

EEE6212 Lecture 14 "Optical Transitions and the Density of States"

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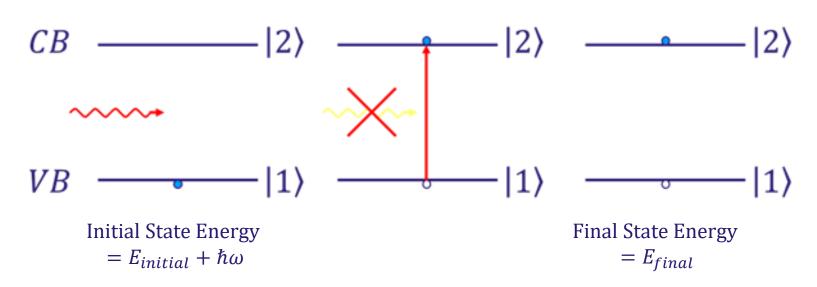


Outline

- Absorption
- Spontaneous emission
- Stimulated emission
- Density of states in 3D and 2D
- Occupancy of states
- Optical transitions in Quantum Wells
- Density of states 3D → 0D



Absorption



Consider a 2 state system with an electron in the lower energy state |1>

An incoming photon of energy $\hbar\omega$ is annihilated and gives energy to the electron promoting it to the higher energy state $|2\rangle$

"Stimulated" absorption – in response to the passing photon

Fundamental absorption – exciting electrons from the valence band to conduction band



Conserve Momentum

Momentum of an electron at the Brillouin Zone edge

$$p = \hbar k = \frac{h}{2\pi} \frac{\pi}{a}$$

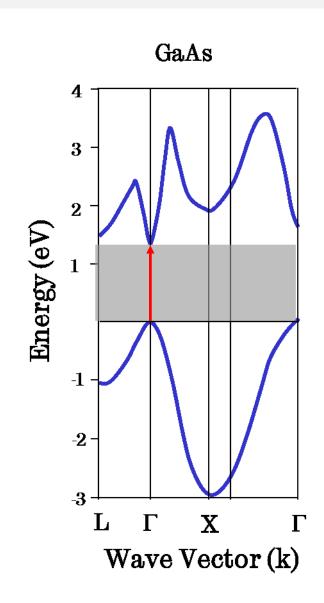
Interatomic spacing $a \sim 3 \times 10^{-10} m$

Momentum of a photon

$$p = \frac{h}{\lambda}$$

E.g.
$$\lambda = 840nm = 8.4 \times 10^{-7}m$$

Photon Momentum ~1000th that of electron so is essentially vertical when plotted on electron E-k graph





Absorption Rate

Will look later at how quantum mechanics can be used to calculate transition rates.

Initially we will use the original Einstein coefficients for these transition rates

Absorption rate

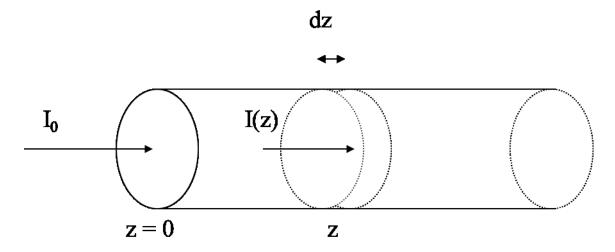
$$= B_{12} \times \begin{pmatrix} photon \\ density \end{pmatrix} \times \begin{pmatrix} density \ of \\ electrons \ in \ VB \end{pmatrix} \times \begin{pmatrix} density \ of \\ holes \ in \ CB \end{pmatrix}$$

Where B_{12} is a rate coefficient for absorption (s^{-1})

Probability proportional to photon density – "stimulated" absorption



Absorption Coefficient



Let I_0 be the number of photons incident / unit area / unit time at z=0, Absorption reduces I_0 to I(z) at z

In a further distance dz number of photons absorbed is $\alpha I(z)dz = -dI$ Hence

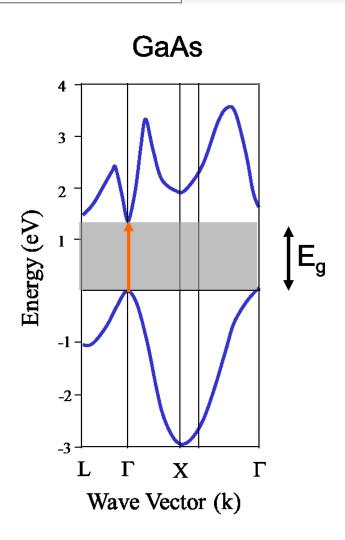
$$\frac{dI}{dz} = -\alpha I(z) \Rightarrow I(z) = I_0 e^{-\alpha z}$$

 α = absorption coefficient, has dimensions 1/L

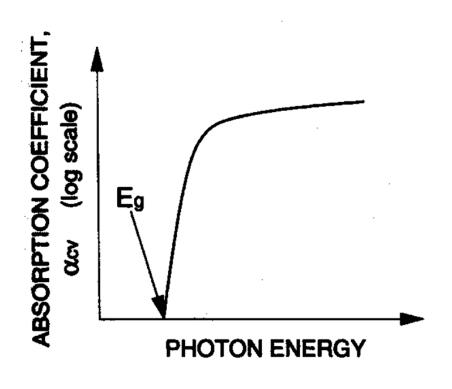
Typically $\alpha = 10^6 m^{-1}$ for GaAs at an energy just above the band gap



Direct Band-gap



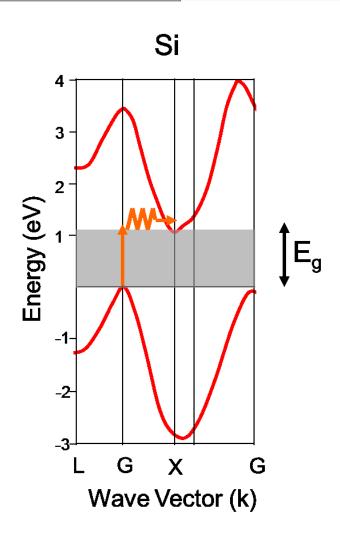
Direct Band-gap



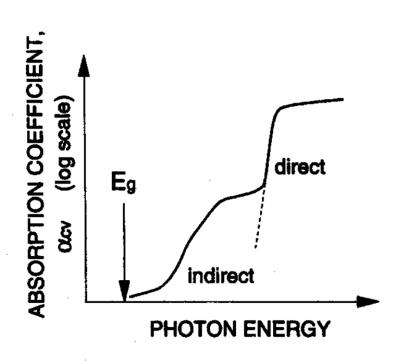
Absorption coefficient $\alpha \propto (\hbar \omega - E_g)^{1/2}$



Indirect Band-gap



Indirect Band-gap



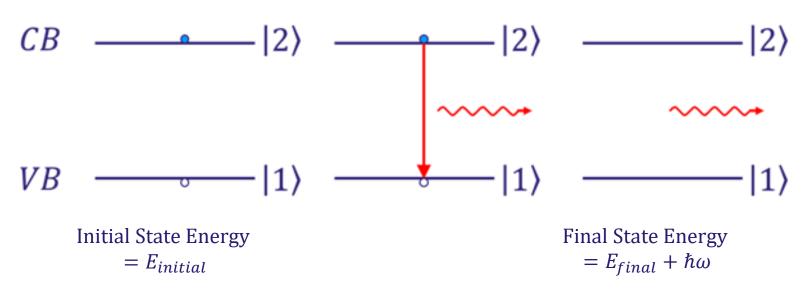
Indirect Absorption process only possible by phonon emission

Absorption coefficient

$$\alpha \propto (\hbar\omega - E_g \pm \hbar\omega_q)^{1/2}$$



Spontaneous Emission



Electron in the conduction band recombines with a hole in the valence band without any apparent provocation, emitting a photon

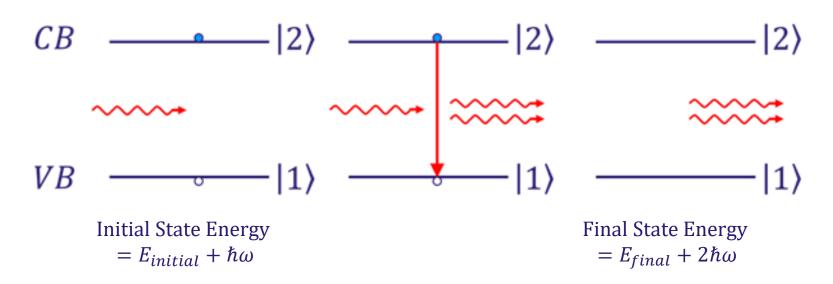
Spontaneous emission rate

$$= A_{21} \times \begin{pmatrix} density \ of \\ electrons \ in \ CB \end{pmatrix} \times \begin{pmatrix} density \ of \\ holes \ in \ VB \end{pmatrix}$$

Photons are emitted isotropically with random direction and phase



Stimulated Emission



Recombination rate proportional to photon density

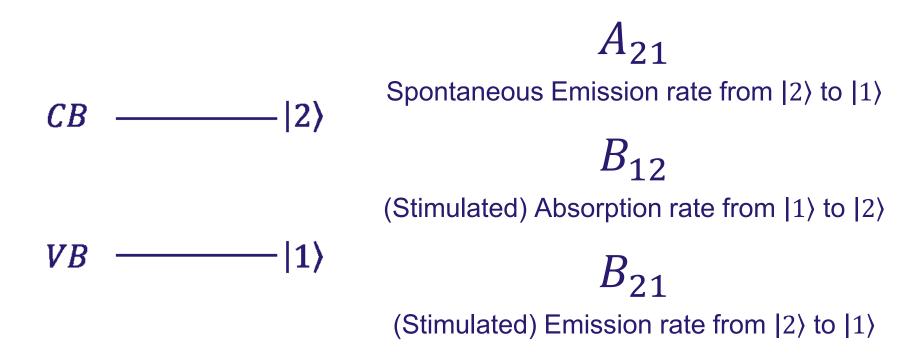
Stimulated emission rate

$$= B_{21} \times \begin{pmatrix} photon \\ density \end{pmatrix} \times \begin{pmatrix} density \ of \\ electrons \ in \ CB \end{pmatrix} \times \begin{pmatrix} density \ of \\ holes \ in \ VB \end{pmatrix}$$

Photons are identical in energy, phase and direction due to stimulated emission



Einstein Coefficients



Formulated before quantum mechanics, A & B coefficients are proportional to "Oscillator Strength"

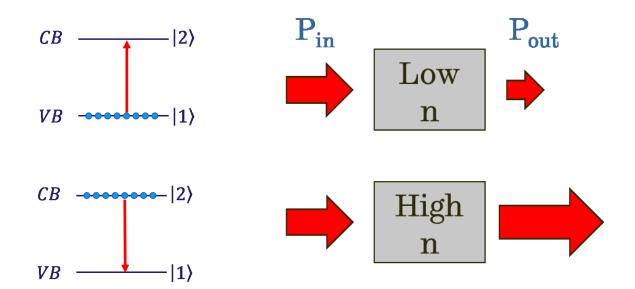
In the quantum picture the rates are governed by Fermi's Golden Rule



Absorption/Stimulated Emission

$$SE\ Rate = B_{21} \times {photon \choose density} \times {density\ of \choose electrons\ in\ CB} \times {density\ of \choose holes\ in\ VB}$$

$$Abs.Rate = B_{12} \times \begin{pmatrix} photon \\ density \end{pmatrix} \times \begin{pmatrix} density \ of \\ electrons \ in \ VB \end{pmatrix} \times \begin{pmatrix} density \ of \\ holes \ in \ CB \end{pmatrix}$$





Fermi's Golden Rule

$$W_{i\to f} = \frac{2\pi}{\hbar} |M_{if}|^2 g(\hbar\omega)$$

Where

W transition rate

i initial state

f final state

ħ reduced Planck constant

 M_{if} matrix element

 $g(\hbar\omega)$ joint density of states

In all cases (Spontaneous/Stimulated) we can see that the density of states is important.

First let's consider the matrix element...



Matrix Element

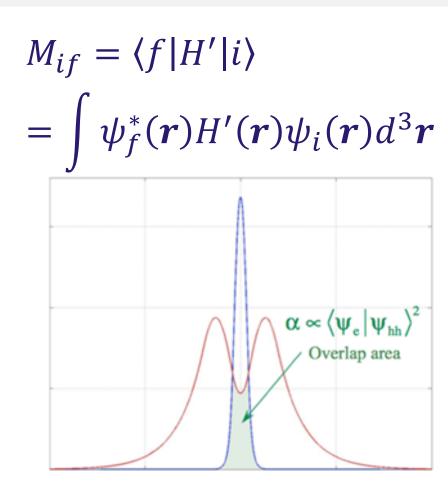
Matrix element describes the effect of the external perturbation of light on electrons

(See Mark Fox's book pages for a detailed description and derivation!)

The most important factor is the degree of overlap between the initial and final state wavefunctions

Large "Oscillator strength" corresponds to a high degree of overlap

Relevant to Type-I vs Type-II bandstructures





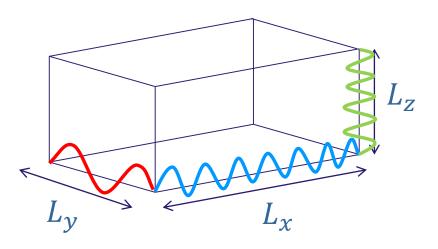
Density of States

We want to evaluate g(E)dE

The density of states g(E) over a given interval dE at energy E

Consider our particle in a box model in 3D And recall that the motion is decoupled

$$\psi(x, y, z) = \psi(x)\psi(y)\psi(z)$$



$$\psi(x, y, z) = \sin(k_x x) \sin(k_y y) \sin(k_z z)$$

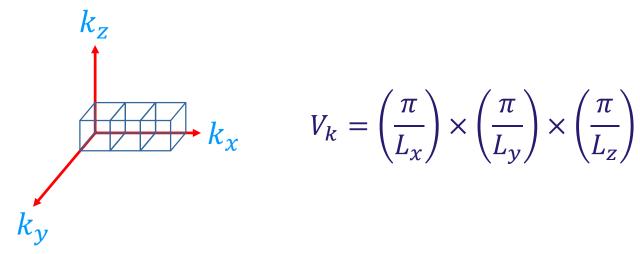
Where

$$k_x = \frac{n_x \pi}{L_x}$$
, $k_y = \frac{n_y \pi}{L_y}$, $k_z = \frac{n_z \pi}{L_z}$, for n_x , $n_y n_z$ integers



k-space

Each state in k-space occupies a k-space volume V_k



Number of states per volume (i.e. density) in k-space is the reciprocal of this

$$=\frac{L_x L_y L_z}{\pi^3} = \frac{V}{\pi^3}$$

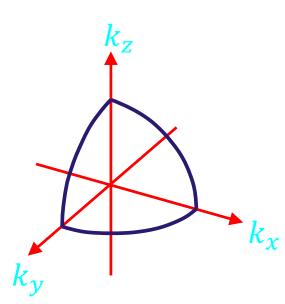
Where *V* is the volume of the semiconductor in real space



Number of states

How many states are there within the octant of the sphere of radius |k|?

Number of states = (Total Volume / Volume of a state) x 2 (for spin)



$$N = \left(\frac{1}{8} \times \frac{4\pi}{3} k_x k_y k_z\right) \times \frac{V}{\pi^3} \times 2$$

$$N = \frac{\pi k^3}{3} \times \frac{V}{\pi^3}$$

Assuming it's a sphere $(k_x = k_y = k_z = k)$



DOS (3D)

Density of States (DOS) = Number of states per unit volume per unit energy

$$DOS_{3D} = \frac{1}{V} \frac{dN}{dE}$$

Using the chain rule

$$= \frac{1}{V} \frac{dN}{dk} \times \frac{dk}{dE}$$

Where

$$N = \frac{\pi k^3}{3} \times \frac{V}{\pi^3}, \qquad \frac{dN}{dk} = \pi k^2 \times \frac{V}{\pi^3}$$

So

$$DOS_{3D} = \frac{k^2}{\pi^2} \frac{dk}{dE}$$

We know that

$$E = \frac{\hbar^2 k^2}{2m^*}$$

So
$$k = \frac{\sqrt{2m^*E}}{\hbar}$$
 and $\frac{dk}{dE} = \frac{m^*}{\hbar^2 k}$



DOS (3D)

So finally

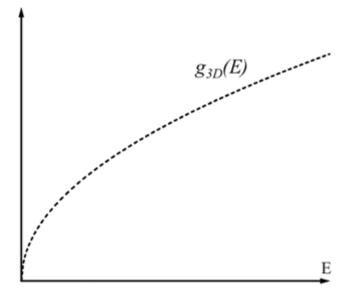
$$DOS_{3D} = \frac{k^2}{\pi^2} \frac{dk}{dE} = \frac{k^2}{\pi^2} \frac{m^*}{\hbar^2 k} = \frac{m^*}{\pi^2 \hbar^2} \frac{\sqrt{2m^* E}}{\hbar}$$

Noting that all energies are relative to the conduction band edge $E-E_c$ This becomes

$$DOS_{3D} = \frac{1}{2\pi^2} \left(\frac{2m^*}{\hbar^2}\right)^{\frac{3}{2}} \sqrt{E - E_c}$$

In 3D the density of states is proportional to the square root of the Energy and also $m^{*^{3/2}}$

Large effective mass → Large density of states

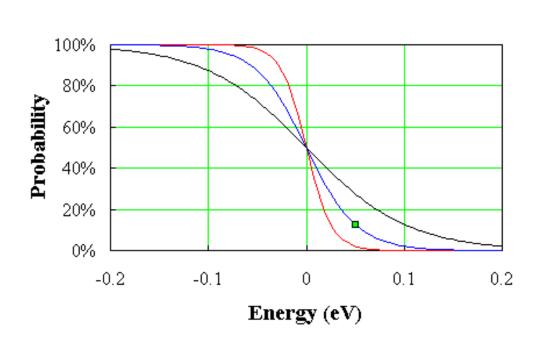


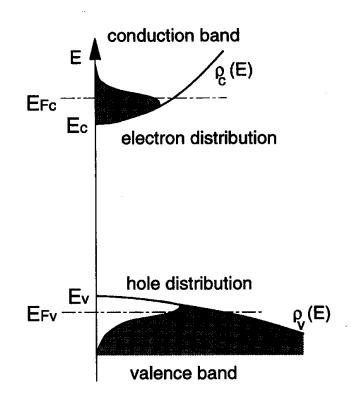


Carrier Distribution

The electrons and holes have thermal energy (Fermi Distribution)

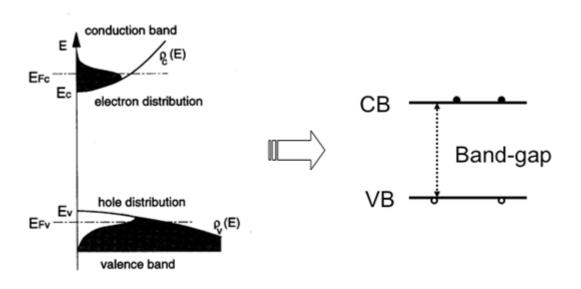
$$f(E) = \frac{1}{1 + e^{(E - E_F)/kT}}$$







Behind the scenes



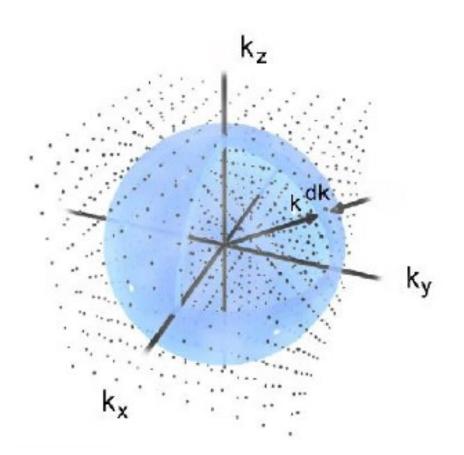
The occupancy of the states is a combination of the density of states and the fermi distribution (but we could modify this electrically or optically)

$$SE\ Rate = B_{21} \times \begin{pmatrix} photon \\ density \end{pmatrix} \times \begin{pmatrix} density\ of \\ electrons\ in\ CB \end{pmatrix} \times \begin{pmatrix} density\ of \\ holes\ in\ VB \end{pmatrix}$$

We will return to this when we discuss LASERs...



Number of states for a given |k|



Slightly different way of calculating the 3D DOS

Construct a spherical shell of radius |k| and thickness dk

Volume of this spherical shell in k-space is

$$4\pi k^2 dk$$

Number of k-states within the shell (k - snace)

$$= \binom{k - space}{volume} \times \binom{k - space}{density}$$

$$g(k)dk = 4\pi k^2 \left[\frac{V}{\pi^3}\right] dk$$



(contd)

Each state can hold 2 spins so x2

$$g(k)dk = 8\pi k^2 \left[\frac{V}{\pi^3}\right] dk$$

Each octant is indistinguishable so x1/8

(can't tell the difference between +k and –k)

$$g(k)dk = \pi k^2 \left[\frac{V}{\pi^3}\right] dk$$

Converting to Energy

$$p = \hbar k, E = \frac{p^2}{2m^*}, \qquad \Rightarrow E = \frac{\hbar^2 k^2}{2m^*}$$

Rearranging

$$k^2 = \frac{2m^*E}{\hbar^2}$$



Differentiating $k^2 = \frac{2m^*E}{\hbar^2}$ implicitly

$$2kdk = \frac{2m^*}{\hbar^2}dE$$

So

$$dk = \frac{m^*}{k\hbar^2} dE$$

Substituting into

$$g(k)dk = \pi k^2 \left[\frac{V}{\pi^3} \right] dk = \frac{V}{\pi^2} \times k^2 \times \frac{m^*}{k\hbar^2} dE$$

Gives

$$g(k)dk = \frac{Vm^*}{\pi^2\hbar^2} \times kdE = \frac{Vm^*[2m^*E]^{1/2}}{\pi^2\hbar^3}dE$$

Noting that all energies are relative to the conduction band

$$E \Rightarrow E - E_c$$



Density of states in 3D

Finally dividing by volume gives the number of states in the conduction band per unit volume in an energy range dE

$$g(E)dE = \frac{m^*[2m^*(E - E_c)]^{1/2}}{\pi^2\hbar^3}dE$$

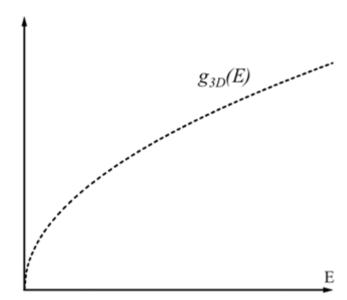
Which can be written more neatly as

$$g(E)dE = \frac{1}{2\pi^2} \left(\frac{2m^*}{\hbar^2}\right)^{\frac{3}{2}} \sqrt{E - E_c} dE$$

In 3D the density of states is proportional to the square root of the Energy and also $m^{*3/2}$

Large effective mass → Large density of states

Same result but a slightly different approach





Density of states in 2D

In 2D you consider the number of states in a circular ring (annulus) of radius |k| and width dk

The ring has area $2\pi kdk$

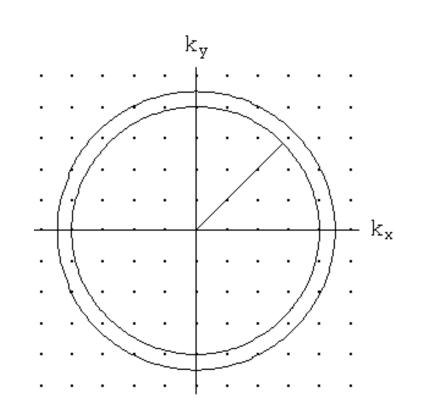
And a state in k-space occupies an area $A_k = \left(\frac{\pi}{L_x}\right) \times \left(\frac{\pi}{L_y}\right)$

So the number of states per unit area in k-space is A/π^2 where A is the area in real space

$$g(k)dk = 2 \times \frac{1}{4} \times 2\pi k \left[\frac{A}{\pi^2}\right] dk = \frac{A}{\pi} k dk$$

2 for spin

1/4 for only the +ve quadrant





Density of states in 2D

Per unit area we then get

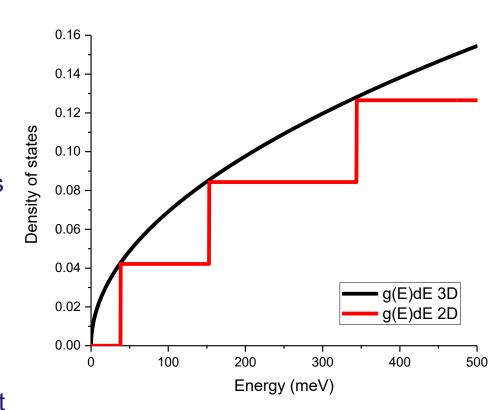
$$g(E)dE = \frac{1}{\pi}kdk = \frac{1}{\pi}\frac{m^*dE}{\hbar^2}$$
$$= \frac{m^*}{\pi\hbar^2}dE$$

This is constant and no longer depends on Energy

There are many available states as soon as the 1st energy level is reached.

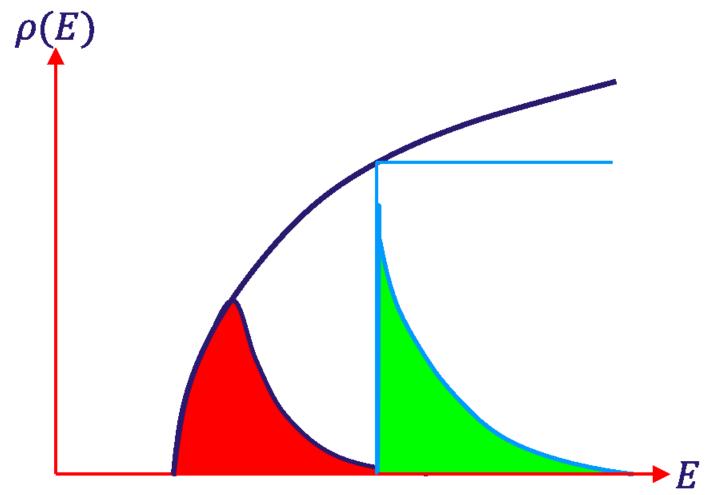
Taking lower energy levels into account the

2D DOS is a staircase of step functions.





2D vs 3D carrier distributions

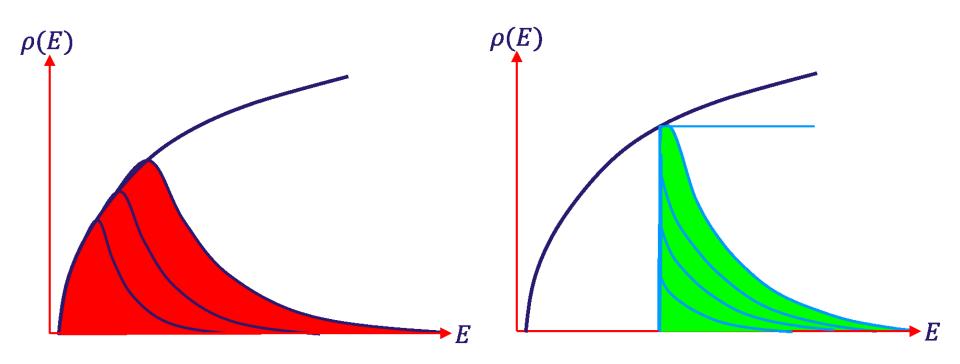


In addition to reducing the volume (and hence total number of states)

Quantum confinement acts to concentrate the carriers at the required wavelength



Increasing occupancy



In the 3D case the peak of the density distribution is shifting as the population increases → shift in emission wavelength

In the Quantum Well case this shift is delayed until higher levels of occupation

→ more stable emission wavelength with Temperature (for example)



Quantum Well – Transitions

Fermi's Golden Rule

$$W_{i\to f} = \frac{2\pi}{\hbar} \left| M_{if} \right|^2 g(\hbar\omega)$$

Where

$$M_{if} = \langle f | H' | i \rangle$$

$$= \int \psi_f^*(\mathbf{r}) H'(\mathbf{r}) \psi_i(\mathbf{r}) d^3 \mathbf{r}$$

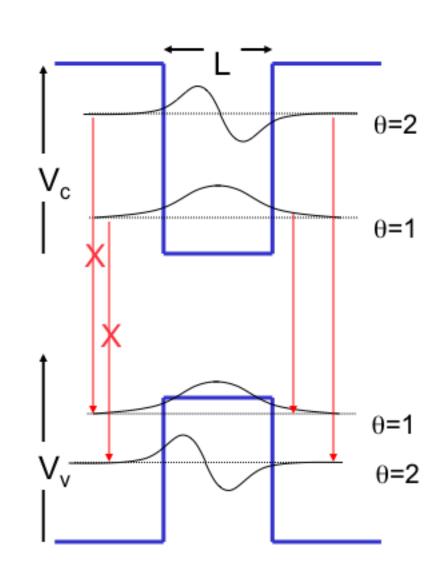
Due to Symmetry only transitions such as

$$\theta = 1 \rightarrow \theta = 1$$
 and $\theta = 2 \rightarrow \theta = 2$

Are allowed whilst

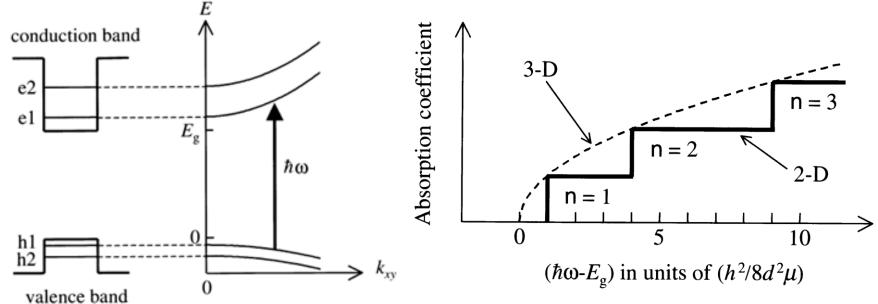
$$\theta = 1.2 \rightarrow \theta = 2.1$$

Are forbidden





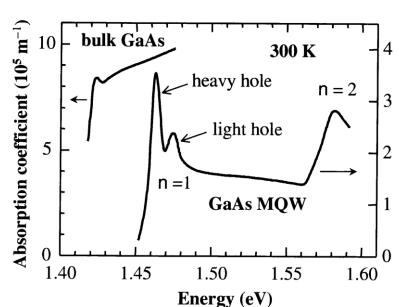
QW Absorption + Excitons



Simple picture involving momentum conservation and QW selection rules is not sufficient to explain absorption seen in 3D or 2D, (need excitons as well)

Quantum confinement forces the electrons and holes closer together than in bulk materials

Leads to $\sim 2.5 \times$ increase in the exciton binding energy \rightarrow Excitonic effects at RT





Density of states in 1D

In 1D consider a line of length L in real space

For
$$k = (k_x, k_y, k_z) = (\frac{\pi n_x}{L_x}, k_y, k_z)$$
 where n is an integer

the volume of the unit cell in k-space is π/L and the space between the end points of a line from |k| to |k+dk| is 2dk

The number of states in k-space is then

$$g(k)dk = 2 \times \frac{1}{2} \times 2dk \times \frac{L}{\pi}$$

Per unit length we then get

$$g(E)dE = \frac{2}{\pi}dk = \frac{(2m^*)^{1/2}}{\pi\hbar}E^{-1/2}dE$$

This has a form similar to the branch of a hyperbola

Again taking into account the lower energy levels the 1D DOS is a staircase of such branches stacked on top of each other



Density of states in 0D

All values of k are quantised in all three directions in k-space

There is no continuous k-space to be filled and all available states exist only at discrete energies

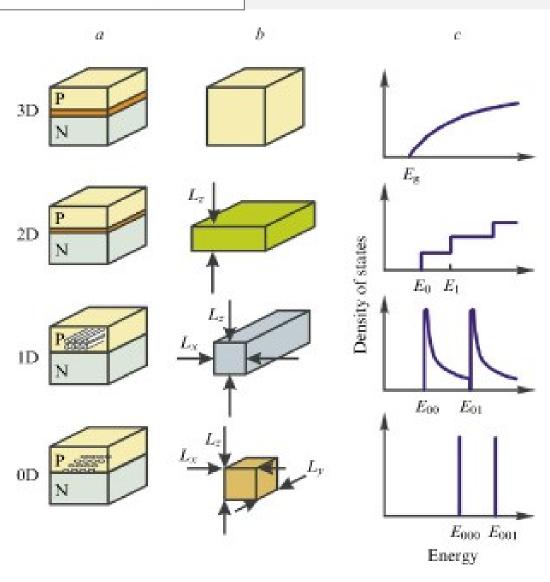
So the DOS in 0D for electrons can be described by a delta function (again with a factor of 2 for the spins)

$$g(E)dE = 2\delta(E - E_c)$$

Taking into account lower energy levels the 0D DOS is a series of delta functions



DOS Overview - 1



Type of confinement

None = 3D

Potential well or Quantum Well confines particles to movement in the (x,y) plane = 2D

A Quantum Wire or nanorod confines the particles to movement along y-direction only = 1D

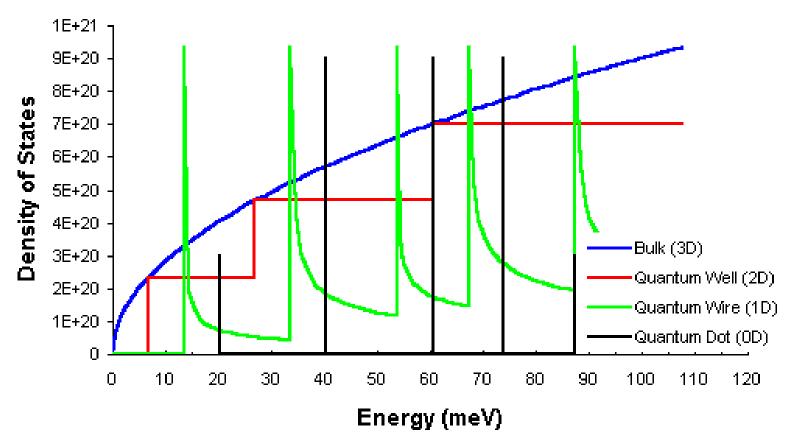
Total confinement by a Quantum Dot, the particle can no longer move at all = 0D



DOS overview - 2

The stronger the confinement is, the more the initially continuous DOS splits up into discrete levels

This concentrates the density of states into an ever narrower a range of wavelengths in optical emission or absorption which is useful for optoelectronic devices





Summary

- Introduced at Absorption, Spontaneous emission, Stimulated emission
- Density of states in 3D and 2D
- Occupancy of states
- Optical Transitions in Quantum Wells
- Density of states 3D → 0D





EEE6212 Lecture 15 "Excitons and Free Carriers"

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Introduction

- Coulomb attraction
- Free exciton binding energies, radius
- Absorption process and spectra
- Electric fields
- Increasing carrier densities
- Frenkel excitons
- Applications



Coulomb Attraction

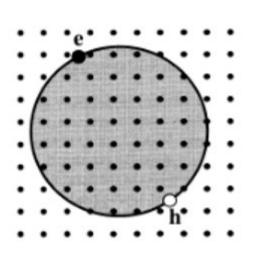
Absorption of a photon by an Interband transition

Electron and hole created in same spatial location

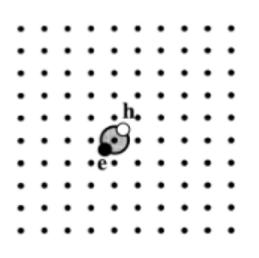
Oppositely charged particles – mutual attraction

Attractive interaction increases transition rate

May form a bound electron-hole pair analogous to hydrogen atom – an exciton



Free Exciton – Wannier-Mott



Bound Exciton – Frenkel



Free Exciton Binding Energy

$$E_H(n) = -\frac{m_0 e^4}{8\epsilon_0^2 h^3 c} \frac{1}{n^2} = -\frac{R_H}{n^2}$$

Apply Bohr model to exciton to determine energies of bound states.

$$E_x(n) = -\left(\frac{\mu}{m_0}\frac{1}{\epsilon_r^2}\right)\frac{R_H}{n^2} = -\frac{R_x}{n^2}$$
 Modified due to e-h pair having reduced effective mass, and being in high dielectric constant material

$$\frac{1}{\mu} = \left(\frac{1}{m_e^*} + \frac{1}{m_h^*}\right)$$

$$R_{x} = -\left(\frac{\mu}{m_{0}\epsilon_{r}^{2}}\right)R_{H}$$

high dielectric constant material

Bound states characterised by principal quantum number n

 μ/m_0 – reduced electron-hole mass R_H – Rydberg energy of Hydrogen atom (13.6eV)

 R_x – exciton Rydberg energy ϵ_r – relative permittivity



Free Exciton Radius

 a_x – exciton Bohr radius

$$r_H(n) = \frac{4\pi\epsilon_0\hbar^2}{m_0e^2}n^2$$

$$r_{\chi}(n) = \left(\frac{m_0}{\mu} \epsilon_r\right) n^2 a_H = n^2 a_{\chi}$$

$$r_n$$
 - radius of electron-hole orbit a_H - Bohr radius of hydrogen atom $(a_H = 5.29 \times 10^{-11} m)$

$$E_x(n) = -\frac{\mu}{m_0} \frac{1}{\epsilon_r^2} \frac{R_H}{n^2} = -\frac{R_x}{n^2}$$

$$a_{x} = \frac{m_0}{\mu} \epsilon_r a_H$$

Ground-state, (n=1)
Largest binding energy
Smallest radius

Excited States (n>1)
Less strongly bound, larger radius



Typical Materials

General trend – as E_g increases, so Rydberg energy increases and a_χ decreases

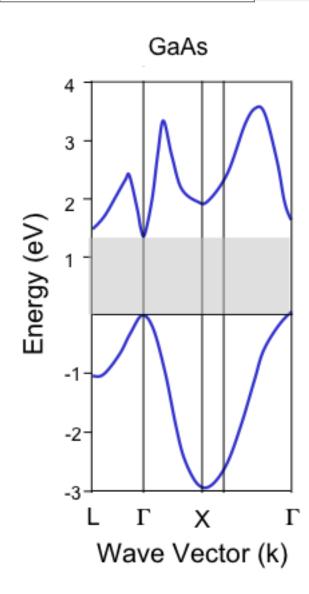
To have a stable exciton at room temperature – one requirement is that we need $R_x \sim kT$

Only possible in wider band-gap materials

Crystal	$E_{\mathbf{g}}$ (eV)	$R_{ m X} m (meV)$	$a_{ m X} m (nm)$
GaN	3.5	23	3.1
\mathbf{ZnSe}	2.8	20	4.5
CdS	2.6	28	2.7
\mathbf{ZnTe}	2.4	13	5.5
CdSe	1.8	15	5.4
CdTe	1.6	12	6.7
GaAs	1.5	4.2	13
InP	1.4	4.8	12
GaSb	0.8	2.0	23
InSb	0.2	(0.4)	(100)



Interband Absorption (excitons)



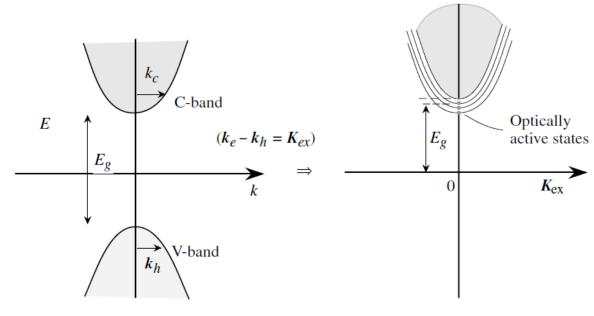
Additional requirement is that electron and hole group velocities are the same

$$v_g = \frac{1}{\hbar} \frac{\partial E}{\partial k}$$

Only occurs at Γ -point, where $v_g=0$ for both

All other k:

electron and hole have opposite group velocities



Independent electron picture + e-h interact ⇒ Excitonic bandstructure



Absorption Spectra - GaAs

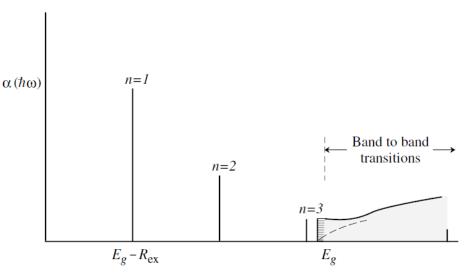
$$E(n) = E_{gap} - \frac{R_{\chi}}{n^2}$$

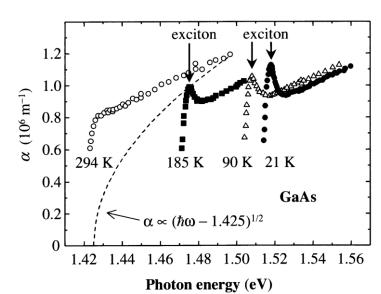
Excitonic peaks are <u>strong/sharp</u> lines in the absorption spectrum <u>below</u> the bandgap energy

Whilst the excitonic absorption peak is lost if $kT \gg R_x$,

the absorption is still modified *even* at Room Temperature

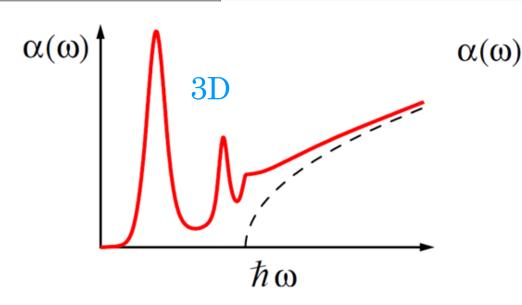
- 1) Reduction in the energy of onset of absorption
- Higher Absorption predicted than by the 3D Density of States

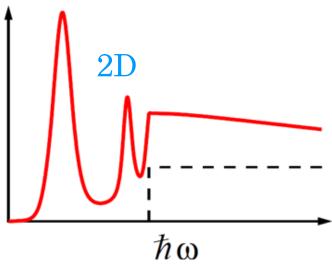






Enhancement above the bandgap

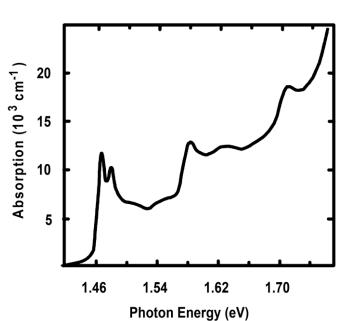




The absorption is enhanced relative to that predicted by the Density Of States when excitonic effects are taken into account in both 3D and 2D

Due to electron-hole correlations in unbound states (Sommerfield factor)

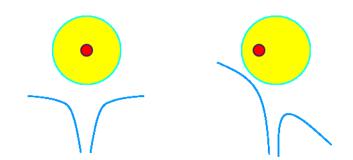
(Right) Absorption spectrum of a GaAs/AlGaAs Quantum Well at *Room Temperature*





Electric Field - Field Ionisation

Electric field accelerates electron and hole in opposite directions dissociating the exciton



Ionisation field is approximated as

$$E_{e-h} = \frac{2R_{\chi}}{ea_{\chi}} \approx 6 \times 10^5 Vm^{-1}$$

Field ionisation occurs when $E > E_{e-h}$

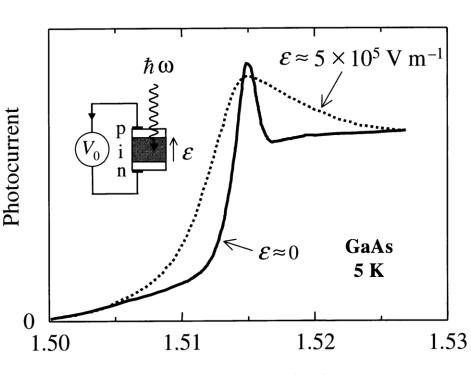
$$E = \frac{|V_{bi} - V_0|}{l_i}$$

Eg.

$$V_{bi} \approx 1.5V, l_i \approx 1\mu m$$

 $\Rightarrow E = 1.5 \times 10^6 V m^{-1}$

It occurs at modest E-fields...



Photon energy (eV)



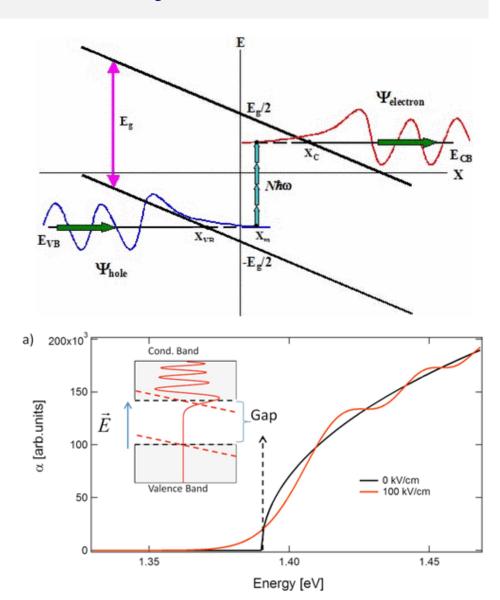
Franz-Keldysh Effect

At higher E-fields....tunnelling

Electron and hole wavefunction take on Airy wavefunction character and "leak" into the band-gap

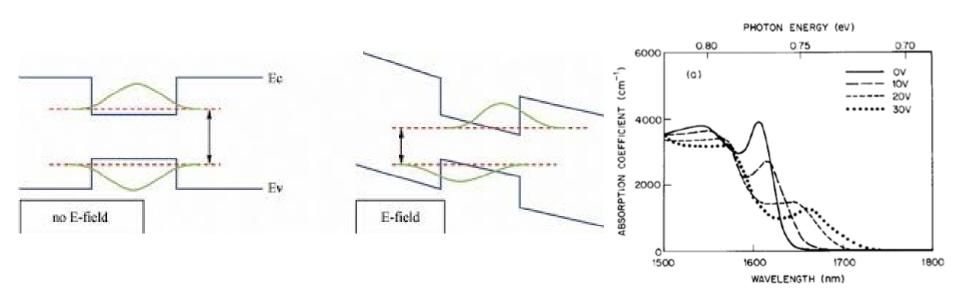
Observe:

Absorption below the band-gap and Oscillations above the band-gap





Quantum Confined Stark Effect



Field causes a decrease in the transition energy

The Quantum Well confines the exciton so it cannot ionize as readily

Reducing wavefunction overlap with increasing bias, absorption becomes weaker

Application as a modulator for optical communications



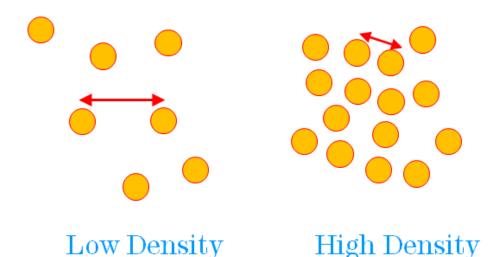
Mott Transition

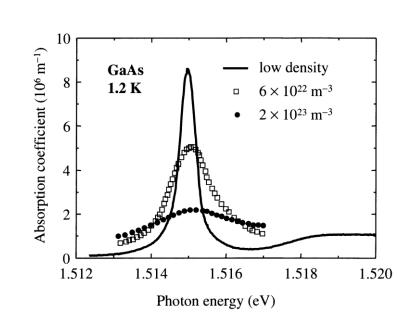
The interaction of excitons can lead to additional effects – new effects depend upon the ratio of R_x and kT

Excitons interact with each other when the number (1 / exciton volume) → Mott Density

$$N_{Mott} \approx \frac{1}{\frac{4}{3}\pi r_n^3}$$

- Creation of electron hole plasmas
 → Loss of excitonic effects
- Biexcitons....
- Bose-Einstein condensates







Higher Carrier Densities

If the electron and/or hole densities are high their own Coulomb potentials may begin to screen out the atomic potential

The weakening of the atomic potential will tend to reduce the band gap (c.f. effect of increasing the inter-atomic spacing)

Band gap shrinkage

$$\Delta E_g = -cN^{1/3}$$

For bulk GaAs

$$c \approx 32 meV/(10^{18} cm^{-3})^{1/3}$$



Moss-Burstein Shift

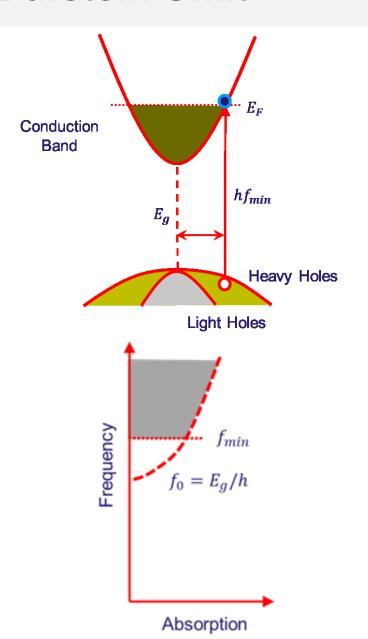
Degenerate semiconductors (where E_F lies in the conduction or valence band)

Observe a shift in the onset of absorption due to the filling of states

Blocking of the absorption transitions due to the Pauli exclusion principle

$$hf_{min} = E_g + \frac{\hbar^2 k_F^2}{2} \left(\frac{1}{m_c} + \frac{1}{m_v} \right)$$
$$= E_g + E_F \left(1 + \frac{m_c}{m_v} \right)$$

$$E_F = \frac{\hbar^2 k_F^2}{2m_c} = \frac{\hbar^2}{2m_c} (3\pi^2 n)^{2/3}$$





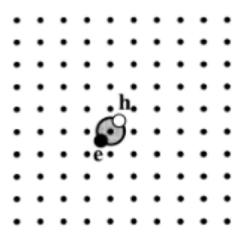
Frenkel Excitons

Observed in large bandgap materials with small dielectric constants and large effective masses

Predicted excitonic radius is comparable to the atomic spacing

→ Large binding energies

Bohr model breaks down



Frenkel exciton is akin to an excited state of the atom which "hops" from atom to atom

Observed in rare gas crystals, alkali halides and some organic crystals

Not covered further



Applications

Bulk excitonic effects

Limited to modification of absorption spectra in photo-diodes

Quantum Well structures

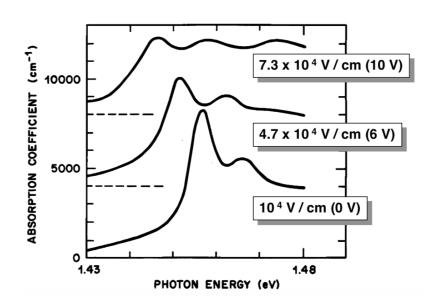
Strong/sharp absorption possible at Room Temperature

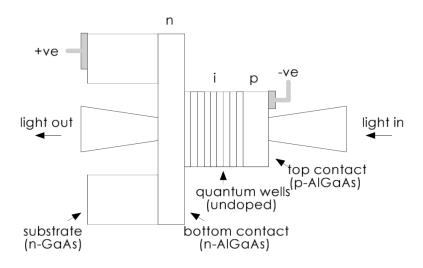
Modulators

Direct absorption

Mach-Zehnder

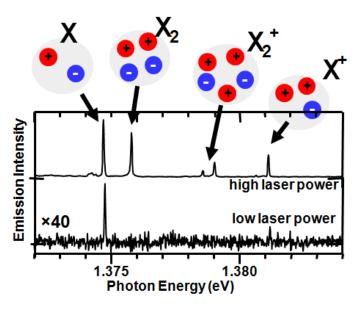
Lots of quantum information processing, quantum optics and non-linear optics applications

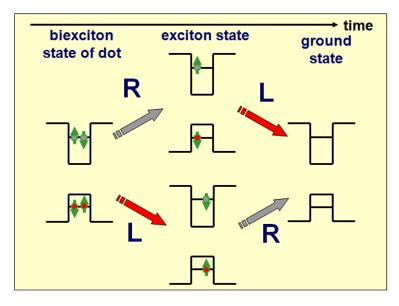






Excitons in Quantum Dots





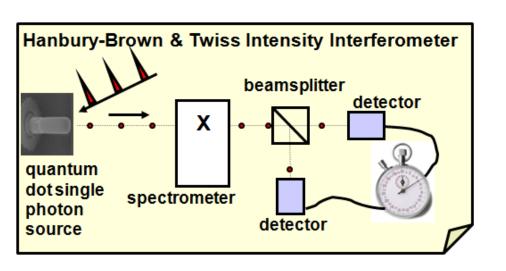
In Quantum Dots it is easy to achieve higher order excitonic states as they are small in size

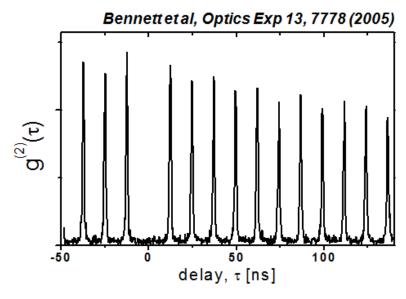
Singe quantum dots are ideal building blocks for Single Photon Emitters for use in Quantum Cryptography / Quantum Optics / Quantum Teleportation

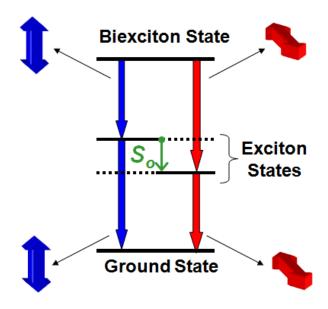
Biexciton → Exciton decay path is the prime candidate, if the dot is symmetric we cannot tell which e-h pair recombined first → an entangled state



Single Photon Emission







If the source emits only a single photon you cannot have a "click" on both detectors at once... excellent for secure communications

If the QD is not symmetric the exciton states are split in energy and we can tell from the polarisation "which path" was taken → destroys *entanglement*



Summary

- Discussed Coulomb effects which give rise to excitons
- Explored Bohr treatment of free excitons
- Looked at absorption process, spectra and effect of electric field
- Discussed effects of high exciton and free carrier density
- Touched upon Frenkel excitons
- Discussed some applications of these effects

