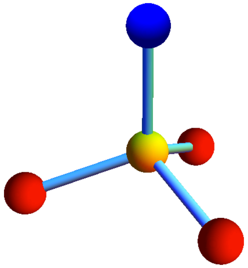

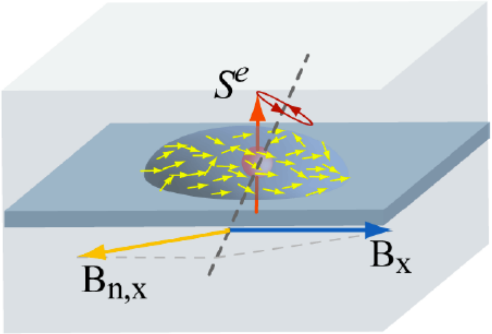

Optical pumping of carrier and nuclear spins in quantum dots

Bernhard Urbaszek

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INSA TOULOUSE

Université de Toulouse

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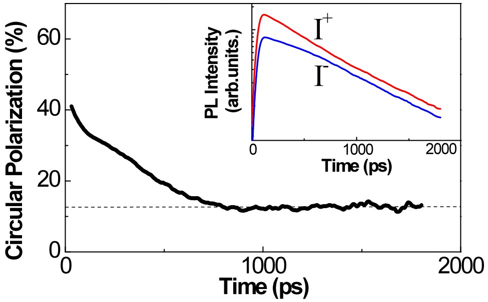
cnrs advancing the frontiers

ILNACS (IAL CNRS-IOFFE RAS) ITN SpinOptronics ANR QUAMOS

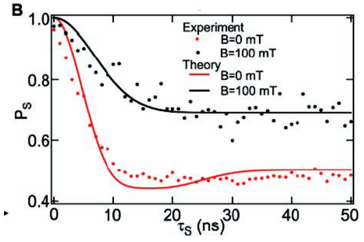
Nuclear Spin effects in Quantum Dots

1. Limitation for electron spin coherence

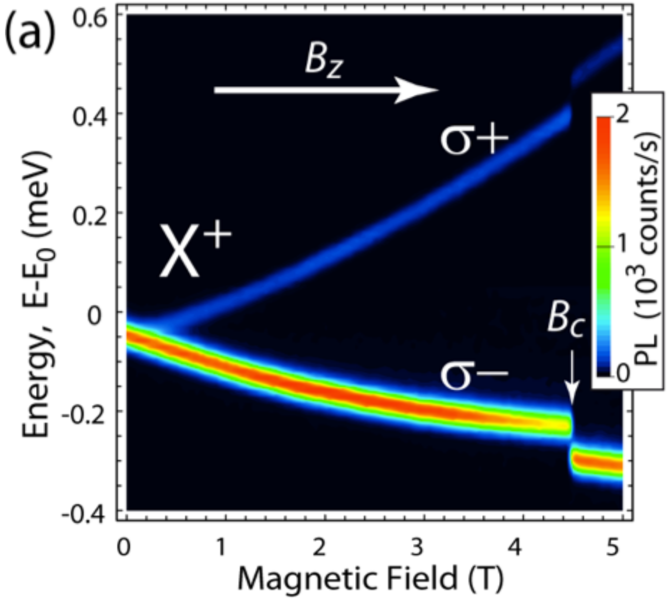
Braun PRL 2005



Petta Science 2005

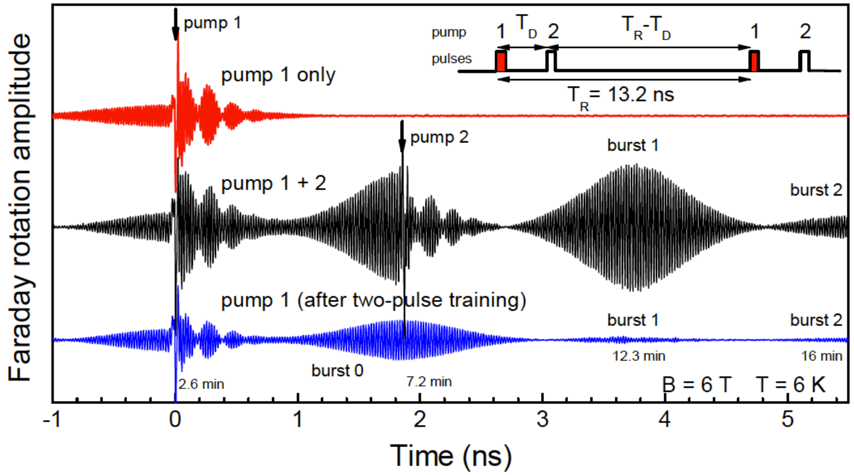


arXiv1202.4637



2. Internal Magnetic fields of several Tesla

Greilich Science 2007



3. Memory effects: Nuclear Spin coherence

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Richard Warburton, Basel University

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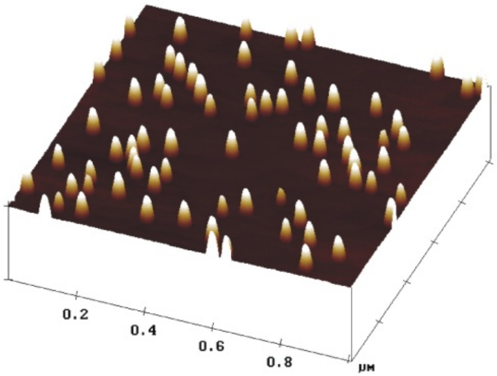
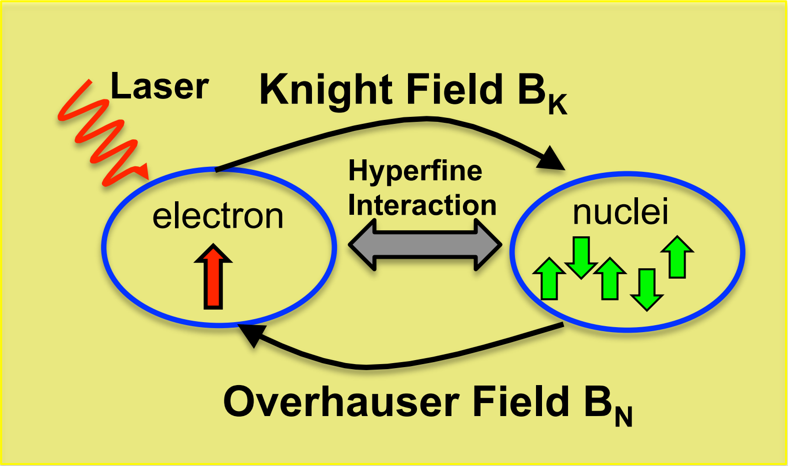
Ioffe
Physico-
Technical
Institute

Degrees of Freedom in Semiconductors: *Optical Manipulation*

Main Part :

Optical pumping of **carrier spins** and **nuclear spins** in quantum dots





Laser

Knight Field B_K

Hyperfine Interaction

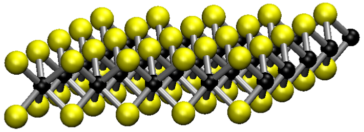
electron

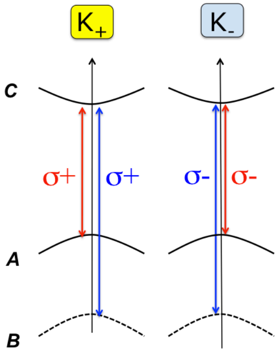
nuclei

Overhauser Field B_N

Outlook:

selective **K-valley** excitation in MoS_2 monolayers





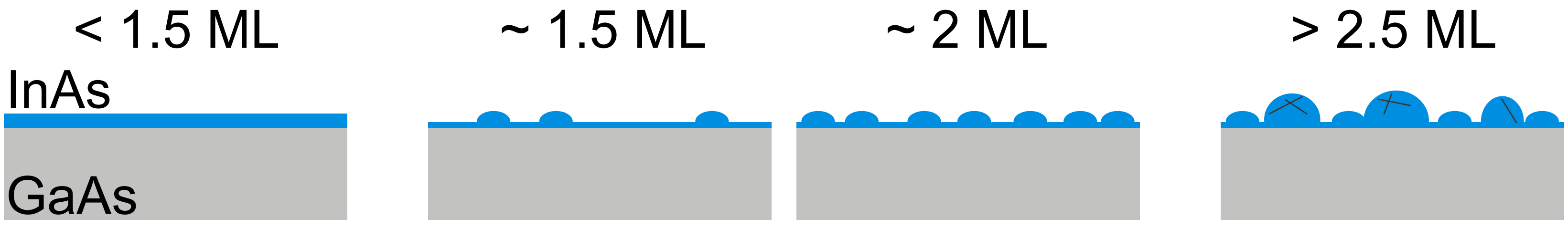
Outline:

- Introduction to semiconductor quantum dots
- Hyperfine interaction between nuclear spins ⬄ carrier spins
- Carrier spin dephasing due to fluctuations of the Nuclear Field
- Optical Pumping of Nuclear Spins
- Nuclear Spins Physics in quantum dots: What’s new ?

quantum dot formation through self-assembly:

band gap: E_G(GaAs) > E_G(InAs)

lattice constants: a_0(InAs) > a_0(GaAs)



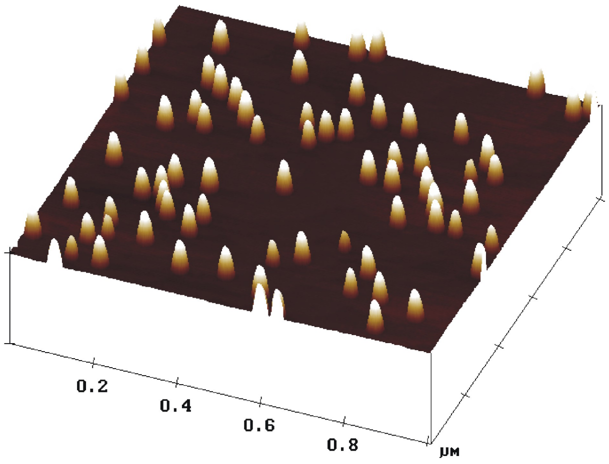
2D-growth

stressed dots and wetting layer

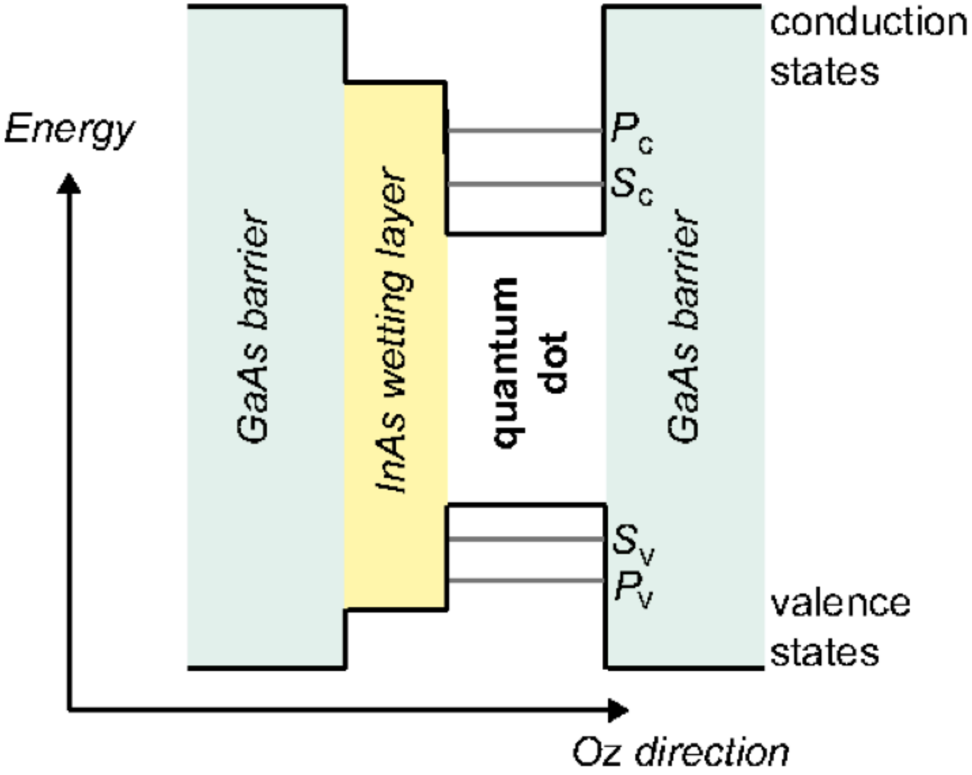
dislocations

dot height ~ 6 nm

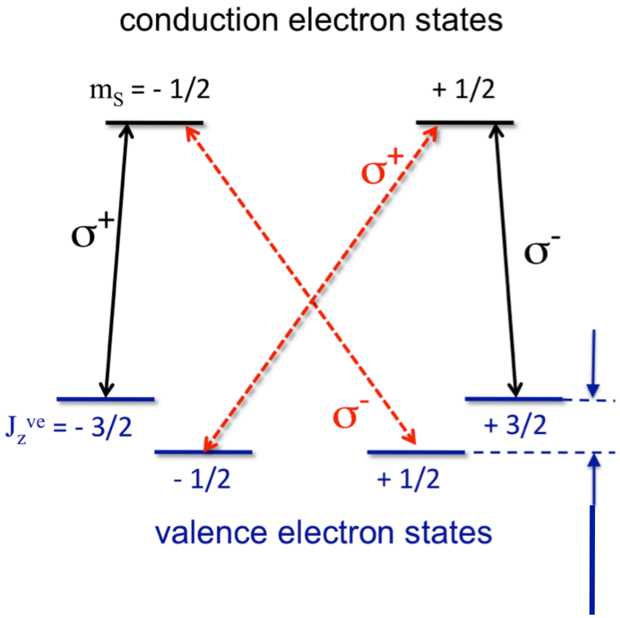
diameter ~ 20 nm



Quantum Dots: *discrete energy states*



Optical Selection rules:
High fidelity Spin preparation
for pure Heavy Hole States $J_z=+/- 3/2$

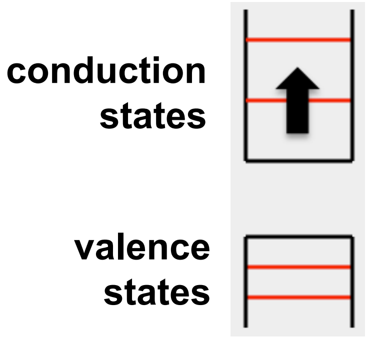


Heavy Hole states
Separated from
Light Hole states due to:

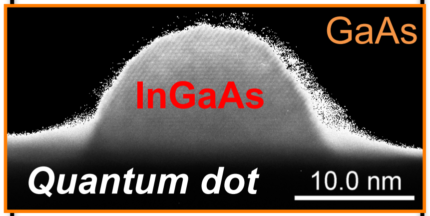
1. *Strain*
2. *Quantum confinement*

Photon Polarization ⇔ 1 Electron Spin ⇔ 10^4 Nuclear Spins

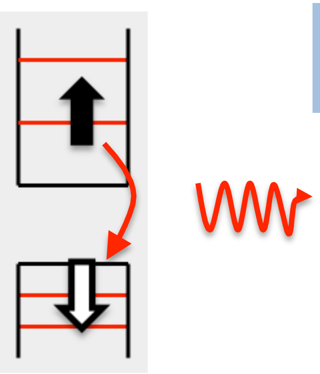
stable Single Spin State



None of the Spin relaxation mechanisms based on movement/collisions applies !



Quantum Emitter



next talk:
J.-M. Gérard

✓ Single Photons
✓ **Polarization** Entangled Photon Pairs

Need: long carrier Spin Coherence times

Limitations for Electron and Hole Spin Coherence time: ***Fluctuating Nuclear Spins***

Target: Optical Control of

1 Electron/Hole Spin

Hyperfine Interaction

10^4 Nuclear Spins in Dot

Outline:

- Introduction to semiconductor quantum dots
- Hyperfine interaction between nuclear spins ⇔ carrier spins
- Carrier spin dephasing due to fluctuations of the Nuclear Field
- Optical Pumping of Nuclear Spins
- Nuclear Spins Physics in quantum dots: What's new ?

Origin of nuclear spin

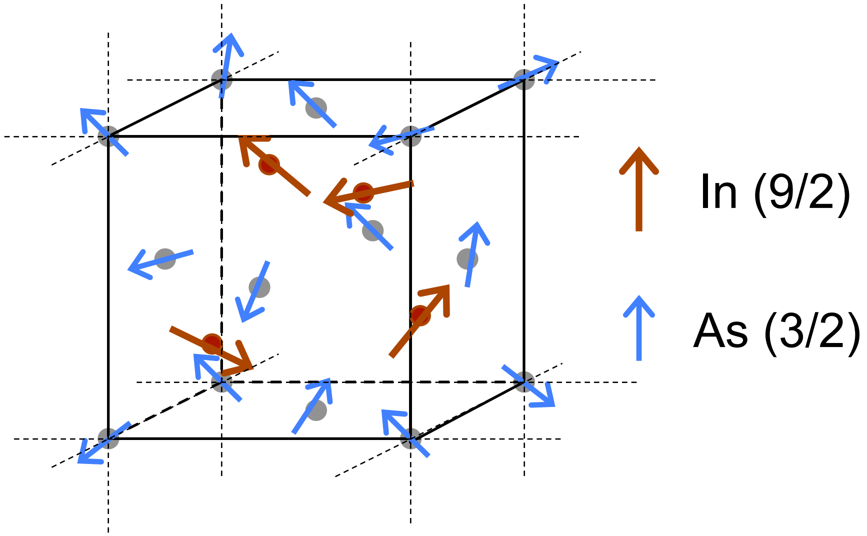
Atom nuclei :

- Collection of protons and neutrons results in a total nuclear spin I

- *number of protons* + *number of neutrons = mass number*

→ *zero, half integer or integer spin I : depends on mass number*

Sublattice of N_L nuclei with spin $\hat{I}^j$



Element abundance	^{27}Al	$^{69(71)}$Ga 60(40)%	^{75}As	^{115}In	^{28}Si	^{29}Si 4.7%	^{12}C	^{13}C 1.1%
Z	13	31	33	49	14	14	6	6
Nuclear spin *I*	5/2	3/2	3/2	9/2	0	1/2	0	1/2

Zeeman effect: energy scales for Nuclei & Electrons

Nuclei

$$\boxed{H_{Zn} = -\hat{\boldsymbol{\mu}}_n.\mathbf{B} = -\mu_N g_n \hat{\mathbf{I}}_n.\mathbf{B}}$$

$$\mu_N = \frac{|e|\hbar}{2M_p}$$

Nuclear magneton

$$\boxed{\mu_N \cong 3.1 \text{ neV T}^{-1}}$$

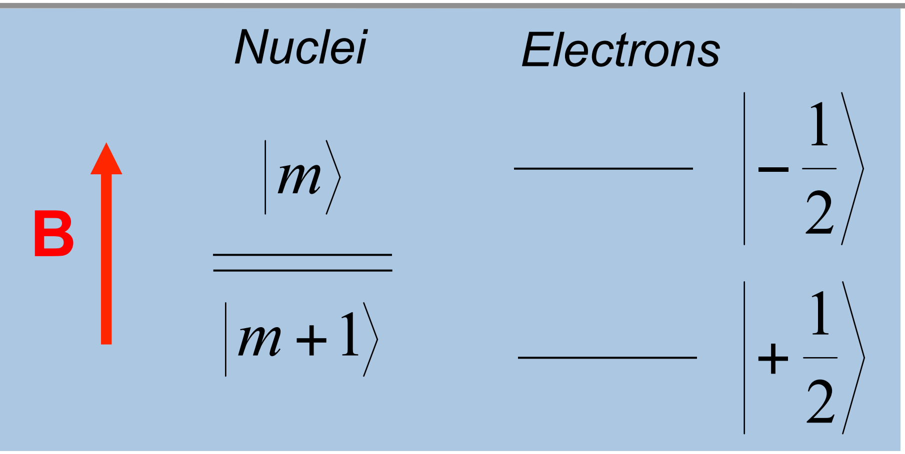
Comparison with electrons :

$$\boxed{H_{Ze} = -\hat{\boldsymbol{\mu}}_e.\mathbf{B} = -\mu_B g_e \hat{\mathbf{S}}.\mathbf{B}}$$

$$\mu_B = \frac{-|e|\hbar}{2m_e}$$

Bohr magneton

$$\boxed{\mu_B \cong 58 \text{ μeV T}^{-1}}$$



$$\boxed{\mu_N << \mu_B}$$

Electron-nuclear magnetic coupling : hyperfine interaction

$$H_{hf} = \underbrace{\frac{\mu_0}{4\pi}\Bigg(\frac{8\pi}{3} g_N \mu_N \; \hat{\mathbf{I}}.\hat{\mathbf{S}} \; \delta(\mathbf{r})}_{\text{Fermi-contact}} + \underbrace{g_N \mu_N \frac{1}{r^3} \hat{\mathbf{I}}.\left[\hat{\mathbf{L}} - \hat{\mathbf{S}} + 3\frac{\mathbf{r}(\hat{\mathbf{S}}.\mathbf{r})}{r^2}\right]\Bigg)}_{\text{Dipolar magnetic}}$$

A. Abragam, Clarendon press, Oxford (1961)

Electron-nuclear magnetic coupling : hyperfine interaction

$$H_{hf} = \underbrace{\frac{\mu_0}{4\pi}\Bigg(\frac{8\pi}{3}\, g_N \mu_N \;\hat{\mathbf{I}}.\hat{\mathbf{S}}\;\delta(\mathbf{r})}_{\text{Fermi-contact}} + \underbrace{g_N \mu_N \,\frac{1}{r^3}\,\hat{\mathbf{I}}.\Bigg[\hat{\mathbf{L}} - \hat{\mathbf{S}} + 3\,\frac{\mathbf{r}(\hat{\mathbf{S}}.\mathbf{r})}{r^2}\Bigg]\Bigg)}_{\text{Dipolar magnetic}}$$

Conduction electrons: “*s*” wave function

$$\longrightarrow \qquad H_{hf}^{c} = \frac{\mu_0}{4\pi}\frac{8\pi}{3}\, g_N \mu_N \;\hat{\mathbf{I}}.\hat{\mathbf{S}}\; \left|\Psi_c(0)\right|^2 = A_e\;\hat{\mathbf{I}}.\hat{\mathbf{S}}$$

A. Abragam, Clarendon press, Oxford (1961)

Electron-nuclear magnetic coupling : hyperfine interaction

$$H_{hf} = \underbrace{\frac{\mu_0}{4\pi}\Bigg(\frac{8\pi}{3} g_N \mu_N \;\hat{\mathbf{I}}.\hat{\mathbf{S}}\;\delta(\mathbf{r})}_{\text{Fermi-contact}} + \underbrace{g_N \mu_N \frac{1}{r^3}\hat{\mathbf{I}}.\left[\hat{\mathbf{L}} - \hat{\mathbf{S}} + 3\frac{\mathbf{r}(\hat{\mathbf{S}}.\mathbf{r})}{r^2}\right]\Bigg)}_{\text{Dipolar magnetic}}$$

Conduction electrons: “*s*” wave function

$$\longrightarrow \qquad H^c_{hf} = \frac{\mu_0}{4\pi}\frac{8\pi}{3} g_N \mu_N \;\hat{\mathbf{I}}.\hat{\mathbf{S}}\;\left|\Psi_c(0)\right|^2 = A_e\;\hat{\mathbf{I}}.\hat{\mathbf{S}}$$

Valence electrons (holes) : “*p*” wave function $l = 1$

$$\longrightarrow \qquad H^v_{hf} = \frac{\mu_0}{4\pi} g_N \mu_N \left\langle \frac{1}{r^3} \right\rangle_{\Psi_h} \frac{l(l+1)}{j(j+1)}\;\hat{\mathbf{I}}.\hat{\mathbf{J}} = A_h\;\hat{\mathbf{I}}.\hat{\mathbf{J}} \qquad (\hat{\mathbf{J}} = \hat{\mathbf{L}} + \hat{\mathbf{S}})$$

A. Abragam, Clarendon press, Oxford (1961)

The Fermi contact hamiltonian

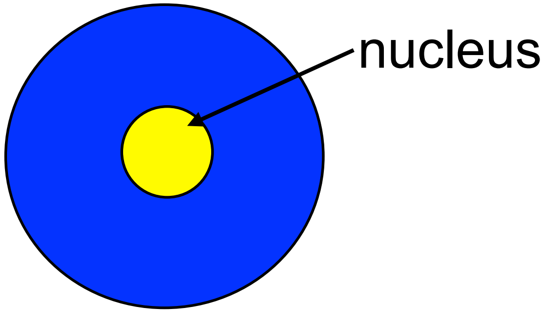
QD: $\Psi_{c(v)}(\mathbf{r}) = \psi(\mathbf{r})u_{c(v)}(\mathbf{r})$

$\psi(\mathbf{r})$ = envelope function

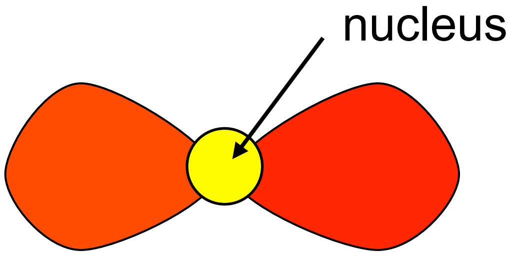
$u_{c(h)}(\mathbf{r})$ = periodic function

$$\hat{H}_{hf}^{fc} = v_0 \sum_j \boxed{A^j} \left|\psi(\mathbf{r}_j)\right|^2 \left[\hat{I}_z^j \hat{S}_z + (\hat{I}_+^j \hat{S}_- + \hat{I}_-^j \hat{S}_+)/2\right]$$

$\boxed{\sim |u_{c(h)}|^2}$ *Periodical part of the carrier Bloch function*



conduction electron:
"s" symmetry
➔ Strong overlap
(Fermi contact)



Valence hole:
"p" symmetry
➔ Weak overlap
(dipolar interaction only)

Weaker interaction of hole with nuclear spins

Gryncharova *et al.*, Sov. Phys. Semicond. **11**, 997 (1977).

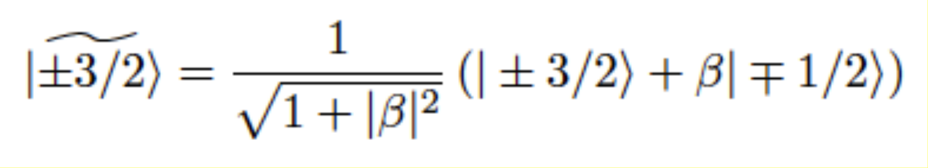
Hyperfine Interaction in Quantum Dots: CB ⇔ VB

For Conduction Electron spins: *Fermi contact Interaction*

$$\hat{H}_{hf}^{fc} = \nu_0 \sum_j A^j \left|\psi(\mathbf{r}_j)\right|^2 \left[\hat{I}_z^j \hat{S}_z + (\hat{I}_+^j \hat{S}_- + \hat{I}_-^j \hat{S}_+)/2 \right]$$

For Valence Hole spins: *Dipolar Interaction*

$$\hat{H}_{hf}^{dip} = \nu_0 \sum_j \frac{A_h^j}{1+\beta^2} \left|\psi(\mathbf{r}_j)\right|^2 \left[\hat{I}_z^j \hat{S}_z^h + \frac{|\beta|}{\sqrt{3}} (\hat{I}_+^j \hat{S}_-^h + \hat{I}_-^j \hat{S}_+^h)/2 \right]$$



$$|\widetilde{\pm 3/2}\rangle = \frac{1}{\sqrt{1+|\beta|^2}} \left(|\pm 3/2\rangle + \beta|\mp 1/2\rangle\right)$$

for pure Heavy Holes: β=0

Eble, PRL 2009

Hyperfine Interaction strength

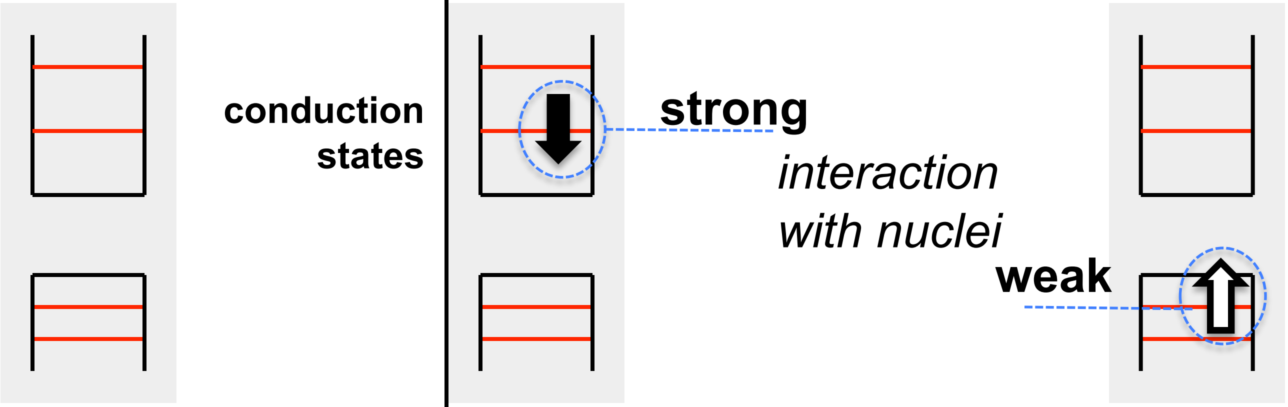
$$\frac{\text{Holes}}{\text{Electrons}} = \frac{\left|A_h^j\right|}{\left|A^j\right|} \approx 0.1$$

Quantum Dots: doping and electrical **charge control**

neutral

n type

p type



before photon absorption

conduction states

strong

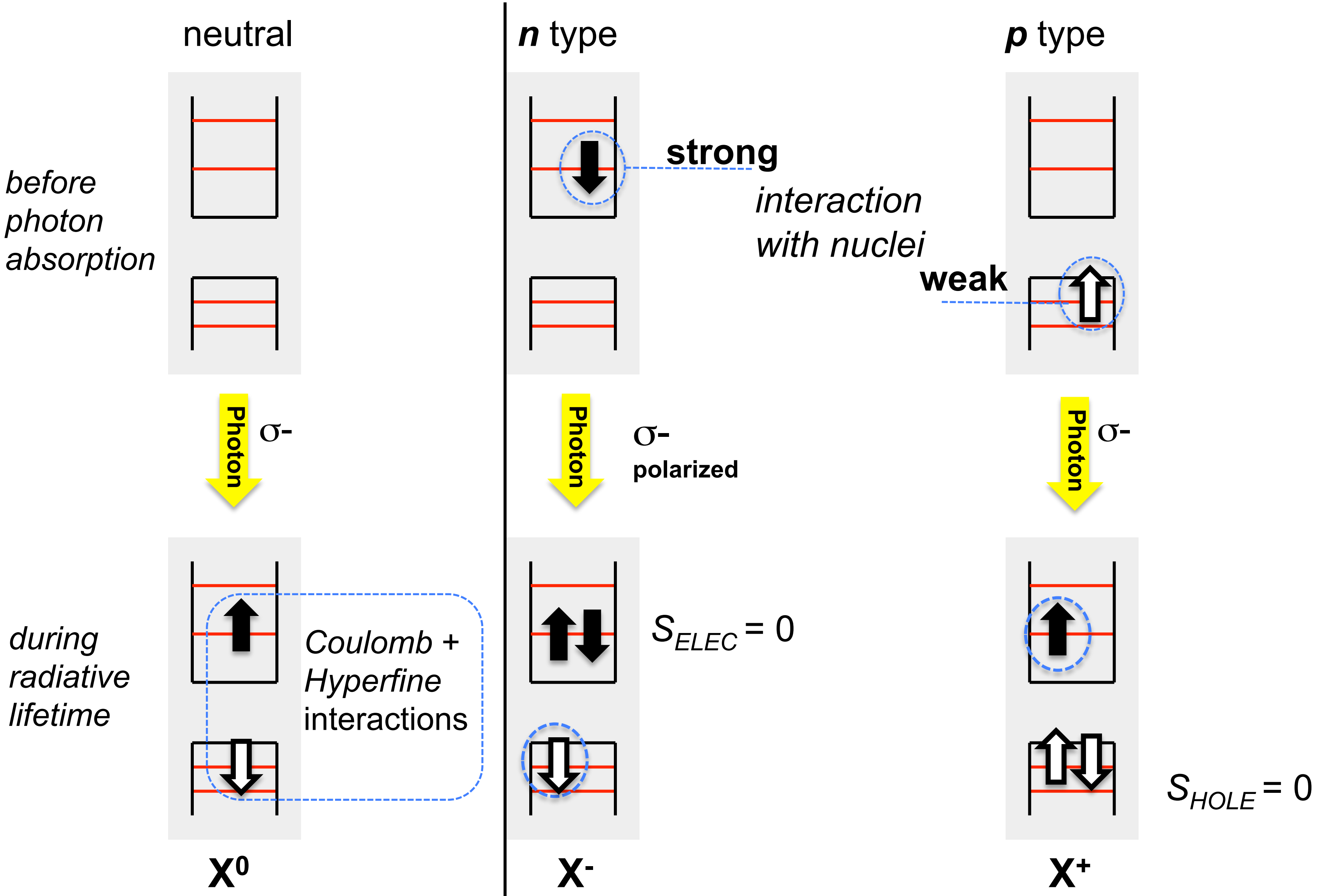
interaction with nuclei

weak

valence states

1. Photon Polarization → **2. Electron Spin Orientation** → 3. Nuclear Spin Orientation

Quantum Dots: doping and electrical **charge control**



neutral

n type

p type

before photon absorption

strong *interaction with nuclei* **weak**

Photon σ-

Photon σ- **polarized**

Photon σ-

during radiative lifetime

Coulomb + Hyperfine interactions

$S_{ELEC} = 0$

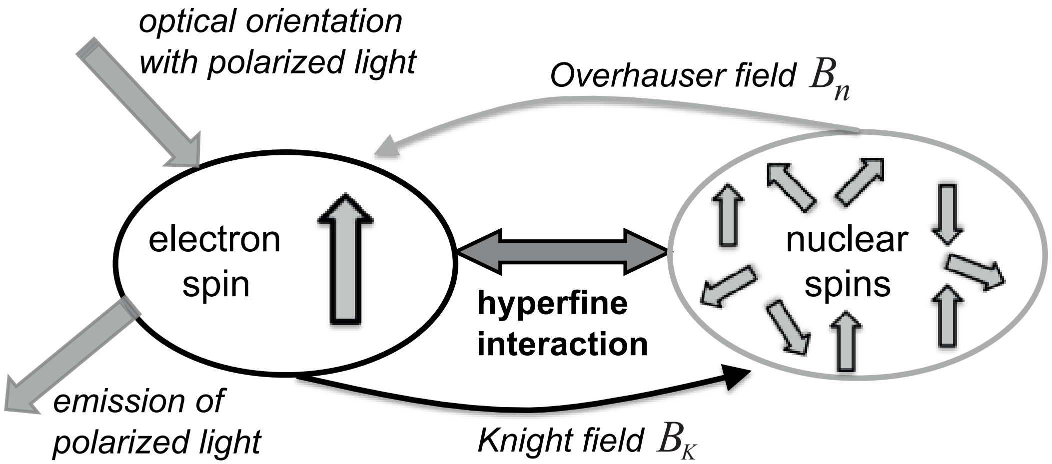
$S_{HOLE} = 0$

X^0

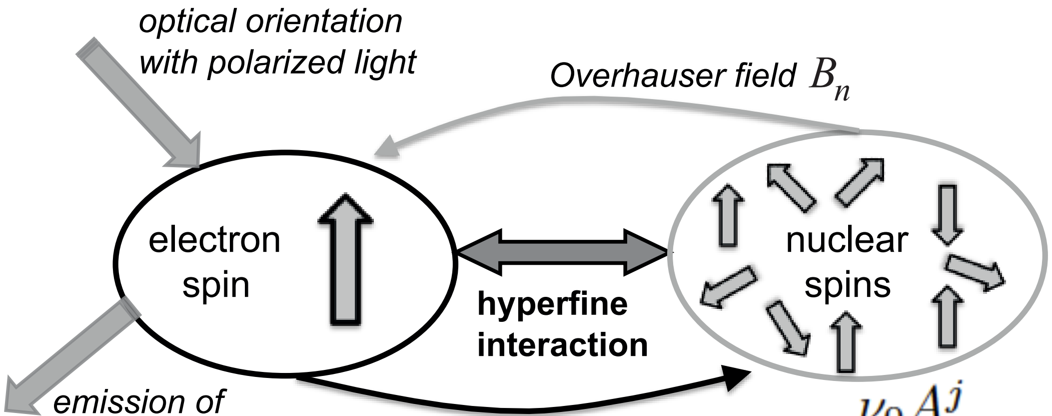
X^-

X^+

Duality of the Hyperfine Interaction



Duality of the Hyperfine Interaction



optical orientation with polarized light

Overhauser field B_n

electron spin

hyperfine interaction

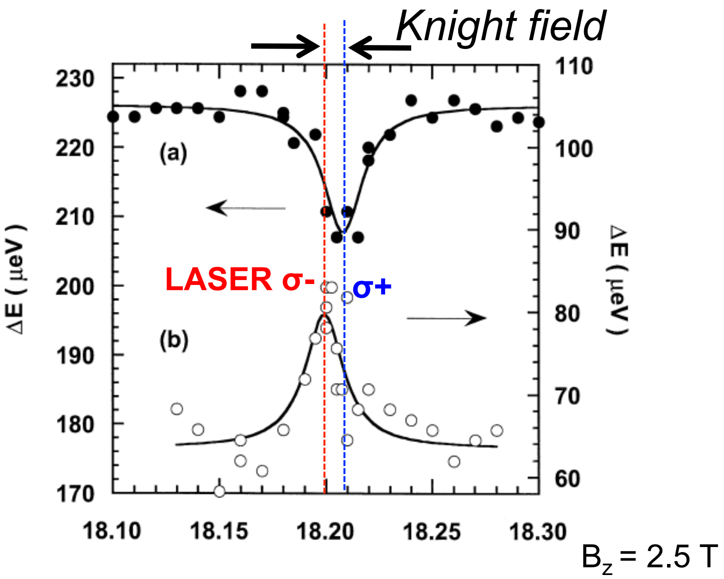
nuclear spins

emission of polarized light

Knight field $B_{K j} = f_e \dfrac{\nu_0 A^j}{g_N \mu_N} |\psi(\boldsymbol{r}_j)|^2 \langle \hat{\boldsymbol{S}}^e \rangle$

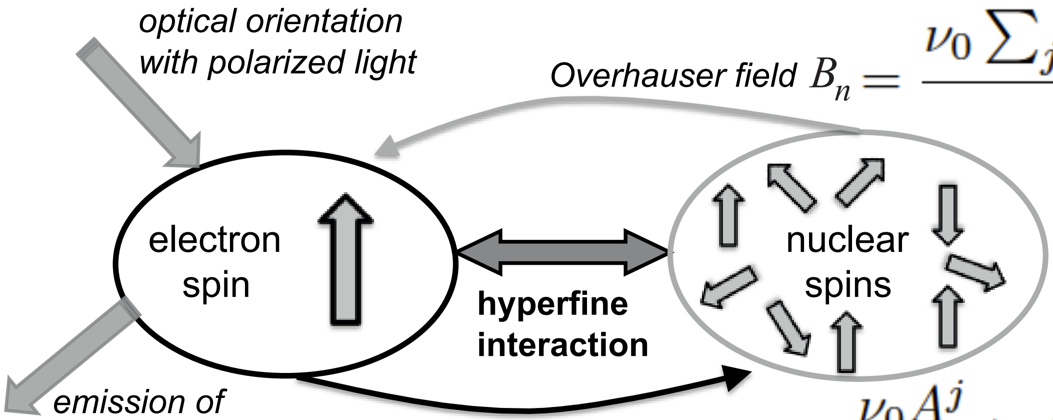
Knight Phys. Rev. 76, 1259 (1949).

Nuclear Zeeman Splitting = NMR frequency ^{75}As (MHz)



S.W. Brown et al. / Solid State Nuclear Magnetic Resonance 11 (1998) 49–58

Duality of the Hyperfine Interaction



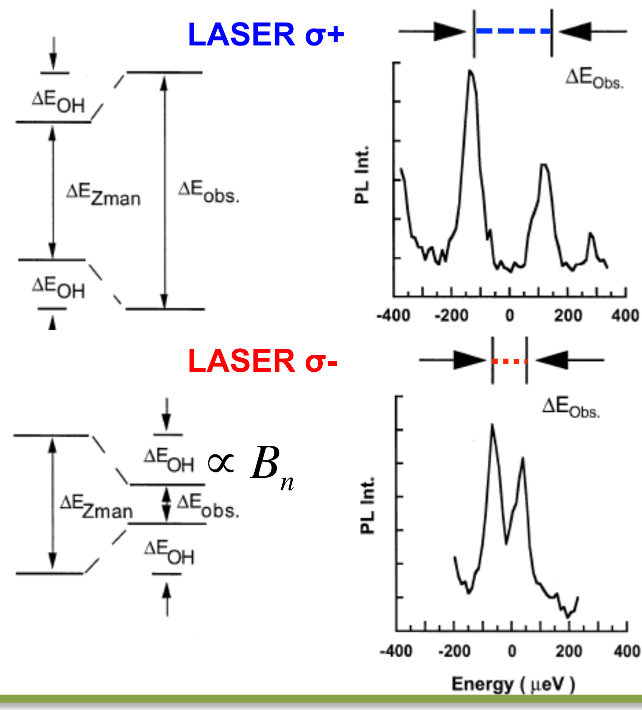
Overhauser field $$B_n = \frac{\nu_0 \sum_j A^j |\psi(\boldsymbol{r}_j)|^2 \langle \hat{\boldsymbol{I}}^j \rangle}{g_e \mu_B}$$

Overhauser Phys. Rev. 92, 411 (1953)

Knight field $$B_{K_j} = f_e \frac{\nu_0 A^j}{g_N \mu_N} |\psi(\boldsymbol{r}_j)|^2 \langle \hat{\boldsymbol{S}}^e \rangle$$

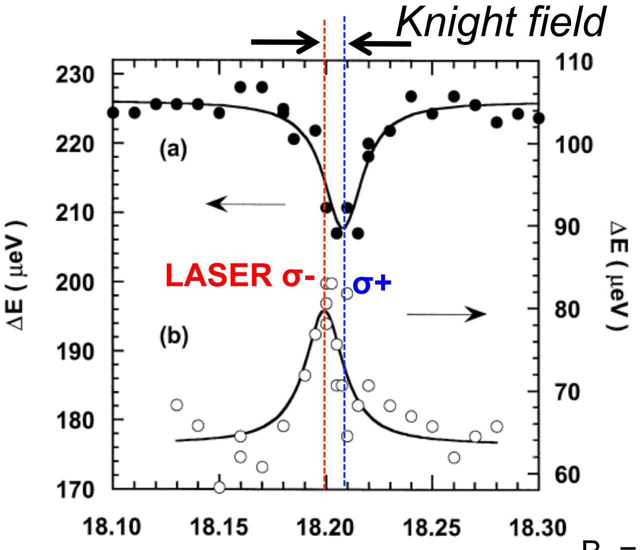
Knight Phys. Rev. 76, 1259 (1949).

Overhauser field + B_{ext}



Electron Zeeman Splitting

Nuclear Zeeman Splitting = NMR frequency ^{75}As (MHz)



$B_z = 2.5$ T

S.W. Brown et al. / Solid State Nuclear Magnetic Resonance 11 (1998) 49–58

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Fermi contact hamiltonian: *consequences*

$$\hat{H}_{hf} = \frac{\nu_0}{2} \sum_j A^j |\psi(\bar{r}_j)|^2 \left(2\hat{I}^j_z \hat{S}^e_z + \underline{[\hat{I}^j_+ \hat{S}^e_- + \hat{I}^j_- \hat{S}^e_+]} \right)$$

Depending on

1) *experimental conditions*
2) *samples …*

a succession of spin flip-flops can lead to:

* **Electron spin *dephasing***

* **Dynamic *polarization* of nuclear spins**

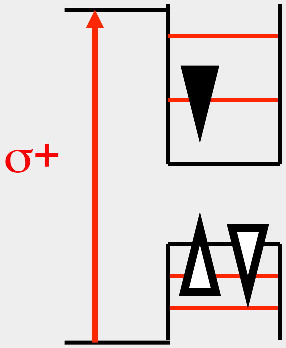
* ***Depolarization* of nuclear spins**

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j|\psi(\bar{r}_j)|^2\left(2\hat{I}_z^j\hat{S}_z^e + [\hat{I}_+^j\hat{S}_-^e + \hat{I}_-^j\hat{S}_+^e]\right)$$

Electron spin dephasing

Time resolved photoluminescence in InAs dots: B_{ext}=0 and T=10K

σ+ laser excitation into wetting layer

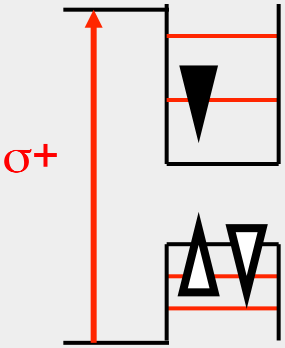


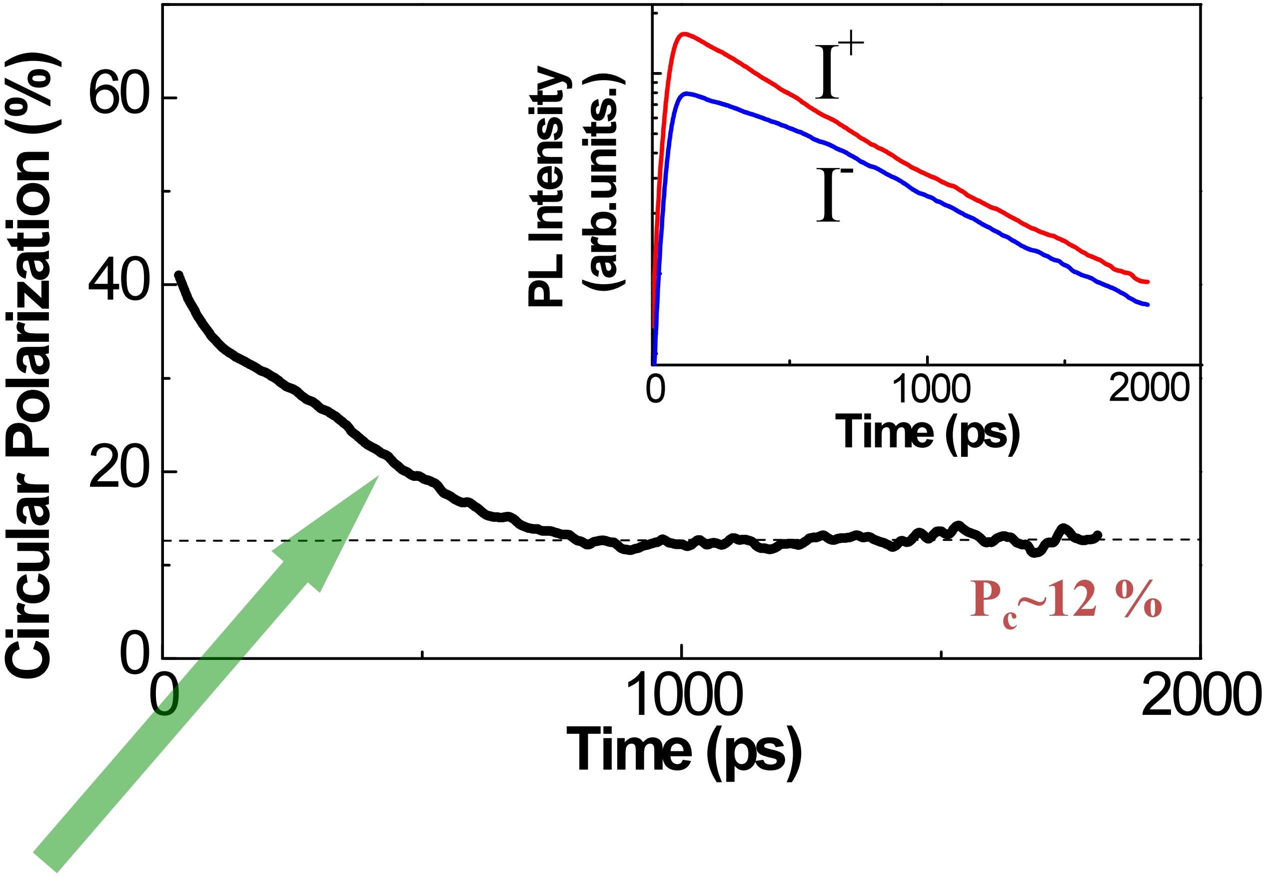
$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j|\psi(\bar{r}_j)|^2 \left(2\hat{I}_z^j\hat{S}_z^e + [\hat{I}_+^j\hat{S}_-^e + \hat{I}_-^j\hat{S}_+^e]\right)$$

Electron spin dephasing

Time resolved photoluminescence in InAs dots: $B_{ext}=0$ and $T=10K$

σ+ laser excitation into wetting layer





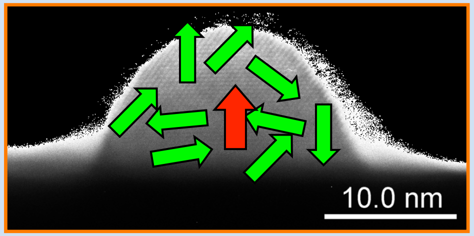
Decay due to hyperfine interaction

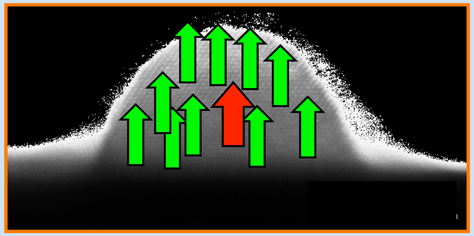
Braun *et al* Phys. Rev. Lett. 94, 116601 (2005)

How precisely can we determine the Overhauser field B_n in a Quantum Dot ?

Merkulov et al, PRB 2002; Dyakonov & Perel Sov. Phys. JETP 1974

Khaetskii et al, PRL 2002





For GaAs dots: *B_n (min) = 0* *B_n (max) = 5 T*

With a root mean square deviation $\delta B_n = \sqrt{\langle B_n^2 \rangle - \langle B_n \rangle^2} = 20 mT$ in InAs dots

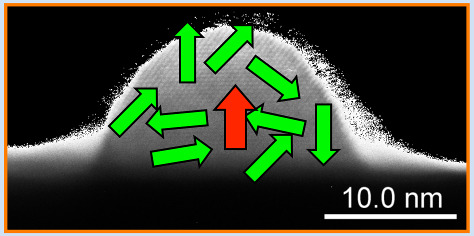
↓

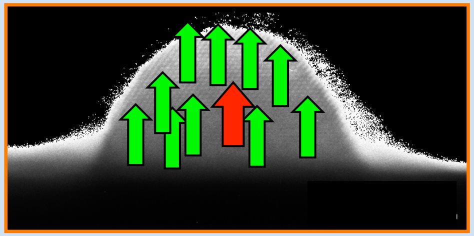
Electron 'feels' small magnetic field of arbitrary orientation, even if $\langle B_n \rangle = 0$

How precisely can we determine the Overhauser field B_n in a Quantum Dot ?

Merkulov et al, PRB 2002; Dyakonov & Perel Sov. Phys. JETP 1974

Khaetskii et al, PRL 2002





For GaAs dots: *B_n (min) = 0* *B_n (max) = 5 T*

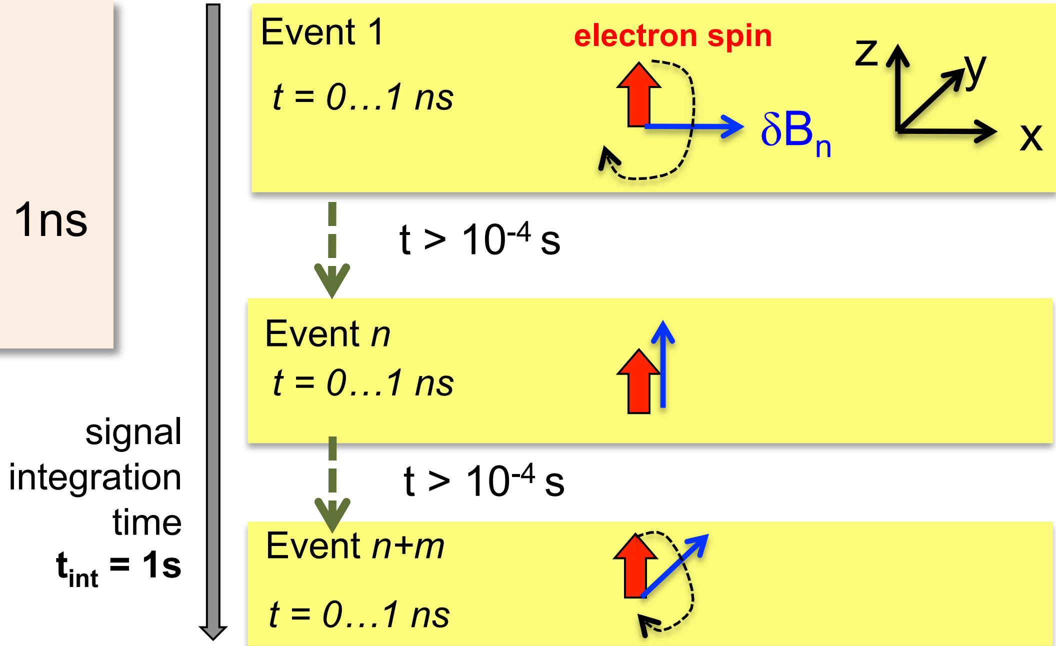
With a root mean square deviation $\delta B_n = \sqrt{\langle B_n^2\rangle - \langle B_n\rangle^2} = 20mT$ in InAs dots

↓

Electron 'feels' small magnetic field of arbitrary orientation, even if $\langle B_n\rangle = 0$

Optical spectroscopy **timescales**:

- electron stays 1ns in dot
- δB_n changes direction every 10^{-4} s >> 1ns
- signal integration time 1s >> 10^{-4} s



Event 1

t = 0…1 ns

t > 10^{-4} s

Event *n*

t = 0…1 ns

t > 10^{-4} s

Event *n+m*

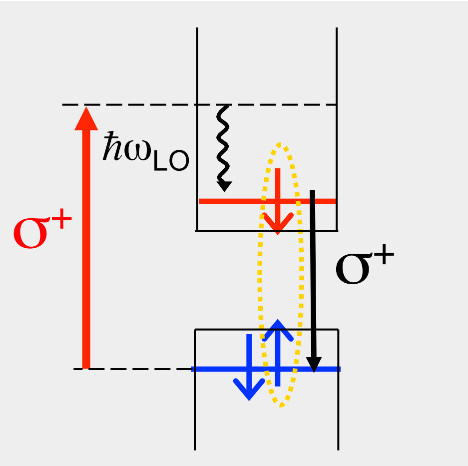
t = 0…1 ns

signal
integration
time
t_{int} = 1s

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j|\psi(\bar{r}_j)|^2 \left(2\hat{I}^j_z\hat{S}^e_z + [\hat{I}^j_+\hat{S}^e_- + \hat{I}^j_-\hat{S}^e_+]\right)$$

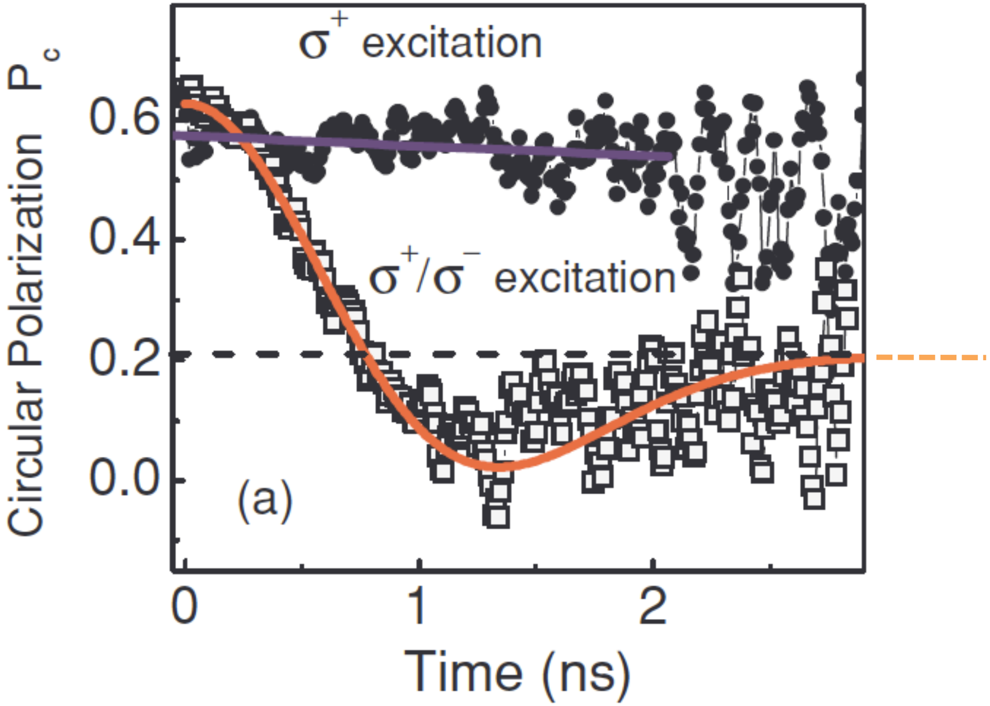
Electron spin dephasing

Time resolved photoluminescence in InAs dots: $B_{ext}=0$ and $T=10K$



Single Dot spectroscopy

Dou et al. Phys. Rev. B **84**, 033302 (2011)

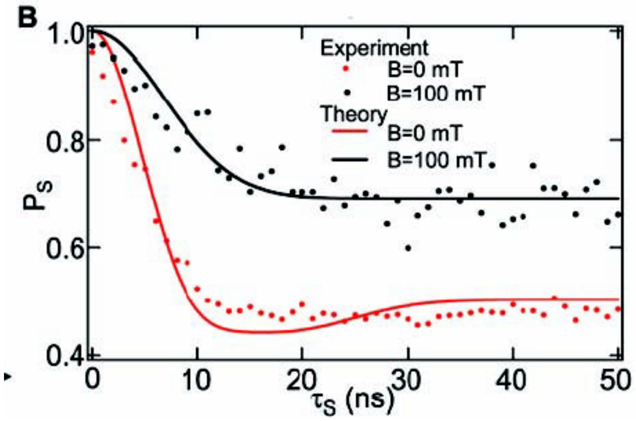


$= 1/3\ P_c(t=0)$

dephasing time T_Δ = *0.5 ns*

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j |\psi(\bar{r}_j)|^2 \left(2\hat{I}_z^j \hat{S}_z^e + [\hat{I}_+^j \hat{S}_-^e + \hat{I}_-^j \hat{S}_+^e]\right)$$

Electron spin dephasing
other systems



Gate defined GaAs double dot

Temperature: *135mK*

bigger dots ⇔ Longer dephasing time T_Δ = *10ns*

$$\delta B_n = \frac{1}{g_e \mu_B}\frac{2\tilde{A}}{\sqrt{N}}\sqrt{I(I+1)}$$

Petta, Science 2005

Bluhm, Nature Physics 2010 *prolonged up to* *200µs*

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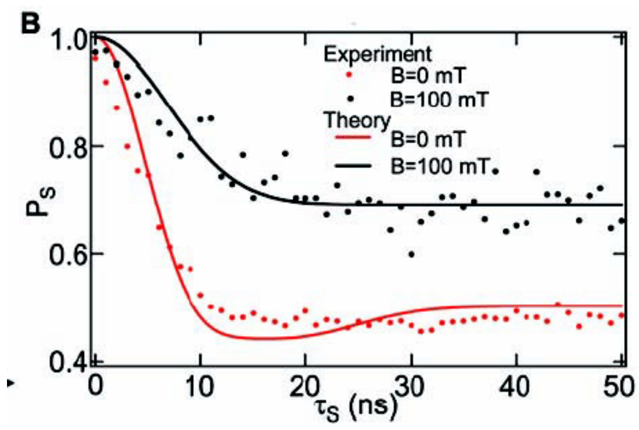
Electron spin dephasing
other systems



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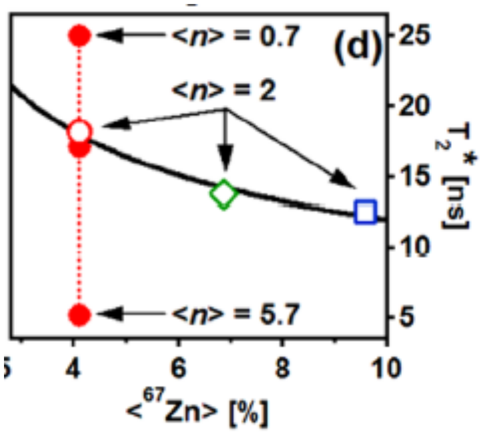


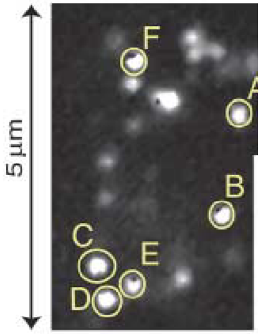
ZnO nano-crystals

Temperature: *300K*

Electron spin dephasing time: *25ns*

Liu, Phys. Rev. Lett. 2007

Whitaker, J. Phys. Chem. 2010



Nitrogen Vacancy centre in Diamond

Temperature: *300K*

Electron spin dephasing time: *µs*

Childress, Science 2006

Balasubramanian, Nature Materials 2009

Outline:

- Introduction to semiconductor quantum dots
- Hyperfine interaction between nuclear spins ⇔ carrier spins
- Carrier spin dephasing due to fluctuations of the Nuclear Field
- Optical Pumping of Nuclear Spins
- Nuclear Spins Physics in quantum dots: What’s new ?

Fermi contact hamiltonian: *consequences*

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j |\psi(\bar{r}_j)|^2 \left(2\hat{I}^j_z\hat{S}^e_z + [\hat{I}^j_+\hat{S}^e_- + \hat{I}^j_-\hat{S}^e_+]\right)$$

Depending on

1) *experimental conditions*
2) *samples …*

a succession of spin flip-flops can lead to:

* **Electron spin *dephasing***

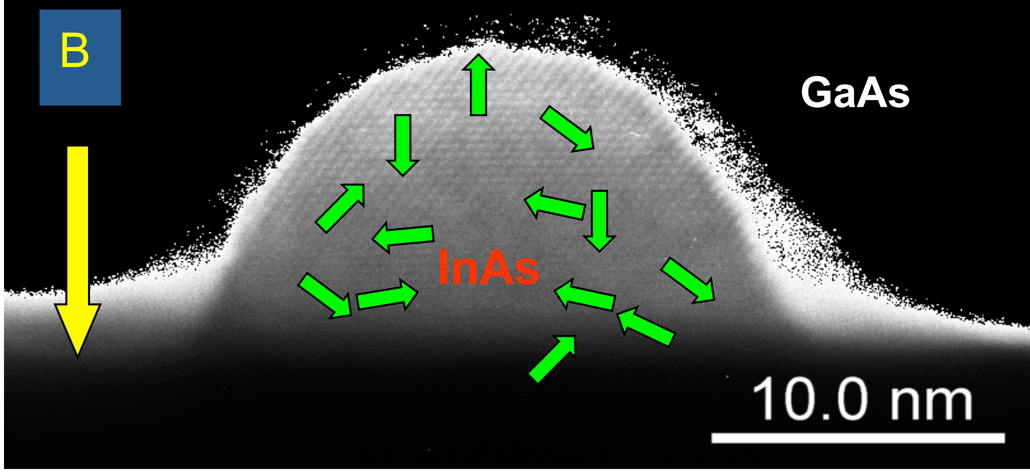
* **Dynamic *polarization* of nuclear spins**

* ***Depolarization* of nuclear spins**

↑ nuclear spin

B applied magnetic field

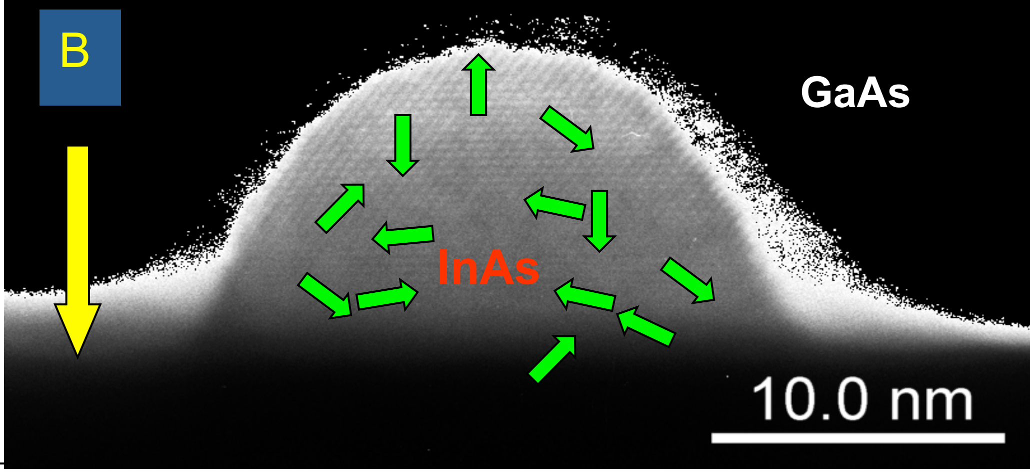
↑ electron spin



↑ nuclear spin

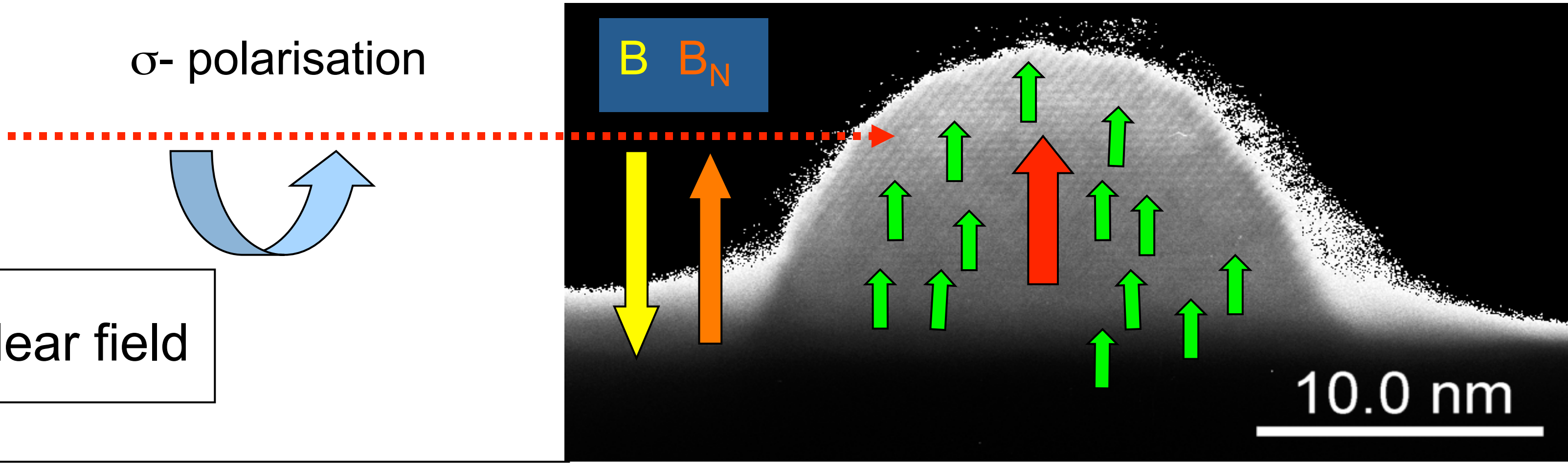
B applied magnetic field

↑ electron spin

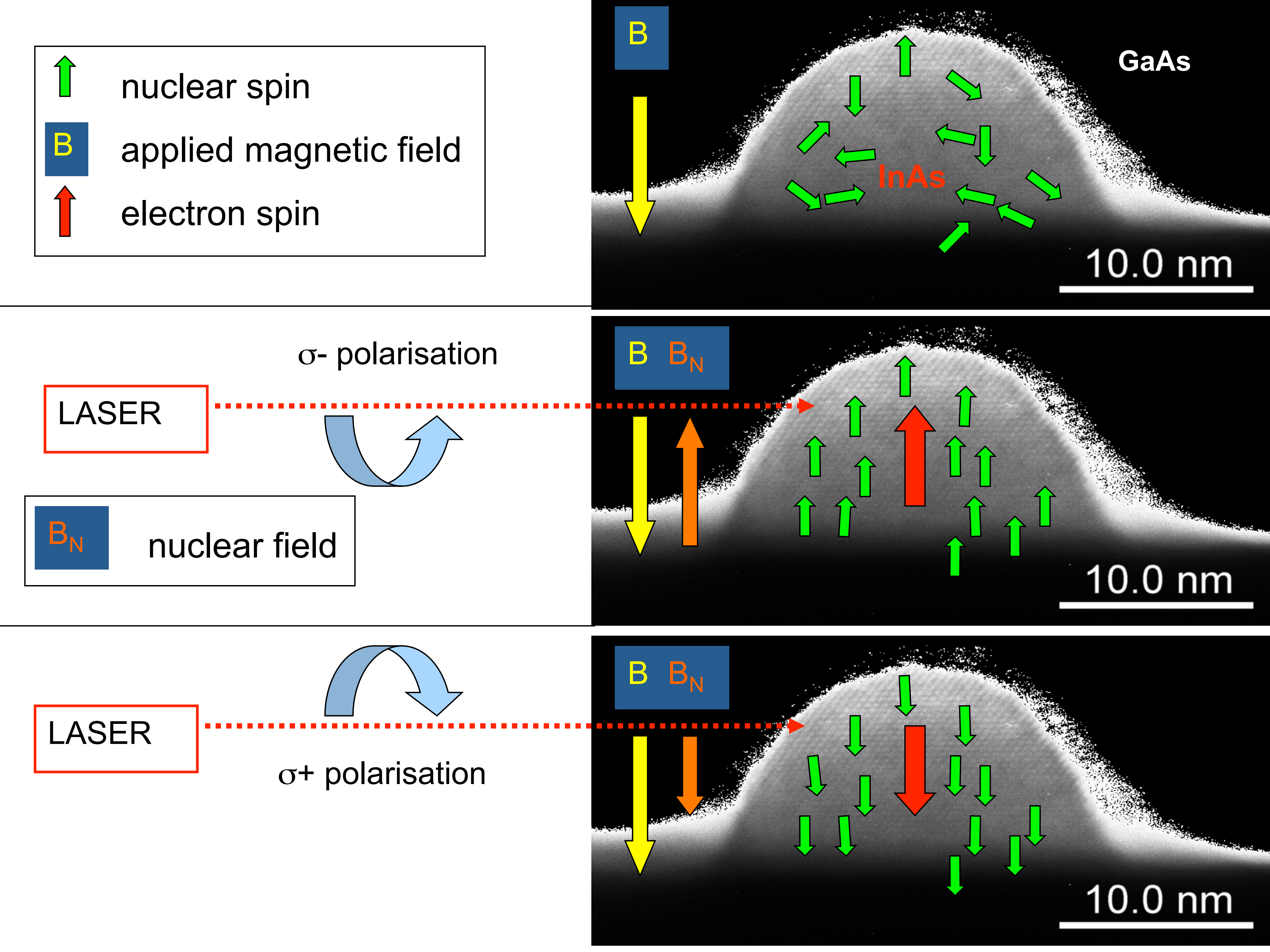


σ- polarisation

LASER



B_N nuclear field



nuclear spin

B applied magnetic field

electron spin

σ- polarisation

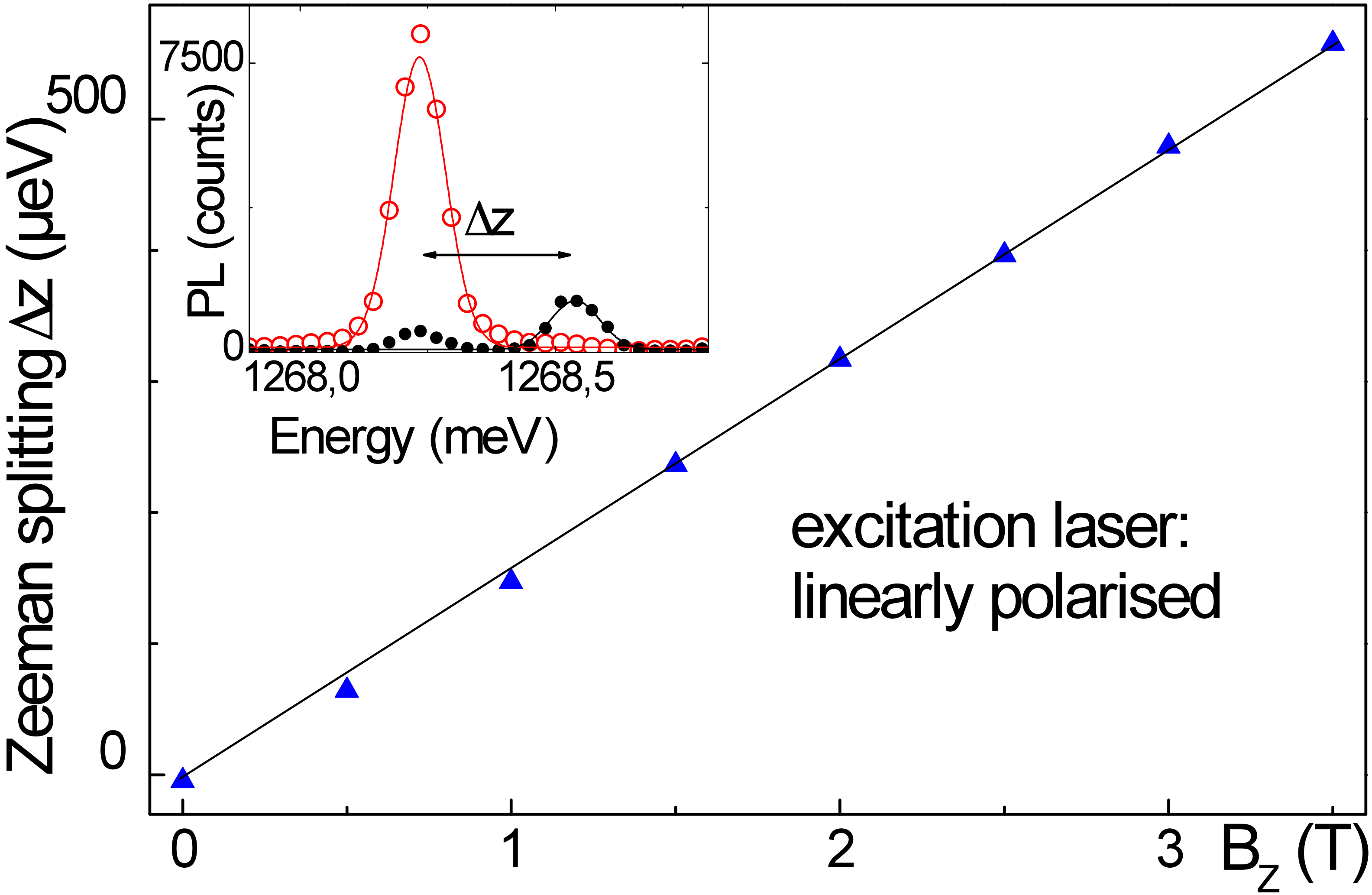
LASER

B_N nuclear field

LASER

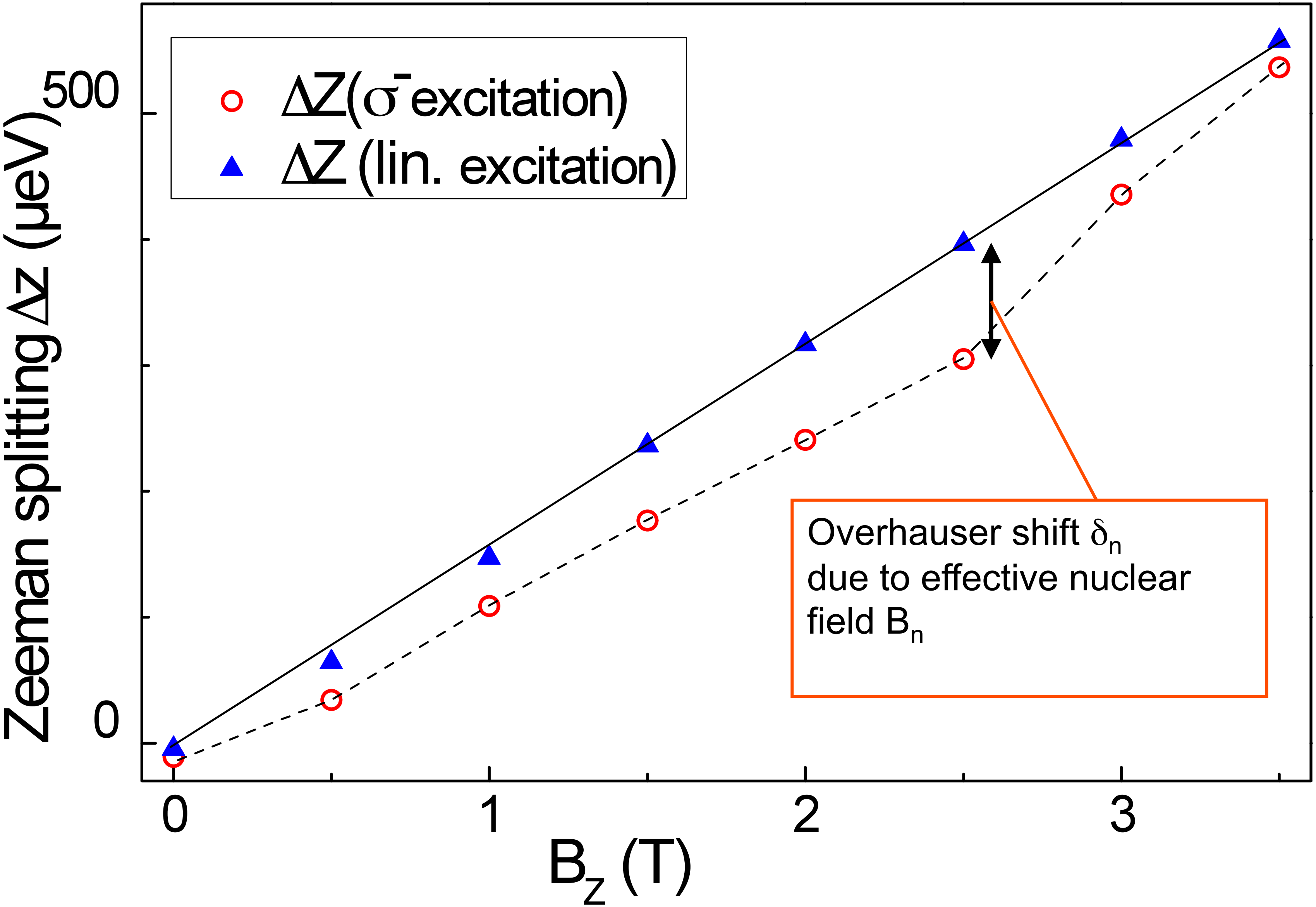
σ+ polarisation

Magnetic field dependence



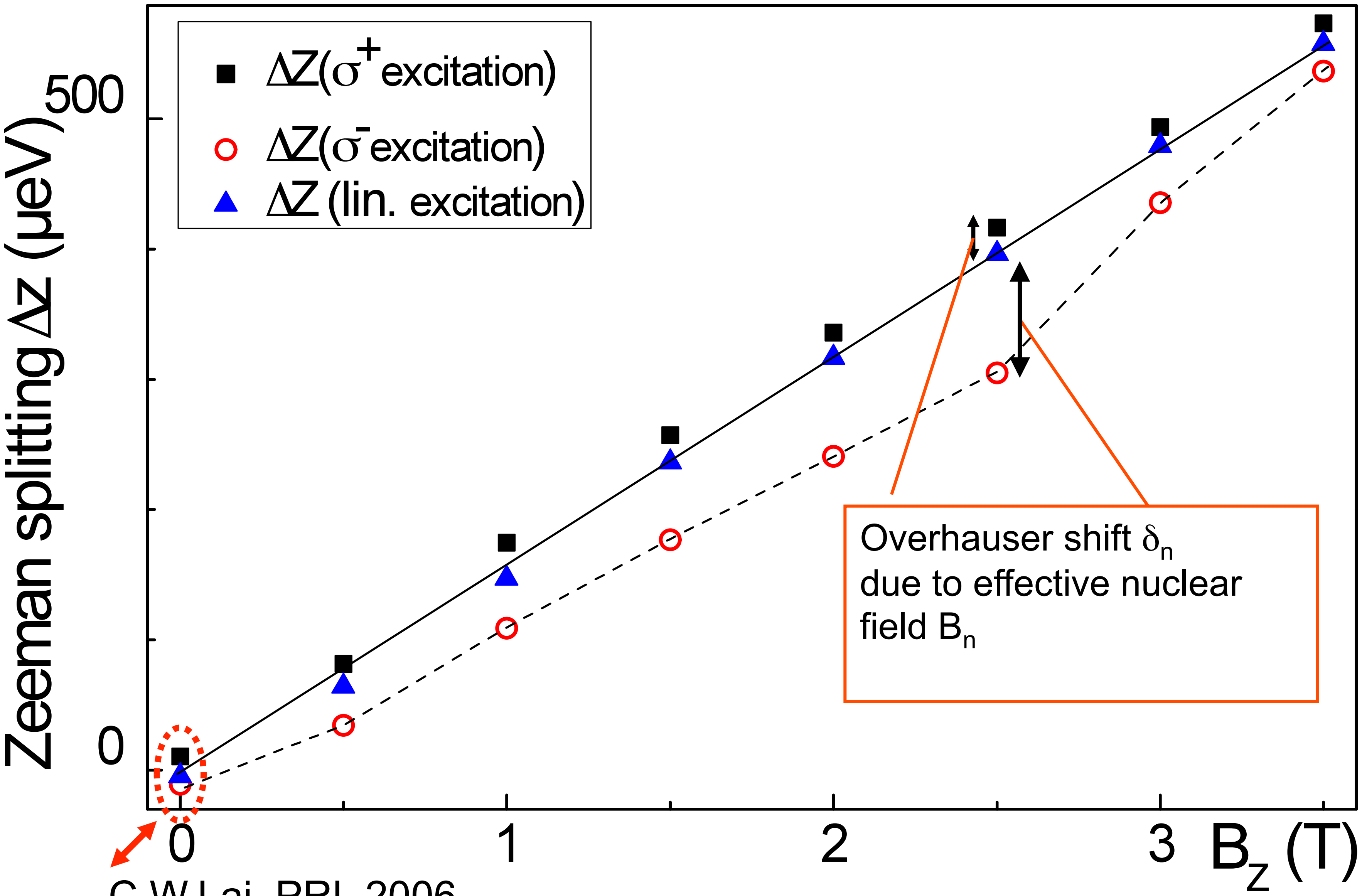
the average electron spin is zero
➔ no dynamical nuclear polarisation

Magnetic field dependence



Overhauser shift δ_n due to effective nuclear field B_n

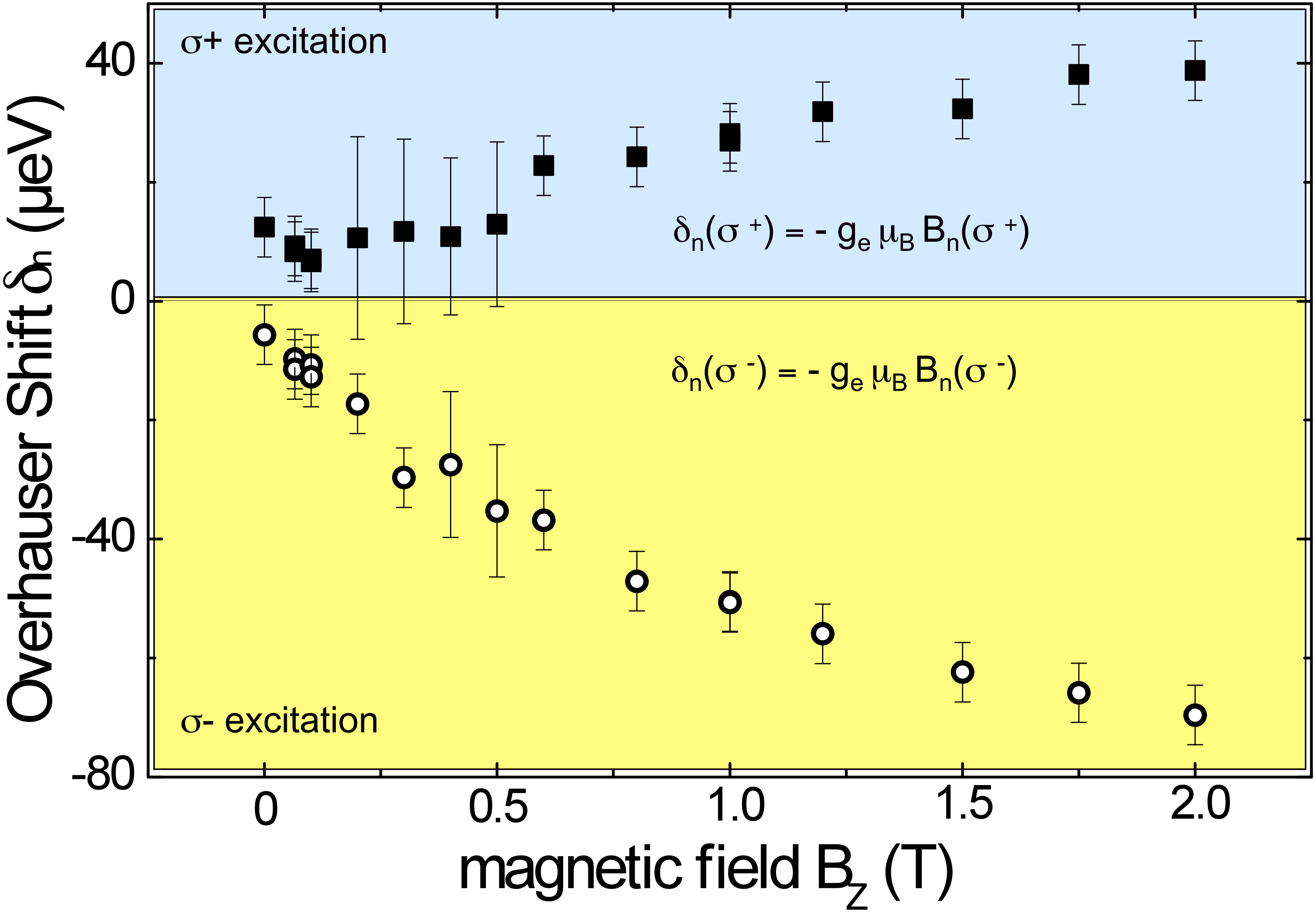
Magnetic field dependence



Overhauser shift δ_n due to effective nuclear field B_n

C.W.Lai, PRL 2006

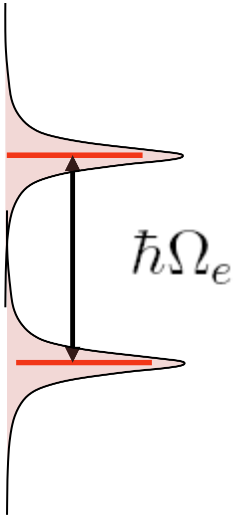
Magnetic field dependence



Dynamical nuclear polarisation in a magnetic field

$Bz >> B_e, B_L$

$$\left|\uparrow^{e}\right\rangle \otimes \left|I_z\right\rangle$$



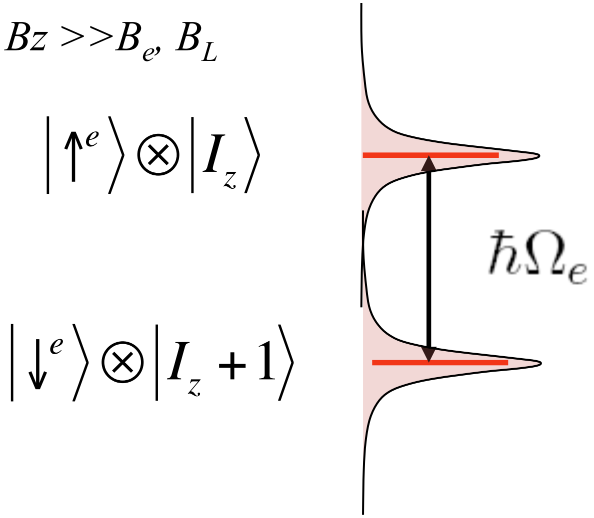
$$\left|\downarrow^{e}\right\rangle \otimes \left|I_z + 1\right\rangle$$

$$\mathcal{H}_0 \approx \hbar\gamma_n B_z \hat{I}_z + \underbrace{g_e \mu_B \left(B_z + B_n\right)}_{\hbar\Omega_e} \hat{S}_z$$

The **flip-flop term** = random perturbation between states split in energy by $\hbar\Omega_e$.

$$\mathcal{H}_1(t) = \frac{A}{N}\left(\hat{S}_+\hat{I}_- + \hat{S}_-\hat{I}_+\right) h_1(t)$$

Dynamical nuclear polarisation in a magnetic field



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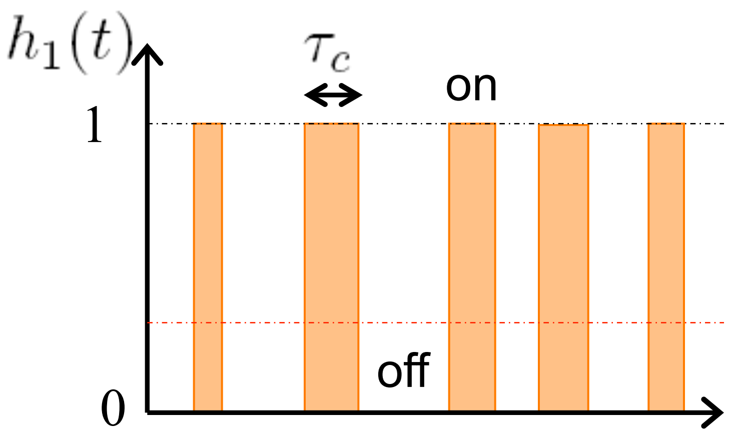
$$\mathcal{H}_1(t) = \frac{A}{N}\left(\hat{S}_+\hat{I}_- + \hat{S}_-\hat{I}_+\right) h_1(t)$$

Its temporal dependence is characterised by a **correlation time** τ_c



$$f_e = \overline{h_1(t)}$$

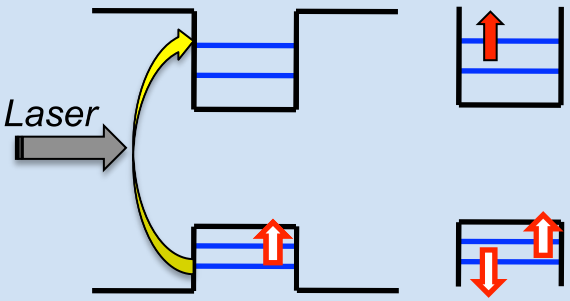
rate of nuclear polarization determined by

(1) total splitting $\hbar\Omega_e = \delta_z + \delta_n$

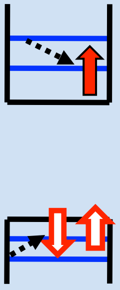
(2) level broadening $\hbar/\tau_c$

Correlation time electron spin – nuclear spin interaction: *several Physical Origins*

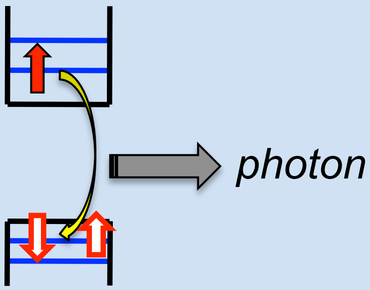
1. Electron spin 'lifetime'



photon
absorption
t = 0



energy relaxation
to ground state
t ≈ 10 ps



radiative
recombination
t ≈ 1 ns

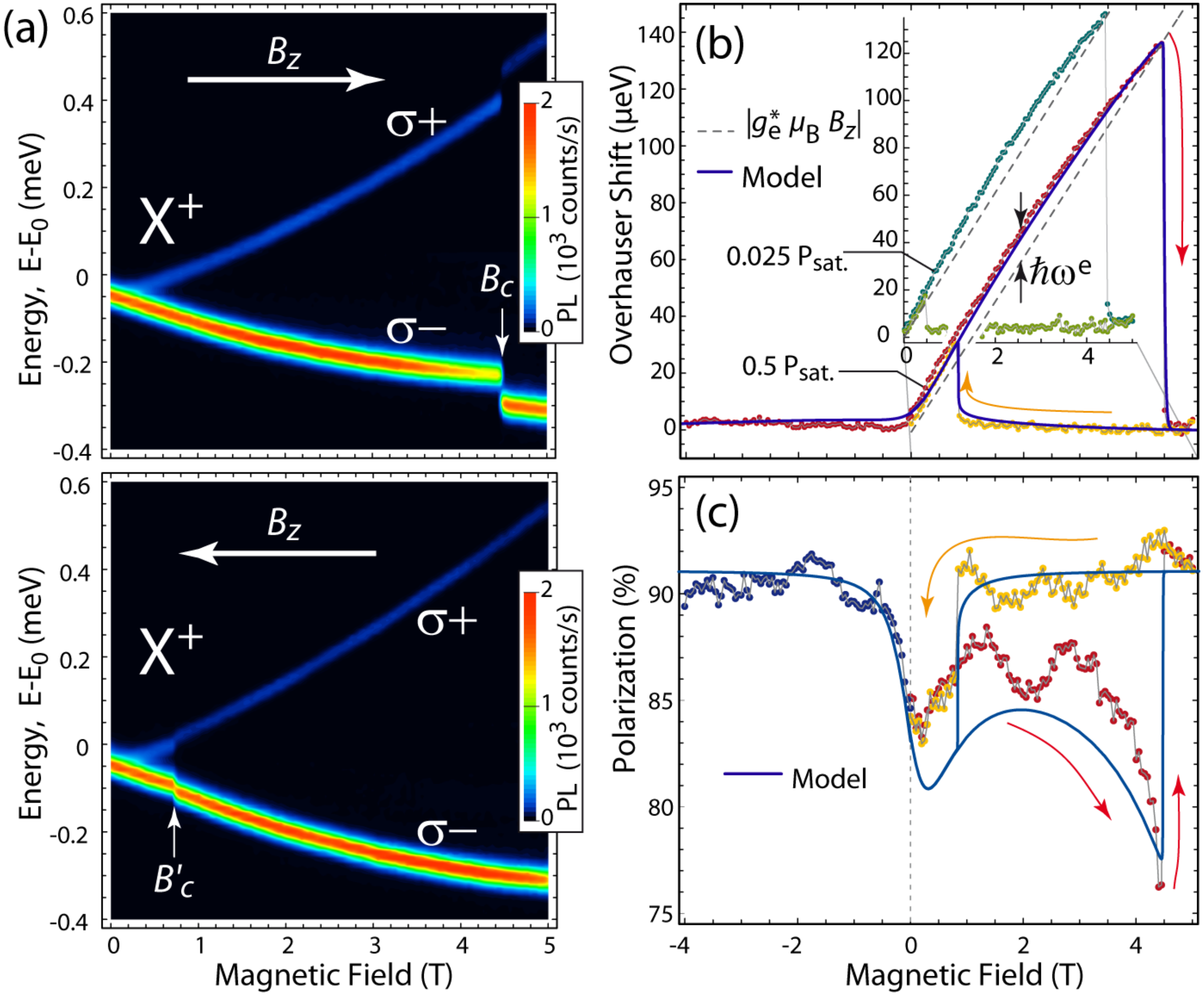
orders of magnitude: $\frac{\hbar}{10\,ps} \approx 65\mu eV$ ↔ electron Zeeman splitting at 2T:
≈ 60 μeV

Other origins:

- co-tunneling in charge tunable structures
- Hyperfine flip-flops themselves
- …

Strong Non-linearities and Bistabilities

as seen in Paris, Zurich, St. Petersburg, Sheffield, Chicago, Edinburgh, Toulouse …



Model:

Eble et al, PRB 2006

A. Abragam, *Principles of Nuclear Magnetism*

We extract:

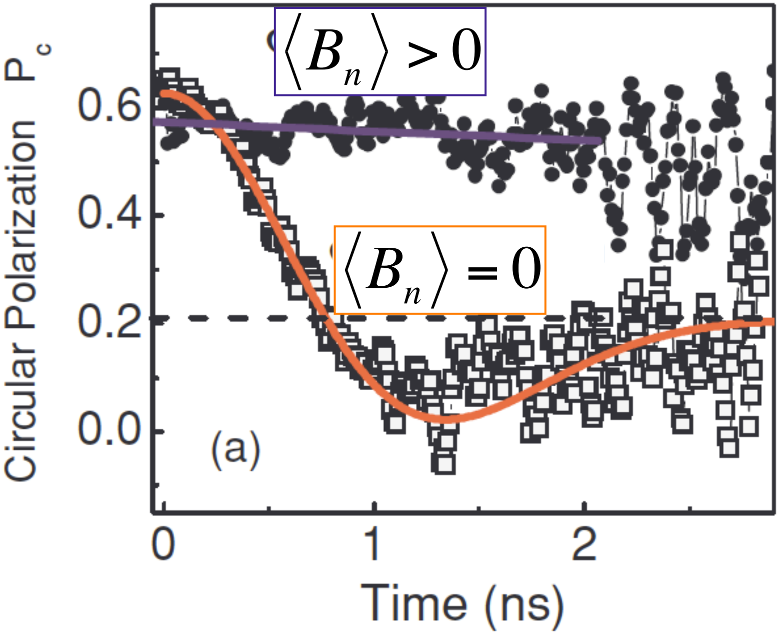
Correlation time τ_c *= 70ps*

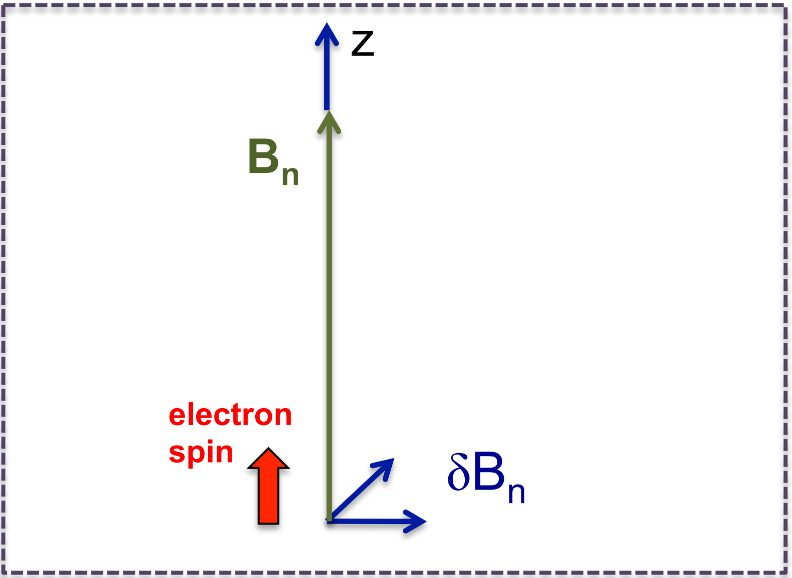
Olivier Krebs in Rev. Mod. Phys. arXiv 1202.4637

In the presence of strong nuclear polarization: *Are the fluctuations δB_n still important ?*

1. For Electron spin lifetime (z-projection): NO

Dou et al. Phys. Rev. B **84**, 033302 (2011)





2. For Electron spin coherence: YES

New Question: What happens when we polarize 99.99 % of all Nuclear spins ?

➔ Experimental Challenge !

Question: Can we polarize Nuclear Spins at zero Magnetic field ?

1 nucleus *n* interacts with other nuclei *n’* via dipole-dipole interaction

$$\hat{H}_{dd} = \frac{\mu_N^2}{2} \sum_{n \neq n'} \frac{g_n g_{n'}}{r_{nn'}^3} \left(\hat{I}^n \hat{I}^{n'} - 3 \frac{(\hat{I}^n \boldsymbol{r}_{nn'})(\hat{I}^{n'} \boldsymbol{r}_{nn'})}{r_{nn'}^2} \right)$$

⇒ *precession in a fluctuation field* **$\delta B_L \approx 0.2$ mT**

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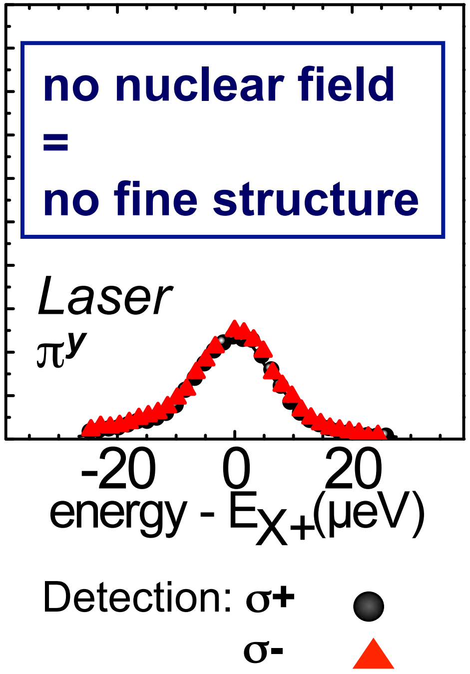
Answer: YES – if we can screen δB_L Lai et al, PRL 2006



Phys. Rev. Lett. 103, 086601 (2009)

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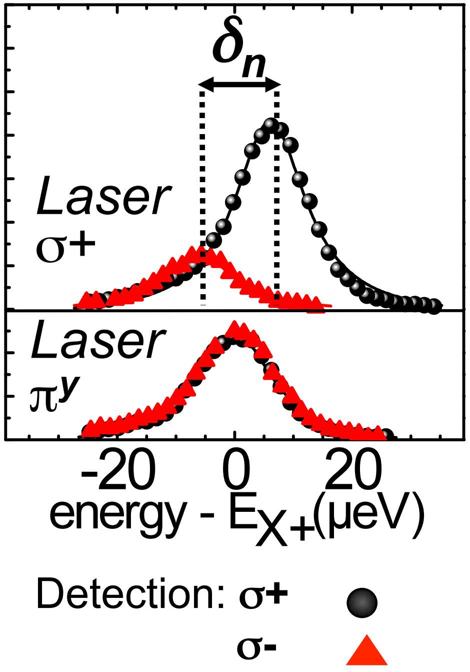
What mechanism screens δB_L ?

1. Knight field B_K : mT range
2. Nuclear Quadrupolar Interaction in strained dots: several 10 to several 100 mT`

   Dzhioev & Korenev, Phys. Rev. Lett. 2007



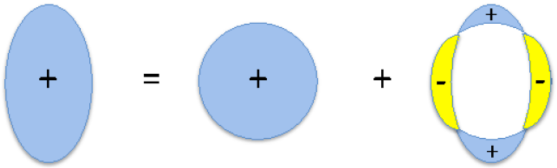
Belhadj et al Phys. Rev. Lett. 103, 086601 (2009)

Nuclear Spins I>1/2 sensitive to electric field gradients: ***nuclear quadrupole effects***

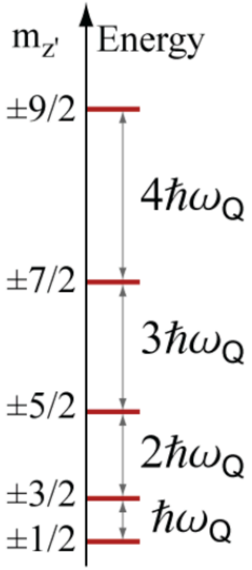
Due to:

- Lattice Strain
- Alloy dissorder

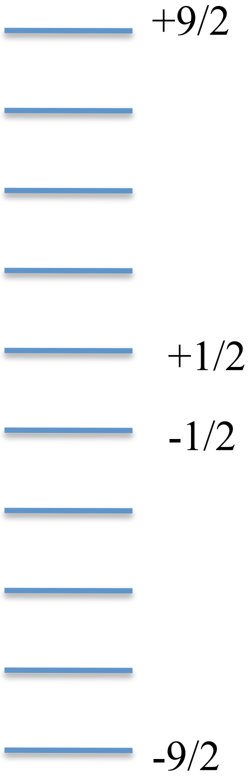
Origin of electric quadrupole moments:
prolate shape of nuclei



Magnetic field = 0



Magnetic field > 0



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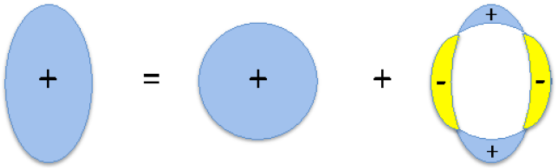

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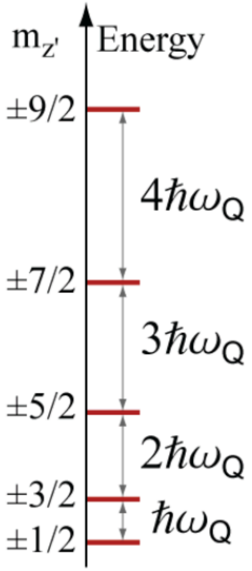
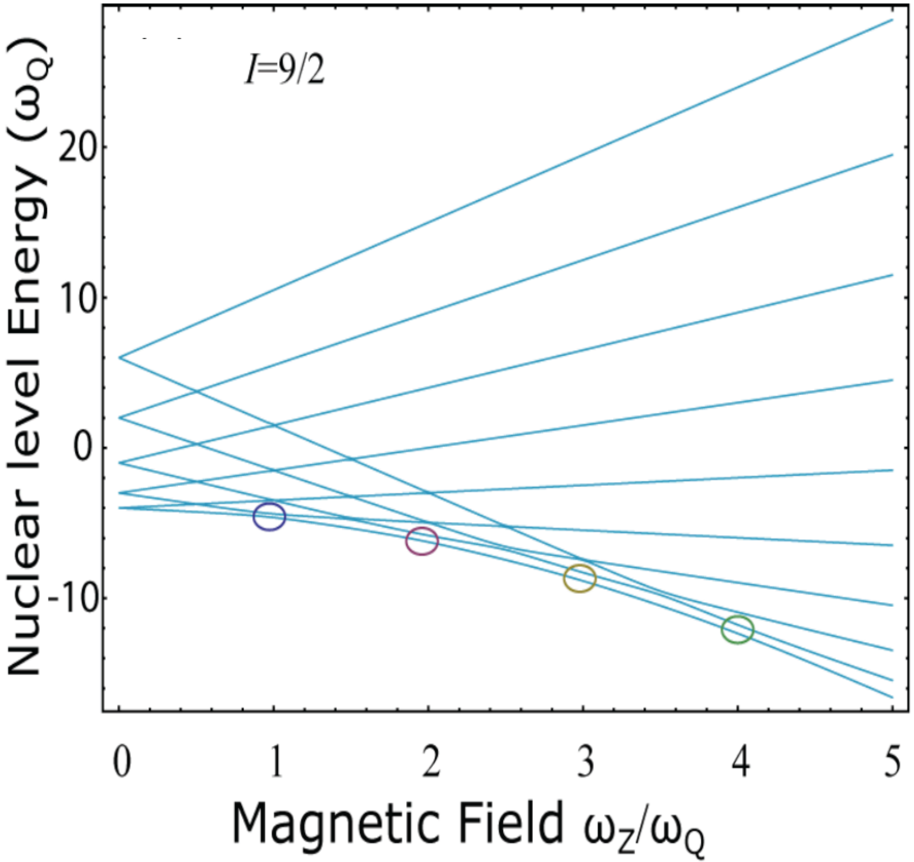
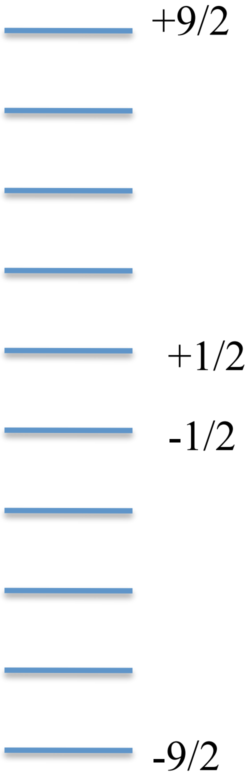




Magnetic field > 0



more atomistic insight: Ceyhun Bulutay PRB 85, 115313 (2012)

see also: Talk E.A. Chekhovich

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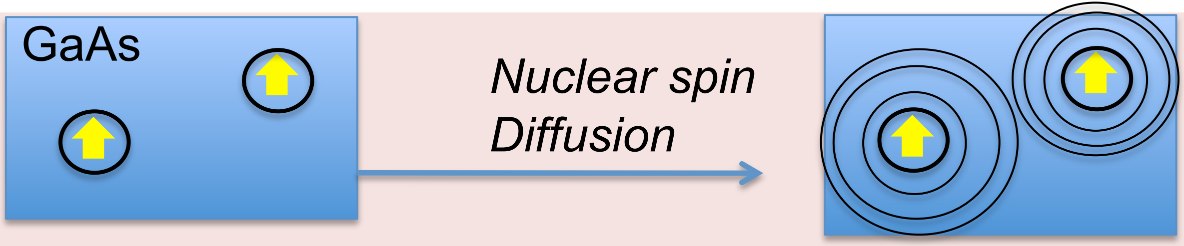
- Optical Pumping of Nuclear Spins
- Nuclear Spins Physics in quantum dots: What's new ?

1 electron Spin Interaction with a mesoscopic nuclear spin ensemble

Electrons bound to Donors



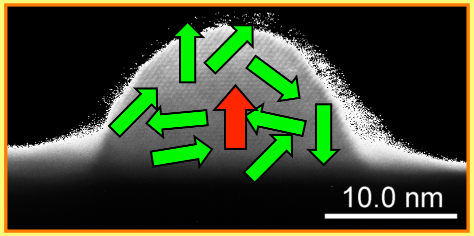
Nuclear spin Diffusion

Optical Orientation, edited by Meier and Zakharchenya (1984)

1 Electron in a single dot →

Different experimental techniques and new Physics: *a small selection*

Rev. Mod. Phys. arXiv1202.4637



- access Nuclear spin polarization (Overhauser shift) directly
- charge tuning: DNP evolution with and without carriers
- Strong chemical contrast dot material ⇔ barrier material
- holes spins can be initialized *(talk Andrew Ramsay)*
- **strong strain = strong nuclear quadrupolar effects** *(talk E.A. Chekhovich)*
  - strong suppression of spin diffusion
  - **D**ynamic **N**uclear **P**olarization at zero magnetic field
  - Anomalous Hanle Effect
  - Line dragging in Absorption …. *(talk Martin Kroner)*

Overhauser effect:

Polarize Nuclear spins via flip-flop

$$\mathcal{H}_1(t) = \frac{A}{N}\left(\hat{S}_+\hat{I}_- + \hat{S}_-\hat{I}_+\right) h_1(t)$$

How can we compensate electron Zeeman energy in strong magnetic fields ?

Overhauser effect:

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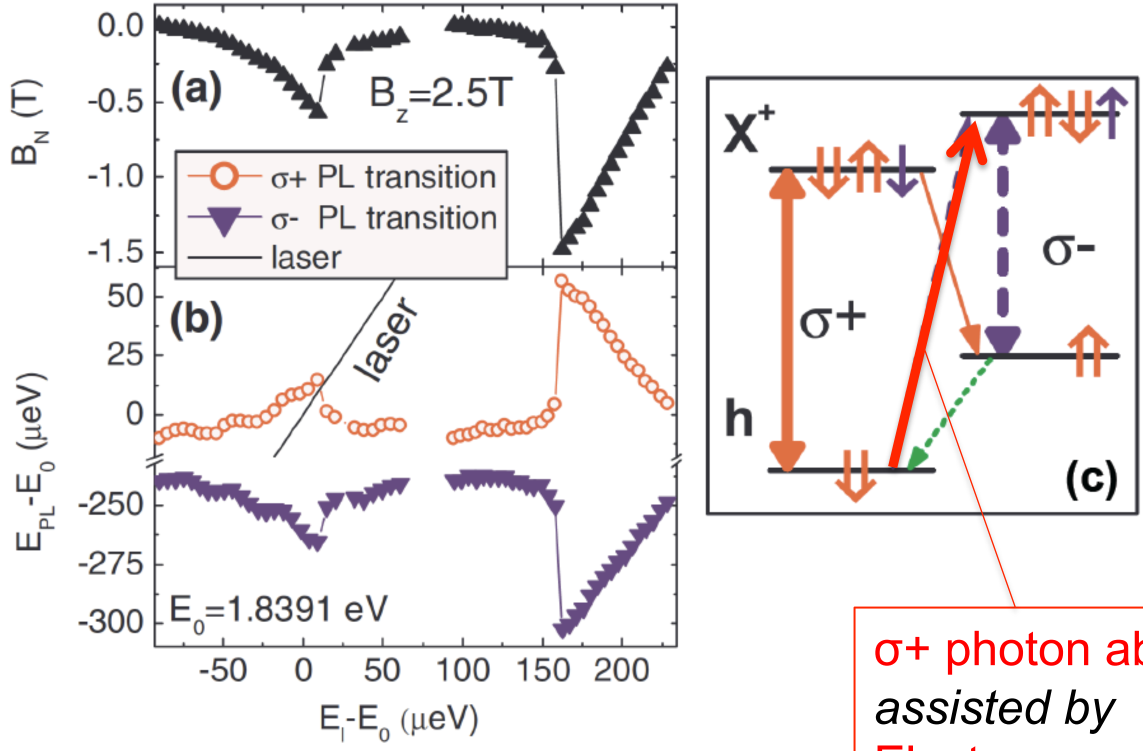
How can we compensate electron Zeeman energy in strong magnetic fields ?

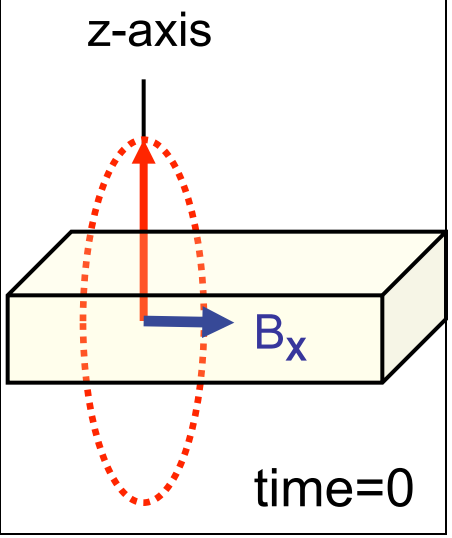
External source: driving laser provides excess energy ➔ **Optical Solid Effect**



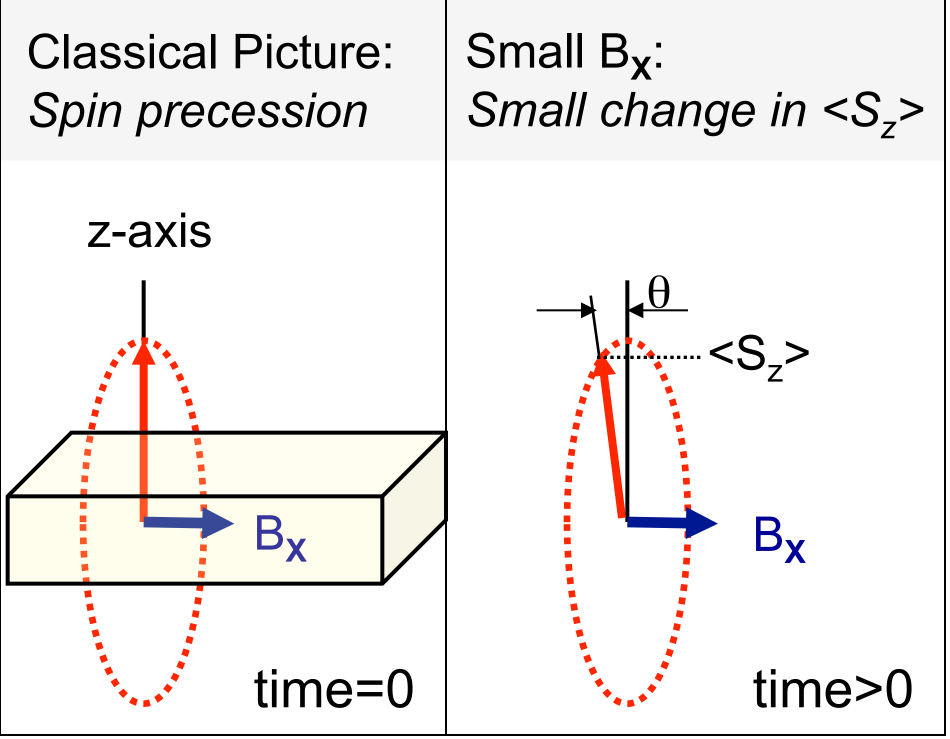
σ+ photon absorption
assisted by
Electron-nuclear spin flip-flop

E. A. Chekhovich, M. N. Makhonin, K. V. Kavokin et al Phys. Rev. Lett. 104 (2010)

Classical Picture:
Spin precession



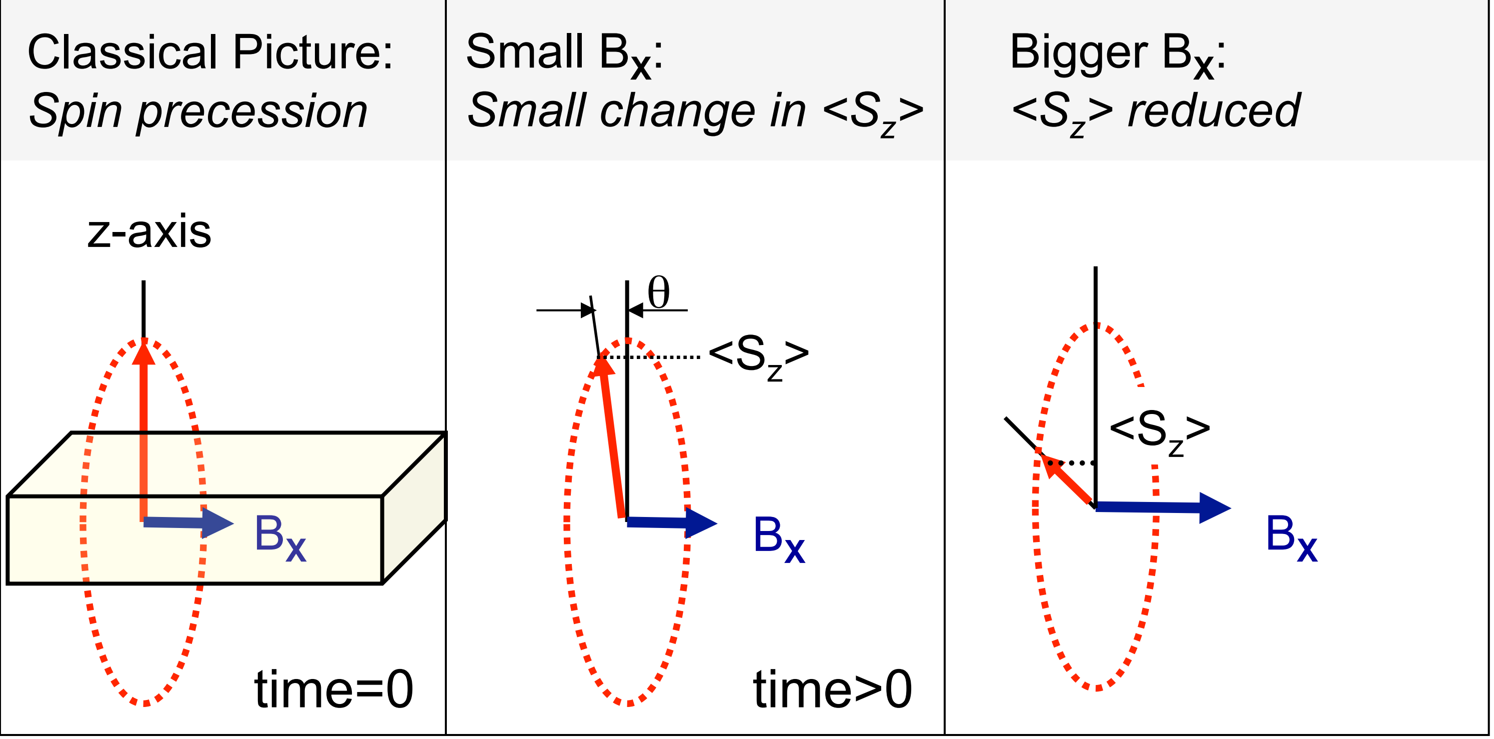
Electron spin precession in transvers magnetic field



Classical Picture:
Spin precession

Small B_x:
Small change in $\langle S_z \rangle$

Electron spin precession in transvers magnetic field



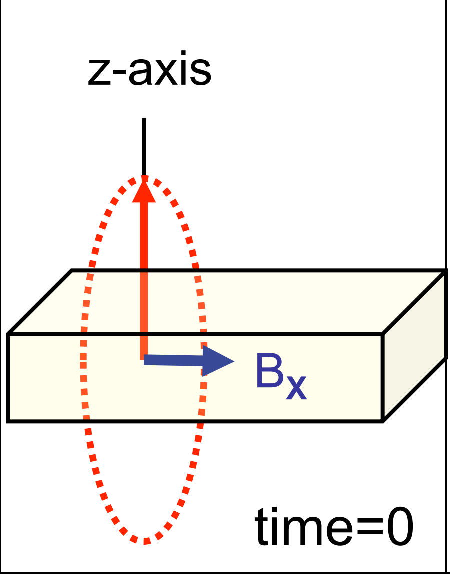
Classical Picture:
Spin precession

Small B_x:
Small change in $\langle S_z \rangle$

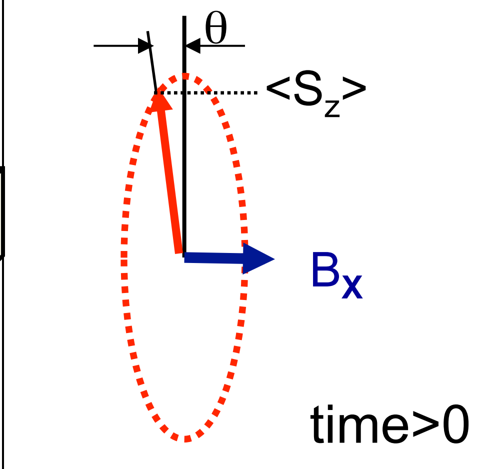
Bigger B_x:
$\langle S_z \rangle$ reduced

Electron spin precession in transvers magnetic field

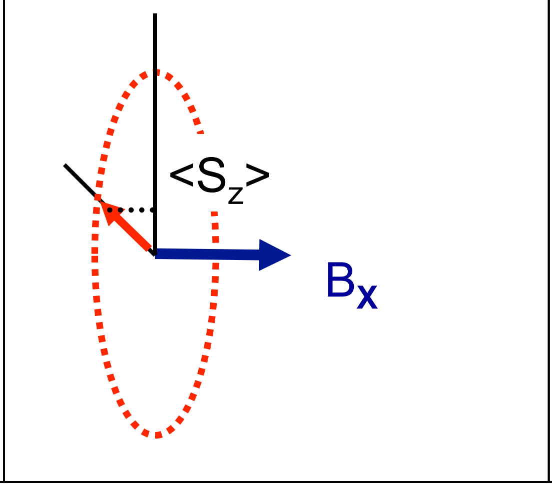
Classical Picture:
Spin precession



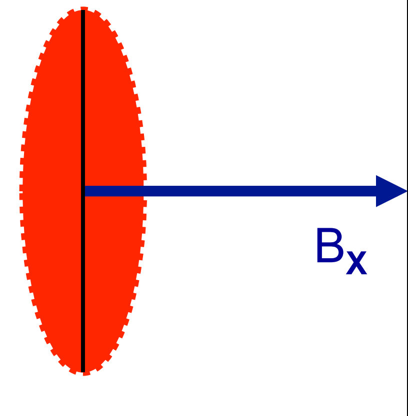
Small B_x:
Small change in $\langle S_z \rangle$



Bigger B_x:
$\langle S_z \rangle$ reduced

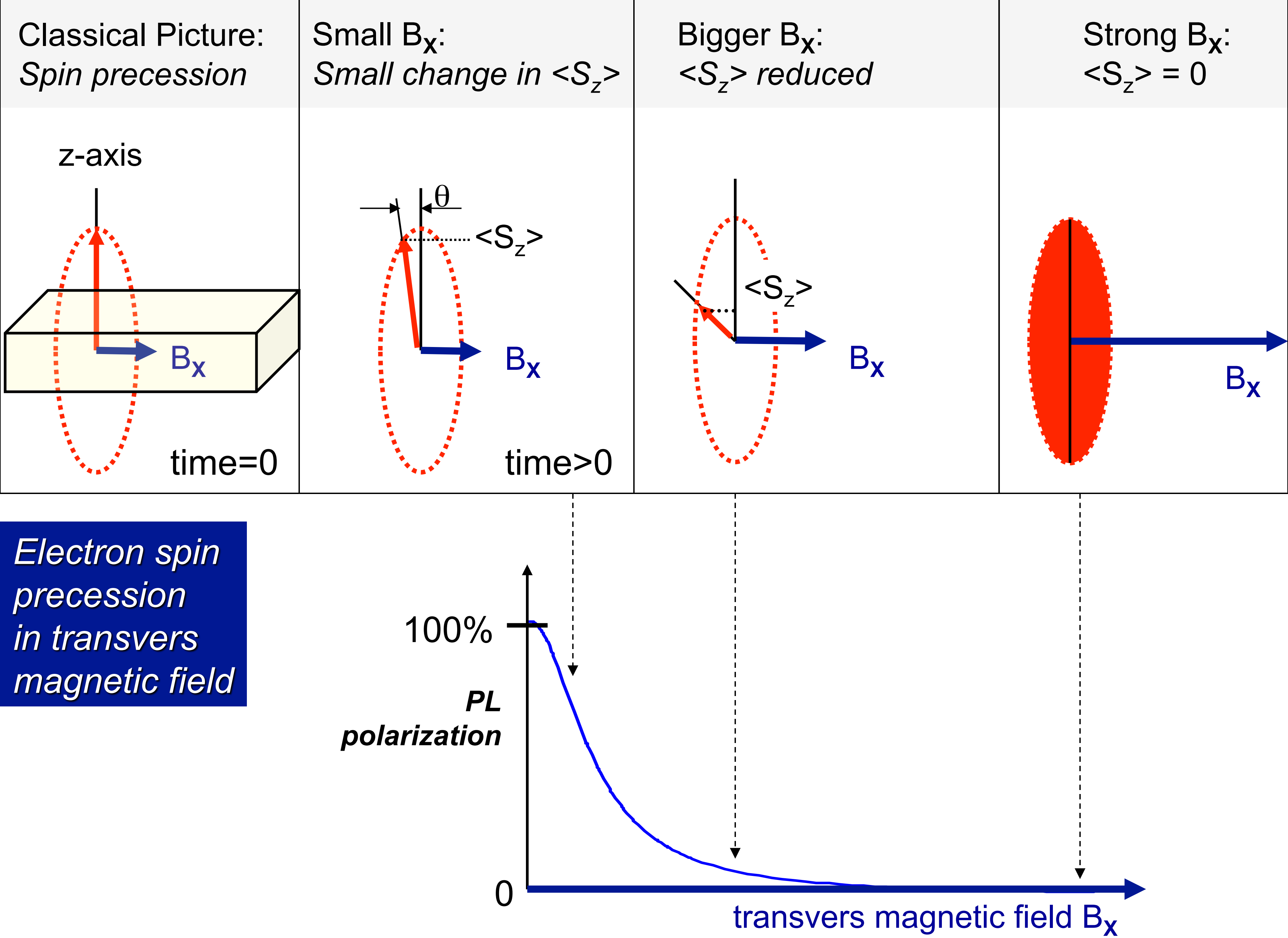


Strong B_x:
$\langle S_z \rangle = 0$



Electron spin precession in transvers magnetic field

Electron spin precession in transvers magnetic field



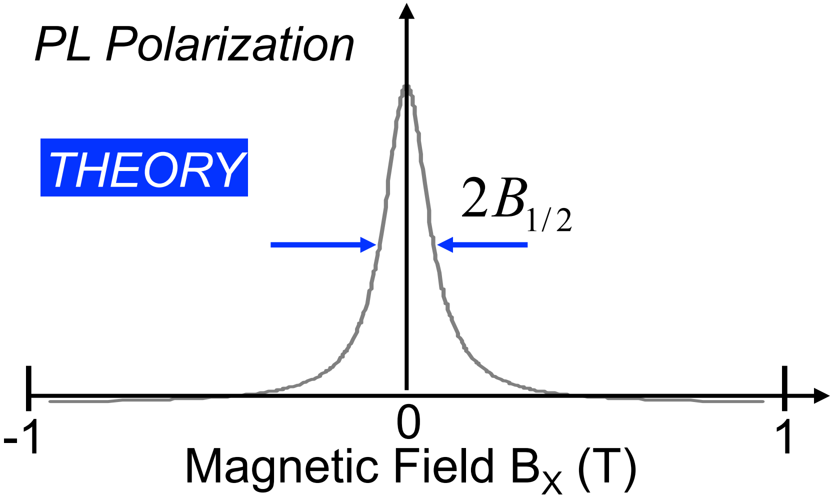
Classical Picture: *Spin precession*

Small B_x: *Small change in $\langle S_z \rangle$*

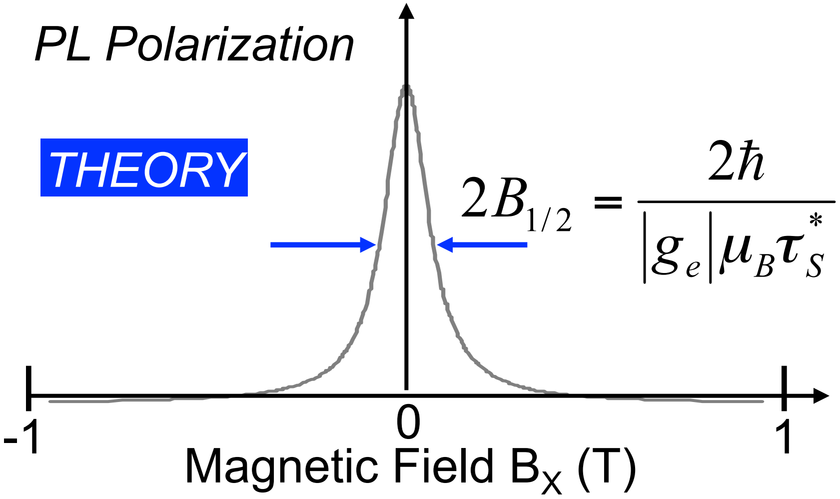
Bigger B_x: *$\langle S_z \rangle$ reduced*

Strong B_x: $\langle S_z \rangle = 0$

Electron spin precession: *Hanle effect*



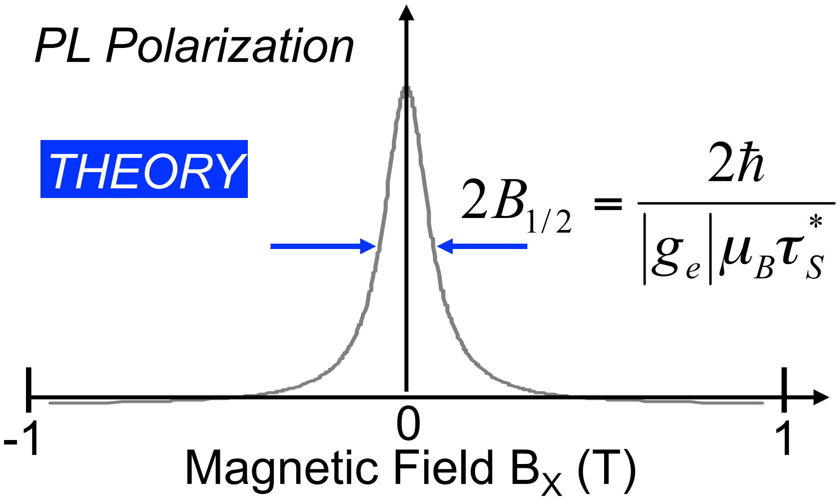
Electron spin precession: *Hanle effect*



$$2B_{1/2} = \frac{2\hbar}{|g_e|\mu_B\tau_S^*}$$

measure electron spin relaxation time τ_s^*

Electron spin precession: *Hanle effect*



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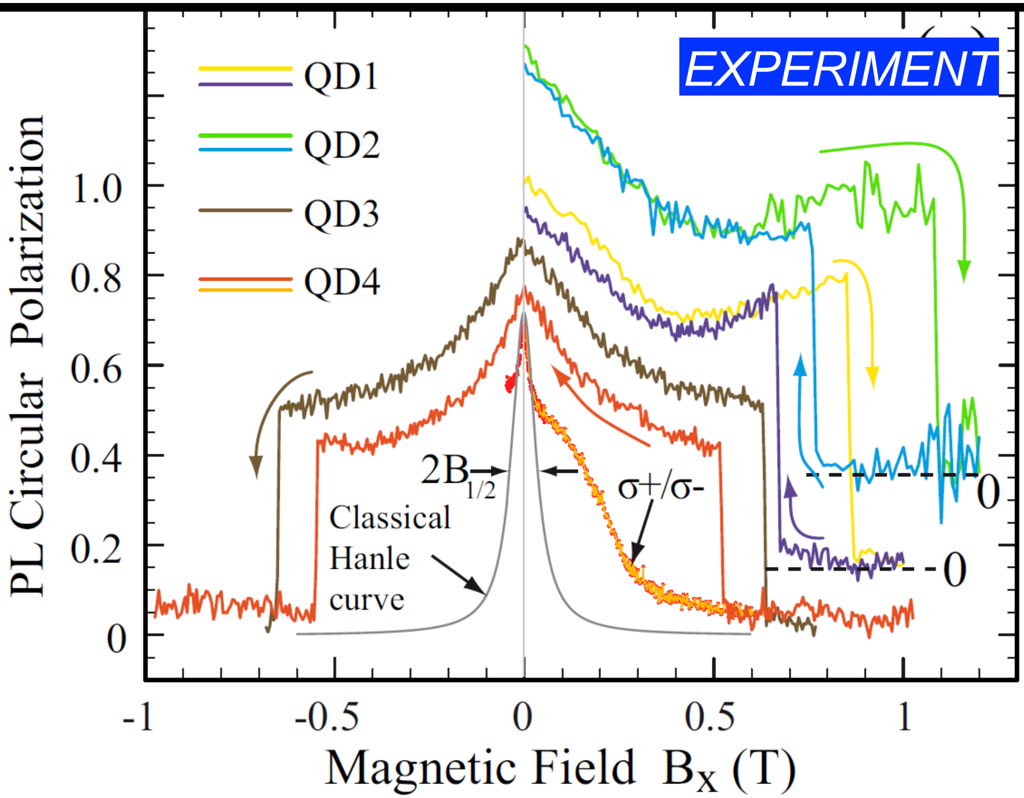
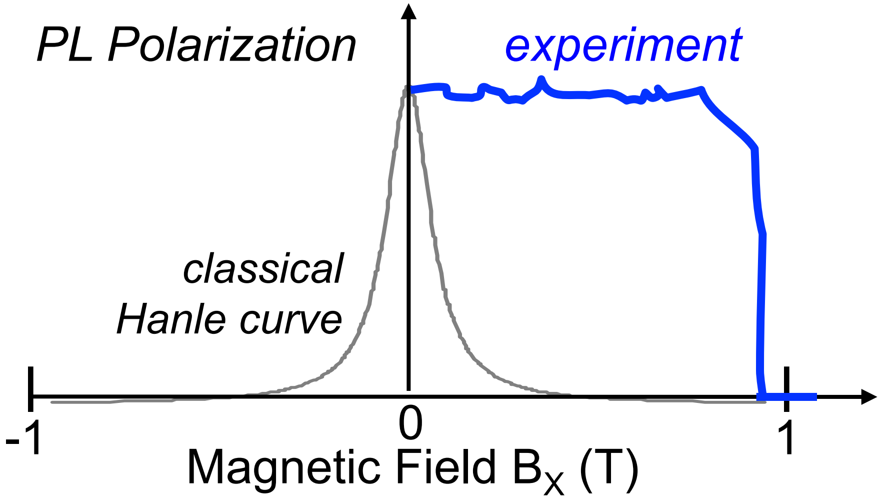
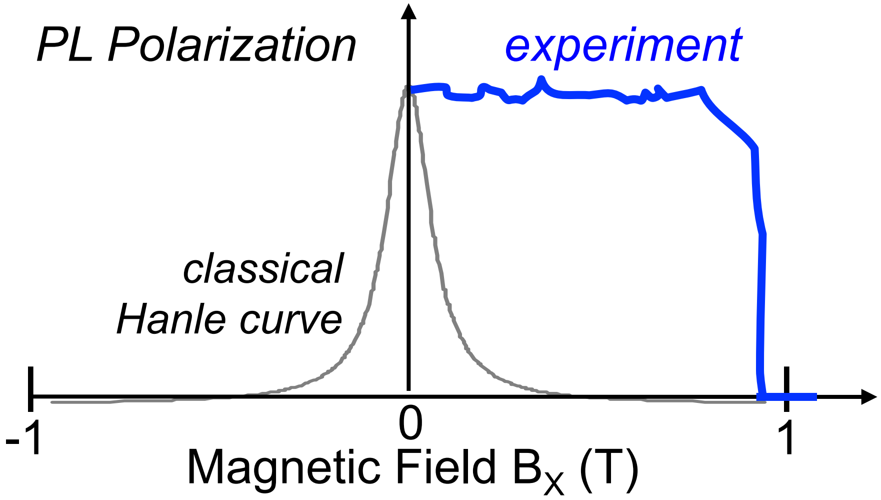


Anomalous Hanle effect

Krebs *et al* Phys.Rev.Lett. 2010

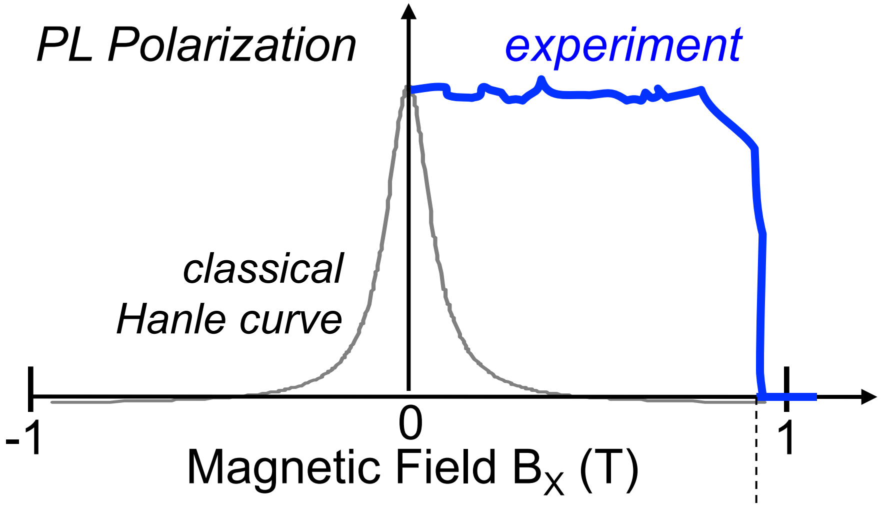
Dzhioev & Korenev, Phys. Rev. Lett. 2007


PL Polarization
experiment
classical
Hanle curve
-1
0
1
Magnetic Field B_X (T)



Electron feels 2 magnetic fields:
(1) applied field $\mathbf{B_X}$
(2) nuclear field $\mathbf{B_N}$

if B_N cancels out B_X:
- *no spin precession*
- *no electron Zeeman splitting*



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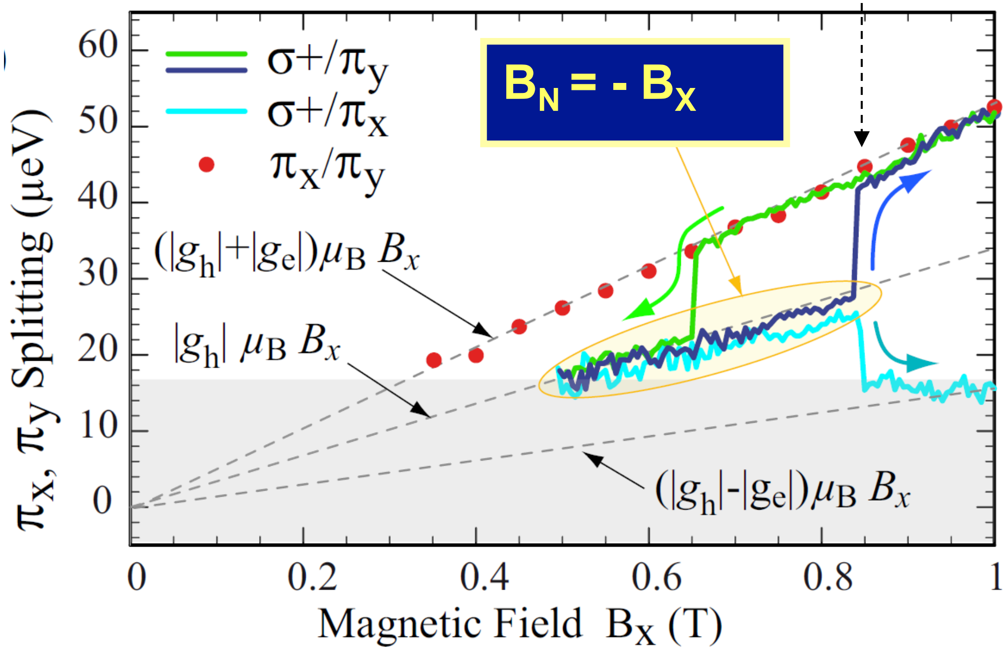

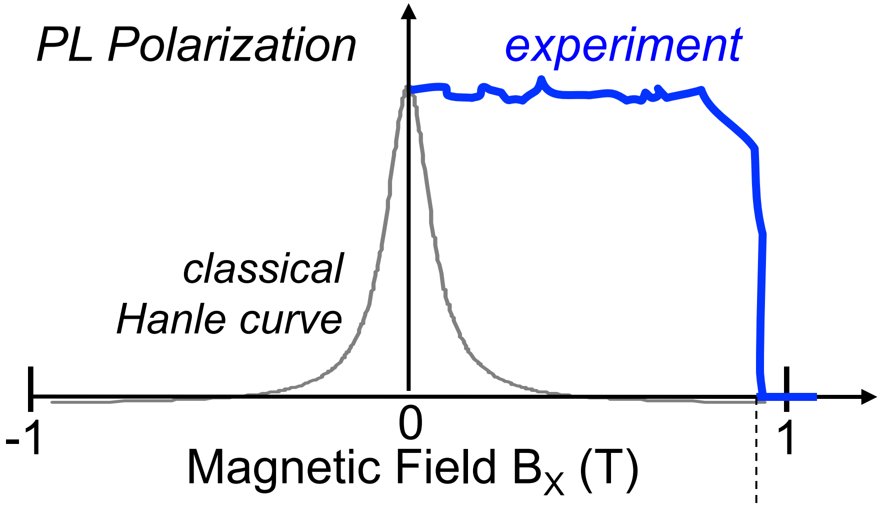

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- *no spin precession*
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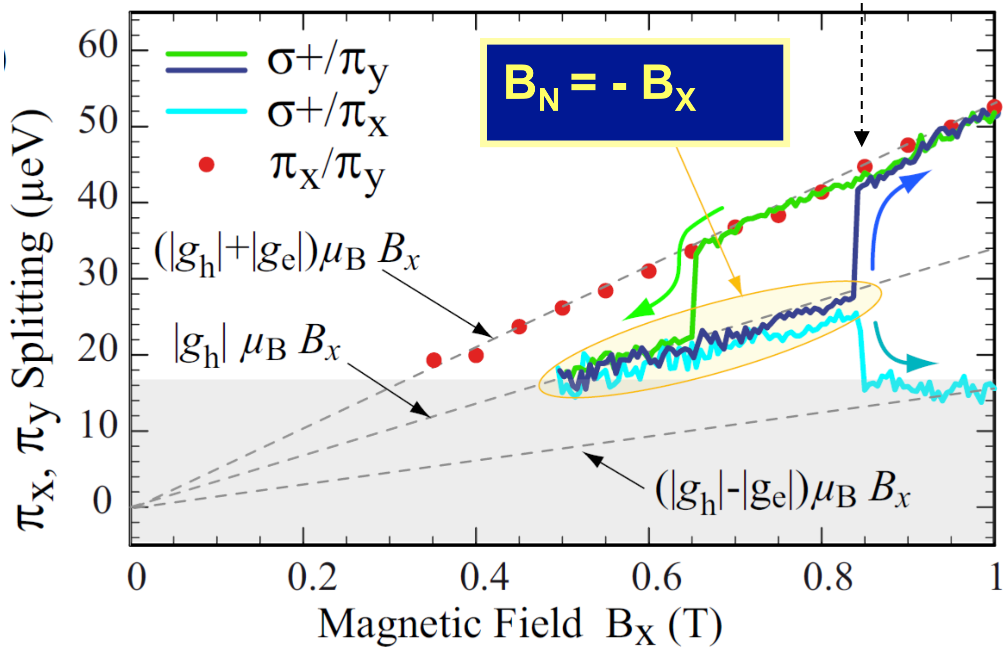
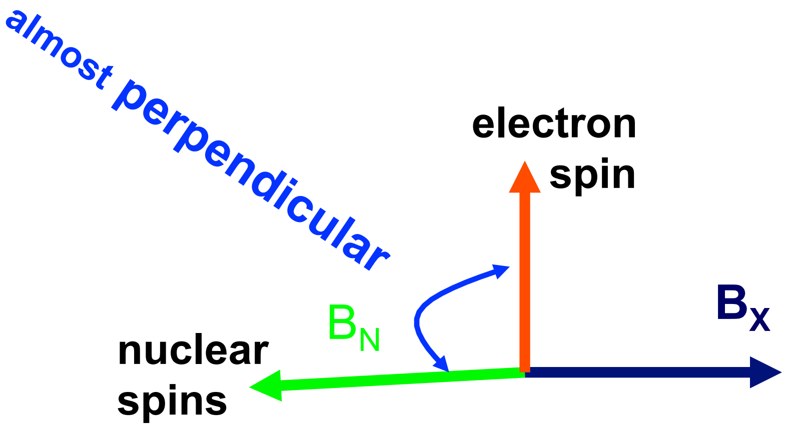
Experimental confirmation:
$\mathbf{B_N = - B_X}$ up to 1 Tesla

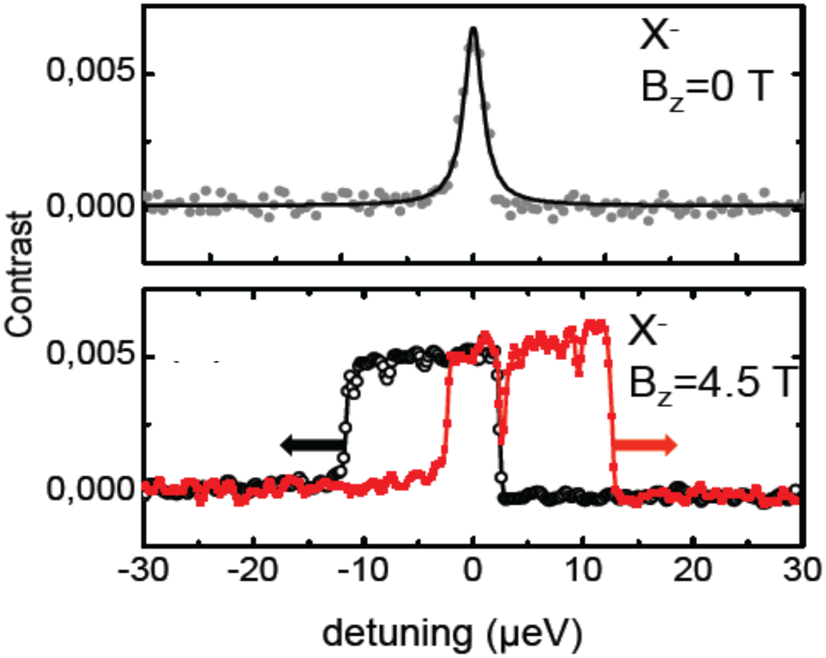


Krebs *et al* Phys.Rev.Lett. 2010



InGaAs dots in GaAs

Differential transmission



Latta et al., Nature Phys. (2009)
Högele et al, PRL (2012)

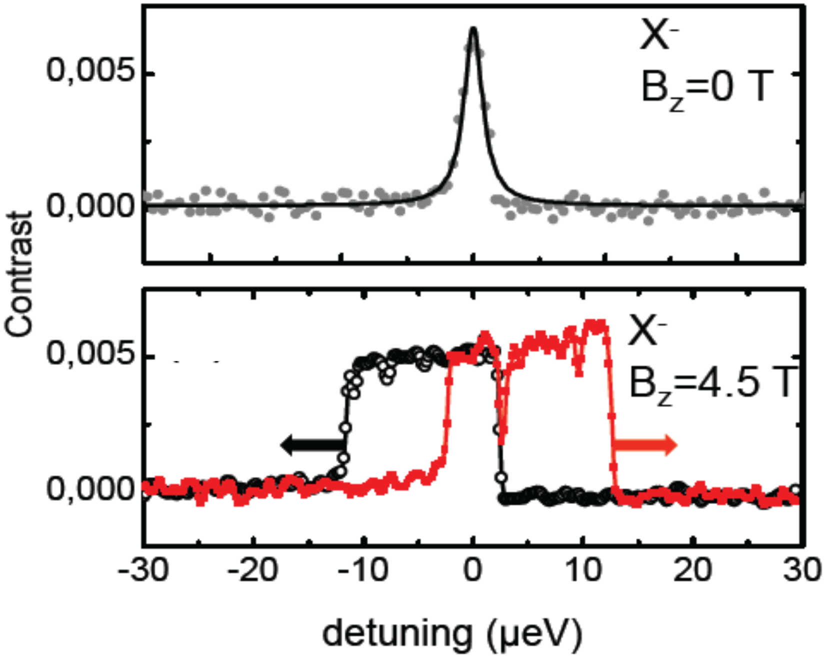
Locking of Quantum Dot transition to Laser Excitation: *‘Dragging’*

talk Martin Kroner at 11am

How can we build up nuclear polarization in high magnetic fields ?

InGaAs dots in GaAs

Differential transmission



Latta et al., Nature Phys. (2009)
Högele et al, PRL (2012)

Locking of Quantum Dot transition to Laser Excitation: *'Dragging'*

talk Martin Kroner at 11am

$$\hat{H}_{hf} = \frac{\nu_0}{2}\sum_j A^j|\psi(\bar{r}_j)|^2 \left(2\hat{I}_z^j\hat{S}_z^e + \cancel{[\hat{I}_+^j\hat{S}_-^e + \hat{I}_-^j\hat{S}_+^e]}\right)$$

Electron – Nuclear spin flip-flops too costly in energy.

1. NEW Non-collinear Hyperfine interaction

$$\boxed{\hat{H}_{hf}^{nc} = \sum_i A_{\text{nc}}^i \hat{I}_x^i \hat{S}_z^e}$$

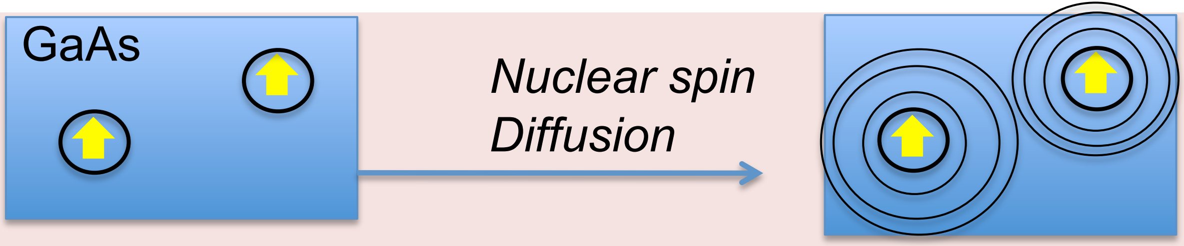
Origin:
Nuclear Quadrupole Interaction

+ 2. optical detuning

= bi-directional Nuclear Polarization

1 electron Spin Interaction with a mesoscopic nuclear spin ensemble

Electrons bound to Donors



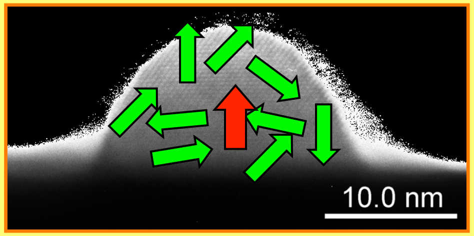
Nuclear spin Diffusion

Optical Orientation, edited by Meier and Zakharchenya (1984)

1 Electron in a single dot →

Different experimental techniques and new Physics: *a small selection*

Rev. Mod. Phys. arXiv1202.4637



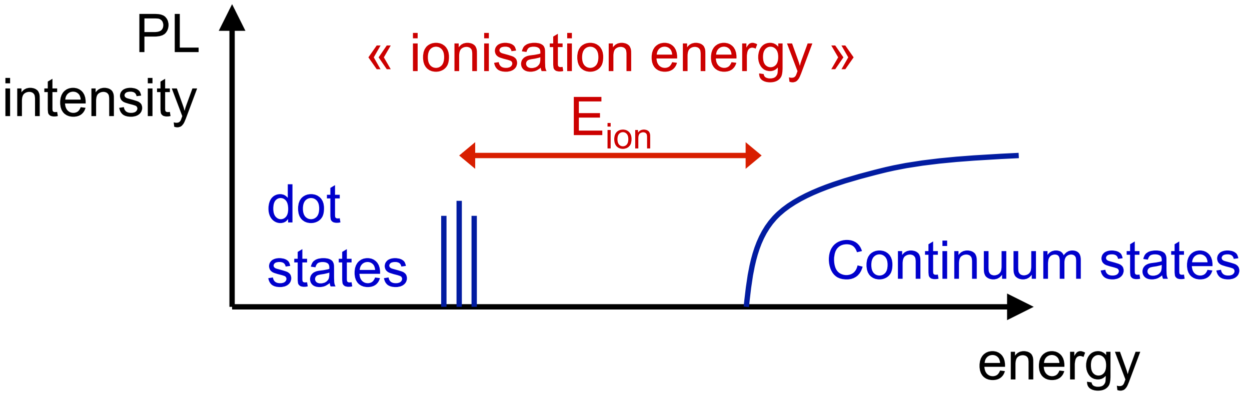
…

strong strain = strong nuclear quadrupolar effects *(talk E.A. Chekhovich)*

- strong suppression of spin diffusion
- **D**ynamic **N**uclear **P**olarization at zero magnetic field
- Anomalous Hanle Effect
- Line dragging in Absorption …. *(talk Martin Kroner)*

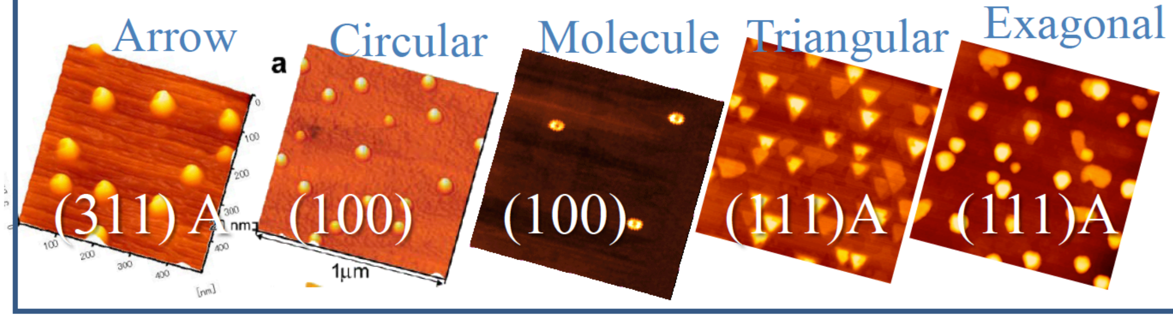
We need strain free dots !

Target: *Spin Physics in unstrained dots*

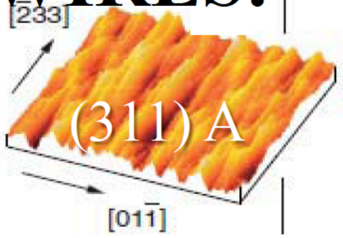


quantum dot system	**strained**	E_{ion}	**references**
GaAs/AlGaAs interface fluctuation dots	no	2..12 meV	NRL Washington Dan Gammon, Science 1996
InAs/GaAs SK dots	yes	~140meV	….Williamson et al, PRB 62, 12963 (2000)
GaAs droplet dots in AlGaAs	no	~100 meV	Belhadj et al PRB 78, 205325 (2008)

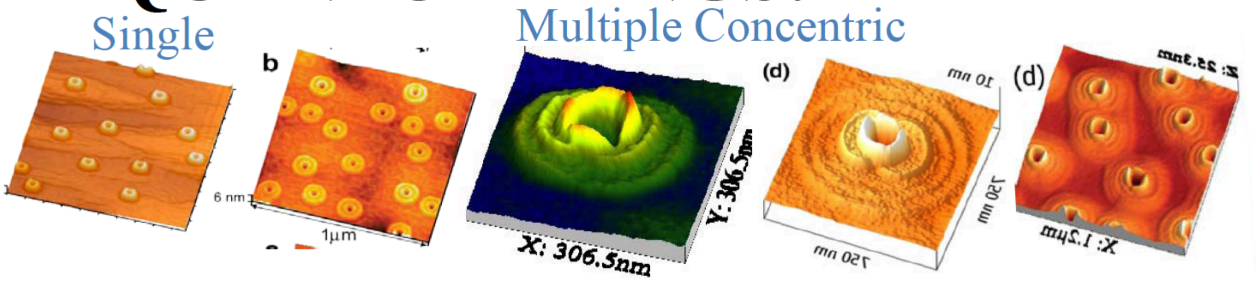
QUANTUM DOTS:



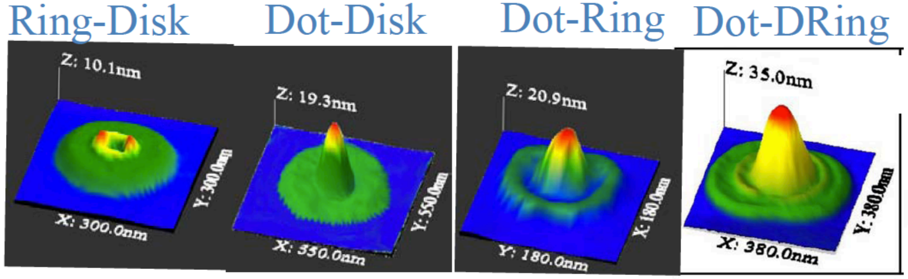
QUANTUM WIRES:



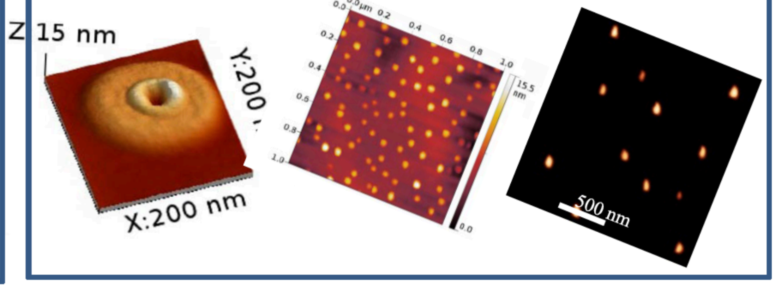
QUANTUM RINGS:



COUPLED NANO-STRUCTURES

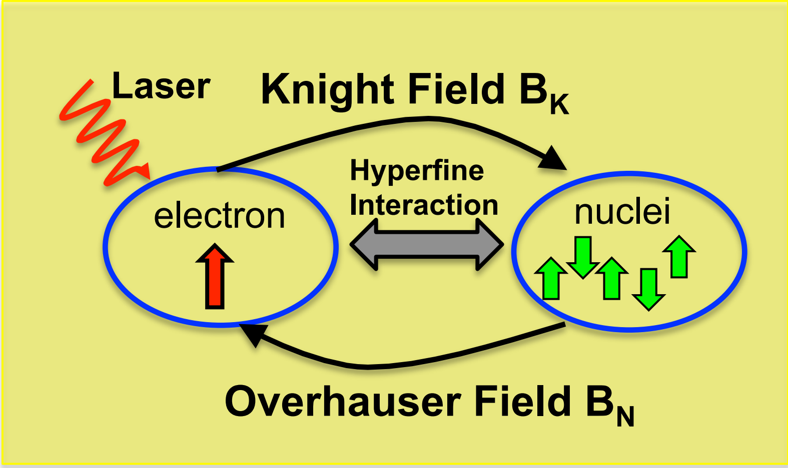


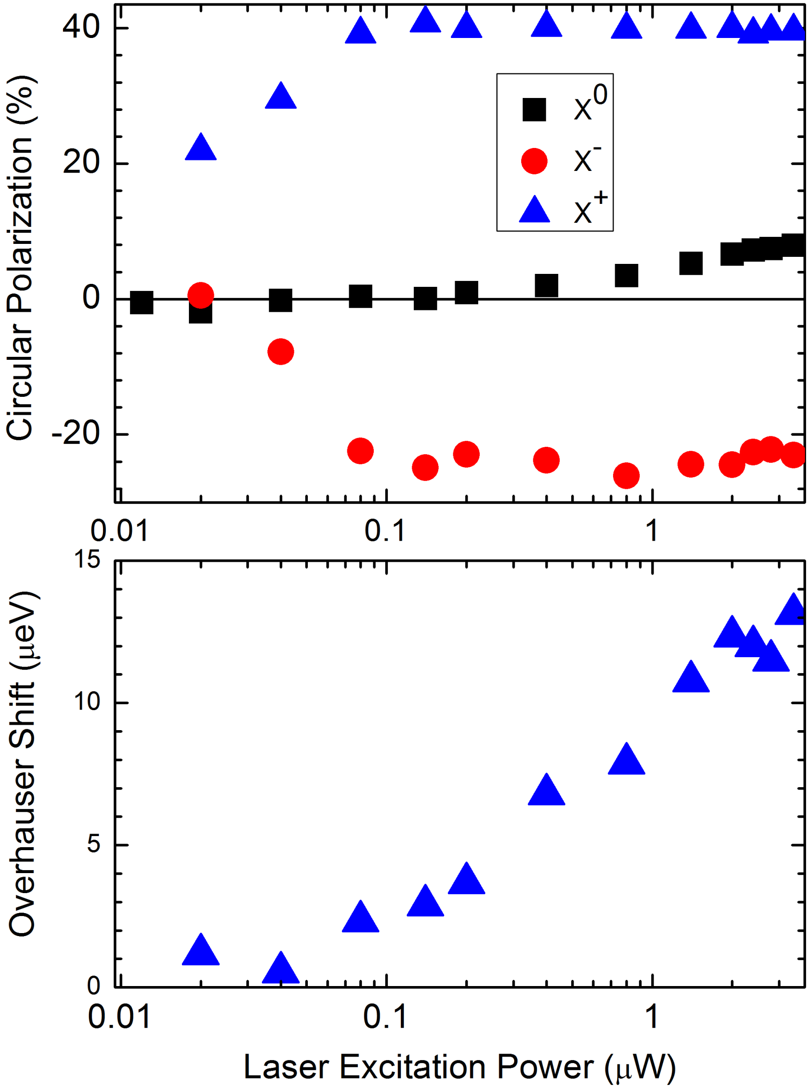
ON Si AND Ge SUBSTRATES



Collaboration M. Abbarchi

Non-resonant excitation: Laser polar. σ^+





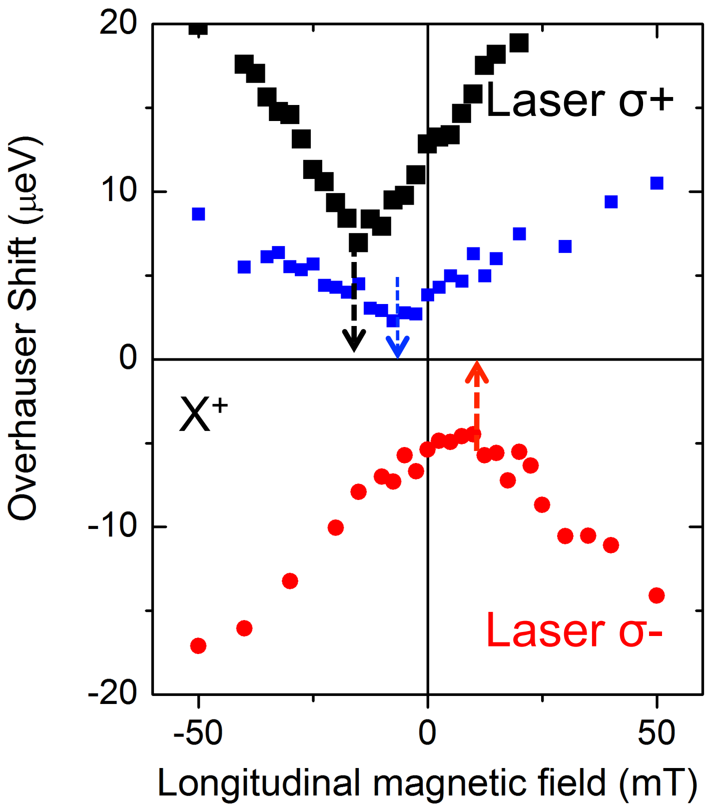
see also:
M. E. Ware et al PRL 2005
S. Laurent et al PRB 2006

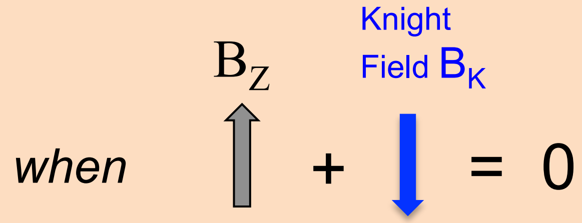
Dynamic **N**uclear **P**olarization at B = 0 ✔

What screens the depolarizing nuclear dipole-dipole interaction ?

- Nuclear Quadrupole Effects ⇔ strain
- Knight field

C. W. Lai et al, PRL 2006
R. I. Dzhioev and V. L. Korenev, PRL 2007
T. Belhadj et al PRL 2009
R. Oulton et al PRL 2007
…

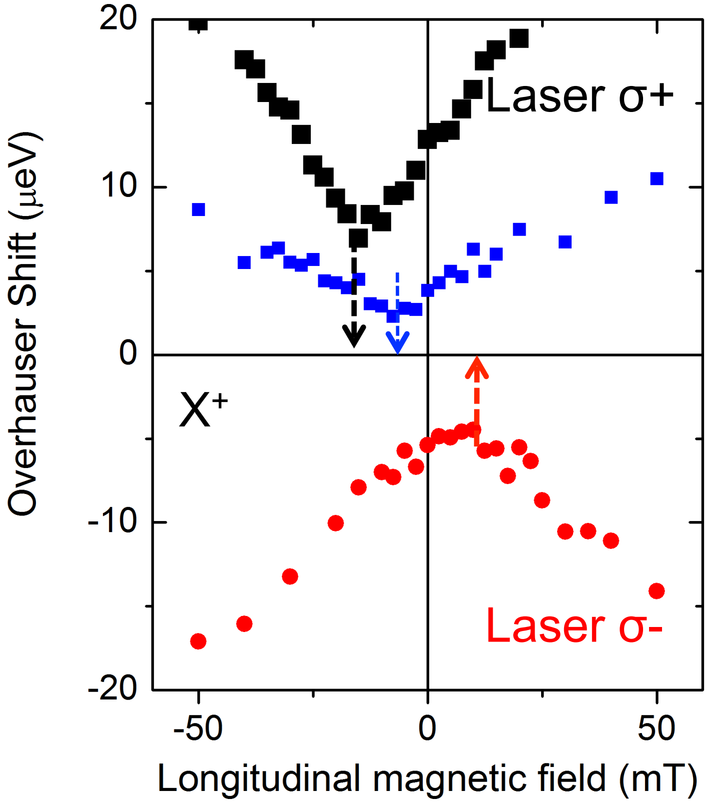




when B_Z + Knight Field B_K = 0

➔ Nuclear Spins get depolarized

see also: Lai et al PRL 2006
Moskalenko et al PRB 2009



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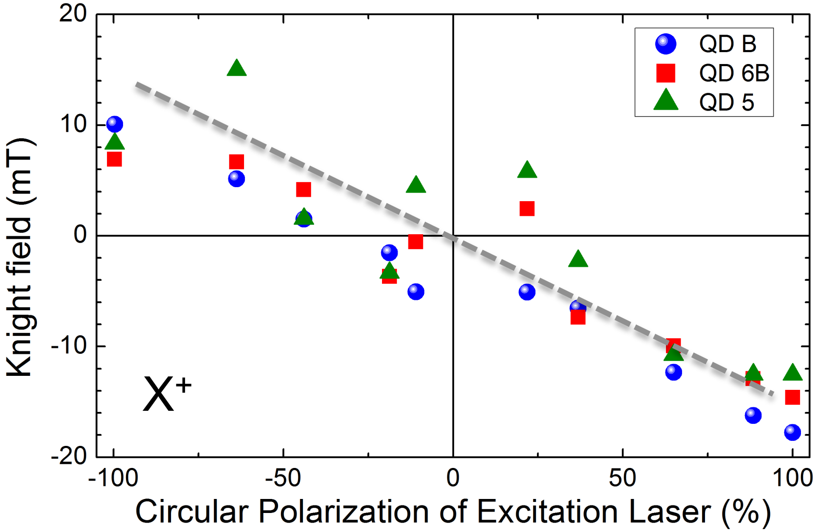
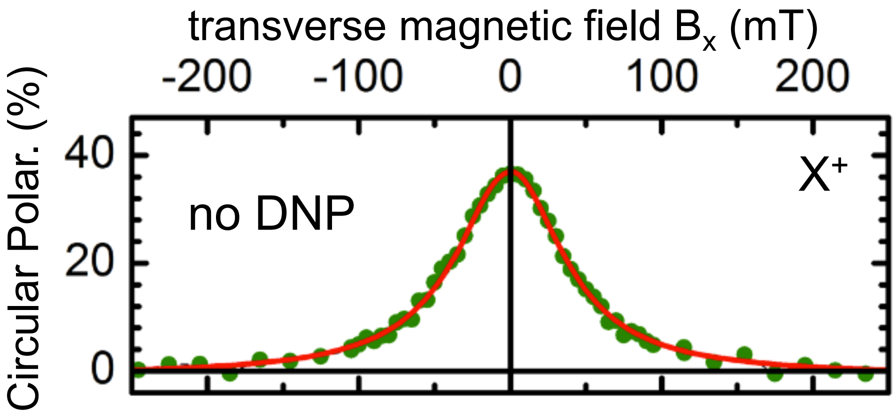


Knight Field for a nucleus *j*

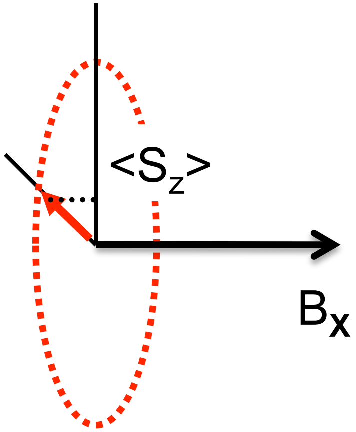
$$B_{Kj} = f_e \frac{\nu_0 A^j}{g_N \mu_N} |\psi(\boldsymbol{r}_j)|^2 \langle \hat{\boldsymbol{S}}^e \rangle$$

can be tuned via laser polarization

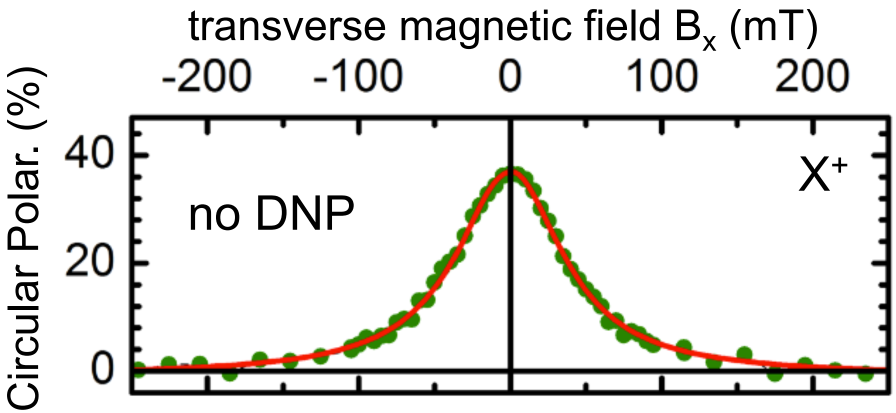


spin precession in transverse magn. field: *Hanle Effect*

$$\tau_s^* = 350 \text{ ps}$$



see also A. Bracker et al, PRL 2005



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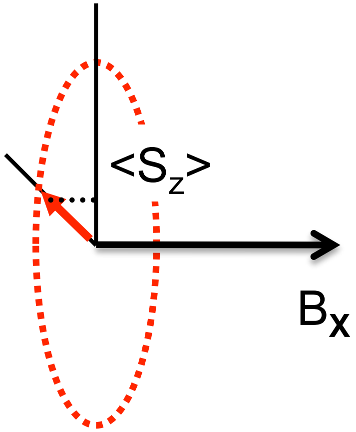

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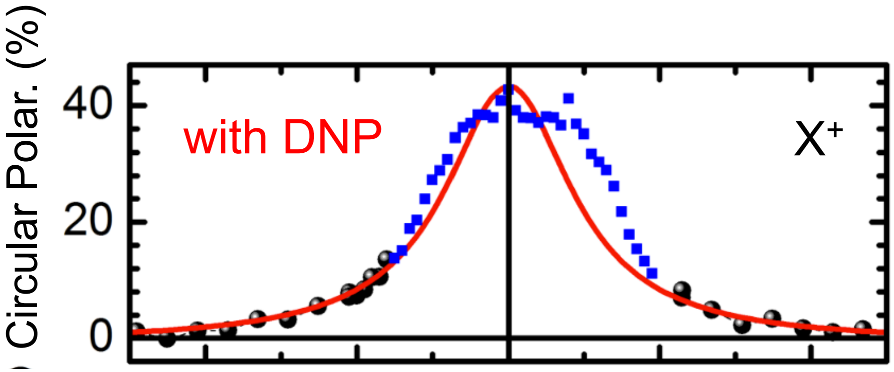
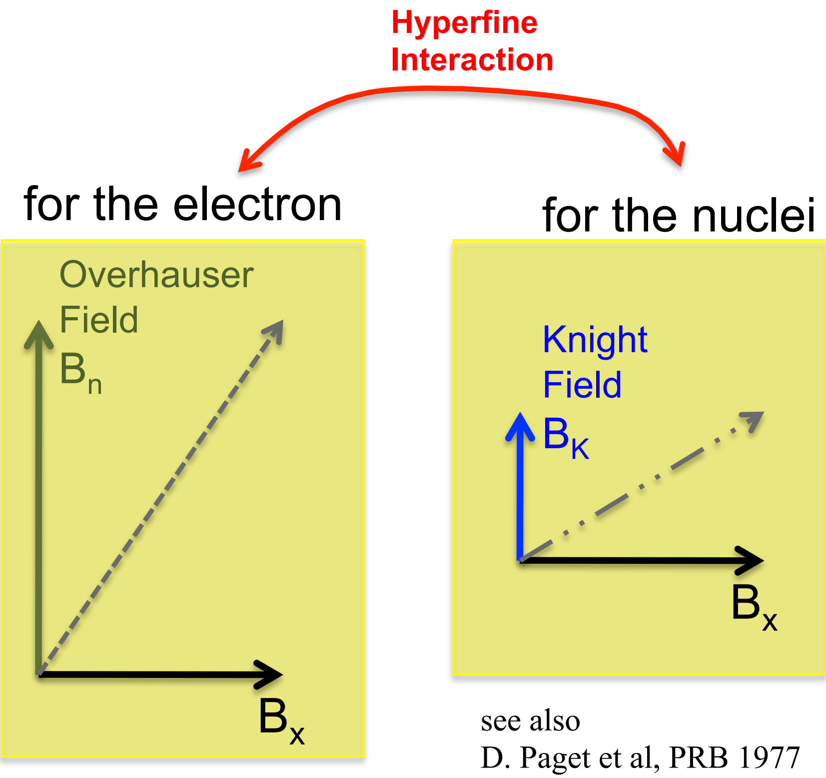


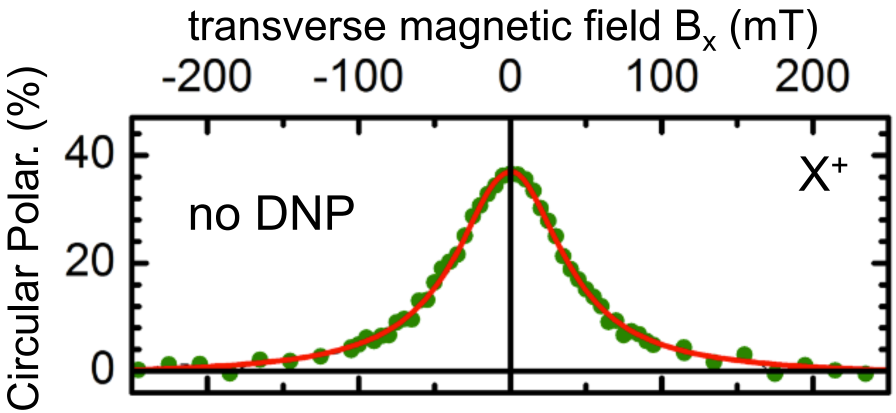


Hyperfine Interaction

for the electron

for the nuclei

see also
D. Paget et al, PRB 1977
O. Krebs et al, PRL 2010



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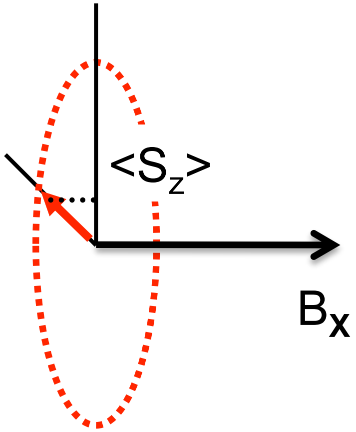

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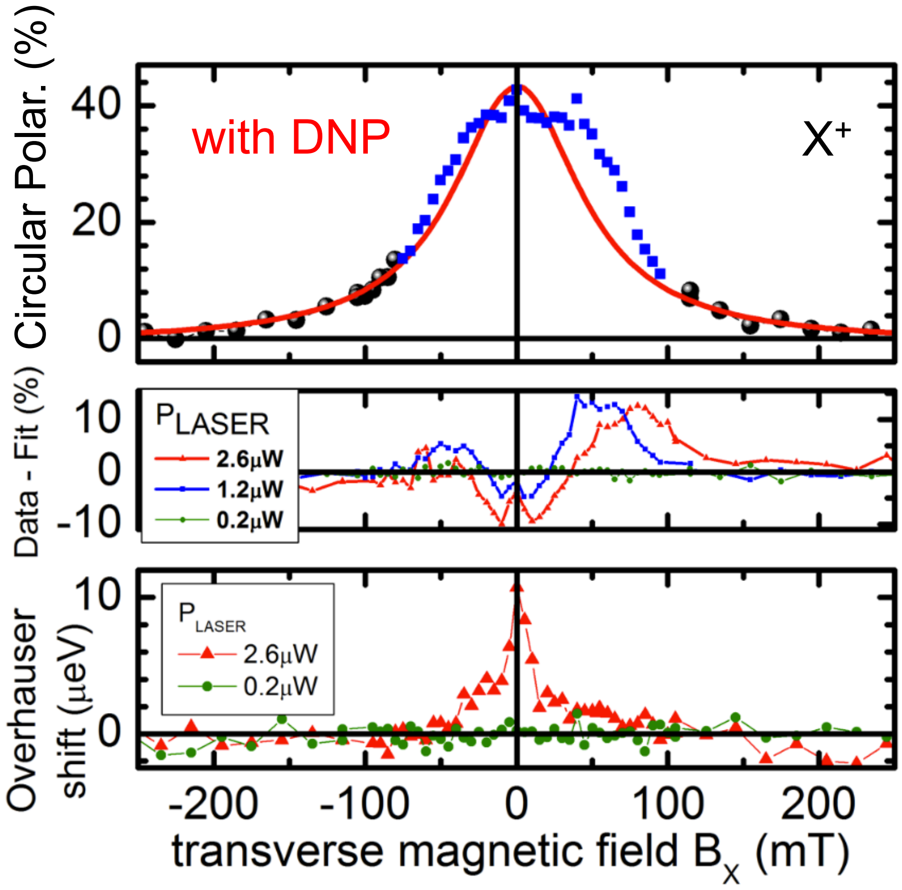

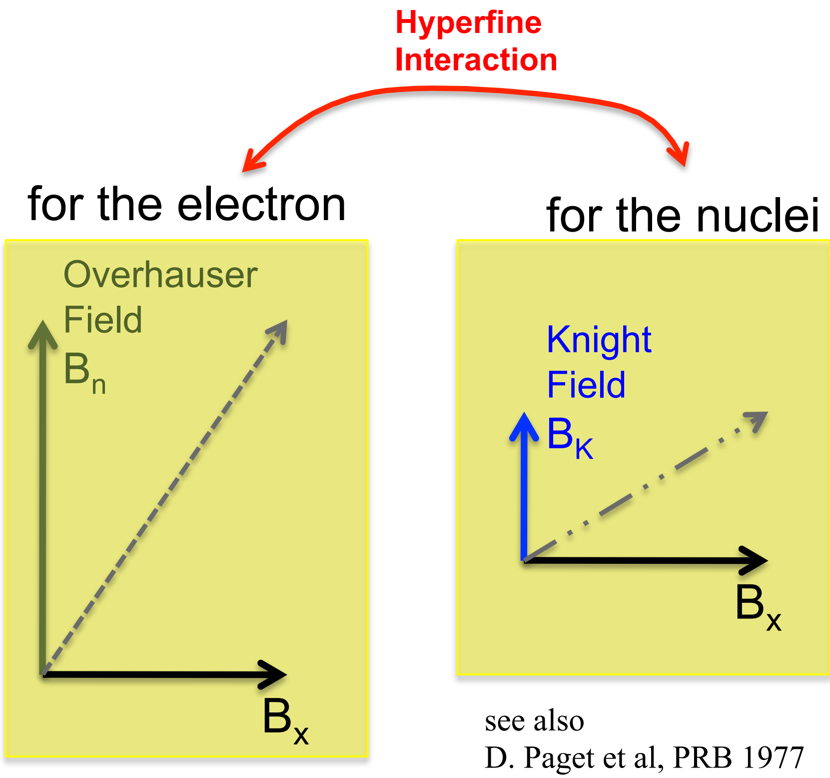

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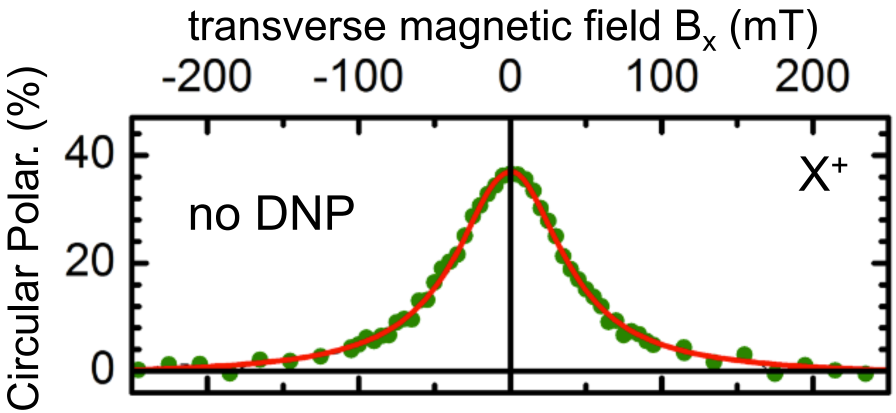

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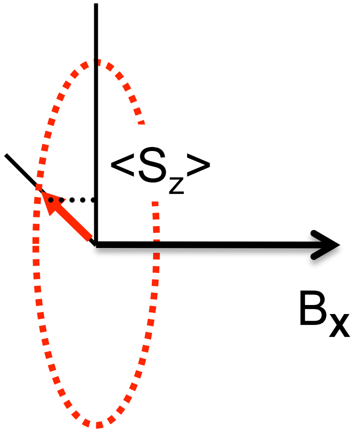

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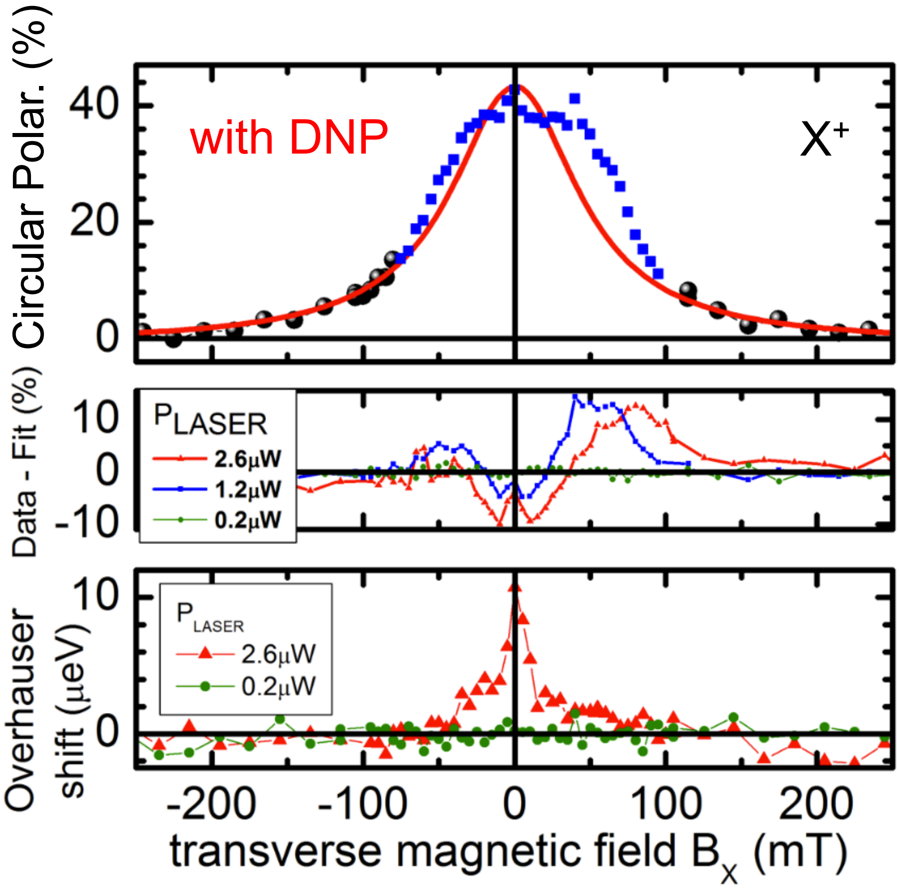
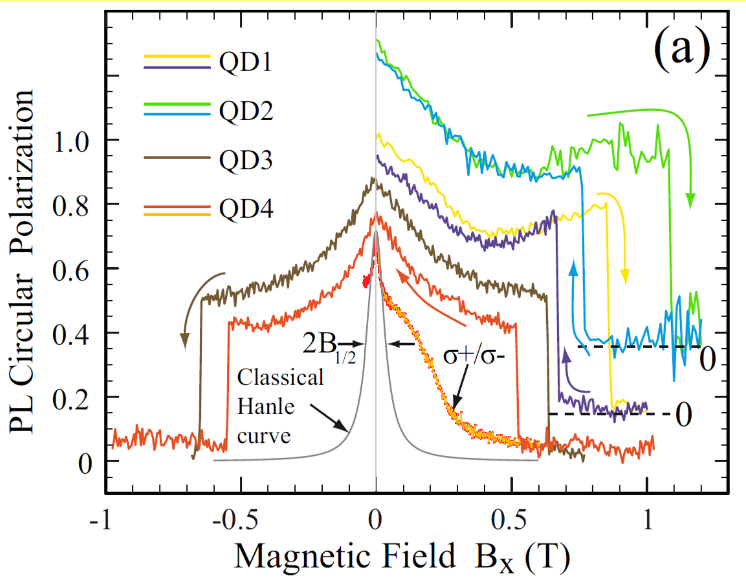


For comparison:
Strained InAs dots



Spin Physics in [111] grown GaAs/AlGaAs dots: *recent results*

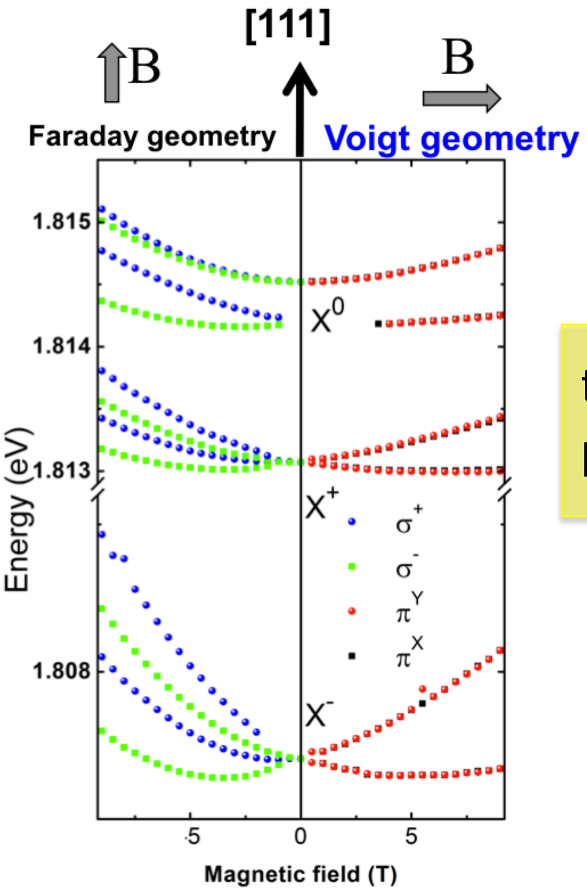
Heavy hole mixing

due to C_{3V} symmetry

Λ system for
coherent hole spin control

X^0 bright splitting changes sign

G. Sallen et al, PRL 2011

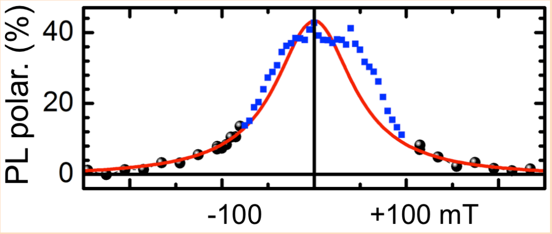


transverse

heavy-hole g-factor ≈ 0

Electron ⬄ Nuclear spin coupling at low fields

- **D**ynamic **N**uclear **P**olarization at B = 0
- strong, tunable **Knight field** B_K = 15 mT
- **Hanle effect**: build-up of transverse Nuclear Spin Polarization



Bibliography:

1961: *The principle of nuclear magnetism, A. Abragam, Clarendon press*

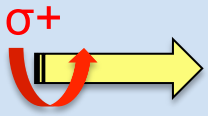
1984: *Optical Orientation, F. Meier and B. Zhakharchenya, North Holland*

2008: *Semicond. Sci. Technol.* ***23*** *(2008) (Special issue about spin)*

2008: *Spin physics in semiconductors, M. Dyakonov, Springer*

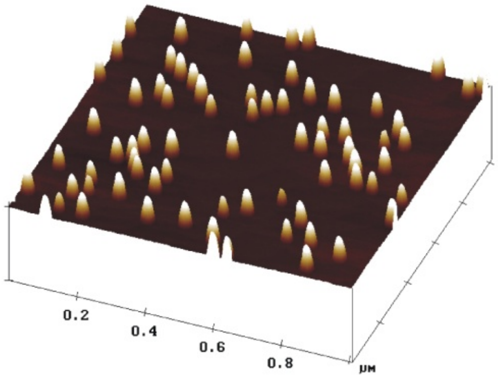
2012: *Nuclear spin physics in quantum dots: an optical investigation*
Rev. Mod. Phys. *in press* arXiv:1202.4637
B. Urbaszek, X. Marie and T. Amand
O. Krebs and P. Voisin
P. Maletinsky, A. Högele, and A. Imamoglu

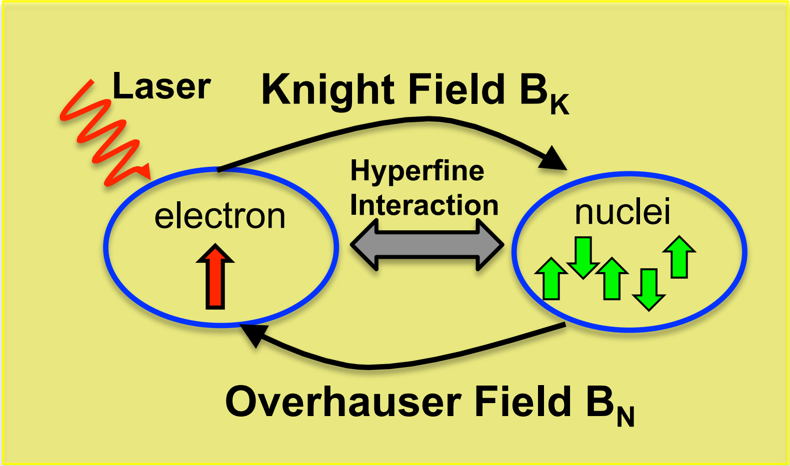
Degrees of Freedom in Semiconductors: *Optical Manipulation*



Main Part :

Optical pumping of **carrier spins** and **nuclear spins** in quantum dots





Laser

Knight Field B_K

Hyperfine Interaction

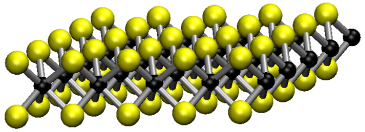
electron

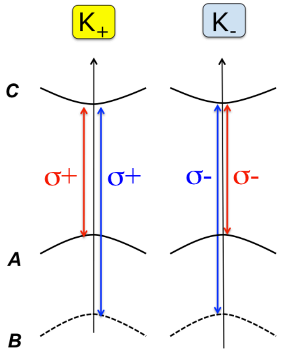
nuclei

Overhauser Field B_N

Outlook:

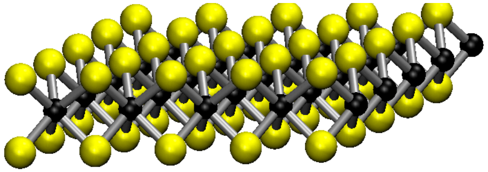
selective **K-valley** excitation in MoS_2 monolayers





arXiv:1206.5128

Robust optical emission polarization in MoS_2 monolayers through selective valley excitation



G. Sallen, L. Bouet, X. Marie, T. Amand, B. Urbaszek
Universite de Toulouse, INSA-CNRS-UPS, LPCNO, France

G. Wang, C.R. Zhu and B.L. Liu
Beijing National Laboratory for Condensed Matter Physics,
Institute of Physics, Chinese Academy of Sciences, China

W.P. Han, Y. Lu and P.H. Tan
State Key Laboratory of Superlattices and Microstructures,
Institute of Semiconductors, Chinese Academy of Sciences, Beijing, China

http://en.wikipedia.org/wiki/Molybdenum_disulfide

MoS_2 **Molybdenum disulfide**

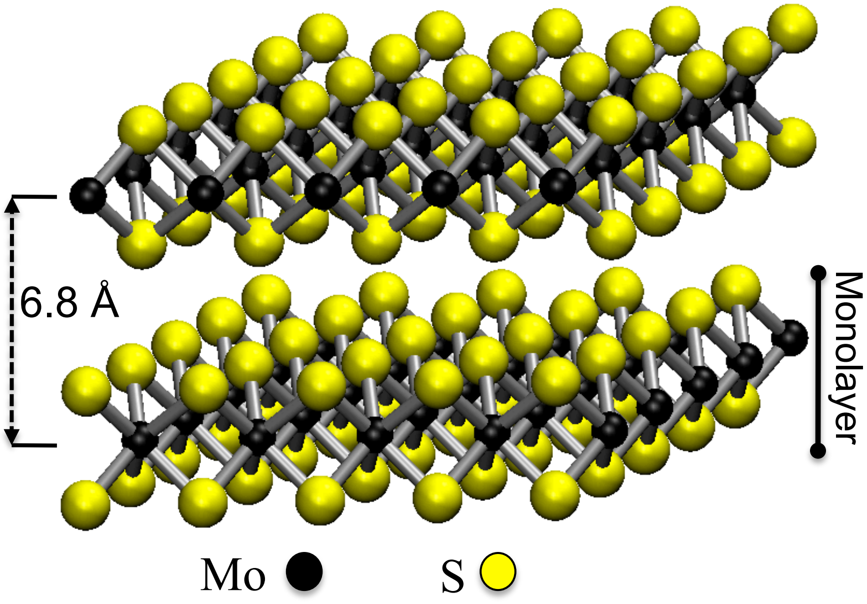
Natural occurance as mineral Molybdenite

current Applications

- lubricant up to 350 °C
- Nylon, Teflon, ski wax
- catalyst in petroleum refineries

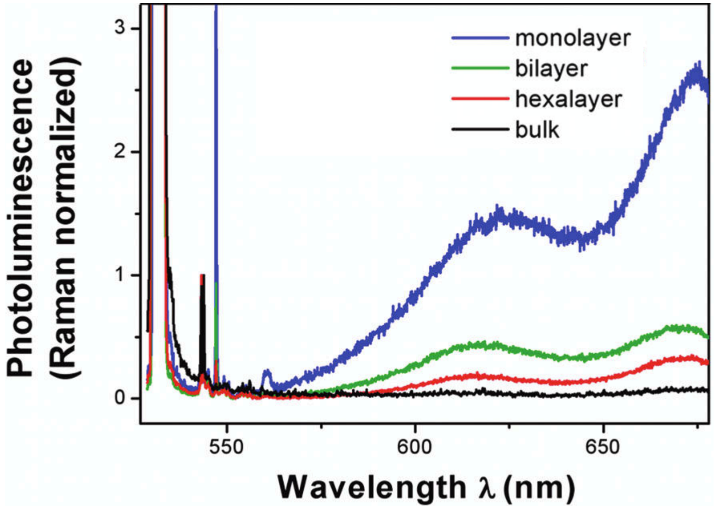
similar to graphite:

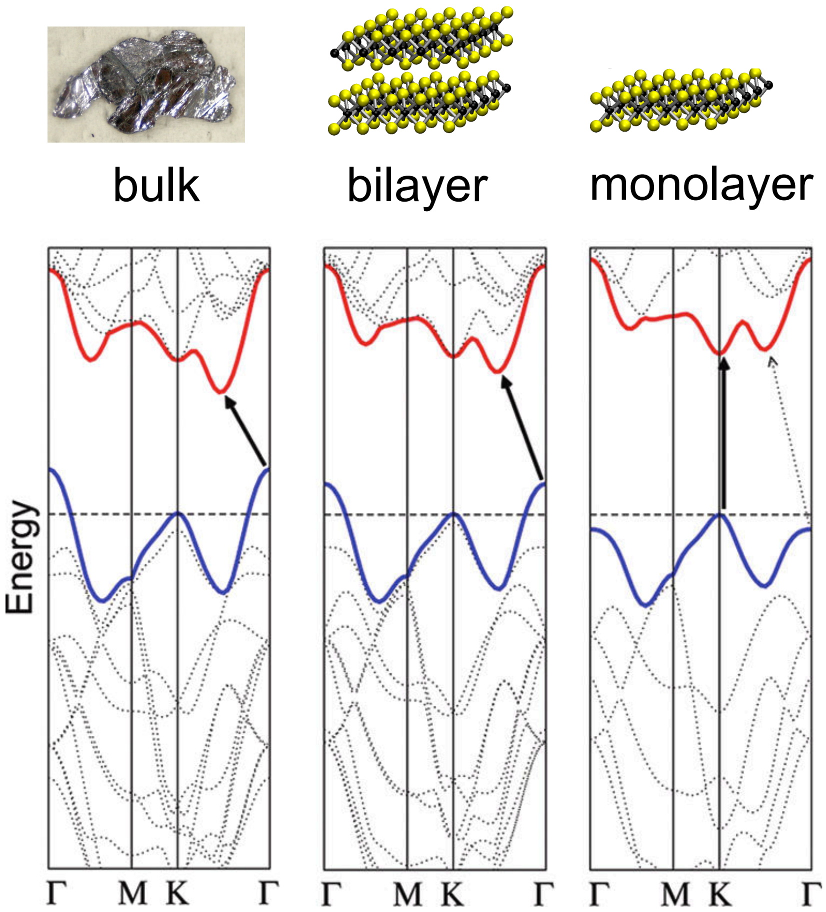
multilayers connected by van der Waals bonding



Semiconductor MoS_2

Bulk MoS_2 : indirect bandgap → 1 Monolayer: direct bandgap

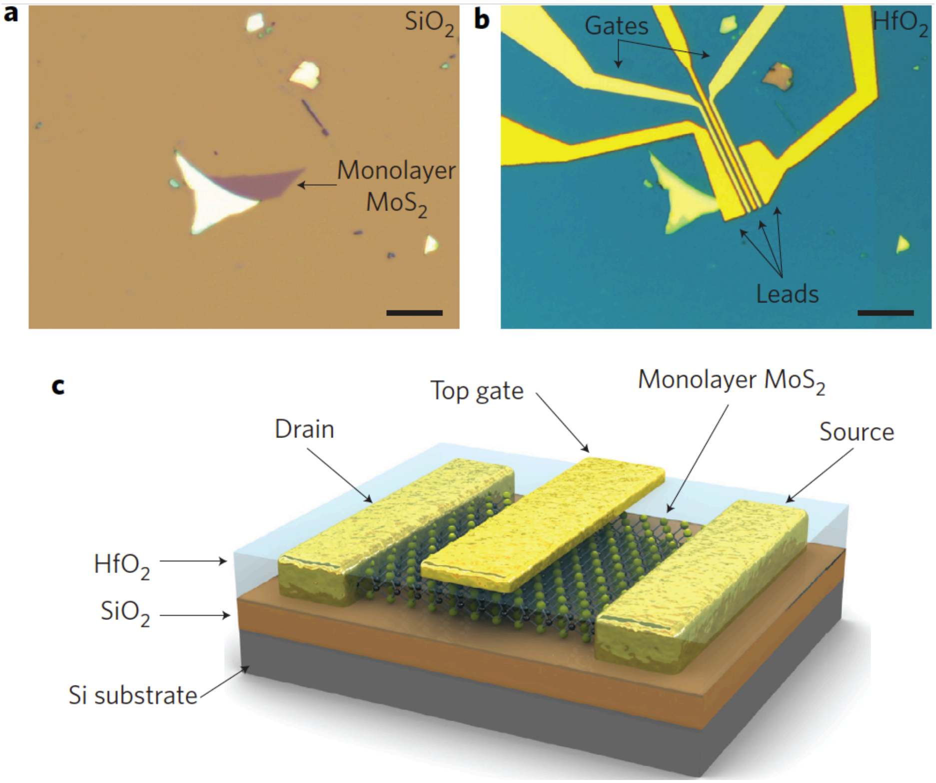




Splendiani, et al, Nano Lett.10, 1271 (2010) & Mak et al, Phys. Rev. Lett. 105, 136805 (2010).

Single-layer MoS_2 transistors

Radisavljevic et al, Nature Nanotech. Vol. 6, p. 147 (2011)

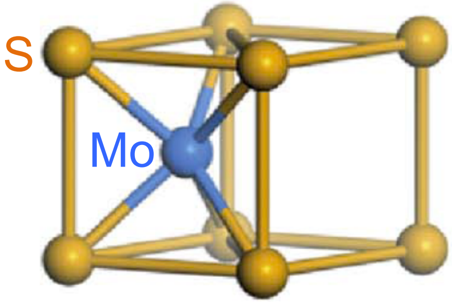


Key parameters:

- MoS_2 direct bandgap 1.8 eV
- Room temperature carrier mobility > 200 $cm^2 V^{-1} s^{-1}$
- transistors with room-temperature current on/off ratios of $1x10^8$

Monolayer MoS_2 ⬄ SpinOptronics Summer School

Theory: Ting Cao et al, Nature Communications ncomms1882 (June 2012)
Di Xiao et al, Phys. Rev. Lett. 108, 196802 (May 2012)



Main difference to Graphene:

- direct bandgap
- broken Inversion symmetry
- strong spin-orbit coupling

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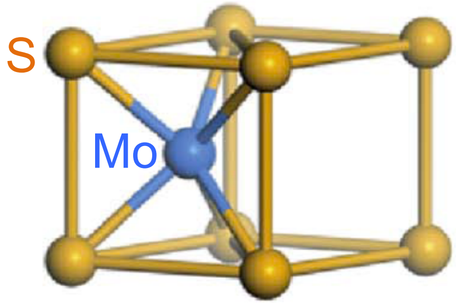
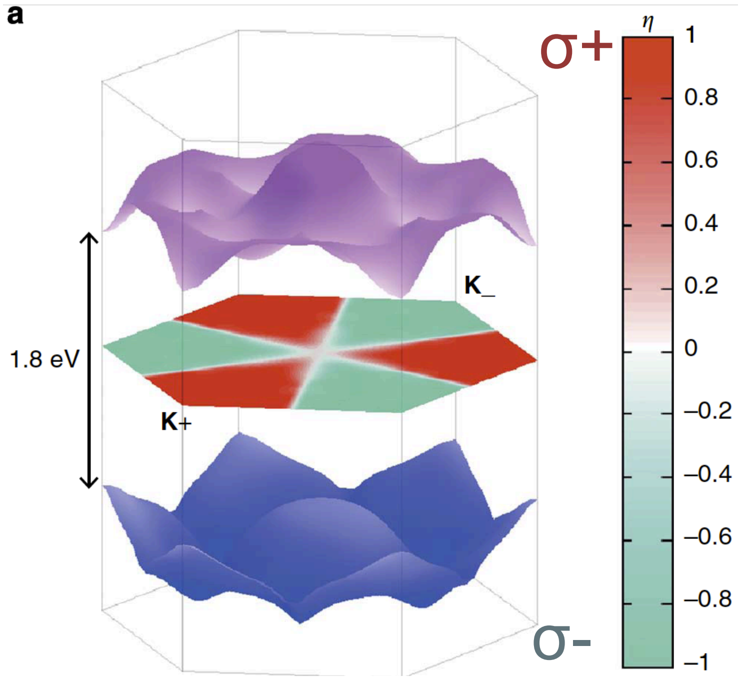




Main difference to <u>Graphene</u>:

- direct bandgap
- broken Inversion symmetry
- strong spin-orbit coupling

Coupled spin and K - valley physics:

Laser Excitation **σ+** ➔ 100% of electrons in K_+ valley

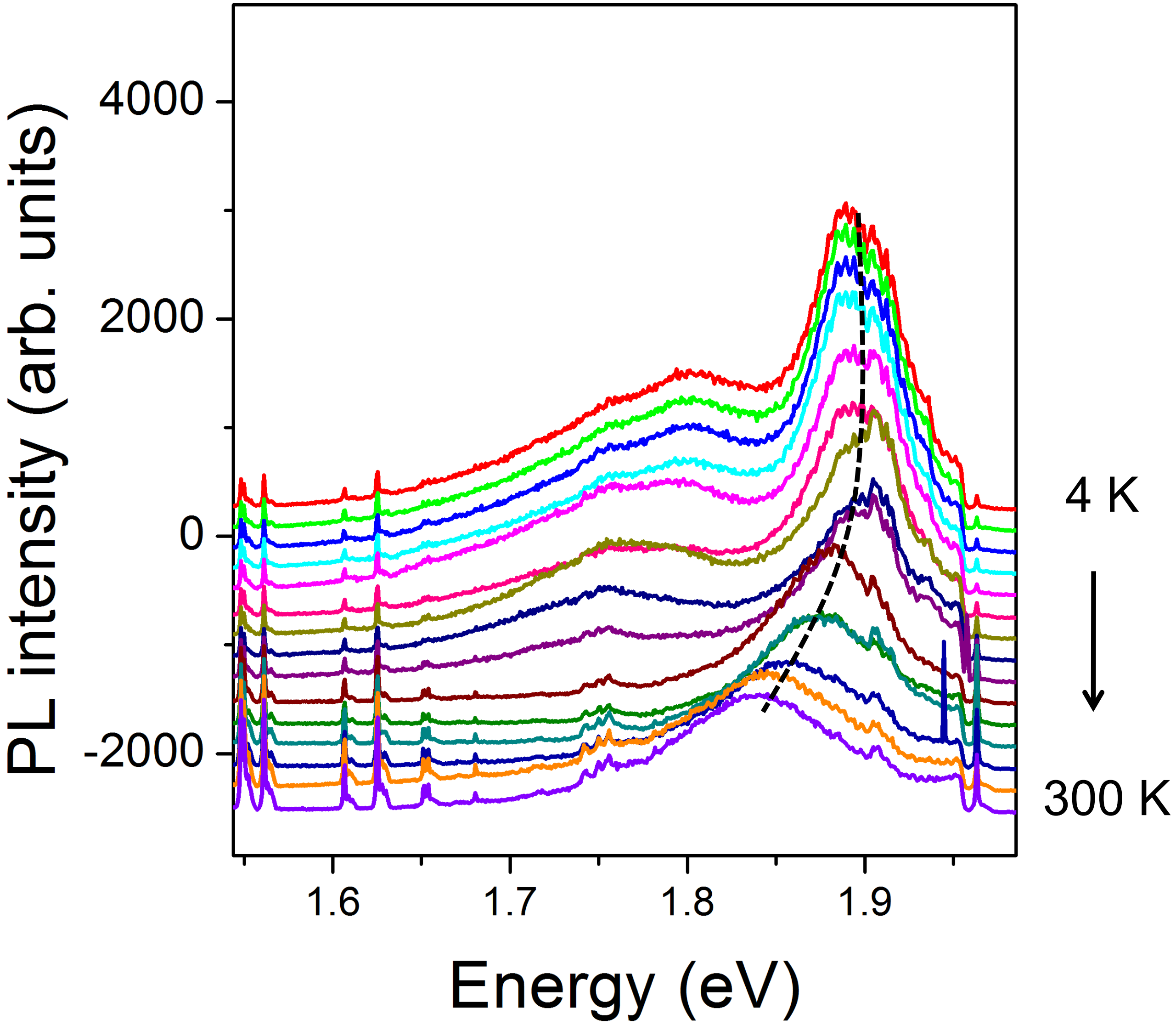
σ- ➔ 100% of electrons in K- valley

Change of valley unlikely !

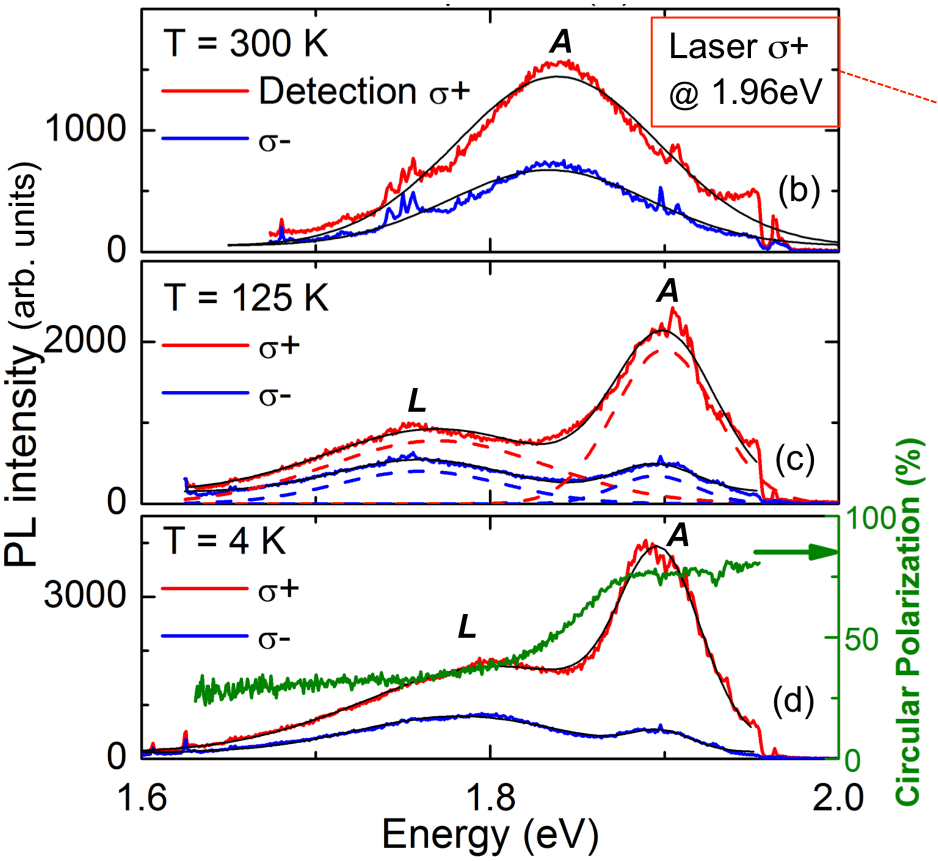
⇩

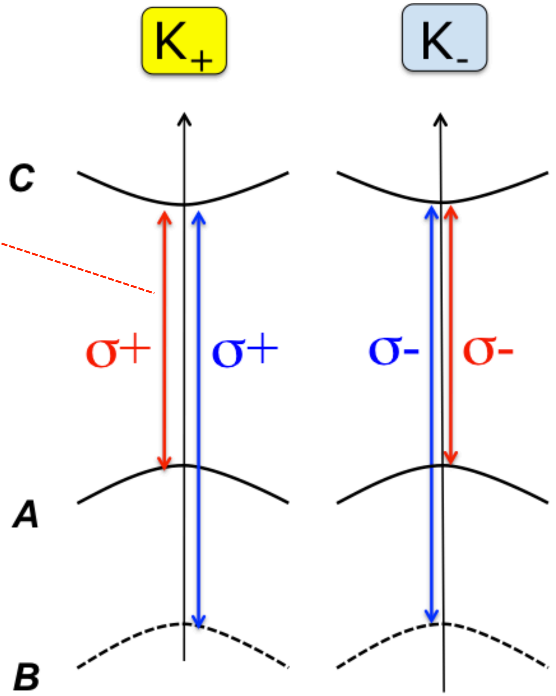
K – valley index (+/-) stable enough for transport (Valley Hall effect) ?

Monolayer MoS_2 Photoluminescence: *temperature 4-300 K*

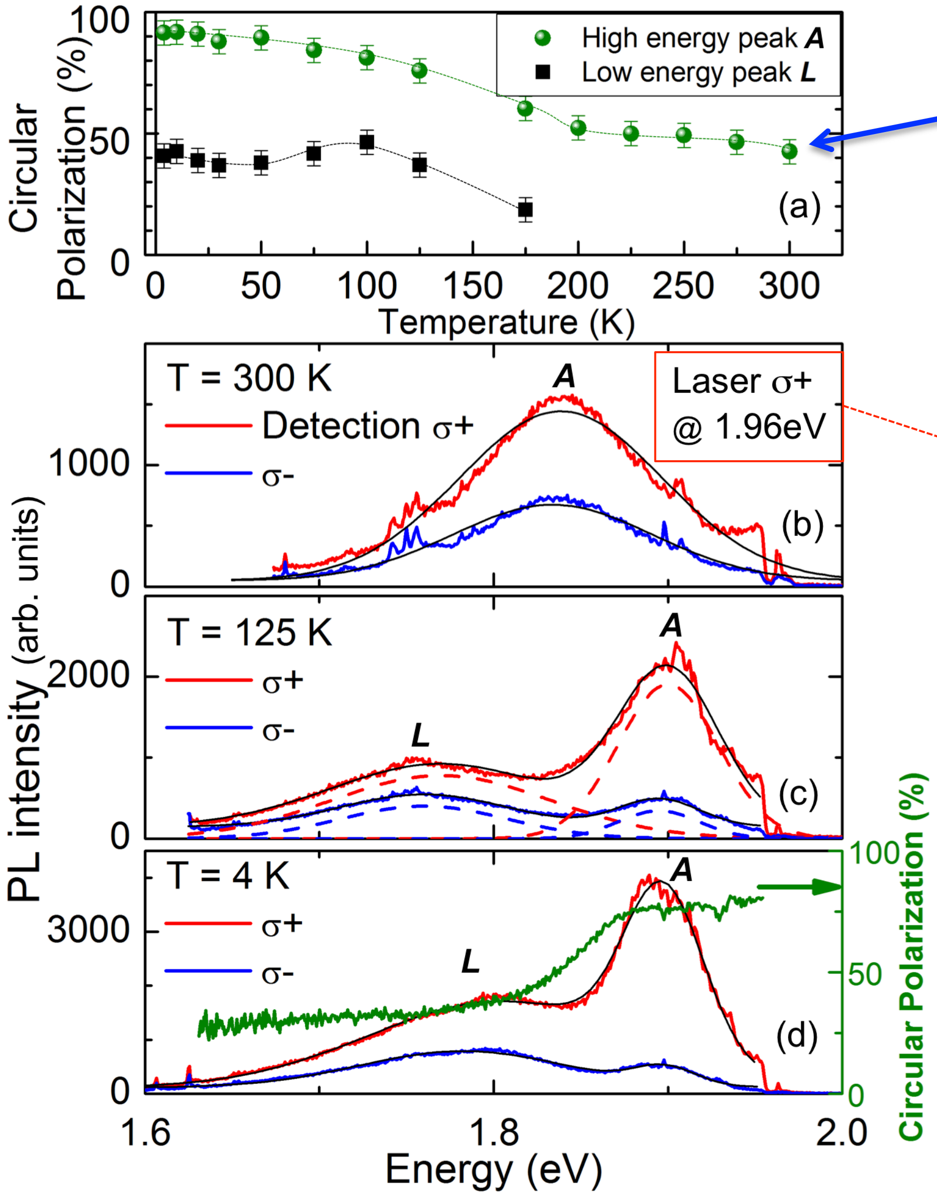


Polarization of PL from MoS_2 monolayers

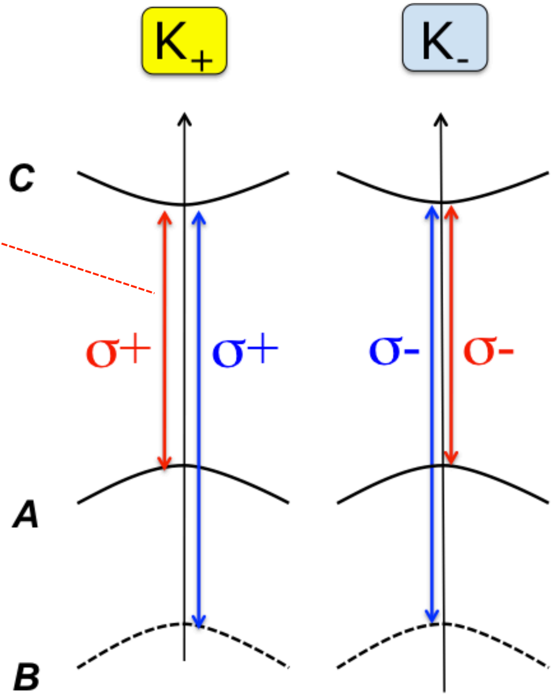




Polarization of PL from MoS_2 monolayers

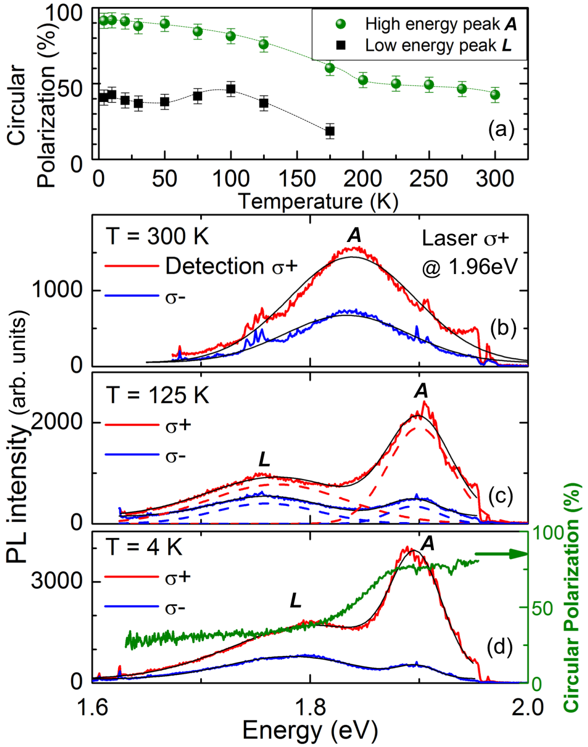


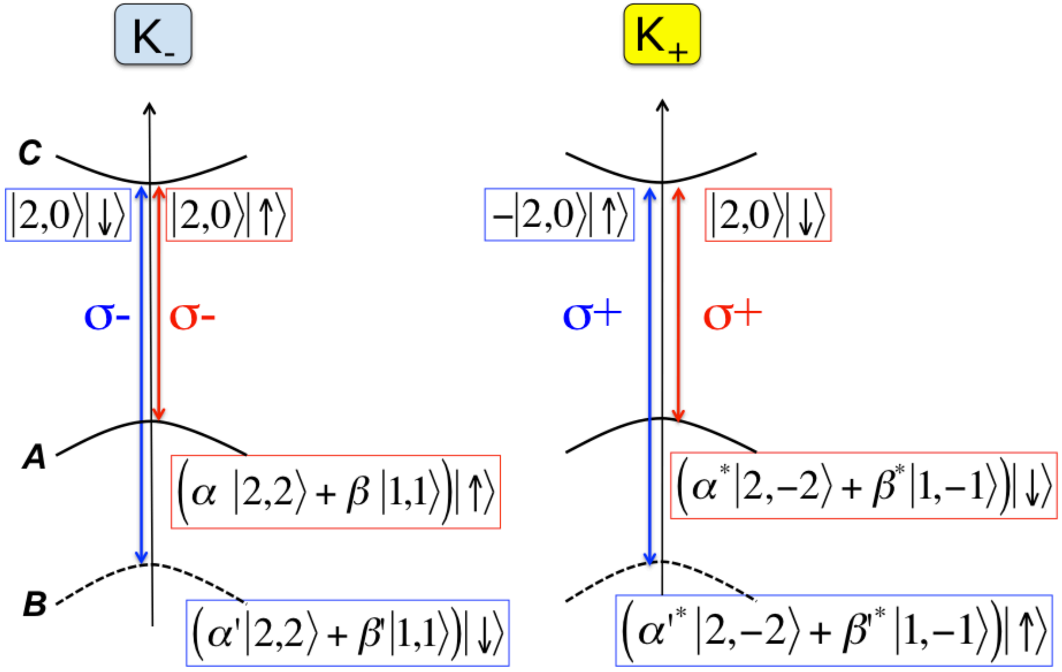
40 % at room temperature



G. Sallen et al, arXiv:1206.5128

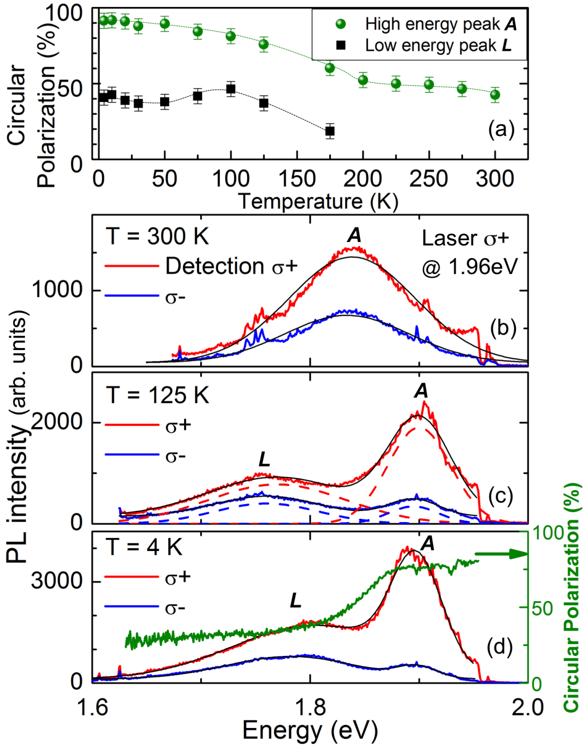
Monolayer MoS_2: optical dipole selection rules

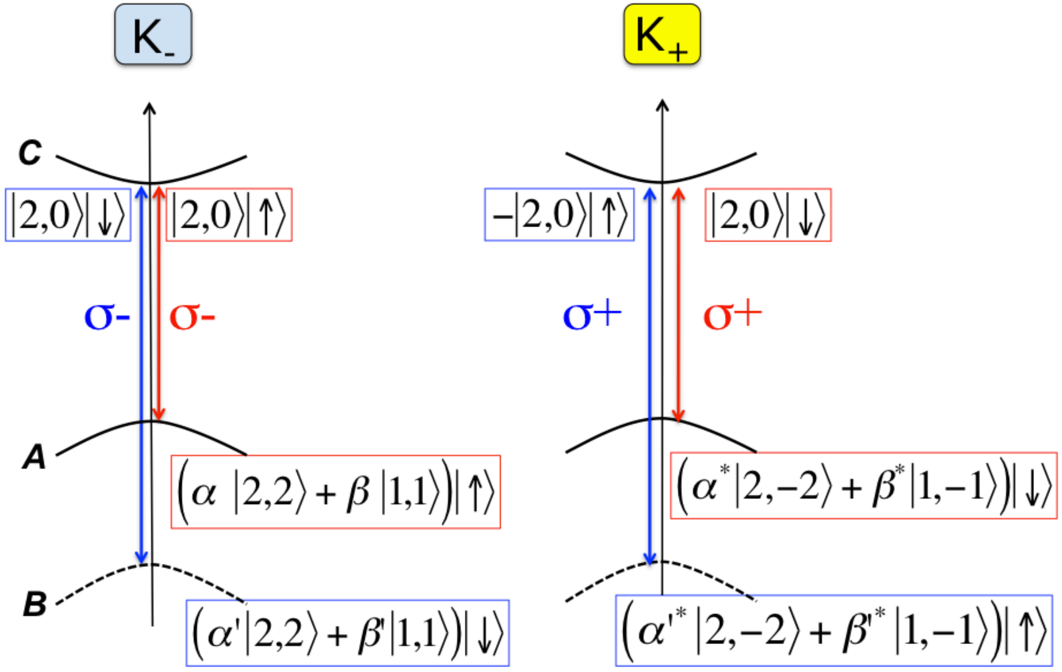




Resonant σ+ excitation: Valley K_+ *and* Spin $|\downarrow\rangle$ initialisation

Monolayer MoS_2: optical dipole selection rules





1. Laser excitation σ+ 2. recombination Conduction Band ➔ Valence Band ***A***

Emission σ+: no change in valley and angular momentum states

Emission σ-: necessary changes

valley $K_+ \rightarrow K_-$

hole spin $|\Uparrow\rangle \rightarrow |\Downarrow\rangle$

electron spin $|\downarrow\rangle \rightarrow |\uparrow\rangle$

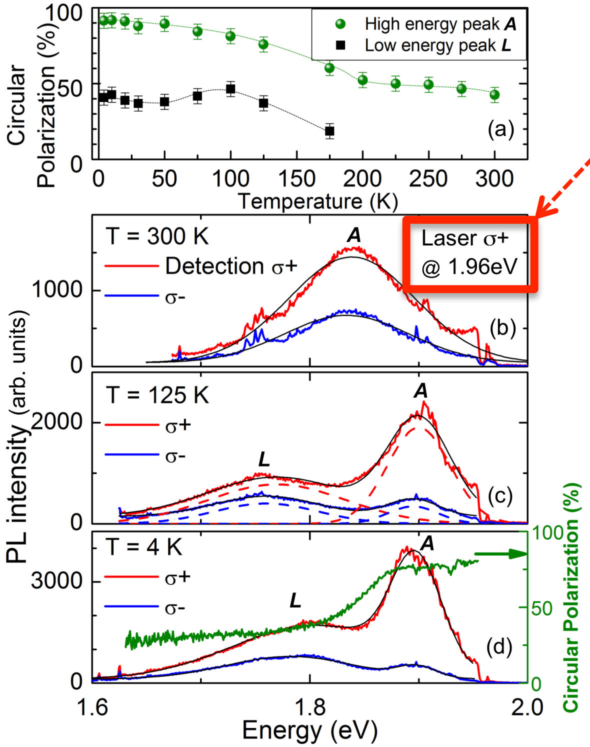
orbital angular momentum

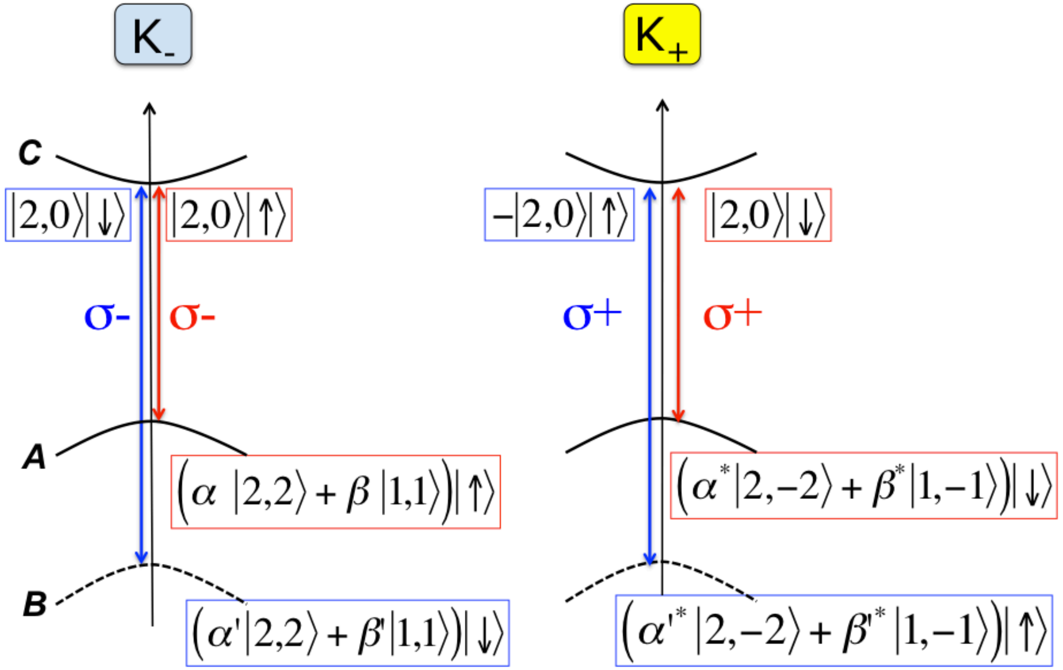
➡ *high PL polarisation*

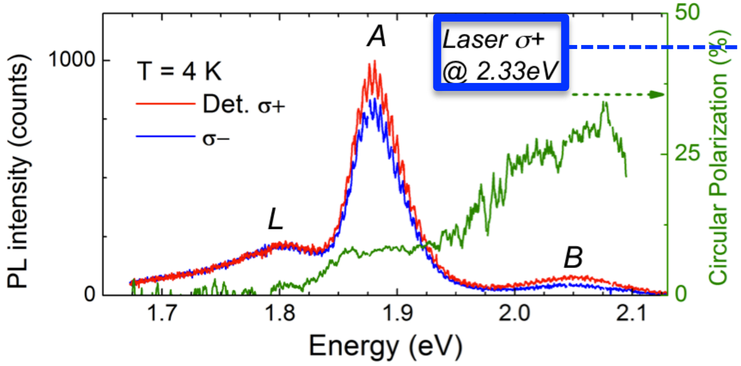
⇔

stable valley index

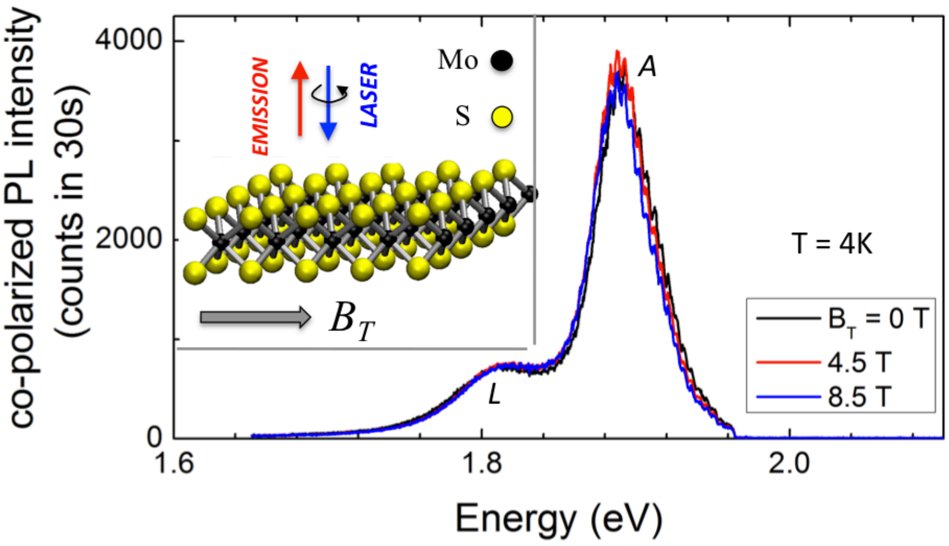
Monolayer MoS_2: high Valley selectivity for quasi-resonant excitation

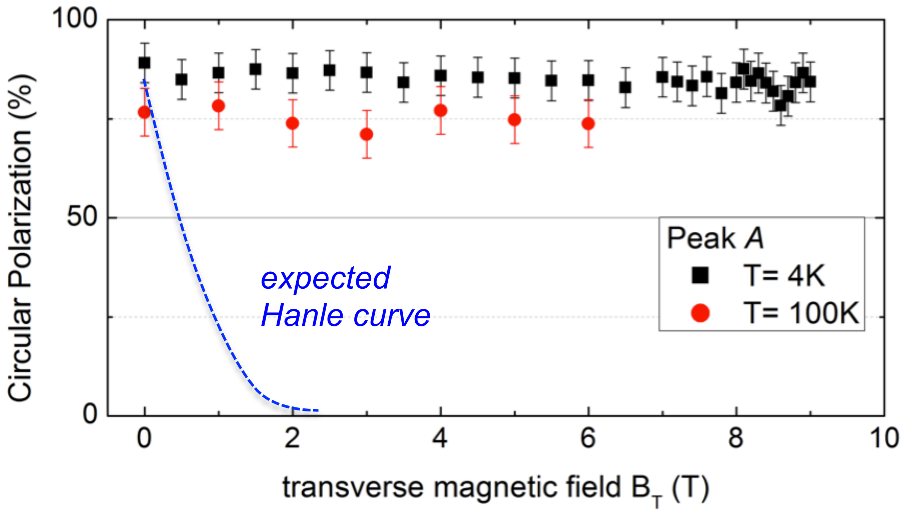






poor Valley selectivity
for highly non-resonant excitation





Stable PL polarization in transverse magnetic fields:

no ‘Hanle’ depolarization curve.

Experiment

⇕

Group theory analysis:

Transverse field B_T **cannot**

couple the ***A*** valence electron

Spin $|\uparrow\rangle$ and $|\downarrow\rangle$ states

from K_- and K_+ valleys

PL Spectroscopy of Monolayer MoS_2: *Main results*

- Initilisation of K+ and K- valley states with σ^+ and σ^- polarized laser
- valley index robust at room temperature and in strong transverse magnetic fields

G. Sallen et al, arXiv:1206.5128

Other recent experimets:
K. F. Mak, T. F. Heinz *et al* Nature Nanotech. NNANO.2012.96 (June 2012).
H. Zeng *et al*, Nature Nanotech. NNANO.2012.95 (June 2012).

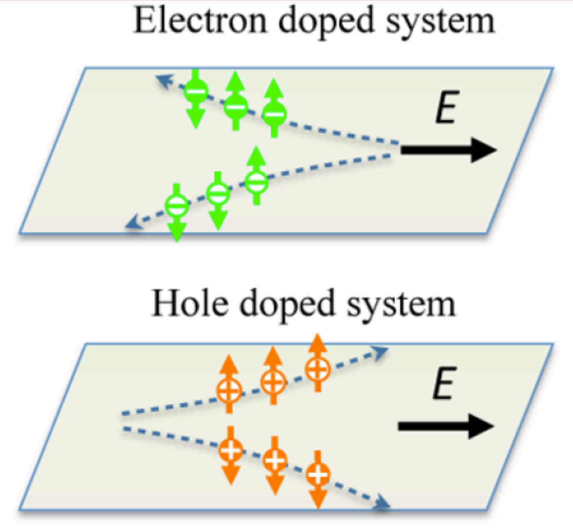
Open Questions:

- Role of strong Coulomb interaction: Exciton binding energy 800meV
- Measure carrier spin and valley lifetimes

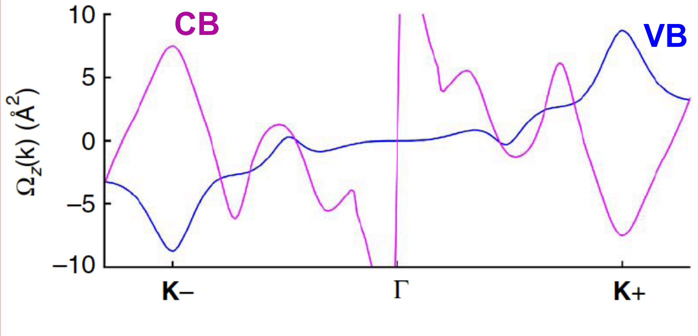
Theory: Olsen et al, arXiv:1107.0600 &
Cheiwchanchamnangij PRB 85, 205302 (2012)

Working towards:

Coupled Spin Hall and Valley Hall effect


Electron doped system

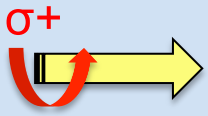
Hole doped system



| Berry curvature, $\Omega_{n,z}(k)$, of bands across the bandgap.

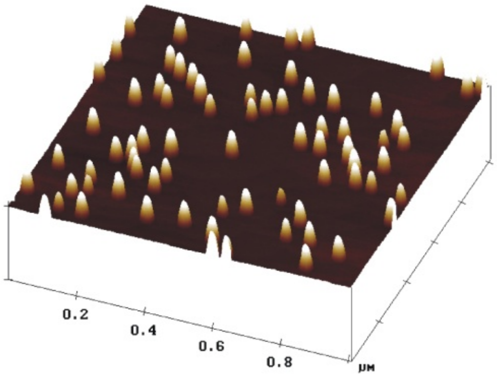
Theory:
Ting Cao et al, ncomms1882 (June 2012) & Di Xiao et al, PRL 108, 196802 (May 2012)

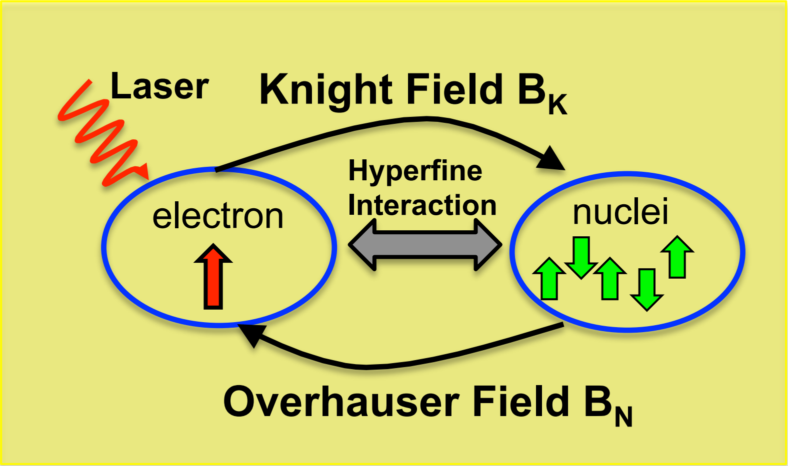
Degrees of Freedom in Semiconductors: *Optical Manipulation*



Main Part :

Optical pumping of **carrier spins** and **nuclear spins** in quantum dots

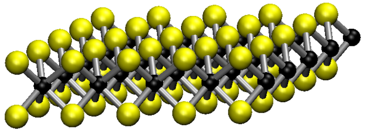


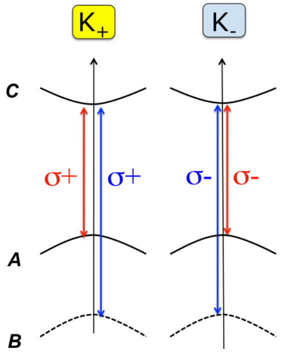


Rev. Mod. Phys. *in press* (arXiv:1202.4637)

Outlook:

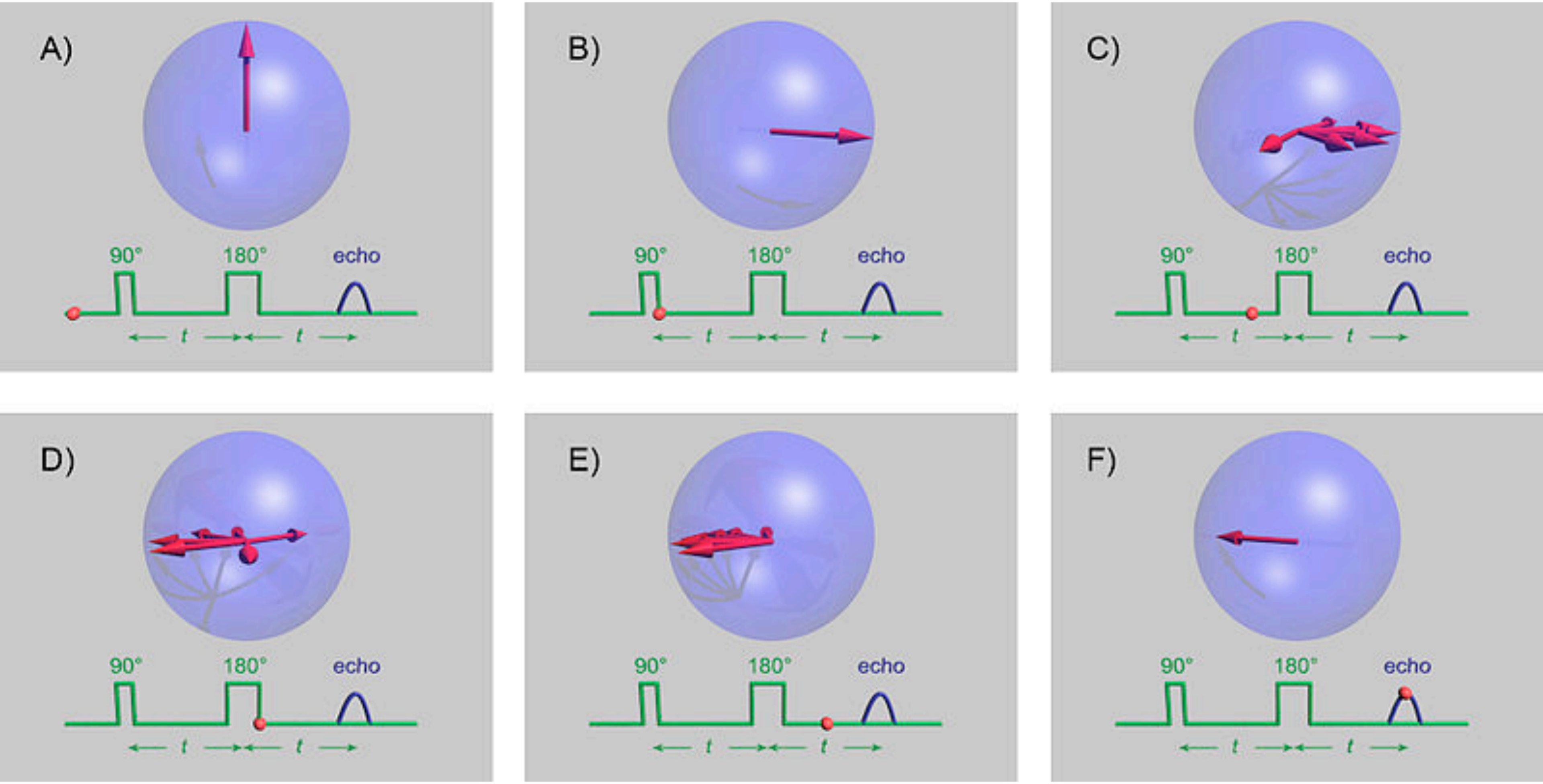
selective **K-valley** excitation in MoS_2 monolayers





arXiv:1206.5128

How can we prolong spin coherence times: *Spin Echo Experiments*



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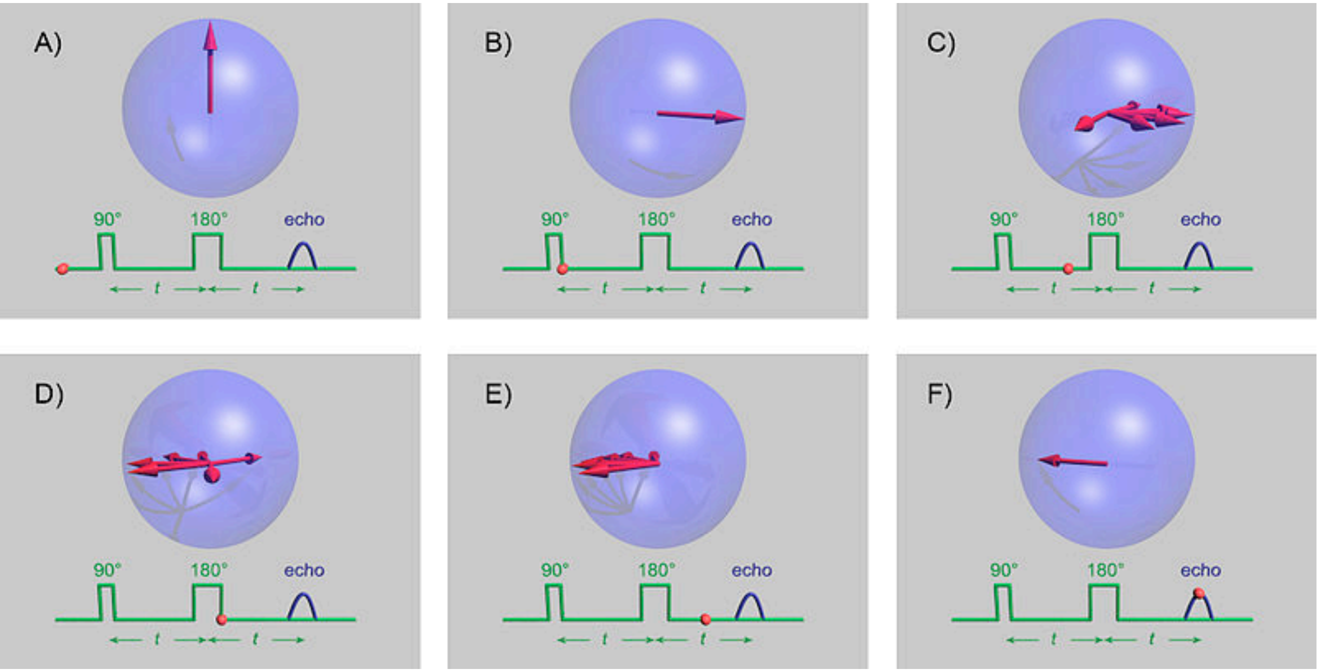
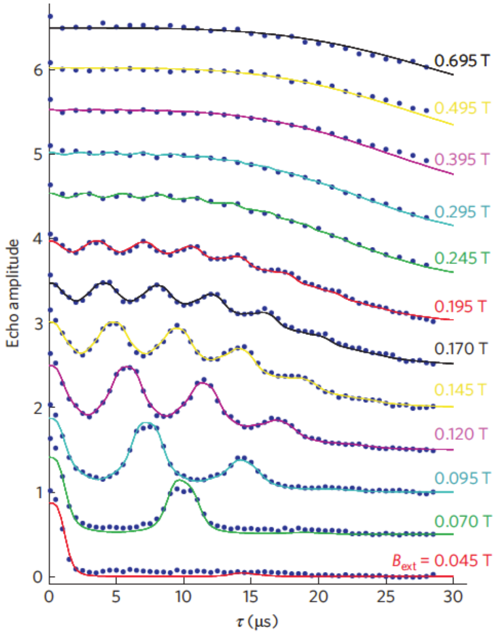


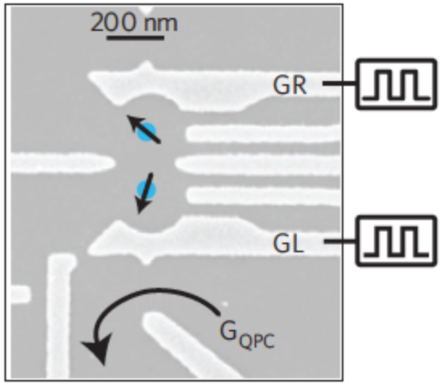
We did not eliminate δB_n

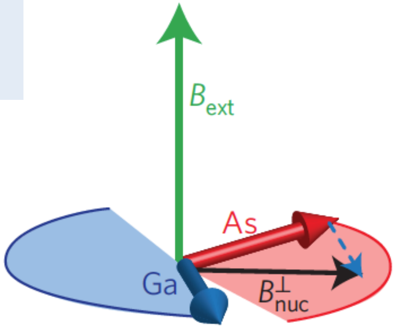
➔ We diminish it's influence



Coherence times in GaAs dots:

- Simple Hahn Spin Echo: 30μs
- Multiple pulses: 200μs



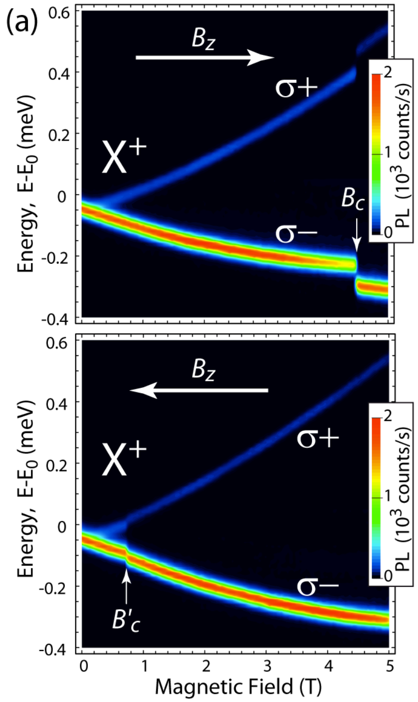


Hahn, E.L. (1950) Physical Review 80;
H. Bluhm Nature Physics 2010

Electron-nuclei interactions in quantum dots

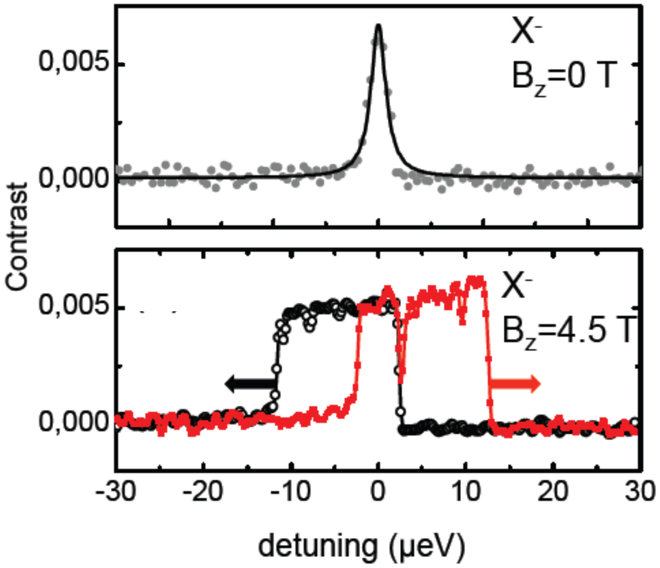
Examples of Nuclear Spin effects in Quantum Dot Optics :

InGaAs/GaAs
Photoluminescence



O. Krebs in Rev. Mod. Phys.
arXiv:1202.4637

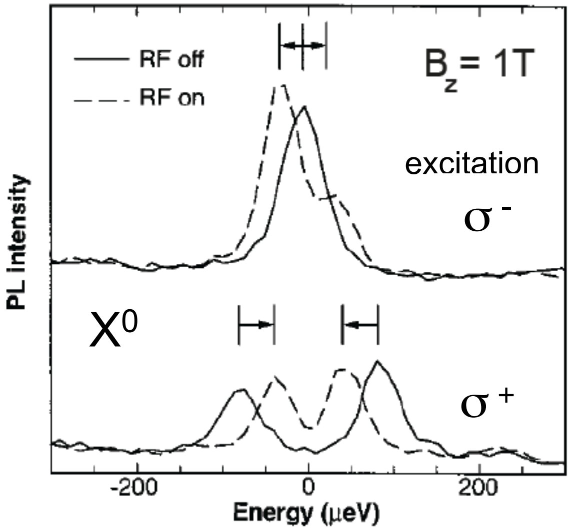
InGaAs/GaAs
Differential transmission



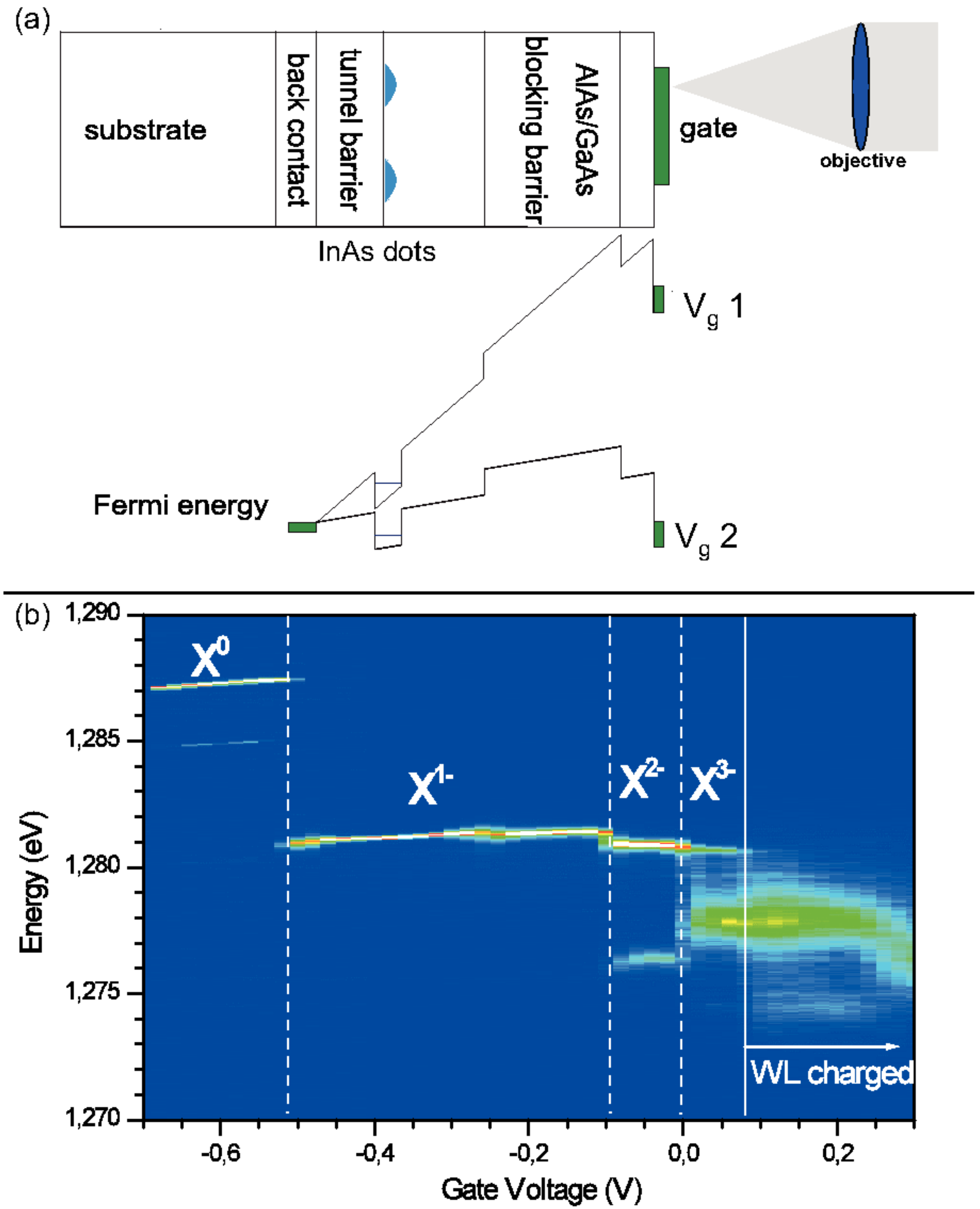
Latta et al., Nature Phys. (2009)

GaAsAlGaAs quantum well
interface fluctuation QD

PL with/out chirped RF pulses



Gammon et al., Science (1997)



Charge tuning

⇔

Paired or unpaired spins