Model of the time-resolved photoluminescence from resonantly excited p-doped InAs/GaAs QDs Towards realistic modelling of a dot-cavity system

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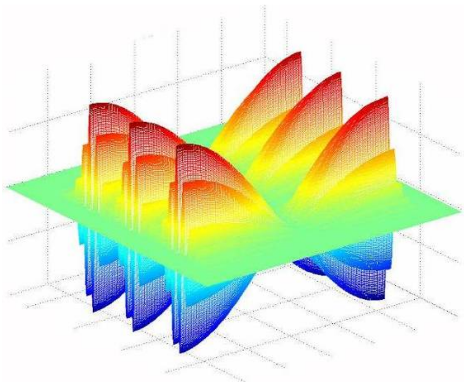

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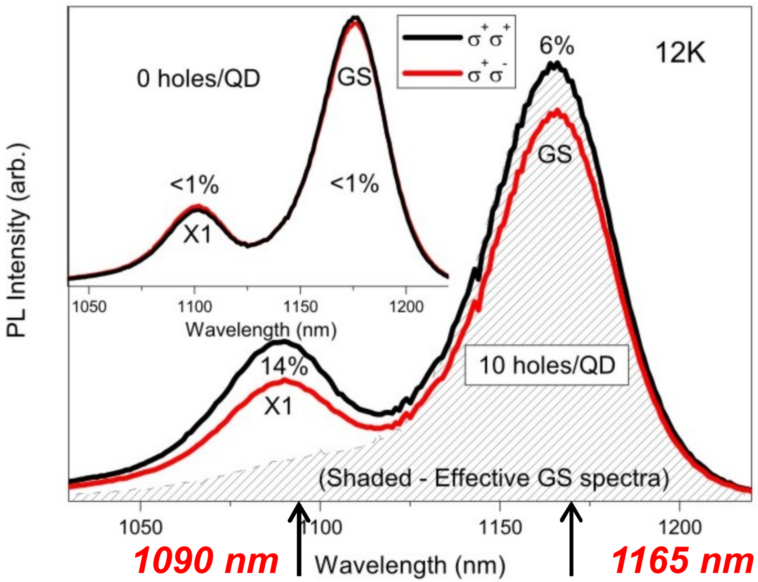
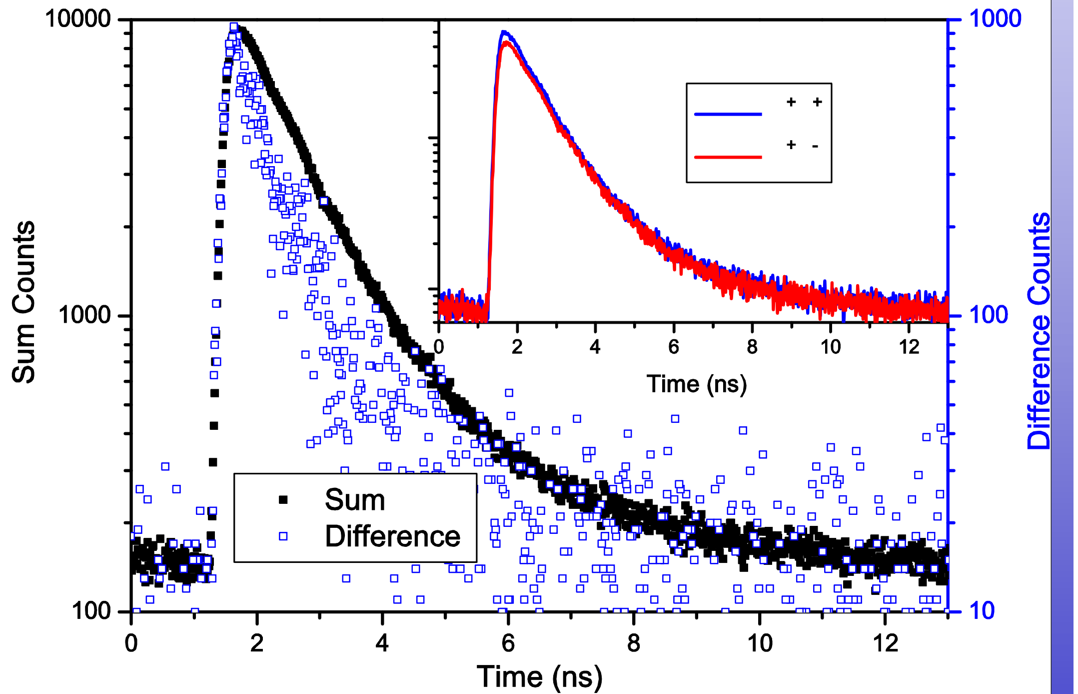
Overview

- Hot-trion energy level structure upon resonant optical excitation into p-shell states
- Spin dynamics under resonant excitation into hot positive-trion states in InAs/GaAs QDs
- Theoretical background and dynamical model
- Simulation results
- Modelling of E-field quantum fluctuations due to exciton spontaneous emission in a semiconductor microcavity
- Summary and outlook

Experimental results for p-doped QD ensembles

- *Experimentally (PL/TRPL) observed increased polarisation contrast in the excited state emission from p-doped QD ensembles compared to the GS emission (spin filter effect)*
- *Non-resonant (into bulk GaAs) circularly polarised excitation GS polarisation ~10%* $\lambda_{exc} = 790 nm, \quad T_p = 2.4\ ps$





M. W. Taylor et al., APL 97, 171907 (2010)

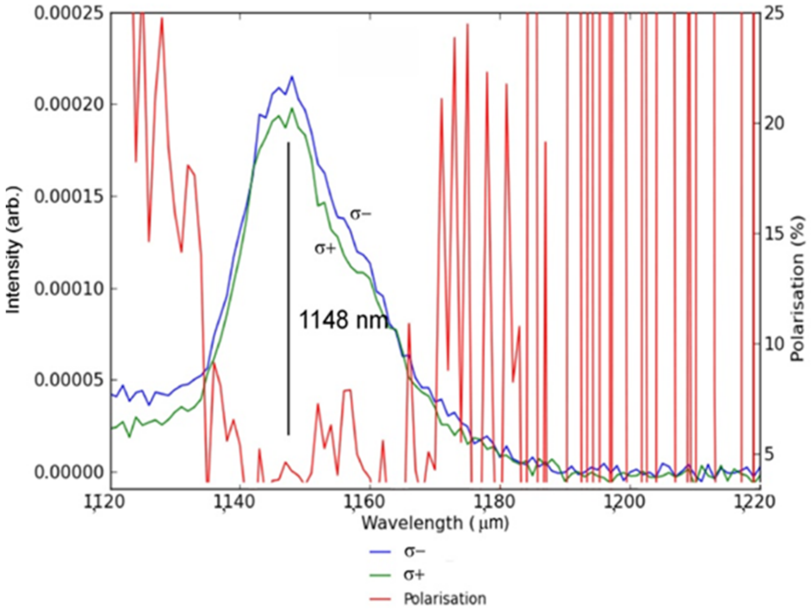
Resonant excitation

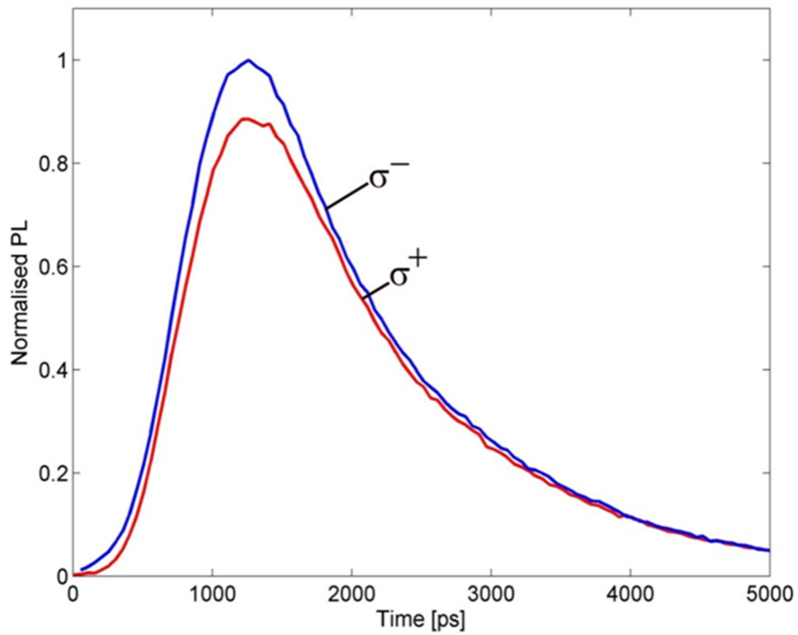
- Resonant excitation: degree of polarisation nearly doubled (increased spin injection efficiency)
- High-fidelity optical spin orientation and detection using hot trion states

PL and TRPL under resonant excitation into excited states

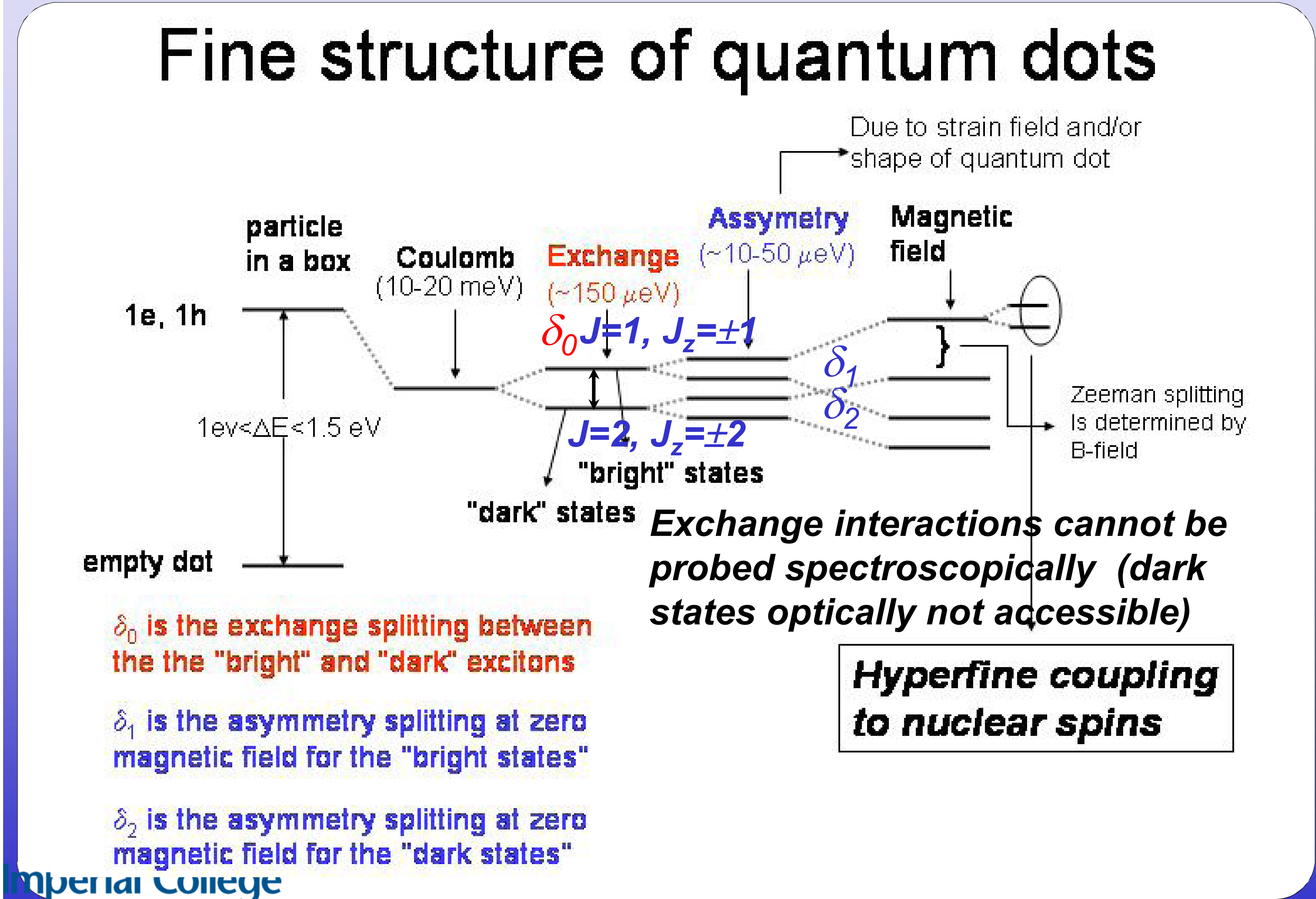
λ_{res}=1065 nm of a p-doped (1-hole) QD ensemble

λ_{det}~ 1148 nm at the X^{+} transition (T_p=50 ps)





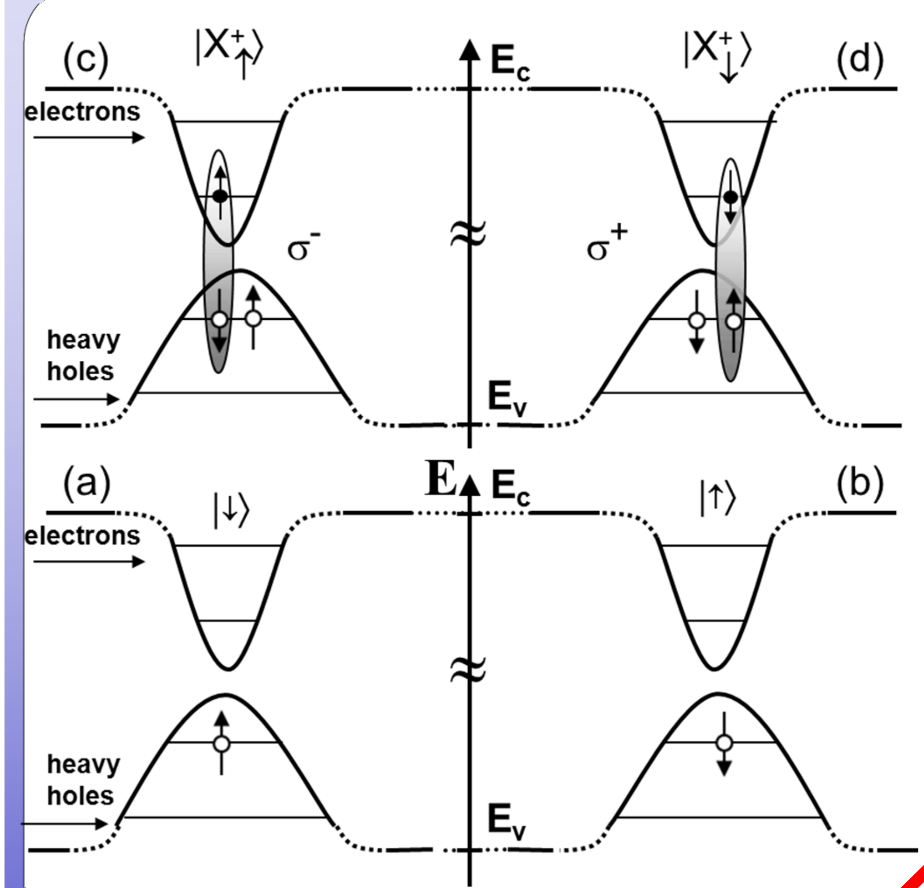
Undoped quantum dots: exciton energy levels



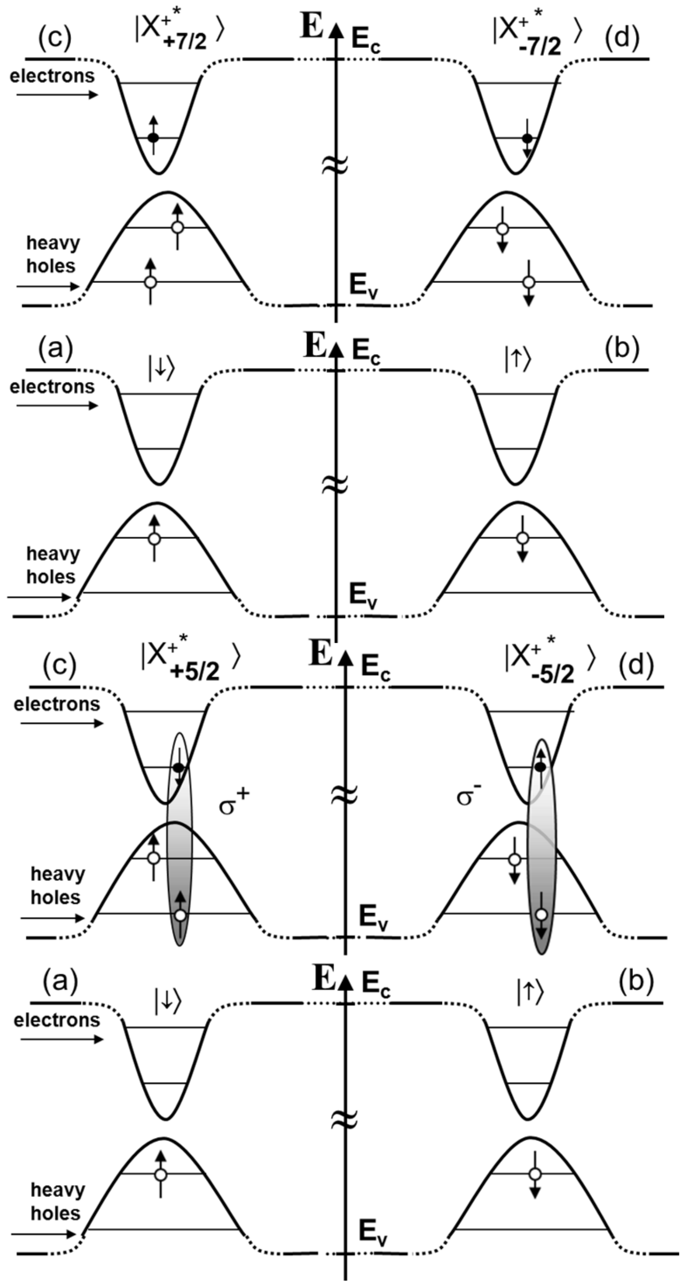
Exchange interactions cannot be probed spectroscopically (dark states optically not accessible)

Hyperfine coupling to nuclear spins

Positively-charged trion X^+

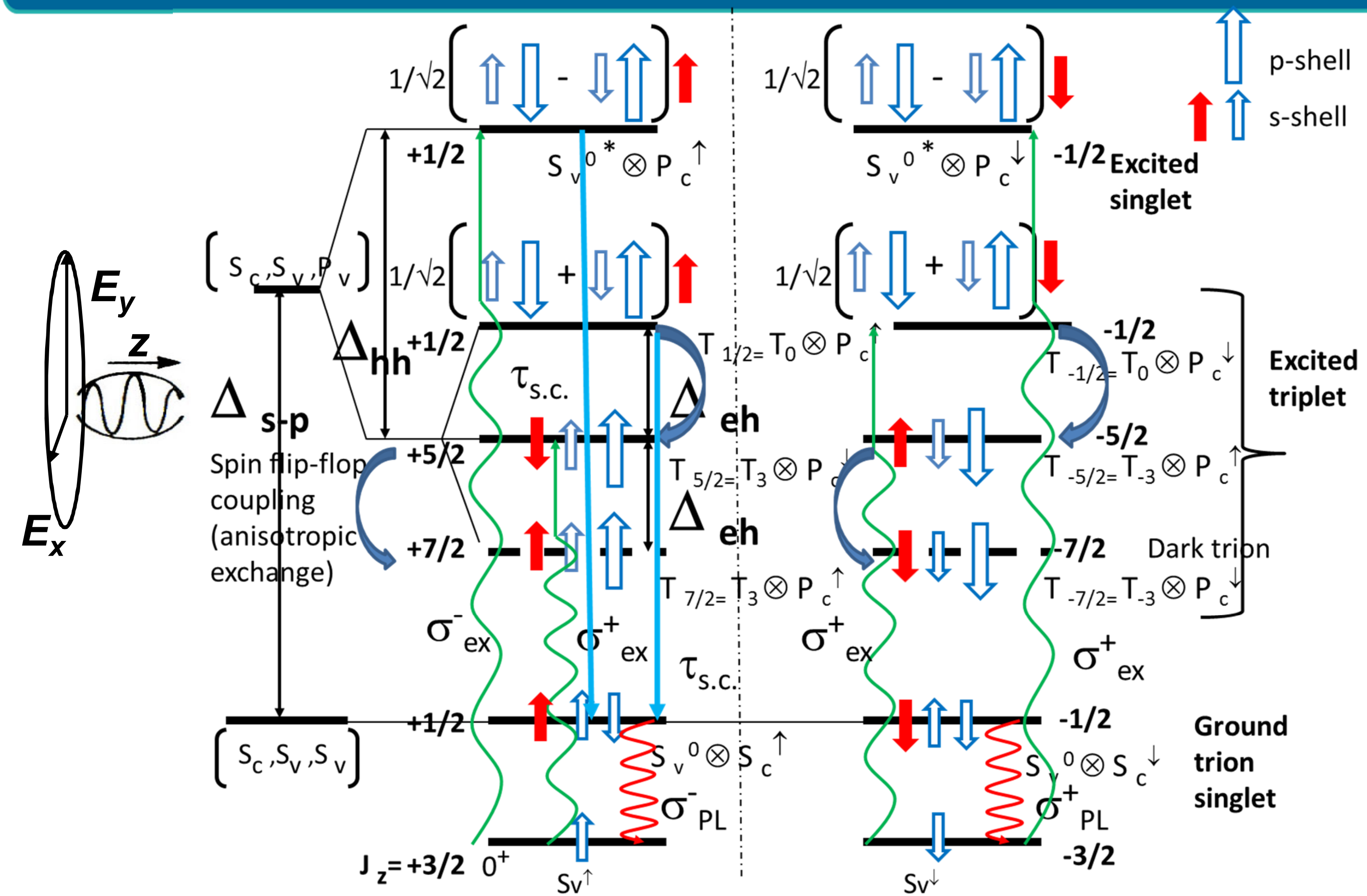


Ground state X^+ trion ($e_0h_0h_0$)



Hot X^{+*} trion ($e_0h_0h_1$) (2^3 spin configurations, half-integer total hot trion spin ⇒ doubly-degenerate (Kramers Theorem)⇒ 4 doublets

Hot X^+ trion energy-level diagram in a p-doped InAs/GaAs QD

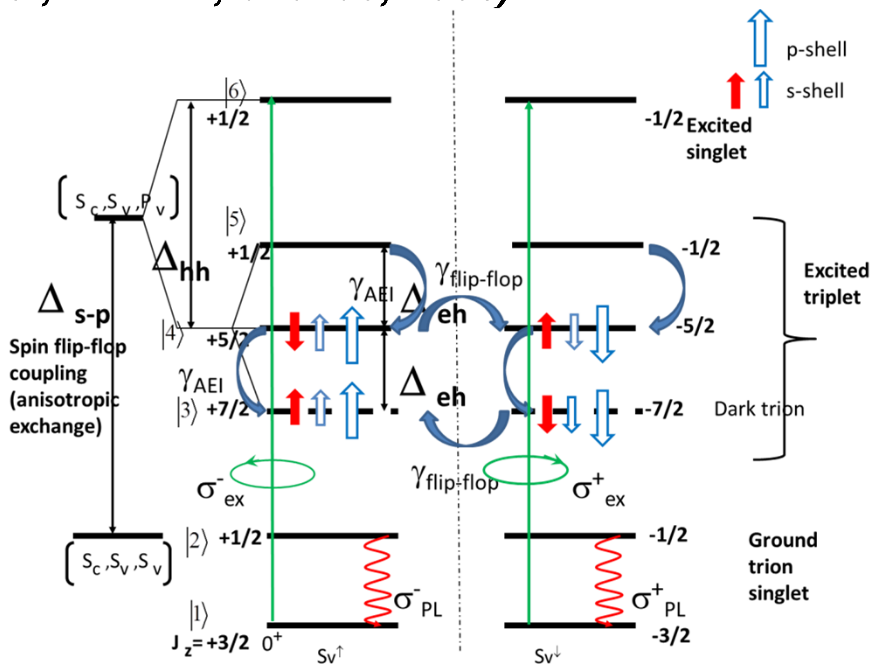


Exchange interactions can be revealed in the excited charged excitonic spectra (trions in particular)

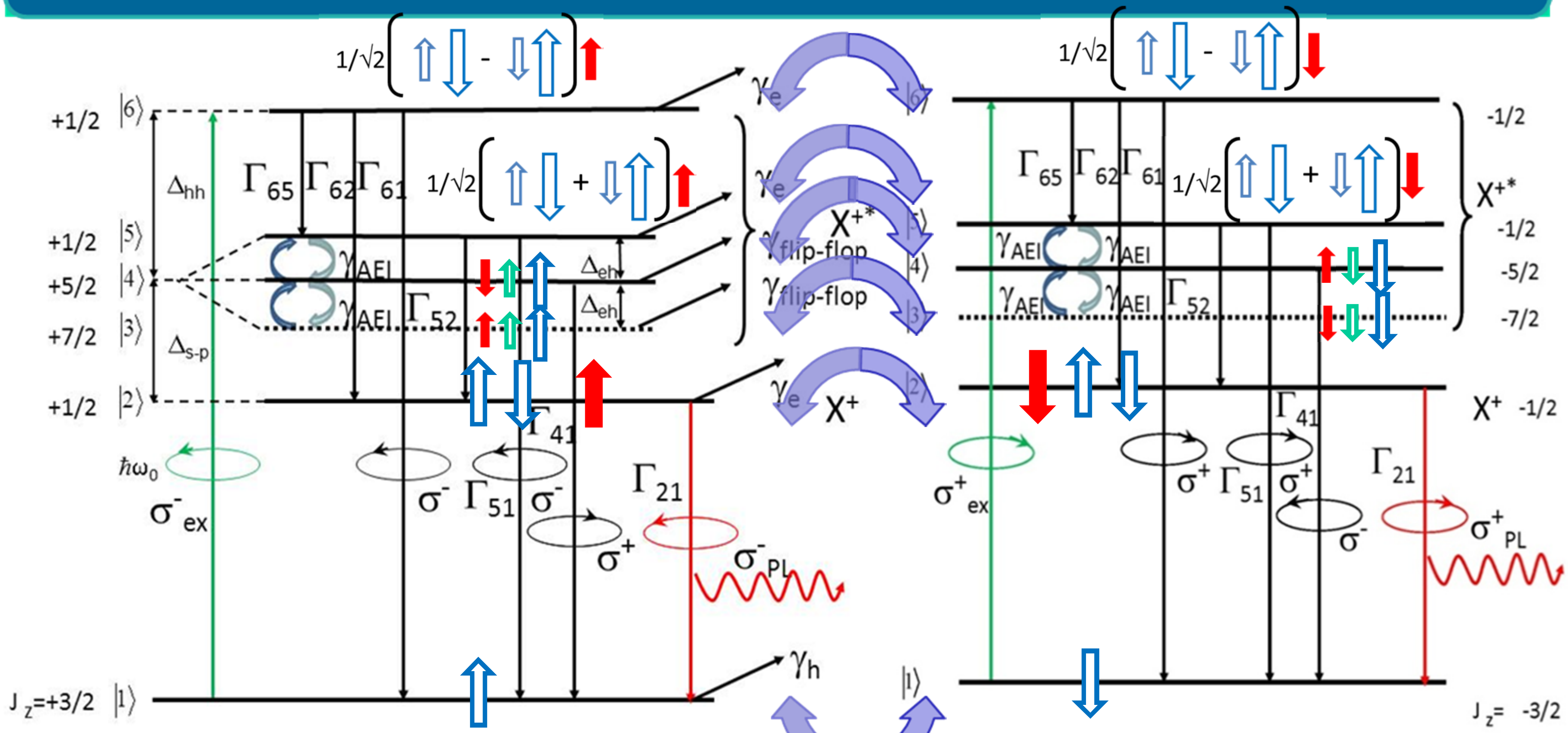
Energy splitting

Splitting controlled by:

- *Isotropic exchange interaction (Δ_{hh}) - splits the two-fold degenerate singlet from the 6-fold degenerate triplet states Δ_{hh} ~h-h Coulomb interaction* ***1-10 meV (K. Kavokin, Phys. Stat Sol. (a) 195, 592, 2003)***
- *Electron-hole exchange interaction:*
  - *Isotropic part (Δ_{eh}) – equally spaced splitting within the triplet states predicted* ***~ 0.1-1 meV (K. Kavokin, ibid.); ~ 0.23-2.1 meV (Warming et al., PRB 79, 125316, 2009) – asymmetric splitting experimentally observed***
  - *Anisotropic part – leads to mixing of the triplet states*
- *Splitting between (ground) s- and (excited) p-shell states, corrected by the direct Coulomb term $\Delta_{s\text{-}p}$~* ***20 meV*** *- comparable to acoustic phonon frequencies* ***(Narvaez, Bester and Zunger, PRB 74, 075403, 2006)***



Discrete-level model of the X+ fine structure



Transverse relaxation (spin decoherence) times:

- ***hyperfine interaction of electron spin with nuclei (γ_e~ 500 ps^{-1})****
- ***hyperfine interaction of hole spin with nuclei (dipole-dipole) (γ_h~ 14 ns^{-1})*****
- ***spin-flip processes mediated by anisotropic e-h exchange interaction (AEI) in the hot trion triplet state ~ tens μeV⇔ γ_{AEI}~125 ps^{-1}***

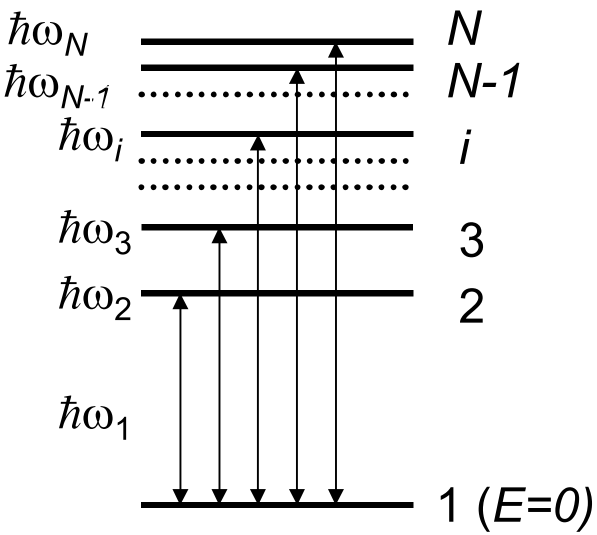
***Braun et al. PRL 94, 116601 (2005)**
**** Eble et al. , PRL 102, 146601 (2009)**

Equation of motion

Dynamical evolution of an N-level quantum system

Liouville equation (Schrödinger picture):

$$i\hbar\frac{\partial\hat{\rho}}{\partial t}=[\hat{H},\hat{\rho}]$$



$\Rightarrow$

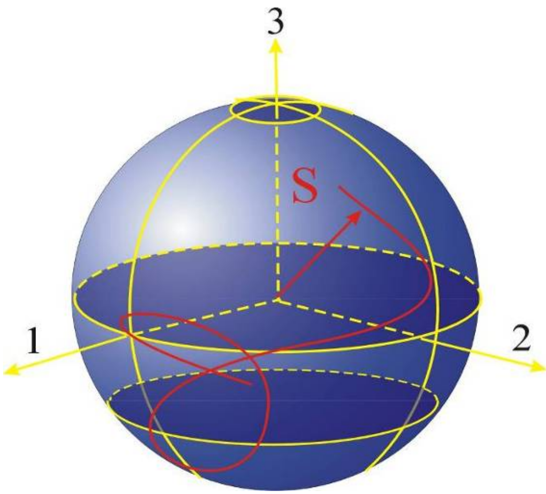
Pseudospin equation for the real state coherence vector S =(S_1, S_2, ... ,S_{N^2-1}) (Heisenberg picture):* $S_j(t)=Tr\left(\hat{\rho}(t)\hat{\lambda}_j\right)$

$$\frac{\partial S_i}{\partial t}=f_{ijk}\Gamma_j S_k$$

$i,j,k=1,\ldots,N^2-1$

***f_{ijk}* guarantees constant length:**

$$\left|\vec{S}\right|=const.$$



Using Gell-Mann's λ-generators of the SU(N) Lie algebra:

$$[\hat{\lambda}_j,\hat{\lambda}_k]=2i\,f_{jkl}\hat{\lambda}_l$$

$$\Gamma_j(t)=\frac{1}{\hbar}Tr\left(\hat{H}(t)\hat{\lambda}_j\right)$$

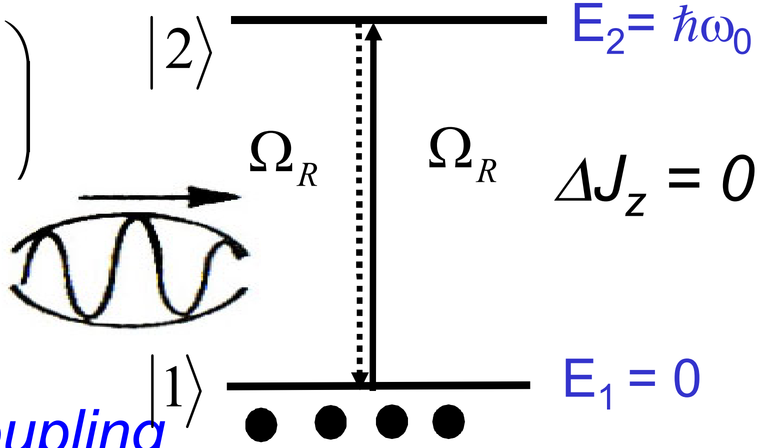
torque vector

Optical Bloch equations for a two-level quantum system

For dipole-coupling interaction: $H_{int} = -e\mathbf{E}.\hat{\mathbf{Q}}$

Electric dipole transitions excited by **linearly polarised** light

$$\hat{H} = -\hbar \begin{pmatrix} 0 & \Omega_R \\ \Omega_R & \omega_0 \end{pmatrix}$$



$\Delta J_z = 0$

$$\Gamma_j(t) = \frac{1}{\hbar} Tr\left(\hat{H}(t)\hat{\lambda}_j\right)$$

$$\Downarrow$$

$$\Gamma = \left(-2\Omega_R, 0, \omega_o\right)$$

Rabi frequency

$$\Omega_R = \frac{\wp}{\hbar} E$$

$\wp = eq_o$ dipole coupling

SU(2) generators (N=2):

$$\hat{\lambda}_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \hat{\lambda}_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \quad \hat{\lambda}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

$$\dot{S}_i = f_{ijk}\Gamma_j S_k \quad i, j, k = 1,2,3$$

$$f_{ijk} = \frac{1}{4} i\left[Tr(\hat{\lambda}_i\, \hat{\lambda}_k\, \hat{\lambda}_j) - Tr(\hat{\lambda}_i\, \hat{\lambda}_j\, \hat{\lambda}_k)\right]$$

$$f_{123} = f_{231} = f_{312} = 1$$

$$f_{132} = f_{213} = f_{321} = -1$$

$$\frac{\partial S_1}{\partial t} = -\omega_o S_2$$

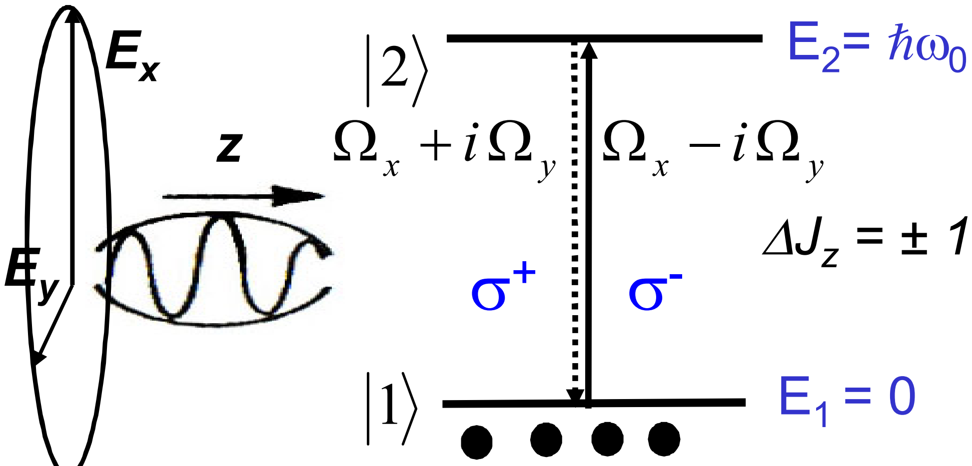
$$\frac{\partial S_2}{\partial t} = \omega_o S_1 + 2\Omega_R S_3$$

$$\frac{\partial S_3}{\partial t} = -2\Omega_R S_2$$

$$\Leftrightarrow \frac{d\mathbf{S}}{dt} = \Gamma \times \mathbf{S}$$

Optical Bloch equations for a two-level quantum system

*Electric dipole transitions excited by **circularly polarised** light*
Let us construct Hamiltonian corresponding to:



$$\hat{H} = \hbar \begin{pmatrix} 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & \omega_0 \end{pmatrix}$$

Rabi frequencies

$$\Omega_x = \frac{\wp}{\hbar} E_x$$

$$\Omega_y = \frac{\wp}{\hbar} E_y$$

Using: $\Gamma_j(t) = \frac{1}{\hbar} Tr\left(\hat{H}(t)\hat{\lambda}_j\right) \Rightarrow \Gamma = \left(-\Omega_x, -\Omega_y, \omega_o\right)$

$$\frac{d\mathbf{S}}{dt} = \Gamma \times \mathbf{S} \quad \Rightarrow \quad \begin{cases} \frac{\partial S_1}{\partial t} = \omega_o S_2 - \Omega_y S_3 \\ \frac{\partial S_2}{\partial t} = -\omega_o S_1 + \Omega_x S_3 \\ \frac{\partial S_3}{\partial t} = \Omega_y S_1 - \Omega_x S_2 \end{cases}$$

Six-level quantum systems

N = 6, ⇒ 35 generators of SU(6) group, $S = (S_1, S_2, \ldots, S_{35})$

f_{ijk} has 750 non-vanishing elements

$$\hat{\lambda}_1 = \begin{pmatrix} 0&1&0&0&0&0\\ 1&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0 \end{pmatrix} \quad \hat{\lambda}_2 = \begin{pmatrix} 0&0&1&0&0&0\\ 0&0&0&0&0&0\\ 1&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0 \end{pmatrix} \quad \hat{\lambda}_{31} = \begin{pmatrix} -1&0&0&0&0&0\\ 0&1&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0 \end{pmatrix}$$

$$\hat{\lambda}_{32} = \begin{pmatrix} -\frac{1}{\sqrt{3}}&0&0&0&0&0\\ 0&-\frac{1}{\sqrt{3}}&0&0&0&0\\ 0&0&\frac{2}{\sqrt{3}}&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0 \end{pmatrix}$$

$$\hat{\lambda}_{33} = \begin{pmatrix} -\frac{1}{\sqrt{6}}&0&0&0&0&0\\ 0&-\frac{1}{\sqrt{6}}&0&0&0&0\\ 0&0&-\frac{1}{\sqrt{6}}&0&0&0\\ 0&0&0&\sqrt{\frac{3}{2}}&0&0\\ 0&0&0&0&0&0\\ 0&0&0&0&0&0 \end{pmatrix} \quad \hat{\lambda}_{34} = \begin{pmatrix} -\frac{1}{\sqrt{10}}&0&0&0&0&0\\ 0&-\frac{1}{\sqrt{10}}&0&0&0&0\\ 0&0&-\frac{1}{\sqrt{10}}&0&0&0\\ 0&0&0&-\frac{1}{\sqrt{10}}&0&0\\ 0&0&0&0&2\sqrt{\frac{2}{5}}&0\\ 0&0&0&0&0&0 \end{pmatrix}$$

$$\hat{\lambda}_{35} = \begin{pmatrix} -\frac{1}{\sqrt{15}}&0&0&0&0&0\\ 0&-\frac{1}{\sqrt{15}}&0&0&0&0\\ 0&0&-\frac{1}{\sqrt{15}}&0&0&0\\ 0&0&0&-\frac{1}{\sqrt{15}}&0&0\\ 0&0&0&0&-\frac{1}{\sqrt{15}}&0\\ 0&0&0&0&0&\sqrt{\frac{5}{3}} \end{pmatrix}$$

$S_{31}, .., S_{35}$ – population terms

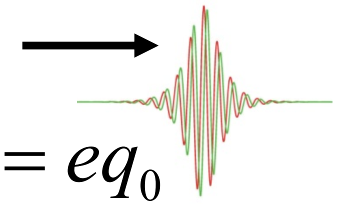
Quasi-resonant circularly polarised pulsed excitation into the p-shell

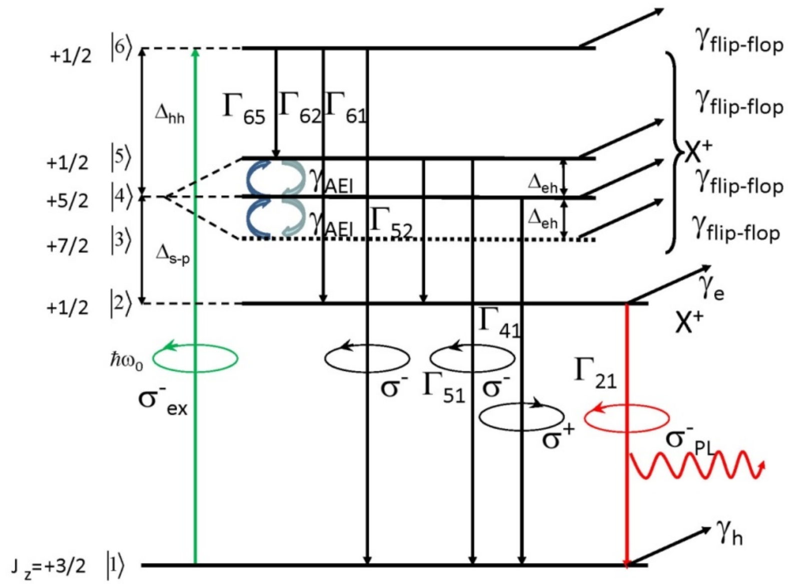
N=6, SU(6) Lie group

$$\hat{\lambda}_1, \hat{\lambda}_2, \ldots, \hat{\lambda}_{35}$$

$$\begin{cases} \Omega_x = \dfrac{\wp}{\hbar} E_x \\ \Omega_y = \dfrac{\wp}{\hbar} E_y \end{cases} \qquad \wp = e q_0$$

τ=50 ps
σ^-
π-pulse





System Hamiltonian

$$\hat{H} = \hbar \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) \\ 0 & \omega_0 - \Delta_{hh} - \Delta_{sp} & 0 & 0 & 0 & 0 \\ 0 & 0 & \omega_0 - \Delta_{hh} - \Delta_{eh} & 0 & 0 & 0 \\ 0 & 0 & 0 & \omega_0 - \Delta_{hh} & 0 & 0 \\ 0 & 0 & 0 & 0 & \omega_0 - \Delta_{hh} + \Delta_{eh} & 0 \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & 0 & 0 & 0 & 0 & \omega_0 \end{pmatrix}$$

Torque vector and coupling to Maxwell's curl eqs

Using: $\Gamma_j(t) = \frac{1}{\hbar} Tr\left(\hat{H}(t)\hat{\lambda}_j\right)$ **N-1= 5 population terms**

$$\mathbf{\Gamma} = \begin{pmatrix} 0,0,0,0,-\Omega_x,0,0,0,0,0,0,0,0,0,0,0,0,0,-\Omega_y,0,0,0,0,0,0,0,0,0,0, \\ -\Delta_{hh}-\Delta_{sp}+\omega_0, \frac{-2\Delta_{eh}-\Delta_{hh}+\Delta_{sp}+\omega_0}{\sqrt{3}}, \frac{\Delta_{eh}-\Delta_{hh}+\Delta_{sp}+\omega_0}{\sqrt{6}}, \frac{5\Delta_{eh}-\Delta_{hh}+\Delta_{sp}+\omega_0}{\sqrt{10}}, \frac{4\Delta_{hh}+\Delta_{sp}+\omega_0}{\sqrt{15}} \end{pmatrix}$$

6-level system excited by circularly polarised pulse:

$$\vec{\mathbf{E}} = E_x \vec{\mathbf{e}}_\mathbf{x} + E_y \vec{\mathbf{e}}_\mathbf{y}$$

Dipole-interaction Hamiltonian:

$$\hat{H}_{\text{int}}(t) = -e\vec{\mathbf{E}}\cdot\hat{Q} = \hbar \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & -\frac{1}{2}(\Omega_x - i\Omega_y) \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ -\frac{1}{2}(\Omega_x + i\Omega_y) & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$$

Decomposition of local displacement operator:

$$\hat{Q} = \hat{Q}_x + \hat{Q}_y = q_0 \left[\hat{\lambda}_5 \vec{\mathbf{e}}_x + \hat{\lambda}_{20} \vec{\mathbf{e}}_y\right]$$

Coupling to Maxwell's curl equations

Macroscopic polarisation: $\mathrm{P} = - eN_a Tr(\hat{\rho}.\hat{Q})$

N_a **Number density of resonant dipoles**

$$\hat{\rho}(t) = \frac{1}{N}\hat{I} + \frac{1}{2}\sum_{j=1}^{N^2-1} S_j(t)\hat{\lambda}_j \Rightarrow \begin{matrix} P_x = -\wp_{1\to 6} N_a S_5 \\ P_y = -\wp_{1\to 6} N_a S_{20} \end{matrix}$$

Maxwell's curl equations

$$\begin{cases} \dfrac{\partial H_x(z,t)}{\partial t} = \dfrac{1}{\mu}\dfrac{\partial E_y(z,t)}{\partial z} \\ \dfrac{\partial H_y(z,t)}{\partial t} = -\dfrac{1}{\mu}\dfrac{\partial E_x(z,t)}{\partial z} \\ \dfrac{\partial E_x(z,t)}{\partial t} = -\dfrac{1}{\varepsilon}\dfrac{\partial H_y(z,t)}{\partial z} - \dfrac{1}{\varepsilon}\dfrac{\partial P_x(z,t)}{\partial t} \\ \dfrac{\partial E_y(z,t)}{\partial t} = \dfrac{1}{\varepsilon}\dfrac{\partial H_x(z,t)}{\partial z} - \dfrac{1}{\varepsilon}\dfrac{\partial P_y(z,t)}{\partial t} \end{cases}$$

Master pseudospin equations

Transverse spin relaxation

$$\frac{\partial S_i}{\partial t} = f_{ijk}\Gamma_j S_k$$

$$\frac{\partial \hat{\rho}}{\partial t} = \frac{i}{\hbar}\left[\hat{\rho}, \hat{H}\right] + \hat{\sigma} - \hat{\Gamma}_t \hat{\rho}$$

$$\hat{\sigma} = diag\left(Tr(\hat{\Gamma}_i \hat{\rho})\right), \quad i = 1,2,\ldots,6$$

$$\frac{\partial S_j}{\partial t} = \begin{cases} f_{jkl}\Gamma_k S_l + \frac{1}{2}Tr\left(\hat{\sigma}\lambda_j\right) - \frac{1}{T_j}\left(S_j - S_{je}\right), & j = 1,2,\ldots,30 \\ f_{jkl}\Gamma_k S_l + \frac{1}{2}Tr\left(\hat{\sigma}\lambda_j\right), & j = 31,\ldots,35 \end{cases}$$

Longitudinal spin relaxation

$$\begin{cases} \frac{\partial H_x(z,t)}{\partial t} = \frac{1}{\mu}\frac{\partial E_y(z,t)}{\partial z} \\ \frac{\partial H_y(z,t)}{\partial t} = -\frac{1}{\mu}\frac{\partial E_x(z,t)}{\partial z} \\ \frac{\partial E_x(z,t)}{\partial t} = -\frac{1}{\varepsilon}\frac{\partial H_y(z,t)}{\partial z} - \frac{1}{\varepsilon}\frac{\partial P_x(z,t)}{\partial t} \\ \frac{\partial E_y(z,t)}{\partial t} = \frac{1}{\varepsilon}\frac{\partial H_x(z,t)}{\partial z} - \frac{1}{\varepsilon}\frac{\partial P_y(z,t)}{\partial t} \end{cases}$$

$$P_x = -\wp_{1\to 6} N_a S_5$$

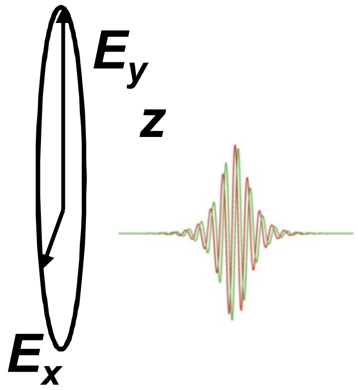
$$P_y = -\wp_{1\to 6} N_a S_{20}$$

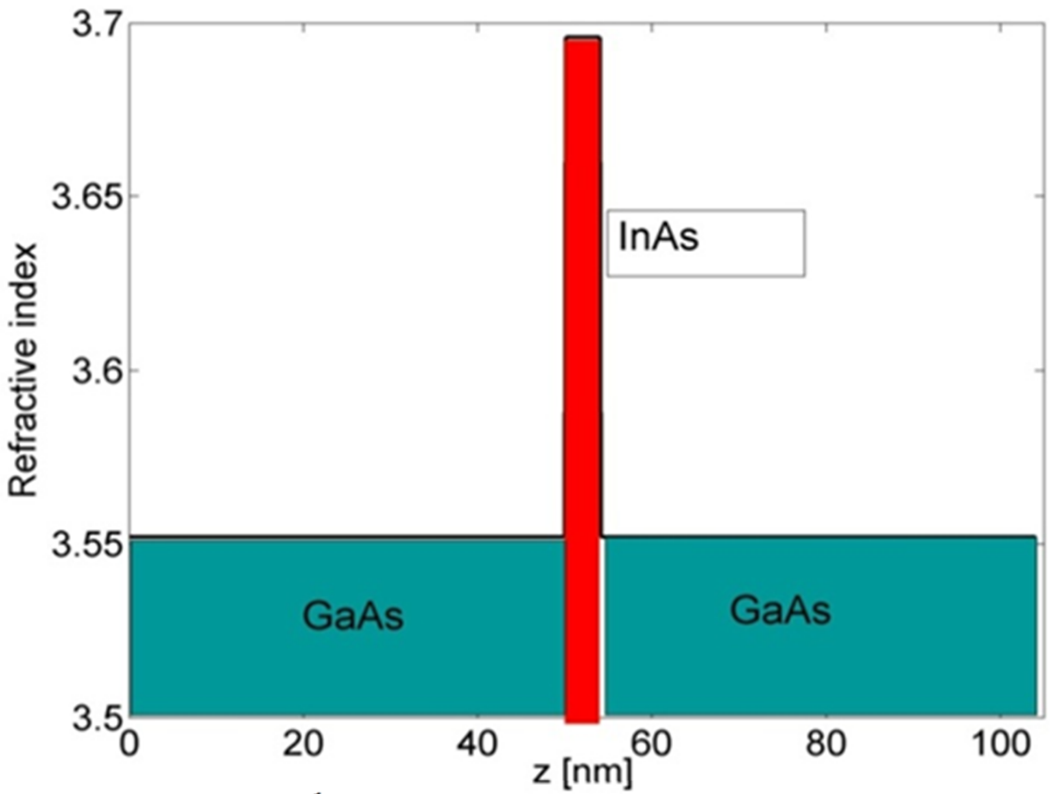
Maxwell's curl equations

G.Slavcheva, PRB 77, 115347 (2008)

Finite-Difference Time-Domain solution

50 nm/4 nm/50 nm





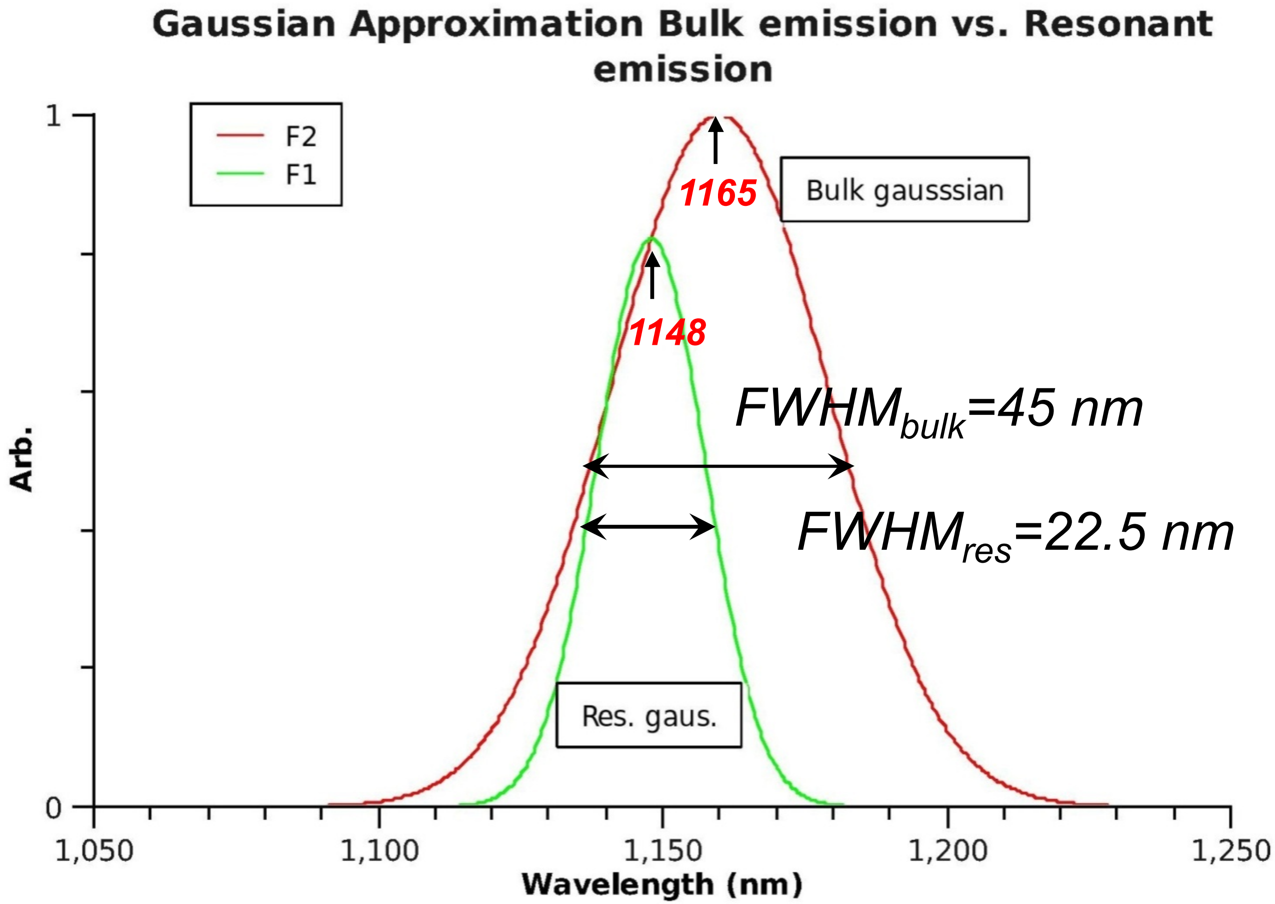
Source optical field (Gaussian pulse)

$$\sigma^- \begin{cases} E_x(z=0,t) = E_o \exp[-(t-t_0)^2/t_d^2]\cos(\omega_o t) \\ E_y(z=0,t) = -E_0 \exp[-(t-t_0)^2/t_d^2]\sin(\omega_o t) \end{cases}$$

$$\sigma^+ \begin{cases} E_x(z=0,t) = E_o \exp[-(t-t_0)^2/t_d^2]\cos(\omega_o t) \\ E_y(z=0,t) = E_0 \exp[-(t-t_0)^2/t_d^2]\sin(\omega_o t) \end{cases}$$

Estimate for resonantly excited dot density

$N_{tot\ dots}$=2×10^{10} cm^{-2}; h~4 nm; inhomogeneously broadened ensemble of QDs $N_{res\ dots}$~0.2-0.3 $N_{tot\ dots}$



Spin relaxation and decoherence timescales

- ***Longitudinal spin-relaxation rates:***

➢ ***Radiative (spontaneous) decay***

$\Gamma_{21} \sim 1.27\ ns^{-1}$ ***- experiment for 1 hole***

Taylor et al., APL 97, 171907 (2010)

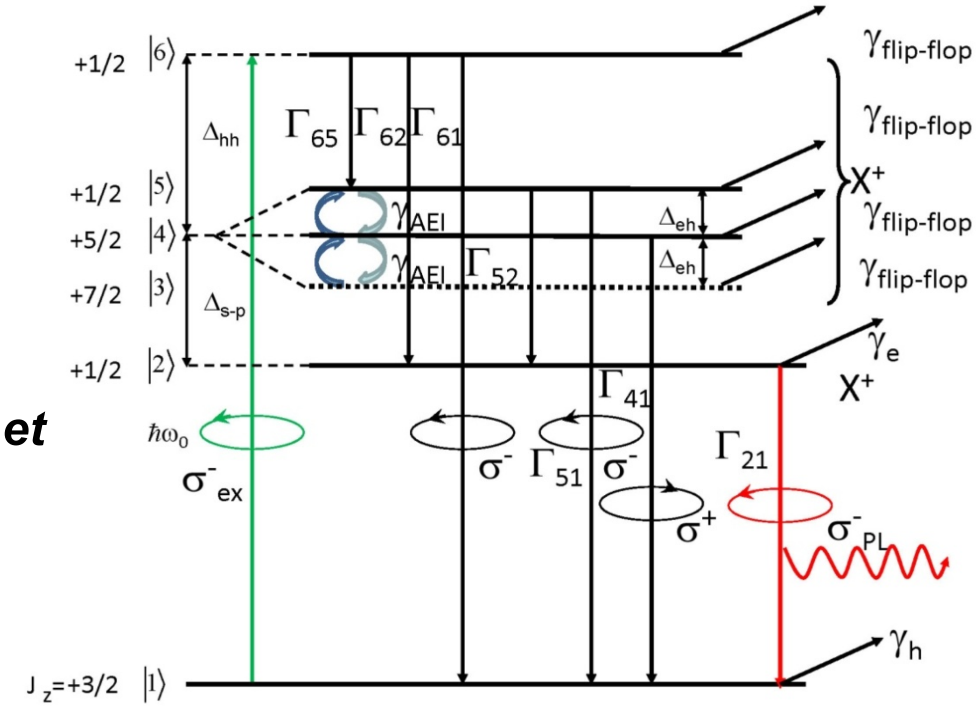
X^+ transition dipole matrix element:

$\mu \sim 8 \times 10^{-29}$ ***Cm***

Fry et al. PRL 84, 733 (2000); Findeis et al. APL 78, 2958 (2001)

(Estimated spontaneous emission times:

$\Gamma_{41} \sim 1.35\ ns^{-1}$; $\Gamma_{51} \sim \Gamma_{61} \sim 1.2 - 1.3\ ns^{-1}$



➢ ***Nonradiative decay***

$\Gamma_{52} \sim$ ***3-7*** ps^{-1} ***p→s intershell relaxation - Narvaez, et al., PRB 74, 075403 (2006) (or very slow ~ 750 ps -7.7 ns)***

$\Gamma_{62} \sim$ ***3-5*** ps^{-1} ***(s*→s intrashell relaxation)***

$\Gamma_{65} \sim$ ***35-50*** ps^{-1} ***(s*→p intershell relaxation via acoustic phonon emission)***

$$\tau_{spont} = \frac{3\pi\varepsilon_0 \hbar c^3}{n\omega_0^3 \wp^2}$$

- ***Transverse spin decoherence rates:*** $\gamma_e \sim 500\ ps^{-1}$; $\gamma_h \sim 14\ ns^{-1}$; $\gamma_{AEI} \sim 125\ ps^{-1}$

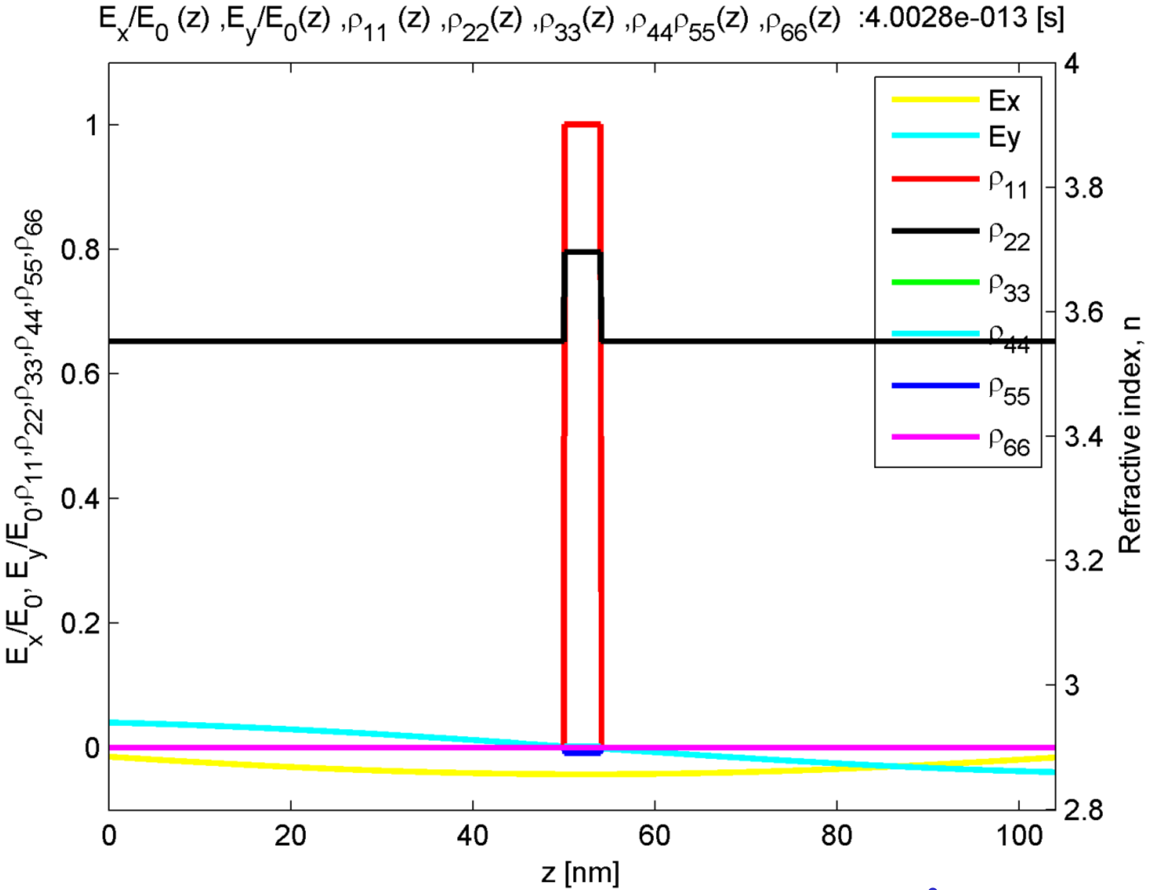
Numerical Simulations

Parameters	Simulation data 1	Simulation data 2	Simulation data 3	Simulation data 4	Simulation data 5	Simulation data 6
τ_{21} [ns]	1.27	1.27	1.27	1.59	1.59	1.27
τ_{41} [ns]	1.354	1.354	1.354	1.3	1.3	1.354
τ_{51} [ns]	1.2	1.2	1.2	1.3	1.3	1.2
τ_{52} [ps]	5	7	5	5	5	5
τ_{61} [ns]	1.2	1.2	1.2	1.27	1.27	1.2
τ_{62} [ps]	5	750	3	5	5	5
τ_{65} [ps]	50	5	50	50	50	50
τ_{AEI} [ps]	125	125	125	125	125	125
λ_{res} [nm]	1127	1065	1065	1065	1125.52	1125.52
E_{trion} [eV] @1148 nm	1.08	1.0815	1.0815	1.0815	1.0815	1.0815
Δ_{sp} [meV]	16	73	73	73	16	16
Δ_{hh} [meV]	5.6	12	12	12	5.6	5.6
Δ_{eh} [meV]	0.5	0.5	0.5	0.5	0.5	0.5
$\gamma_{1\rightarrow 6}$[C.m]	8×10^{-29}	9.78×10^{-29}	9.78×10^{-29}	9.56×10^{-29}	9.56×10^{-29}	9.83×10^{-29}
N_A [m^{-3}]	5×10^{22}	5×10^{19}	5×10^{19}	1.5×10^{22}	3×10^{22}	7×10^{22}
E_0 (Gauss π-pulse) [V/m]	3.3×10^5 @1060 nm	2.7×10^5 @1065 nm	2.7×10^5 @1065 nm	2.765×10^5 @1065 nm	2.765×10^5 @1065 nm	2.765×10^5 @1065 nm
T_p [ps]	**53.38**	**50**	**50**	**50**	**50**	**50**

Key parameters: N_{dots}, optical dipole matrix element $\wp_{1\rightarrow 6}$, nonradiative intra- and intershell decay τ_{62}, τ_{52}

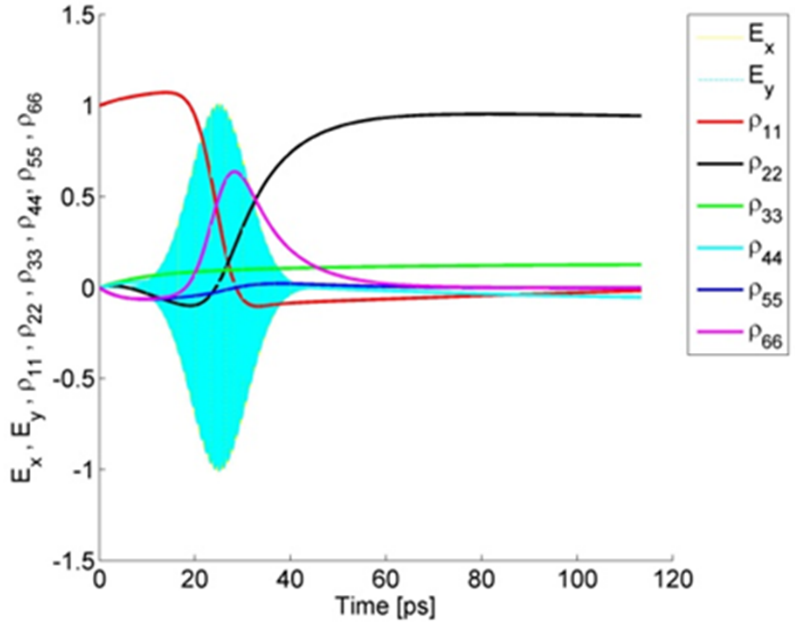
Simulated spatially resolved dynamics

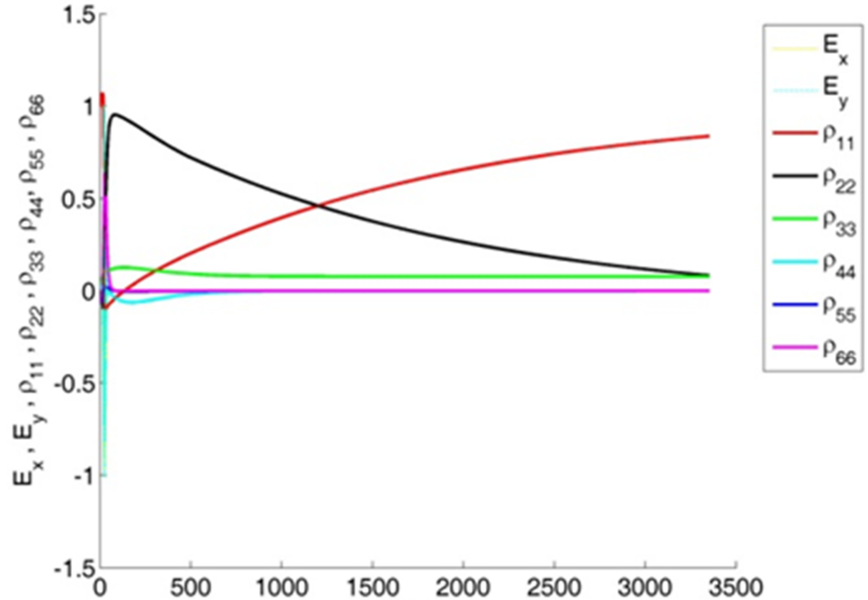
InAs/GaAs self-assembled modulation doped QD layer 50/4/50 nm



Spatial and temporal discretisation: Δz=1 Å, Δt=3.3×10^{-4} fs

Simulated temporal dynamics under σ^- - excitation

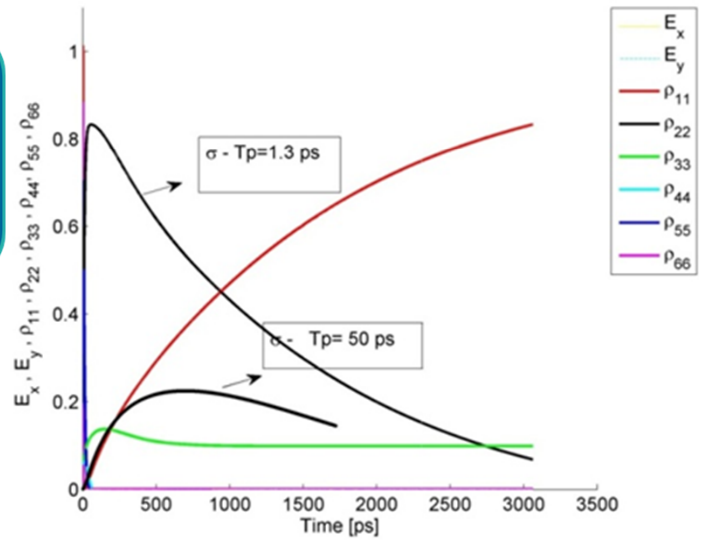




Simulated temporal dynamics

σ^- - Gaussian π-pulse (Tp=50 ps) excitation

Snapshots t=120 ps, 3.5 ns ;comparison with Tp=1.3 ps



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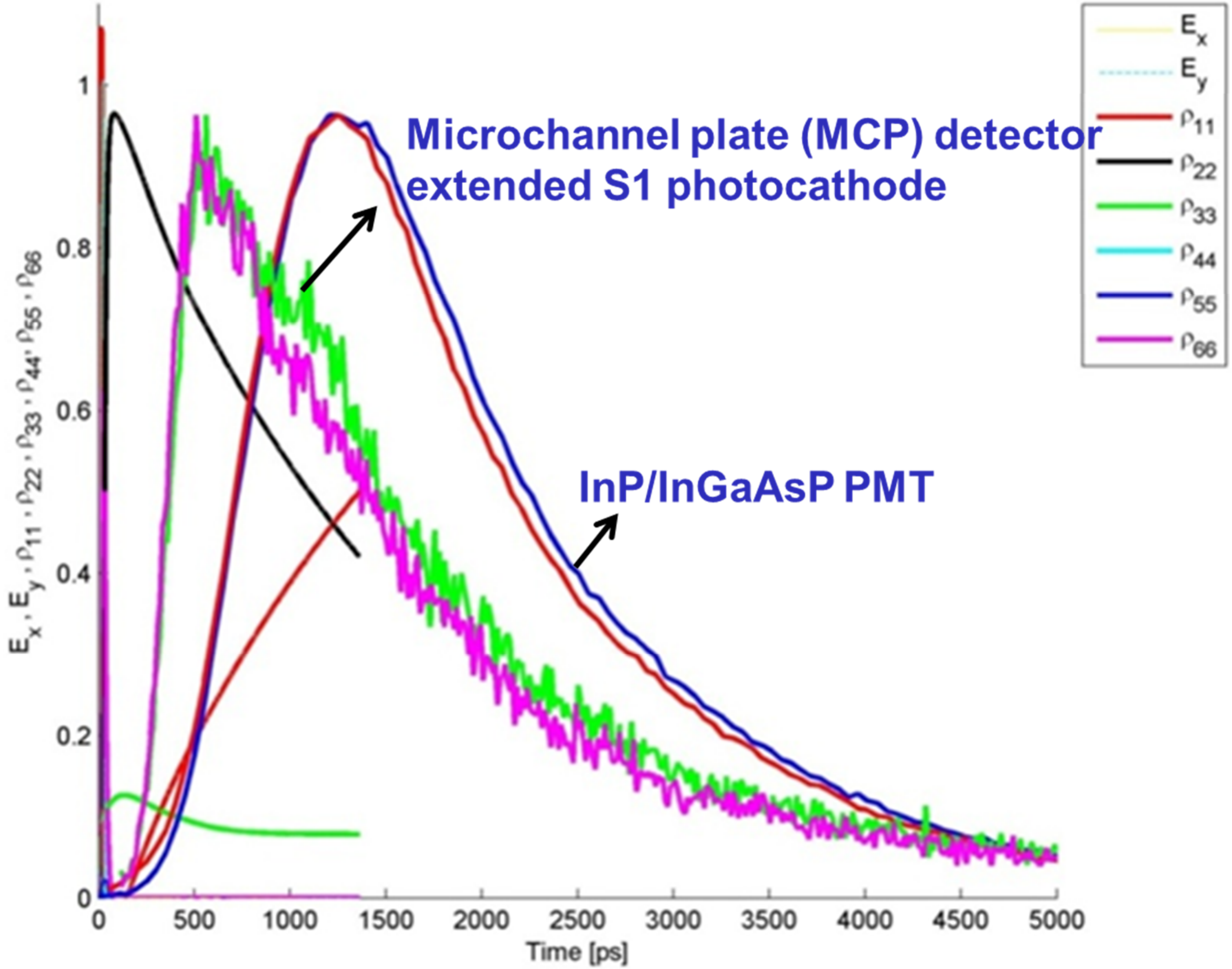
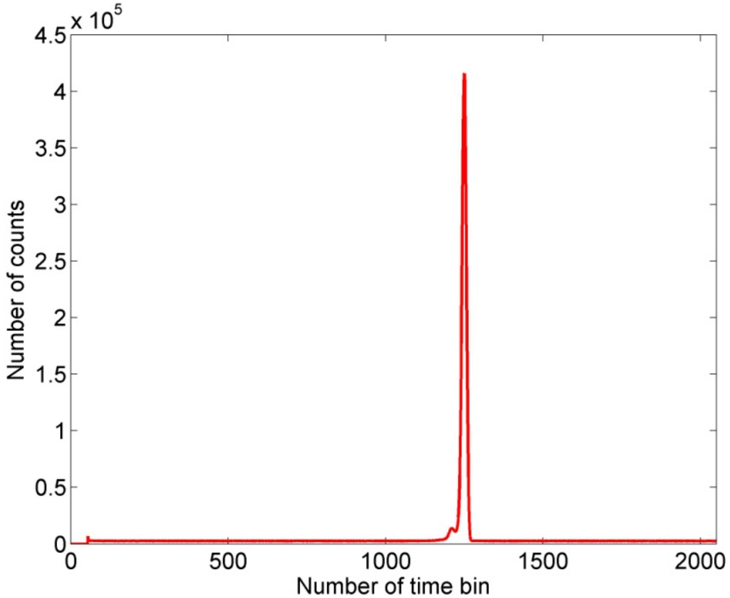
Comparison between theory and experiment before convolution with detector response function

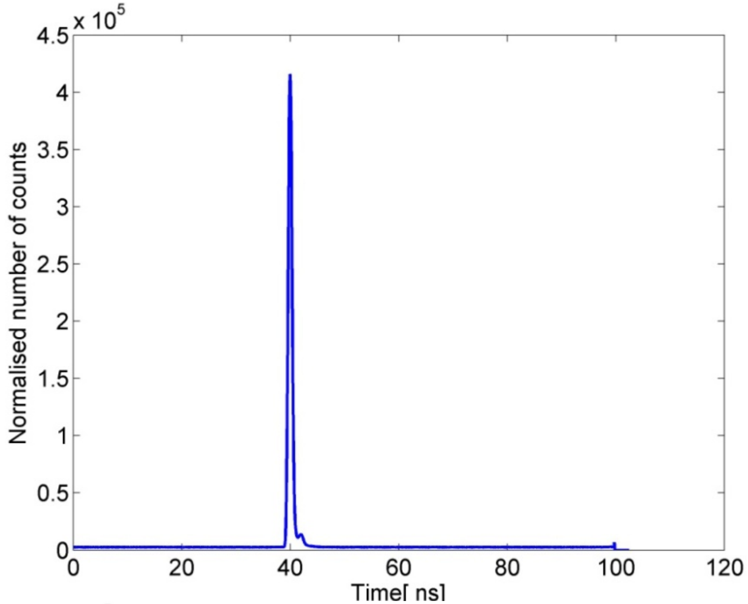


Detector response function

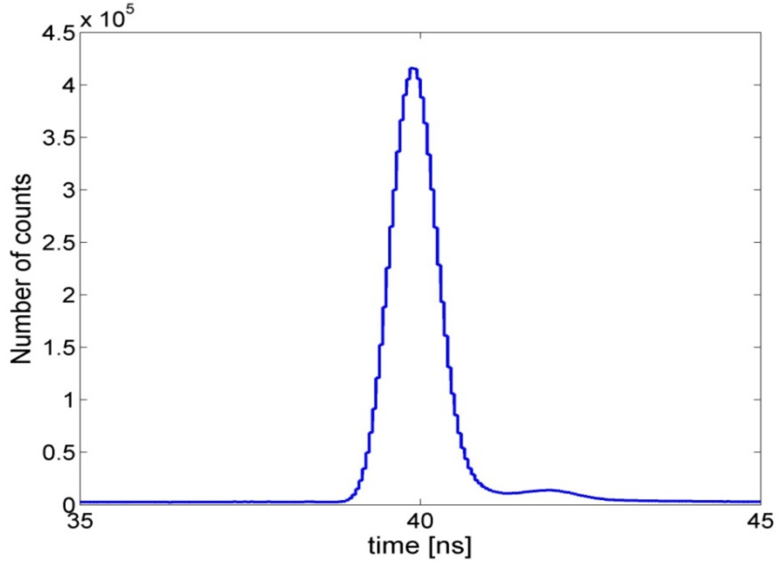
InP/InGaAsP detector response function



Discretised detector response function



Detector sampling rate $\Delta t_1 = 0.05$ ns



Simulations sampling rate

$\Delta t_2 = 3.33 \times 10^{-4}$ fs

Number of points added within each bin $N = INT(\Delta t_1 / \Delta t_2)$

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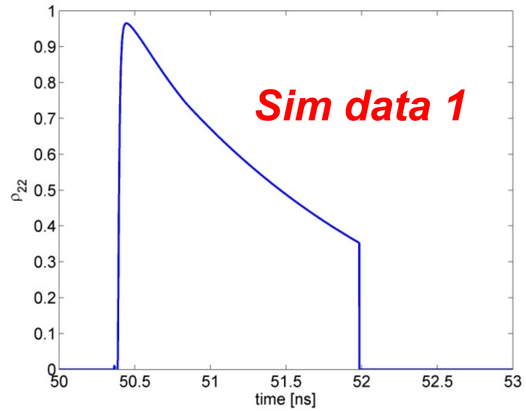
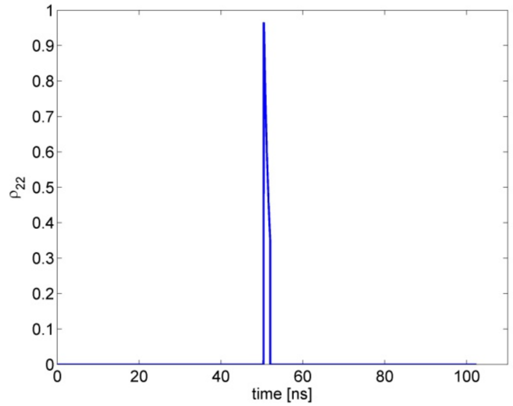
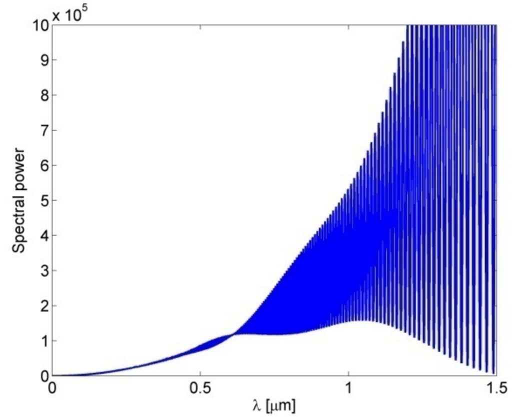
Convolution of sim. data with detector response function



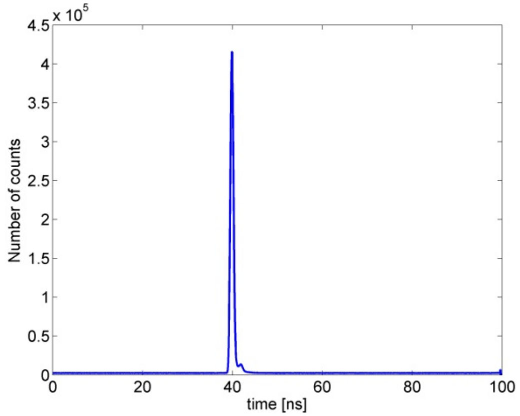


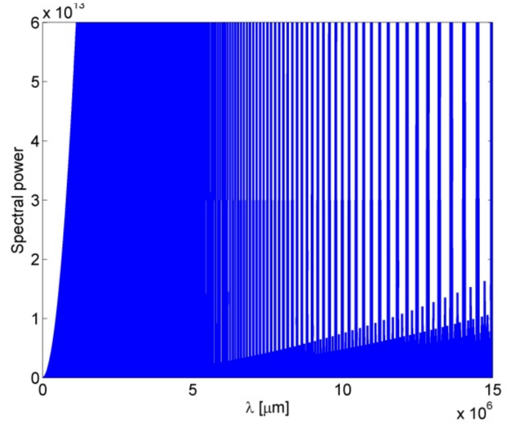


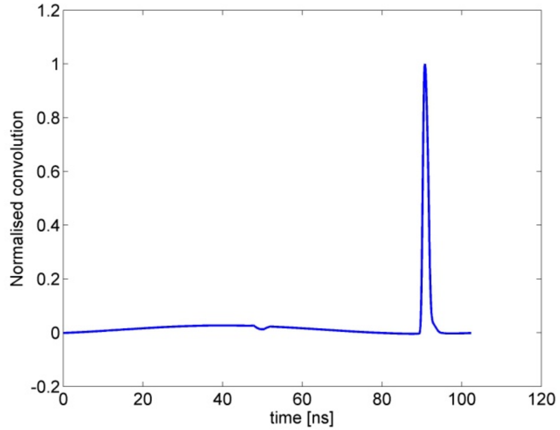
Convolution Theorem:

$$g * h = \int_{-\infty}^{\infty} g(\tau)h(t-\tau)d\tau$$

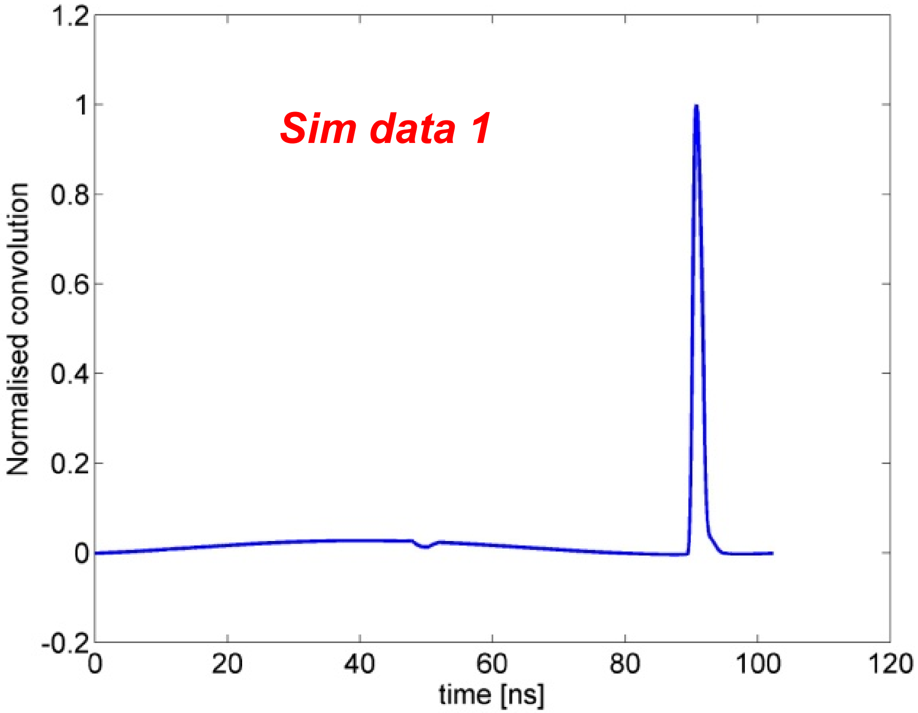
$$F(g * h) \Leftrightarrow G(f)H(f)$$

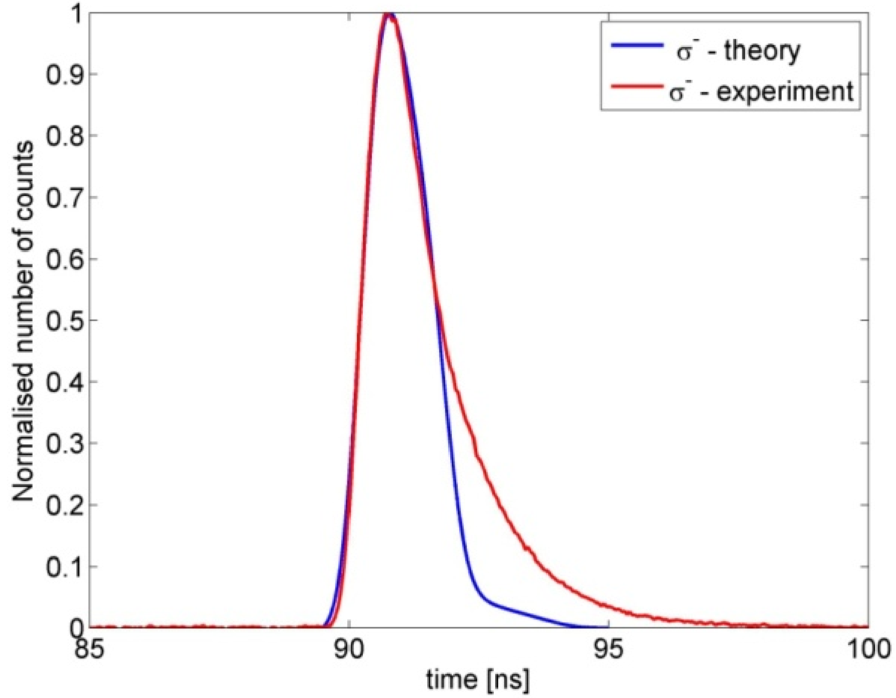






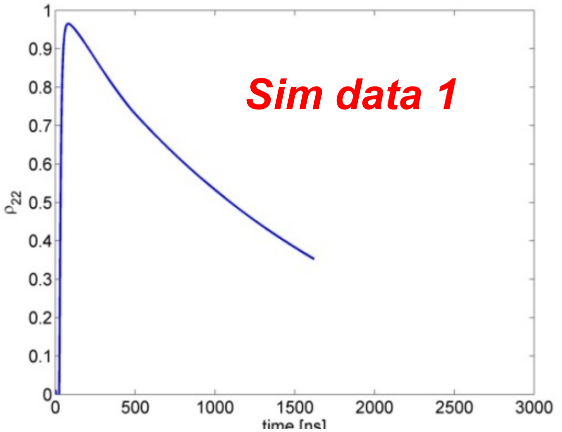
Comparison with experimental TRPL trace

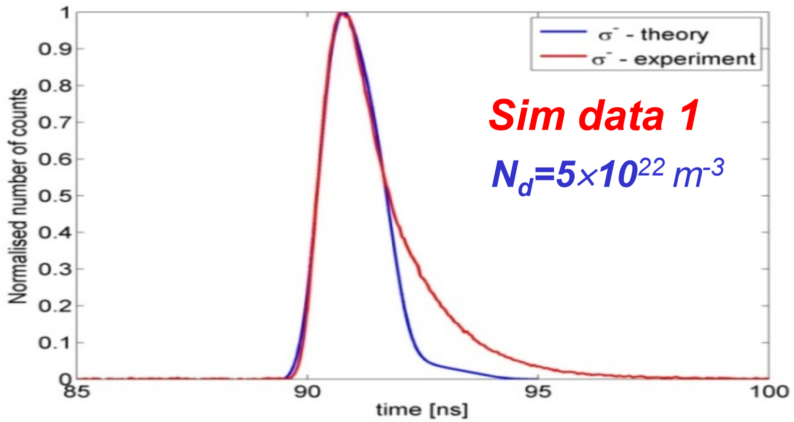


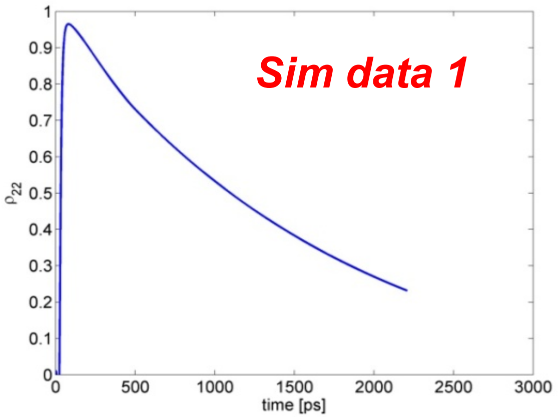


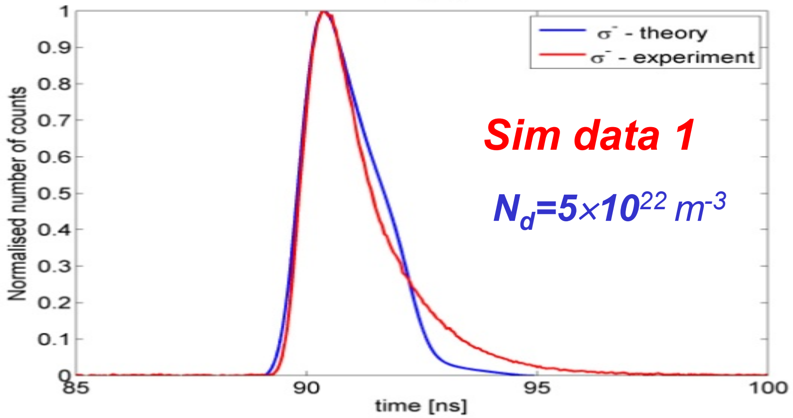
TRPL simulation convolved with detector response function

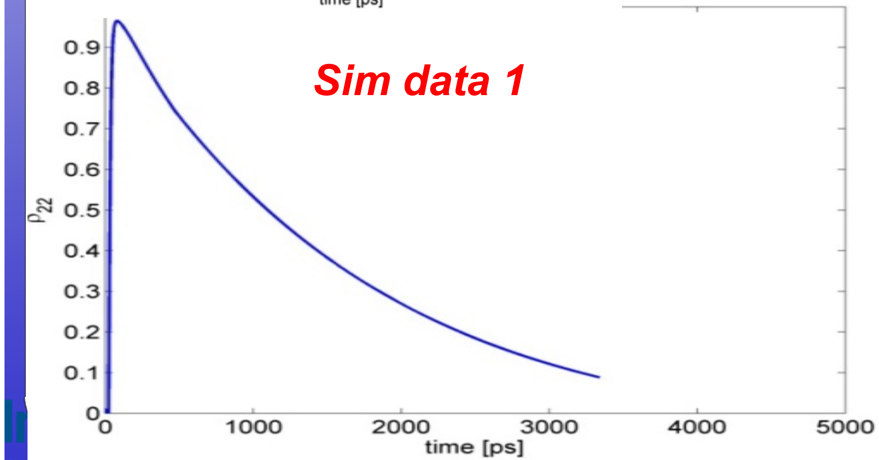
Comparison with simulations of different length in time

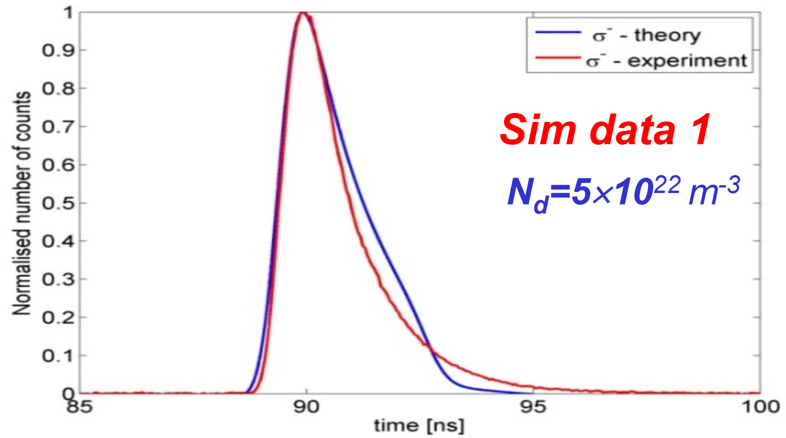




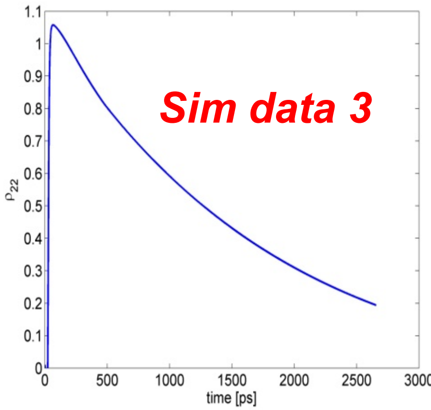


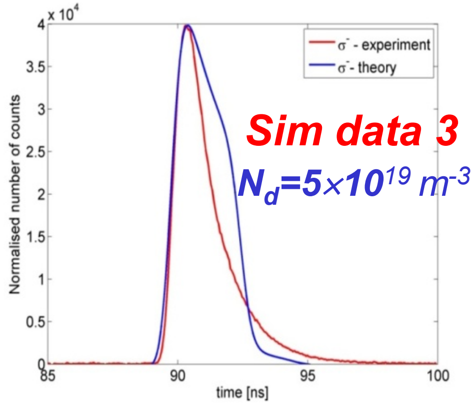


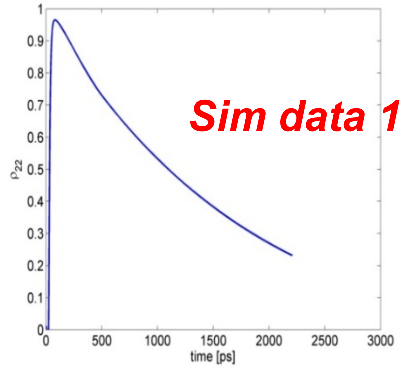


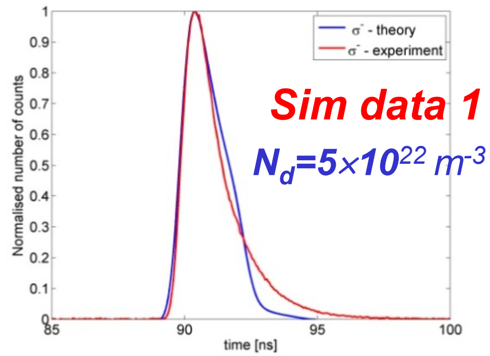


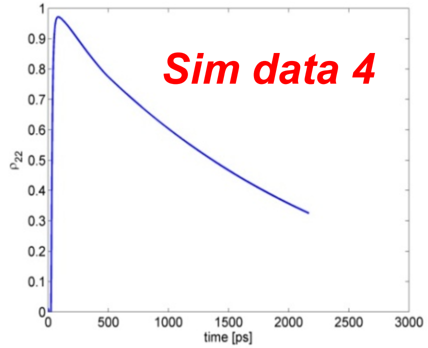
Comparison theory/experiment (N_{dot} – parameter)

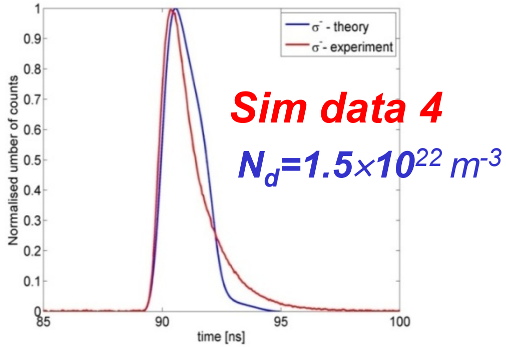


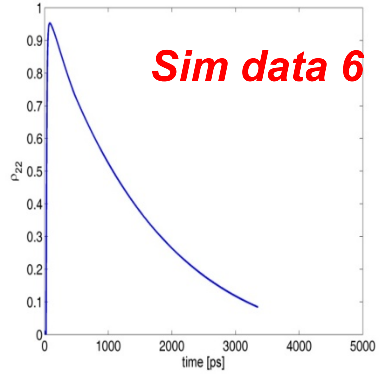


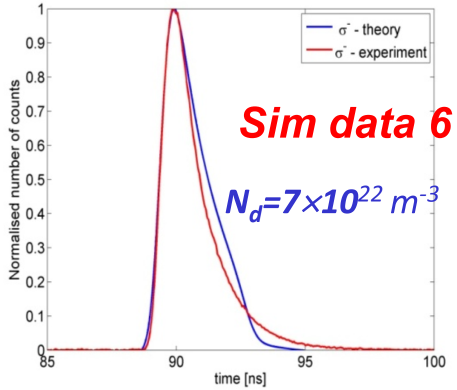


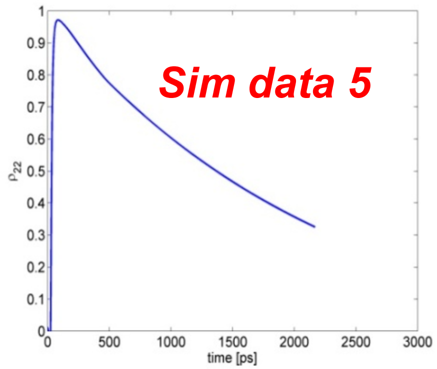


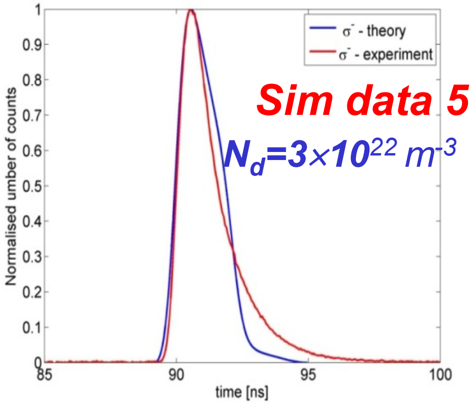




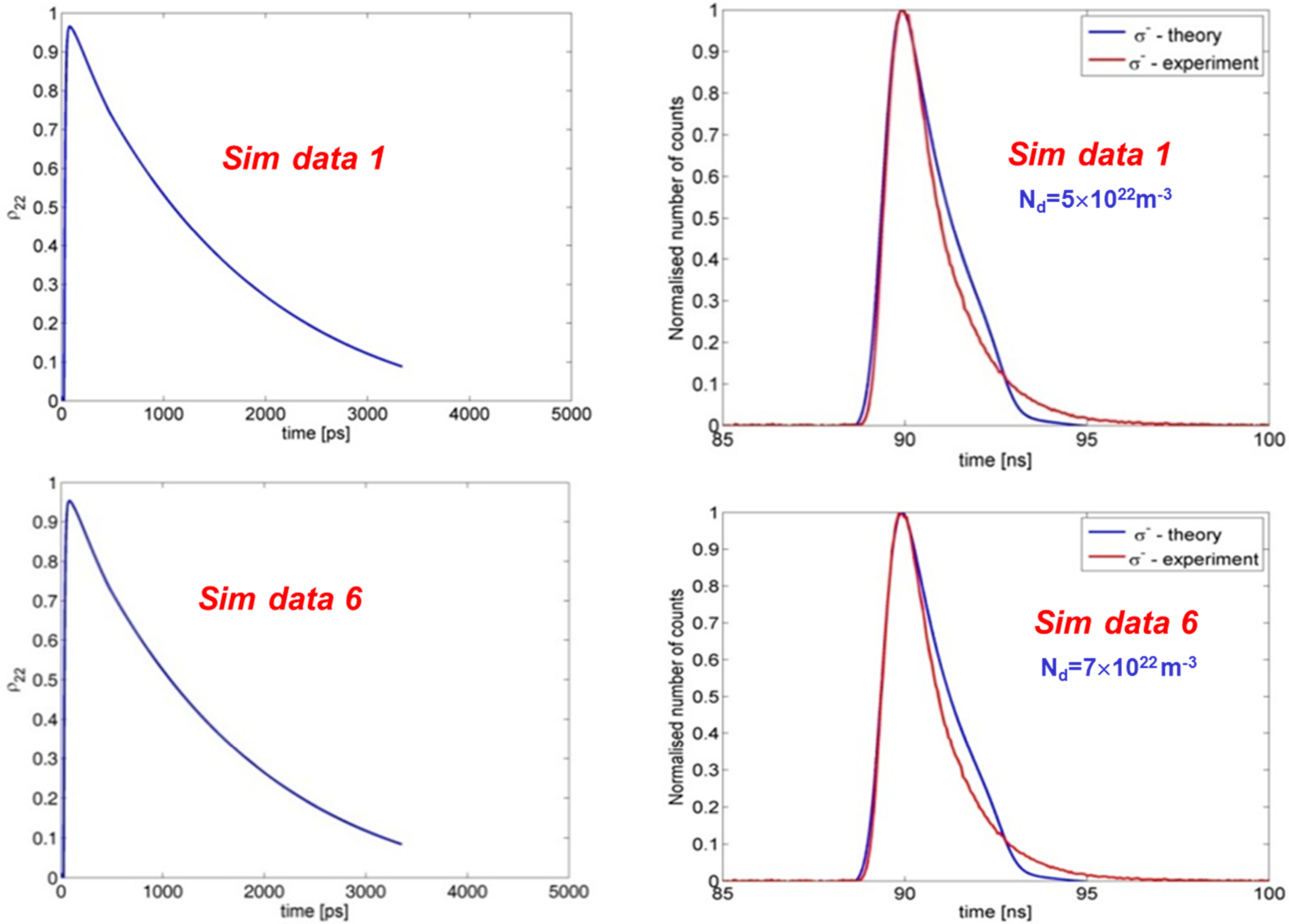








Comparison σ^- TRPL / theory after convolution



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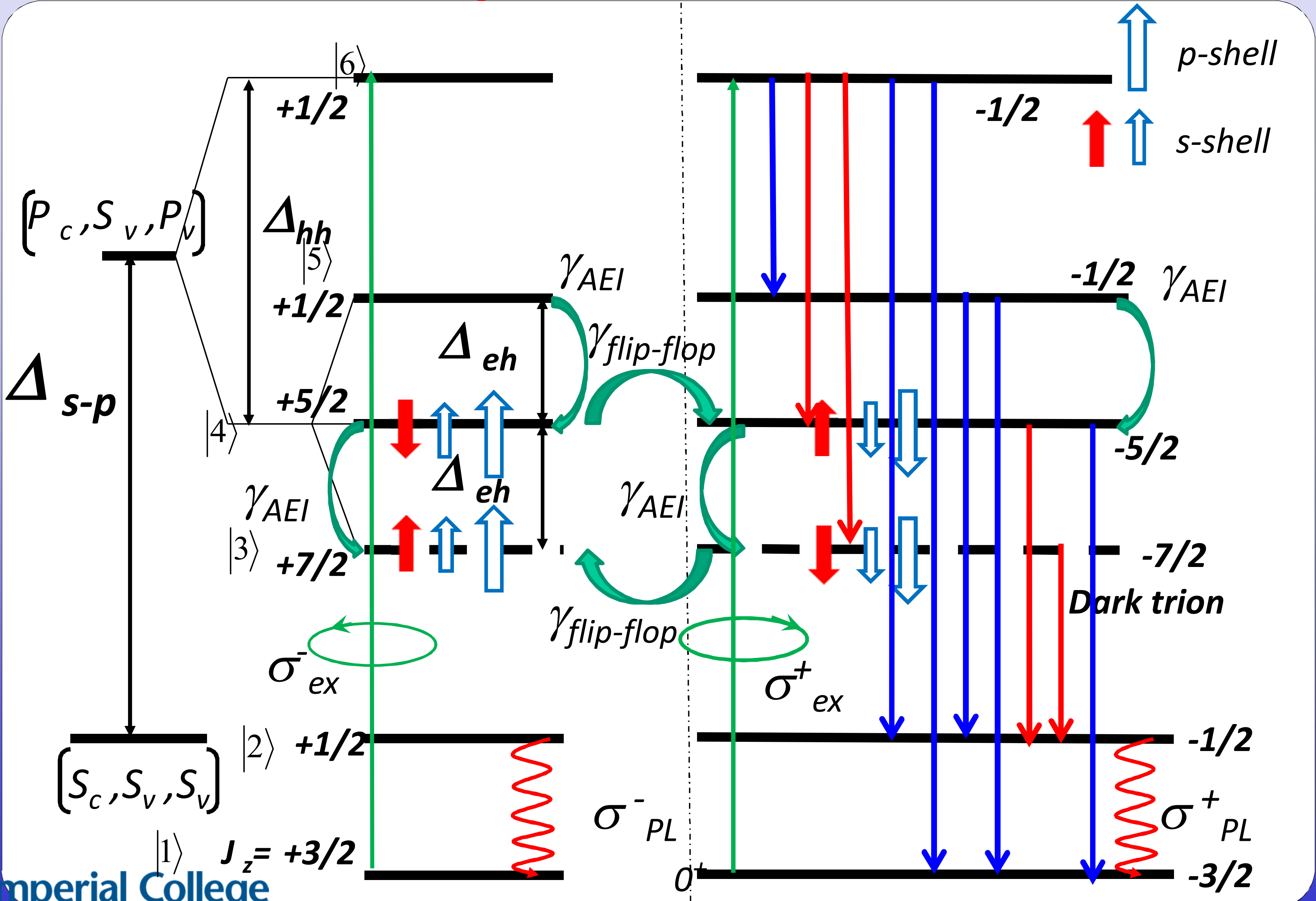
Relaxation dynamics under σ^+ - excitation



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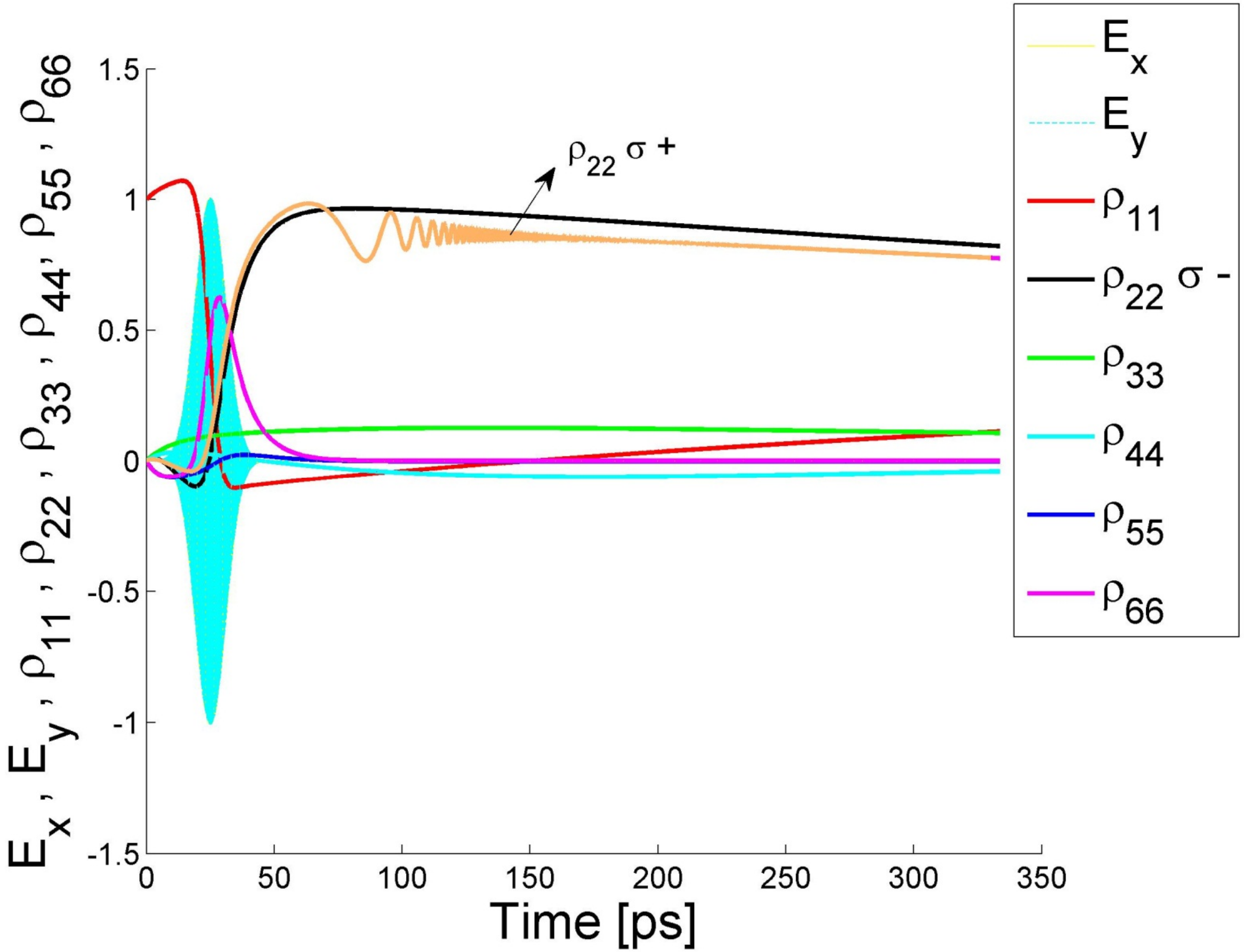
Simulation parameters for σ^+ - excitation

Key parameters:
N_{dots},
optical dipole matrix element
$\wp_{1\to 6}$, ,
radiative decay times τ_{51},τ_{41}

Parameters	Simulation data 1	Parameters	Simulation data 1
τ_{21} [ns]	1.27	Δ_{sp} [meV]	16
τ_{32} [ns]	1.0	Δ_{hh} [meV]	5.6
τ_{41} [ns]	1.6	Δ_{eh} [meV]	0.5
τ_{42} [ns]	1.5	λ_{res} [nm] (E1→6)	1125.5
τ_{51} [ps]	8	E_{trion} [eV] @1148 nm **(ground state)**	1.0815
τ_{52} [ps]	500	γ [C.m]	9.83×10^{-29}
τ_{61} [ns]	1.42	N_{dots} [m^{-3}]	7×10^{22}
τ_{62} [ps]	5	E_0 (Gauss π-pulse) [V/m]	2.689×10^5@1060 nm
τ_{63} [ps]	20	T_p [ps]	50.0
τ_{64} [ps]	35		
τ_{65} [ps]	50		
τ_{AEI} [ps]	125		

Simulated temporal dynamics under σ^+ - excitation



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Summary

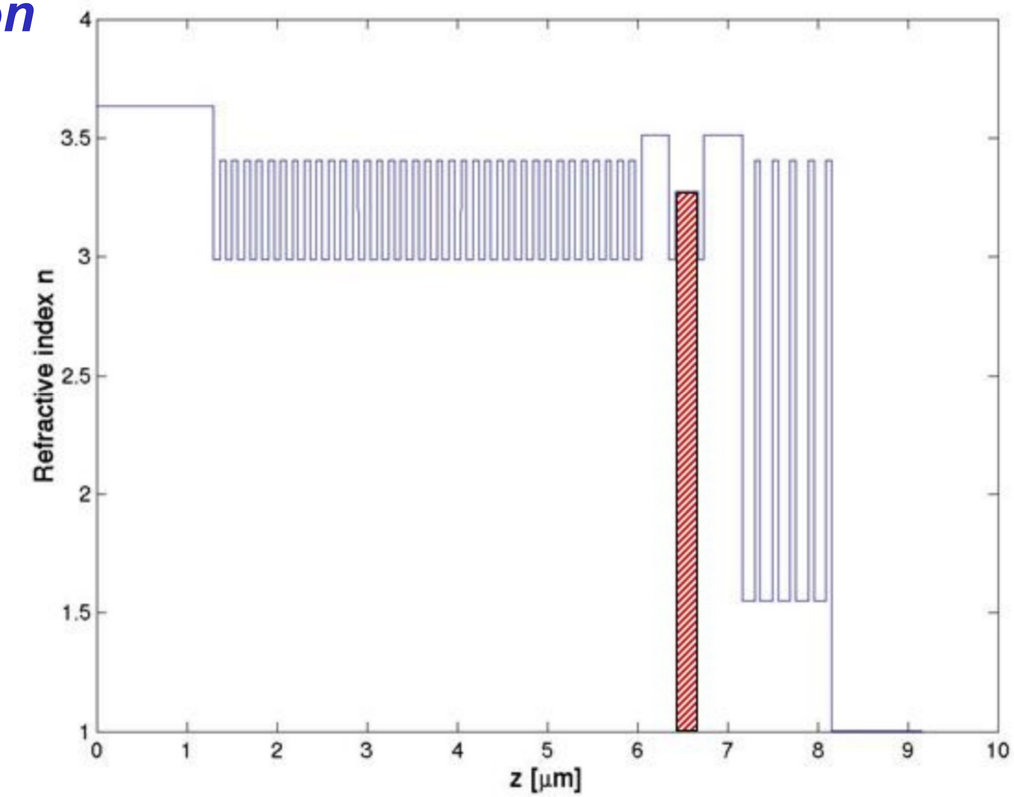
Model of the polarisation dynamics following a resonant circularly polarised pulsed excitation of the hot X^+ excited states

- ***hot trion fine structure mapped onto a discrete level system***
- ***possible spin relaxation and decoherence channels included***
- ***dipole optical selection rules applied for radiative decay processes***
- ***X^+ ground (singlet) state population dynamics compared with TRPL directly in the time domain, taking into account detector response function***
- ***excellent agreement between theory and experiment obtained***
- ***identified key parameters governing spin dynamics***
- ***extracted time scales of dominant spin relaxation and decoherence processes by comparison with experiment***

Outlook: Investigation of the nonmonotonic pulse power / duration dependence of the polarisation contrast

Quantum noise in a semiconductor microcavity

- ***Quantum fluctuations in the light field – increasingly important with scaling down device dimensions***
- ***Comprehensive model of quantum noise effects is indispensable for correct simulation of the optical field evolution***



Cavity entirely filled with gain medium ($Al_{0.5}Ga_{0.5}As$)

FDTD simulation of a semiconductor microcavity designed at λ_{res}=850 nm

Coherent optical Maxwell-Bloch equations

1D Maxwell-Bloch equations at resonance for a plane wave propagating along z and polarised along x including the damping:

$$E(\mathbf{r},t) = E_x(z,t)\hat{\mathbf{x}}$$

$$H(\mathbf{r},t) = H_y(z,t)\hat{\mathbf{y}}$$

$$P_x(t) = -\wp N_a \rho_1$$

$$\frac{\partial H_y}{\partial t} = -\frac{1}{\mu}\frac{\partial E_x}{\partial z}$$

$$\frac{\partial E_x}{\partial t} = -\frac{1}{\varepsilon}\frac{\partial H_y}{\partial z} - \frac{N_a \wp}{\varepsilon T_2}\rho_1 + \frac{N_a \wp\, \omega_o}{\varepsilon}\rho_2$$

$$\frac{\partial \rho_1}{\partial t} = -\frac{1}{T_2}\rho_1 + \omega_o \rho_2$$

$$\frac{\partial \rho_2}{\partial t} = -\omega_o \rho_1 - \frac{1}{T_2}\rho_2 + 2\frac{\wp}{\hbar}E_x \rho_3$$

$$\frac{\partial \rho_3}{\partial t} = -2\frac{\wp}{\hbar}E_x \rho_2 - \frac{1}{T_1}(\rho_3 - \rho_{30})$$

T_1, T_2 – population relaxation time, dephasing time

ρ_{30} = 1 initial population profile

Random E-field fluctuations and quantum noise
Langevin formalism

Spontaneous emission modelled by Langevin random noise term in Maxwell's equations within the cavity:

$$\frac{\partial}{\partial t}\left(E_x(z,t)+\delta E_x(z,t)\right)=-\frac{1}{\varepsilon}\frac{\partial H_y}{\partial z}-\frac{N_A\wp}{\varepsilon T_2}\rho_1+\frac{N_A\wp\,\omega_0}{\varepsilon}\rho_2$$

Pseudorandom number generator (Box-Müller method) for generating random deviates with a normal (Gaussian) distribution from uniformly distributed in the interval (0,1) random numbers : a,b

White Gaussian noise with variance ($\xi_E = \sigma^2 = 0.001\ V^2 m^{-2}$) implemented at each time step:

$$E_j(z)=E_j(z)+\sqrt{-2\,\xi_E\ln(a)}\cos(2\pi b)$$

G. Slavcheva et al., J of Sel. Top. in Quantum Electronics 10, 1052 (2004)

$$E_j(z)=E_j(z)+\sqrt{-2\,\xi_E\ln(a)}\cos(2\pi b)$$

- **Moments of the random distribution:**

$$\langle \delta E_x(z,t) \rangle = 0$$

$$\langle \delta E_x(z,t)\delta E_x(z,t') \rangle = \tilde{\xi}_E \delta(t-t') = \xi_E R_{sp} \delta\left(\frac{t-t'}{\Delta t}\right)$$

Spontaneous emission rate inferred from comparison with the stochastic rate equations *(G. Gray et al., PRA 40, 2425, 1989)*:

$$\langle F_E(t) F_E(t') \rangle = R_{sp}\delta(t-t')$$

$$R_{sp} = \frac{\sqrt{\xi_E}(\varepsilon_0\varepsilon)}{N_A \wp T_2}$$

Parameters: λ=0.85 μm, n=3.2736, T_1=10 ps, T_2 =70 fs, $\wp$=4.8×10^{-28} Cm, resonant dipole density N_A^{3D}=1.0×10^{24} m^{-3}, E_0= 700 V/m

For $\xi_E=1\times10^{-3}\ V^2m^{-2} \Rightarrow R_{sp}\sim2.84026\times10^{10}\ s^{-1}$; $\tau_{sp}\sim 35$ ps

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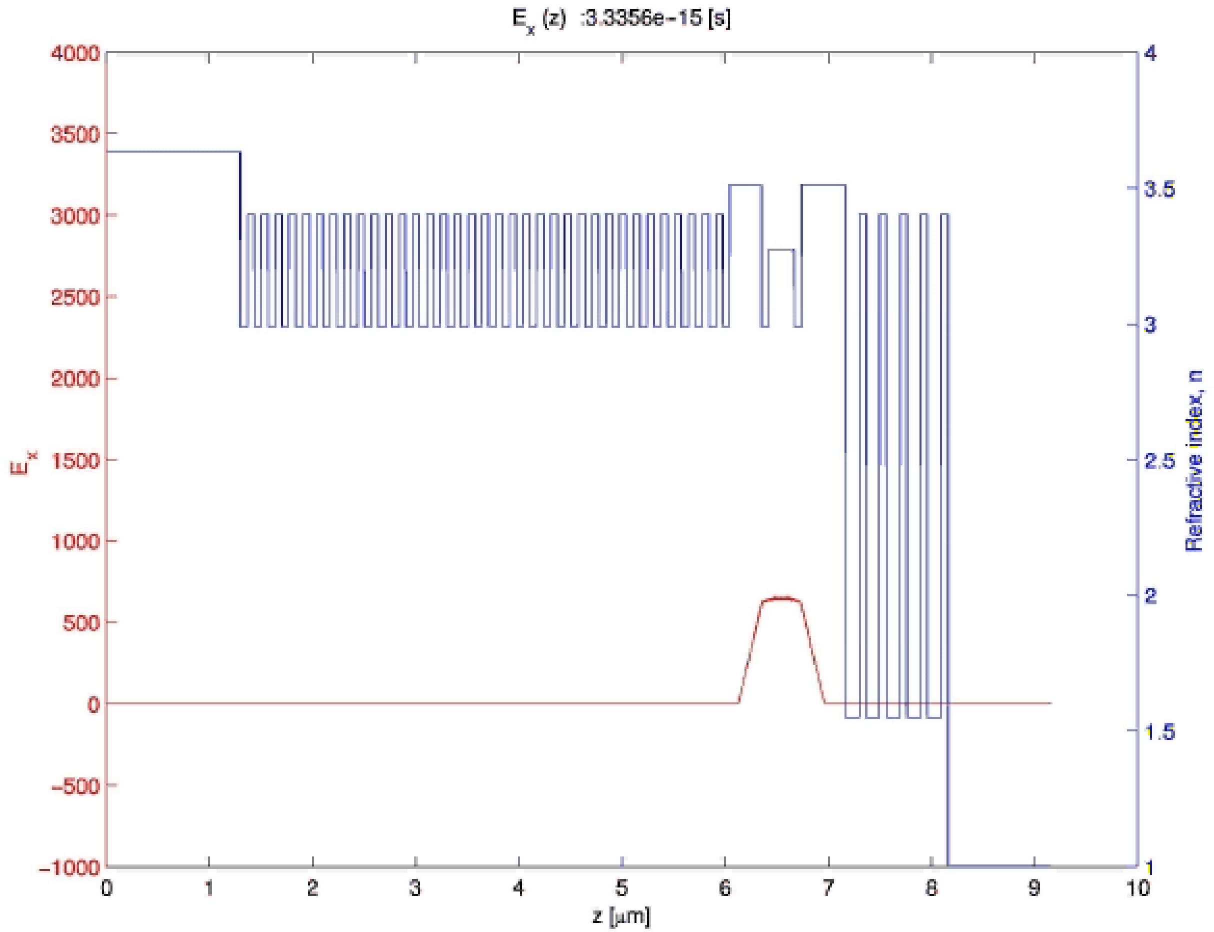
Intracavity dynamics

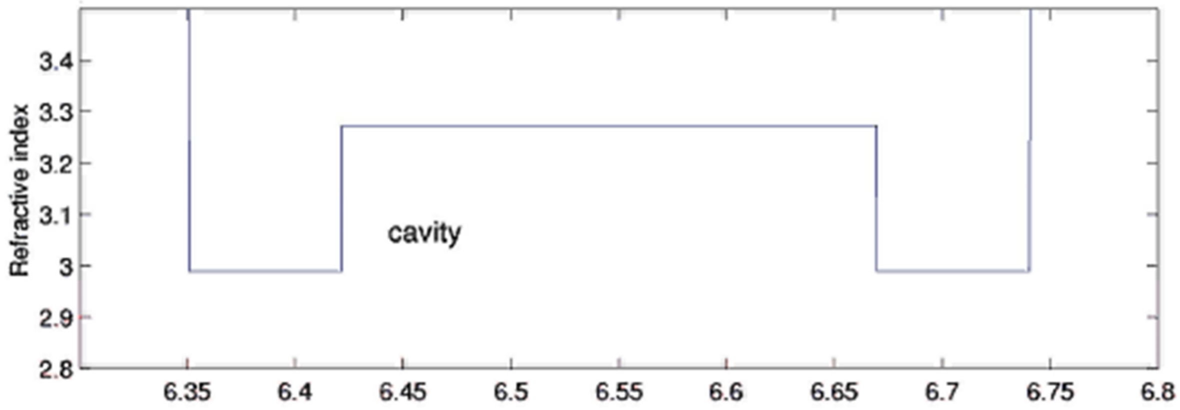
Electric field build up within the cavity due solely to the noise background as a function of time elapsed

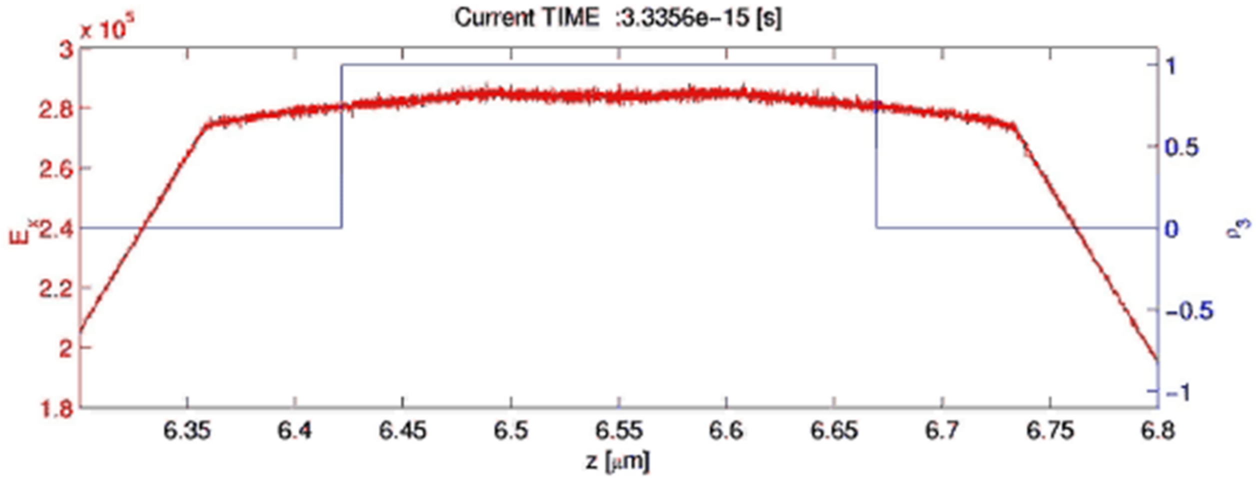
ρ_{30} **=1 (gain) within the cavity**



DBRs: bottom – 35.5 pairs AlAs/$Al_{0.3}Ga_{0.7}As$, top – 5 pairs AlO/$Al_{0.3}Ga_{0.7}As$, cavity – $Al_{0.5}Ga_{0.5}As$; n=3.27

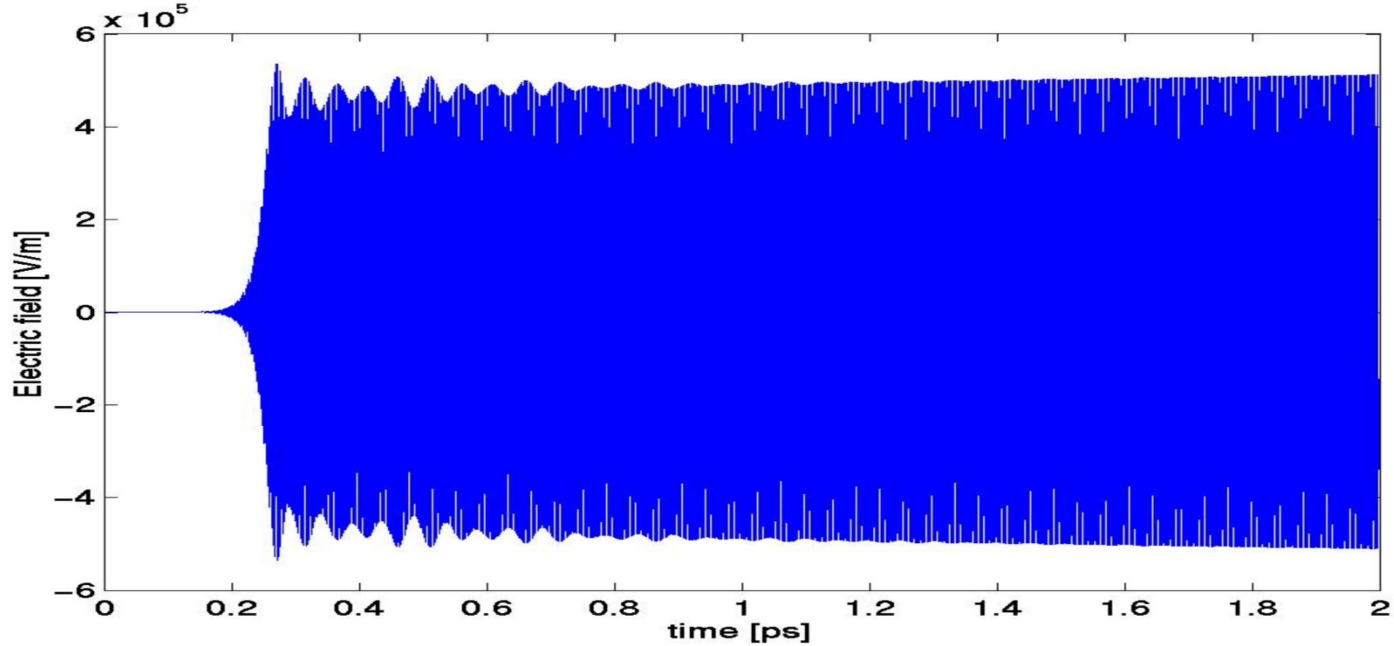
Time evolution of the intracavity field and population inversion



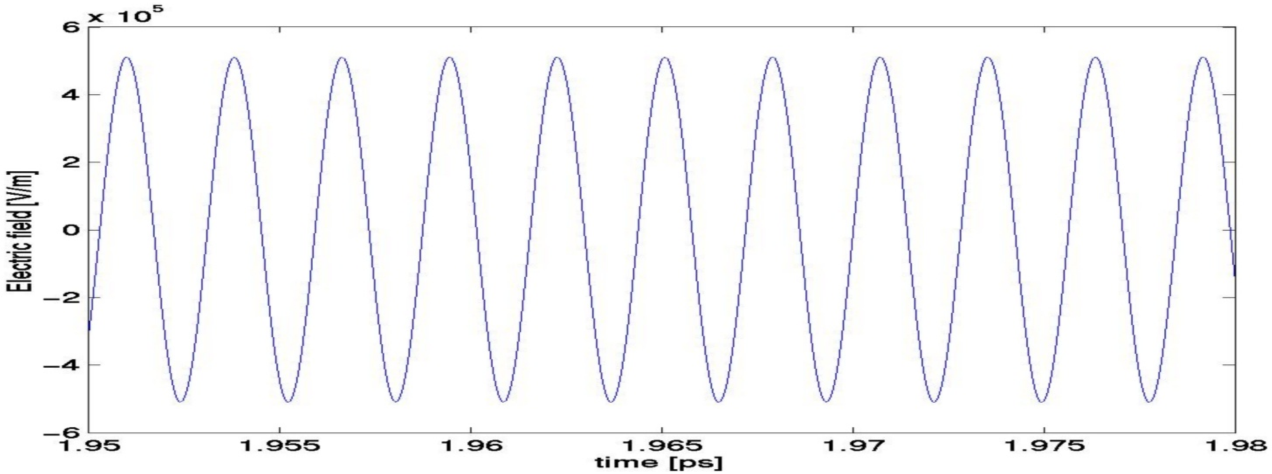


Fast Rabi oscillations of the population inversion resulting in relaxation oscillations of the electric field envelope

Generation of coherent oscillations (lasing)



λ=844.216 nm (lasing wavelength)

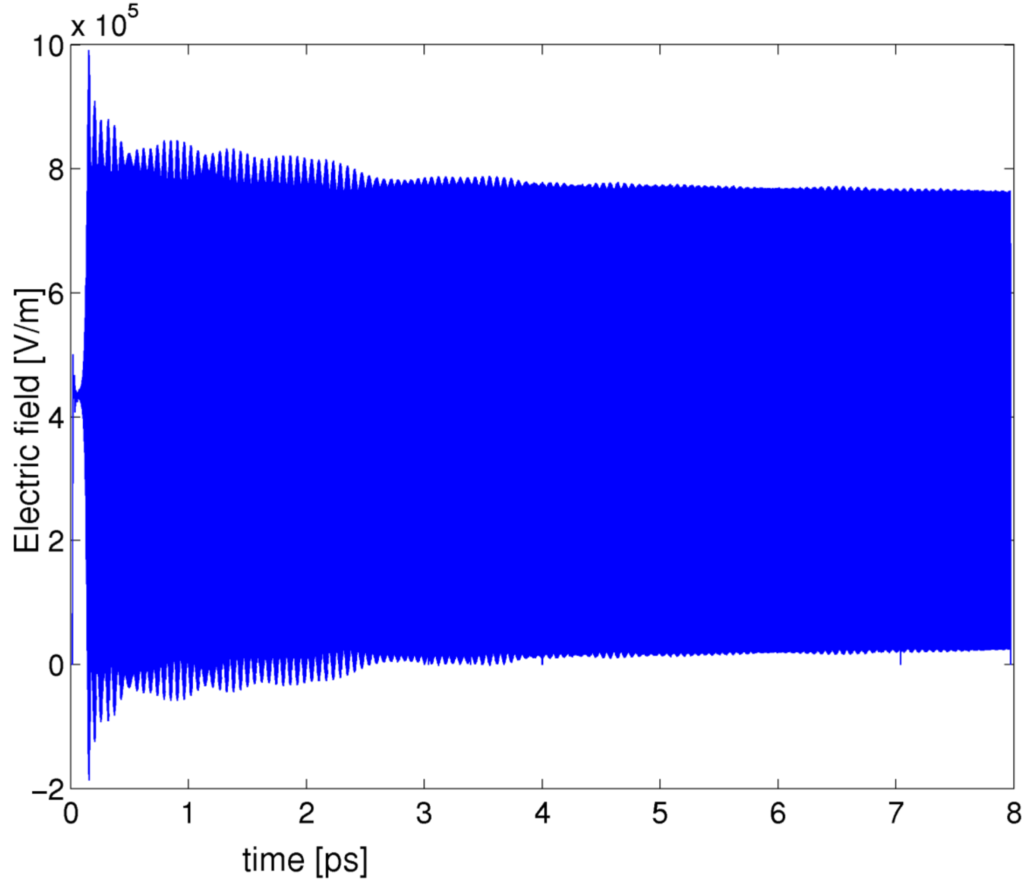


Gain saturation to a steady state

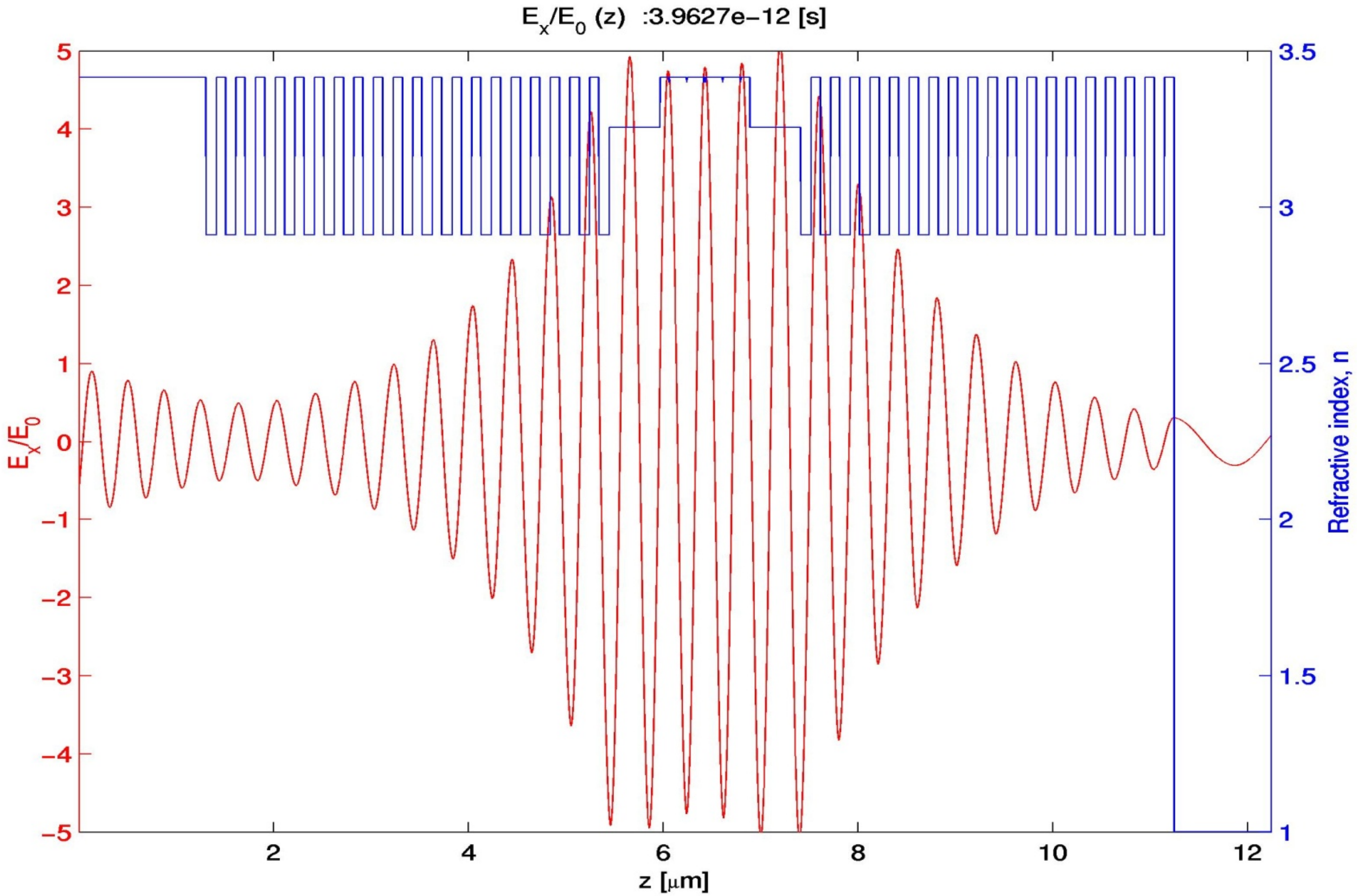
λ=0.85 μm, n=3.2736, T_1=1 ns, T_2 =10 ps, ℘=4.8×10^{-28} Cm, resonant dipole density N_A =1.0×10^{24} m^{-3}, E_0=2.8 ×10^5 V/m

Saturation condition:

$$\Omega_R^2 T_1 T_2 \approx 16241 >> 1$$

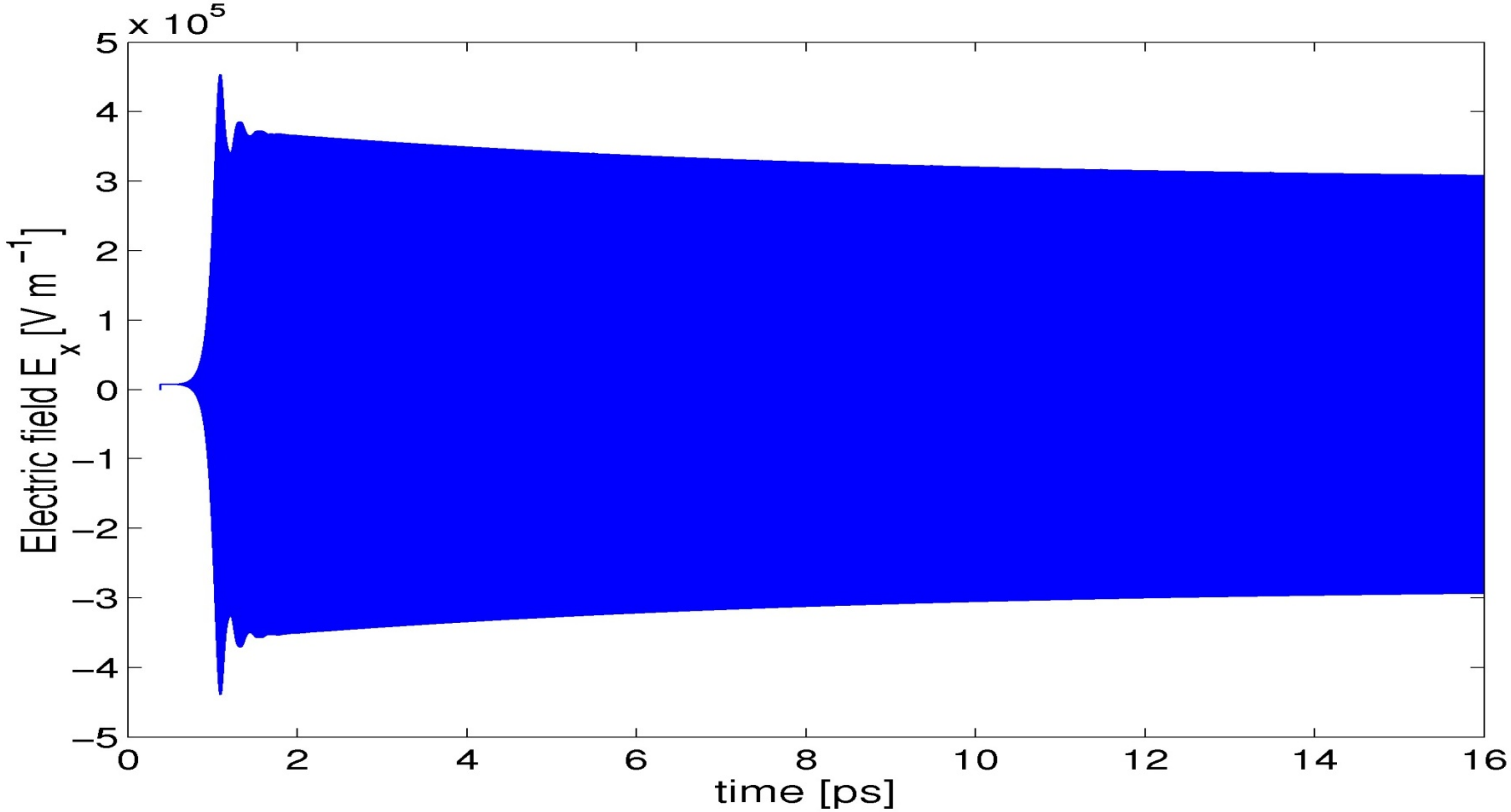


Standing wave profile
including QW resonant absorption λ=1.29187 μm



DBRs: bottom – 20.5 pairs AlAs/GaAs, top – 19 pairs AlAs/GaAs, cavity – 5-λ with 6 $Ga_{0.63}In_{0.37}AsN_{0.012}$ QWs; n=3.4

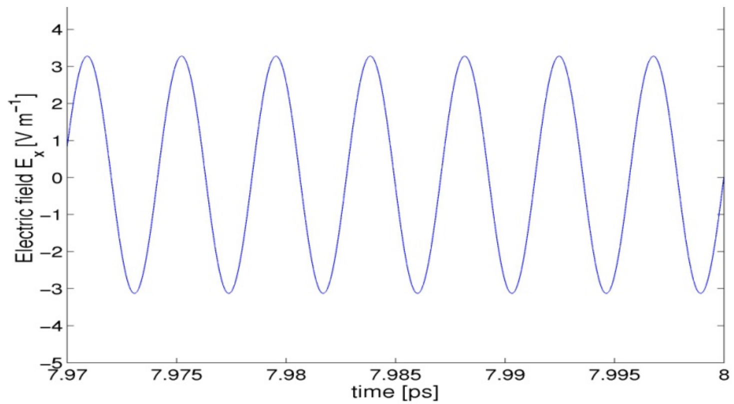
Build up of coherent oscillations



Relaxation-oscillation decay rate:

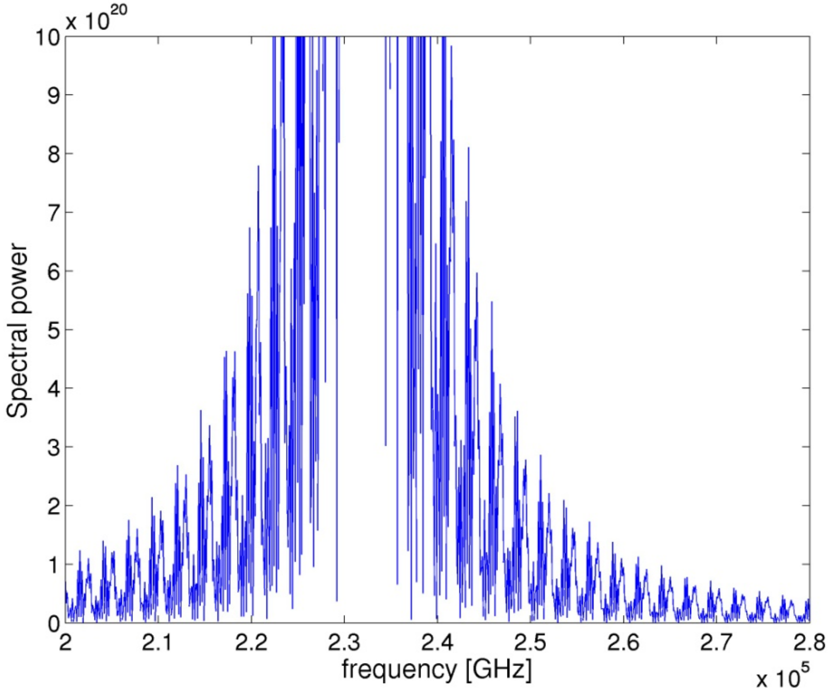
$\Gamma_R \sim 1.5 \times 10^{11}\ s^{-1}$

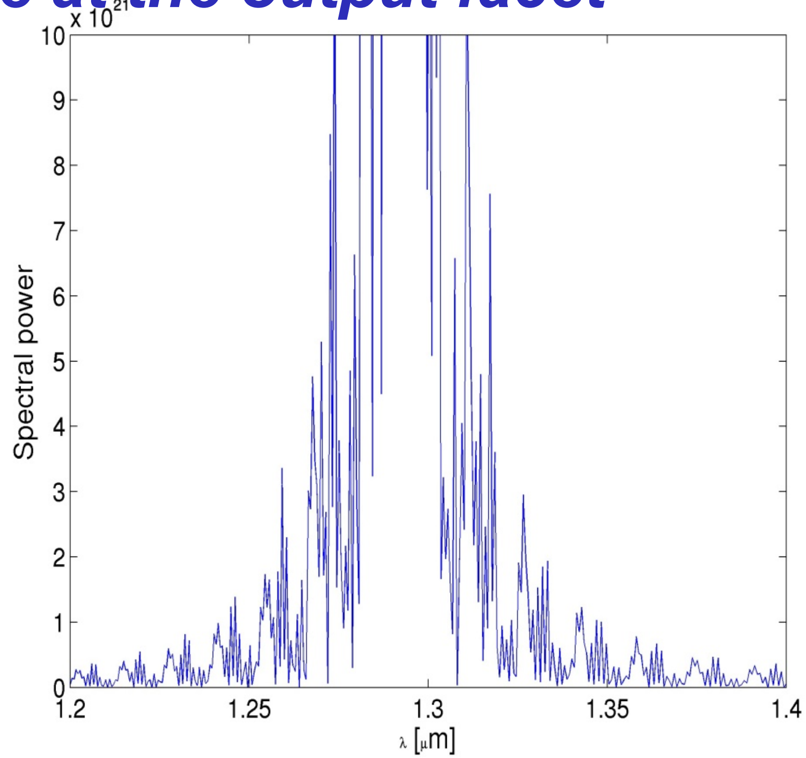
$\lambda = 1.289\ \mu m$



FDTD-computed laser line width and ultrafast relaxation oscillations spectrum

Discrete FFT of the time trace at the output facet





$\omega_0 \pm n\Omega_R$ **Relaxation oscillation sidebands**

Frequency of relaxation oscillations $\Omega_R/2\pi \sim 2.54$ THz

Relaxation-oscillation decay rate: $\Gamma_R \sim 1.5 \times 10^{11}$ s^{-1}

Outlook

Quantum Stochastic Approach to Non-classical Dot-Nanocavity Radiation

A strongly-coupled quantum dot (QD) in a cavity: a solid-state analogue of the atom-cavity system in quantum optics

Reports of signatures of non-classical light in solid-state systems have just started to emerge: evidence remains inconclusive,

Kasprzak et al., Nature Mater. 9, 304 (2010);
Faraon et al., Nature Physics, 4, 859 (2008)
Hennessy et al. , Nature 445, 896 (2007)

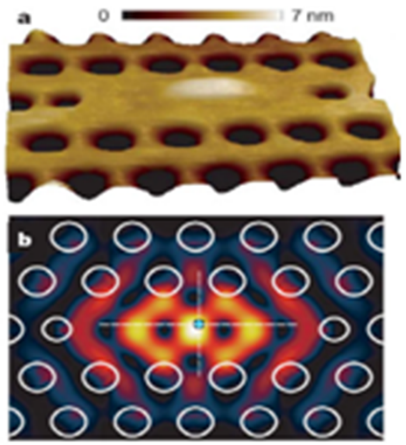


Fig.1. (a) AFM of a QD buried in a L3 PhC nanocavity; (b) Cavity mode showing a QD positioned at the electric field maximum (from *Hennessy et al, Nature 445,896, 2007)*

Extreme few-photon strong coupling regime

- *Large photon densities: quantum noise is necessary to explain laser line width and threshold behaviour*
- *Low photon densities: light is generated by spontaneous emission, described in terms of single photon emission events within the single-particle Dirac picture* *P.A.M. Dirac, Proc. R. Soc. Lond. A 11, 243 (1927)*
- *Control over such noise effects crucial in the design of devices for QIP*

Quantum Stochastic Equations

- **Resonance fluorescence** of an atomic system (a single QD) displays a sequence of bright and dark periods
- **Quantum jump approach** needed *: taking into account the results of a quantum mechanical measurement (photon counting)
- **Quantum jump approach** ⇔ **Quantum Stochastic Equations**, shown to be identical to the semiclassical Maxwell-Bloch equations + Langevin terms
- Quantum theory of propagation of non-classical radiation in a near-resonant system: introduce Langevin noise source terms in Maxwell's equations and pseudospin equations for the field and polarisation
- FDTD solution directly in the time domain for a few-photon excitation (ultralow photon intensity limit)

*M. B. Plenio, P. L. Knight, Rev. Mod. Phys. 70, 101 ,1998
C. W. Gardiner, A. S. Parkins, P. Zoller, Phys. Rev. A 46, 4363 (1992); C. W. Gardiner, Quantum Noise (Springer, Berlin), 1992.
P.D. Drummond and M. G. Raymer, Phys. Rev. A, 44, 2072 (1991)