

## Lecture 5

### 4). Exciton photoluminescence (1h)

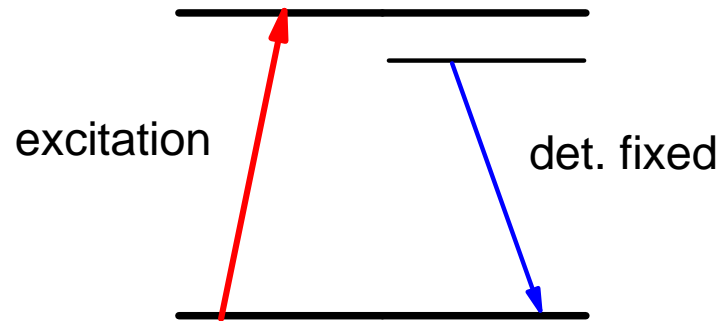
(exciton lifetime in bulk, QW, QWW and QD, homogeneous linewidth, interaction with phonons)

### 5). Optical orientation and alignment (1h)

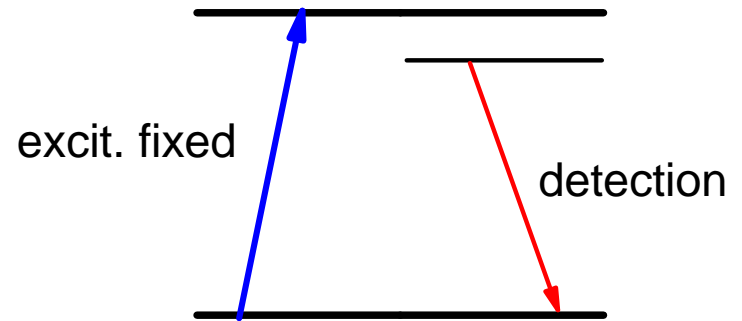
(the idea of the optical orientation; mechanisms of exciton spin relaxation, the alignment of the exciton dipole momenta, polarization conversion)

# Photoluminescence

Photoluminescence excitation



Photoluminescence



- Normally the exciton PL spectrum determine by recombination of localized exciton states.
- The main mechanism for the localization is monolayer fluctuation of the interfaces.

## Delocalized states

At high temperature excitons become delocalized

$$f(K_{\parallel}) = C \exp(-\hbar^2 K_{\parallel}^2 / 2Mk_B T)$$

here  $C \propto T^{d/2} N$        $N$  – Number of excitons

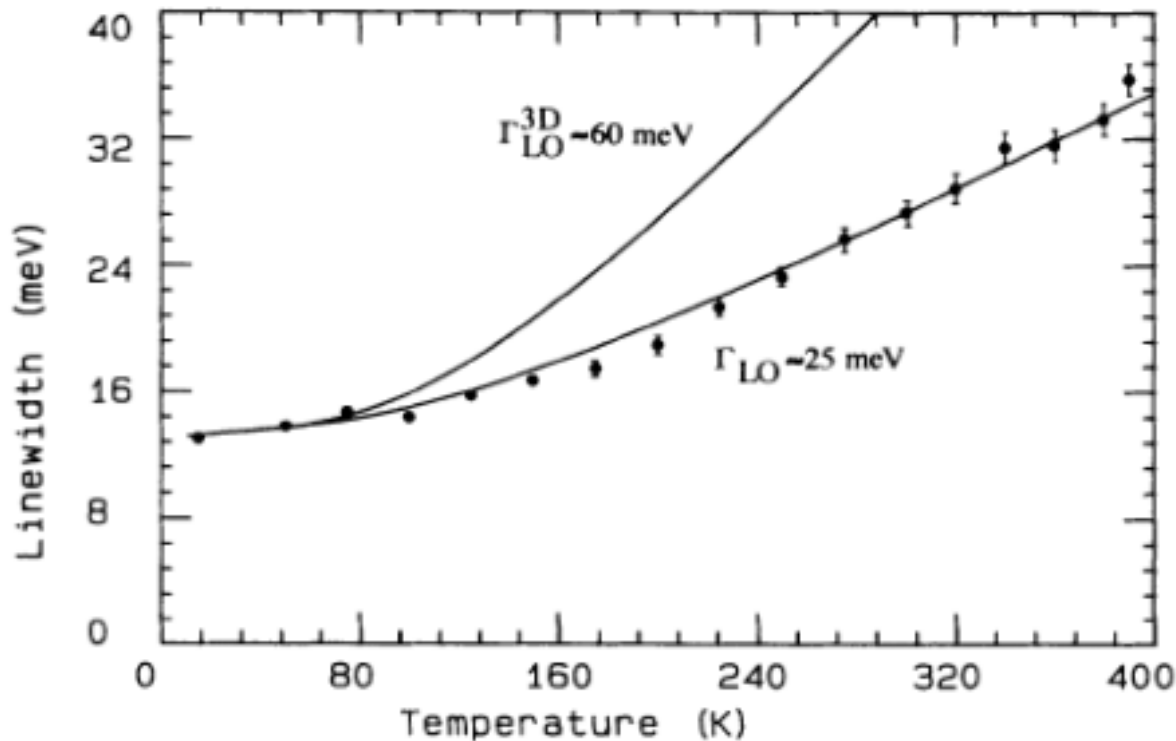
Optically active are only excitons with  $K_{\parallel} < (\omega_0 / c) \sqrt{\epsilon_b}$

The excitons with  $K_{\parallel} > (\omega_0 / c) \sqrt{\epsilon_b}$  are out of the light cone

# Efficiency of the radiative recombination

$$\langle \tau_r^{-1} \rangle = \frac{\int d\mathbf{K}_{\parallel} \tau_r^{-1}(K_{\parallel}) f(K_{\parallel})}{\int d\mathbf{K}_{\parallel} f(K_{\parallel})} \propto \frac{C}{N} \propto T^{d/2}$$

Homogeneous linewidth at high temperature



Line width QWs

$$\frac{\Delta E}{E} \propto 2 \frac{\Delta L}{L}$$

# Resonant fluorescence and Raman scattering

Two level systems. Spectral intensity:

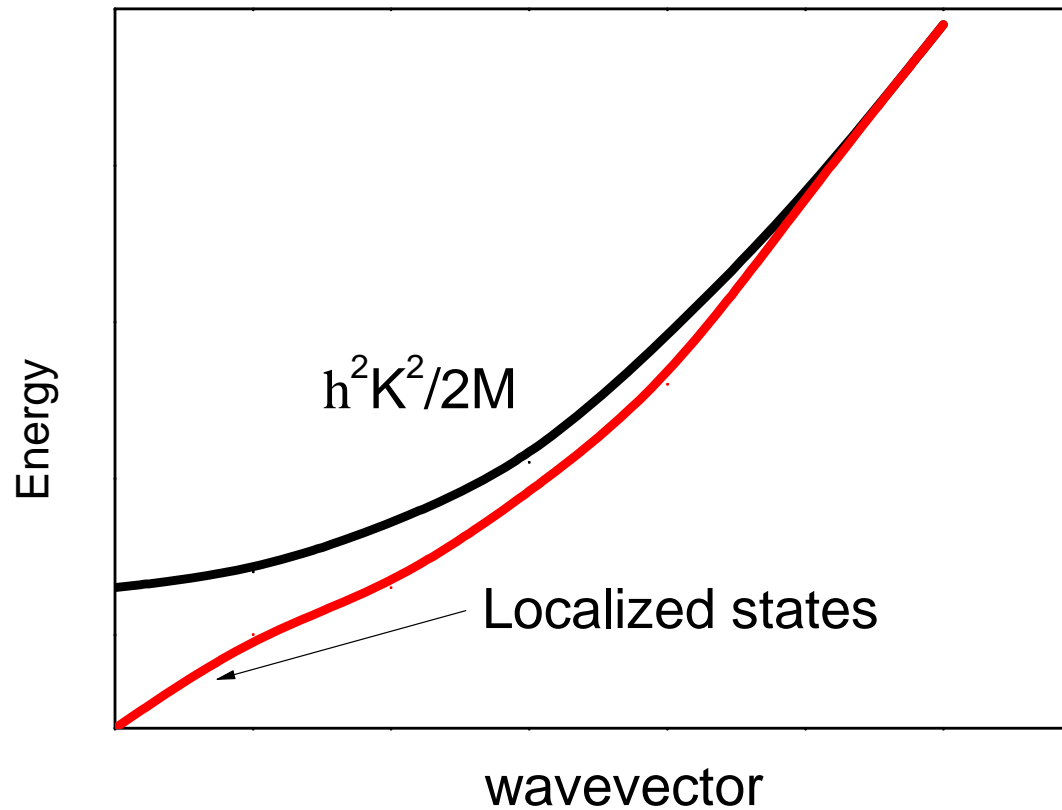
$$I(e_2, \omega_2 | e_1, \omega_1) \propto |M|^2 \delta(\omega_2 - \omega_1)$$

$$M \propto \frac{(\vec{e}_2^* \vec{d}_{if})(\vec{e}_1 \vec{d}_{fi})}{\omega_0 - \omega - i\Gamma} E_0$$

$$\Gamma = \frac{1}{2\tau_f} \quad \tau_f \text{ is the lifetime of the excited state}$$

- Delocalized excitons can recombine radiatively only inside light cone  $K < (\omega/c)\sqrt{\epsilon_b}$
- At high temperature excitons delocalized and linewidth becomes homogeneous
- At resonant excitation two processes are undistinguishing: Raman scattering and resonant fluorescence

# Localized states



- There are jumps of excitons over these states
- The PL spectrum forms due to competition between recombination and relaxation



## Photoluminescence, Stokes shift

Stokes shift:  $E = E_0 - \mathcal{E}$  ; Mobility edge  $E_0$

Mobility edge separates extended and localized states

1. The density of localized states is exponential  
(number of shallow wells is large)

$$g(\mathcal{E}) = g_0 \exp(-\mathcal{E} / \mathcal{E}_0)$$

2. Exciton hopping transfer rate between localized states  
(probability of the jump defined by distance only)

$$\omega(\mathcal{E}, \mathcal{E}', r) = \theta(\mathcal{E} - \mathcal{E}') \omega_h(r)$$

3. The rate  $\omega_h(r)$  is independent of  $\mathcal{E}$  and  $\mathcal{E}'$ , and decays exponentially because it is the tale of wavefunction

$$\omega_h(r) = \omega_0 e^{-2r/L}$$

The PL peak  $E_{\max} = E - \mathcal{E}_{\max}$  and the delay time  $t$  (at pulse excitation) is

$$\tau_{\mathcal{E}_{\max}} = \omega_0^{-1} \exp(2\bar{r}_\varepsilon / L) = t$$

$$\mathcal{E}_{\max}(t) = \varepsilon_0 \ln \left[ g_0 \varepsilon_0 V_d (L/2) \ln(\omega_0 t) \right] = d\varepsilon_0 \ln \ln(\omega_0 t) + A$$

where  $A = \varepsilon_0 \ln \left[ g_0 \varepsilon_0 V_d (L/2) \right]$

At CW excitation  $t \leftrightarrow \tau_0$  recombination time

$$\mathcal{E}_{\max}^{cw}(t) = d\varepsilon_0 \ln \ln(\omega_0 \tau_0) + A$$

4. The recombination time is independent of  $\mathcal{E}$

Concentration of the states with energy exceeds  $\mathcal{E}$  is

$$\rho(\mathcal{E}) = \int_{\mathcal{E}}^{\infty} d\mathcal{E}' g(\mathcal{E}') = g_0 \mathcal{E}_0 e^{-\mathcal{E}/\mathcal{E}_0}$$

Localized exciton onto the site  $\mathcal{E}$  hops within  $\bar{r}_{\mathcal{E}}$

Average distance  $\bar{r}_{\mathcal{E}}$  between localized states with  $\mathcal{E}' > \mathcal{E}$

$$V_d(\bar{r}_{\mathcal{E}}) \rho(\mathcal{E}) = 1$$

Lifetime with respect to hopping

$$\tau_{\mathcal{E}} = \omega_h^{-1}(\bar{r}_{\mathcal{E}}) = \omega_0 e^{2\bar{r}_{\mathcal{E}}/L}$$

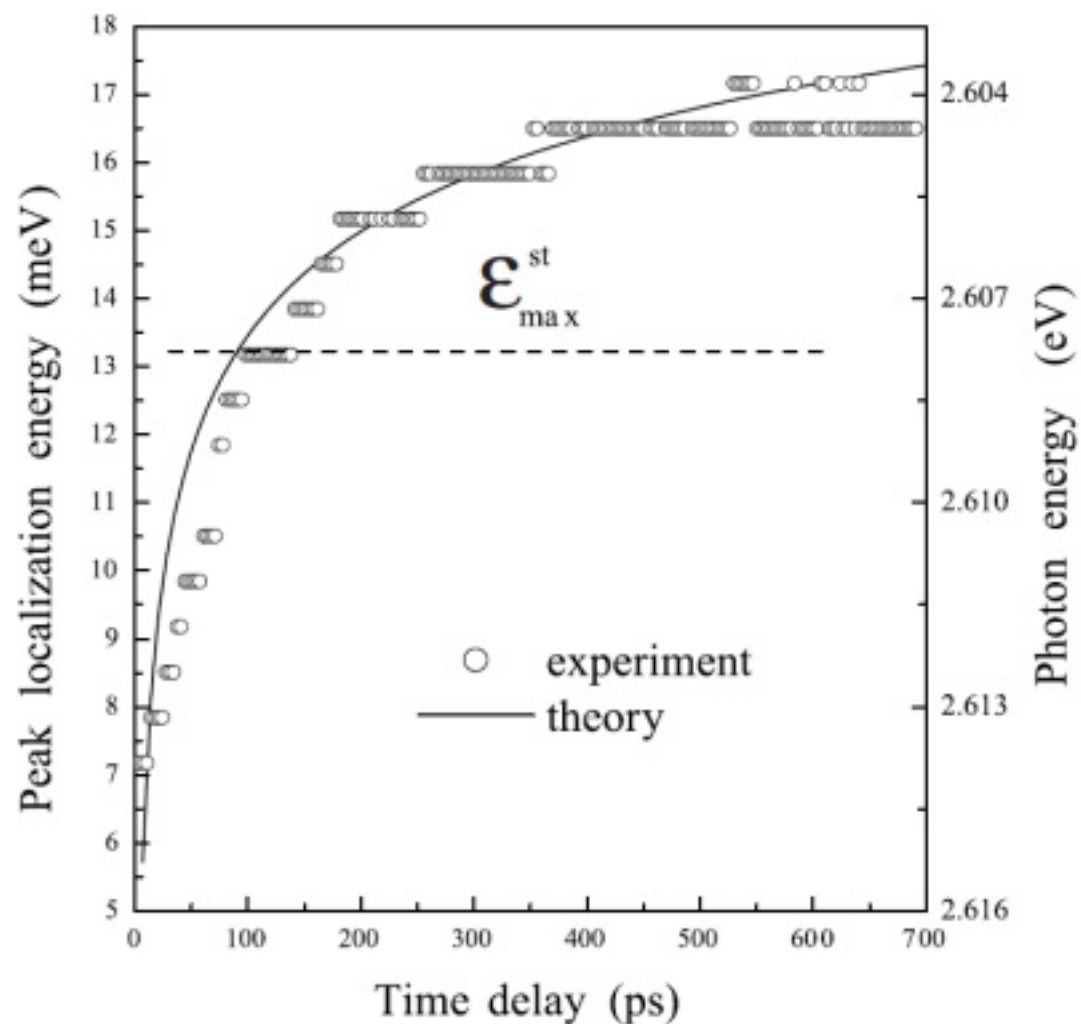


Fig. 5.1. The temporal shift of the PL peak measured at  $T = 2$  K in the  $20 \text{ \AA}$   $\text{Zn}_{0.8}\text{Cd}_{0.2}\text{Se}/\text{ZnSe}$  QW and calculated from (5.30) with the values of parameters indicated in the text. The dashed horizontal line indicates the position of the PL peak under steady-state excitation. [5.3]

- In the PL always Stokes shift
- The Stokes shift defined by competition between Relaxation and recombination

# Principle of electron optical orientation

1). At interband optical transitions the angular momentum of photon transforms into the angular momentum of exciton

$$\frac{n_+}{\tau_0} + \frac{1}{2\tau_s} (n_+ - n_-) = G_+$$

$$\frac{n_-}{\tau_0} + \frac{1}{2\tau_s} (n_- - n_+) = G_-$$

Here  $G_{\pm}$  is generation rate

$\tau_0$  is lifetime

$\tau_s$  is spin relaxation time

Notify	$n = n_+ + n_-$	Total number of electrons
	$G = G_+ + G_-$	Generation rate
	$s_z = (n_+ + n_-) / 2$	Average spin
	$p = \frac{n_+ - n_-}{n_+ + n_-}$	Degree of polarization
	$S_z = s_z / n = p / 2$	Average spin of one electron

The solution of the equations

$$p = p_0 \left( \frac{T}{\tau_0} \right)$$

$$p_0 = \frac{G_+ - G_-}{G_+ + G_-}, \quad \frac{1}{T} = \frac{1}{\tau_0} + \frac{1}{\tau_s} \quad T \text{ is spin lifetime}$$

2). Oriented spins conserve their polarization (in part)  
at a steady state pumping

$$S_z = \frac{\tau_s}{\tau_0 + \tau_s} S_z^0, \quad S_z^0 = \frac{1}{2} p_0$$

The initial spin polarization is proportional to the polarization of excited light  $p_0 = \kappa P_c^0$

$\kappa$  Depends on the selection rules

For bulk GaAs  $\kappa = -1/2$

For QW  $\kappa = -1$



3). Due to the same selection rules the photoluminescence is partially polarized

$$P_c = \kappa p = \kappa^2 P_c^0 \frac{T}{\tau_0}$$

4). Transverse magnetic field lead to depolarization of the PL (Hanle effect)

The kinetic equations  $\frac{n}{\tau_0} = G, \quad \left( \frac{1}{\tau_0} + \frac{1}{\tau_s} \right) \mathbf{s} + \mathbf{s} \times \boldsymbol{\Omega}_L = \dot{\mathbf{s}}$

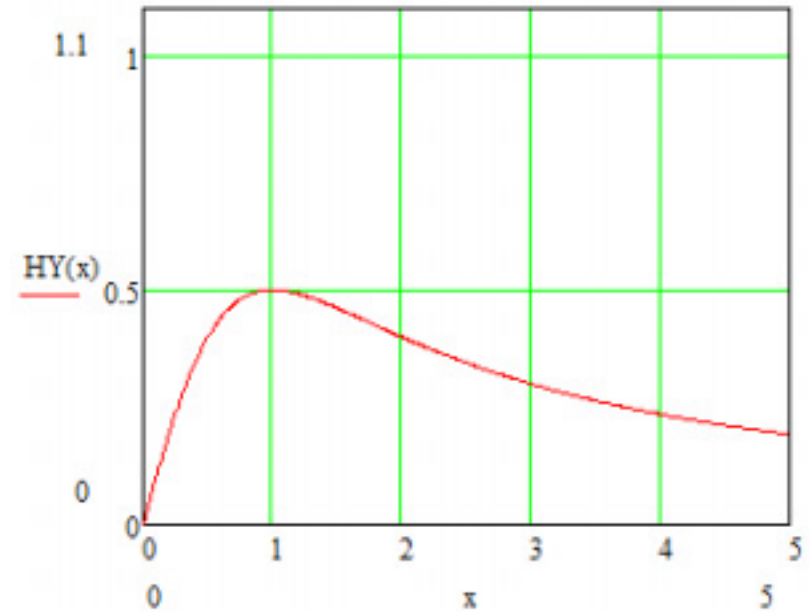
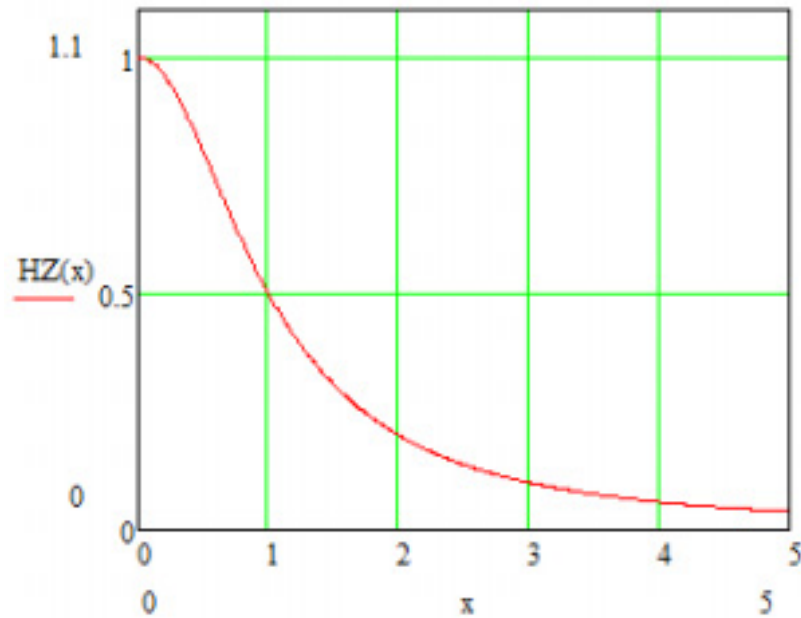
$\boldsymbol{\Omega}_L$  Is Larmour frequency

$$\frac{s_x}{T} = \dot{s}_x, \quad \frac{s_y}{T} + \boldsymbol{\Omega}_L s_z = \dot{s}_y, \quad \frac{s_z}{T} - \boldsymbol{\Omega}_L s_y = \dot{s}_z$$

# Hanle Effect

$$s_z(B) = \frac{s_z(0)}{1 + (\Omega_L T)^2}$$

$$s_y(B) = \frac{\Omega_L T}{1 + (\Omega_L T)^2} s_z(0)$$



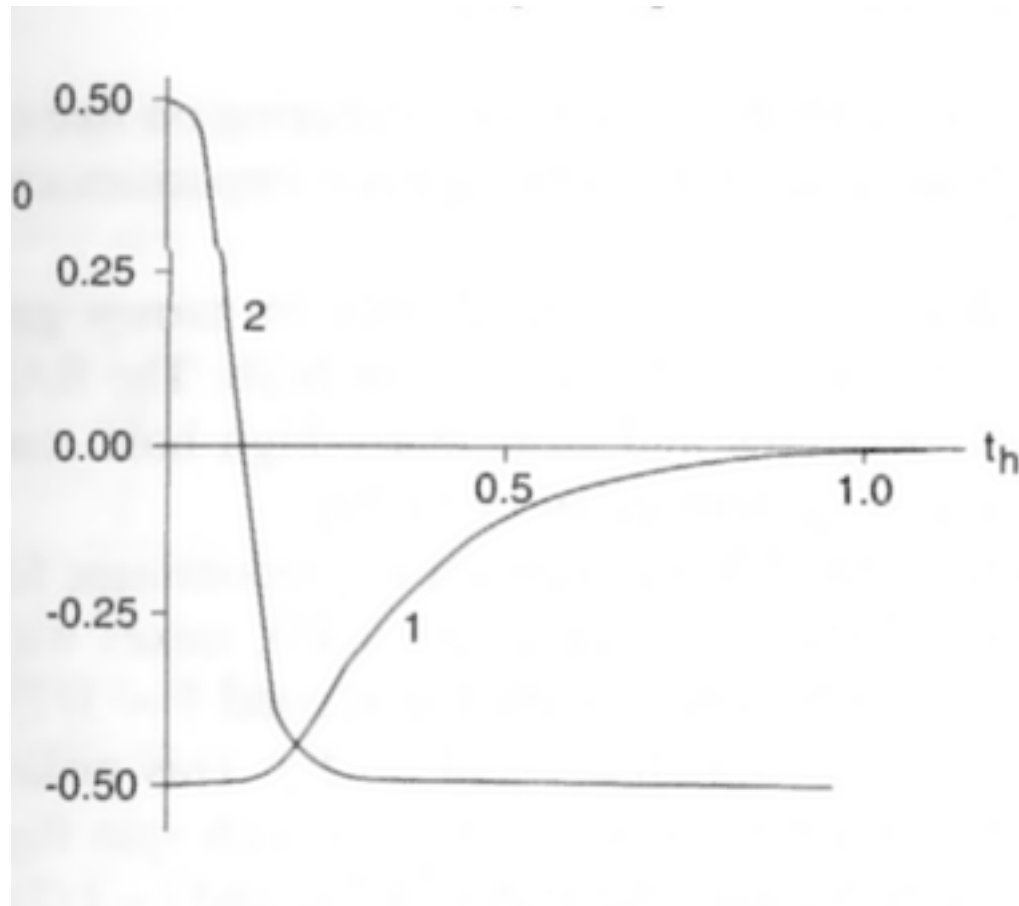
- In optical orientation an angular momentum of the photon transfer to the electron
- Effect Hanle is a stroboscopic effect and allows to determine lifetime and spin relaxation time in CW experiment
- This method is used for optically detected paramagnetic resonance, optically detected nuclear magnetic resonance, optical cooling, etc.

# Electron spin relaxation mechanisms

- 1). Elliott – Yaffet (EY) mechanism. Mixing opposite spin states from other bands by  $(\mathbf{K} \cdot \mathbf{p})$  perturbation
- 2). Beer-Ahronov-Pikus (BDP). Electron hole scattering with spin flip
- 3). D'yakonov and Perel (DP). Precession of the electron spin in an effective quasi-magnetic field without inversion symmetry
- 4). Fluctuation of nuclear spins in QDs

- EY is important in narrow gap semiconductors like InSb  
In QWs EY is important for holes.
- BAP is important for doped structures at low temperatures.
- DP is important in wide array of optical nanostructures

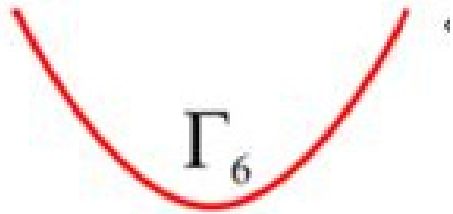
# Dependence of the main spin on the excitation energy in QWs



**Fig. 9.7.** Dependence of the mean spin of excited electrons on the quantity  $t_h = k_{\perp}^2 / (k_{\perp}^2 + k_z^2)$  for transitions to the first electronic level from the lowest levels of heavy (curve 1) and light (curve 2) holes (after [9.32])

Remain the band structure of cubic semiconductors

In the conductivity band  
(diamond)

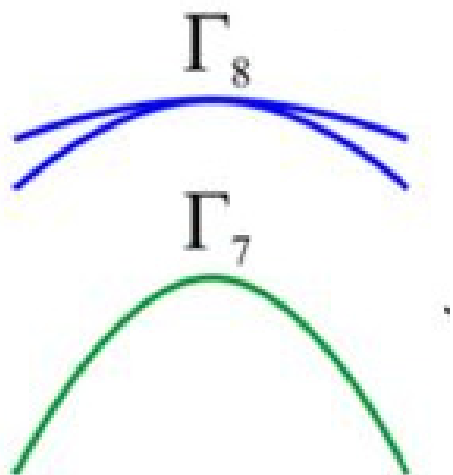


$$\varphi_{\mathbf{k},m}^e = u_m e^{i\mathbf{k}\mathbf{r}}, \quad m = \pm 1/2$$

$$u_{1/2} = S(r) \uparrow, \quad u_{-1/2} = S(r) \downarrow$$

At  $\Gamma$  point

$$\mathcal{E}_e(k) = \hbar^2 k^2 / 2m_e$$



No inversion symmetry

$$\mathcal{E}_e(k) = \frac{\hbar^2 k^2}{2m_e} + \frac{\hbar^2}{2} (\vec{\sigma} \vec{\Omega})$$

$$\Omega_x(\mathbf{k}) = \alpha \hbar^2 \left( m_e \sqrt{2m_e E_g} \right)^{-1} k_x (k_y^2 - k_z^2)$$

Electron Hamiltonian includes linear term in QWs

$$H_s(k) = \frac{1}{2} \vec{\sigma} \cdot \vec{\Omega}_1$$

$\Omega_1 \equiv \Omega_1(-\cos \phi, \sin \phi, 0)$        $\phi$  is an angle between  $k$  and  $x$

$$\hbar\Omega_1 = 2\gamma k \left( \langle k_z^2 \rangle - \frac{1}{2} k^2 \right)$$



## Spin relaxation time

$$\frac{1}{\tau_{s,zz}^e} = \frac{2}{\tau_{s,xx}^e} = \frac{2}{\tau_{s,yy}^e} = \Omega_1^2 \tau_1$$

$$\frac{1}{\tau_{s,ij}^e} = 0$$

$$\Omega_1 \tau_p \ll 1$$

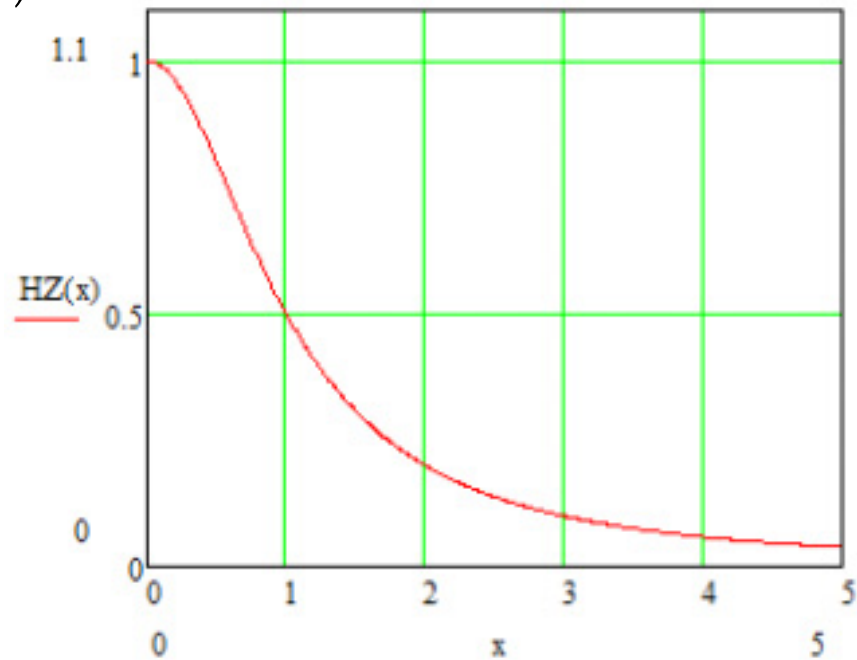
$$s_z(t) = s_z(0) \exp\left(-\frac{t}{\tau_s}\right)$$

Taking average on magnetic field

# Suppression of the spin relaxation in longitudinal magnetic fields

$$\frac{1}{\tau_{s,zz}^e} = \frac{\Omega_1^2 \tau_p}{1 + (\omega_c \tau_p)^2}$$

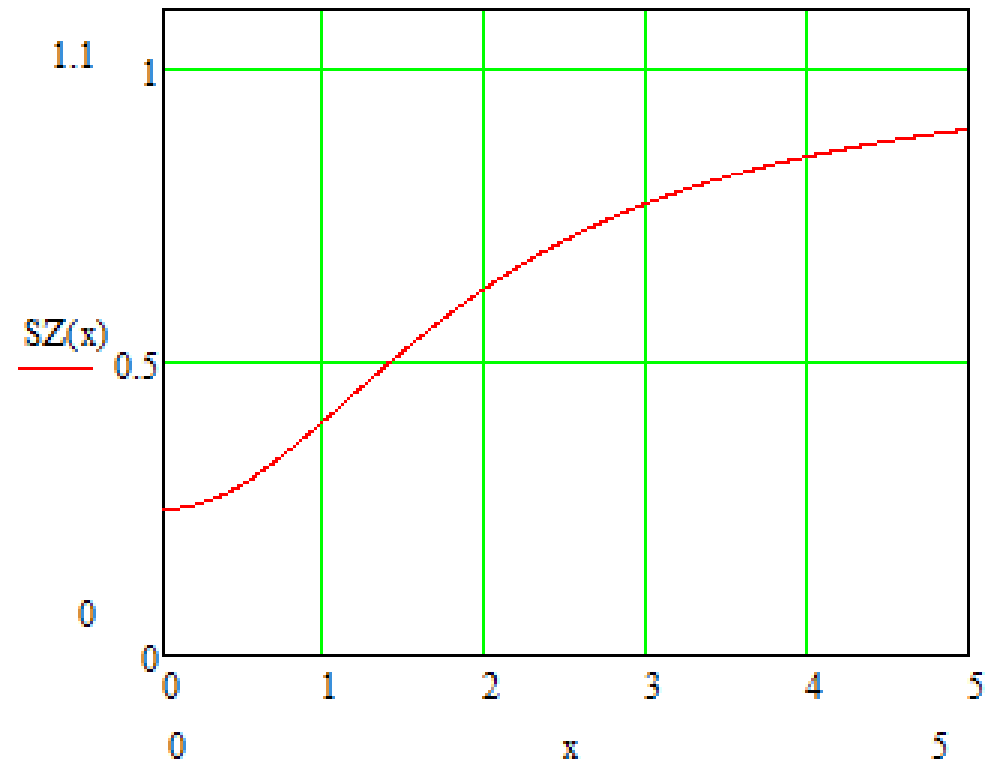
$$\left(\tau_{s,zz}^e\right)^{-1} \quad \omega_c = eB / mc$$



Magnetic field

## Optical orientation in the longitudinal magnetic field

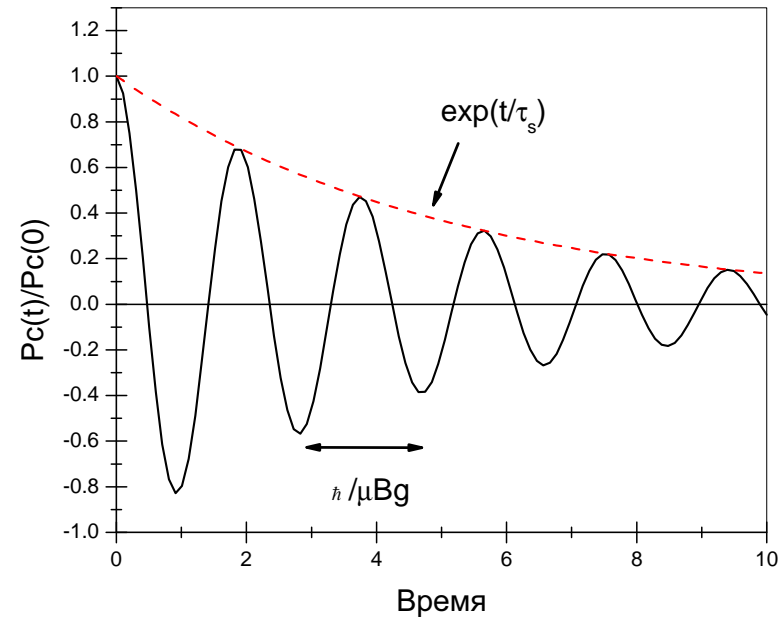
$$s_z = \frac{\tau_s}{\tau_0 + \tau_s} s_z(0)$$



# Spin quantum bits

$$\vec{S}(t) = \vec{S}(0) \cos(\omega_L t) e^{-t/\tau_s}$$

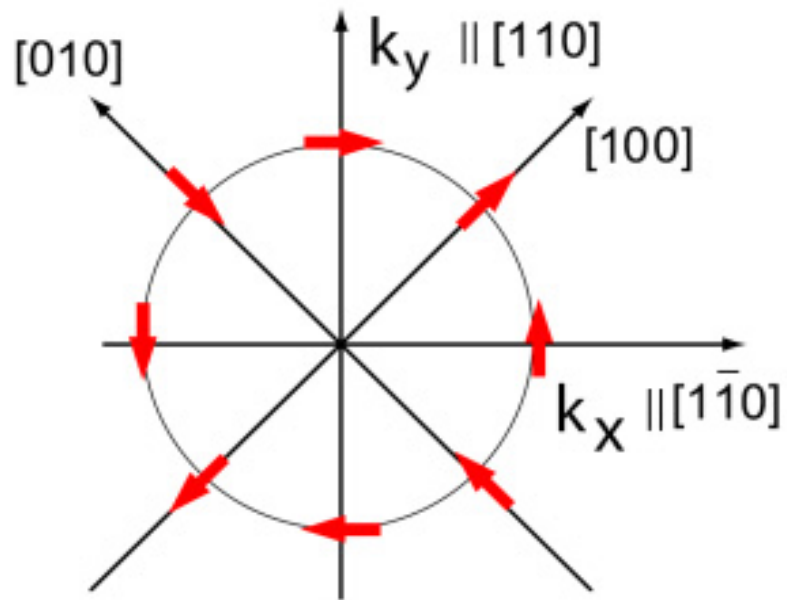
$$\left\langle \vec{S}(t) \right\rangle_t = \frac{\vec{S}_0}{1 + (\omega_L \tau_s)^2}$$



One can separate anisotropy of g-factor and relaxation time

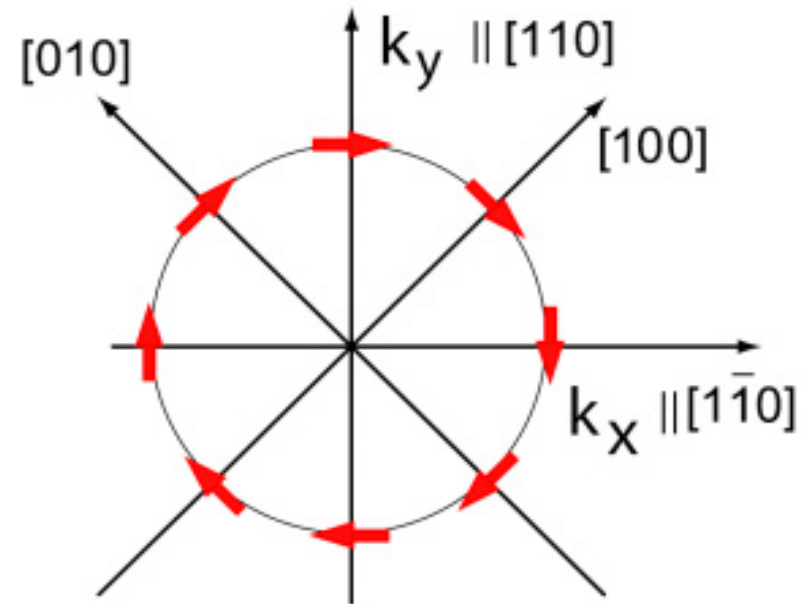
# Dresselhaus and Rashba effective field

*Dresselhaus*



$$\mathcal{H}_{\text{BIA}} = \beta(\sigma_x k_x - \sigma_y k_y)$$

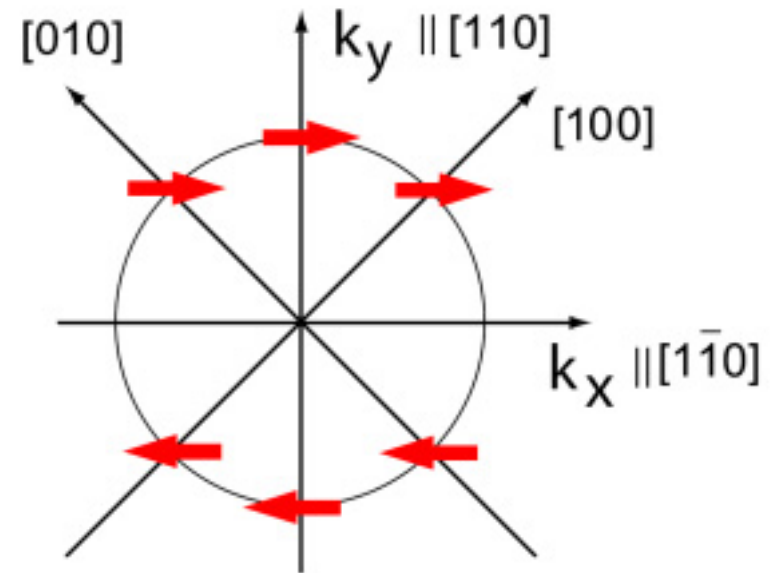
*Rashba*



$$\mathcal{H}_{\text{SIA}} = \alpha(\sigma_x k_y - \sigma_y k_x)$$

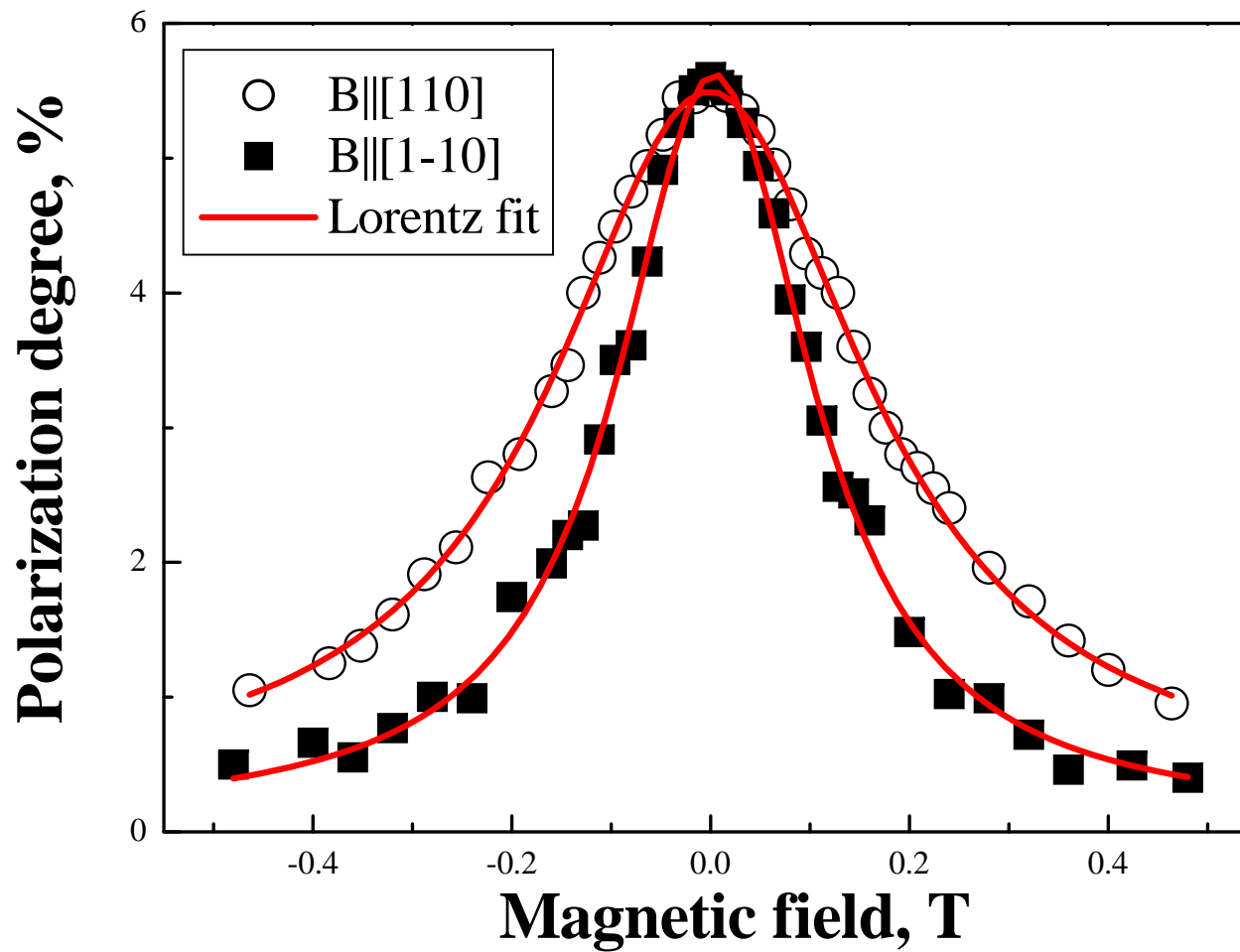
# *Rashba + Dresselhaus*

$$\alpha = \beta$$



N.S. Averkiev and L.E. Golub, *Phys. Rev. B* (1999)

# Hanle $\Rightarrow$ different SRT



• There are a lot of mechanisms for energy relaxation but there are only few mechanisms of angular momentum relaxation.

(For example relaxation between hole states  $+\frac{3}{2}$  to  $-\frac{3}{2}$   $\Delta M = 3$  is forbidden. But LH HH mixing leads to allowed transition  $\Delta M = 1$  )



# Optical orientation and optical alignment of excitons

At linearly polarized excitation in longitudinal magnetic field the polarization plane turns on

$$\theta = \frac{1}{2} \operatorname{arctg} (\Omega_{\parallel} \tau_0)$$

The degree of polarization  $P_{lin} = (P'_{lin} + P''_{lin})^{1/2}$  decreases

in  $\left[ 1 + (\Omega_{\parallel} \tau_0)^2 \right]^{1/2}$  times

$$P'_{lin} = \frac{\bar{d}_x^2 - \bar{d}_y^2}{\bar{d}_x^2 + \bar{d}_y^2}, \quad P''_{lin} = \frac{\bar{d}_{x'}^2 - \bar{d}_{y'}^2}{\bar{d}_{x'}^2 + \bar{d}_{y'}^2}$$

Consider the exciton as a classic dipole .  $\mathbf{d}_0$  Let it excited in  $x$  direction. In the longitudinal  $K \parallel B$  magnetic field it rotates around  $z$  with velocity

$$\Omega_{\parallel} / 2, \quad \Omega_{\parallel} = \Omega_e + \Omega_h, \quad \hbar\Omega = \mu g B$$

Created at  $t = 0$  components of the dipole moment at  $t > 0$

$$d_x(t) = d_0 \cos(\Omega_{\parallel} t / 2)$$

$$d'_x(t) = d_0 \cos(\Omega_{\parallel} t / 2 - \pi / 4)$$

$$d_y(t) = d_0 \sin(\Omega_{\parallel} t / 2)$$

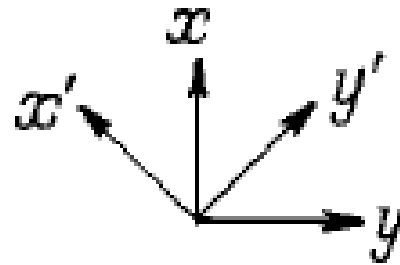
$$d'_y(t) = d_0 \sin(\Omega_{\parallel} t / 2 - \pi / 4)$$

$$\bar{d}_\alpha^2 = \frac{\int_0^\infty dt \exp(-t / \tau_0) d_\alpha^2(t)}{\int_0^\infty dt \exp(-t / \tau_0)}$$

$$d_x^2 - d_y^2 = d_0^2 \cos^2 \Omega_{\parallel} t$$

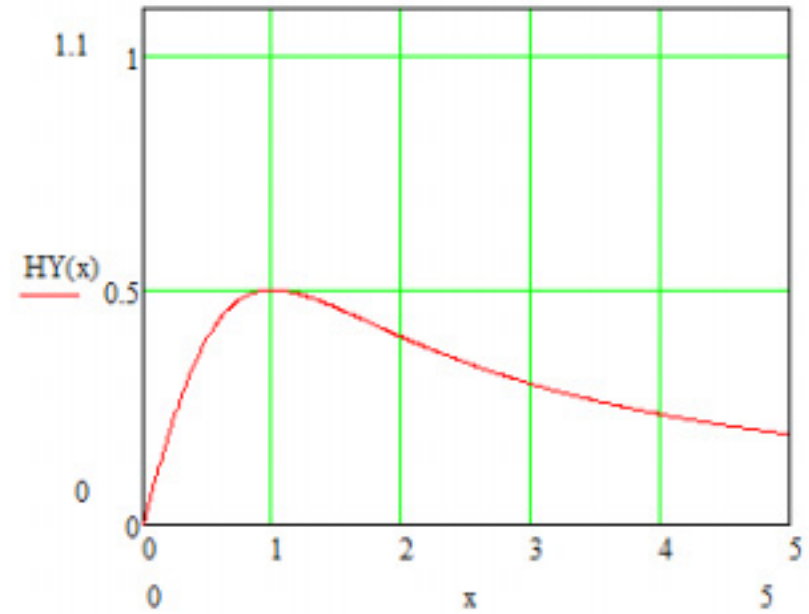
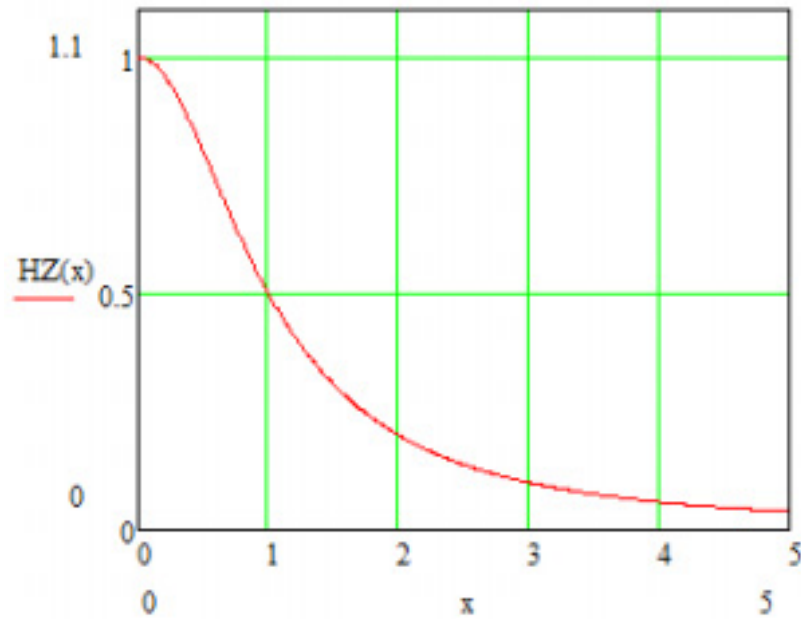
$$d_{x'}^2 - d_{y'}^2 = d_0^2 \sin^2 \Omega_{\parallel} t$$

$$d_x^2 + d_y^2 = d_{x'}^2 + d_{y'}^2 = d_0^2$$

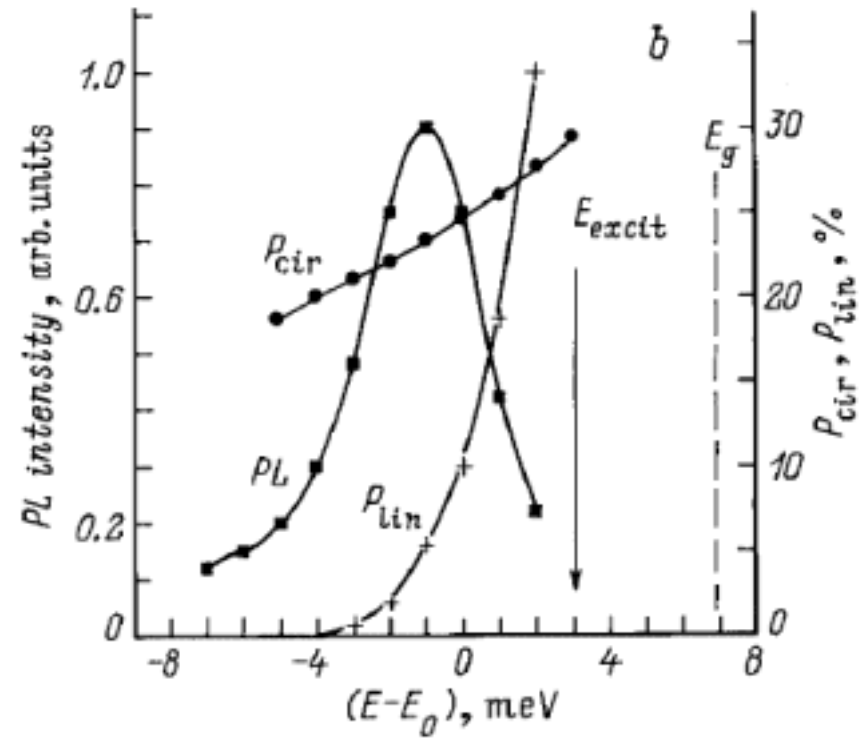
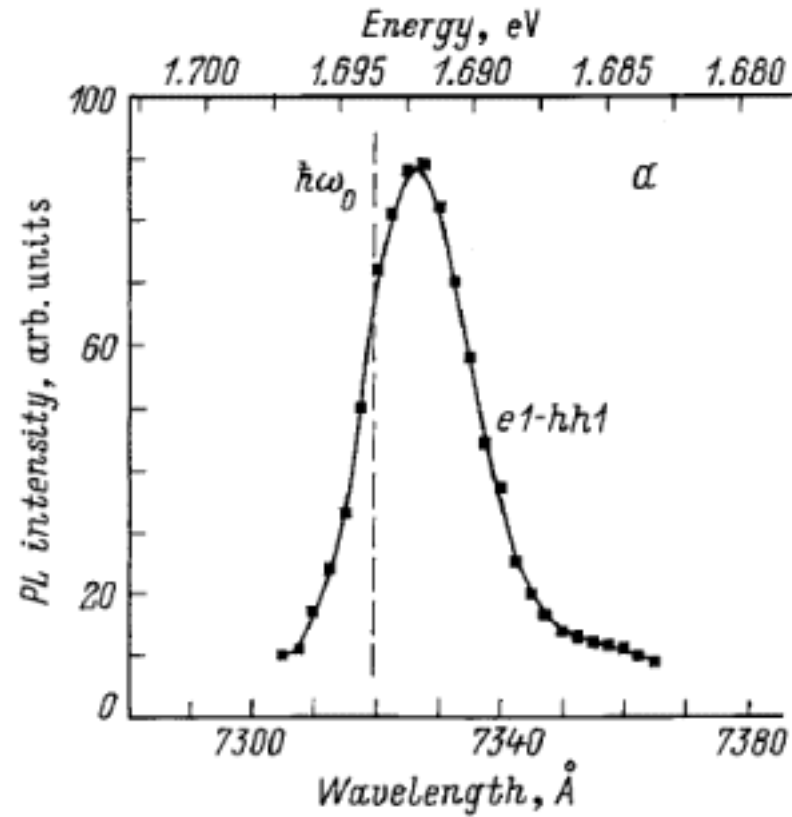


## Degree of polarization

$$P'_{lin}(B_{\parallel}) = \frac{1}{1 + (\Omega_{\parallel} \tau_0)^2}, \quad P''_{lin}(B_{\parallel}) = \frac{\Omega_{\parallel} \tau_0}{1 + (\Omega_{\parallel} \tau_0)^2}$$



# Resonant optical orientation and alignment of excitons in QWs



At circularly polarized excitation longitudinal fields have no effect on the exciton orientation

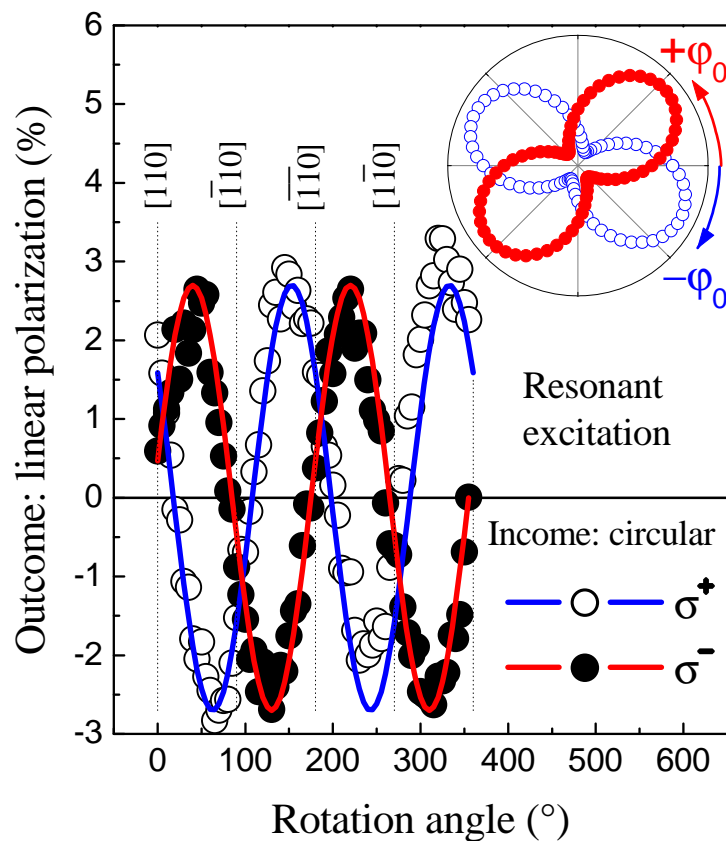
Transversal field suppresses the orientation and can induce linear polarization

$$P_c(\mathbf{B}_\perp) = \frac{1}{1 + \frac{3}{4}(\Omega_\perp \tau_0)^2}$$

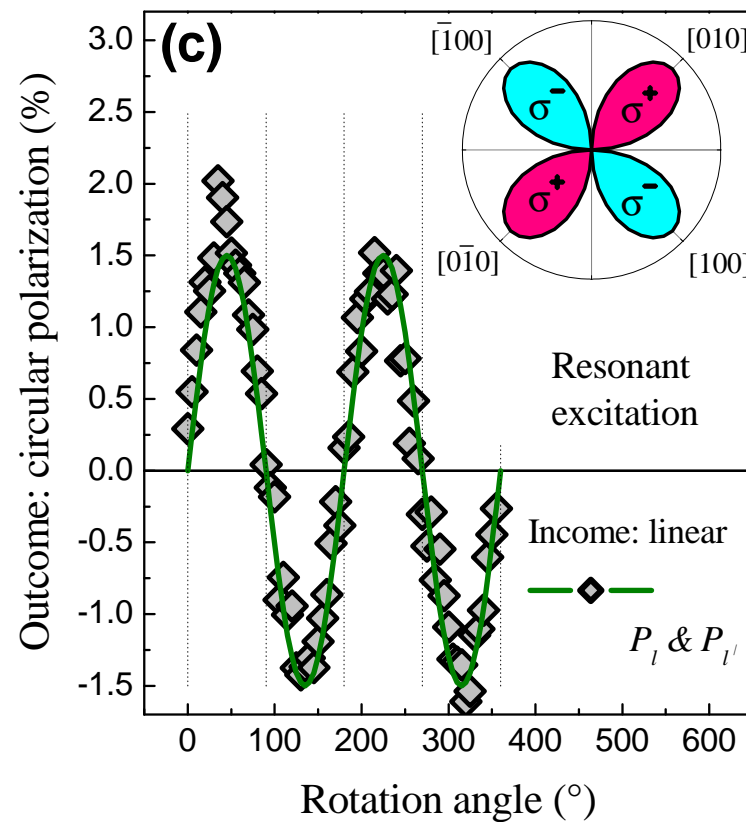
$$P'_{lin} = \frac{(\Omega_\perp \tau_0)^2}{4 + 3(\Omega_\perp \tau_0)^2}$$

# Transformation of polarizations in QDs

$$P_c \Rightarrow \rho_l$$



$$P_l \Rightarrow \rho_c$$



- Specific of excitons is that for exciton additionally to optical orientation we have optical alignment of dipole momenta.
- The mechanisms for exciton spin relaxation are different from that for electrons











