Theoretical seminar on optical properties of semiconductors

EXCITONS IN NANOSTRUCTURES

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CONTENT

I. Repetition

II. Excitons in quantum wells

III. Excitons in quantum wires

IV. Excitons in quantum dots

OPTICAL POLARISATION

Polarisation in second quantisation:

$$\mathbf{P}(t) = \int d^3r \, \langle \hat{\psi}^\dagger(\mathbf{r},t) \, e\mathbf{r} \, \hat{\psi}(\mathbf{r},t) \rangle$$

With the field operators in the Bloch functions basis: $\hat{\psi}(\mathbf{r},t) = \sum_{\lambda,\mathbf{k}} a_{\lambda,\mathbf{k}}(t) \psi_\lambda(\mathbf{k},\mathbf{r})$

$$\mathbf{P}(t) = \sum_{\lambda,\lambda',\mathbf{k},\mathbf{k}'} \langle a^\dagger_{\lambda,\mathbf{k}} a_{\lambda',\mathbf{k}'} \rangle \underbrace{\int d^3r \, \psi^*_{\lambda,\mathbf{k}}(\mathbf{r}) \, e\mathbf{r} \, \psi_{\lambda',\mathbf{k}'}(\mathbf{r})}_{\simeq \delta_{\mathbf{k},\mathbf{k}'} \mathbf{d}_{\lambda,\lambda'}} = \sum_{\lambda,\lambda',\mathbf{k}} \langle a^\dagger_{\lambda,\mathbf{k}} a_{\lambda',\mathbf{k}}(t) \rangle \, \mathbf{d}_{\lambda,\lambda'}$$

Pair function:

$$P_{\lambda\lambda',\mathbf{k},}(t) = \langle a^\dagger_{\lambda,\mathbf{k}} a_{\lambda',\mathbf{k}}(t) \rangle \to P_{vc,\mathbf{k},}(t) = \langle a^\dagger_{v,\mathbf{k}} a_{c,\mathbf{k}}(t) \rangle$$

$$\text{with } \lambda = v \,\&\, \lambda' = c$$

EQUATION OF MOTION FOR THE PAIR FUNCTION

Interaction with light in dipole approximation:

$$\mathcal{H}_I = \int d^3r\, \hat{\psi}^\dagger(\mathbf{r})(-e\mathbf{r})\cdot\mathcal{E}(\mathbf{r},t)\hat{\psi}(\mathbf{r}) \simeq -\sum_{\mathbf{k}} \mathcal{E}(t)(a^\dagger_{c,\mathbf{k}}a_{v,\mathbf{k}}d_{cv} + h.c.)$$

Electron Hamiltonian:

$$\mathcal{H}_{el} = \sum_{\lambda,\mathbf{k}} E_{\lambda,k}a^\dagger_{\lambda,\mathbf{k}}a_{\lambda,\mathbf{k}} + \frac{1}{2}\sum_{\substack{\mathbf{k},\mathbf{k}'\\ \mathbf{q}\neq\mathbf{0}\\ \lambda,\lambda'}} V_q a^\dagger_{\lambda,\mathbf{k}+\mathbf{q}}a^\dagger_{\lambda',\mathbf{k}'-\mathbf{q}}a_{\lambda',\mathbf{k}'}a_{\lambda,\mathbf{k}} \underset{\substack{\lambda = v\\ \lambda' = c}}{=} \sum_{\mathbf{k}}(E_{c,k}a^\dagger_{c,\mathbf{k}}a_{c,\mathbf{k}} + E_{v,k}a^\dagger_{v,\mathbf{k}}a_{v,\mathbf{k}})$$

$$+\frac{1}{2}\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}\neq\mathbf{0}} V_q\Big(a^\dagger_{c,\mathbf{k}+\mathbf{q}}a^\dagger_{c,\mathbf{k}'-\mathbf{q}}a_{c,\mathbf{k}'}a_{c,\mathbf{k}} + a^\dagger_{v,\mathbf{k}+\mathbf{q}}a^\dagger_{v,\mathbf{k}'-\mathbf{q}}a_{v,\mathbf{k}'}a_{v,\mathbf{k}} + 2a^\dagger_{c,\mathbf{k}+\mathbf{q}}a^\dagger_{v,\mathbf{k}'-\mathbf{q}}a_{v,\mathbf{k}'}a_{c,\mathbf{k}}\Big)$$

Dynamics of interband polarisation function (from Haisenberg equation of motion):

$$\boxed{\hbar\left[\boldsymbol{i}\frac{\boldsymbol{d}}{\boldsymbol{dt}} - (\boldsymbol{e}_{\boldsymbol{c},\boldsymbol{k}} - \boldsymbol{e}_{\boldsymbol{v},\boldsymbol{k}})\right]\boldsymbol{P}_{\boldsymbol{vc},\mathbf{k}}(\boldsymbol{t}) = \Big[\boldsymbol{n}_{\boldsymbol{c},\mathbf{k}}(\boldsymbol{t}) - \boldsymbol{n}_{\boldsymbol{v},\mathbf{k}}(\boldsymbol{t})\Big]\left[\boldsymbol{d}_{\boldsymbol{cv}}\boldsymbol{\mathcal{E}}(\boldsymbol{t}) + \sum_{\mathbf{q}\neq\mathbf{k}}\boldsymbol{V}_{|\mathbf{k}-\mathbf{q}|}\boldsymbol{P}_{\boldsymbol{vc},\mathbf{q}}\right]}$$

shifted frequencies

$n_{\lambda,\mathbf{k}} = \langle a^\dagger_{\lambda,\mathbf{k}}a_{\lambda,\mathbf{k}}\rangle$

„MASSAGING“ THE EQUATION FOR POLARISATION

Quasi-equilibrium: $n_{c,\mathbf{k}}(t) \to f_{c,k}$ $n_{v,\mathbf{k}}(t) \to f_{v,k}$

$$\hbar\left[i\frac{d}{dt} - (e_{c,k} - e_{v,k})\right] P_{vc,\mathbf{k}}(t) = \left(f_{c,\mathbf{k}} - f_{v,\mathbf{k}}\right)\left[d_{cv}\mathcal{E}(t) + \sum_{\mathbf{q}\neq\mathbf{k}} V_{|\mathbf{k}-\mathbf{q}|} P_{vc,\mathbf{q}}\right]$$

Recipe to solve:
make Fourier transform into frequency domain,
solve equation,
find the result by back transformation into time domain
(already seen in free carrier case)

Unexcited crystal: $f_{c,k} \equiv 0$ $f_{v,k} \equiv 1$

Fourier transform into frequency domain:

$$\left[\hbar\omega - E_g - \frac{\hbar^2 k^2}{2m_r}\right] P_{vc,\mathbf{k}}(\omega) = -\left[d_{cv}\mathcal{E}(\omega) + \sum_{\mathbf{q}\neq\mathbf{k}} V_{|\mathbf{k}-\mathbf{q}|} P_{vc,\mathbf{q}}(\omega)\right]$$

Fourier transform into real space:

$$\left[\hbar\omega - E_g + \frac{\hbar^2 \nabla_{\mathbf{r}}^2}{2m_r} + V(r)\right] P_{vc}(\mathbf{r},\omega) = -d_{cv}\mathcal{E}(\omega)\delta(\mathbf{r})L^3$$

WANNIER EQUATION IN 2D CASE

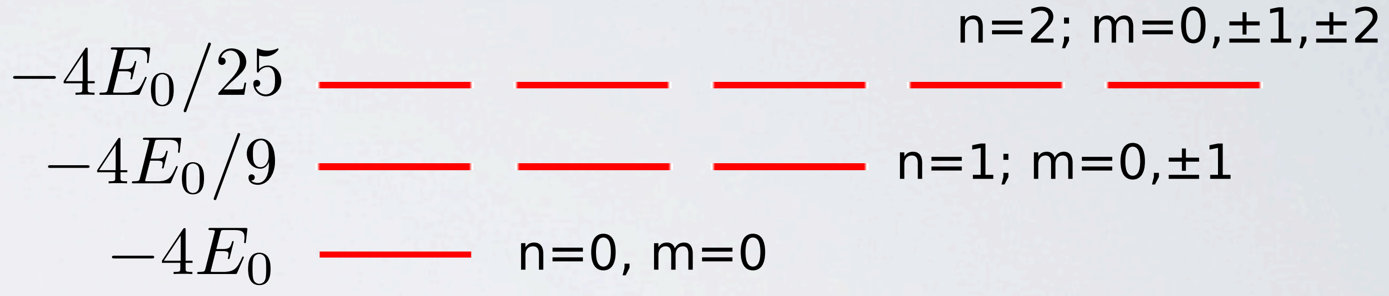
Solve first homogeneous equation:

$$-\left[\frac{\hbar^2\nabla_{\mathbf{r}}^2}{2m_r}+V(r)\right]\psi_\nu(\mathbf{r})=E_\nu\psi_\nu(\mathbf{r})$$

Wannier equation

2D exciton bound state energies:

$$E_n=-E_0\frac{1}{(n+1/2)^2}\quad\text{with } n=0,1,\ldots$$



2D exciton wave functions:

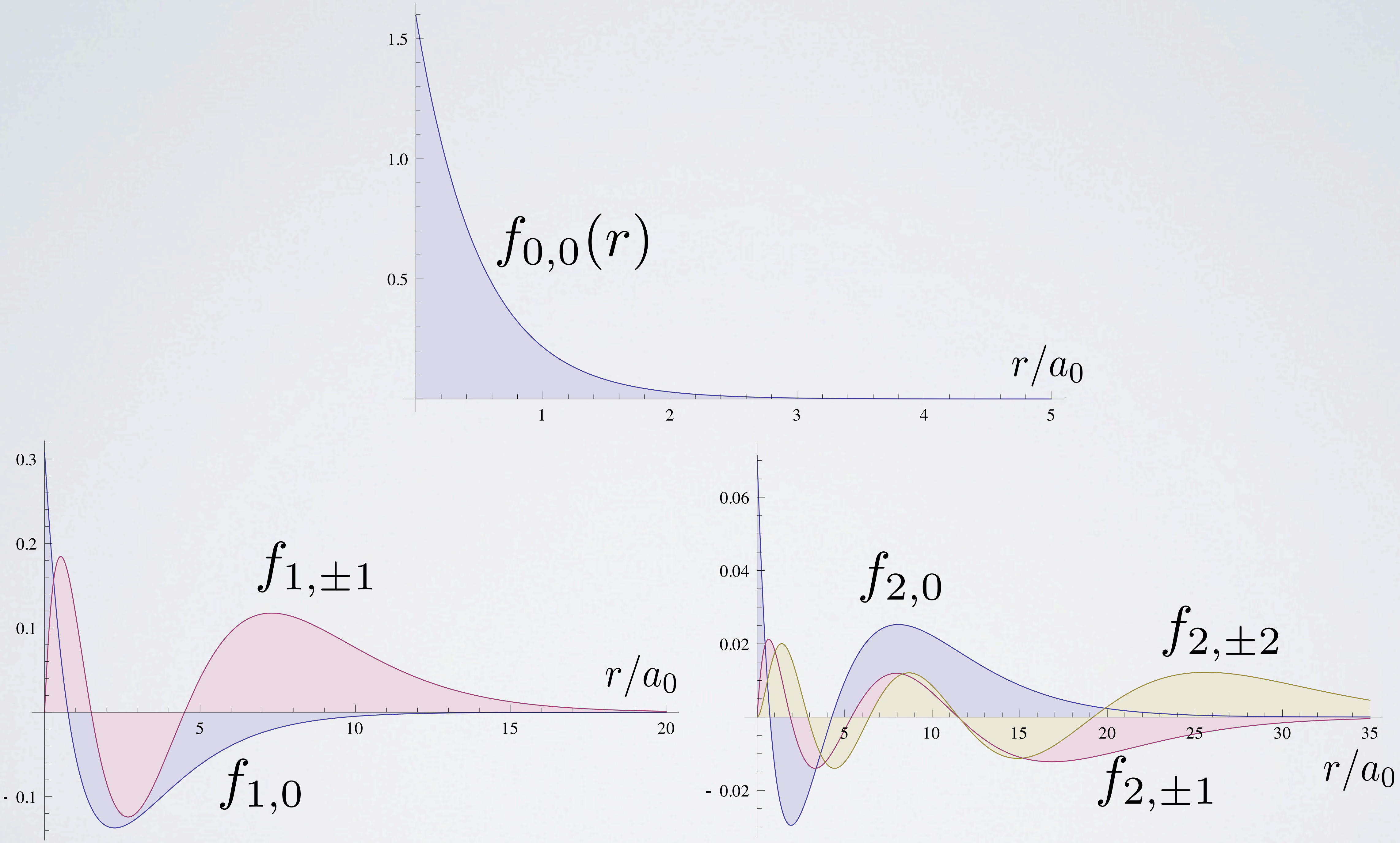
$$\psi_{n,m}(\mathbf{r})=\sqrt{\frac{1}{\pi a_0^2(n+\frac{1}{2})^3}\frac{(n-|m|)!}{[(n+|m|)!]^3}}\rho^{|m|}e^{-\frac{\rho}{2}}L_{n+|m|}^{2|m|}(\rho)\,e^{im\phi}$$

The normalised wave function for ionisation continuum in 2D:

$$\psi_{k,m}(\mathbf{r})=\frac{(i2kr)^{|m|}}{(2|m|)!}\sqrt{\frac{\pi k}{\mathcal{R}(1/4+|\lambda|^2)\cosh(\pi|\lambda|)}\prod_{j=0}^{|m|}\left[\left(j-\frac{1}{2}\right)^2+|\lambda|^2\right]}$$

$$\times\quad e^{\frac{\pi|\lambda|}{2}}e^{-ikr}F\left(|m|+\frac{1}{2}+i|\lambda|;\,2|m|+1;\,2ikr\right)\frac{e^{im\phi}}{\sqrt{2\pi}}$$

WAVE FUNCTIONS FOR EXCITONS IN 2D



OPTICAL SPECTRUM

To solve the inhomogeneous equation for the polarisation: $P_{vc}(\mathbf{r},\omega) = \sum_{\nu} b_\nu \, \psi_\nu(\mathbf{r})$

Put the ansatz into inhomogeneous equation for P, find coefficients b, make many Fourrier transforms:

$$P(\omega) = -2L^3 \sum_{\nu} |d_{cv}|^2 |\psi_\nu(\mathbf{r}=0)|^2 \mathcal{E}(\omega) \left[\frac{1}{\hbar\omega - E_g - E_\nu} - \frac{1}{\hbar\omega + E_g + E_\nu} \right]$$

Known relation:

$$\chi(\omega) = \frac{\mathcal{P}(\omega)}{\mathcal{E}(\omega)} = \frac{P(\omega)}{L^3 \mathcal{E}(\omega)}$$

Electron-Hole Pair susceptibility:

$$\chi(\omega) = -2 \sum_{\nu} |d_{cv}|^2 |\psi_\nu(\mathbf{r}=0)|^2 \left[\frac{1}{\hbar\omega - E_g - E_\nu} - \frac{1}{\hbar\omega + E_g + E_\nu} \right]$$

resonant

non-resonant

probability to find electron and hole in the same unit cell

OPTICAL SPECTRUM IN 2D CASE

The resonant part of the optical susceptibility in 2D case:

$$\chi(\omega) = -\frac{|d_{cv}|^2}{L_c \pi a_0^2 E_0} \left[\sum_{n=0}^{\infty} \frac{2}{(n+1/2)^3} \frac{E_0}{\hbar\omega - E_g - E_n} + \int dx \frac{x \mathrm{e}^{\pi/x}}{\cosh(\pi x)} \frac{E_0}{\hbar\omega - E_g - E_0 x^2} \right]$$

2D Elliott formula (absorption spectrum):

$$\alpha(\omega) = \alpha_0^{2D} \frac{\hbar\omega}{E_0} \left[\sum_{n=0}^{\infty} \frac{4}{(n+1/2)^3} \delta\left(\Delta + \frac{1}{(n+1/2)^2} \right) + \Theta(\Delta) \frac{\mathrm{e}^{\pi/\sqrt{\Delta}}}{\cosh(\pi\sqrt{\Delta})} \right]$$

Normalised detuning:

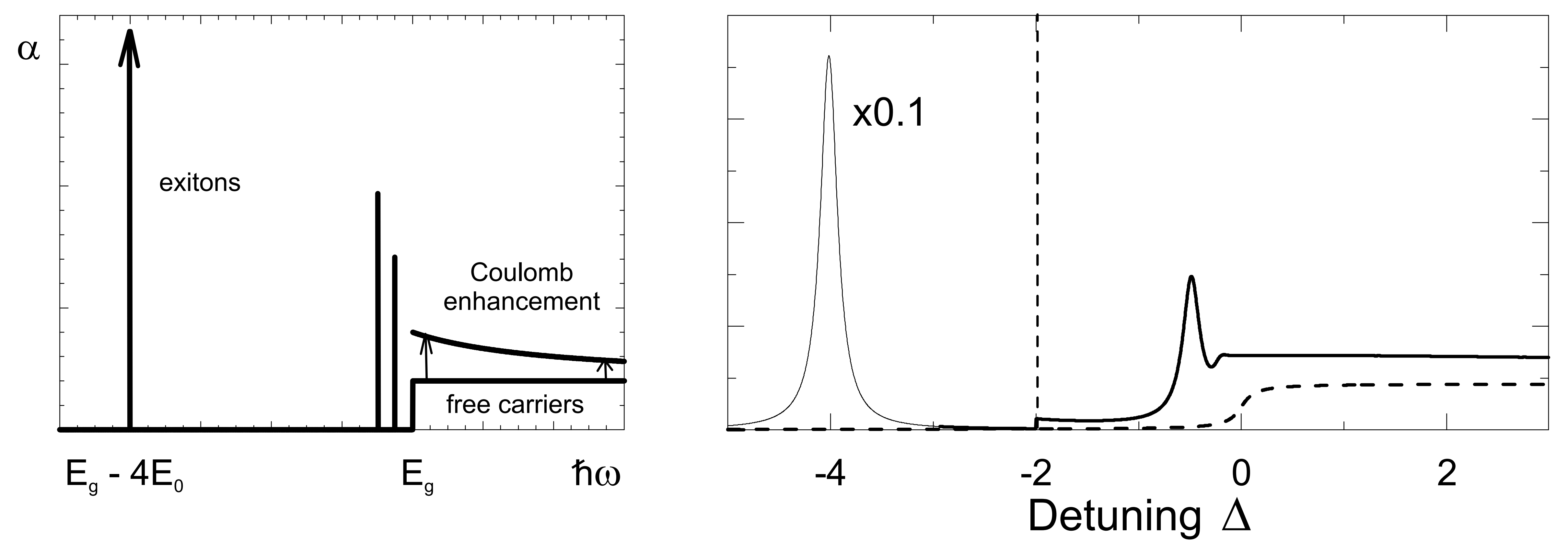
$$\Delta = (\hbar\omega - E_g)/E_0$$

Coulomb enhancement factor

ABSORPTION SPECTRUM FOR 2D SEMICONDUCTORS

Coulomb enhancement factor:

$$C(\omega) = \frac{e^{\pi/\sqrt{\Delta}}}{\cosh(\pi/\sqrt{\Delta})} \xrightarrow{\Delta \to 0} 2$$



NOTES ON EXCITONS IN 2D NANOSTRUCTURES

- ✴ The theoretical description is very close to the 3D case
- ✴ The absorption line of the 1s exciton is better resolved than in 3D case, but the further excited exciton states are more „dissolved“ in the absorption spectrum of free carriers
- ✴ The absorption at the band gap edge is enhanced by the Coulomb interaction

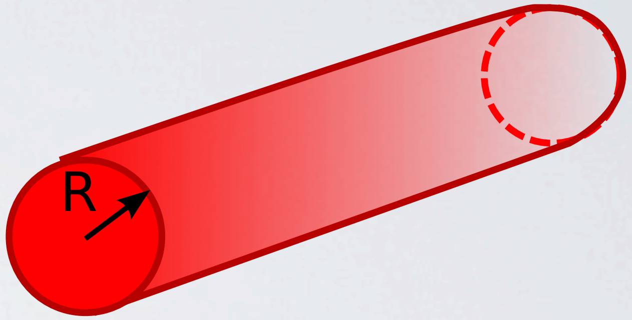
WANNIER EQUATION IN 1D CASE

$$-\left[\frac{\hbar^2 \nabla_{\mathbf{r}}^2}{2m_r} + V(r)\right]\psi_\nu(\mathbf{r}) = E_\nu \psi_\nu(\mathbf{r})$$

Wannier equation

In 1D case we have to replace the Coulomb potential with envelope averaged potential in a quantum wire (radius R):

$$V(\mathbf{r}) \to V^{1D}(z) = \frac{e^2}{\epsilon_0}\frac{1}{|z| + \gamma R}$$



1D exciton bound state energies:

$$E_\lambda = -E_0 \frac{1}{\lambda^2}$$

from boundary conditions

e. g. GaAs/GaAlAs wire: $E_{\lambda_0} \simeq 5E_0$

1D exciton wave functions:

$$f_\lambda(|z|) = N_\lambda W_{\lambda,1/2}\left(\frac{2(|z| + \gamma R)}{\lambda a_0}\right)$$

Whittaker functions

The normalised wave function for ionisation continuum in 1D:

$$f_k(\zeta) = \left(\frac{e^{\pi|\lambda|}}{2\pi}\right)^{1/2} \frac{D_0^{(2)} W^{(1)}(\zeta) - D_0^{(1)} W^{(2)}(\zeta)}{(|D_0^{(1)}|^2 + |D_0^{(2)}|^2)^{1/2}}$$

OPTICAL SPECTRUM FOR 1D SEMICONDUCTORS

Optical susceptibility in 1D case:

$$\chi(\omega) = -2|d_{cv}|^2 \sum_{\lambda} |f_\lambda(\alpha\gamma R)|^2 \left[\frac{1}{\hbar\omega - E_g - E_\lambda} - \frac{1}{\hbar\omega + E_g + E_\lambda} \right]$$

With only resonant contributions:

$$\chi(\omega) = -\frac{2}{E_0}|d_{cv}|^2 \left[\sum_{\lambda} |N_\lambda W^2_{\lambda,1/2}(2\gamma R/\lambda a_0)|^2 \frac{E_0}{\hbar\omega - E_g - E_\lambda} + \right.$$

$$\left. + \frac{2}{a_0} \int_0^\infty dx\, \frac{\mathrm{e}^{\pi/x}}{2\pi} \frac{|D_0^{(2)} W^{(1)} - D_0^{(1)} W^{(2)}|^2}{|D_0^{(1)}|^2 + |D_0^{(2)}|^2} \frac{E_0}{\hbar\omega - E_g - E_0 x^2} \right]$$

Absorption coefficient:

$$\alpha(\omega) = \frac{4\pi\omega}{nc} \frac{2}{E_0} |d_{cv}|^2 \left[\sum_{\lambda} |N_\lambda W^2_{\lambda,1/2}(2\gamma R/\lambda a_0)|^2 \pi\delta(\Delta - E_\lambda/E_0) + \right.$$

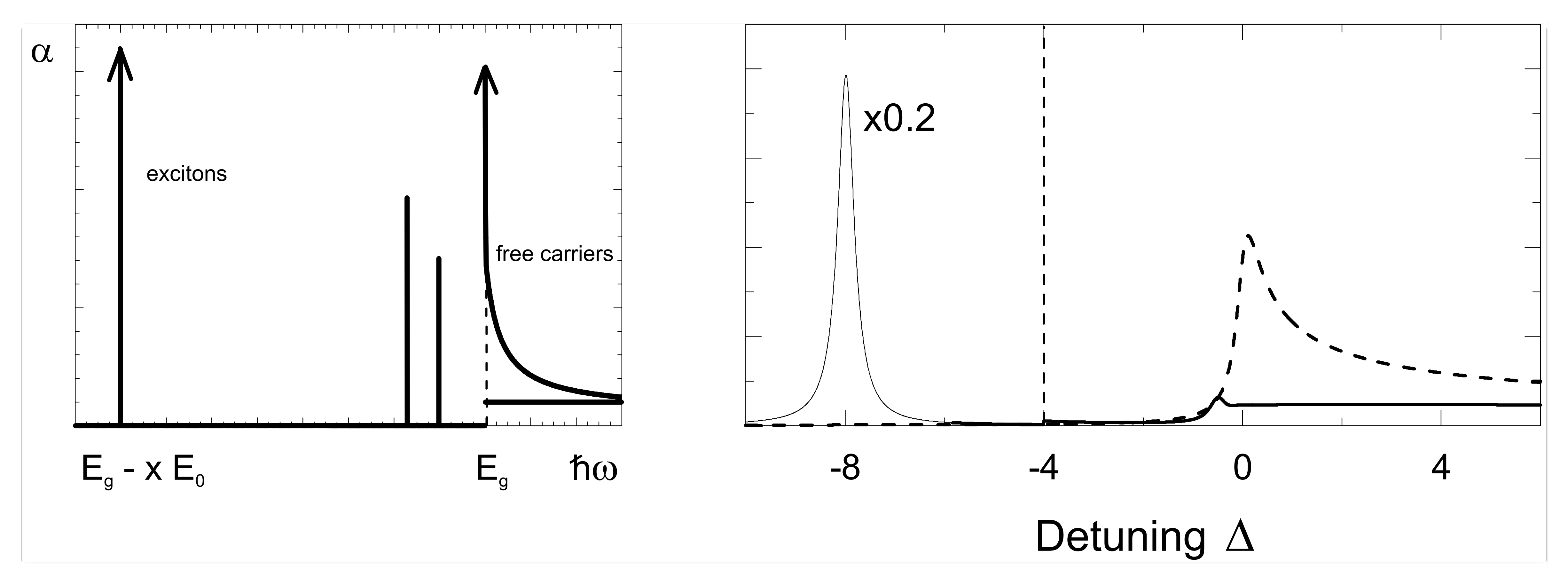
$$\left. + \frac{1}{\pi a_0} \frac{|D_0^{(2)} W^{(1)} - D_0^{(1)} W^{(2)}|^2}{|D_0^{(1)}|^2 + |D_0^{(2)}|^2} \frac{\mathrm{e}^{\pi/\sqrt{\Delta}}}{2\sqrt{\Delta}} \right]$$

ABSORPTION SPECTRUM FOR 1D SEMICONDUCTORS

Sommerfeld factor:

$$C(\omega) = \frac{e^{\pi/\sqrt{\Delta}}}{8} \frac{|D_0^{(2)} W^{(1)} - D_0^{(1)} W^{(2)}|^2}{|D_0^{(1)}|^2 + |D_0^{(2)}|^2} \quad < 1 \text{ for all } \hbar\omega > E_g$$

The pick in absorption from 1D free carrier of states is suppressed due to Cpulomb interaction. The band gap energy cannot be defined from absorption spectra



NOTES ON EXCITONS IN 1D NANOSTRUCTURES

- The theoretical description starts from general Wannier equation, but requieres special treatment

- The absorption line of the 2s exciton is well resolved, the 1s state does not contribute to the optical spectrum due to odd parity.

- The high excited states in a quantum wire are described by odd wave functions, therefore do not have any fingerprints in the spectra.

- The absorption at the band gap edge is reduced by the Coulomb interaction

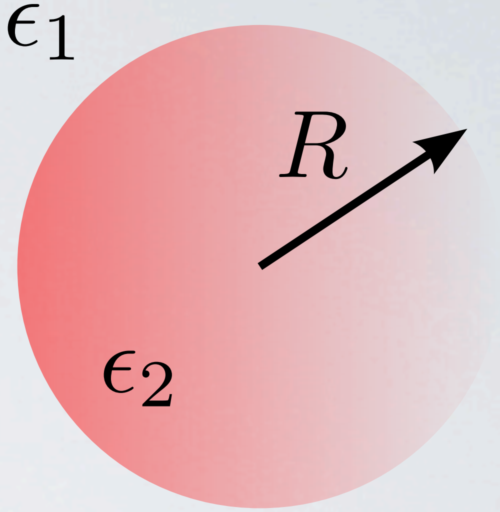
QUANTUM DOTS: 0D CASE

Approximation for the electron wave function:

$$\psi(\mathbf{r}) = \zeta(\mathbf{r})\, \mathbf{u}_\lambda(\mathbf{k} \simeq \mathbf{0}, \mathbf{r})$$

envelope function

Bloch function in bulk material



Boundary conditions: $\psi(r \geq R) = 0$

Hamiltonian for excitons in quantum dots:

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_h + V_{ee} + V_{hh} + V_{eh} \qquad (***)$$

$$\mathcal{H}_e = -\frac{\hbar^2}{2m_e} \int d^3r\, \hat{\psi}_e^\dagger(\mathbf{r})\, \nabla^2 \hat{\psi}_e(\mathbf{r}) + E_g \int d^3r\, \hat{\psi}_e^\dagger(\mathbf{r})\, \hat{\psi}_e(\mathbf{r})$$

$$\mathcal{H}_h = -\frac{\hbar^2}{2m_h} \int d^3r\, \hat{\psi}_h^\dagger(\mathbf{r}) \nabla^2 \hat{\psi}_h(\mathbf{r})$$

$$V_{ee} = \frac{1}{2} \int\int d^3r\, d^3r'\, \hat{\psi}_e^\dagger(\mathbf{r})\, \hat{\psi}_e^\dagger(\mathbf{r}')\, V(\mathbf{r}, \mathbf{r}')\, \hat{\psi}_e(\mathbf{r}')\, \hat{\psi}_e(\mathbf{r}) \qquad V_{hh} = V_{ee}(e \to h)$$

$$V_{eh} = -\int\int d^3r\, d^3r'\, \hat{\psi}_e^\dagger(\mathbf{r})\, \hat{\psi}_h^\dagger(\mathbf{r}')\, V(\mathbf{r}, \mathbf{r}')\, \hat{\psi}_h(\mathbf{r}')\, \hat{\psi}_e(\mathbf{r})$$

EXCITON STATES IN QUANTUM DOTS

Ansatz for exciton wave function:

$$|\psi_{eh}\rangle = \int\int d^3r_e\, d^3r_h\, \psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)\hat{\psi}_e^\dagger(\mathbf{r}_e)\hat{\psi}_h^\dagger(\mathbf{r}_h)\,|0\rangle$$

Inserting this state representation into Hamiltonian (***) gives:

$$\mathcal{H}_e|\psi_{eh}\rangle = -\frac{\hbar^2}{2m_e}\int d^3r\,[\nabla^2\hat{\psi}_e{}^\dagger(\mathbf{r})]\underline{\hat{\psi}_e(\mathbf{r})}\int d^3r_e\int d^3r_h\,\psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)\underline{\hat{\psi}_e{}^\dagger(\mathbf{r}_e)}\hat{\psi}_h{}^\dagger(\mathbf{r}_h)|0\rangle$$

$$+E_g\int d^3r\,\hat{\psi}_e{}^\dagger(\mathbf{r})\underline{\hat{\psi}_e(\mathbf{r})}\int d^3r_e\int d^3r_h\,\psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)\underline{\hat{\psi}_e{}^\dagger(\mathbf{r}_e)}\hat{\psi}_h{}^\dagger(\mathbf{r}_h)|0\rangle$$

General note:

$$\hat{\psi}_e(\mathbf{r})\hat{\psi}_e{}^\dagger(\mathbf{r}_e) = \delta(\mathbf{r}-\mathbf{r}_e) - \hat{\psi}_e{}^\dagger(\mathbf{r}_e)\hat{\psi}_e(\mathbf{r}) \quad \text{and } \hat{\psi}_e(\mathbf{r})|0\rangle = 0$$

$$\Rightarrow \mathcal{H}_e|\psi_{eh}\rangle = \iiint dr\,dr_e\,dr_h\,[\nabla^2\hat{\psi}_e^\dagger(\mathbf{r})]\delta(\mathbf{r}-\mathbf{r}_e)\psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)\hat{\psi}_h^\dagger(\mathbf{r}_h)$$

$$+E_g\iiint dr\,dr_e\,dr_h\,\hat{\psi}_e^\dagger(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}_e)\psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)\hat{\psi}_h^\dagger(\mathbf{r}_h) =$$

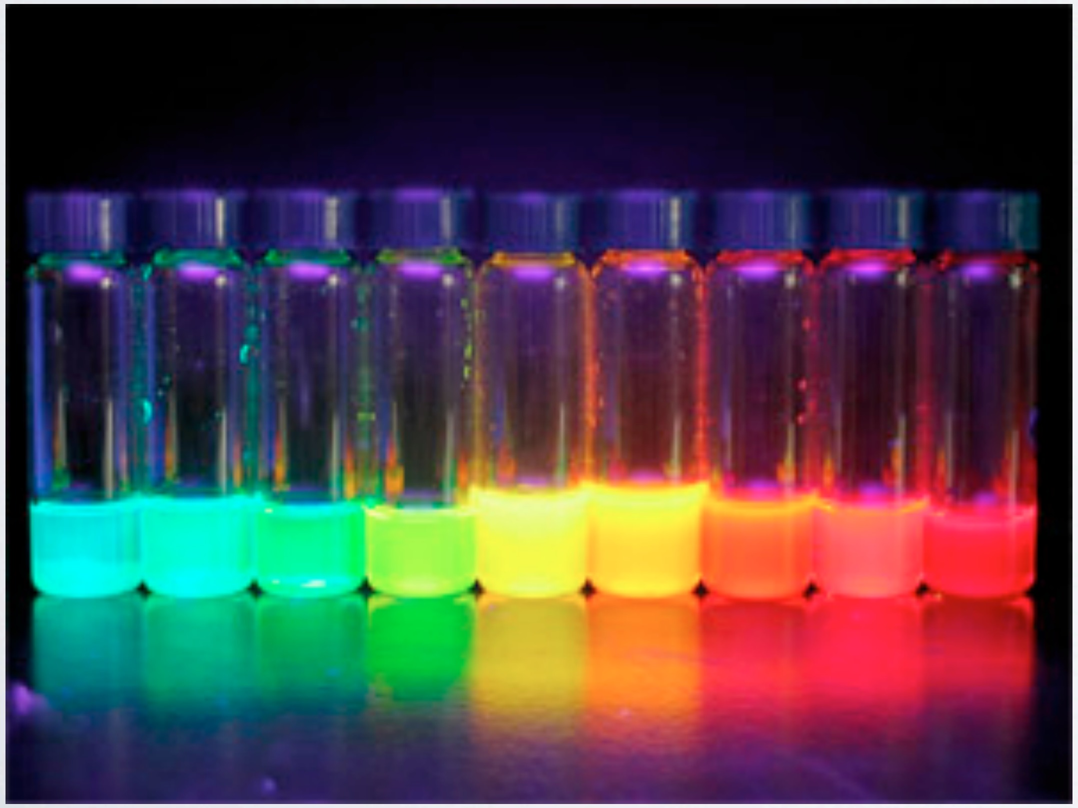
$$\iint dr_e\,dr_h\,[(\nabla^2+E_g)\psi_{eh}(\mathbf{r}_e,\mathbf{r}_h)]\hat{\psi}_e^\dagger(\mathbf{r_e})\hat{\psi}_h^\dagger(\mathbf{r}_h)$$

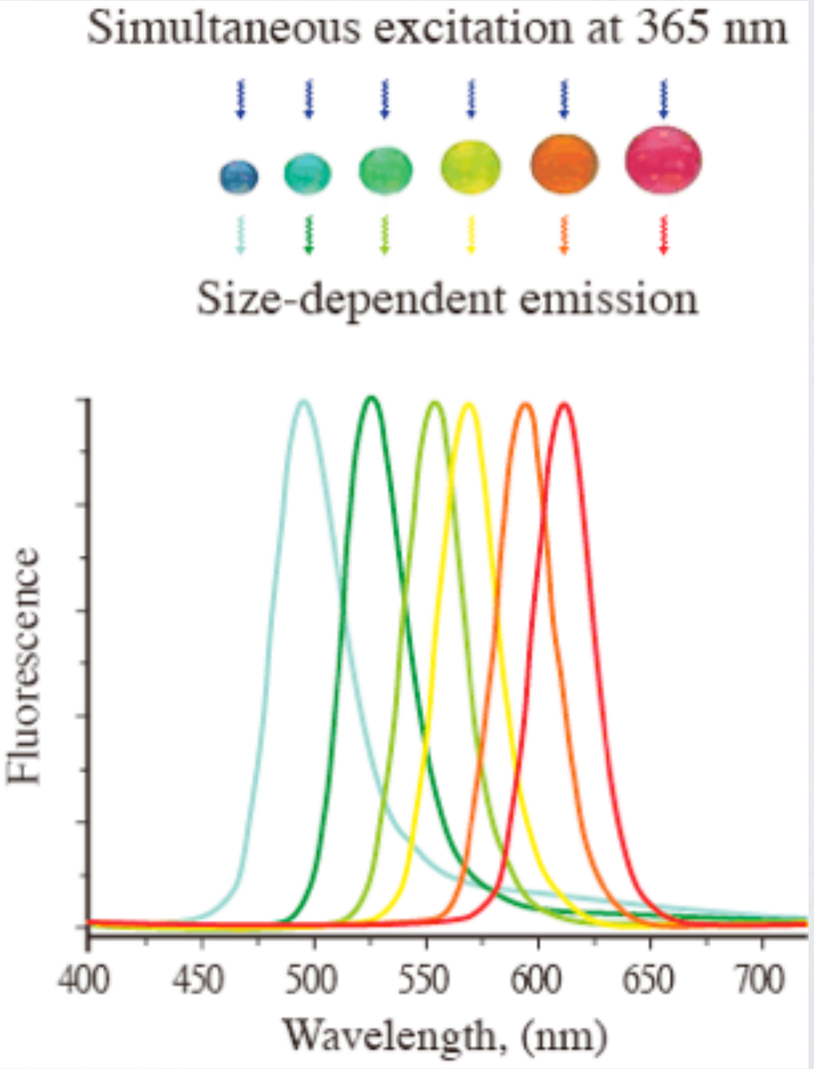
EQUATION FOR THE EXCITON WAVE FUNCTION

$$\Bigg[\underbrace{-\frac{\hbar^2}{2m_e}\nabla_e^2-\frac{\hbar^2}{2m_h}\nabla_h^2}_{\sim 1/R^2}\underbrace{-V(\mathbf{r_e},\mathbf{r_h})}_{\sim 1/R}\Bigg]\psi_{eh}(\mathbf{r_e},\mathbf{r_h})=(E-E_g)\psi_{eh}(\mathbf{r_e},\mathbf{r_h})$$

$$\psi_{eh}(\mathbf{r_e},\mathbf{r_h}) = 0 \text{ if } |\mathbf{r}_e| > R \quad \text{or} \quad |\mathbf{r}_h| > R$$

Blueshift of absorption frequency for smaller quantum dot sizes





SINGLE PARTICLES STATES

Approximation for exciton wave function: $E_{eh,nlm} = E_{e,nlm} + E_{h,nlm}$

Use Schrödinger equation to find eigenvalues: $\mathcal{H}|\psi_e\rangle = E_e\,|\psi_e\rangle$

Apply ansatz:

$$|\psi_e\rangle = \int d^3r\,\zeta_e(\mathbf{r})\hat{\psi}_e^\dagger(\mathbf{r})\,|0\rangle$$

Single electron eigenvalue equation:

$$-\frac{\hbar^2}{2m_e}\nabla^2\zeta_e(\mathbf{r}) = (E_e - E_g)\,\zeta_e(\mathbf{r})$$

Wave functions for single electron in a quantum dot (the same applies for single hole states, just exchange the index and exclude band gap energy from calculations)

$$\boldsymbol{\zeta_{e,nlm}(\mathbf{r}) = \sqrt{\frac{2}{R^3}}\,\frac{j_l(\alpha_{nl}r/R)}{j_{l+1}(\alpha_{nl})}\,Y_{l,m}(\Omega)}$$

$$j_l(\alpha_{nl}) = 0 \quad \text{for} \quad n = 1, 2, \cdots$$

$$\left\{\begin{aligned} E_{e,nlm} &= E_g + \frac{\hbar^2}{2m_e}\,\frac{\alpha_{nl}^2}{R^2} \\ E_{h,nlm} &= \frac{\hbar^2}{2m_h}\,\frac{\alpha_{nl}^2}{R^2} \end{aligned}\right.$$

$$\psi_{eh}(r_e, r_h) \simeq \zeta_{100}(r_e)\,\zeta_{100}(r_h) + \text{ other states}$$

DIPOLE TRANSITIONS

To know optical response, we need to know dipole moment matrix elements:

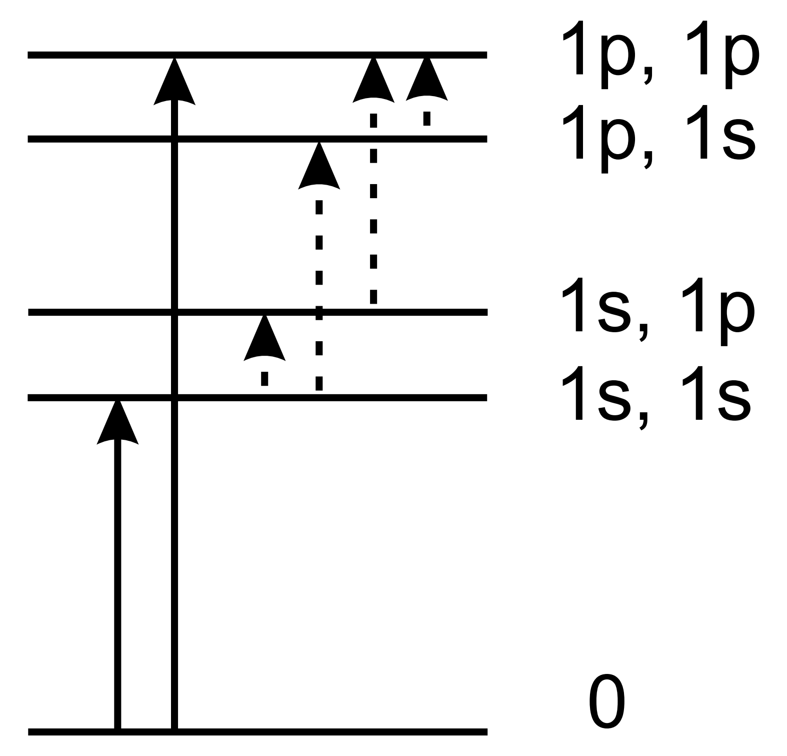
$$\hat{\boldsymbol{P}} = \int d^3r \sum_{i,j=e,h} \hat{\psi}_i^\dagger(\mathbf{r})\, e\mathbf{r}\, \hat{\psi}_j(\mathbf{r}) = \int d^3r e\mathbf{r} \Big[\underbrace{\hat{\psi}_e^\dagger(\mathbf{r})\hat{\psi}_e(\mathbf{r}) + \hat{\psi}_h(\mathbf{r})\hat{\psi}_h^\dagger(\mathbf{r})}_{\text{Intraband transitions}} + \underbrace{\hat{\psi}_e^\dagger(\mathbf{r})\hat{\psi}_h^\dagger(\mathbf{r}) + \hat{\psi}_h(\mathbf{r})\hat{\psi}_e(\mathbf{r})}_{\substack{\text{Interband transitions:} \\ \text{creation and annihilation} \\ \text{of exciton pair states}}} \Big]$$

$$\int d^3r\, e\mathbf{r}\, \hat{\psi}_e^\dagger(\mathbf{r})\, \hat{\psi}_h^\dagger(\mathbf{r}) = \mathbf{d}_{c\nu} \sum_{\substack{nlm \\ n'l'm'}} a^\dagger_{nlm}\, b^\dagger_{n'l'm'} \int d^3R\, \zeta^*_{nlm}(\mathbf{R})\, \zeta_{n'l'm'}(\mathbf{R}) = \mathbf{d}_{c\nu} \sum_{nlm} a^\dagger_{nlm}\, b^\dagger_{nlm}$$

$$\hat{\psi}_e(\mathbf{r}) = \sum_{nlm} \psi^e_{nlm}(\mathbf{r})\, a_{nlm}$$

$$\mathbf{d}_{c\nu} = \int d^3r\, e\mathbf{r}\, u_c^*(\mathbf{r}) u_v(\mathbf{r})$$

$$\hat{\psi}_h(\mathbf{r}) = \sum_{nlm} \psi^h_{nlm}(\mathbf{r})\, b_{nlm}$$



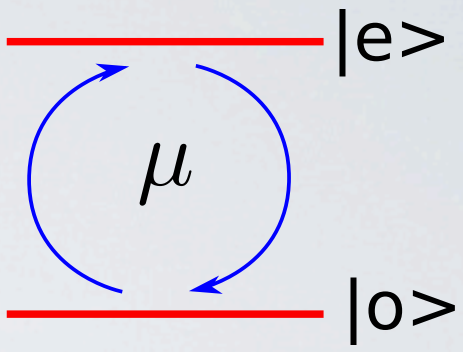
Intraband transition dipole moment matrix elements:

$$\boxed{n \neq n' \,; \quad l - l' = 0 \;,\; \pm 1 \,; \quad m - m' = 0 \,,\; \pm 1}$$

BLOCH EQUATIONS FOR SINGLE EXCITON

Assume two level system - ground state and exciton state:

$$H = \hbar\omega_e|e\rangle\langle e| \qquad (\omega_o = 0)$$



Interaction with light:

$$H_I = -\mu_{eo}|e\rangle\langle o| - \mu_{oe}|o\rangle\langle e|$$

$$\mu_{ij} = \mathbf{d}_{ij} \cdot \boldsymbol{\mathcal{E}}(t)$$

Density matrix:

$$\rho = \rho_{oo}|o\rangle\langle o| + \rho_{ee}|e\rangle\langle e| + \rho_{eo}|e\rangle\langle o| + \rho_{oe}|o\rangle\langle e|$$

Dynamics of density matrix elements:

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\rho = [H + H_I, \rho]$$

Solution:

$$\rho_{ee} = 1 - \rho_{oo}$$

$$\mathrm{i}\hbar\frac{\partial}{\partial t}\rho_{eo} = \mu_{eo}(\rho_{ee} - \rho_{oo}) + \hbar\omega_e\rho_{eo} = \mathrm{i}\hbar\frac{\partial}{\partial t}\rho^*_{oe}$$

OPTICAL SPECTRA FOR QUANTUM DOTS

Linear polarisation:

$$P_{lin} = d_{oe}\rho_{eo} + c.c. \qquad\qquad \chi_{lin} = P_{lin}(\omega)/\mathcal{E}(\omega)$$

To the first order of the field and with phenomenological damping constant:

$$\frac{\partial}{\partial t}\rho_{oe}^{(1)} = -(i\omega_e + \gamma_e)\rho_{oe}^{(1)} + id_{oe}\frac{\mathcal{E}(t)}{\hbar}$$

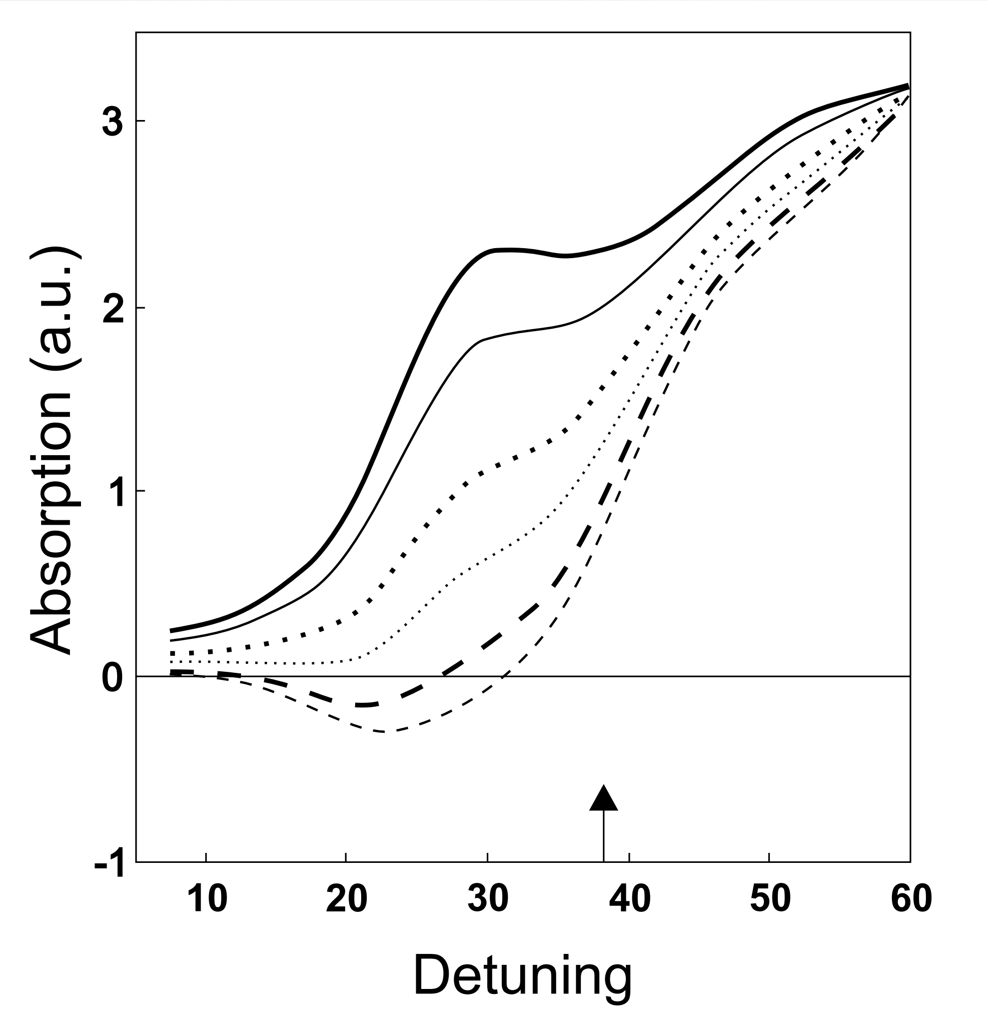
Linear optical susceptibility: $\chi_{lin} = \frac{i}{\hbar}\sum_e |d_{oe}|^2 \left[\frac{1}{\gamma_e + i(\omega_e - \omega)} + \frac{1}{\gamma_e - i(\omega_e + \omega)}\right]$

Linear absorption coefficient $\boldsymbol{\alpha_l(\omega) = \frac{4\pi\omega}{\hbar c\sqrt{\epsilon_2}}\sum_e |d_{oe}|^2 \frac{\gamma_e}{\gamma_e^2 + (\omega_e - \omega)^2}}$

(a) Helium–"real atom"

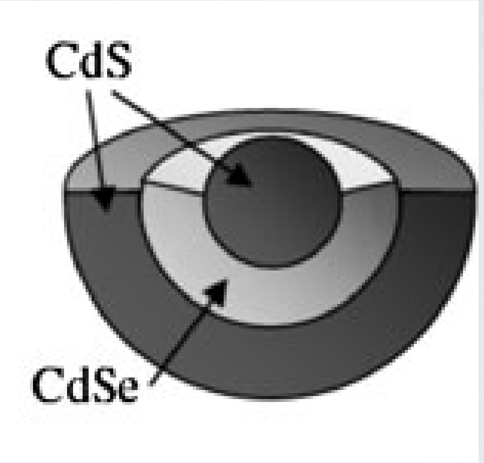
(b) Quantum dot–"artifical atom"

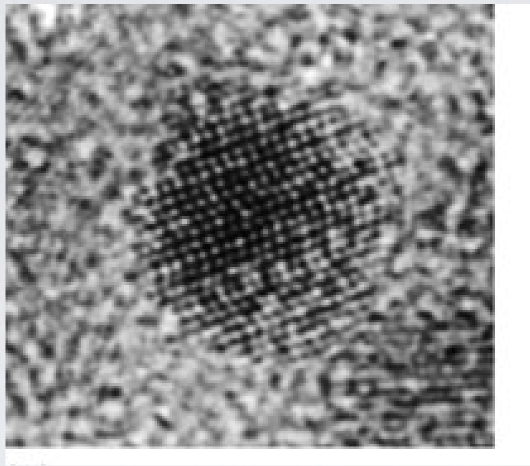
NEGATIVE OPTICAL ABSORPTION



Non-linear regime:
by certain light intensity
optical gain can be
obtained.
Application:
quantum dot laser

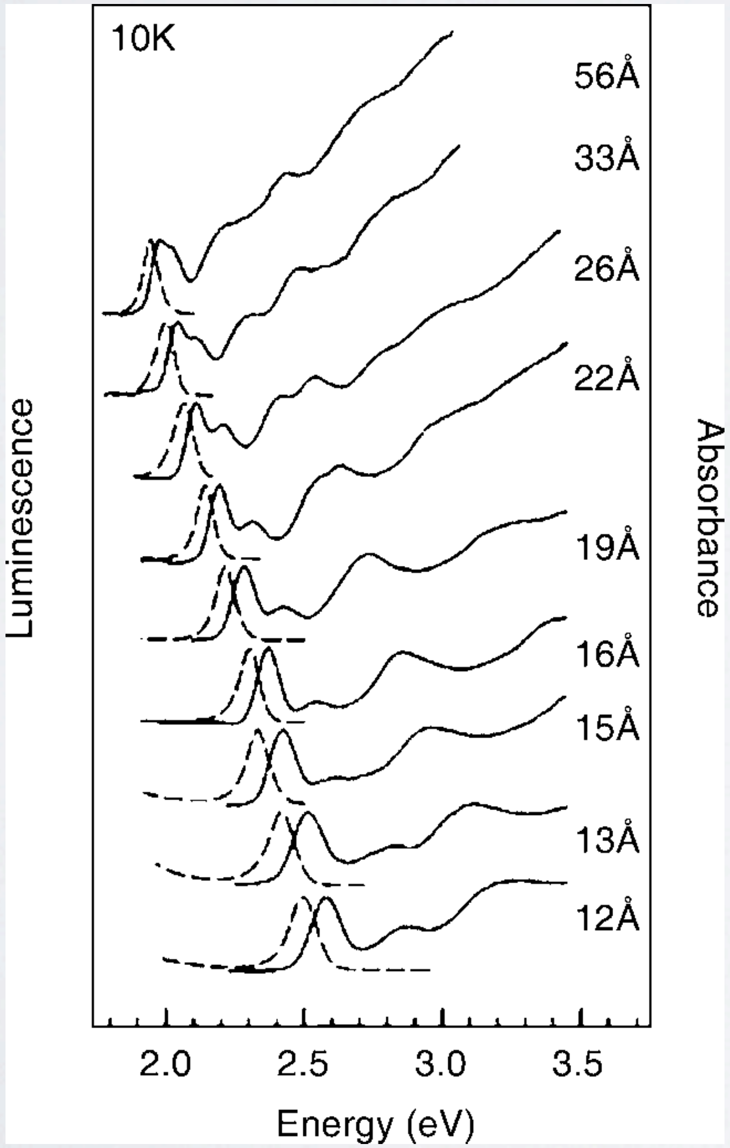
COLLOIDAL QUANTUM DOTS

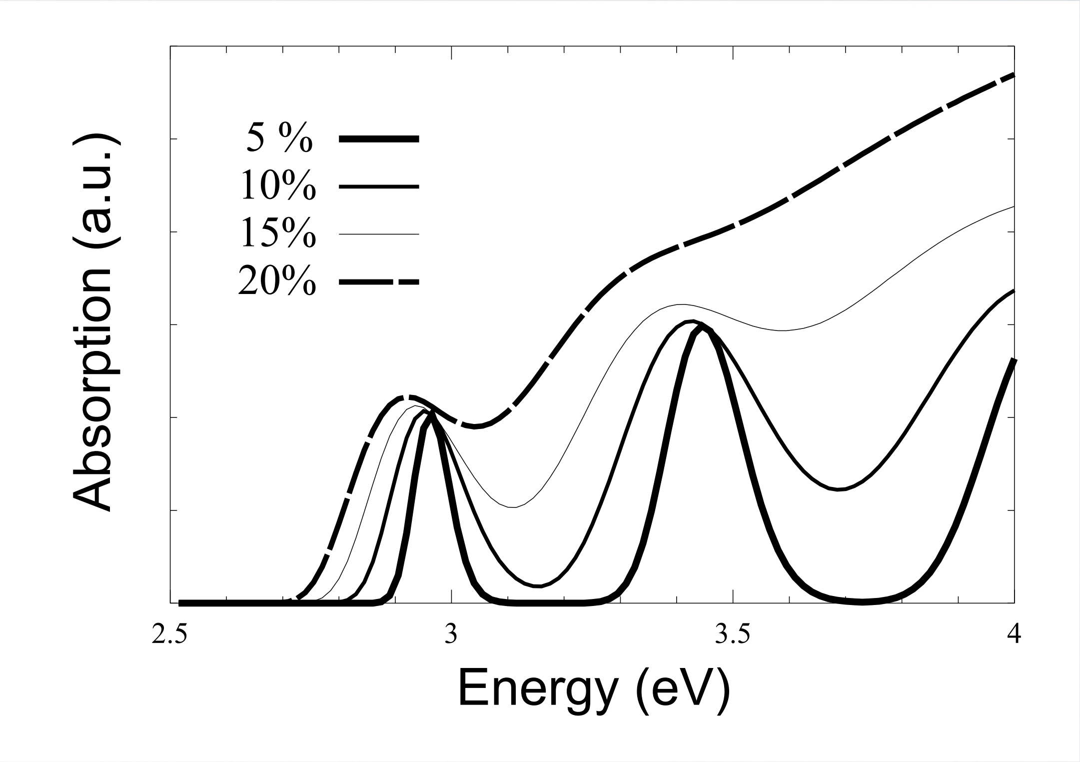




Broadening of the absorption spectrum due to variation of the radii of the dots (Gaussian distribution)

Shift of the exciton energy in different size quantum dots

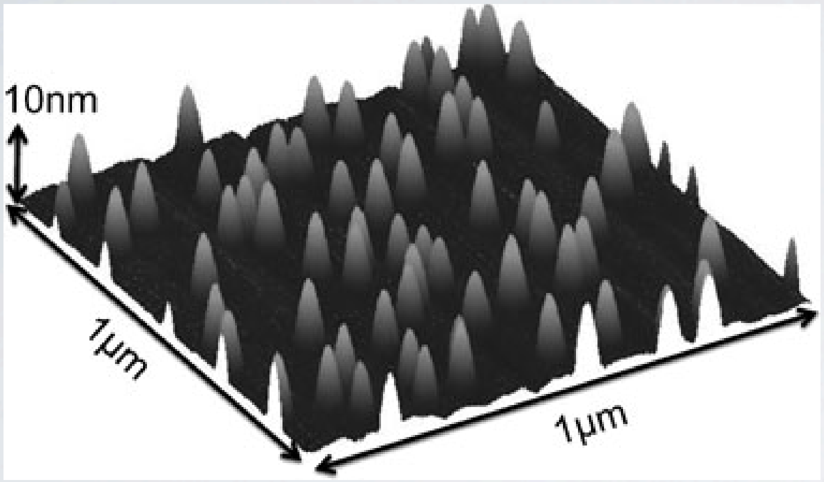




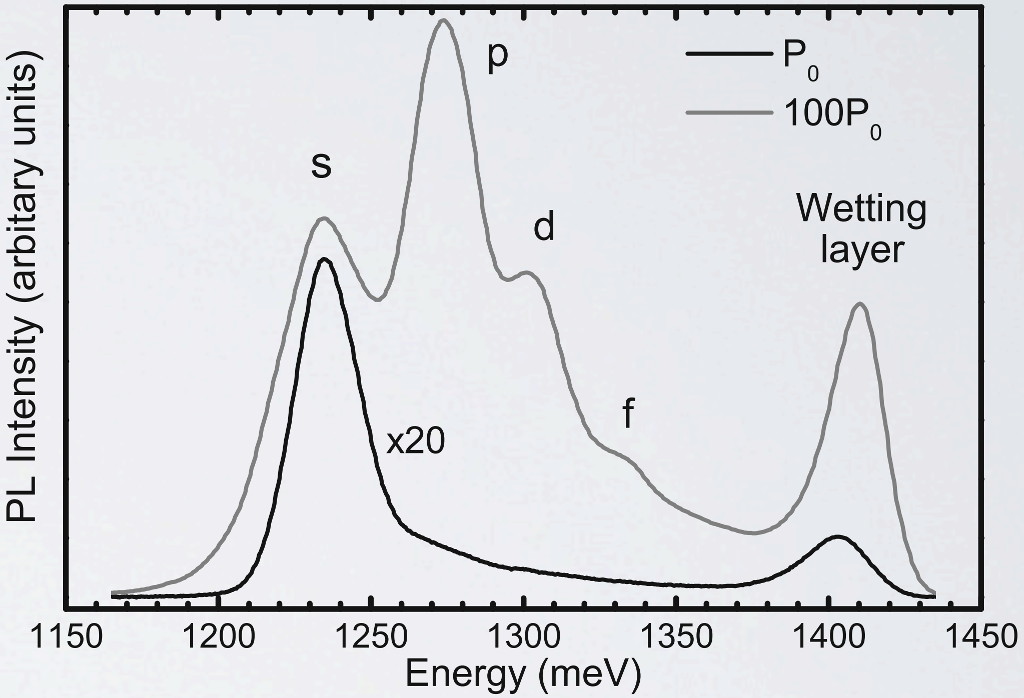
$$\alpha_l(\omega)|_{a\nu} = \int_0^\infty dR\, f(R)\, \alpha_l(\omega)|_R$$

SELF-ASSEMBLED QUANTUM DOTS

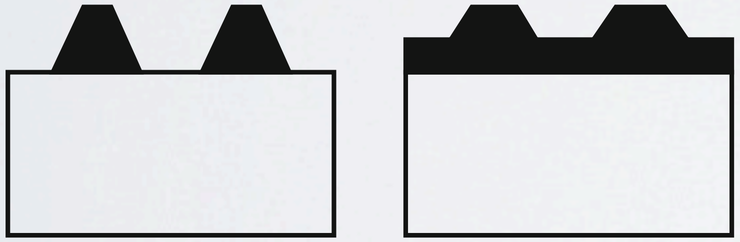
In(Ga)As/GaAs, In(Ga)As/InP, SiGe/Si or CdSe/ZnSe



Photoluminescence spectrum



Heteroepitaxical growth methods:

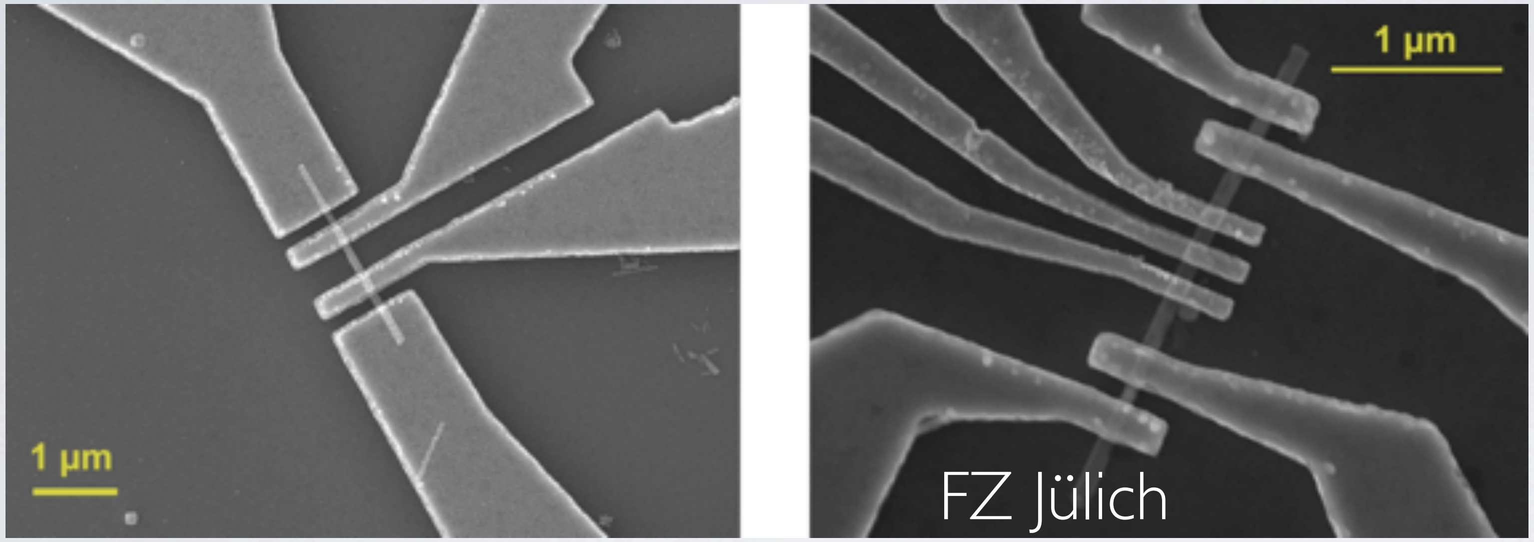


Volmer–Weber

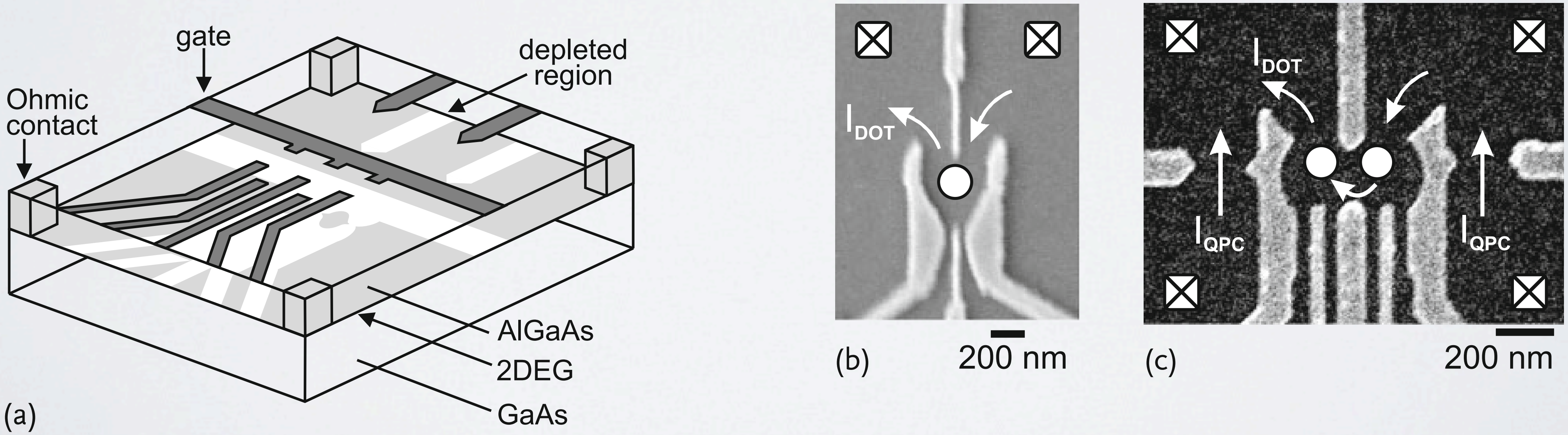
Stanski–Krastanow

GATE DEFINED QUANTUM DOTS

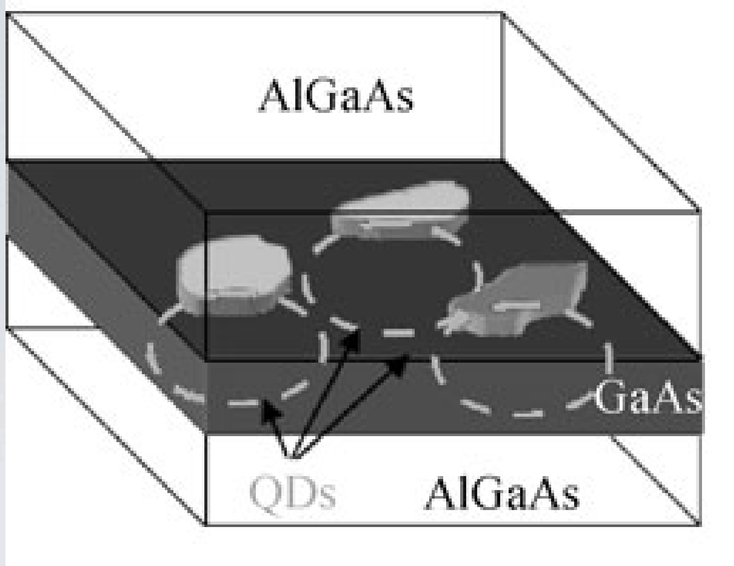
Quantum dots in quantum wires:

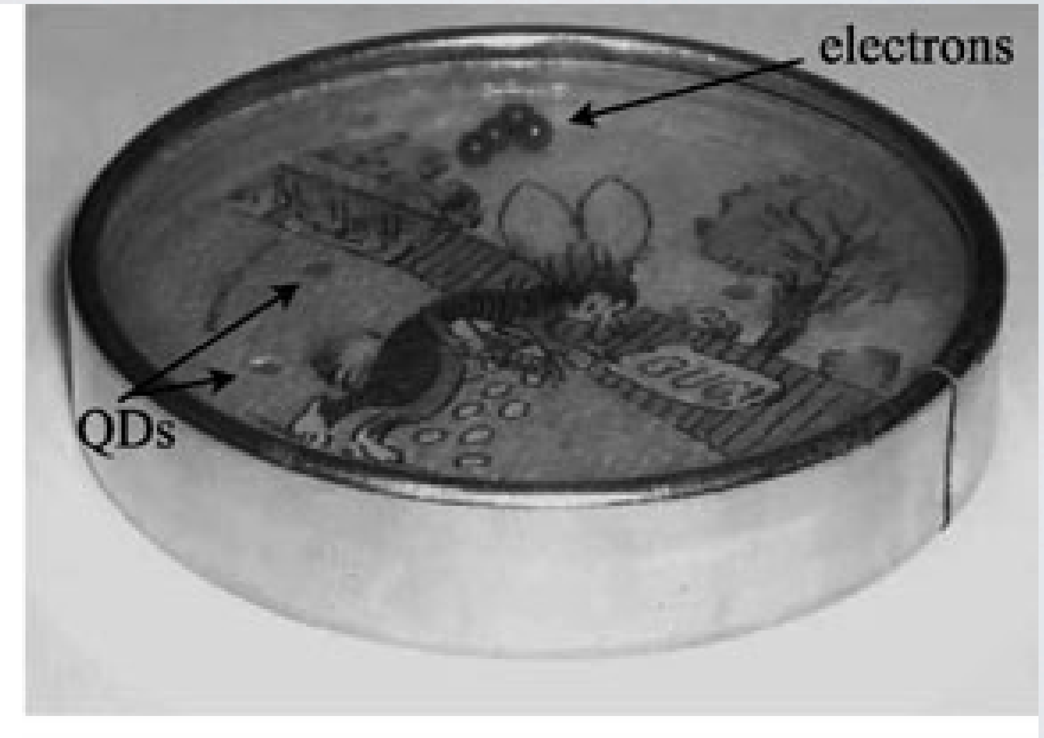


Quantum dots in a 2D heterostructure:

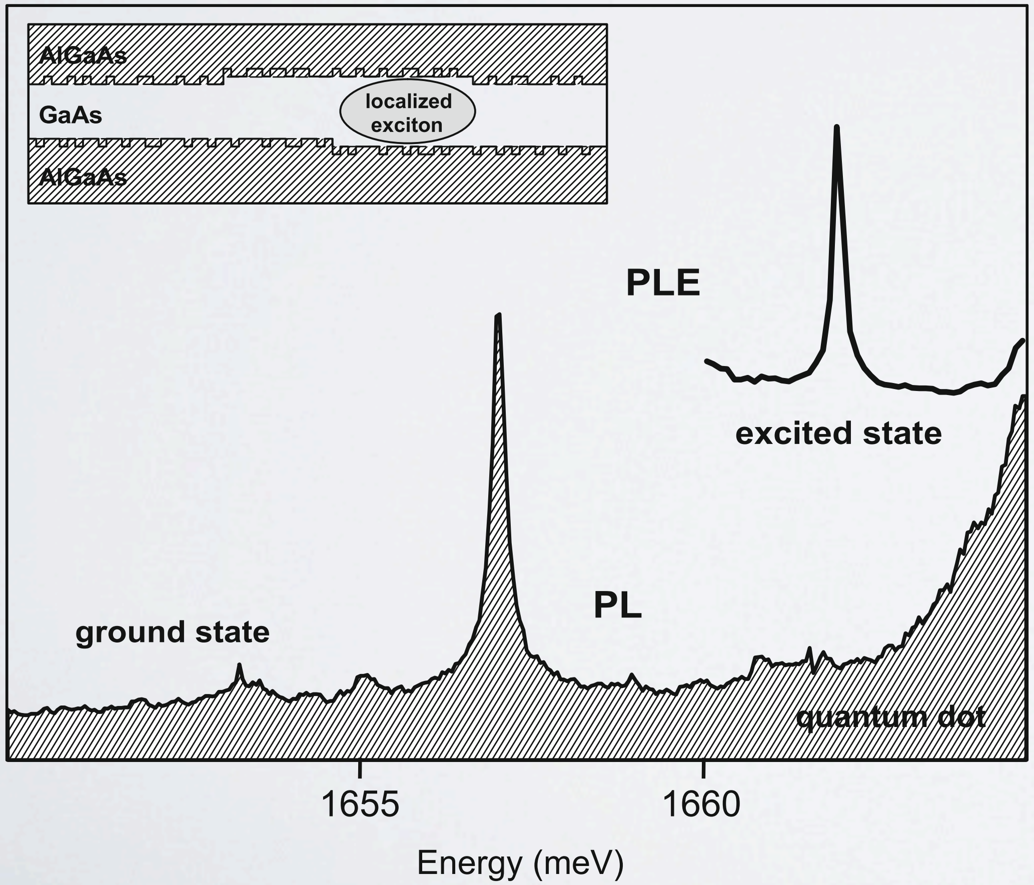


INTERFACE FLUCTUATION QUANTUM DOTS





Photoluminescense spectrum



NOTES ON EXCITONS IN QUANTUM DOTS

- ✴ Quantum dots exhibit atom-like optical properties: well-defined absorption peaks
- ✴ The size and form (confining potential) of a quantum dot have significant influence on optical properties
- ✴ The energy eigenvalues for exciton states can be defined analytically in a strict approximation, otherwise only numerically

WANNIER EQUATION IN 2D CASE

Solve first homogeneous equation:

$$-\left[\frac{\hbar^2\nabla_{\mathbf{r}}^2}{2m_r}+V(r)\right]\psi_\nu(\mathbf{r})=E_\nu\psi_\nu(\mathbf{r})$$

Wannier equation

With scaled radius $\rho = r\alpha$ and $\lambda=\frac{e^2}{\hbar\epsilon_0}\sqrt{-\frac{m_r}{2E_\nu}}=\frac{2}{\alpha a_0}$ $\quad E_0=\frac{\hbar^2}{2m_r a_0^2}$

we get: $\left(-\nabla_\rho^2-\frac{\lambda}{\rho}\right)\psi(\rho)=-\frac{1}{4}\psi(\rho)$

$\lambda > 0$ bound states

$\lambda < 0$ ionisation continuum

2D Laplace operator, polar coordinates:

$$\nabla_\rho^2=\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho}-\frac{\mathcal{L}_z^2}{\rho^2} \qquad \mathcal{L}_z^2=-\frac{\partial^2}{\partial\phi^2} \qquad \mathcal{L}_z\frac{1}{\sqrt{2\pi}}e^{im\phi}=m\frac{1}{\sqrt{2\pi}}e^{im\phi}$$

Ansatz:

$$\psi(\rho)=f_m(\rho)\frac{1}{\sqrt{2\pi}}\mathrm{e}^{im\phi}$$

ELECTRON WAVE FUNCTION IN 2D CASE

Equation for radial part:

$$\left(\frac{1}{\rho}\frac{\partial}{\partial\rho}\rho\frac{\partial}{\partial\rho}+\frac{\lambda}{\rho}-\frac{1}{4}-\frac{m^2}{\rho^2}\right)f_m(\rho)=0 \qquad (*)$$

Ansatz:

$$f_m(\rho)=\rho^{|m|}e^{-\frac{\rho}{2}}R(\rho)$$

assymptotic behaviour for $\rho\rightarrow 0$ and $\rho\rightarrow\infty$

Inserting in (*) gives:

$$\rho\frac{\partial^2 R}{\partial\rho^2}+(2|m|+1-\rho)\frac{\partial R}{\partial\rho}+\left(\lambda-|m|-\frac{1}{2}\right)R=0$$

Solution:

$$R(\rho)=\sum_{\nu=0}\beta_\nu\rho^\nu \qquad \nu_{max}+|m|+\frac{1}{2}=\lambda\equiv n+\frac{1}{2}$$

2D exciton bound state energies:

$$\boxed{\boldsymbol{E_n=-E_0\frac{1}{(n+1/2)^2}}\quad \text{with } \boldsymbol{n=0,1,\ldots}}$$

2D EXCITON WAVE FUNCTIONS

2D radial wave functions:

ν_{max}	n	m	$f_{n,m}(\rho) = C\rho^{\lvert m\rvert}e^{-\frac{\rho}{2}}\sum_{\nu}\beta_{\nu}\rho^{\nu}$	E_n
0	0	0	$f_{0,0}(r) = \frac{1}{a_0}4e^{-2r/a_0}$	$E_{n=0} = -4E_0$
1	1	0	$f_{1,0}(r) = \frac{4}{a_0 3\sqrt{3}}\left(1 - \frac{4r}{3a_0}\right)e^{-\frac{2r}{3a_0}}$	$E_1 = -\frac{4E_0}{9}$
0	1	± 1	$f_{1,\pm 1}(r) = \frac{16}{a_0 9\sqrt{6}}\frac{r}{a_0}e^{-2r/3a_0}$	$E_1 = -\frac{4E_0}{9}$.

2D exciton wave functions:

$$\psi_{n,m}(\mathbf{r}) = \sqrt{\frac{1}{\pi a_0^2(n+\frac{1}{2})^3}\frac{(n-|m|)!}{[(n+|m|)!]^3}}\rho^{|m|}e^{-\frac{\rho}{2}}L_{n+|m|}^{2|m|}(\rho)\,e^{im\phi}$$

IONISATION CONTINUUM IN 2D

λ negative

The same ansatz for radial part: $f_m(\rho) = \rho^{|m|} e^{-\frac{\rho}{2}} R(\rho)$

The equation for R:

$$\rho \frac{\partial^2 R}{\partial \rho^2} + (2|m| + 1 - \rho) \frac{\partial R}{\partial \rho} - \left(i|\lambda| + |m| + \frac{1}{2} \right) R = 0$$

The normalised wave function:

$$\psi_{k,m}(\mathbf{r}) = \frac{(i2kr)^{|m|}}{(2|m|)!} \sqrt{\frac{\pi k}{\mathcal{R}(1/4+|\lambda|^2)\cosh(\pi|\lambda|)} \prod_{j=0}^{|m|} \left[\left(j - \tfrac{1}{2}\right)^2 + |\lambda|^2 \right]}$$

$$\times \quad e^{\frac{\pi|\lambda|}{2}} e^{-ikr} F\left(|m| + \tfrac{1}{2} + i|\lambda|;\ 2|m| + 1;\ 2ikr \right) \frac{e^{im\phi}}{\sqrt{2\pi}}$$

WANNIER EQUATION IN 1D CASE

$$-\left[\frac{\hbar^2\nabla_{\mathbf{r}}^2}{2m_r}+V(r)\right]\psi_\nu(\mathbf{r})=E_\nu\psi_\nu(\mathbf{r})$$

Wannier equation

In 1D case we have to replace the Coulomb potential with envelope averaged potential in a quantum wire (radius R):

$$V(\mathbf{r})\rightarrow V^{1D}(z)=\frac{e^2}{\epsilon_0}\frac{1}{|z|+\gamma R}$$

general scaled Wannier equation $\left(-\nabla_\rho^2-\frac{\lambda}{\rho}\right)\psi(\rho)=-\frac{1}{4}\psi(\rho)$ (**)

in 1D case $\rho\rightarrow\zeta=\alpha(|z|+\gamma R)$ $\nabla_\rho^2=\frac{\partial^2}{\partial\zeta^2}$ $\psi(\rho)=f(\zeta)$

Equation (**) in 1D case:

Assymptotic behavior for large radii (distances):

$$\left(\frac{\partial^2}{\partial\zeta^2}+\frac{\lambda}{\zeta}-\frac{1}{4}\right)f(\zeta)=0 \longleftarrow f(\zeta)=e^{-\frac{\zeta}{2}}R(\zeta)$$

$$\left(\frac{\partial^2}{\partial\zeta^2}+\frac{\partial}{\partial\zeta}+\frac{\lambda}{\zeta}\right)R=0$$

ELECTRON WAVE FUNCTION IN A QUANTUM WIRE

Wannier equation in 1D case is Whittaker equation:

$$\left(\frac{\partial^2}{\partial\zeta^2}+\frac{\lambda}{\zeta}-\frac{1}{4}+\frac{1/4-\mu^2}{\zeta^2}\right)W_{\lambda,\mu}(\zeta)=0 \qquad \mu=\pm 1/2$$

Whittaker functions

1D exciton bound state energies:

$$E_\lambda=-E_0\frac{1}{\lambda^2} \qquad \lambda$$ from boundary conditions

1D exciton wave functions:

$$f_\lambda(|z|)=N_\lambda W_{\lambda,1/2}\left(\frac{2(|z|+\gamma R)}{\lambda a_0}\right)$$ (even: $df(\zeta)/dz\,|_{z=0}=0$)

Eigenvalue for the ground state:

$$\frac{1}{2}+\lambda_0\,\ln\left(\frac{2\gamma R}{\lambda_0 a_0}\right)=0 \quad \rightarrow \lambda_0 \ll 1 \qquad E_{\lambda_0}\gg E_0$$

e. g. GaAs/GaAlAs wire: $E_{\lambda_0}\simeq 5E_0$

IONISATION CONTINUUM IN 1D

Scaled Wannier equation with λ negative:

$$\left[\frac{d^2}{d\zeta^2} - \left(\frac{1}{4} + i\frac{|\lambda|}{\zeta}\right)\right] f(\zeta) = 0$$

Two independent solutions (Whittaker functions):

$$W^{(1)}_{-i|\lambda|,1/2}(\zeta) = \Gamma(1+i|\lambda|)\zeta e^{-\zeta/2}[F(1+i|\lambda|,2;\zeta) + G(1+i|\lambda|,2;\zeta)]$$

$$W^{(2)}_{-i|\lambda|,1/2}(\zeta) = \Gamma(1-i|\lambda|)\zeta e^{-\zeta/2}[F(1+i|\lambda|,2;\zeta) - G(1+i|\lambda|,2;\zeta)]$$

Even electron wave function combined of these two solutions and normalised:

$$f_k(\zeta) = \left(\frac{e^{\pi|\lambda|}}{2\pi}\right)^{1/2} \frac{D_0^{(2)} W^{(1)}(\zeta) - D_0^{(1)} W^{(2)}(\zeta)}{(|D_0^{(1)}|^2 + |D_0^{(2)}|^2)^{1/2}} \qquad D_0^{(j)} = \frac{dW^{(j)}(\zeta)}{d\zeta}|_{\zeta=2ik\gamma R}$$