Lecture Series: Part 1
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.

NM STATE BE BOLD. Shape the Future.

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Biography

Regensburg/Stuttgart
Germany

Motorola (Mesa, Tempe)
Arizona, 1997-2005

Freescale, IBM
New York, 91-92; 07-10

NMSU
Las Cruces, NM
Since 2010

Motorola, Freescale
Texas, 2005-2007

Where is Las Cruces, NM ???

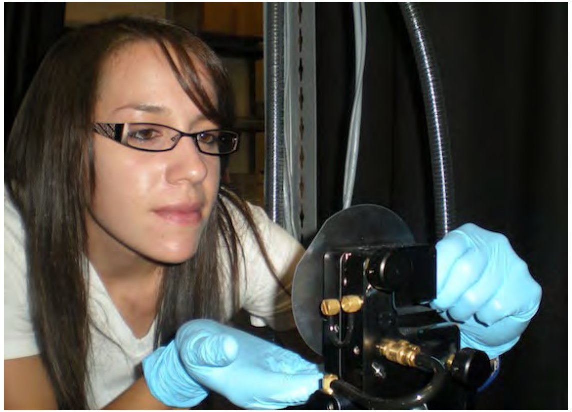
White Sands NP

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 Lattice Vibrations (Optical Phonons)
3. Interband Electronic Transitions:
   Electronic Band Structure of Crystalline Solids

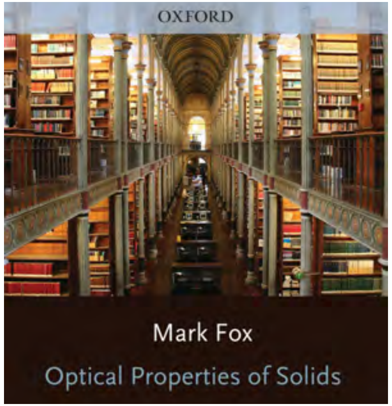


Lecture 1 Outline: Polarized Light and the Dielectric Tensor

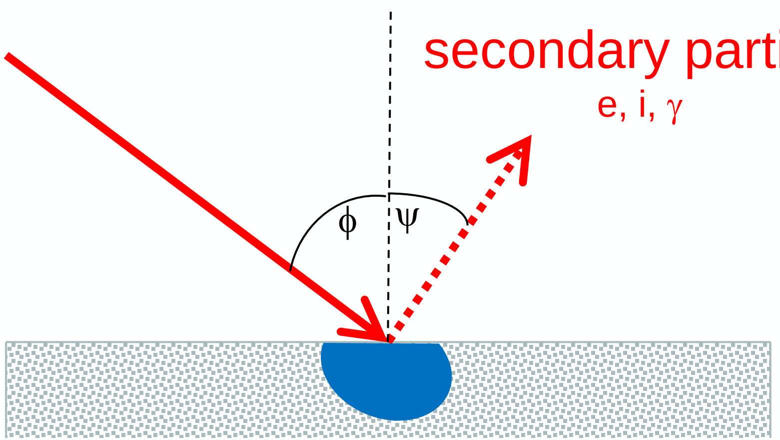
- Spectroscopy, Instrumentation, Bohr Model, Band Structure of Germanium
- Maxwell's Equations in Fourier Space
- Propagation of Light in **Vacuum**: Plane Waves
- Jones and Stokes Vectors
- Reflection of Light: Jones and Mueller Matrices
- **Dielectrics**: Electrodynamics of Continuous Media; Optical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

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). Company data bases.
- Short Course Lectures from instrument suppliers and other sources.



Classification Schemes for Surface Spectroscopy I



Ellipsometry:
Photon in,
Photon out

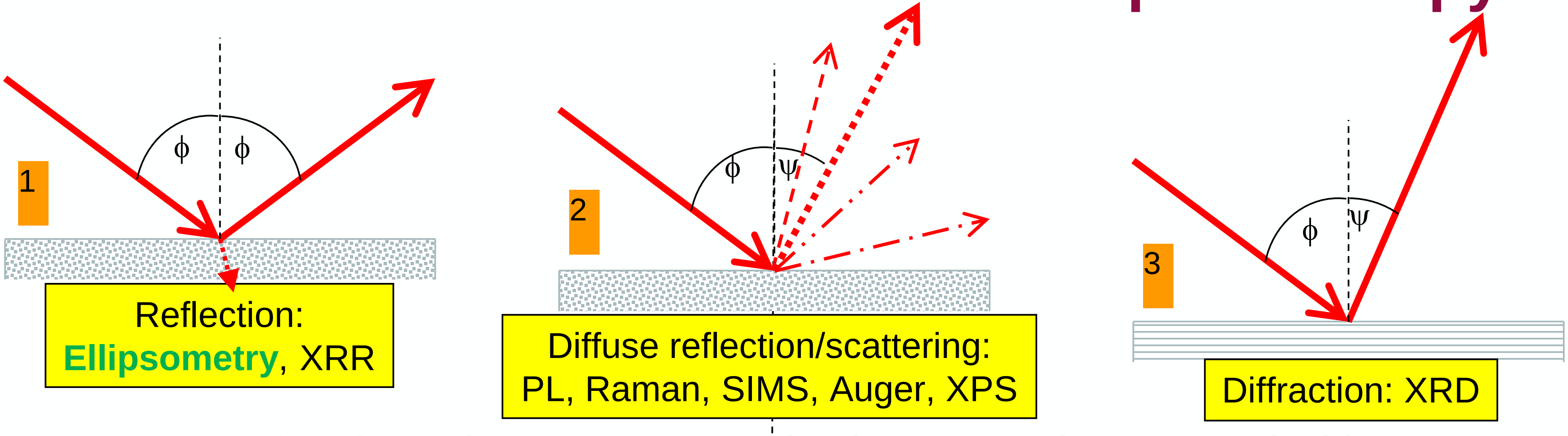
Particles: Electron (e), ion (i), or photon (γ)

The term **spectroscopy** implies that we prepare, vary, or measure the **energy (wavelength)** and/or **momentum** (**direction**) of the primary and/or secondary particle.

For **photons**, we can also measure the **polarization** of the primary and/or secondary photon.

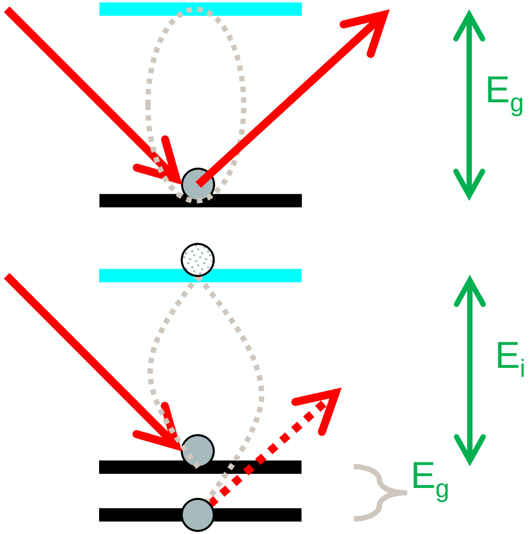
The **interaction depth** for thin films depends on the **penetration depth** of the primary particle and the **escape depth** of the secondary particle. (This can be nanometers to micrometers, depends on each technique.)

Classification Schemes for Surface Spectroscopy II



1. **Specular reflection:** The angle of reflection is equal to the angle of incidence. For some spectroscopies, the angles are measured relative to the surface (XRR), for others relative to the surface normal (SE).
2. **Diffuse reflection or scattering:** There is no well-defined direction, in which the secondary particle exits. The scattering probability may depend on the angles.
3. **Diffraction:** Requires a periodic (crystalline) layer. There is a well-defined angular relationship between the spacing of the diffraction (Bragg) planes and the momentum of the incident/diffracted beams.

Classification Schemes for Surface Spectroscopy III



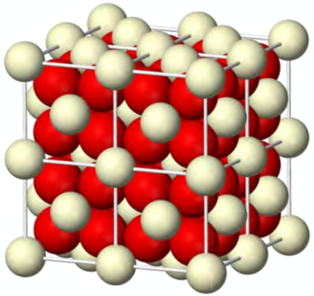
Elastic: The intensity of the reflected (relative to the incident) beam depends on the excited states of the system (band gaps).

Inelastic: The energy difference (gain or loss) provides information about vibrational (Raman) or electronic (Auger) energy states. The strength of the scattering process depends on the interaction with an intermediate state.

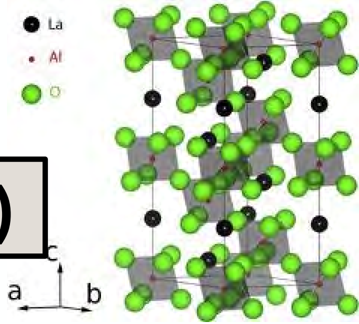
- **Elastic scattering:**
  The energy of the incident particle equals that of the scattered particle.
- **Inelastic scattering:** The two energies are different, depending on the energy gained or lost by the interaction with the thin film.

Classification Schemes for Surface Spectroscopy IV

- Spectroscopic Ellipsometry: Elastic, specular, $\gamma \to \gamma$
  Thickness, Energy (band gap), refractive index, composition
- X-ray reflectivity: Elastic, specular, $\gamma \to \gamma$
  Thickness, density, surface/interface roughness
- X-ray diffraction: Elastic, diffracted, $\gamma \to \gamma$
  Lattice constant, stress/strain, composition
- UV Raman Spectroscopy: Inelastic, scattered, $\gamma \to \gamma$
  Vibrational (phonon) energy, composition, stress/strain
- Secondary Ion Mass Spectrometry: Inelastic, scattered, i $\to$ i
  Composition, depth profile (sputtering), doping
- Auger Electron Spectrometry: Inelastic, scattered, e $\to$ e
  Composition, depth profile (sputtering)
- Rutherford backscattering: Inelastic, scattered, $\alpha \to \alpha$
  Composition, some depth information, primary standard



Solid State Physics (Crystalline)



Crystal Structure (Point & Space Group)

Electrons ↔ **Phonons** | **Defects**

- Electrons: 0.2-10 eV — Near-IR, VIS, UV
- Phonons: 10-80 meV — Far-IR to mid-IR

Ashcroft & Mermin: Solid-State Physics

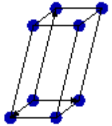
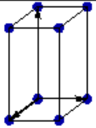
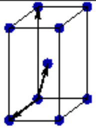
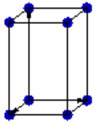
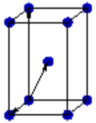
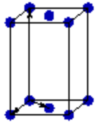
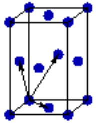
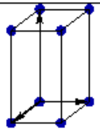
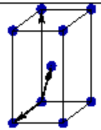
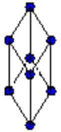
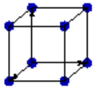
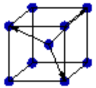
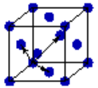
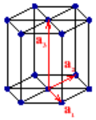
Magnetism | Superconductivity | Excitons | Plasmons

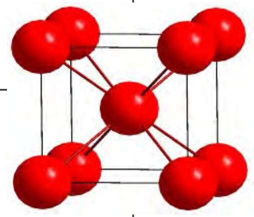
Surfaces | Topological Insulators | Transport | Polaritons

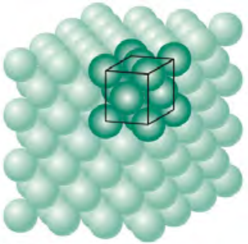
CMOS | RF | Power | Analog | Magnetic Storage | Catalysis

Photovoltaics | Energy Conversion | Lasers | Sensors

Crystallography: Fourteen Bravais Lattices

Bravais lattice	Parameters	Simple (P)	Volume centered (I)	Base centered (C)	Face centered (F)
Triclinic	$a_1 \neq a_2 \neq a_3$ $\alpha_{12} \neq \alpha_{23} \neq \alpha_{31}$				
Monoclinic	$a_1 \neq a_2 \neq a_3$ $\alpha_{23} = \alpha_{31} = 90°$ $\alpha_{12} \neq 90°$				
Orthorhombic	$a_1 \neq a_2 \neq a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Tetragonal	$a_1 = a_2 \neq a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Trigonal	$a_1 = a_2 = a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} < 120°$				
Cubic	$a_1 = a_2 = a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Hexagonal	$a_1 = a_2 \neq a_3$ $\alpha_{12} = 120°$ $\alpha_{23} = \alpha_{31} = 90°$				





P	simple
I	body-centered
F	face-centered
C	base-centered

Seven crystal systems become 14 Bravais lattices with centering.

Crystal = Lattice + Basis

(Wyckoff positions)

32 point groups

230 space groups (Intl. Tables)

Group Theory, Symmetry

Rohrer: Structure and Bonding in Crystalline Materials

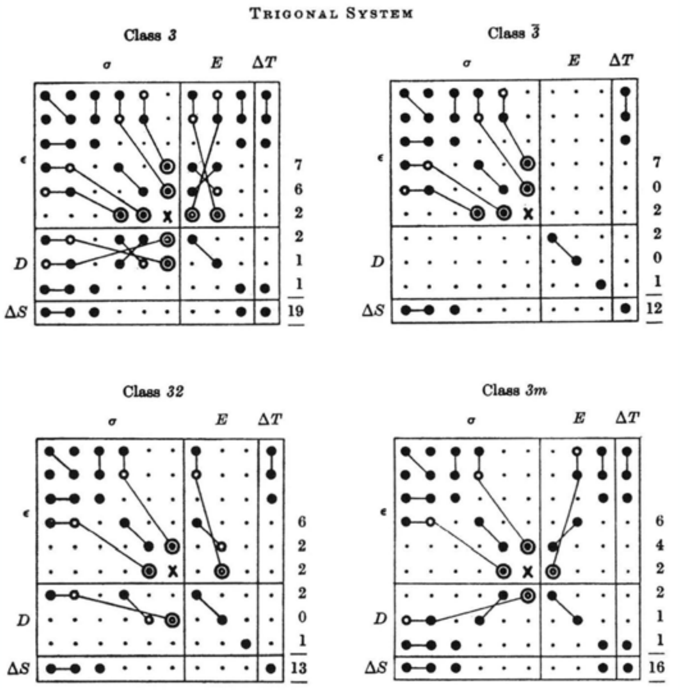
Nye: Physical Properties of Crystals

D Dielectric displacement
E electric field
ε **dielectric tensor**

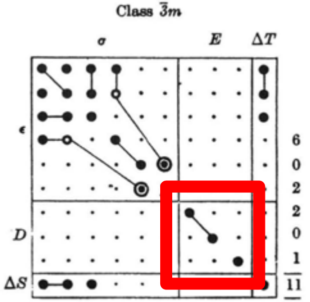
$$\vec{D} = \varepsilon \vec{E}$$

For crystal class -3m,
the dielectric tensor
- has **two independent diagonal** components.
- **off-diagonal components are zero**.

Also: Stress/strain, magnetic, piezo, …
Many different tensor properties.



LaAlO$_3$



Nye: Physical Properties of Crystals (Appendix E)

Matrix Elements: Selection Rules

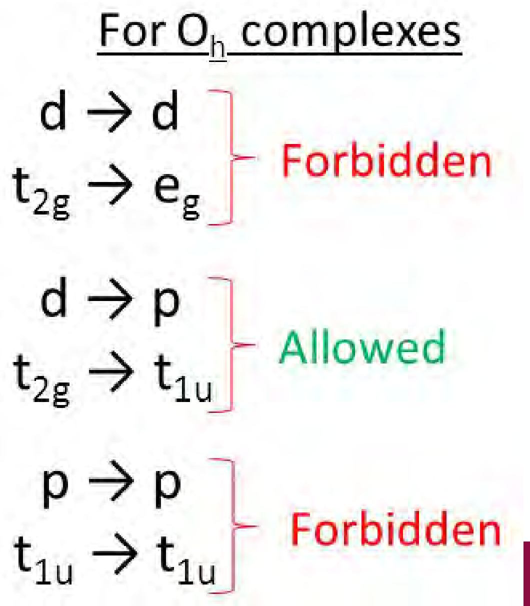
Problem Statement:

- Initial state <i|: symmetry Γ_i
- Final state <f|: symmetry Γ_f
- Interaction Hamiltonian: symmetry Γ_H

Symmetry produces selection rules.
H-atom: $\Delta l = \pm 1$

Question:
Is the transition from <i| to <f| allowed?
Is the matrix element <f|H|i> zero (i.e., transition forbidden).

Answer: The transition is forbidden, unless the final state symmetry Γ_f is contained in the product of Γ_i and Γ_H.
This calculation uses character tables (or similar tools).



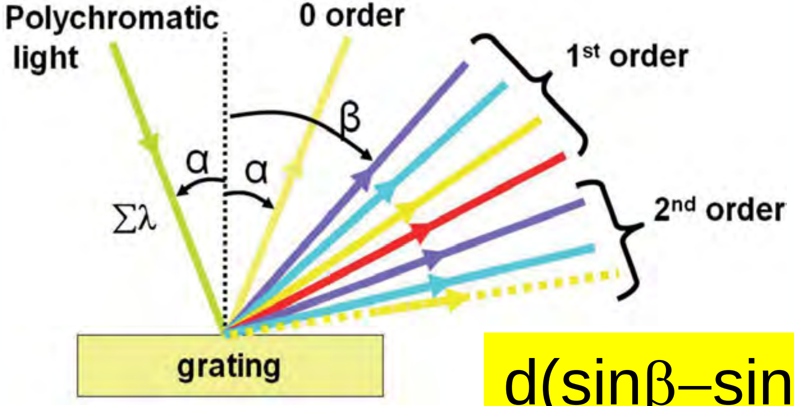
Example:
Optical transition from Γ_7^+ to Γ_7^- ($E_0'+\Delta_0$) forbidden in Ge.

Note: Selection rules are relaxed, if symmetry is lowered.
(If we lose the inversion symmetry, parity rules go away.)

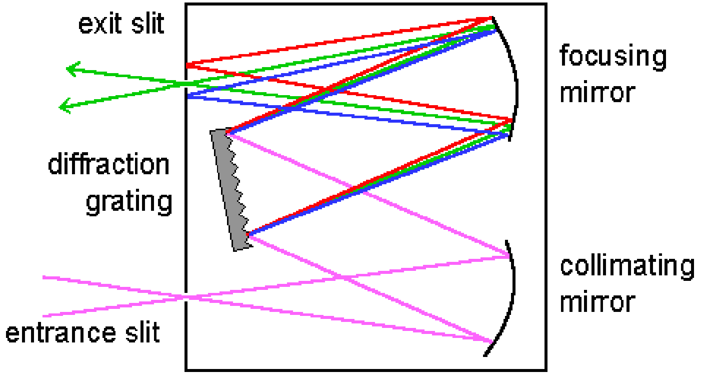
Materials Properties Accessible by Optical Spectroscopy

- Mid-infrared spectral range: (FTIR-VASE)
  - Insulator/semiconductor:
    Lattice vibrations (optical long-wavelength phonons)
  - Metal: Free carrier properties (density, scattering rate)
- Near-IR to visible to UV range: (RC-2, VASE, M-2000)
  - Electronic excitations
  - Band gap, interband transitions
- Ellipsometry allows study of semiconductors, insulators, and metals.
- Thin films and surfaces can be investigated with proper data analysis (fitting).

Grating Monochromator

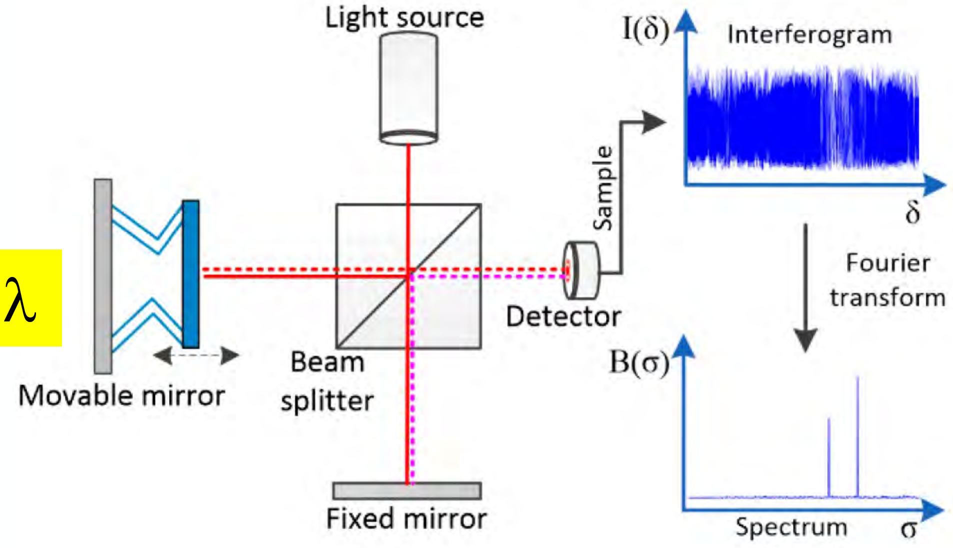


$$d(\sin\beta - \sin\alpha) = N\lambda$$



Diffracted intensity depends on angle and polarization.

Fourier-Transform Infrared Spectrometer

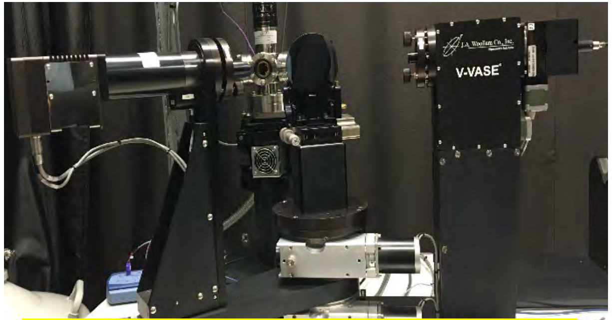


Constructive interference: $\Delta x = N\lambda$

Destructive interference: $\Delta x = (2N+1)\lambda/2$

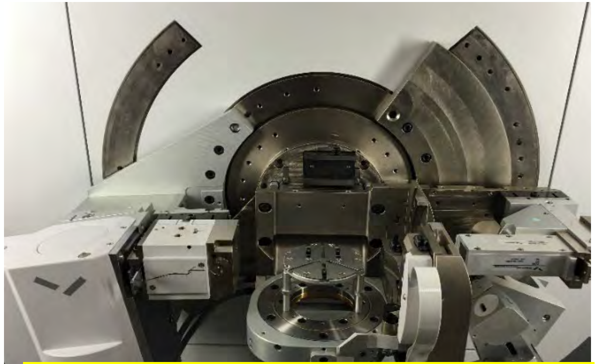
Common for mid-infrared spectroscopy (50-500 meV).

Reflectance Spectroscopy Instrumentation



Spectroscopic Ellipsometer
190 to 2500 nm (0.5 to 6.5 eV)

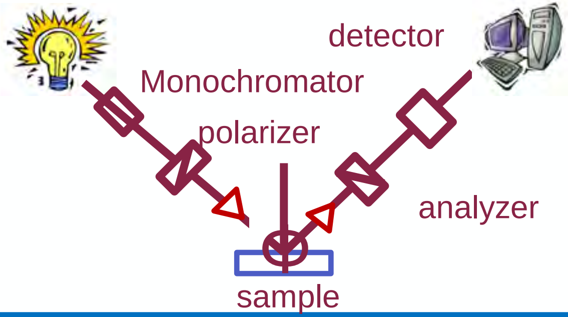
Infrared Ellipsometer
1.25 to 40 μm (250 to 8000 cm^{-1})



X-ray diffraction & reflectance

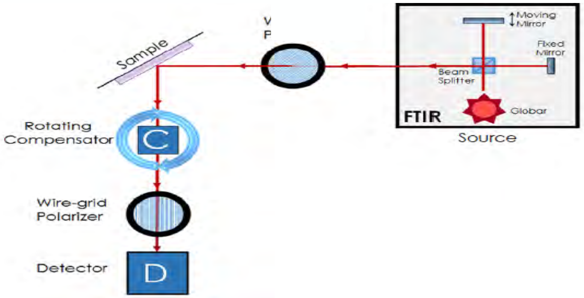
Spectroscopic Ellipsometry:

- **Thickness (100 to 10000 Å)**
- **Absorption, band gap**
- **Refractive index**



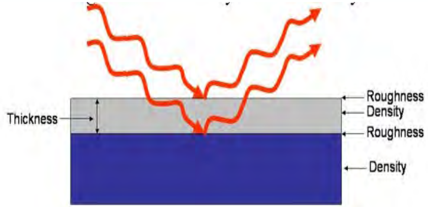
FTIR ellipsometry:

- **Very thick films (> 5000 Å)**
- **Phonon absorption**
- **Optical Constants**

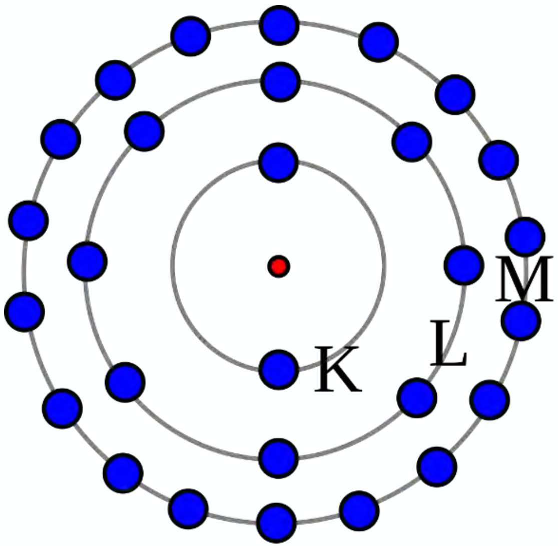


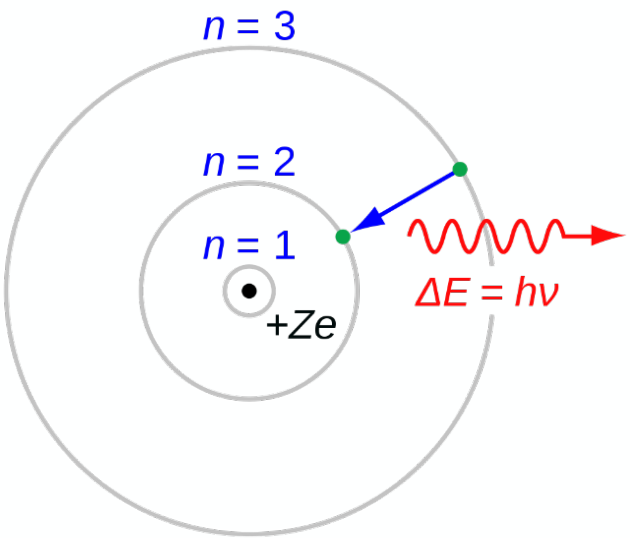
XRD/XRR:

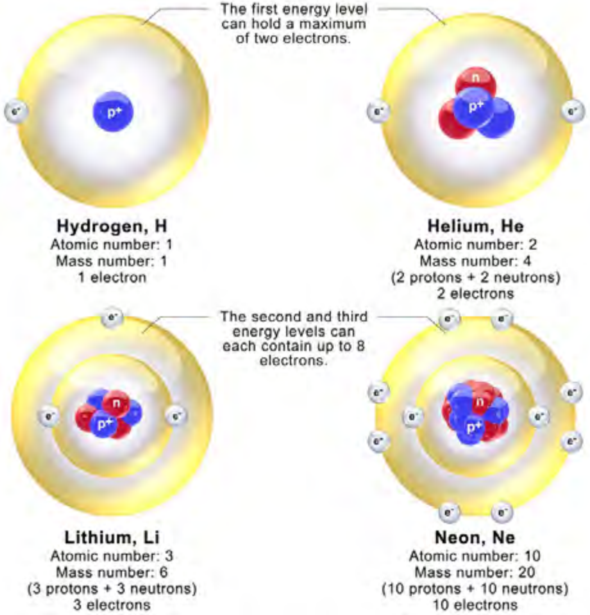
- **Crystal structure**
- **Lattice spacings** (strain)
- **Thickness** (5 Å to 1000 Å)
- **Surface, roughness** layer
- **Density**



Bohr Model for the Hydrogen Atom







Quantum Numbers:

n	1,2,3,…
l	0,…,n-1
m	-l,…,l
s	+/- 1/2

$E(n) = -R/n^2$

$R = 13.6$ eV

Relativistic corrections:

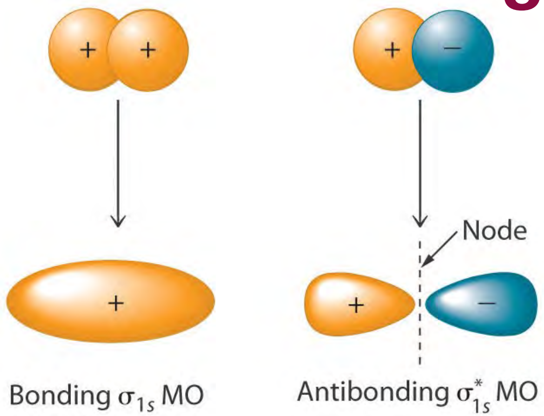
s electrons (l=0) close to the core

J=**L**+**S** total angular momentum

Spin-orbit coupling **L·S**

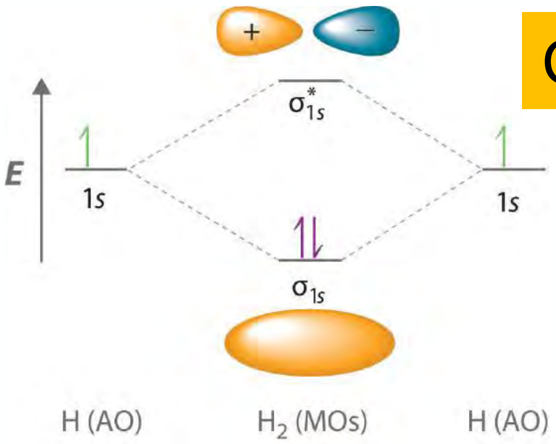
L=1, S=1/2 J=1/2 or 3/2

Bonding and Anti-Bonding Orbitals



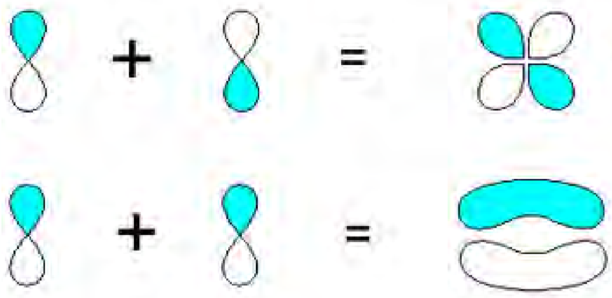
s: $\Psi=\psi_1+\psi_2$

s*: $\Psi=\psi_1-\psi_2$



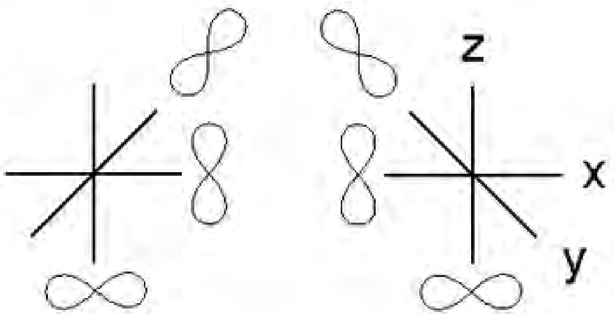
Conduction Band

Valence Band



p*

p

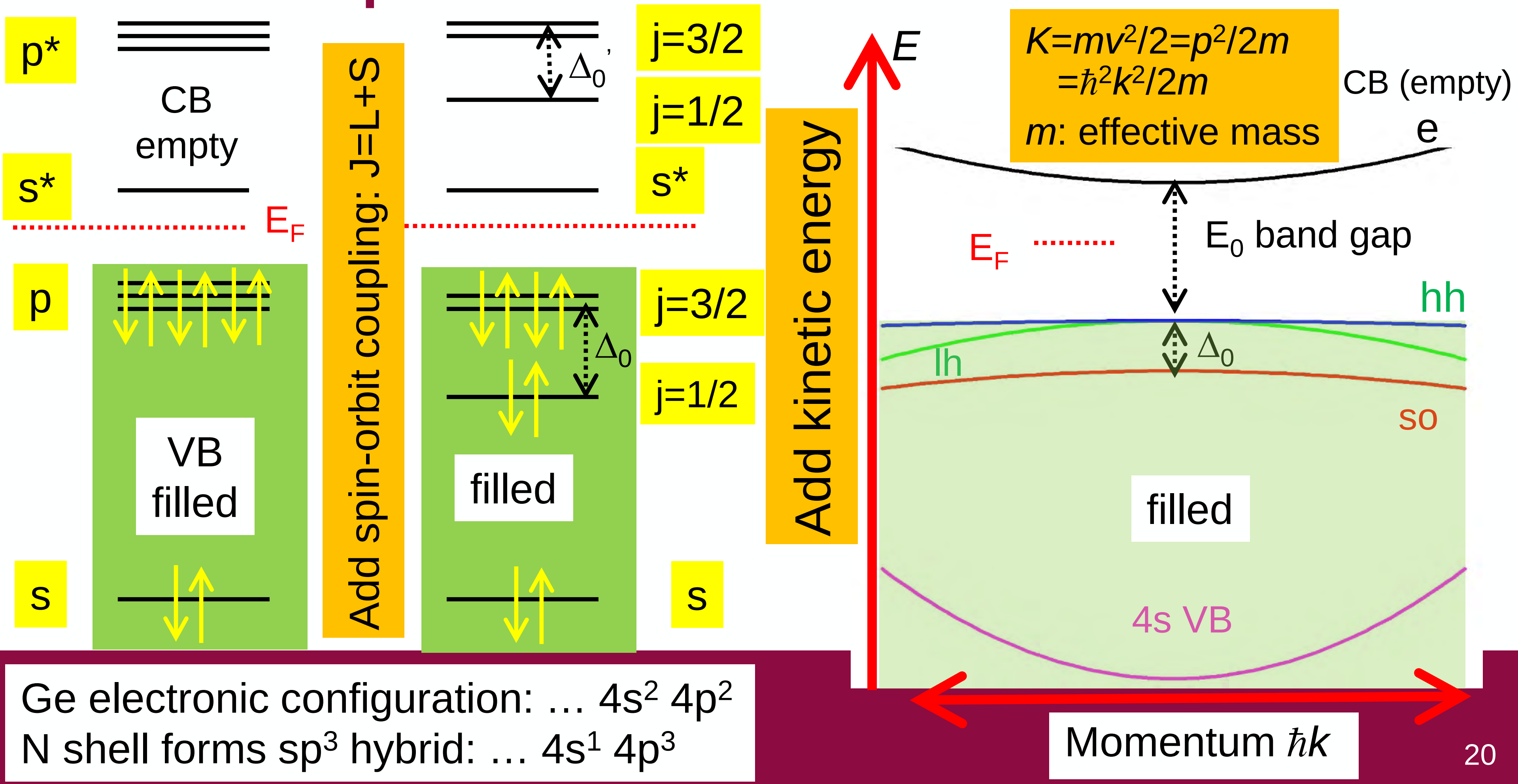


p_x, p_y, p_z

C electronic configuration: $1s^2\ 2s^2\ 2p^2$

L shell forms sp³ hybrid: $1s^2\ 2s^1\ 2p^3$

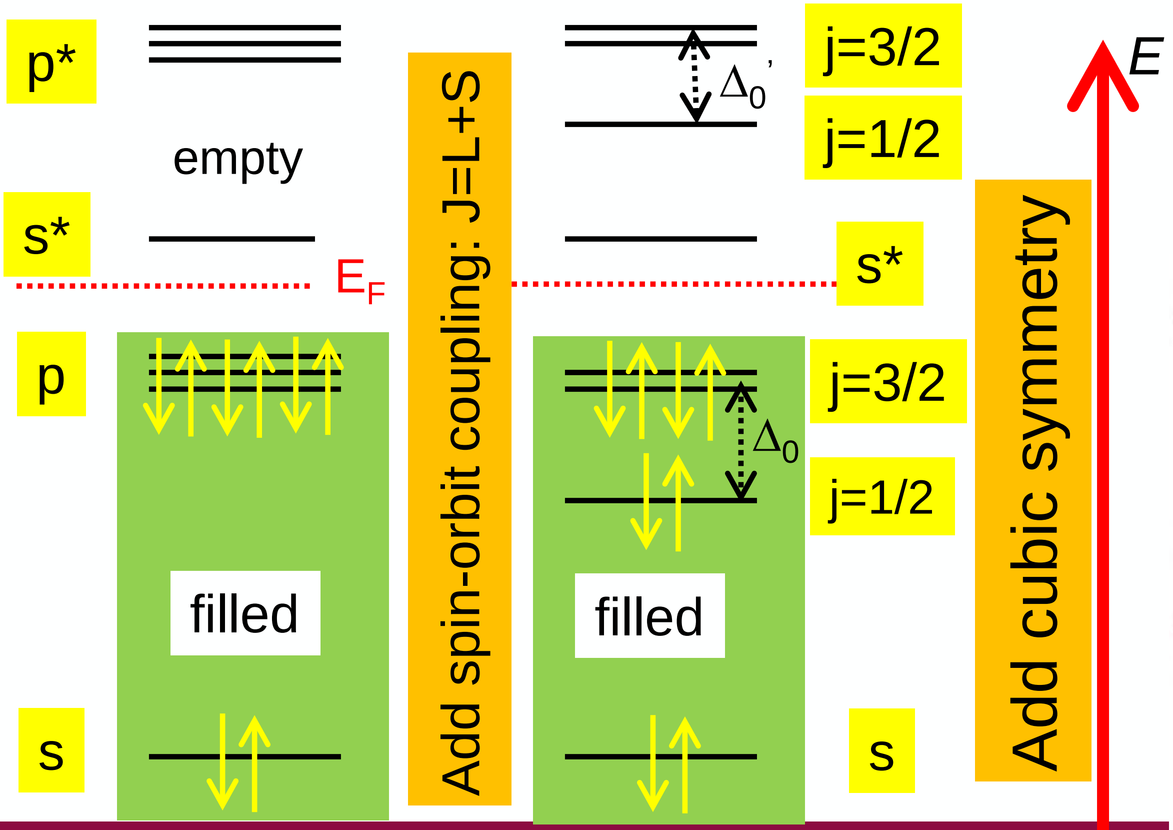
A simple band structure for Germanium

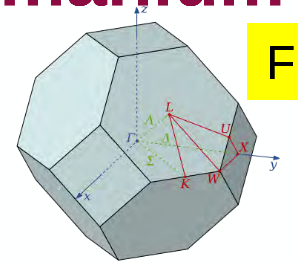


Ge electronic configuration: … $4s^2$ $4p^2$

N shell forms sp^3 hybrid: … $4s^1$ $4p^3$

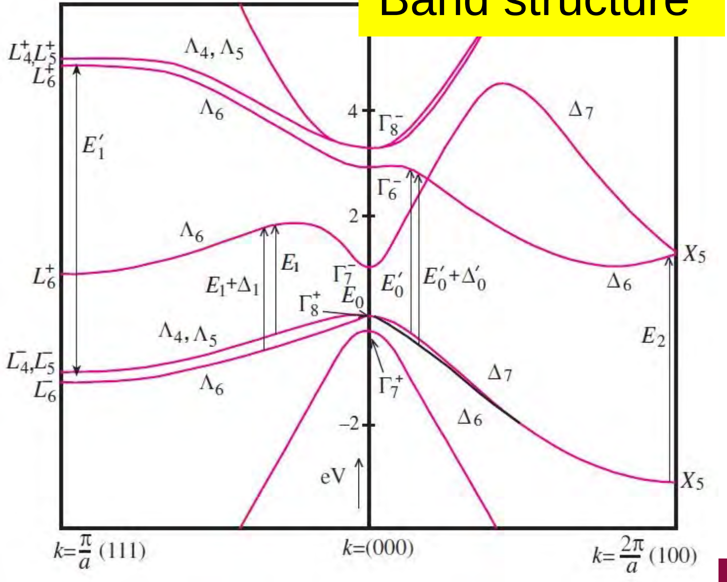
Band structure of Germanium





FCC Brillouin zone

Band structure

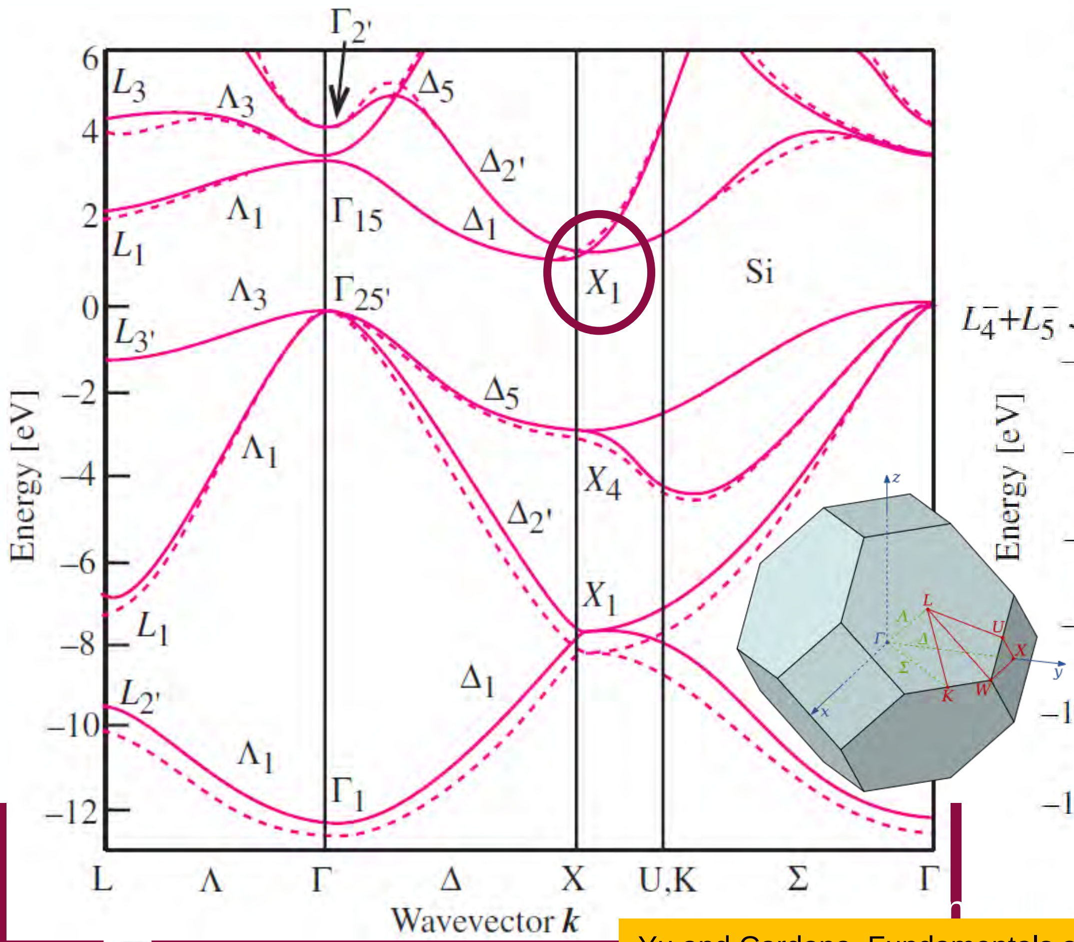


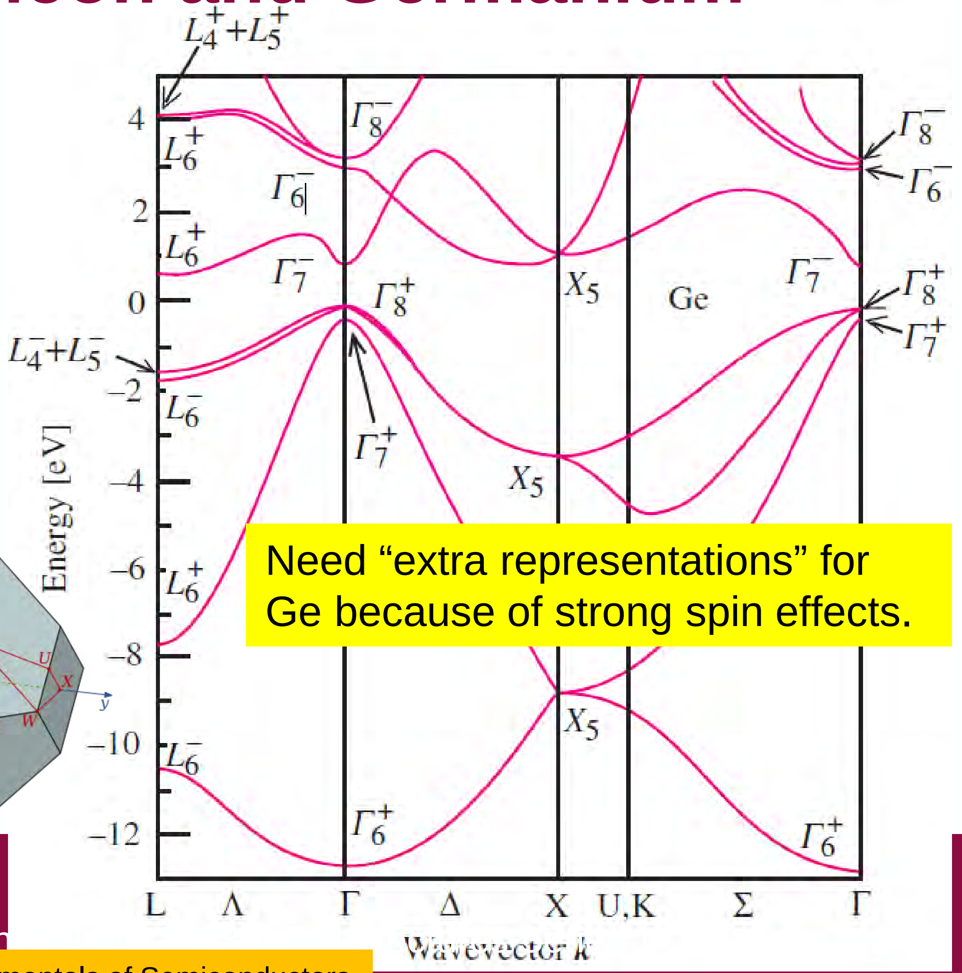
Momentum $\hbar k$

Ge electronic configuration: … $4s^2\ 4p^2$

N shell forms sp^3 hybrid: … $4s^1\ 4p^3$

Band Structure of Silicon and Germanium

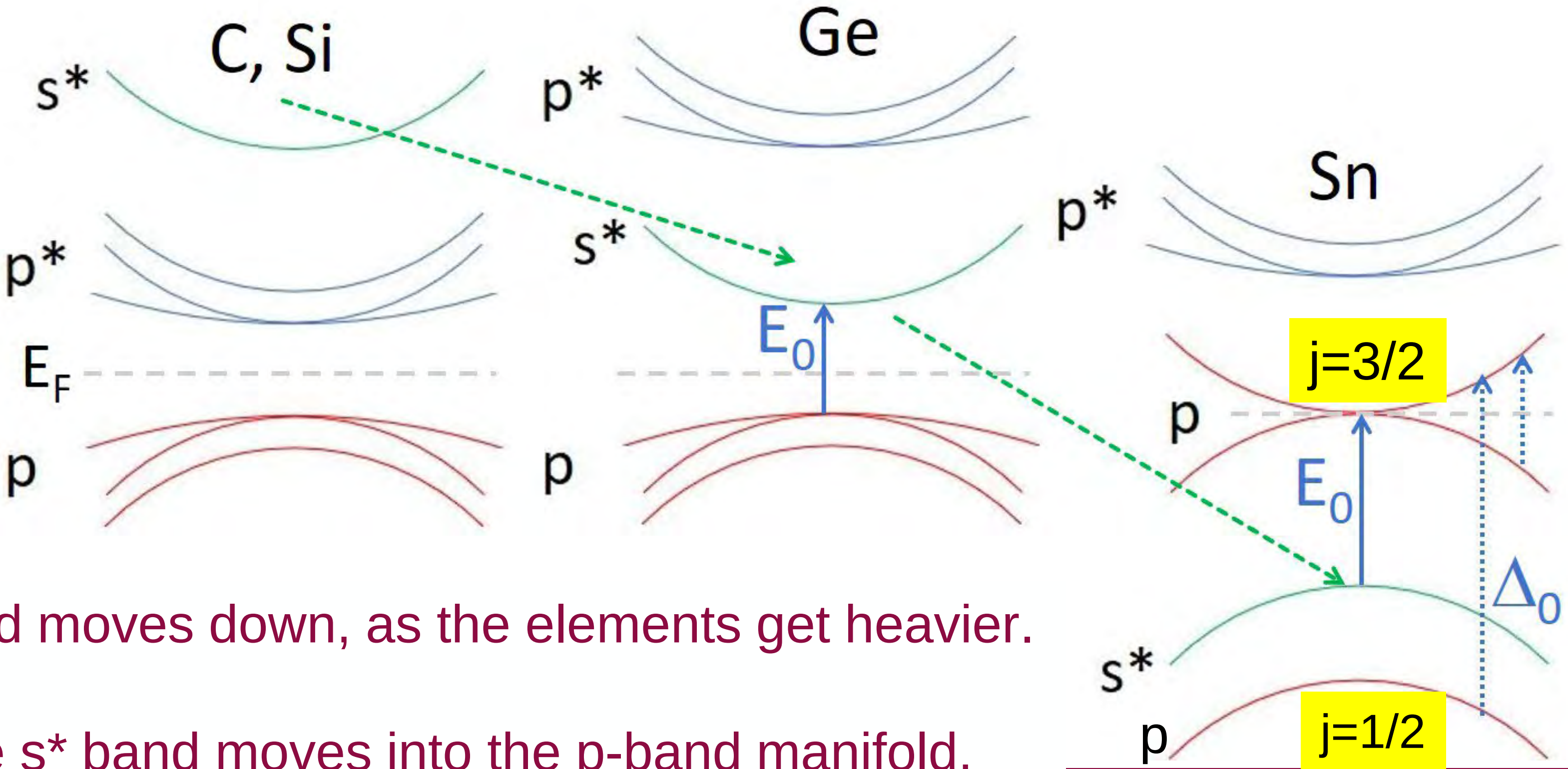




Need “extra representations” for Ge because of strong spin effects.

Yu and Cardona, Fundamentals of Semiconductors

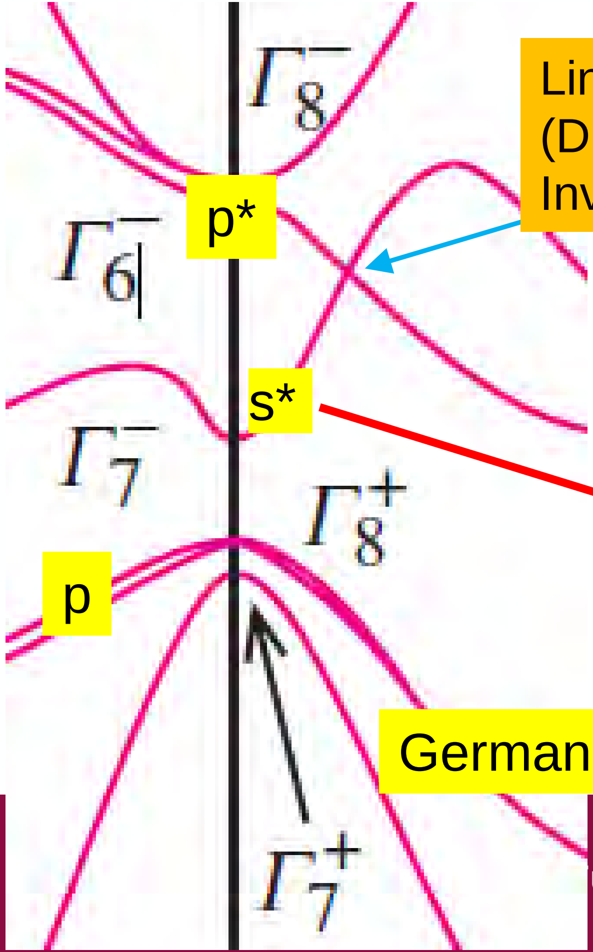
Relativistic Effects: Darwin Shift: C, Si, Ge, Sn



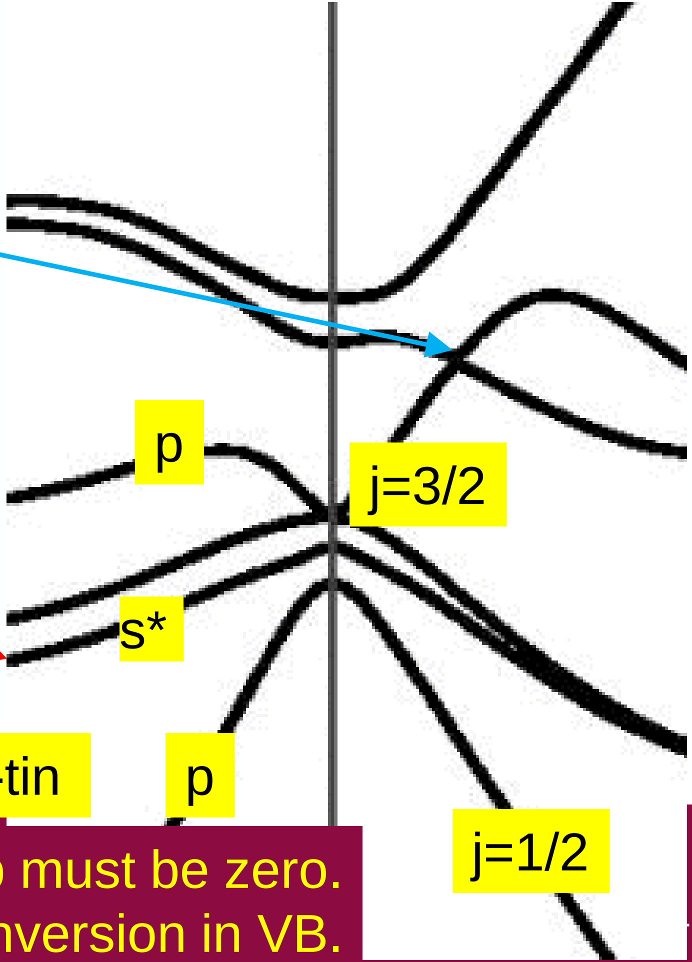
The s* band moves down, as the elements get heavier.

In α-tin, the s* band moves into the p-band manifold, between the j=1/2 and j=3/2 states.
This makes α-tin an (inverted) zero-gap semiconductor.

Band Inversion: Topological Insulators



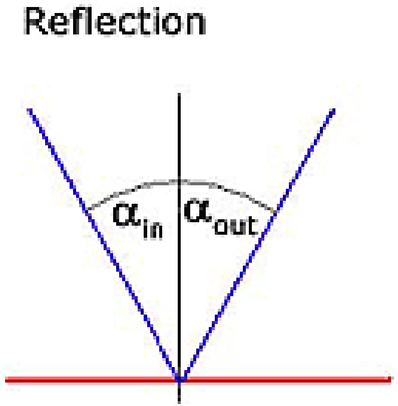
Linear crossing
(Dirac point)
Inversion Symmetry

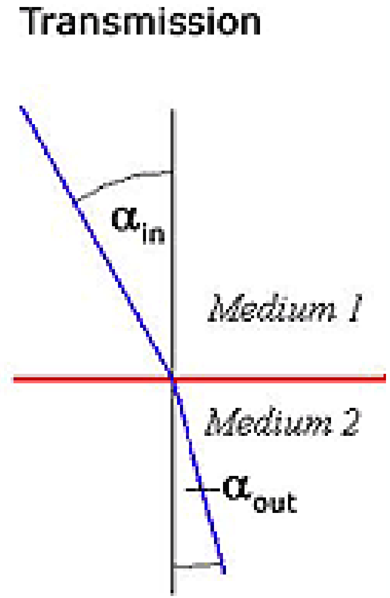


Band gap must be zero.
Symmetry inversion in VB.

Reflection and Transmission

Also have diffuse scattering.

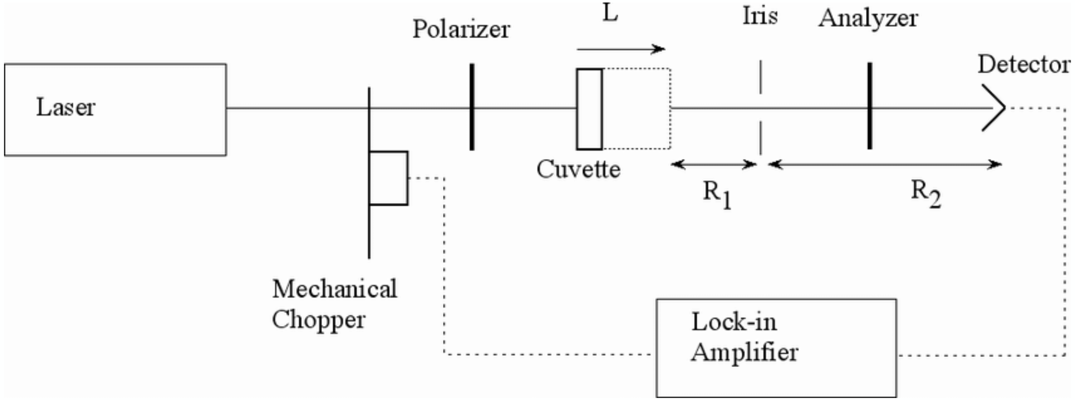




$$R = \left(\frac{n-1}{n+1}\right)^2$$

Law of reflection: $\alpha_{in}=\alpha_{out}$
Snell's Law: $n_1 \sin\alpha_{in} = n_2 \sin\alpha_{out}$
n: Refractive Index

Beer's Law: $I(L)=I_0\exp(-\alpha L)$



Absorption coefficient α (cm^{-1})
Consider reflection losses

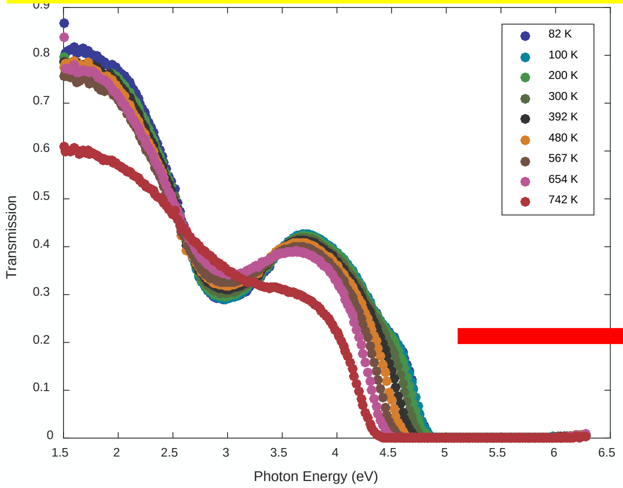
$$\exp(-\alpha L) \approx \frac{T}{(1-R)^2}$$

Transmission: LSAT or $(LaAlO_3)_{0.3}(SrAlTaO_6)_{0.35}$

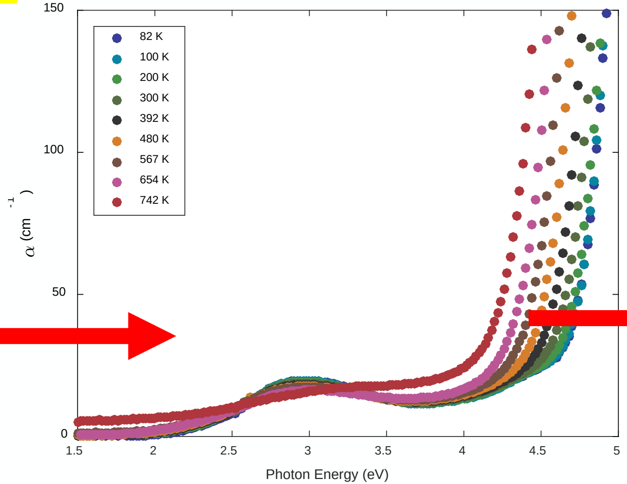
$$T = \frac{(1-R)^2 e^{-\alpha d}}{1 - R^2 e^{-2\alpha d}}$$

We need to know the complex refractive index to calculate the reflection losses. Very good for small absorption coefficients.

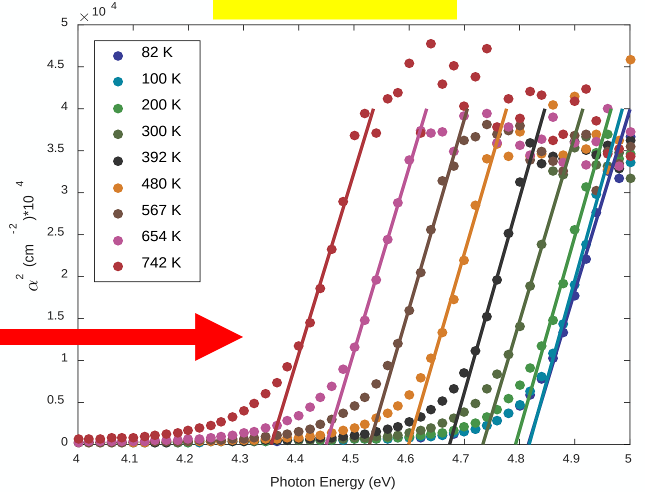
Transmission Measurement



Absorption Coefficient



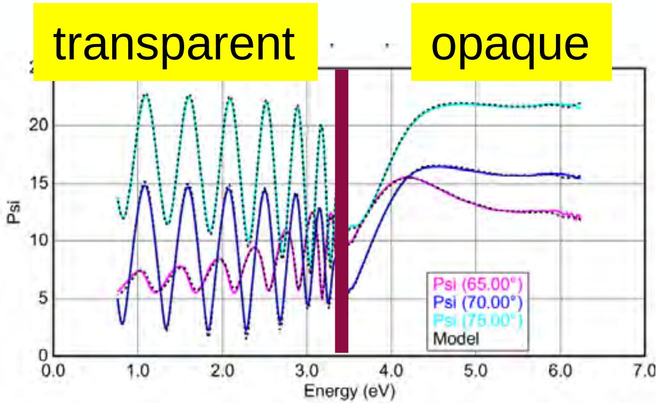
Tauc Plot

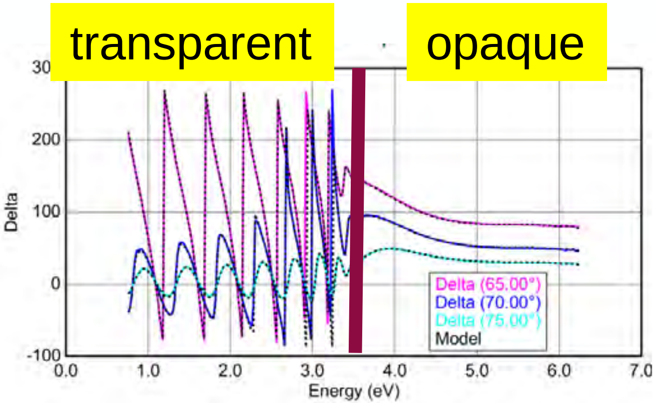


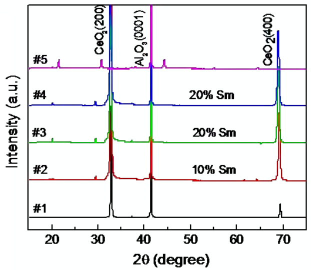
Jacqueline Cooke, 2016 AVS Meeting (Nashville)

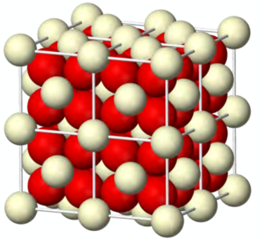
David C. Look and Jacob H. Leach JVST B **34**, 04J105 (2016).

Crystalline CeO_2 on sapphire (liquid deposition)

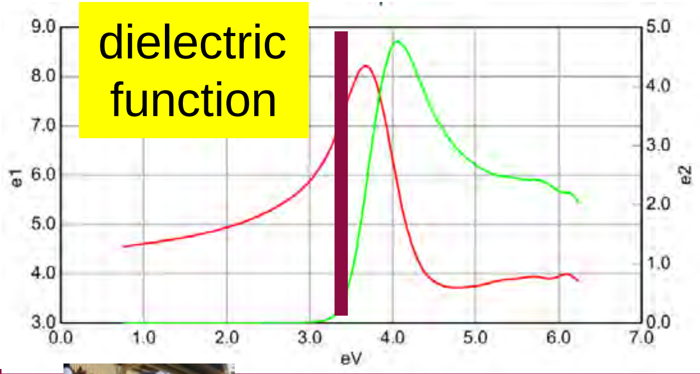




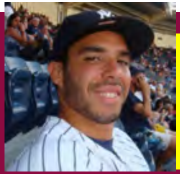




Fluorite (O_h^5)

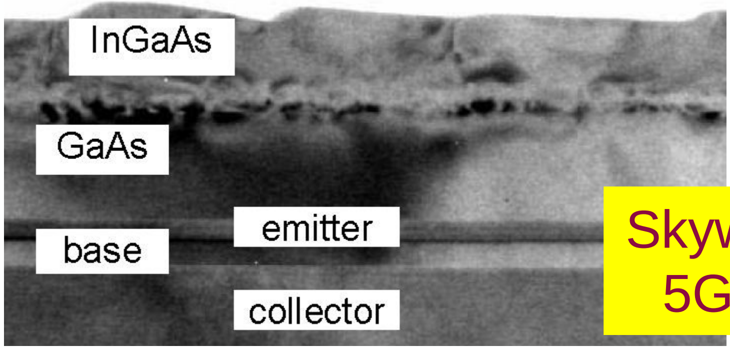


- Insulating CeO_2 film on **<u>single-side polished</u>** sapphire, with **band gap near 3.7 eV.**
- Determine **film thickness** from interference fringes in transparent region.
- Fit **optical constants** with basis spline polynomials.

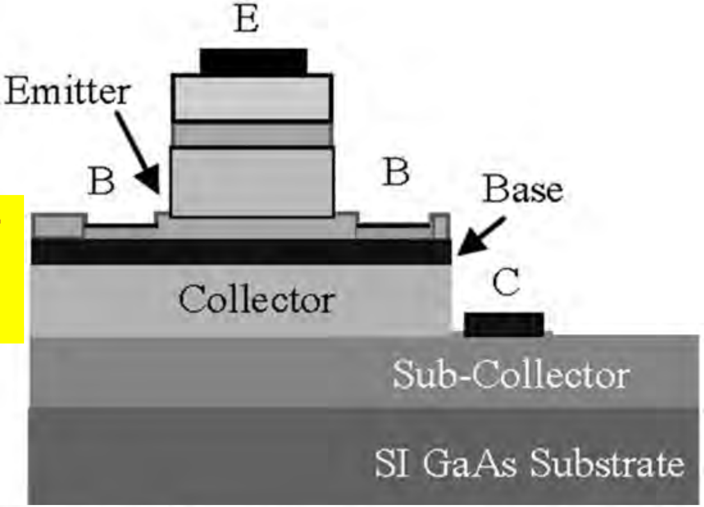


K. Mitchell, C.O. Rodriguez, Y. Li, 2013; X. Guo, Boston Applied Technologies, Inc.

Thickness Fringes or Band Structure ???



Skyworks InGaP HBT
5G cell phone chip



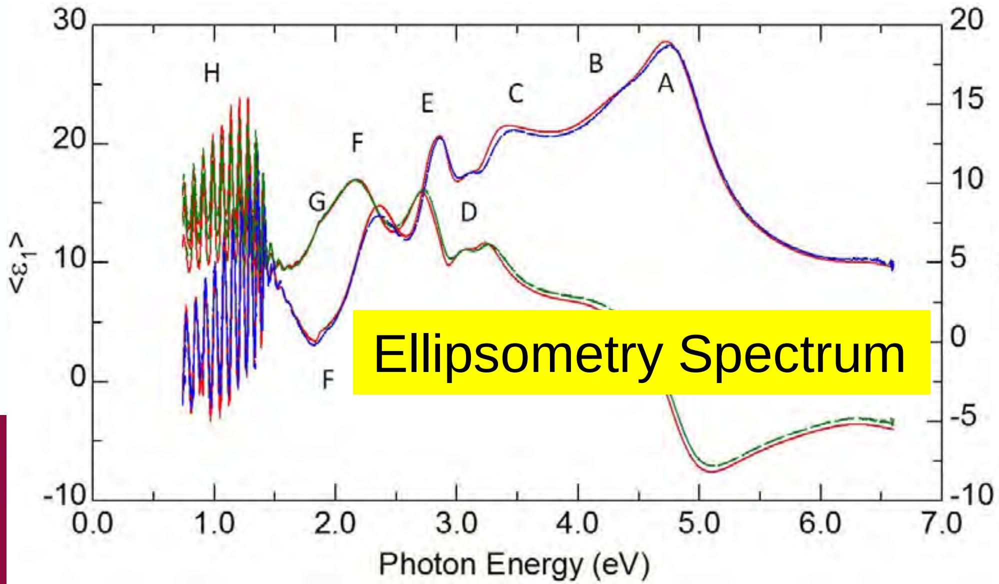


Table 2 Doping and layer content profile of a typical InGaP double heterojunction bipolar transistor (DHBT). Compare Ref. [28]. See also Ref. [42].

Layer	Material	Doping	Concentration (cm^{-3})	Thickness	Function
11	InGaAs	n+	$>10^{19}$	100 nm	emitter contact
10	GaAs	n+	5×10^{18}	120 nm	contact buffer layer
9	InGaP	n	3×10^{17}	40 nm	emitter
8	GaAs	p+	5×10^{19}	70 nm	base
7	GaAs	n	3×10^{16}	30 nm	collector
6	GaAs	n+	2×10^{18}	5 nm	dopant spike for DHBT
5	InGaP	n	3×10^{16}	10 nm	DHBT collector
4	GaAs	n	3×10^{16}	155 nm	collector layer
3	GaAs	n	7.5×10^{15}	400 nm	collector layer
2	GaAs	n+	5×10^{18}	1000 nm	subcollector
1	AlAs (?)	?	?	30 nm	substrate isolation
0	GaAs	?	?	NA	semi-insulating substrate

Scalar and Vector Waves

- **Field:** Scalar or vector depends on position **r**.
- **Physical quantities are always real.**
  Scalar: energy, charge, etc.
  Vector: momentum, current density, electric field, etc.

- Scalar wave

$$s(\vec{r},t) = A\cos\left(\vec{k}\cdot\vec{r} - \omega t + \varphi\right)$$

- Vector wave

$$\vec{E}(\vec{r},t) = \vec{E}_0 A\cos\left(\vec{k}\cdot\vec{r} - \omega t + \varphi\right)$$

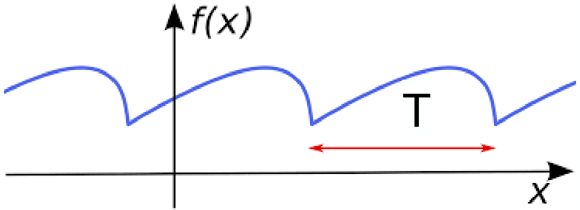
- **Where do the complex notations come from?**

Fourier Series of Periodic Functions

- A real-valued scalar function f(t) is called **periodic** with period T, if f(t)=f(t+T) for all values of t.

- A periodic scalar function with period T can be written as a **Fourier Series**

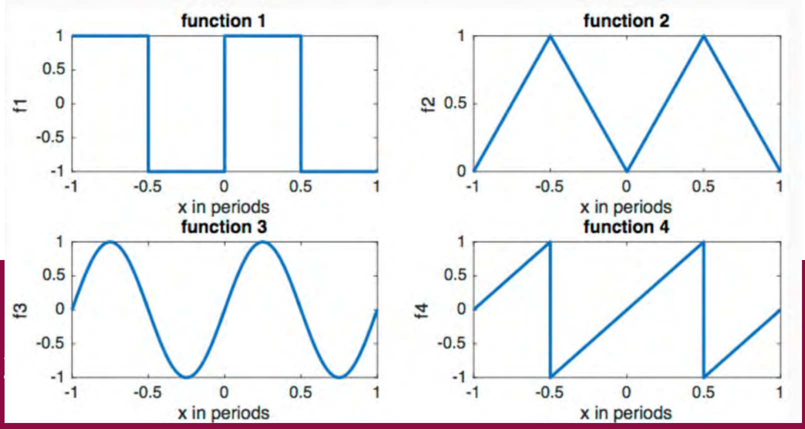
$$f(t) = \frac{1}{2}A_0 + \sum_{m=1}^{\infty}\left[A_m \cos(m\omega t) + B_m \sin(m\omega t)\right]$$



with angular frequency $\omega=2\pi/T$ and Fourier coefficients

$$A_m = \frac{\omega}{\pi}\int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t)\cos(m\omega t)\,dt$$

$$B_m = \frac{\omega}{\pi}\int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t)\sin(m\omega t)\,dt$$



Jackson, E&M, 1975

Fourier Series of Periodic Functions

- Dealing with harmonic functions (sin, cos) is not convenient, because
  - We need two functions for each harmonic.
  - Taking derivatives is not easy, because sin and cos switch at each order.

- A periodic scalar function with period T can be written as a **Fourier Series**

$$f(t) = \sum_{m=-\infty}^{\infty} c_m \exp(-im\omega t)$$

Ellipsometry analyzes complex Fourier coefficients.

with **complex** Fourier coefficients

$$c_m = \frac{\omega}{\pi} \int_{-\frac{\pi}{\omega}}^{\frac{\pi}{\omega}} f(t) \exp(im\omega t)\, dt = \begin{cases} \frac{A_0}{2} & m = 0 \\ \frac{1}{2}(A_m + iB_m) & m > 0 \\ \frac{1}{2}(A_{-m} - iB_{-m}) & m < 0 \end{cases}$$

Jackson, E&M, 1975

- The Fourier coefficients are now complex, but the function f(t) is still real.
- The imaginary parts all cancel, if the complex coefficients c_m are defined correctly.

Fourier Transforms of Non-Periodic Functions

- If the function f(t) is not periodic, then the period T becomes infinite and the frequency spacing ω between overtones becomes very small.
- The Fourier series now becomes a **Fourier Integral**.

$$f(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} F(\omega) \exp(-i\omega t) d\omega$$

with the **Fourier transform F(ω)**

$$F(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} f(t) \exp(i\omega t) dt$$

- The prefactors $1/\sqrt{2\pi}$ before the integral can vary (depends on convention).
- **The Fourier transform function F(ω) is allowed to be complex, because it is not a meaningful physical quantity.**
- Orthogonality and completeness:

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i(\omega - \omega')t] \, dt = \delta(\omega - \omega')$$

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i\omega(t - t')t] \, d\omega = \delta(t - t')$$

$$\frac{1}{\sqrt{2\pi}} \exp(i\omega t)$$

Orthonormal basis of
Hilbert Space of real functions

Jackson, E&M, 1975

Math with Fourier Transforms

- Convolution theorem:
  The Fourier transform of a convolution equals the product of the Fourier transforms.

$$(f * g)(t) = \int_{-\infty}^{\infty} f(t')g(t - t')dt'$$

$$\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} (f * g)(t) \exp(i\omega t)dt = \sqrt{2\pi} F(\omega) G(\omega)$$

- The Fourier transform of the derivative of f(t) equals –iωF(ω).

$$\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} f'(t) \exp(i\omega t)dt = -i\omega F(\omega)$$

- The complex conjugate of the Fourier transform equals F(–ω).

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

Jackson, E&M, 1975

Fourier Series in Multiple Dimensions

- A real-valued scalar field s(**r**) in a Bravais lattice (with Bravais lattice vectors **T** and reciprocal lattice vectors **G**) is called periodic, if s(**r**+**T**)=s(**r**) for all Bravais lattice vectors **T.**
- A real-valued periodic scalar field s(**r**) in a Bravais lattice can be written as a Fourier sum in reciprocal space

$$s(\vec{r}) = \sum_{\vec{G}} s_{\vec{G}} \exp\left(i\vec{G}\cdot\vec{r}\right)$$

with complex Fourier coefficients

$$s_{\vec{G}} = \frac{1}{V}\int_C s(\vec{r}) \exp\left(-i\vec{G}\cdot\vec{r}\right) d^3\,\vec{r}$$

where C is the unit cell with volume V. **G** is a reciprocal lattice vector.
- The same equations apply to a real-valued periodic vector field **E**(**r**).

$$\vec{E}(\vec{r}) = \sum_{\vec{G}} \vec{E}_{\vec{G}} \exp\left(i\vec{G}\cdot\vec{r}\right)$$

$$\vec{E}_{\vec{G}} = \frac{1}{V}\int_C \vec{E}(\vec{r}) \exp\left(-i\vec{G}\cdot\vec{r}\right) d^3\,\vec{r}$$

Ashcroft & Mermin, Appendix D

Fourier Transforms in Multiple Dimensions

- Fourier transforms can also be generalized to multiple dimensions for scalar fields

$$s(\vec{r}) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} S(\vec{k}) \exp(i\vec{k}\cdot\vec{r}) d^3\vec{k}$$

$$S(k) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} s(\vec{r}) \exp(-i\vec{k}\cdot\vec{r}) d^3\vec{r}$$

- and vector fields

$$\vec{E}(\vec{r}) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} \vec{E}(\vec{k}) \exp(i\vec{k}\cdot\vec{r}) d^3\vec{k}$$

$$\vec{E}(k) = \left(\frac{1}{\sqrt{2\pi}}\right)^3 \iiint_{-\infty}^{\infty} \vec{E}(\vec{r}) \exp(-i\vec{k}\cdot\vec{r}) d^3\vec{r}$$

- The fields s(**r**) and **E**(**r**) in real space have real values.
- **The Fourier transforms *S*(*k*) and *E*(*k*) have complex values, but their imaginary parts cancel out in the summation.**

Microscopic Maxwell's Equations (in Vacuum)

- Electric field strength **E**(**r**)
- Magnetic field strength **H**(**r**)
- Current density **j**(**r**), charge density ρ(**r**)
- Permittivity of free space ε_0, permeability of free space μ_0.

$$\vec{\nabla} \cdot \vec{E} = \frac{\rho}{\varepsilon_0} = 0$$ Gauss' Law (Coulomb)

$$\vec{\nabla} \cdot \vec{H} = 0$$ Gauss' Law (magnetic field)

$$\vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial \vec{H}}{\partial t}$$ Faraday's Law (Lenz)

$$\vec{\nabla} \times \vec{H} = \vec{j} + \varepsilon_0 \frac{\partial \vec{E}}{\partial t} = \varepsilon_0 \frac{\partial \vec{E}}{\partial t}$$ Ampere's Law

- Homogeneous (in vacuum), linear, first-order, constant coefficients, partial DEQ.
- Vector analysis can be used (Stokes' Theorem) to transform Maxwell's equations into integral form.
- Introduce speed of light

$$c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$$

- Units: MKSA (SI).

Jackson, E&M, 1975

Wave Equations (in Vacuum)

- Electric field strength **E**(**r**); Magnetic field strength **H**(**r**).
- Maxwell's equations can be combined to obtain the vacuum wave equations (second order, linear, homogeneous, constant coefficients).

$$\vec{\nabla}^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = 0$$

$$\vec{\nabla}^2 \vec{H} - \frac{1}{c^2}\frac{\partial^2 \vec{H}}{\partial t^2} = 0$$

$$c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$$

- **Plane wave solutions:**

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

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

- Why are the solutions complex ?
  Plane wave is not physical (infinite, monochromatic). Form Gaussian wave packets.
- Poynting vector indicates energy flow: $\vec{S} = \vec{E} \times \vec{H}$

Jackson, E&M, 1975

Plane-Wave Solutions to Maxwell's Equations (Vacuum)

- Electric field strength **E**(**r**); Magnetic field strength **H**(**r**).
- Any electric and magnetic field strength can be written as a Fourier-transform

$$\vec{E}(\vec{r},t) = \left(\frac{1}{2\pi}\right)^2 \int d\omega \iiint d^3\vec{k}\, \vec{E}(\vec{k},\omega) \exp[i(\vec{k}\cdot\vec{r}) - \omega t]$$

$$\vec{E}(\vec{k},\omega) = \left(\frac{1}{2\pi}\right)^2 \int dt \iiint d^3\vec{r}\, \vec{E}(\vec{r},t) \exp[-i(\vec{k}\cdot\vec{r}) - \omega t]$$

- The Fourier transforms are complex, but the **E**(**r**) and **H**(**r**) fields are not.
- **Signs: Nebraska convention as modified by Aspnes.**
  Kinetic energy of free particle in quantum mechanics is positive. Classical wave travels along **k**.
- The complex plane waves

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

  are just one term in the Fourier transform. The entire integral is real.
  (Add complex conjugate.)
- **Solutions to Maxwell's equations are superpositions of plane waves.**

Jackson, E&M, 1975

Fourier-transform Maxwell's Equations

- Substitute plane wave solutions into the differential form of Maxwell's Equations:

$\vec{\nabla} \cdot \vec{E} = 0$	Gauss' Law (Coulomb)
$\vec{\nabla} \cdot \vec{H} = 0$	Gauss' Law (magnetic field)
$\vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial \vec{H}}{\partial t}$	Faraday's Law
$\vec{\nabla} \times \vec{H} = \varepsilon_0 \frac{\partial \vec{E}}{\partial t}$	Ampere's Law

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

$\vec{k} \cdot \vec{E}_0 = 0$	Gauss' Law (Coulomb)
$\vec{k} \cdot \vec{H}_0 = 0$	Gauss' Law (magnetic field)
$\vec{k} \times \vec{E}_0 = \omega \mu_0 \vec{H}_0$	Faraday's Law
$\vec{k} \times \vec{H}_0 = -\omega \varepsilon_0 \vec{E}_0$	Ampere's Law

Fourier-transform Maxwell's Equations

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

$\vec{k}\cdot\vec{E}_0 = 0$	Gauss' Law (Coulomb)
$\vec{k}\cdot\vec{H}_0 = 0$	Gauss' Law (magnetic field)
$\vec{k}\times\vec{E}_0 = \omega\mu_0\vec{H}_0$	Faraday's Law
$\vec{k}\times\vec{H}_0 = -\omega\varepsilon_0\vec{E}_0$	Ampere's Law
$k^2 = \frac{\omega^2}{c^2}$	Wave equation (Dispersion relation)

Any solution to Maxwell's equation in vacuum can be written as a superposition of plane waves.

Electromagnetic waves are **transverse** (**E**, **H** **perpendicular** to **k**).

E ⊥ H, $\mathbf{E_0=Z_0H_0}$, $Z_0=\sqrt{(\mu_0/\varepsilon_0)}=377\ \Omega$ impedance of vacuum.

Polarized Light; Jones Vectors

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

- Select **k** along the z-axis. Then two field components E_x and E_y are sufficient.

$$\vec{E}(\vec{r},t) = \begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix} \exp[i(kz - \omega t)]$$

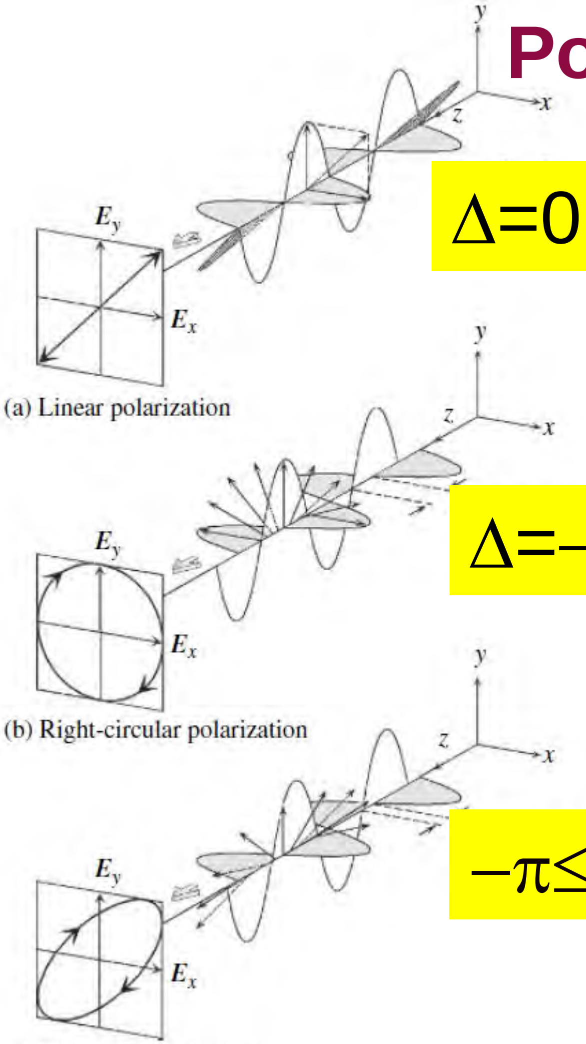
- **An EM wave is described by seven (7) real quantities:**
  - Direction of wave vector (two angles ϕ and θ).
  - Magnitude of wave vector (and angular frequency).
  - Two complex amplitudes E_{0x} and E_{0y} (**Jones vector**).
  - One of these (**absolute phase**) cannot be measured; leaving six parameters.

J. Humlicek, in Tompkins & Irene (Handbook of Ellipsometry)

$$\begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix} = E_0 \begin{pmatrix} X \exp i\Delta_X \\ Y \exp i\Delta_Y \end{pmatrix} = E_0 \begin{pmatrix} \sin\psi \exp i\Delta \\ \cos\psi \end{pmatrix} \exp i\Delta_y$$

  - We don't care about the **light intensity** and the **absolute phase**.
  - **ψ and Δ are called the ellipsometric angles; describe polarization of wave.**
  - $\psi=\arctan(X/Y)$; $\Delta=\Delta_X-\Delta_Y$; $\rho=\tan\psi\exp(i\Delta)$;

Polarized Light; Jones Vectors



(a) Linear polarization

(b) Right-circular polarization

(c) Elliptical polarization

$\Delta=0$

$\Delta=-\pi/2$

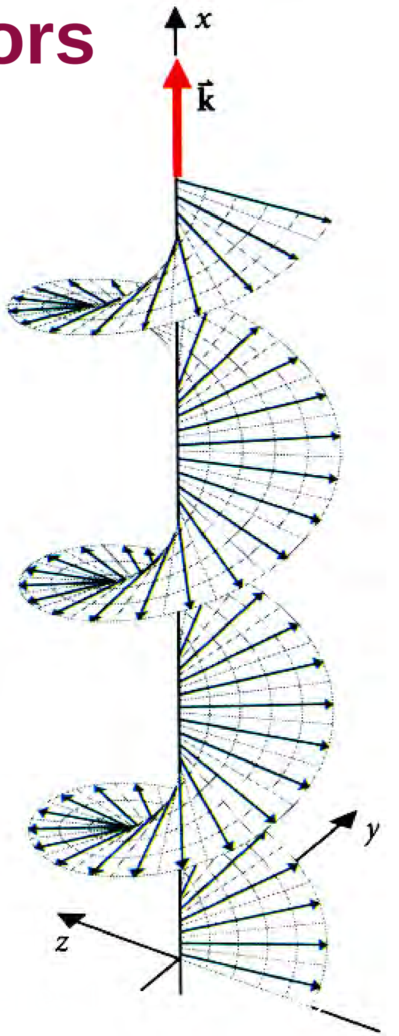
$-\pi\leq\Delta\leq\pi$

Jones Vector

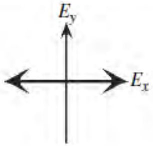
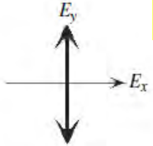
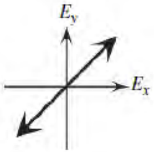
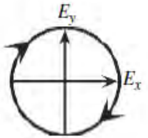
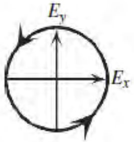
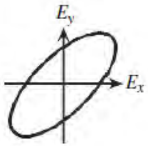
$$\begin{pmatrix} \sin\psi \exp i\Delta \\ \cos\psi \end{pmatrix}$$

Angle ψ:
Direction of linear polarization

H. Fujiwara



Polarized Light; Jones Vectors

Polarization	Polarization state	Jones vector	Stokes vector		
Linear polarization parallel to x axis		$\begin{bmatrix}1\\0\end{bmatrix}$	$\begin{bmatrix}1\\1\\0\\0\end{bmatrix}$	p-polarized	$\Delta=0$ $\psi=\pi/2$
Linear polarization parallel to y axis		$\begin{bmatrix}0\\1\end{bmatrix}$	$\begin{bmatrix}1\\-1\\0\\0\end{bmatrix}$	s-polarized	$\Delta=0$ $\psi=0$
Linear polarization oriented at 45°		$\frac{1}{\sqrt{2}}\begin{bmatrix}1\\1\end{bmatrix}$	$\begin{bmatrix}1\\0\\1\\0\end{bmatrix}$	45° linear	$\Delta=0$ $\psi=\pi/4$
Right-circular polarization		$\frac{1}{\sqrt{2}}\begin{bmatrix}1\\i\end{bmatrix}$	$\begin{bmatrix}1\\0\\0\\1\end{bmatrix}$	right-circular	$\Delta=-\pi/2$ $\psi=\pi/4$
Left-circular polarization		$\frac{1}{\sqrt{2}}\begin{bmatrix}1\\-i\end{bmatrix}$	$\begin{bmatrix}1\\0\\0\\-1\end{bmatrix}$	left-circular	$\Delta=\pi/2$ $\psi=\pi/4$
Elliptical polarization		$\begin{bmatrix}\sin\psi\exp(i\Delta)\\\cos\psi\end{bmatrix}$	$\begin{bmatrix}1\\-\cos 2\psi\\\sin 2\psi\cos\Delta\\-\sin 2\psi\sin\Delta\end{bmatrix}$	elliptical	

The polarization state of polarized light can be described with two parameters **ψ and Δ** called **ellipsometric angles**.

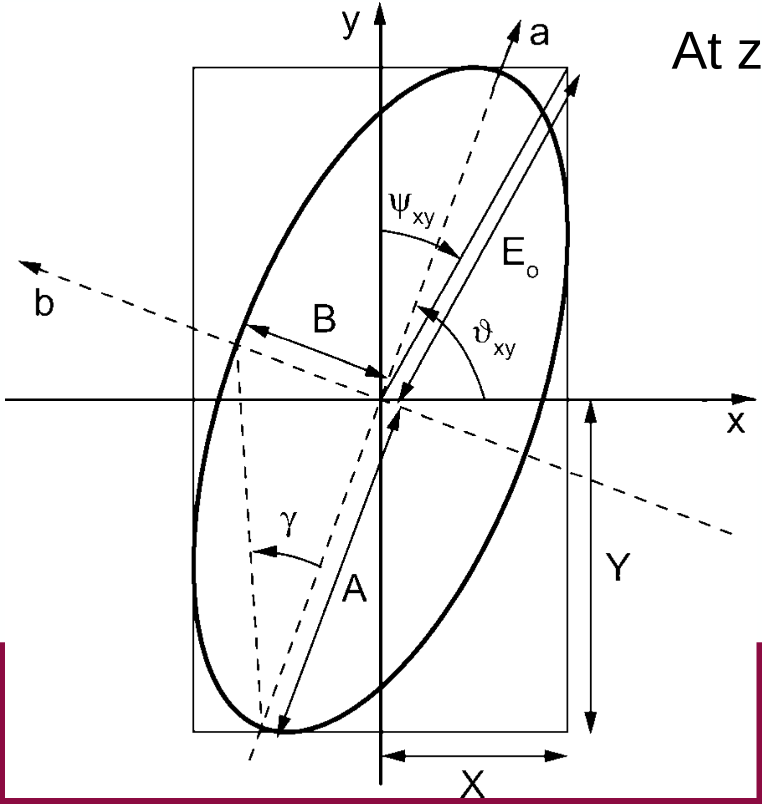
$-\pi\leq\Delta\leq\pi$

$0\leq\psi\leq\pi/2$

H. Fujiwara

Polarization Ellipse

$$\vec{E}(z=0,t) = E_0 \begin{pmatrix} \sin\psi \exp i\Delta \\ \cos\psi \end{pmatrix} \exp[-i\omega(t-\tau)t]$$



At z=0, the electric field vector traces out an ellipse.

Parameters of the ellipse:

- **Azimuth** ϑ
- Ratio tanγ major/minor axis

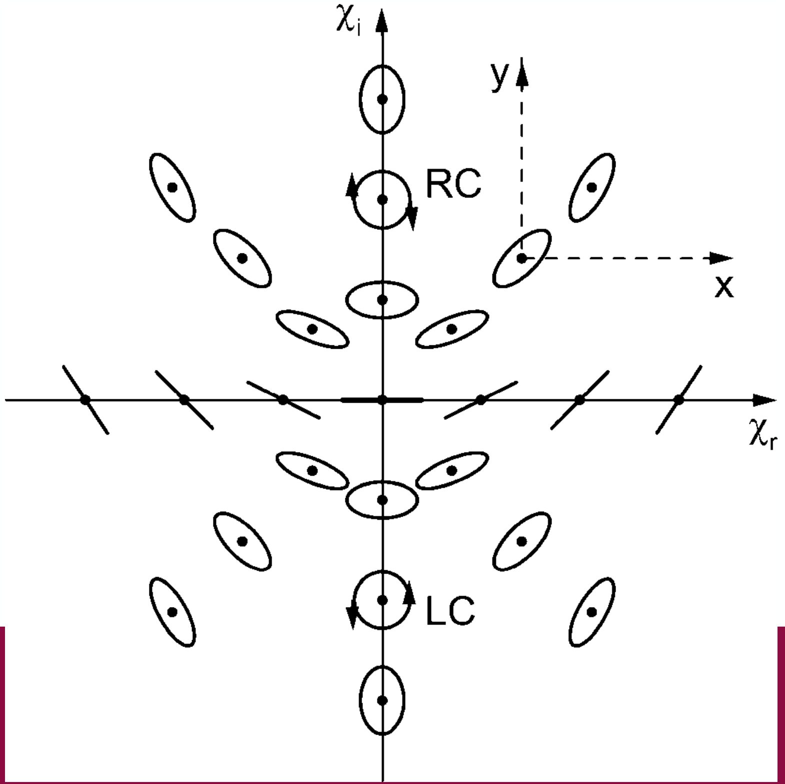
  Ellipticity e=tanγ=B/A

can be calculated from ψ,Δ.

J. Humlicek

Representation of Polarized Light by Complex Numbers

$$\vec{E}(z=0,t) = E_0 \begin{pmatrix} \sin\psi \exp i\Delta \\ \cos\psi \end{pmatrix} \exp[-i\omega(t-\tau)t]$$



$$\chi = \frac{\exp i\Delta}{\tan\psi} = \frac{\tan\vartheta + i\tan\gamma}{1 - i\tan\vartheta\tan\gamma}$$

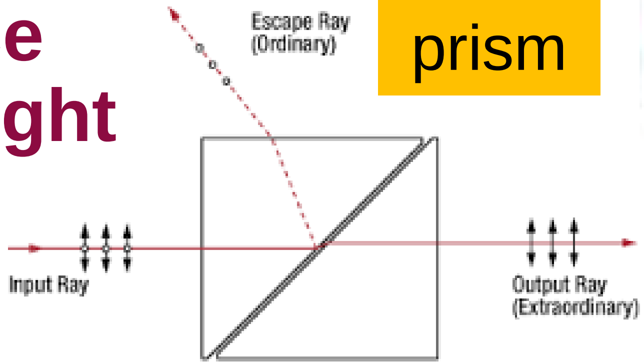
Complex number χ is related to ellipticity and azimuth of the polarization ellipse.

Also **Jones ratio**:

$$\rho = \tan\psi \exp i\Delta = \frac{E_x}{E_y}$$

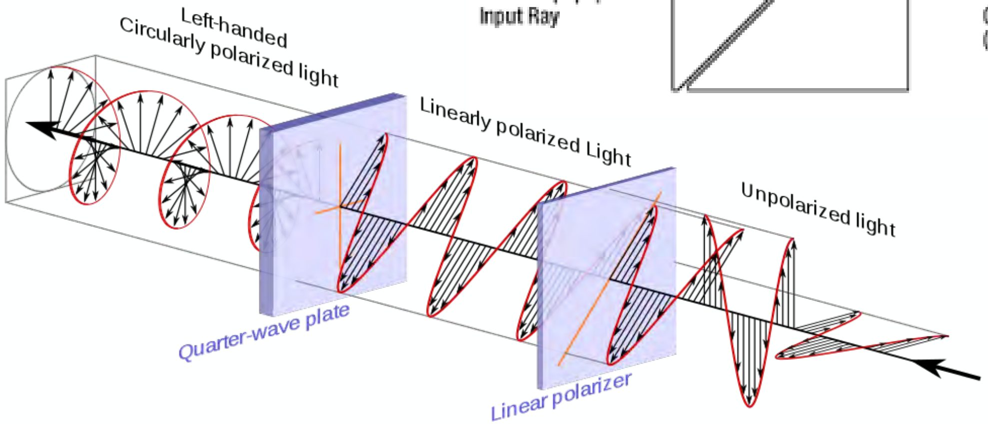
J. Humlicek

Optical Elements to Produce Polarized Light



calcite prism

wire grid (IR)



Linear polarizer:

Mica sheet, Prism (VIS/UV), wire grid (IR), metallic mirrors (VUV)

Compensator/retarder:

Quarter-wave plate

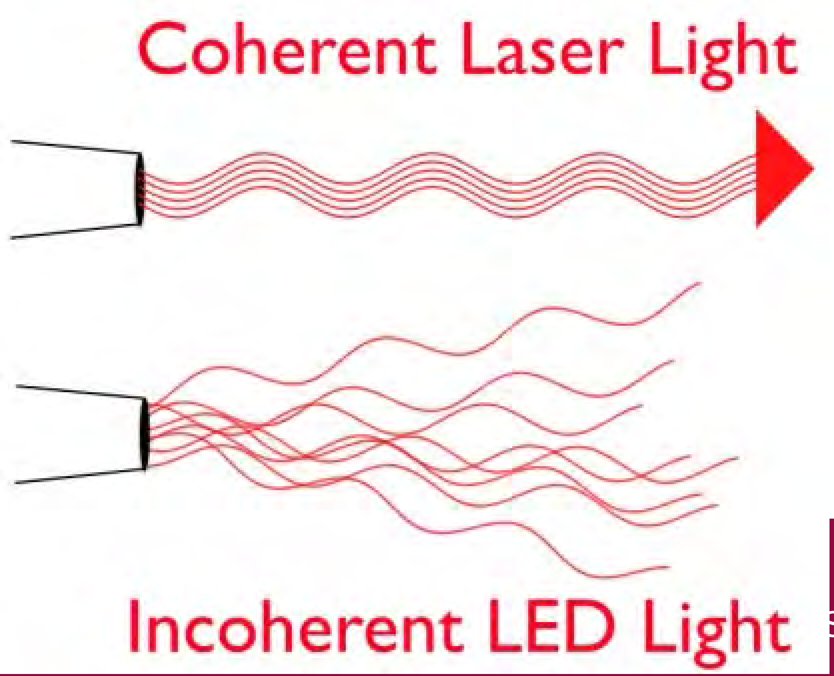
$$\Delta = \frac{2\pi}{\lambda}|n_e - n_o|d = \frac{\pi}{2}$$

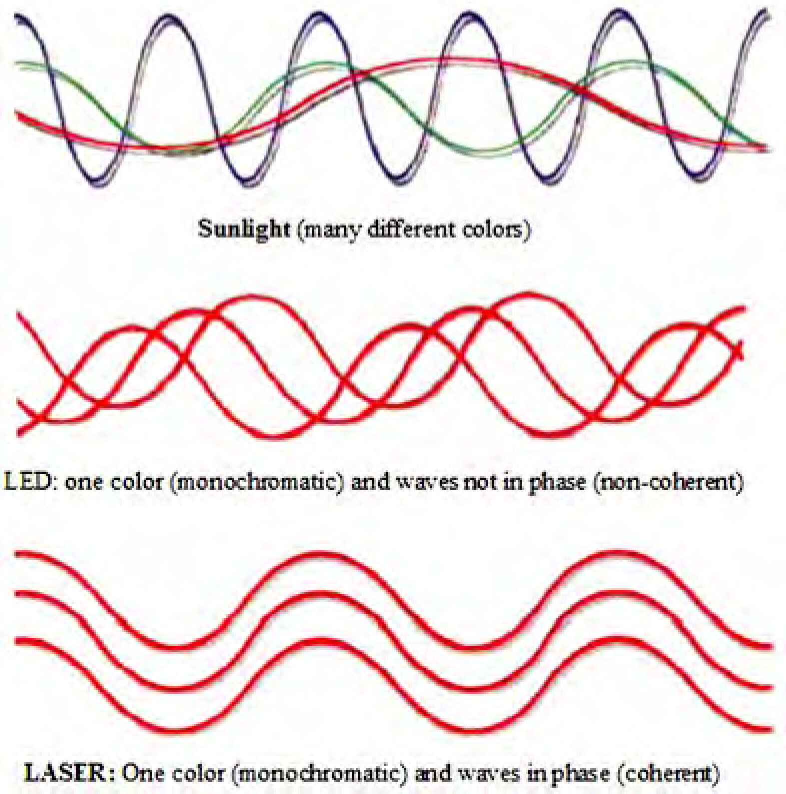
H. Fujiwara

Decoherence and Depolarization

In practice, light sources are superpositions with several frequencies, called **wave packets**. Similarly, light sources have **mixed polarization states**.

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





Stokes Parameters

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

$S_0 = I_x + I_y = E_x E_x^* + E_y E_y^*$ Total intensity

$S_1 = I_x - I_y = E_x E_x^* - E_y E_y^*$ s-polarized minus p-polarized

$S_2 = I_{45°} - I_{-45°} = E_x E_y^* + E_x^* E_y$ Diagonal difference

$S_3 = I_R - I_L = 2\,\mathrm{Re}\left(E_x^* E_y\right)$ Right minus left circular

$$0 \leq p = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \leq 1$$

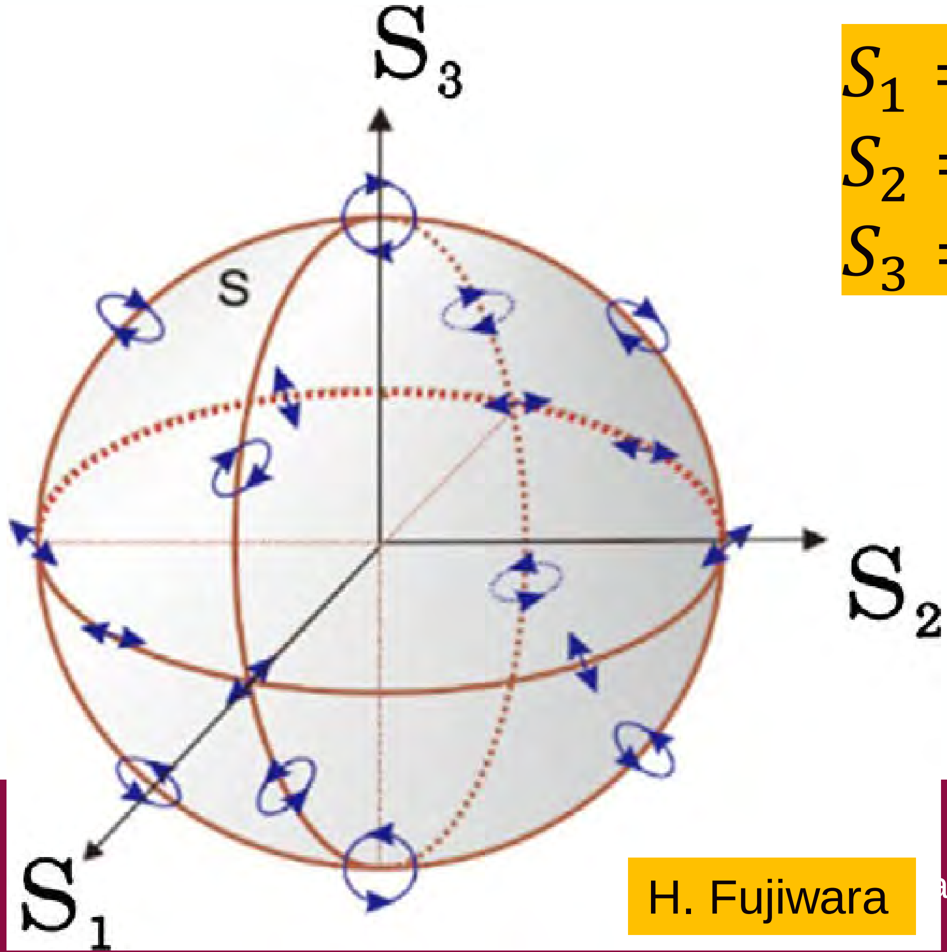
Degree of Polarization (%)

$$\tan(2\vartheta) = \frac{S_2}{S_1}$$

$$\tan(2\gamma) = \frac{S_3}{\sqrt{S_1^2 + S_2^s}}$$

The Stokes parameters are related to the azimuth ϑ and ellipticity γ of the polarization ellipse.

Poincare Sphere



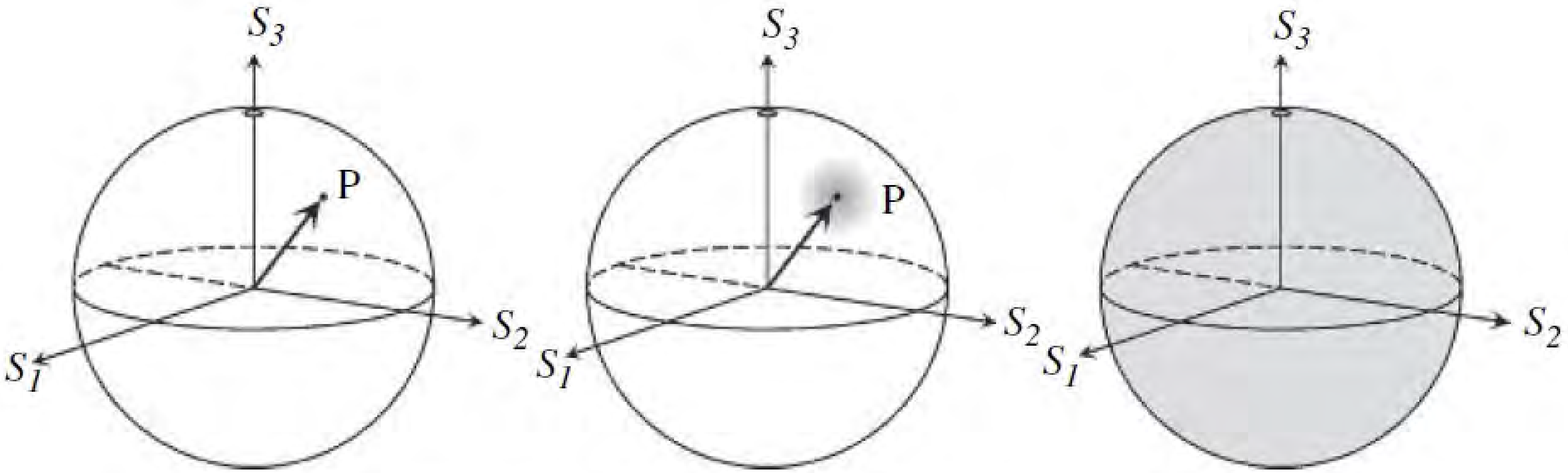
H. Fujiwara

$$S_1 = \cos 2\gamma \cos 2\vartheta = -\cos 2\psi$$
$$S_2 = \cos 2\gamma \sin 2\vartheta = \sin 2\psi \cos \Delta$$
$$S_3 = \sin 2\gamma = -\sin 2\psi \sin \Delta$$

The Stokes parameters for **completely polarized light**, taken as coordinates, define a **point on the surface** of a unit sphere (Poincare sphere).

Poles: Circularly polarized
Equator: Linearly polarized

Partially Polarized Light



(a) Totally polarized light (b) Partially polarized light (c) Unpolarized light

Totally polarized light: point on the surface of Poincare sphere.
Partially polarized light: point inside the sphere.
Completely unpolarized light: point in the center of the sphere.

H. Fujiwara

Origins of Depolarization

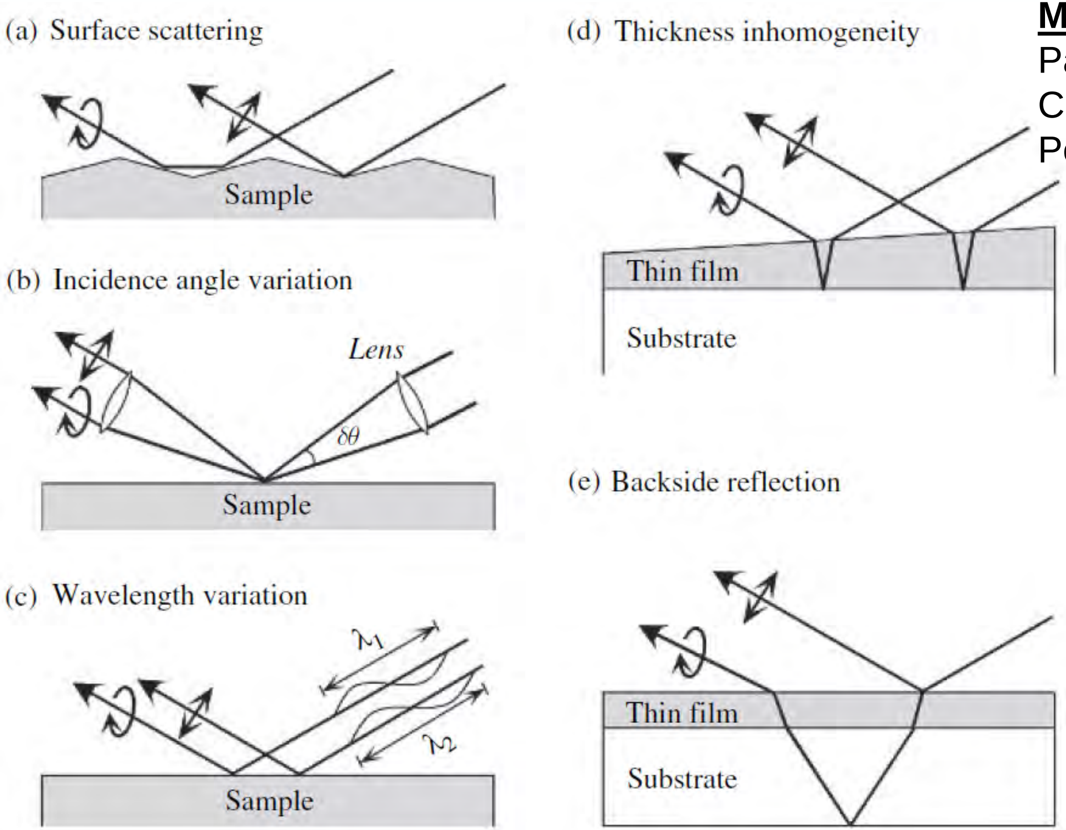


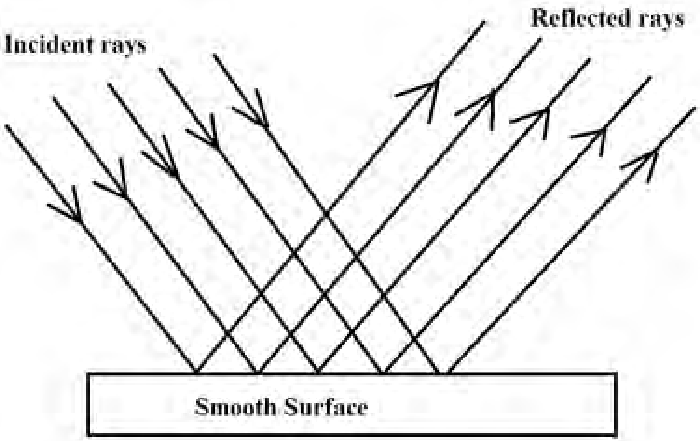
Figure 4.30 Depolarization of incident light by (a) surface scattering, (b) incidence angle variation, (c) wavelength variation, (d) thickness inhomogeneity, and (e) backside reflection.

More general:
Patterned substrate, contacts,
Contaminated substrate
Peeling layers, etc.

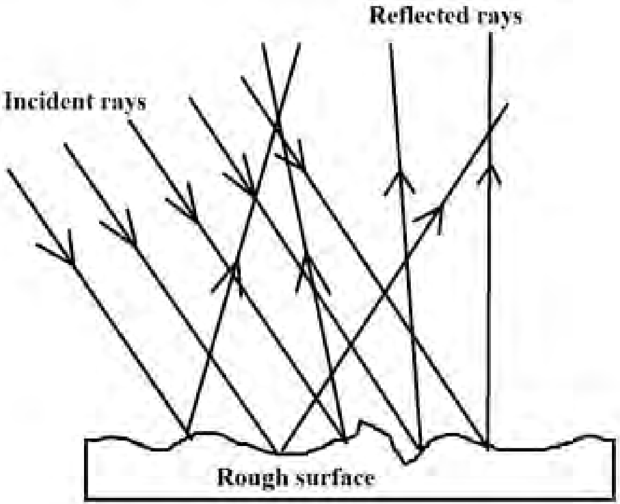
My strategy:
Always acquire full Mueller matrix.
Inspect off-diagonal blocks.
If there is nothing to see, deselect Mueller matrix items, analyze isotropic ellipsometric angles and depolarization.

Measurement time is the same, but file size increases.

Reflection from a Rough Surface



Specular Reflection



Specular+Diffuse
Usually still polarized

<u>Debye-Waller correction:</u>

Assumes sinusoidal roughness

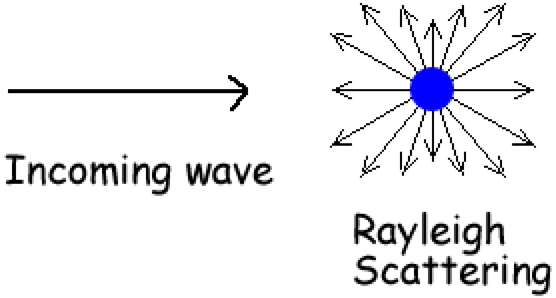
$R_{rough}=R_0\exp[-(4\pi\sigma n\cos\theta/\lambda)^2]$

σ: rms surface roughness parameter

Also: I. Ohlidal, F. Lukes, and K. Navratil, Surf. Sci. **45**, 91 (1974). 98 citations.

D.K.G. de Boer, Phys. Rev. B **49**, 5817 (1994).

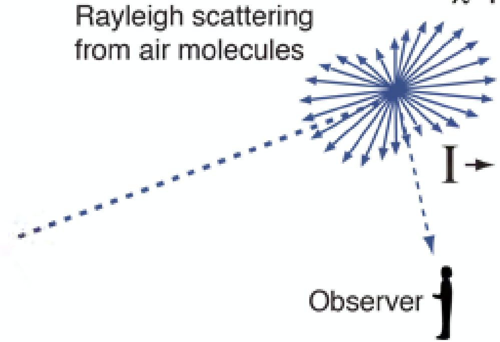
Raleigh Scattering (Elastic)



Why is the sky blue?

$$I = I_0 \frac{8\pi^4 N\alpha^2}{\lambda^4 R^2}(1 + \cos^2\theta)$$

Scattering at right angles is half the forward intensity for Rayleigh scattering



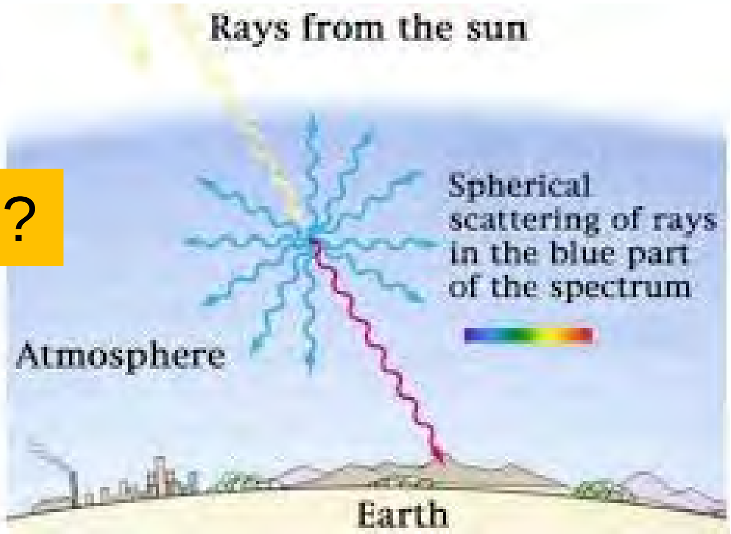
$$I \rightarrow \frac{1}{\lambda^4}$$

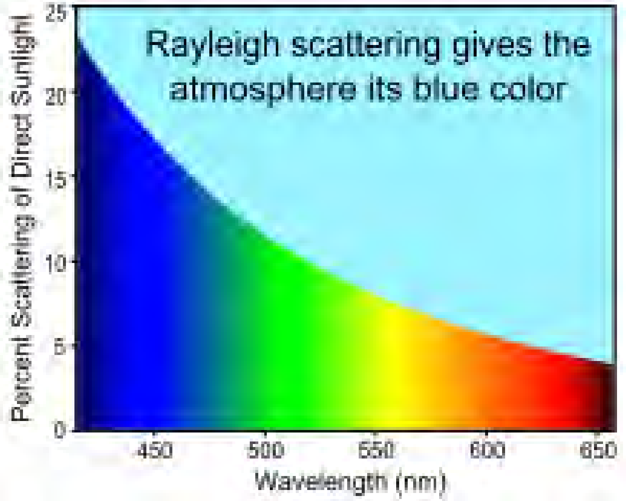
N = # of scatterers

α = polarizability

R = distance from scatterer

The strong wavelength dependence of Rayleigh scattering enhances the short wavelengths, giving us the blue sky.





Ellipsometry Measurement

Polarization State
Jones Vector

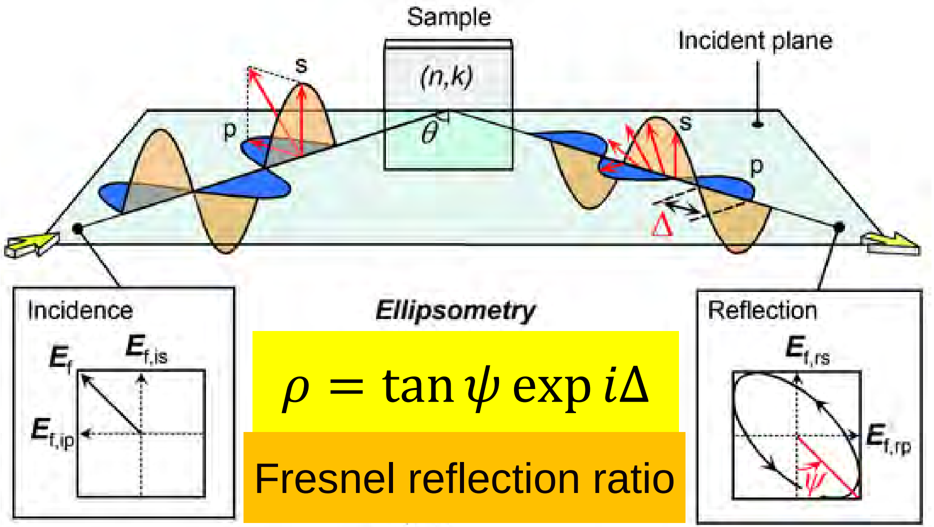
$$J = \begin{pmatrix} E_{0x} \\ E_{0y} \end{pmatrix}$$

Ellipsometry Experiment

$$J_{\text{out}} = \begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} J_{\text{in}}$$

Fresnel reflection coefficients

$$\begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} = r_{ss} \begin{pmatrix} \rho & \rho_{ps} \\ \rho_{sp} & 1 \end{pmatrix}$$



$$\rho = \tan\psi \exp i\Delta$$

Fresnel reflection ratio

Anisotropy or depolarization (not both)

Isotropic surface:
Off-diagonal elements vanish.

$$\begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} = r_{ss} \begin{pmatrix} \rho & 0 \\ 0 & 1 \end{pmatrix}$$

What is an Isotropic Surface?

- Surface of a cubic crystal (any crystal orientation)
- Surface of an amorphous material
- Surface of a **uniaxial crystal**, if
- the optical axis is perpendicular to the surface
- the optical axis is perpendicular to the plane of incidence
- the optical axis is in the plane of incidence
- Result: Diagonal Jones Matrix

$$\begin{pmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{pmatrix} = r_{ss} \begin{pmatrix} \rho & 0 \\ 0 & 1 \end{pmatrix}$$

- See Case A in **O. Arteaga**, Thin Solid Films **571**, 584 (2014).

Azzam/Bashara

Jones Matrix for Optical Elements

Optical element	Corresponding Jones matrix
Linear polarizer with axis of transmission horizontal[1]	$\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$
Linear polarizer with axis of transmission vertical[1]	$\begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}$
Linear polarizer with axis of transmission at ±45° with the horizontal[1]	$\frac{1}{2}\begin{pmatrix} 1 & \pm 1 \\ \pm 1 & 1 \end{pmatrix}$
Quarter-wave plate with fast axis vertical[2][note 1]	$e^{\frac{i\pi}{4}}\begin{pmatrix} 1 & 0 \\ 0 & -i \end{pmatrix}$
Quarter-wave plate with fast axis horizontal[2]	$e^{-\frac{i\pi}{4}}\begin{pmatrix} 1 & 0 \\ 0 & i \end{pmatrix}$
Half-wave plate with fast axis at angle θ w.r.t the horizontal axis[3]	$e^{-\frac{i\pi}{2}}\begin{pmatrix} \cos^2\theta - \sin^2\theta & 2\cos\theta\sin\theta \\ 2\cos\theta\sin\theta & \sin^2\theta - \cos^2\theta \end{pmatrix}$

Ellipsometry of a flat surface (bulk sample)

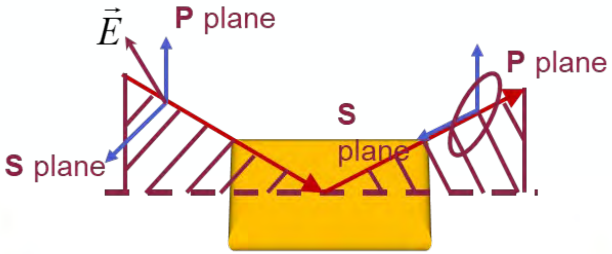
We measure the change in the polarization state of light, when reflected by a flat surface (bulk).

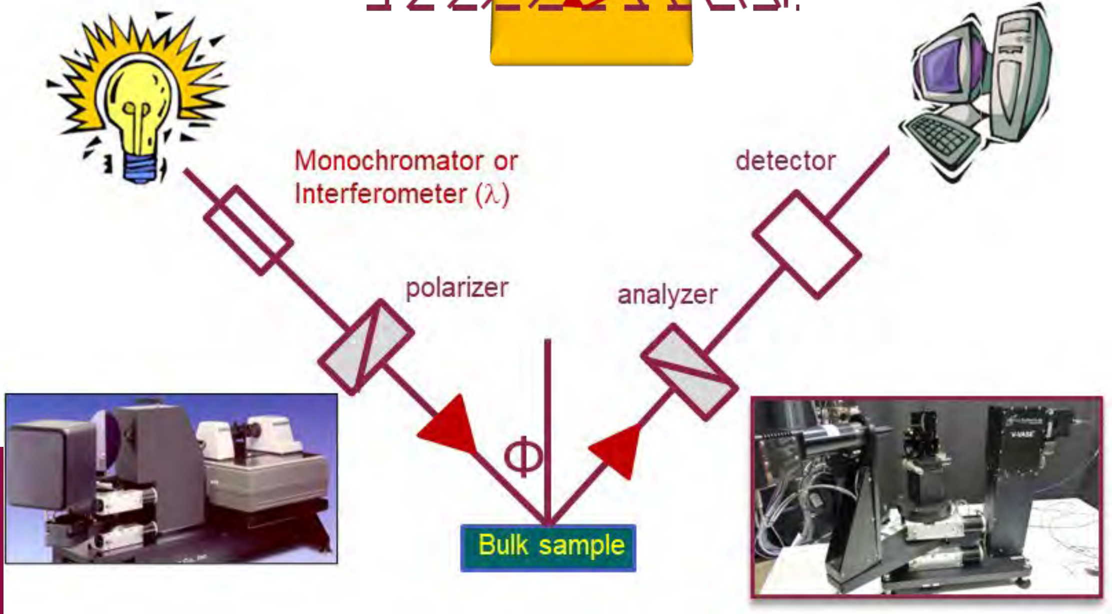
Fresnel Reflectance ratio

$$\rho = \frac{R_p}{R_s} = \frac{E_{rp}}{E_{ip}} \cdot \frac{E_{is}}{E_{rs}} = \tan \Psi e^{i\Delta}$$

$$\tilde{\varepsilon} = \sin^2 \phi \left[1 + \tan^2 \phi \cdot \left(\frac{1-\rho}{1+\rho} \right)^2 \right]$$

Angle of incidence





Result:

$$\tilde{\varepsilon} = \varepsilon_1 + i\varepsilon_2$$

Optical constants versus photon energy

Ellipsometry Instrumentation

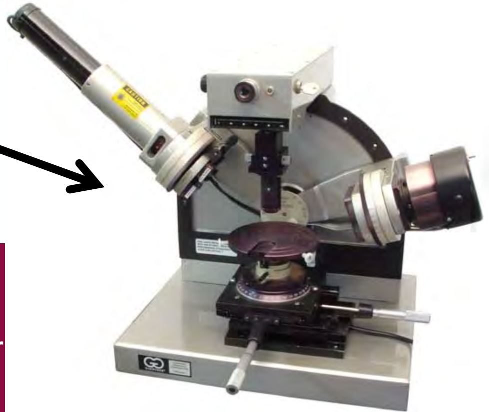
VASE: One wavelength at a time:
Calculate derivatives.
Resolve narrow line shapes.

Dual rotating compensator (**RC2**):
Full 4 by 4 Mueller matrix

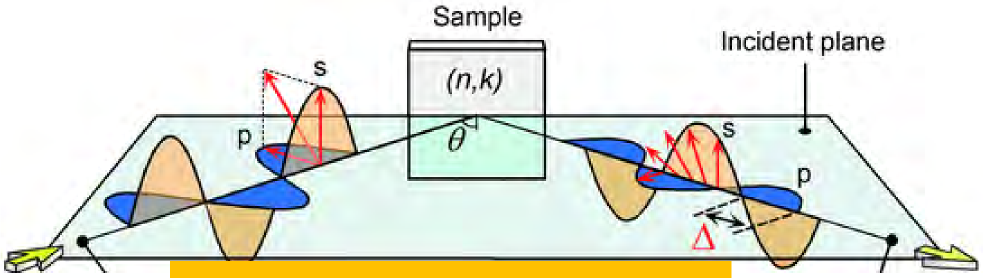
Fourier-Transform Infrared (**FTIR**)

Single-wavelength ellipsometer

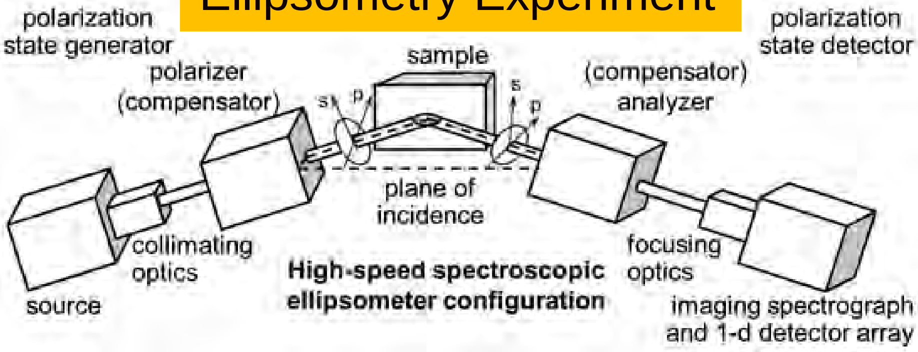
Far-infrared ellipsometer (not shown)
Terahertz ellipsometer (not shown)
VUV ellipsometer (not shown)
Inline (fab) metrology tools
Imaging ellipsometer (Accurion)
What's next ???



Mueller Matrix Ellipsometry



Ellipsometry Experiment



$$S = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix}$$

Polarization States for Incident and Reflected Beams Described by Stokes Vectors

$$S_{\text{out}} = \hat{M} S_{\text{in}}$$

$$\hat{M} = \begin{pmatrix} M_{00} & M_{01} & M_{02} & M_{03} \\ M_{10} & M_{11} & M_{12} & M_{13} \\ M_{20} & M_{21} & M_{22} & M_{23} \\ M_{30} & M_{31} & M_{32} & M_{33} \end{pmatrix}$$

Measures **anisotropy and depolarization.**
Requires two rotating compensators.
Beetles, cancer research, magnetic field.

Mueller Matrix for Isotropic Non-Depolarizing Surfaces

$$\mathbf{J}_{sample} = \begin{bmatrix} r_p & 0 \\ 0 & r_s \end{bmatrix}$$

$$\mathbf{M}_{sample} = \begin{bmatrix} 1 & -N & 0 & 0 \\ -N & 1 & 0 & 0 \\ 0 & 0 & C & S \\ 0 & 0 & -S & C \end{bmatrix}$$

$$N = \cos(2\psi)$$
$$S = \sin(2\psi)\sin(\Delta)$$
$$C = \sin(2\psi)\cos(\Delta)$$
$$N^2 + S^2 + C^2 = 1$$

Standard ellipsometry:

- Thickness measurements of thin films
- Optical functions of isotropic materials

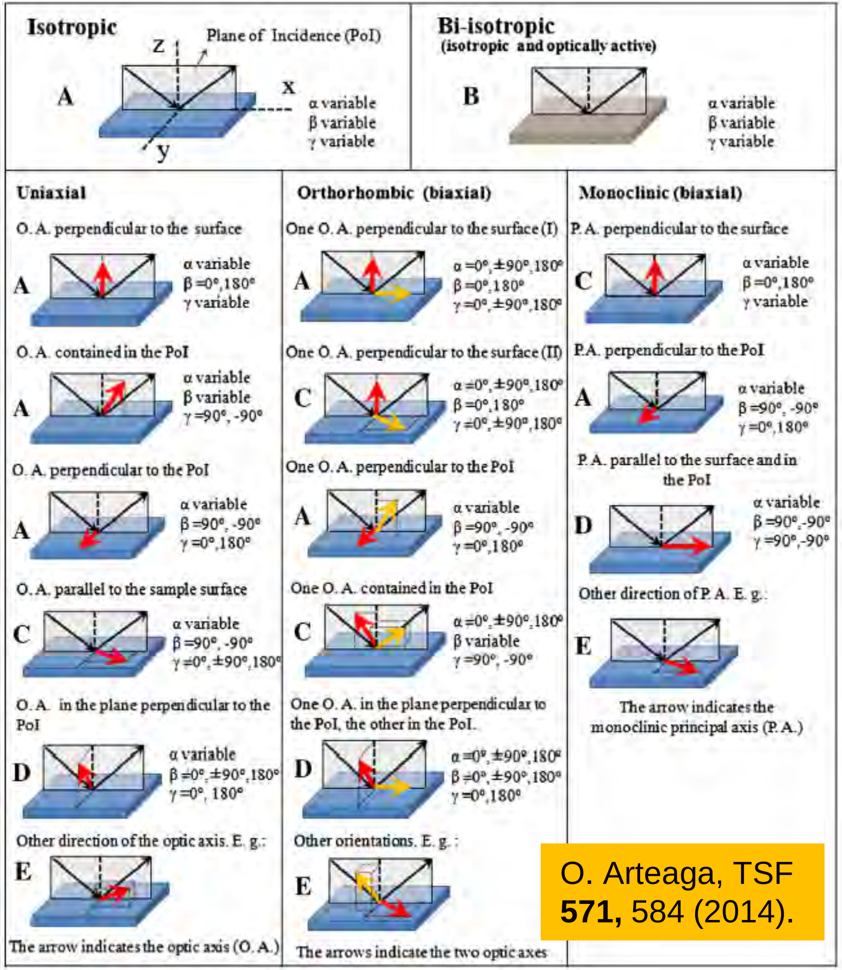
$$\rho = (\rho_{real} + i\rho_{imag}) = \frac{r_p}{r_s} = \tan(\psi)e^{i\Delta} = \frac{C + iS}{1 + N}$$

This Mueller matrix depends only on 2 parameters

O. Arteaga

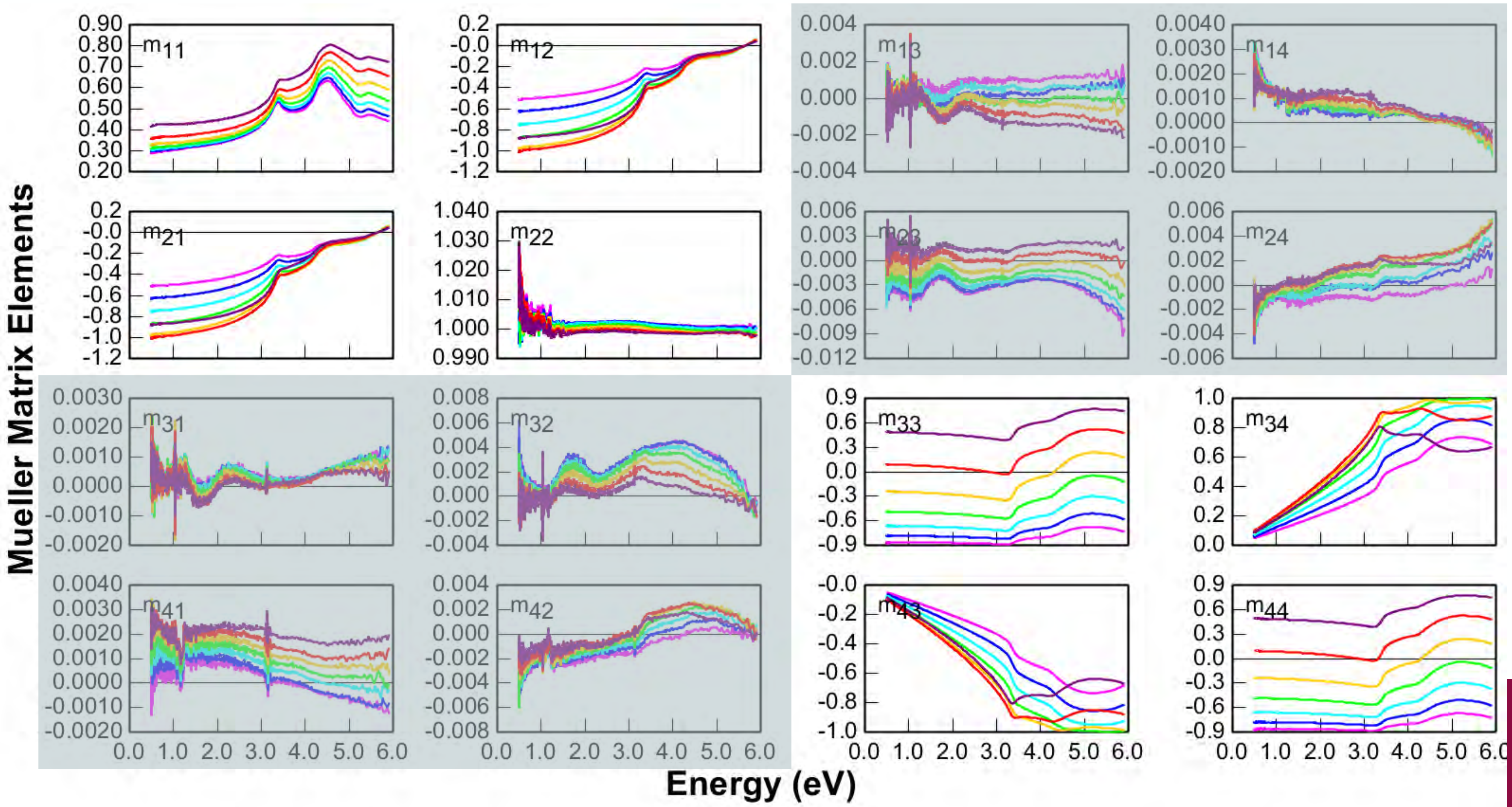
Symmetries of Non-Depolarizing Mueller Matrices

Symmetry	Mueller	Jones
A	$\begin{bmatrix} 1 & m_{01} & 0 & 0 \\ m_{01} & 1 & 0 & 0 \\ 0 & 0 & m_{22} & m_{23}^* \\ 0 & 0 & -m_{23}^* & m_{22} \end{bmatrix}$	$\begin{bmatrix} \rho & 0 \\ 0 & 1 \end{bmatrix}$
B	$\begin{bmatrix} 1 & m_{01} & m_{02}^* & m_{03} \\ m_{01} & m_{11} & m_{12}^* & m_{13} \\ -m_{02}^* & -m_{12}^* & m_{22} & m_{23}^* \\ m_{03} & m_{13} & -m_{23}^* & m_{33} \end{bmatrix}$	$\begin{bmatrix} \rho & \rho_{ps} \\ -\rho_{ps} & 1 \end{bmatrix}$
C	$\begin{bmatrix} 1 & m_{01} & m_{02} & m_{03}^* \\ m_{01} & m_{11} & m_{12} & m_{13}^* \\ -m_{02} & -m_{12} & m_{22} & m_{23}^* \\ m_{03}^* & m_{13}^* & -m_{23}^* & m_{33} \end{bmatrix}$	$\begin{bmatrix} \rho & \rho_{ps} \\ -\rho_{ps} & 1 \end{bmatrix}$
D	$\begin{bmatrix} 1 & m_{01} & m_{02} & m_{03}^* \\ m_{01} & m_{11} & m_{12} & m_{13}^* \\ m_{02} & m_{12} & m_{22} & m_{23}^* \\ -m_{03}^* & -m_{13}^* & -m_{23}^* & m_{33} \end{bmatrix}$	$\begin{bmatrix} \rho & \rho_{ps} \\ \rho_{ps} & 1 \end{bmatrix}$
E	$\begin{bmatrix} 1 & m_{01} & m_{02} & m_{03}^* \\ m_{10} & m_{11} & m_{12} & m_{13}^* \\ m_{20} & m_{21} & m_{22} & m_{23}^* \\ m_{30}^* & m_{31}^* & m_{32}^* & m_{33} \end{bmatrix}$	$\begin{bmatrix} \rho & \rho_{ps} \\ \rho_{sp} & 1 \end{bmatrix}$

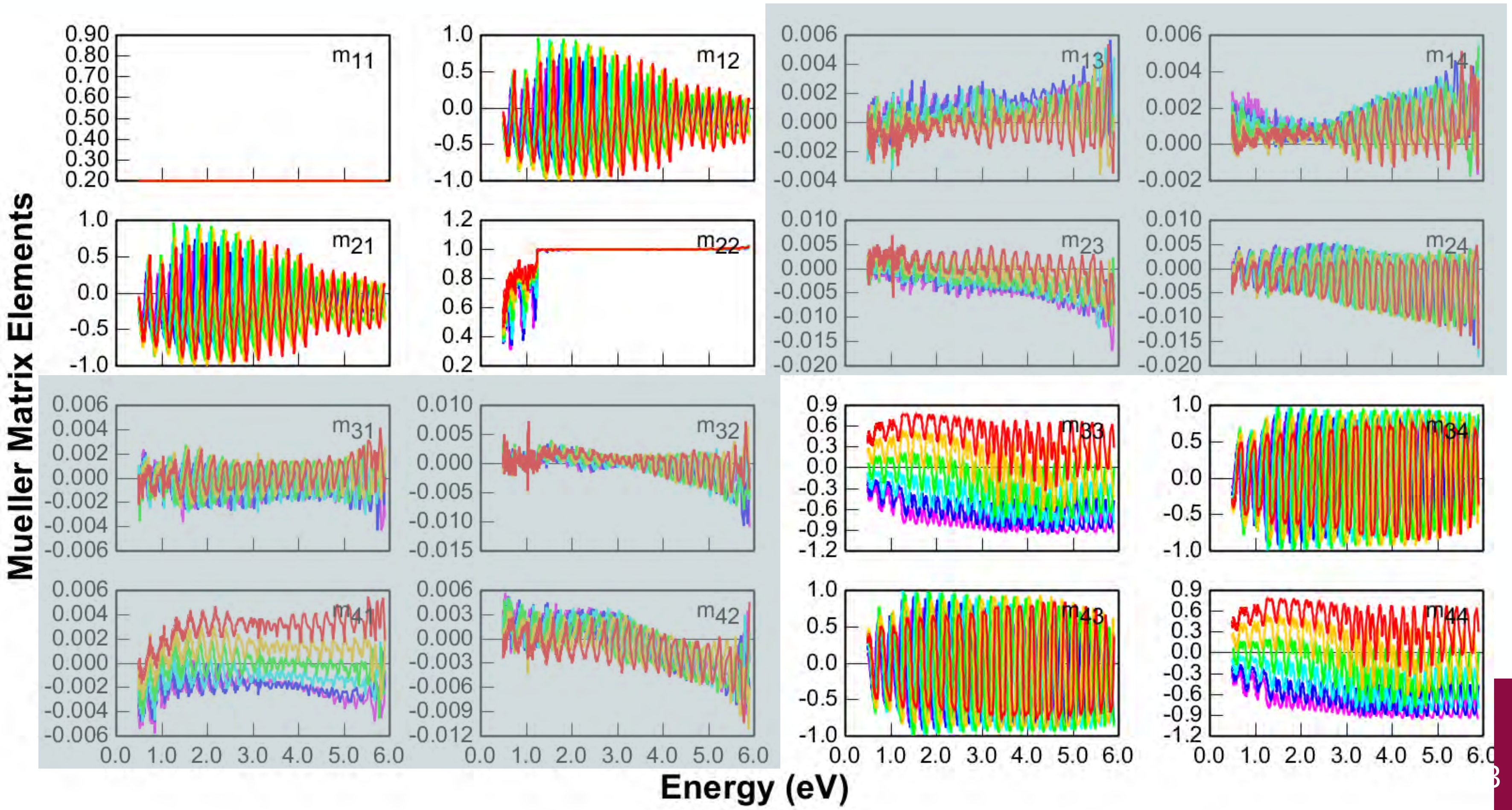


O. Arteaga, TSF **571,** 584 (2014).

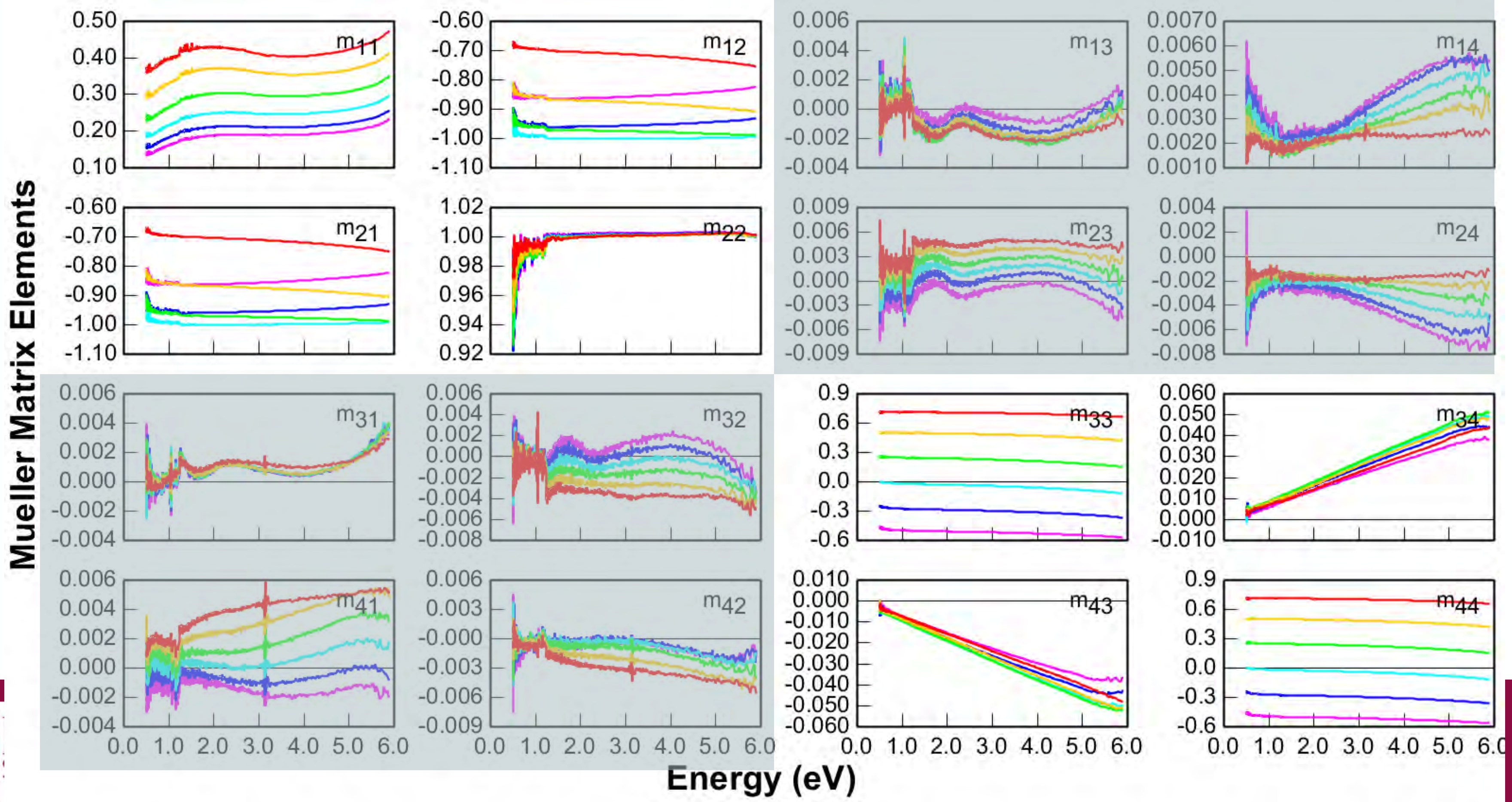
Mueller Matrix Examples: Si Calibration Wafer



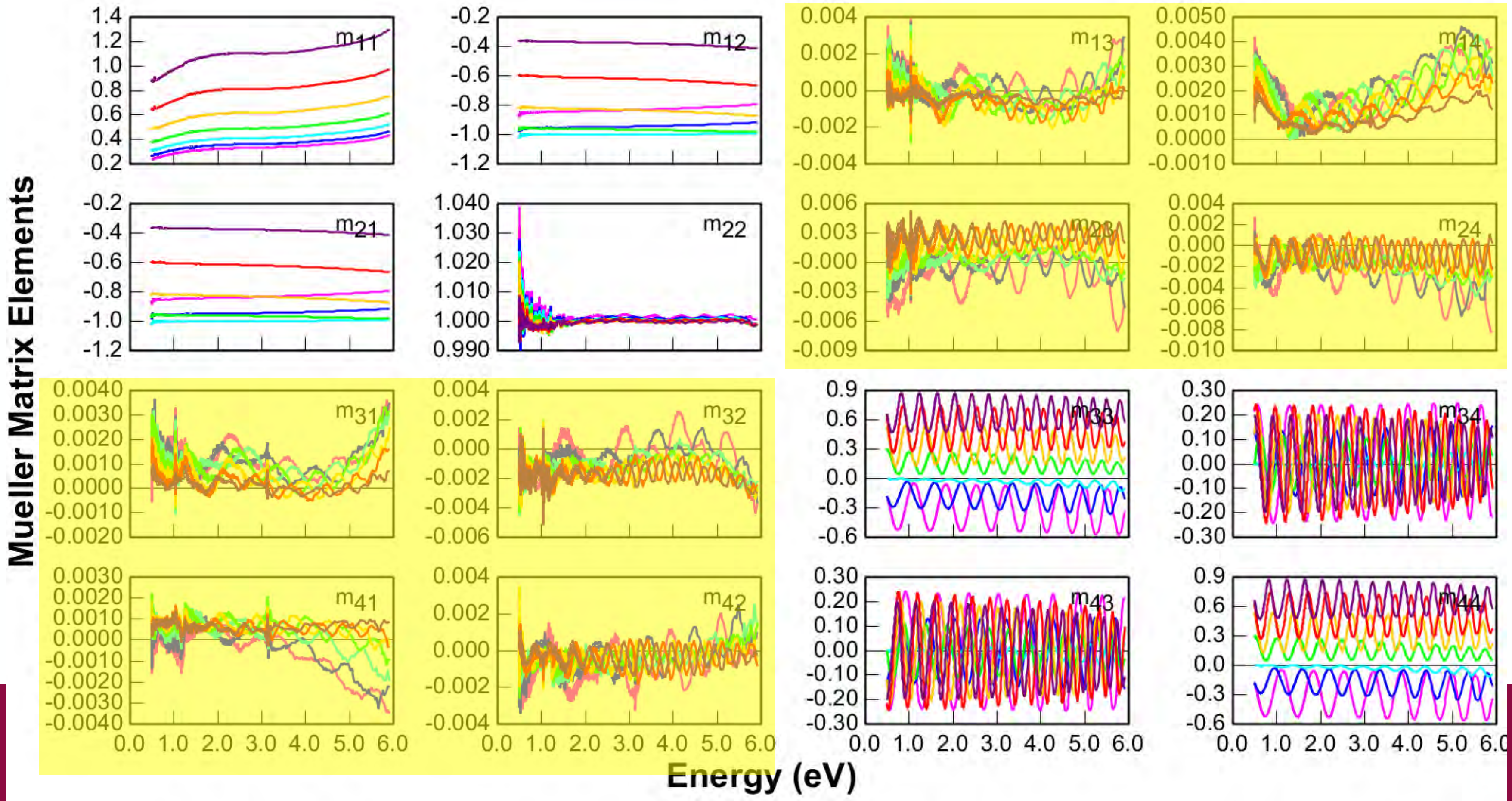
Mueller Matrix Examples: Si with 198 nm Oxide



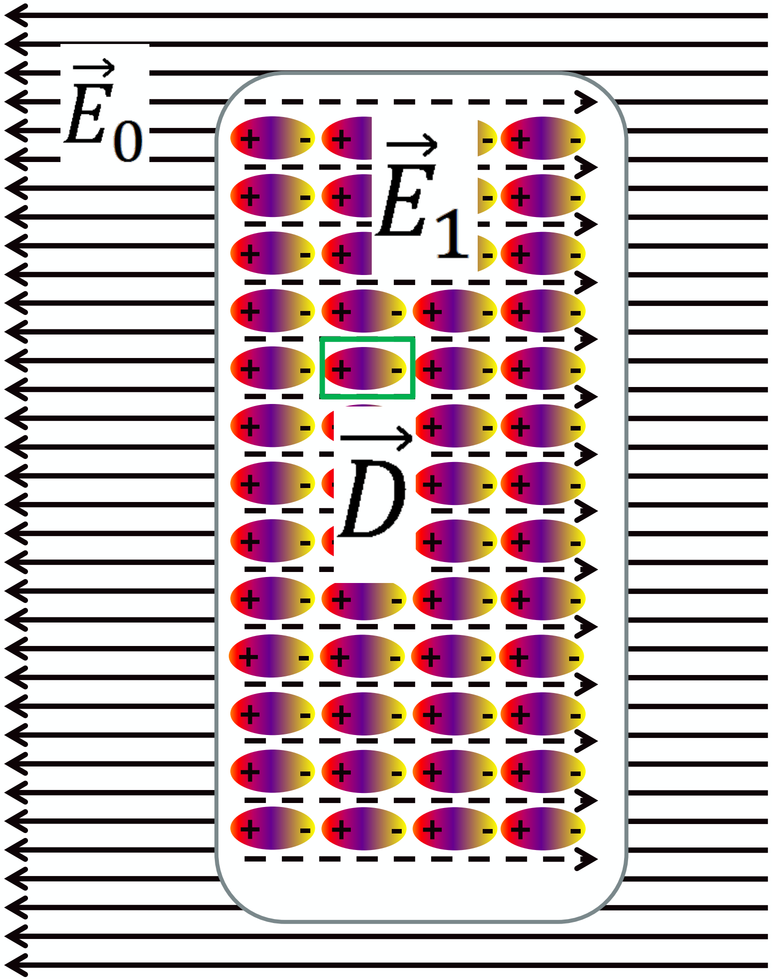
MM Examples: c-oriented Sapphire (SSP)



MM Examples: c-oriented Sapphire (DSP)



Dielectric in Static Electric and Magnetic Fields



Applied external electric field $\mathbf{E}_0$
(homogeneous, constant)
Infinite dielectric
(ignore boundary effects)

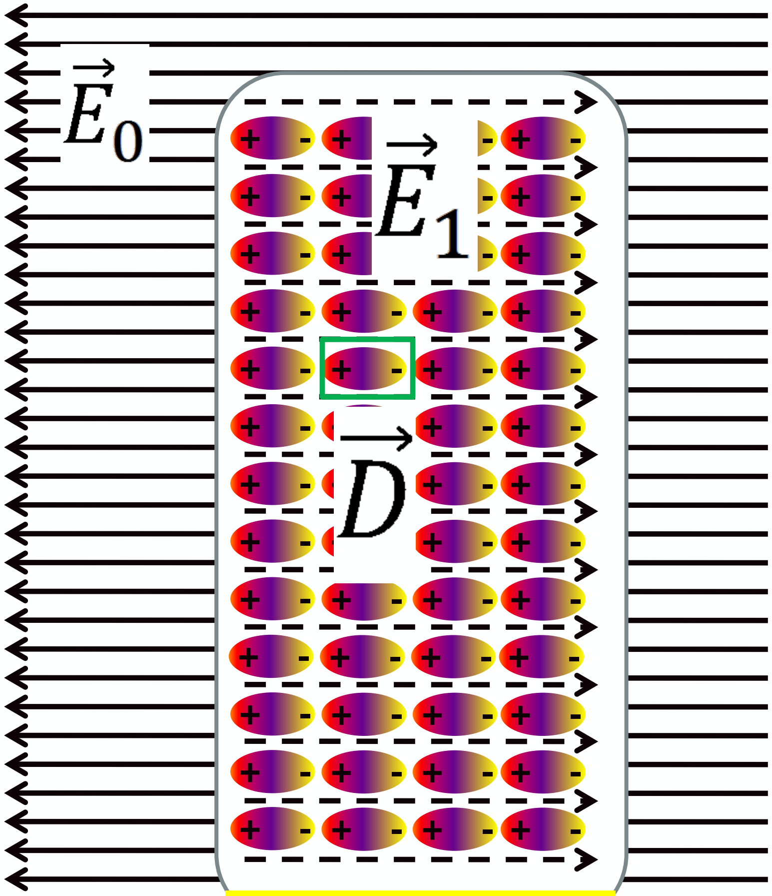
Charges move in response to $\mathbf{E}_0$
Average charge density still zero.
Induced (depolarizing) electric field $\mathbf{E}_1$
weakens applied field $\mathbf{E}_0$.

Local electric field (inside)

$\mathbf{E}=\mathbf{E}_{local}=\mathbf{E}_0+\mathbf{E}_1$
Metal: $\mathbf{E}_{local}=0$ (for $\omega=0$)
Dielectric: $E_{local}<E_0$ (screening)
E_{local} depends on crystal shape (boundary conditions)

Nye, Physical Properties of Crystals

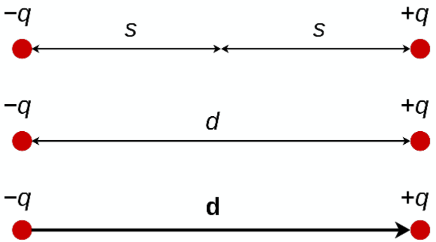
Dielectric Polarization, Dielectric Displacment



$E = E_{local} = E_0 + E_1$

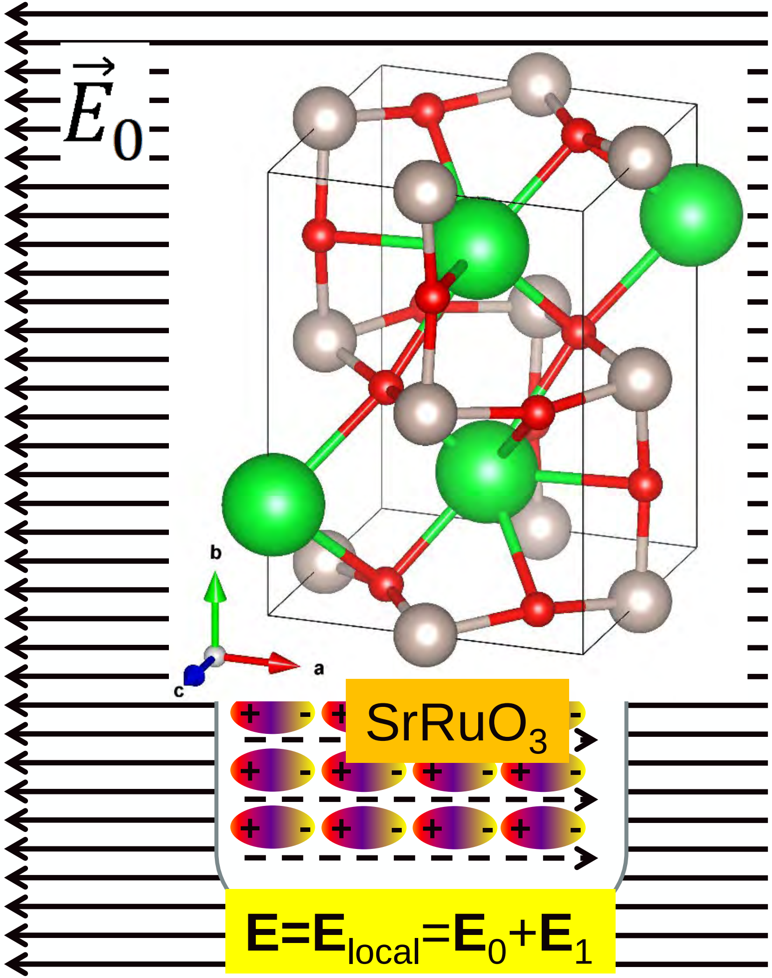
Applied external electric field $\mathbf{E}_0$
(homogeneous, constant)
Infinite dielectric (ignore boundary effects)
Total electric field $\mathbf{E}=\mathbf{E}_0+\mathbf{E}_1$

Charges move:
Dipole moment
$\mathbf{p}=q\mathbf{d}$
($\mathbf{d}$ from –q to +q)

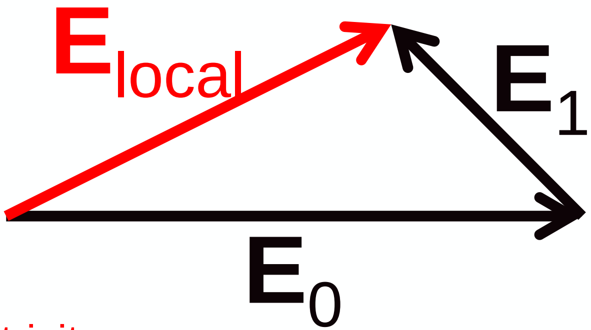


Dielectric polarization P
Dipole moment per unit volume
Dielectric Displacement: $\mathbf{D}=\varepsilon_0\mathbf{E}+\mathbf{P}$
Linear dielectric susceptibility
$\mathbf{P}=\varepsilon_0\chi_e\mathbf{E}$
Dielectric constant: $\varepsilon=1+\chi_e$, $\mathbf{D}=\varepsilon_0\varepsilon\mathbf{E}$

Complications



$\mathbf{E}_{local}=\mathbf{E}_0+\mathbf{E}_1$



Anisotropy
requires tensors.

Ferro-/Pyro-/Piezoelectricity

Non-zero polarization for zero field ($\mathbf{E}_0$=0).

$\mathbf{P}(E_0=0)=\mathbf{P}_r+\mathbf{p}\Delta T+d_{ijk}X_{jk}$

$\partial\mathbf{P}_r/\partial t=0$

Nonlinear effects

$\mathbf{P}(\mathbf{E})=\mathbf{P}_r+\varepsilon_0\chi_e\mathbf{E}+\varepsilon_0\chi_e^{(2)}\mathbf{E}\otimes\mathbf{E}+\varepsilon_0\chi_e^{(3)}{}_{ijkl}E_jE_kE_l+\ldots$

Magneto-electric effects

$\mathbf{P}=\mathbf{P}_r+\varepsilon_0\chi_e\mathbf{E}+\varepsilon_0\delta H$

$\varepsilon=1+\chi_e$

$\boldsymbol{D}=\varepsilon_0\varepsilon\boldsymbol{E}$

Dielectric Displacement:

$\mathbf{D}=\mathbf{P}_r+\varepsilon_0\mathbf{E}+\varepsilon_0\chi_e\mathbf{E}+\varepsilon_0\delta\mathbf{H}$

$\mathbf{D}=\mathbf{P}_r+\varepsilon_0\varepsilon\mathbf{E}+\varepsilon_0\delta\mathbf{H}$

Dielectric constant ε

Nye, Physical Properties of Crystals

Magnetostatics and Magnetization

Electric field strength **E**
Dielectric polarization **P**: electric dipole moment per unit volume
Dielectric displacement $\mathbf{D}=\varepsilon_0\mathbf{E}+\mathbf{P}=\mathbf{P}_r+\varepsilon_0\mathbf{E}+\varepsilon_0\chi_e\mathbf{E}+\varepsilon_0\delta\mathbf{H}$
Magnetic field strength **H**
Magnetization **M**: magnetic dipole moment per unit volume
$\mathbf{M}=\mathbf{M}_r+\mu_0\chi_m\mathbf{H}+\mu_0\gamma\mathbf{E}$ ($\mathbf{M}_r$ remanence, $\partial\mathbf{M}_r/\partial t=0$)
Magnetic susceptibility χ_m

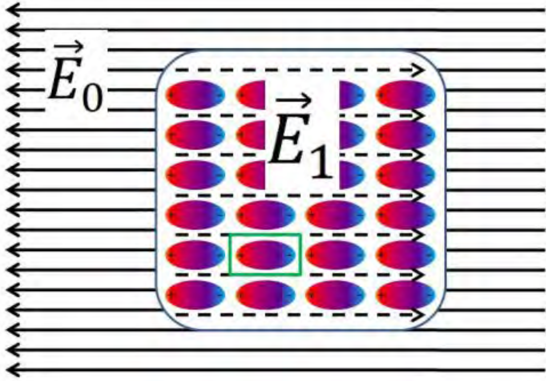
Magnetic flux density **B**
$\mathbf{B}=\mu_0\mathbf{H}+\mathbf{M}=\mathbf{M}_r+\mu_0\mu\mathbf{H}+\mu_0\gamma\mathbf{E}$
$\mu=1+\chi_m$ magnetic permeability ($\mu=1$ unless $\omega=0$)

For electromagnetic waves with $\omega\neq0$, we can set $\mu=1$.

AC Response Function: Dispersion, Nonlocality

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

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

Nonlocal effects scale like $2\pi an/\lambda_0$

Dielectric function ε depends on frequency ω (dispersion).

Nonlocality Example: Birefringence in Cubic Crystals

$$\Delta\varepsilon_{ij}(\vec{k}) = \alpha_{ijkl}k_k k_l$$

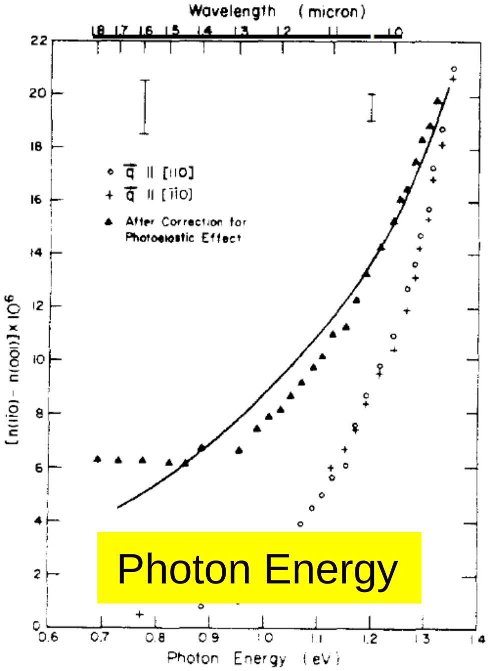
vanishes along [001], but not along [110]

Birefringence

$\Delta n = n_{110} - n_{100}$

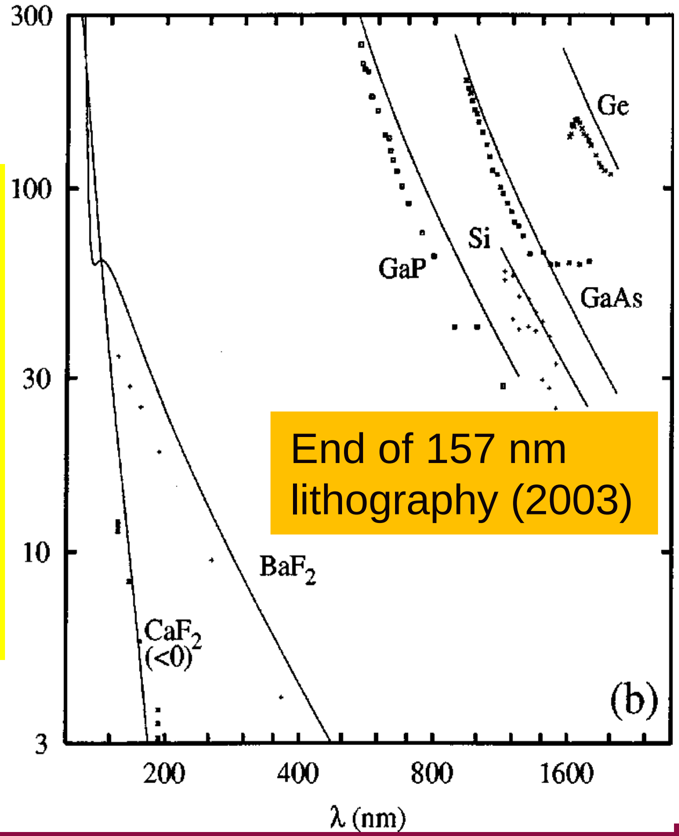
Strong near absorption edge.

$\Delta n = (n_{110} - n_{100}) \times 10^6$



Photon Energy

$\Delta n = (n_{110} - n_{100}) \times 10^7$



End of 157 nm lithography (2003)

Birefringence in GaAs near band gap
Model from k.p theory

Yu & Cardona, SSC **9**, 1421 (1971)

Agranovitch & Ginzburg, Crystal Optics
J.H. Burnett, PRB **64**, 241102 (2000)

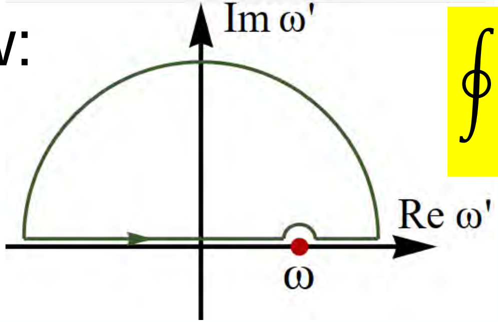
Causality: Charge Movement Follows the Field

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

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

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

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

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

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.

Maxwell's Equations for Continuous Media

$\vec{\nabla}\cdot\vec{D}=\rho=0$	Gauss' Law (Coulomb)
$\vec{\nabla}\cdot\vec{B}=0$	Gauss' Law (magnetic field)
$\vec{\nabla}\times\vec{E}=-\frac{\partial\vec{B}}{\partial t}$	Faraday's Law
$\vec{\nabla}\times\vec{H}=\vec{j}+\frac{\partial\vec{D}}{\partial t}=\frac{\partial\vec{D}}{\partial t}$	Ampere's Law

Anisotropic wave equation:

Take curl on both sides in Ampere's Law and Faraday's Law

$$\Delta\vec{E}-\vec{\nabla}(\vec{\nabla}\cdot\vec{E})=\mu_0\frac{\partial}{\partial t}\vec{\nabla}\times\mu\vec{H}$$

$$\Delta\vec{H}-\vec{\nabla}(\vec{\nabla}\cdot\vec{H})=-\varepsilon_0\frac{\partial}{\partial t}\vec{\nabla}\times\varepsilon\vec{E}$$

The terms in red do not vanish (cannot be simplified) in anisotropic media.

Isotropic wave equation:

$$\Delta\vec{E}=\frac{\epsilon\mu}{c^2}\frac{\partial^2}{\partial t^2}\vec{E}$$

$$v_{\text{phase}}=\frac{c}{\sqrt{\varepsilon\mu}}=\frac{c}{n\sqrt{\mu}}$$

Refractive index n=√ε

Macroscopic Optical Constants

n: **refractive index**, $n=c/v$
k: **extinction coefficient**
n+ik: **complex refractive index**
R: **reflectance** at normal incidence (I_{refl}/I_0)
T: **transmittance** (I_{trans}/I_0)
R+T+A+S=1
α: **absorption coefficient**
$\alpha=4\pi k/\lambda$
ε: **complex dielectric function**
$\varepsilon=\varepsilon_1+i\varepsilon_2=(n+ik)^2$
σ: **complex optical conductivity**
$\sigma=-i\varepsilon_0\omega(\varepsilon-1)$
$\eta=\mathrm{Im}(-1/\varepsilon)$ **loss function**

Why not n–ik?
Wave goes like $\exp[i(kx-\omega t)]$
EE: $j=-i$

Absorbed power per unit volume:

$$\frac{P}{V}=\vec{j}\cdot\vec{E}=\sigma E^2$$

All are connected through Maxwell's equations.

Assume μ=1: Crystal Optics

$\vec{\nabla}\cdot\vec{D}=\rho=0$	Gauss' Law (Coulomb)
$\vec{\nabla}\cdot\vec{B}=0$	Gauss' Law (magnetic field)
$\vec{\nabla}\times\vec{E}=-\frac{\partial\vec{B}}{\partial t}$	Faraday's Law
$\vec{\nabla}\times\vec{H}=\vec{j}+\frac{\partial\vec{D}}{\partial t}=\frac{\partial\vec{D}}{\partial t}$	Ampere's Law

Anisotropic wave equation:

Take curl on both sides in Ampere's Law and Faraday's Law

$$\Delta\vec{E}-\vec{\nabla}(\vec{\nabla}\cdot\vec{E})=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\epsilon\vec{E}$$

$$\Delta\vec{H}=-\varepsilon_0\frac{\partial}{\partial t}\vec{\nabla}\times\varepsilon\vec{E}$$

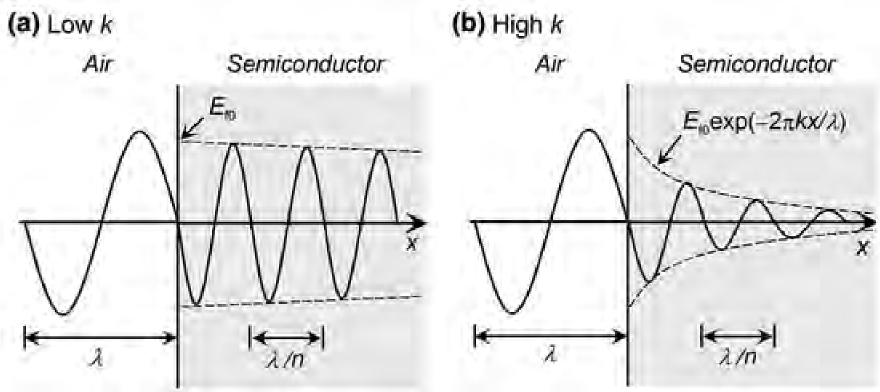
For μ=1 we get a single wave equation for ***E***, from which ***H*** can be calculated as well. ***E*** and ***H*** are decoupled.

Use Berreman / Yeh 4x4 matrix formalism for (***E***,***H***).

Agranovitch & Ginzburg, Crystal Optics

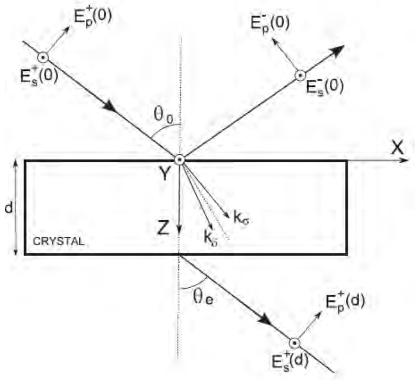
Inhomogeneous Plane Waves

Plane waves do not solve Maxwell's equations, if Im(ε)≠0.



The amplitude of the plane wave decays in the medium due to absorption.

Snell: $$\frac{\sin\theta_1}{\sin\theta_2} = \frac{n_1}{n_2}$$



Inhomogeneous plane wave (aka generalized plane waves):

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

Allow complex wave vector: $\vec{k} = \vec{k}_1 + i\vec{k}_2 = k_1\vec{u} + ik_2\vec{v}$

$$\vec{E}(\vec{r},t) = \vec{E}_0 \exp\left[-\vec{k}_2\cdot\vec{r}\right] \exp\left[i\left(\vec{k}_1\cdot\vec{r} - \omega t\right)\right]$$

Attenuation

Propagation

Mansuripur, *Magneto-Optical Recording*, 1995
Stratton, *Electromagnetic Theory*, 1941/2007
Orteaga, TSF 571, 701 (2014).

Maxwell's Equations in Continuous Media

$\vec{\nabla} \cdot \vec{D} = 0$ Gauss' Law (Coulomb)

$\vec{\nabla} \cdot \vec{B} = 0$ Gauss' Law (magnetic field)

$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$ Faraday's Law

$\vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$ Ampere's Law

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

etc. for other fields

Inhomogeneous plane waves with complex wave vectors

$\vec{k} \cdot \vec{D}_0 = 0$ Gauss' Law (Coulomb)

$\vec{k} \cdot \vec{B}_0 = 0$ Gauss' Law (magnetic field)

$\vec{k} \times \vec{E}_0 = \omega \vec{B}_0$ Faraday's Law

$\vec{k} \times \vec{H}_0 = -\omega \vec{D}_0$ Ampere's Law

Stratton, *Electromagnetic Theory*, 1941/2007

Anisotropic Wave Equations in Continuous Media

$\vec{k}\cdot\vec{D}_0 = 0$	Gauss' Law (Coulomb)
$\vec{k}\cdot\vec{B}_0 = 0$	Gauss' Law (magnetic field)
$\vec{k}\times\vec{E}_0 = \omega\vec{B}_0$	Faraday's Law
$\vec{k}\times\vec{H}_0 = -\omega\vec{D}_0$	Ampere's Law

$$\vec{D}_0(\vec{k},\omega) = \varepsilon_0\varepsilon(\vec{k},\omega)\vec{E}_0(\vec{k},\omega)$$
$$\vec{B}_0(\vec{k},\omega) = \mu_0\mu(\vec{k},\omega)\vec{H}_0(\vec{k},\omega)$$

Constitutive Relations

Anisotropic wave equation:

$$\left|\vec{k}\right|^2\vec{E}_0 - (\vec{k}\cdot\vec{E}_0)\vec{k} = -\mu_0\omega\vec{k}\times\mu\vec{H}_0$$
$$\left|\vec{k}\right|^2\vec{H}_0 - (\vec{k}\cdot\vec{H}_0)\vec{k} = -\varepsilon_0\omega\vec{k}\times\varepsilon\vec{E}_0$$

D and **B** are transverse, but **E** and **H** are not.

Isotropic wave equation:

$$\left|\vec{k}\right|^2 = \varepsilon\mu\frac{\omega^2}{c^2}$$

$$v_{\text{phase}} = \frac{c}{\sqrt{\varepsilon\mu}} = \frac{c}{n\sqrt{\mu}}$$

Refractive index n=√ε

Assume μ=1: Crystal Optics

$\vec{k}\cdot\vec{D}_0=0$	Gauss' Law (Coulomb)
$\vec{k}\cdot\vec{B}_0=0$	Gauss' Law (magnetic field)
$\vec{k}\times\vec{E}_0=\omega\vec{B}_0$	Faraday's Law
$\vec{k}\times\vec{H}_0=-\omega\vec{D}_0$	Ampere's Law

$$\vec{D}_0(\vec{k},\omega)=\varepsilon_0\varepsilon(\vec{k},\omega)\vec{E}_0(\vec{k},\omega)$$
$$\vec{B}_0(\vec{k},\omega)=\mu_0\mu(\vec{k},\omega)\vec{H}_0(\vec{k},\omega)$$

For μ=1: Algebraic equation for ***E***, from which ***H*** can be calculated.

Anisotropic wave equation:

$$\left|\vec{k}\right|^2\vec{E}_0-\left(\vec{k}\cdot\vec{E}_0\right)\vec{k}=\frac{\omega^2}{c^2}\varepsilon\vec{E}_0$$
$$\left|\vec{k}\right|^2\vec{H}_0=-\varepsilon_0\omega\vec{k}\times\varepsilon\vec{E}_0$$

Berreman & Yeh 4x4 transfer matrix for (***E***,***H***).

Isotropic wave equation:

$$\left|\vec{k}\right|^2=\varepsilon\mu\frac{\omega^2}{c^2}$$

$$v_{\text{phase}}=\frac{c}{\sqrt{\varepsilon\mu}}=\frac{c}{n\sqrt{\mu}}$$

Refractive index n=√ε

Agranovitch & Ginzburg, Crystal Optics

Berreman Transfer Matrix Formalism

- Tompkins & Hilfiker: *Spectroscopic Ellipsometry*. Very simple, only isotropic layers.
- G.E. Jellison, *Data Analysis for Spectroscopic Ellipsmetry*, in Tompkins & Irene: *Handbook of Ellipsometry*. More formal treatment, but mostly focused on isotropic layers.
- **M. Schubert, *Theory and Application of Generalized Ellipsometry*, in Tompkins & Irene: *Handbook of Ellipsometry*. This is the theory behind the CompleteEase and WVASE32 software.**
- M. Born and E. Wolf, *Principles of Optics* (Cambridge University Press, Cambridge, 1980).
- P. Yeh, *Optical Waves in Layered Media* (Wiley, NY, 1988).
- R.M.A. Azzam and N.M. Bashara, *Ellipsometry and Polarized Light* (North-Holland, Amsterdam, 1977).
- M. Mansuripur, *The Physics of Magneto-Optical Recording* (Cambridge University Press, Cambridge, 1995).

- Original references:
- F. Abeles, Ann. de Physique **5**, 596 (1950).
- **D.W. Berreman, *Optics in stratified and anisotropic media: 4x4 matrix formulation*, JOSA 62, 502 (1972).**
- P. Yeh, *Optics of anisotropic layered media: A new 4x4 matrix algebra*, Surf. Sci., 96, 41 (1980).
- H. Wöhler, G. Hass, M. Fritsch, and D. A. Mlynski, *Faster 4x4 matrix method for uniaxial inhomogeneous media*, JOSA A **5**, 1554 (1988). Also JOSA A **8**, 536 (1991).
- M. Schubert, *Polarization-dependent optical parameters of arbitrarily anisotropic homogeneous layered systems*, Phys. Rev. B **53**, 4265 (1996).

Longitudinal Solutions to Maxwell's Equations (μ=1)

$\vec{k}\cdot\varepsilon\vec{E}_0 = 0$	Gauss' Law (Coulomb)
$\vec{k}\cdot\vec{B}_0 = 0$	Gauss' Law (magnetic field)
$\vec{k}\times\vec{E}_0 = \omega\vec{B}_0$	Faraday's Law
$\vec{k}\times\vec{H}_0 = -\omega\varepsilon\vec{E}_0$	Ampere's Law

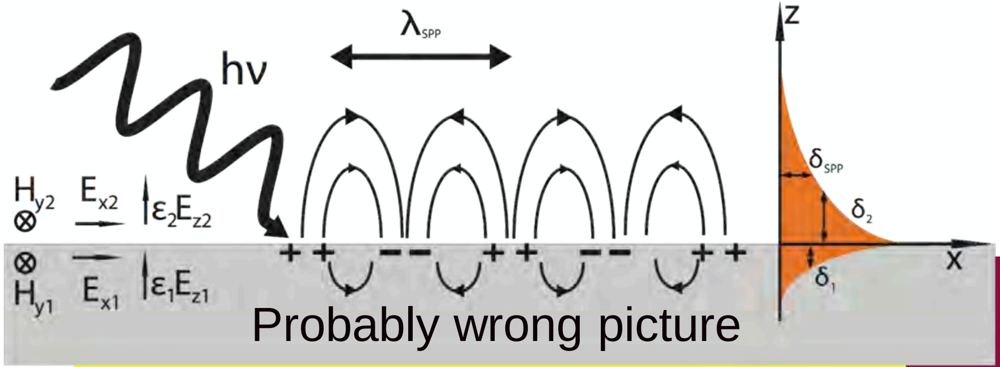
Transverse solution: **D** is transverse

□ 0 and $\left|\vec{k}\right|^2\vec{E}_0 - \left(\vec{k}\cdot\vec{E}_0\right)\vec{k} = \frac{\omega^2}{c^2}\varepsilon\vec{E}_0$ and $\left|\vec{k}\right|^2\vec{H}_0 = -\varepsilon_0\omega\vec{k}\times\varepsilon\vec{E}_0$

Longitudinal solution:

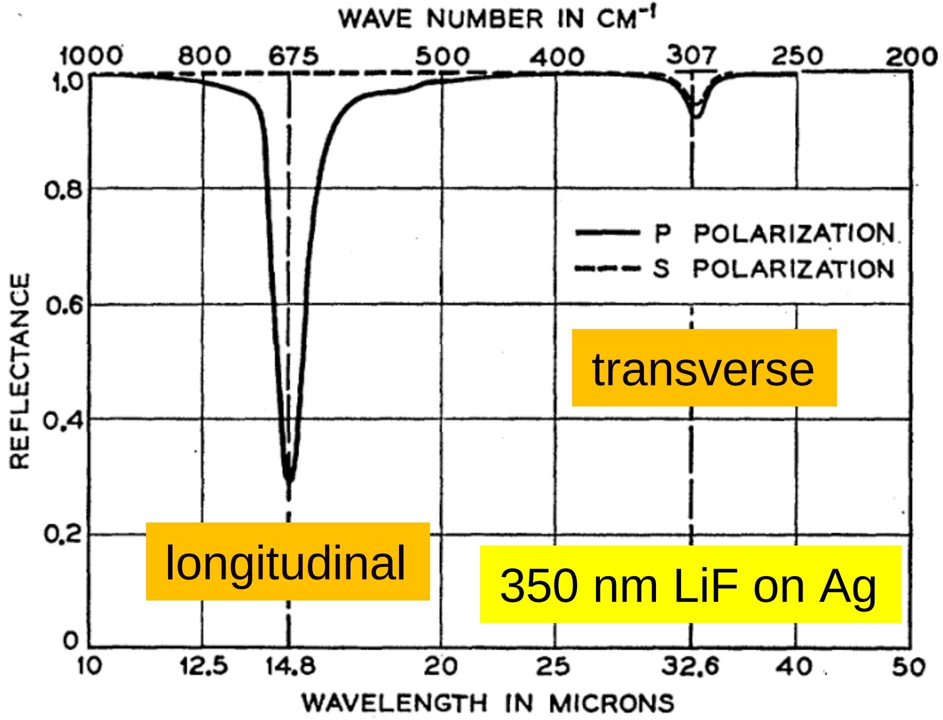
□ 0 and $\vec{E}_0 \parallel \vec{k}$ and $\vec{H}_0 = 0$

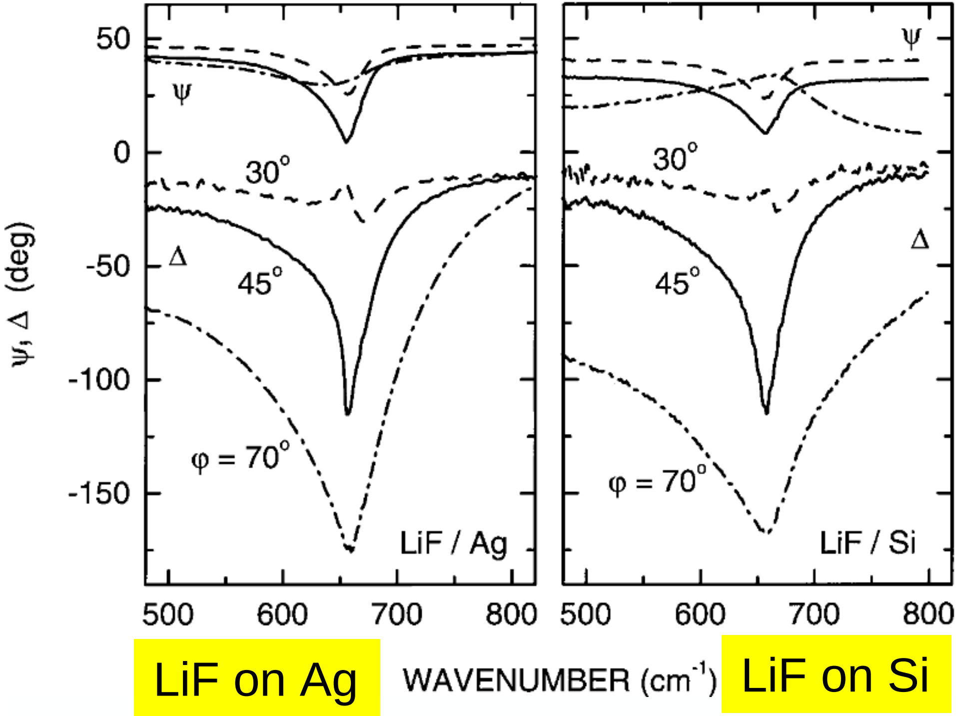
Longitudinal solutions are also called **plasmons.**



Agranovitch & Ginzburg, Crystal Optics

Berreman Modes: Insulator (LiF) on Metal (Ag)





Humlicek: The Berreman peak is an interference effect, which occurs when $\varepsilon_{film}=0$. It is not a longitudinal mode.

D.W. Berreman, Phys. Rev. **130**, 2193 (1963)

J. Humlicek, phys. stat. sol. (b) **215**, 155 (1999)

Energy density, Poynting Vector

Energy density:

$$u = \frac{1}{2}\left(\vec{E}\cdot\vec{D} + \vec{H}\cdot\vec{B}\right) = \frac{1}{2}\left(\vec{E}\cdot\varepsilon_0\epsilon\vec{E} + \vec{H}\cdot\mu_0\mu\vec{H}\right)$$

$$\frac{\partial^2 u}{\partial E_i \partial E_j} = \frac{\varepsilon_0}{2}\varepsilon_{ij}$$

Implies ε_{ij} symmetric tensor (B=0).

Onsager relation

in isotropic medium: $u = \frac{\epsilon\epsilon_0}{2}\left|\vec{E}\right|^2$

Poynting's theorem (energy flow): $\frac{\partial u}{\partial t} = -\vec{\nabla}\cdot\vec{S} - \vec{j}\cdot\vec{E}$

EM wave has no Ohmic power **j•E**

$$\frac{\partial u}{\partial t} = -\vec{\nabla}\cdot\vec{S} = -\vec{\nabla}\cdot\frac{1}{\mu_0}\left(\vec{E}\times\vec{B}\right) = \frac{1}{\mu_0}\left(\vec{B}\cdot\vec{\nabla}\times\vec{E} - \vec{E}\cdot\vec{\nabla}\times\vec{B}\right)$$

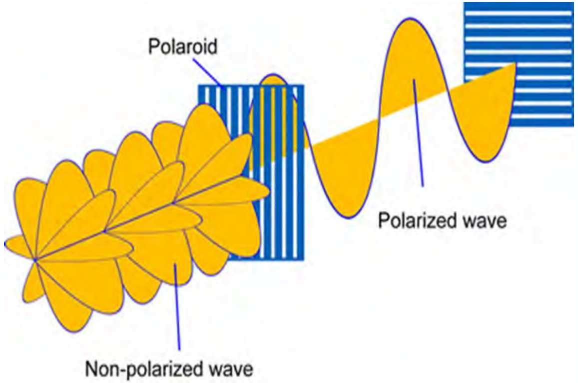
$$\frac{P}{V} = \vec{j}\cdot\vec{E} = \sigma E^2$$

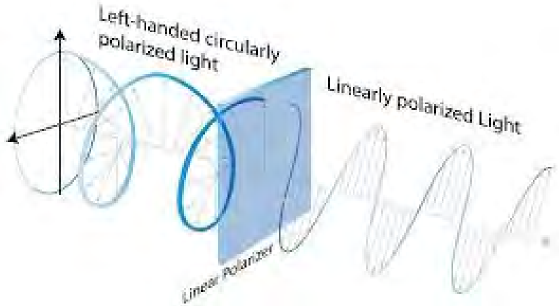
Longitudinal modes carry no energy.

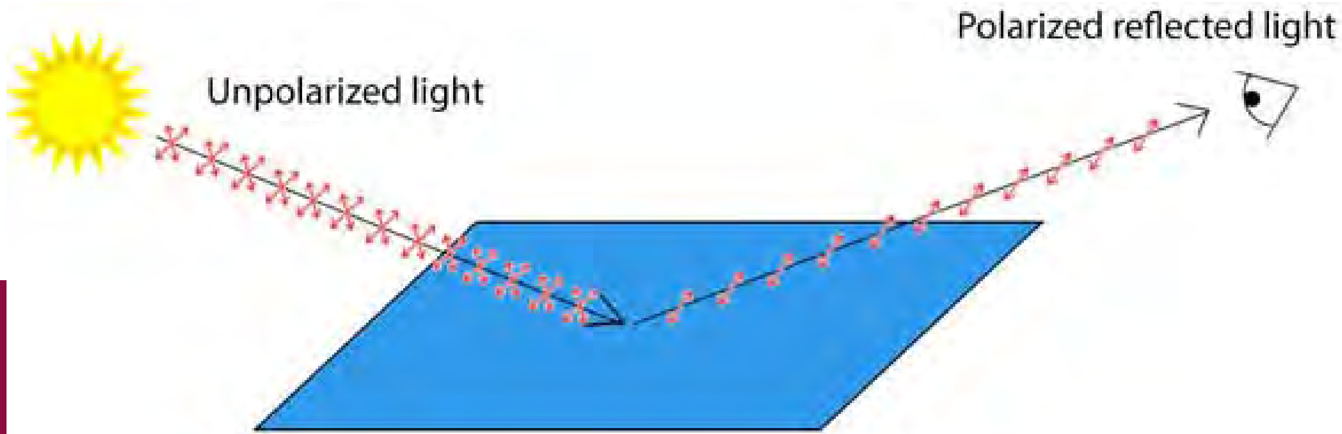
Agranovitch & Ginzburg, Crystal Optics

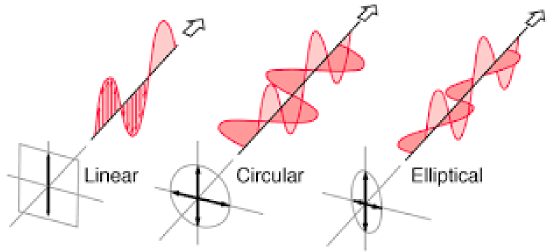
Summary

- Fourier series and Fourier transforms: Plane waves
- Maxwell's Equations in vacuum (general and plane-wave format)
- Polarized light
  - Jones and Stokes vectors
  - Polarization ellipse, Poincare sphere
- Ellipsometry experiments
  - Jones and Mueller matrices
- Electrodynamics of continuous media
- Crystal optics: Maxwell's equations in crystals









Thank you!

Questions?