Optical Properties of Solids: Lecture 6

Stefan Zollner

New Mexico State University, Las Cruces, NM, USA
and Institute of Physics, CAS, Prague, CZR (Room 335)
zollner@nmsu.edu or zollner@fzu.cz

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http://ellipsometry.nmsu.edu

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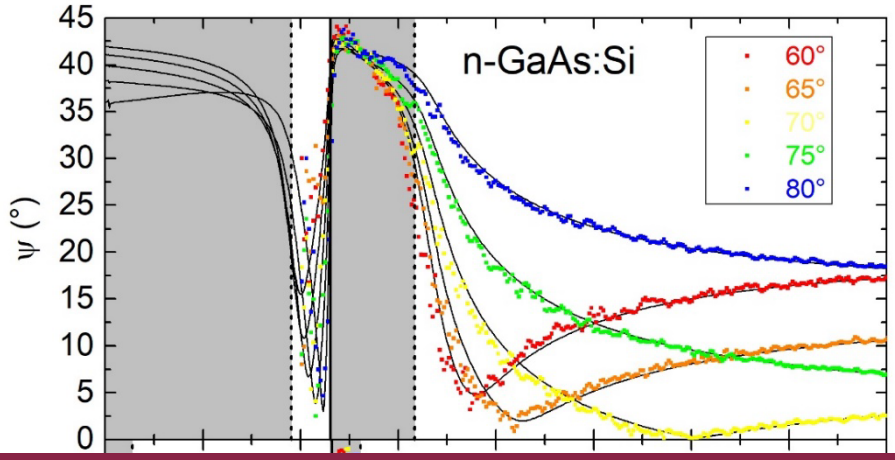
Optical Properties of Solids: Lecture 5+6

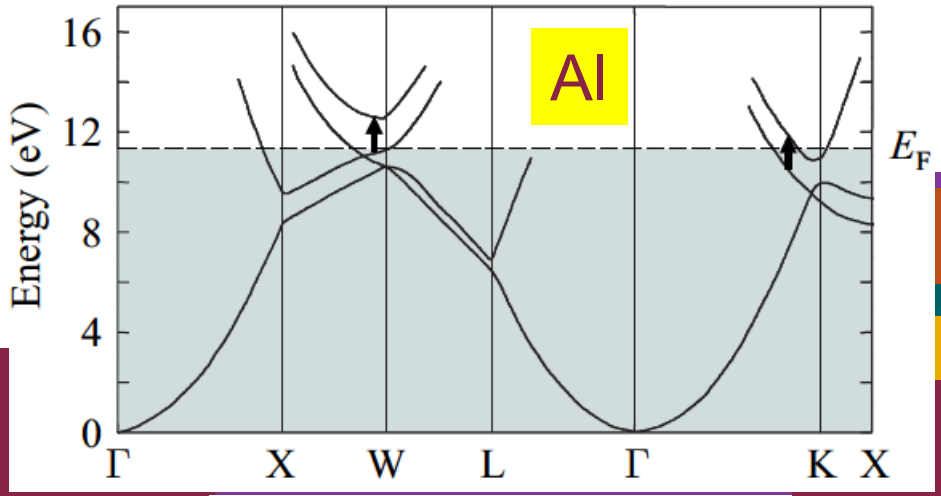
Lorentz and **Drude** model: Applications

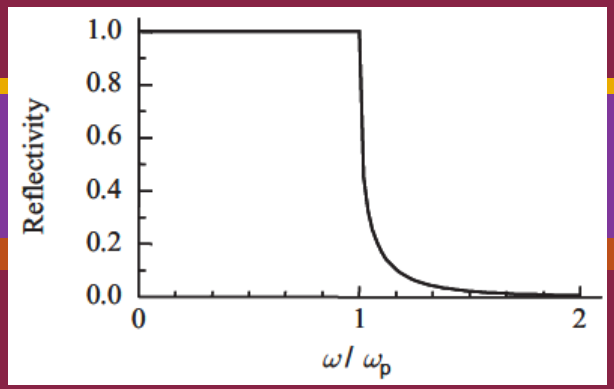
1. Metals, doped semiconductors
2. Insulators

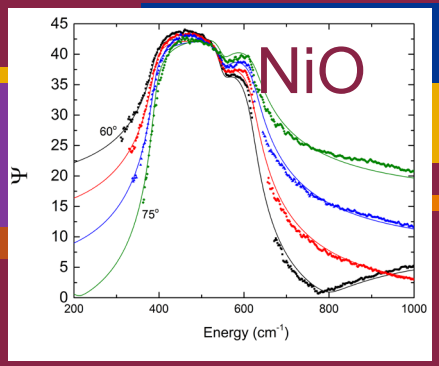
Sellmeier equation, Poles, Cauchy dispersion

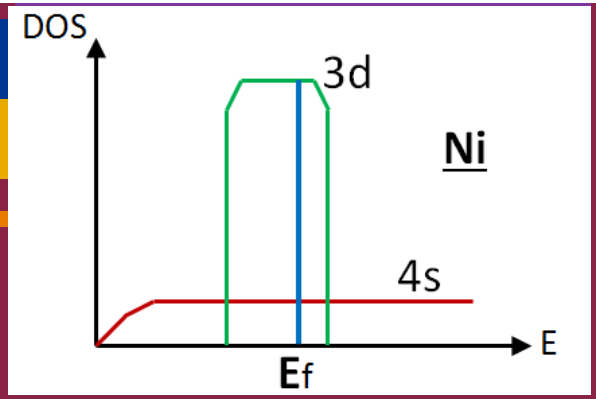
Analytical properties of ε











References: Dispersion, Analytical Properties

Standard Texts on Electricity and Magnetism:

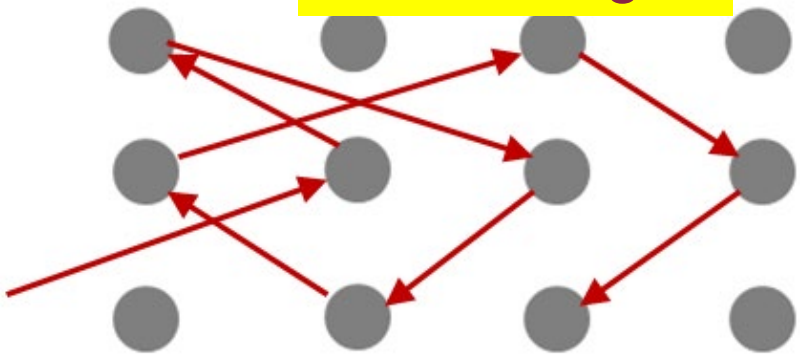
- J.D. Jackson: *Classical Electrodynamics*
- L.D. Landau & J.M. Lifshitz, Vol. 8: *Electrodynamics of Cont. Media*

Ellipsometry and Polarized Light:

- R.M.A. Azzam and N.M. Bashara: *Ellipsometry and Polarized Light*
- **H.G. Tompkins and E.A. Irene: *Handbook of Ellipsometry* (chapters by Rob Collins and Jay Jellison)**
- **H. Fujiwara, *Spectroscopic Ellipsometry***
- **Mark Fox, *Optical Properties of Solids***
- H. Fujiwara and R.W. Collins: *Spectroscopic Ellipsometry for PV* (Vol 1+2)
- Zollner: *Propagation of EM Waves in Continuous Media* (Lecture Notes)
- Zollner: *Drude and Kukharskii mobility of doped semiconductors extracted from FTIR ellipsometry spectra*, J. Vac. Sci. **37**, 012904 (2019).

Drude-Lorentz Model: Free and Bound Charges

Drude:
Free Charges



Lorentz:
Bound Charges

$t) = E_0 \exp(-i\omega t)$

qE

v

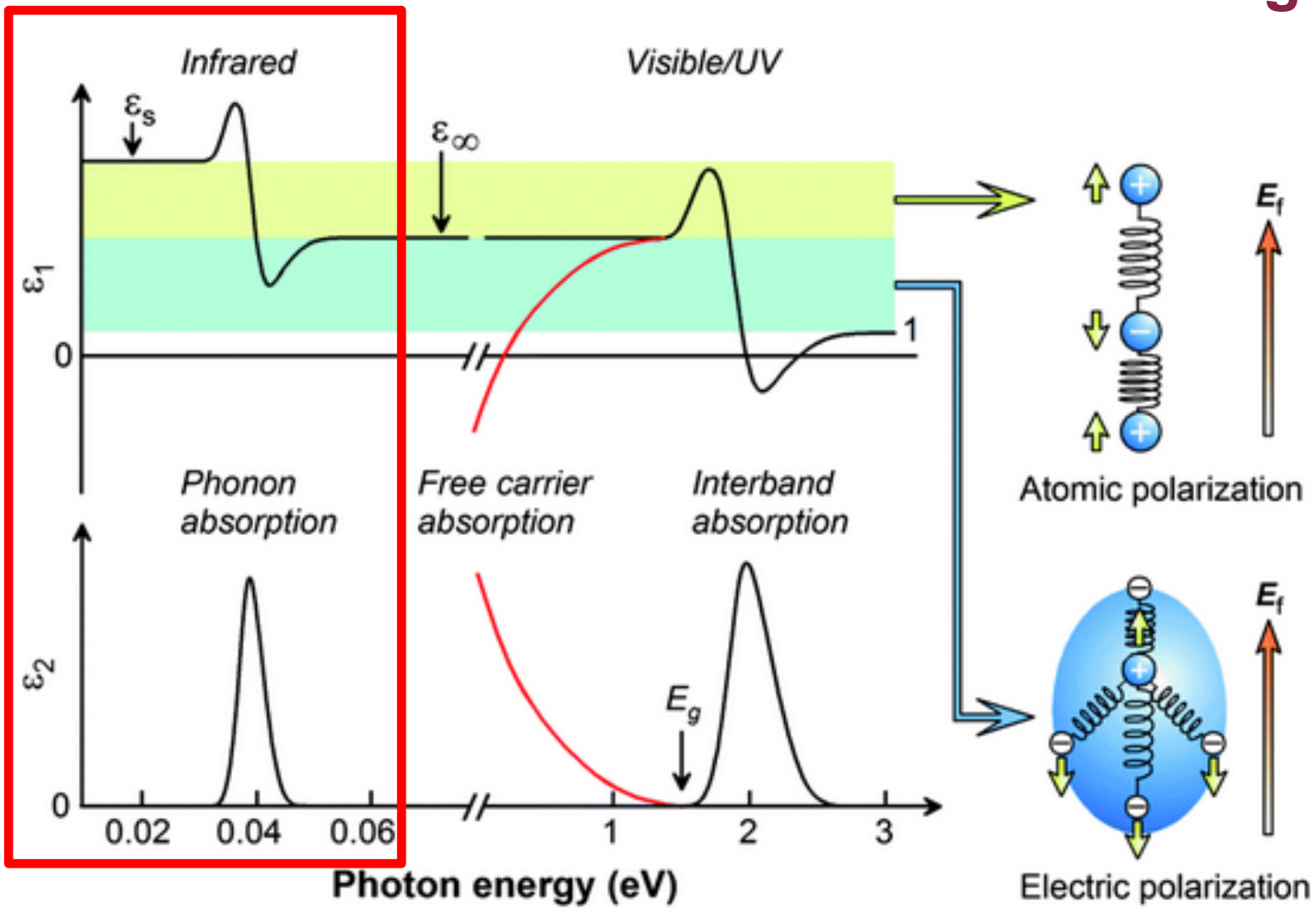
$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

w_P	(unscreened) **plasma frequency** of free charges
w_0	**resonance frequency** of bound charges
g_D, g_0	**broadenings** of free and bound charges
A	**amplitude** of bound charge oscillations (density, strength)

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

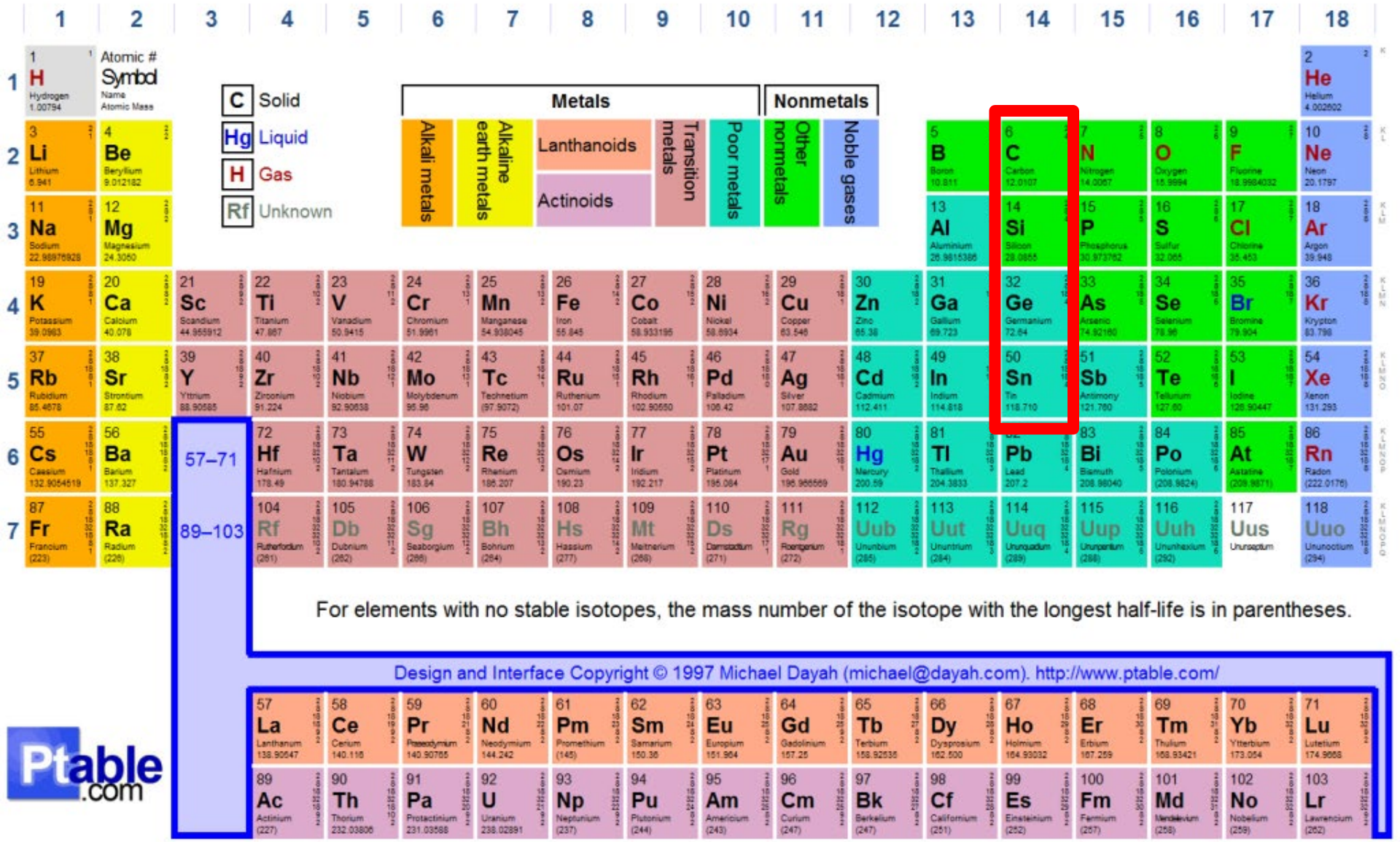
Discuss plasma frequency trends.

Drude-Lorentz Model: Free and Bound Charges

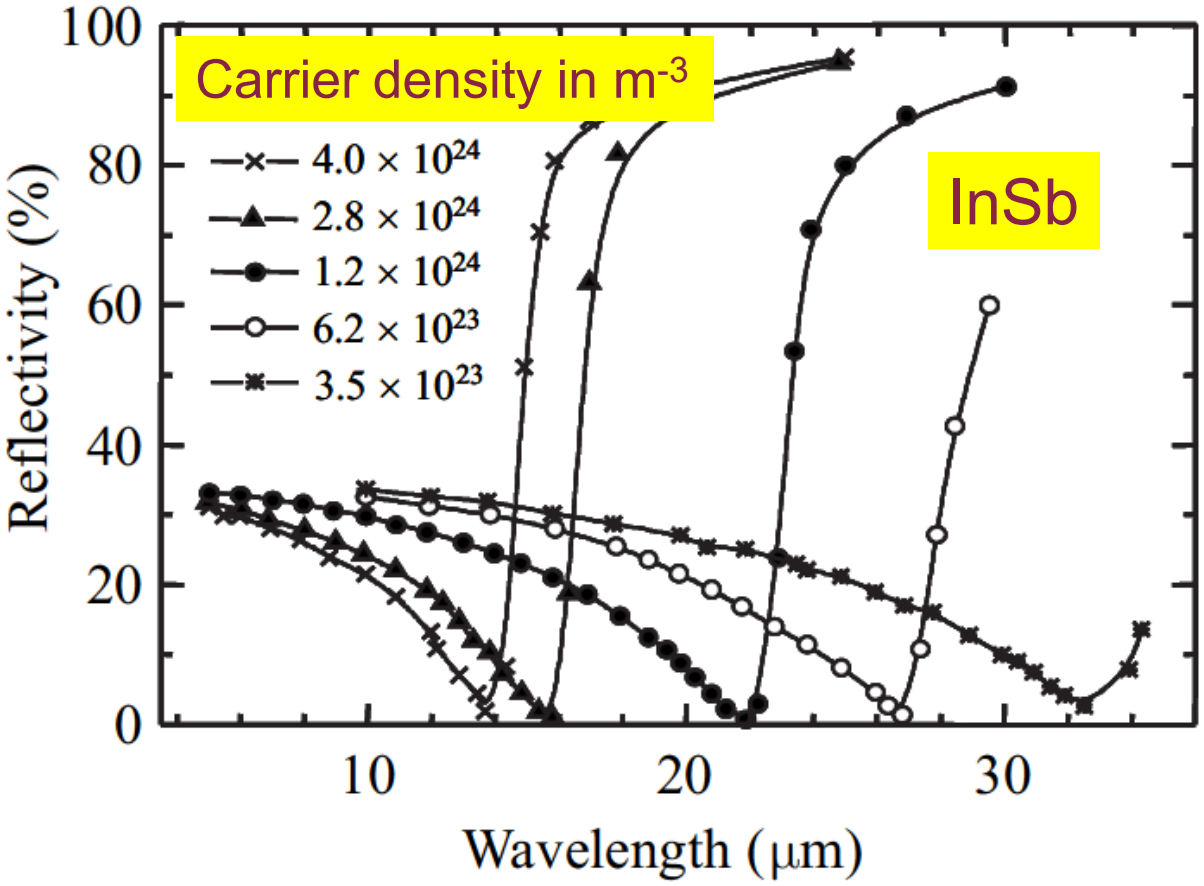


$$\varepsilon(\omega) = 1 - \sum_i \frac{\omega_{P,i}^2}{\omega^2 + i\gamma_{D,i}\omega} + \sum_i \frac{A_i \omega_{0,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_{0,i}\omega}$$

Semiconductors



Free-Carrier Reflection in doped semiconductors



$$\omega_P^2 = \frac{n_f e^2}{m^* m_0 \varepsilon_0}$$

Reflectance minimum near plasma frequency

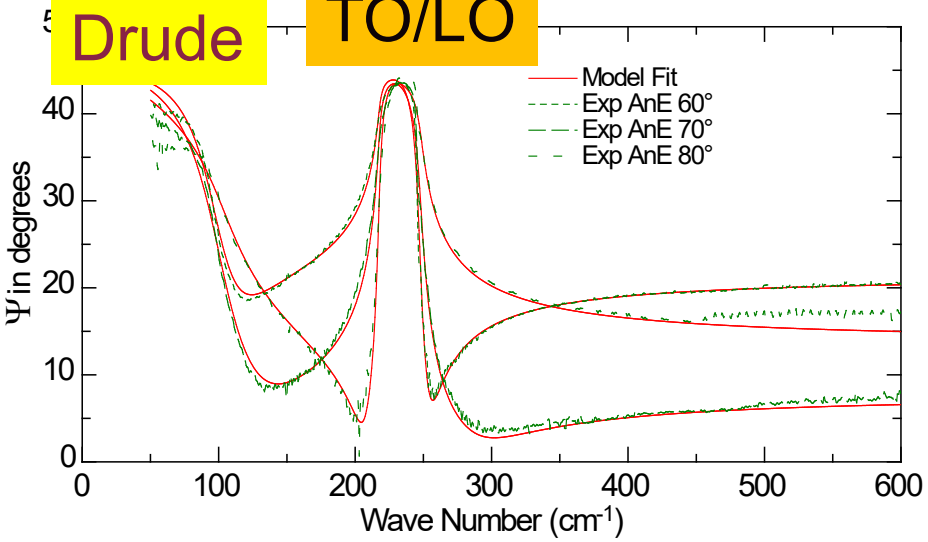
Doped semiconductors behave just like a metal, except for the lower carrier density; **plasma frequency in infrared region.**

Fox, *Optical Properties of Solids*

Infrared ellipsometry of doped semiconductors

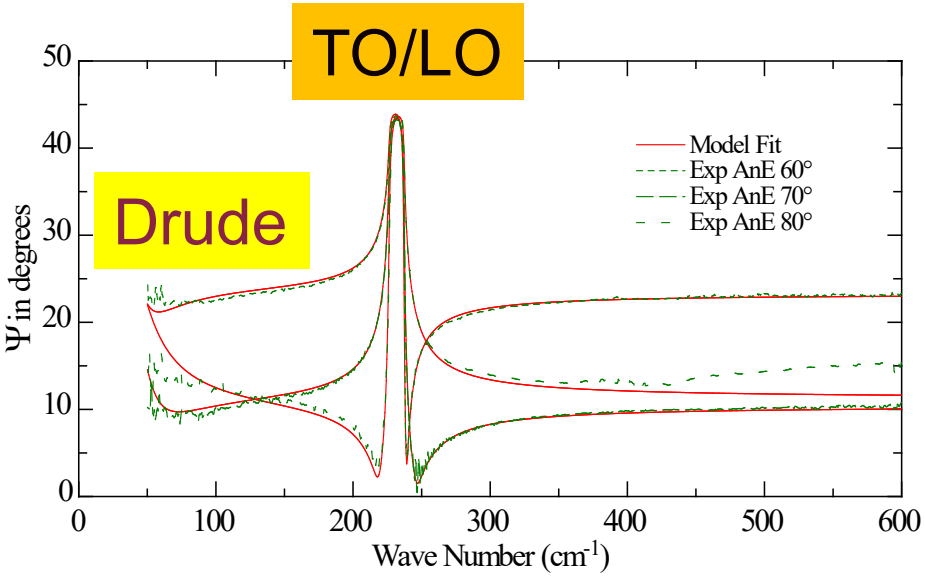
Drude

TO/LO



TO/LO

Drude



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}

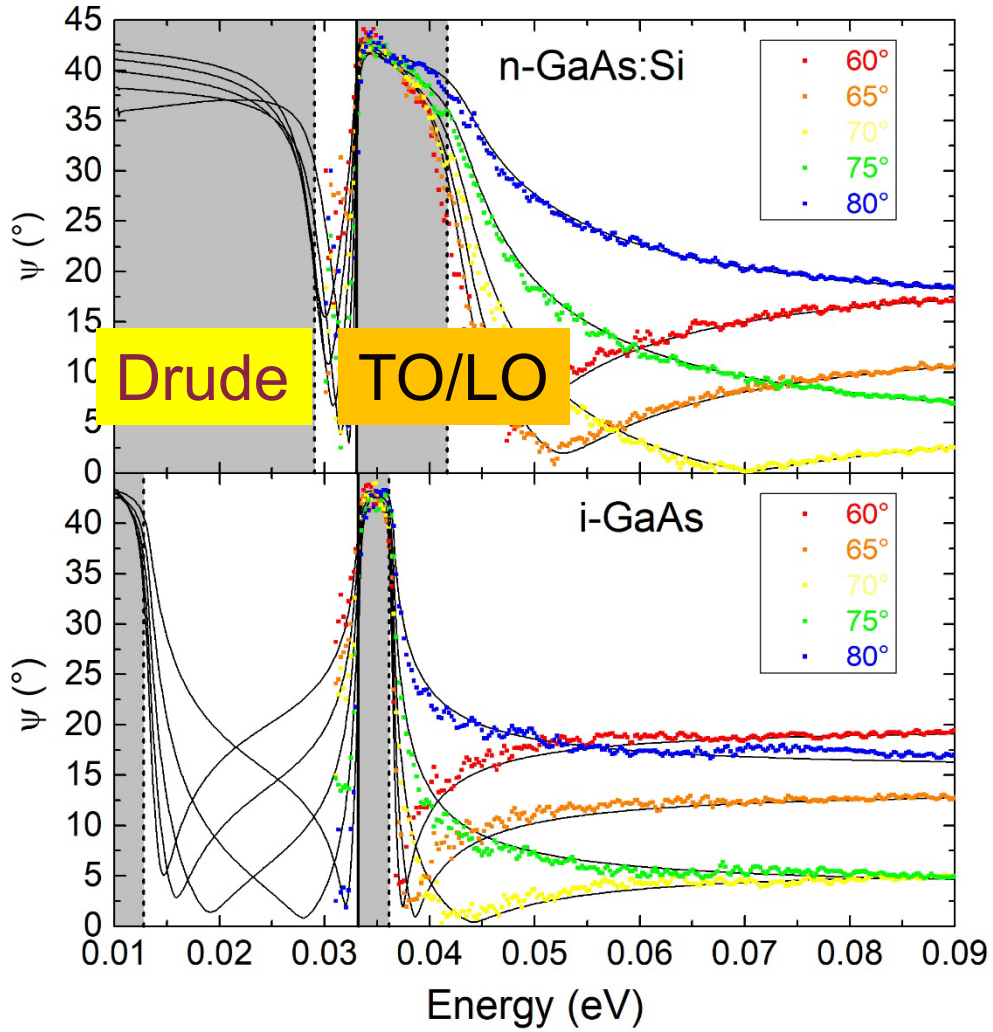
γ= 71 cm^{-1}

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).**

Abadizaman, Emminger, Knight, Schubert

Infrared ellipsometry of doped semiconductors



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

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

Plasmon effect:

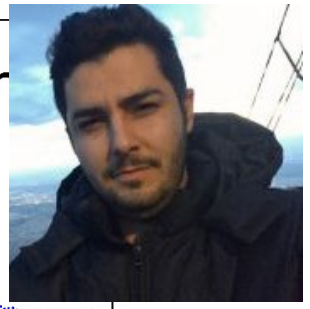
Doping pushes LO phonon to higher energies.

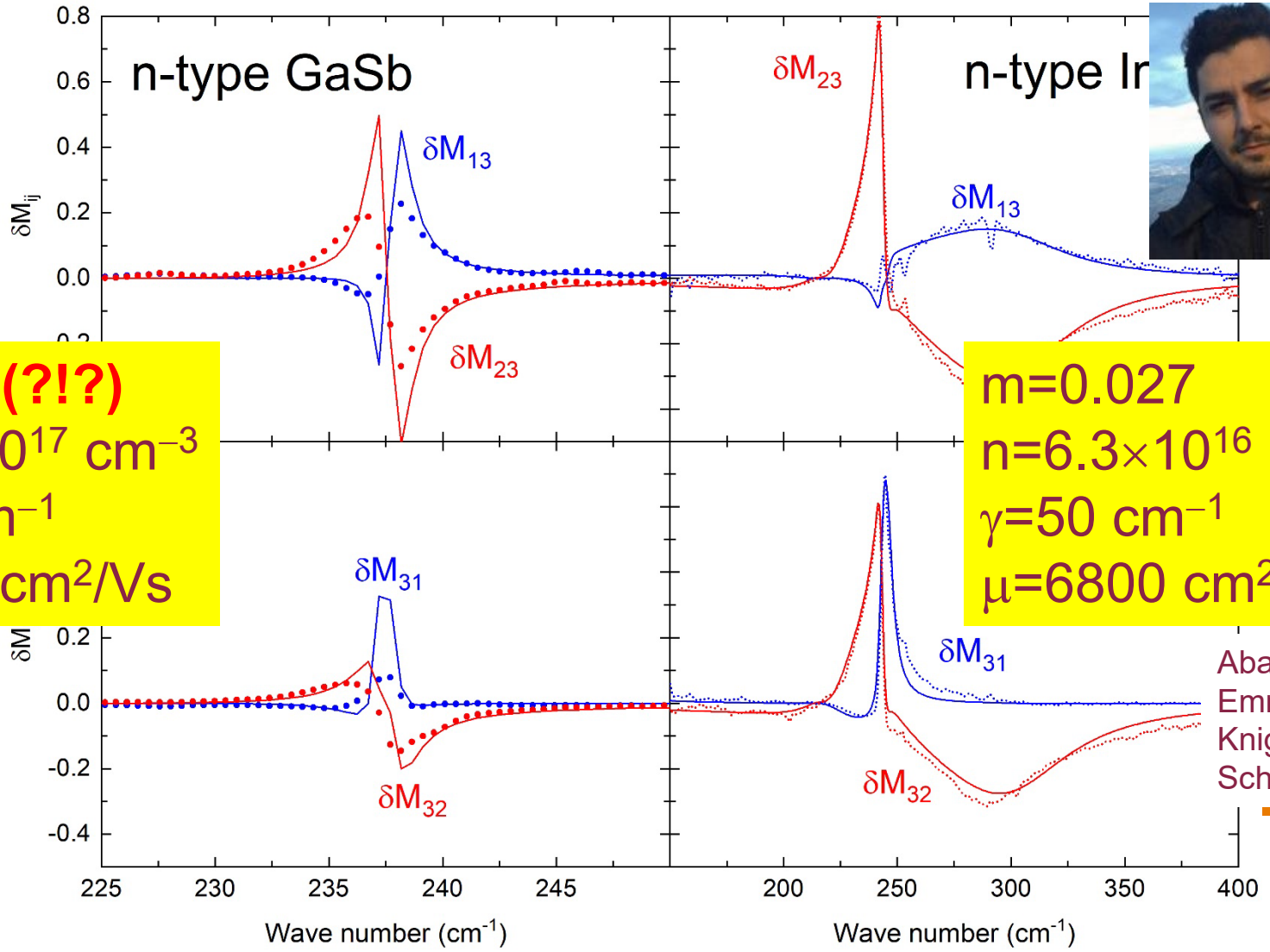
Doped semiconductors behave just like a metal, except for the lower carrier density; **plasma frequency in infrared region.**

Zollner, JVSTB **37**, 012904 (2019)

Optical Hall Effect: Ellipsometry with magnetic field

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





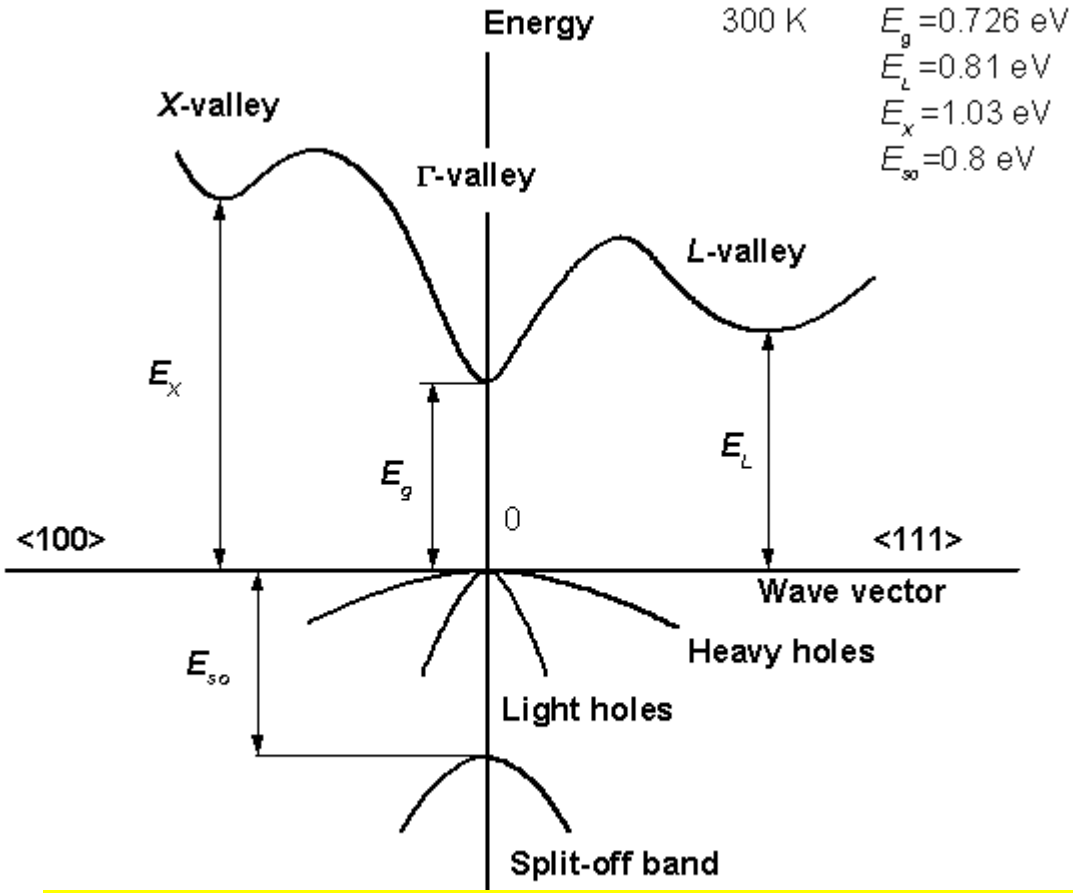
m=0.13 (?!?)
n=1.3×10^{17} cm^{-3}
γ= 71 cm^{-1}
μ=1000 cm^2/Vs

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.



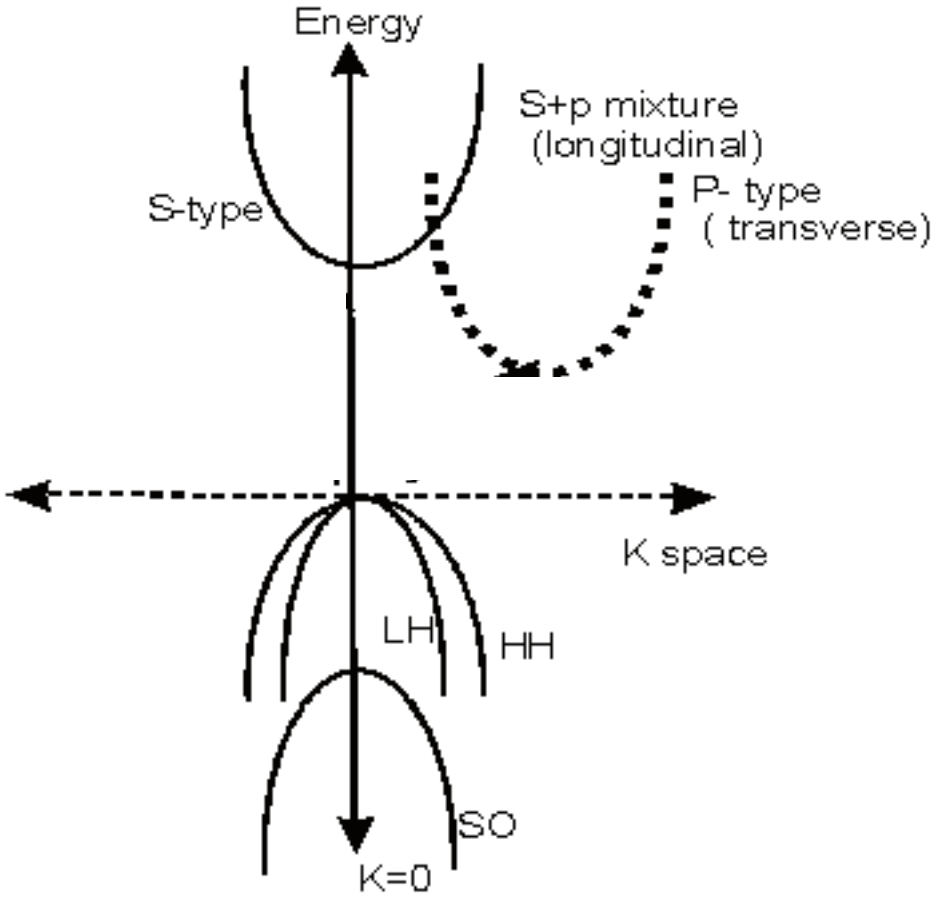
k_BT=26 meV
E_Γ=730 meV
Effective masses
m_Γ=0.041
$m_L^\perp$=0.11
$m_L^\parallel$=0.95
4 L-valleys
m_L^{DOS}=0.57
$m_{transport}$=0.15 (later)

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)

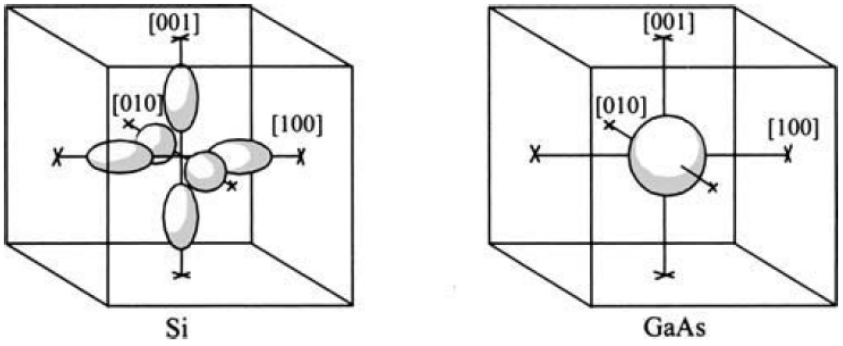
CB: Conduction band (empty)

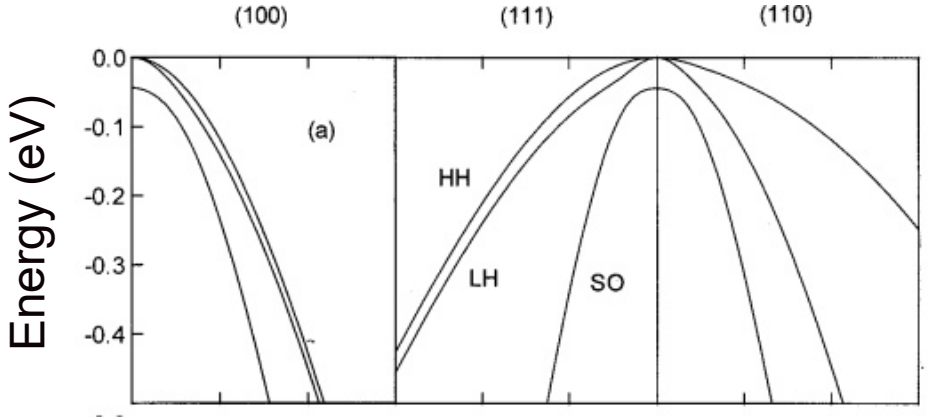


VB: Valence band (filled)

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

Drude Model for Anisotropic Free Carriers





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

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

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

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

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

Δ-valley
Drude mass
Harmonic mean

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

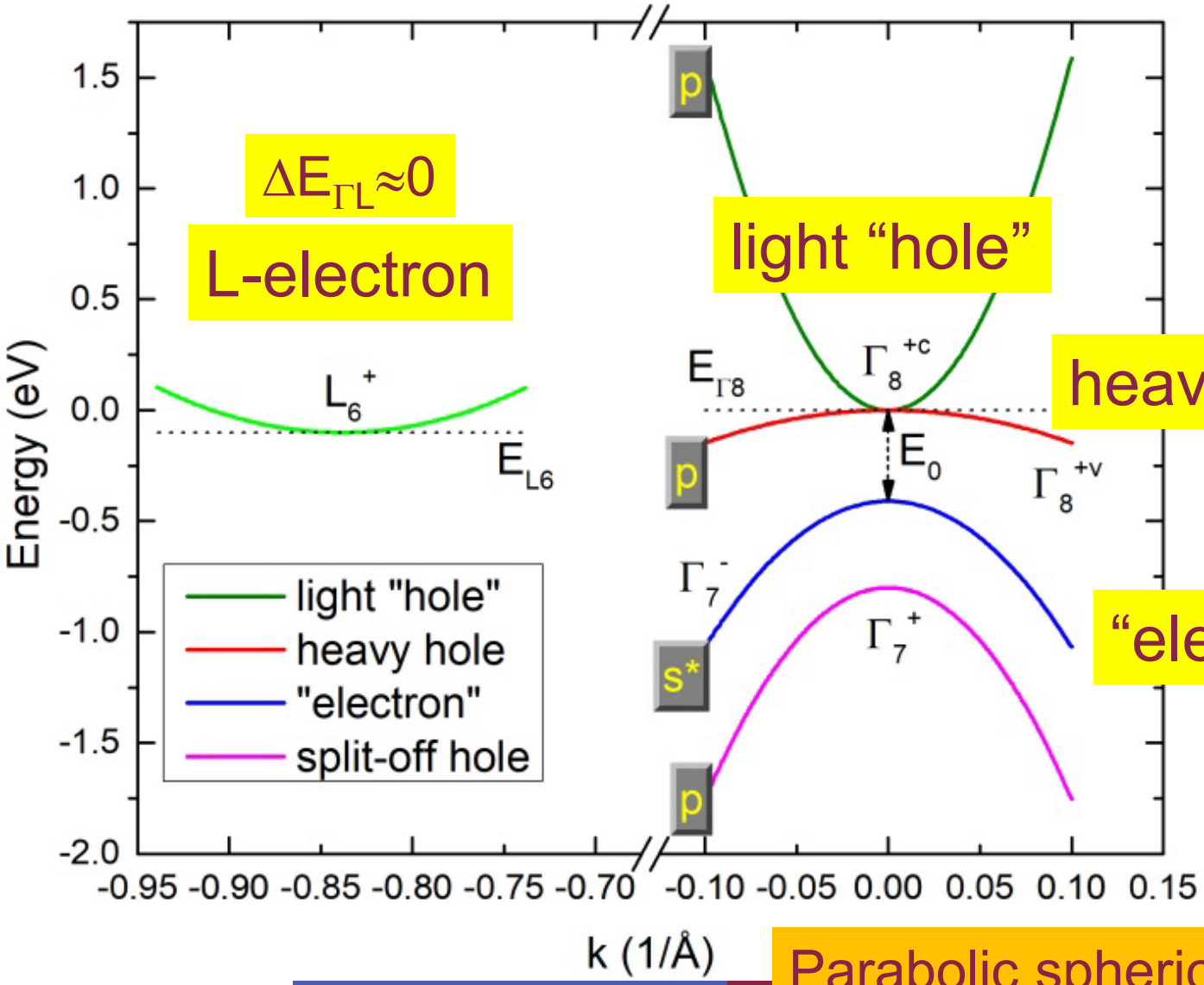
$$\omega_P^2 = \frac{nq^2}{m\varepsilon_0}$$

Charge density

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

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

Grey α-tin is even more complicated



light "hole"

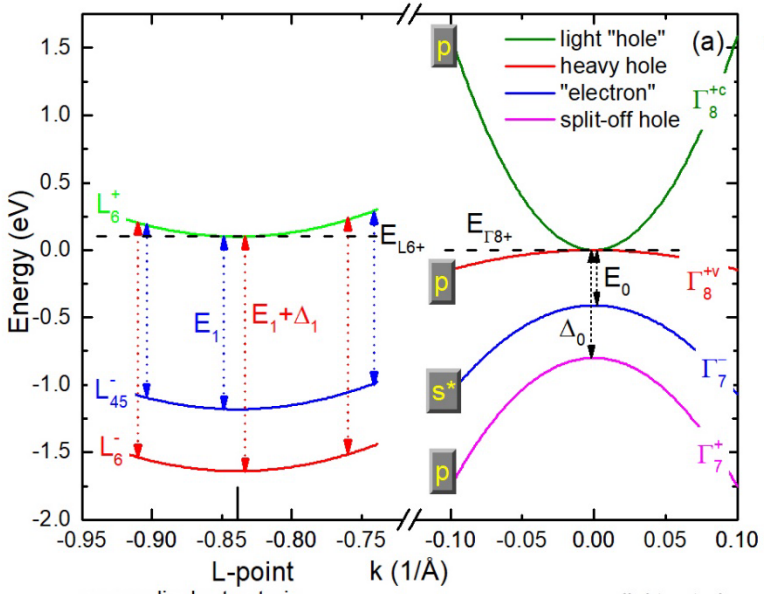
heavy hole

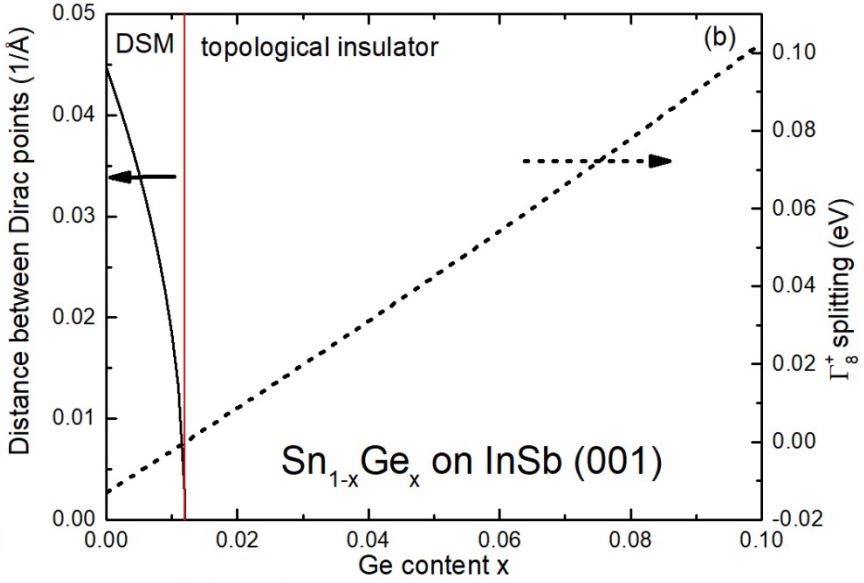
"electron"

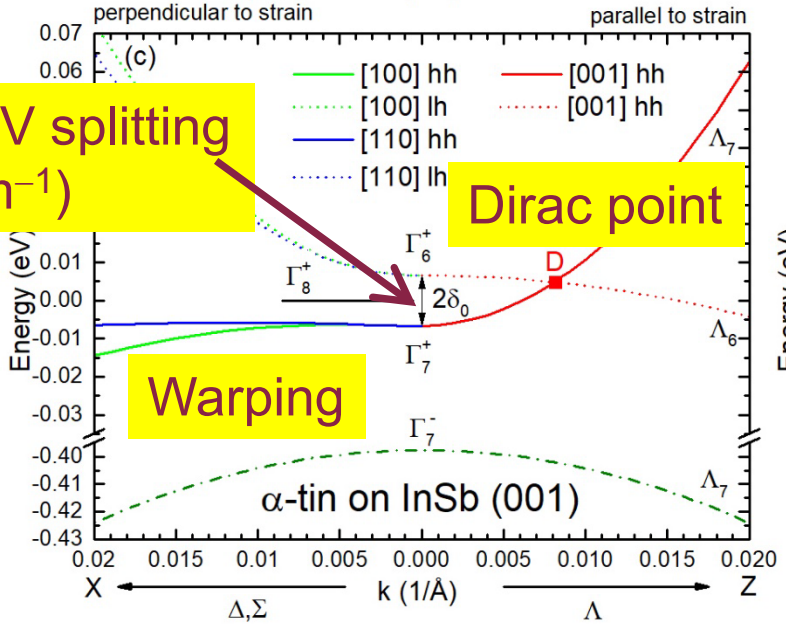
Parabolic spherical bands

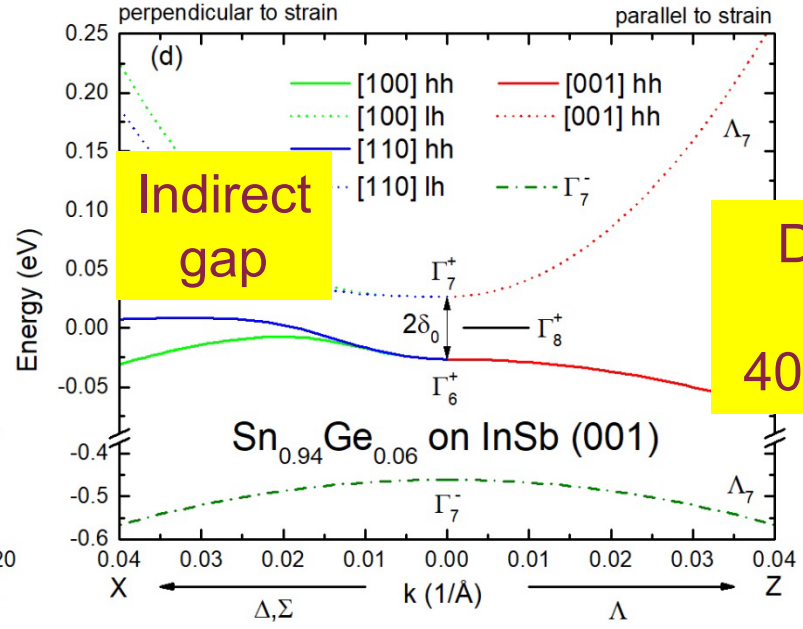
We need a model with THREE carrier species, one of them anisotropic.

Add strain and warping in $Sn_{1-x}Ge_x$ alloys









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

Dirac point

Warping

Indirect gap

Direct gap 400 cm^{-1}

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

Semiclassical Model of Electron Dynamics

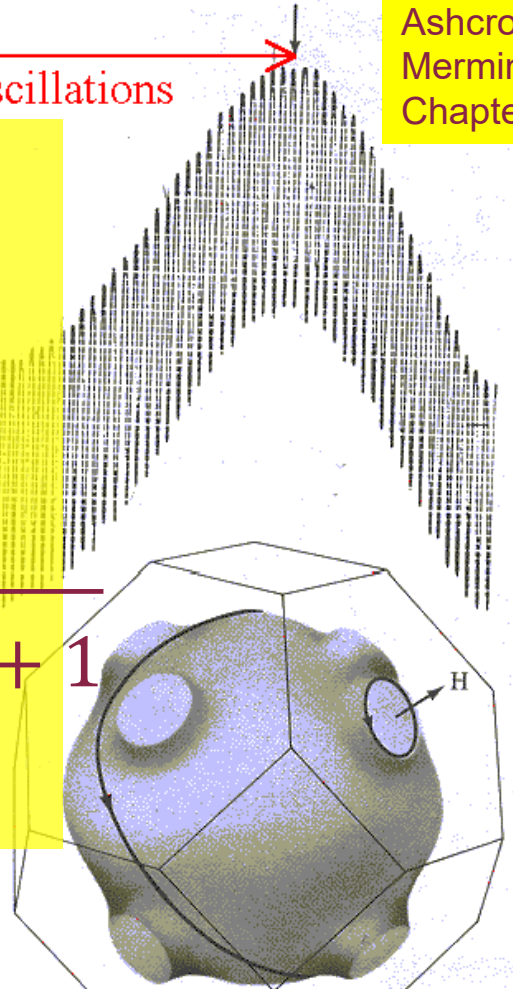
Ashcroft & Mermin, Chapter 12

$$\dot{\vec{r}} = \vec{v}_n\left(\vec{k}\right) = \frac{\hbar\vec{k}}{m} = \frac{1}{\hbar}\frac{\partial E_n\left(\vec{k}\right)}{\partial\vec{k}}$$

$$\hbar\dot{\vec{k}} = q\left[\vec{E}(\vec{r},t) + \vec{v}_n\left(\vec{k}\right)\times\vec{B}(\vec{r},t)\right]$$

$$f\left(E_n\left(\vec{k}\right)\right) = \frac{1}{\exp\left[\left(E_n\left(\vec{k}\right) - E_F\right)/k_BT\right] + 1}$$

$$m_c(E,k_z) = \frac{\hbar^2}{2\pi}\frac{\partial A(E,k_z)}{\partial E}$$



"Electrons move along curves given by the intersection of surfaces of constant energy with planes perpendicular magnetic field."

Is there an **optical analog to Shubnikov-de Haas effect? (Ge-Sn, GaSb)**

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

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**

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

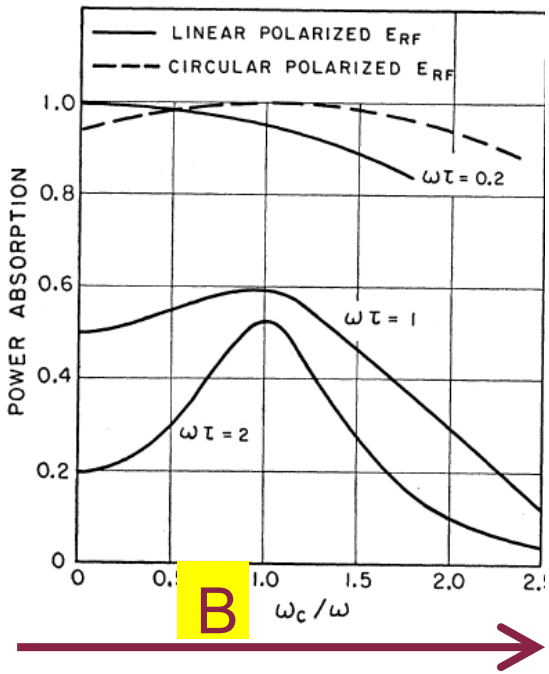
Cyclotron mass

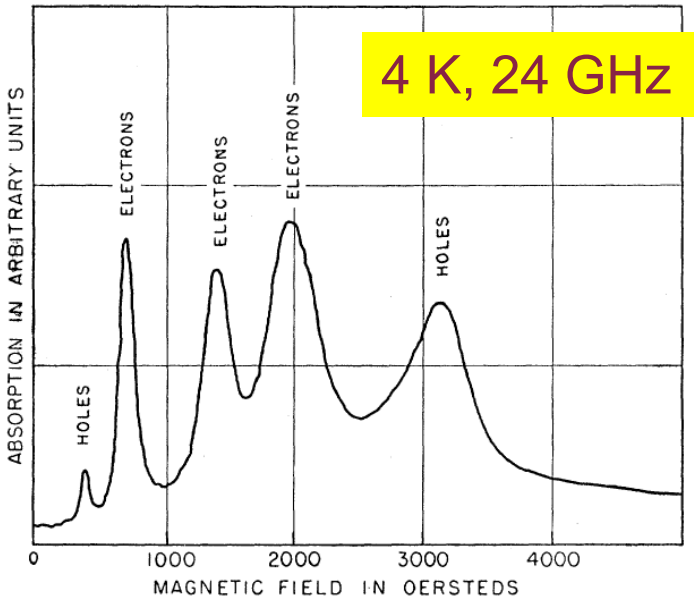
$$m_c = \sqrt{\frac{\det \overleftrightarrow{m}}{\vec{b} \cdot \overleftrightarrow{m} \cdot \vec{b}}}$$

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

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

Cyclotron Resonance

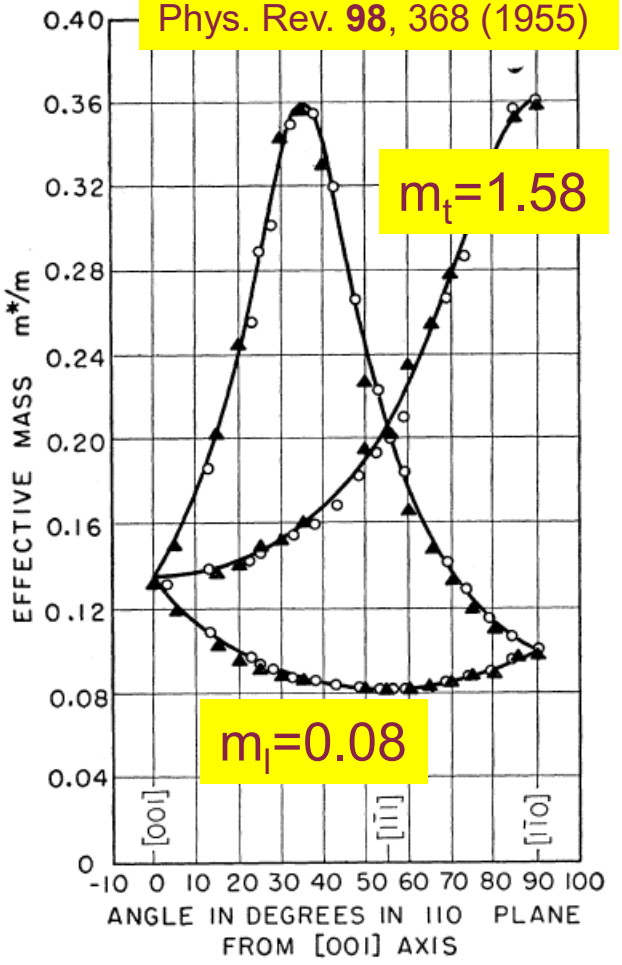




4 K, 24 GHz

10000 Oe=1 T

Dresselhaus, Kip, Kittel, Phys. Rev. **98**, 368 (1955)



$$\omega_c = \frac{qB}{m}$$

Measure microwave absorption as a function of the magnetic field. Information about VB warping.

Insulators

CB: Conduction band (empty)

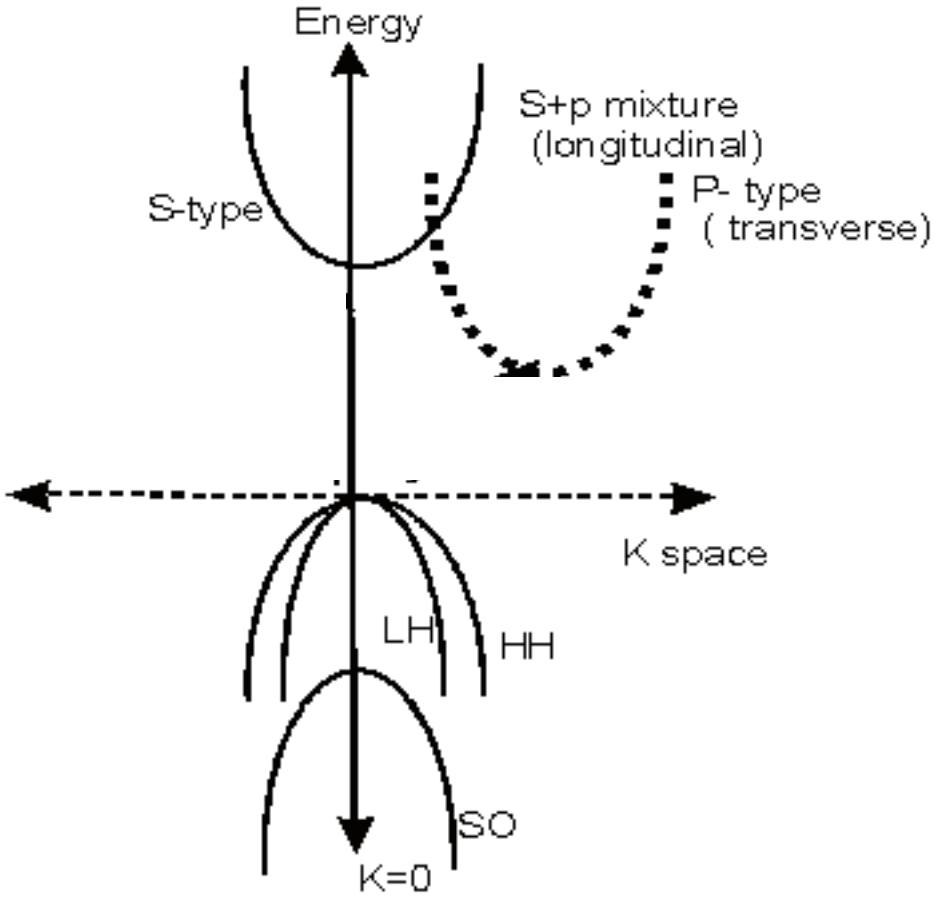
Transparent region below the band gap (VIS/UV).

Forbidden "energy gap".

Semiconductors are insulators with small band gap (IR).

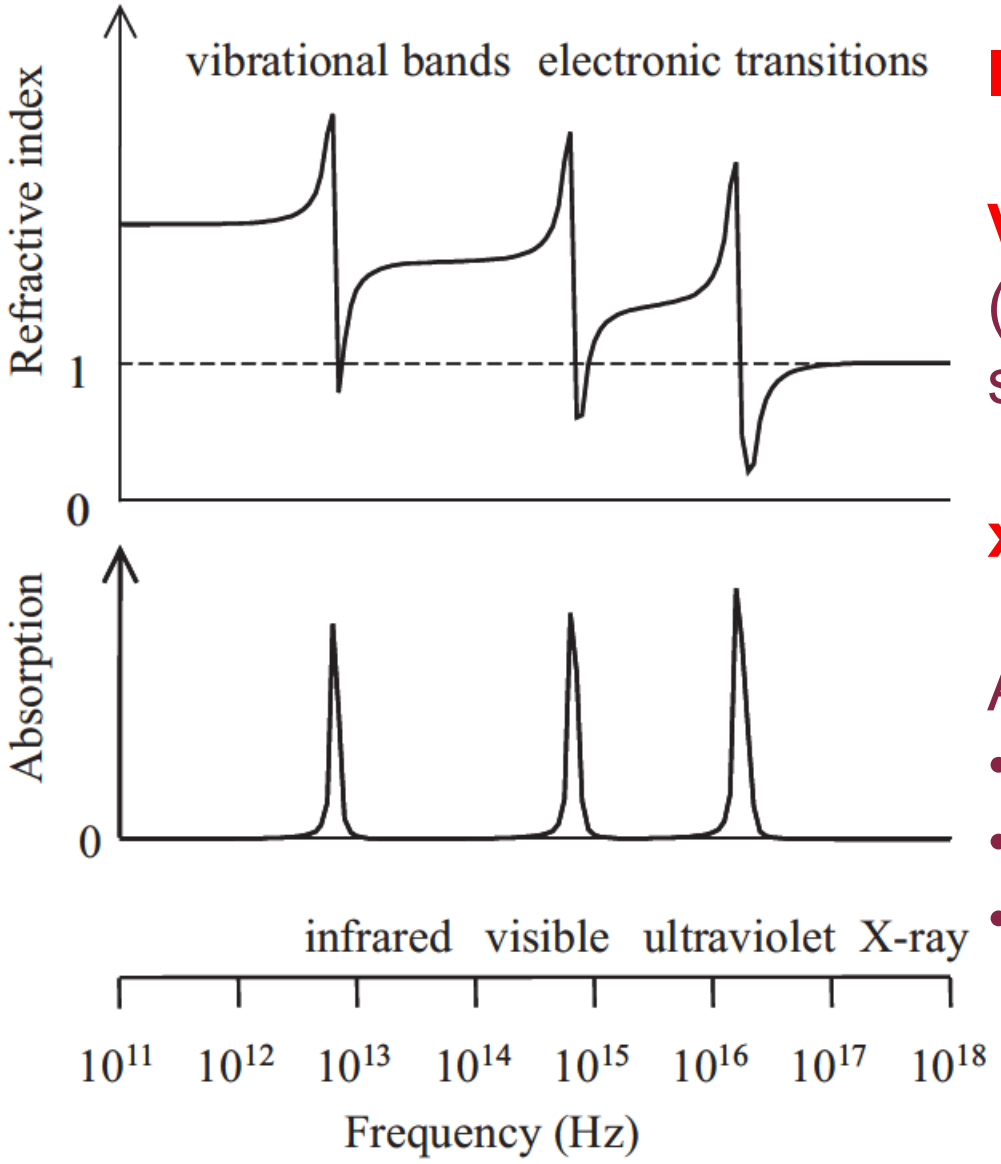
Valence band filled.
Conduction band empty.

Filled bands do not carry current



VB: Valence band (filled)

Multiple Lorentz Contributions: IR, UV, x-ray



IR: lattice vibrations

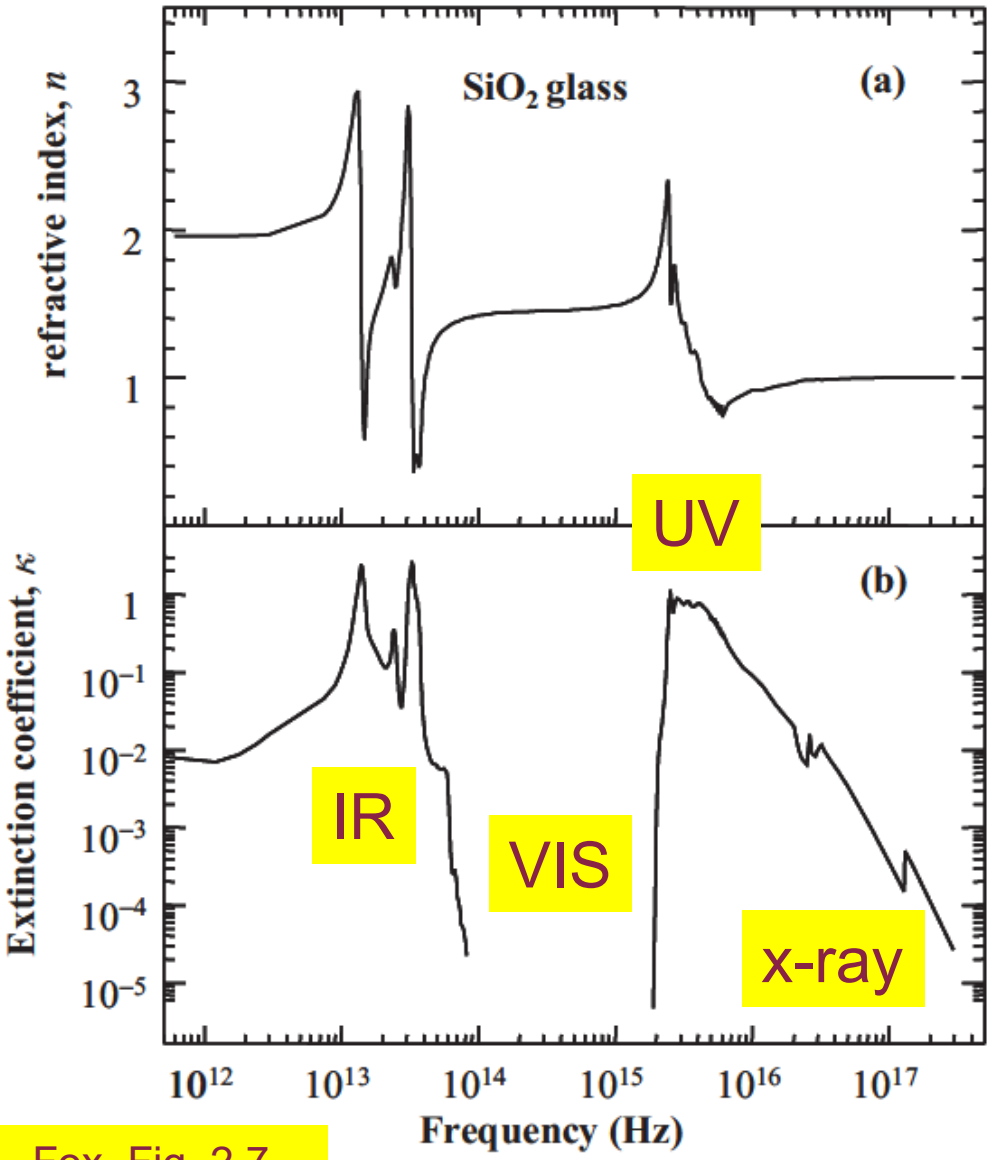
VIS/UV: valence electrons
(usually broadened by band structure effects)

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



Fox, Fig. 2.7

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

Poles, Sellmeier Approximation

Set g=0 far from resonance.
Lorentz oscillator becomes
a **pole**

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

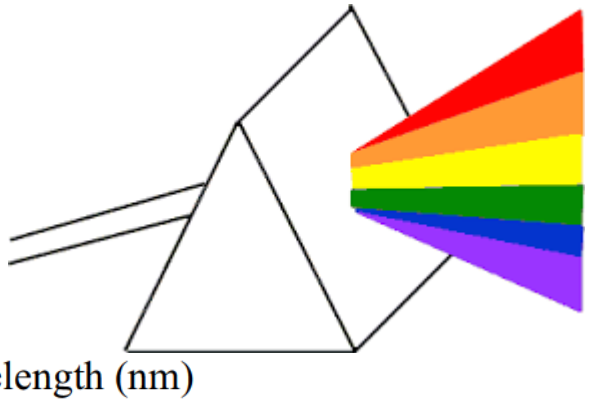
Rewrite as a function of l

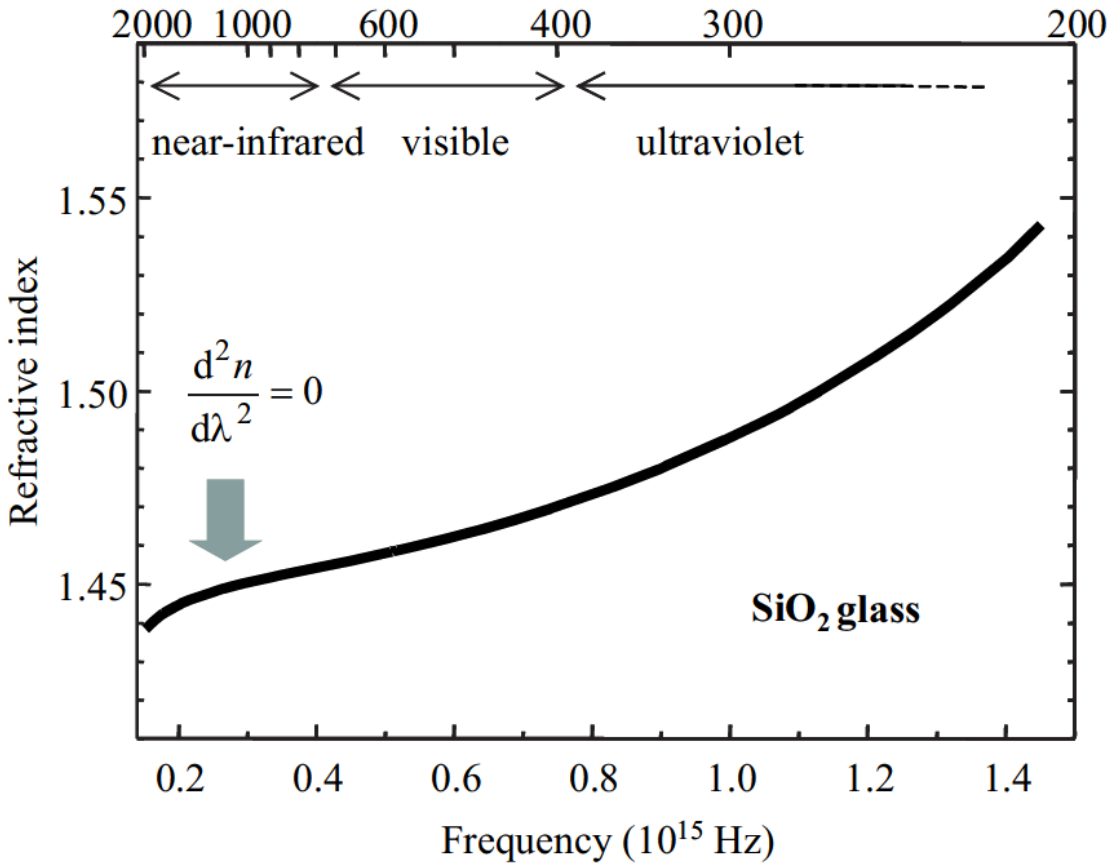
$$\varepsilon(\lambda) = 1 + \frac{B\lambda^2}{\lambda^2 - C}$$

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

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

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

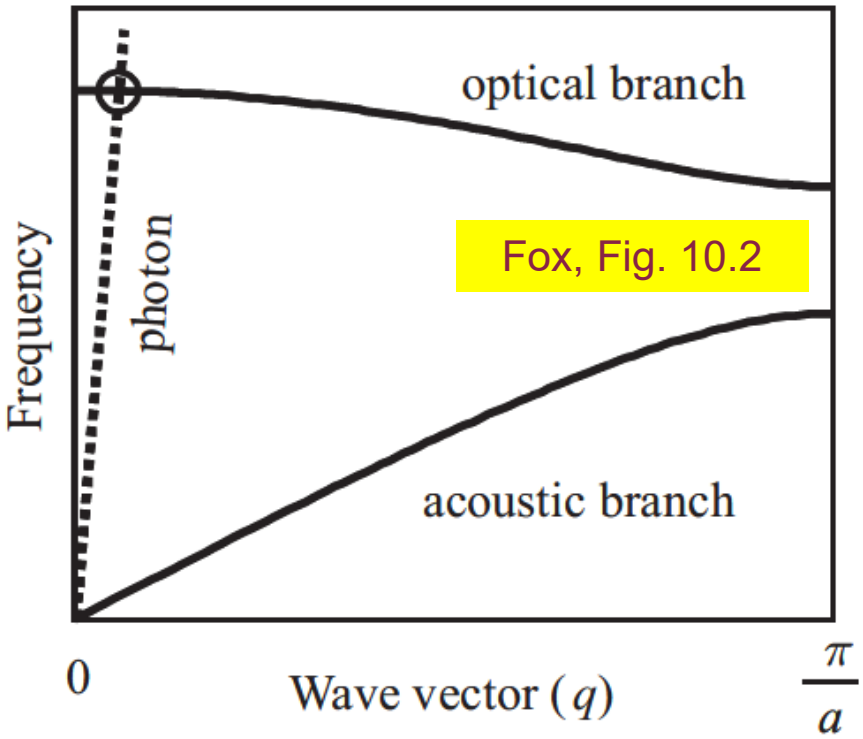
Comments:

The Cauchy equation does not include absorption and therefore is not Kramers-Kronig consistent. Absorption is often included with an Urbach tail

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

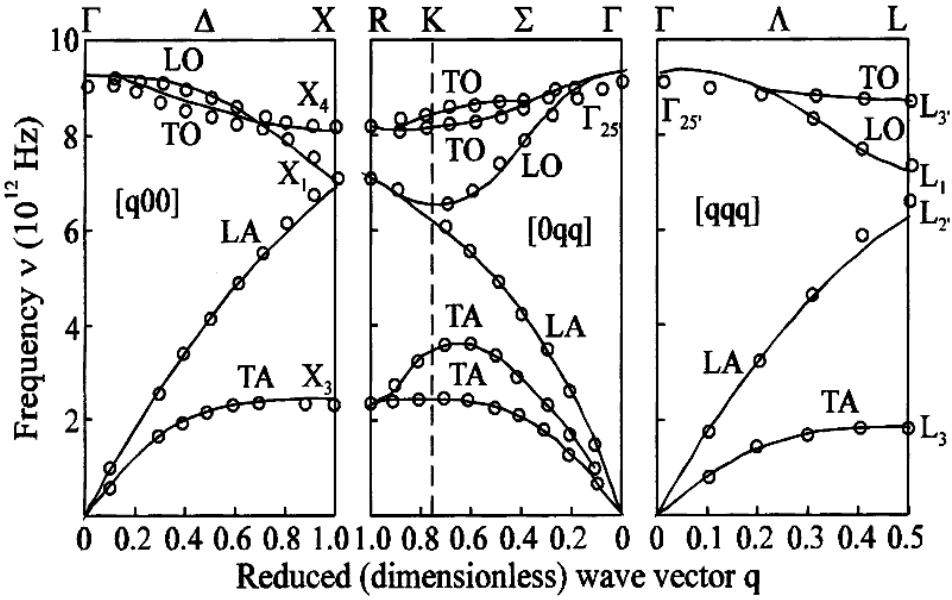
but this still does not make it Kramers-Kronig consistent. Not recommended, use Tauc-Lorentz model instead.

Insulator Phonon Spectra (Ge)



Fox, Fig. 10.2

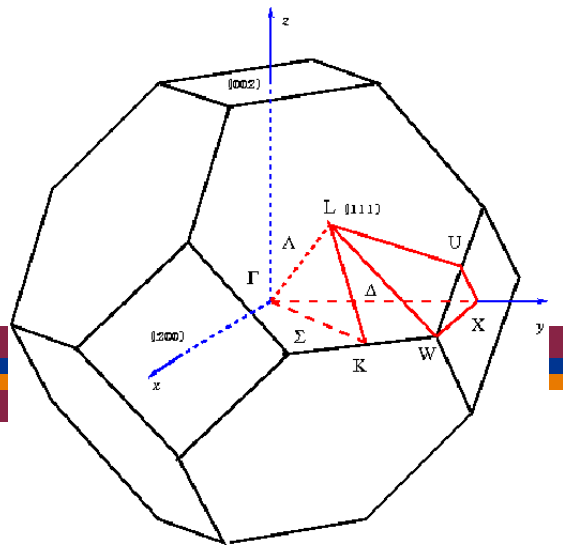
Neutron scattering



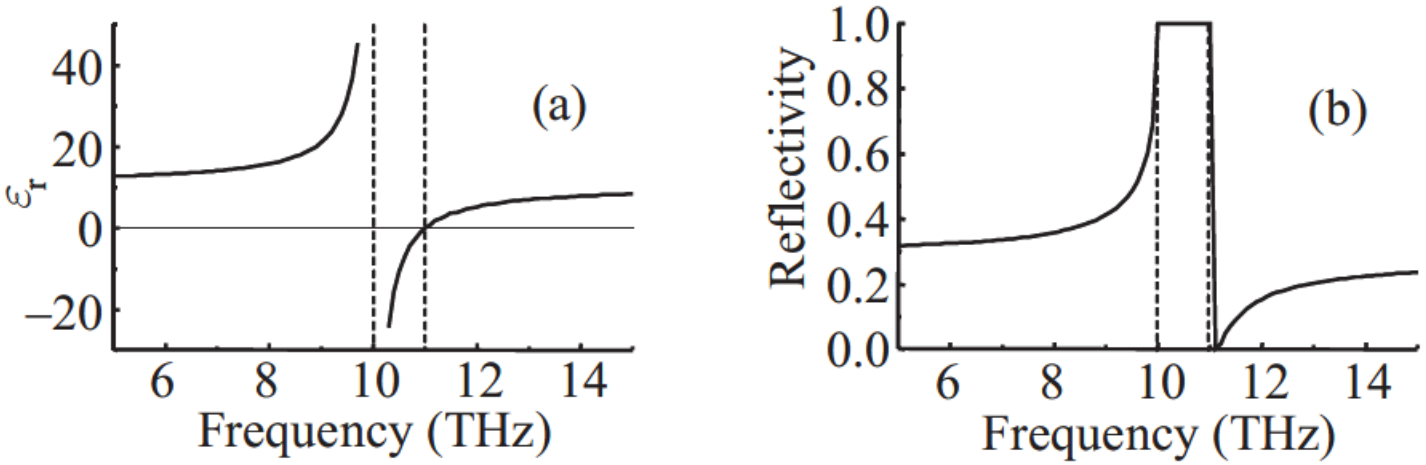
Inelastic neutron scattering measures entire phonon dispersion.

Optical experiments only probe k≈0, because λ≫a.

Ge is IR-inactive (no dipole moment)



Infrared Lattice Vibrations (Lorentz model)

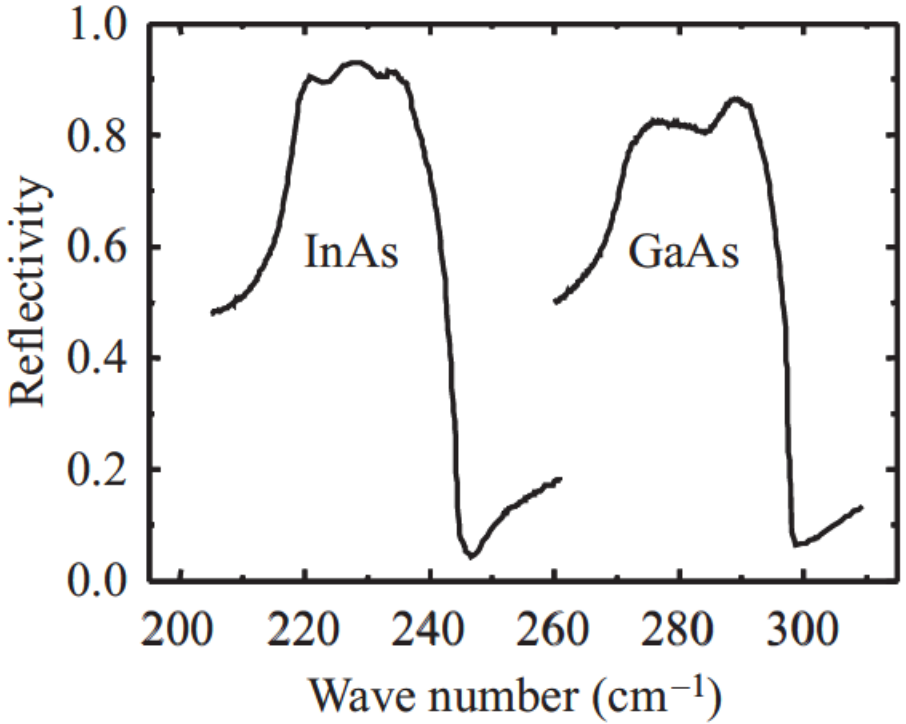


Fox, Fig. 10.4

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

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)



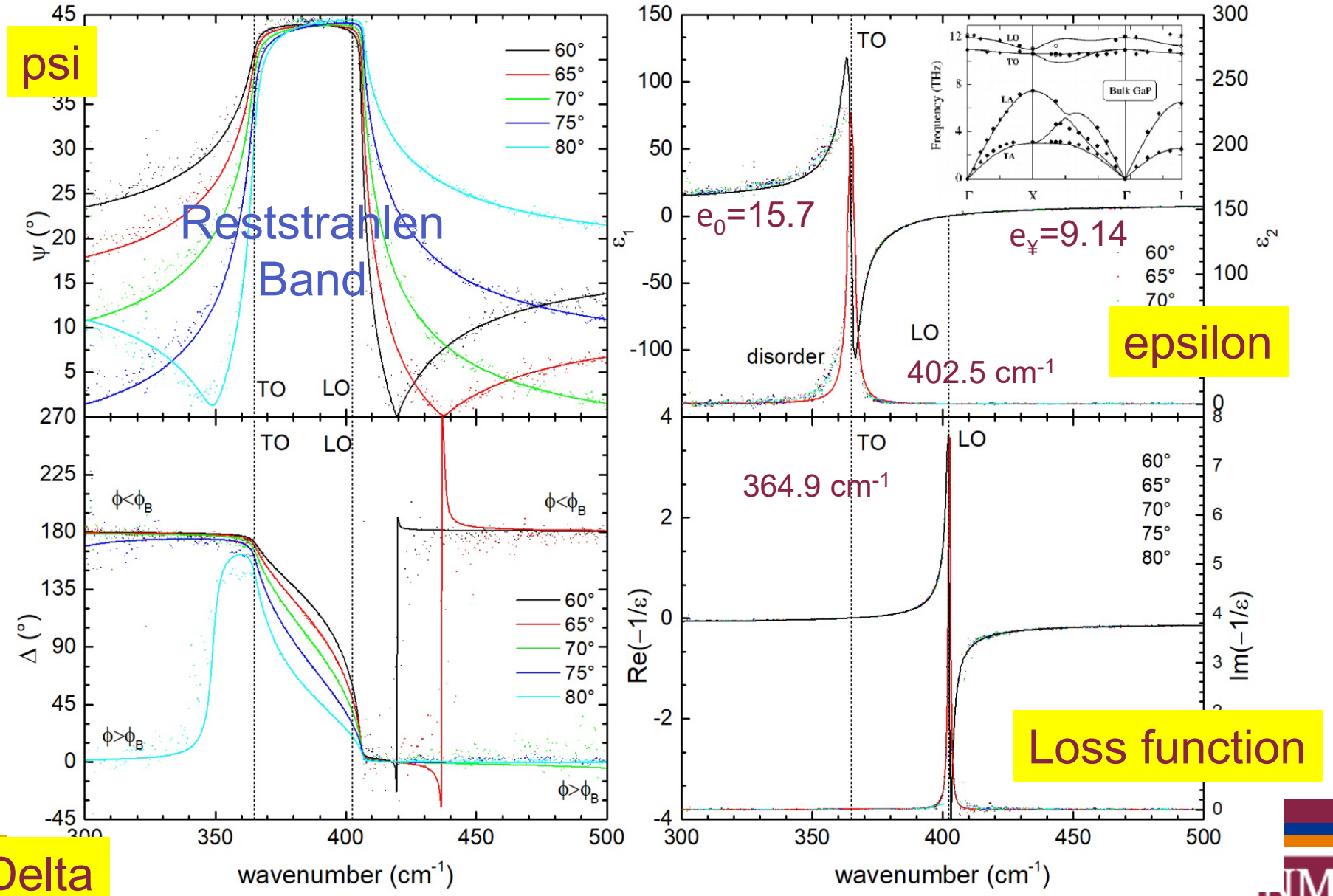
Reststrahlen Band

Fox, Fig. 10.5

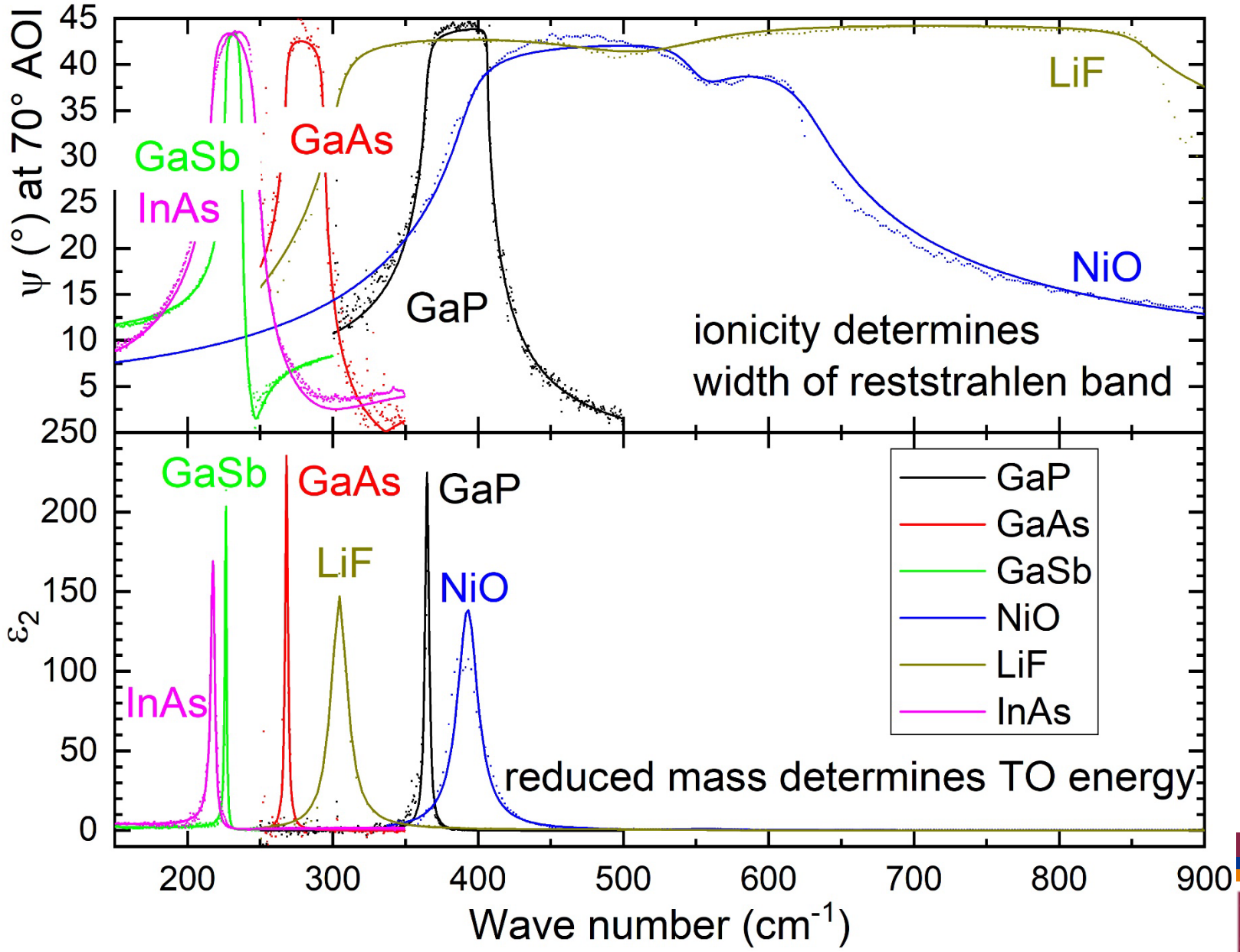
In polar materials (Born effective charge), TO and LO modes are split.
TO: transverse optical (atomic displacement perpendicular to **k**)
LO: longitudinal optical (atomic displacement along **k**)
ε_2 has peak at TO frequency
ε_1 is negative from TO to LO frequency (reflectance is 1)
Restrahlen band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

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

Infrared Lattice Vibrations in GaP



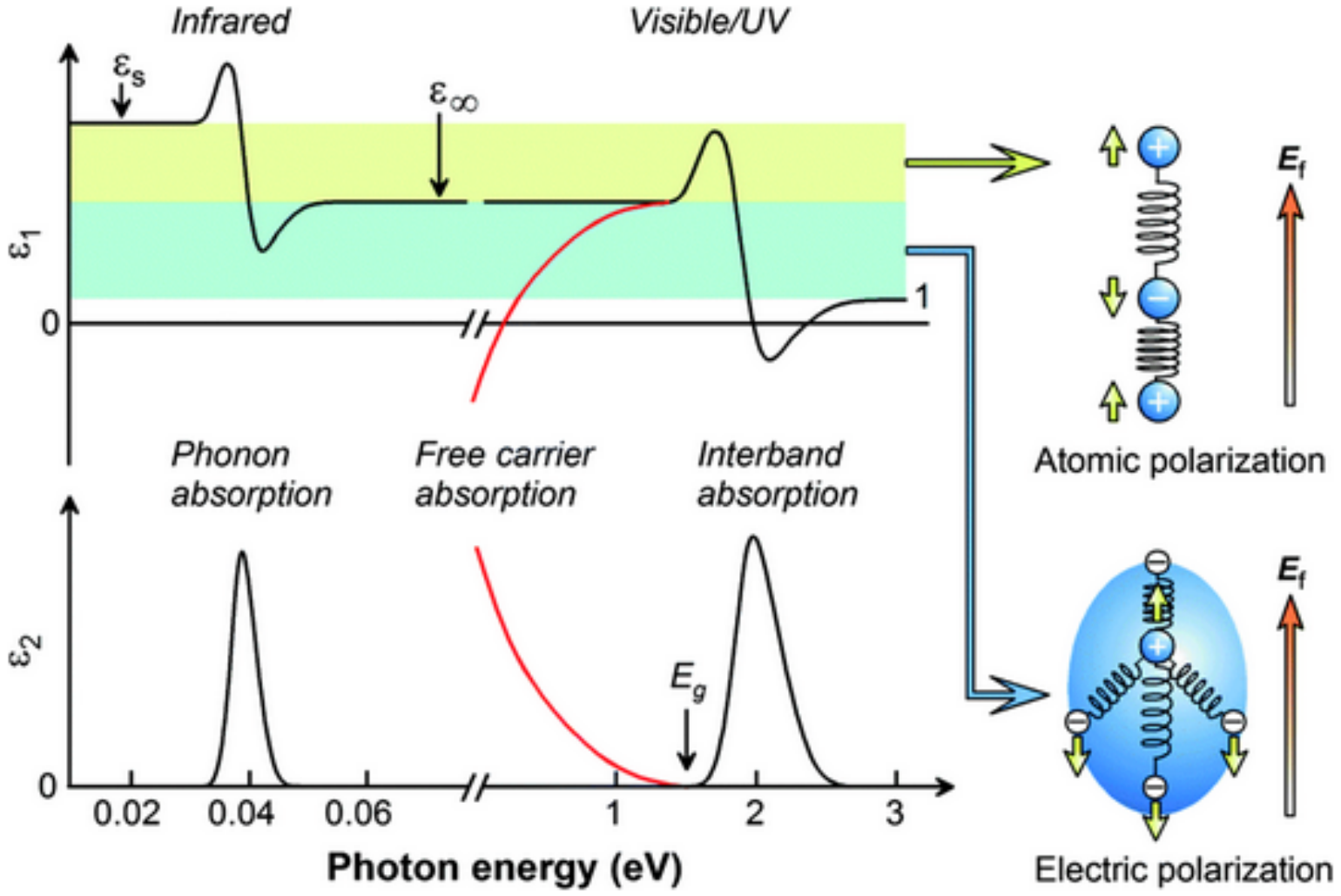
Infrared Lattice Vibrations (Lorentz model)



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

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

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).

$$0 = \varepsilon_\infty + \frac{A\omega_0^2}{\omega_0^2 - \omega_{LO}^2}$$

or

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

LST relation

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

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

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

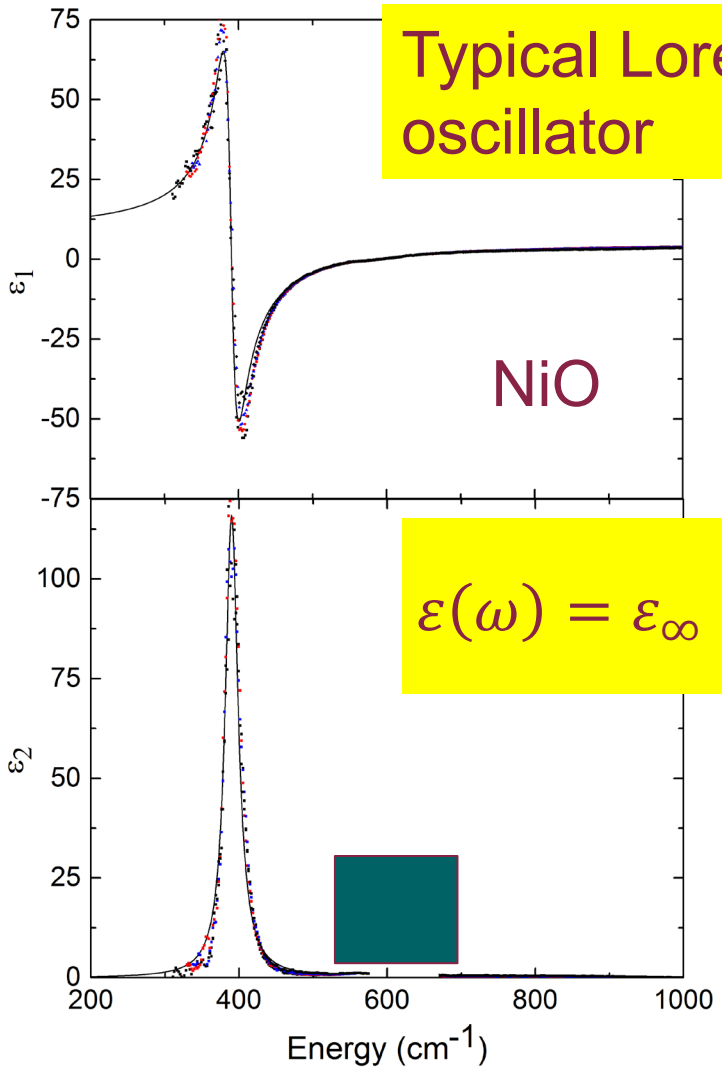
Mathias Schubert, Phys. Rev. Lett. **117**, 215502 (2016)

Amorphous materials and liquids

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

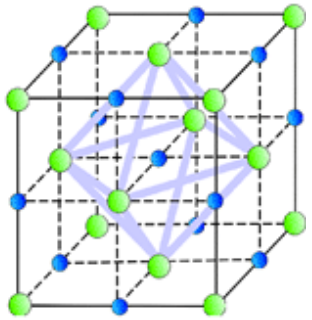
A.J. Sievers and J.B. Page, Infrared Physics **32**, 425 (1991)

Infrared Lattice Vibrations (NiO)



Typical Lorentz oscillator

NiO or NaCl or LiF: Rocksalt lattice



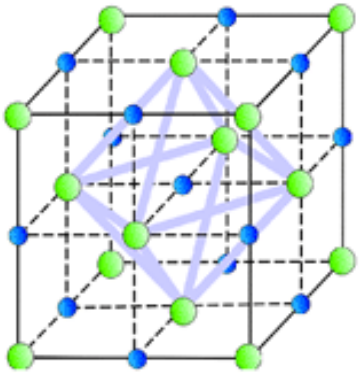
- Ni^{2+}-O^{2-} bonds are polar.
- Ni-O vibration has dipole moment.

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

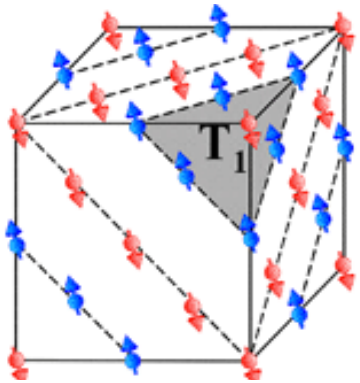
FTIR ellipsometry

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.

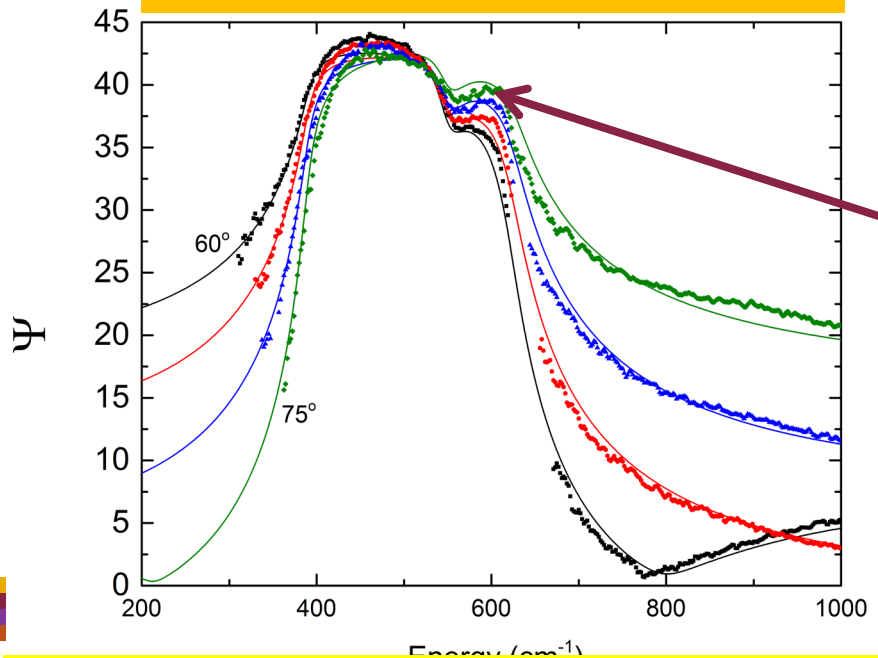


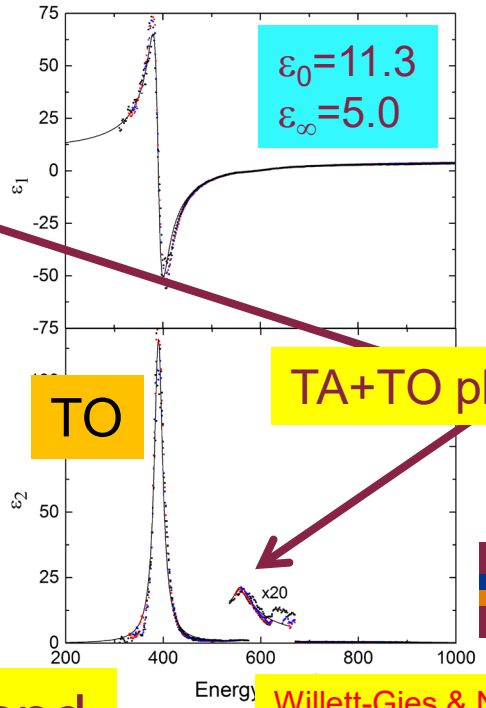
NiO cell



Rooksby, Nature, 1943

NiO Reststrahlen Band

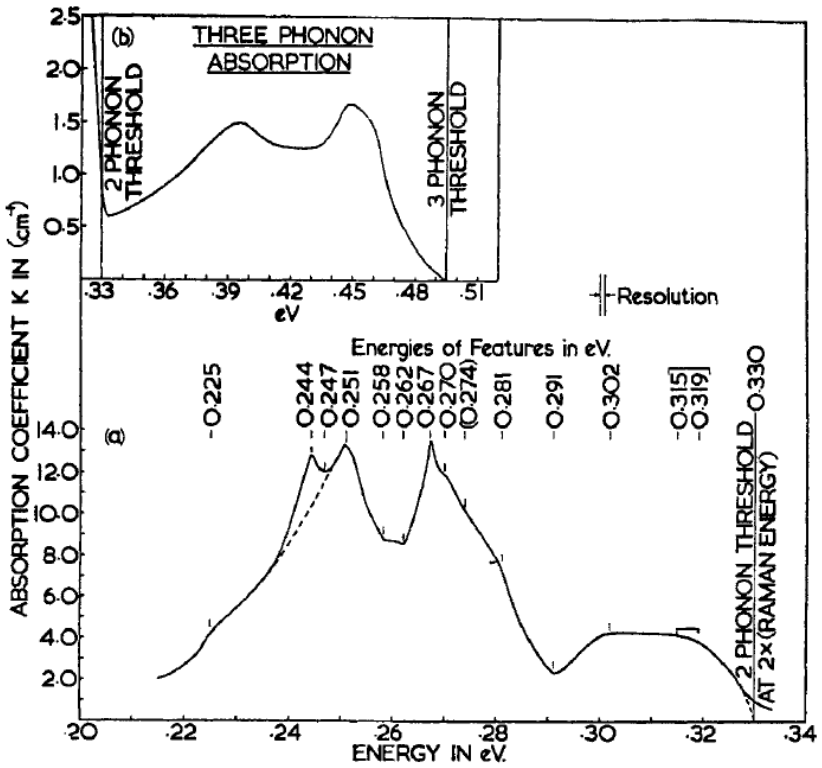


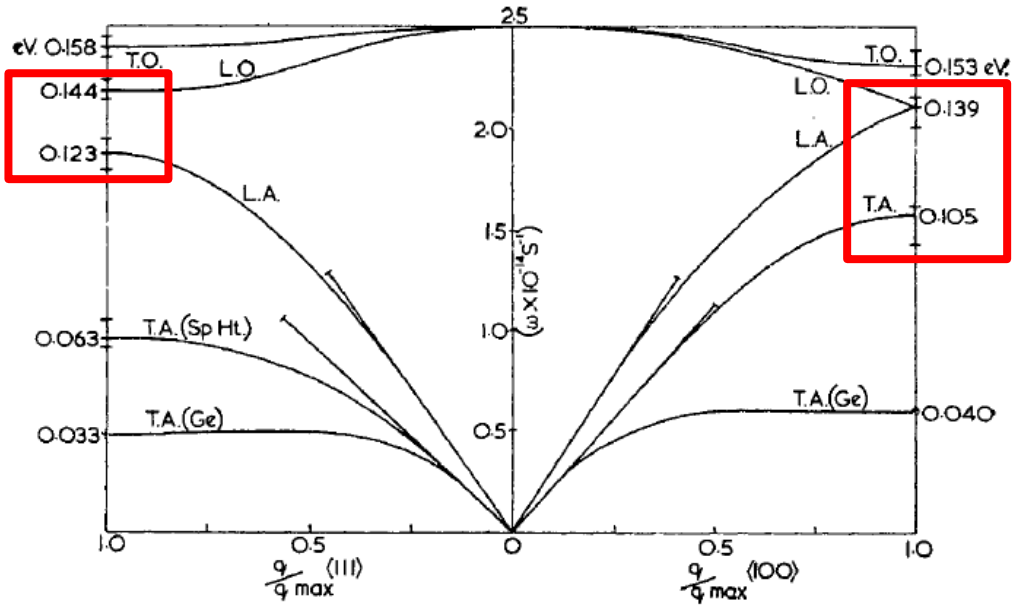


Absorption in the restrahlen band

Willett-Gies & Nelson, JVST A **33**, 061202 (2015)
Also Humlicek PSSB **215**, 155 (1999) for LiF.

Two-phonon absorption (diamond)



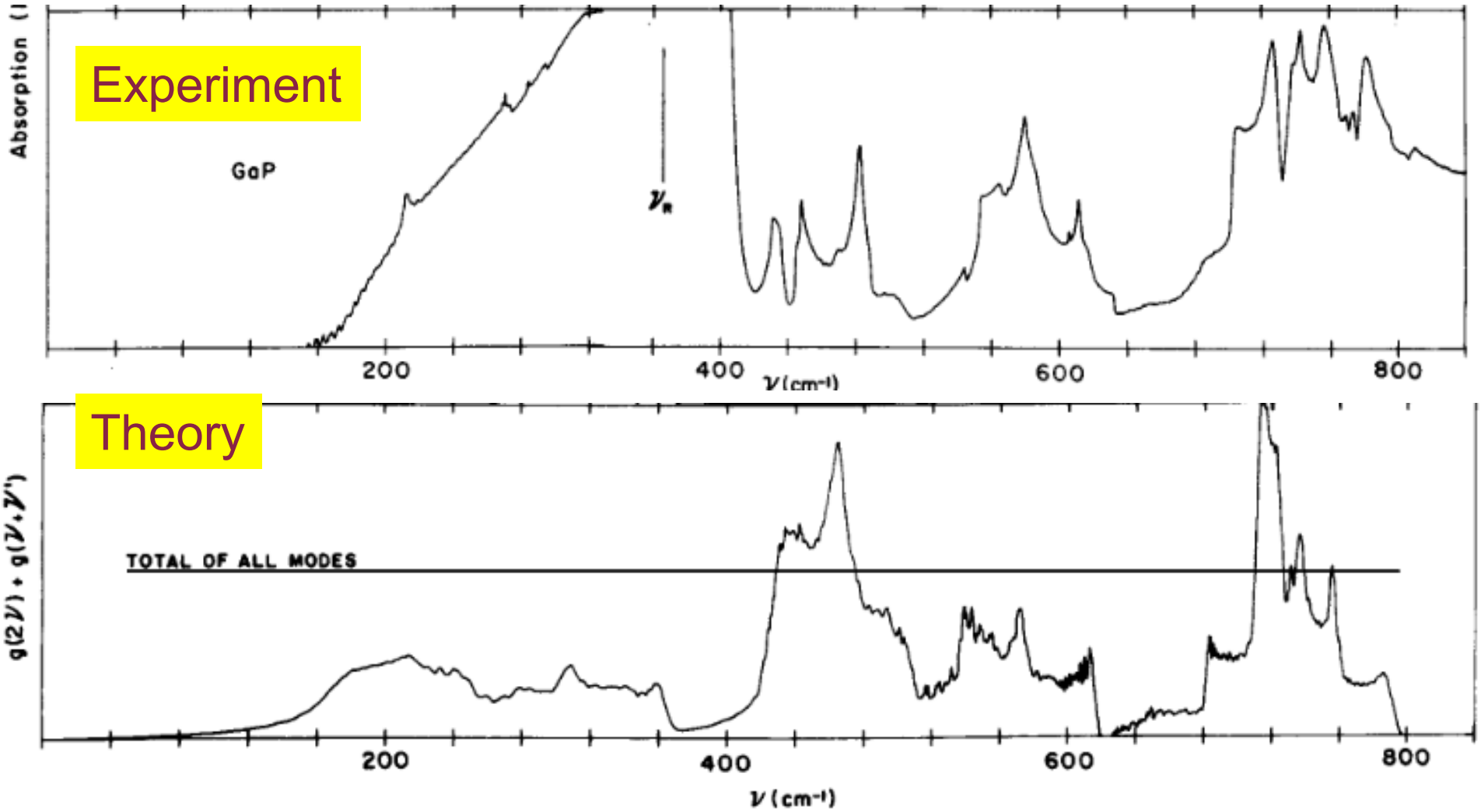


TA+LAO (X): 105+139=244 meV

A single photon excites two phonons. Energy and wave vector conserved.
Two-phonon absorption is weak (lower probability than TO phonon absorption).
Most likely to occur near Brillouin zone boundary (high density of states).

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

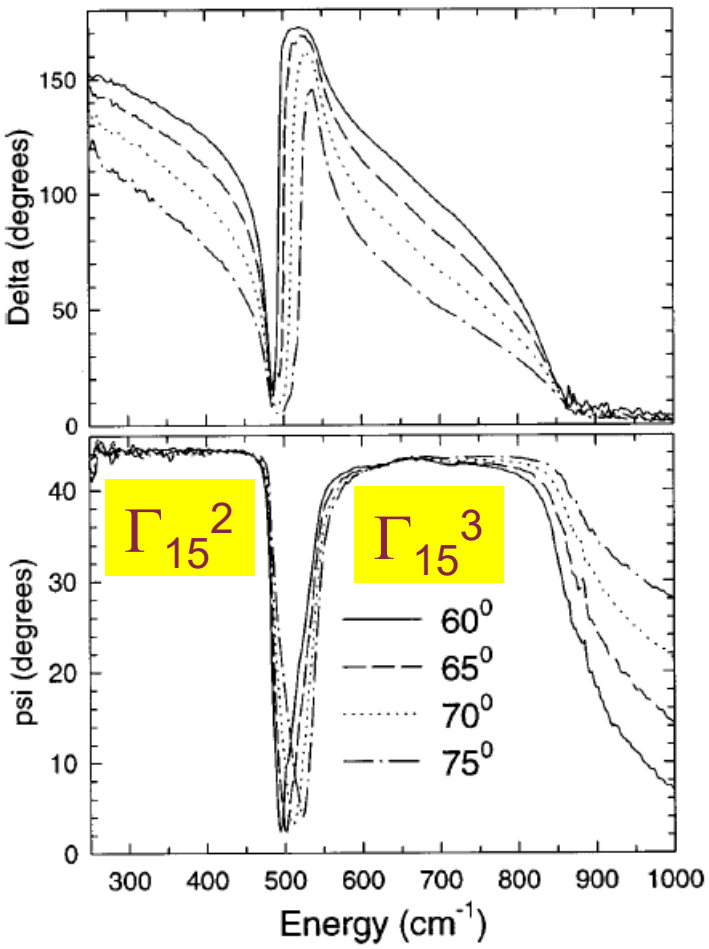
Two-phonon absorption (GaP)

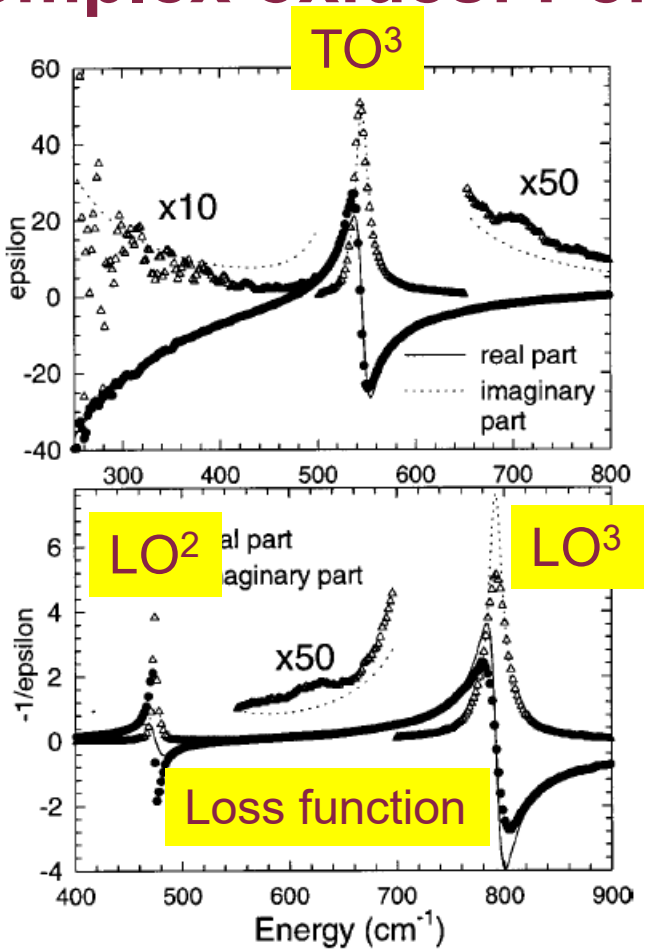


Alternative to neutron scattering to determine zone-edge phonon energies.

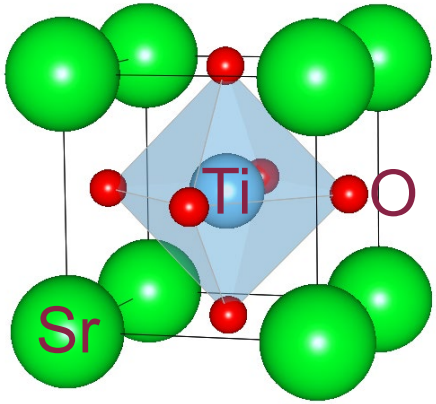
E.S. Koteles, Solid State Commun. **19**, 221 (1976)

Phonons in complex oxides: Perovskites





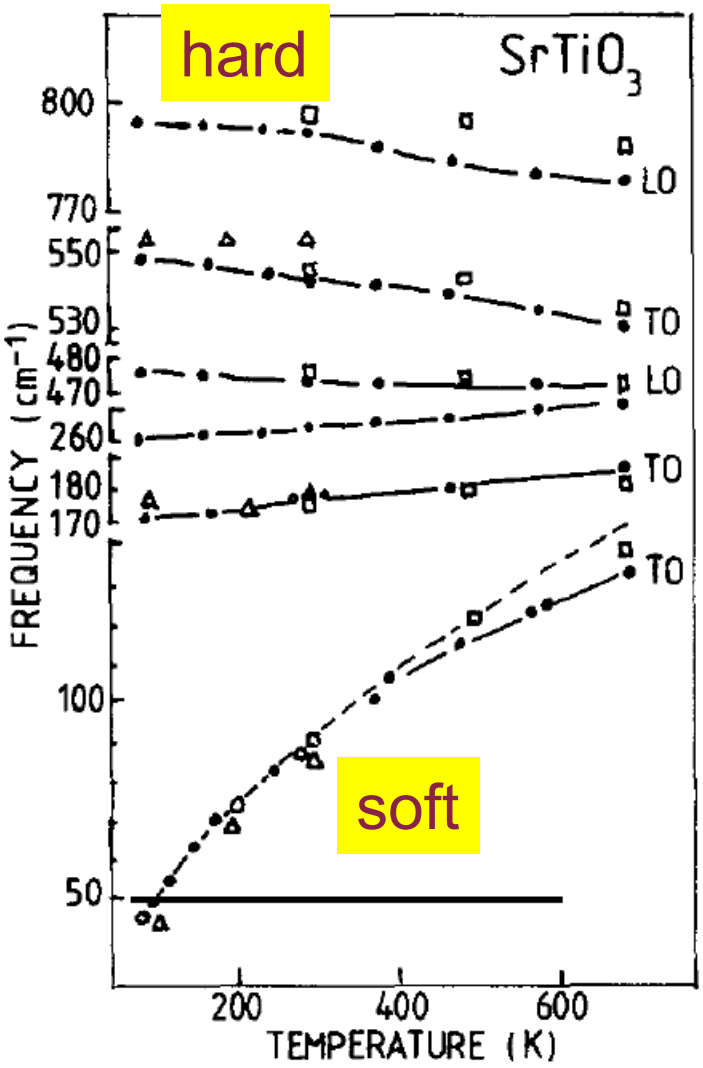
ε shows TO phonons



–1/ε shows LO phonons

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

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)

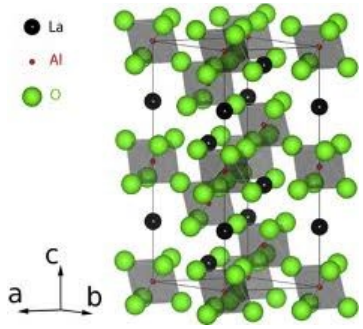
$SrTiO_3$ is nearly ferroelectric (T_C~0 K).

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

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

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

Phonons in more complex oxides (bulk)



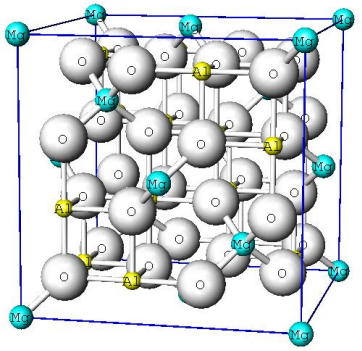
$LaAlO_3$

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

Space Group

O_h^7 or $Fd3m$

$MgAl_2O_4$



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

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

Raman Active: A_{1g}, $3A_{2u}$, $4E_g$

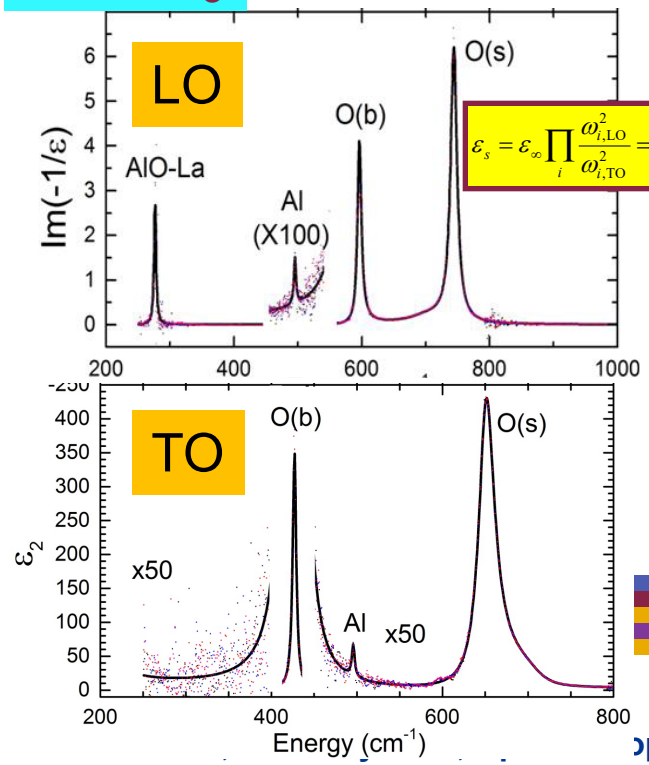
IR Active: $5E_u$

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

Raman Active: A_{1g}, E_g, $3T_{2g}$

IR Active: $4T_{1u}$

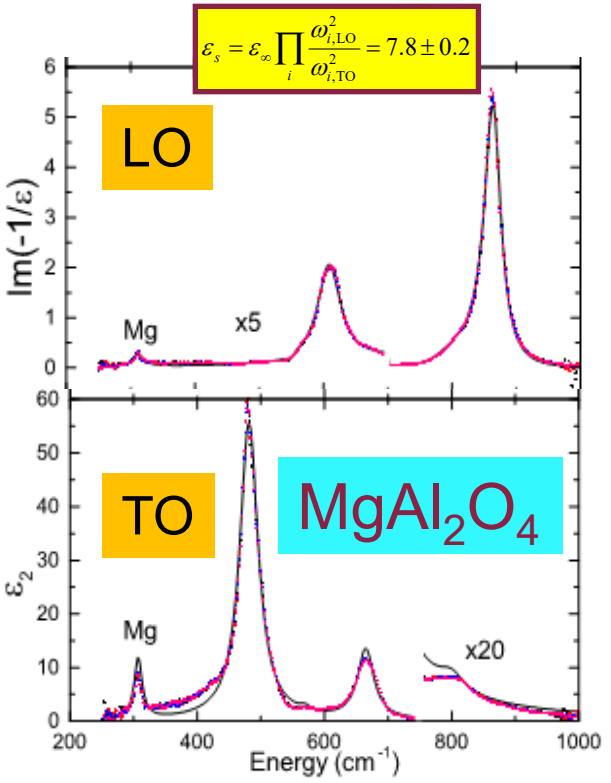
$LaAlO_3$



FTIR Ellipsometry

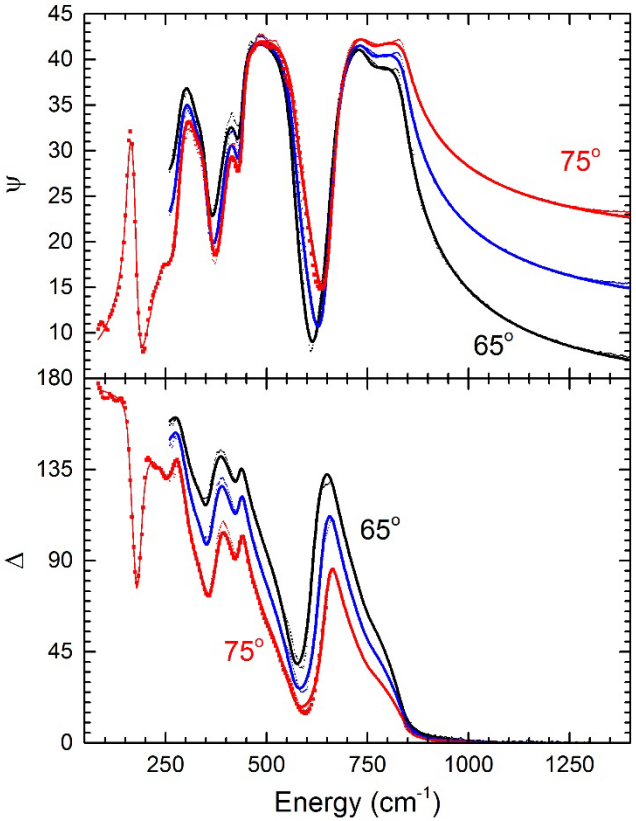
Loss function:
LO phonons

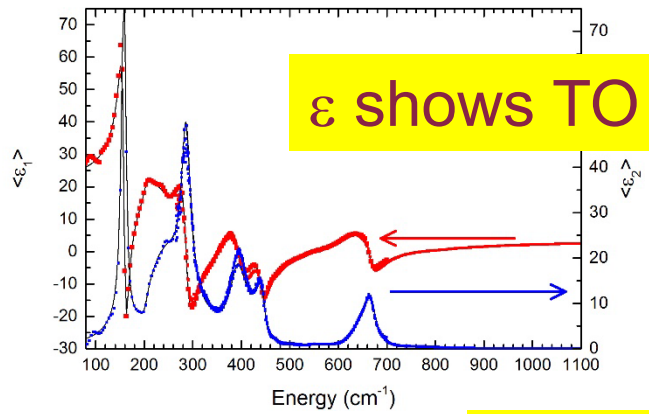
Dielectric function:
TO phonons



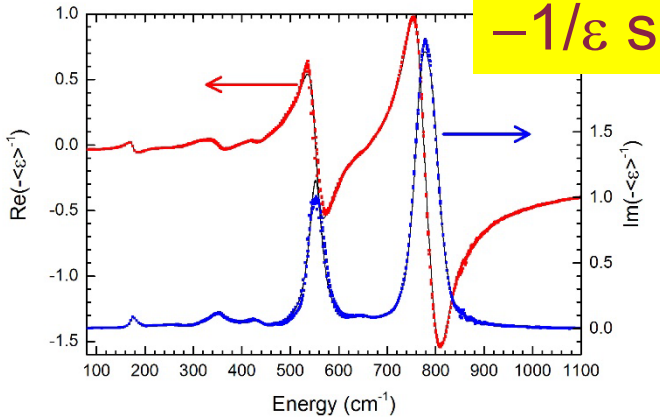
Willett-Gies, Thin Solid Films, 2013
Zollner, Thin Solid Films, 2013

Far-infrared ellipsometry (bulk LSAT)

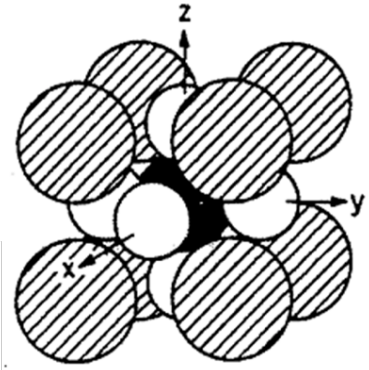




ε shows TO phonons



–1/ε shows LO phonons

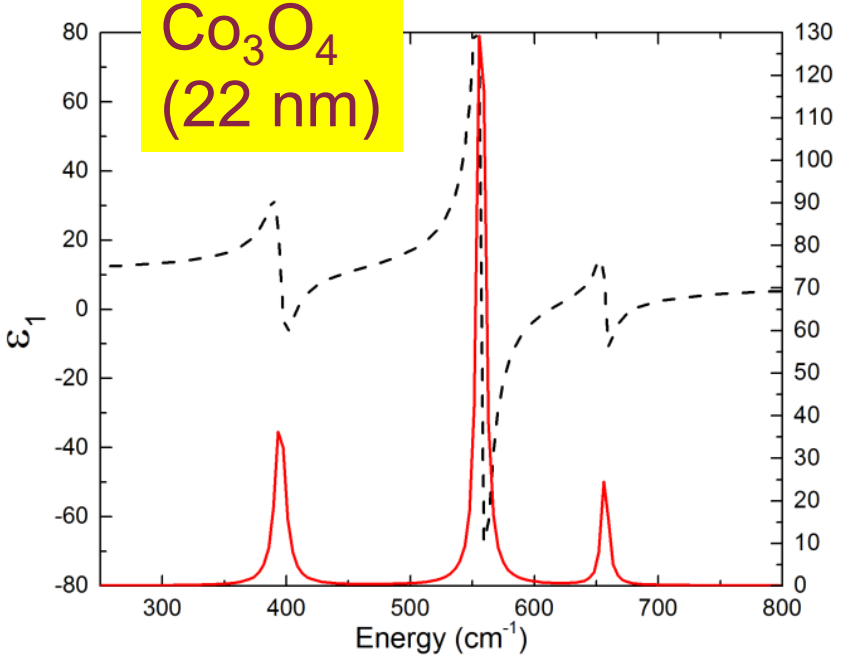


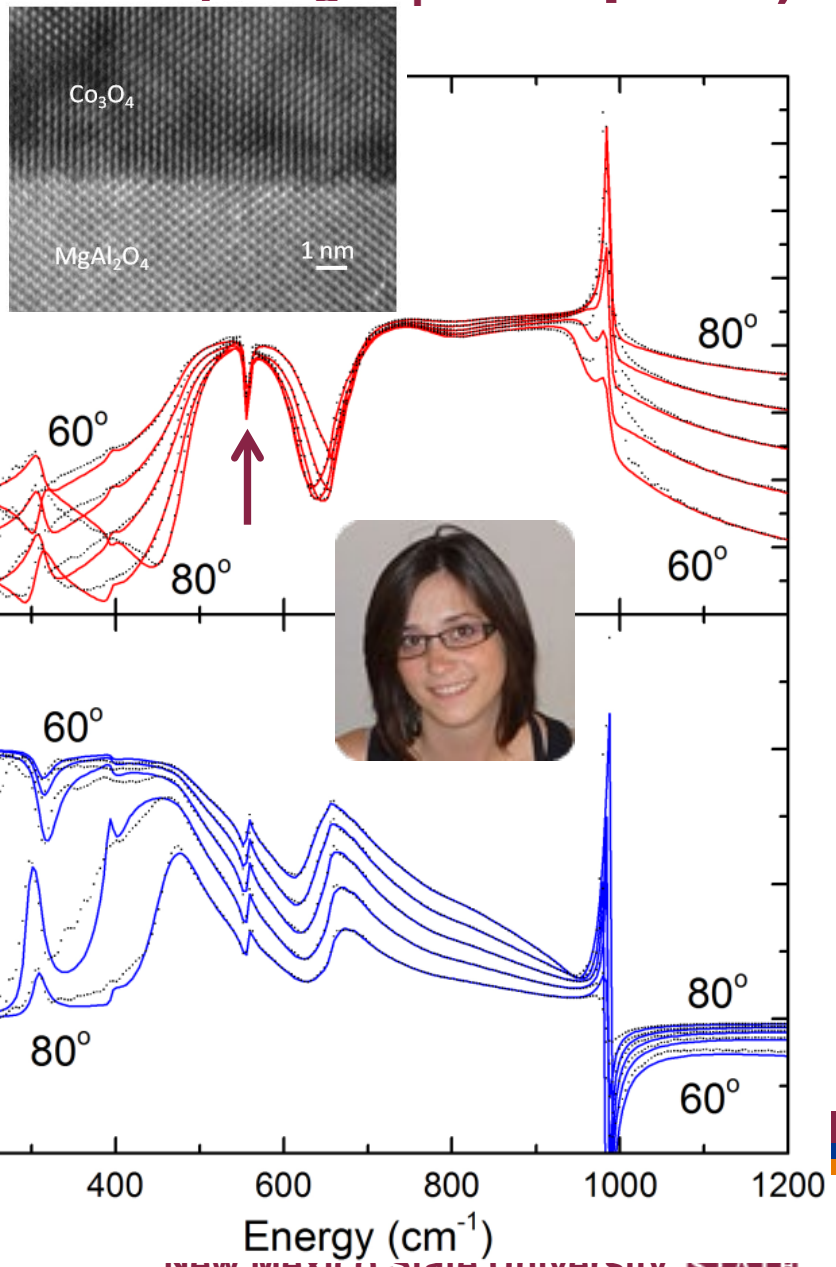
Disordered double perovskite $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.35}$
Many phonon modes. Several reststrahlen bands.

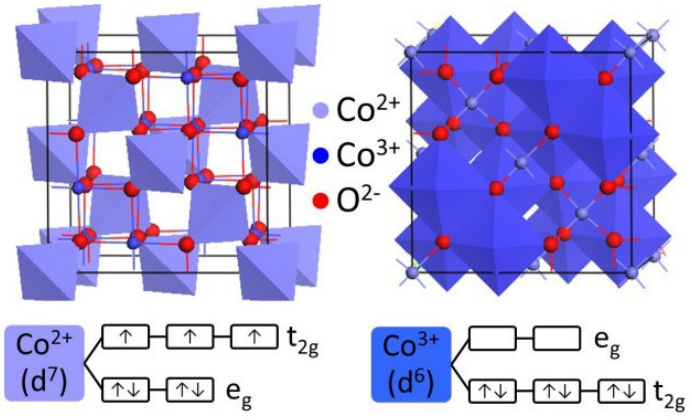
T.N. Nunley, JVSTA 34, 051507 (2016)

Phonons in more complex oxides (Co_3O_4 on spinel)

Co_3O_4 (22 nm)







Cubic field splitting

K.N. Mitchell, Kormondy, SZ, Demkov, JAP **115**, 243708 (2014)

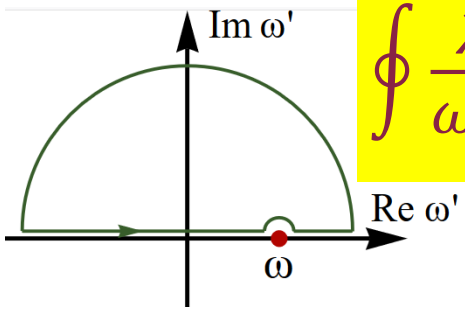
Kramers-Kronig Relations

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

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:



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

Cauchy

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

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

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

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.

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

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

Dielectric tensor symmetric (B=0)

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

Also from energy density.

Passive materials (no optical gain)

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

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

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

R. P. Lowndes,
PRB **1**, 2754 (1970).

γ>0 (causality)

Works well for phonons and plasmons.

Also: 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

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

R. P. Lowndes, PRB **1**, 2754 (1970).

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

Berreman & Unterwald, Phys. Rev. **174**, 791 (1968).

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.