Optical Properties of Solids: Lecture 8

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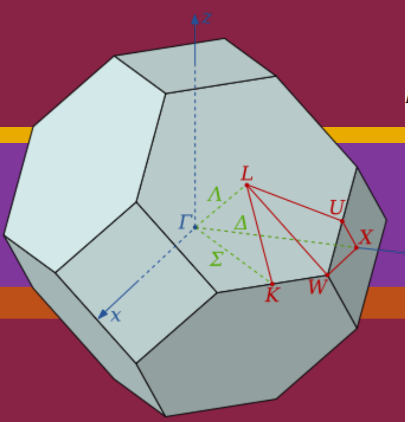
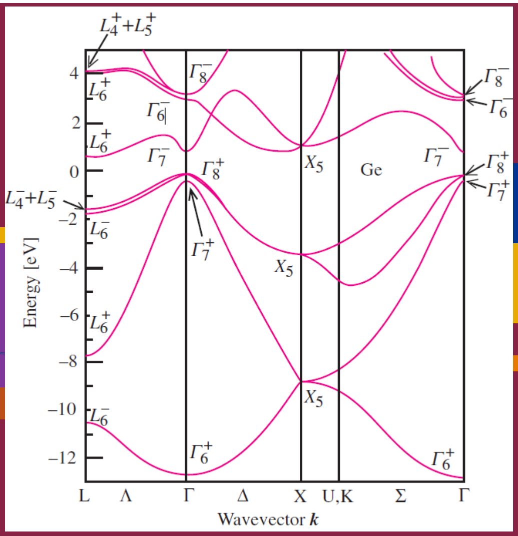
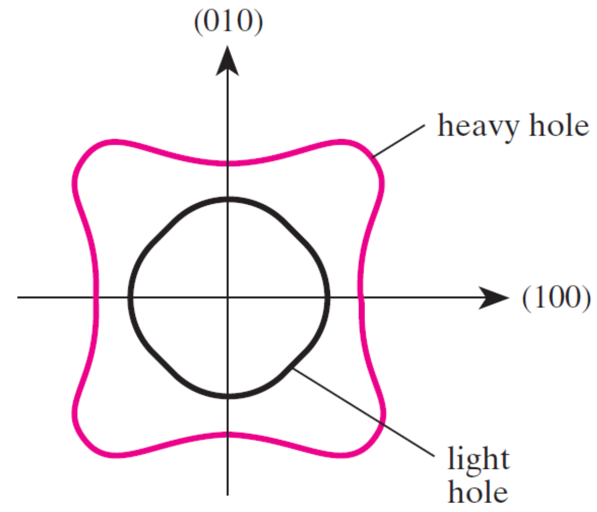
Optical Properties of Solids: Lecture 7+8+9

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







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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

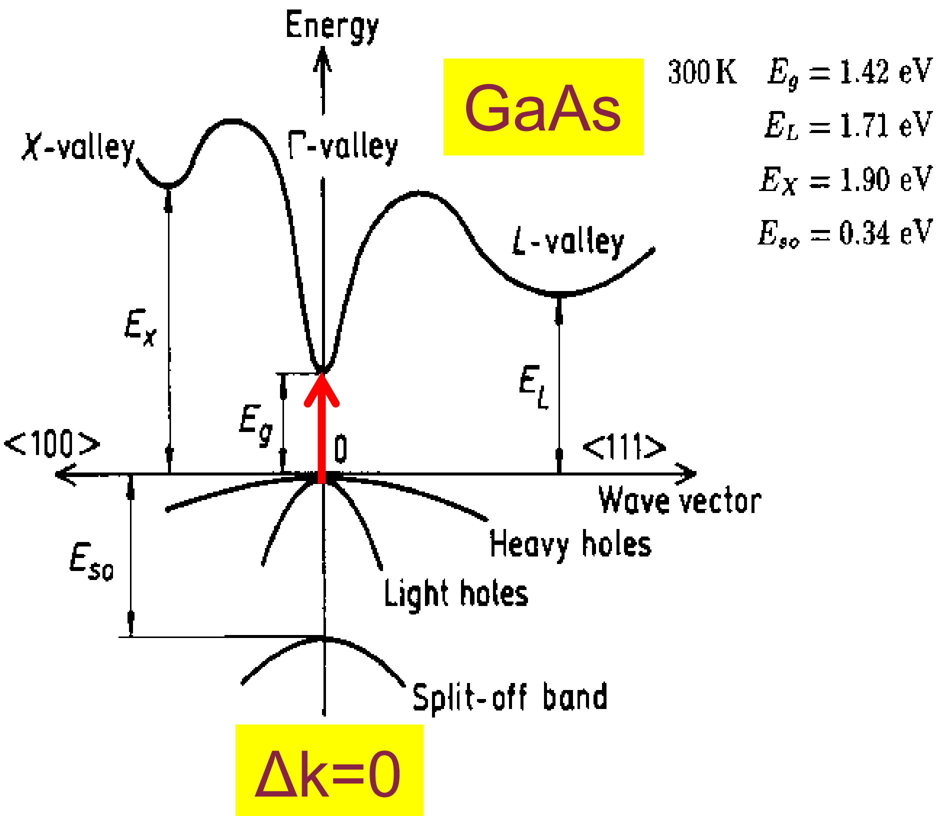
Van Hove singularities

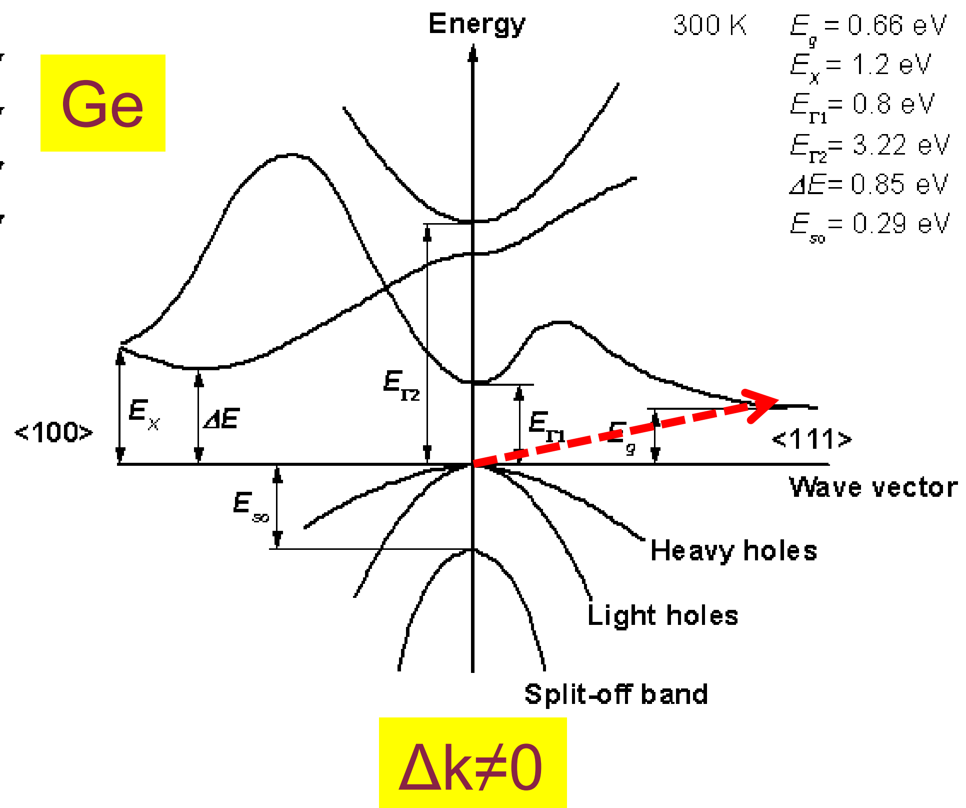
Critical points in the dielectric function

Analytical lineshapes to fit Savitzky-Golay derivative

Parametric oscillator model

Semiconductor Band Structures





Direct transition:

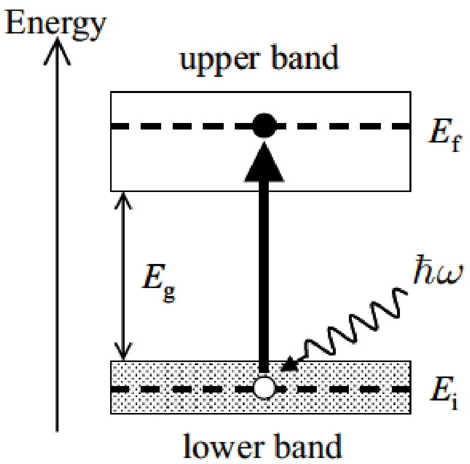
Initial and final electron state have **same** wave vector.

Indirect transition:

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

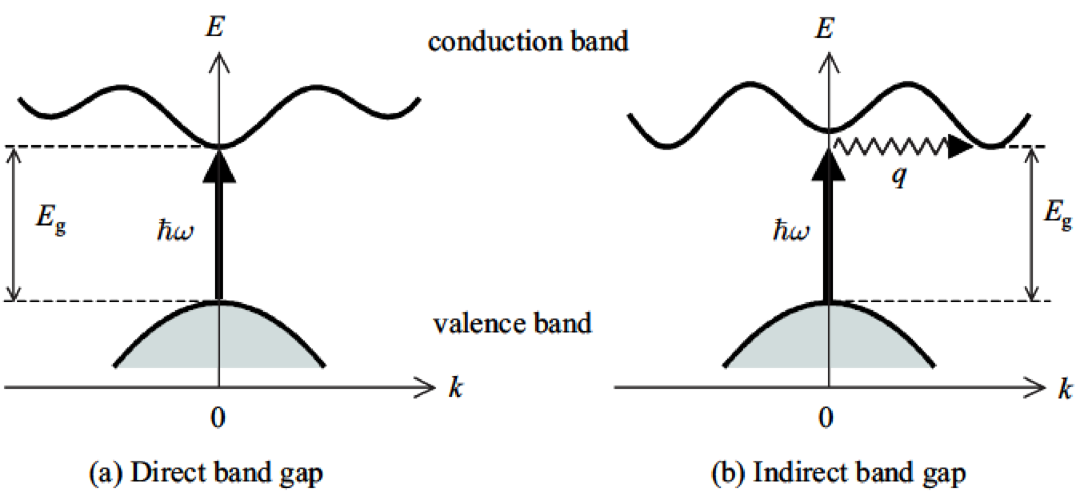
http://www.ioffe.ru/SVA/NSM/Semicond/index.html

Optical Interband Transitions



Absorption:
Incoming photon creates electron-hole pair
Recombination:
Electron-hole pair creates a photon

Energy and crystal momentum conserved (within Heisenberg uncertainty)



Indirect transitions require phonon to **conserve crystal momentum k**.
Consider *Umklapp* processes ($\pm$**G**).

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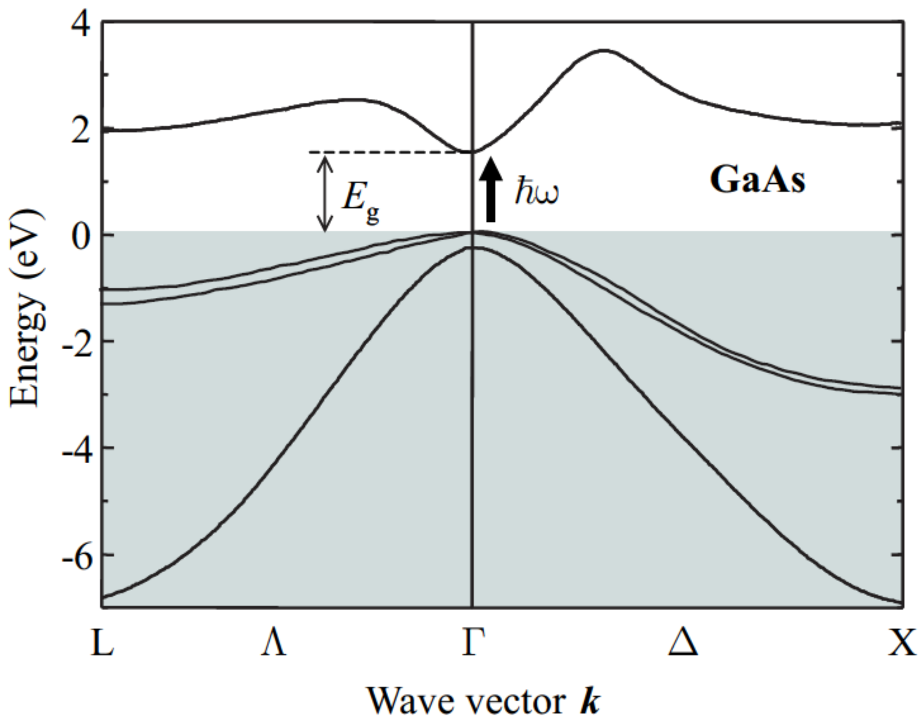
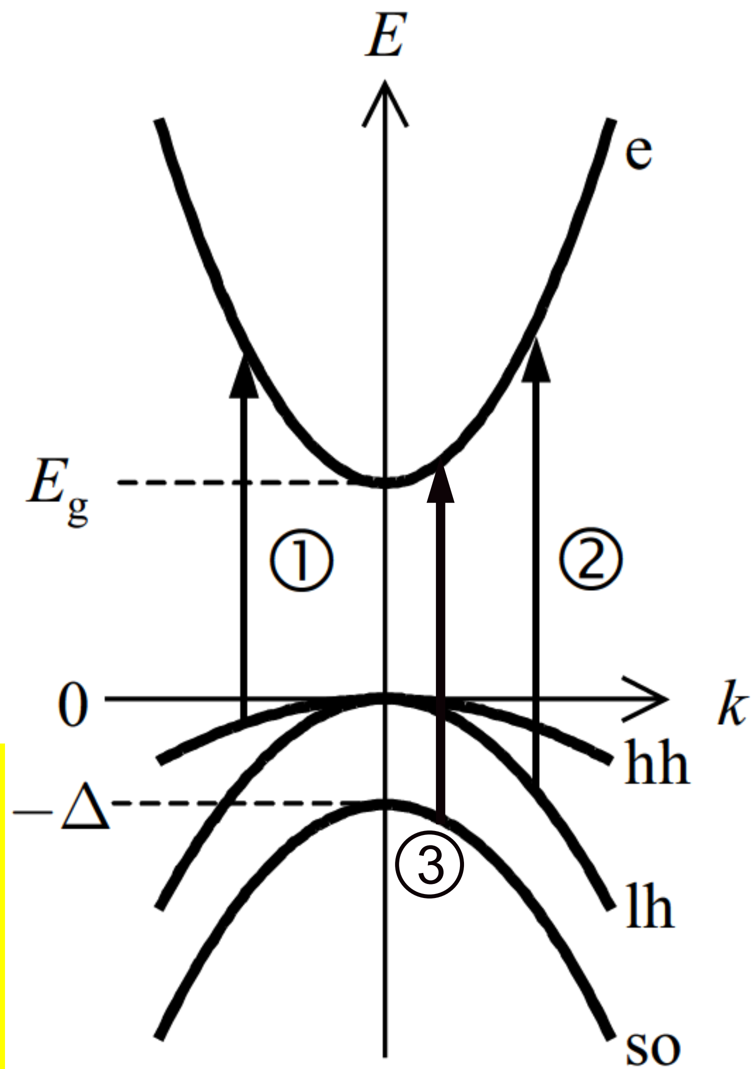
Optical Interband Transitions in GaAs





Various transitions are possible.
Consider non-parabolicity and warping.

How does absorption cross-section depend on energy and wave vector?
How do we describe absorption and emission?

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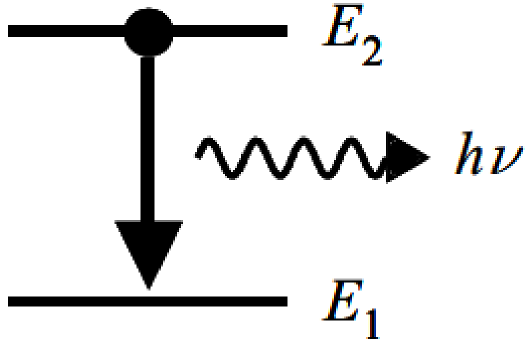
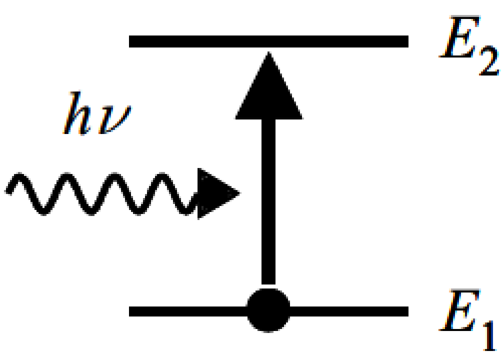
Einstein coefficients: Two-level system



(a) Emission (spontaneous)



(b) Absorption (stimulated)

Conservation of energy, no broadening

$$\hbar\omega = E_2 - E_1$$

Lifetime: $\tau = \frac{1}{A_{21}}$

$$\frac{dN_2}{dt} = -A_{21} N_2$$

(Einstein did not know about fermions and Pauli exclusion in 1917.)

$$\frac{dN_1}{dt} = -B_{12} N_1 u(\hbar\omega)$$

Paradox: For sufficiently high light intensity (or long lifetime), all electrons will end up in the excited state.

Fox, Appendix B
R.C. Hilborn, Am. J. Phys. **50**, 982 (1982).
A. Einstein, Phys. Z. **18**, 121 (1917).

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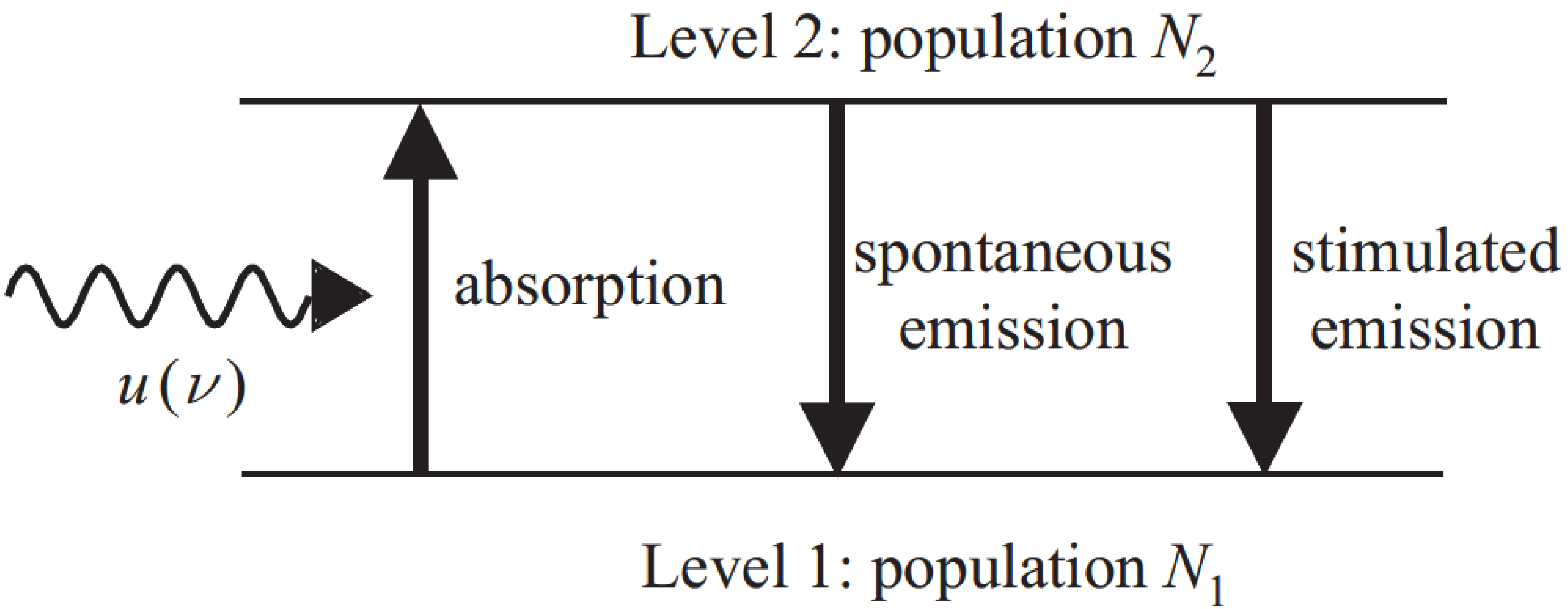
Einstein coefficients: Stimulated Emission



$$\frac{dN_2}{dt} = -B_{21}N_2u(\hbar\omega)$$

In equilibrium: N_1, N_2 constant. Absorption and emission balance.

$$B_{12}N_1u(\hbar\omega) = A_{21}N_2 + B_{21}N_2u(\hbar\omega)$$

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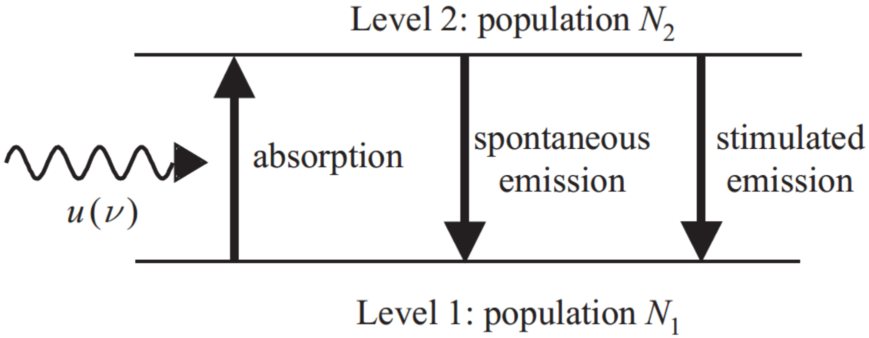

Einstein coefficients

$$B_{12}N_1u(\hbar\omega) = A_{21}N_2 + B_{21}N_2u(\hbar\omega)$$

In thermal equilibrium

$$\frac{N_2}{N_1} = \frac{g_2}{g_1}e^{-\frac{\hbar\omega}{kT}}$$

with black-body radiation

$$u(\hbar\omega) = \frac{2\hbar\omega^3}{\pi c^3}\frac{1}{e^{\frac{\hbar\omega}{kT}} - 1}$$

Einstein coefficients:

$$g_1B_{12} = g_2B_{21}$$

(let T→∞)

$$A_{21} = \frac{2\hbar\omega^3}{\pi c^3}B_{21}$$

One coefficient is sufficient, calculate from Fermi's Golden Rule.

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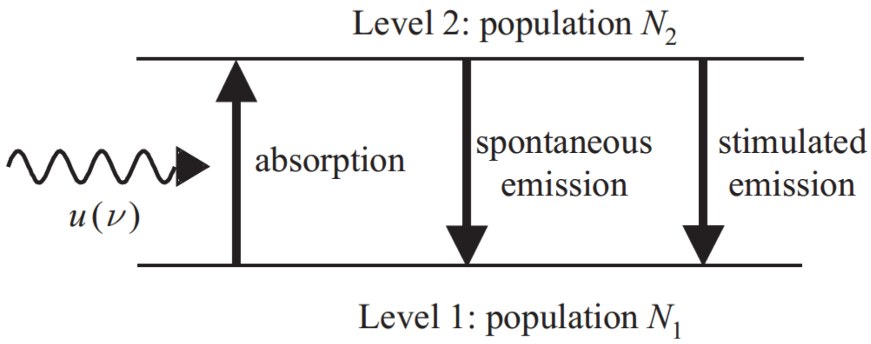
Population Inversion: Laser



$$B_{12}N_1u(\hbar\omega) = A_{21}N_2 + B_{21}N_2u(\hbar\omega)$$

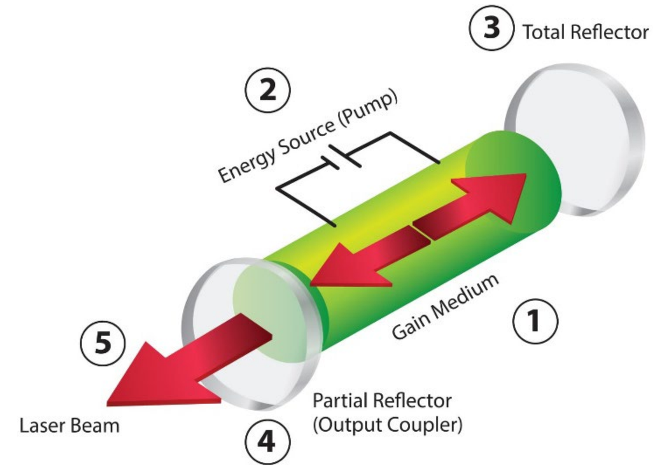
Lasing occurs if stimulated emission exceeds absorption.

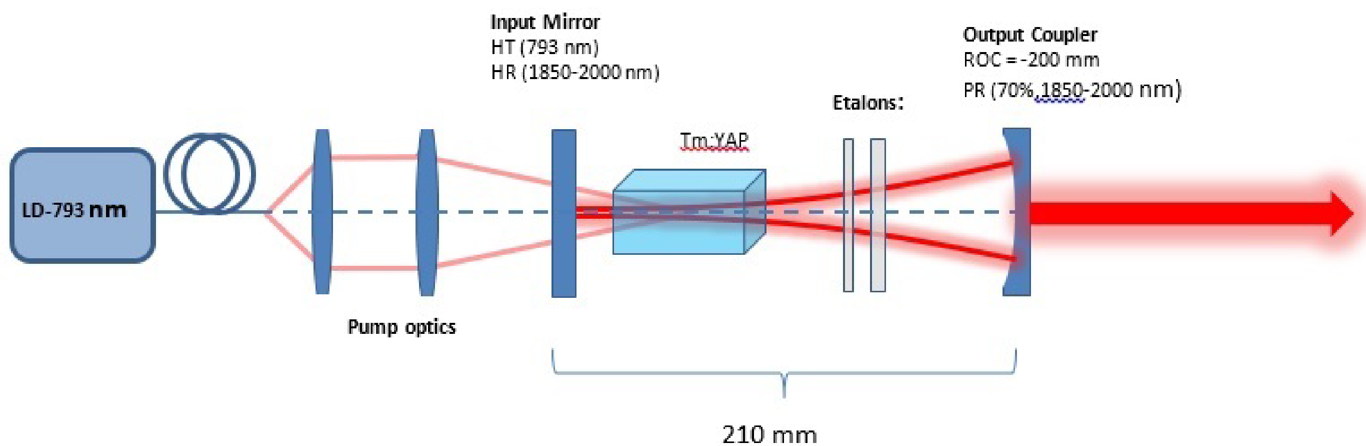
$$B_{21}N_2u(\hbar\omega) > B_{12}N_1u(\hbar\omega)$$

$$g_1B_{12} = g_2B_{21}$$

This requires **population inversion**

$$N_2 > \frac{g_2}{g_1}N_1$$





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Fermi's Golden Rule: Momentum, dipole matrix element

$$\frac{1}{\tau} = \frac{2\pi}{\hbar}\int_{i,f} |\langle f|H_{eR}|i\rangle|^2 \delta\left(E_f - E_i - \hbar\omega\right) = \frac{2\pi}{\hbar}|\langle f|H_{eR}|i\rangle|^2 g_{fi}(\hbar\omega)$$

Constant matrix element
Joint density of states

Interaction Hamiltonian:

Replace ***p*** with ***p***–q***A***
Only keep linear terms in ***A***
Coulomb gauge: div ***A***=0.
Long-wavelength limit: λ>>a
Expand exponential exp(i**k**·**r**)=1
E(t)=–d***A***/dt=iω***A*** (***A*** plane wave)

$$H_{eR} = \frac{e}{m_0}\vec{p}\cdot\vec{A} = \frac{e}{m_0}\vec{p}\cdot\vec{A}_0$$

$$\langle f|H_{eR}|i\rangle = \frac{e}{m_0}\langle \boldsymbol{f}|\vec{\boldsymbol{p}}|\boldsymbol{i}\rangle\cdot\vec{A}_0$$

Use **k·p** matrix element *P*

$$\vec{p} = m_0\vec{v} = m_0\frac{d\vec{r}}{dt} = \frac{im_0}{\hbar}[H_0,\vec{r}] = \frac{im_0}{\hbar}(H_0\vec{r} - \vec{r}H_0)$$

Ehrenfest theorem

$$\frac{e}{m_0}\langle f|\vec{p}|i\rangle = \frac{ie}{\hbar}\langle f|H_0\vec{r} - \vec{r}H_0|i\rangle = \frac{ie}{\hbar}\left\langle f\left|E_f\vec{r} - \vec{r}E_i\right|i\right\rangle = i\omega_{fi}\langle \boldsymbol{f}|\boldsymbol{e}\vec{\boldsymbol{r}}|\boldsymbol{i}\rangle$$

Electric dipole interaction

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Absorption selection rules for single electrons

$\langle f|e\vec{r}|i\rangle$

Quantum number	Selection rule	Polarization
Parity	changes	
l	$\Delta l = \pm 1$	
m	$\Delta m = +1$	circular: σ^+
	$\Delta m = -1$	circular: σ^-
	$\Delta m = 0$	linear: $\parallel z$
	$\Delta m = \pm 1$	linear: $\parallel (x, y)$
s	$\Delta s = 0$	
m_s	$\Delta m_s = 0$	

Selection rules are approximate in low-symmetry crystals for k≠0.

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Matrix element for direct transitions in a solid

$$\langle f|H_{eR}|i\rangle = \frac{e}{m_0}\langle f|\vec{p}|i\rangle\cdot\vec{A} = i\omega_{fi}\langle f|e\vec{r}|i\rangle\cdot\vec{A} = \langle f|e\vec{r}|i\rangle\cdot\vec{E}_0$$

Bloch's Theorem:

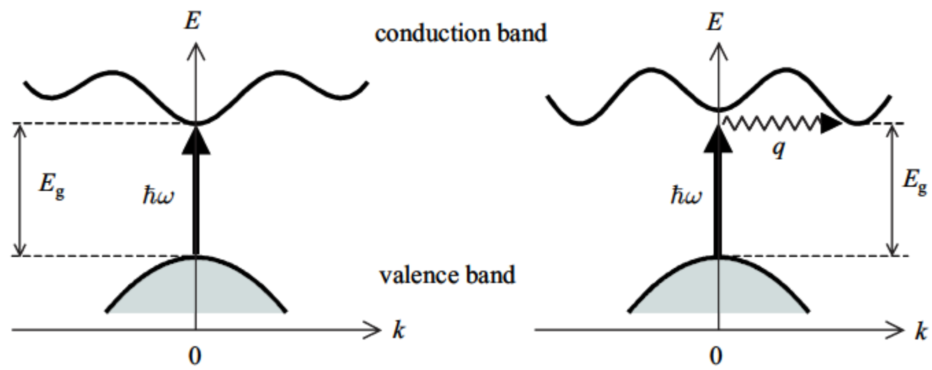
$$\vec{E} = -\frac{d\vec{A}}{dt} = i\omega\vec{A}$$

$$\psi_f(\vec{r}) = e^{i\vec{k}_f\cdot\vec{r}}u_f(\vec{r}) \qquad \psi_i(\vec{r}) = e^{i\vec{k}_i\cdot\vec{r}}u_i(\vec{r})$$

$$\langle f|H_{eR}|i\rangle = \langle u_f|e\vec{r}|u_i\rangle\cdot\vec{E}_0\delta\left(\vec{k}_f-\vec{k}_i\right)$$

Optical interband transitions must be direct: Δ**k**=0

Indirect transitions (Δ**k**≠0) require another particle (phonon, surface, defect, etc) to carry momentum. Indirect transitions require another matrix element.



(a) Direct band gap (b) Indirect band gap

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Joint density of states (effective mass approximation)

Assume constant matrix element (independent of **k**)

$$\frac{1}{\tau}=\frac{2\pi}{\hbar}\int_{i,f}|\langle f|H_{eR}|i\rangle|^2\delta\left(E_{fi}-\hbar\omega\right)=\frac{2\pi}{\hbar}|\langle f|H_{eR}|i\rangle|^2 g_{fi}(\hbar\omega)$$

$$g_{fi}(\hbar\omega)=\int_{i,f}\frac{d^3\vec{k}}{8\pi^3}\delta\left(E_{fi}-\hbar\omega\right)=\int_{i,f}dk\,\frac{4\pi k^2}{8\pi^3}\delta\left(E_{fi}-\hbar\omega\right)$$

$$E=\frac{\hbar^2k^2}{2m},\qquad dE=\frac{\hbar^2 k\,dk}{m}$$

Consider two spin states

$$g_{fi}(\hbar\omega)=\int_{i,f}dE\,\frac{1}{2\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2}\sqrt{E}\,\delta\left(E_{fi}-\hbar\omega\right)$$

$$g_{fi}(\hbar\omega)=\frac{1}{2\pi^2}\left(\frac{2m_{fi}}{\hbar^2}\right)^{3/2}\sqrt{\hbar\omega-E_{fi}}$$

for $\hbar\omega>E_{fi}$, 0 otherwise

Joint density of states

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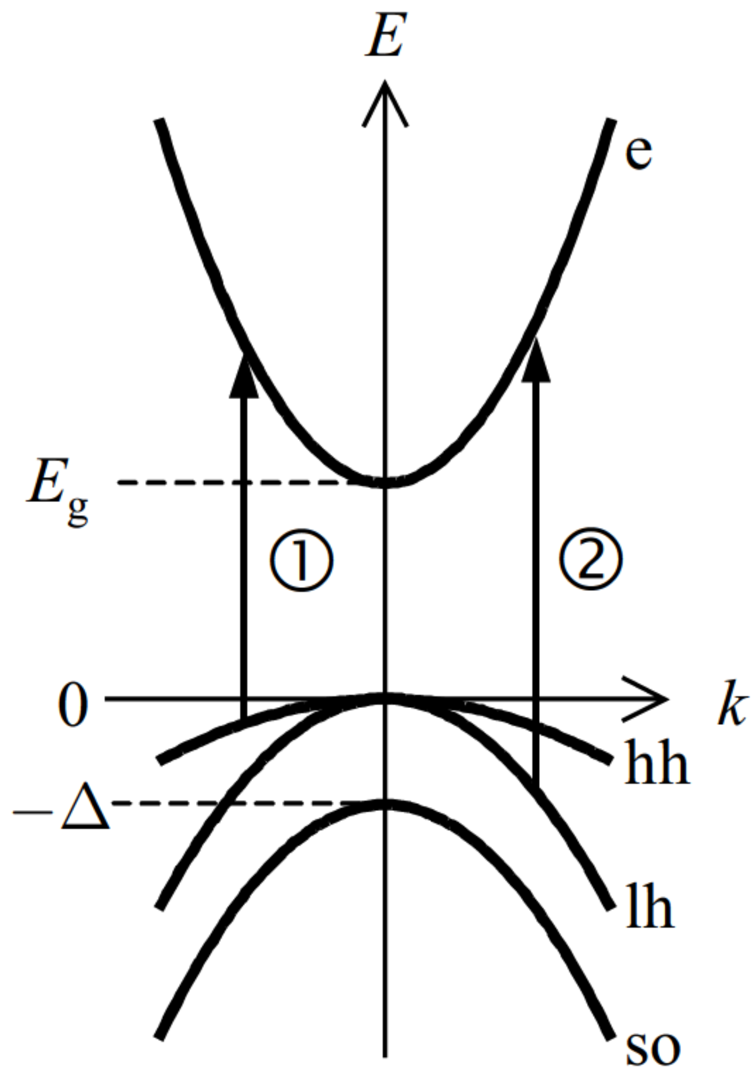
Optical (reduced) mass

$$E_c(k) = E_0 + \frac{\hbar^2 k^2}{2m_e^*}$$

$$E_{hh}(k) = -\frac{\hbar^2 k^2}{2m_{hh}^*}$$

$$E_{lh}(k) = -\frac{\hbar^2 k^2}{2m_{lh}^*}$$

$$E_{so}(k) = -\Delta_0 - \frac{\hbar^2 k^2}{2m_{so}^*}$$



$$\hbar\omega = E_c(k) - E_h(k) = E_0 + \frac{\hbar^2 k^2}{2\mu}$$

$$\frac{1}{\mu} = \frac{1}{m_e^*} + \frac{1}{m_h^*}$$

Optical mass μ

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Direct band gap absorption

For $\hbar\omega < E_g$: $\alpha = 0$

For $\hbar\omega > E_g$: $\alpha(\hbar\omega) \propto \sqrt{\hbar\omega - E_g}$

Yu & Cardona (6.58a)

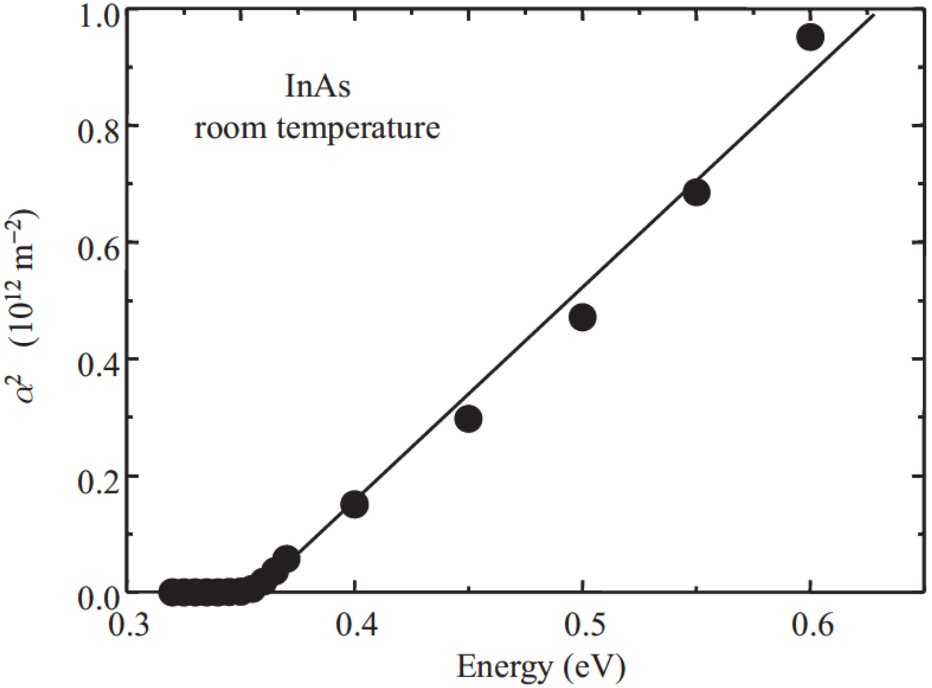
$$\varepsilon_2(\omega) = \begin{cases} 0, & x < 1 \\ A x^{-2}\sqrt{x-1}, & x \geq 1 \end{cases}$$

$$x = \frac{\hbar\omega}{E_g}$$

$$A = \frac{e^2 \mu^{3/2}}{3\sqrt{2}\pi\varepsilon_0 m\hbar} E_P E_g^{-3/2}$$

E_P is the **k·p** matrix element.

Tauc plot for direct band gap



Excitonic corrections needed for low temperatures and large band gaps.

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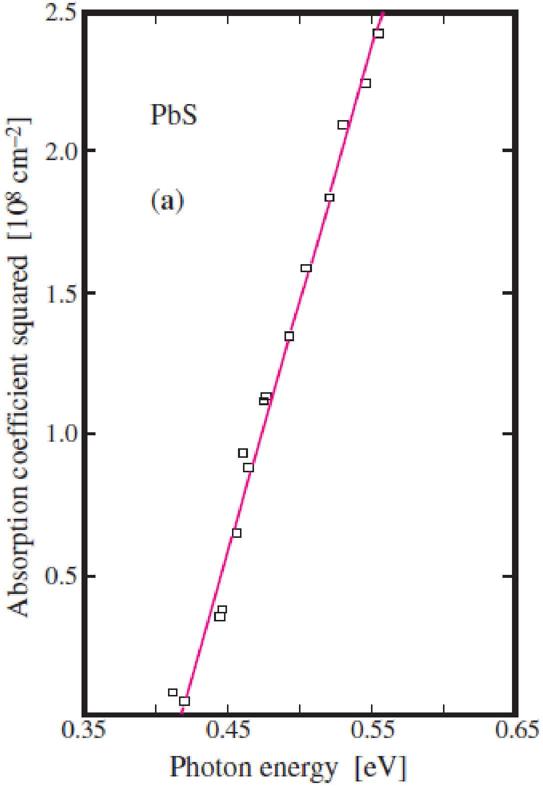
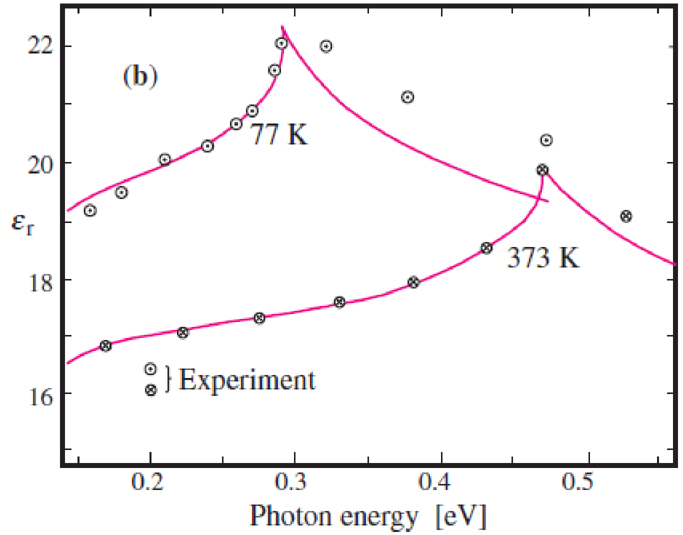
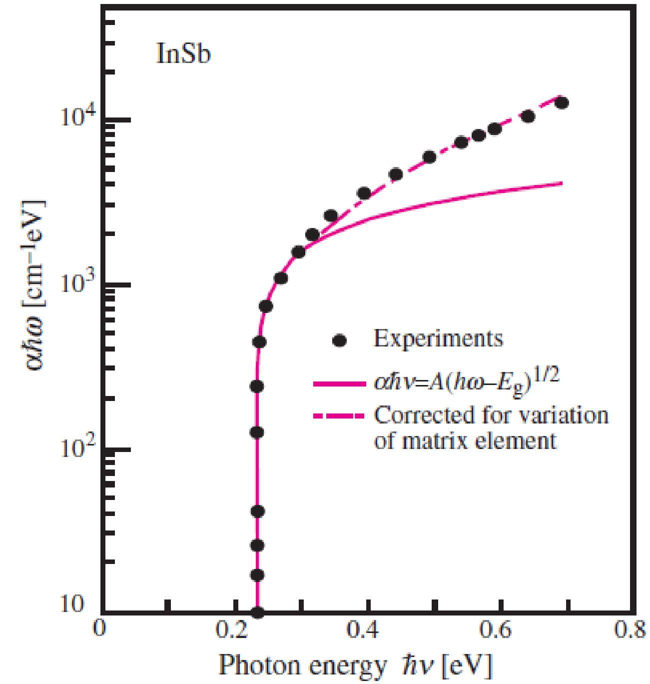
Yu & Cardona

Direct band gap absorption







PbS

PbS: real part ε_1 by Kramers-Kronig transform

InSb: Need k-dependent matrix elements

Yu & Cardona

Summary (Lecture 8)

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Fermi's Golden Rule

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