Optical Properties of Solids: Lecture 9

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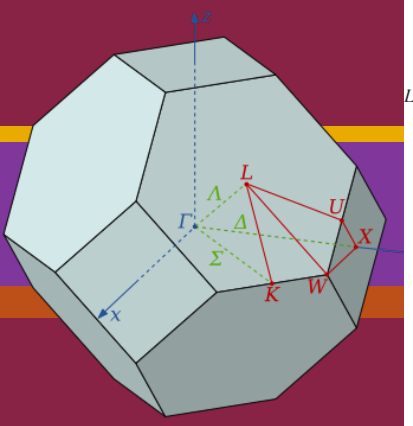
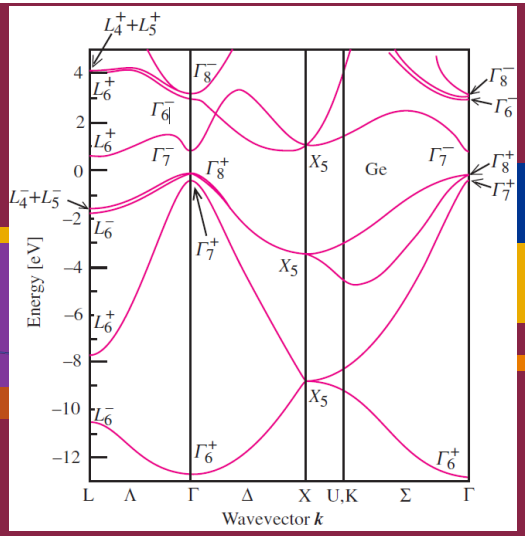
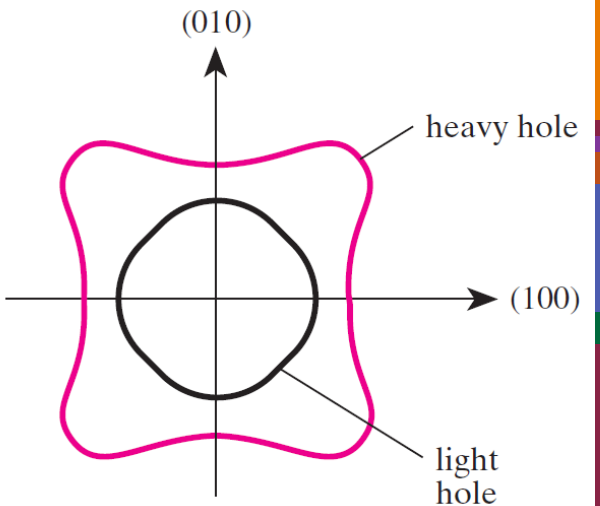
Optical Properties of Solids: Lecture 7+8+9

Electronic Band Structure
Direct and indirect band gaps
Empty lattice, pseudopotential, k.p band structures
Optical interband transitions, Fermi's Golden Rule
Absorption coefficient for direct and **indirect gaps**
Tauc plot
Van Hove singularities







References: Band Structure and Optical Properties

Solid-State Theory and Semiconductor Band Structures:

- Mark Fox, Optical Properties of Solids
- Ashcroft and Mermin, Solid-State Physics
- Yu and Cardona, Fundamentals of Semiconductors
- Dresselhaus/Dresselhaus/Cronin/Gomes, Solid State Properties
- Cohen and Chelikowsky, Electronic Structure and Optical Properties
- Klingshirn, Semiconductor Optics
- Grundmann, Physics of Semiconductors
- Ioffe Institute web site: NSM Archive
  http://www.ioffe.ru/SVA/NSM/Semicond/index.html

Outline

Band structure and optical interband transitions

Einstein coefficients, population inversion, optical gain, lasers

Fermi's Golden Rule

Joint density of states, optical mass

Direct gap absorption in InAs, PbS, and InSb; Tauc plot

Indirect gap absorption in Si and Ge

Experimental techniques to measure absorption

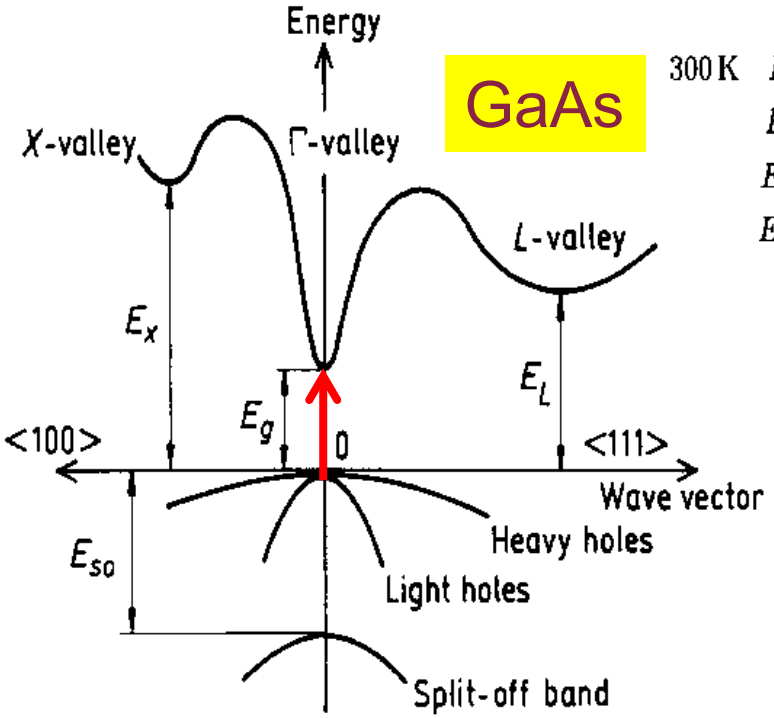
Van Hove singularities

Critical points in the dielectric function

Analytical lineshapes to fit Savitzky-Golay derivative

Parametric oscillator model

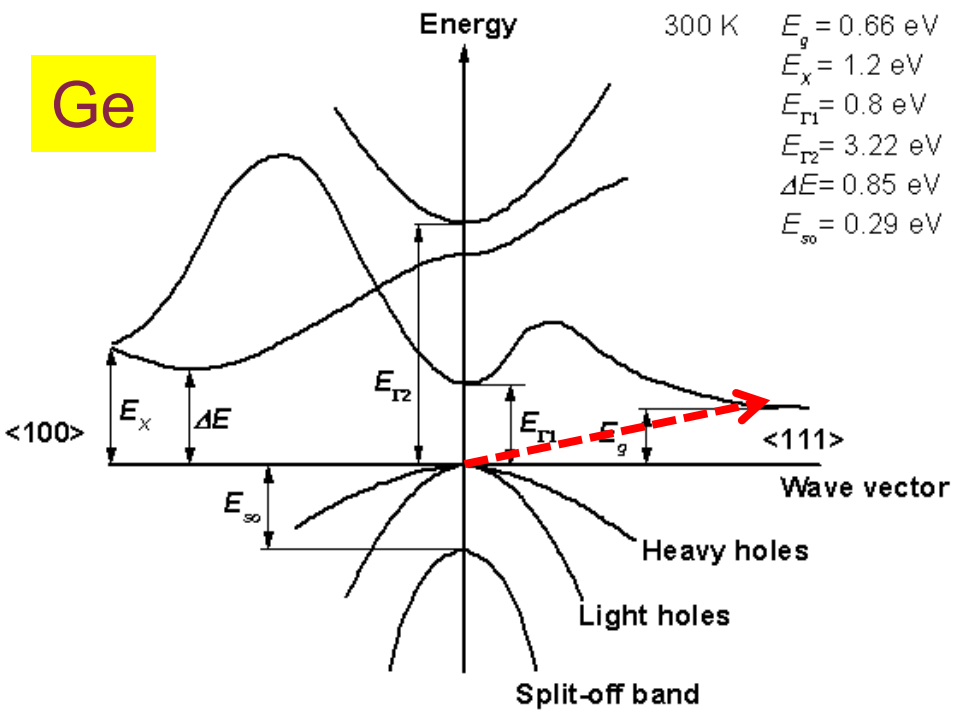
Semiconductor Band Structures



GaAs

300 K $E_g = 1.42$ eV
$E_L = 1.71$ eV
$E_X = 1.90$ eV
$E_{so} = 0.34$ eV

Δk=0



Ge

300 K $E_g = 0.66$ eV
$E_X = 1.2$ eV
$E_{Γ1} = 0.8$ eV
$E_{Γ2} = 3.22$ eV
$\Delta E = 0.85$ eV
$E_{so} = 0.29$ eV

Δk≠0

Direct transition:

Initial and final electron state have **same** wave vector.

Indirect transition:

Initial and final electron state have **different** wave vector.

http://www.ioffe.ru/SVA/NSM/Semicond/index.html

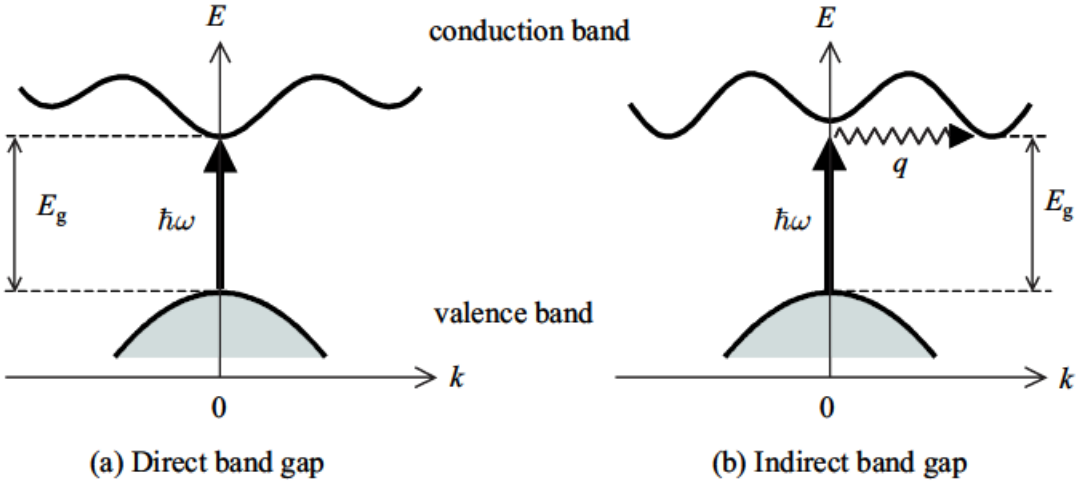
Indirect Interband Transitions

Indirect transitions require phonon absorption or emission to conserve **crystal momentum k**.

Consider *Umklapp* processes (±**G RLV**).

Also possible: **Impurity**-assisted or **alloy** scattering

Also must conserve energy.

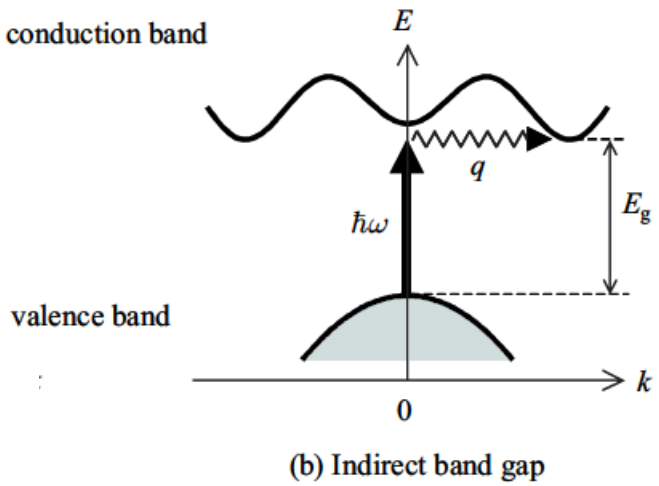


$$\vec{k}_f = \vec{k}_i \pm \vec{Q} \pm \vec{G}$$

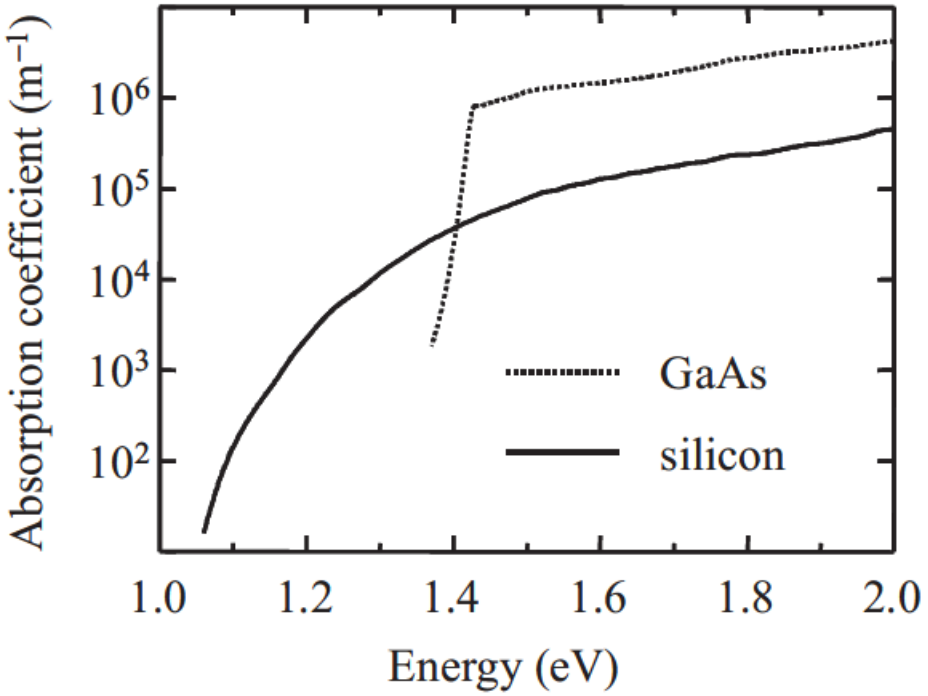
$$\vec{k}_f = \vec{k}_i$$

Fox, Chapter 3

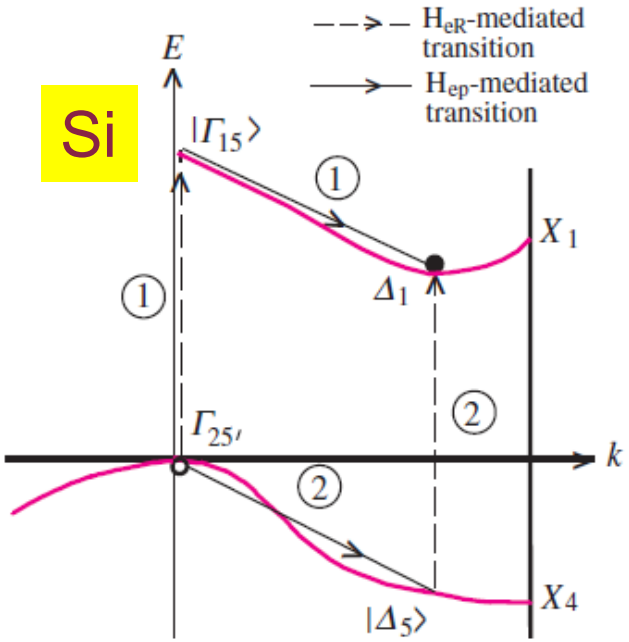
Indirect transitions in Si



(b) Indirect band gap



Si



$$E_f = E_i + \hbar\omega \pm \hbar\Omega$$

energy

$$\hbar\vec{k}_f = \hbar\vec{k}_i \pm \hbar\vec{Q} \pm RLV$$

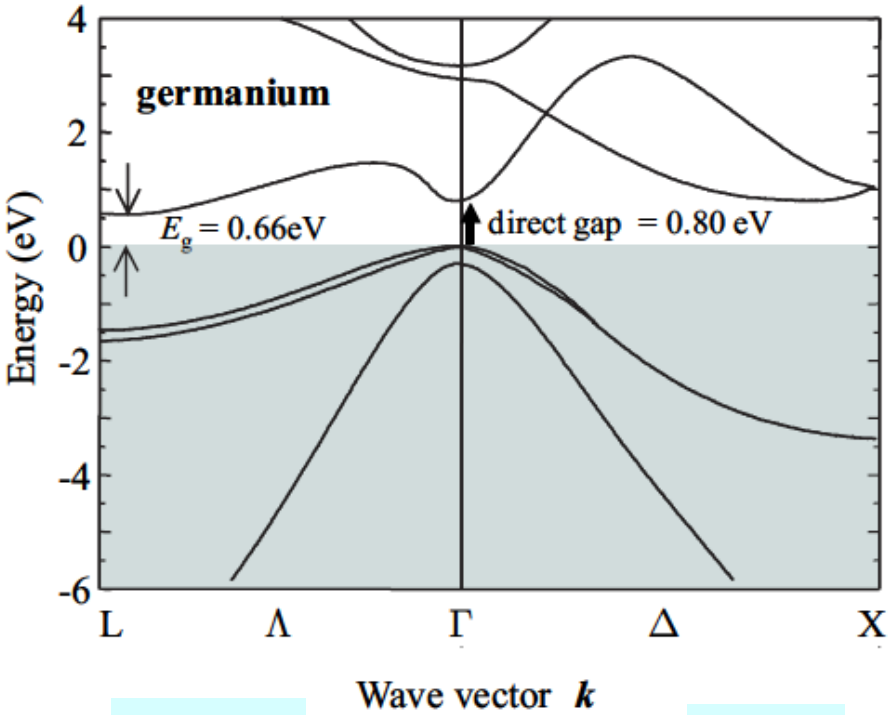
crystal momentum

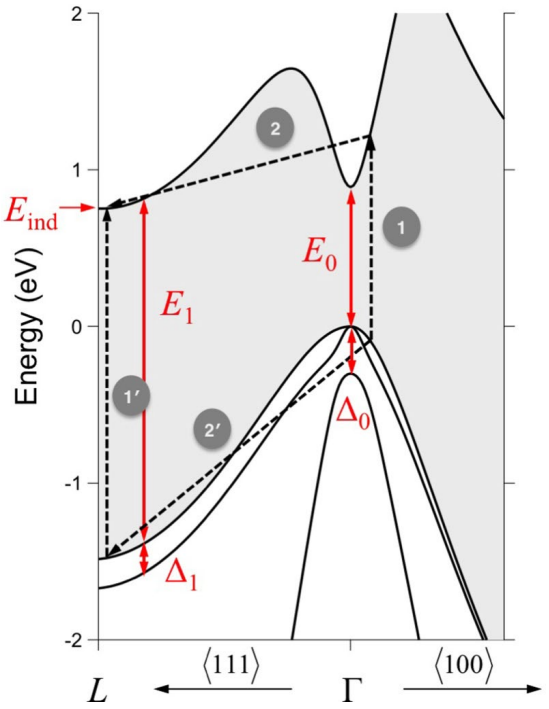
$$\alpha(\hbar\omega) \propto \left(\hbar\omega - E_g \mp \hbar\Omega\right)^2$$

Tauc plot: $\alpha^{1/2}$ increases linearly with energy

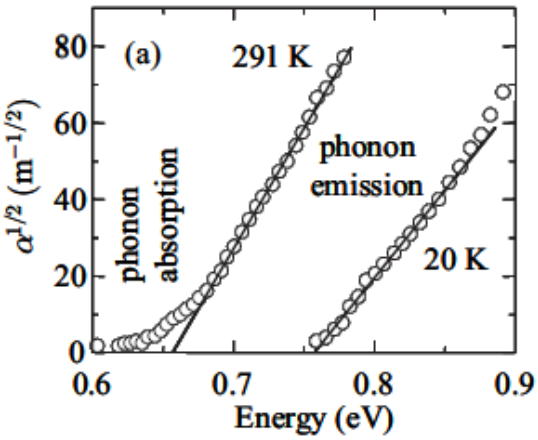
Fox, Chapter 3
Yu & Cardona

Indirect transitions in Germanium

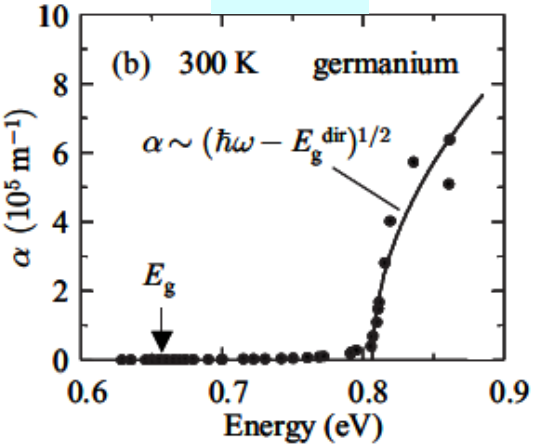




indirect



direct



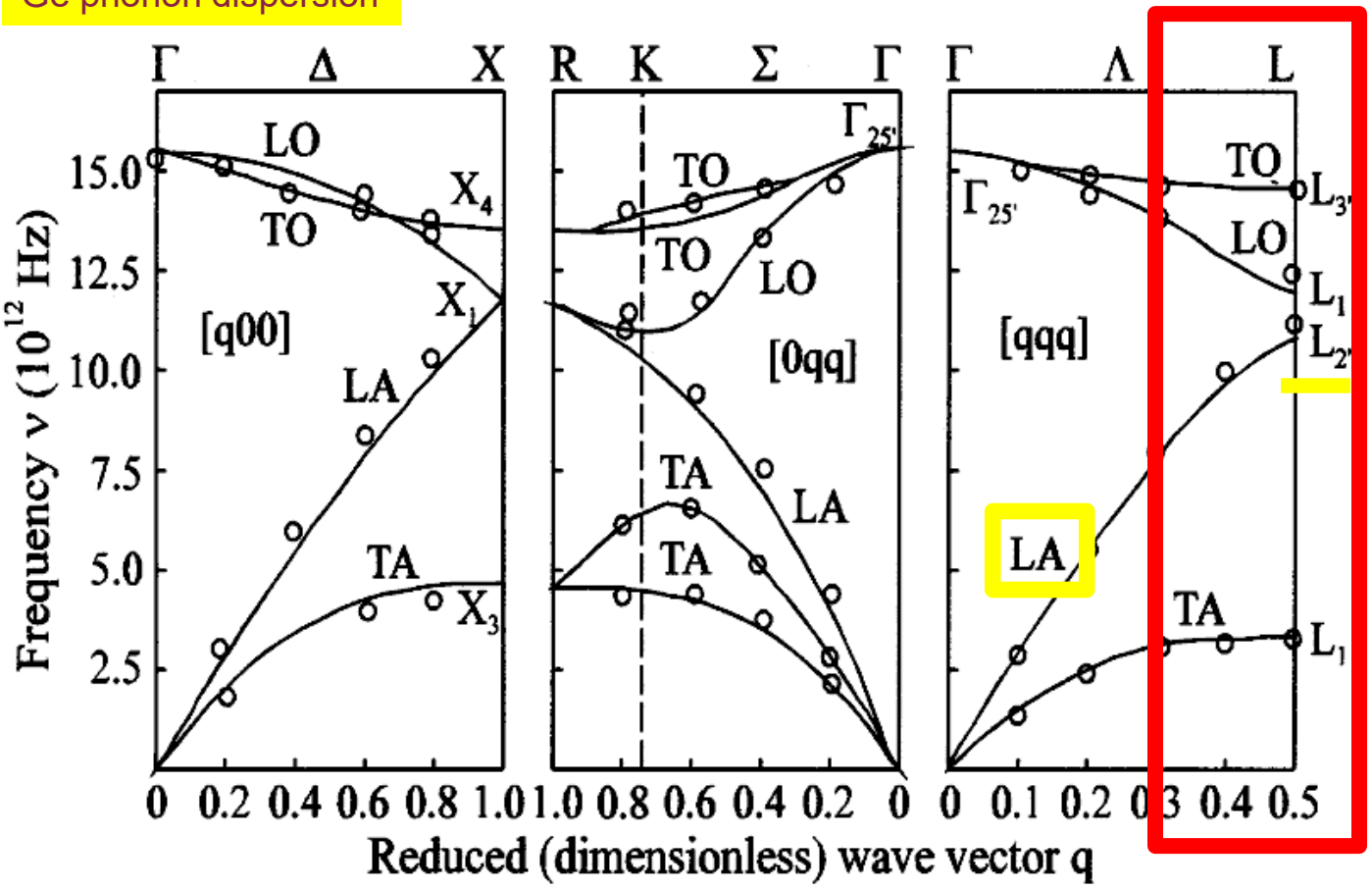
$$\hbar\vec{k}_f = \hbar\vec{k}_i \pm \hbar\vec{Q} \pm RLV$$

$$E_f = E_i + \hbar\omega \pm \hbar\Omega$$

Fox, Chapter 3
Yu & Cardona

Which phonons assist with indirect transitions?

Ge phonon dispersion



Several branches at each k-point:

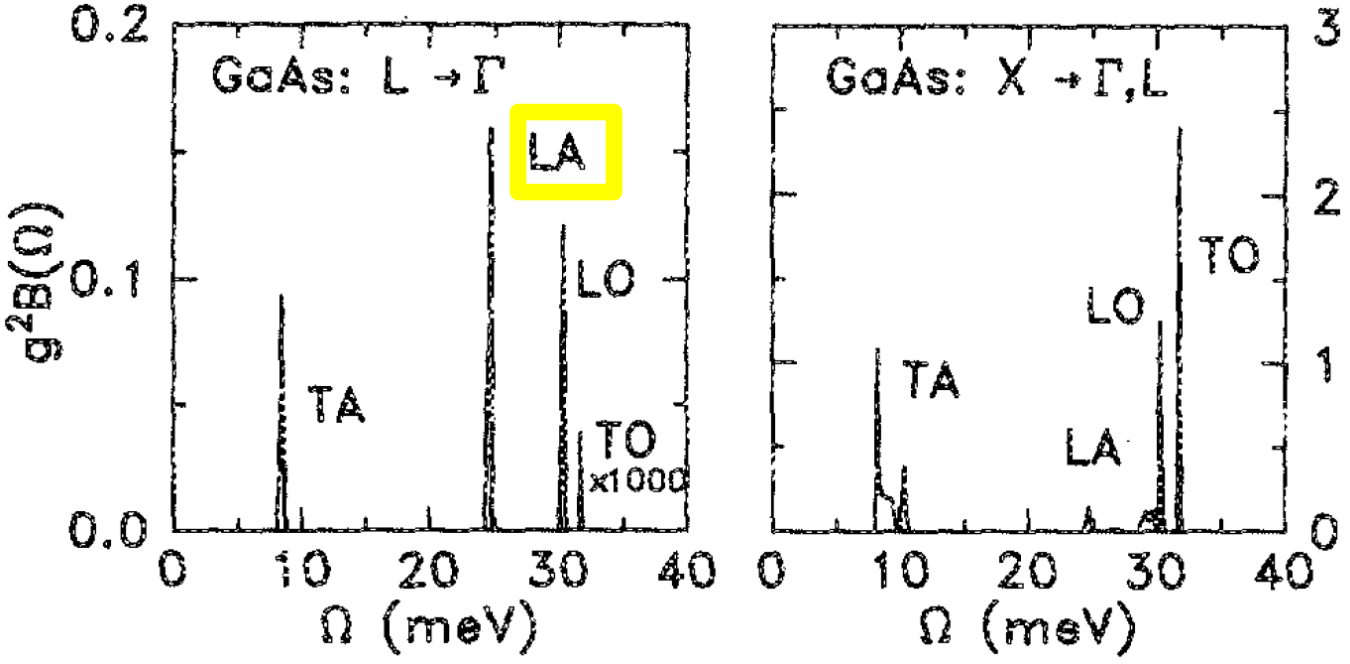
Transverse Optical
Longitudinal Optical
Longitudinal Acoustic
Transverse Acoustic

We know that **q**=L, because the CB minimum is at the L-point.

$\Gamma_{2'} \otimes L_1 = L_{2'}$

J. Birman, PR **127**, 1093 (1962)

Which phonons assist with indirect transitions?



Several branches at each k-point:

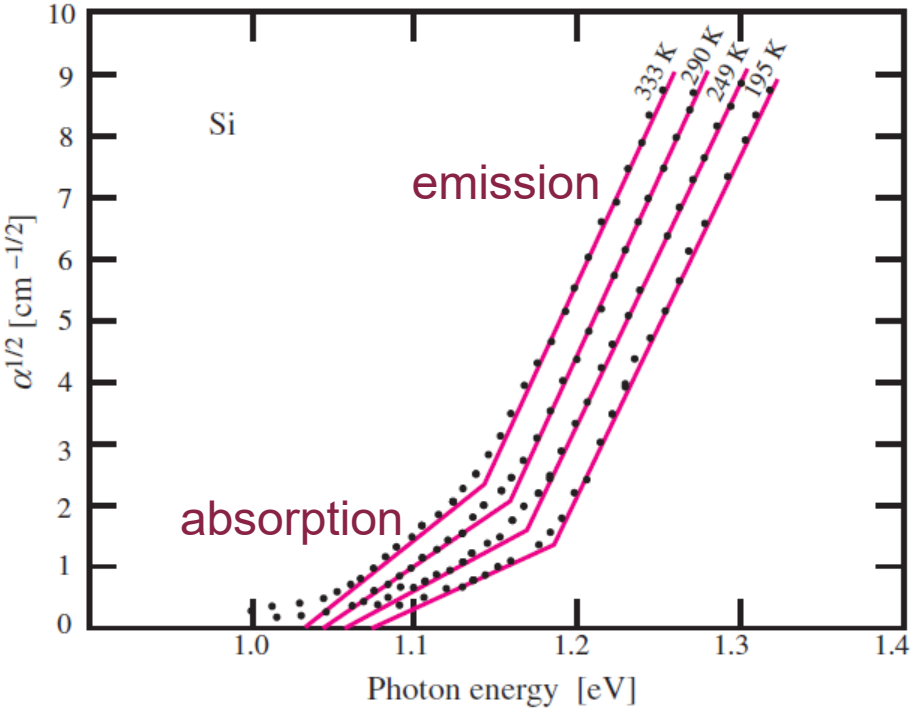
Transverse Optical
Longitudinal Optical
Longitudinal Acoustic
Transverse Acoustic

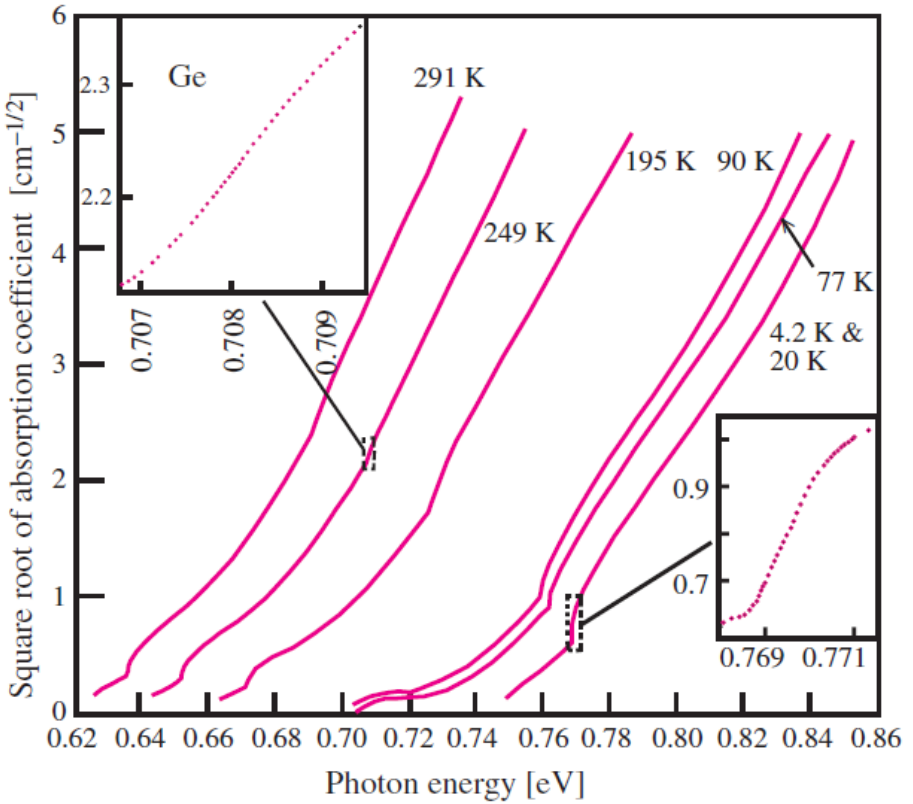
Quantitative description with intervalley spectral functions.

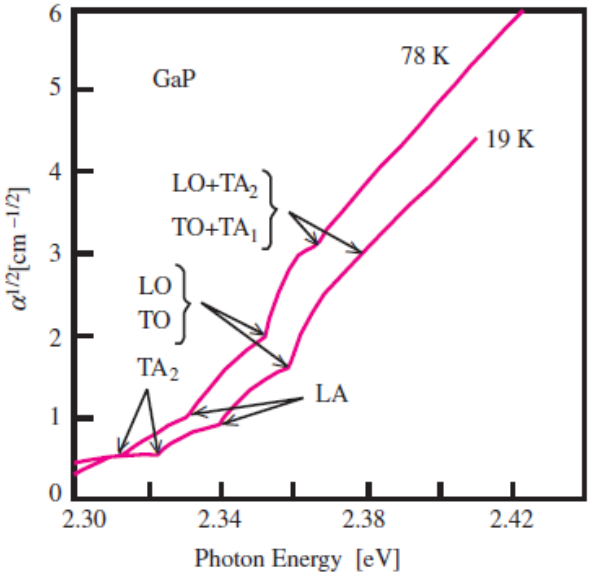
LA-assisted scattering is dominant in Ge for electrons, but we need LO phonons for holes. This ignores the k-dependence of the intervalley scattering matrix element.

Zollner, JAP **68**, 1682 (1990)
Krishnamurthy, JAP **74,** 2117 (1993)

Indirect transitions: Phonon replica





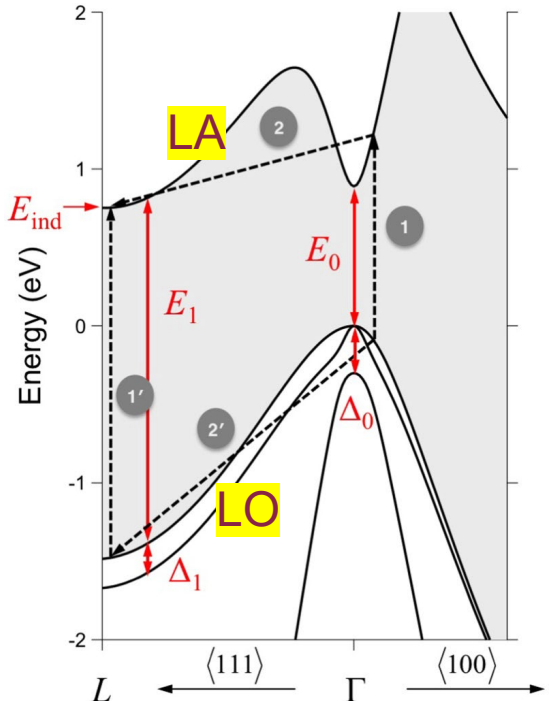


Phonon absorption and emission.
Different phonons can contribute.
Excitonic corrections may be needed.

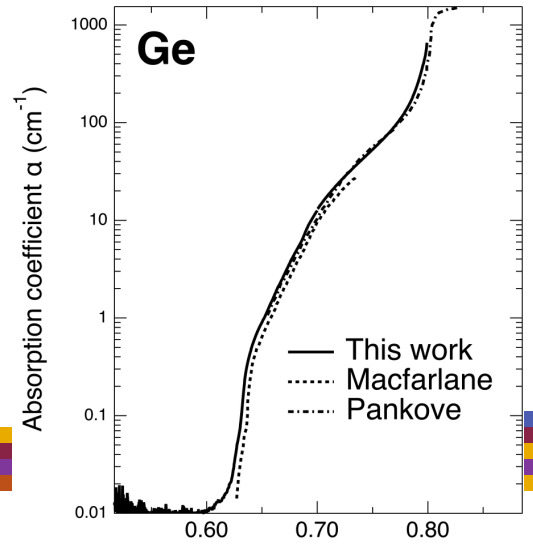
$$\hbar\vec{k}_f = \hbar\vec{k}_i \pm \hbar\vec{Q} + RLV$$

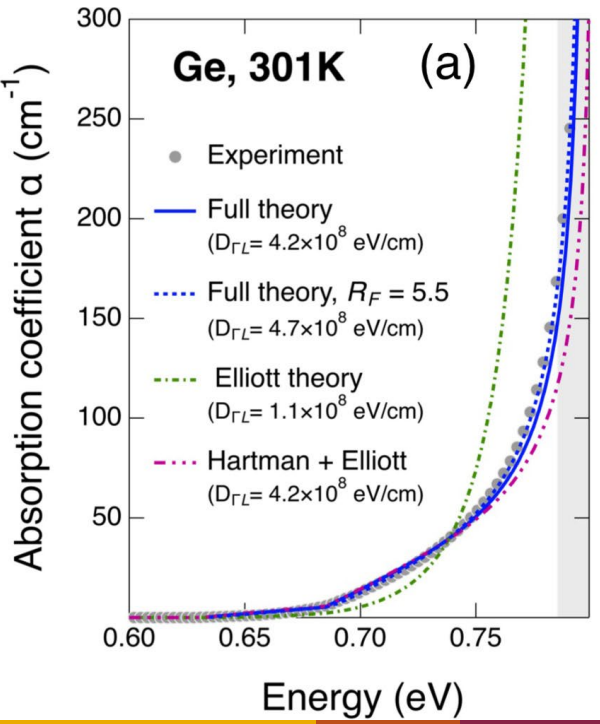
Yu & Cardona
Nishino, JPSJ **37** (1974): wavelength modulation

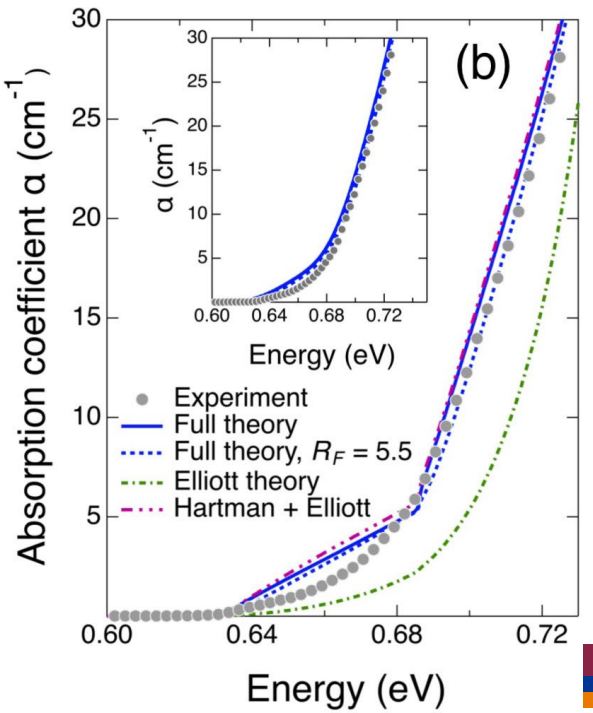
Indirect transitions in Germanium



New experiments (α of Ge at 301 K)
New theory: all details of band structure, excitonic corrections
Only fit parameter: electron-phonon coupling parameter $D_{\Gamma L}$=4.2 eV/Å.

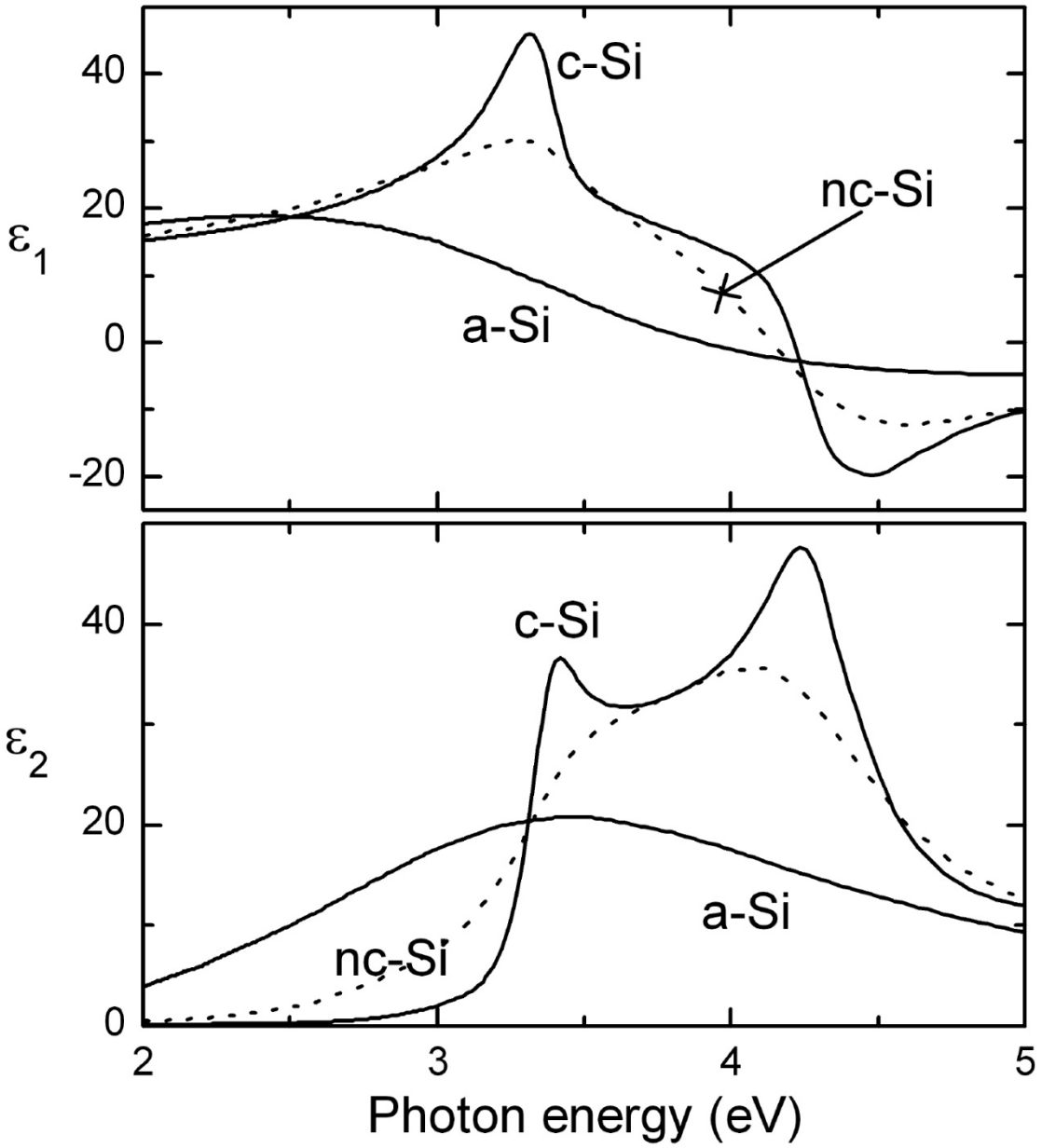






Menendez, PRB (2018); Hartman, PR (1962); Elliot, PR (1957)

Indirect transitions: Weak or Strong ???



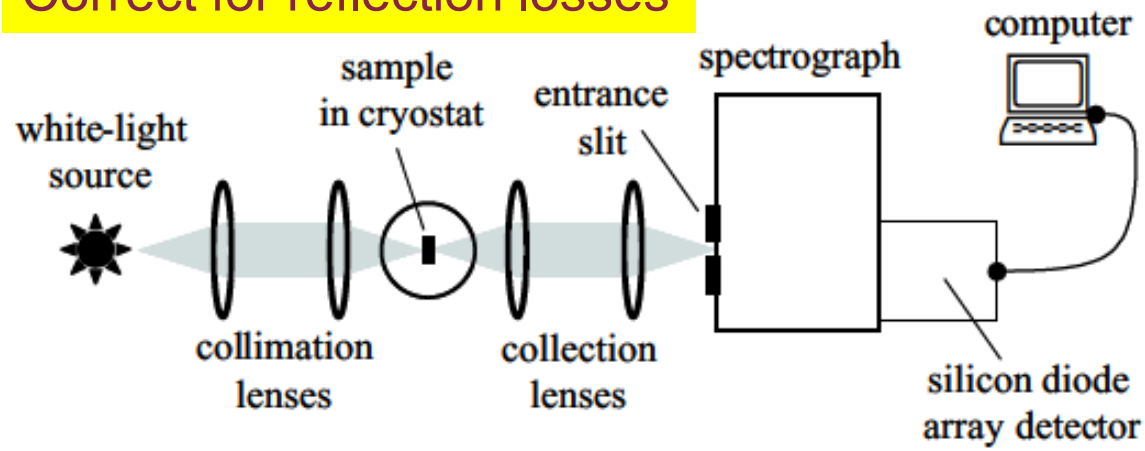
c-Si: All absorption below 3.4 eV is indirect.

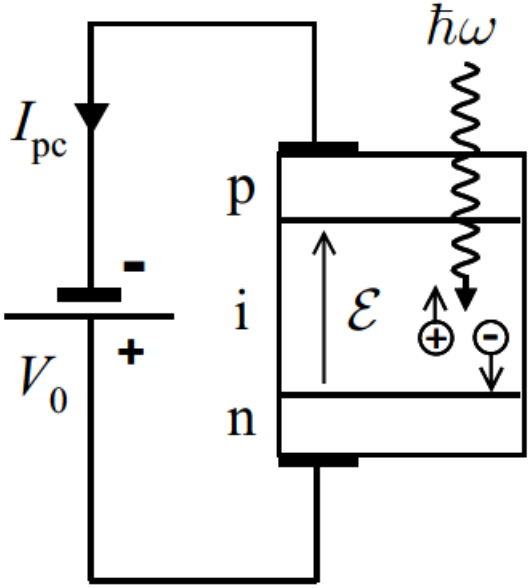
a-Si, nc-Si: Loss of periodicity increases strength of indirect absorption.

Peter Petrik, 2012

Absorption measurements

Correct for reflection losses

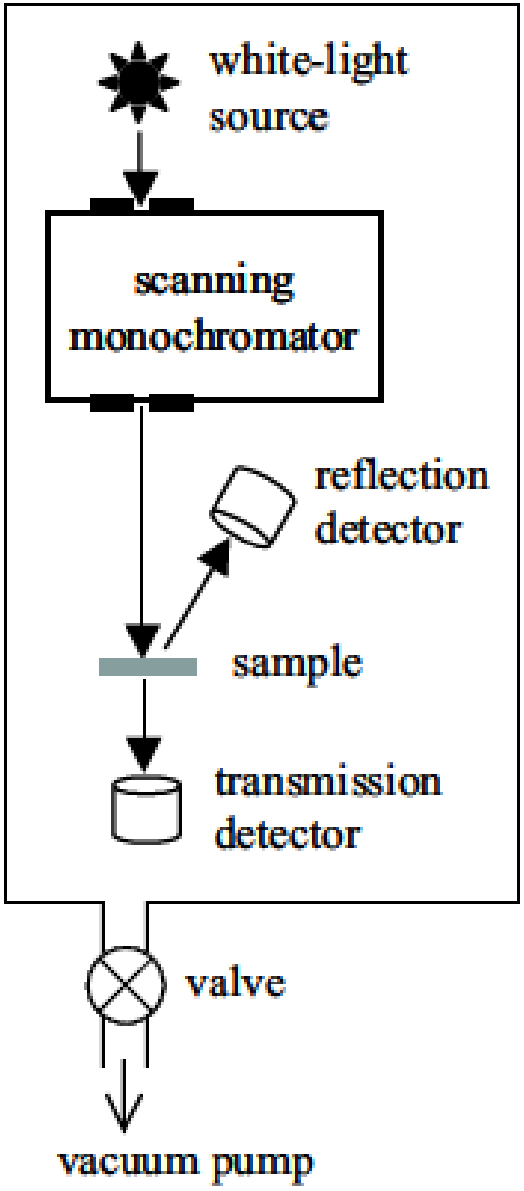




Best for small α

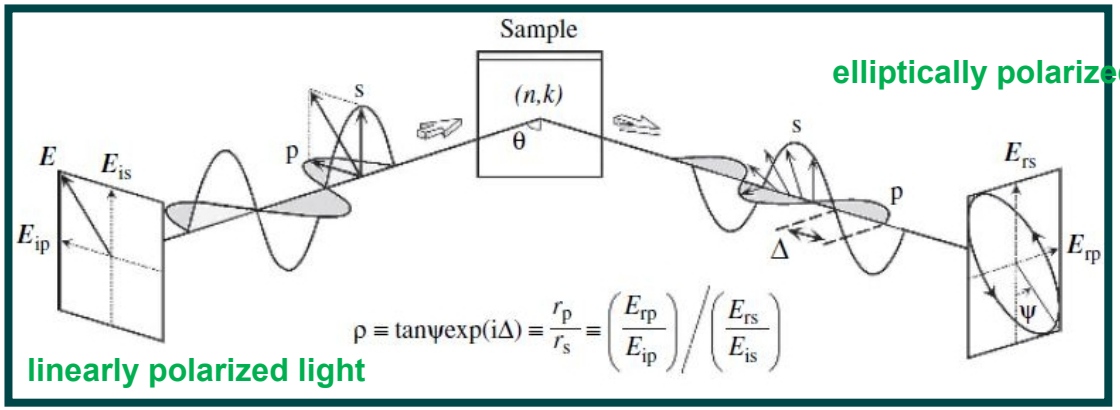
$$I_{PC} = e\eta \frac{P}{\hbar\omega}\left(1 - e^{-\alpha l}\right)$$

Photocurrent



Fox, Chapter 3

Absorption measurements: Ellipsometry



Best for large α

Fresnel reflectance ratio, Ellipsometric angles

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

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

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

Complex dielectric function

Monochromator or Interferometer (λ)

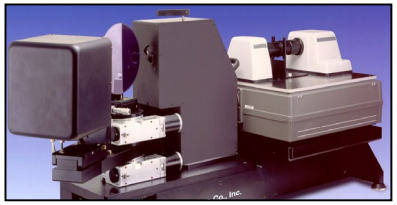
detector

polarizer

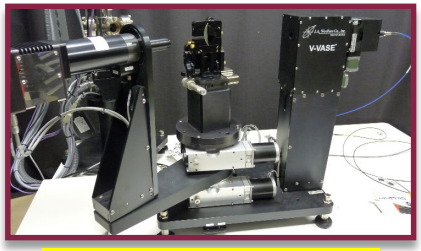
analyzer

Φ

Bulk sample

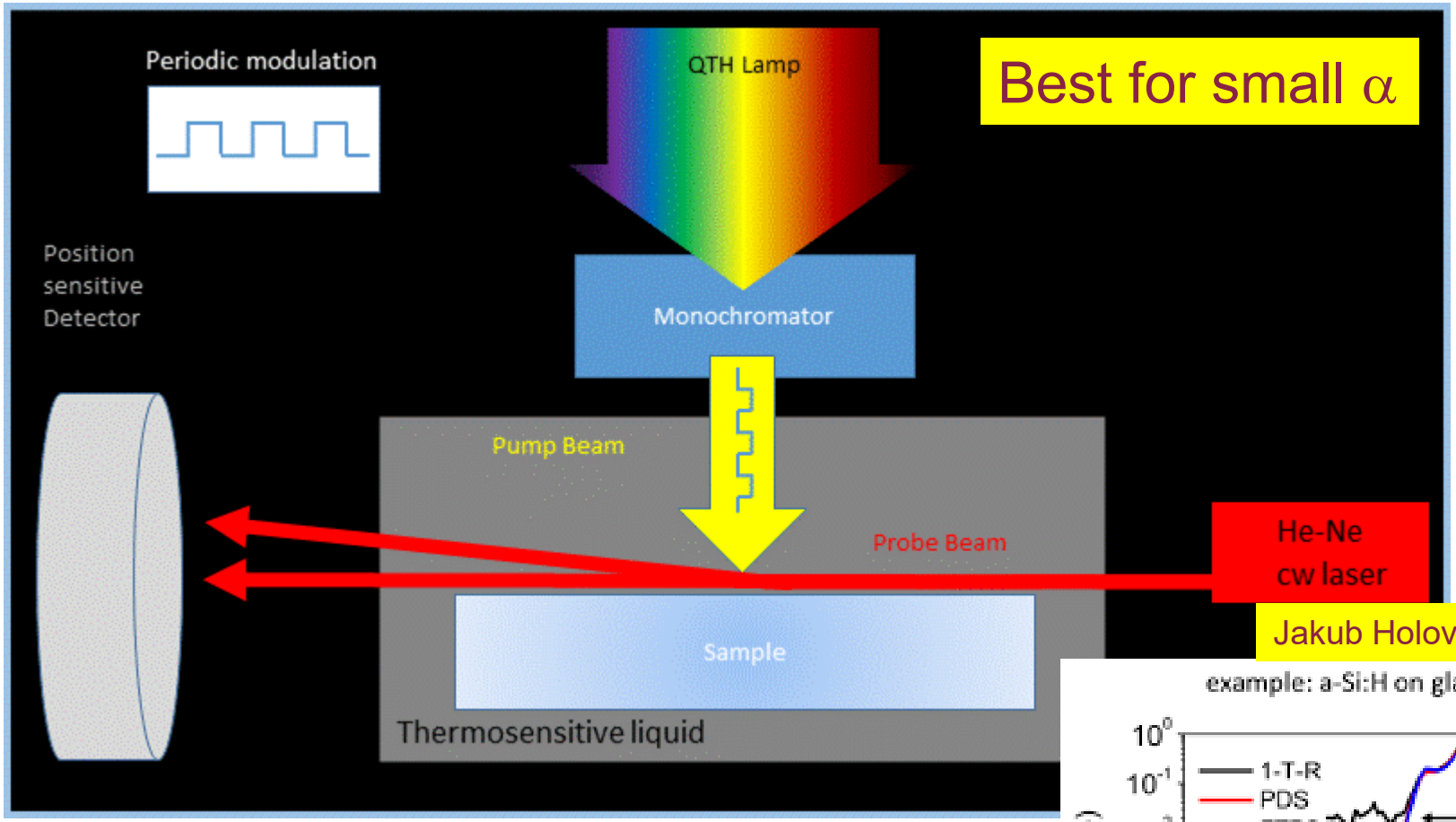


FTIR

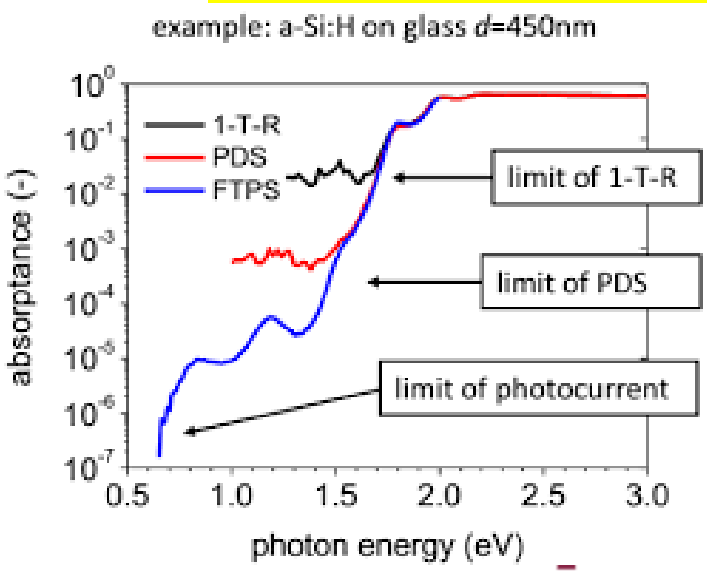


NIR-VIS-QUV

Absorption measurements: Photothermal deflection



Jakub Holovsky, CTU Praha

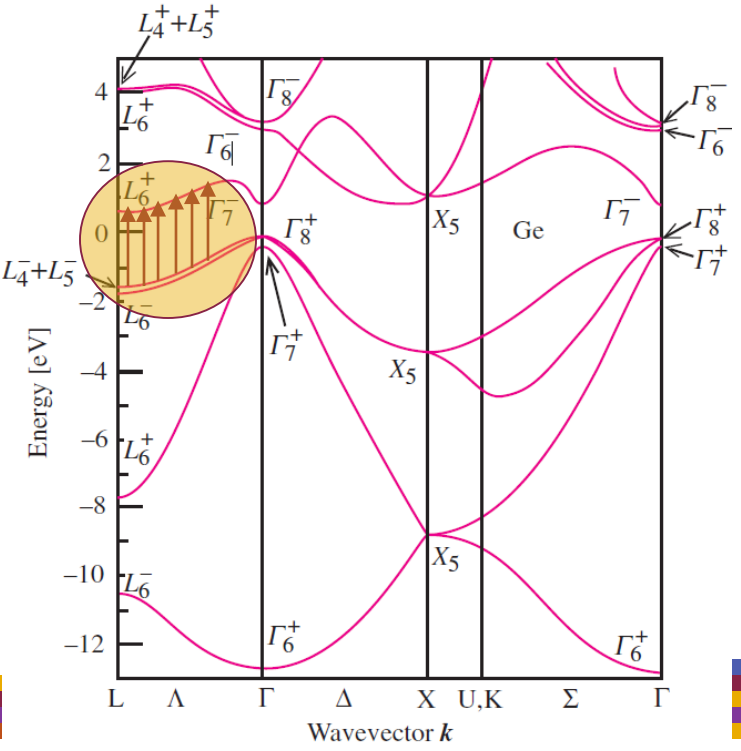


Van Hove Singularities: Beyond effective-mass approximation

$$\frac{1}{\tau(\hbar\omega)} = \frac{2\pi}{\hbar}|\langle f|H_{eR}|i\rangle|^2 g_{fi}(\hbar\omega)$$

Consider two spin states for each **k**.

$$g_{fi}(\hbar\omega) = \iiint_{i,f} \frac{d^3\vec{k}}{4\pi^3}\delta\left(\hbar\omega - E_{fi}(\vec{k})\right) = \frac{1}{4\pi^3}\oiint_{\hbar\omega=E_{fi}} \frac{dS}{|\vec{\nabla}_{\vec{k}} E_{fi}(\vec{k})|}$$



Van Hove singularity: Parallel bands

Use Taylor expansion around k_0:

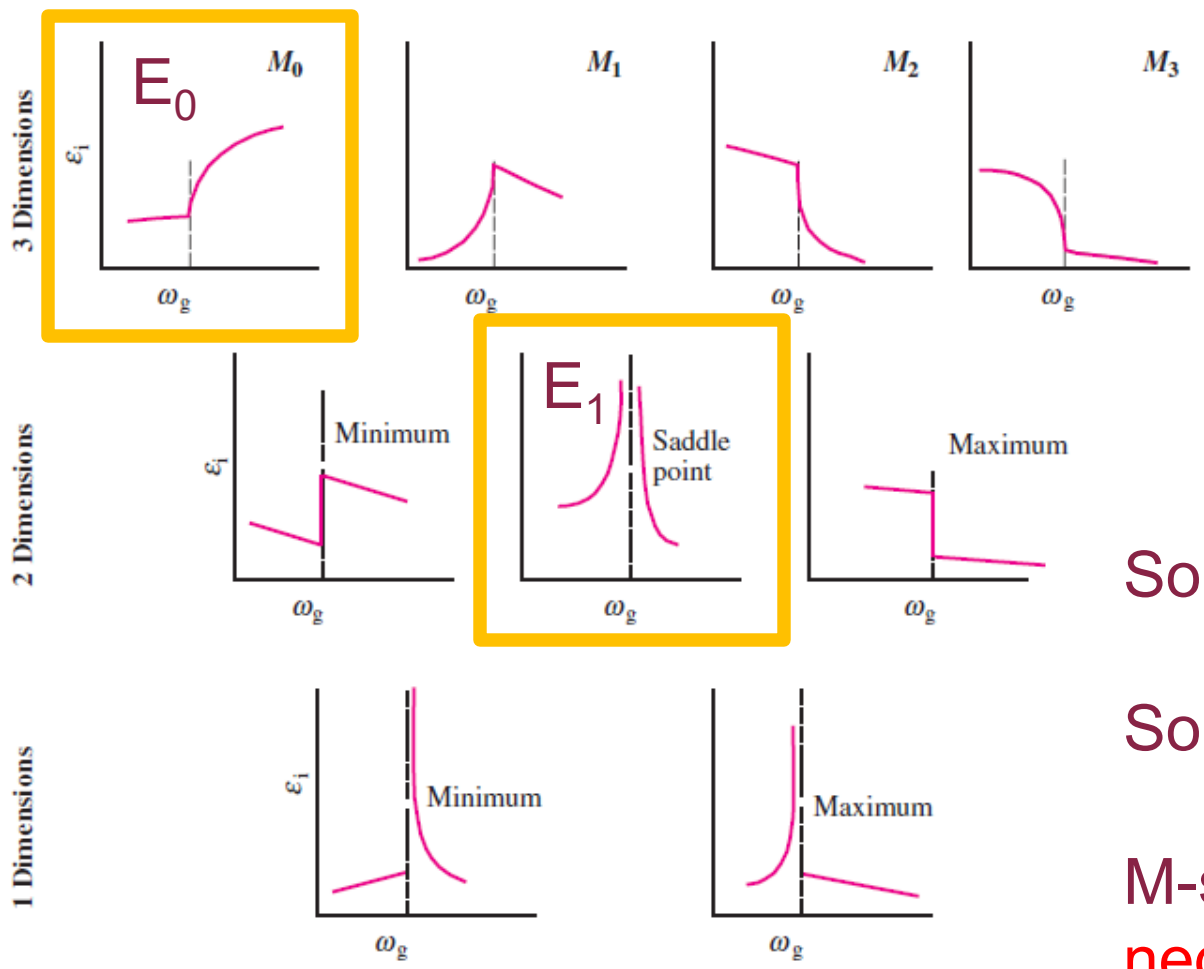
$$E_{fi}(\vec{k}) = E_{fi}(\vec{k}_0) + \sum_{i=1}^{3} a_i (k_i - k_{0i})^2$$

Some a_i small or zero: 1D, 2D, 3D

Some a_i positive, some negative

Ashcroft & Mermin, Chapter 8
Dresselhaus, Chapter 17

Critical Points



$$E_{fi}(\vec{k}) = E_{fi}(\vec{k}_0) + \sum_{i=1}^{3} a_i (k_i - k_{0i})^2$$

Some a_i small or zero:
1D, 2D, 3D

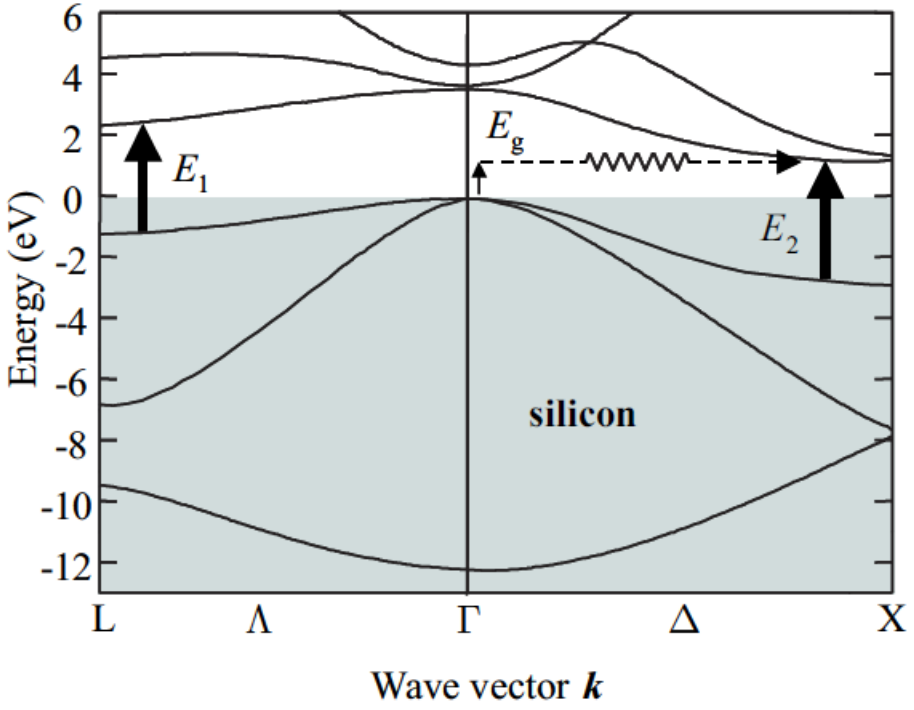
Some a_i positive,
some negative

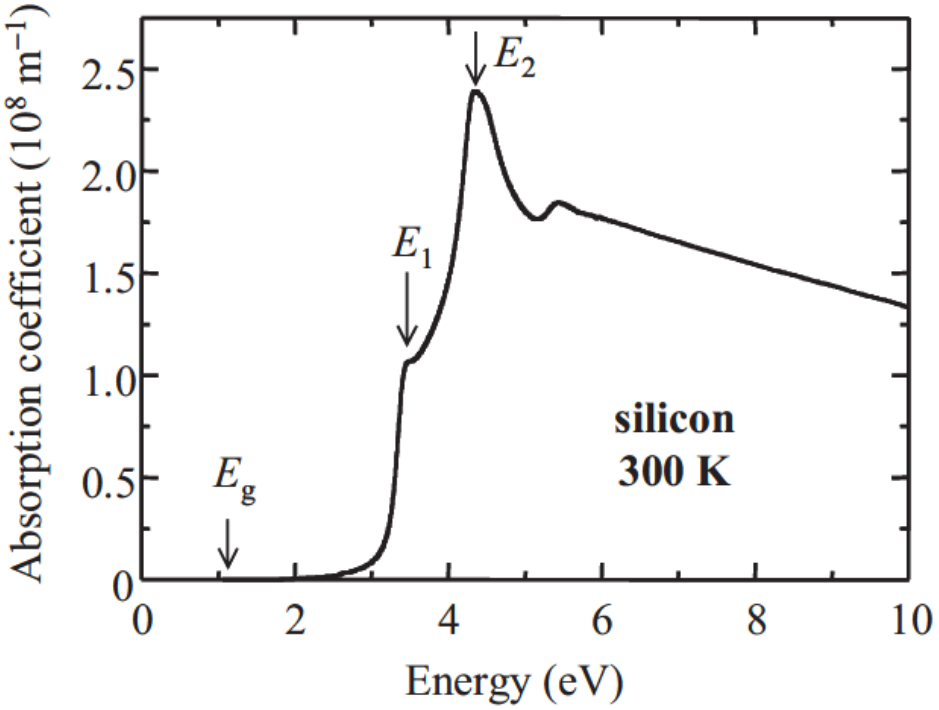
M-subscript: Number of negative mass parameters

	Type	D_j	
		$E < E_0$	$E > E_0$
Three dimensions	M_0	0	$(E - E_0)^{1/2}$
	M_1	$C - (E_0 - E)^{1/2}$	C
	M_2	C	$C - (E - E_0)^{1/2}$
	M_3	$(E_0 - E)^{1/2}$	0
Two dimensions	M_0	0	C
	M_1	$-\ln(E_0 - E)$	$-\ln(E - E_0)$
	M_2	C	0
One dimension	M_0	0	$(E - E_0)^{-1/2}$
	M_1	$(E_0 - E)^{-1/2}$	0

Yu&Cardona
Dresselhaus, Chapter 17

Critical points in silicon





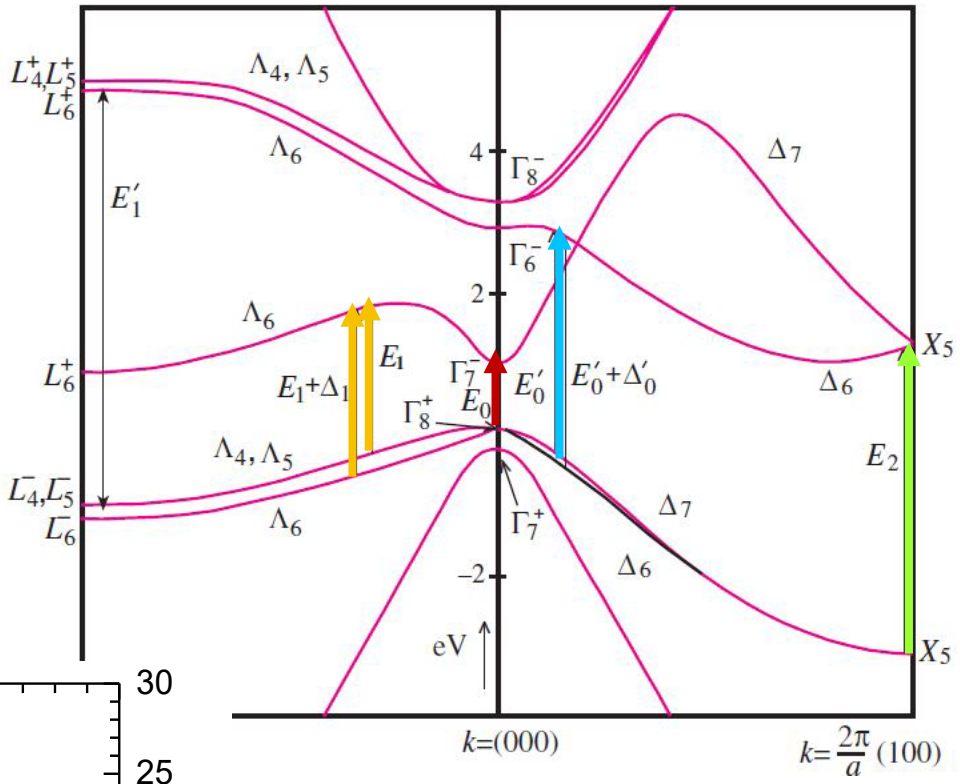
- Direct gap (3.4 eV) is much larger than indirect gap (1.1 eV).
- Si is almost transparent up to 3 eV.
- Spin-orbit splitting is small (no splittings).

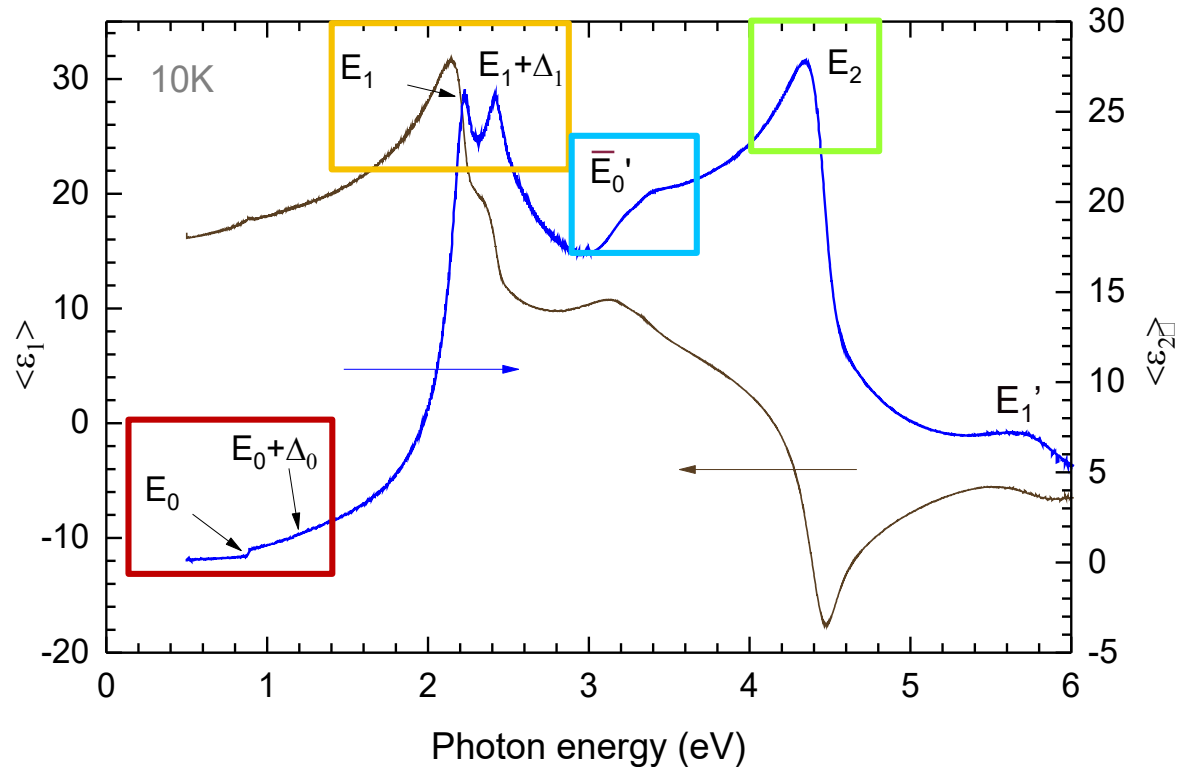
Fox, Chapter 3

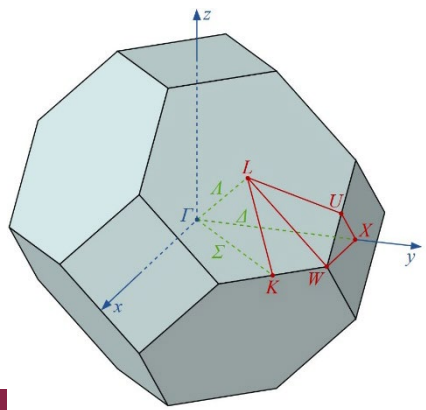
Critical Points in Germanium

- Structures in the dielectric function due to interband transitions
- Joint density of states
- Van Hove singularities

$$D_j(E_{CV}) = \frac{1}{4\pi^3}\int \frac{dS_k}{|\nabla_k(E_{CV})|}$$





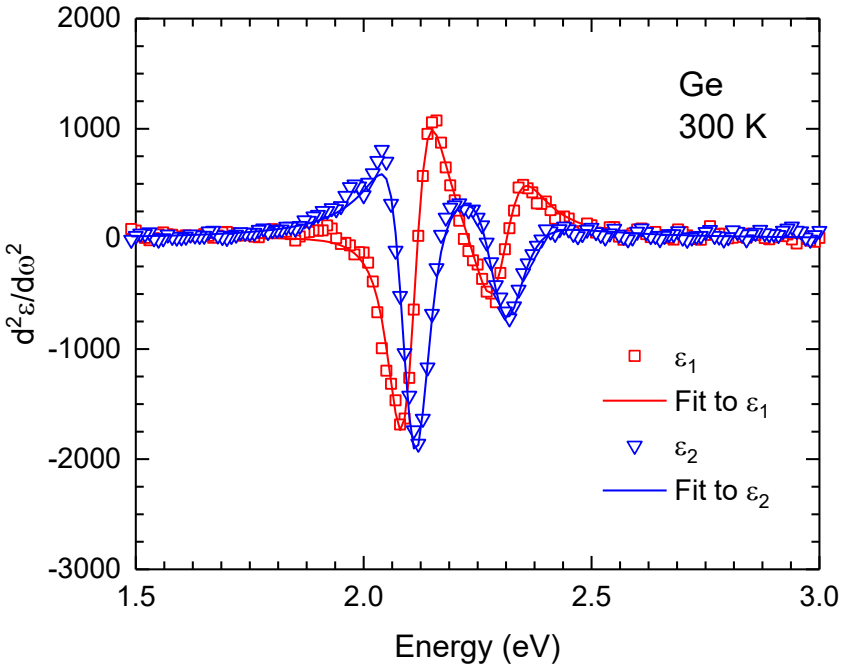


C. Emminger, ICSE-2019

Critical-point lineshapes

Type		D_j	
		$E < E_0$	$E > E_0$
Three dimensions	M_0	0	$(E - E_0)^{1/2}$
	M_1	$C - (E_0 - E)^{1/2}$	C
	M_2	C	$C - (E - E_0)^{1/2}$
	M_3	$(E_0 - E)^{1/2}$	0
Two dimensions	M_0	0	C
	M_1	$-\ln(E_0 - E)$	$-\ln(E - E_0)$
	M_2	C	0
One dimension	M_0	0	$(E - E_0)^{-1/2}$
	M_1	$(E_0 - E)^{-1/2}$	0

Savitzky-Golay derivative of ellipsometry spectra



General expression (1D, 3D):

$$\varepsilon(\hbar\omega) = C + Ae^{i\Phi}(\hbar\omega - E_0 + i\Gamma)^{(n-2)/2}$$

General expression (2D):

$$\varepsilon(\hbar\omega) = C + Ae^{i\Phi}\ln(\hbar\omega - E_0 + i\Gamma)$$

A	amplitude
Φ	excitonic phase angle
Γ	broadening

E_1 and $E_1+\Delta_1$ critical points, fitted with analytical lineshapes

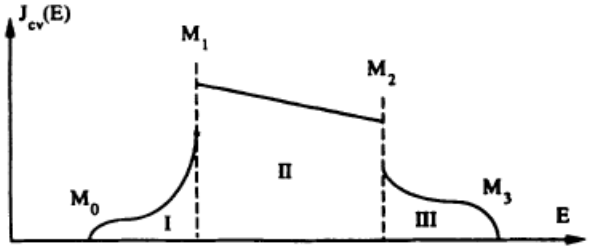
M. Cardona, Modulation Spectroscopy (Academic Press, 1966)
D.E. Aspnes, Surf. Sci. **37**, 418 (1973)

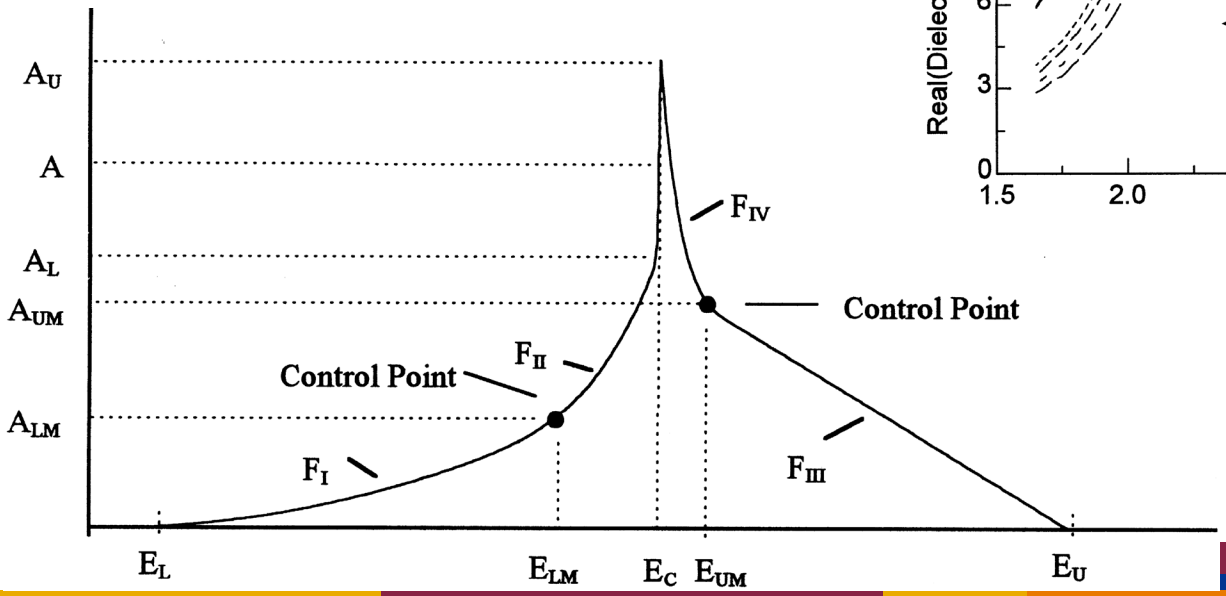
Parametric oscillator (PSEMI) model

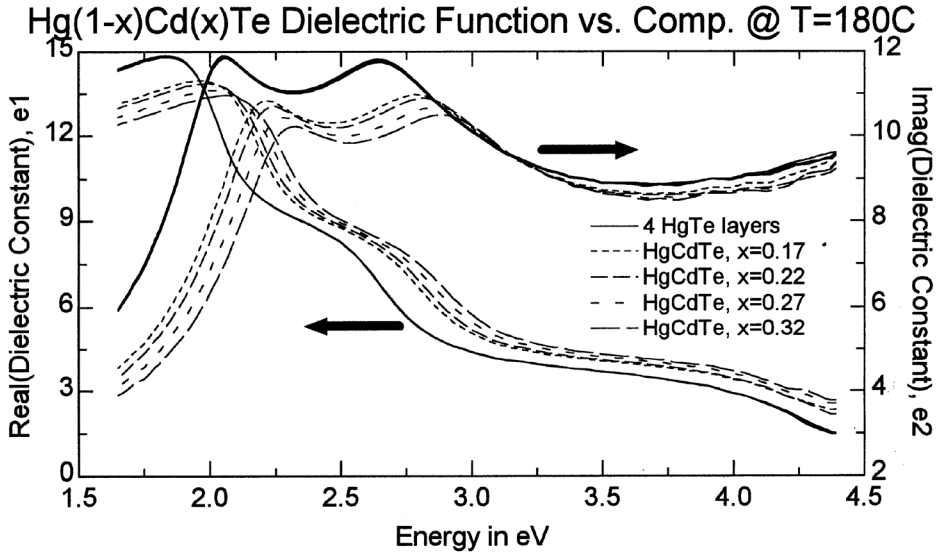
Critical points only valid near E_{CP} (Taylor expansion around E_{CP}, parabolic bands, constant matrix elements), work well to fit derivatives.

To fit ε, we need to add more parameters in the wings.

Many parameters.







B. Johs, C.M. Herzinger, Thin Solid Films **313-314**, 137 (1998).
C.C. Kim, J.W. Garland, Phys. Rev. B **45**, 11749 (1992).

Summary

Indirect gap absorption in Si and Ge

Experimental techniques to measure absorption

Van Hove singularities

Critical points in the dielectric function

Analytical lineshapes to fit Savitzky-Golay derivative

Parametric oscillator model

What's next ???

10: Excitons,
photoluminescence,
quantum confinement, wells, wires, dots

11: Applications I
What would you like to see ?
Please send email to zollner@fzu.cz

12: Applications II
Properties of thin films,
stress/strain, deformation potentials