) [“Easy”, incomplete]
- Ellipsometry (In two weeks) [Complete, indirect]

Advanced material properties

- Non-linearity $D = \varepsilon E + \chi_{(2)} E^2 + \ldots$
  - e.g. asymmetric potentials
  - Strong fields allow frequency conversion
  - Exploited in optical amplifiers (e.g. lasers – green DPSS)
- Anisotropy

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

  - Properties **vary with direction**
  - **Polarizing** activity
  - Technologically important, e.g. LCD, (more in Polarization lecture)
- Permanent dipoles
  - Some crystals have dipole frozen in (e.g. electret microphone)
  - Associated with other special properties, e.g. piezoelectric

Metamaterials

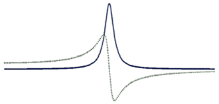
- *Problem*: finite selection of materials limit choice
- *Solution*: engineer materials with artificial atoms

(e.g. Instead of 1Å period, choose size closer to, but still smaller than λ)

Dielectrics
~ averaging
"Effective medium"

Metals
~ resonances

Resonant → -ve phase

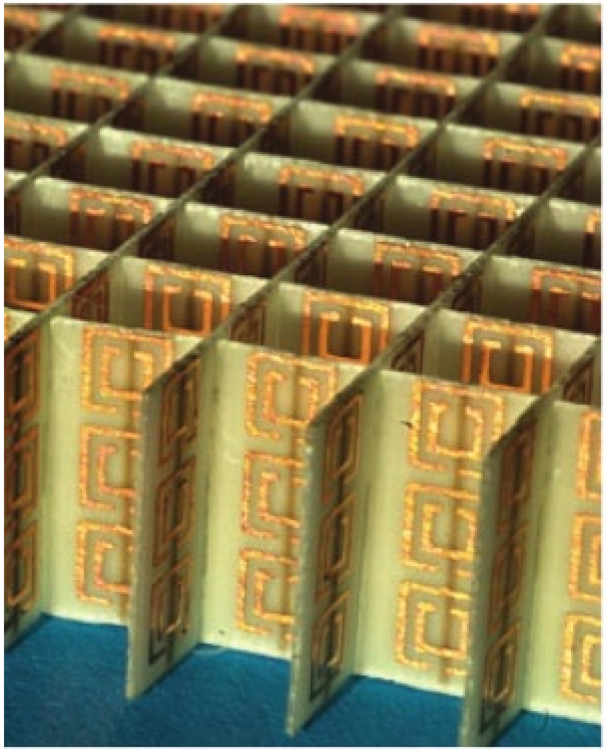


Dipoles
Resonant ε
"Plasmonics"

Current loops
Resonant μ

Negative refraction

Original PCB (for microwaves)

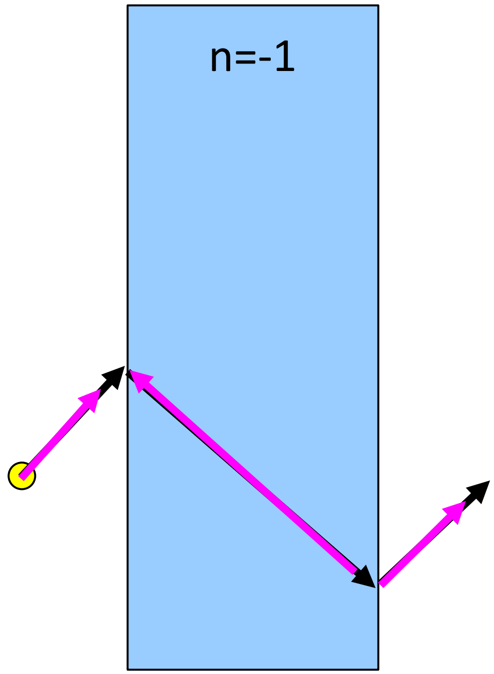


Schelby et al (2001), Science 292 77

http://en.wikipedia.org/wiki/Metamaterial

Negative refraction example

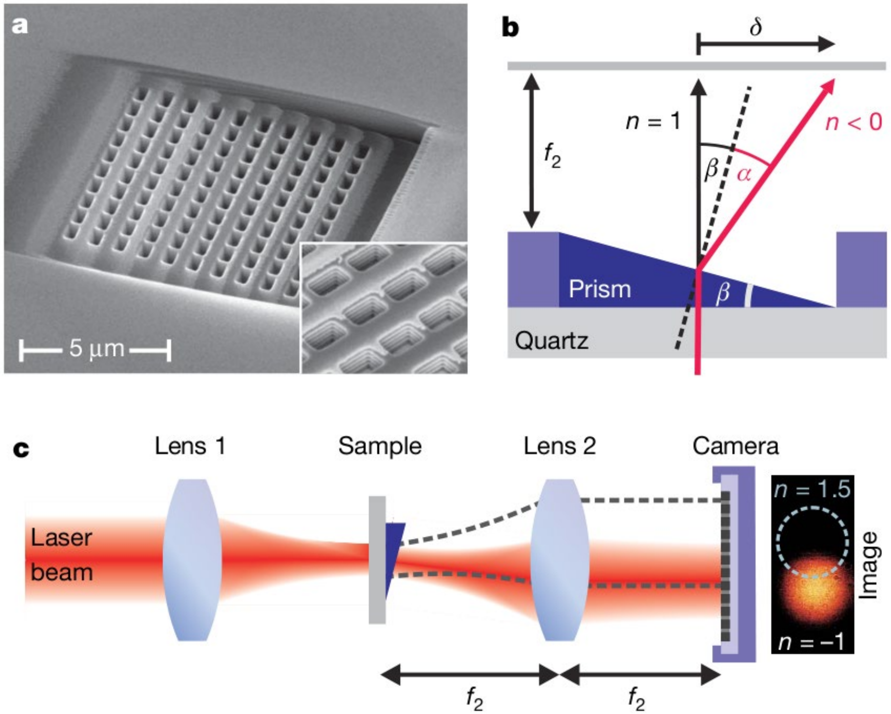
Ray is bent back on opposite side of normal



Group and phase velocities are opposite

Optical frequency example:

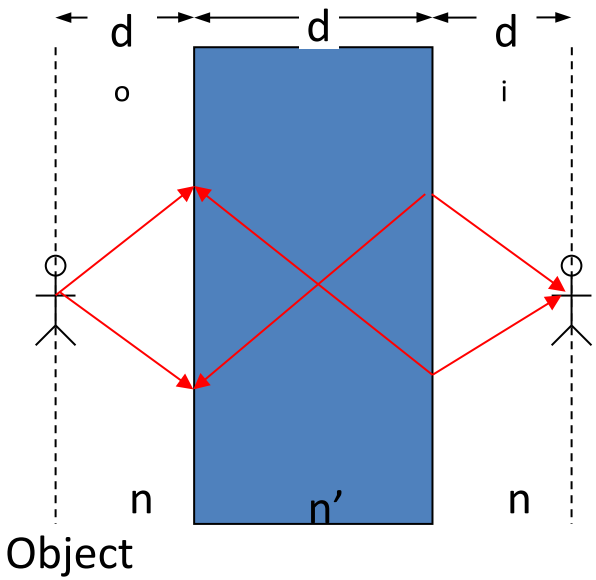
Multilayer fishnet prism



Valentine et al (2008), Nature

https://en.wikipedia.org/wiki/Negative_refraction

Imaging with negative refraction



Rays bent back to opposite angle

- **Flat slab = lens**
- **Beats the diffraction limit**

Imagine the object is a grating

Usually only *diffracted* orders captured

High-frequency evanescent waves decay

Negative refraction “undoes” this decay!

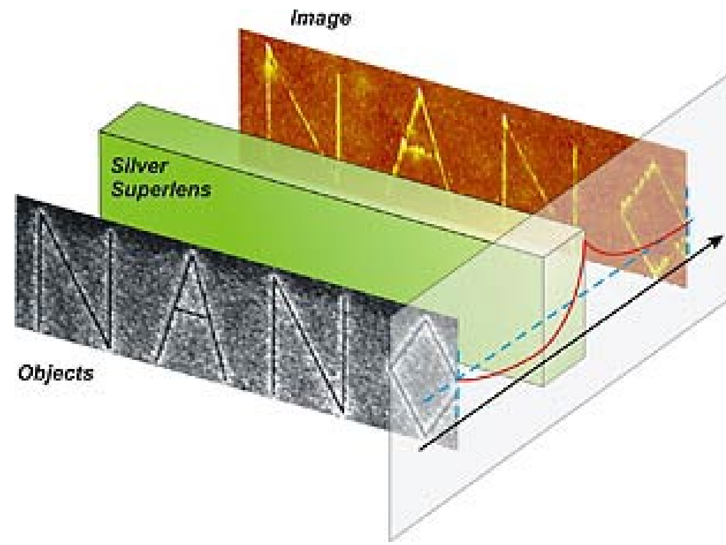
Single-layer metal lens

Not strictly negative refraction

Evanescent waves are “amplified”

Actually complicated

Internal reflection important



[Science](https://www.science.org) 308(5721):534-7. Fang et al (2005)

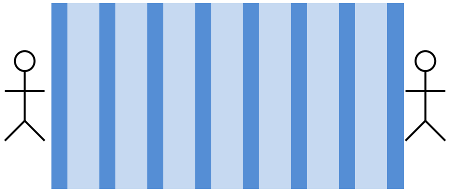
https://en.wikipedia.org/wiki/Superlens

Hyperbolic lenses

- Image reconstruction limited by lens thickness

Thinner lens less blurry but short working-distance

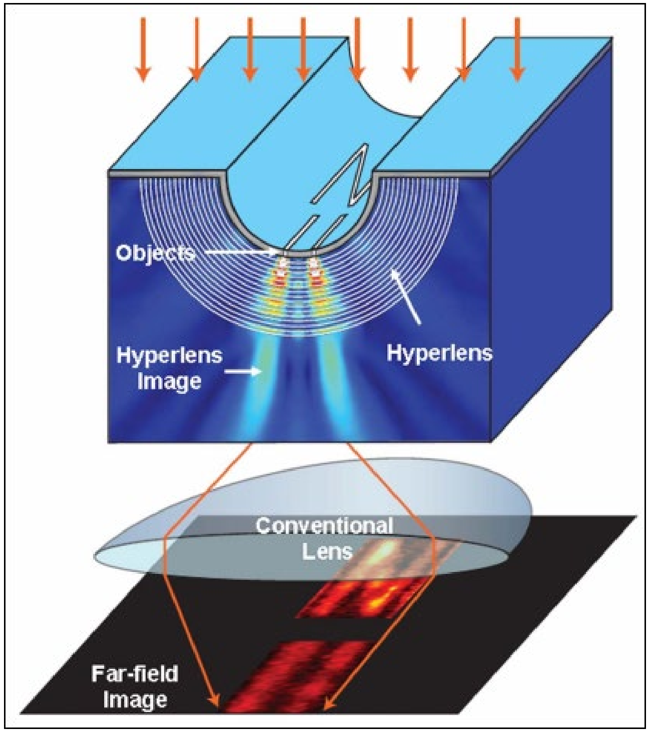
- **Solution: stack up thin lenses**
- Hard to make well (surface roughness)
- Object/observer must be at surface



Lens highly anisotropic

Very different effective ε for E fields ↔ ↕

- Observer must be close to flat lens
- **Solution: curved hyperbolic lens**
- Object must still be close to lens



Science 315, 1686. Liu et al (2007)

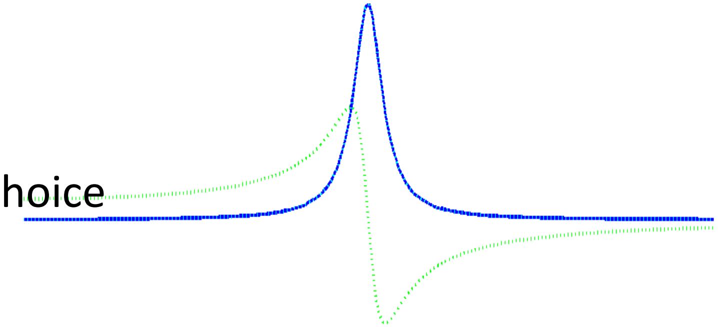
Summary

- Optical properties offer insight into materials
- Refractive index *n*+iκ affects wave propagation: **real=phase, imag decay**
- Single wave (ignoring reflection): $I \sim exp(-4\pi\kappa x/\lambda_0)$
- Interfering waves (ignoring absorption): $\Delta I \sim cos(4\pi n x/\lambda_0)$
- Proper calculation for film or stack more complicated (see next week).

$$\boxed{n + i\kappa = \sqrt{\frac{\varepsilon\mu}{\varepsilon_0\mu_0}}}$$

- **Optical materials dominated by electrical properties** (mostly non-magnetic)
- Electrical properties affected by **free & bound electrons**
- Electron models (**Lorentz=bound, Drude=free**) with resonance ω_0 & damping γ

$$\varepsilon - \varepsilon_0 = \frac{Ne^2/m}{\omega_0^2 - \omega^2 - i\omega\gamma}$$



- **Metamaterials use artificial** atoms to offer more choice