# *Dynamics and optical control of individual Mn spins in a quantum dot*

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# *Introduction*

- Ultimate limit in solid state data storage and processing: Single spin electronics

  - Single carrier spin in a QD
  - N-V centers in diamond
  - P nuclei in Si
  -…
  - Mn atoms in III-V and II-VI semiconductors

- Important properties for a **spin based memory**:

  *Need variable coupling to outside word:*

  - Switching time: *Tunable strong coupling for initialization, manipulation & read-out.*
  - Stability (magnetic anisotropy): *Long relaxation time*.
  - Coherence time: *Isolation to preserve superposition of states.*

- Control the interaction between localized spins for information processing

# *Mn atoms in semiconductors*

- Long relaxation time (ms) for diluted Mn spins under magnetic field (no orbital momentum, weak interaction with phonons)

*Dietl et al. Phys Rev Lett. 74, 474 (1995) ; Scalbert et al. Solid State Com. 66, 571 (1988)*

- Large exchange interaction with free carriers:

  - Tool to interact with a localized spin

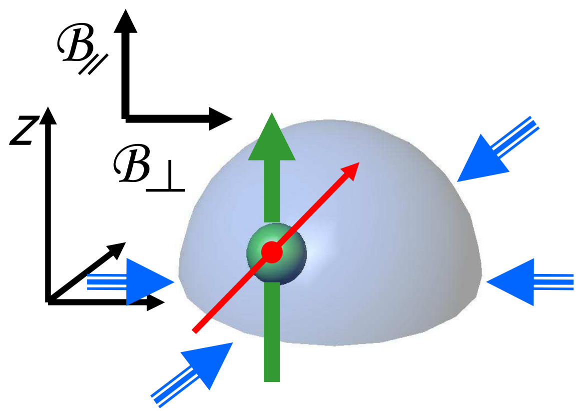
*L. Besombes et al. , Phys. Rev. Lett. 93, 207403 (2004); A. Kudelski et al, Phys. Rev. Lett. 99, 247209 (2007)*

  - Carrier controlled ferromagnetism

*Ohno et al. Nature 408, 944 (2000); Boukari et al. Phys. Rev. Lett. 88, 207204 (2002)*

Control of individual magnetic atoms and their interaction

- But, Mn atoms ($^{55}$Mn, S=5/2, I=5/2) have also a nuclear spin and are sensitive to their solid state environment: Can we deal with this complexity?



# *Outline*

1. *A II-VI quantum dot as a tool to optically probe the spin state of individual magnetic atoms (1 or 2 Mn).*

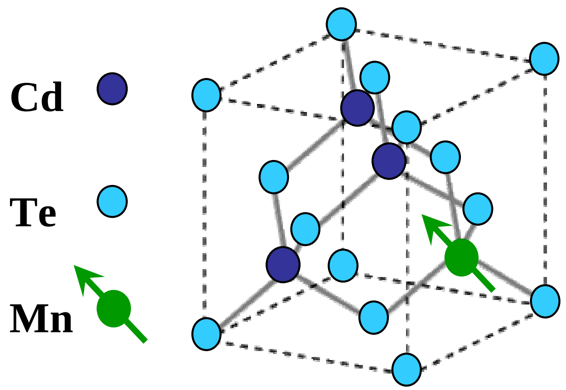
2. *Single Mn spin dynamics:*
   *Mn spin memory: strained induced magnetic anisotropy*
   *Optical initialization and readout of an individual Mn spin*

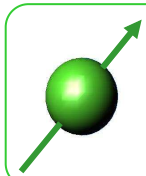
3. *Spin dynamics of optically dressed Mn atoms:*
   *Optical Stark effect on an individual Mn spin*
   *Spin population trapping.*

# *Mn atoms in a II-VI quantum dot*

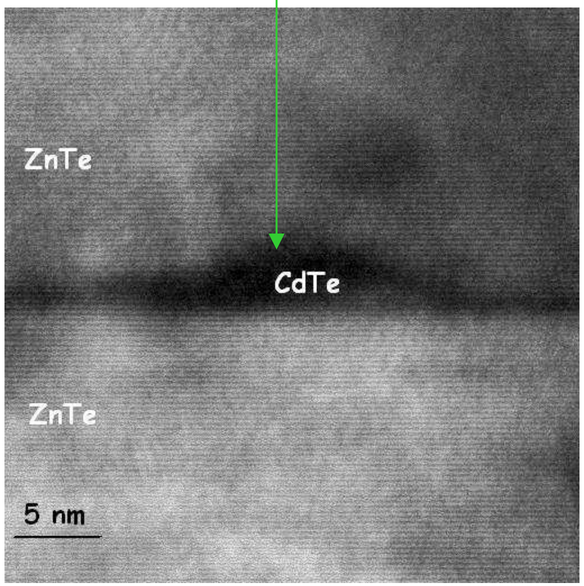
Cd: $3d^{10}$ $4s^2$
Mn: $3d^5$ $4s^2$

Mn replace Cd: $Mn^{2+}$
Isoelectronic doping



Mn: 5 *d* electrons S=5/2




Exchange interaction: contact interaction

•Mn – electron: $\alpha|\psi_e(r_I)|^2 \vec{S}_I \cdot \vec{\sigma}_e$

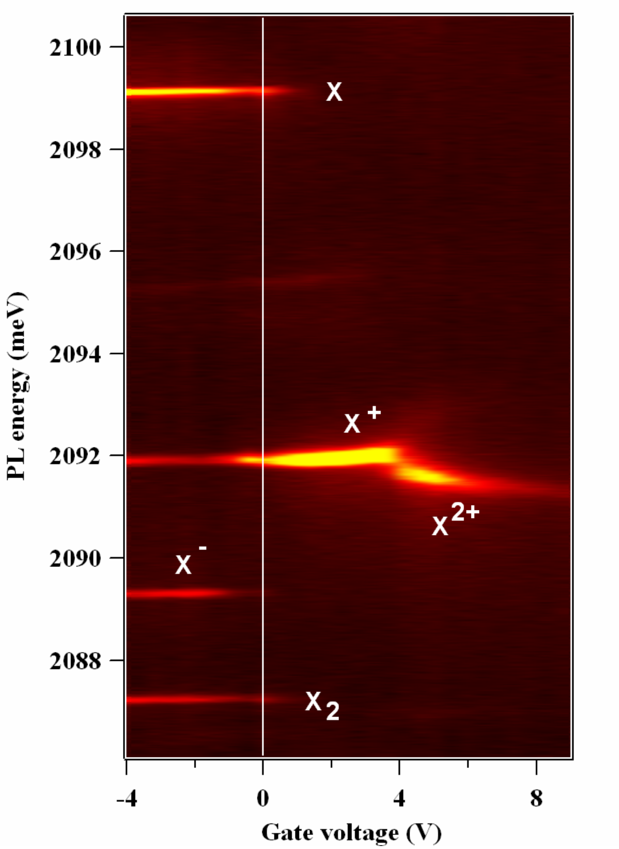
•Mn – hole: $\beta|\psi_h(r_I)|^2 \vec{S}_I \cdot \vec{J}_h$

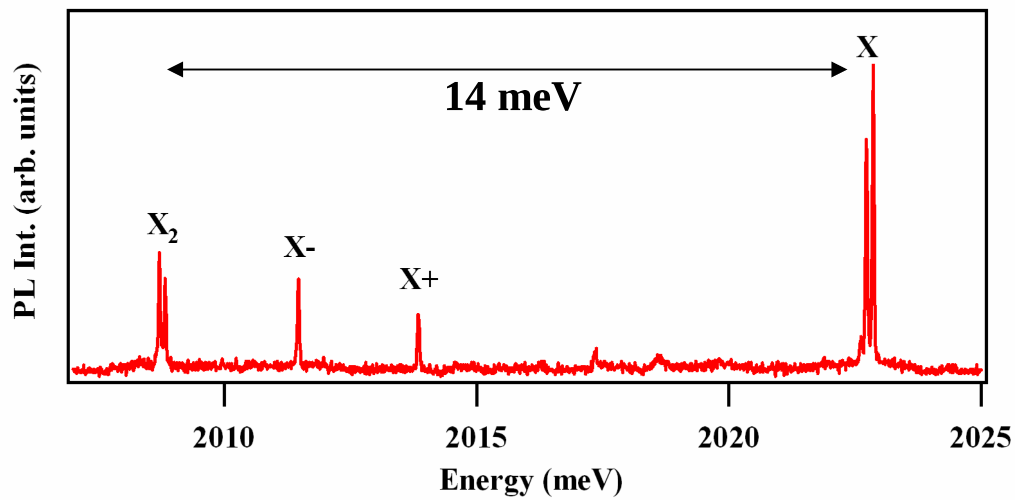
$\beta \approx -4\alpha$ Enhanced by the carrier confinement in a QD

Low density of Mn in the CdTe layer during MBE growth

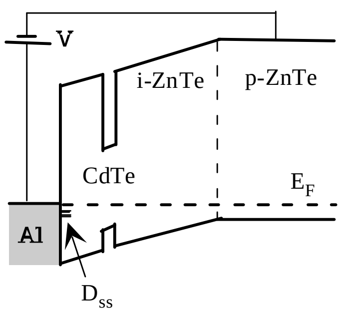
# *Few particles Coulomb effects in a single II-VI QD*

Excitonic species in a single CdTe/ZnTe QD:



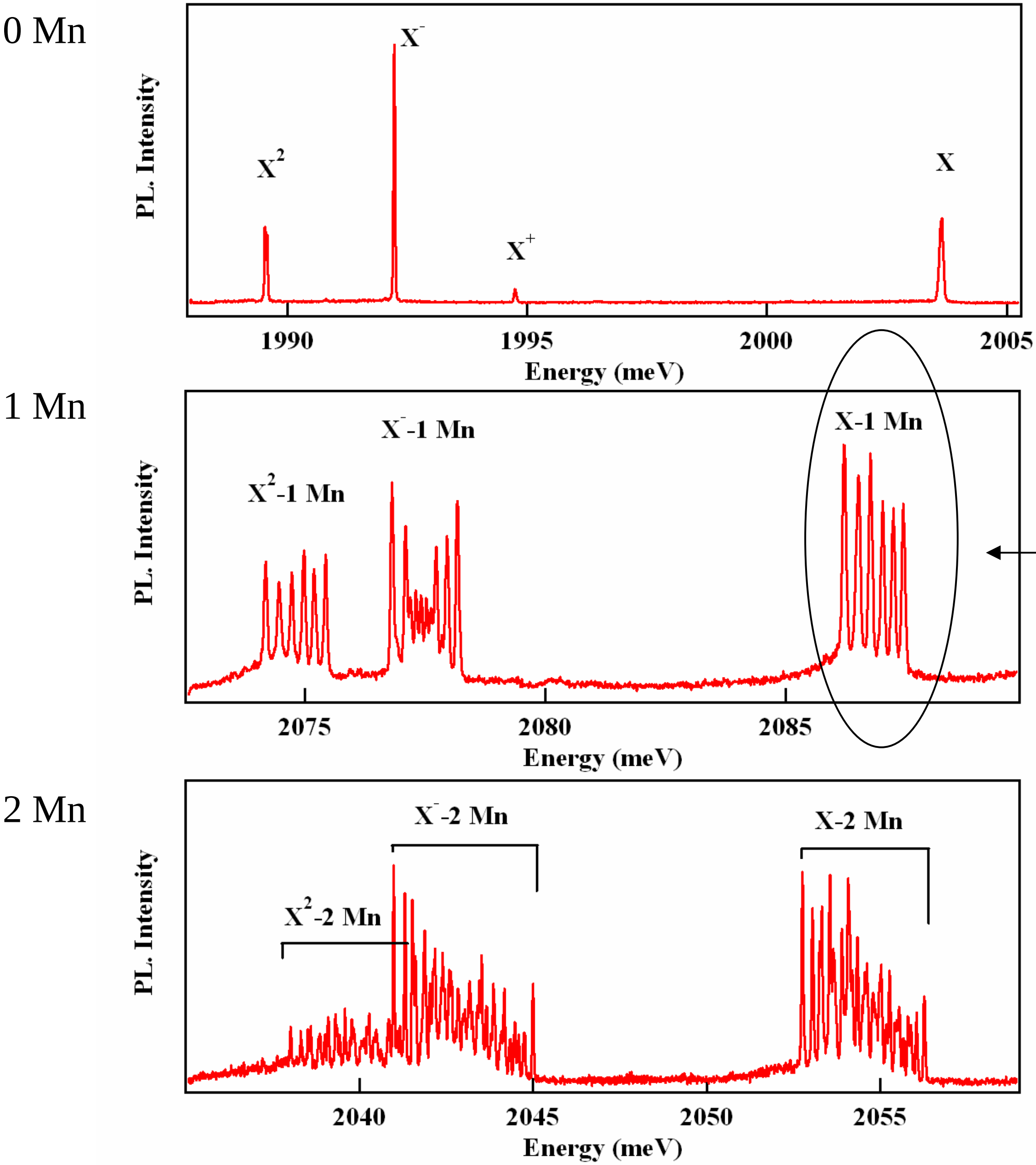



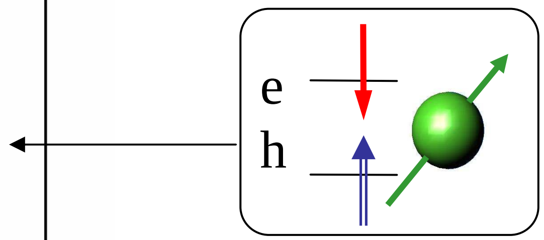

Modulation doping of CdTe / ZnTe QDs by surface states.




- Large Coulomb interaction (direct and exchange)

# *Mn atoms in a II-VI quantum dot*




- Excitonic species are split by the exchange interaction with Mn spin.

# *Heavy-hole exciton / Mn coupling*

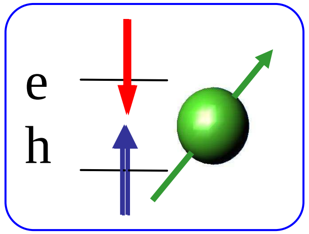
$$I_{h-Mn}(j_z.S_z)$$

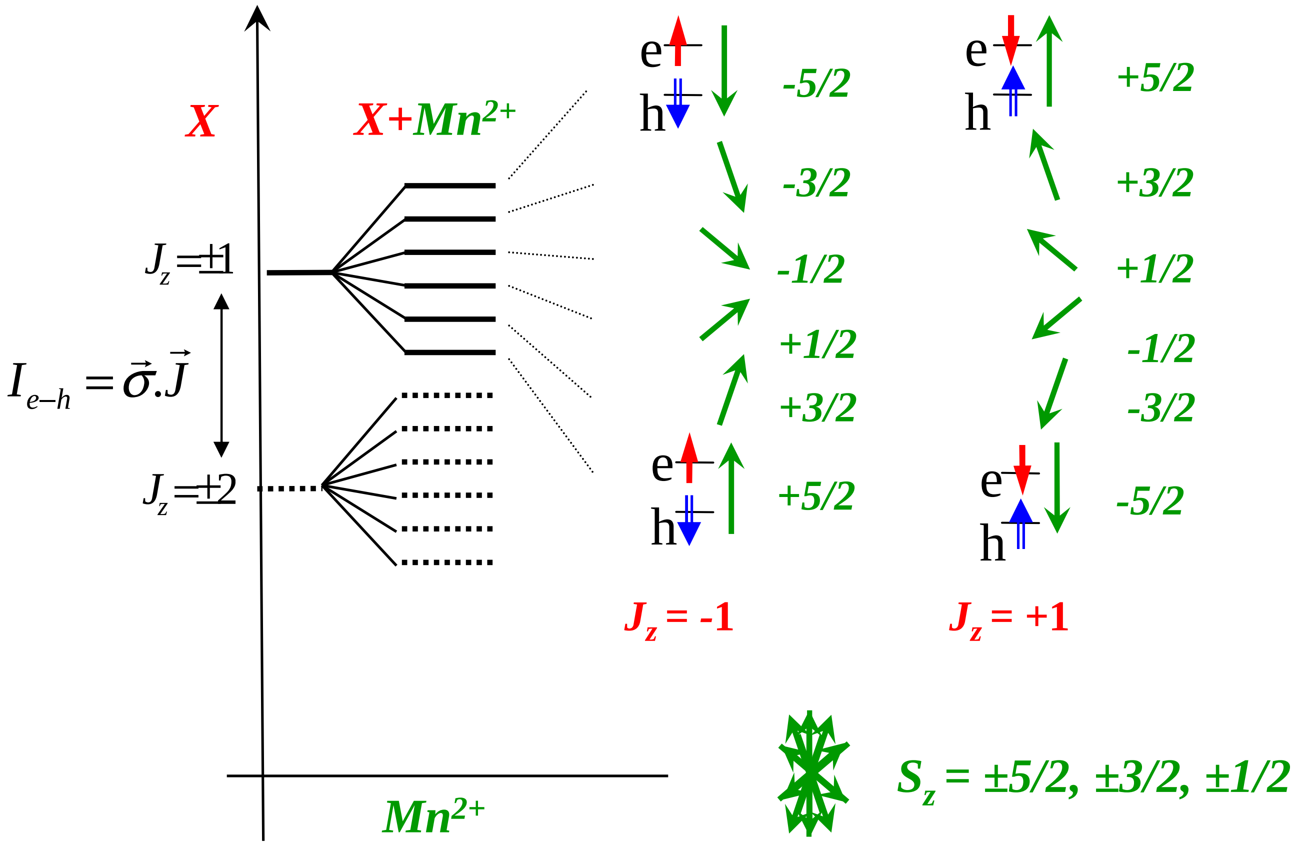
$J_z = \pm 3/2$

$$I_{e-Mn}(\sigma_z.S_z + 1/2(\sigma_+.S_- + \sigma_-.S_+))$$

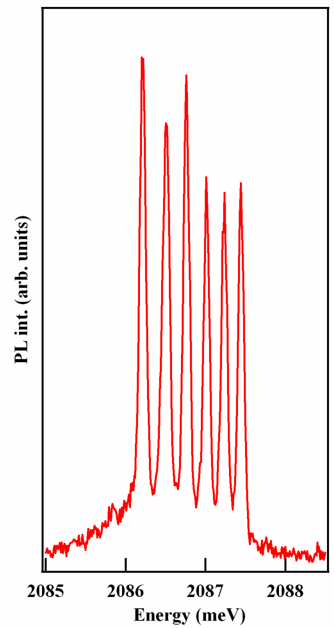
$$|I_{e-Mn}| << |I_{h-Mn}|$$

$$|I_{e-Mn}| << |I_{e-h}|$$

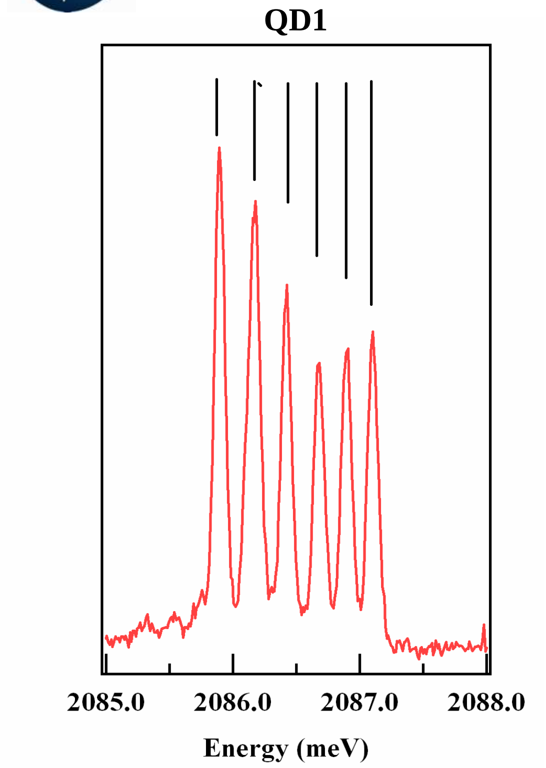



1 photon
=
1 Mn spin state

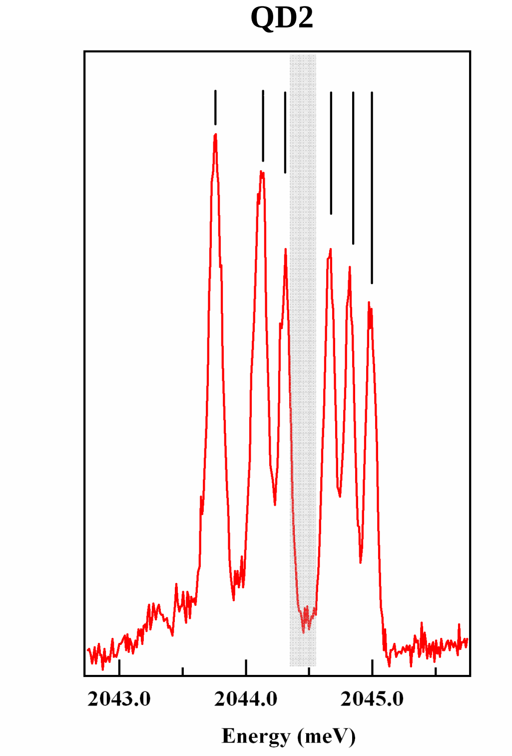


# *Influence of QDs structural parameters*

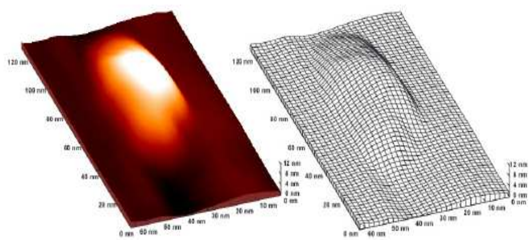



Heavy-hole + Mn

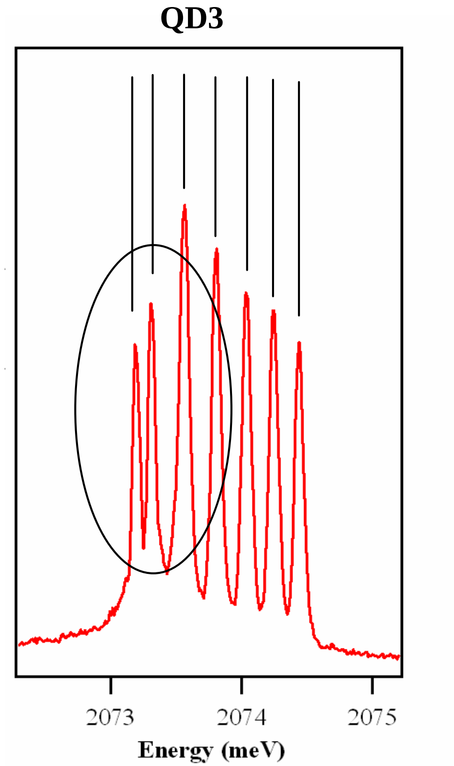
*Phys Rev Lett. 93, 207403 (2004)*



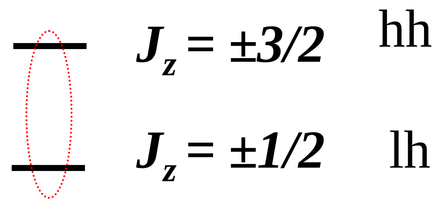

Influence of the QD shape:



*Phys Rev Lett. 95, 047403 (2005)*

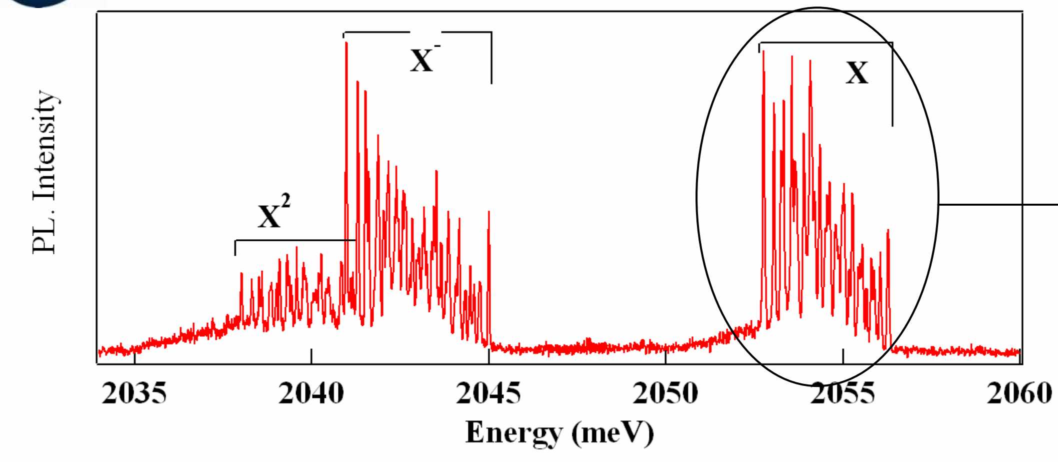


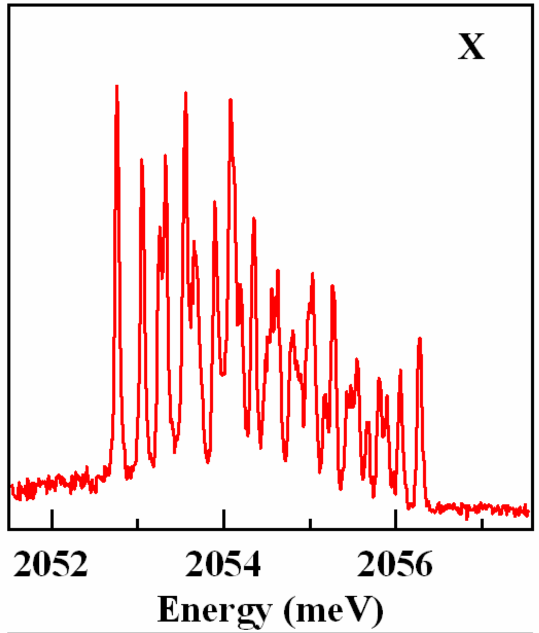

Influence of the valence band mixing:




*Phys Rev B. 72, 241309(R) (2005)*

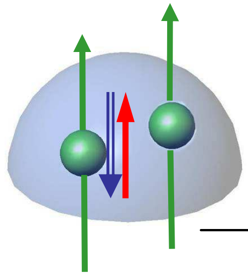
# *Optical probing of two Mn spins*

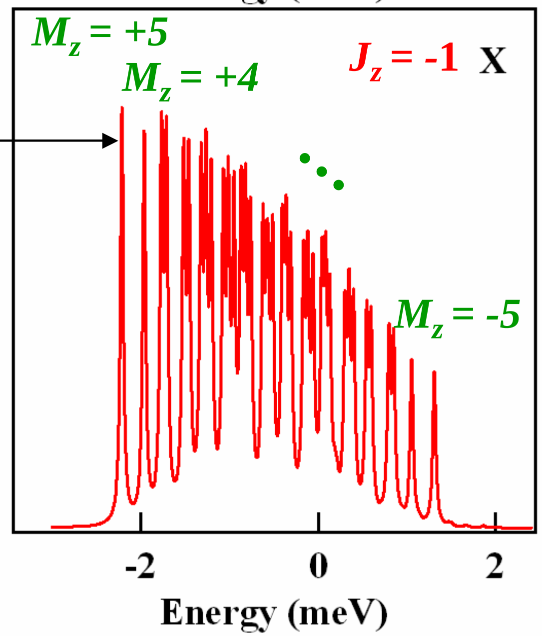



- Mediate the $S_1$ $S_2$ interaction through a carrier …

**$M_z$ = +5: ferromagnetic coupling mediated by the exciton.**






- Spin effective Hamiltonian:

$$\mathcal{H} = \vec{\sigma}\cdot\left(I_{e,1}\vec{S_1} + I_{e,2}\vec{S_2}\right) + \vec{J}\cdot\left(I_{h,1}\vec{S_1} + I_{h,2}\vec{S_2}\right)$$
$$+ \; I_{eh}\vec{\sigma}.\vec{J} + I_{12}\vec{S_1}.\vec{S_2}$$

Exchange interaction of the bright exciton with the 2 Mn spins give rise to 36 emission lines.

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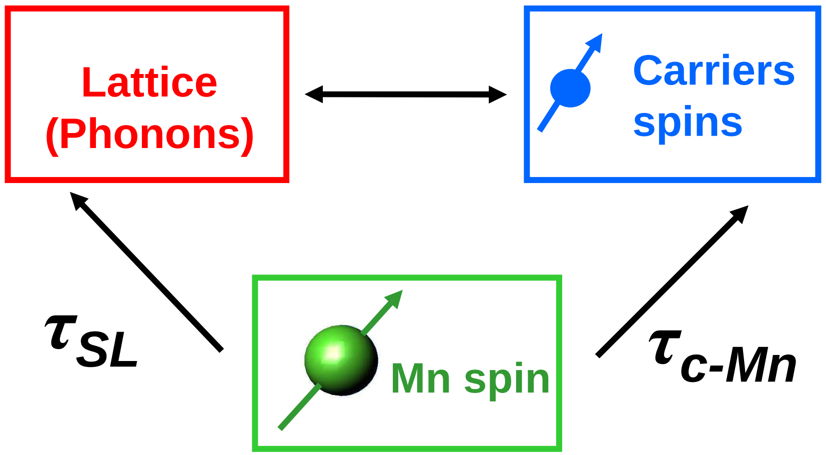
# *Isolated Mn spin dynamics*

Spin relaxation mechanism under magnetic field…




- $\tau_{e\text{-}Mn}$ : spin-spin coupling with the surrounding carriers
- $\tau_{sl}$ very long for an isolated Mn atom

… relaxation time at vanishing Mn density in the ms range.

At zero magnetic field

$^{55}\mathcal{M}n$ $\mathcal{S}=5/2$, $I=5/2$

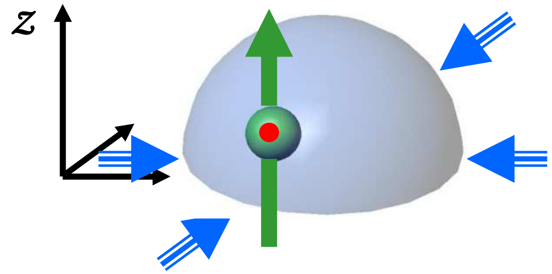
Need to consider the $Mn^{2+}$ **fine structure:**

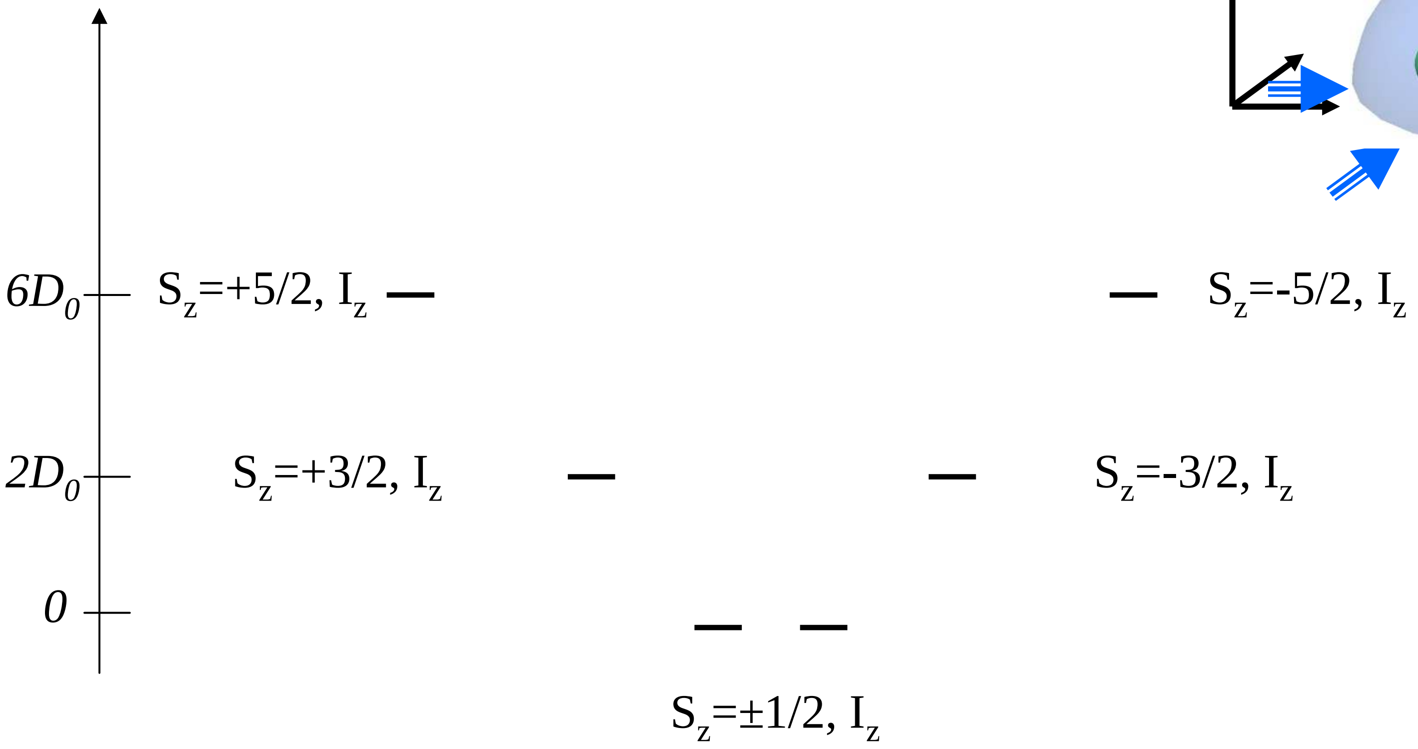
- Hyperfine coupling with its nuclei
- Strained induced magnetic anisotropy (crystal field).

# *$Mn^{2+}$ fine and hyperfine structure in a strained QD*

$\mathcal{H}_{Mn} = D_0 S_z^2$

$^{55}\mathcal{M}n \ \ \mathcal{S}=5/2, \ I=5/2$



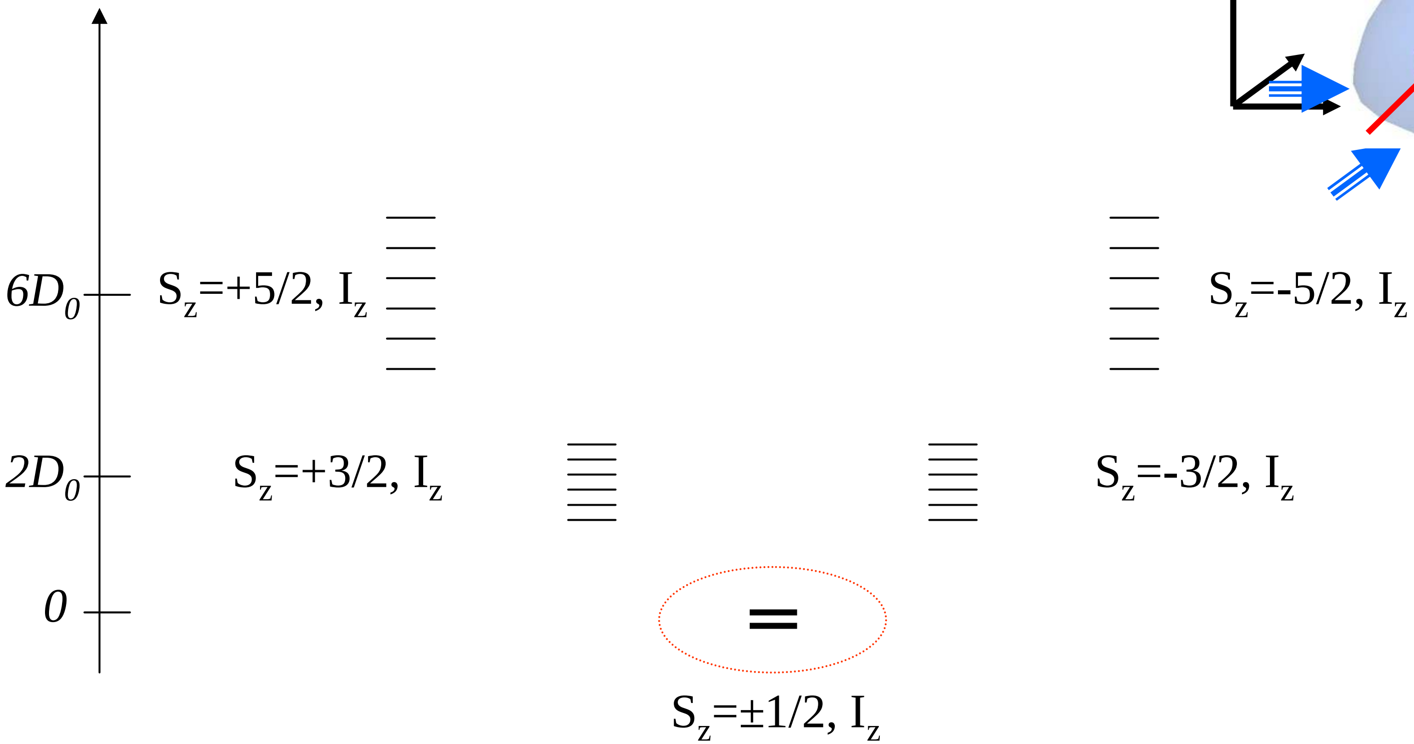

$D_0$ is controlled by the biaxial strain in the QD plane

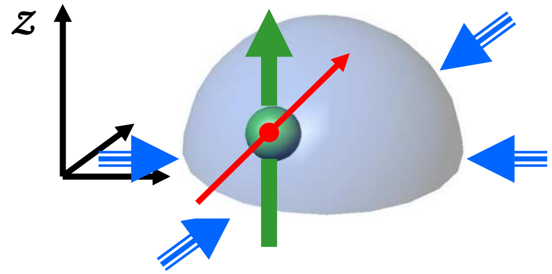
# $Mn^{2+}$ fine and hyperfine structure in a strained QD

$\mathcal{H}_{Mn} = D_0 S_z^2 + A\, I.S$

$^{55}Mn$ $S=5/2$, $I=5/2$

$D_0 > A$



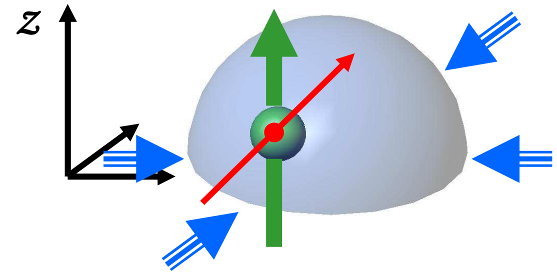
# *$Mn^{2+}$ fine and hyperfine structure in a strained QD*

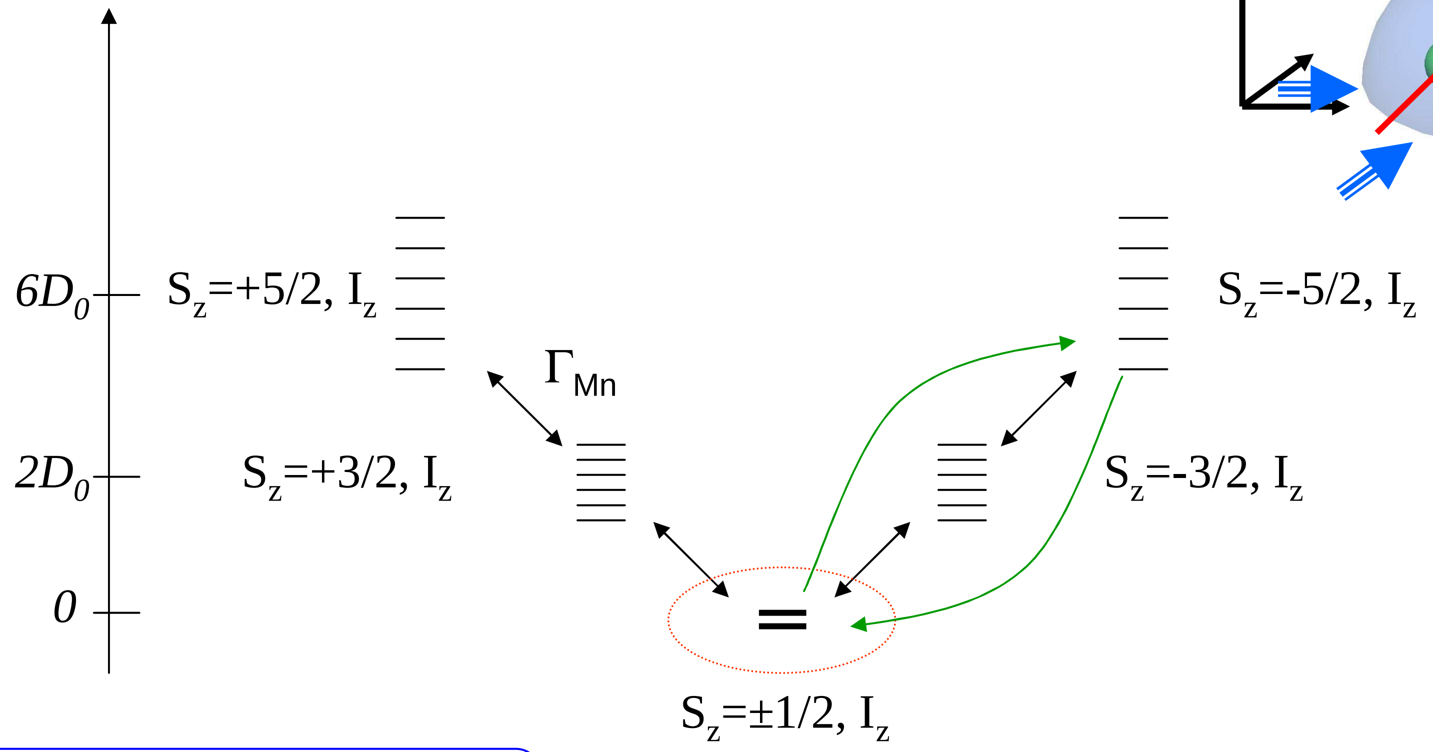
$$\mathcal{H}_{Mn} = D_0 S_z^2 + A\, I.S + a\,(S_z^4+S_y^4+S_x^4) + E\,(S_x^2 - S_y^2)$$

$^{55}\mathcal{M}n$ $\mathcal{S}=5/2$, $I=5/2$

$D_0 > A$, $D_0 > a$, $D_0 > E$ Spin relaxation: Phonons

Free carriers

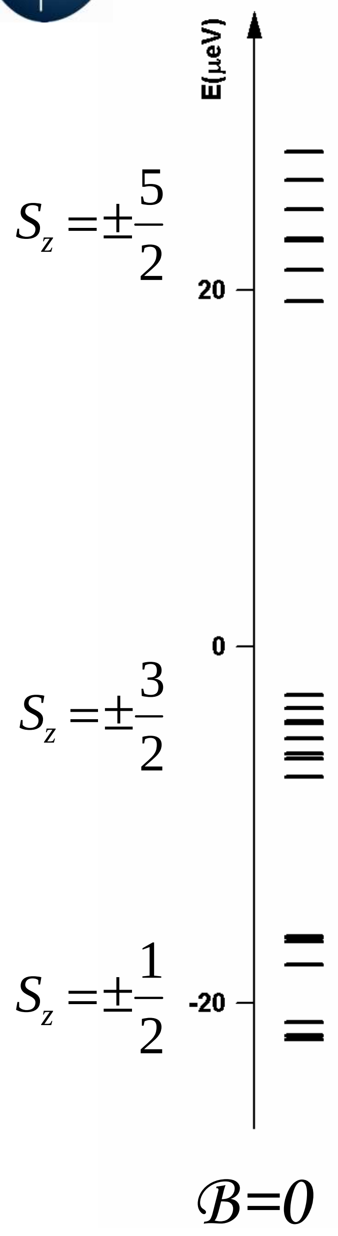



$S_z = \pm\frac{5}{2}$ Biaxial strain suppress the ν-Mn spin-flips at B=0T

$S_z = \pm\frac{1}{2}$ Precess in the ν field

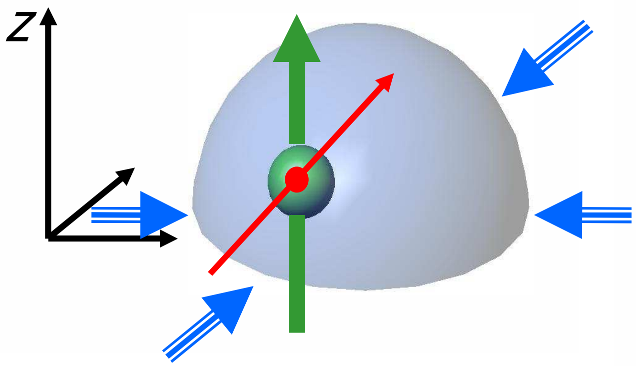
# *$Mn^{2+}$ spin dynamics in a QD at B=0T*

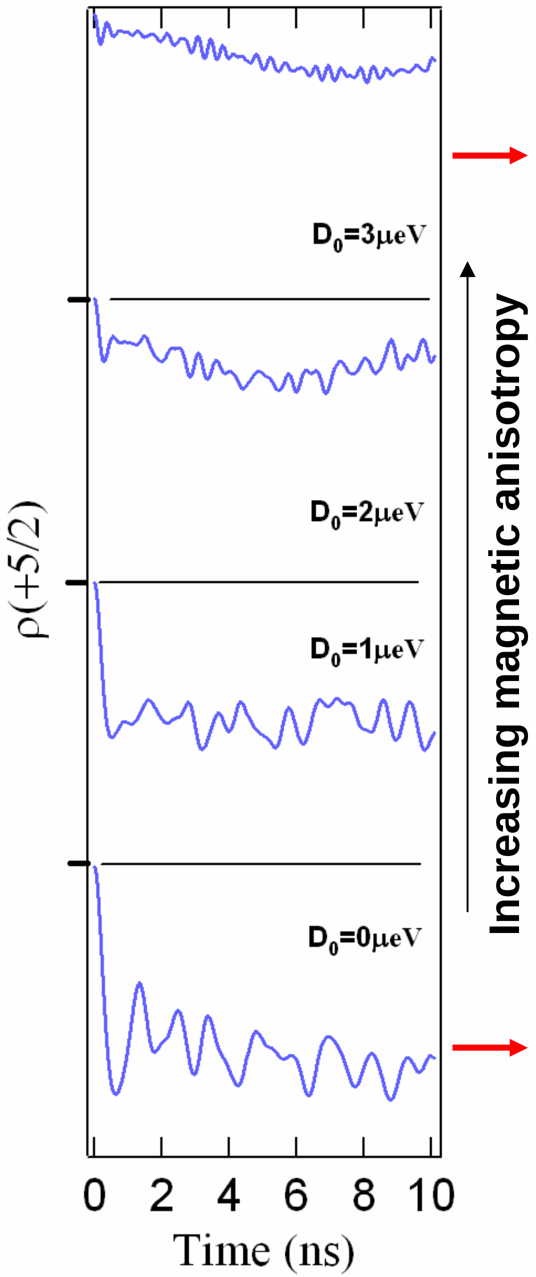



$$\frac{d\varrho_S(t)}{dt} = \frac{1}{i\hbar}[\mathcal{H}_{Mn}, \varrho_S(t)] + \mathcal{L}\varrho_S(t)$$

$$S_z(t=0) = +\frac{5}{2}$$

$$T_1^{Mn} = 1\mu s$$

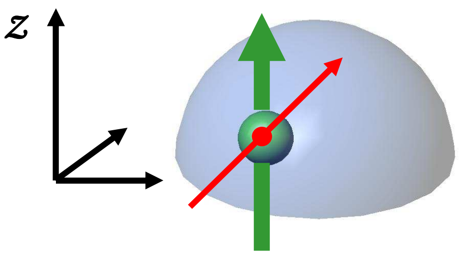


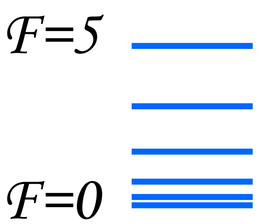

$S_z$ is conserved

$S_z$ precess with the nuclear spin

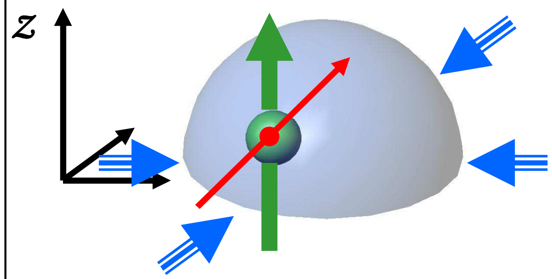
# *Isolated Mn spin relaxation mechanism*



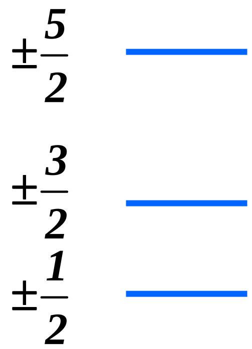

**Hyperfine coupling**



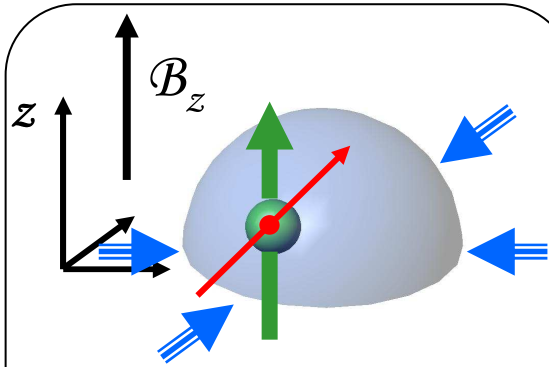

$$\mathcal{H}_{Mn} = \mathcal{A}\,\vec{I}\,.\,\vec{S}$$



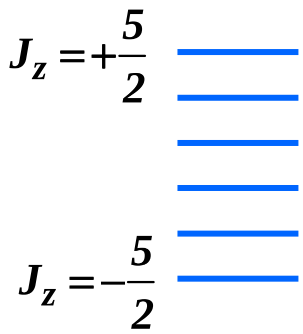

**Biaxial strain in the QD plane**




$$\mathcal{H}_{Mn} = \mathcal{D}_0 S_z^2$$




**Magnetic field or exchange interaction with a hole**




$$\mathcal{H}_{Mn} = g\mu_B H_z.S_z$$

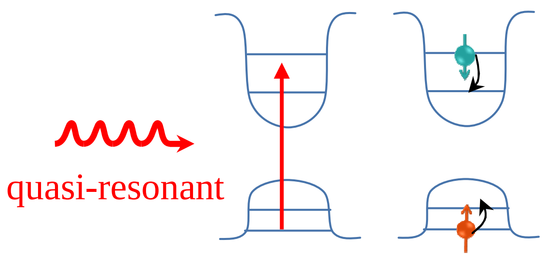
Coherent evolution in the hyperfine field:
No spin memory

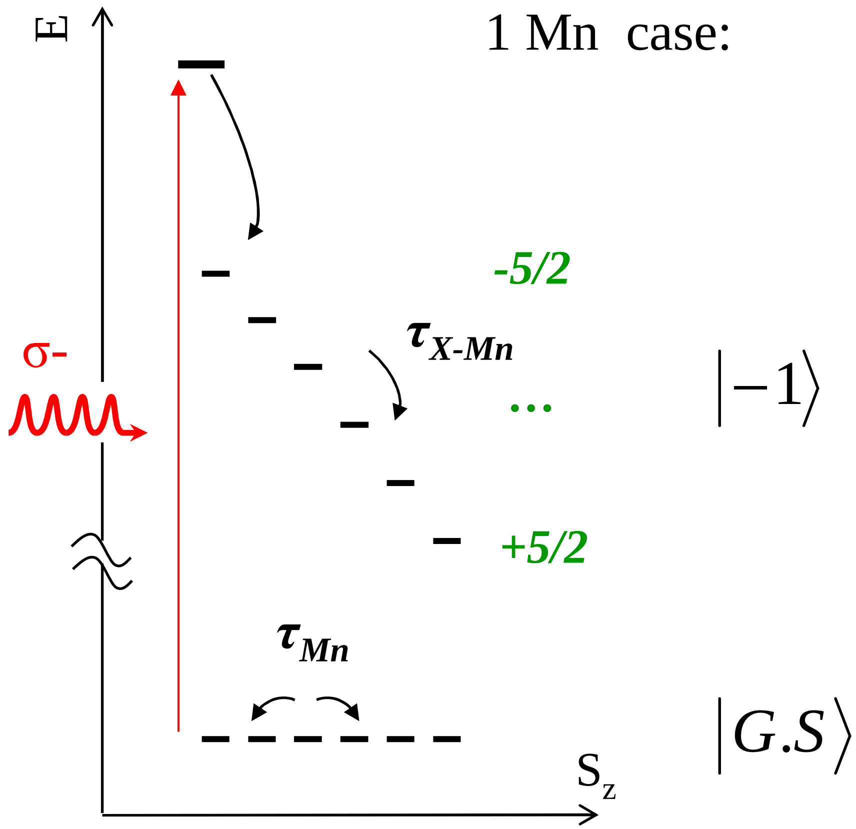
Dynamics controlled by **single phonon process**

**Mn spin relaxation strongly depend on the splitting**

# *Optical orientation of Mn spins*

- Optical orientation of the Mn in the exchange field created by spin polarized carriers:

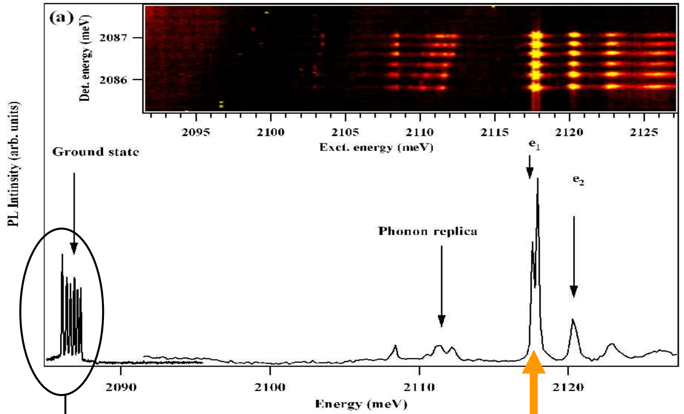


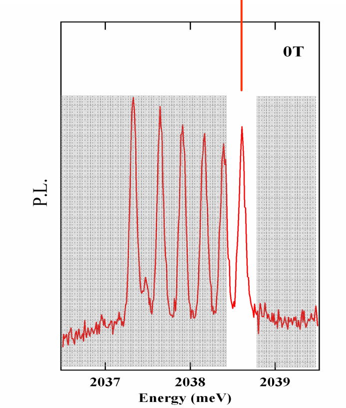

$\tau_{XMn} < \tau_{Mn}$

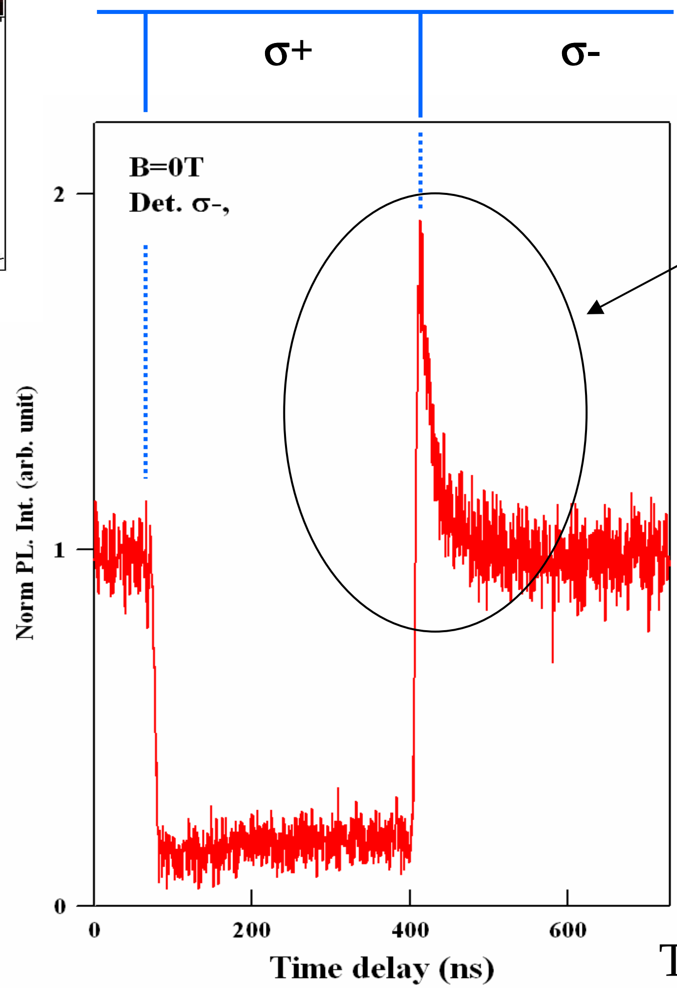
Dynamic optical orientation

# *Optical orientation of a single Mn spin*

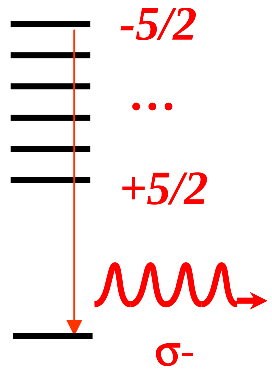



- Time resolved optical orientation under quasi-resonant excitation:



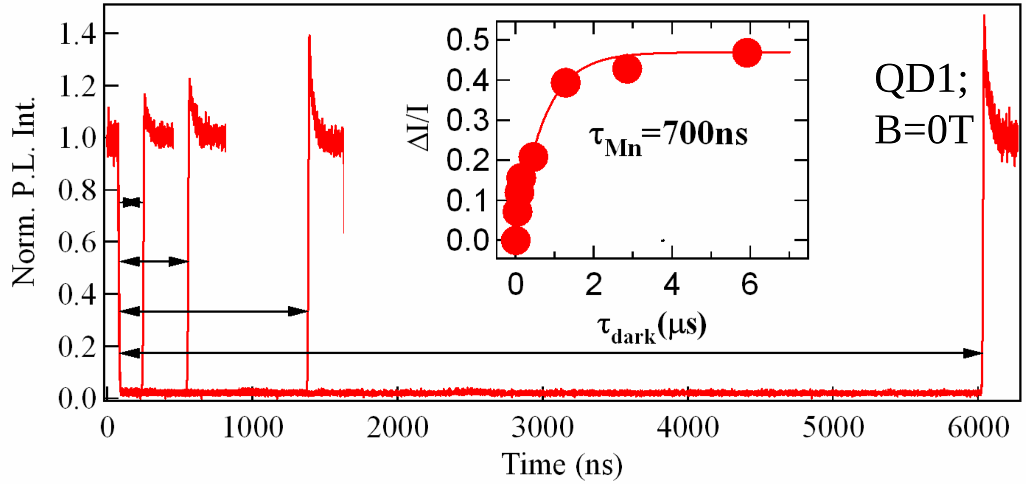

$\tau_{pump} \approx 50ns$




The injection of σ- exciton “empty” the -5/2 state

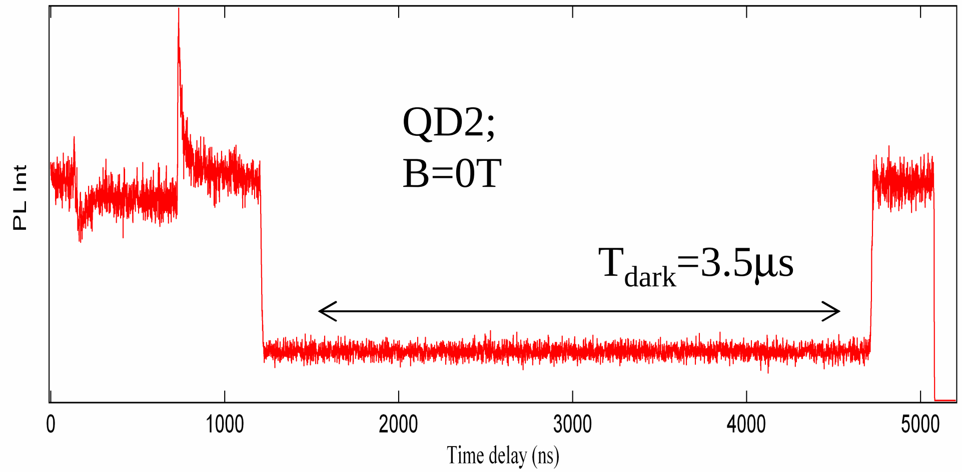
*Phys. Rev. Lett. 102, 127402 (2009)*

# *Single Mn spin memory at B=0T*



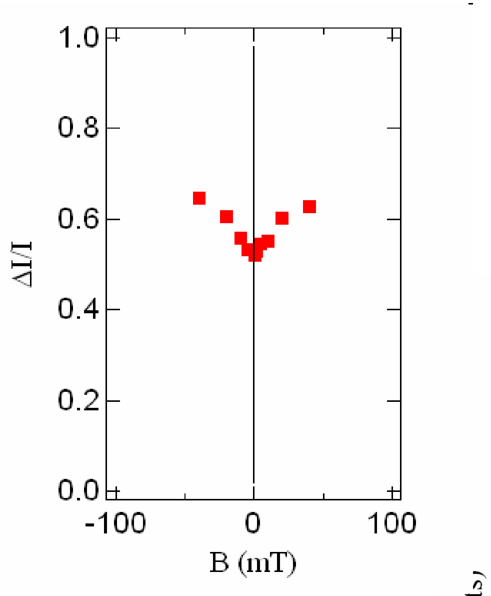

- If the relaxation time is shorter than the dark time the optical pumping signal reappears after the dark time

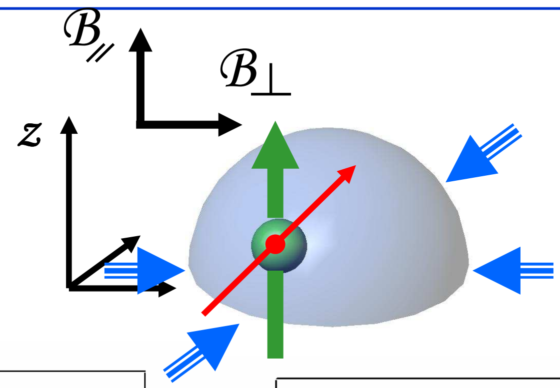
- Most of the dots:
Relaxation time longer than the accessible delay.

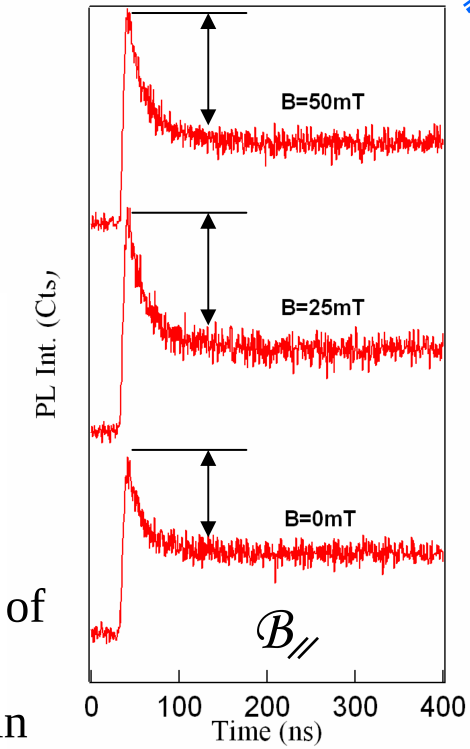


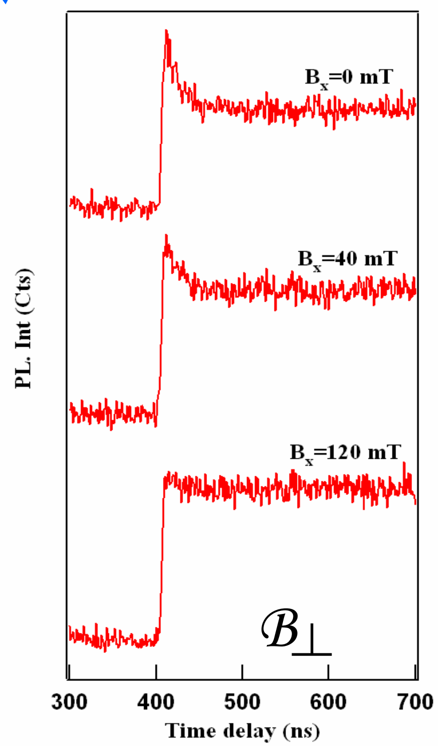

- Spin relaxation time is **not an intrinsic property** of the Mn atom…
influence of the **local Mn environment.**

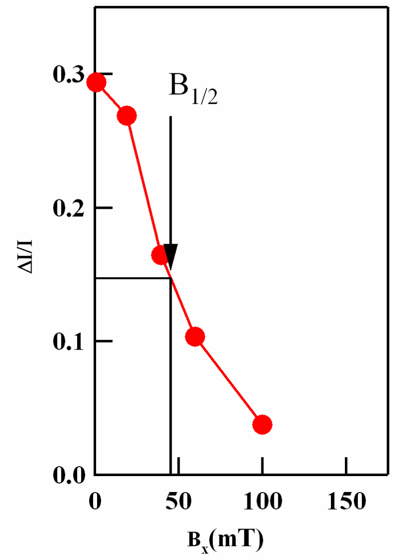
# *Optical evidence of the Mn fine structure*




● B Faraday:

Destroy the influence of tetragonal c.f and or small anisotropic strain

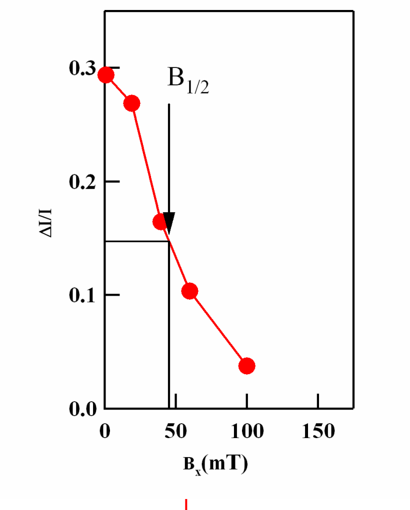
$S_z$ already stabilized by $D_0$

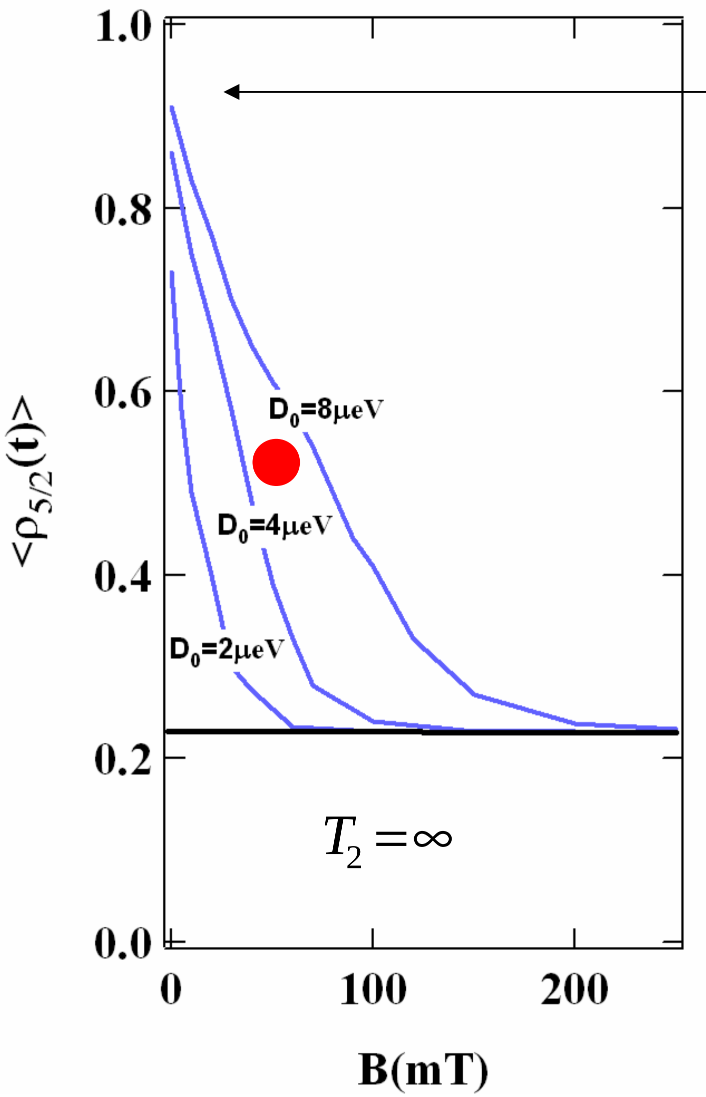
● B Voight:

Decrease of the pumping efficiency: precession of the Mn spin in the transverse B field.

BUT: precession blocked by $\mathcal{H} = D_0 S_z^2$

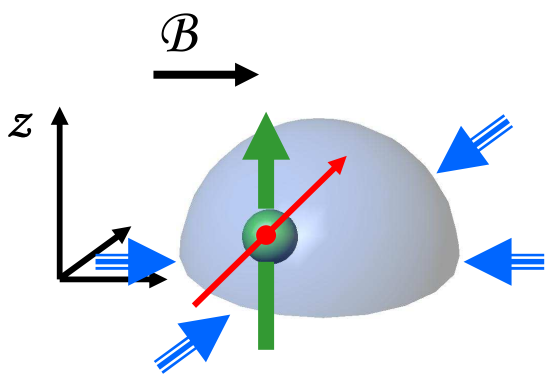
# *Optical measurement of the magnetic anisotropy*




$S_z$ remain eigenstates at weak transverse B field.

Average for a free precessing spin




$\mathcal{H} = D_0 S_z^2 + AI.S + c.f$

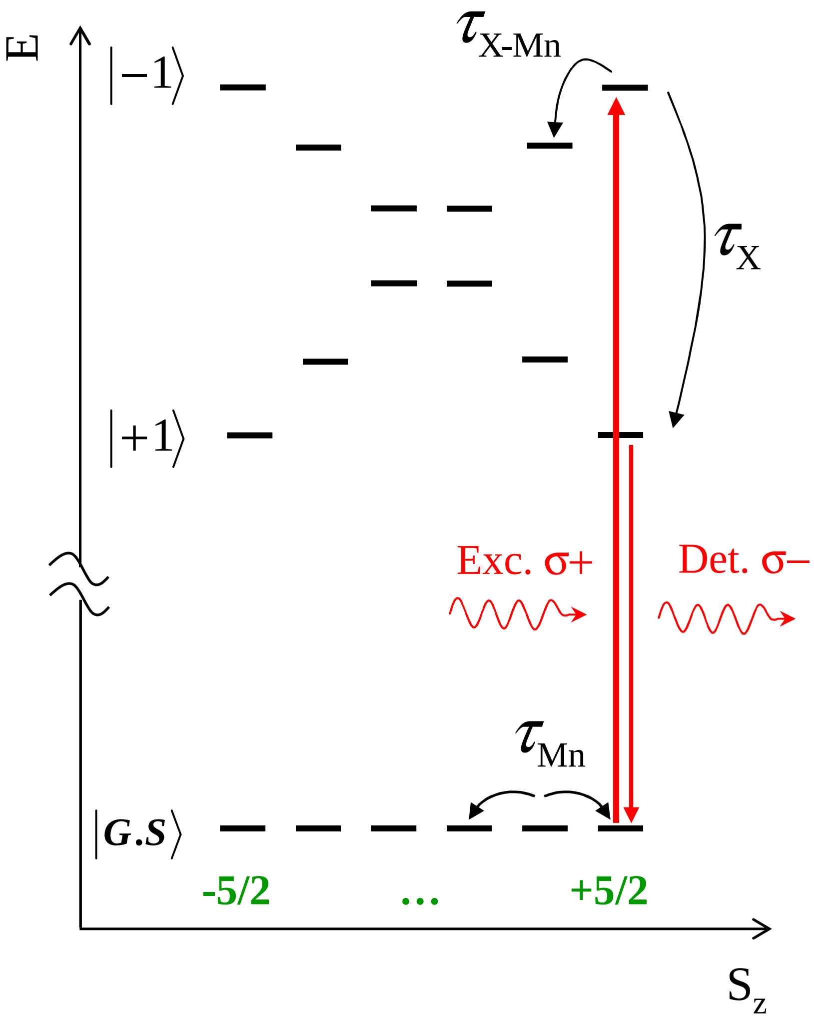
$D_0 \approx 6\mu eV$

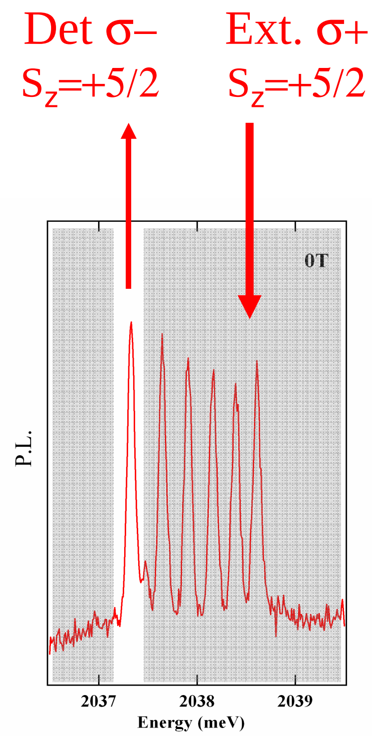
($D_0$ max for CdTe on ZnTe:12μeV)

Resonant excitation on a X-Mn level: Spin selectivity.

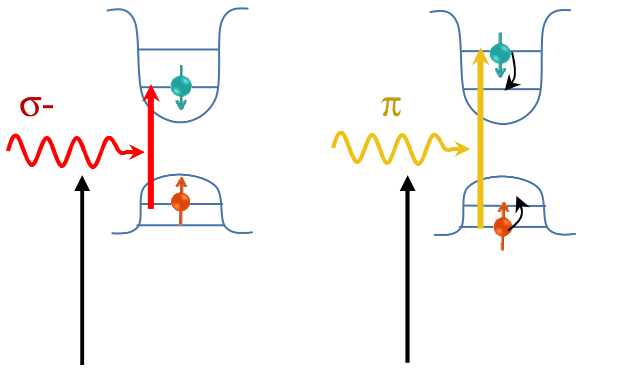
Pumping condition: $\tau_{X\text{-}Mn} < \tau_{Mn}$

Reading condition: $\tau_{X} < \tau_{X\text{-}Mn}$



# *Resonant optical pumping*

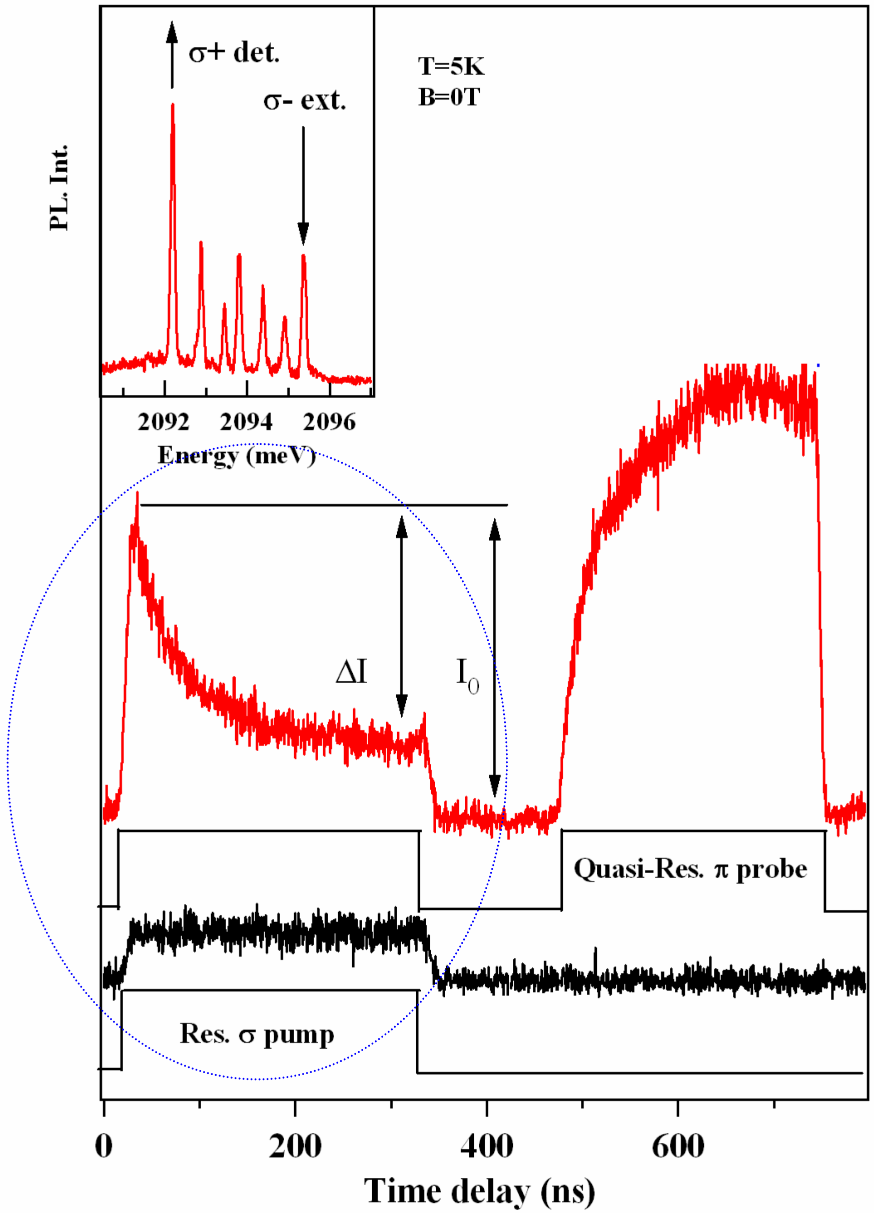



Resonant excitation:
- empty $S_z$=-5/2

Quasi resonant excitation:
- destroy the pumping

Excitation sequence

Optical pumping signal detected on the resonant fluorescence



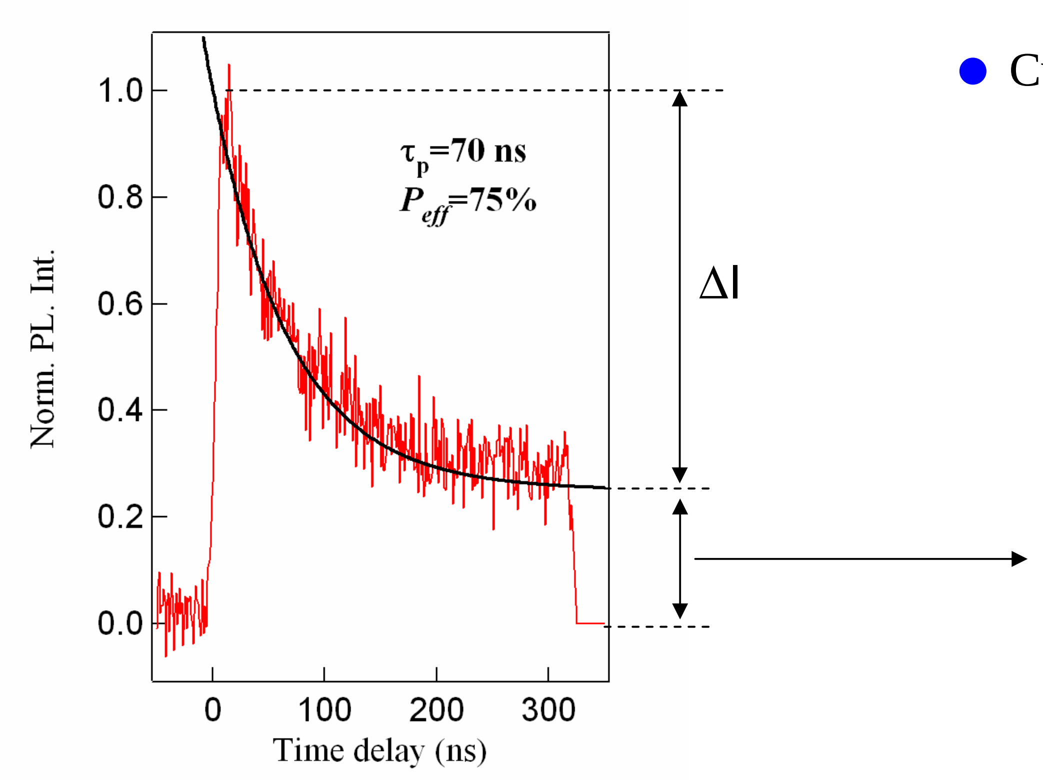

Quasi Res ON

Quasi Res OFF

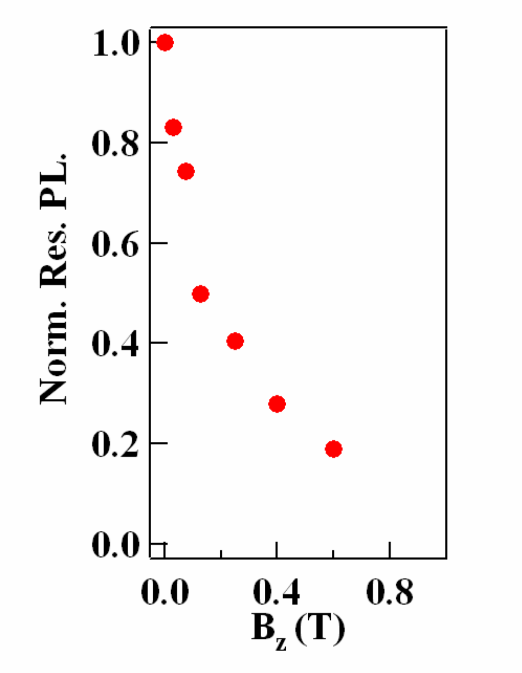
*Phys. Rev. B 81, 245315 (2010)*

# *Resonant optical pumping efficiency*

- Time resolved resonant fluorescence:




- Cw resonant fluorescence under $B_z$:



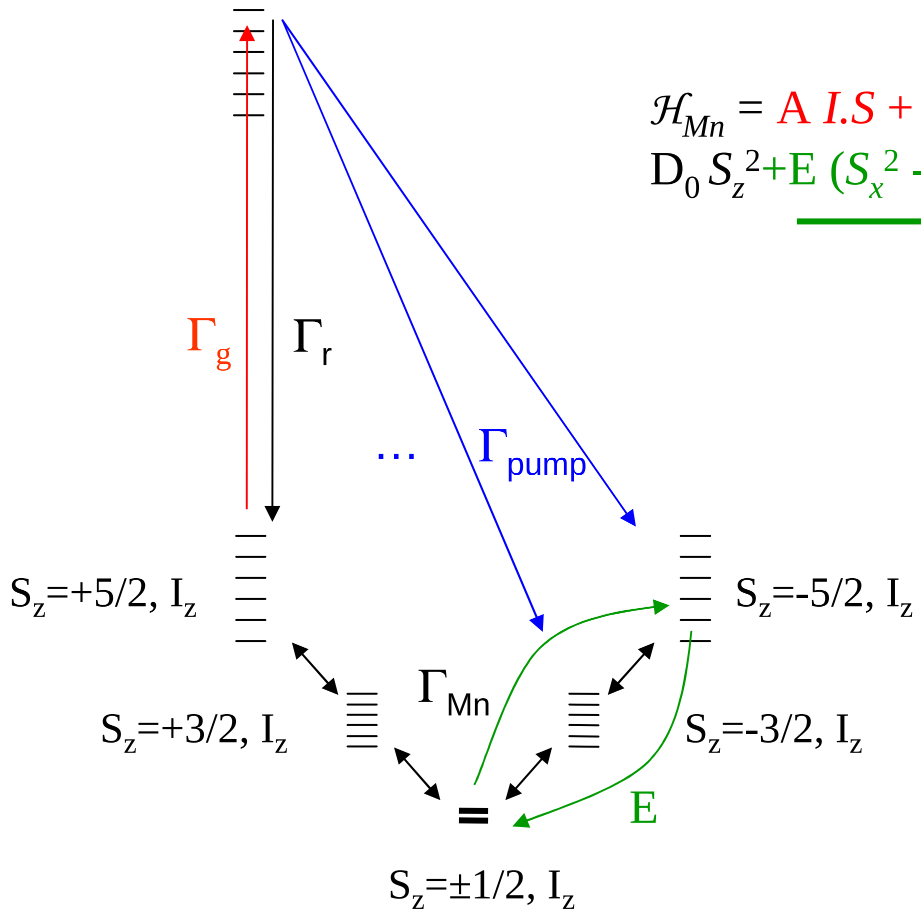

Maximum pumping efficiency: around 75% in the best case?

Decrease of the res. fluo. intensity under $B_z$ of a few tens of mT:
Enhancement of the optical pumping efficiency.

# *Influence of coherent dynamics of the Mn spin on the optical pumping*

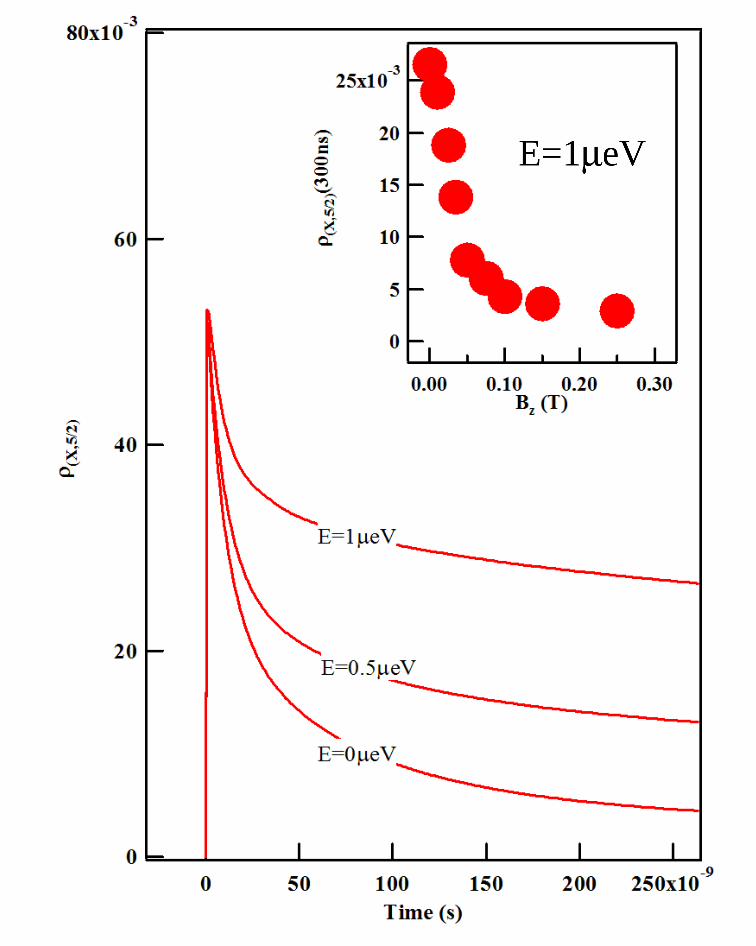
- Model of optical pumping including the coherent dynamics of the Mn:

$$\mathcal{H}_{Mn} = A\, I.S + a\,(S_z^4+S_y^4+S_x^4) + D_0 S_z^2 + E\,(S_x^2 - S_y^2) + g_{Mn}\mu_B B_z.S_z$$



*Influence of coherent dynamics of the Mn spin on the optical pumping*

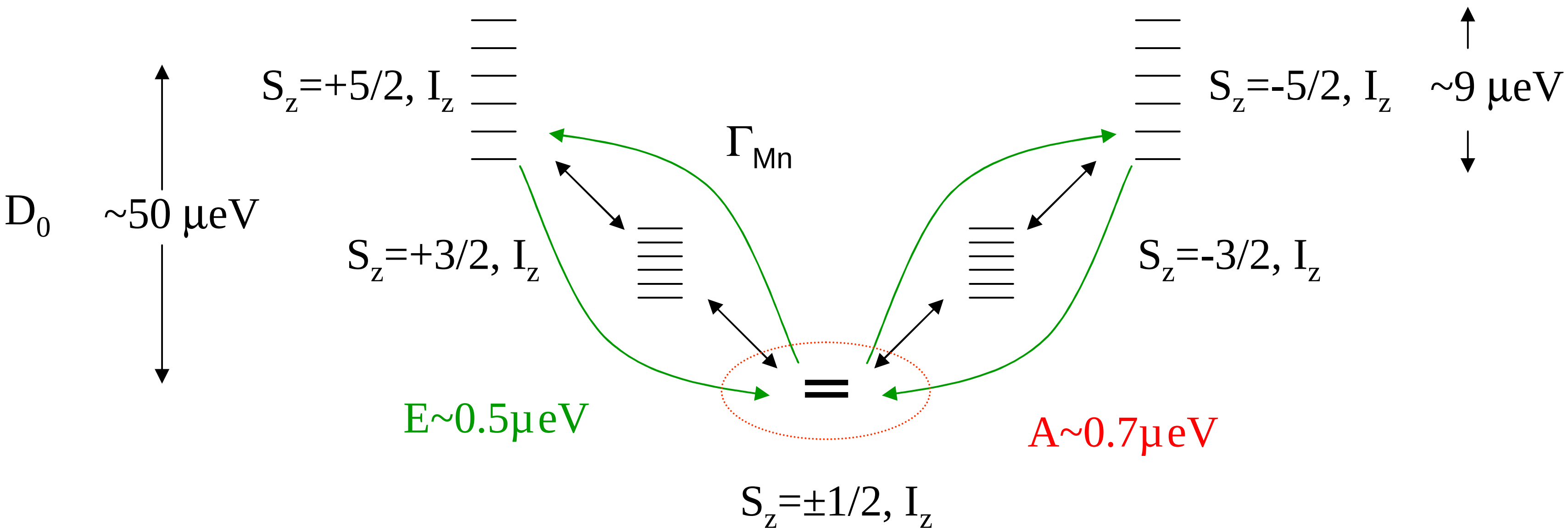
$D_0$=6μeV,
a=0.32μeV,
A=0.7 μeV
$T_{pump}$=20ns,
$T_g$=0.5ns,
$T_{Mn}$=1μs,
$T_r$=0.25ns




E around 0.5 μeV with $D_0$=6μeV

- Strain in-plane anisotropy (E) is the main parameter responsible for the limit of the optical pumping efficiency.

# *$Mn^{2+}$ fine and hyperfine structure in a strained QD*




Mn local environment:

$D_0$: biaxial strain

E: in-plane strain anisotropy

A: hyperfine coupling

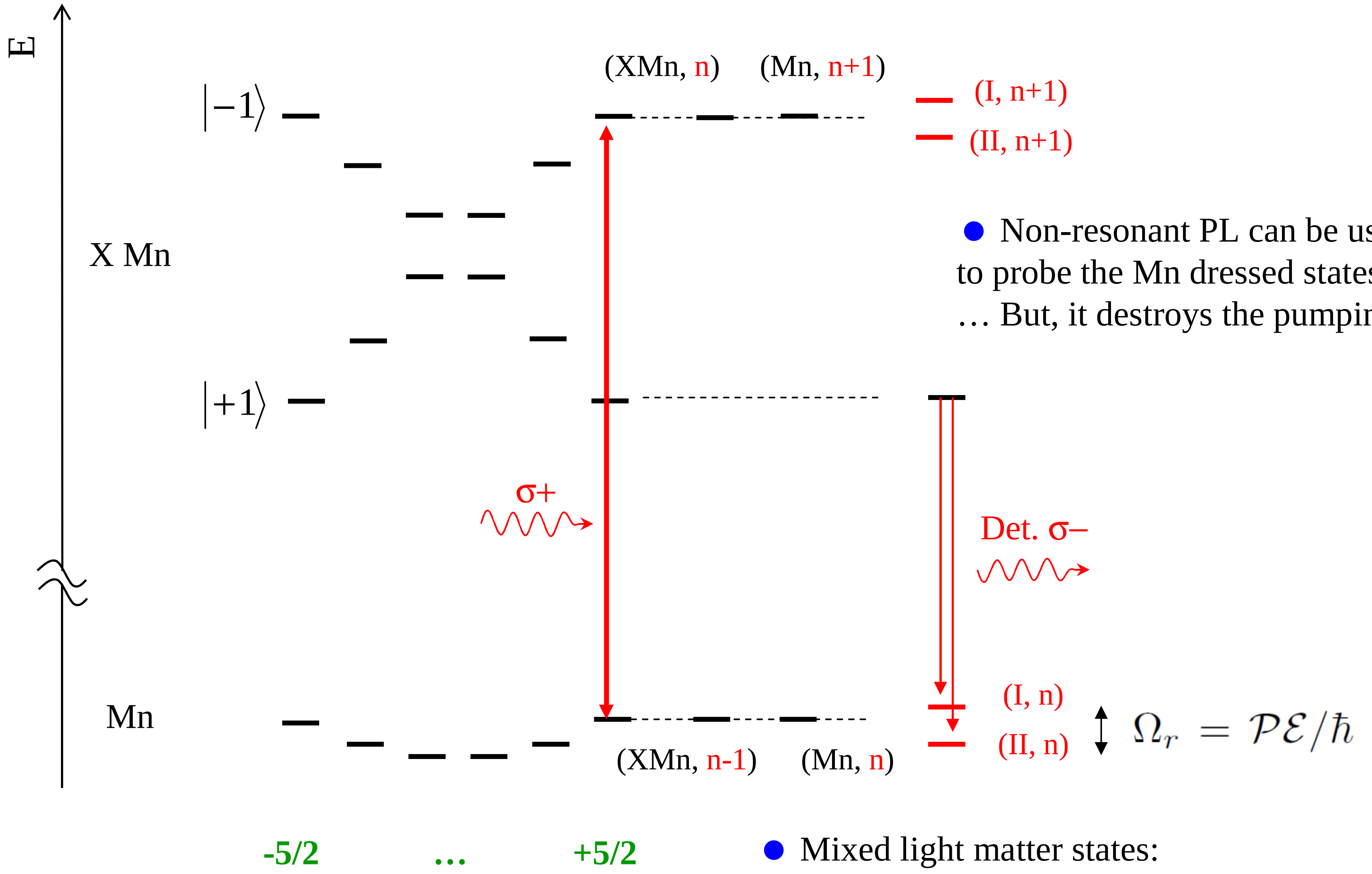
# *Outline*

1. *A II-VI quantum dot as a tool to optically probe the spin state of individual magnetic atoms (1 or 2 Mn).*

2. *Single Mn spin dynamics:*
   *Mn spin memory: strained induced magnetic anisotropy*
   *Optical initialization and readout of an individual Mn spin*

3. *Spin dynamics of optically dressed Mn atoms:*
   *Optical Stark effect on an individual Mn spin*
   *Spin population trapping.*

# *Optically dressed state in a Mn doped quantum dot*

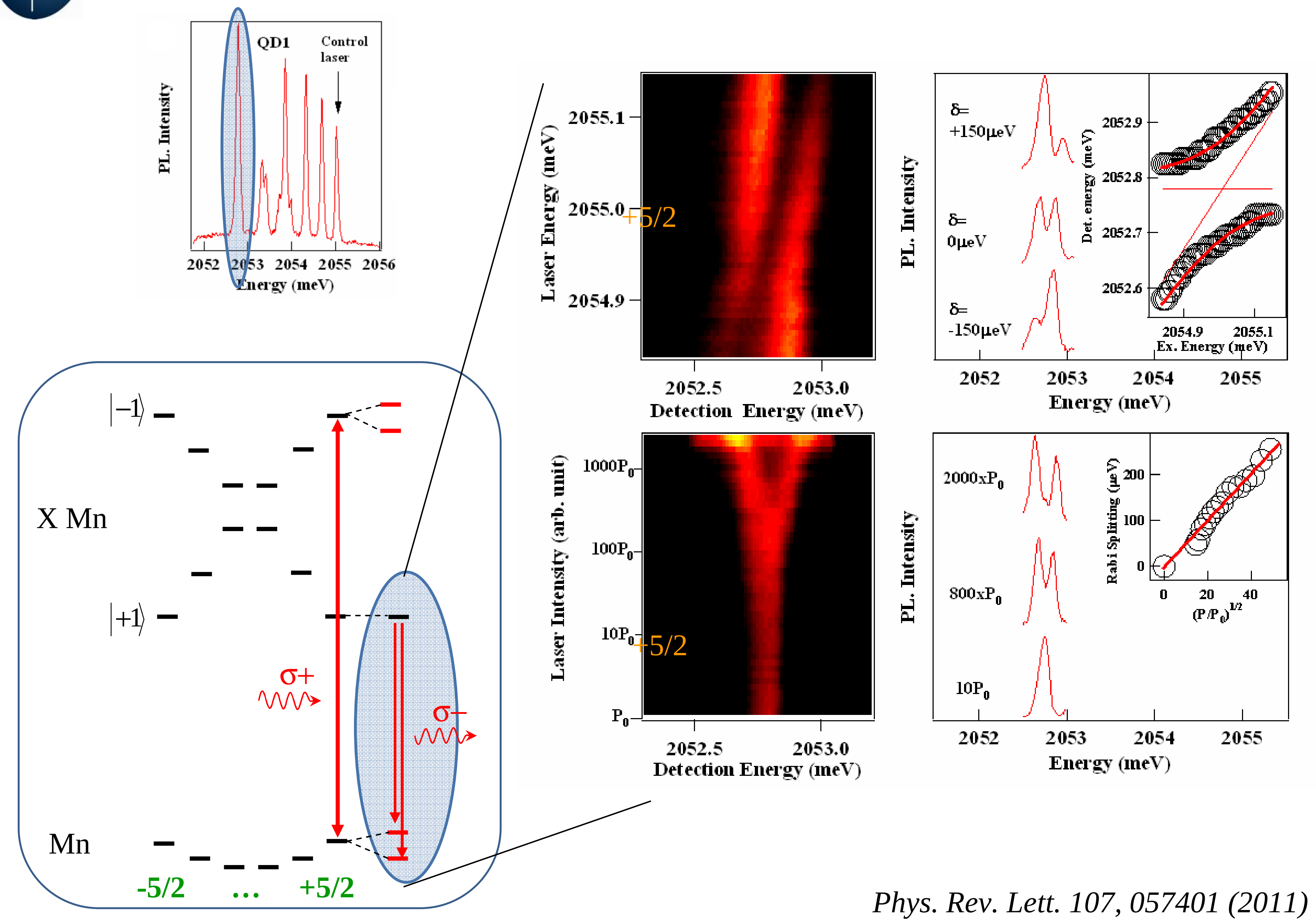



- Non-resonant PL can be used to probe the Mn dressed states. … But, it destroys the pumping

- Mixed light matter states:

$$|I,n\rangle = c|Mn\rangle \otimes |n\rangle - s|XMn\rangle \otimes |n-1\rangle$$
$$|II,n\rangle = s|Mn\rangle \otimes |n\rangle + c|XMn\rangle \otimes |n-1\rangle$$

# *Optically dressed Mn-doped QD*

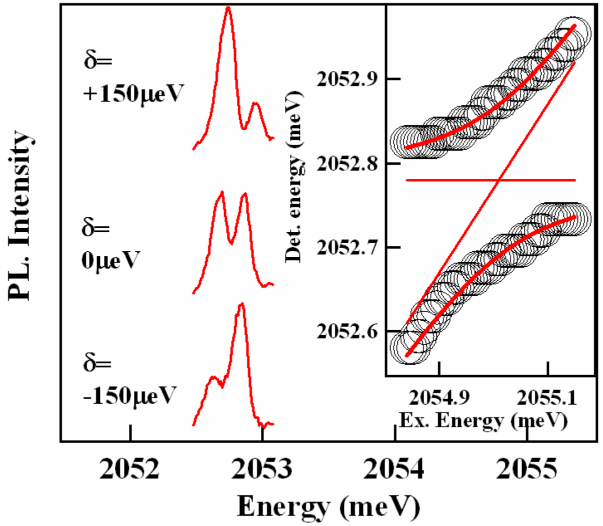


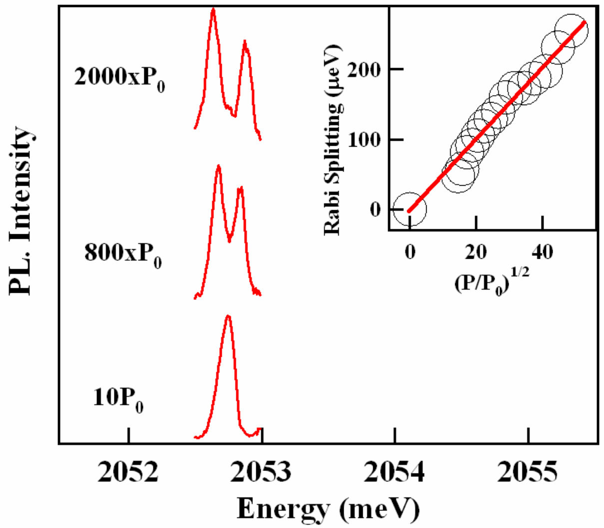

*Phys. Rev. Lett. 107, 057401 (2011)*

# Optically dressed Mn-doped QD

- Large X-Mn exchange interaction:

Each X-Mn transition behaves like a two levels system.




Laser detuning:

$$\delta = \omega_L - \omega_0$$

$$\Delta E_{\pm} = \frac{\hbar}{2}\left( -\delta \pm \sqrt{\delta^2 + \Omega_r^2} \right)$$

Power dependence

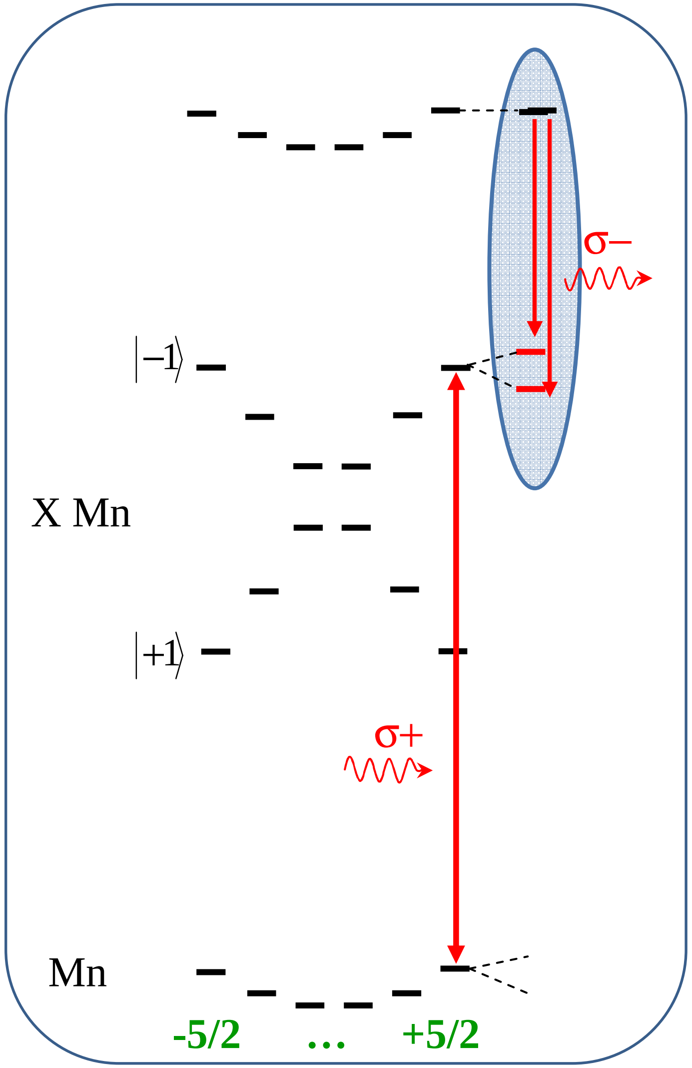
$$\hbar\Omega_r = -d.E$$

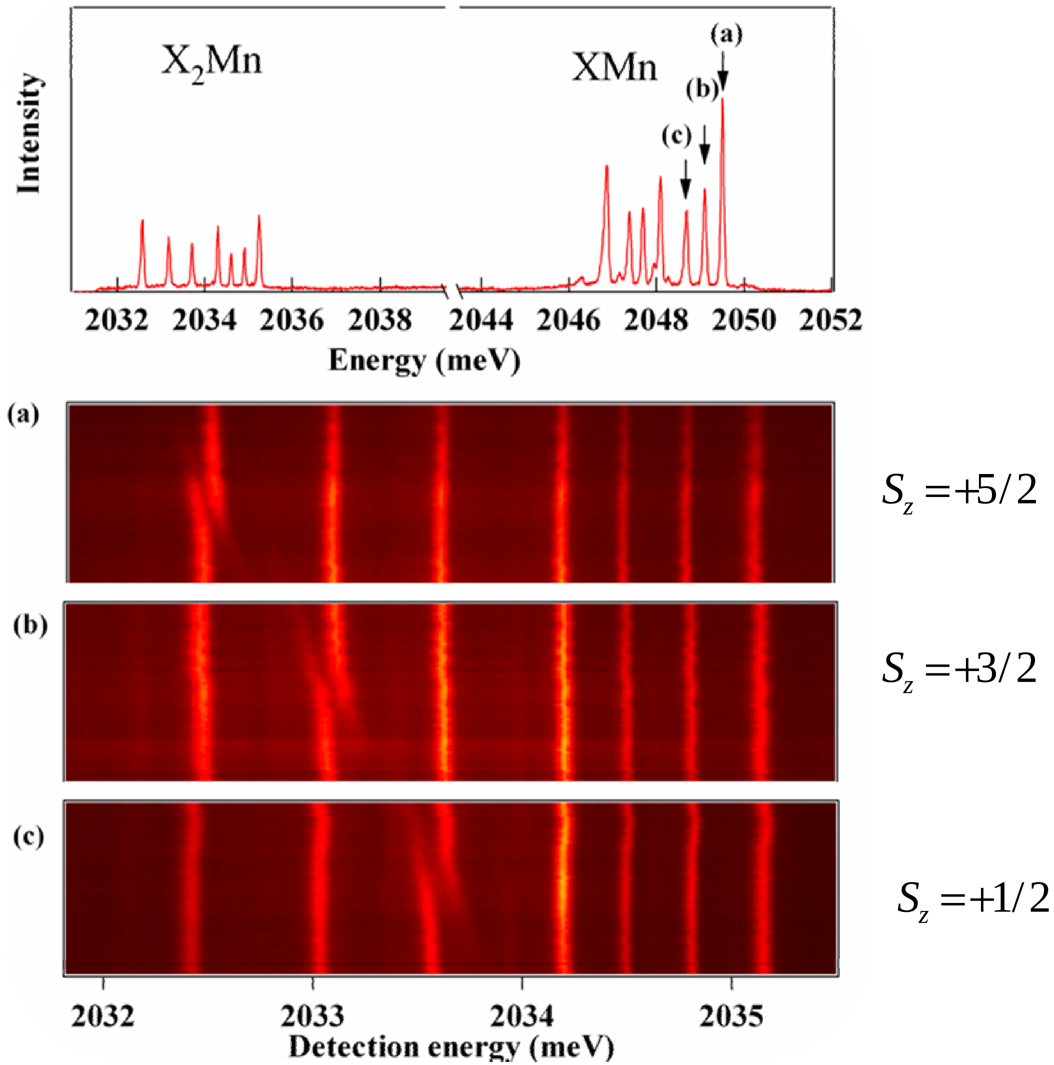
$$\hbar\Omega_r \propto P^{1/2}$$

Can reach 250µeV… much larger than the Mn fine structure

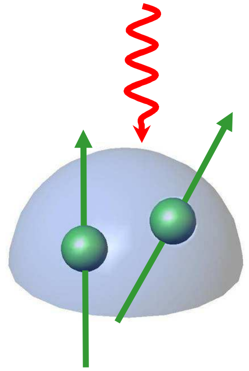
# *Optical addressing of any spin state of the Mn*

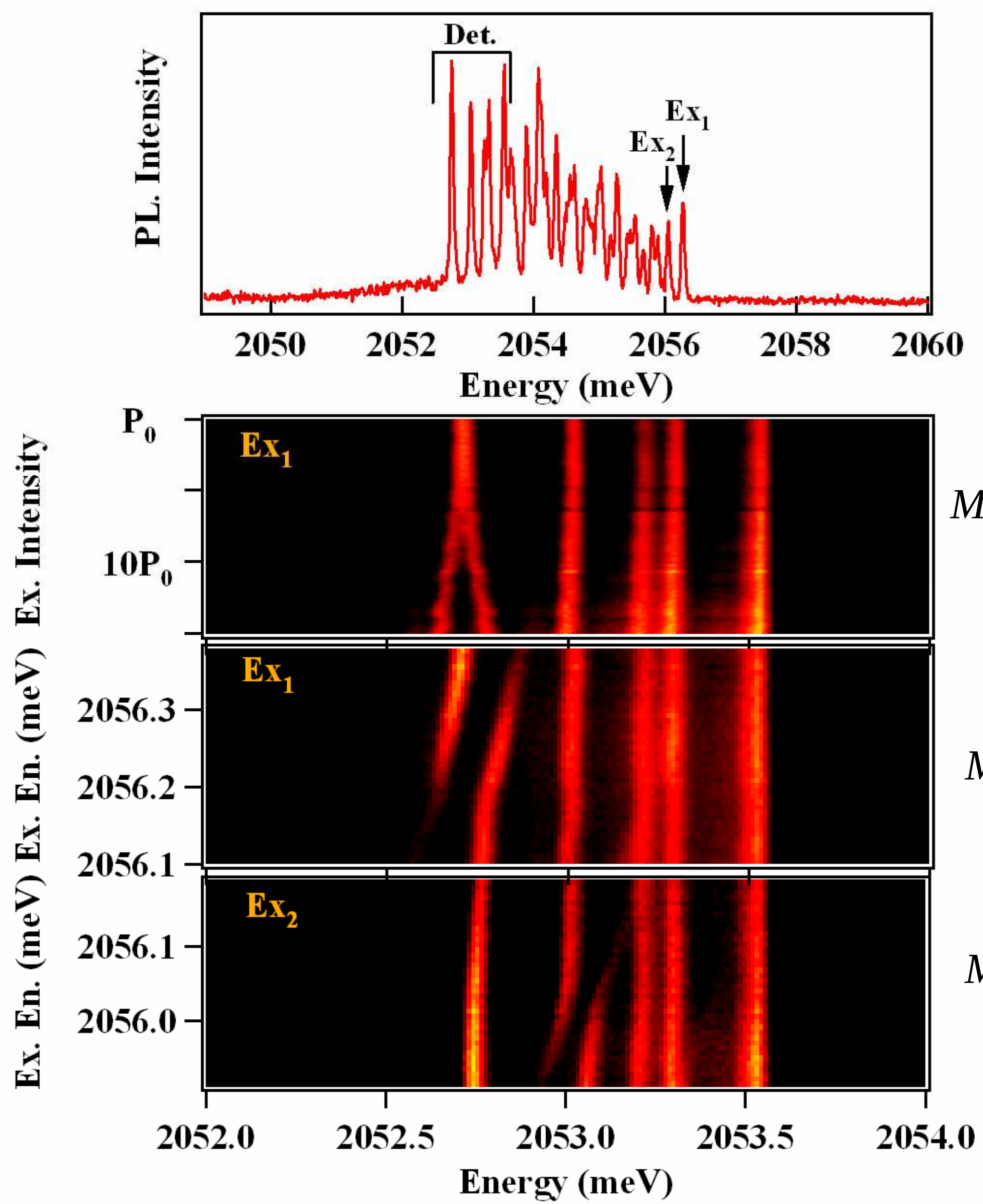
- Large X-Mn exchange interaction: Optically address any spin state of the Mn.



# *Optical Stark effect on the spin states of two Mn atoms*






- Large X-Mn exchange interaction for the two Mn: Optically shift any spin state of the 2 Mn.

- Intensity:
- Detuning:

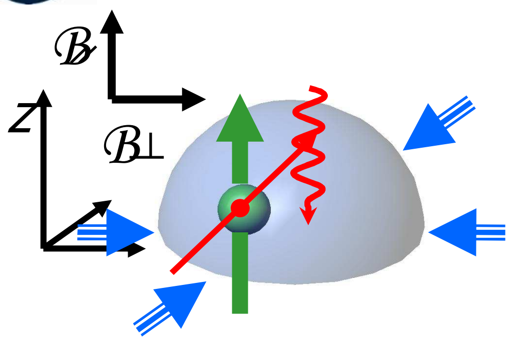
$M_z = +5$

$M_z = +5$

$M_z = +4$

Influence of an optical field on the Mn spins structure?

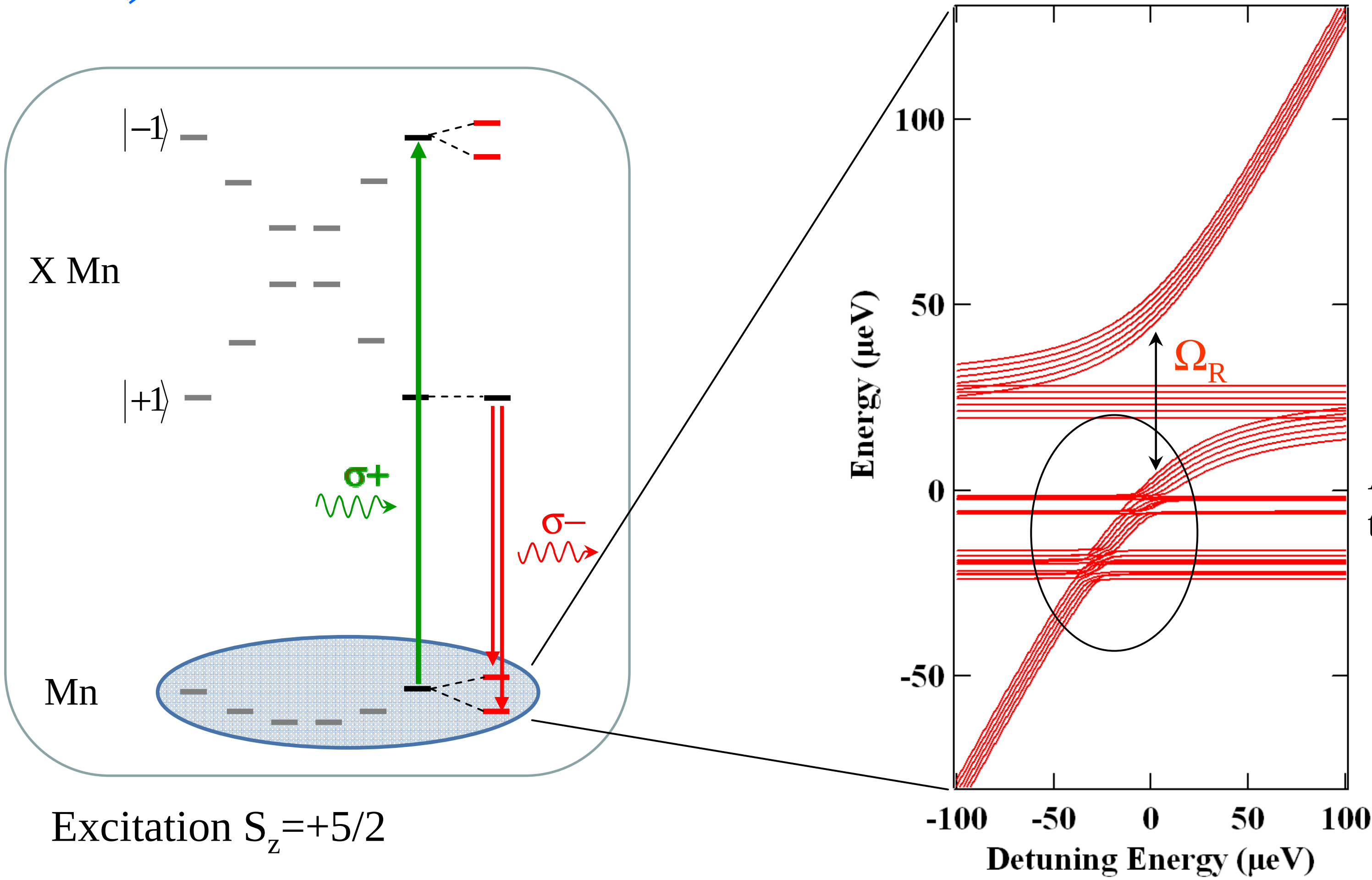
# *Laser controlled fine structure of the Mn*




$$\mathcal{H}_{Mn} = \mathcal{A}\,\vec{I}.\vec{S} + a[S_x^4 + S_y^4 + S_z^4] + \mathcal{D}_0[S_z^2] + E[S_x^2 - S_y^2]$$

$$\mathcal{H}_{af} = \hbar\Omega_R(ad^\dagger + a^\dagger d)$$

- Influence of an optical field resonant on $S_z=+5/2$: Consequence on the Mn spin dynamics?




Anti-crossing with the undressed states

Excitation $S_z=+5/2$

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   *Mn spin memory: strained induced magnetic anisotropy*
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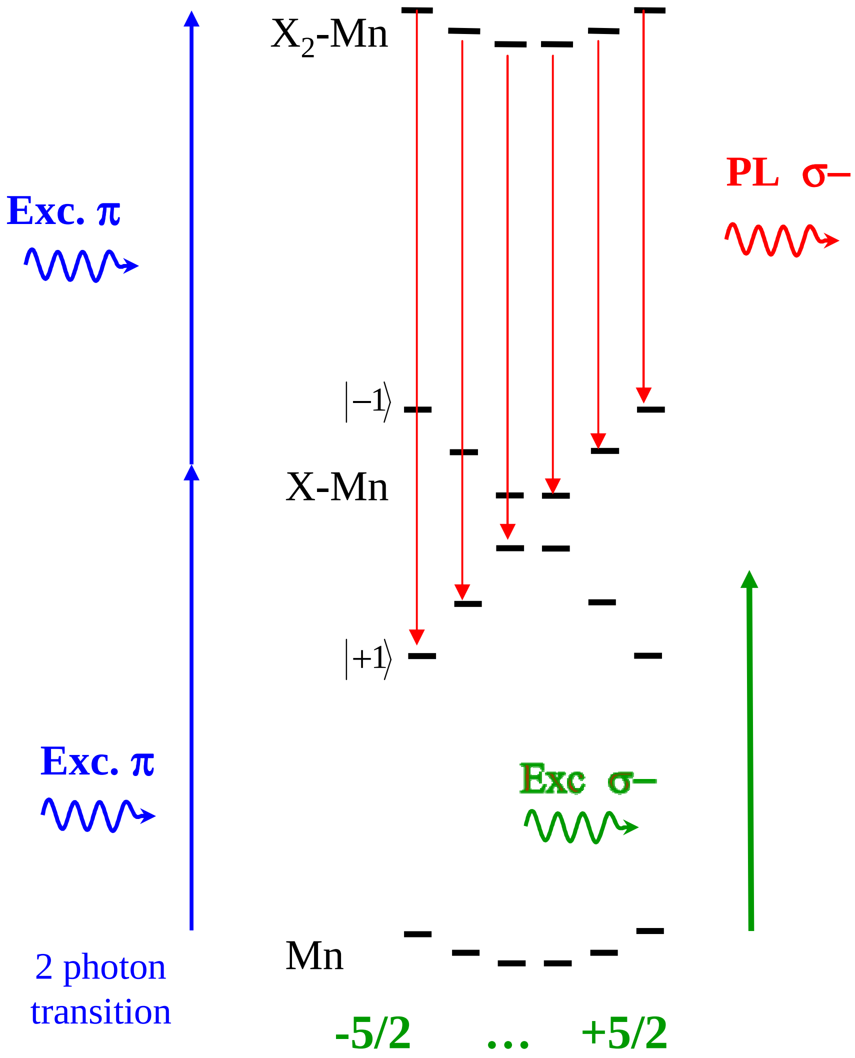
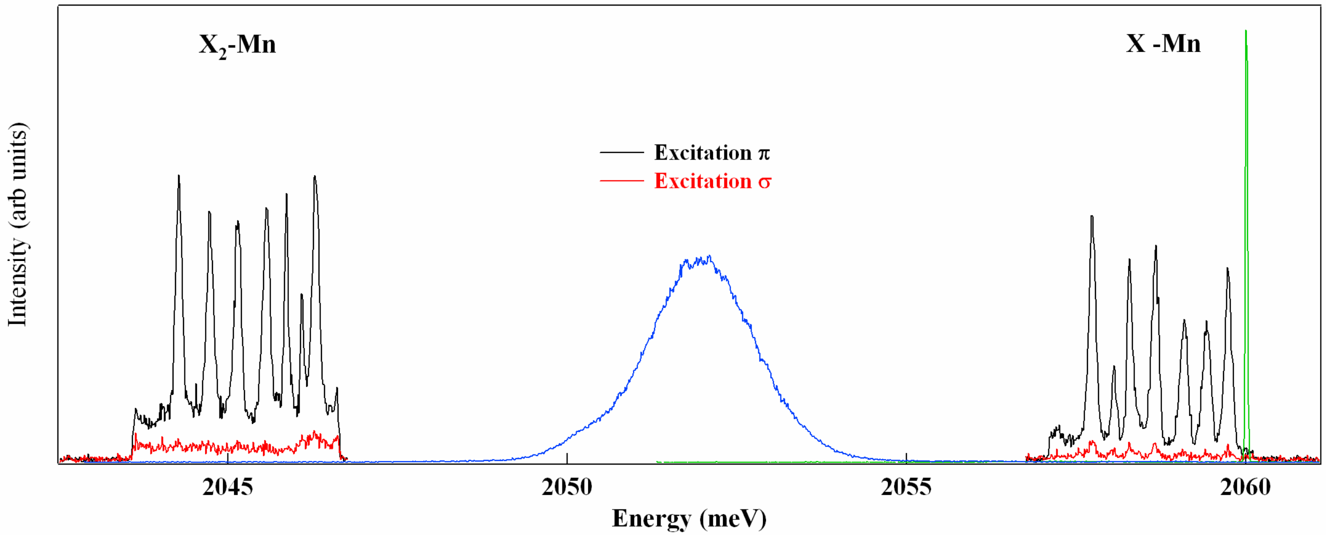
   *Optical Stark effect on an individual Mn spin*
   *Spin population trapping.*

# *Two photons detection of the Mn spin states population*




Resonant $X_2$ is a good probe: - Does not interact with the Mn.
- No free carriers.

# *Two photons detection of the Mn spin states population*

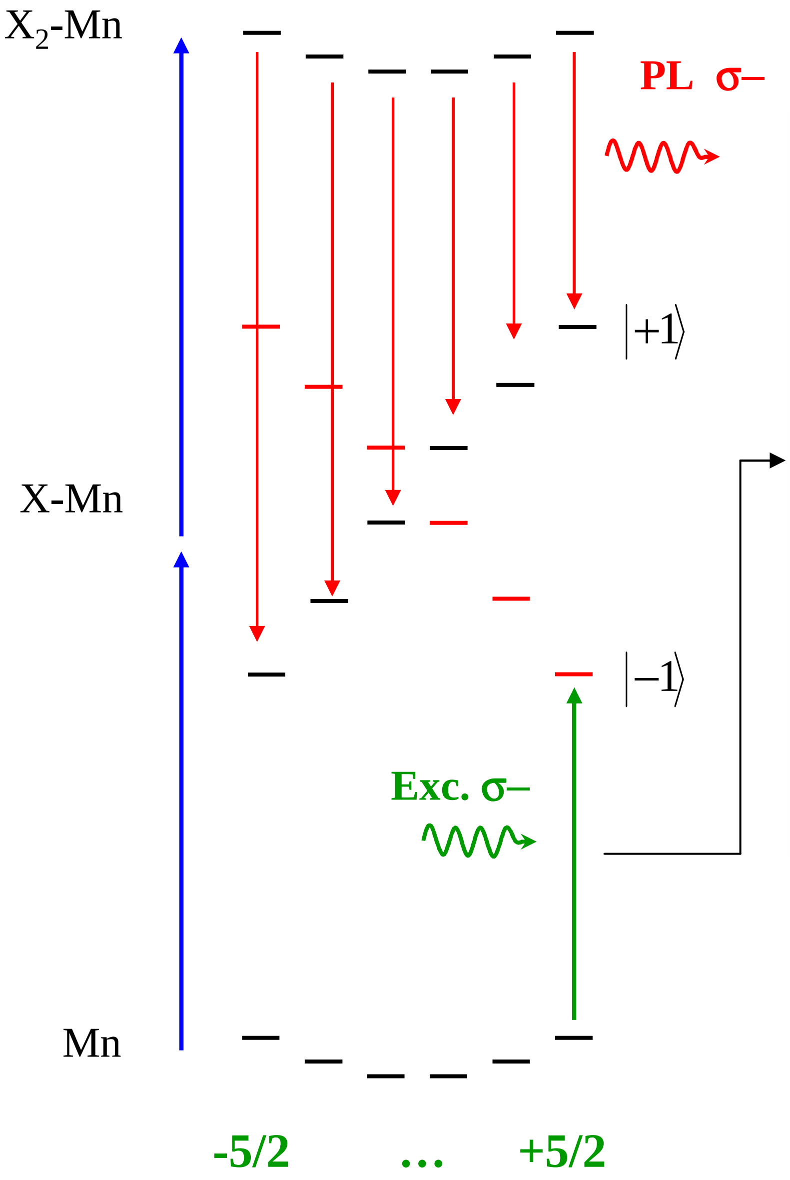



Two photons pulsed (ps) resonant excitation.

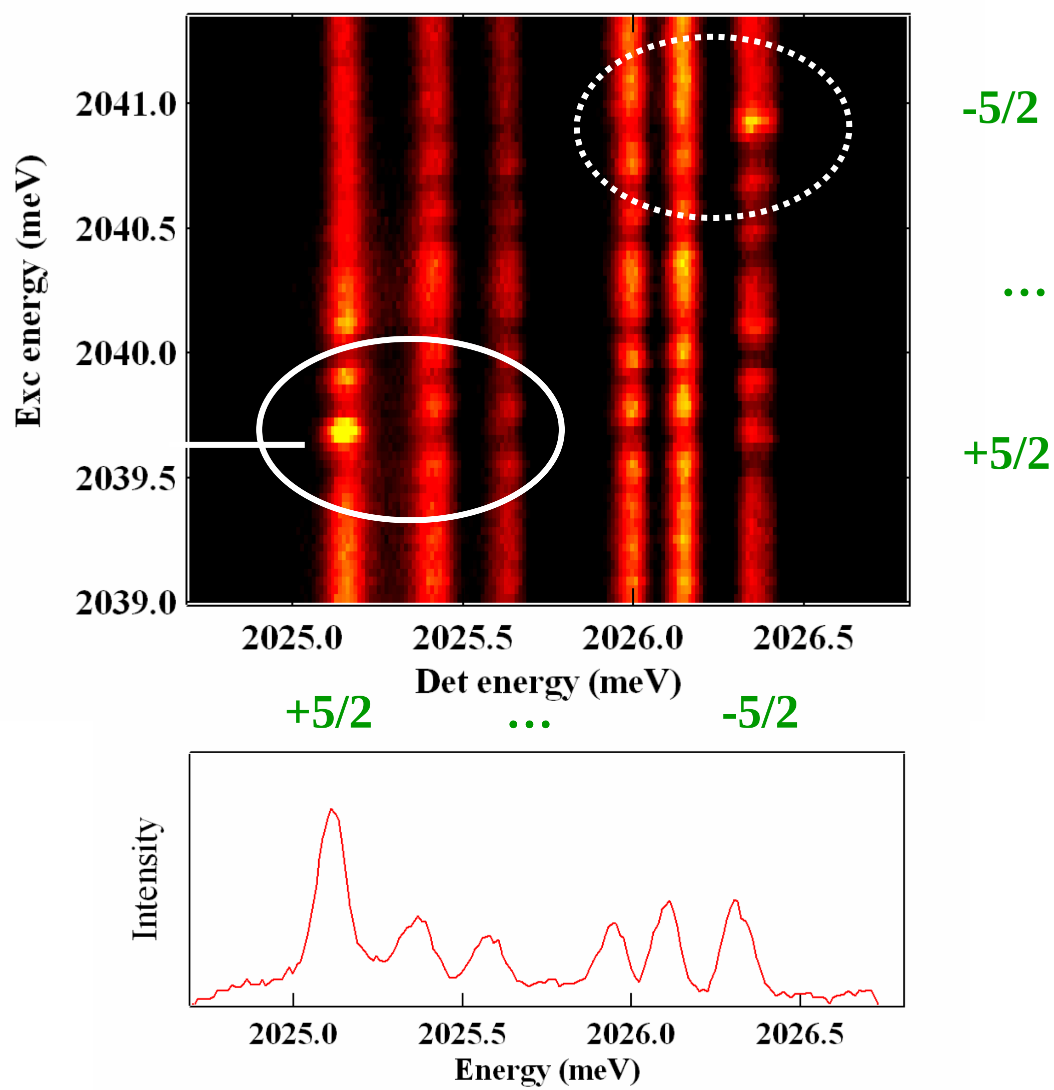
Tuneable cw resonant excitation

Mn spin population:
Intensity distribution in the two-photon PL of the biexciton

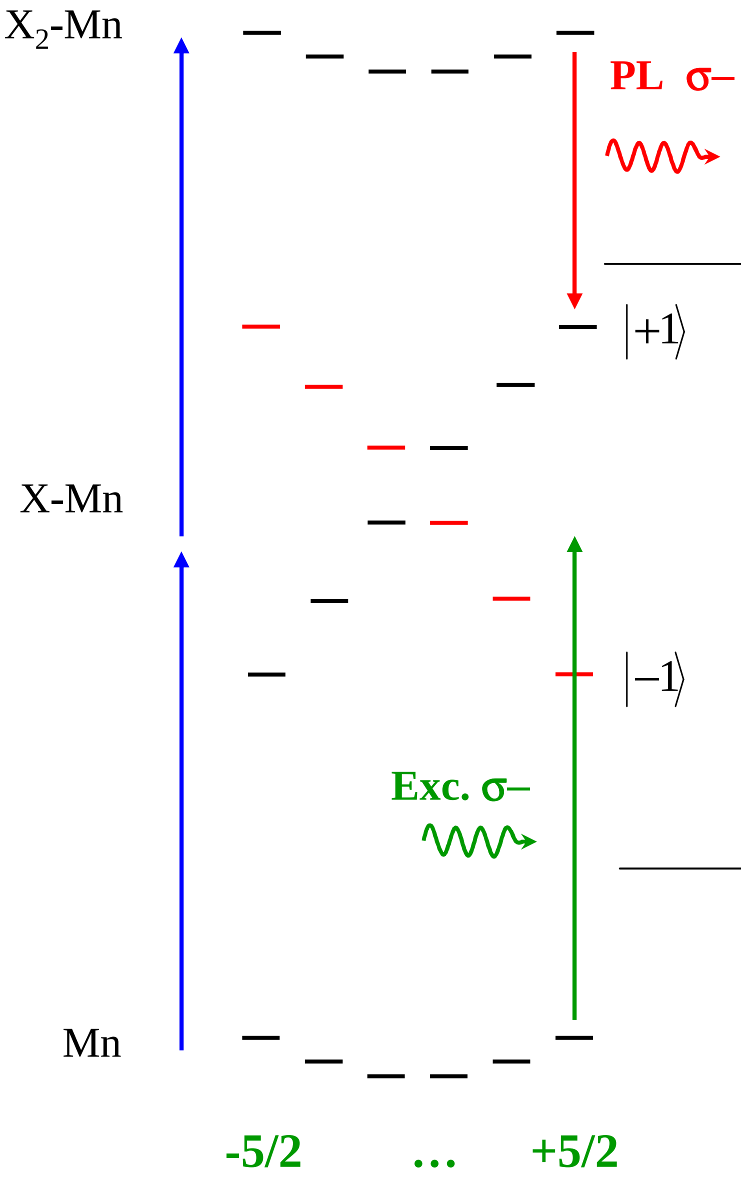
# Two-photon detection of the Mn spin states population



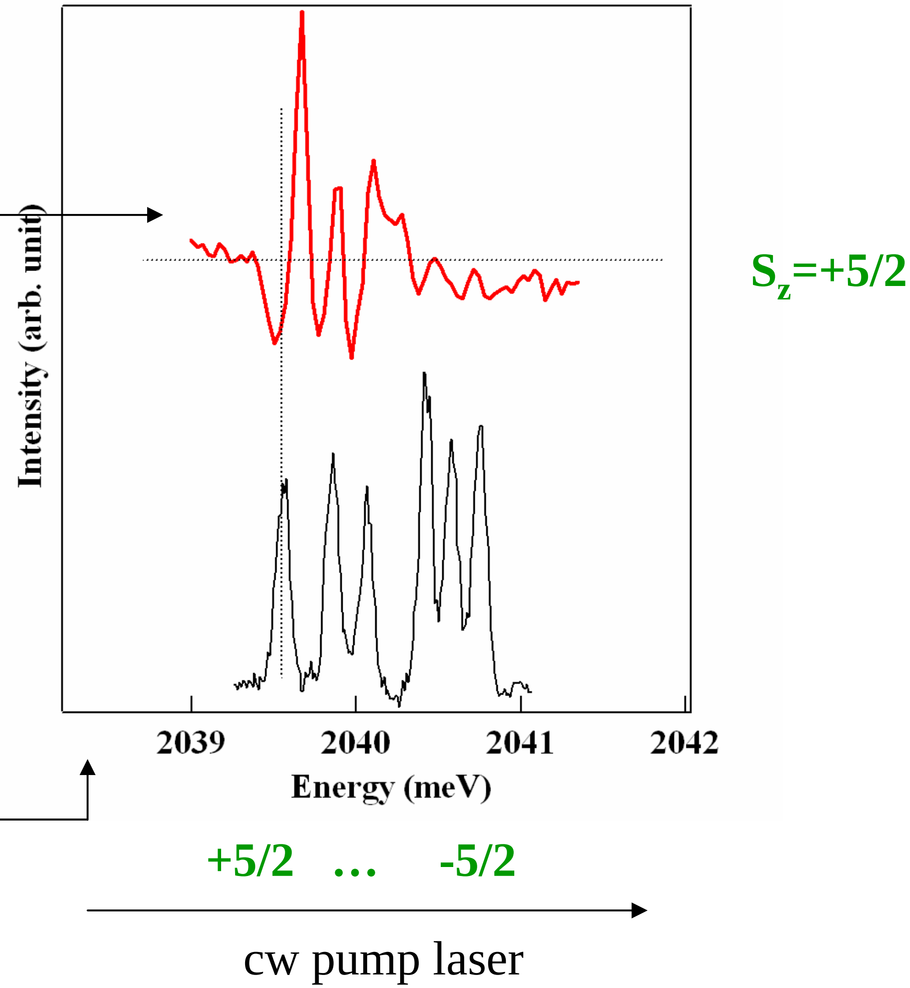

- Two photons PL of $X_2$:



# *Mn spin redistribution under resonant excitation*

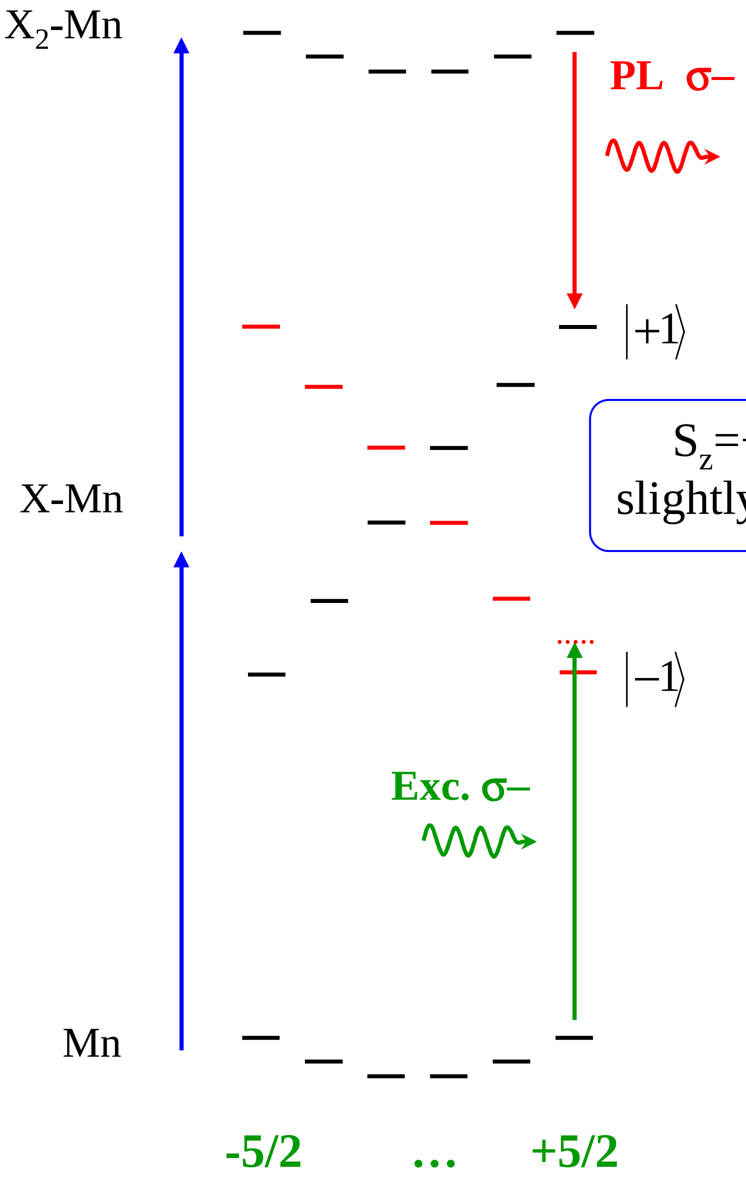



- Two photons detection of $S_z$=+5/2 population:

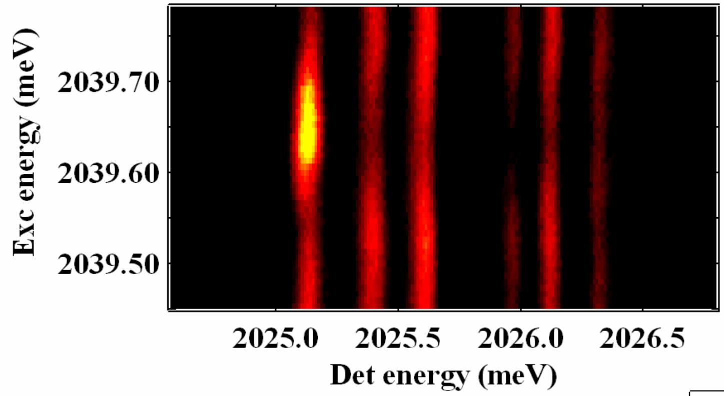


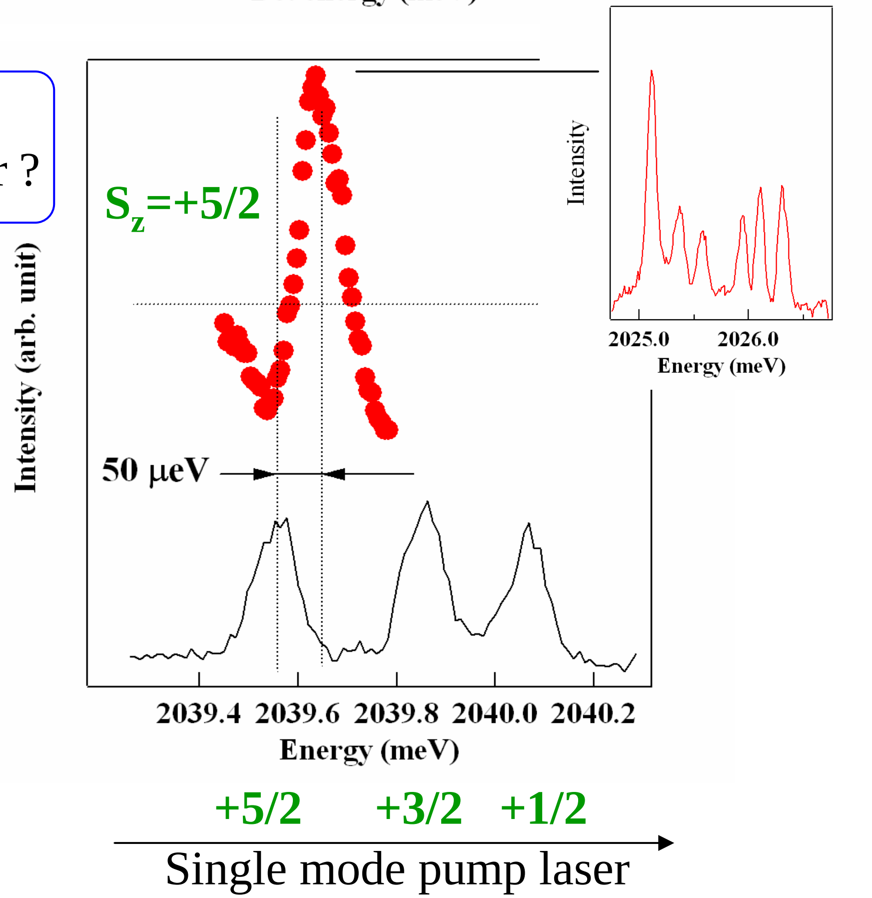

Population of $S_z$=+5/2 increases when we excite near $S_z$=+5/2?

# *Mn spin redistribution under resonant excitation*




$S_z$=+5/2 max for slightly detuned laser ?



# *Optical pumping*

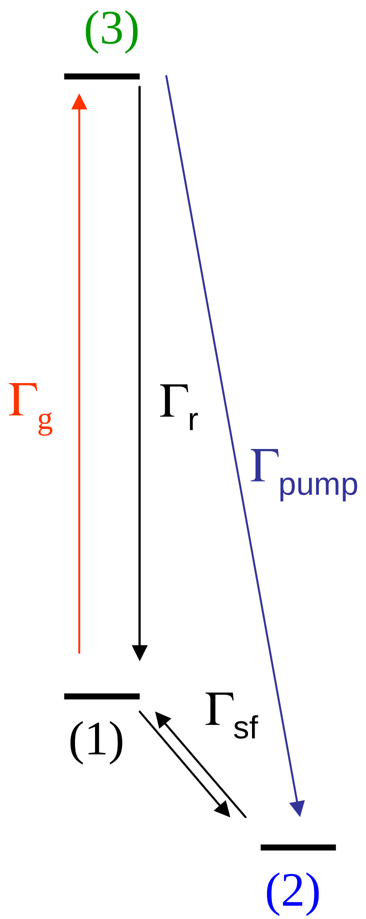
Optical pumping: empty the state resonantly excited (even with incoherent light).

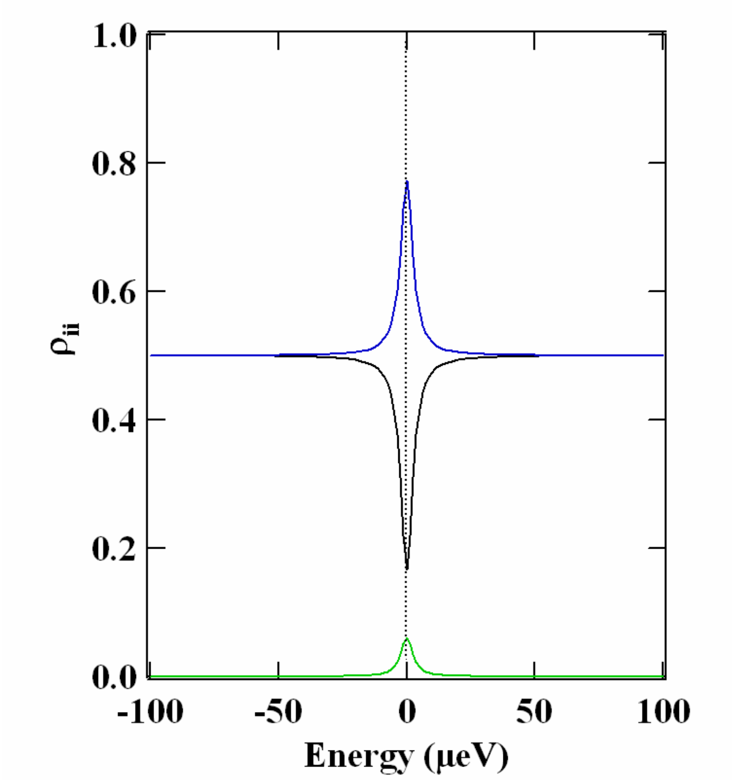
$\Gamma_{sf}$ spin flip rate

$\Gamma_{pump}$ optical pumping rate

$\Gamma_r$ optical recombination rate

$\Gamma_g$ optical generation rate




Condition for optical pumping:
$\Gamma_{pump} > \Gamma_{sf}$

# *Spin population trapping*

Spin coherent dynamics in the strong coupling regime:

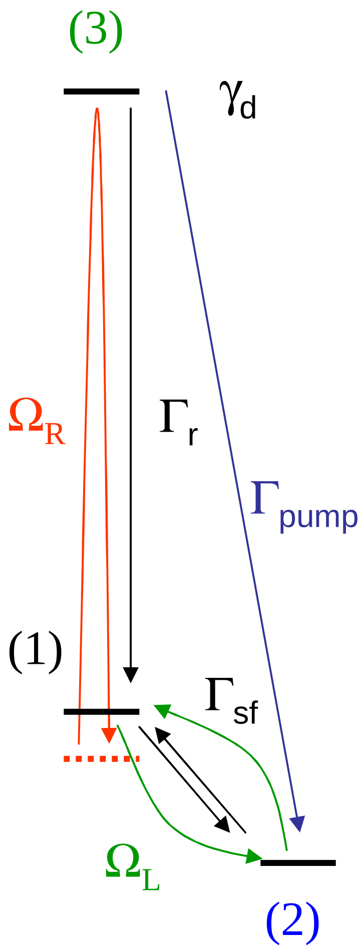
$\Omega_R$ Rabi frequency

$\gamma_d$ pure dephasing rate

$\Gamma_{sf}$ spin flip rate

$\Gamma_{pump}$ optical pumping rate

$\Gamma_r$ optical recombination rate



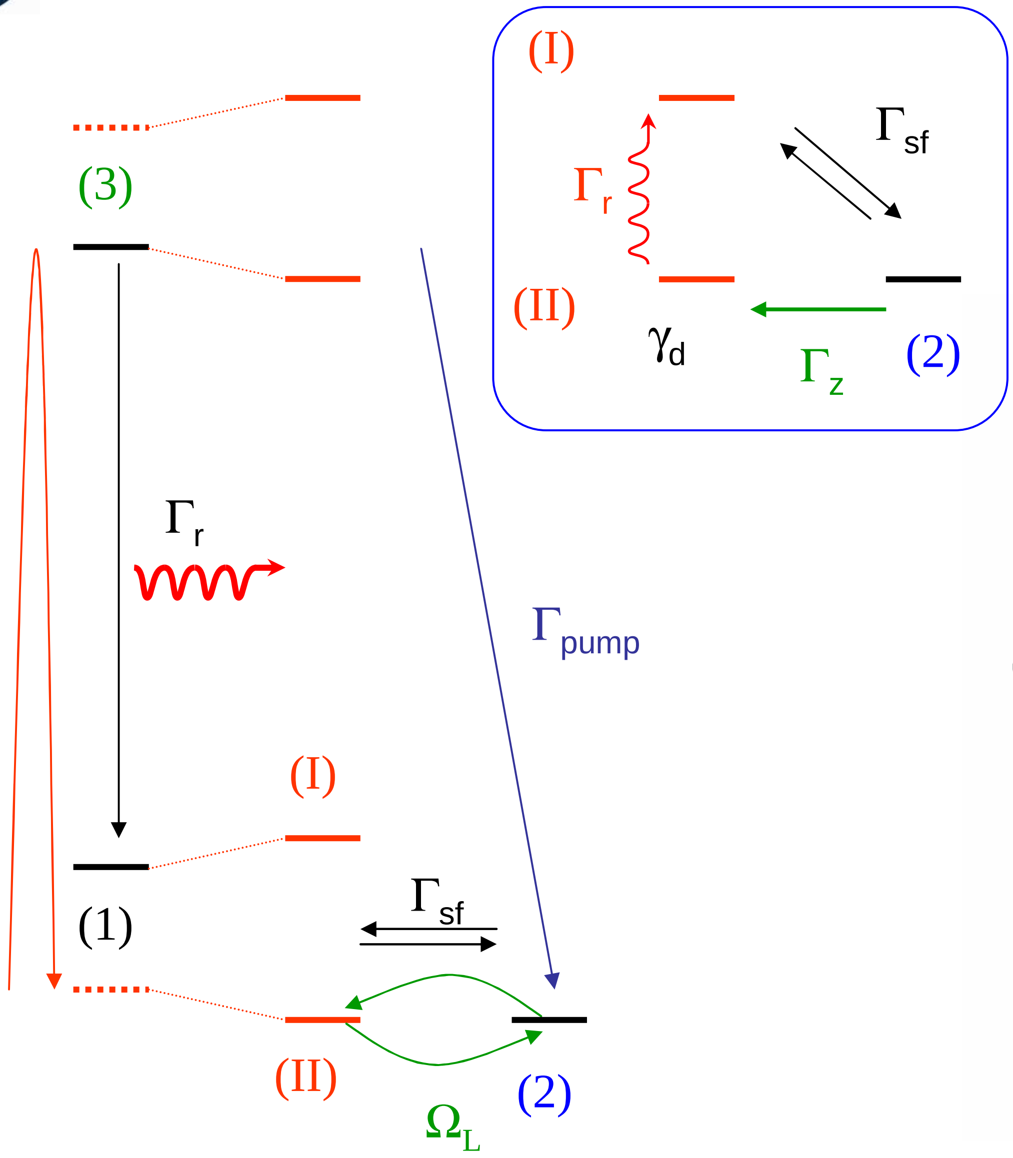

(3): fast dephasing

(1)&(2): long coherence time

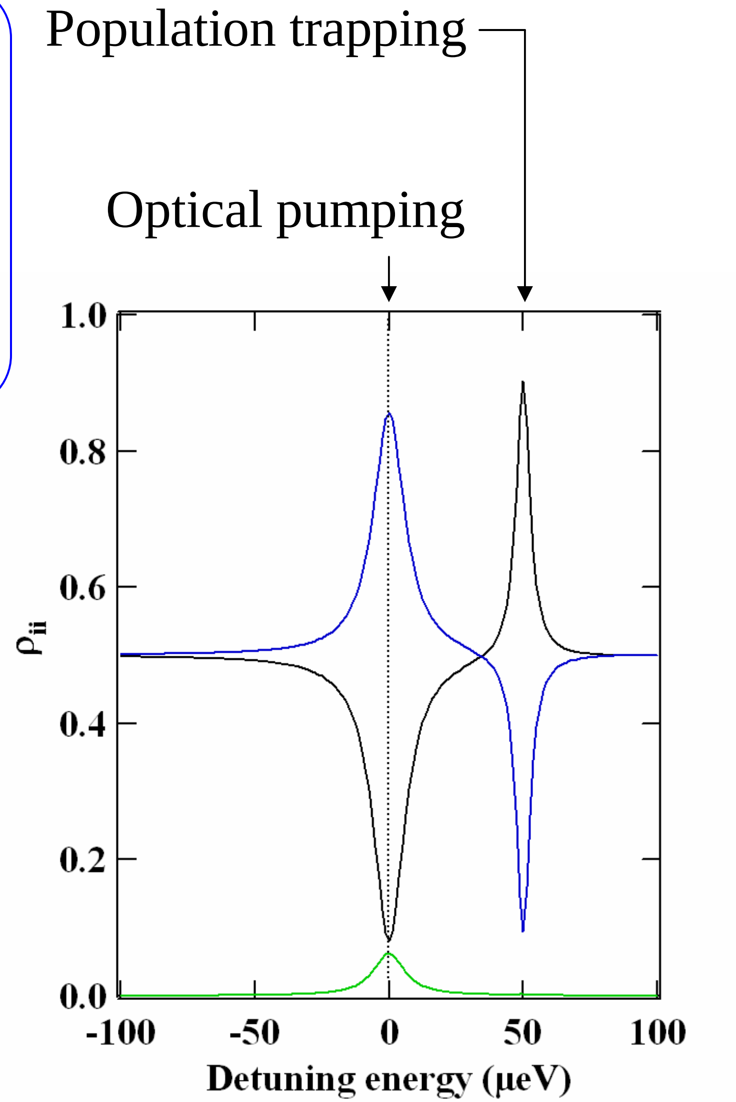
(1)-(3): optically allowed transition

$\Omega_L$ : Coherent coupling

# *Spin population trapping*




(II): fast dephasing




Optical initialization with off resonant light

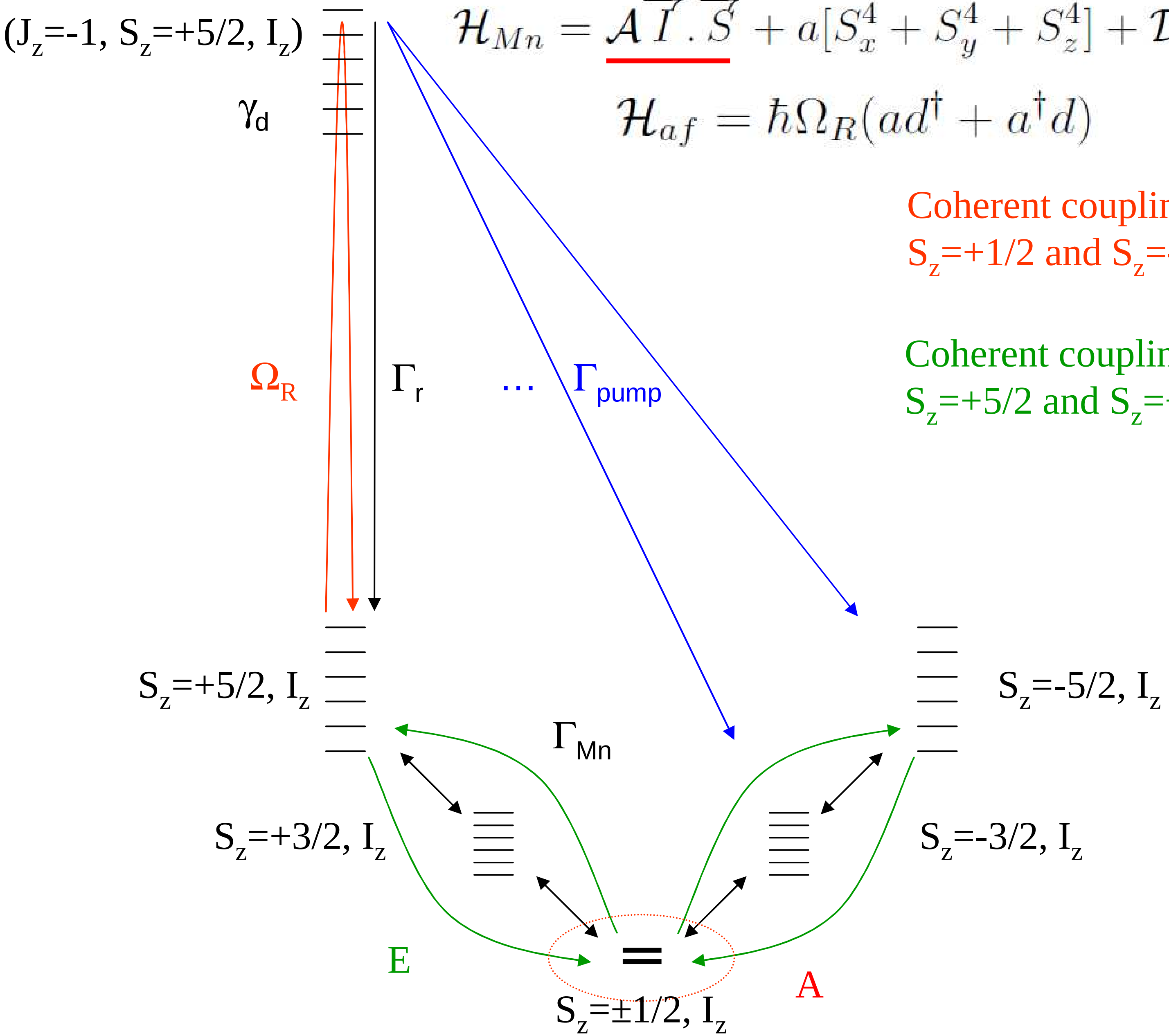
# *An optically dressed Mn atom (S=5/2,I=5/2)*

$$\mathcal{H}_{Mn} = \underline{\mathcal{A}\overrightarrow{I}.\overrightarrow{S}} + a[S_x^4 + S_y^4 + S_z^4] + \mathcal{D}_0[S_z^2] + \underline{E[S_x^2 - S_y^2]}$$

$$\mathcal{H}_{af} = \hbar\Omega_R(ad^\dagger + a^\dagger d)$$

Coherent coupling between $S_z$=+1/2 and $S_z$=-1/2

Coherent coupling between $S_z$=+5/2 and $S_z$=+1/2 …



# *Coherent dynamics of a multi-level system*

Master equation (Lindblad form):

$$\frac{\partial \varrho}{\partial t} = -i/\hbar[H, \varrho] + L\varrho$$

$[H, \rho]$: Hamiltonian evolution

$L\,\rho$ : coupling with the environment

Incoherent coupling:

$$L_{inc,j\to i}\varrho = \frac{\Gamma_{j\to i}}{2}(2|i\rangle\langle j|\varrho|j\rangle\langle i| - \varrho|j\rangle\langle j| - |j\rangle\langle j|\varrho)$$

Pure dephasing:

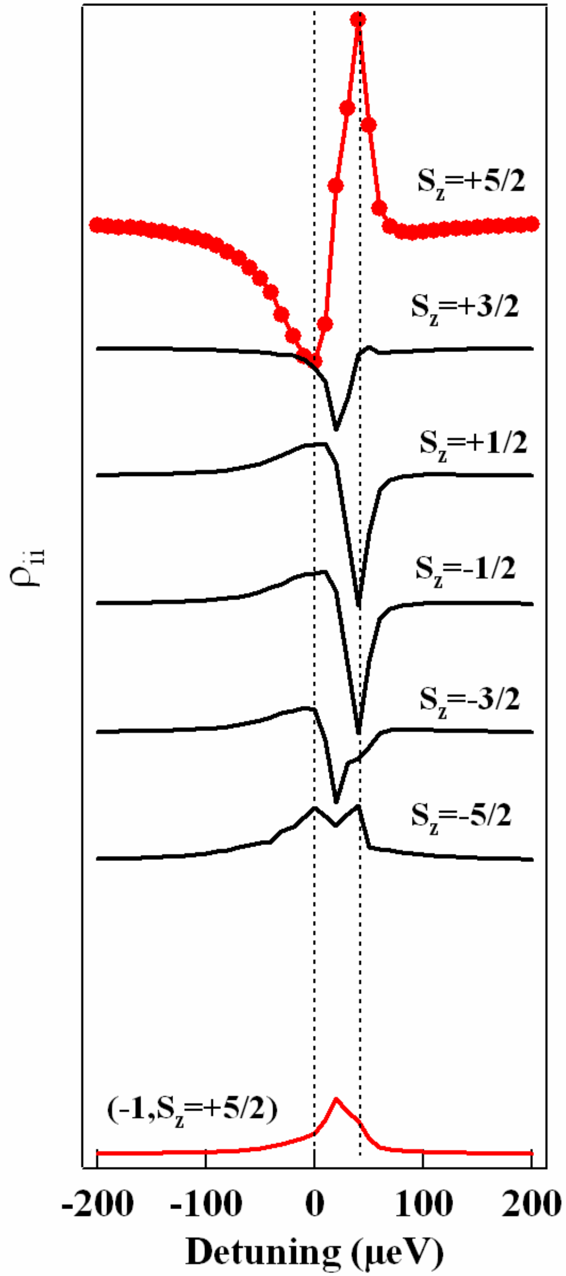
$$L_{deph,jj}\varrho = \frac{\gamma_{jj}}{2}(2|j\rangle\langle j|\varrho|j\rangle\langle j| - \varrho|j\rangle\langle j| - |j\rangle\langle j|\varrho)$$

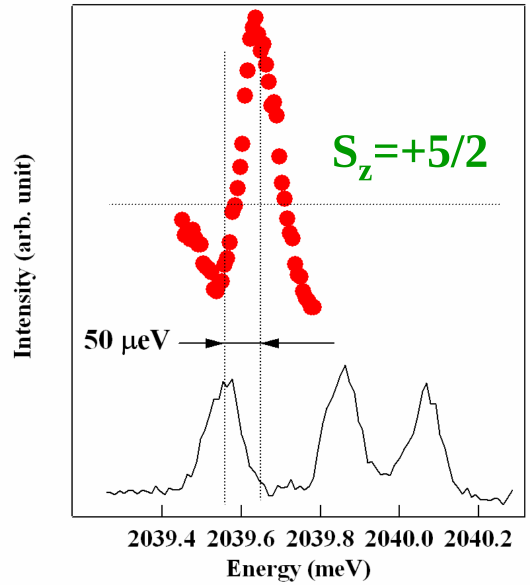
Coherent coupling:

$$L_{coh,i\leftrightarrow j}\varrho = i\frac{\Omega_{ij}}{2}(|j\rangle\langle i|\varrho + |i\rangle\langle j|\varrho - \varrho|j\rangle\langle i| - \varrho|i\rangle\langle j|)$$

# *Spin population trapping on a Mn atom*

- Resonant excitation around $S_z=+5/2$:




+5/2 +3/2 +1/2

$D_0=7\mu eV$
$E=0.7\ \mu eV$

$a=0.32\mu eV$
$A=0.7\mu eV$

$T_{pump}=60ns$
$T_{Mn}=250ns$
$T_r=0.25ns$
$\gamma_d=0.1ns$

$\Omega_r=25\mu eV$

The Mn spin is “trapped” in $S_z=+5/2$ when the dressed states are on resonance with $S_z=\pm1/2$

# *Effect of in-plane strain anisotropy*

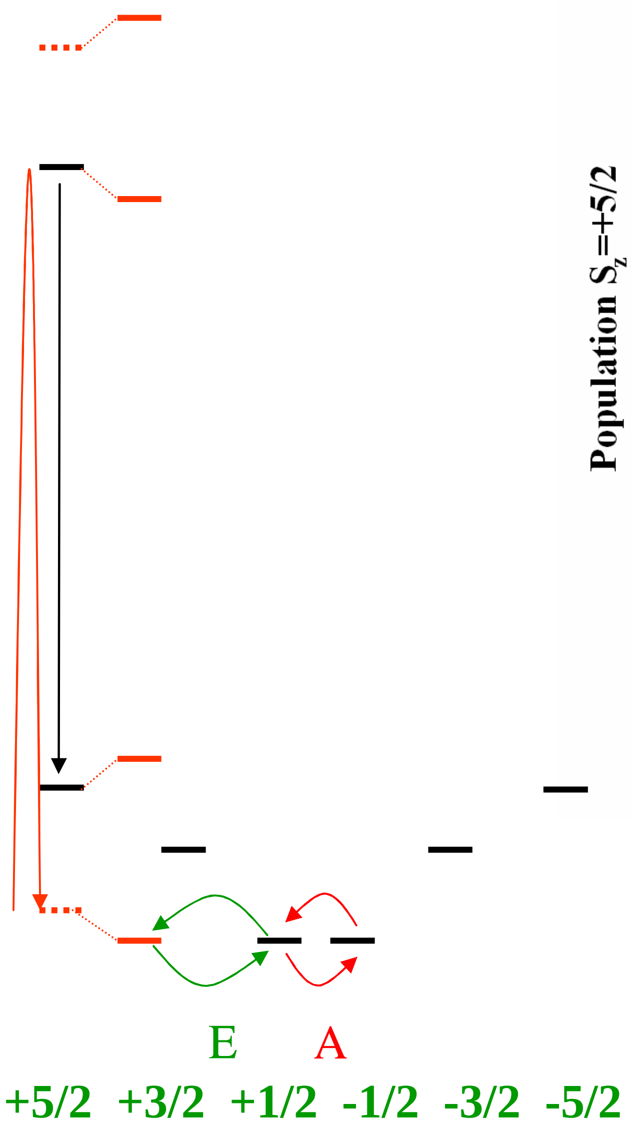
- Resonant excitation around $S_z=+5/2$:

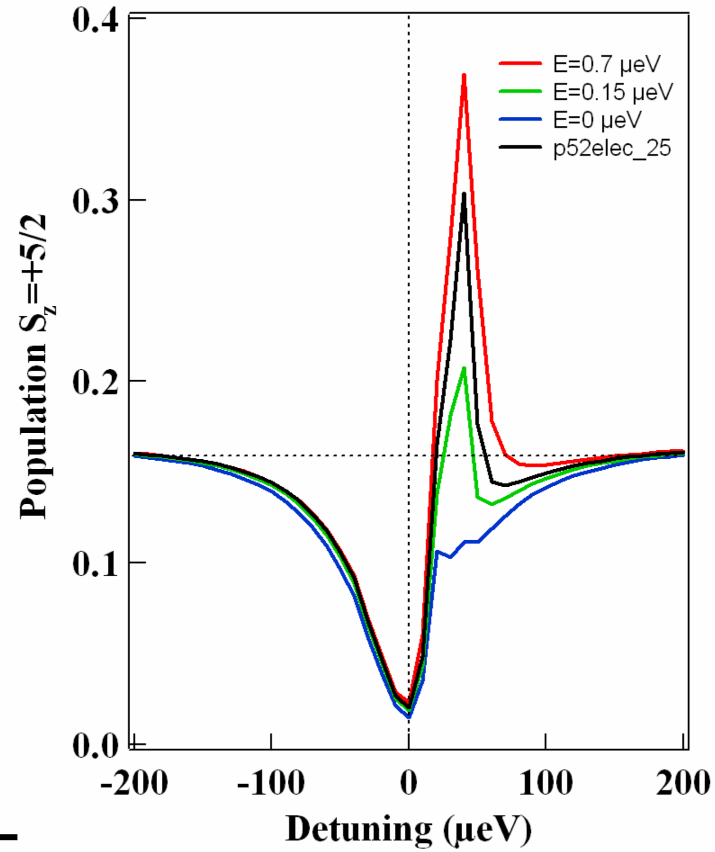
$D_0=7\mu eV$

$a=0.32\mu eV$
$A=0.7\mu eV$

$T_{pump}=60ns$
$T_{Mn}=250ns$
$T_r=0.25ns$
$\gamma_d=0.1ns$

$\Omega_r=25\mu eV$



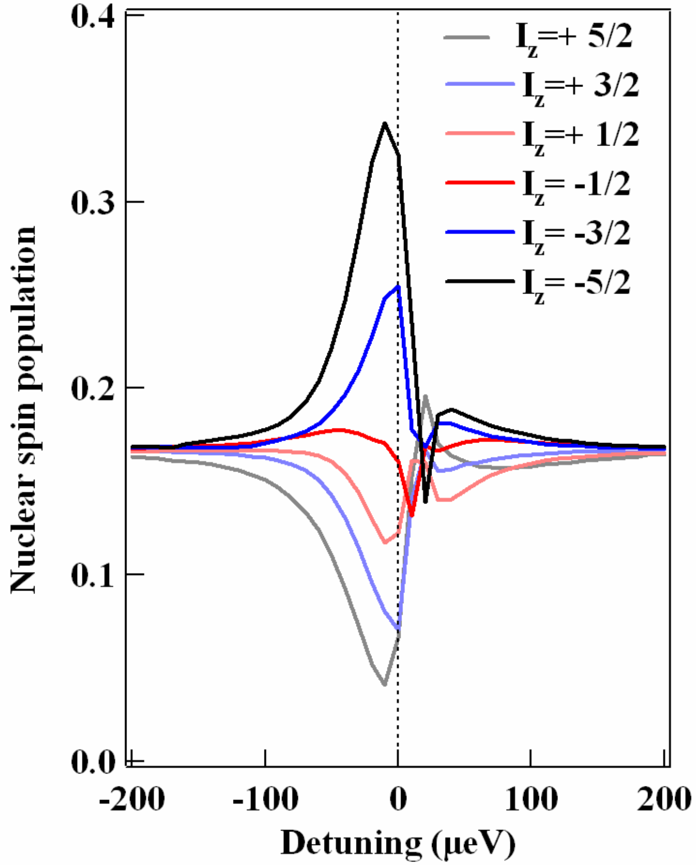




- Increase of the population trapping effect with the increase of the coherent coupling term E.

# *Mn nuclear spin optical pumping*

- Resonant excitation around $S_z$=+3/2 with a fixed Rabi energy $\Omega_r$=25µeV

- Hyperfine coupling can lead to pumping of the Mn nuclear spin.

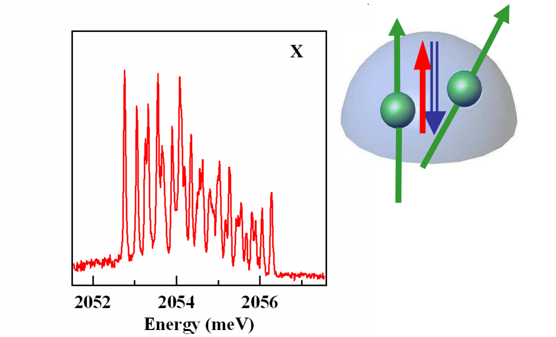



- Optical preparation of an individual nuclear spin

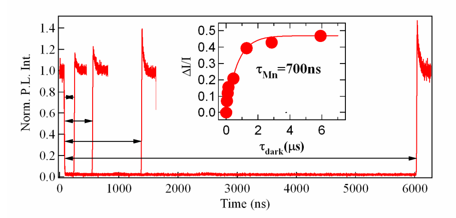
$S_z$=+3/2 resonantly excited ⇨ Dressed states of $S_z$=+3/2 on resonance with $S_z$=+1/2 ⇨ Transfer from $S_z$=+1/2 to $S_z$=+3/2 though an electron/nuclei flip-flop ⇨ Transfer from $S_z$=-1/2 to $S_z$=+1/2 through an electron/nuclei flip-flop ⇨ Pumping of the nuclear spin

# *Conclusion & Perspectives*

- Probe the spin state of one or two Mn atoms using the optical properties of a II-VI QD:
Optically control the interaction between two Mn spins




- Mn dynamics controlled by its fine structure: sensitive to the local strain environment. Spin relaxation in the µs range.




- Optically dressed Mn atom: Modified spin dynamics leads to a “spin population trapping”.
Could be used to optically access the nuclear spin of the Mn
… ongoing time resolved population trapping experiments.