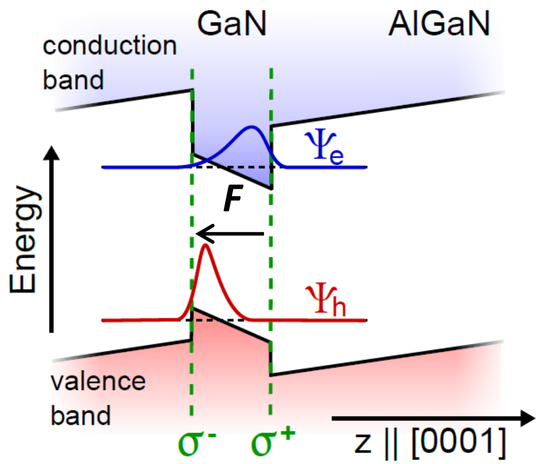
Lecture 4 – 12/03/2025

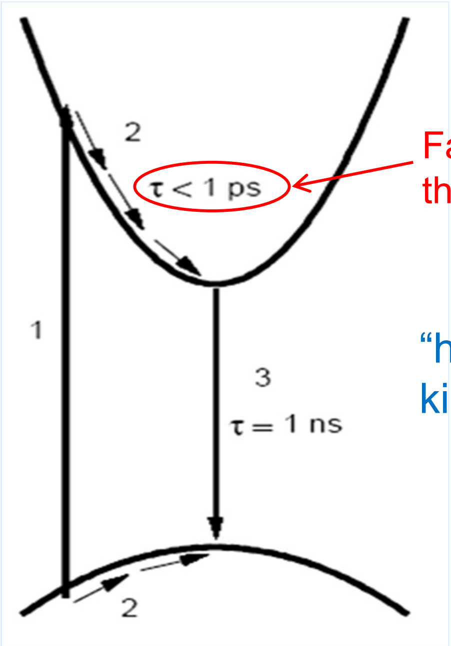
Quantum nanostructures

- Spontaneous emission: bulk vs quantum wells
- Quantum confined Stark effect



Spontaneous emission

Photoluminescence



Fast intraband relaxation time due to the efficient electron-phonon interaction

“hot” carriers (electrons and holes) release their kinetic energy *via* LO and acoustic phonon emission

Electron-photon interaction

Spontaneous emission in a two-level system (Fermi's golden rule)

Dipolar matrix element

$$P_{sp} = q^2 \frac{\pi\omega}{\varepsilon_0 L^3} \left| \langle 2 | \boldsymbol{\varepsilon}_l \hat{\mathbf{r}} | 1 \rangle \right|^2 \delta(\hbar\omega = E_2 - E_1)$$

Further details are given in Lecture 14 of the fall semester

$\boldsymbol{\varepsilon}_l$ *unit polarization vector of the electric field*

The **spontaneous emission rate** is calculated over all the cavity modes

$$\Gamma_{sp} = \iiint P_{sp} \mathrm{d}^3\mathbf{k}$$

+ substitute $\varepsilon_0 \rightarrow \varepsilon_r \varepsilon_0$

Section 3.6 Rosencher-Vinter

$$\Gamma_{sp} = \frac{q^2 r_{12}^2 \omega^3 n_{op}}{3\pi c^3 \hbar \varepsilon_0} = 1/\tau_{sp}$$

τ_{sp} *radiative lifetime*

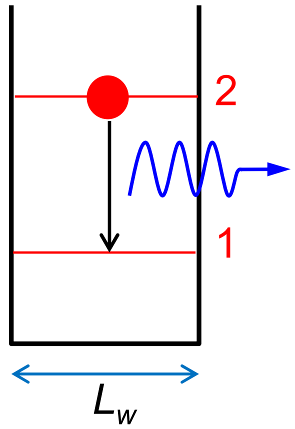
First derivation: Weisskopf-Wigner theory 1930

V. Weisskopf and E. Wigner, Z. Phys. **63**, 54 (1930) (> 1570 citations)

Spontaneous emission: case of discrete levels

Application:

Recombination lifetime in a quantum well with infinite barriers



1) Energy level

$$E_n = n^2 \frac{\hbar^2 \pi^2}{2m^* L_w^2}$$

Then,

$$\hbar\omega_{12} = E_2 - E_1 = 3\frac{\hbar^2 \pi^2}{2m^* L_w^2}$$

2) Dipole element r_{12}

$$r_{12} = \langle \Psi_2(x) | x | \Psi_1(x) \rangle \quad \text{with} \quad \Psi_1(x) = \sqrt{\frac{2}{L_w}} \cos(\pi x / L_w) \quad \text{and} \quad \Psi_2(x) = \sqrt{\frac{2}{L_w}} \sin(2\pi x / L_w)$$

Finally, $r_{12} = \frac{2^4 L_w}{3^2 \pi^2}$ and $r_{12}^2 = \frac{2^7 \hbar^2}{3^3 \pi^2 m^*} 1 / E_{12}$

Spontaneous emission: case of discrete levels

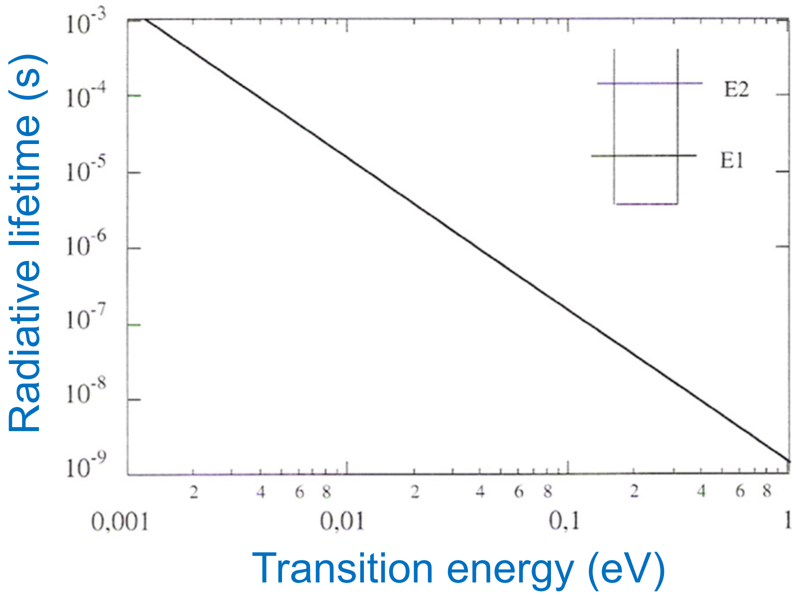
Radiative lifetime:

$$1/\tau_{sp} = \frac{q^2 r_{12}^2 \omega^3 n_{op}}{3\pi c^3 \hbar \varepsilon_0}$$

Optical refractive index

Dipolar matrix element

$$\tau_{sp} = \frac{3^4 \pi^3 m^* c^3 \hbar^2 \varepsilon_0}{2^7 q^2 n_{op}} \frac{1}{E_{12}^2}$$

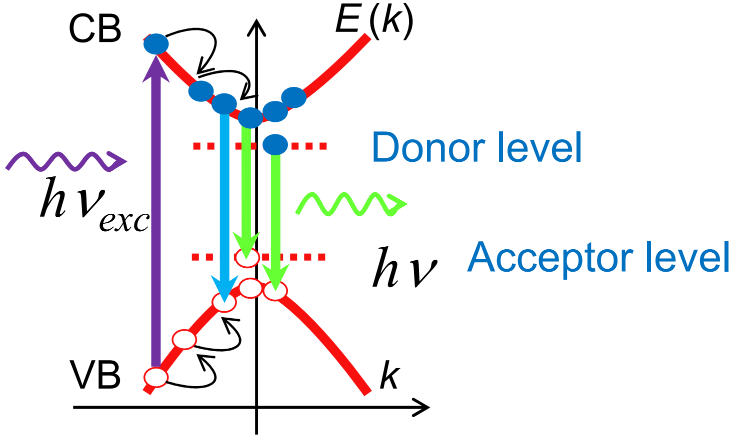


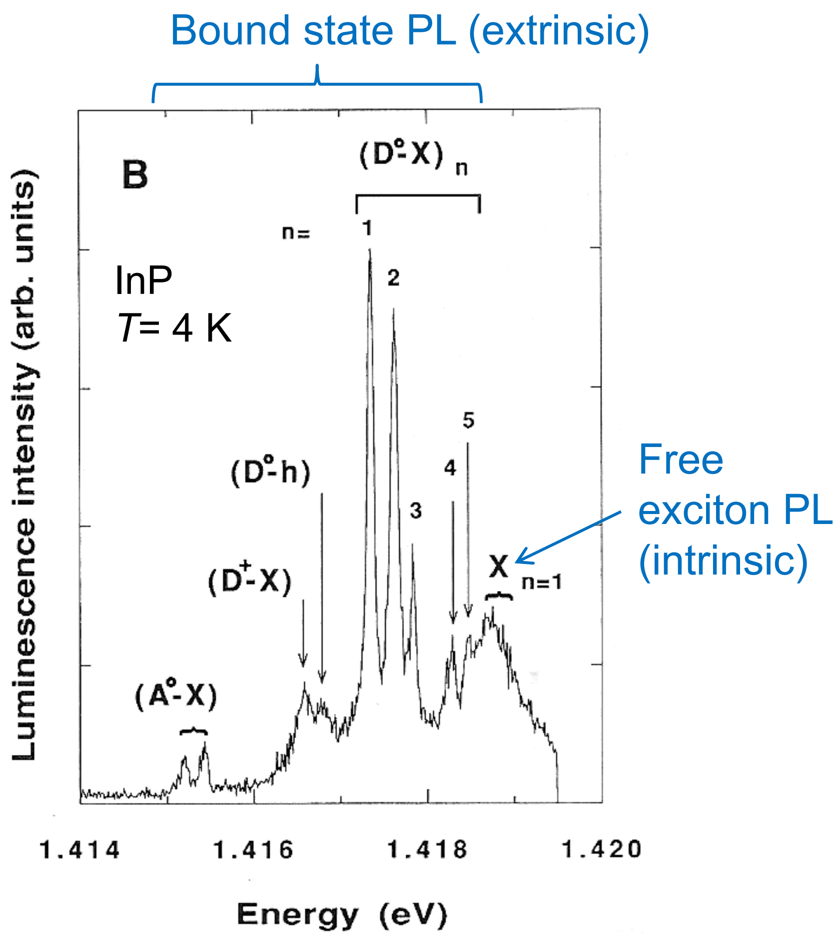
Transition energies > 1 eV
⇒ radiative lifetime ~1 ns

Intersubband optical transitions are expected to be slower than interband ones!

Spontaneous emission

Photoluminescence (case of bulk InP)

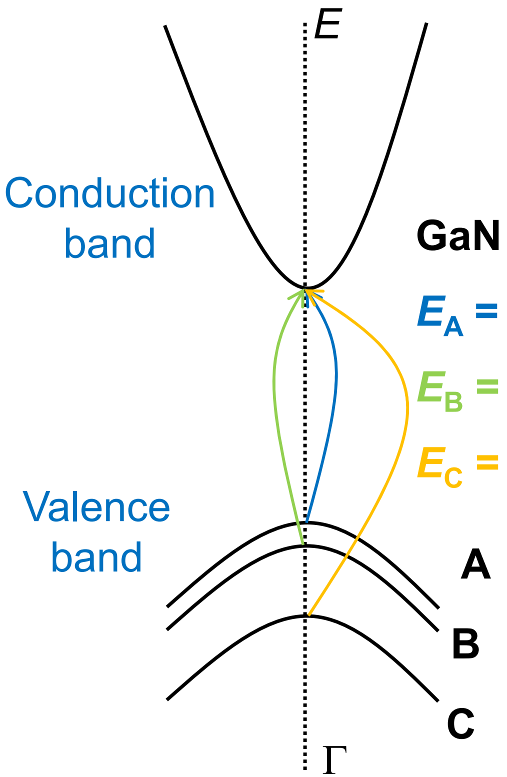




⇒ *How can we discriminate intrinsic PL from extrinsic one?*

Spontaneous emission bulk samples

Photoluminescence and reflectivity (case of GaN)



GaN (10 K):

E_A = 3.478 eV

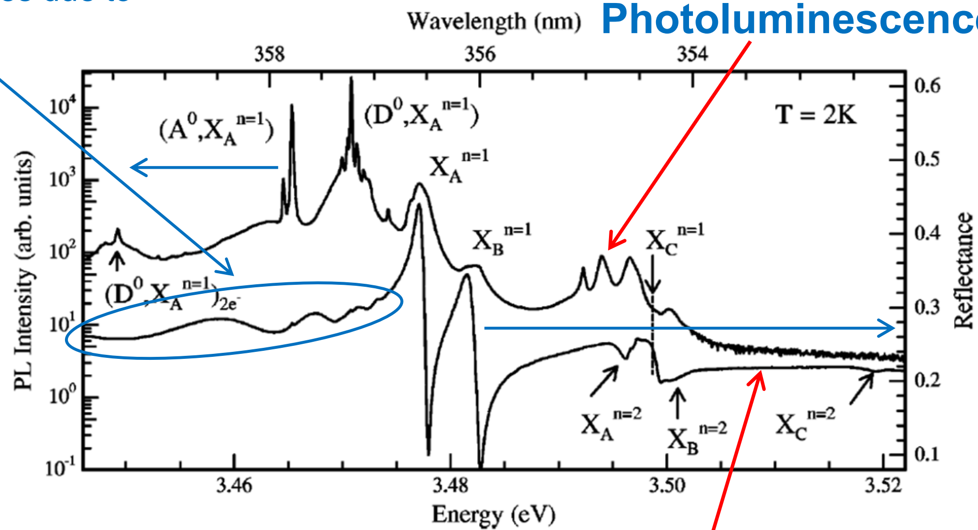
E_B = 3.485 eV

E_C = 3.503 eV

Expected values in relaxed (strain-free) bulk GaN

Interference fringes due to cavity effect

Photoluminescence



Reflectivity (∝ JDOS, i.e., *A* or f_{osc})

$R + T + A = 1$

In thick samples $R \cong 1\text{-}A$

Exciton binding energy can be determined from reflectivity spectra!

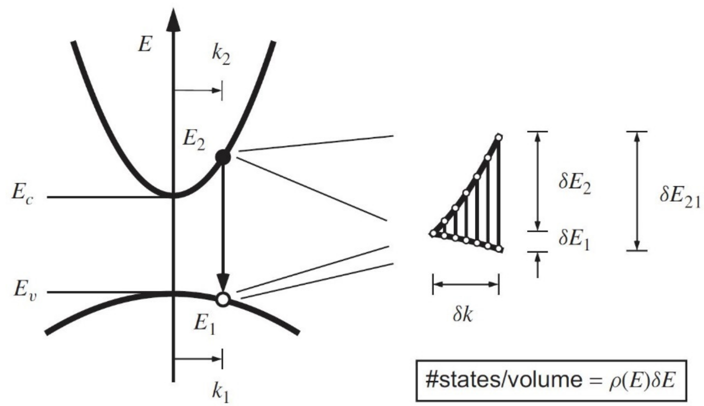
Joint density of states in semiconductors

$\rho_j(\omega)$ **Joint/reduced density of states** (3D case) $\Rightarrow$ the number of transition pairs within δk is equal to the number of states in either the conduction or valence band

$$\rho_j(\omega) = \frac{1}{2\pi^2}\left(\frac{2m_\mathrm{r}}{\hbar}\right)^{3/2}(\omega - E_\mathrm{g}/\hbar)^{1/2}$$

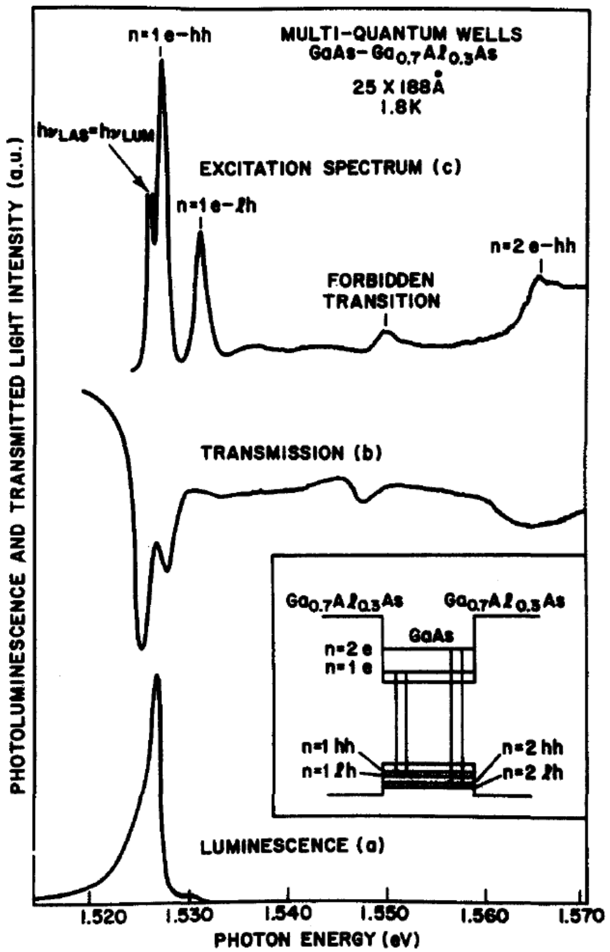
with $\frac{1}{m_\mathrm{r}} = \frac{1}{m_\mathrm{e}} + \frac{1}{m_\mathrm{h}}$ *and m_r is the reduced mass*

Cf. chapter 4 Coldren-Corzine



Spontaneous emission quantum wells

Photoluminescence and transmission spectra (GaAs/AlGaAs QWs)



- Narrow PL linewidth (FWHM) at low temperature

- Disappearance of optical transitions due to bound excitations ascribed to:
  - smearing of the related luminescence bands (spread in the impurity binding energy due to confinement)[1]
  - change in the symmetry of the impurity ground-state wavefunction ⇒ reduced oscillator strength

⇒ QW PL spectra dominated by instrinsic radiative transitions in high quality QWs

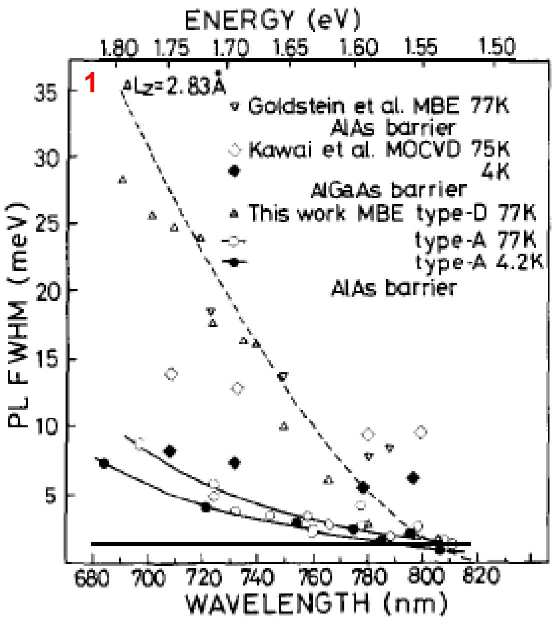
PL spectra of QW heterostructures are usually much simpler than their bulk counterparts

[1] C. Weisbuch *et al.*, Solid State Commun. **37**, 219 (1981) (> 310 citations).

Excitons in quantum wells: role of interfaces

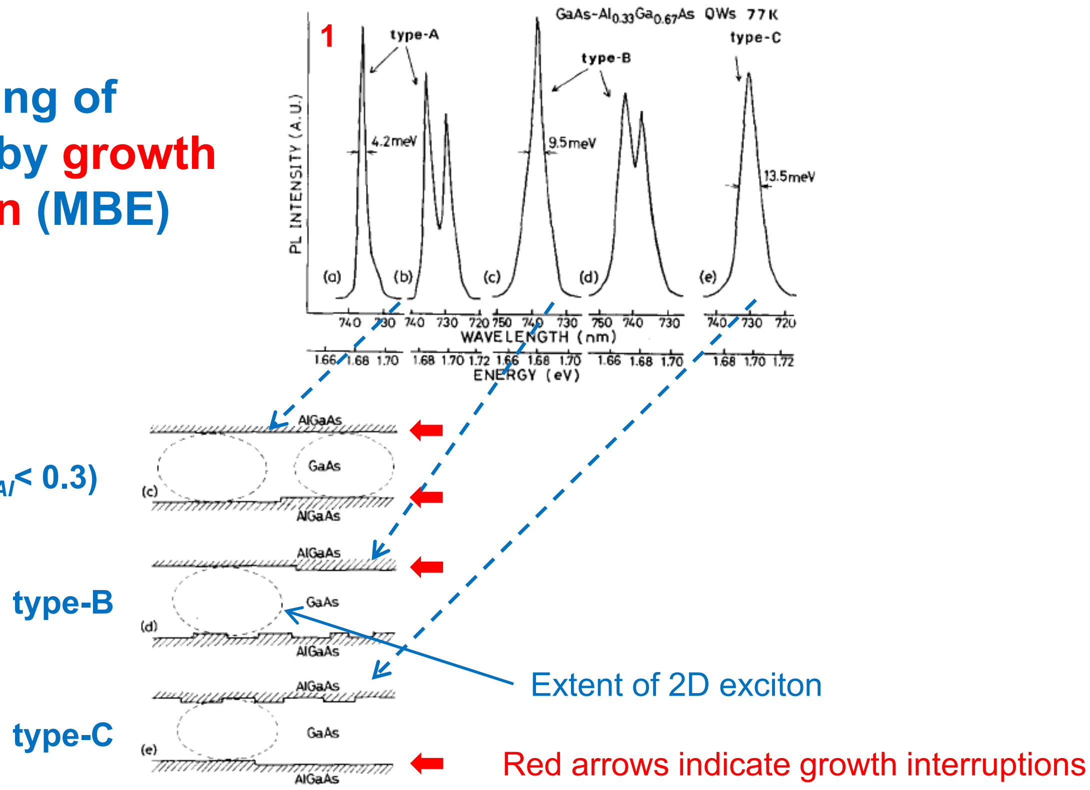
Various types of atomic-scale heterointerfaces lead to different PL lineshapes → impact of roughness and step intervals on excitons

← Decreasing well width



QW thickness fluctuations responsible for inhomogeneous broadening (≡ extrinsic broadening due to disorder)

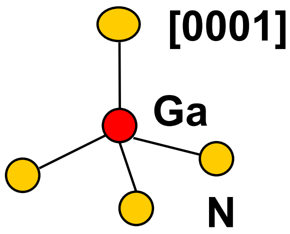
Smoothening of interfaces by growth interruption (MBE)



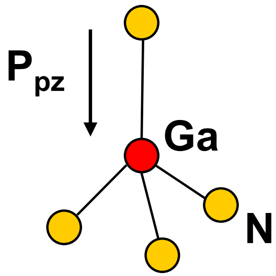
[1]M. Tanaka and H. Sakaki, J. Cryst. Growth **81**, 153 (1987) (> 160 citations).

Quantum confined Stark effect (QCSE)

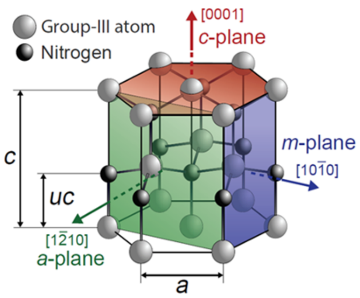
Piezoelectric (cubic & wurtzite)



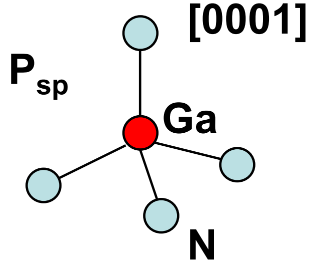
Equilibrium $P_{pz} = 0$



Strain (ϵ) $P_{pz} \neq 0$



Spontaneous (wurtzite)



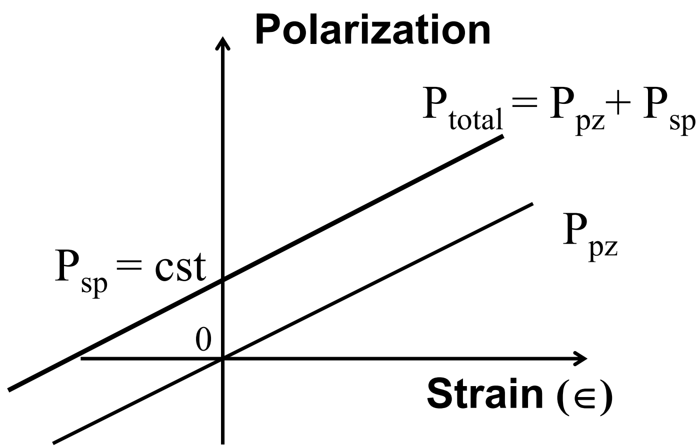
Equilibrium $P_{sp} \neq 0$

Keep in mind that polarization is a bulk vectorial quantity!

Macroscopic polarization:

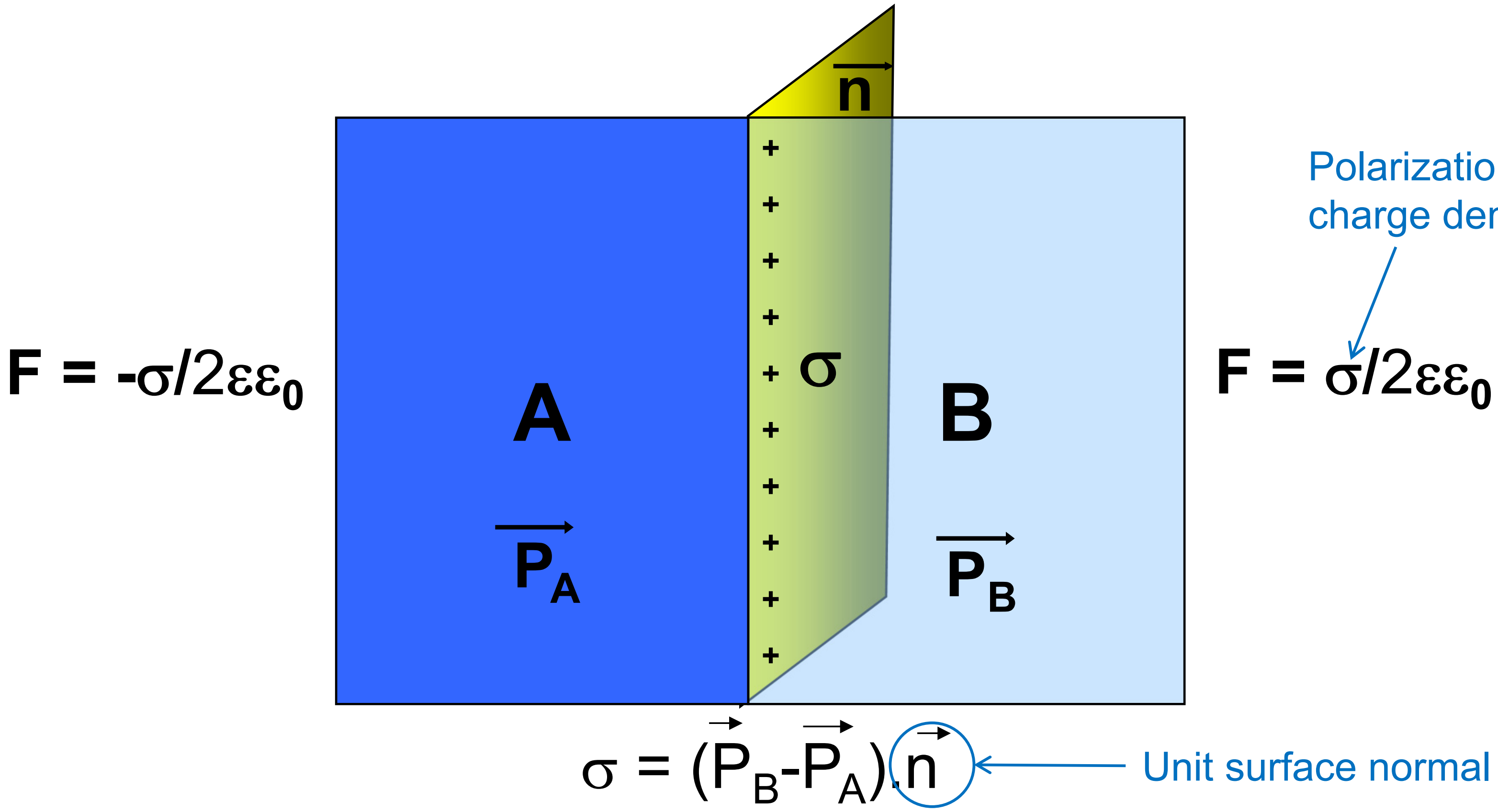
Difference between the barycenters of positive and negative charges

$$P_{total} = P_{pz} + P_{sp} \text{ // } c\text{-axis}$$



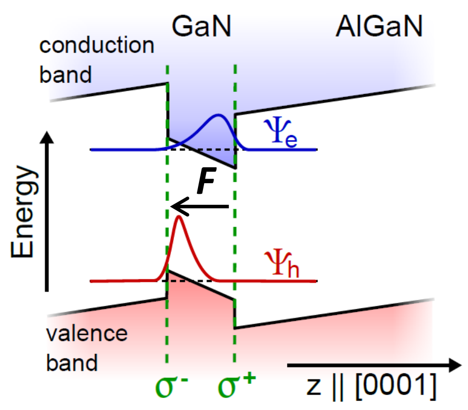
Quantum confined Stark effect

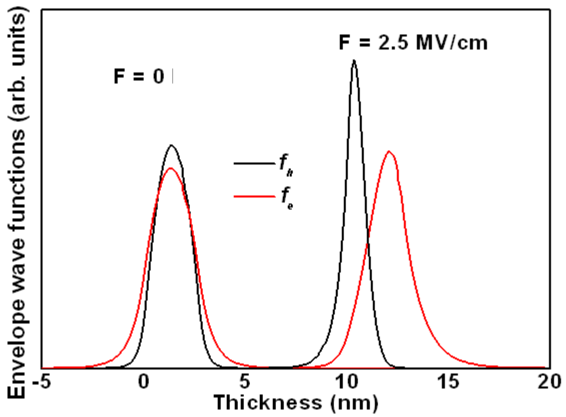
Electric field created by a surface charge



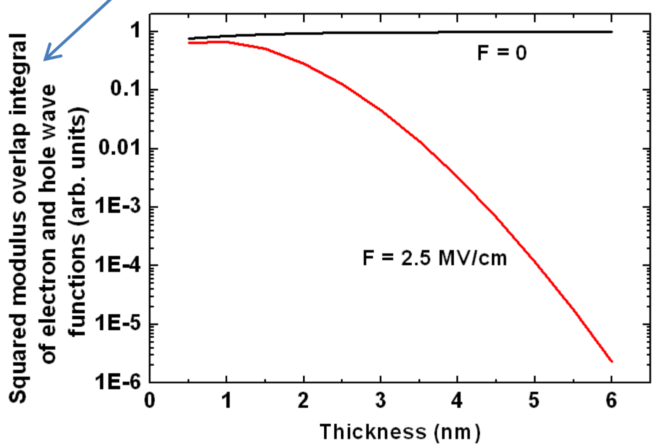
Spontaneous and piezoelectric polarization discontinuities at heterointerfaces

Quantum confined Stark effect





Quantity ∝ oscillator strength



Triangular shape of confining potential, QCSE ⇒

(1) redshift of fundamental optical transition

(2) spatial separation of electron and hole wavefunctions

$$\tau_{rad} = \frac{2\pi\varepsilon_0 m_0 c^3}{n_{op} e^2 \omega_{CV}^2 f_{osc}}$$

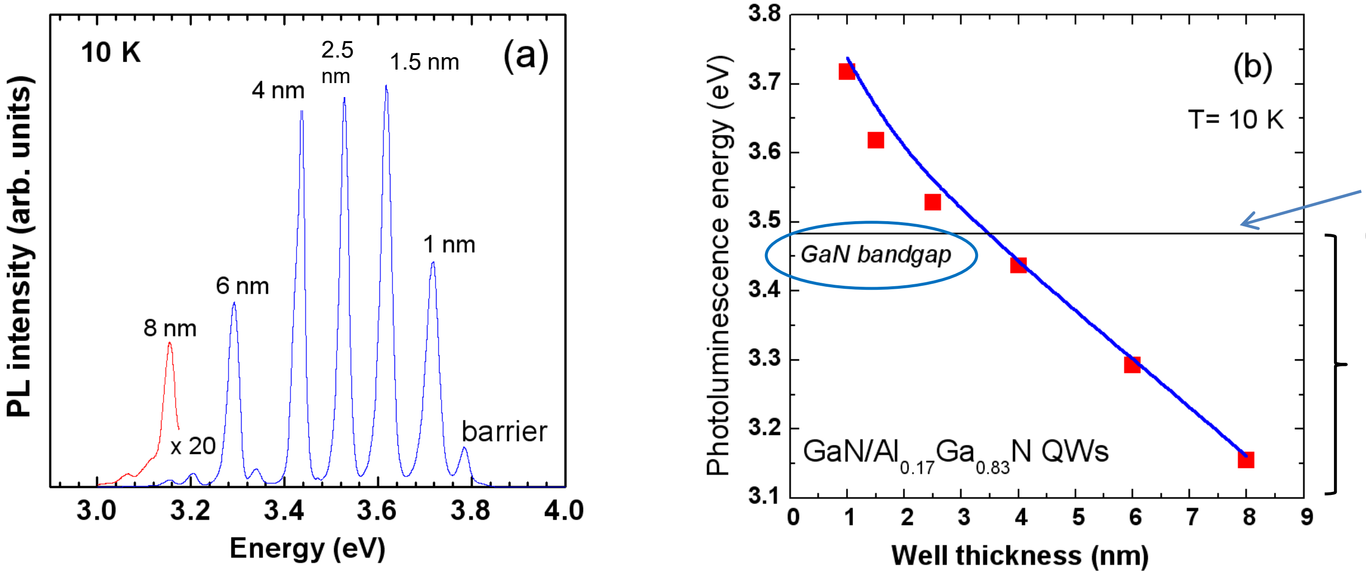
(3) decrease in oscillator strength of optical transition and exciton binding energy

and

(4) increase in radiative lifetime[1]

[1]J. Feldmann *et al.*, Phys. Rev. Lett. **59**, 2337 (1987) (> 1010 citations).

QCSE: experimental facts[1]



Single GaN/AlGaN QWs

Rectangular quantum well limit (≡ bulk SC bandgap)

Decrease in f_{osc} + increase in τ_{rad}

Bulk Bohr radius of GaN ~3 nm

Stark shift due to permanent dipole

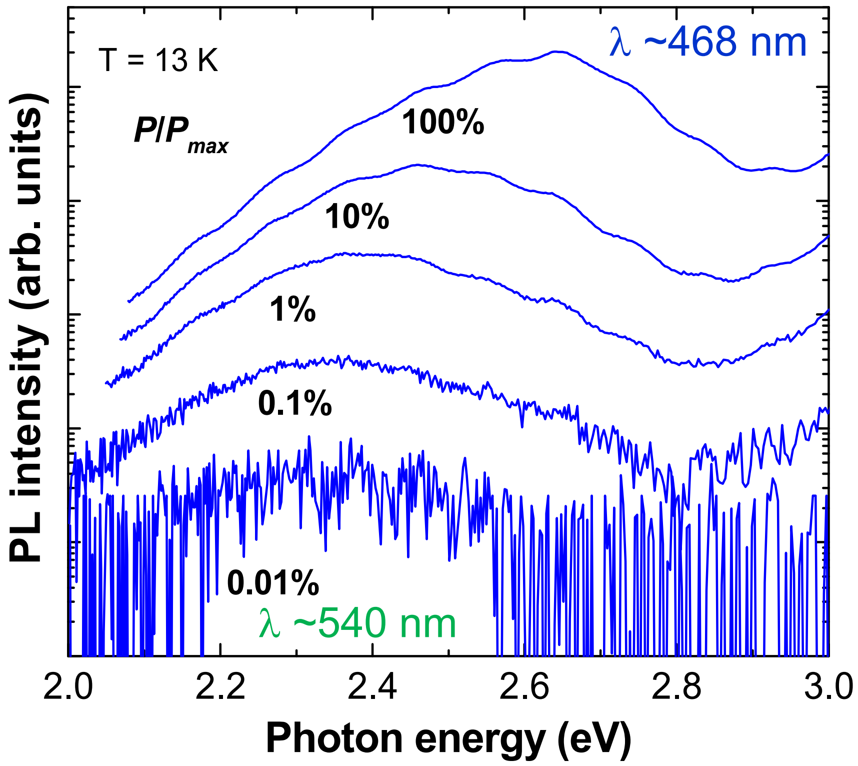
$$E_{e_1-hh_1} = E_{g,W} + e_1 + hh_1 - E_B - eFL_{QW}$$

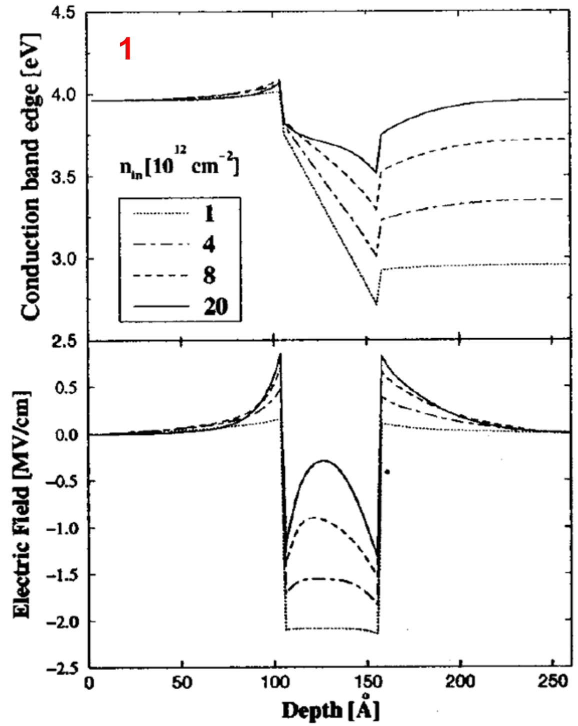
QW transition energies occurring below the bandgap of GaN ⇒ QCSE

[1] N. Grandjean *et al.*, J. Appl. Phys. **86**, 3714 (1999) (> 260 citations).

Electric field screening

Electric field screening by photo-generated free carriers



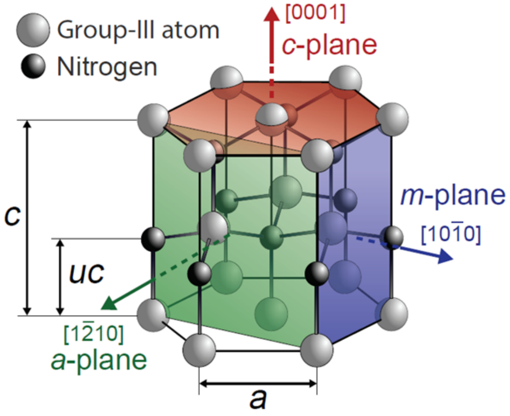


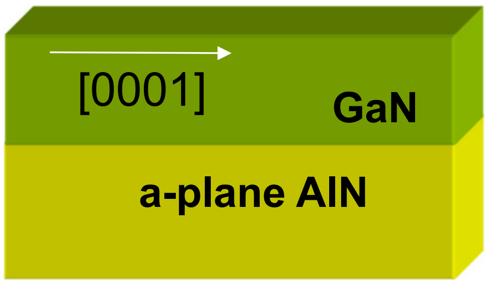
Self-consistent Schrödinger-Poisson calculations

$$\frac{d}{dz}\left[\varepsilon_0\varepsilon_r\frac{d}{dz}V(z)\right]=-e\left[-n_{3D}(z)+p_{3D}(z)-n_{2D,i}(z)\right]$$

[1]V. Fiorentini *et al.*, Phys. Rev. B **60**, 8849 (1999) (> 500 citations).

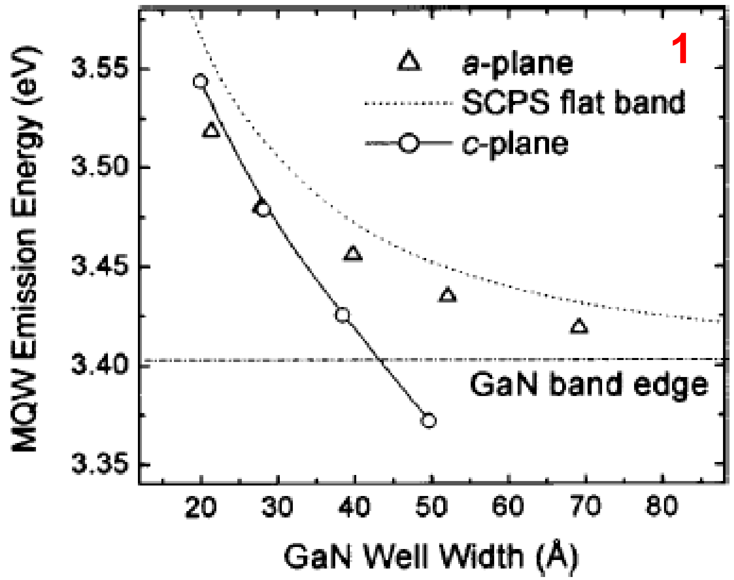
Non-polar quantum wells





Growth on non-polar substrates ⇒ macroscopic polarization // QW plane ⇒ no fixed charge plane

GaN/AlGaN QWs



[1]Craven *et al.*, APL **84**, 496 (2004) (> 120 citations).

⇒ no built-in polarization field in QWs

The transverse Stark effect in 2D systems[1]

Total energy for a carrier External *E*-field // QW plane

$$E = E_x + E_y + E_z = \frac{p_x^2}{2m^*} + eFx + \frac{p_y^2}{2m^*} + \frac{p_z^2}{2m^*} + V_b H\left(z^2 - \frac{L^2}{4}\right)$$

Allowed classical motion such that $p_x^2 \geq 0$

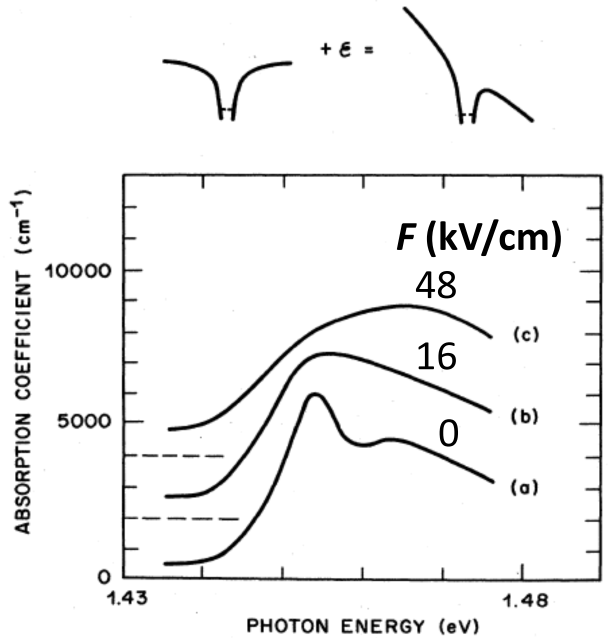
$\Rightarrow x$ motion limited to $x < \frac{1}{eF} E_x$

For an exciton in a perfect 2D system, at $F = 0$ the ground state is bound by $4Ry^*$. If the potential energy difference over one Bohr radius ($0.5eFa_B$) ~ $4Ry^*$, the relative motion becomes unbound on the negative *x* side (⇔ critical field in bulk materials that ionizes the exciton (Franz-Keldysh effect $F_c \sim \frac{Ry^*}{ea_B}$)).

Quasi-2D excitons more tightly bound than bulk ones

$\Rightarrow F_c$ increased by a factor 2-4!

GaAs/$Al_{0.32}Ga_{0.68}As$ MQWs

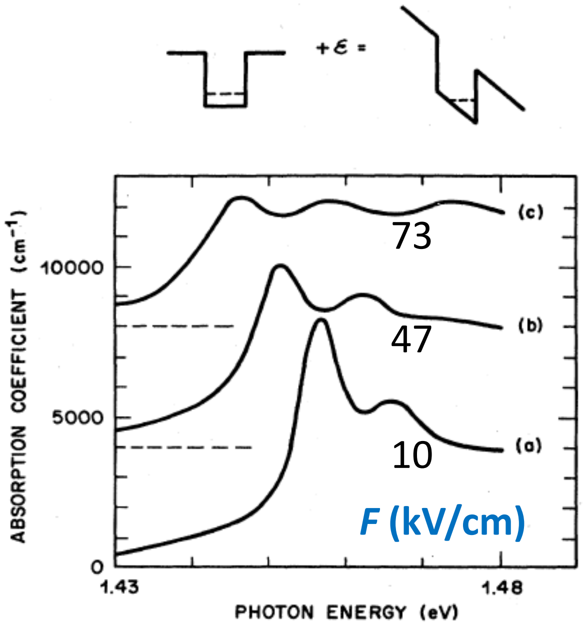


[1]D. A. B. Miller *et al.*, Phys. Rev. B **32**, 1043 (1985) (> 1830 citations).

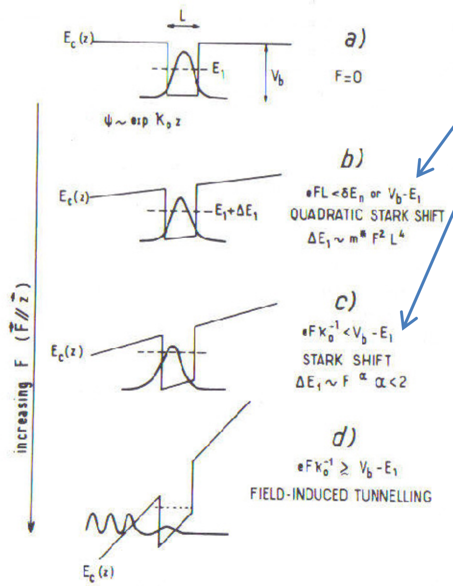
QCSE in non-polar quantum wells[1]

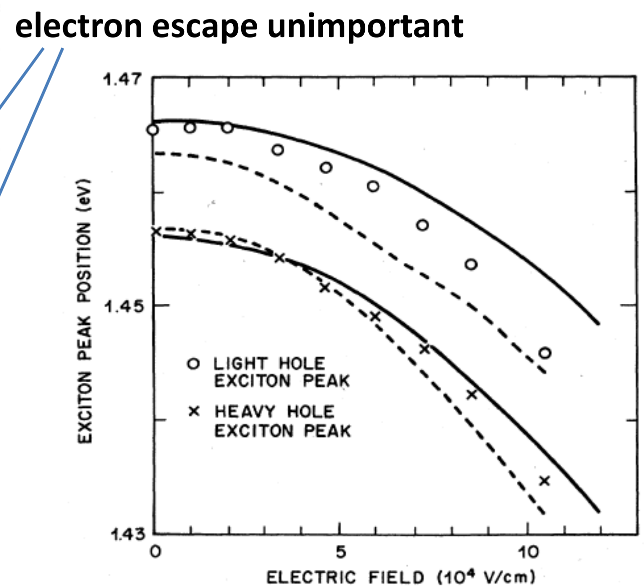
Appearance of a finite dipole between electron and hole $D = |e|(z_h - z_e)$ (can be > L_{QW})
Exciton dissociation considerably hindered by CB and VB potential barriers

GaAs/$Al_{0.32}Ga_{0.68}$As MQWs in a *p-i-n* junction



External *E*-field ⊥ QW plane





$(\kappa_0)^{-1}$: characteristic penetration length of the ground CB state in the barrier at zero field

[1]D. A. B. Miller *et al.*, Phys. Rev. B **32**, 1043 (1985) (> 1830 citations).