Lection 3

3). Optics of excitons in nanostructures (4h)
(exciton -photon interaction, bulk polaritons, exciton -photon interaction in a SQW; array of QWs; short-period SR;
Bragg QW structures, diffraction from an array of QWWs and QDs;
polaritons in microcavities)

Reflectivity and transmitivity of a single quantum well

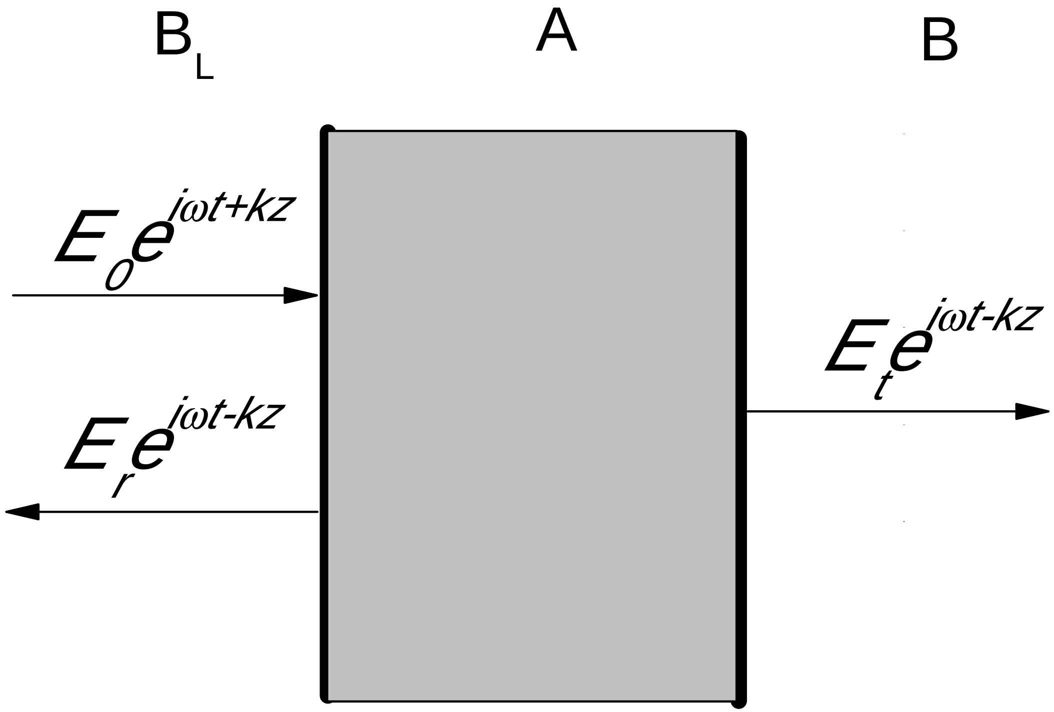
$$D_i(\mathbf{r},t) = \int_{-\infty}^{t} dt \int dV \varepsilon_{ij}(\mathbf{r}-\mathbf{r}', t-t') E_j(\mathbf{r}', t')$$

$$\tau = (t - t') \qquad \tau > 0 \qquad \mathbf{R} = \mathbf{r} - \mathbf{r}'$$

$$\varepsilon_{ij}(\omega, \mathbf{k}) = \int_{0}^{\infty} d\tau \int d\mathbf{R} e^{-i(\mathbf{kR}-\omega\tau)} \varepsilon_{ij}(\mathbf{R}, \tau)$$

$$D_i(\omega, \mathbf{k}) = \varepsilon_{ij}(\omega, \mathbf{k}) E_j(\omega, \mathbf{k})$$

Sandwich B-A-B



Normal incidence

$$k_x = k_y = 0$$

At the left – incident wave with the amplitude E_0
and reflected wave with the amplitude E_r

At the right – transmitted wave E_t

The only difference between **A** and **B** is that there are no excitons in in **B =** barrier, there is exciton in **A =** quantum well

$$r = \frac{E_r}{E_0}, \quad t = \frac{E_t}{E_0}$$ Reflection and transmission

If no absorption $\Rightarrow |r|^2 + |t|^2 = 1$

Let

ω_0 - Exciton resonance frequency

τ_0 - Exciton radiative lifetime

τ - Exciton nonradiative lifetime

After creation of the exciton its wavefunction $\Psi(t) \sim e^{-i\Omega_0 t}$

where $\Omega_0 = \omega_0 - i(\Gamma + \Gamma_0)$ $\quad \Gamma_0 \equiv (2\tau_0)^{-1}$ $\quad \Gamma \equiv (2\tau)^{-1}$

Coefficients $r(\omega)$ and $t(\omega)$ are response functions

Reflection and transmission coefficients describes linear response of the layer A on the incident electromagnetic field

These values should have poles on the eigenfrequencies $\Rightarrow$

$$r(\omega) = C_r + \frac{d_r}{\omega_0 - i\left({}_0 + \right) - \omega}$$

$$t(\omega) = C_t + \frac{d_t}{\omega_0 - i\left({}_0 + \right) - \omega}$$

As usually in optics, incident field induced polarization that Is coherent with this field $E_{exc} e^{-i\omega t}$

Let the wave in $\mathbf{B}_R$ is a sum of incident $E_0 e^{-i\omega t+ikz}$ and

emitted coherently by exciton waves $E_{exc} e^{-i\omega t+ikz}$

Due to mirror symmetry in z=0 ⇨ $E_{exc} = E_r$

⇨ $t(\omega) = 1 + r(\omega)$ ⇨ $d_r = d_t$

Fare from the resonance ⇨ $r = 0, \quad t = 1$ ⇨ $C_r = 0, \quad C_t = 1$

If absorption is absent

$$\frac{|d_r|^2}{(\omega_0 - \omega)^2 + \Gamma^2} + \left| \frac{d_t}{\omega_0 - \omega - i\Gamma} + 1 \right|^2 = 1$$

Solve this square equation

Will get for reflectivity and transmission of QW

$$r(\omega) = \frac{i\Gamma_0}{\omega_0 - \omega - i\left(\Gamma + \Gamma_0\right)}$$

$$t(\omega) = 1 + \frac{i\Gamma_0}{\omega_0 - \omega - i\left(\Gamma + \Gamma_0\right)}$$

Absorption by QW

$$A = 1 - |r|^2 - |t|^2 = \frac{2\Gamma\Gamma_0}{\left(\omega_0 - \omega\right)^2 + \left(\Gamma + \Gamma_0\right)^2}$$

- Quantum well can not be considered as a dielectric media.
- It is a 2D molecule.
- We can’t use usual dielectric function.
- Nonlocal response.

Exciton in a QW

Remaining

QW Exciton Schrödinger equation

$$(H_0^e + H_0^h + H^{eh})\Psi_{exc}(r_e, r_h) = \left(E - E_g - \frac{\hbar^2 K_{II}^2}{2M}\right)\Psi_{exc}(r_e, r_h)$$

$$H_0^e = -\frac{\hbar^2}{2M_{II}^e}\frac{\partial^2}{\partial z^2} + V_e(z_e) \quad (electrons) \qquad H_0^h = -\frac{\hbar^2}{2M_{II}^h}\frac{\partial^2}{\partial z^2} + V_h(z_e) \quad (holes)$$

$$H^{eh} = -\frac{\hbar^2}{2\mu}\left(\frac{\partial^2}{\partial \rho_e^2} + \frac{\partial^2}{\partial \rho_h^2}\right) - \frac{e^2}{\epsilon r} \qquad r = \sqrt{(\rho_e - \rho_h)^2 + (z_e - z_h)^2}$$

2D approximation $\frac{e^2}{r} \approx \frac{e^2}{\rho}$

$$\Psi_{exc}(r_e, r_h) \approx \frac{1}{\sqrt{S}} e^{iK_{II}R_{II}} \varphi(\rho, z_e, z_h), \quad \varphi(\rho, z_e, z_h) = f(\rho)\varphi_e(z_e)\varphi_h(z_h)$$

Radial trial function, 2D hydrogen-like

$$f(\rho) = \sqrt{\frac{2}{\pi a_B^2}} e^{-\rho/a_B} \qquad \rho \equiv \sqrt{(x_e - x_h)^2 + (y_e - y_h)^2}$$

Bloch envelop functions are solutions of electron and hole 1D equations

Exciton photon interaction

System of equation for electromagnetic field and exciton polarization

Wave equation for electromagnetic field $\Delta\vec{E} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{D} = 0$

For plane waves $\frac{d^2}{dz^2}E = -\left(\frac{\omega}{c}\right)^2 D = -\left(\frac{\omega}{c}\right)^2\left[\varepsilon_b E + 4\pi P_{exc}(z)\right]$

$$\Rightarrow \quad \frac{d^2}{dz^2}E(z,t) + k^2 E(z,t) = -k_0^2 4\pi P_{exc}(z,t) \qquad k_0 = \omega/c \qquad k^2 = \varepsilon_b k_0^2$$

Equation for exciton polarization

$$\left[i\hbar\frac{\partial}{\partial t} - \hat{H}_0\right]P_{exc}(z,\omega) = \alpha E(z,\omega) \qquad \alpha = \left(\frac{e|p_{cv}|}{\omega_0 m_0}\right)^2 \frac{1}{\pi a_B^3} = \frac{\hbar\varepsilon_b\omega_{LT}}{4\pi}$$

H_0 - Hamiltonian of the mechanical pert of the exciton

Solutions:

For electromagnetic field

$$E(z,\omega) = E_0(z,\omega)e^{ikz} + 4\pi k_0^2 \int_{-\infty}^{\infty} P_{exc}(z',\omega)G(z,z',\omega)dz'$$

$G(z,z',\omega)$ Is Green function of the wave equation

In our case of 3D it is $\sim e^{iK|z-z'|}$

For the exciton polarization

$$4\pi P_{exc}(z,\omega) = \int \chi(z,z',\omega)E(z',\omega)dz'$$

Here $\chi(z,z',\omega)$ is susceptibility

For QW exciton $$\chi(z,z',\omega)=\pi a_B^3\varepsilon_b\omega_{LT}\sum_n\frac{\Phi_n(z)\Phi_n^*(z')}{\omega_n^0-\omega-i\Gamma_n}$$

Consider lowers exciton states

$$\chi(z,z',\omega)=\pi a_B^3\varepsilon_b\omega_{LT}\frac{\Phi(z)\Phi^*(z')}{\omega_0-\omega-i\Gamma}$$

Substituting P_{exc} into eq. for E , we obtain

$$E\left(z,\omega\right)=E_0e^{ikz}+2\pi i\frac{k_0^2}{k}\int dz'e^{ik|z-z'|}P_{exc}\left(z',\omega\right)$$

Taking into account

$$r=\left.\frac{E(z)-E_0(z)e^{ikz}}{E_0(z)e^{-ikz}}\right|_{z\to\infty}\qquad t=\left.\frac{E(z)}{E_0(z)e^{ikz}}\right|_{z\to\infty}$$

Reflection and Transmission coefficients

$$r(\omega) = \frac{i\Gamma_0}{\tilde{\omega}_0 - \omega - i(\Gamma + \Gamma_0)}$$

$$t(\omega) = \frac{\tilde{\omega}_0 - \omega - i\Gamma}{\tilde{\omega}_0 - \omega - i(\Gamma + \Gamma_0)}$$

here

$$\Gamma_0 = \frac{2\pi k_0}{\hbar\varepsilon_b}\left[\frac{e|p_{cv}|}{m_0\omega_0}\right]^2 \left[\int \Phi(z)\cos(kz)\,dz\right]^2$$

Overlap of the EM field with electron and hole wavefunctions

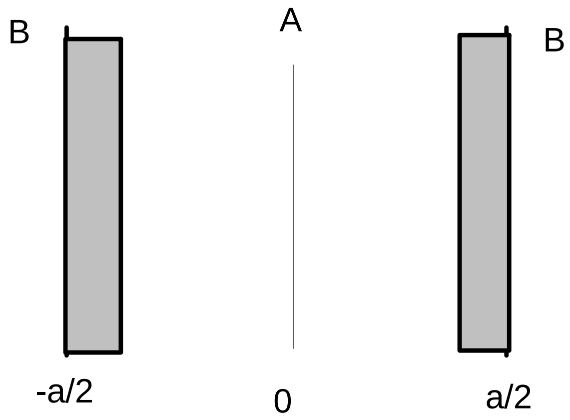
$$\tilde{\omega}_0 = \omega_0 + \frac{1}{2}k\omega_{LT}\pi a_B^3 \iint dz\,dz' \sin k|z - z'|\,\Phi(z)\Phi(z')$$

Renormalization of the resonant frequency

To describe exciton photon interaction we have to consider equation for the exciton polarization induced by the electromagnetic field and equation for the electromagnetic field induced by exciton polarization.

Local respond instead of nonlocal

Nonlocal $= P_{exc}(z) = \int dz' \chi(z,z') E(z')$



Introduce formally dielectric function

$$\varepsilon(\omega, z) = \begin{cases} \varepsilon_A(\omega) & |z| < a/2 \\ \varepsilon_B & |z| > a/2 \end{cases}$$

ω_{LT}^{eff} effective oscillator strength

$$\varepsilon_A(\omega) = \varepsilon_0 + \frac{\varepsilon_0 \omega_{LT}^{eff}}{\omega_0 - \omega - i\Gamma}$$

Reflectivity from a plate A

Summarizing all multiple reflections in **A** ⟹ $r_A = \frac{1-e^{2i\phi}}{1-e^{2i\phi}r_{BA}^2} e^{-i\phi} r_{BA}$

here $r_{BA} = \frac{n_B - n_A}{n_B + n_A}, \quad n_B = \sqrt{\varepsilon_B}, \quad n_A = \sqrt{\varepsilon_A}$

$\phi = k_A a$ phase shift in the layer **A**

In the case if $\omega_{LT}^{eff} << \Gamma$

$$r_A = \frac{i}{2} \frac{\omega_{LT}^{eff} \sin \Phi}{\omega_0 - \omega - i\Gamma}$$

consequently $\omega_{LT}^{eff} = \frac{2\Gamma_0}{\sin k_0 a} \approx \frac{2\Gamma_0}{k_0 a}$

Reflectivity from a real structures

Summarizing multiple contributions from surface and QW

$$r = r_{01} + \frac{t_{01}t_{10}e^{2i\phi}}{1 - r_{10}r_{QW}e^{2i\phi}} r_{QW} \qquad \text{here} \qquad \phi = kb + k(a/2)$$

$$R = \frac{r_{01}^2 + 2\,\mathrm{Re}\left\{r_{01}r_{QW}e^{2i\phi}\right\} + \left|r_{QW}\right|^2}{1 + 2\,\mathrm{Re}\left\{r_{01}r_{QW}e^{2i\phi}\right\} + r_{01}^2\left|r_{QW}\right|^2}$$

$$R = \bar{R}\left[1 + 2\frac{t_{01}t_{10}}{r_{01}}\mathrm{Re}\left\{r_{QW}e^{2i\phi}\right\}\right]$$

$$\bar{R} = r_{01}^2 = \left(\frac{n-1}{n+1}\right)^2, \quad \frac{t_{01}t_{10}}{r_{01}} = \frac{4n}{n^2-1}$$

Reflection from quantum well defined by interference of the light reflected from the surface and from the quantum well

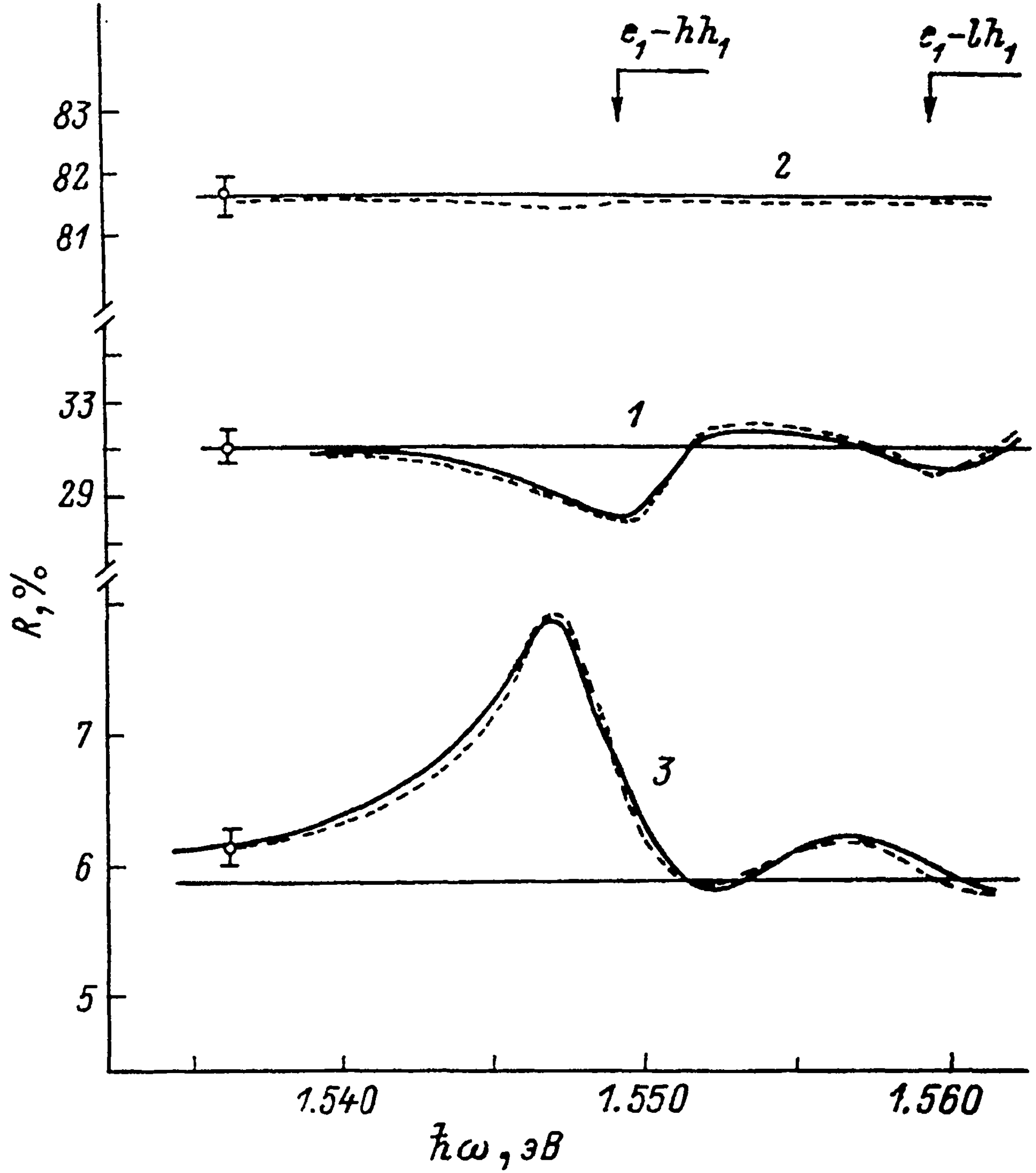
Oblique incidence

$$R = \bar{R}(\varphi_0)\left[1 + S(\varphi_0)\, f(x,\Phi)\right]$$

$$S(\varphi_0) = \frac{8\sqrt{\varepsilon_0 - \sin^2\varphi_0}}{\varepsilon_0 - 1}\begin{cases} \cos\varphi_0 & -s \\ \dfrac{\varepsilon_0 \cos\varphi_0}{\varepsilon_0 \cos^2\varphi_0 - \sin^2\varphi_0} & -p \end{cases}$$

$$f(x,\Phi) = \frac{\omega_{LT}^{eff}}{\Gamma}\,\frac{\sin\Phi + x\cos\Phi}{1 + x^2}$$

here $\quad x = \dfrac{(\omega - \omega_0)}{\Gamma}, \qquad \Phi = 2\Phi_1 + \Phi_2 + \dfrac{\pi}{2}$



$e_1 - hh_1$
$e_1 - lh_1$
83
82
81
2
33
29
1
R, %
7
6
5
3
1.540
1.550
1.560
$\hbar\omega$, эВ

Using oblique geometry we can suppress reflectivity from the surface and by this way increase the contribution from the QW

Exciton photon interaction as a function of QW with

$$\Gamma_0 = \frac{2\pi k_0}{\hbar\varepsilon_b}\left[\frac{e|p_{cv}|}{m_0\omega_0}\right]^2\left[\int \Phi(z)\cos kz dz\right]^2$$

Narrow wells $ak \ll 1$ or $a \ll \lambda$

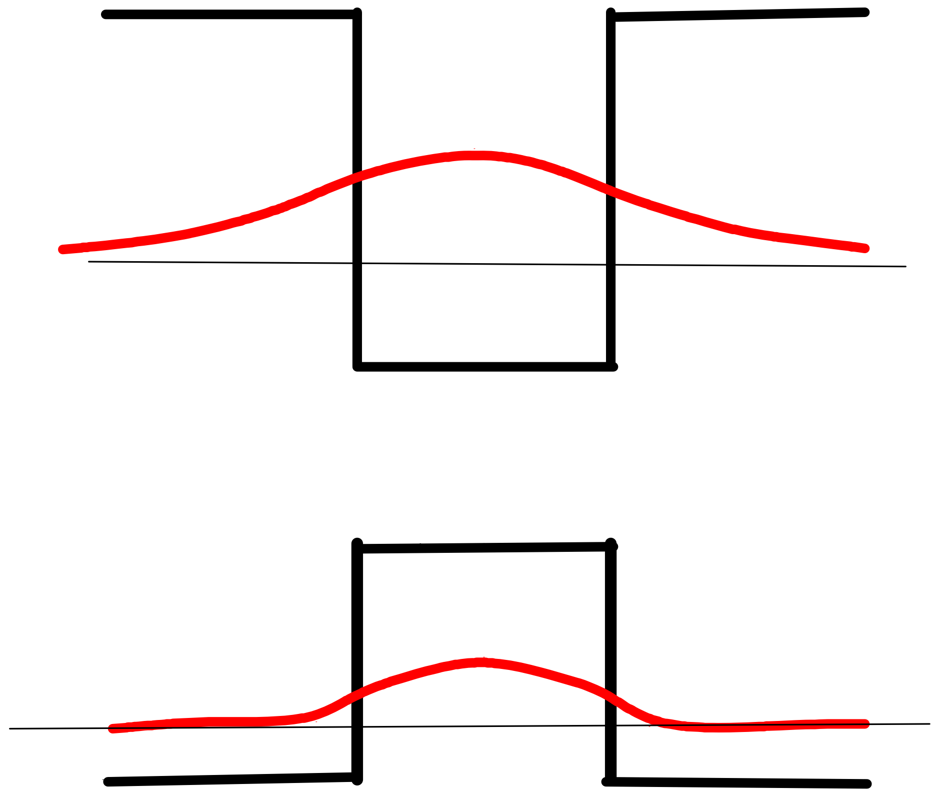
$$\int_{-\infty}^{\infty}\Phi(z)\cos kz dz \rightarrow \int \Phi(z)dz$$

$$\Gamma_0 = \frac{1}{2}k\omega_{LT}\pi a_B^3\left[\int \Phi(z)dz\right]^2$$

Using trial function $f(\rho)$ $\Rightarrow$ $\int \Phi(z)dz = \sqrt{\frac{2}{\pi}}\frac{1}{a}I_{11}^0$

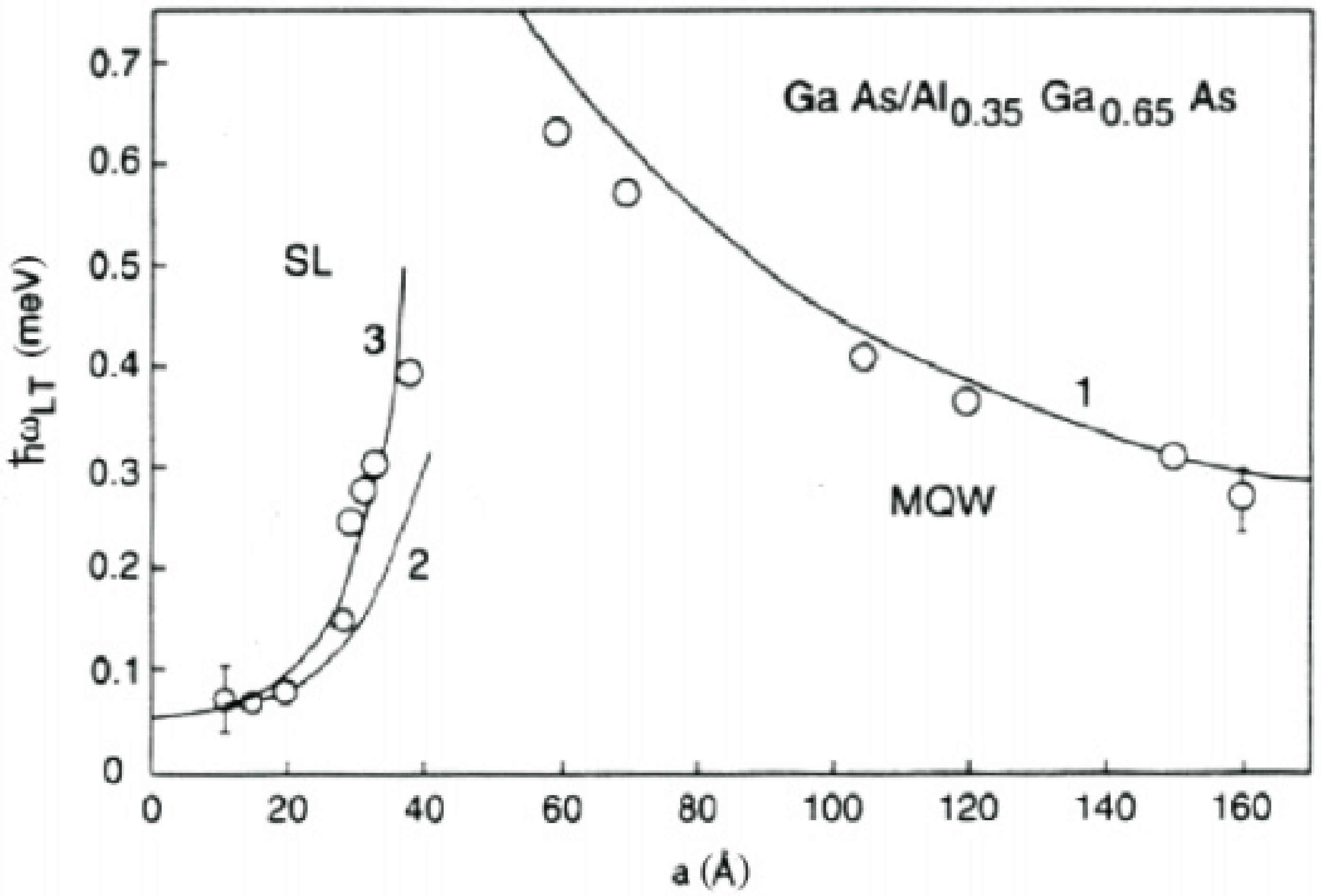
$$I_{11}^0 = \int \varphi_e(z)\varphi_h(z)dz$$

Overlap of electron and hole functions



Exciton oscillator strength depends on the nanostructure size

Oscillator strength as a function of the QW width



Short remarks related to oscillator strength

QW exciton wavefunction

$$\Phi_{2D} = \left(\frac{1}{\sqrt{N_{2D}}}\right) e^{iK_{\|}R_{\|}} f_{2D}(\boldsymbol{\rho})\varphi_e(z_e)\varphi_h(z_h)u_e(\mathbf{r}_e)u_h(\mathbf{r}_h)$$

Radiative damping

$$\Gamma_0 \sim \left| f_{2D}(0)\right| \left[\int \varphi_e(z_e)\,\varphi_h(z_h)\cos kz dz\right]^2 \qquad \text{here} \qquad \left| f_{2D}(0)\right|^2 = \frac{1}{\pi a_0^2} \equiv \frac{1}{A_x}$$

Oscillator strength f_x^{2D}: $\Gamma_0 = N\, f_x^{2D}$

Because all unit cells can participate in absorption $N = A \cdot L_z$

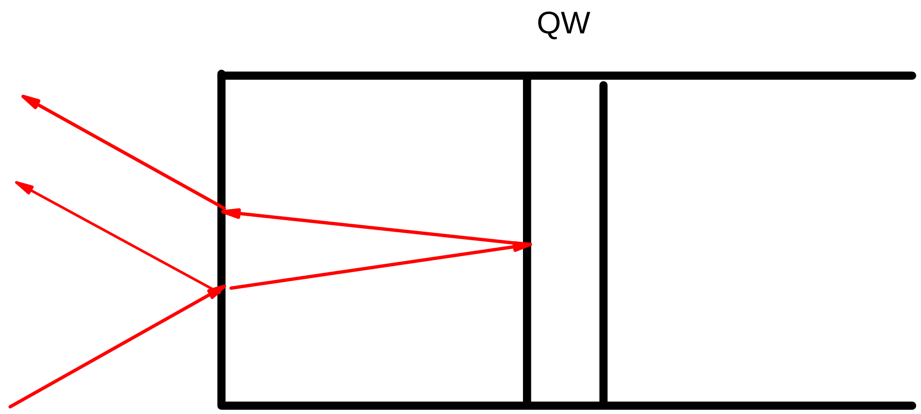
Here A is square of the sample $\Rightarrow$ $\Gamma_0^{2D} \sim \left| p_{cv}\right|^2 \dfrac{A}{A_x}$

For the bound (or localized) exciton $\Gamma_0^{Bx} \sim \dfrac{A_{Bx}}{A_x}$

The oscillator strength of the bound exciton normalized on one impurity (or localization) center is equal to the oscillator strength of the free exciton normalized on unit cell.

The exciton radiative lifetime is equal to the inverse exciton radiative damping.
It is maximal for bulk and minimal for quantum dots

Oscillator strength as a function of the environment



$$r = r_{01} + \frac{t_{01}t_{10}e^{2i\varphi}}{1 - r_{01}r_{QW}e^{2i\varphi}} r_{QW}$$

Phase shift $\varphi = n_0 \frac{\omega}{c}(b + \frac{a}{2})$

let $$r'_{QW} \equiv \frac{r_{QW}}{1 - r_{01} r_{QW} e^{2i\varphi}}$$

⇩

$$r'_{QW} = \frac{i\Gamma'_0}{\omega'_0 - \omega - i(\Gamma + \Gamma'_0)}$$

$$\omega'_0 = \omega_0 + r_{10} \sin 2\varphi$$
$$\Gamma'_0 = \Gamma_0 \left(1 + r_{01} \cos 2\varphi\right)$$

$$r_{01} = \frac{n-1}{n+1} \sim \frac{3-1}{3+1} = 0.5 \quad \Rightarrow \quad \Gamma_0 \text{ from } 0.5\Gamma_0 \text{ to } 1.5\Gamma_0$$

Exciton oscillator strength depends on the environment

Exciton reflectivity line-shape as a function of the environment

reflection $$R(\omega) = |r(\omega)|^2 = R_0 + \frac{A + Bx}{1 + x^2}$$

here $$x = \frac{\omega - \omega_0}{\Gamma}, \qquad R_0 = |r_{01}|^2$$

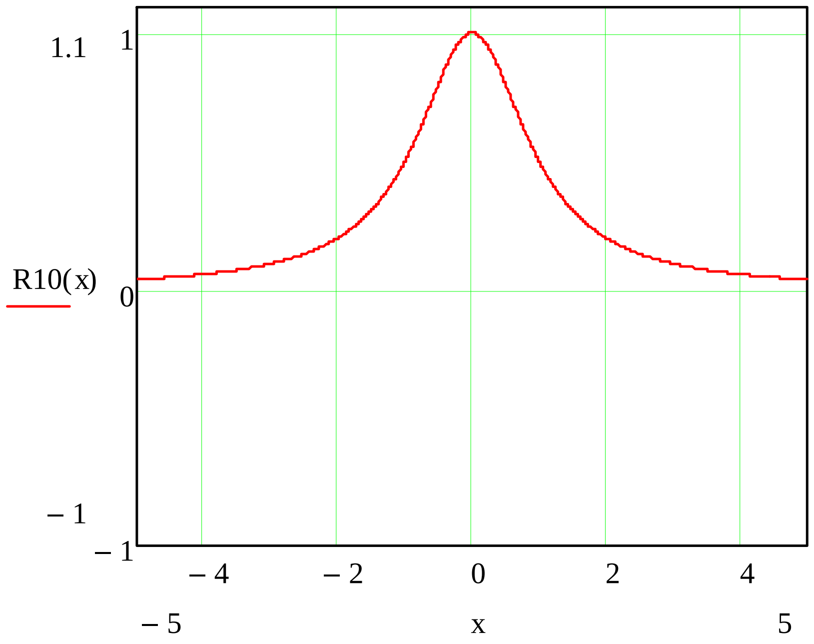
$$A = t_{01} t_{10} s \left[t_{01} t_{10} s - 2 r_{01} (1 - s') \cos 2\varphi \right]$$

$$B = 2 r_{01} t_{01} t_{10} s \sin 2\varphi \qquad s \equiv \frac{\Gamma_0}{\Gamma}, \quad s' \equiv \frac{\Gamma_0'}{\Gamma}$$

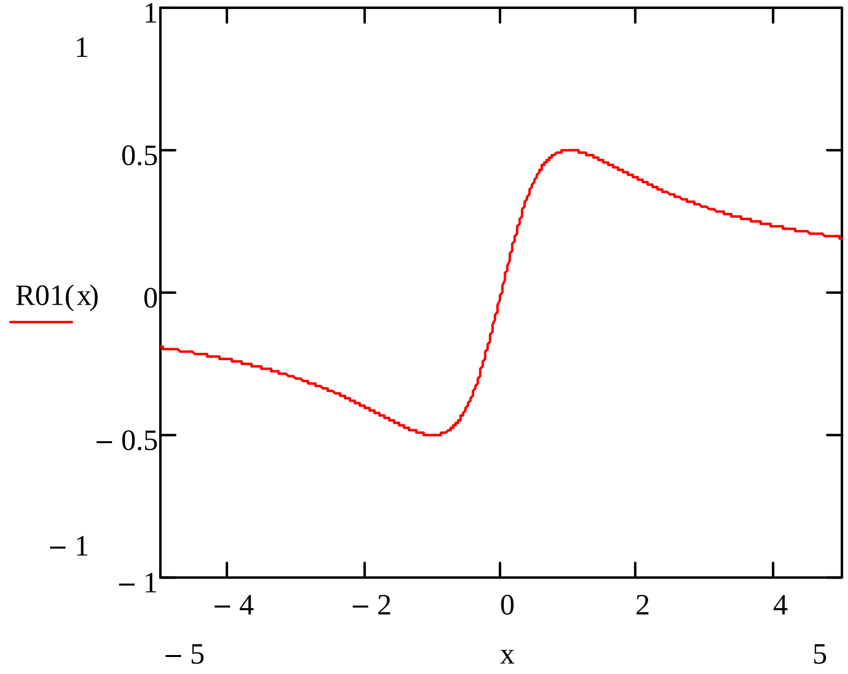
At normal incidence $$r_{01} = -r_{10} = \frac{n_b - 1}{n_b + 1}, \quad t_{01} t_{10} = \frac{4 n_b}{(n_b + 1)^2}$$

consider $$R(x) = R_0 + \frac{A + Bx}{1 + x^2}$$

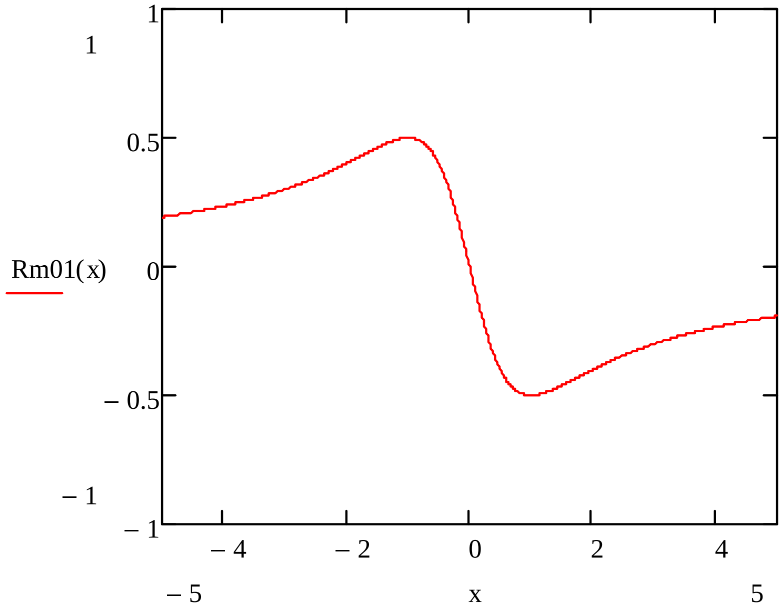
$$A > 0, \quad B = 0$$



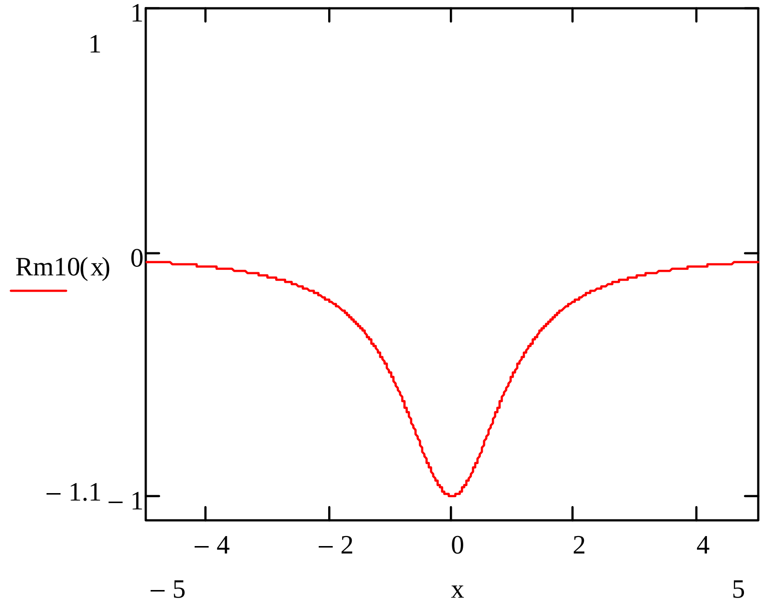
$$A = 0, \quad B > 0$$

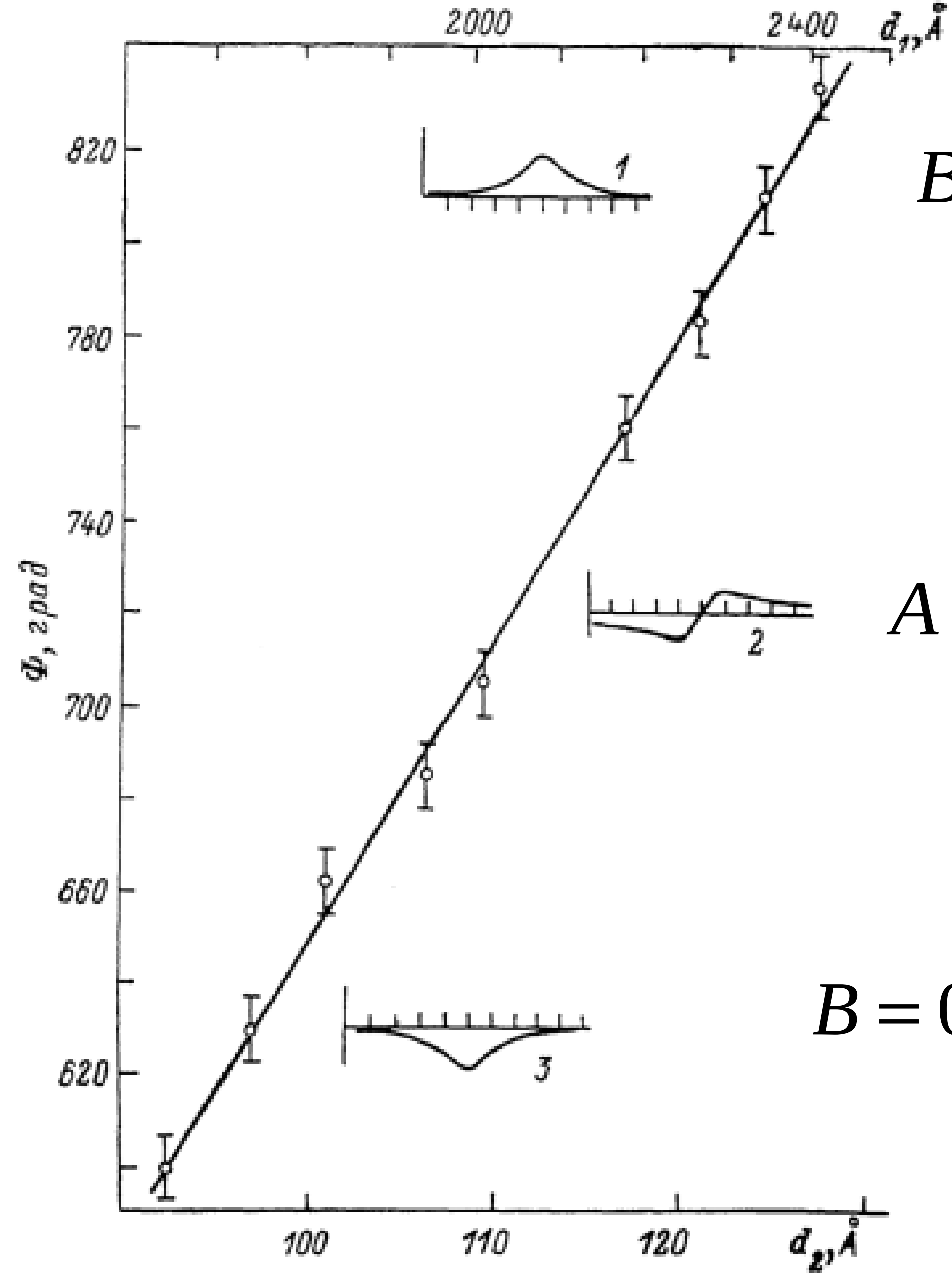


$$A = 0, \quad B < 0$$



$$A < 0, \quad B = 0$$





$B = 0, \quad A > 0$

$A = 0, \quad B > 0$

$B = 0, \quad A < 0$

Exciton line-shape depends strongly on the cap layer thickness