Optical Manipulation of an Electron Spin in Quantum Dots

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Electron Spin Manipulation for Quantum Computer

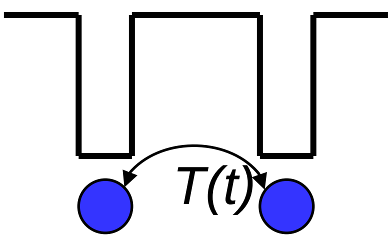
- General physical interest to this problem (R. Feynman, 1983)
- Shor's algorithm for large number factorization (1994)

The major difference from a computer is replacement of a bit ("on" or "off" states) by a qubit (a coherent superposition of "on" and "off" states)

A single electron spin in a QD is a natural qubit [Loss and DiVincenzo, *PRA*(1998)]

Universal quantum computation requires:

- Arbitrary 1-qubit rotations (single spin rotation)
  - Performed by turning on local magnetic field
- A single 2-qubit gate operation (spin-spin interaction)
  - spin-spin exchange interaction controlled by gates



Coulomb repulsion U in each dot. Tunneling $T(t)$ between the dots. Effective coupling $J(t) \sim T^2(t)/U$. Tunneling $T(t)$ controlled by varying gate voltage.

- Fast operations in order to keep a coherent superposition

Optical Manipulation of Electron Spin

Critical Steps for Implementation of a Solid State Based Quantum Computer

- **Initialization**
- **One qubit operation**
- Entanglement of 2 qubits
- Read-out

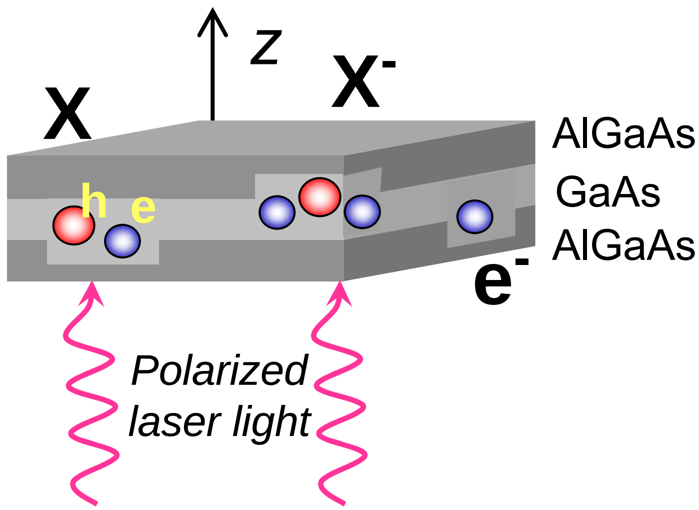
Optical methods provide high speed techniques of spin control and manipulation and allow to access an electron spin locally.

• Non-resonant optical pumping of an electron spin in negatively charged quantum dots [Phys. Rev. Lett. **94**, 047402 (2005)]

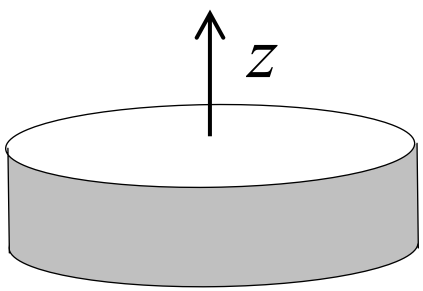
•Optical initialization of electron spins by resonant π-pulses of $\sigma^{\pm}$ -polarized light [Phys. Rev. B **68,** 201305(R) (2003)]

•Optical control of spin coherence in charged QDs [condmat/0603020]

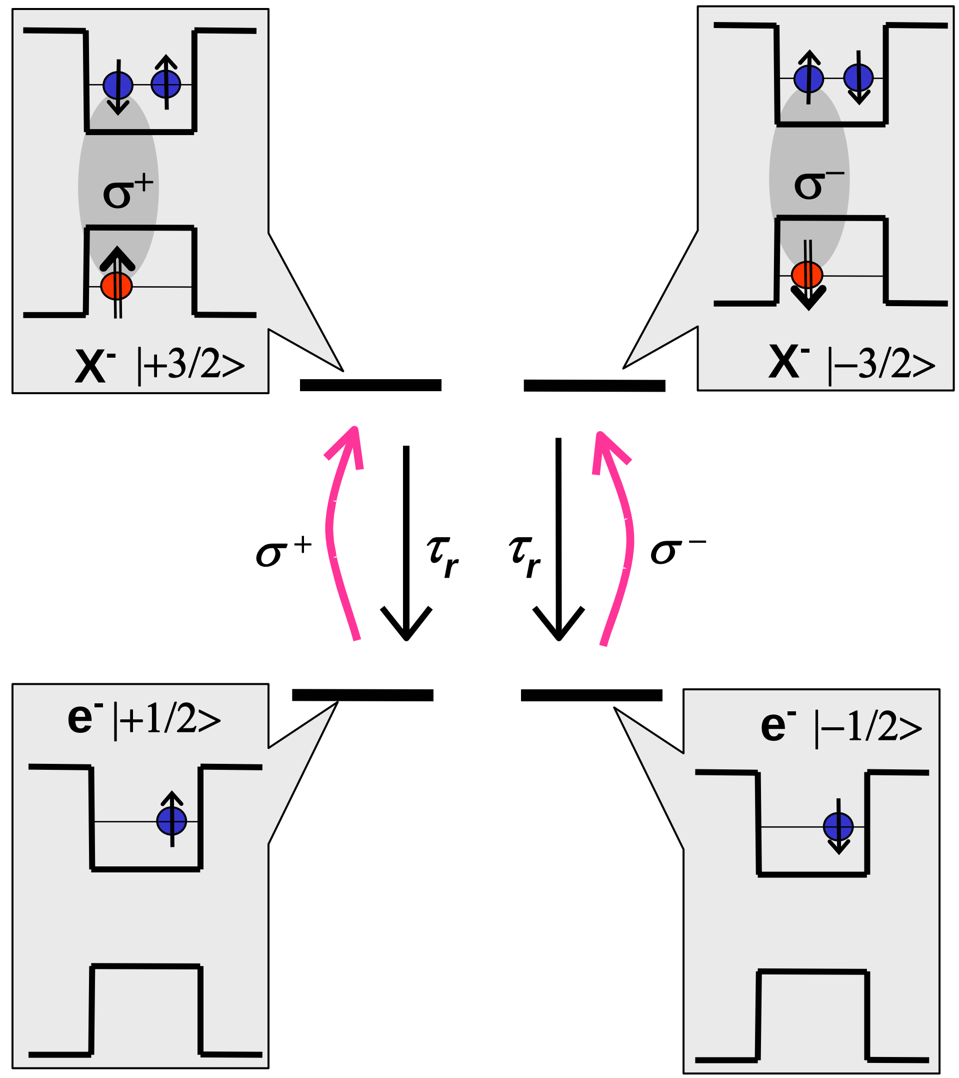
Optical excitations in singly charged QD



Disk-shaped QDs



The ground electron and hole states are |±1/2> and |±3/2>, respectively.

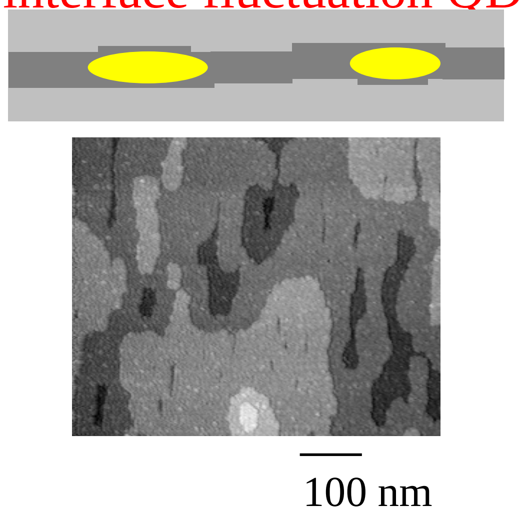


PL polarization is controlled by the hole spin projection

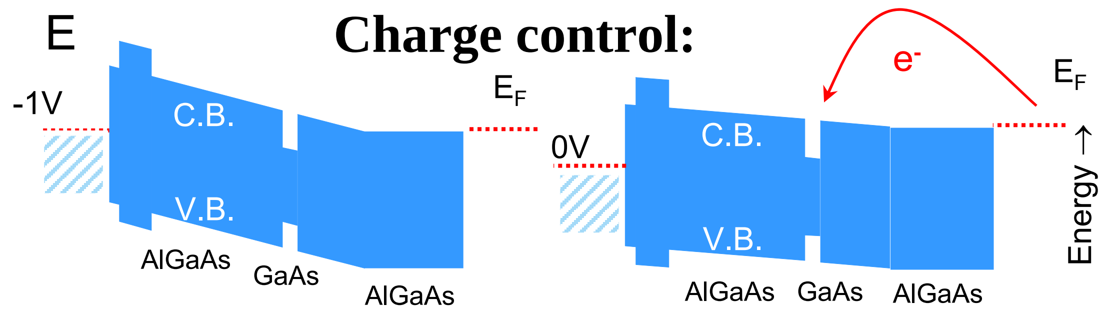
Single Charge-Tunable QD Spectroscopy

Exp: on non-resonant pumping

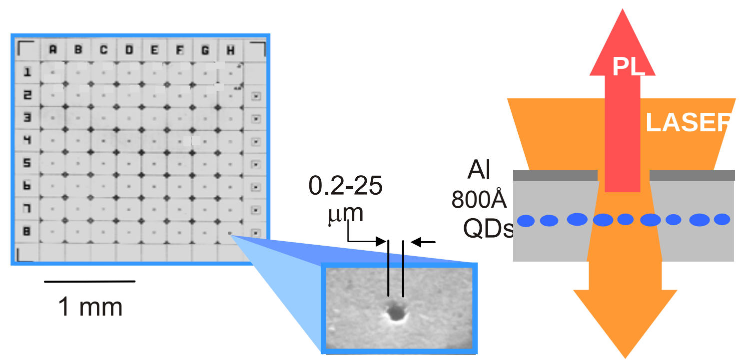
interface fluctuation QDs



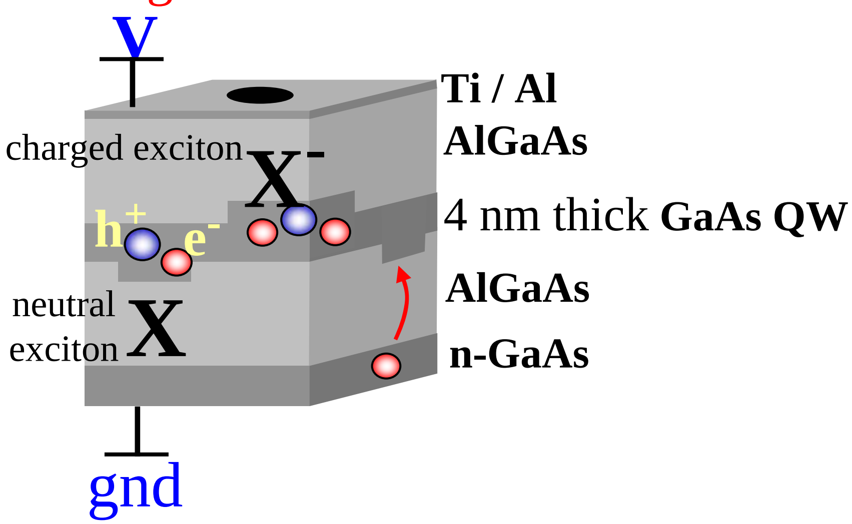
Charge control:

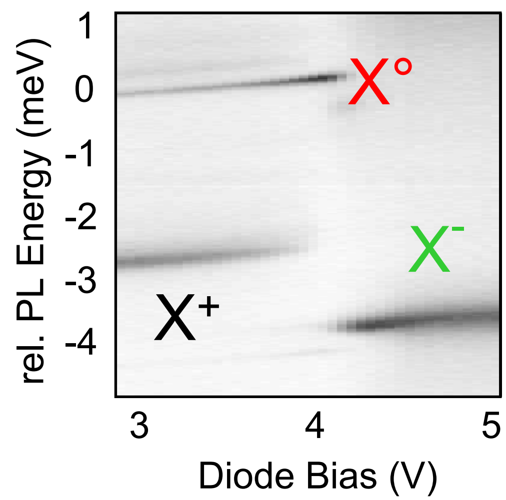


Schottky diode heterostructure



charge state control

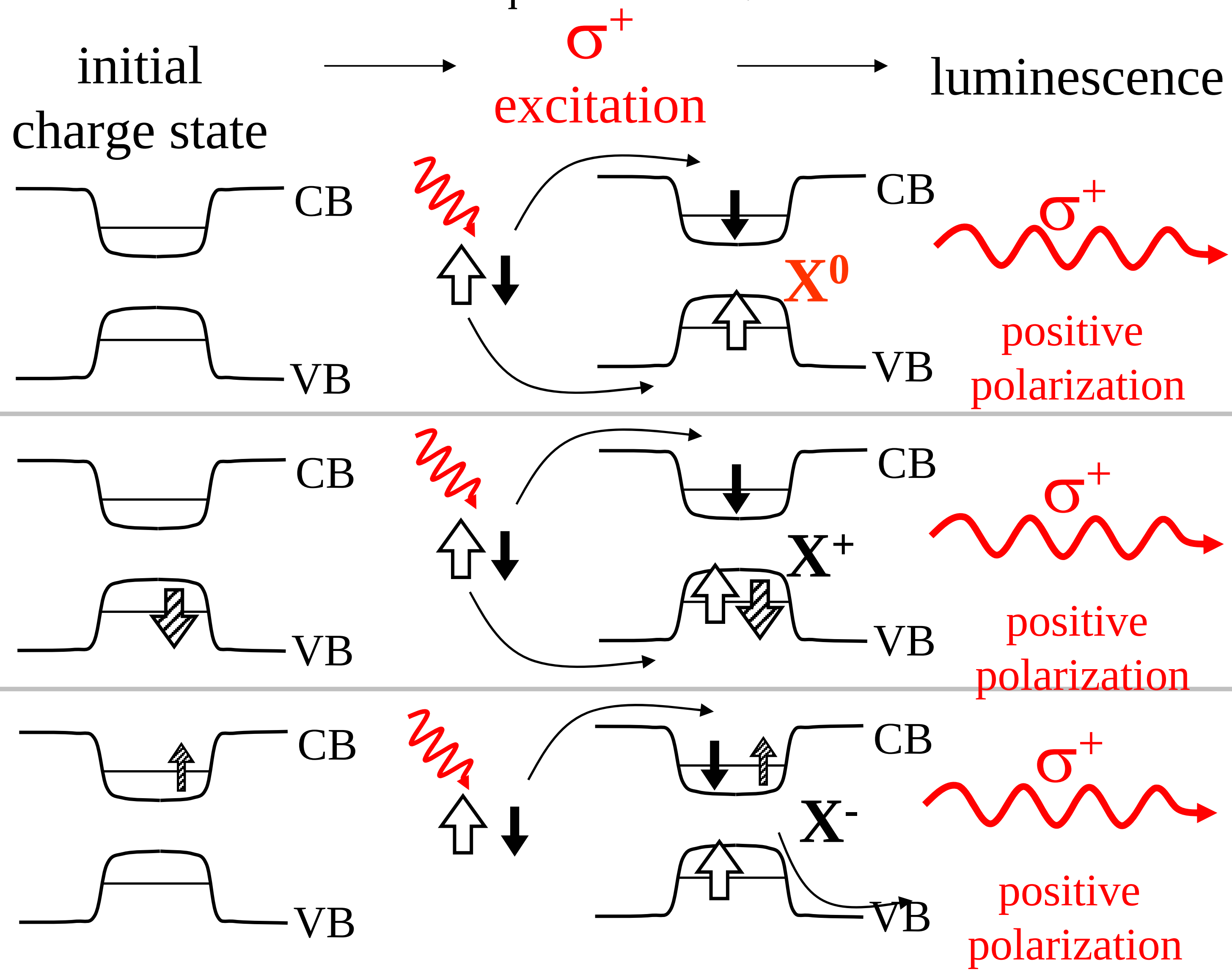




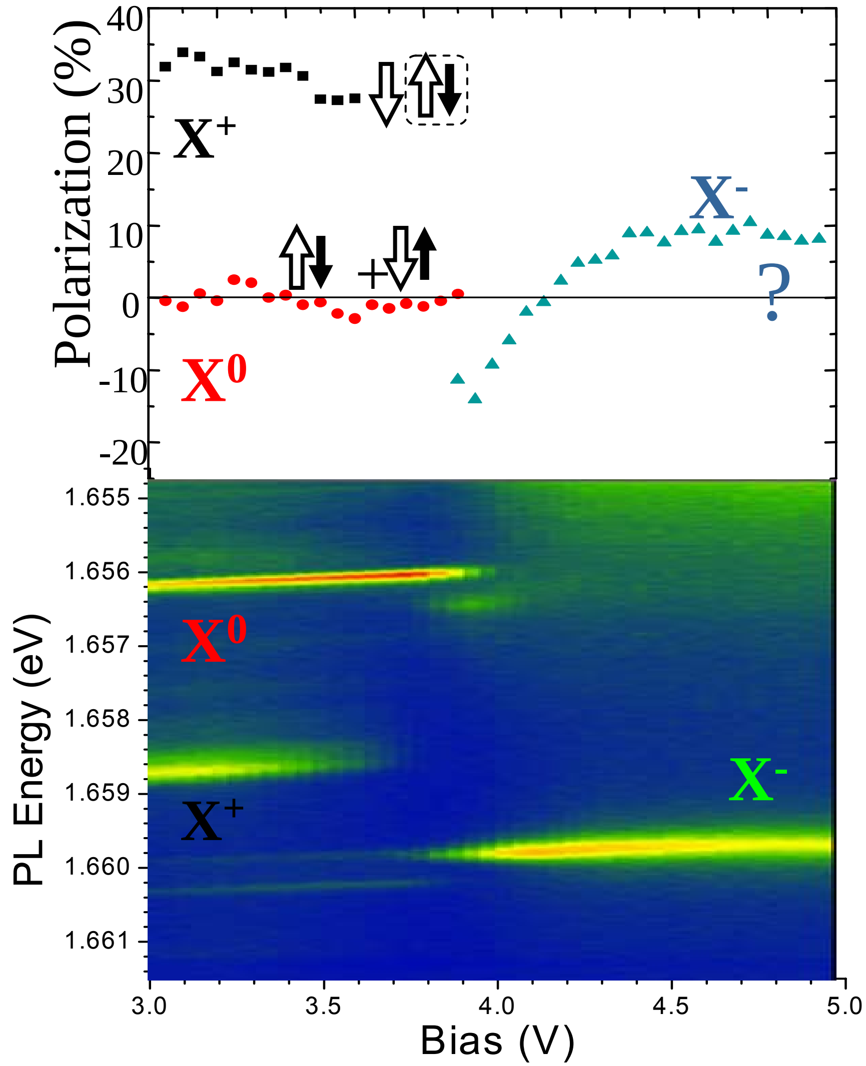
Photoluminescence Polarization Memory Effect

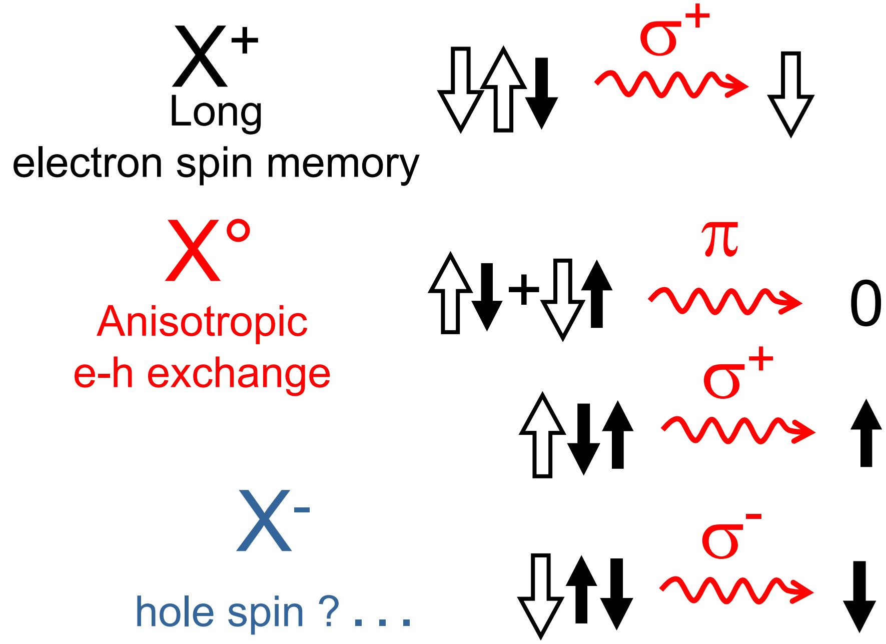
(initial expectations)

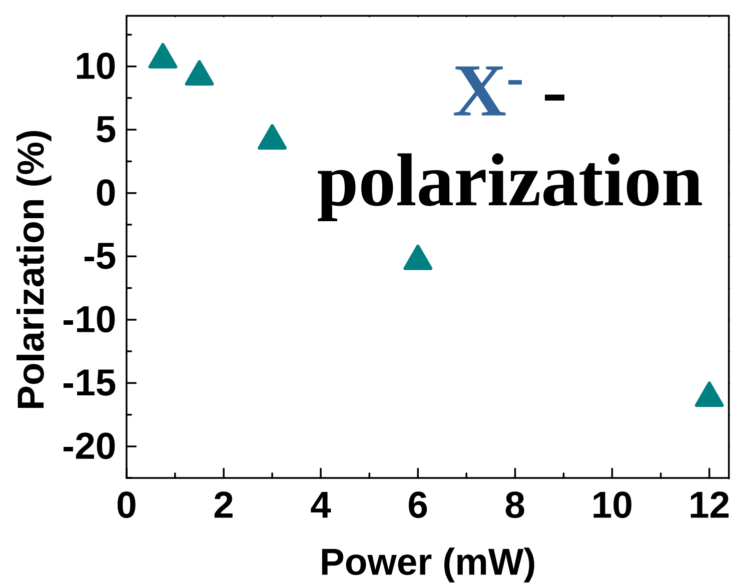
Nonresonant excitation in GaAs quantum well:



PL Polarization Memory Effect: Experiment







Negative PL Polarization Degree of X^- in QD

Why is X^- polarization degree negative, why does it grow with pumping intensity and changes sign with bias?

Negative polarization was observed in ensembles of charged QDs:

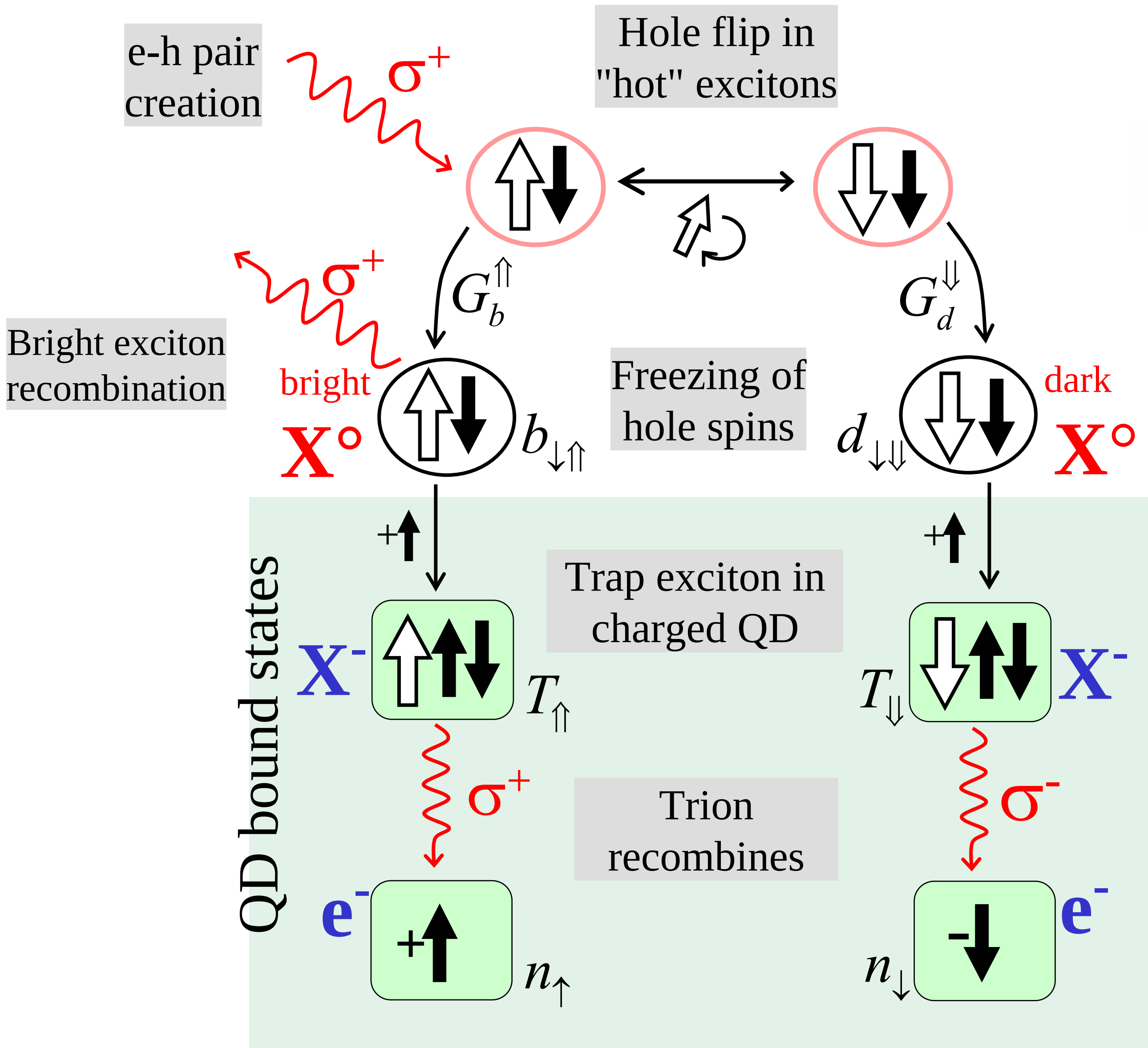
Dzhioev et al. *Phys. Solid State* **40**, 1587 (1998)
Cortez et al. *Phys Rev. Lett.* **89**, 207401 (2002)
Kalevich et al. *phys. stat. sol.* “b” **238**, 250 (2003)

Spectroscopy of a single charge QD [Bracker et al. *Phys. Rev. Lett.* 94, 047402 (2005)] allows us to suggest a model that

1. Describes **unusual PL polarization properties** of charged quantum dots.

2. Explains mechanism responsible for **optical pumping of electron spins**

Mechanism for X$^-$ polarization



$$G_b^{\Uparrow} > G_d^{\Downarrow}$$

Dark exciton can only form trion

X$^-$ polarization:

$$\eta = \frac{T_{\Uparrow} - T_{\Downarrow}}{T_{\Uparrow} + T_{\Downarrow}} = \frac{b_{\downarrow\Uparrow} - d_{\downarrow\Downarrow}}{b_{\downarrow\Uparrow} + d_{\downarrow\Downarrow}}$$

These Processes are Described:

$$\frac{dT_{\Uparrow}}{dt} = W\left(n_{\uparrow} \times b_{\downarrow\Uparrow} + n_{\downarrow} \times d_{\uparrow\Uparrow}\right) - \frac{T_{\Uparrow}}{\tau_T}$$

$$\frac{dT_{\Downarrow}}{dt} = W\left(n_{\downarrow} \times b_{\uparrow\Downarrow} + n_{\uparrow} \times d_{\downarrow\Downarrow}\right) - \frac{T_{\Downarrow}}{\tau_T}$$

$$\frac{dn_{\uparrow}}{dt} = -\Omega S_x - Wn_{\uparrow}\left(b_{\downarrow\Uparrow} + d_{\downarrow\Downarrow}\right) + \frac{T_{\Uparrow}}{\tau_T} - \frac{n_{\uparrow} - n_{\downarrow}}{\tau_S}$$

$$\frac{dn_{\downarrow}}{dt} = \Omega S_x - Wn_{\uparrow}\left(b_{\uparrow\Downarrow} + d_{\uparrow\Uparrow}\right) + \frac{T_{\Downarrow}}{\tau_T} - \frac{n_{\downarrow} - n_{\uparrow}}{\tau_S}$$

$$\frac{dS_x}{dt} = \frac{\Omega}{2}\left(n_{\uparrow} - n_{\downarrow}\right) - \frac{S_x}{\tau_S} - \frac{W}{2} S_x\left(b_{\uparrow\Downarrow} + b_{\downarrow\Uparrow} + d_{\downarrow\Downarrow} + d_{\uparrow\Uparrow}\right)$$

$$\frac{db_{\uparrow\Downarrow}}{dt} = G_b^{\Downarrow} - \frac{b_{\uparrow\Downarrow}}{\tau_b} - Wn_{\downarrow} \times b_{\uparrow\Downarrow} - \frac{b_{\uparrow\Downarrow} - d_{\uparrow\Uparrow}}{\tau_H} - \frac{b_{\uparrow\Downarrow} - b_{\downarrow\Uparrow}}{\tau_{EX}}$$

$$\frac{db_{\downarrow\Uparrow}}{dt} = G_b^{\Uparrow} - \frac{b_{\downarrow\Uparrow}}{\tau_b} - Wn_{\uparrow} \times b_{\downarrow\Uparrow} - \frac{b_{\downarrow\Uparrow} - d_{\downarrow\Downarrow}}{\tau_H} - \frac{b_{\downarrow\Uparrow} - b_{\uparrow\Downarrow}}{\tau_{EX}}$$

$$\frac{dd_{\uparrow\Uparrow}}{dt} = G_d^{\Uparrow} - Wn_{\downarrow} \times d_{\uparrow\Uparrow} - \frac{d_{\uparrow\Uparrow} - b_{\uparrow\Downarrow}}{\tau_H}$$

$$\frac{dd_{\downarrow\Downarrow}}{dt} = G_d^{\Downarrow} - Wn_{\uparrow} \times d_{\downarrow\Downarrow} - \frac{d_{\downarrow\Downarrow} - b_{\downarrow\Uparrow}}{\tau_H}$$

$$N = n_{\uparrow} + n_{\downarrow} + T_{\Uparrow} + T_{\Downarrow}$$

Steady State Solution

If the hole spin relaxation time τ_H is very long the steady state concentrations of Bright, $\boldsymbol{b}_{\downarrow\Uparrow}$, and Dark, $\boldsymbol{d}_{\downarrow\Downarrow}$, excitons is determined:

$$d_{\downarrow\Downarrow} = \frac{G_d^{\Downarrow}}{Wn_{\uparrow}}$$

$$b_{\uparrow\Downarrow} = \frac{G_b^{\Uparrow}}{Wn_{\uparrow} + 1/\tau_b}$$

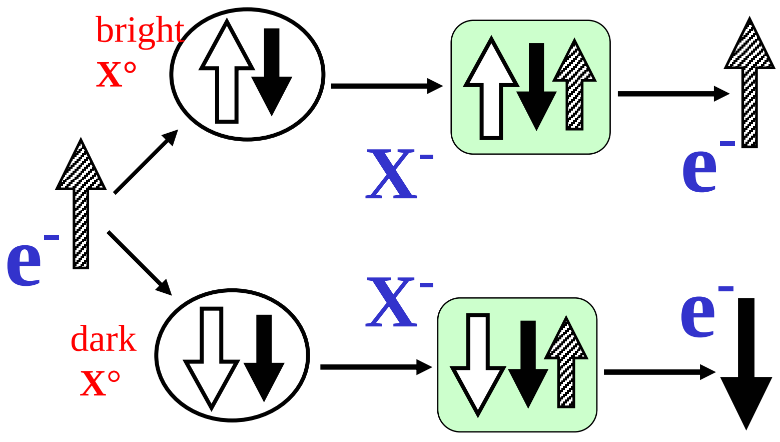
where $Wn_{\uparrow}$ is the rate of exciton capture on charged quantum dots

The concentrations of spin ↑ and spin ↓ electrons ($n_{\uparrow}$ and $n_{\downarrow}$) depends on the pumping intensity. At low intensity:

$$n_{\uparrow} = n_{\downarrow} - G_d^{\Downarrow}\tau_{SE}$$

where τ_{SE} is the electron spin relaxation time

Optical Pumping of Electron Spin



Bright exciton leaves electron unchanged

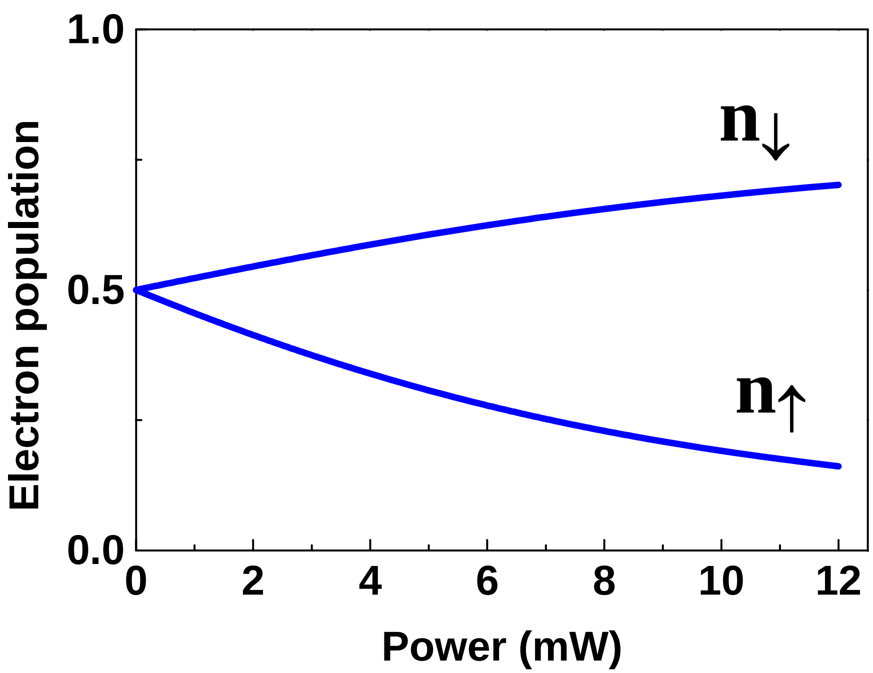
Dark exciton leaves behind electron with spin down
Optical pumping

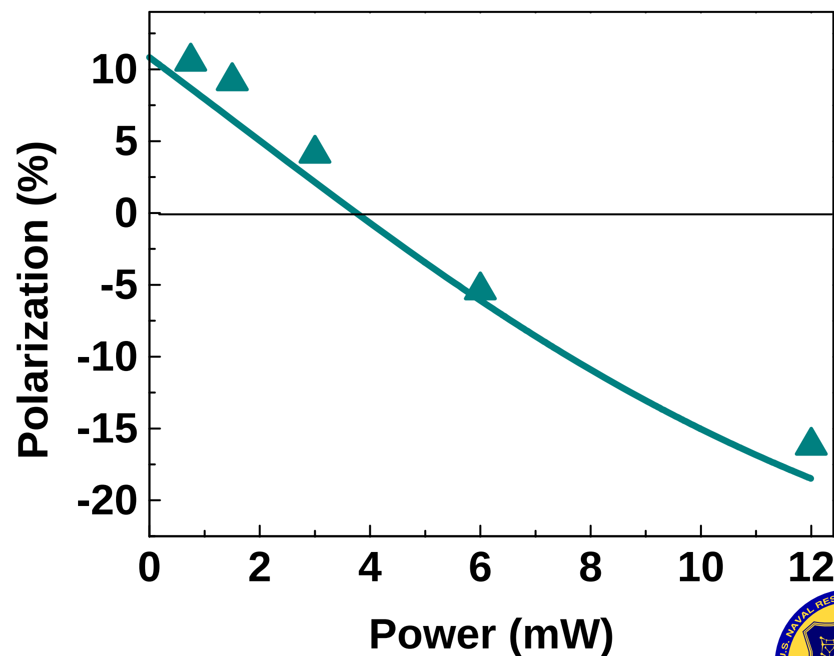
Pumping loop:

excitons capture rate ~ $n_\uparrow$

DE recombination decreases $n_\uparrow$

leading to DE accumulation

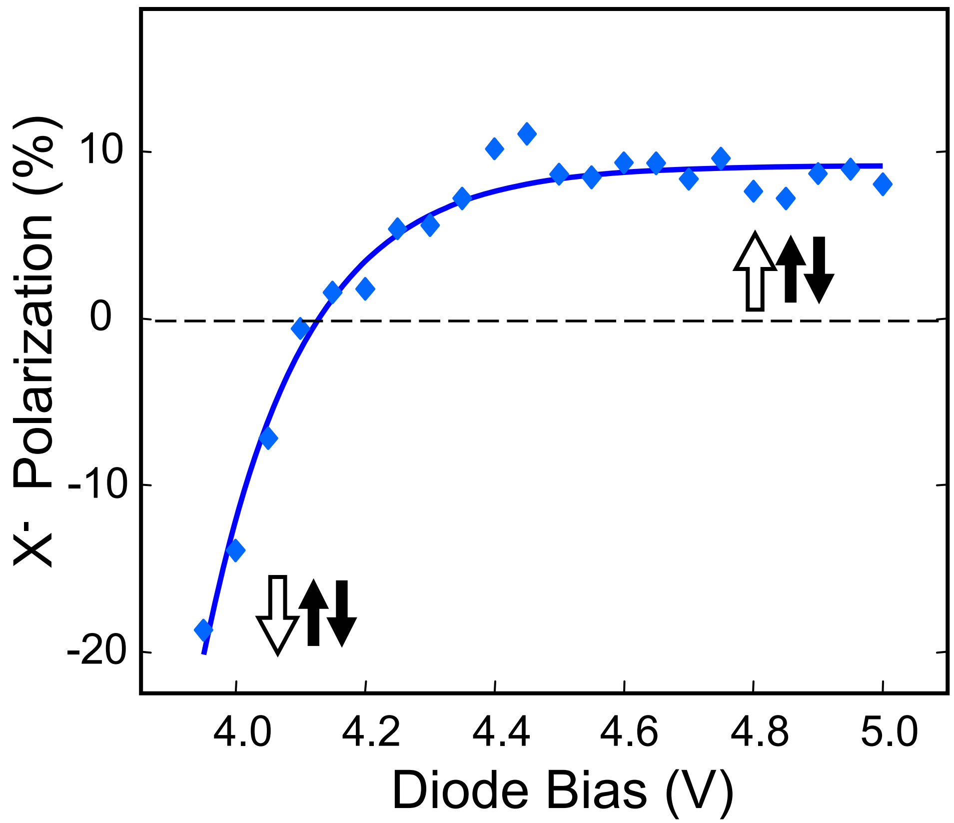




X^- polarization: bias dependence

Electrons in QW: few → many

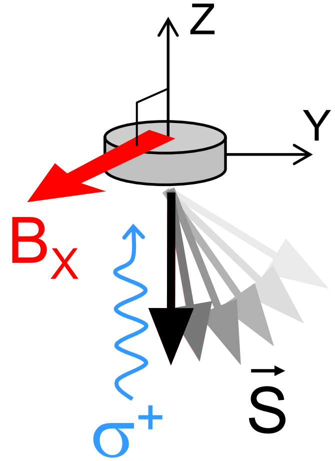
- many excitons per charged QD
- pumping of electron spins
- accumulation of Dark Excitons
- negative PL polarization



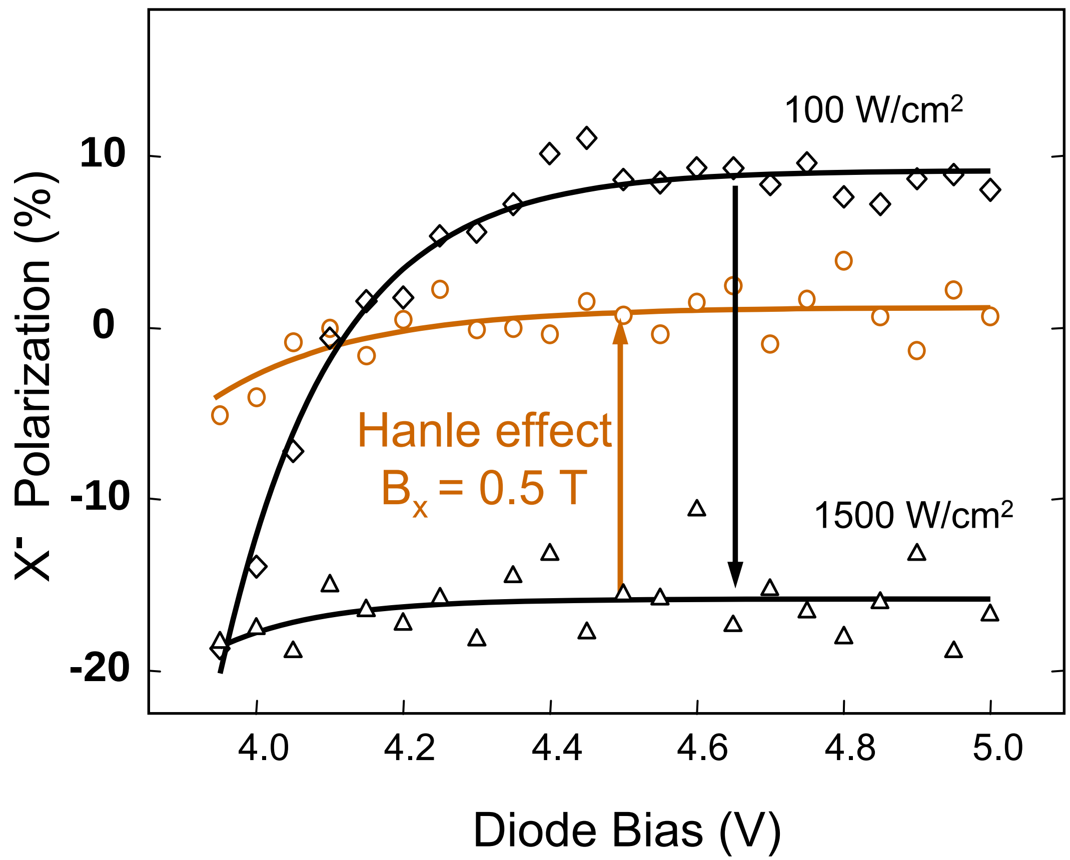
- few excitons per charged QD
- no pumping of electron spins
- no accumulation of Dark Excitons
- positive PL polarization

Erase Electron Spin Polarization

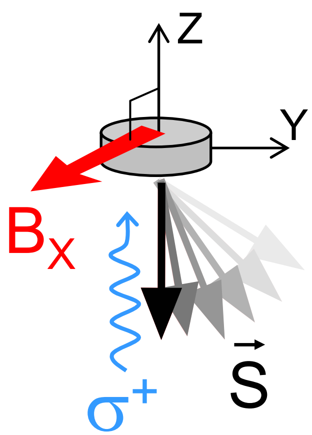
Hanle effect: Erase spin polarization through precession in magnetic field



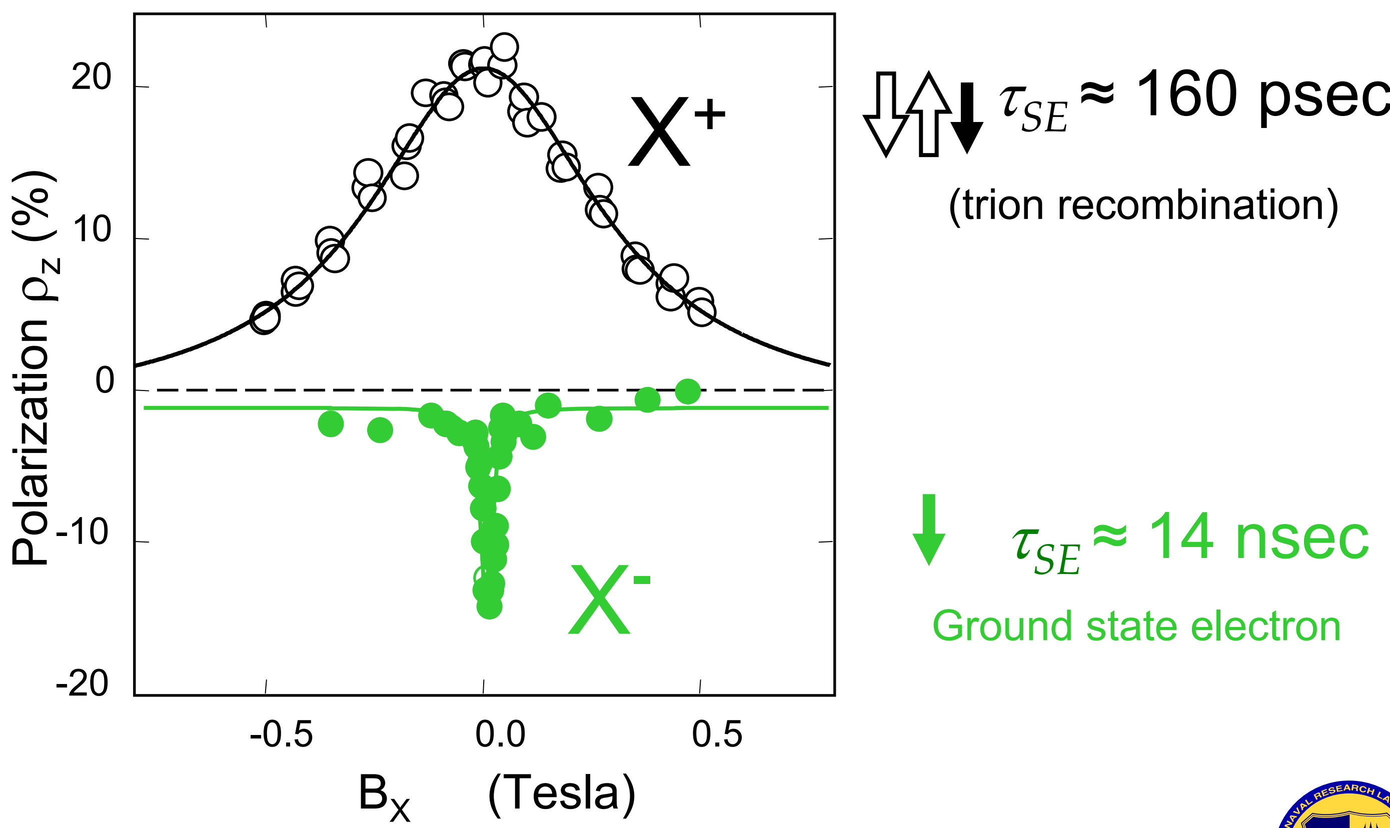
(Holes are not affected by B_x)



Hanle effect: electron spin lifetimes

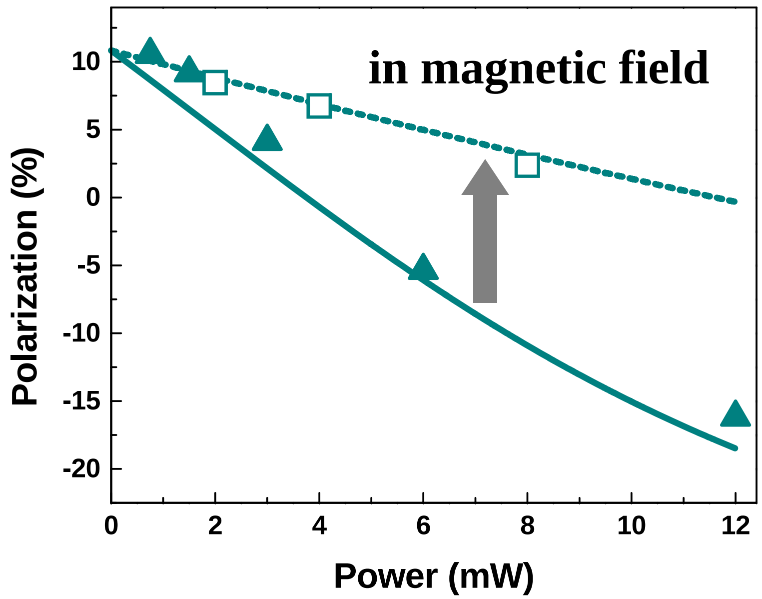


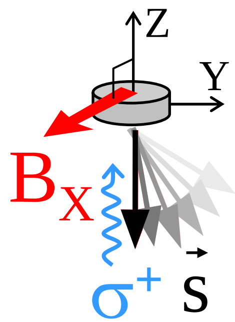
- Depolarize electrons but not holes with magnetic field in the QD plane
- Smaller depolarization field implies longer electron spin lifetime



Theory of Erasing Electron Spin Polarization

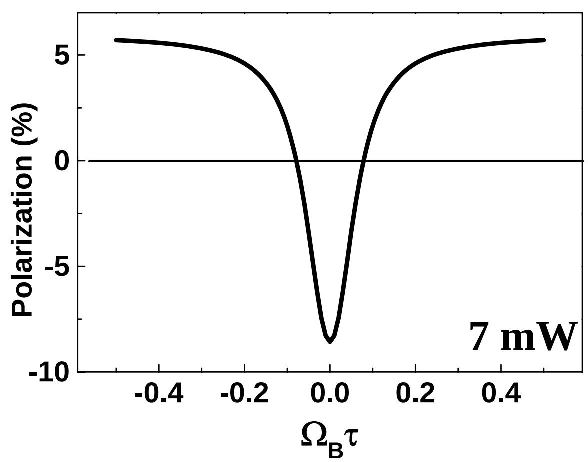
X^- - polarization measurement



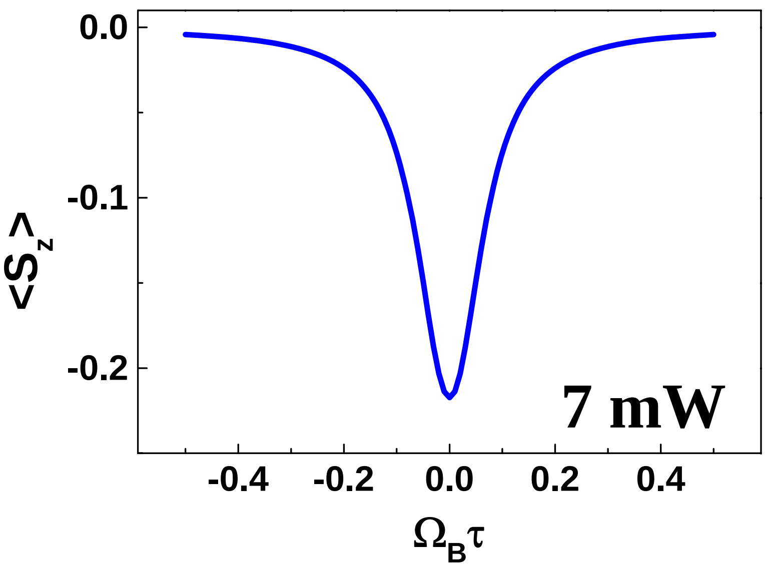


$$\Omega_B = g_e \mu B_x$$

Hanle effect:



Average electron spin:



45% polarization

Summary

– Optical orientation is observed in a single, charge-tunable QD.

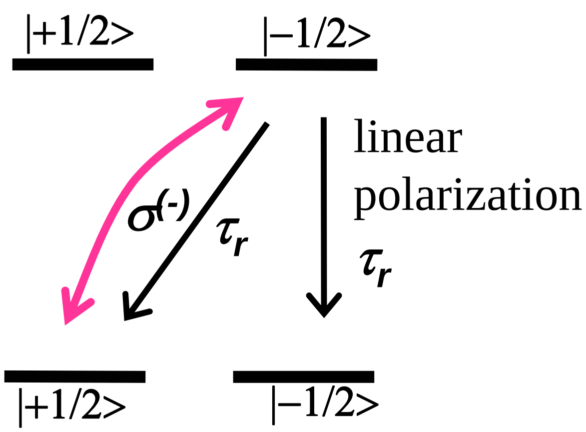
– Theory explains the negative optical polarization in negatively charged QDs resulting from the electron spin pumping, which leads to accumulation of dark excitons.

– Theory quantitatively describes amplitude and sign of Hanle effect, as well as the polarization power dependence in negatively charged QDs.

– However the non-resonant optical pumping is not the best way of the electron spin initialization.

Resonant Optical Spin Orientation: Atoms vs. QD's

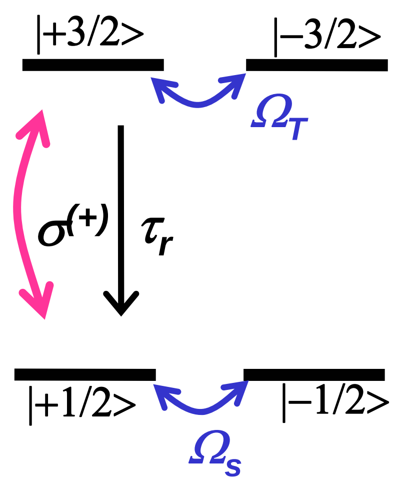
In atoms



(Brossel and Kastler, Comp. Rend. **229,** 1213 (1949).)

Selection rules allow luminescence in both spin states. It leads to accumulation of S_z = -1/2.

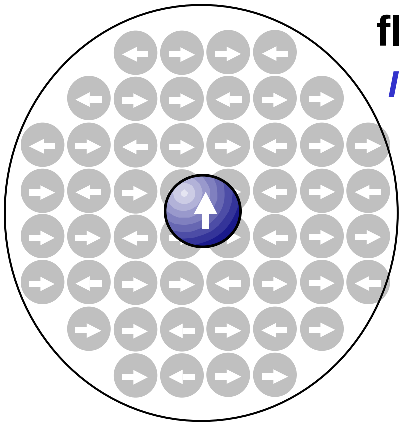
In "artificial" atoms (quantum dots)



Luminescence returns electron to the same initial spin state. Optical initialization is not possible!!

Electron spin can change due to either electron or hole spin relaxation.

Spin flip transitions

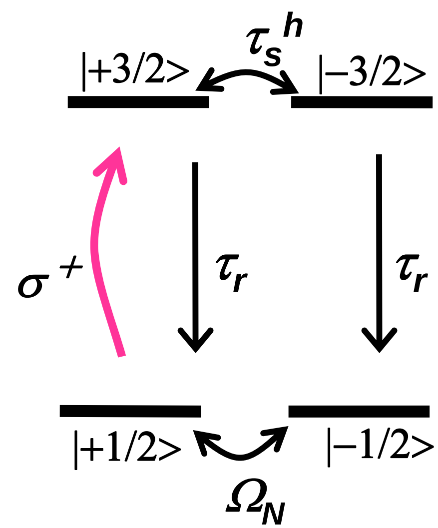


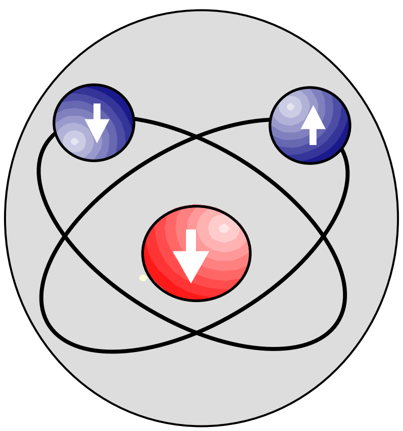
fluctuation of nuclear spin directions in a finite size QD

I.A. Merkulov, Al. L. Efros, and M. Rosen Phys. Rev. B 65, 205309 (2002).

precession of electron spin in effective hyperfine B-field of nuclei

$1/\Omega_N \sim$ 1-10 ns





Net electron spin of trion = 0
Hole is not affected by nuclei.

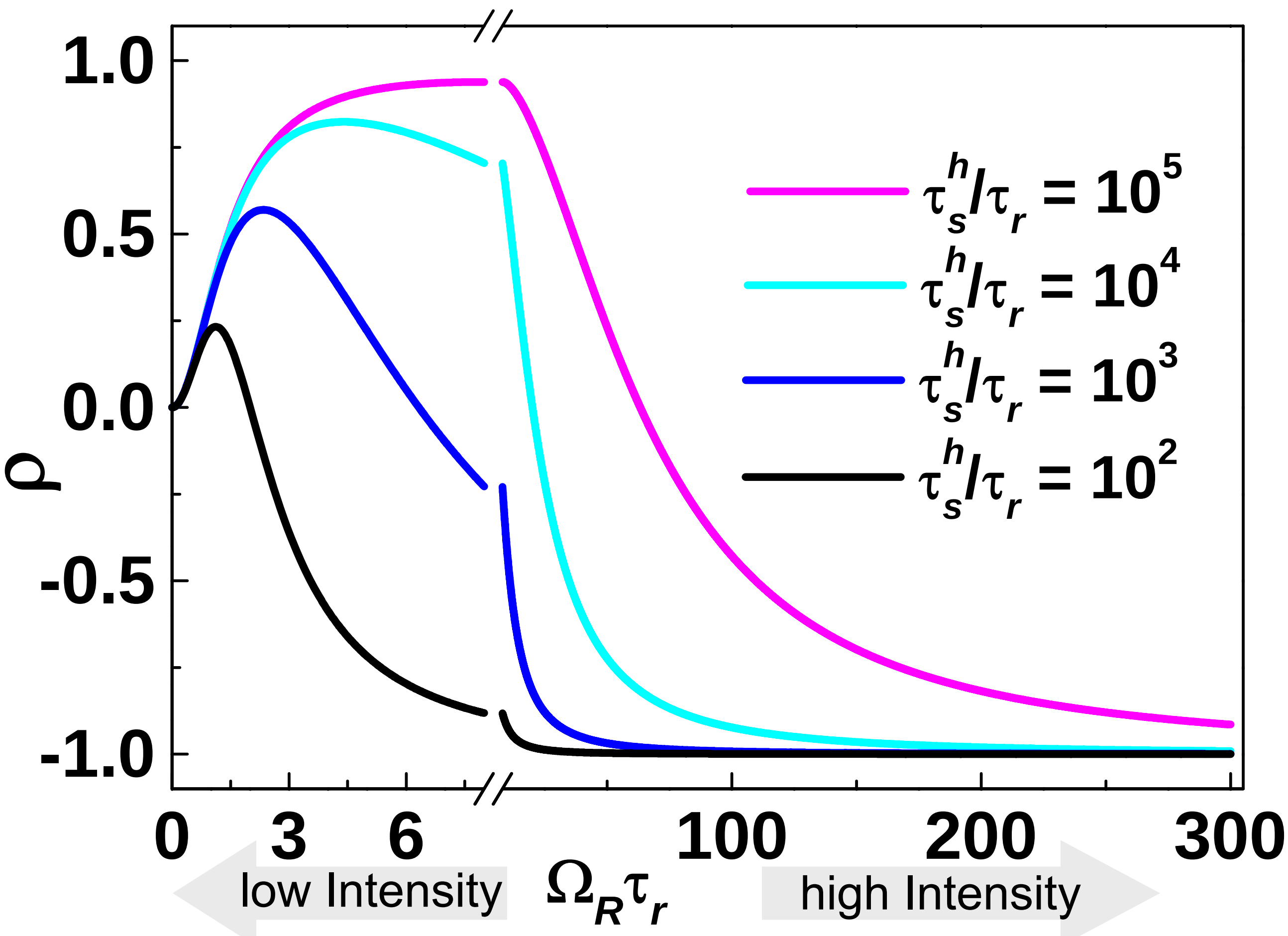
Two-phonon process flips the spin of hole.
It requires phonons: $1/\tau_s^h \rightarrow 0$ at low temperature.

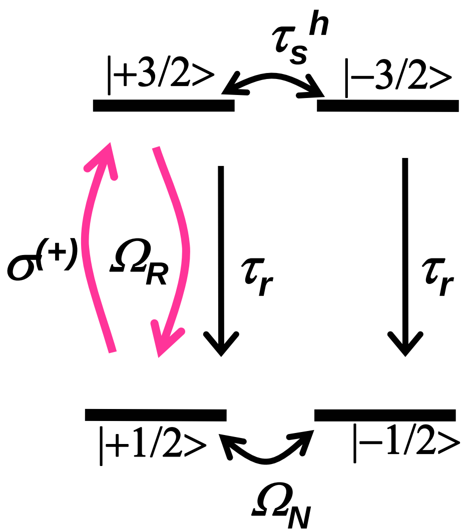
T. Takagahara Phys. Rev. B 62, 16840 (2000).

Spin initialization by polarized light

Steady state electron spin polarization at t → ∞ is independent of initial spin.

$$\rho = (P_{1/2} - P_{-1/2}) / (P_{1/2} + P_{-1/2})$$





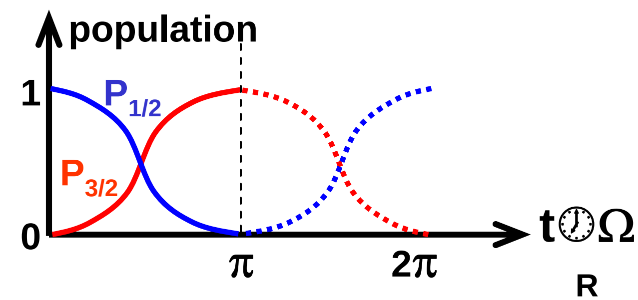
$\Omega_N\, \tau_r = 0.1$

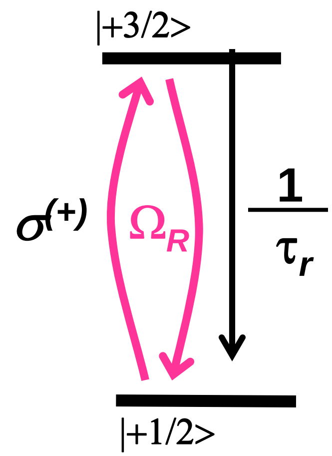
$\Omega_R \sim (Intensity)^{1/2}$

Effect of σ^+ Polarized Optical π- Pulses

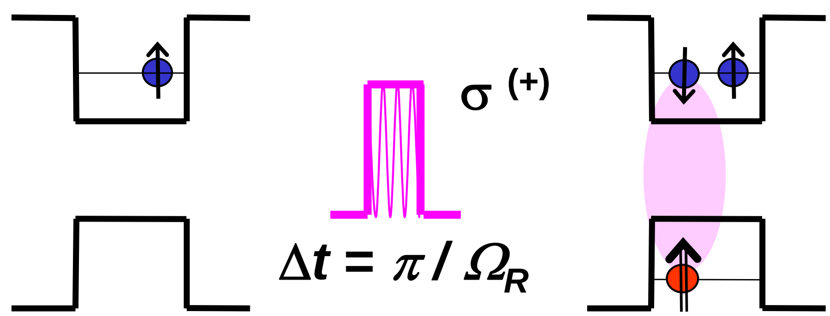
The intense ♦$^{(+)}$ polarized light drives the +1/2 electron into the +3/2 trion states and back with Rabi frequency: $\Omega_R = 2\,(E \cdot d/\hbar)$

Electron $P_{1/2}$ and trion $P_{3/2}$ population:



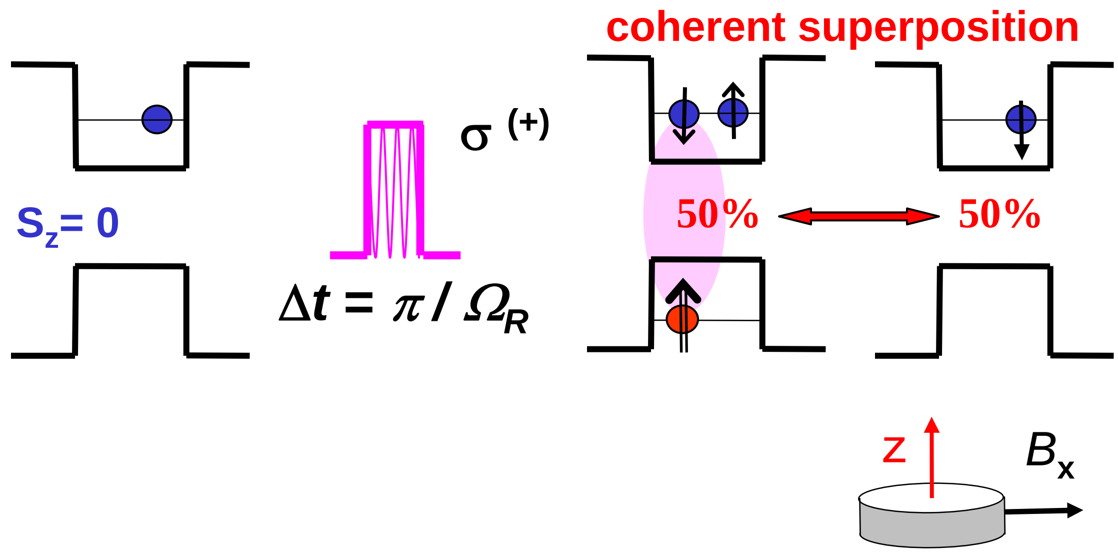


Optical ❐-pulse of ♦$^{(+)}$ polarization creates the +3/2 trion from the 100% polarized S_z= +1/2 electron, but it does not affect the 100% polarized S_z= -1/2 electron.

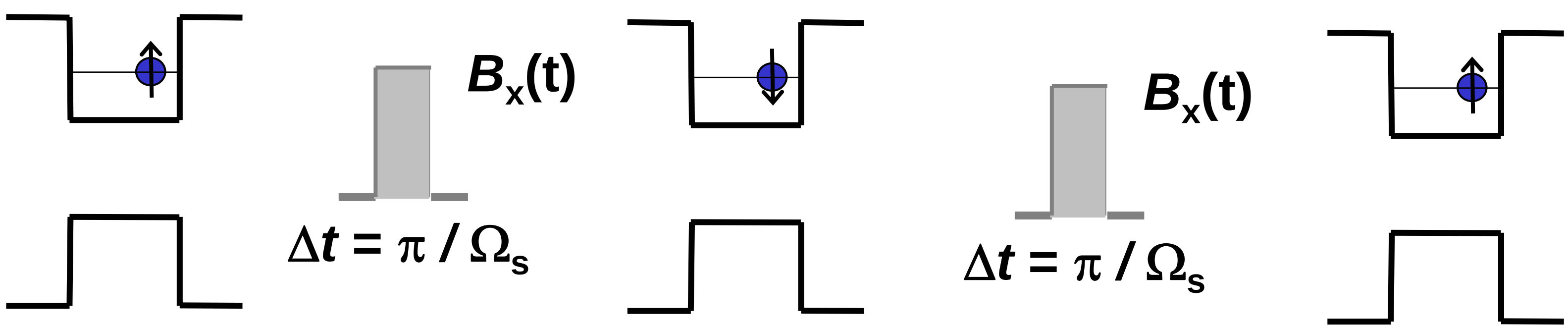


Effect of π- pulses of optical and magnetic field

If an electron is unpolarized S_z= 0 optical π-pulse of $\sigma^{(+)}$ polarization creates a coherent superposition of the +3/2 trion and the optically passive S_z= -1/2 electron.

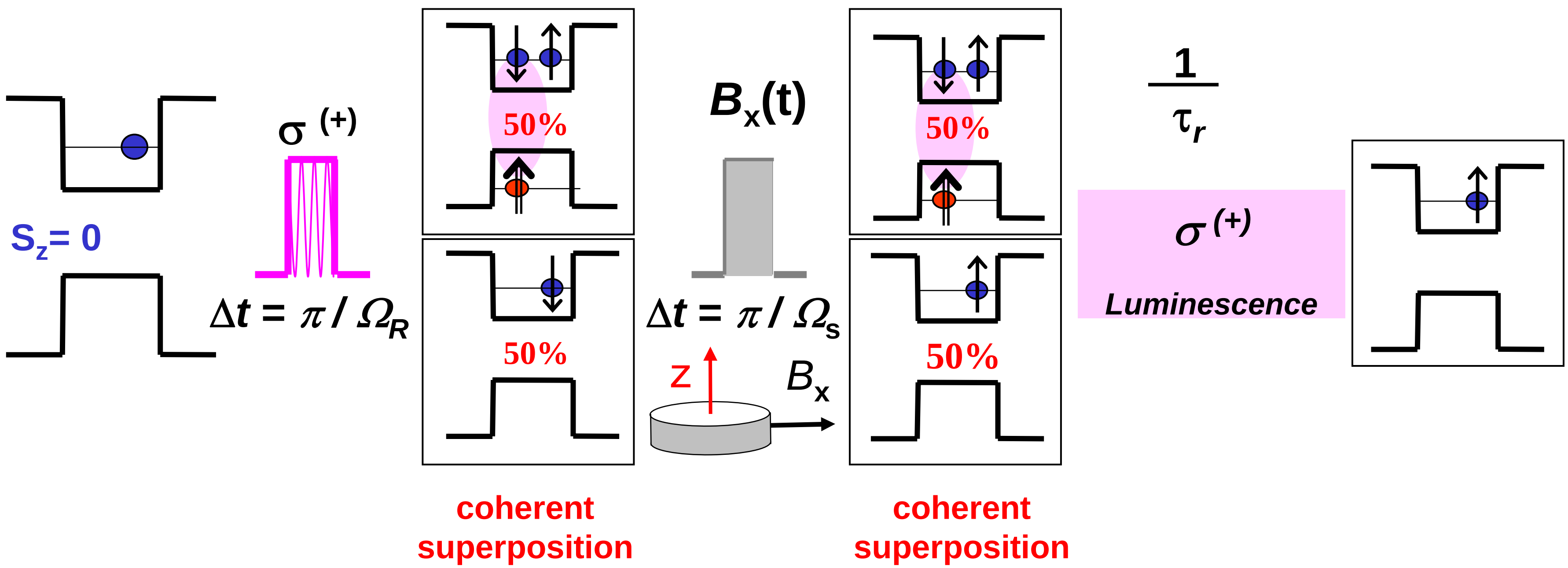


Transverse magnetic field rotates electron spin: $\Omega_s = \mu_B g_e B_x / \hbar$



Initialization by optical and magnetic □ -pulses

Short magnetic □-pulse flips the electron spin but it does not affect the trion.



100% electron spin polarization can be achieved!

<u>Problem</u>: It is difficult to generate short magnetic pulses ($\tau_r >> \Delta t$) .

Summary

•Electron spin in QDs can be optically initialized by intense polarized light.

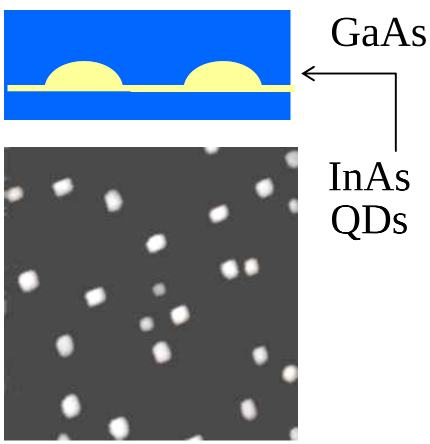
•We propose optical initialization of an electron spin that using a combination of optical and transverse magnetic field π-pulses.

Controlled Initialization of Spin Coherence in Ensemble of Singly charged QDs

…. M. Bayer, Dortmund

resonant optical excitation of charged QDs

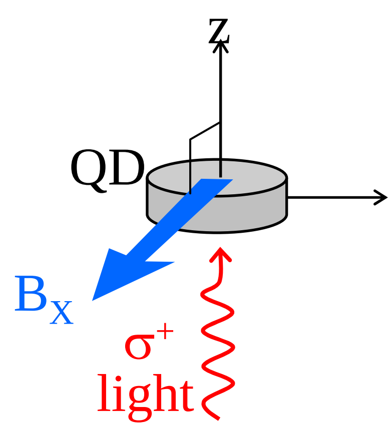
Pump-probe Faraday and Kerr rotation measurements*



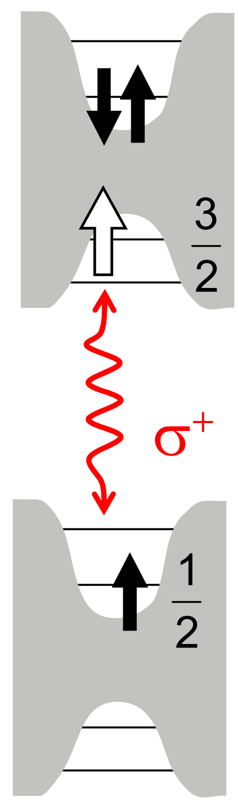
self-assembled QDs

Samples: 20 layers of QDs separated by 60 nm wide barriers,

QD density ~ 10^{10} cm^{-2}, *n* -modulation doped 20nm below each layer with the same Si density.



Charged exciton or "trion"

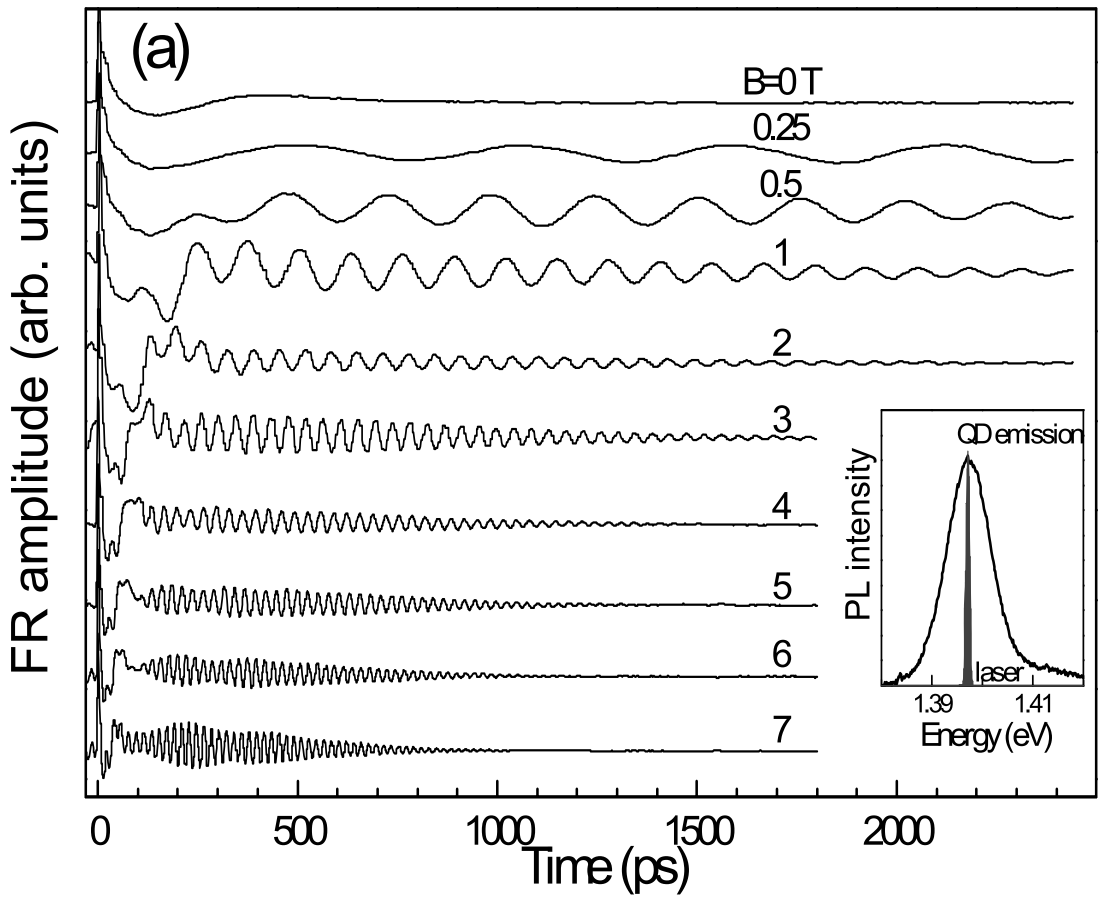


Ground state electron

“Semiconductor Spintronics and Qauntum Computation” Eds. D.Awschalom, D. Loss…(2002), J. Kikkawa & D. Awschalom, Science **287**, 473 (2000), J. Gupta & D. Awschalom PRB **59**, 10421 (1999)….

Pump-Probe Faraday Rotation in (In,Ga)As/GaAs QDs

Traces of FR signal, excited at *T*= 2K, pulses duration of 1ps resonant excitation.

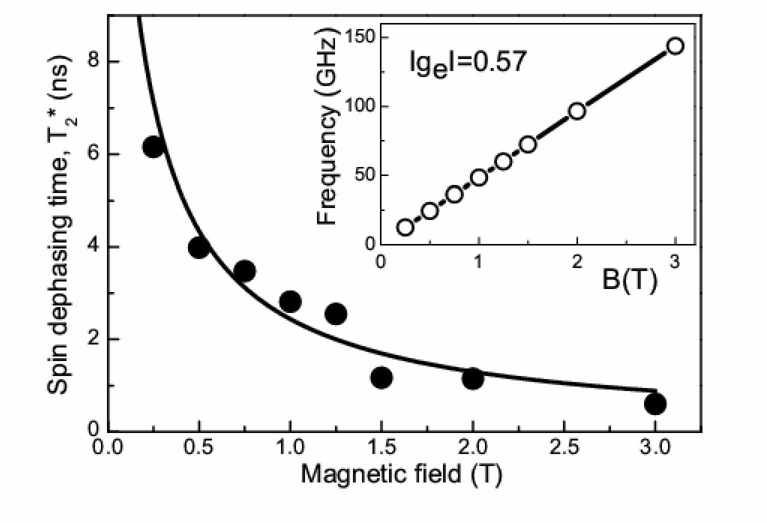


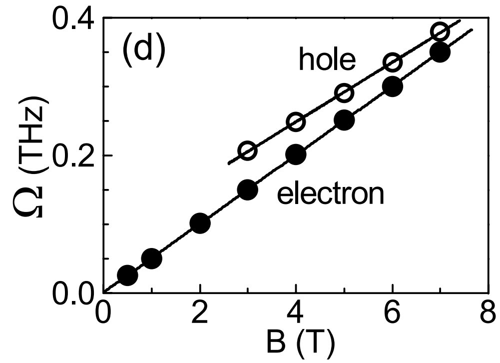
1. Pronounced oscillations
2. Duration longer than τ_r= 400 ps
3. Oscillation frequencies increases with magnetic field, *B*
4. Decay increases with *B*
5. Additional modulations at high *B*

Long-Lived Electron Spin Coherent State

Increase of frequencies $\hbar\Omega_e = g_e \mu_B B$

$A_{FR} \sim \exp(-t/T_2^*)\cos(\Omega_e t)$





Additional modulations: $\hbar\Omega_h = g_h \mu_B B$

$|g_h|$=0.66 at B= 5 T with

T_2^*=170 ps vs τ_r= 400 ps

g-factor dispersion Δg_e

$$\frac{1}{T_2^*(B)} = \frac{1}{T_2^*(0)} + \frac{\Delta g_e \mu_B B}{\sqrt{2}\hbar}$$

best fit: Δg_e=0.004

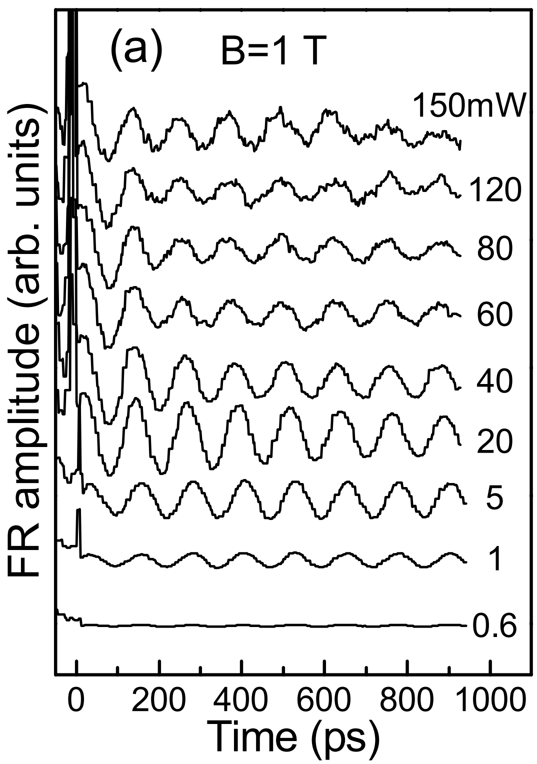
The optical intialization of **LL-ESC** was observed in GaAs/AlGaAs interface QDs: Gurudev Dutt et al. PRL **94**, 227403 (05)

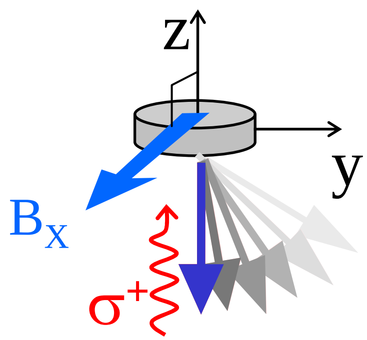
Theory of this effect: S. E. Economou et al., Phys. Rev. B **71**, 195327 (2005)

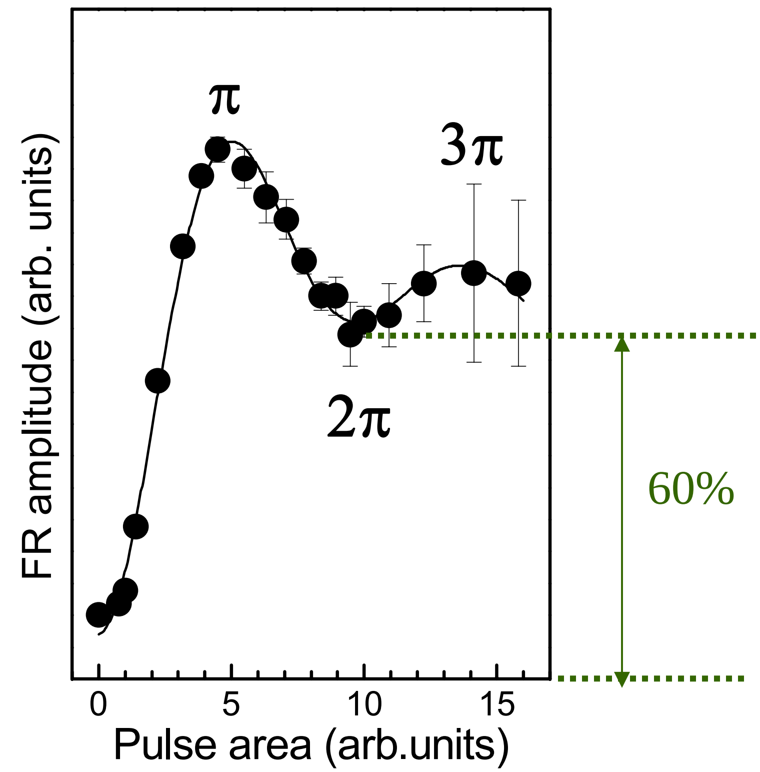
Pump Power Dependence of FR Amplitude

Nonmonotonic dependence

Pulse area: $\Theta = (2/\hbar)\int(\vec{d} \bullet \vec{E}(t))dt$







Rabi-like oscillations of the FR amplitude:

⇓ origin of the spin coherence

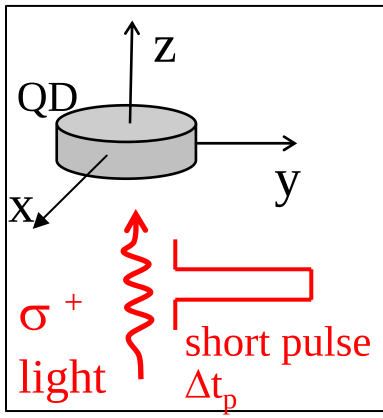
Coherent Spin Superposition

A short pulse of resonant circularly polarized light with external transverse magnetic field creates a CS of the electron and trion states. If pulse length $\Delta t \ll \tau_r, \tau_s^h$, and τ_s^e :

coherent spin state

$\alpha^0\uparrow + \beta^0\downarrow$

electron ground state



electron/trion coherent superposition

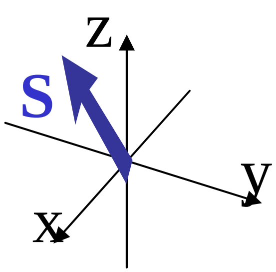
trion state

$\alpha^0\cos(\Theta/2)\uparrow + \beta^0\downarrow - i\,\alpha^0 \sin(\Theta/2)\uparrow\downarrow\Uparrow$

electron ground state

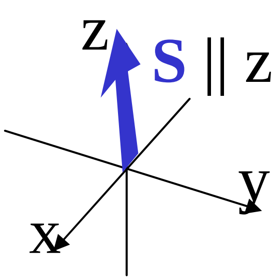
pulse area $\Theta = \left(2\vec{d} \bullet \vec{E} / \hbar\right)\Delta t_p$

Spin polarization vector



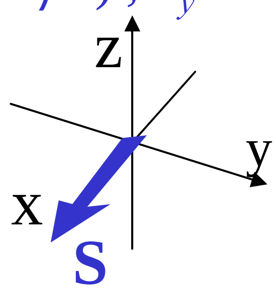
Arbitrary spin orientation: $|\psi\rangle = \alpha|\uparrow\rangle + \beta|\downarrow\rangle$, where $|\alpha|^2 + |\beta|^2 = 1$ in the ground state, and $|\alpha|^2 + |\beta|^2 < 1$ in the excited state.

spin vector: $S_x = \mathrm{Re}(\alpha\,\beta^*)$, $S_y = -\mathrm{Im}(\alpha\,\beta^*)$, $S_z = (|\alpha|^2 - |\beta|^2)/2$



$|\alpha| = 1, \;\; \beta = 0$,

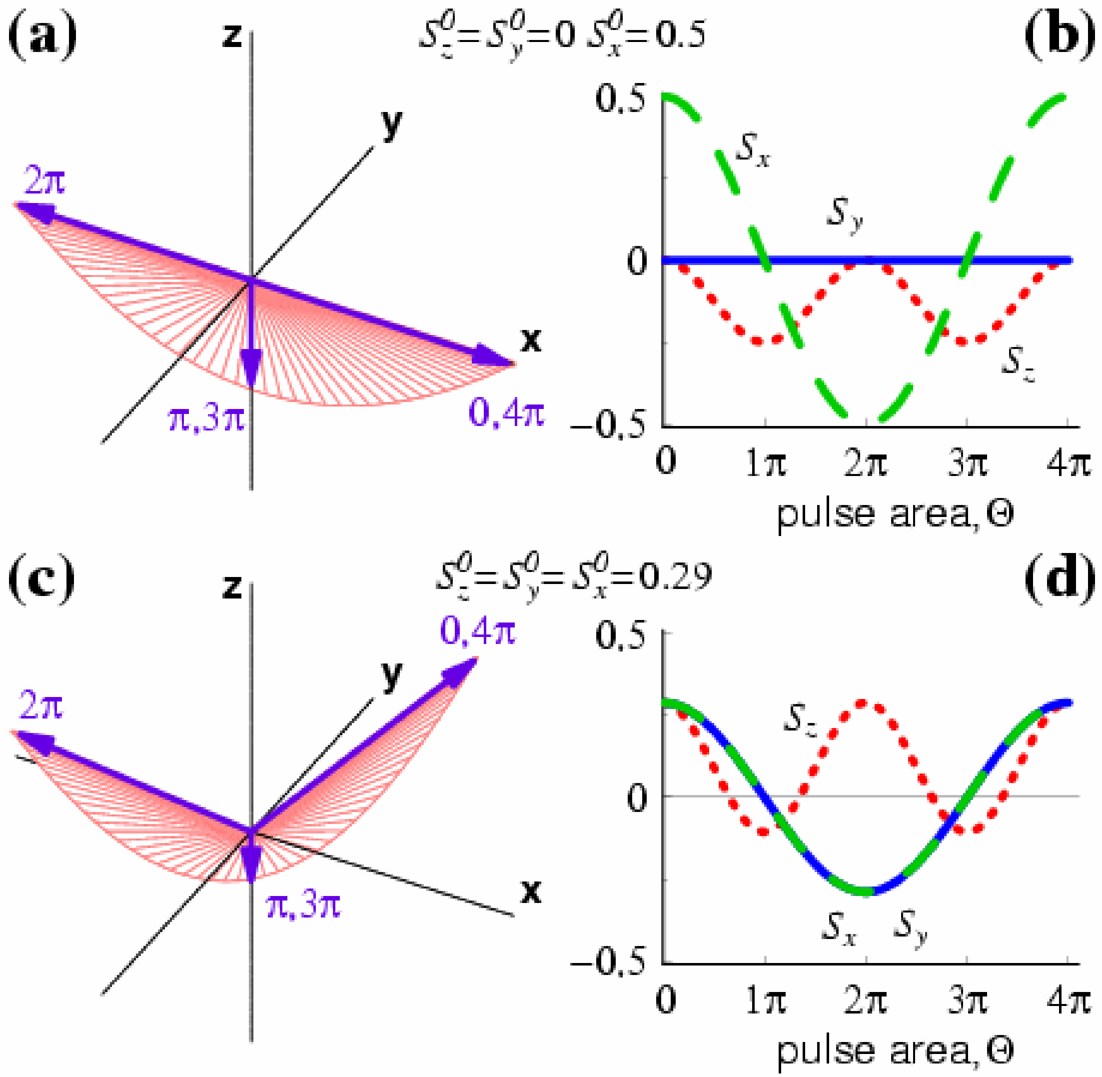
$S_x = S_y = 0, S_z = 1/2$



$\alpha = \beta = 1/2^{1/2}$,

$S_x = 1/2, S_y = S_z = 0$

Optical Control of Electron Spin



σ^+ polarized pulse decreases S_z:

$|S_z - S^0_z| = 0.5|\alpha^0|^2 \sin^2(\Theta/2)$

and it reaches maximum at $\Theta=(2n+1)\pi$

S_x and S_y component change sign with period 2π, $\Theta=2n\pi$-pulses can be used

Spin Dynamics After Pulse

After the pulse the electron and trion spin vectors in a single QD:

$$\frac{d\boldsymbol{J}}{dt} = [\boldsymbol{\Omega}_h \times \boldsymbol{J}] - \frac{\boldsymbol{J}}{\tau_s^h} - \frac{\boldsymbol{J}}{\tau_r},$$

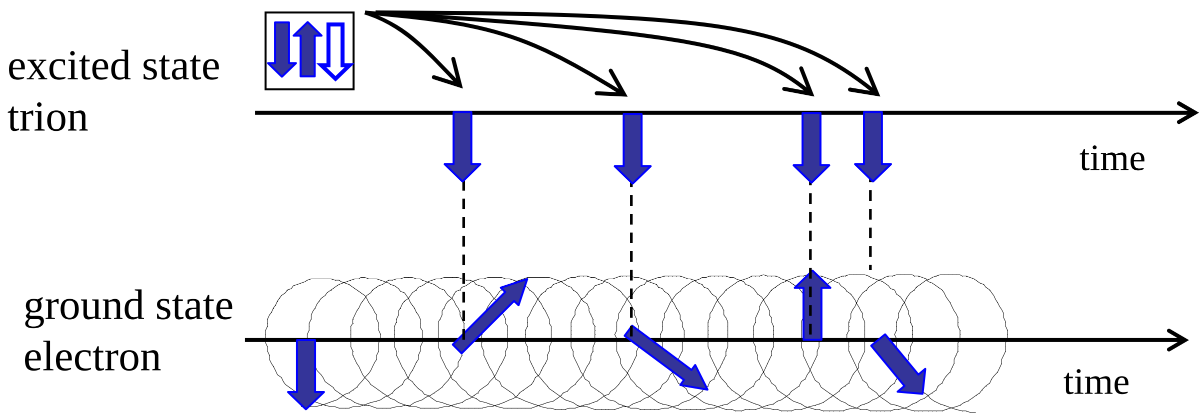
$$\frac{d\boldsymbol{S}}{dt} = [(\boldsymbol{\Omega}_e + \boldsymbol{\Omega}_N) \times \boldsymbol{S}] + \frac{(\hat{\boldsymbol{J}}\hat{\boldsymbol{z}})\,\hat{\boldsymbol{z}}}{\tau_r},$$

where $\boldsymbol{\Omega}_{e,h} \parallel \boldsymbol{e}_x$ and $\boldsymbol{\Omega}_N = g_e \mu_B \boldsymbol{B}_N / \hbar$

Trion spin vector: $\boldsymbol{J} = (J_x, J_y, J_z)$, describes polarization of the trion state: $|\psi_{tr}$ ⇧$=\alpha_{tr}|$✸✵⬆⇧$+\beta_{tr}$ |✸✵←⇧

$J_x=\mathrm{Re}(\alpha_{tr}\,\beta^*_{tr})$, $J_y=-\mathrm{Im}(\alpha_{tr}\,\beta^*_{tr})$, $J_z=-(1/2)(|\alpha_{tr}|^2-|\beta_{tr}|^2)$.

The long lived electron spin polarization at $t >> \tau_r$: $S_z(t) = S_{z\infty} \cos[(\Omega_e + \Omega_{N,x})\, t]$,



If $\Omega_e >> 1/\tau_r$, $S_{z\infty} = S_z(0)$ is the electron polarization created by the pulse ONLY

Faraday Rotation Amplitude in a QD Ensemble

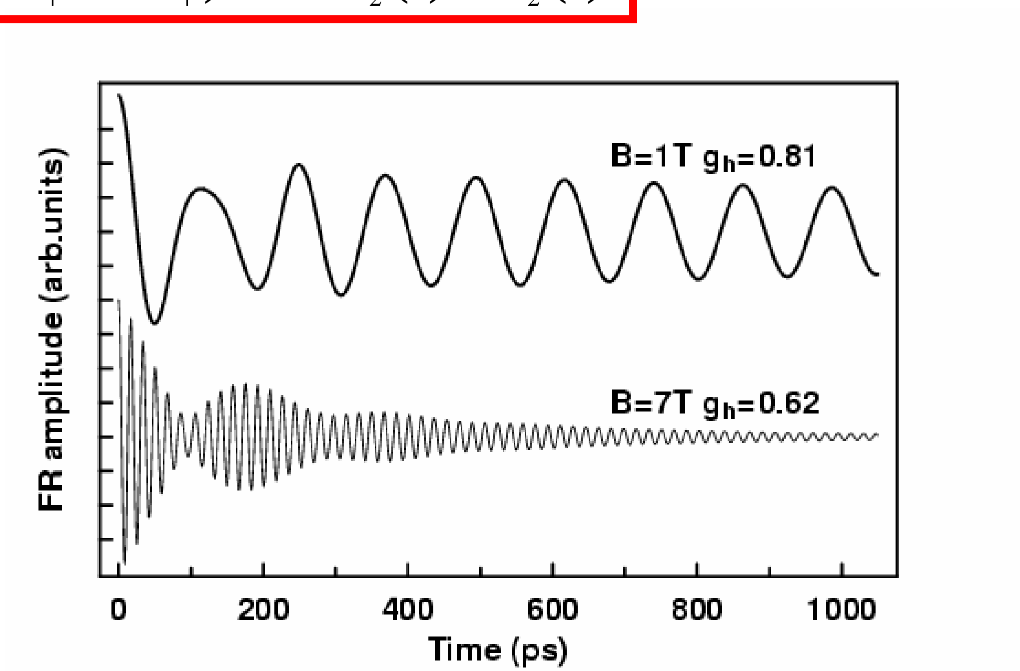
In a QD ensemble ***J****(t)* and ***S****(t)* should be averaged over Δg_e=0.004

Optically induced FR amplitude:

Proportional to the population difference of states involved in σ^+ and σ^- transitions: $\Delta n_+ = n_\downarrow - n_\Uparrow$ and $\Delta n_- = n_\uparrow - n_\Downarrow$. The FR angle:

$$\phi(t) \sim (\Delta n_+ - \Delta n_+)/2 = S_z(t) - J_z(t).$$

Theoretical time dependence of FR signal for ensemble of QDs, which use parameters from experiment.



Summary

1. We have shown experimentally and theoretically that short pulses of circularly polarized light with transverse magnetic field allow initialization a complete control of electron spin coherence in a single quantum dot.

2. For resonant excitation, the pulse area uniquely determines the electron spin coherence.

- The spontaneous decay of the trion does not affect the spin coherence at $\Omega_e \tau_r >> 1$.

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