Lecture Series: Part 2
Spectroscopic Ellipsometry and Optical Constants of Crystalline Solids

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Thanks for support from AFOSR SFFP!
Thanks to AFRL/RYDH for hosting my student and me.

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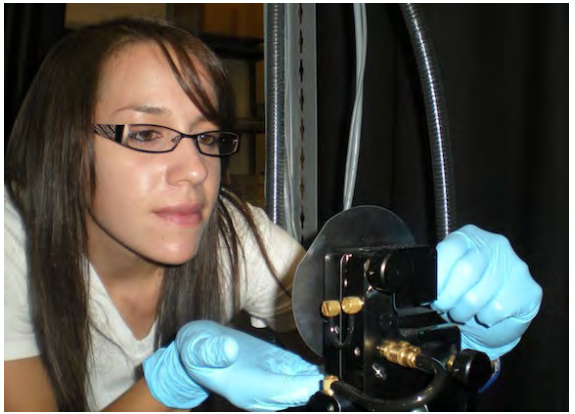
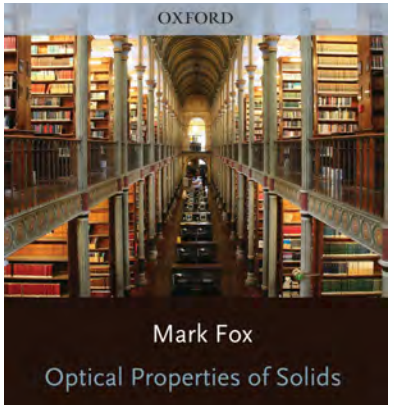
Outline: Ellipsometry Lecture Series

1. Polarized Light and the Dielectric Tensor
2. ~~*Analyzing Ellipsometric Angles and Mueller matrices*~~
   ~~*Reflection of Light by Stratified Planar Structures*~~
   ~~*Berreman 4 by 4 formalism (including anisotropy)*~~
   See slides from other sources.
2. **Lorentz and Drude Models:**
   Infrared Response of Free Carriers and Metals;
   and Lattice Vibrations (Optical Phonons)
3. Interband Electronic Transitions:
   Electronic Band Structure of Crystalline Solids



References:

- Landau/Lifshitz: Electrodynamics of Continuous Media; or Jackson: E&M
- Ashcroft & Mermin: Solid-State Physics
- Mildred Dresselhaus *et al.*: Solid-State Properties
- Yu and Cardona: Fundamentals of Semiconductors
- **Mark Fox: Optical Properties of Solids**
- Cohen/Chelikowsky: Electronic Structure and Optical Properties
- Azzam/Bashara, Fujiwara, Tompkins/Hilfiker, Tompkins/Irene, Fujiwara/Collins: Several good textbooks on Spectroscopic Ellipsometry
- Palik: Handbook of Optical Constants (three volumes)
- Short Course Lectures from instrument suppliers and other sources.



Lecture 1 Outline:
Polarized Light and the Dielectric Tensor

- Spectroscopy, Instrumentation, Bohr Model, Band Structure of Germanium
- **Maxwell's Equations in Fourier Space**
- **Propagation of Light in Vacuum: Plane Waves**
- Jones and Stokes Vectors
- Reflection of Light: **Jones and Mueller Matrices**
- Dielectrics: Electrodynamics of Continuous Media; Optical Constants
- **Propagation of Light in Solids: Inhomogeneous Plane Waves, Crystal Optics**

Lecture 2 Outline: Lorentz and Drude Models: Infrared Response of Free Carriers and Lattice Vibrations (Optical Phonons)

- Lorentz Model
- Drude Model
- Classification of Phonons based on Crystal Symmetry
- Infrared-Active Optical Phonons (Lattice Vibrations)
- Free-Carrier Infrared Absorption
- Optical Properties of Metals (Interband Transitions)

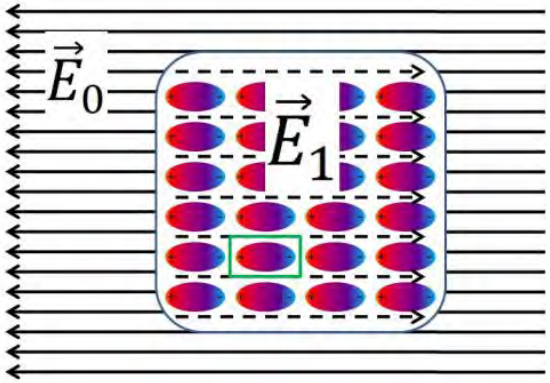
AC Response Function, Dielectric Function

How does a dielectric respond to an electromagnetic wave?

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[i\left(\vec{k}\cdot\vec{r} - \omega t\right)\right]$$

Polarization may be delayed.

~~Polarization may be non-local.~~



$$\vec{P}(\vec{r},t) = \varepsilon_0 \int_{-\infty}^{t} \chi_e(\vec{r}',\vec{r},t',t)\vec{E}(\vec{r}',t')dt'd^3\vec{r}'$$

Time invariance

Infinite homogeneous crystal

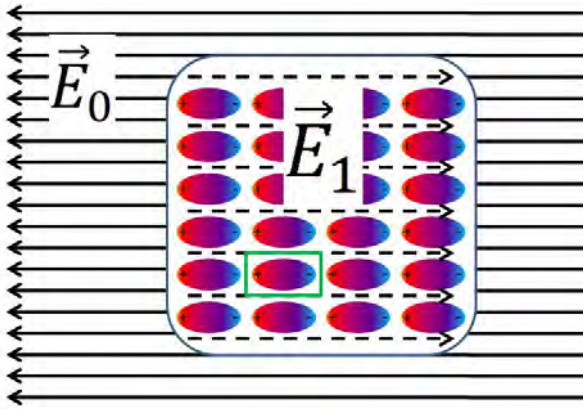
$$\vec{P}(\vec{r},t) = \varepsilon_0 \int_{-\infty}^{t} \chi_e(\vec{r}'-\vec{r},t'-t)\vec{E}(\vec{r}',t')dt'd^3\vec{r}'$$

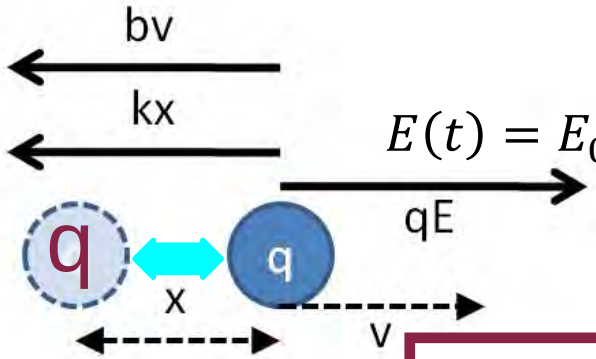
Use convolution theorem for Fourier transforms

$$\vec{P}(\vec{k},\omega) = \varepsilon_0\chi_e(\vec{k},\omega)\,\vec{E}(\vec{k},\omega)$$
$$\vec{D}(\vec{k},\omega) = \varepsilon_0\varepsilon(\vec{k},\omega)\,\vec{E}(\vec{k},\omega)$$

Dielectric function ε depends on frequency ω (dispersion).

Lorentz Model for Oscillating Charges

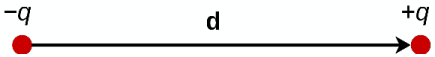




Static screening.
Friction proportional to velocity.
Small displacement (harmonic).

$$F = ma$$
$$qE - b\dot{x} - kx = m\ddot{x}$$
$$\text{Try } x(t) = x_0 \exp(-i\omega t)$$
$$x(t) = \frac{-qE_0}{m\omega^2 + ib\omega - k}\exp(-i\omega t)$$
$$P(t) = \chi_e E(t) = \frac{qx(t)}{V}$$
$$\varepsilon = 1 + \chi_e$$

dipole moment



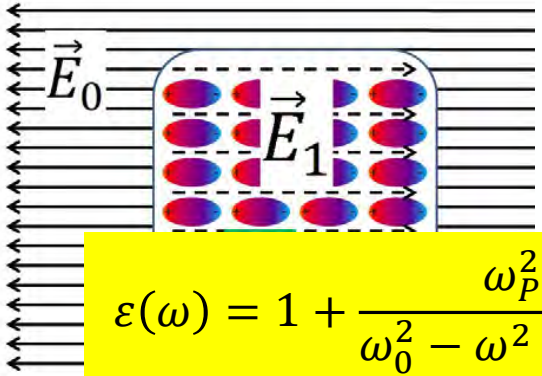
$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$\omega_P^2 = \frac{nq^2}{m\varepsilon_0}$$ Charge density

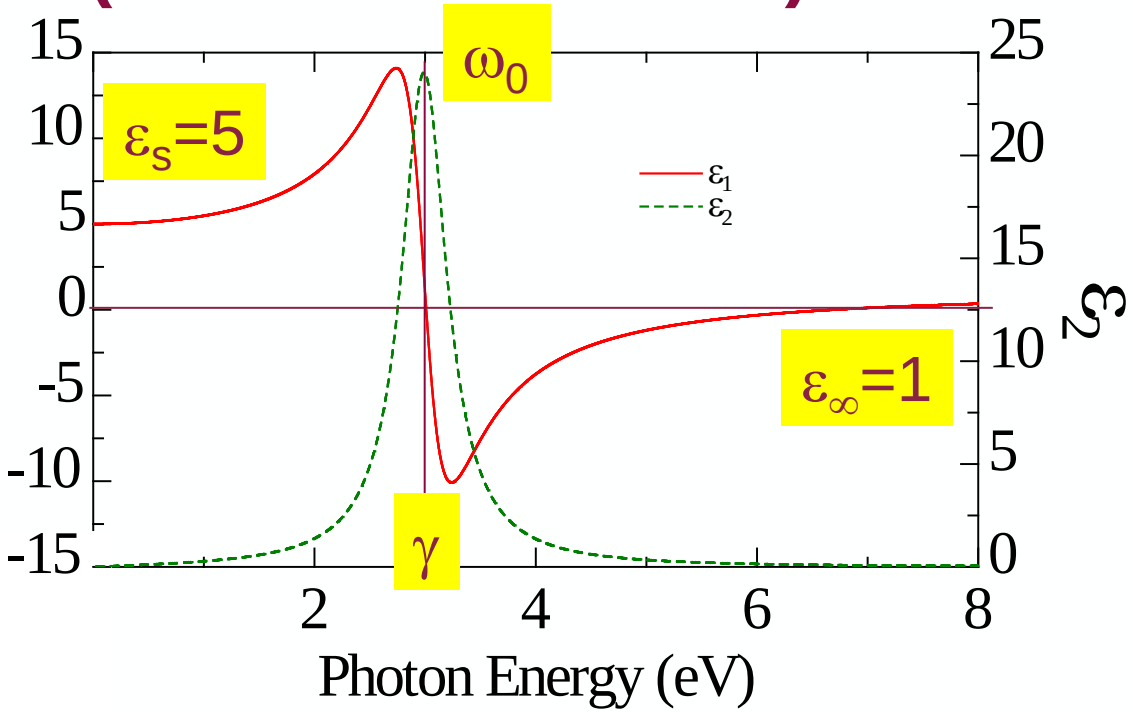
$$\omega_0^2 = \frac{k}{m}$$ Resonance frequency

H. Helmholtz, Ann. Phys **230**, 582 (1875)
F. Wooten, *Optical Properties of Solids*, 1972

Lorentz Model (Dielectric Function)



$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$



ω_0=3 eV, γ=0.5 eV, ω_p=6 eV

- Peak of ε_2 at ω_0
- Broadening γ
- Amplitude $\omega_P^2 = A\omega_0^2$
- Dimensionless $A = \varepsilon_s - \varepsilon_\infty$
- ε_2 is never negative
- ε_1 has a wiggle at ω_0
- Longitudinal solution for

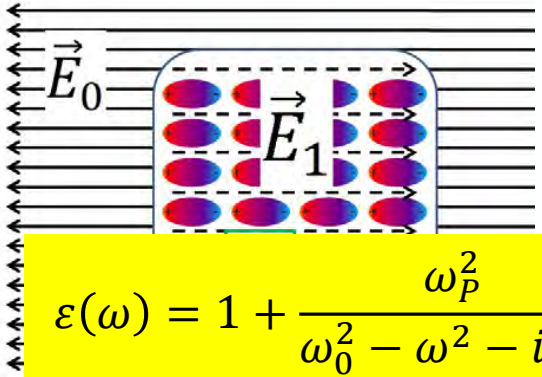
$$\omega_L = \sqrt{\omega_0^2 + \omega_P^2 - i\gamma} \approx 6.7 \text{ eV}$$

- ε_1 negative from ω_0 to ω_L

ıture.

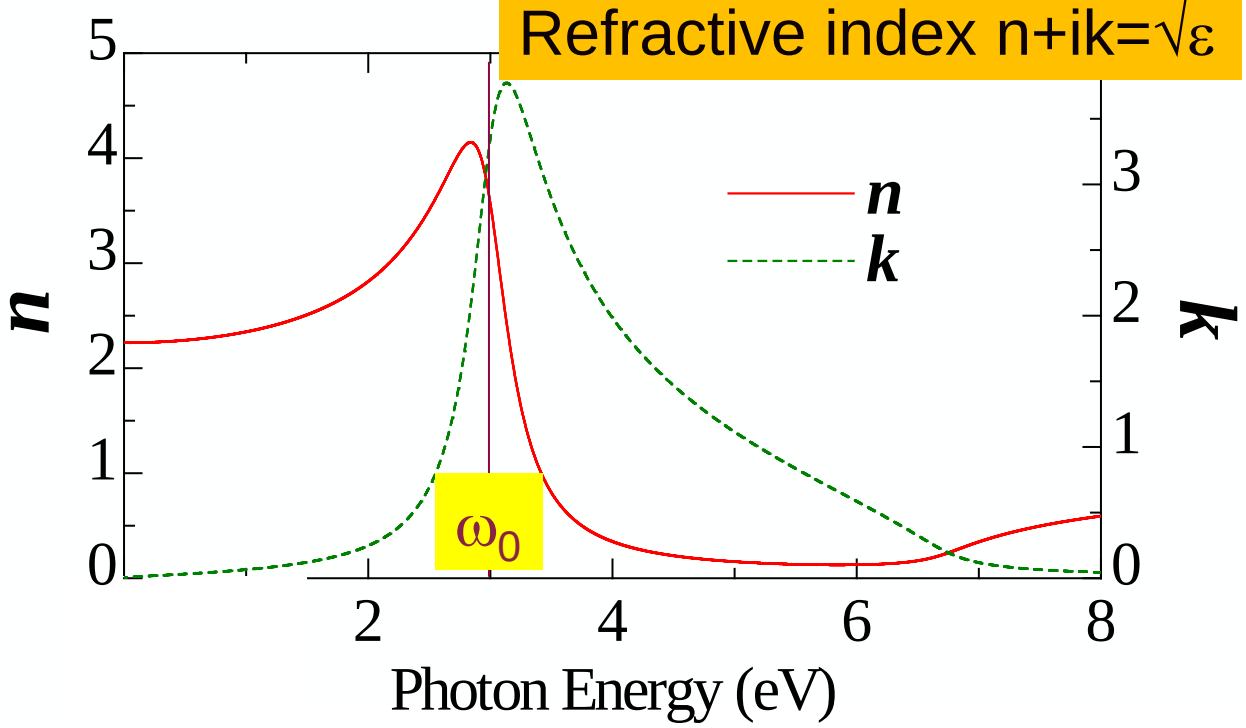
F. Wooten, *Optical Properties of Solids*, 1972

Lorentz Model (Complex Refractive Index)



$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

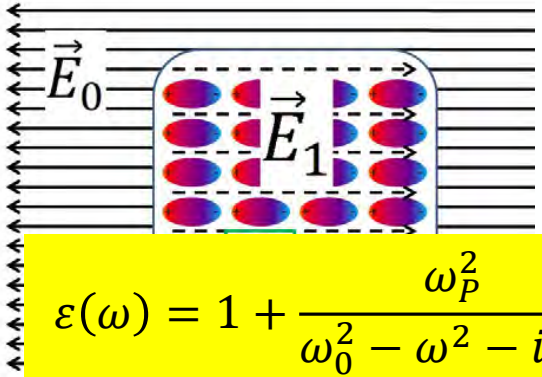
Refractive index n+ik=√ε



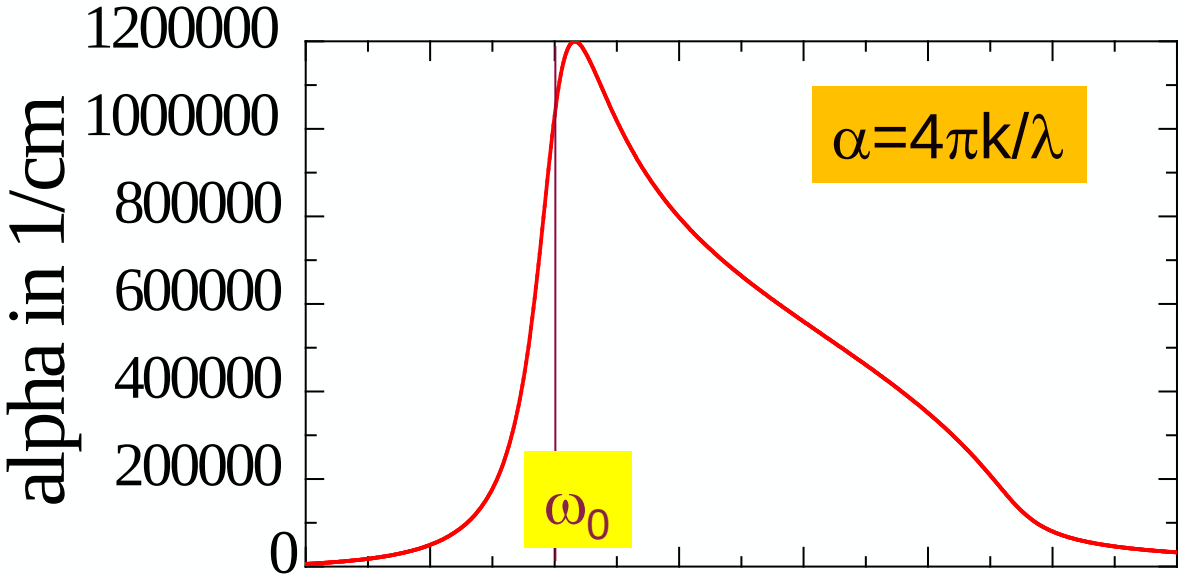
ω_0=3 eV, γ=0.5 eV, ω_p=6 eV

Peak of k shifted (>ω_0)
k is asymmetric
n and k always positive
n→1 at large energies
n<1 above ω_0, below ω_L
(Reststrahlen band,
high reflectance, plasmonics)
Normal dispersion: dn/dE>0
Anomalous dispersion: dn/dE<0

Lorentz Model (Absorption Coefficient)



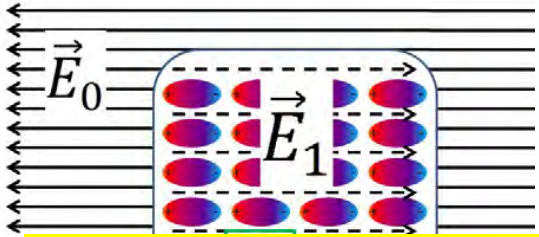
$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$



Peak of α shifted ($>\omega_0$).
α is always positive.
α is asymmetric.
Fast rise, slow drop.

ω_0=3 eV, γ=0.5 eV, ω_p=6 eV

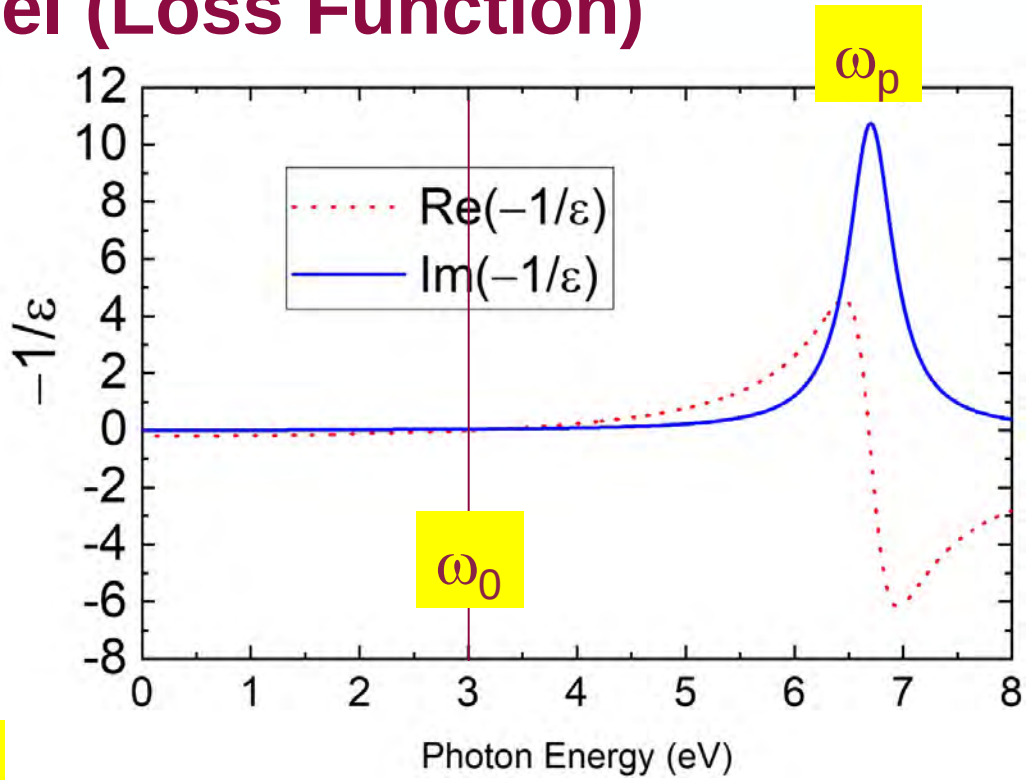
Lorentz Model (Loss Function)



$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

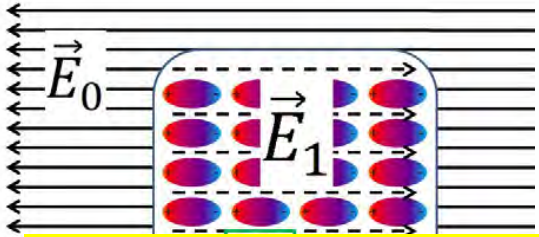
The loss function **Im(–1/ε)** peaks at the longitudinal frequency.

$$\omega_L = \sqrt{\omega_0^2 + \omega_P^2 - i\gamma} \approx 6.7 \text{ eV}$$



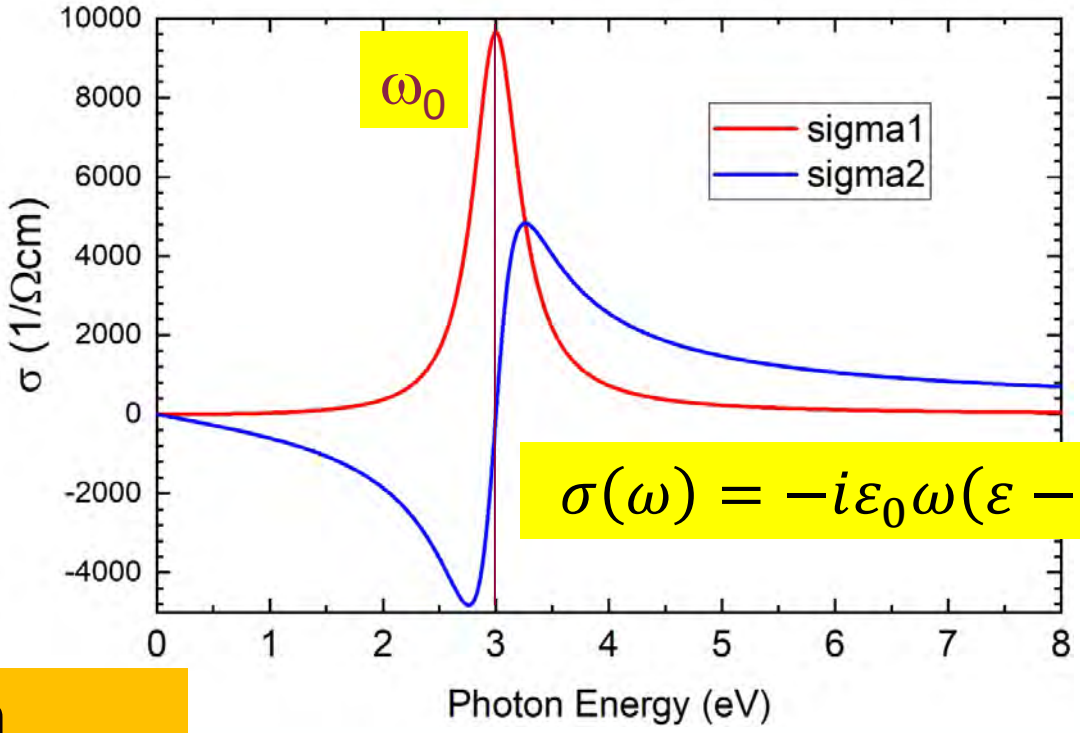
ω_0=3 eV, γ=0.5 eV, ω_p=6 eV

Lorentz Model (Optical Conductivity)



$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

The optical conductivity has a peak at the resonance frequency.



$$\sigma(\omega) = -i\varepsilon_0\omega(\varepsilon - 1)$$

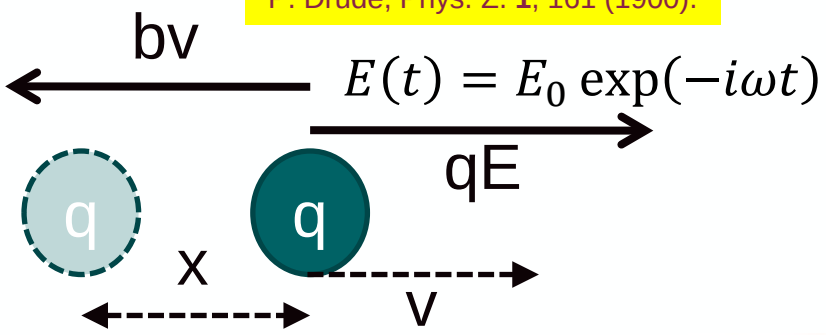
ω_0=3 eV, γ=0.5 eV, ω_p=6 eV

Re(σ), Im(ε): Dissipation
Im(σ), Re(ε): Dispersion
j=σE
Absorption is a resonant current.

Drude Model for Free Carriers

Drude: Free Charges

P. Drude, Phys. Z. **1**, 161 (1900).



Static screening.
Friction proportional to velocity.

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

$$\omega_P^2 = \frac{nq^2}{m\varepsilon_0}$$

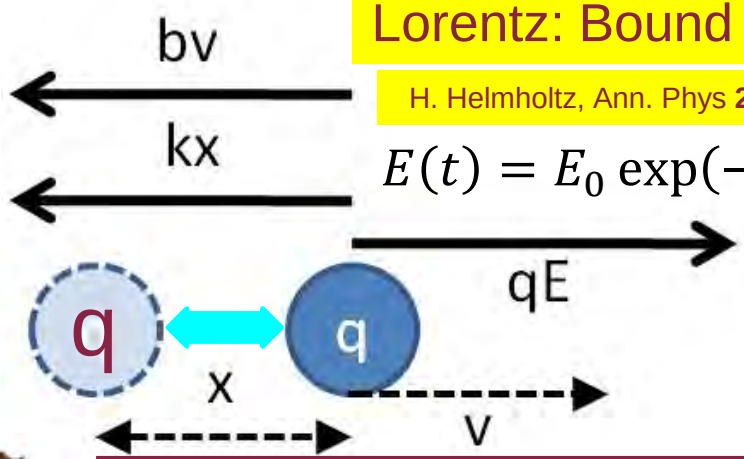
Charge density

$$\omega_0^2 = 0$$

Resonance frequency

Lorentz: Bound Charges

H. Helmholtz, Ann. Phys **230**, 582 (1875)



$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_0^2 - \omega^2 - i\gamma\omega}$$

$$\omega_P^2 = \frac{nq^2}{m\varepsilon_0}$$

Charge density

$$\omega_0^2 = \frac{k}{m}$$

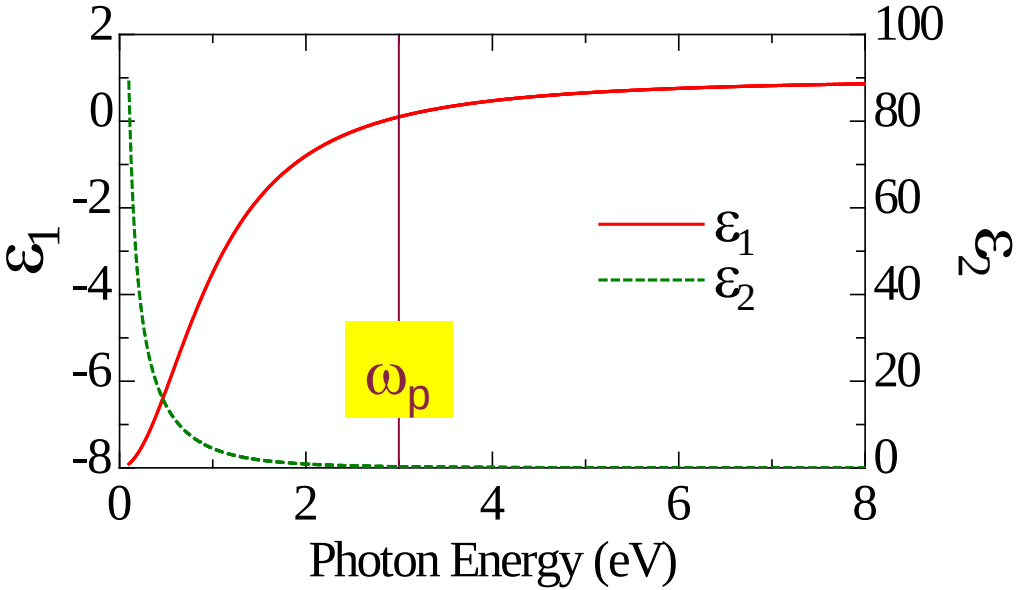
Resonance frequency

Drude Model for Free Carriers (Dielectric Function)

Both ε_1 and ε_2 **diverge** at $\omega=0$
Broadening γ
$\varepsilon_1 \rightarrow 1$ at large energies
$\varepsilon_2 \rightarrow 0$ at large energies

$$\omega_L = \sqrt{\omega_P^2 - i\gamma} \approx \omega_P = 3 \text{ eV}$$

ε_1 negative from ω_0 to ω_L
Real/imaginary part has factor γ/ω



$$\varepsilon(\omega) = 1 - \frac{\omega_P^2}{\omega^2 + i\gamma\omega} = 1 - \frac{\omega_P^2}{\omega^2 + \gamma^2} + i\frac{\omega_P^2}{\omega^2 + \gamma^2} \times \frac{\gamma}{\omega}$$

ω_p=3 eV, γ=1 eV, $\tau=1/\gamma$=0.6 fs

Bad metal

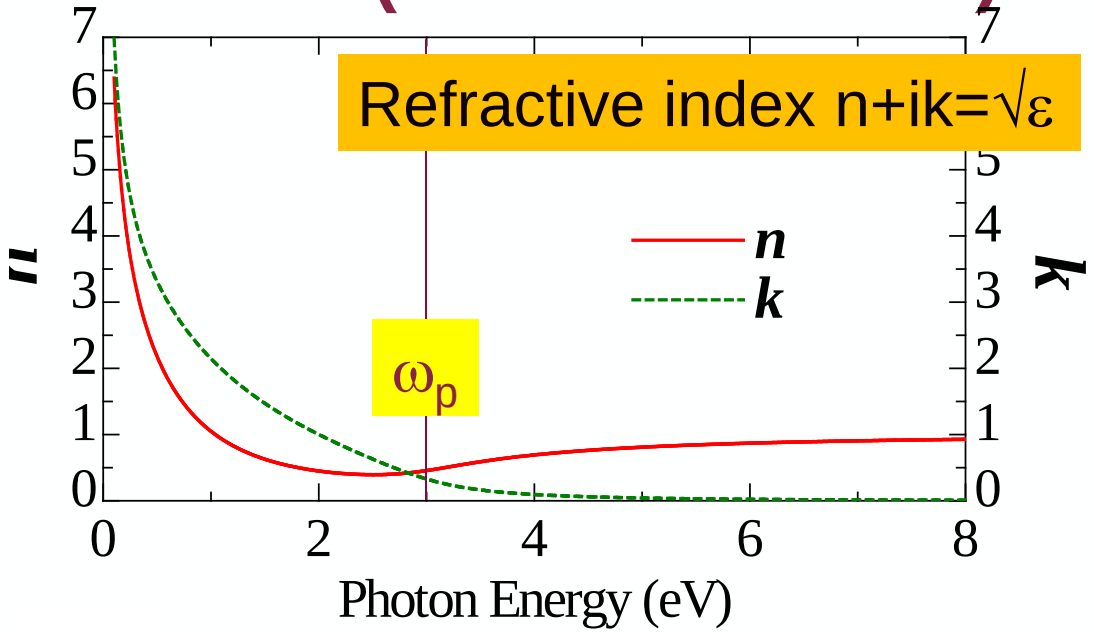
$$n = \frac{\omega_P^2 \epsilon_0 m_0}{\hbar^2 e^2} = 6.5 \times 10^{21} \text{cm}^{-3}$$

Drude Model for Free Carriers (Refractive Index)

Both n and k diverge at $\omega=0$
Broadening γ
n drops off faster than k
n, k always positive

$n \rightarrow 1$ at large energies
$n<1$ at large energies
(important for XRR)
$v_{phase}>c$ if $n<1$
n drops up to ω_P, then rises.

$k \rightarrow 0$ at large energies



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

Drude Model (Absorption Coefficient)

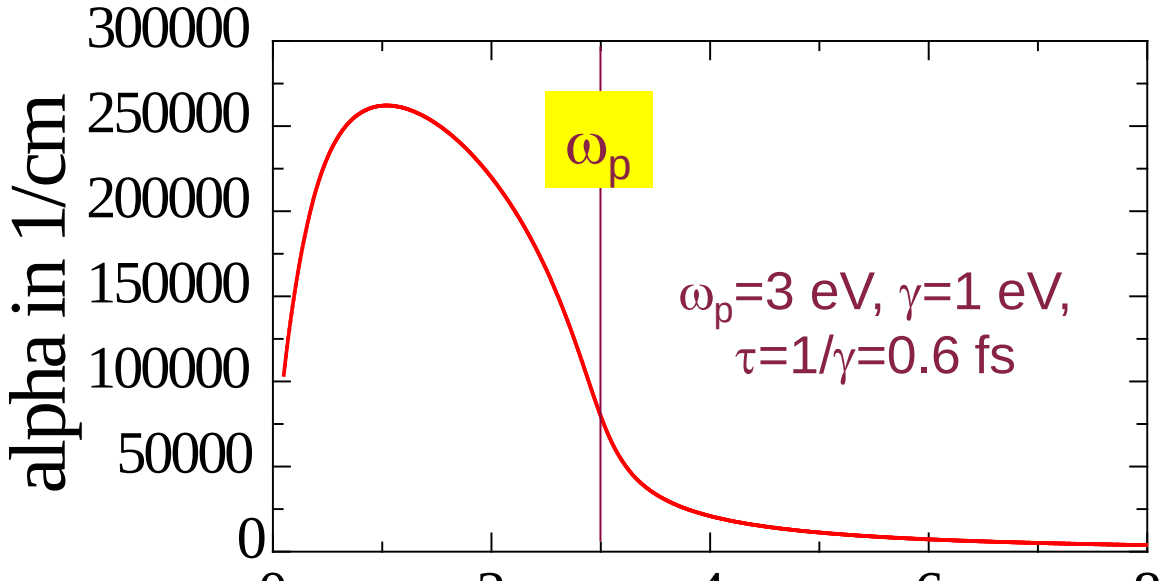
$\alpha \to 0$ as $E \to 0$.

Peak around $\omega_p/2$

Small α above ω_P.
$\alpha \to 0$ as $E \to \infty$

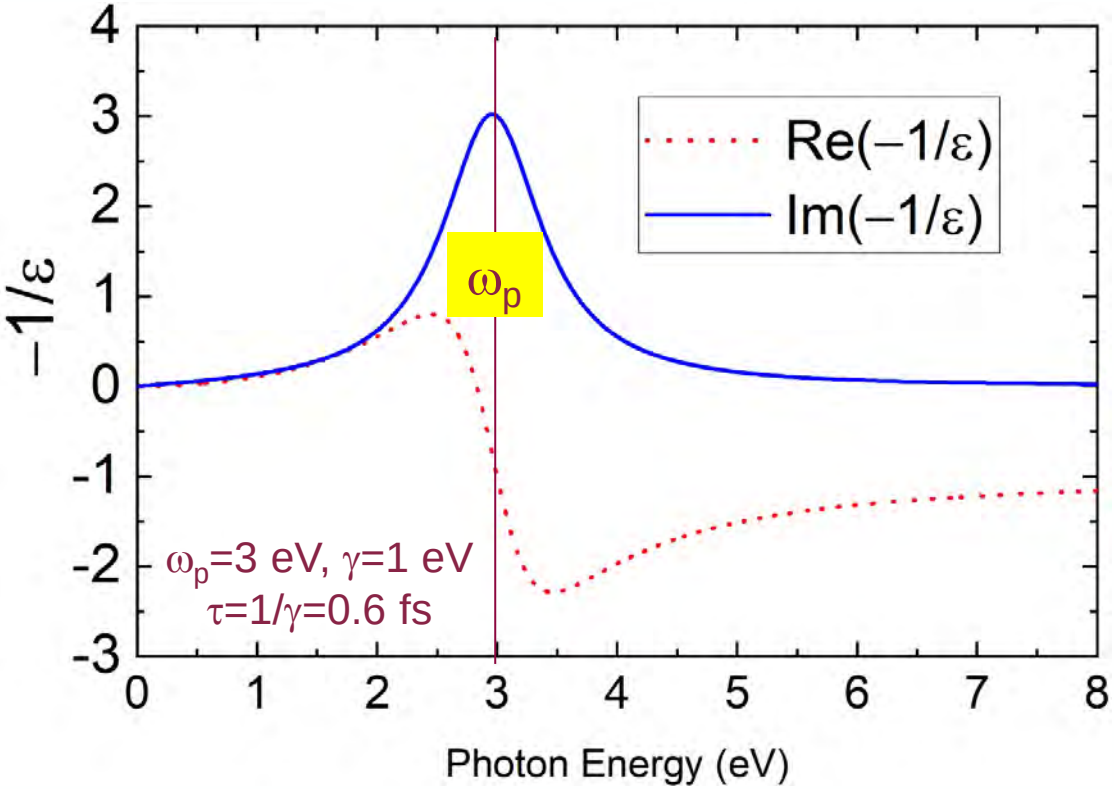
Metals become nearly transparent above the plasma frequency.

Reflectance minimum at ω_P.



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

Drude Model for Free Carriers (Loss Function)



ε peak:
Dissipation (TO)

Im(–1/ε) peak:
Plasmon solution (LO)

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

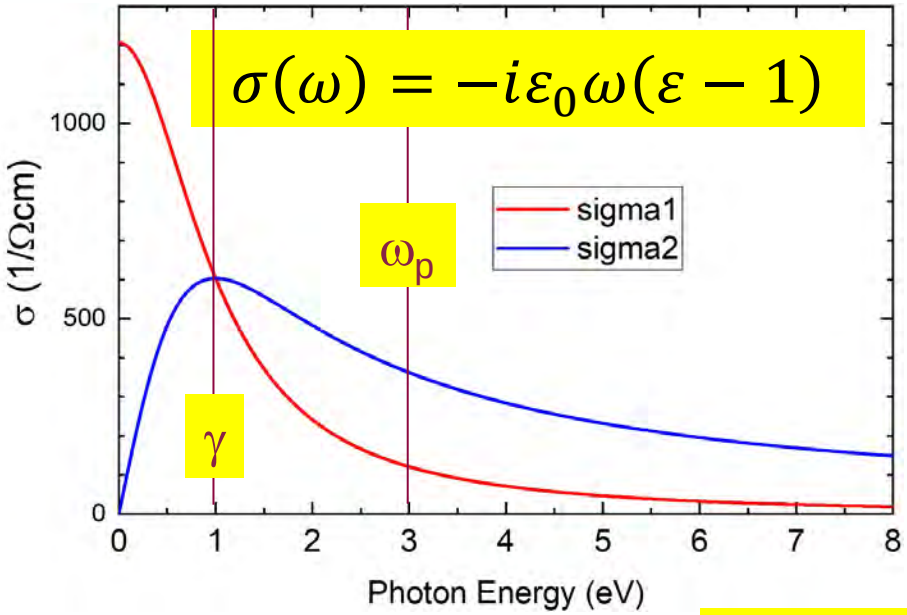
ω_p=3 eV, γ=1 eV

The loss function **Im(–1/ε)** peaks at the longitudinal frequency

$$\omega_L = \sqrt{\omega_P^2 - i\gamma} \approx \omega_P = 3 \text{ eV}$$

Drude Model (Optical Conductivity)

$$\sigma(\omega) = -i\varepsilon_0\omega(\varepsilon - 1)$$



Multiplying by ω cancels the divergence.

$\sigma_2 \to 0$ as $E \to 0$

σ_2 peaks at $\omega=\gamma$

Finite $\sigma_{DC}=\sigma_1(\omega=0)$

$\sigma_{DC}=ne\mu=ne^2\tau/m_0m^*$

$\tau=1/\gamma$ scattering time

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

Re(σ), Im(ε): Dissipation
Im(σ), Re(ε): Dispersion
j=σ**E**

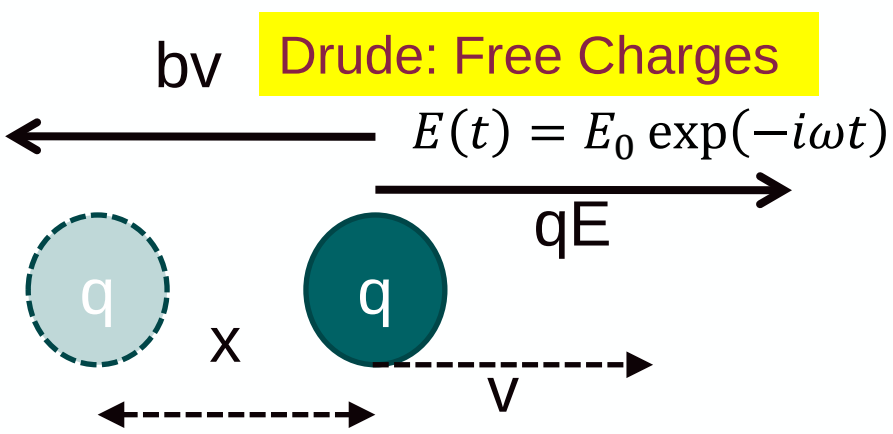
σ_{DC}=1200 1/Ωcm

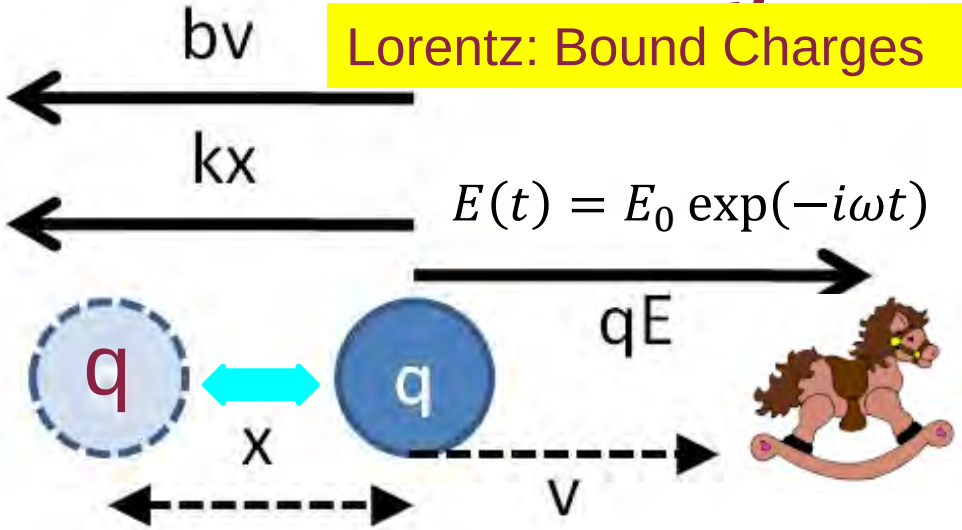
Bad metal

ω_p=3 eV, γ=1 eV,
μ_0=1.1 cm²/Vs

Dresselhaus, *Solid-State Properties*

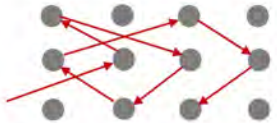
Drude-Lorentz Model: Free and Bound Charges





Discuss plasma frequency trends.

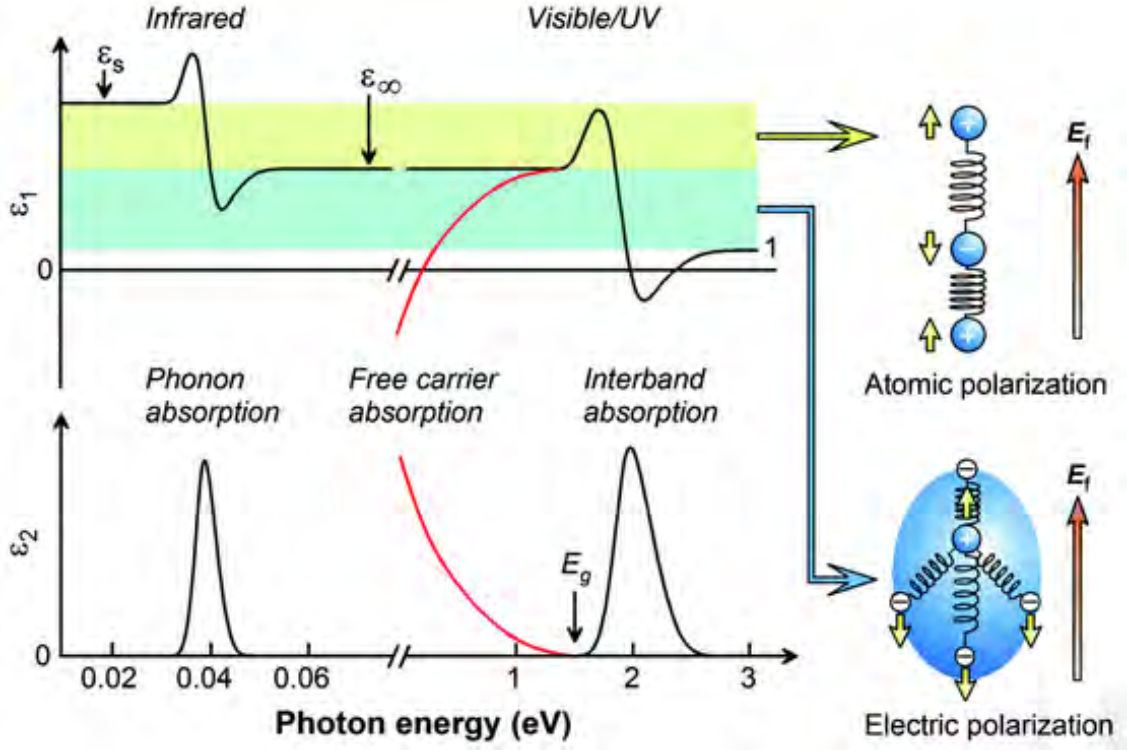
$$\omega_P^2 = \frac{n_f e^2}{m\varepsilon_0}$$



$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

ω_P	(unscreened) **plasma frequency** of free charges
ω_0	**resonance frequency** of bound charges
γ_D, γ_0	**broadenings** of free and bound charges
A	**amplitude** of bound charge oscillations (density, strength)

Drude-Lorentz Model: Free and Bound Charges



Mid-infrared spectral range:

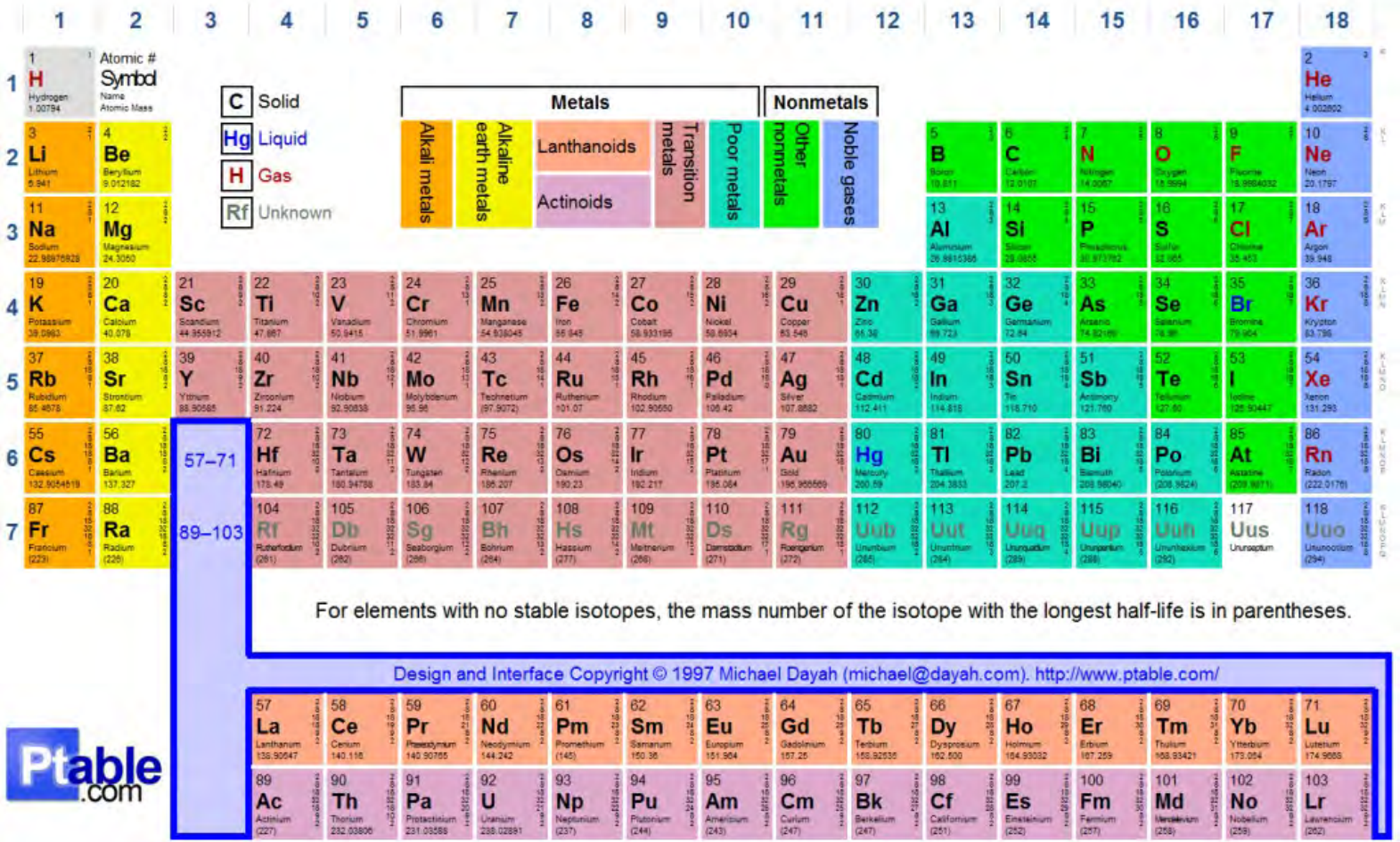
- Insulator/semiconductor: Lattice vibrations (optical long-wavelength phonons)
- Metal: Free carrier properties (density, scattering rate)

Near-IR to visible to UV range:

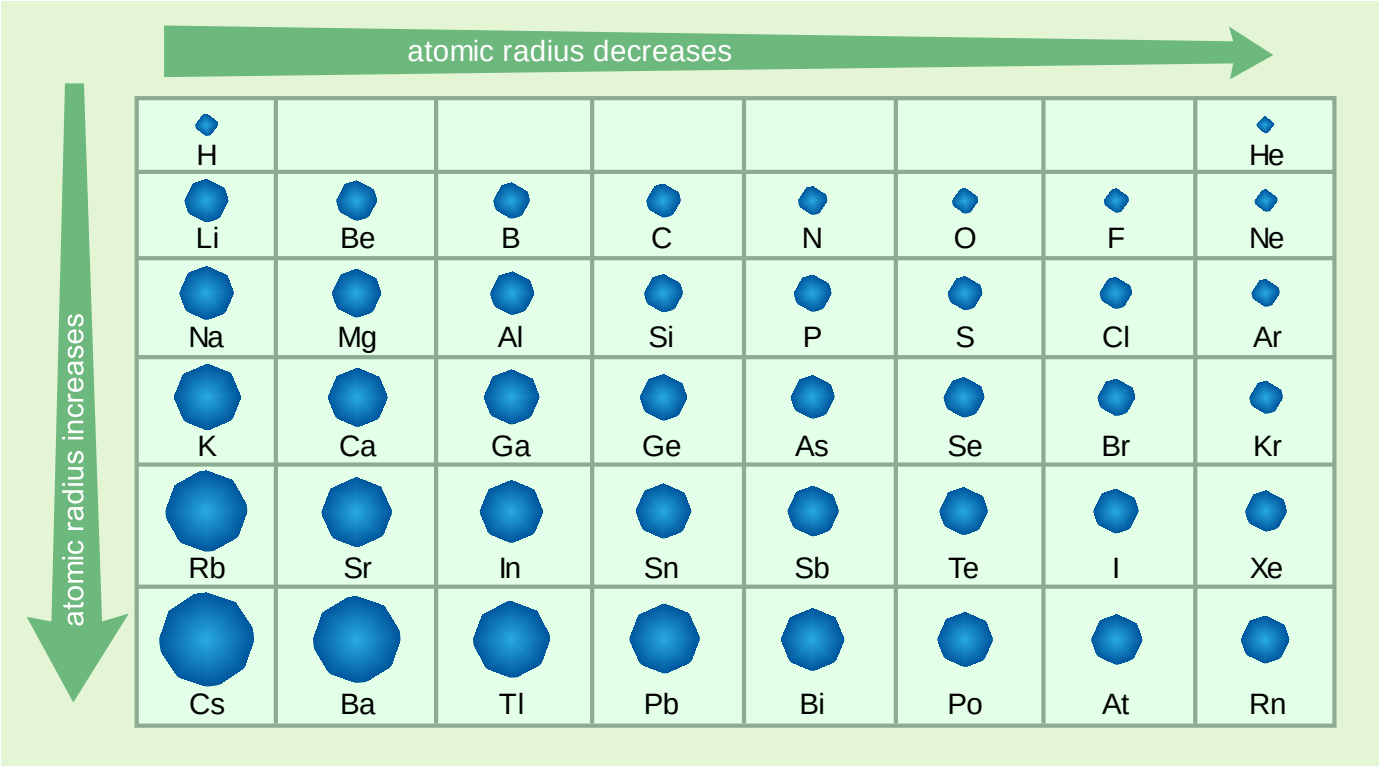
- Electronic excitations
- Band gap, interband transitions

$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

Metals: Calculate Plasma Frequency

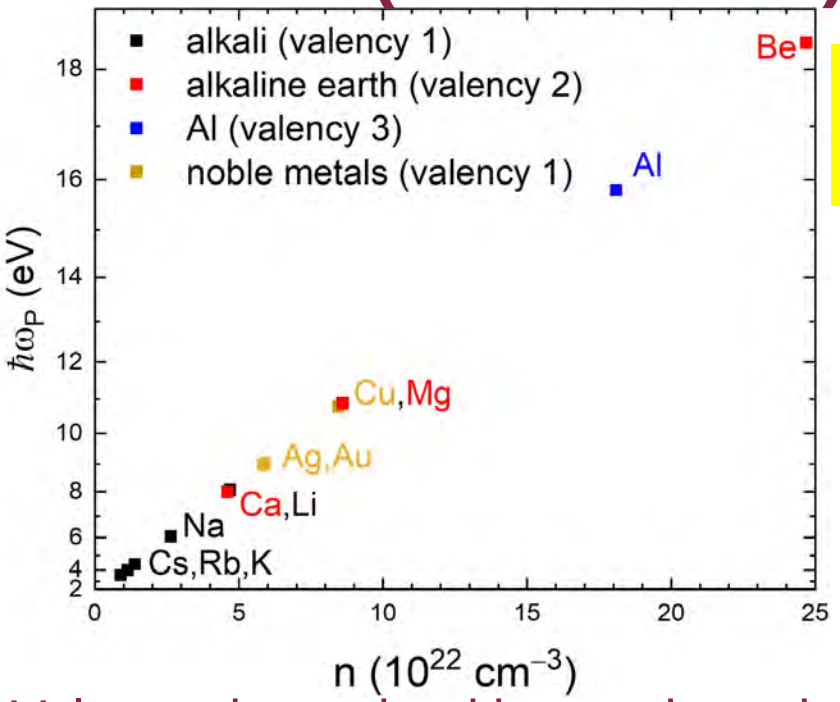


Atomic Radius



Atomic radius decreases to the right, increases downward.

(Unscreened) Plasma Frequency



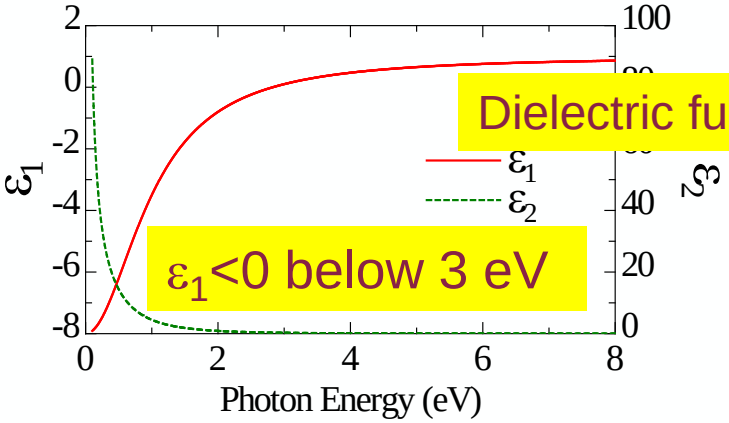
$$\omega_P^2 = \frac{n_f e^2}{m\varepsilon_0}$$

Metal	Valency	N (10^{28} m^{-3})	$\omega_p/2\pi$ (10^{15} Hz)	λ_p (nm)
Li (77 K)	1	4.70	1.95	154
Na (5 K)	1	2.65	1.46	205
K (5 K)	1	1.40	1.06	282
Rb (5 K)	1	1.15	0.96	312
Cs (5 K)	1	0.91	0.86	350
Cu	1	8.47	2.61	115
Ag	1	5.86	2.17	138
Au	1	5.90	2.18	138
Be	2	24.7	4.46	67
Mg	2	8.61	2.63	114
Ca	2	4.61	1.93	156
Al	3	18.1	3.82	79

Valency determined by row in period table.
Atomic radius decreases from K to Ca to Cu.

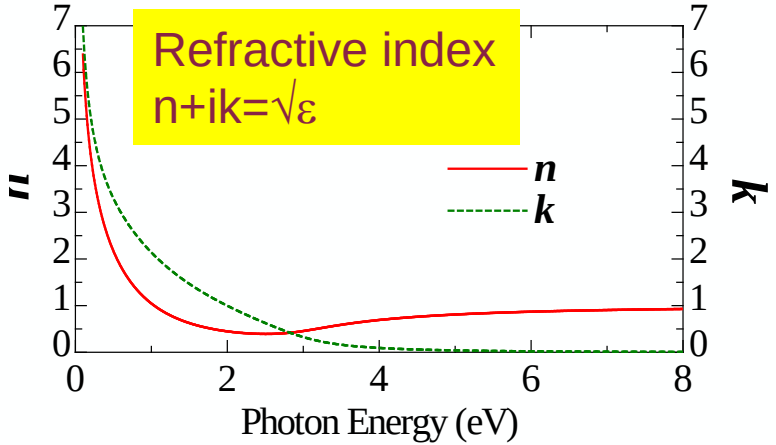
Fox, Table 7.1

Free-Carrier Reflection/Absorption in Metals



Dielectric function ε

ε_1<0 below 3 eV



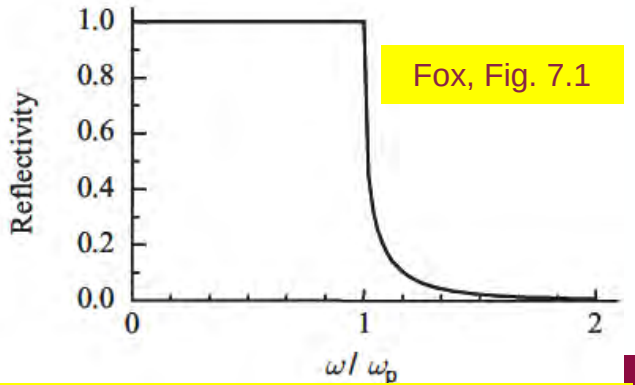
Refractive index
n+ik=√ε

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

ω_p=3 eV, γ=1 eV

$$R_{90}(\omega) = \left|\frac{n + ik - 1}{n + ik + 1}\right|^2$$

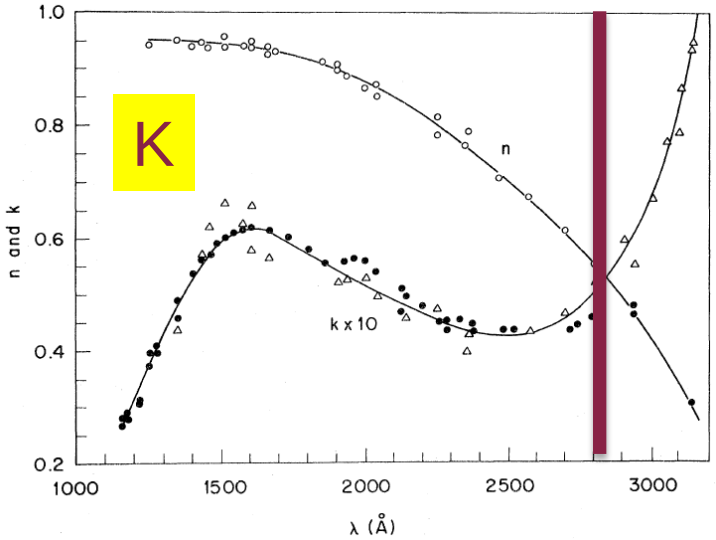
R=1 if n is purely imaginary (γ=0) below ω_P.



Fox, Fig. 7.1

Metals reflect below ω_P (plasma edge)

Transparent Alkali Metals Above ω_P

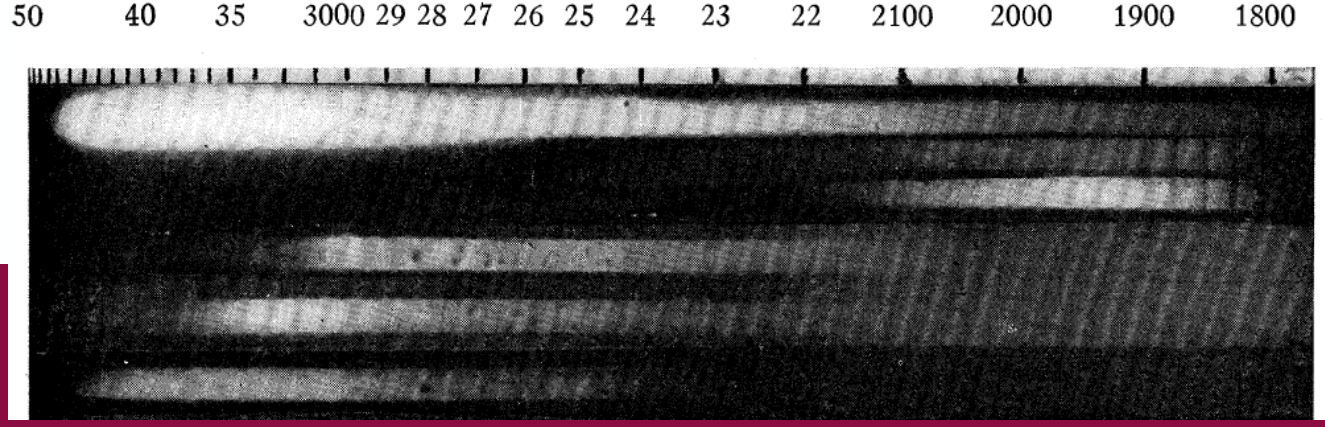


U.S. Whang *et al.*, PRB **6**, 2109 (1972)

Metal	λ_{UV} (nm)
Li	205
Na	210
K	315
Rb	360
Cs	440

Fox, Table 7.2

ω_P=4.4 eV (280 nm)



transmission

R.W. Wood, Phys. Rev. **44**, 353 (1933)

Bands of Total Reflection

Occur below plasma frequency and between TO/LO energies.
Increased sensitivity to weak absorption processes.

Drude model: $\varepsilon(\omega) = 1 - \frac{\omega_P^2}{\omega^2 + i\gamma\omega}$

Small damping ($\gamma << \omega_P$): $\varepsilon(\omega) = 1 - \frac{\omega_P^2}{\omega^2}$ (real, negative)

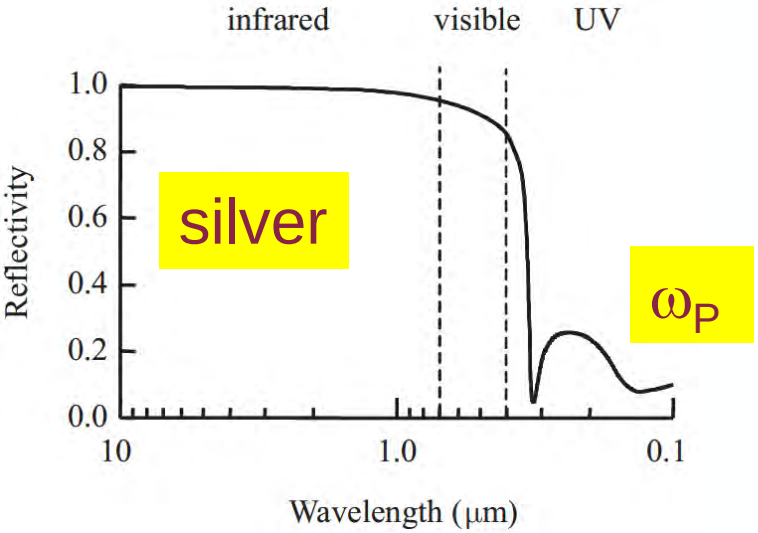
Low frequency ($\omega < \omega_P$): $\varepsilon(\omega) < 0$

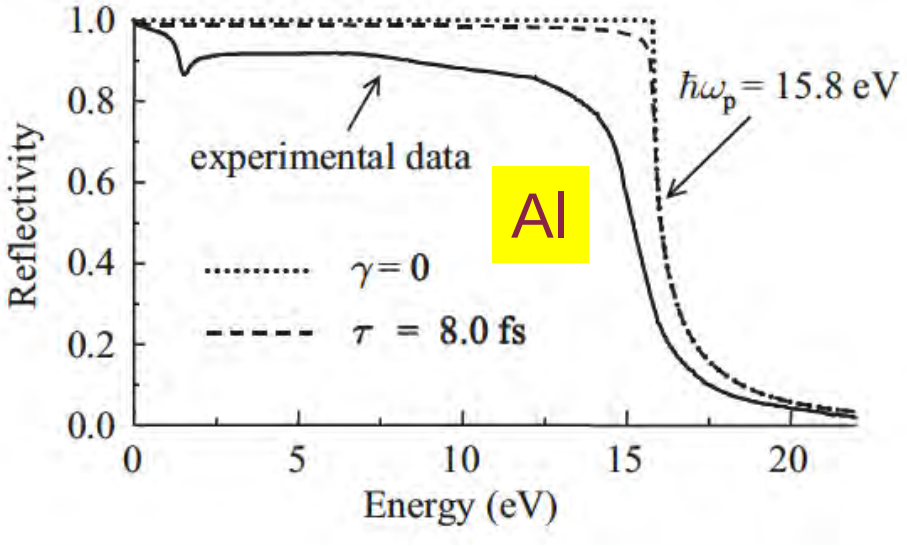
Refractive index ($\omega << \omega_P$): $\tilde{n}(\omega) = \sqrt{\varepsilon(\omega)} \approx ik$ (purely imaginary)

Reflectance at 90° ($\omega < \omega_P$):

$$R_{90}(\omega) = \left|\frac{n + ik - 1}{n + ik + 1}\right|^2 = \left|\frac{ik - 1}{ik + 1}\right|^2 = \frac{(ik - 1)(-ik - 1)}{(ik + 1)(-ik + 1)} = 1$$

Free-Carrier Reflection in Ag and Al

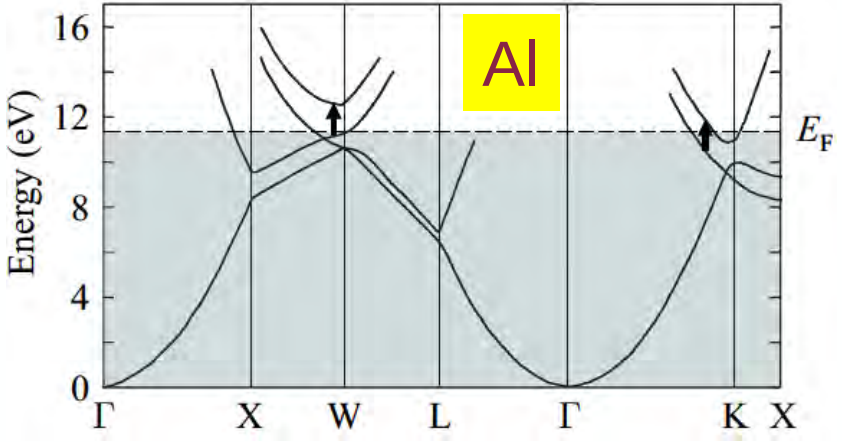


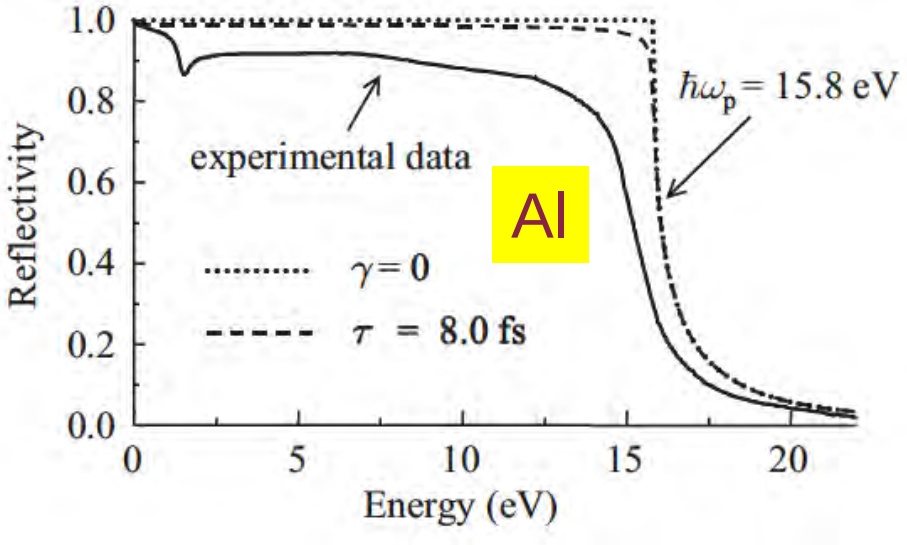


Ag is a noble metal.
Filled 4d-shell, $5s^1$
High reflectance
below ω_P=9 eV (above 138 nm)
Sharp drop above ω_P. Damping.

Al has three electrons ($3s^2$, $3p^1$)
High reflectance
below ω_P=15.8 eV (above 78 nm)
Sharp drop above ω_P.
Damping, interband absorption.

Free-Carrier Reflection in Al



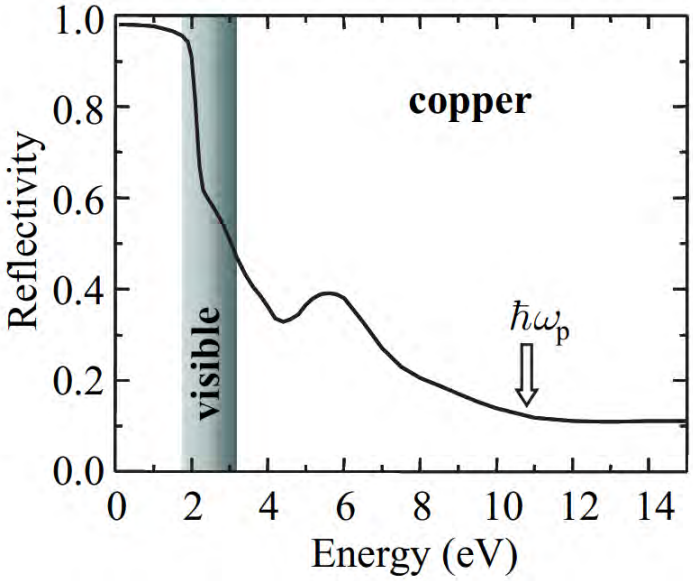


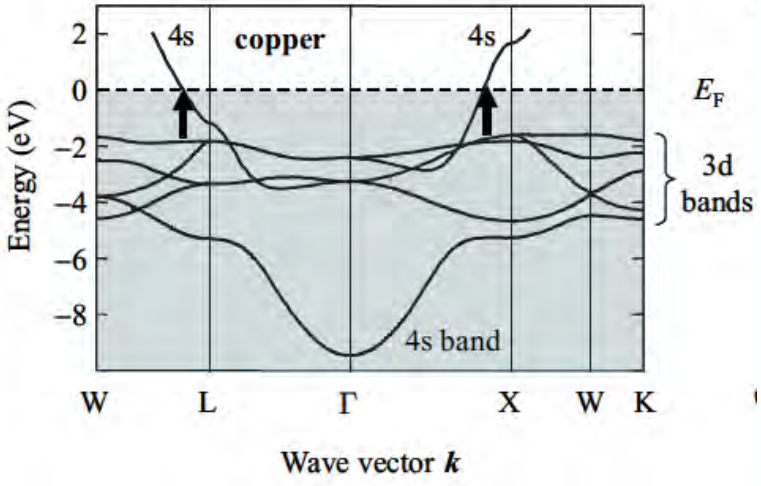
Interband transitions at W cause absorption band at 1.5 eV, lowers reflectivity.

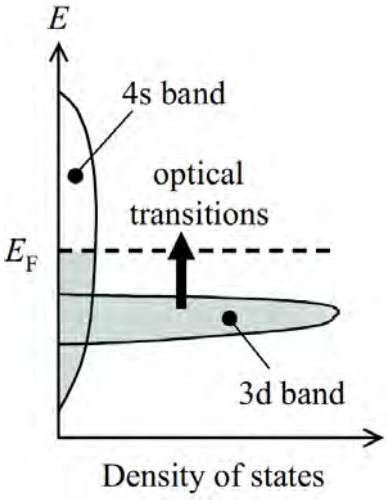
See also: G. Jungk, Thin Solid Films **234**, 428 (1993).

Al has three electrons ($3s^2$, $3p^1$)
High reflectance
below ω_P=16 eV (above 78 nm)
Sharp drop above ω_P.
Damping, interband absorption.

Free-Carrier Reflection in Cu





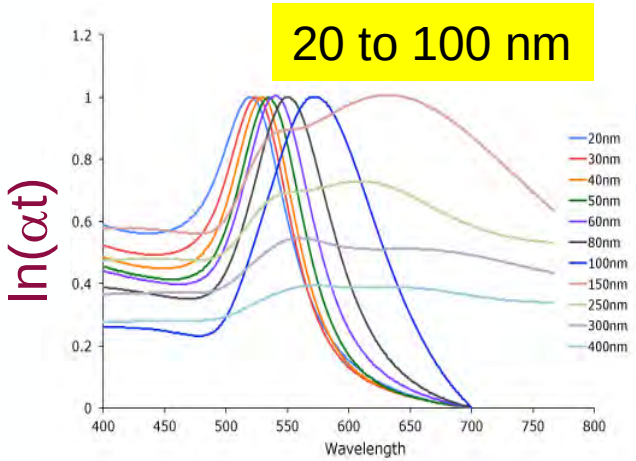


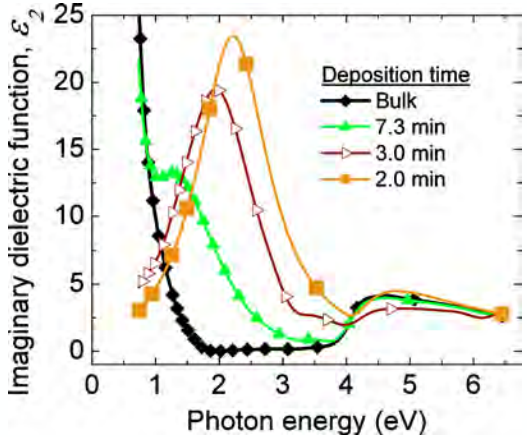
Noble metal, $4s^1$, ω_P=10.8 eV
Transitions from 3d to 4s at 2 eV (near L and X). Similar for Ag, Au.

Fox, *Optical Properties of Solids*

Plasmon Resonance in Gold Nanoparticles

20 to 100 nm





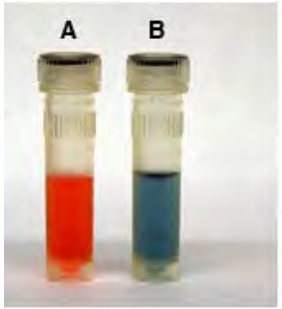
Gold is not always yellow.

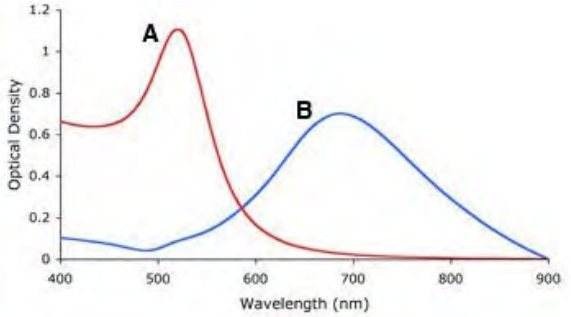
Nanoparticle radius a<λ

$$\alpha = 4\pi a^3 \frac{\varepsilon_m - \varepsilon_d}{\varepsilon_m + 2\epsilon_d}$$

m: metal, d: dielectric

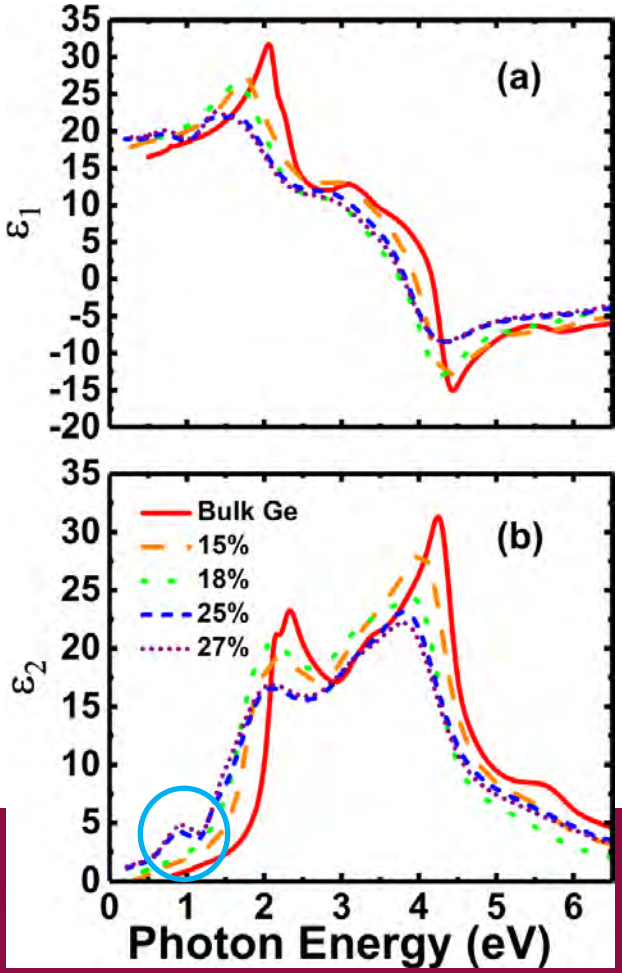
Enhance molecular absorption.





Fox, *Optical Properties of Solids*
Little, *APL* **98**, 101910 (2011)

Plasmon Resonance in β-Tin Nanoparticles



Ge-Sn alloys with high tin content
"have a broadened peak in ε_2, near 0.9 eV."

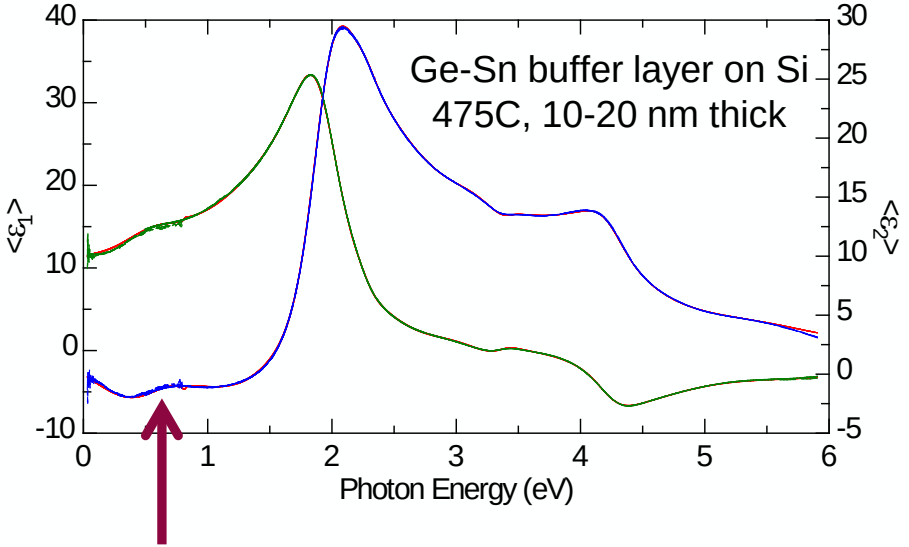
"possibly caused by an intrinsic feature such as from the **band structure**, an extrinsic one such as from a **defect**, or an **interference fringe** from the substrate".

Answer:
None of the above.
This peak is likely a **plasmon resonance** of metallic β-Sn precipitates.

D. Imbrenda, APL **113**, 122104 (2018)

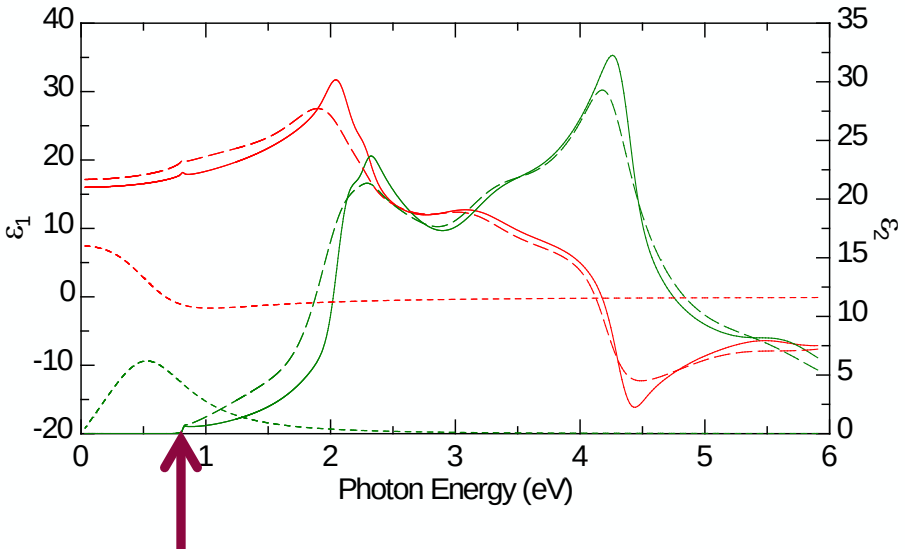
Plasmon Resonance in β-Tin Nanoparticles

Pseudodielectric function



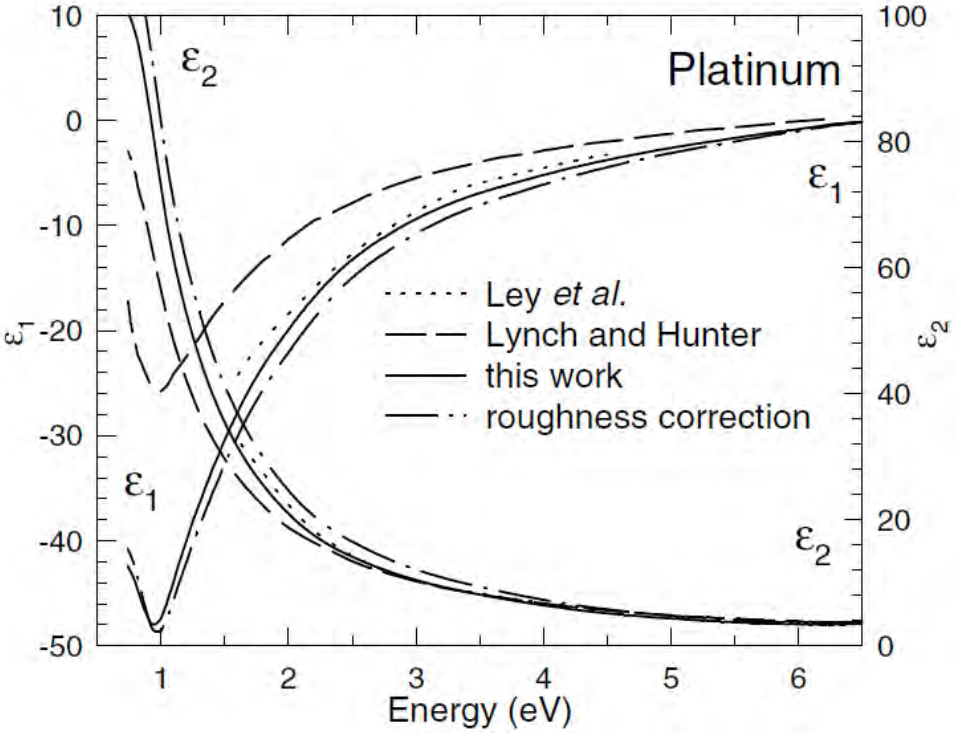
Not an interference fringe.

Dielectric function

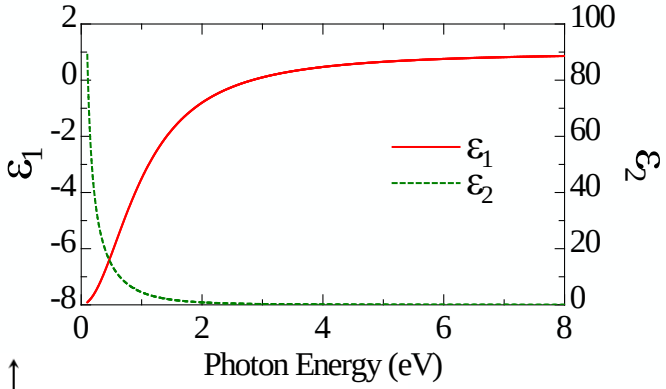


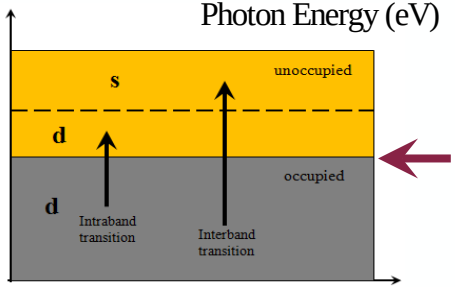
This Lorentzian peak is likely a
plasmon resonance of metallic β-Sn precipitates.

Dielectric Function of Transition Metals (Pt)



Drude-like ε above 1 eV



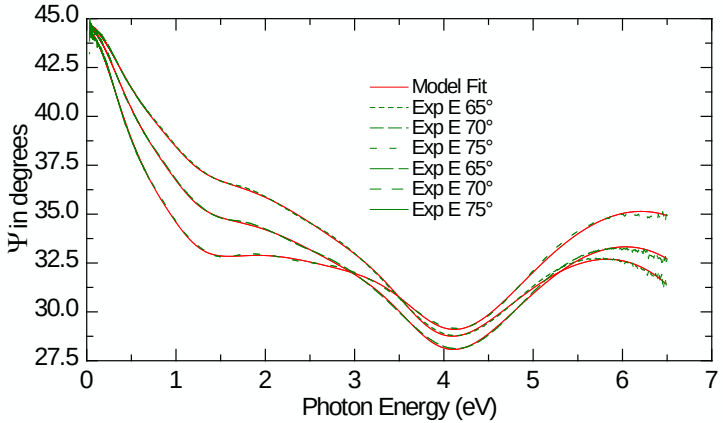


E
Fermi

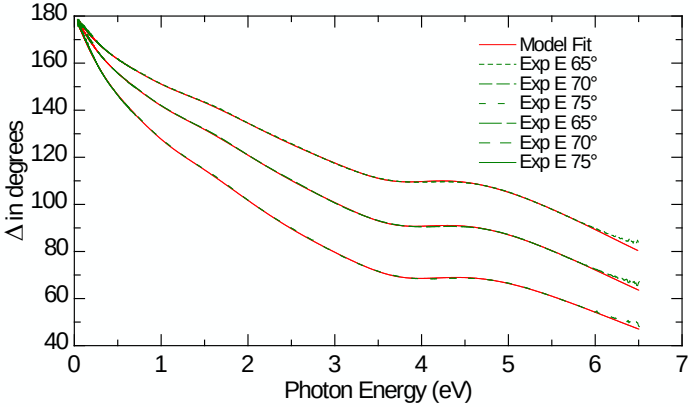
The dielectric function of Pt deviates from the Drude model below 1 eV due to d-interband transitions. Pt is **not a noble metal**, partially filled d-shell.

S. Zollner, phys. stat. solidi (a) **177**, R7 (2000)

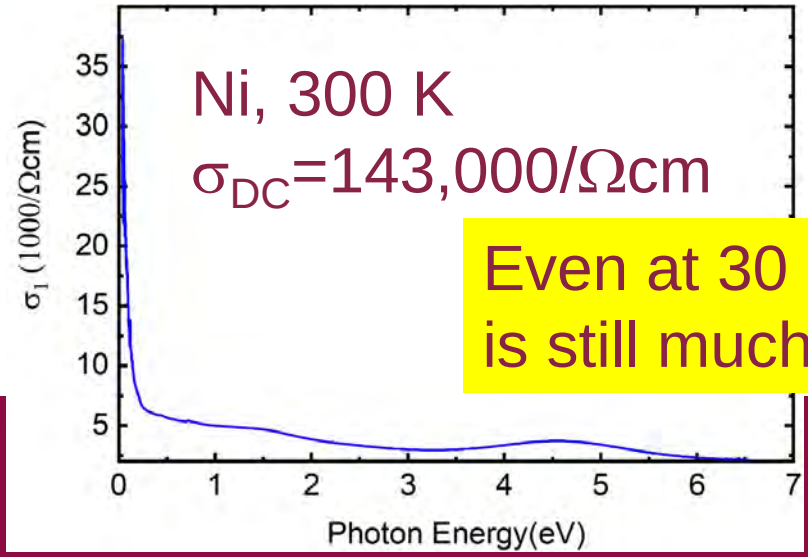
Dielectric Function of Transition Metals (Ni)



Drude response:
Low frequency:
$R \rightarrow 1$
$\psi \rightarrow 45^\circ$
$\Delta \rightarrow 180^\circ$

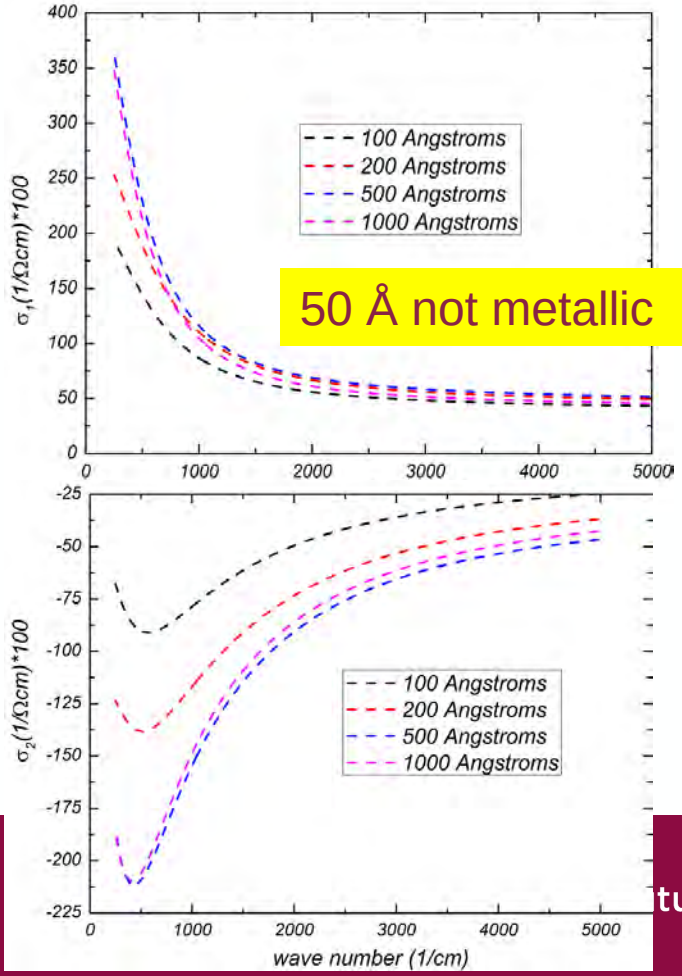


Spatial dispersion:
Anomalous skin effect ???

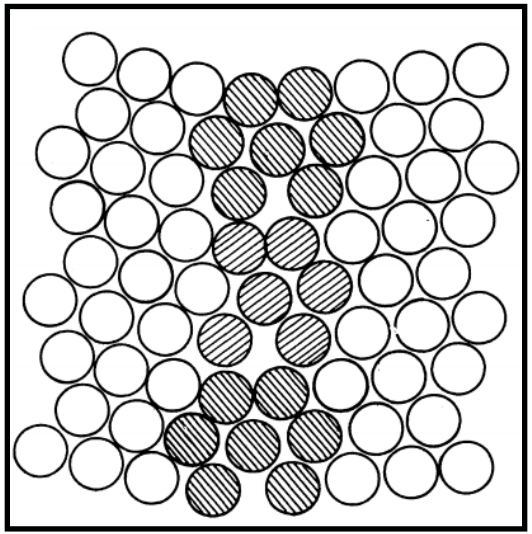


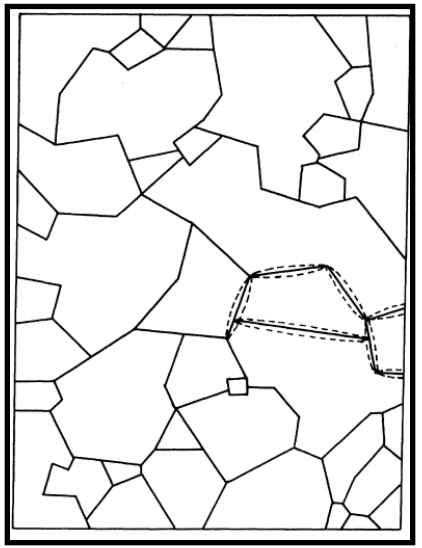
Even at 30 meV, the optical σ is still much smaller than σ_{DC}.

Thickness Dependence of Dielectric Function (Ni)



50 Å not metallic





Ola Hunderi, PRB, 1973

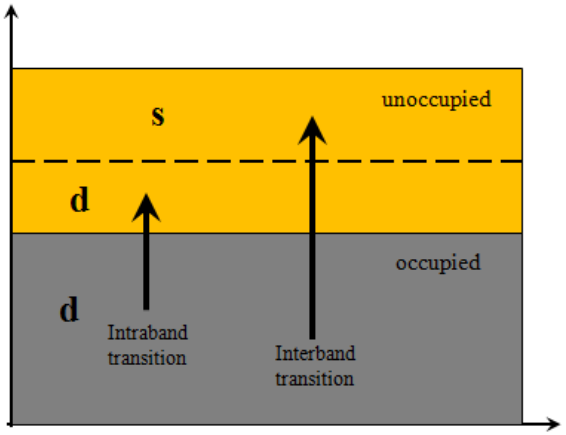
$\sigma_1 \uparrow$ with $t \uparrow$

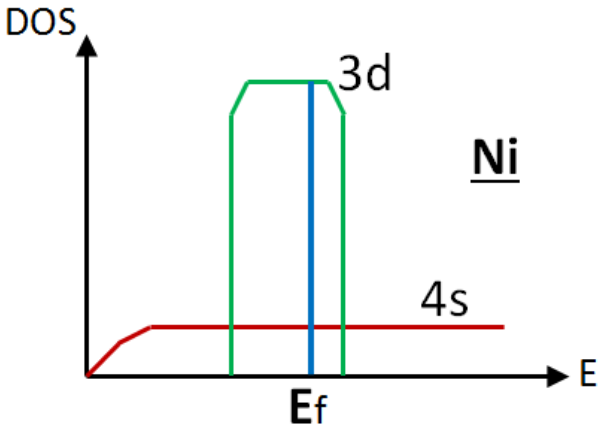
reduced grain boundary scattering in thicker films

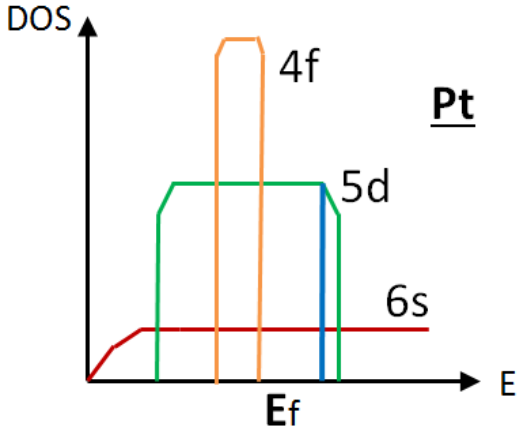
L. Abdallah, AIP Adv. **4**, 017102 (2014)

ture.

Difference between Ni and Pt







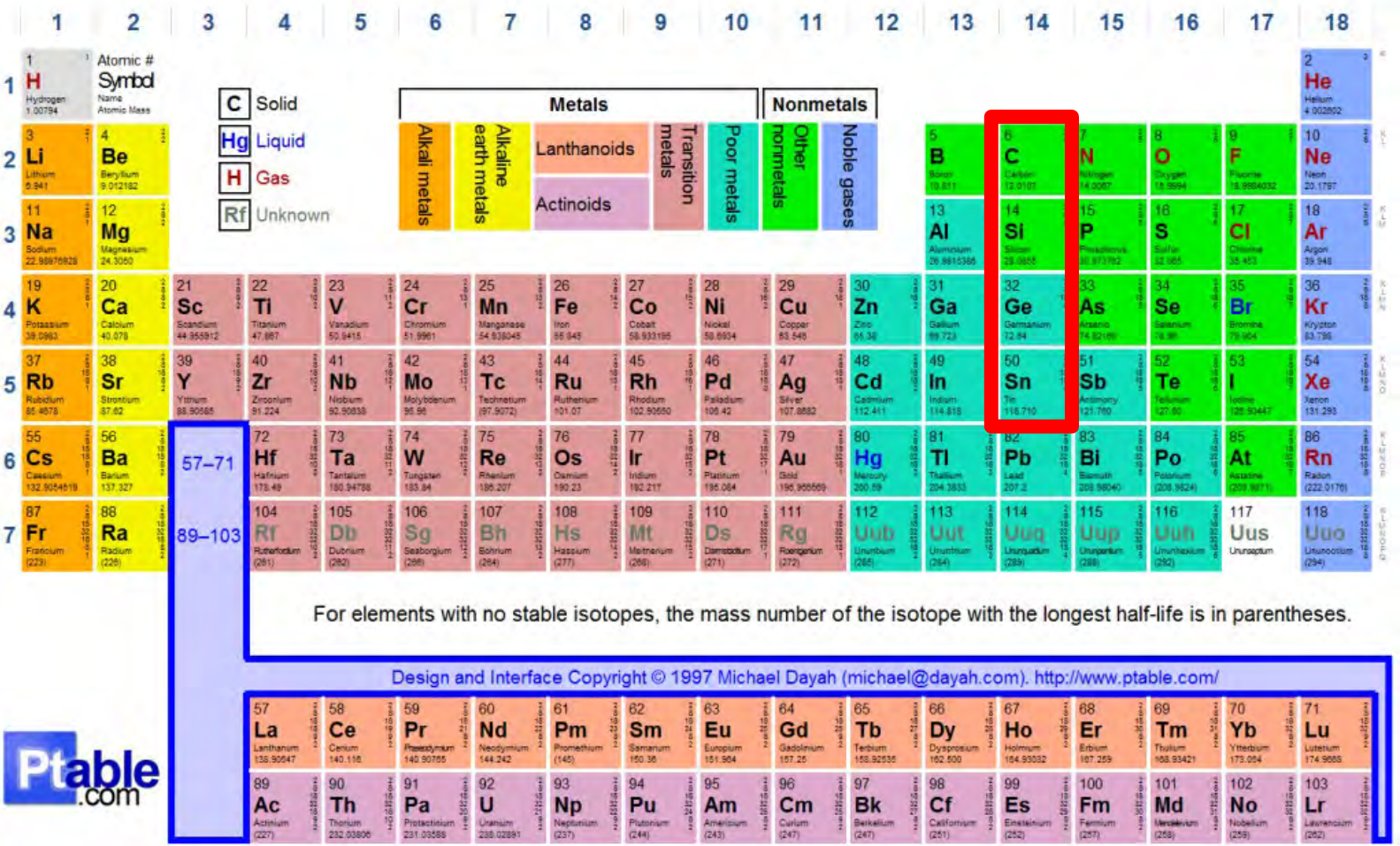
Ni 3d states are more localized.
Pt 5d states are broader, more dispersive.

Ni-Pt alloys have broader transitions than pure Ni.

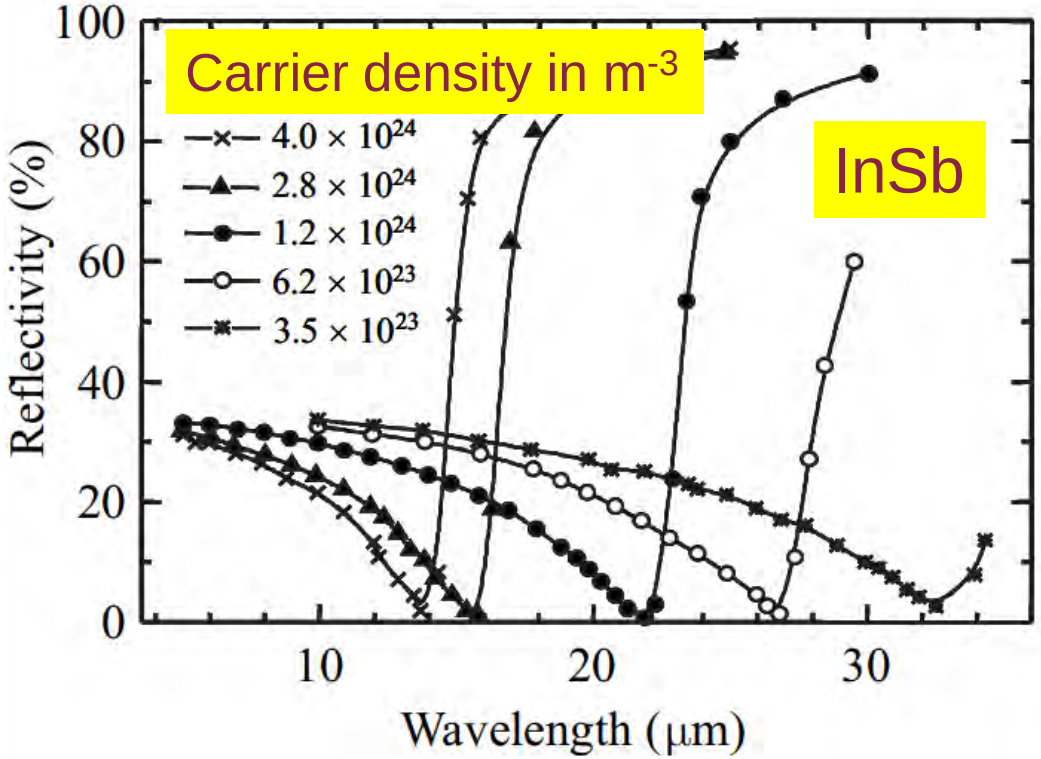
- Alloy broadening: Potential fluctuations
- Initial Pt 5d states broader than Ni 3d states.

L. Abdallah, AIP Adv. **4**, 017102 (2014)

Semiconductors



Free-Carrier Reflection in Doped Semiconductors



Reflectance minimum near plasma frequency

Fox, *Optical Properties of Solids*

Doped semiconductors behave just like a metal, except for the lower carrier density; **plasma frequency in infrared region.**

Why Infrared Ellipsometry ?

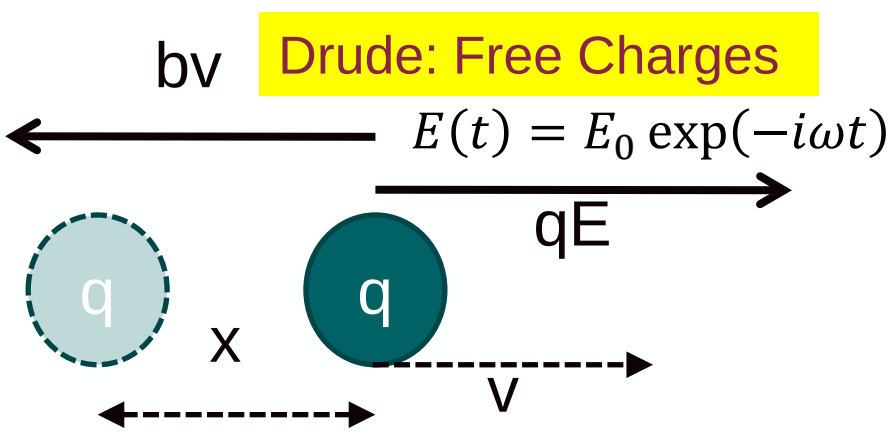
Advantages:

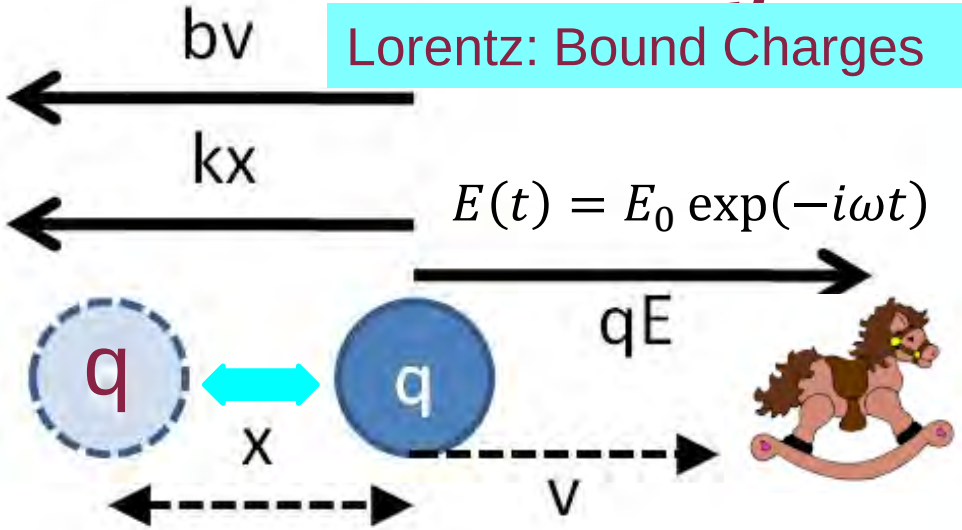
- Measures amplitude ψ and phase Δ.
- Direct access to complex ε (no Kramers-Kronig transform).
- Modeling may contain depth information.
- No need to subtract substrate reference data.
- Anisotropy information (off-diagonal Jones and MM data)
- Possible measurements in a magnetic field (optical Hall effect)
- Obtain plasma frequency and scattering rate (B=0)
- Obtain *carrier density*, scattering rate, *effective mass* (B≠0).

Disadvantages:

- Time-consuming (15 FTIR reflectance spectra)
- Requires polarizing elements (polarizer, compensator)
- Requires **large samples** (no focusing), at least 5 by 10 mm^2
- Requires modeling for thin layer on substrate.
- Commercial instruments **only down to 30 meV (250 cm^{-1})**

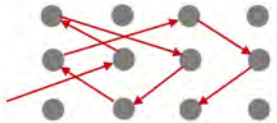
Drude-Lorentz Model: Free and Bound Charges





Discuss plasma frequency trends.

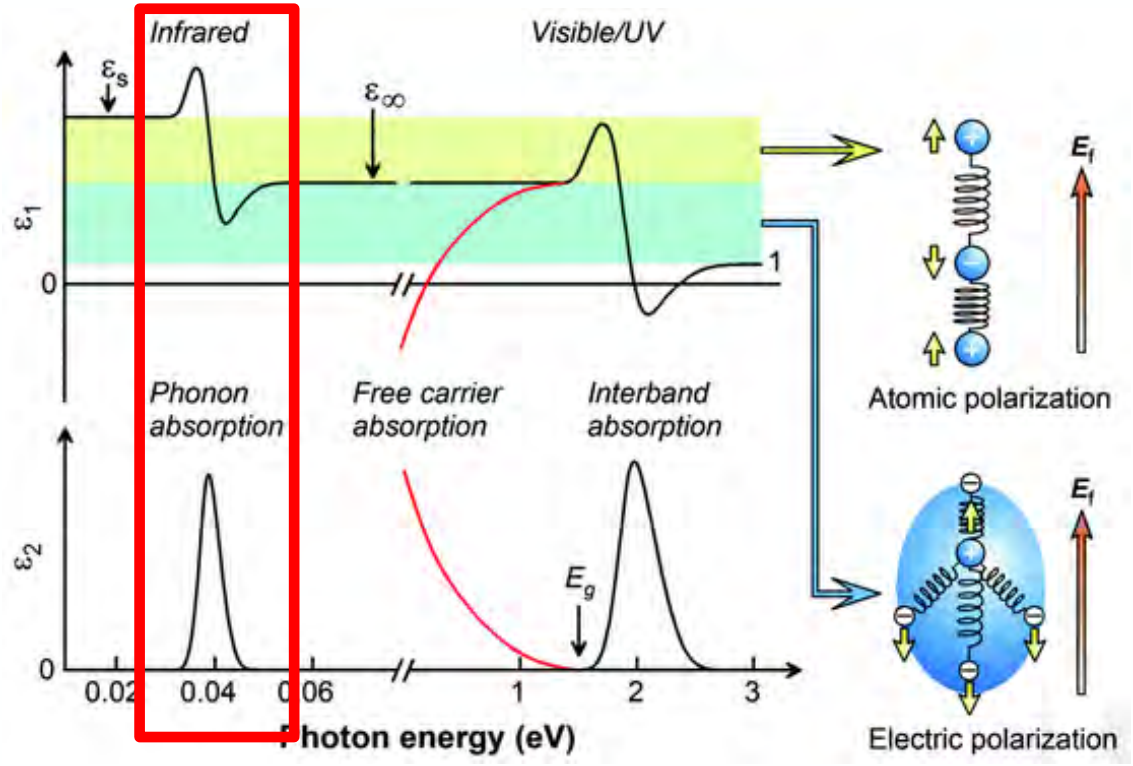
$$\omega_P^2 = \frac{n_f e^2}{m\varepsilon_0}$$



$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

ω_P	(unscreened) **plasma frequency** of free charges
ω_0	**resonance frequency** of bound charges
γ_D, γ_0	**broadenings** of free and bound charges
A	**amplitude** of bound charge oscillations (density, strength)

Drude-Lorentz Model: Free and Bound Charges



Mid-infrared spectral range:

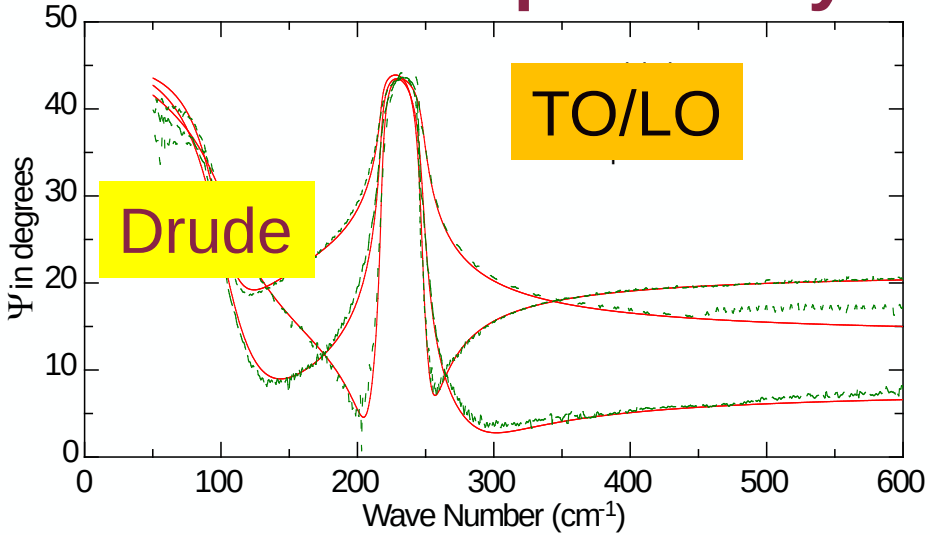
- **Insulator/semiconductor: Lattice vibrations (optical long-wavelength phonons)**
- Metal: Free carrier properties (density, scattering rate)

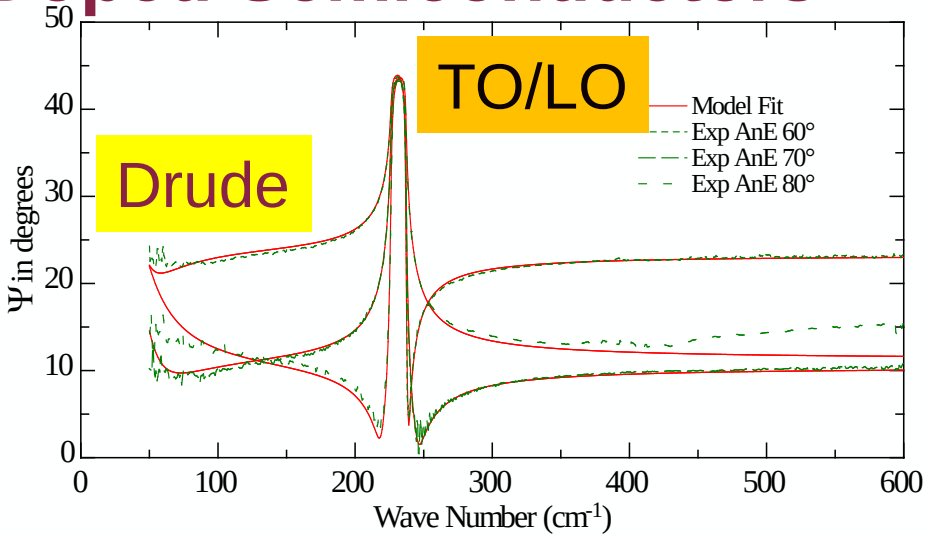
Near-IR to visible to UV range:

- Electronic excitations
- Band gap, interband transitions

$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

Infrared Ellipsometry of Doped Semiconductors





n-type InAs
m=0.027
n=6.3×10^{16} cm^{-3}
γ=50 cm^{-1}
μ=6800 cm^2/Vs

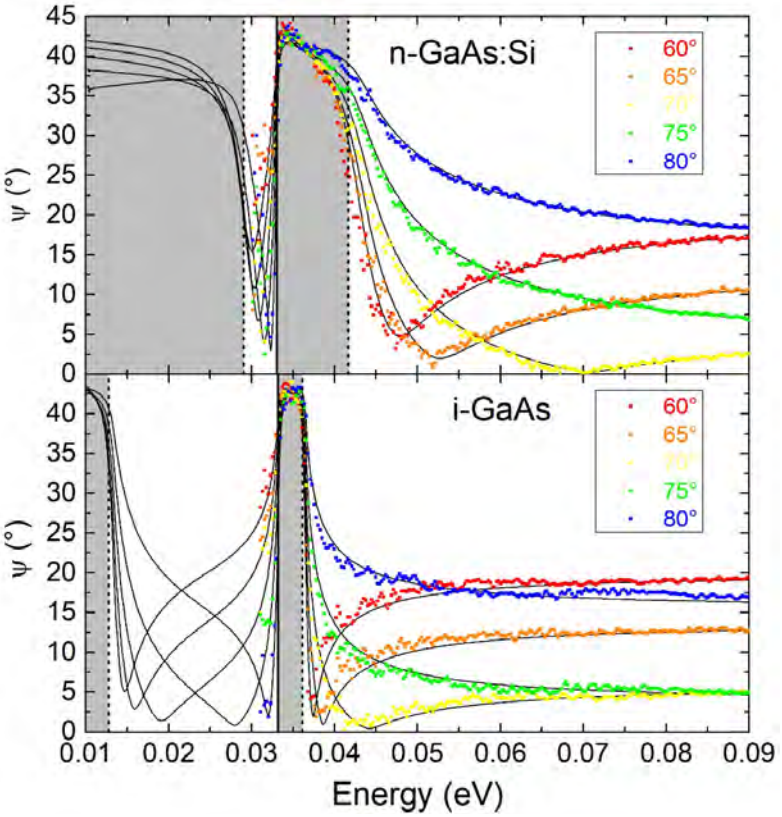
$$\varepsilon(\omega) = 1 - \frac{\omega_P^2}{\omega^2 + i\gamma\omega}$$

n-type GaSb:Te
m=0.13 (?!?)
n=1.3×10^{17} cm^{-3}
γ= 71 cm^{-1}
μ=1000 cm^2/Vs

Doped semiconductors behave just like a metal, except for the lower carrier density; **plasma frequency in infrared region. Only visible for electrons (small mass).**

Coupled Phonon-Plasmon Bands

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



Doped GaAs:

Plasmon effect:

Doping pushes LO phonon to higher energies.
TO phonon not affected by plasma oscillations.

Undoped GaAs:
30 meV lower cut-off insufficient to see Drude term.

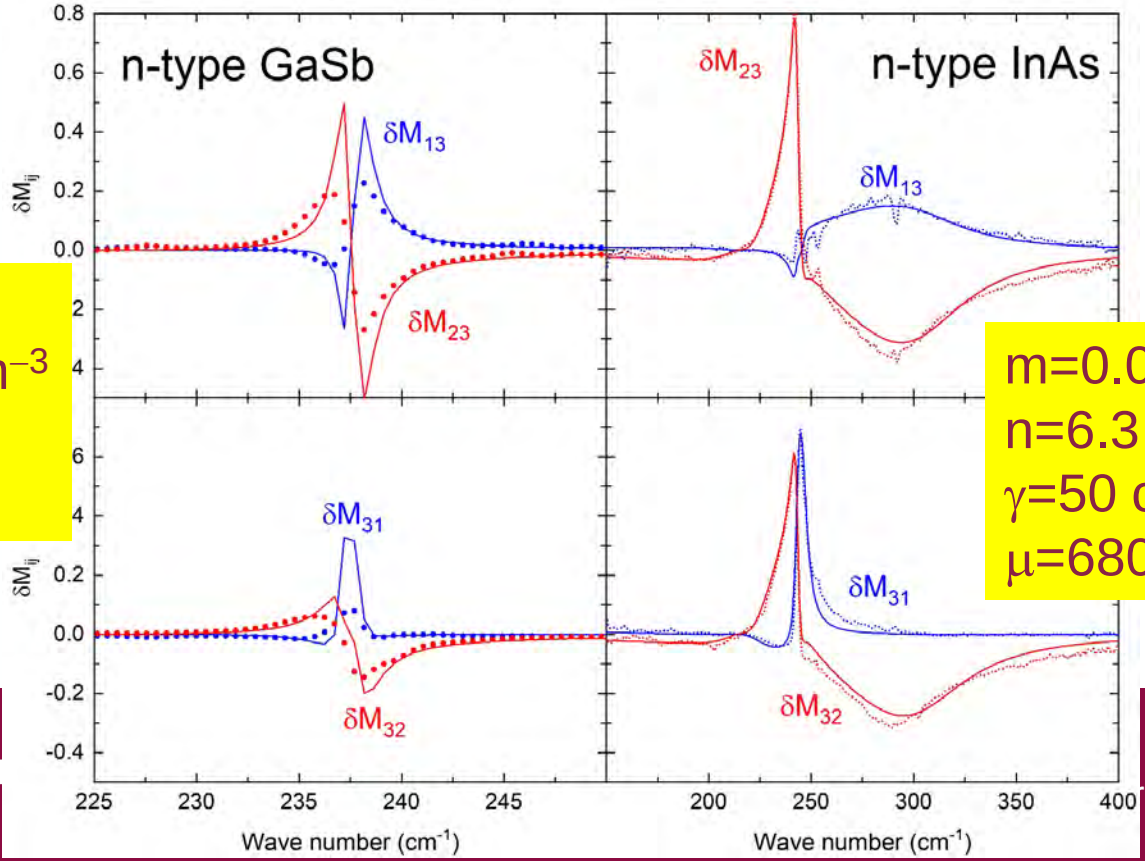
Zollner, J. Vac. Sci. Technol. B **37**, 012904 (2019).
Kukharskii, Fiz. Tverd. Tela **14**, 1744 (1972)
[Sov. Phys. Solid State **14**, 1501 (1972).
Mooradian, Phys. Rev. Lett. **16**, 999 (1966).

Doped semiconductors behave just like a metal, except for the lower carrier density; **plasma frequency in infrared region.**

Optical Hall Effect: Ellipsometry with Magnetic Field

If we measure Mueller matrix spectra in a magnetic field,
we get **carrier concentrations, mobilities, effective masses**.

m=0.13 (?!?)
n=1.3 × 10^{17} cm^{-3}
γ= 71 cm^{-1}
μ=1000 cm^2/Vs

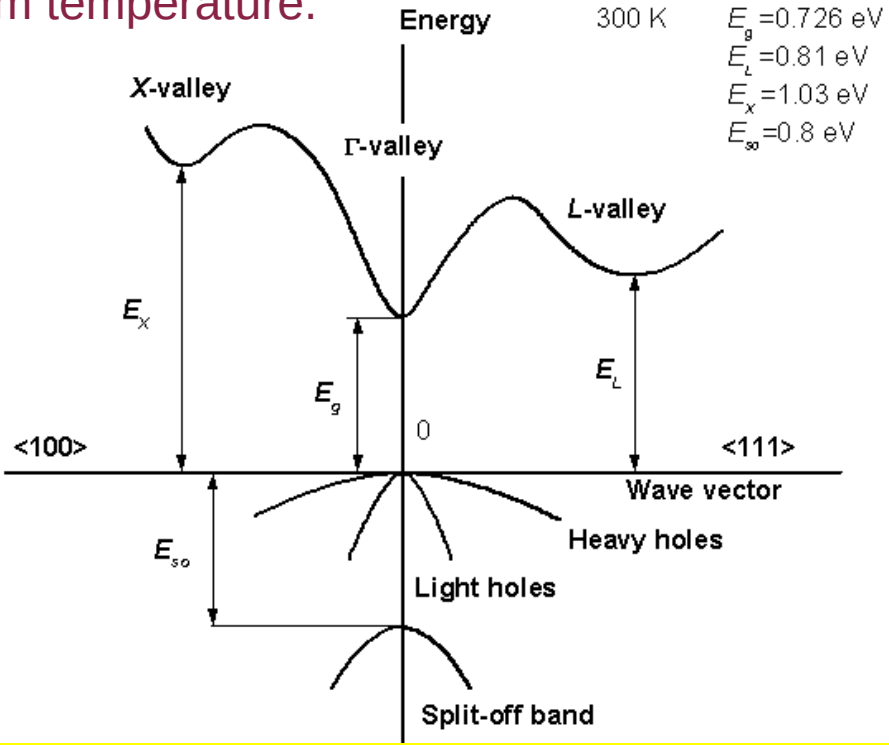


m=0.027
n=6.3 × 10^{16} cm^{-3}
γ=50 cm^{-1}
μ=6800 cm^2/Vs

Abadizaman, Emminger, Knight, Schubert

Multi-Valley Semiconductors

GaSb is a direct semiconductor (like GaAs), but ALMOST indirect. The L-valley in GaSb is only **80 meV** above the Γ-valley. Almost all electrons are in the L-valley at room temperature.



k_BT=26 meV

E_Γ=730 meV

Effective masses

m_Γ=0.041

$m_L^\perp$=0.11

$m_L^{||}$=0.95

4 L-valleys

m_L^{DOS}=0.57

$m_{transport}$=0.15 (later)

We need a model with two carrier species, one of them anisotropic.

Multiple Drude Contributions

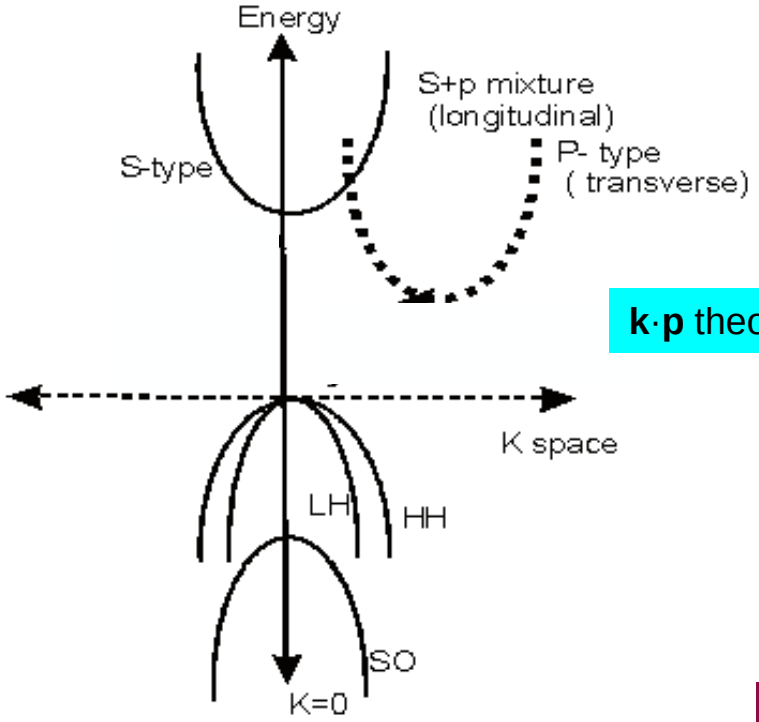
- **Electron mass depends on orbital (s, p, d, f)**
  s: light (small mass m*<<1)
  p: intermediate (m*~0.3 to 1)
  d,f: heavy (large mass m*>>1)
  p,d,f: usually anisotropic

- Electrons and holes

- Different CB minima (Γ,L,X)

- Different VB hole bands (light, heavy, split-off)

CB: Conduction band (empty)

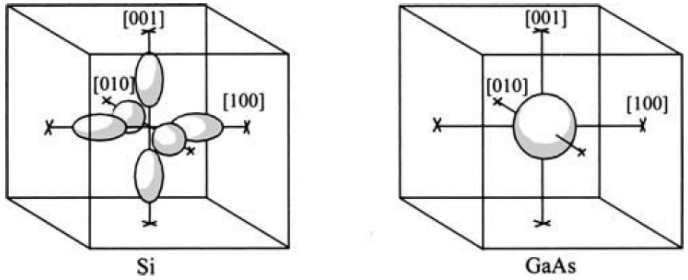


VB: Valence band (filled)

P. Drude, Phys. Z. **1**, 161 (1900).

fan Zoll

Drude Model for Anisotropic Free Carriers



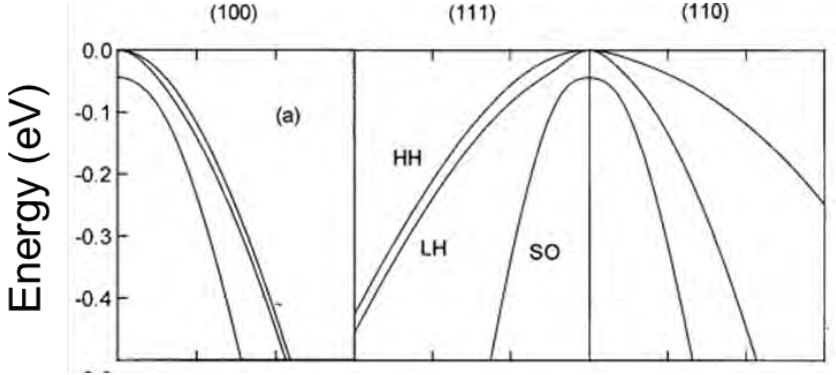
Conduction band minima in Ge and Si are anisotropic. Ge: m_l=1.59, m_t=0.0815.

$$E(\vec{k}) = \frac{\hbar^2 \vec{k}^2}{2m}$$

$$m^{-1} = \frac{1}{\hbar^2}\frac{\partial^2 E(\vec{k})}{\partial \vec{k}^2} = \begin{pmatrix} m_l^{-1} & 0 & 0 \\ 0 & m_t^{-1} & 0 \\ 0 & 0 & m_t^{-1} \end{pmatrix}$$

Δ-valley
Drude mass
Harmonic mean

$$m = \frac{3 m_l m_t}{m_t + 2 m_l}$$



Valence band maxima in semiconductors are warped (Luttinger parameters).

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

$$\omega_P^2 = \frac{nq^2}{m\varepsilon_0}$$

Drude formula still valid, but ε, ω_p^2, m^{-1}, and γ are tensors.

Anisotropic Masses (GaSb L-Valley)

Longitudinal mass at L m_l=0.95

Transverse mass at L m_t=0.11

Density of states mass (geometric mean) m_d=0.57

$$m_d = \sqrt[3]{N_V^2 m_l m_t^2}$$

Drude transport mass (harmonic mean) **m_D=0.15**

$$\frac{3}{m_D} = \frac{1}{m_l} + \frac{2}{m_t}$$

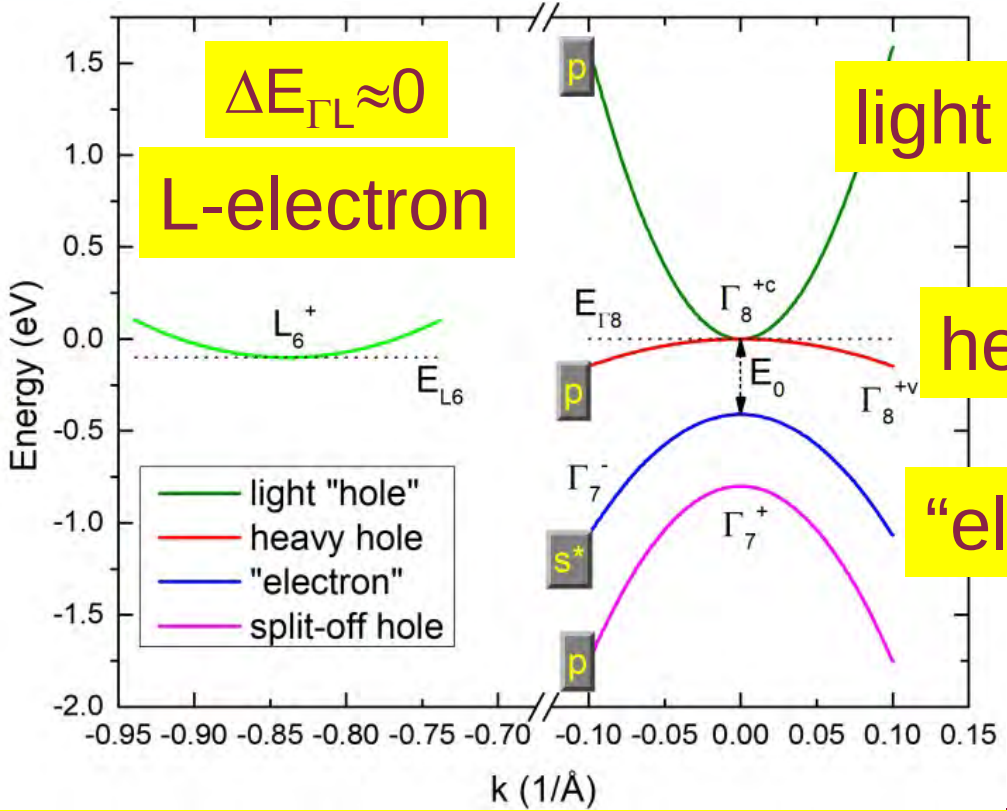
Cyclotron mass

$$m_c = \sqrt{\frac{\det \overleftrightarrow{m}}{\vec{b} \cdot \overleftrightarrow{m} \cdot \vec{b}}}$$

Optical Hall effect measurements on anisotropic materials are sparse. Need measurements on bulk Si, Ge, GaP, SiC with different orientations (direction of magnetic field ***b***).

Dresselhaus, Dresselhaus, Cronin, Gomes, Solid-State Properties (10.50)

Grey α-tin is Even More Complicated



light "hole"

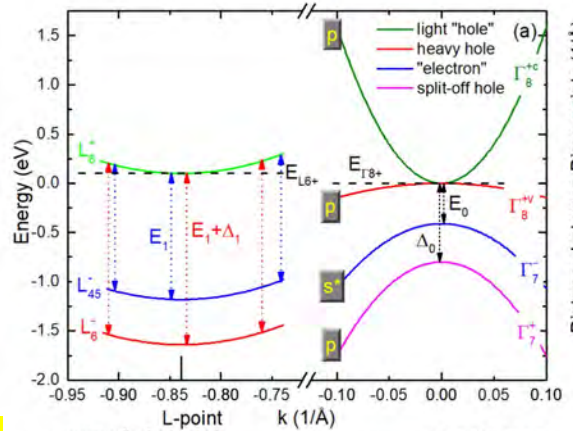
heavy hole

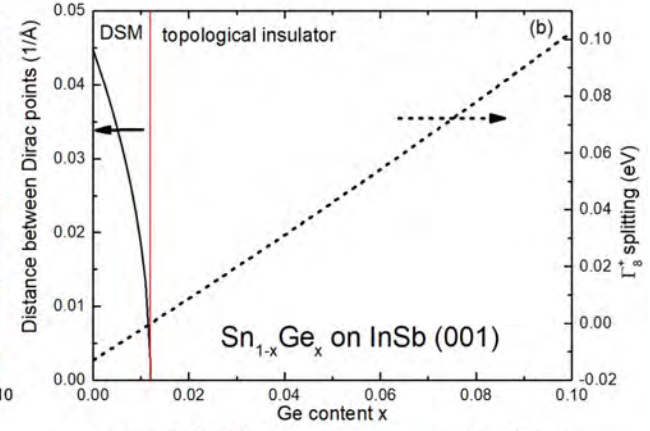
"electron"

C.A. Hoffman, PRB **40**, 11693 (1989).
R.A. Carrasco, APL **113**, 232104 (2018).

We need a model with THREE carrier species, one of them anisotropic.

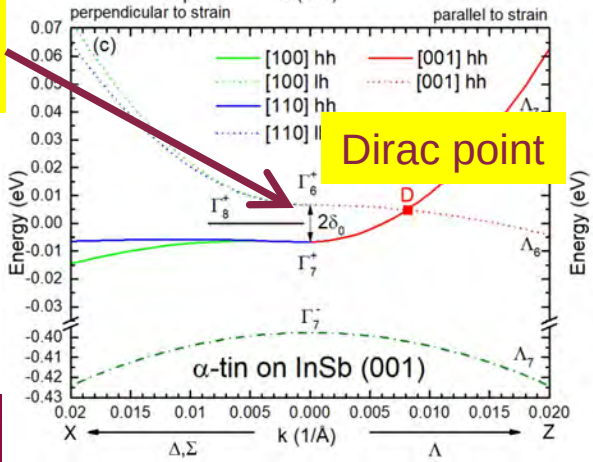
Add Strain and Warping in $Sn_{1-x}Ge_x$ Alloys





12 meV splitting (97 cm^{-1})

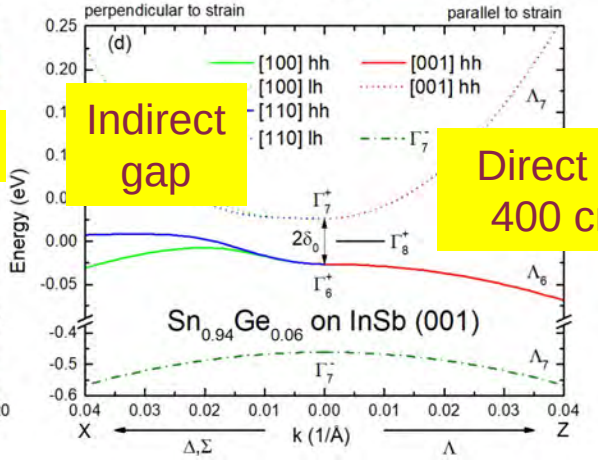
Warping



Dirac point

Indirect gap

Direct gap 400 cm^{-1}



R.A. Carrasco, APL **114**, 062102 (2019).

How do we model Drude response of free carriers with realistic band structures?

Insulators

Transparent region below the band gap (VIS/UV).

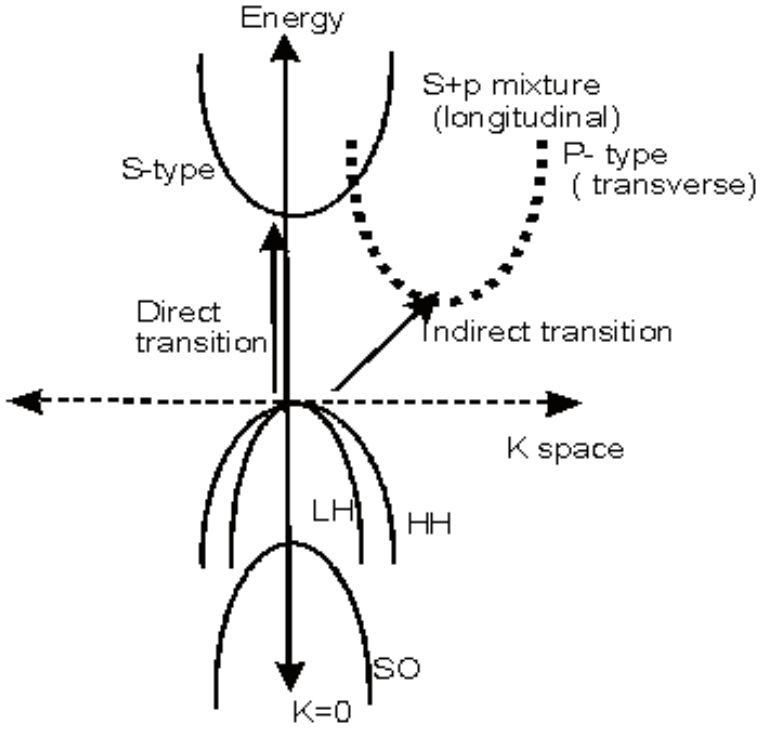
Forbidden “energy gap”.

Semiconductors are insulators with small band gap (IR).

Valence band filled.
Conduction band empty.

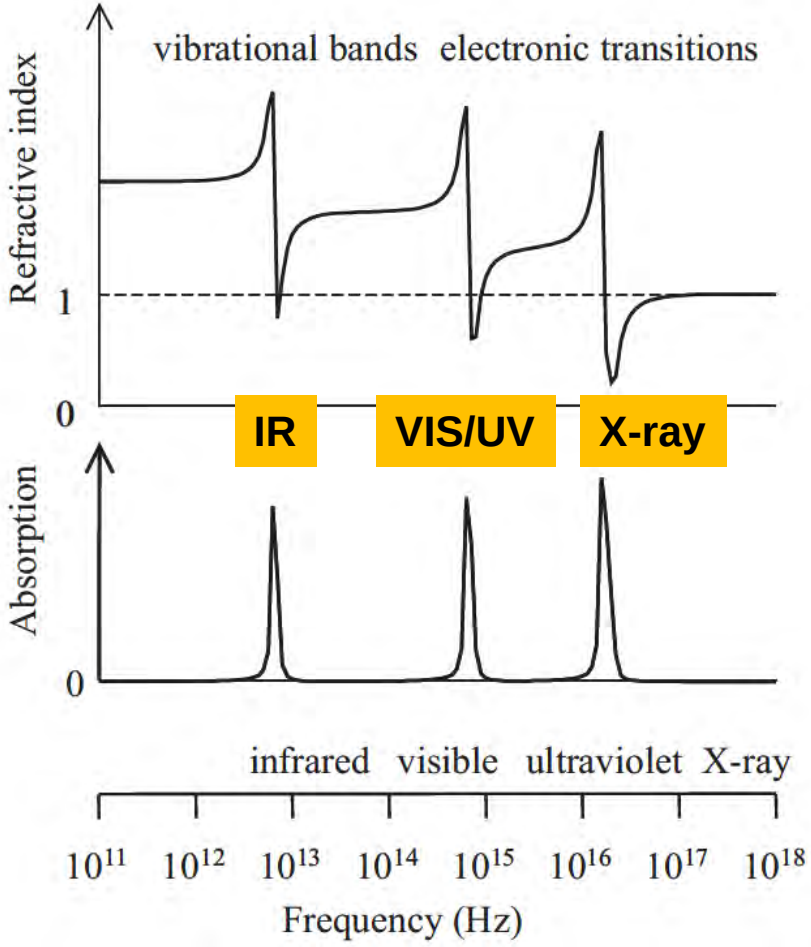
Filled bands do not carry current (no absorption).

CB: Conduction band (empty)



VB: Valence band (filled)

Multiple Lorentz Contributions: IR, UV, x-ray



IR: lattice vibrations

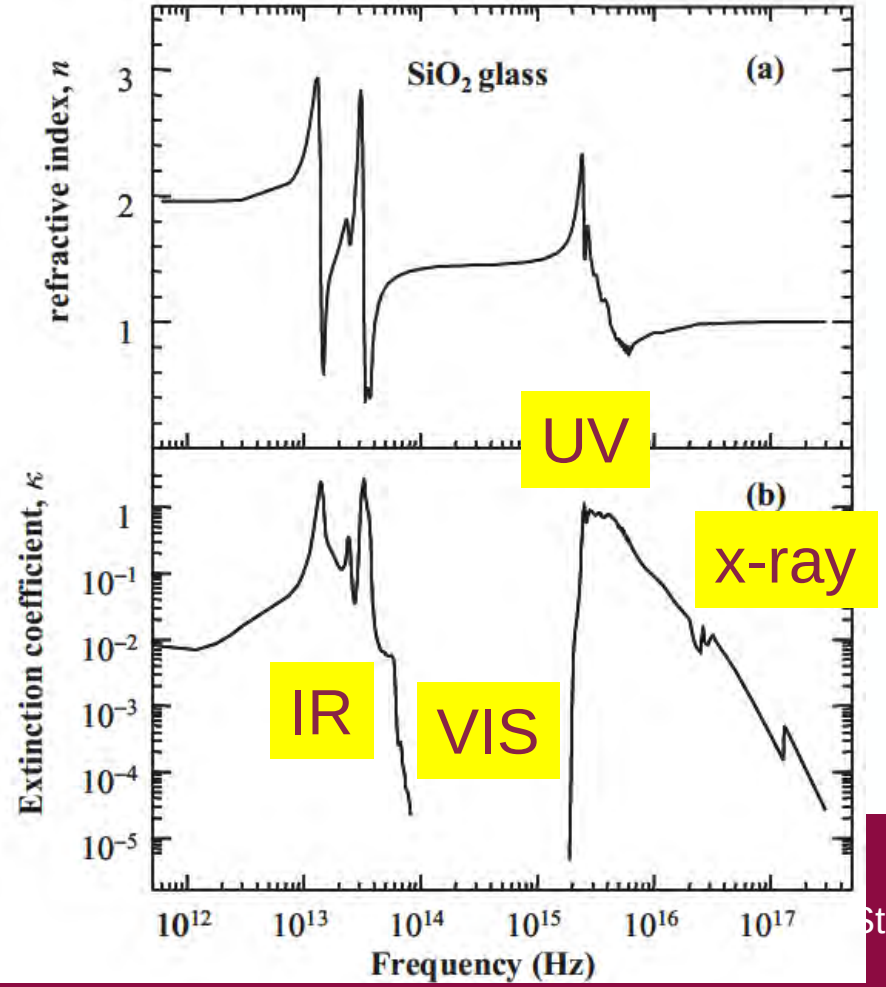
VIS/UV: valence electrons
(broadened by band structure effects)

x-ray: core electrons

Amplitude depends on
- Density of oscillators
- Matrix elements
- Born effective charge

Fox, Fig. 2.6

Multiple Lorentz Contributions: SiO_2 as an Example



IR: lattice vibrations
(Si-O bend, stretch)

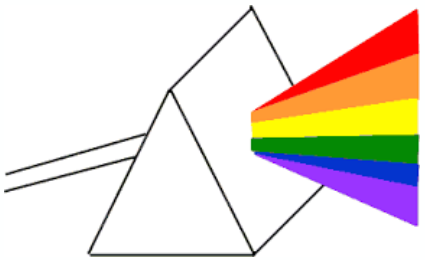
VIS: Nothing happens

UV: valence electrons
(interband transitions)

x-ray: core electrons
(absorption edges)

Amplitude depends on
- Density of oscillators
- Matrix elements
- Born effective charge

Sellmeier Approximation (Poles)



Set γ=0 **far from resonance**.
Lorentz oscillator becomes a **pole (Woollam SW)**

$$\varepsilon(\omega) = 1 + \frac{A\omega_0^2}{\omega_0^2 - \omega^2}$$

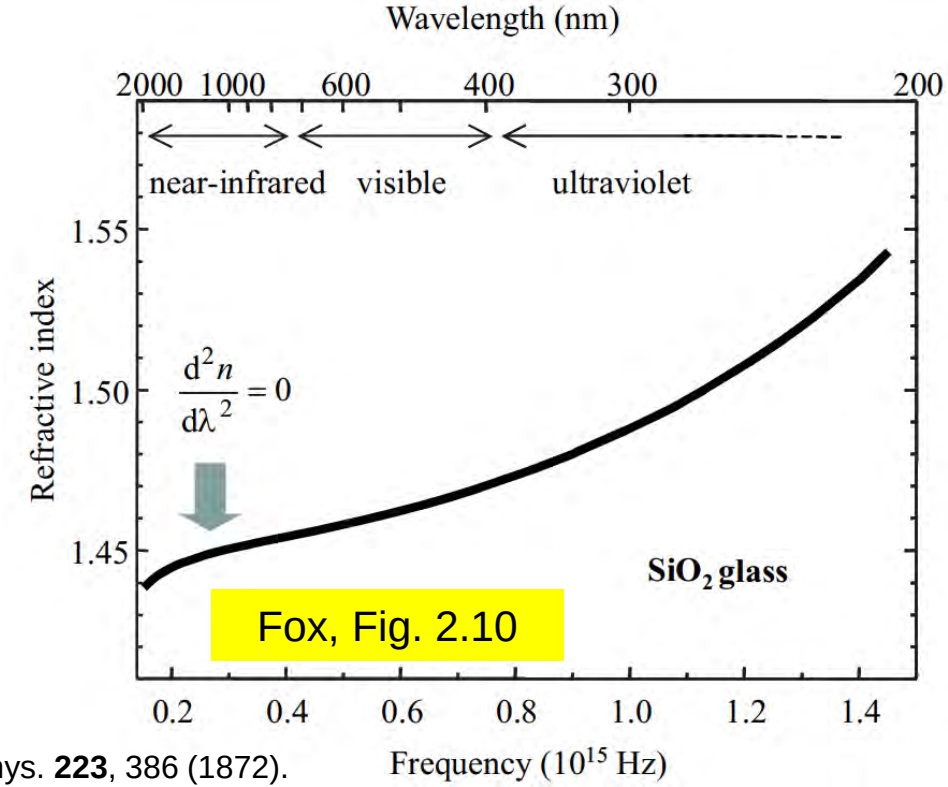
Rewrite as a function of λ

$$\varepsilon(\lambda) = 1 + \frac{B\lambda^2}{\lambda^2 - C}$$

Several Lorentz oscillators
(one in IR, two in UV)

$$\varepsilon(\lambda) = 1 + \sum_i \frac{B_i\lambda^2}{\lambda^2 - C_i}$$

Sellmeier approximation.



Fox, Fig. 2.10

W. Sellmeier, Ann. Phys. **223**, 386 (1872).

Cauchy Equation (Urbach Tail)

The Cauchy equation

$$n(\lambda) = \sqrt{\varepsilon(\lambda)} = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4}$$

can be viewed as a Laurent series expansion of the Sellmeier equation

$$n(\lambda) = \sqrt{\varepsilon(\lambda)} = \sqrt{1 + \frac{B\lambda^2}{\lambda^2 - C}}$$

Comments:

The Cauchy equation does not include absorption.

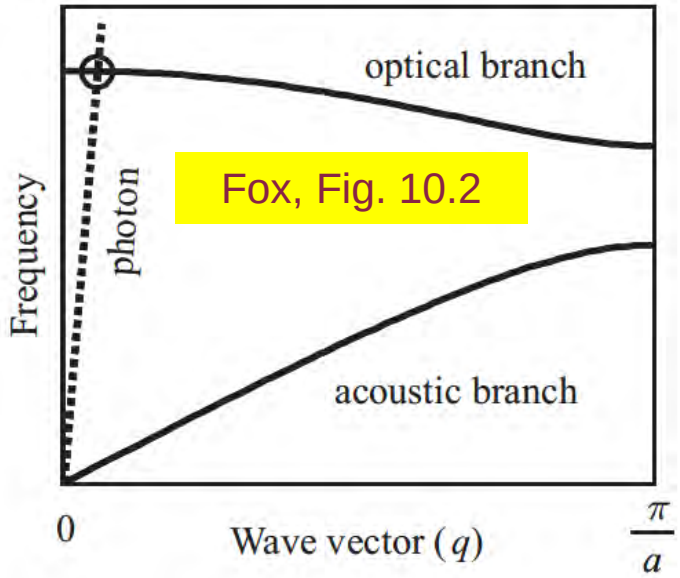
Absorption is often included with an **Urbach tail**

$$k(E) = \alpha e^{\beta(E-\gamma)}$$

but this does not make it Kramers-Kronig consistent.

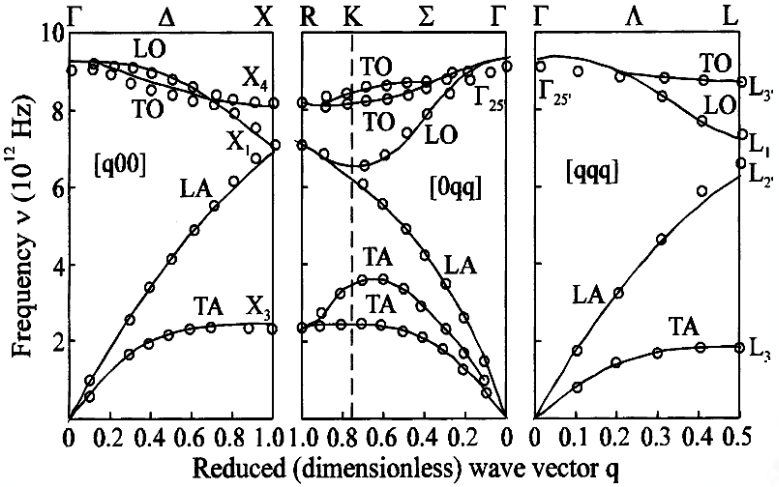
Not recommended, use Tauc-Lorentz model instead.

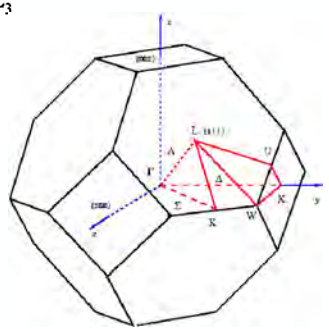
Insulator Phonon Spectra (Ge)



Fox, Fig. 10.2

Inelastic neutron scattering

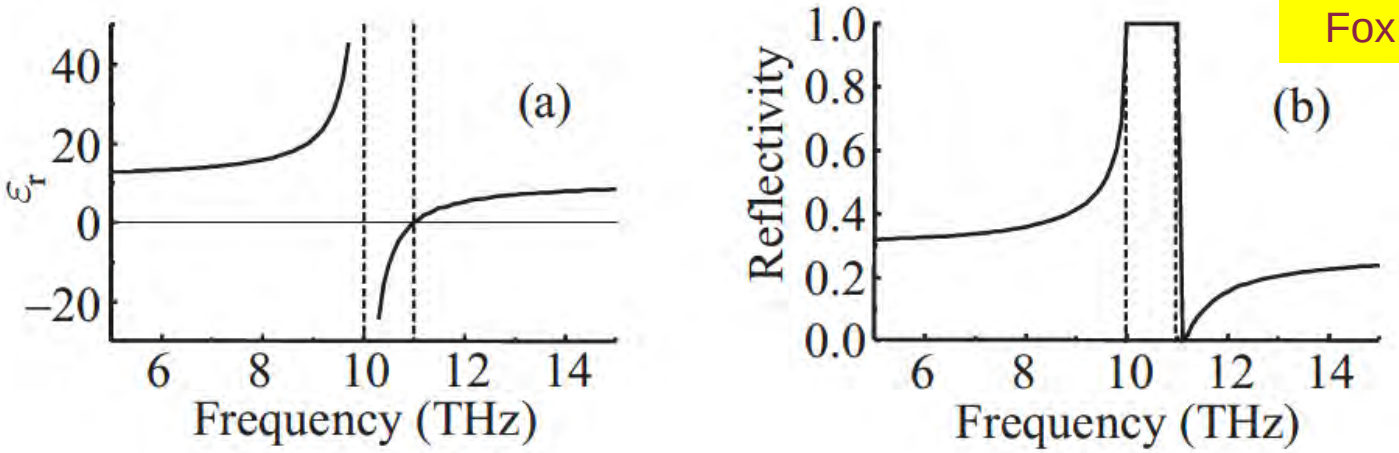




Inelastic neutron scattering measures entire phonon dispersion.
Optical experiments only probe k≈0, **because λ>>a**.
C, Si, Ge, α-Sn are IR-inactive (no dipole moment)

Infrared Lattice Vibrations (Lorentz model)

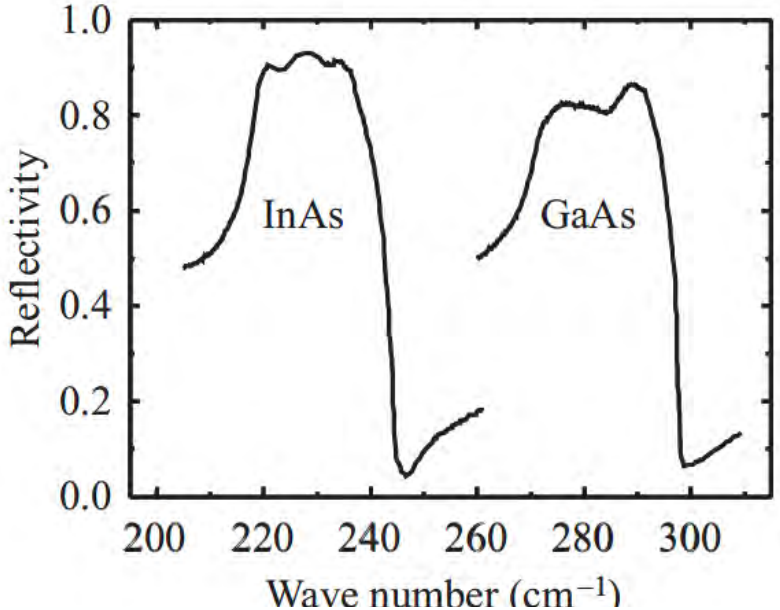
Fox, Fig. 10.4



$$\varepsilon(\omega) = \varepsilon_\infty + \sum_i \frac{A_i \omega_{TO,i}^2}{\omega_{TO,i}^2 - \omega^2 - i\gamma_{TO,i}\omega}$$

In **polar materials** (Born effective charge), TO and LO modes are split.
TO: transverse optical (displacement perpendicular to **k**)
LO: longitudinal optical (displacement along **k**)
ε_2 has peak at TO frequency
ε_1 is negative from TO to LO frequency (reflectance is 1)

Infrared Lattice Vibrations (Lorentz model)



Reststrahlen Band

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{A\omega_0^2}{\omega_{TO}^2 - \omega^2 - i\gamma_{TO}\omega}$$

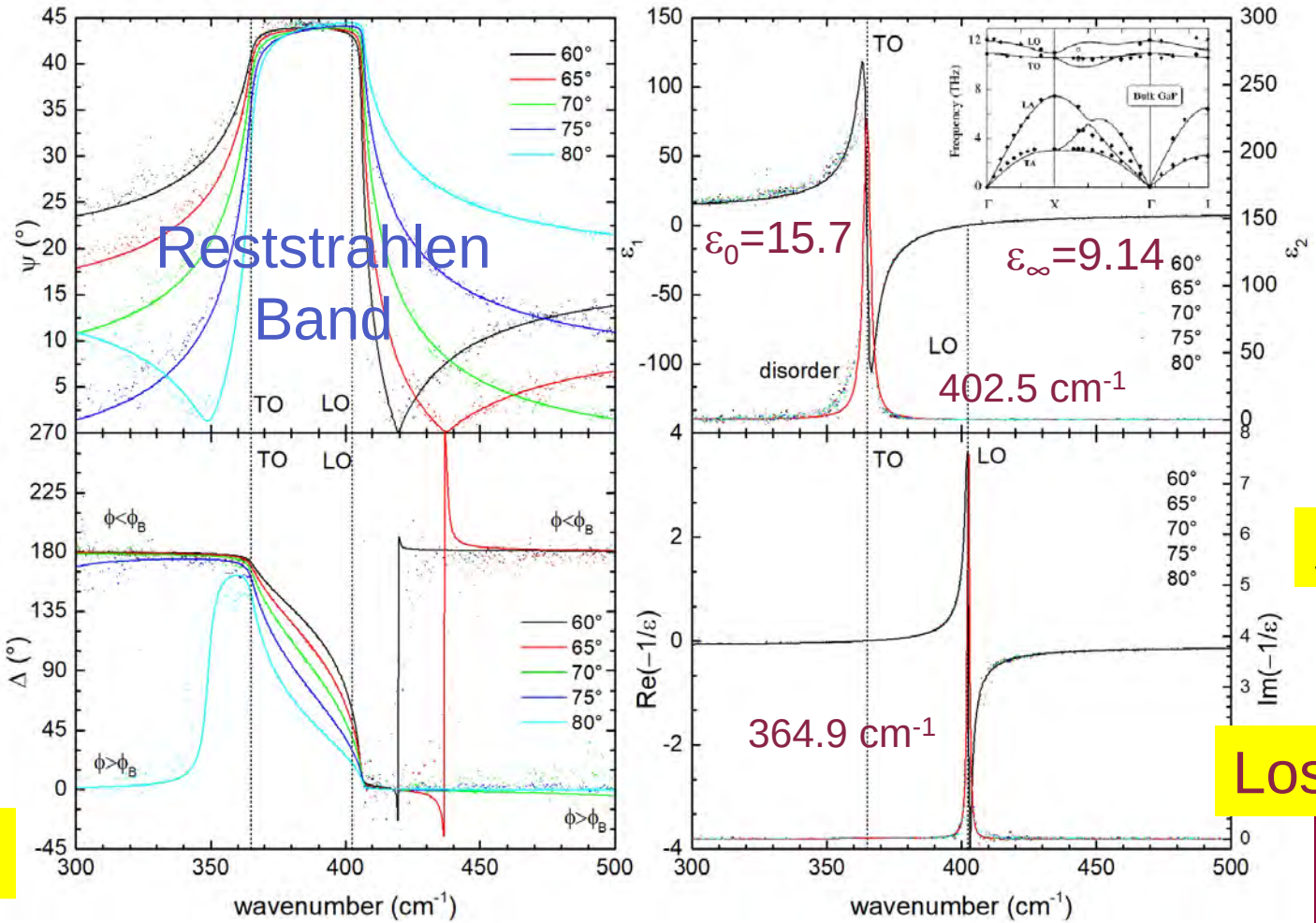
In polar materials (Born effective charge), TO and LO modes are split.
TO: transverse optical (atomic displacement perpendicular to **k**)
LO: longitudinal optical (atomic displacement along **k**)
ε_2 has peak at TO frequency
ε_1 is negative from TO to LO frequency (reflectance is 1)
Restrahlen band extends from TO to LO energy.

Fox, Fig. 10.5

Infrared Lattice Vibrations in GaP

psi

epsilon

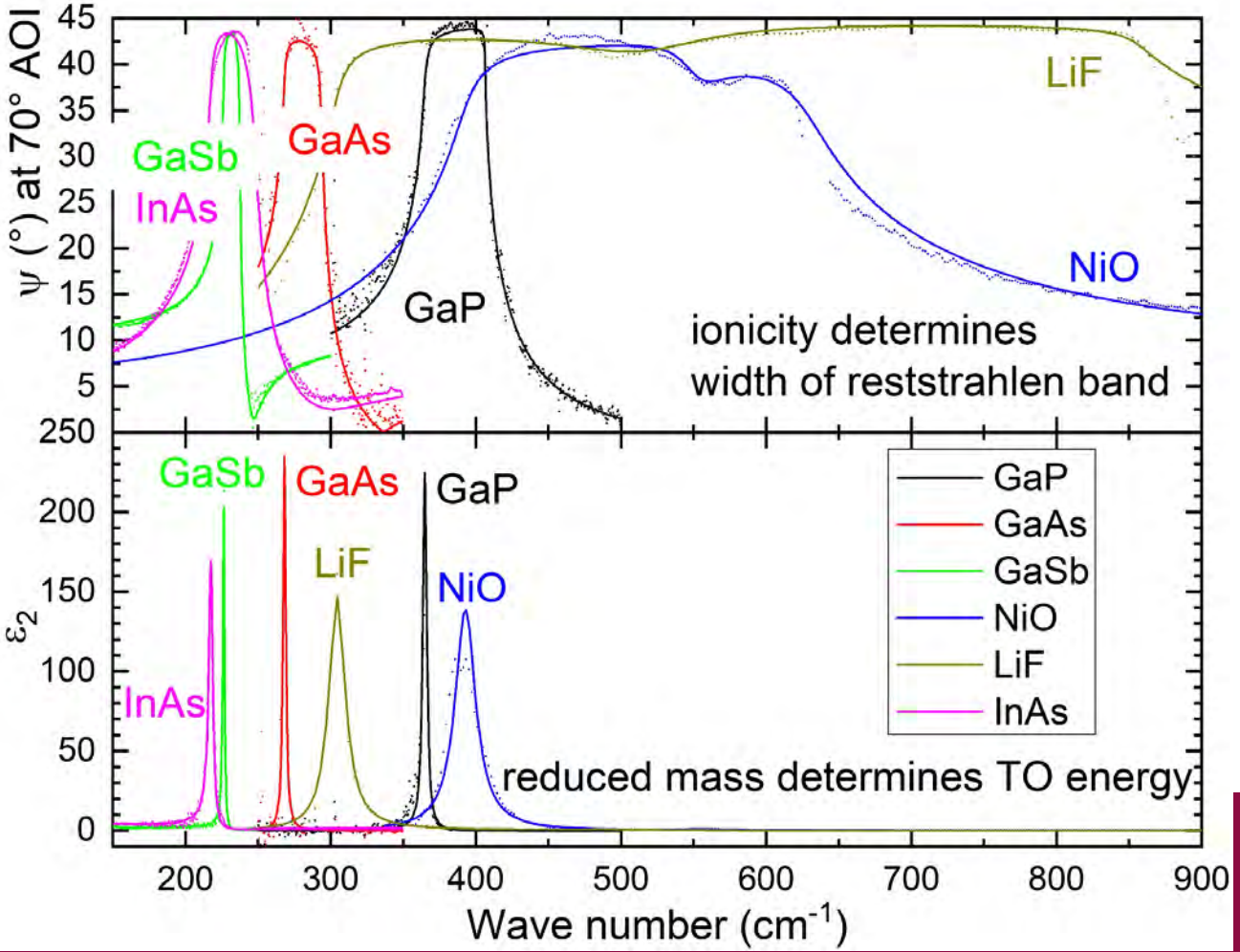


N. Samarasingha, JVSTB **39**, 052201 (2021)

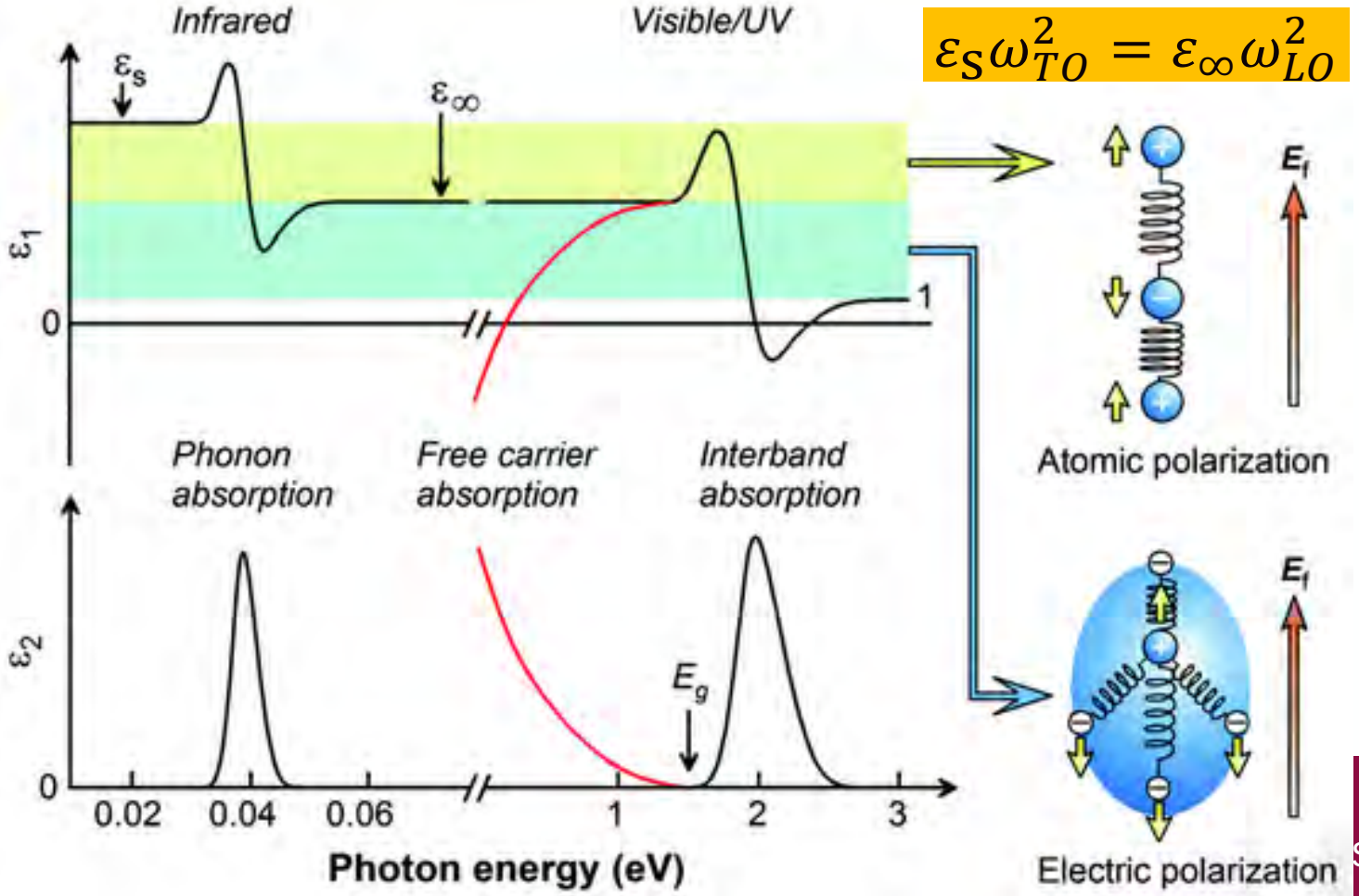
Delta

Loss function

Infrared Lattice Vibrations (Lorentz model)



Lyddane Sachs Teller Relation (Lorentz Model)



$$\varepsilon_S \omega_{TO}^2 = \varepsilon_\infty \omega_{LO}^2$$

LST relation

Lyddane, Sachs, Teller, Phys. Rev. **59**, 673 (1941)

Lyddane Sachs Teller Relation (Lorentz Model)

Lorentz model (γ=0) for one TO/LO phonon mode

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{A\omega_{TO}^2}{\omega_{TO}^2 - \omega^2}$$

At zero frequency (ω=0) define static dielectric constant ε_s

$$\varepsilon_S = \varepsilon(\omega = 0) = \varepsilon_\infty + A$$

Define ω_{LO} through $\varepsilon(\omega_{LO})$=0
(Longitudinal modes require ε=0).

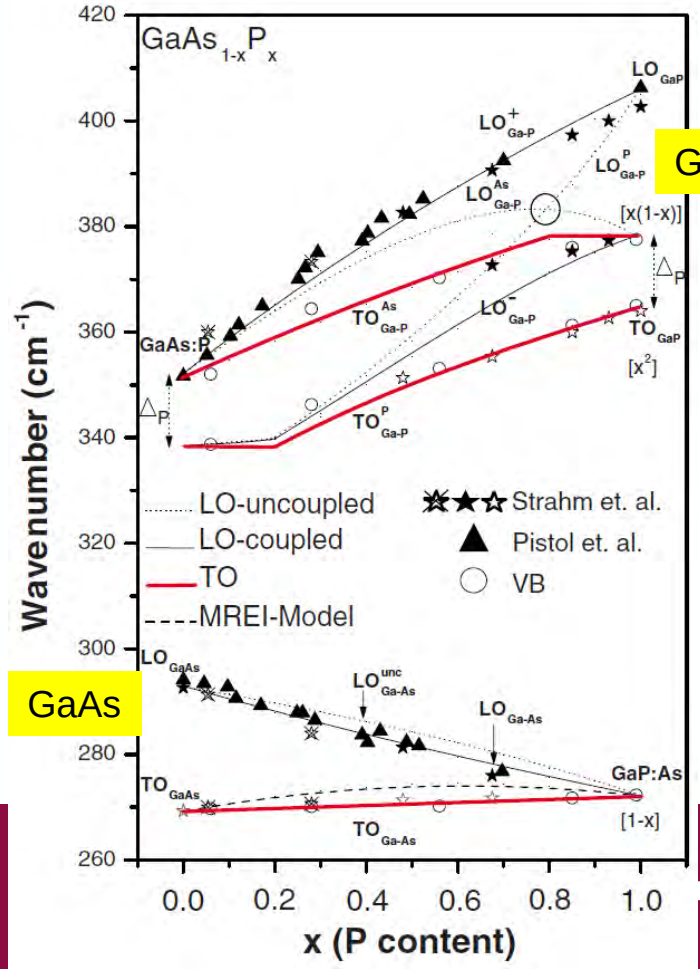
$$0 = \varepsilon_\infty + \frac{A\omega_{TO}^2}{\omega_{TO}^2 - \omega_{LO}^2}$$

$$A = \varepsilon_\infty \frac{\omega_{LO}^2 - \omega_{TO}^2}{\omega_{TO}^2}$$

or $\varepsilon_S \omega_{TO}^2 = \varepsilon_\infty \omega_{LO}^2$ LST relation

Lyddane, Sachs, Teller, Phys. Rev. **59**, 673 (1941)

Optical Phonons in Semiconductor Alloys

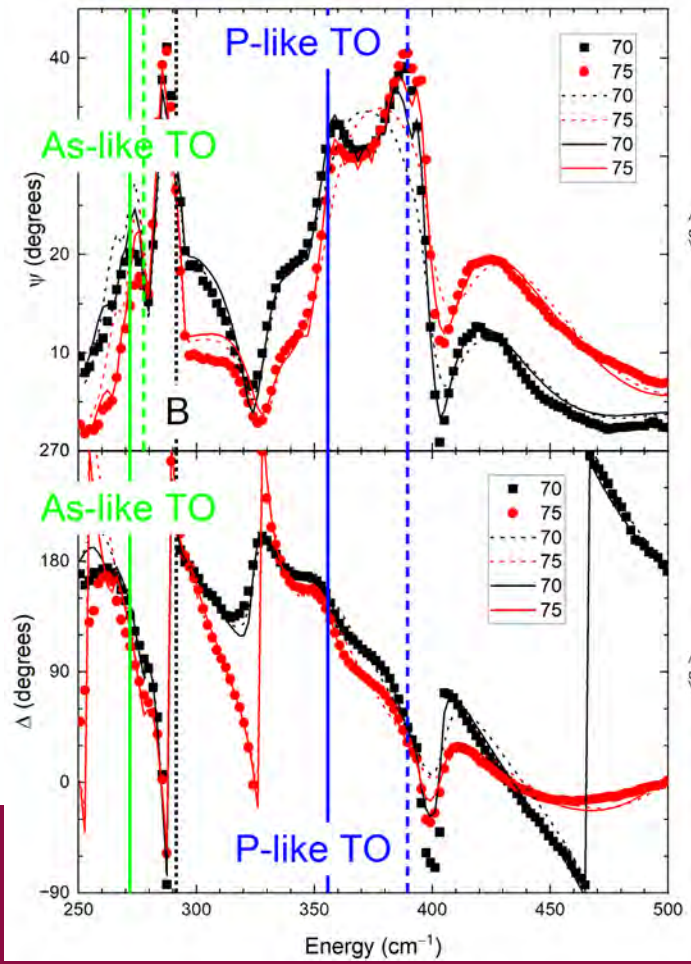


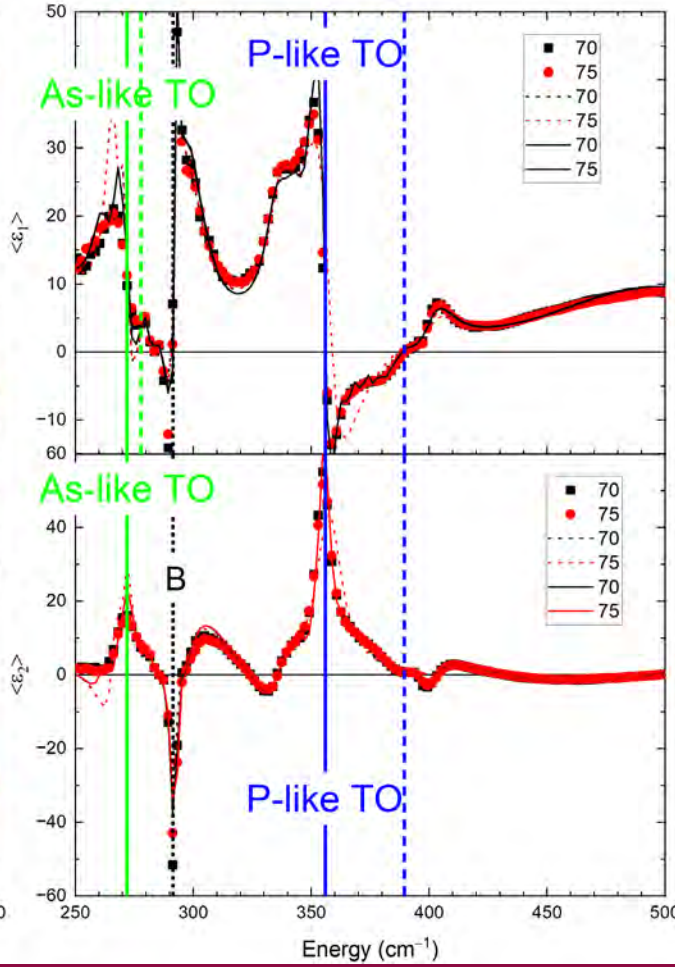
Two-mode behavior:

- GaP and GaAs reststrahlen bands do not overlap.
- P cannot vibrate in GaAs host.
- As cannot vibrate in GaP host.
- **Outcome:**
  In a GaAsP alloy, there are two phonon bands:
  GaAs-like (low frequency reststrahlen band)
  GaP-like (high-frequency reststrahlen band)
- The width of these phonon bands decreases near the binary endpoints.
- The TO amplitude is related to the TO/LO splitting.
- Additional modes appear, which makes the phonon spectra very complex.

L. Genzel, phys. stat. sol. **62**, 83 (1974).
O. Pages, PRB **80**, 035204 (2009).

Optical Phonons in GaAsP Alloys (on GaAs)

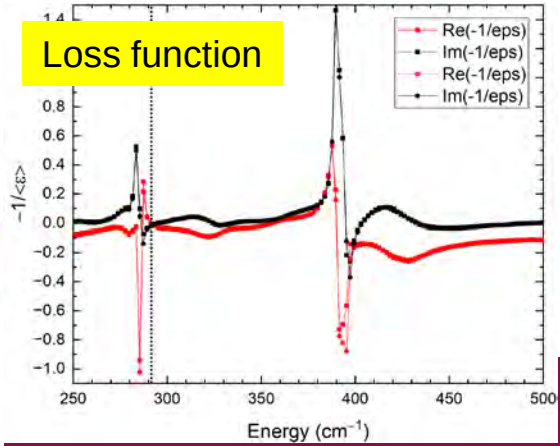




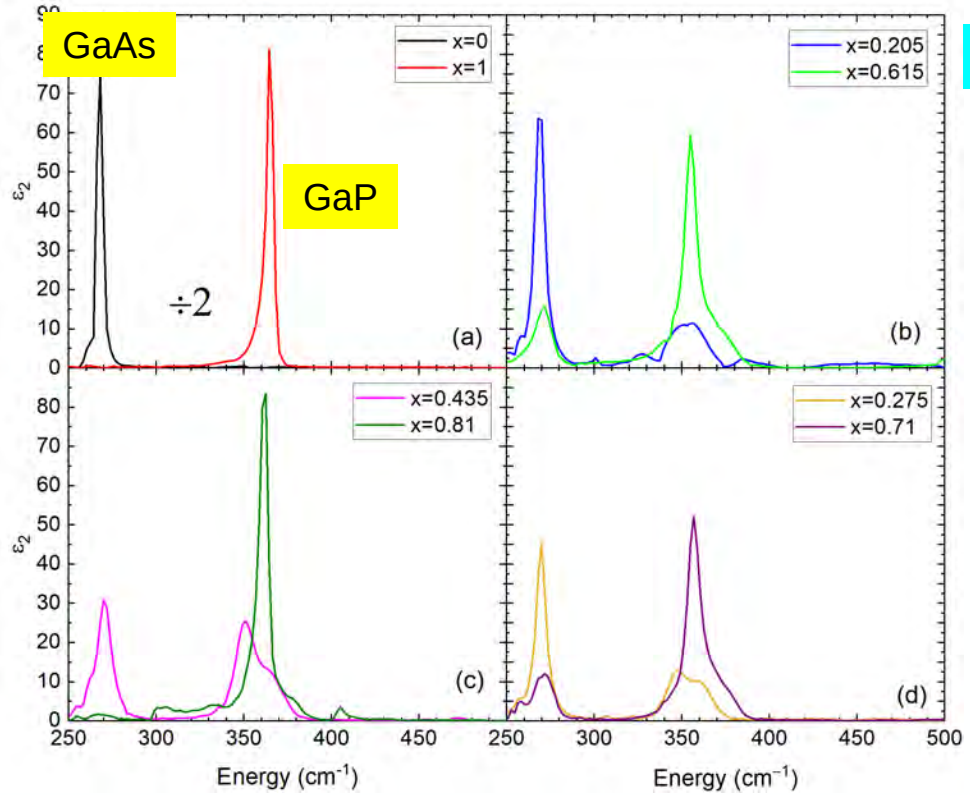
TO: solid
LO: dashed

Note Berreman mode of the GaAs substrate (dotted).

TO: peaks of $<\varepsilon_2>$.
LO: peaks of loss function



Optical Phonons in GaAsP Alloys (on GaAs)



x: P content

TO: peaks of $<\varepsilon_2>$.
LO: peaks of loss function

In a GaAsP alloy, there are two phonon bands:
GaAs-like (low frequency reststrahlen band)
GaP-like (high-frequency reststrahlen band)
Amplitude scales with composition.

Additional weak modes (side bands).

Verleur & Barker, PR **149**, 715 (1966).
J. Cebulski, PSSB **250**, 1614 (2013).

Generalized Lyddane Sachs Teller Relations

Multiple phonon modes (isotropic)

$$\frac{\varepsilon_S}{\varepsilon_\infty} = \prod_i \frac{\omega_{LO,i}^2}{\omega_{TO,i}^2}$$

Kurosawa, J. Phys. Soc. Jpn. **16**, 1298 (1961)

Anisotropic crystals

$$\frac{\det(\varepsilon_S)}{\det(\varepsilon_\infty)} = \prod_i \frac{\omega_{LO,i}^2}{\omega_{TO,i}^2}$$

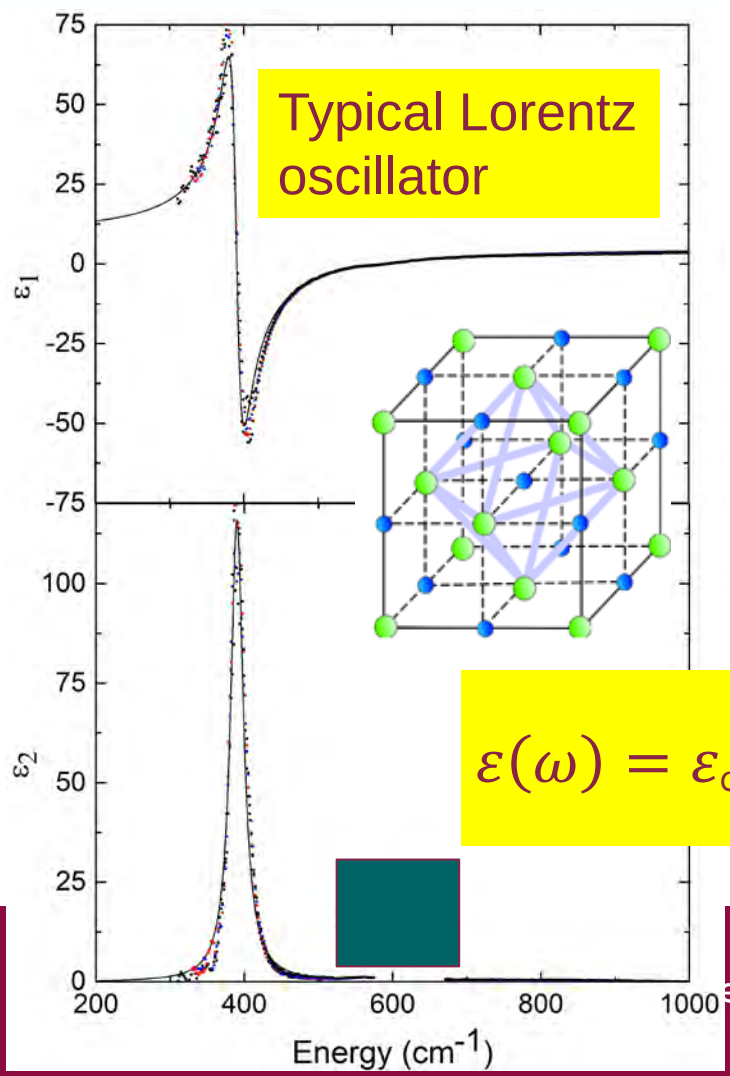
Mathias Schubert, Phys. Rev. Lett. **117**, 215502 (2016)

Amorphous materials and liquids

$$\frac{\varepsilon_S}{\varepsilon_\infty} = \frac{\langle \omega^2 \rangle_l}{\langle \omega^2 \rangle_t}$$

A.J. Sievers and J.B. Page, Infrared Physics **32**, 425 (1991)

Infrared Lattice Vibrations (NiO)



NiO or NaCl or LiF: Rocksalt lattice

- Ni^{2+}-O^{2-} bonds are polar.
- Ni-O vibration has dipole moment.

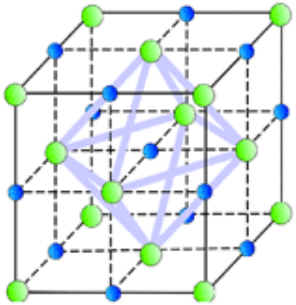
$$\varepsilon(\omega) = \varepsilon_\infty + \frac{A\omega_0^2}{\omega_0^2 - \omega^2 - i\gamma_0\omega}$$

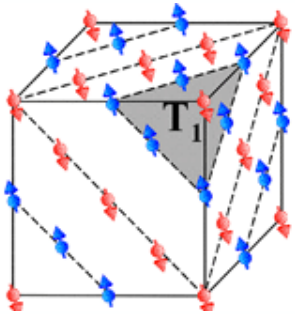
FTIR ellipsometry

Infrared Lattice Vibrations (NiO)

- Rocksalt Crystal Structure (FCC), Space Group 225 (Fm-3m).
- Single TO/LO phonon pair.
- Antiferromagnetic ordering along (111), should cause phonon splitting (8-30 cm^{-1})
- **Second-order phonon absorption.**

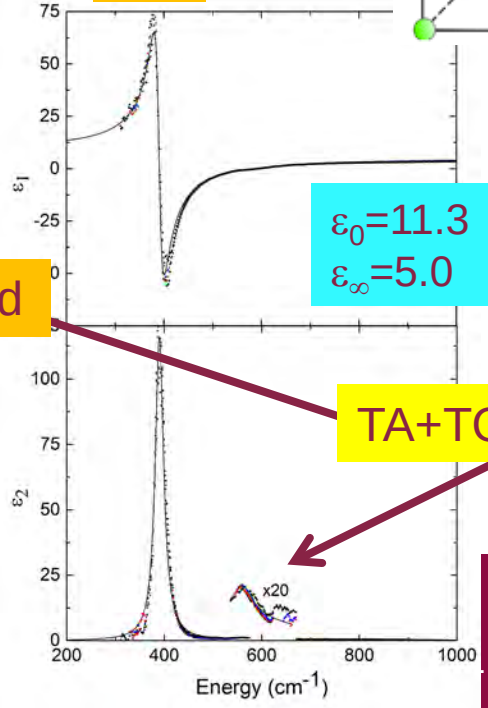
NiO cell



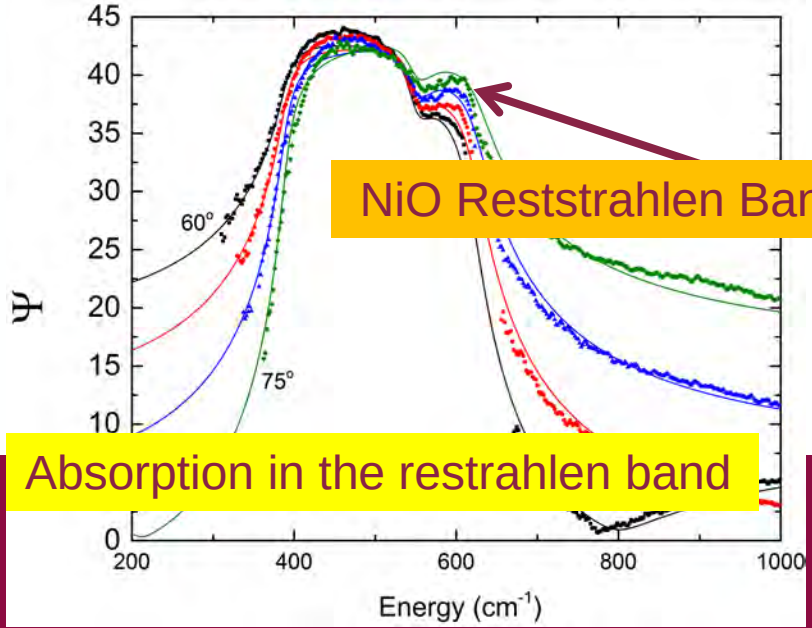


Rooksby, Nature, 1943

TO



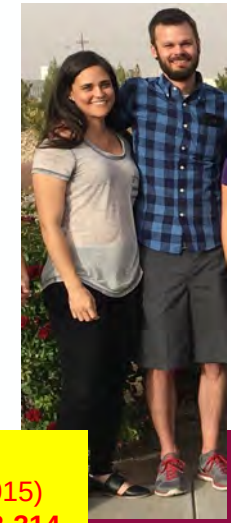
ε_0=11.3
ε_∞=5.0



NiO Reststrahlen Band

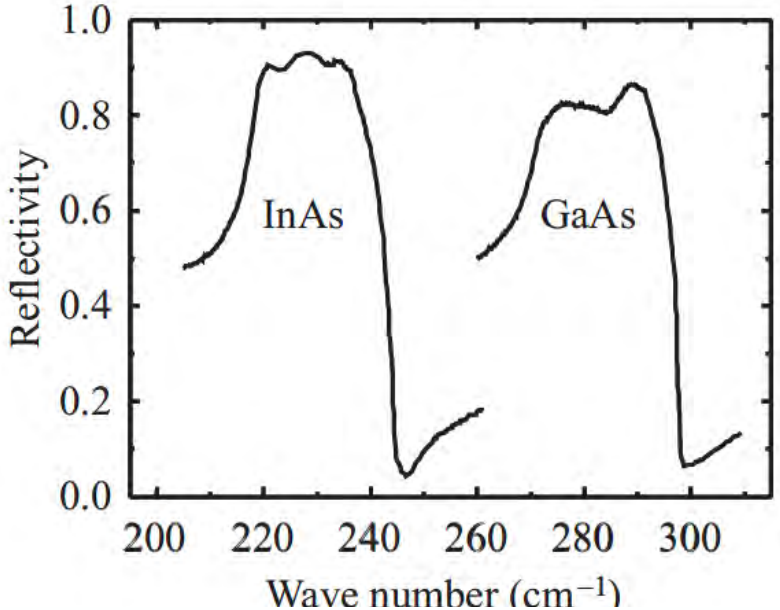
TA+TO phonon

Absorption in the restrahlen band



Willett-Gies & Nelson, JVST A **33**, 061202 (2015) Also Humlicek TSF **313-314**, 687 (1998) for LiF.

Infrared Lattice Vibrations (Lorentz Model)



Reststrahlen band

with two-phonon absorption

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{A\omega_0^2}{\omega_0^2 - \omega^2 - i\gamma_0\omega}$$

In polar materials (Born effective charge), TO and LO modes are split.

TO: transverse optical (atomic displacement perpendicular to **k**)

LO: longitudinal optical (atomic displacement along **k**)

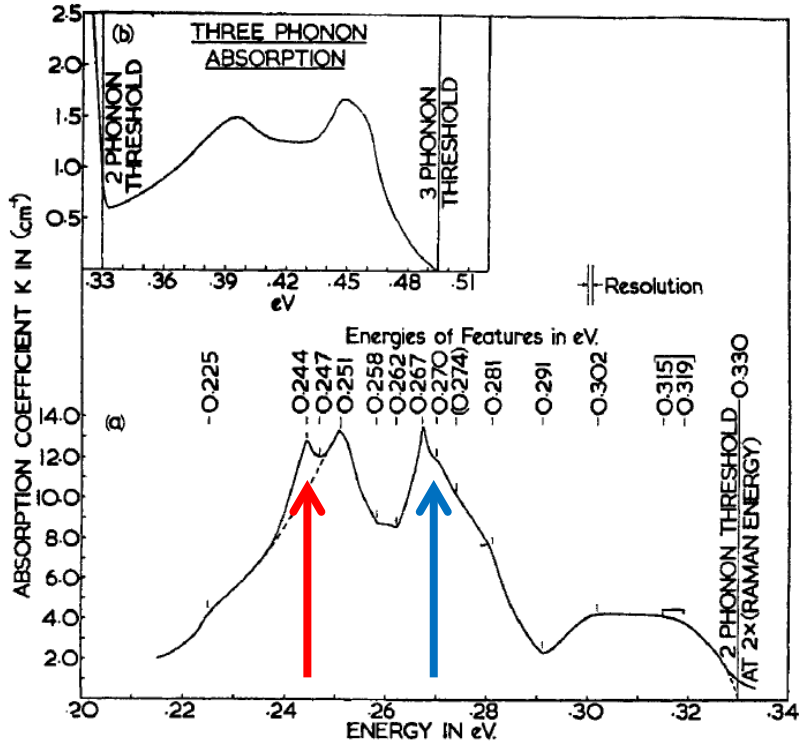
ε_2 has peak at TO frequency

ε_1 is negative from TO to LO frequency (reflectance is 1)

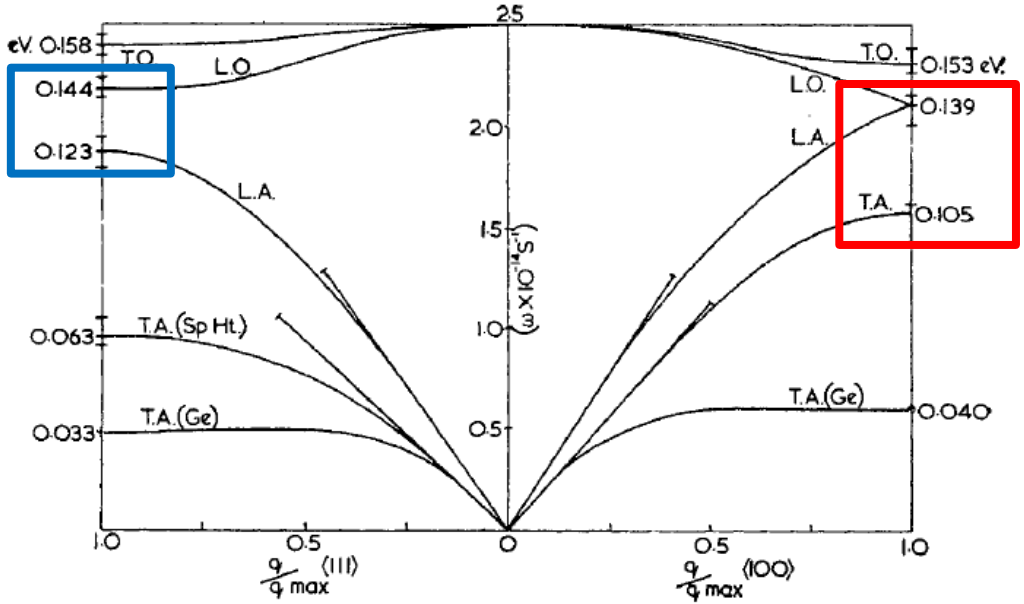
Restrahlen band extends from TO to LO energy.

Fox, Fig. 10.5

Two-Phonon Absorption (Diamond)



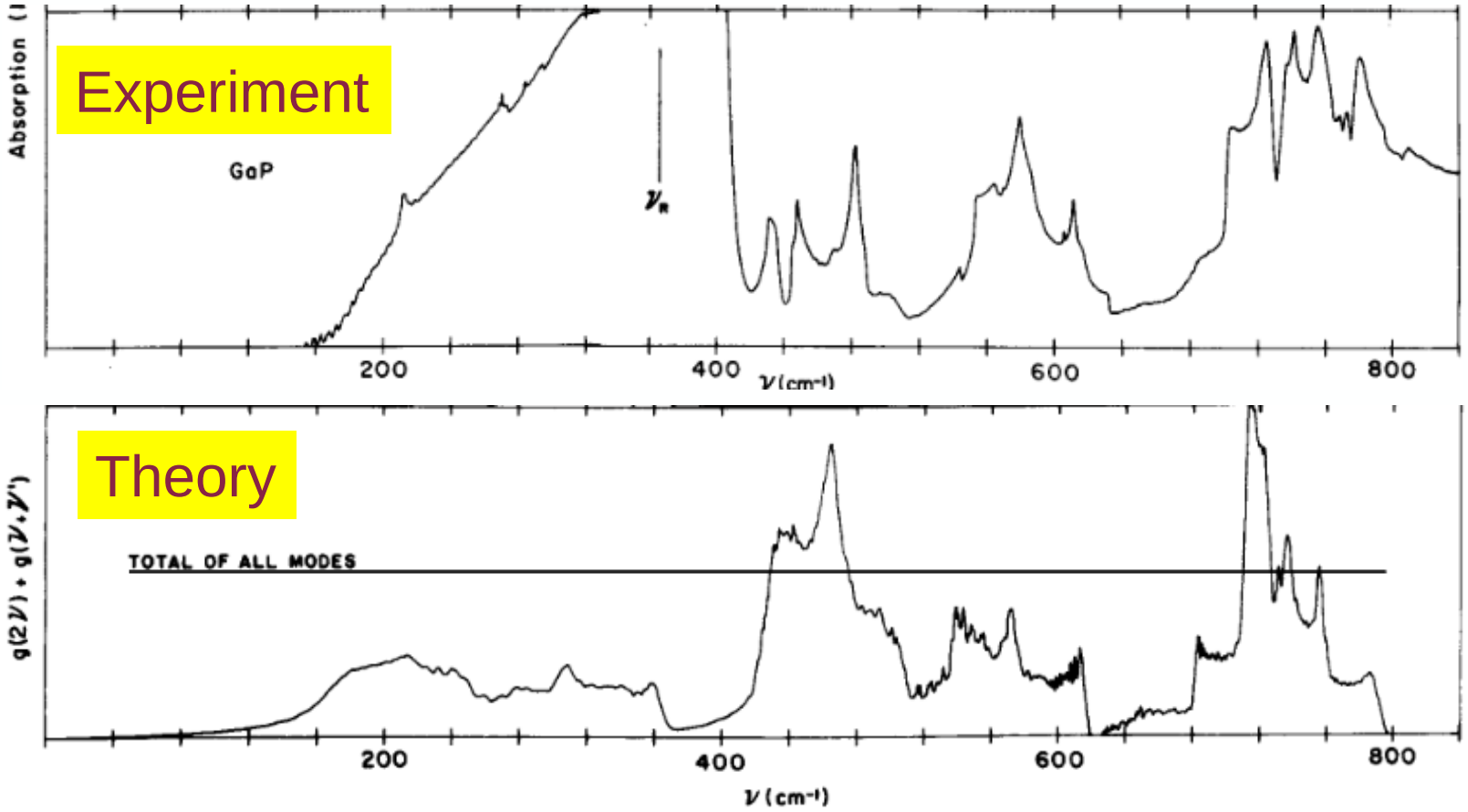
J.R. Hardy and S.D.Smith, Phil. Mag. **69**, 1163 (1961)



TA+LAO (X): 105+139=244 meV

A single photon excites two phonons. Energy and wave vector conserved.
Two-phonon absorption is weak (lower probability than TO phonon absorption).
Usually too weak to be studied with ellipsometry outside of TO/LO band.
Most likely to occur near Brillouin zone boundary (high density of states).

Two-Phonon Absorption (GaP)

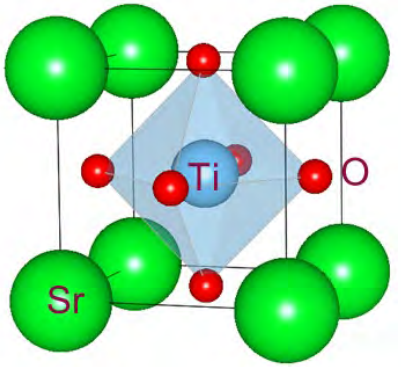


E.S. Koteles, Solid State Commun. **19**, 221 (1976)

Classification of Lattice Vibrations (k=0)

Long-wavelength (zone-center) lattice vibrations can be

- Infrared-active (transform like x, y, z)
- Raman-active (transform like xy, yz, zx or x^2, y^2, z^2)
- Silent
- Transformation property can be found from point group character table.



If there are N atoms per primitive cell, there are 3N degrees of freedom.

- 3 acoustic phonons (translation of crystal), zero energy
- 3(N-1) optical phonons

Find representations for optical phonons!

Need to know:

- Bravais lattice+basis, point group, space group
- Wyckoff positions (where are the atoms?)
- How do the symmetry operations act on the atoms? Check International Tables.
- Call N_R the number of invariant atoms for symmetry operator R
- Calculate character

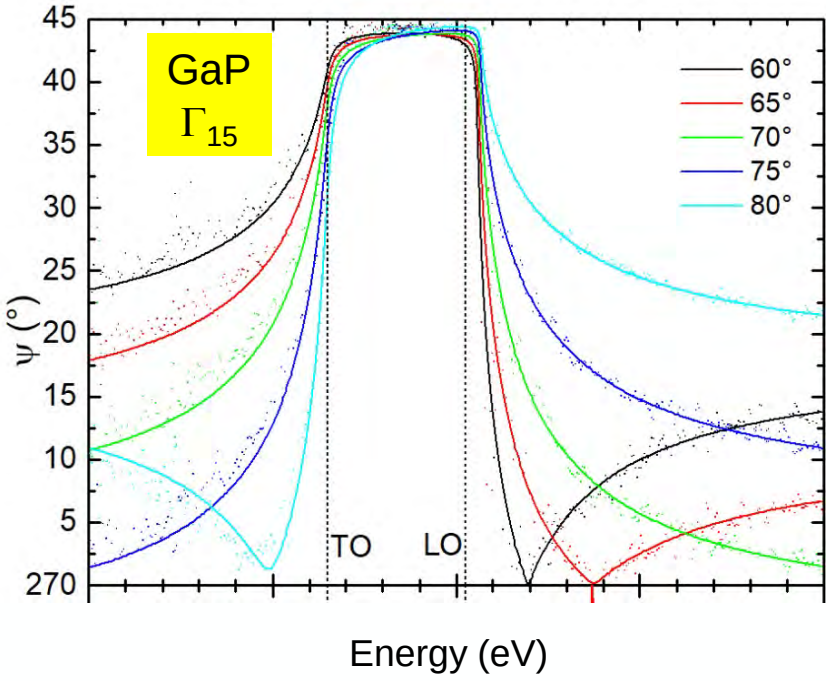
$$\chi(R) = N_R(\det R + 2\cos\phi)$$

- Decompose χ(R) into irreducible representations (using characters).

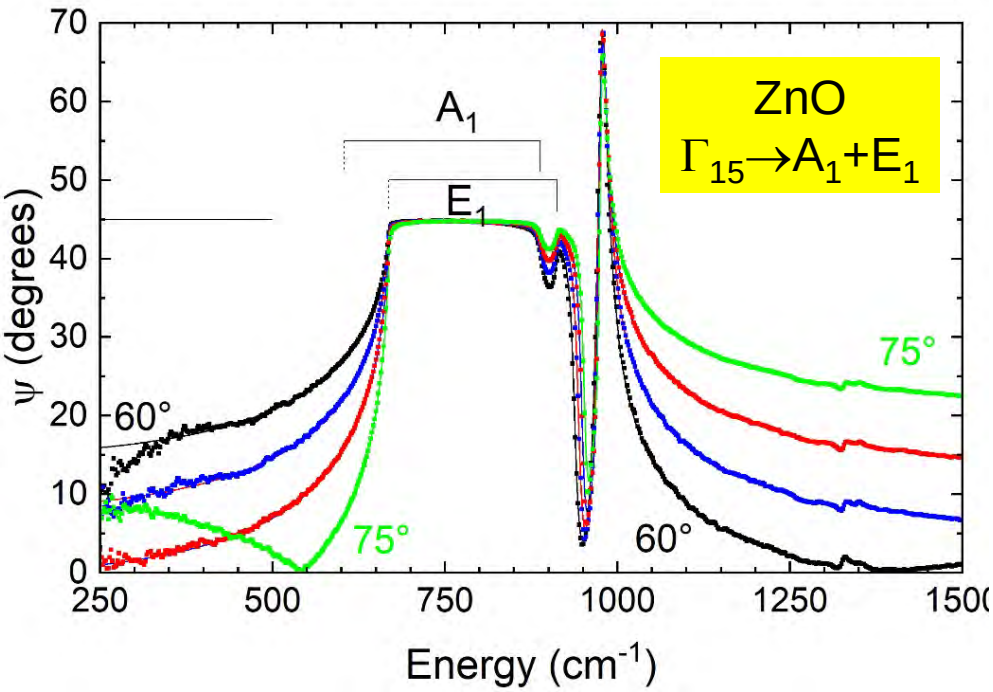
Dresselhaus, Dresselhaus, and Jorio, Group Theory (Springer, 2008)

Optical Phonons in Zincblende and Wurtzite Crystals

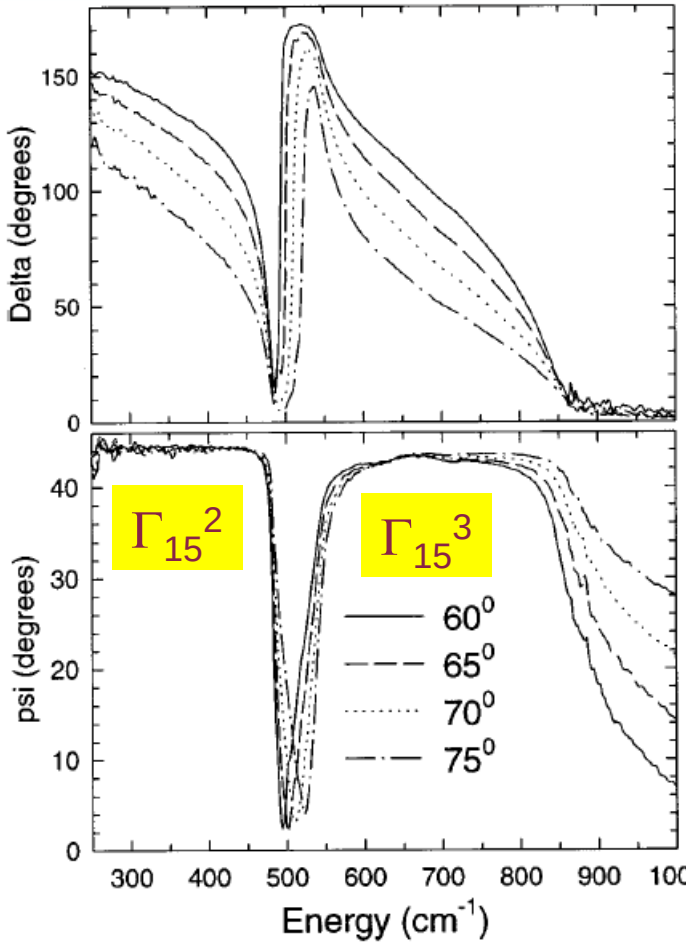
ψ near 45 degrees in reststrahlen band

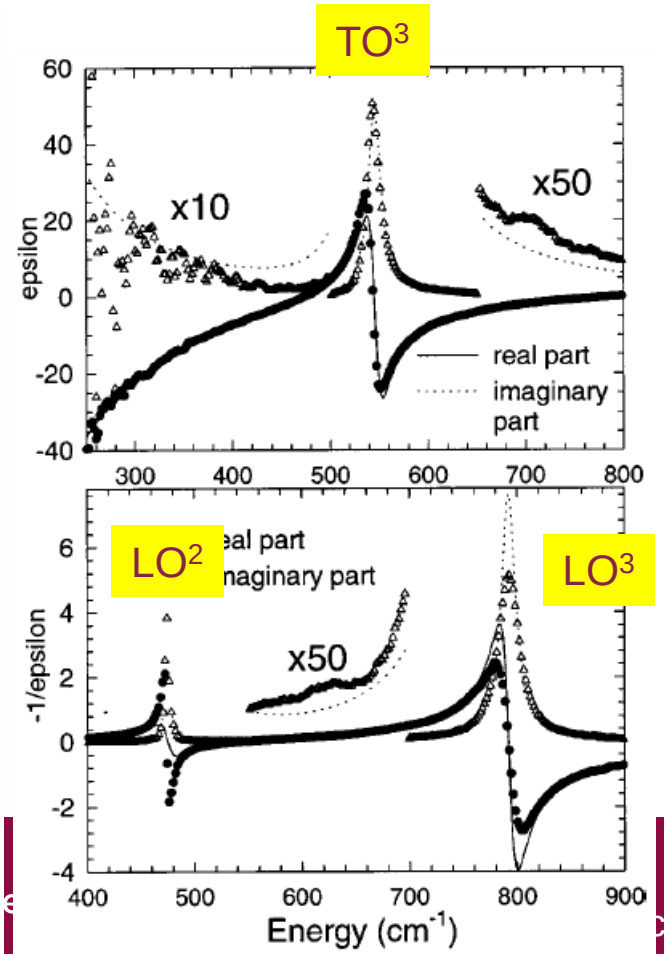


ψ above 45 degrees: LO phonon anisotropy

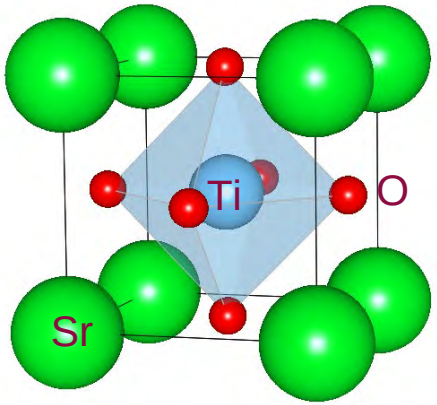


Phonons in Complex Oxides: Perovskites



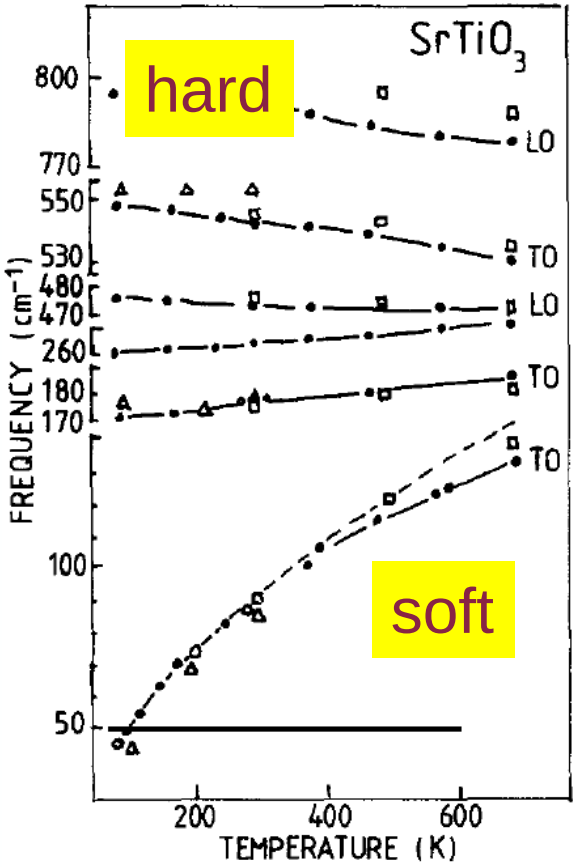


5 atoms ($SrTiO_3$)
4 optical phonons at Γ
$3\Gamma_{15}$(IR)+Γ_{25}(silent)



ε TO phonons
$-1/\varepsilon$ LO phonons

Hard and Soft Phonons



Typical behavior: **Hard**
Phonon energy decreases with temperature.

Anomalous: **Soft**
Phonon energy goes to zero at low temperature, **drives a phase transition** (collective movement of atoms)

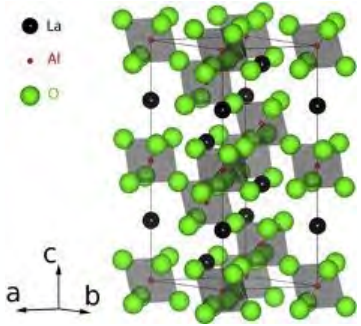
$SrTiO_3$ is nearly ferroelectric (T_C~0 K).

LST relation $\varepsilon_S \omega_{TO}^2 = \varepsilon_\infty \omega_{LO}^2$

$\omega_{\mathrm{TO}} \to 0$ at T_C implies $\varepsilon_s \to \infty$

V.N. Denisov, Physics Reports **151**, 1 (1987)

Classification of Phonons in Metal Oxides



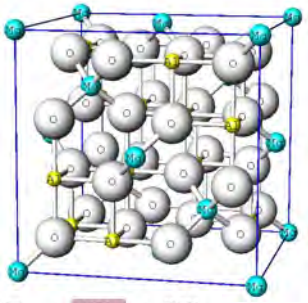
$LaAlO_3$

D_{3d}^6 or $R\bar{3}c$

Space Group
Wyckoff positions

$MgAl_2O_4$

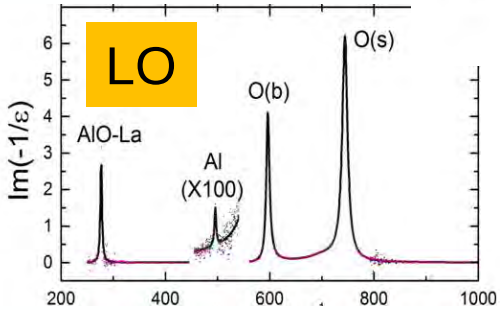
O_h^7 or $Fd3m$

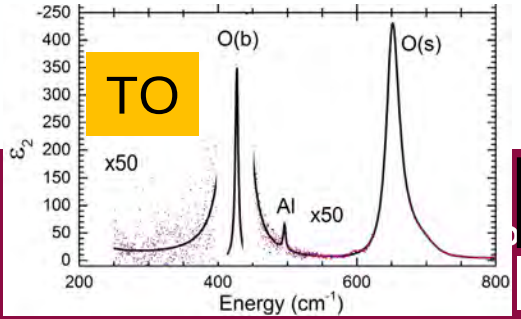


$$\chi(R) = N_R(\det R + 2\cos\phi)$$

$\Gamma(D_{3d}^6) = 2A_{1u} + 3A_{2g} + A_{1g} + 3A_{2u} + 4E_g + 5E_u$ (Raman Active; IR Active)

$\Gamma(O_h^7) = A_{1g} + E_g + T_{1g} + 3T_{2g} + 2A_{2u} + 2E_u + 4T_{1u} + 2T_{2u}$ (Raman Active; IR Active)



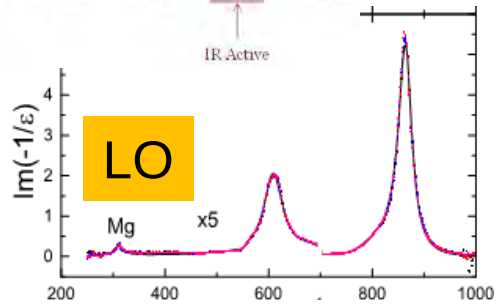


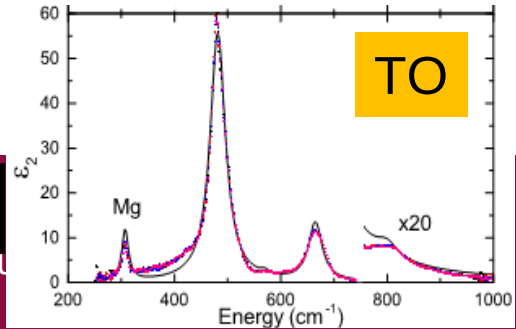
FTIR Ellipsometry

Loss function: **LO phonons**

Dielectric function: **TO phonons**

Raman exp: Raman modes



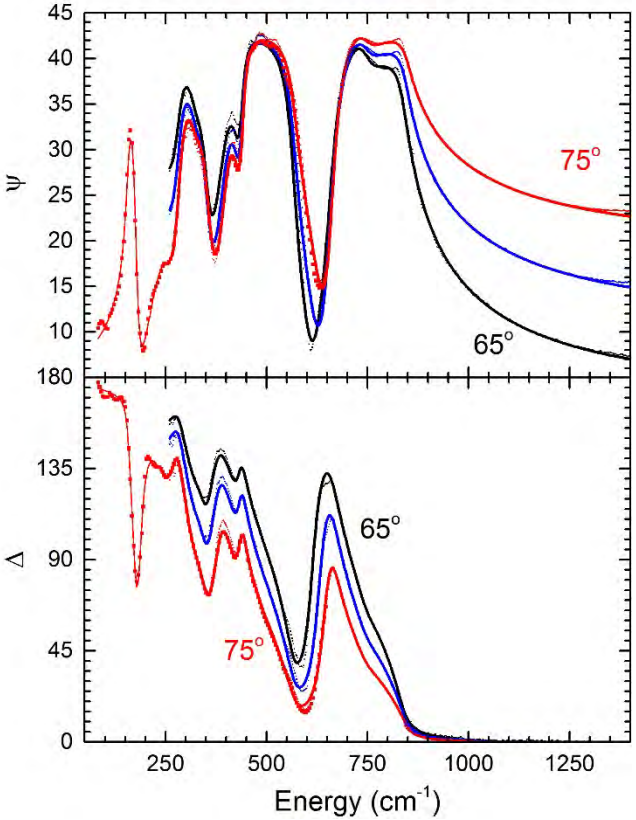


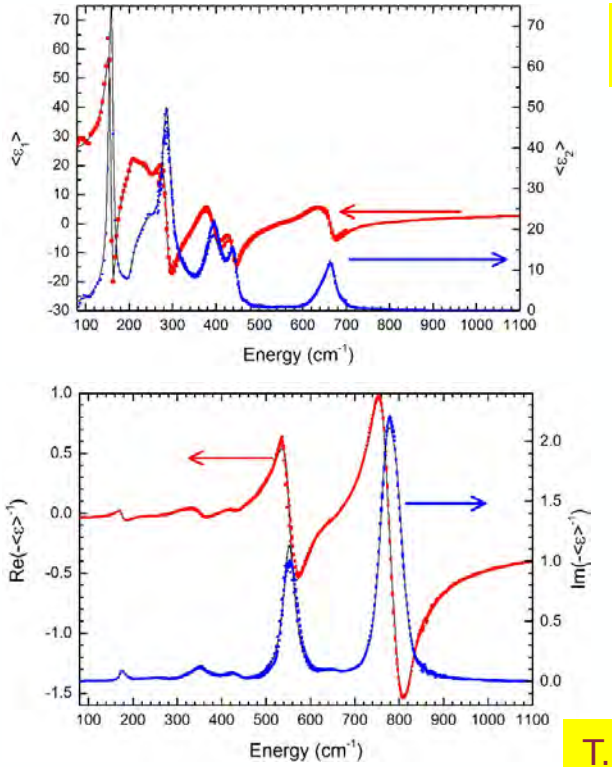
$$\varepsilon_s = \varepsilon_\infty \prod_i \frac{\omega_{i,\mathrm{LO}}^2}{\omega_{i,\mathrm{TO}}^2} = 22.3 \pm 0.3$$

$$\varepsilon_s = \varepsilon_\infty \prod_i \frac{\omega_{i,\mathrm{LO}}^2}{\omega_{i,\mathrm{TO}}^2} = 7.8 \pm 0.2$$

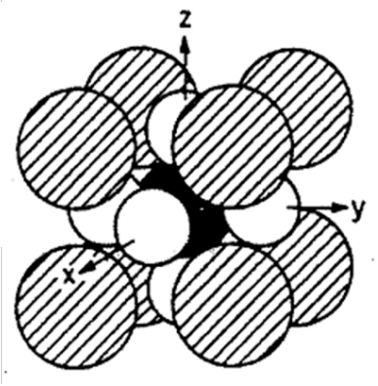
Willett-Gies, Thin Solid Films, 2013; Zollner, Thin Solid Films, 2013

Far-Infrared Ellipsometry (bulk LSAT)





ε shows TO phonons

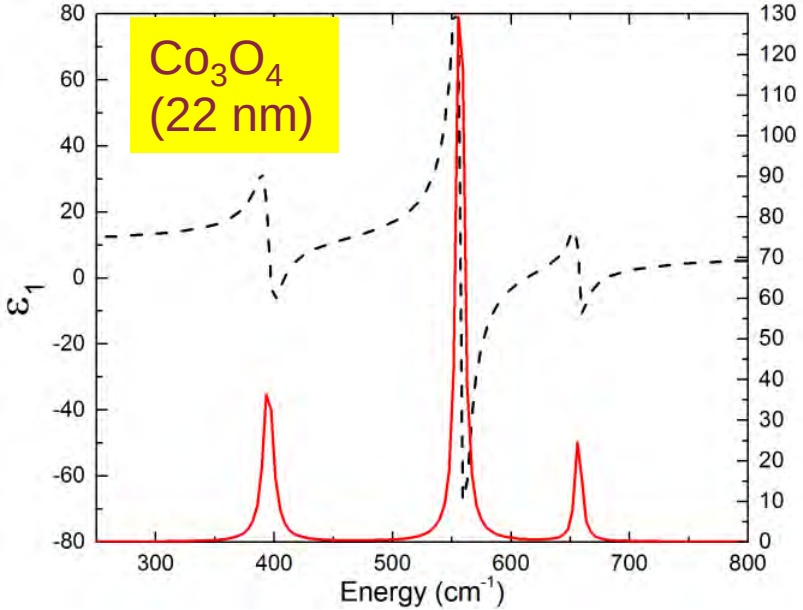


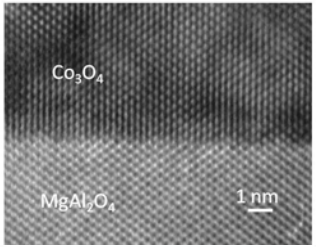
–1/ε shows LO phonons

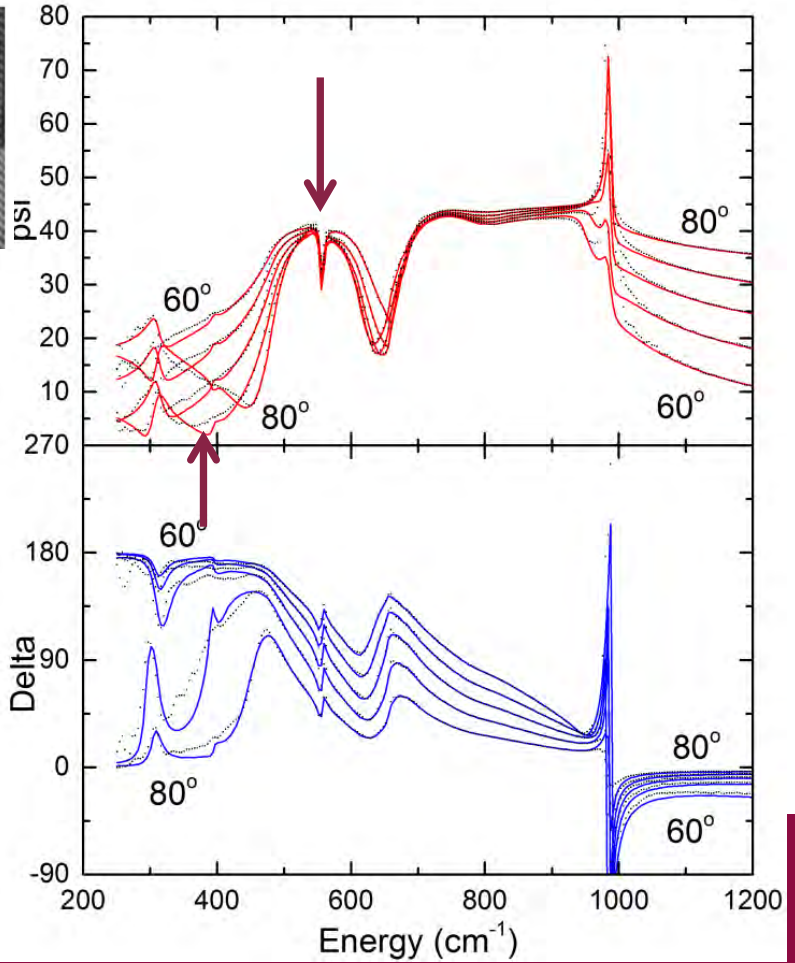
T.N. Nunley, JVSTA 34, 051507 (2016)

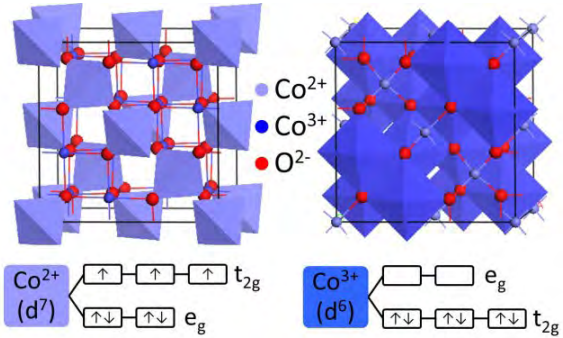
Disordered double perovskite $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.35}$
Many phonon modes. Several reststrahlen bands.

Phonons in More Complex Oxides (Co_3O_4 on Spinel)



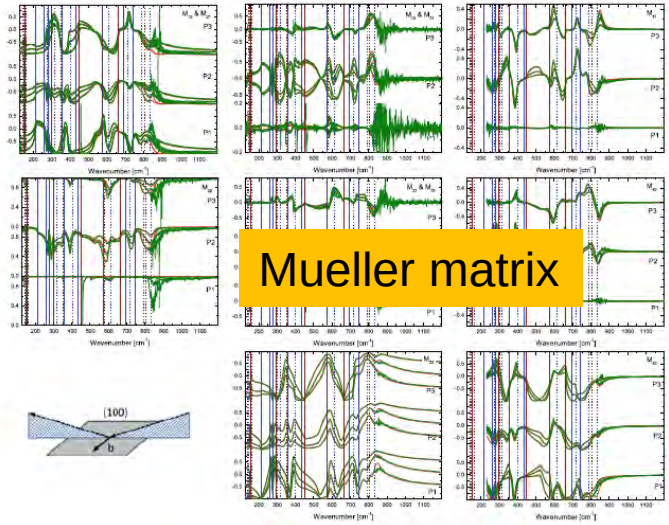




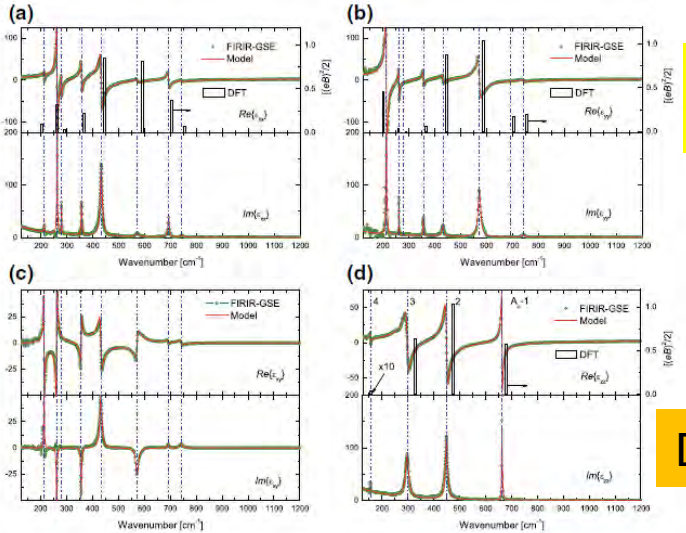


K. Kormondy, JAP **115**, 243708 (2014)

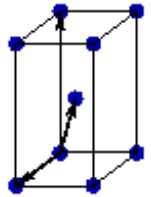
Phonon Anisotropy in β-Gallium Oxide



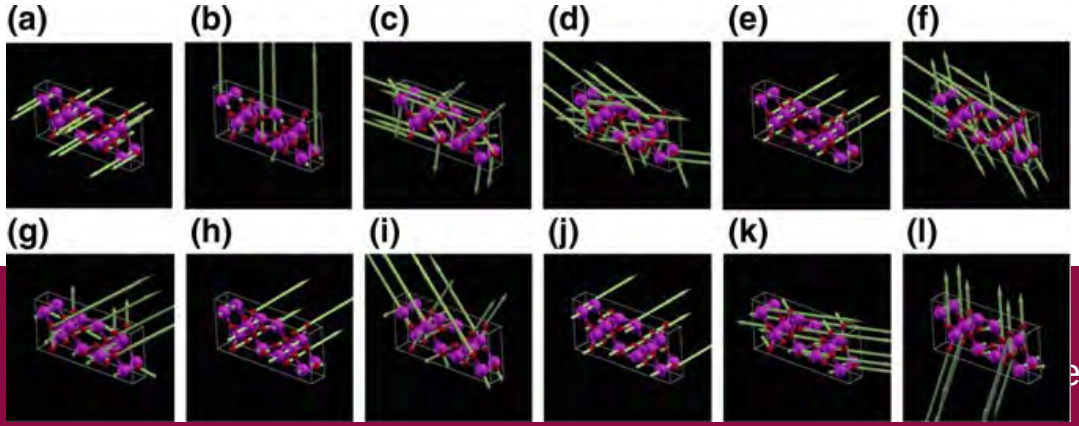
Mueller matrix



Monoclinic
Space Group C2/m



Dielectric tensor



Phonon vibrations

M. Schubert in M. Higashiwaki, *Gallium Oxide*, 2020

Kramers-Kronig Relations

$$\vec{P}(\vec{r},t) = \varepsilon_0 \int \chi_e(\vec{r}' - \vec{r}, t' - t)\vec{E}(\vec{r}', t')dt'd^3\vec{r}'$$

Response function $\chi_e(\vec{r}' - \vec{r}, t' - t) = 0 \text{ for } t' > t$

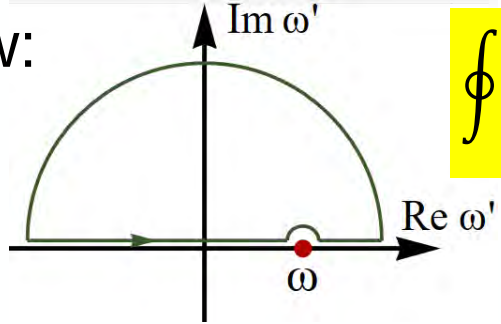
The charges cannot move before the field has been applied.

Kramers-Kronig relations follow:

$$\vec{D}(\vec{k},\omega) = \varepsilon_0 \varepsilon(\vec{k},\omega)\, \vec{E}(\vec{k},\omega)$$

$$\varepsilon_1(\omega) - 1 = \frac{2}{\pi} \wp \int_0^\infty \frac{\omega' \varepsilon_2(\omega') d\omega'}{\omega'^2 - \omega^2}$$

$$\varepsilon_2(\omega) = -\frac{2\omega}{\pi} \wp \int_0^\infty \frac{\varepsilon_1(\omega') d\omega'}{\omega'^2 - \omega^2}$$



$$\oint \frac{\chi(\omega)}{\omega' - \omega} d\omega' = 0$$

Cauchy

Contour integrals in complex plane:
The real part of ε can be calculated if the imaginary part is known (and vice versa). Similar Kramers-Kronig relations for other optical constants.

Lucarini, Springer, 2005

Analytical Properties of the Dielectric Function

Fields **E**(**r**,t) are real quantities

$$\varepsilon\left(-\vec{k},-\omega\right)=\overline{\varepsilon\left(\vec{k},\omega\right)}$$

$$\varepsilon(-\omega)=\overline{\varepsilon(\omega)}$$

Onsager relation

Dielectric tensor symmetric (B=0)

Also from energy density.

Passive materials (no optical gain)

$$\varepsilon\left(-\vec{k},\omega\right)={}^{t}\varepsilon\left(\vec{k},\omega\right)$$

$$\varepsilon(\omega)={}^{t}\varepsilon(\omega)$$

$$\varepsilon_2(\omega)\geq 0$$

Like any analytic complex function, $\varepsilon(\omega)$ is defined by its zeroes and poles in the complex plane (below real axis). This implies

$$\varepsilon(\omega)=\varepsilon_\infty\prod_{j=1}^{N}\frac{\omega_{\mathrm{LO},j}^2-\omega^2-i\gamma_{\mathrm{LO},j}\omega}{\omega_{\mathrm{TO},j}^2-\omega^2-i\gamma_{\mathrm{TO},j}\omega}$$

γ>0 (causality)

Works well for phonons and plasmons.

R. P. Lowndes, PRB 1, 2754 (1970).
Berreman & Unterwald, Phys. Rev. **174**, 791 (1968).
Zollner, JVST B (2019).

Comparison of Lorentz and Lowndes Models

Drude-Lorentz Model

$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

Lowndes Model

$$\varepsilon(\omega) = \varepsilon_\infty \prod_{j=1}^{N} \frac{\omega_{\mathrm{LO},j}^2 - \omega^2 - i\gamma_{\mathrm{LO},j}\omega}{\omega_{\mathrm{TO},j}^2 - \omega^2 - i\gamma_{\mathrm{TO},j}\omega}$$

Drude terms: Poles on the imaginary axis (ω_0=0)
Additional broadening parameter for LO phonon.
Lorentz identical to Lowndes model, if $\omega_{TO}=\omega_{LO}$.
Otherwise: **Frictional force allowed to vary with velocity.**
Complex Lorentz amplitude, frequency-dependent damping.
Lowndes model makes no assumptions about forces.

R. P. Lowndes, PRB 1, 2754 (1970).
Berreman & Unterwald, Phys. Rev. **174**, 791 (1968).
Zollner, JVST B (2019).

Anharmonically Broadened Lorentzian

Drude-Lorentz Model

$$\varepsilon(\omega) = 1 + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

Lowndes Model

$$\varepsilon(\omega) = \varepsilon_\infty \prod_{j=1}^{N} \frac{\omega_{\mathrm{LO},j}^2 - \omega^2 - i\gamma_{\mathrm{LO},j}\omega}{\omega_{\mathrm{TO},j}^2 - \omega^2 - i\gamma_{\mathrm{TO},j}\omega}$$

Anharmonically broadened Lorentzian

$$\varepsilon(\omega) = 1 + \sum_i \frac{A_i \omega_{0,i}^2 - ib\omega}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

GenOsc2 layer in Woollam software.

$$b = \varepsilon_\infty(\gamma_{LO} - \gamma_{TO})$$

Schubert group
ICSE 2022 (Beijing)

Summary

- **Drude model** explains optical response of **metals**.
- High reflectance below the plasma frequency.
- Interband transitions overlap with Drude absorption.

- Doped semiconductors have infrared plasma frequencies.

- **Lorentz model** explains **infrared lattice absorption**.
- TO/LO modes result in **reststrahlen band**.
- Multiple modes for complex crystal structures or alloys.

Thank you!

Questions?