Experimental aspects of spin carrier interaction in II-VI DMSs: from carrier induced ferromagnetism to single spin quantum dots

Joël Cibert
Laboratoire Louis Néel, Grenoble

H.Boukari, Y. Léger, L. Maingault,
L. Besombes, D. Ferrand, H. Mariette, S. Tatarenko...
Nanophysics and Semiconductors Group,
Laboratoire de Spectrométrie Physique, Grenoble

P.Kossacki, W.Maslana, J.Gaj
Institute of Physics, Warsaw University

joel.cibert@grenoble.cnrs.fr

1. II-VI diluted magnetic semiconductors
   - why II-VI's
   - spectroscopy

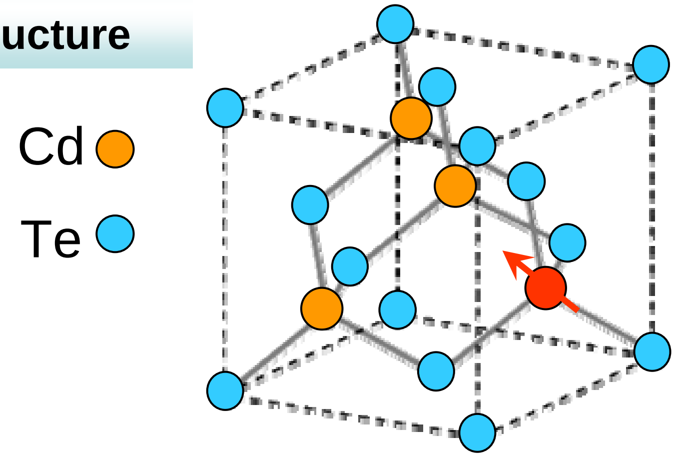
2. 2D carrier induced ferromagnetism:
   - experimental evidences of disorder?

3. Quantum dots with Mn
   - one single spin in a quantum dot

joel.cibert@grenoble.cnrs.fr

Electronic and spectroscopic properties of semiconductors

Zinc-blende structure



diluted magnetic semiconductors

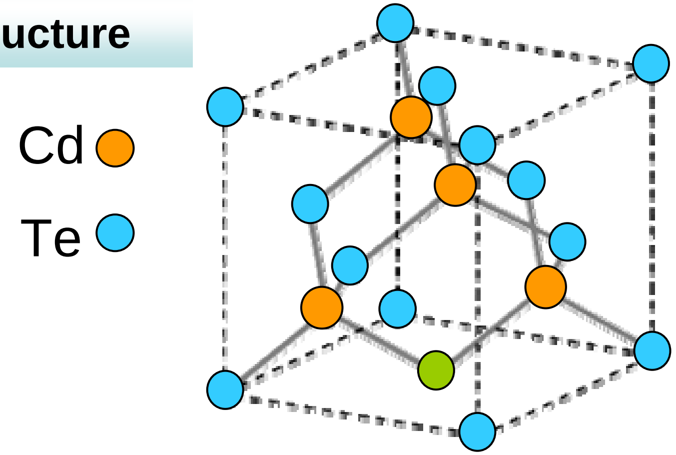
Zn: $4s^2\ 3d^{10}$
Cd: $5s^2\ 4d^{10}$
Mg: $3s^2$
Mn: $4s^2\ 3d^5$ $\Rightarrow$ spin 5/2

8 electrons brought by each pair of atoms

I	II	II										III	IV	V	VI	VII	VIII
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo		Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

Electronic and spectroscopic properties of semiconductors

Zinc-blende structure

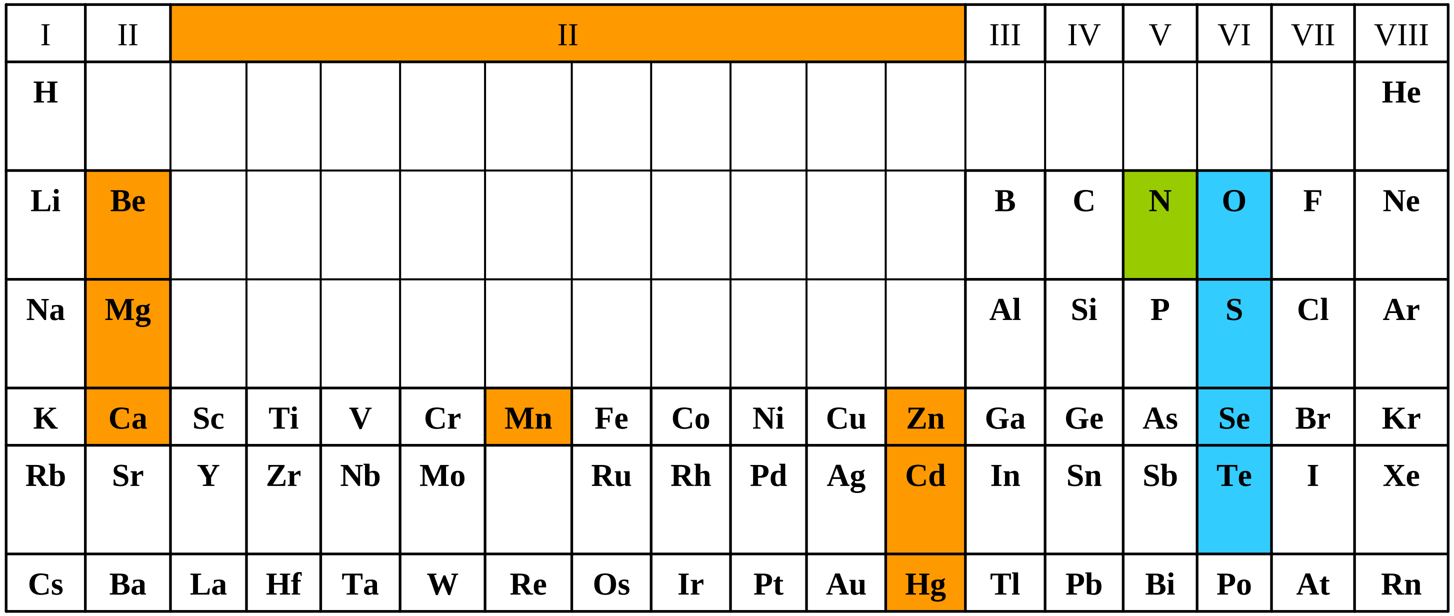


p-type doping

Te: $5s^2\ 4d^{10}\ 5p^6$

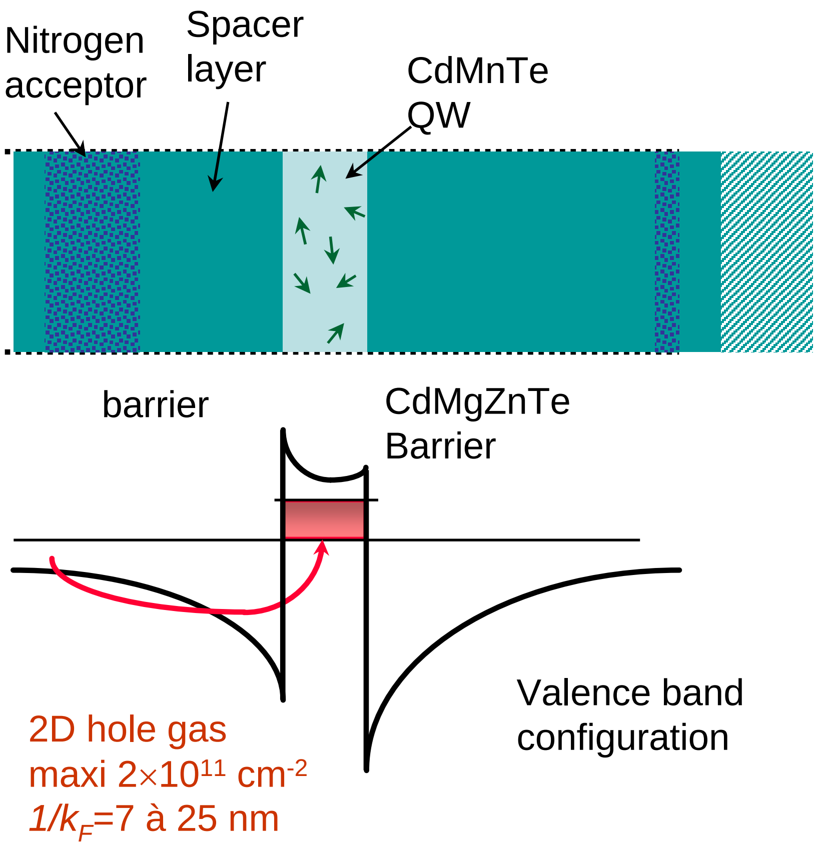
N: $2s^2\ 2p^5$ ⇒ acceptor

8 electrons brought by each pair of atoms



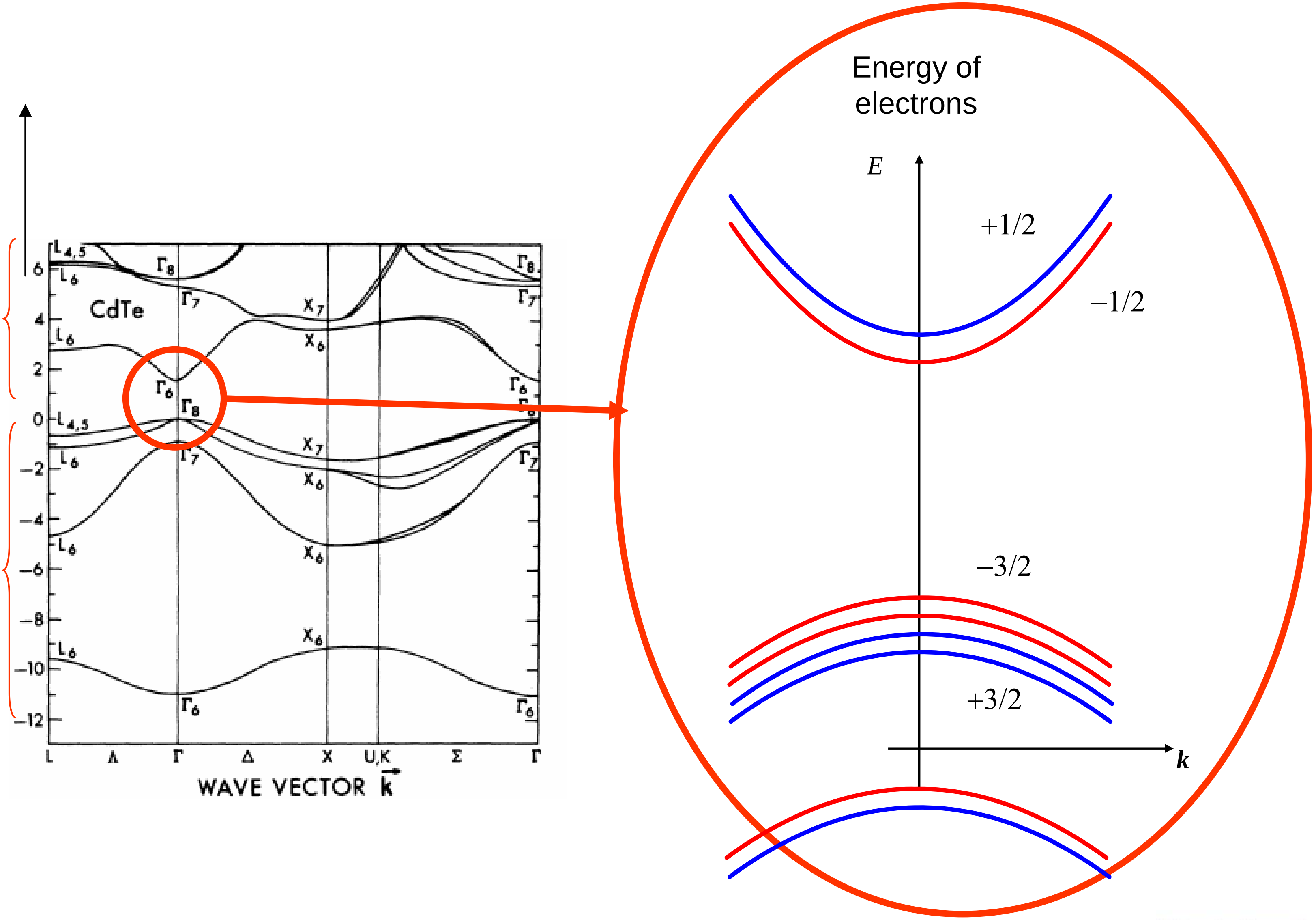
I	II	II										III	IV	V	VI	VII	VIII
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo		Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

II-VIs modulation-doped magnetic quantum well



2D hole gas
maxi 2×10^{11} cm^{-2}
$1/k_F$=7 à 25 nm

Electronic and spectroscopic properties of semiconductors



Energy of electrons

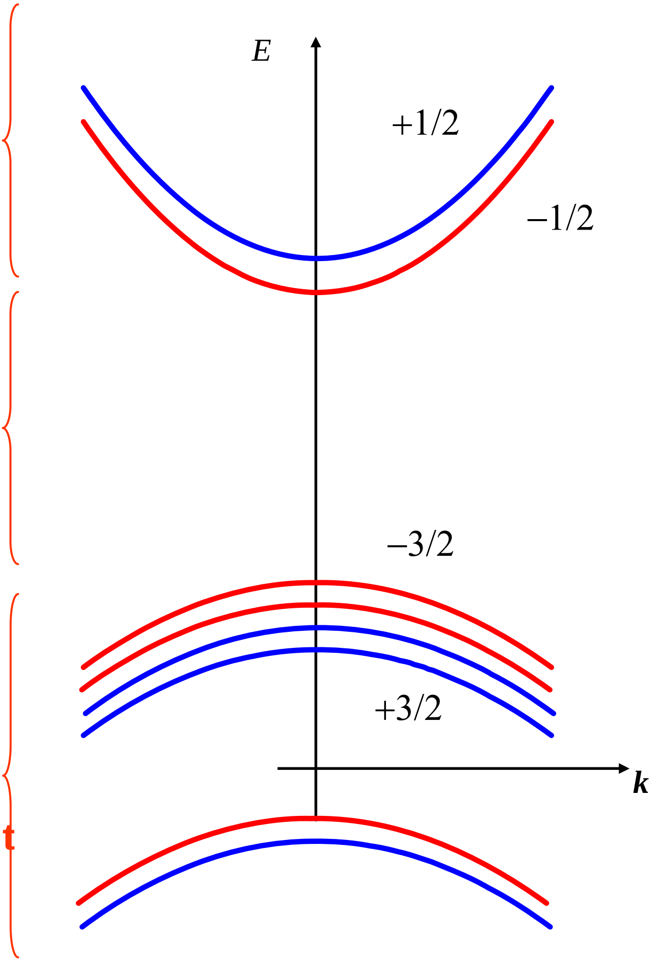
conduction band

valence band

Energy of electrons

Electronic and spectroscopic properties of semiconductors

Energy of
electrons

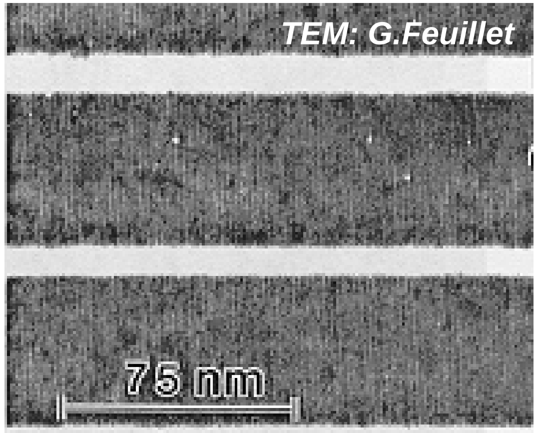


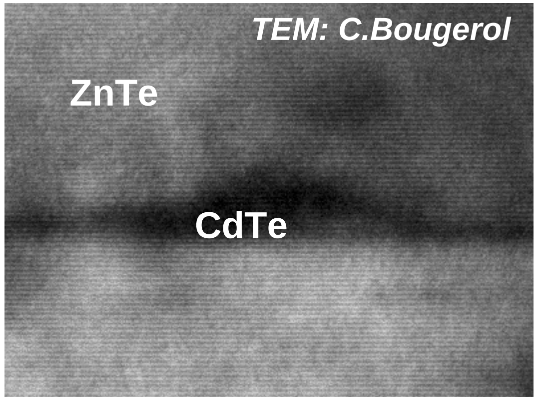
conduction band (s states)

bandgap

valence band (p states and spin-orbit coupling)

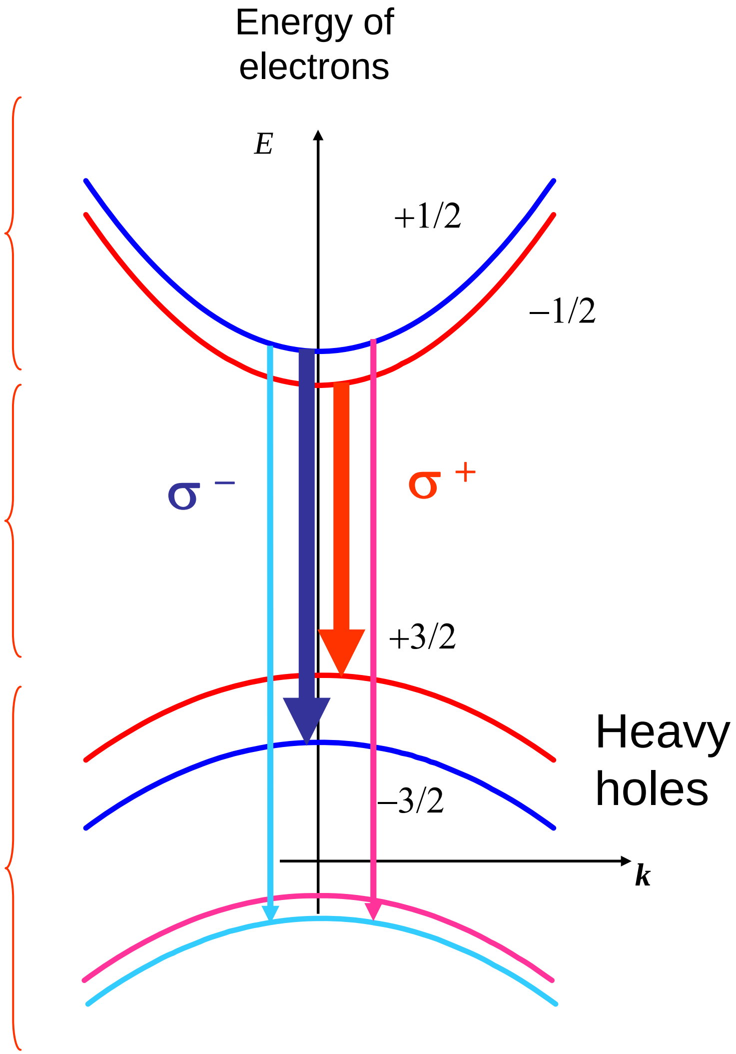
Electronic and spectroscopic properties of semiconductors





spin-orbit coupling
⇒ light holes and heavy holes

confinement in a quantum well
or a quantum dot
⇒ light-hole - heavy-hole splitting



conduction band (s states)

bandgap

valence band (p states)

Electronic and spectroscopic properties of semiconductors

z
(=quantization axis)

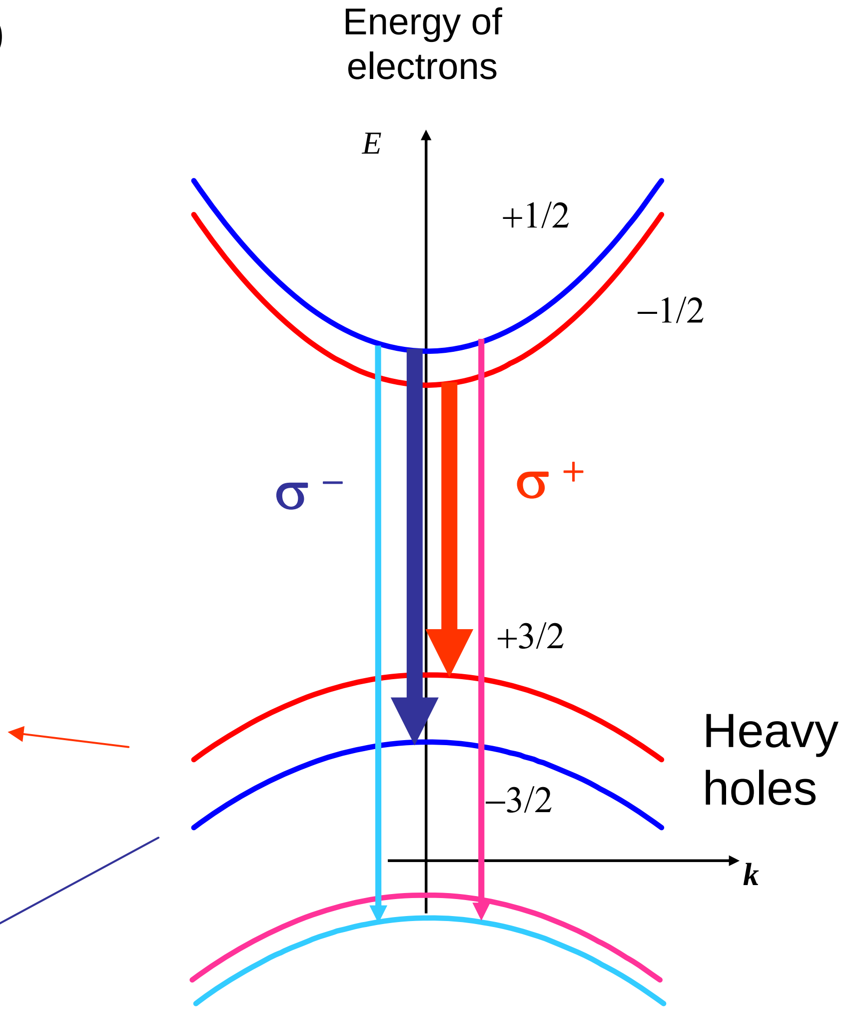
due to the orbit part (p-state)
optical selection rules
no off-diagonal spin (Ising)

orbit

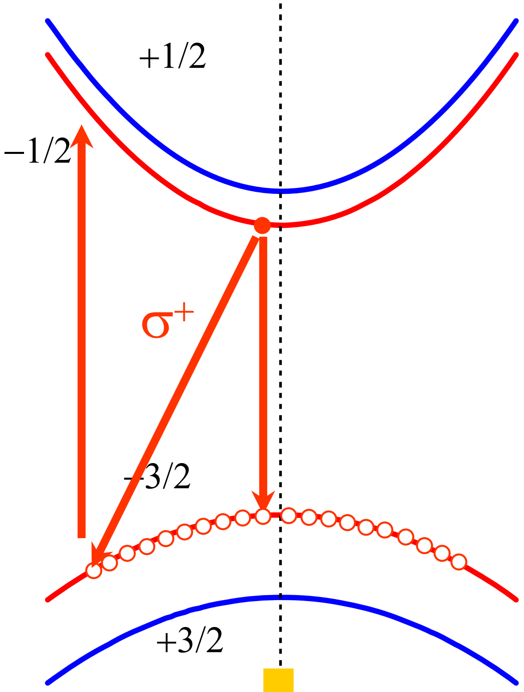
spin

$$\left|+\frac{3}{2}\right\rangle = \left|+1\right\rangle \left|+\frac{1}{2}\right\rangle$$

$$\left|-\frac{3}{2}\right\rangle = \left|-1\right\rangle \left|-\frac{1}{2}\right\rangle$$

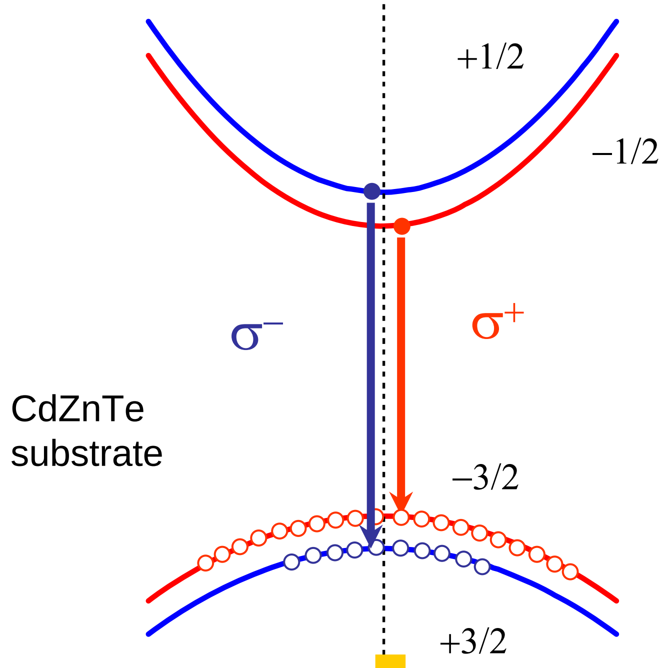


in the presence of a hole gas:



kinetic energy of carriers

carrier density



Zeeman splitting or shift

in a dilute magnetic semiconductor:

- **Mn magnetization**
- **spin density**

1. II-VI diluted magnetic semiconductors
    - why II-VI's
    - spectroscopy

2. 2D carrier induced ferromagnetism:
    - **experimental evidences of disorder?**

3. Quantum dots with Mn
    - one single spin in a quantum dot

Electronic and spectroscopic properties of semiconductors

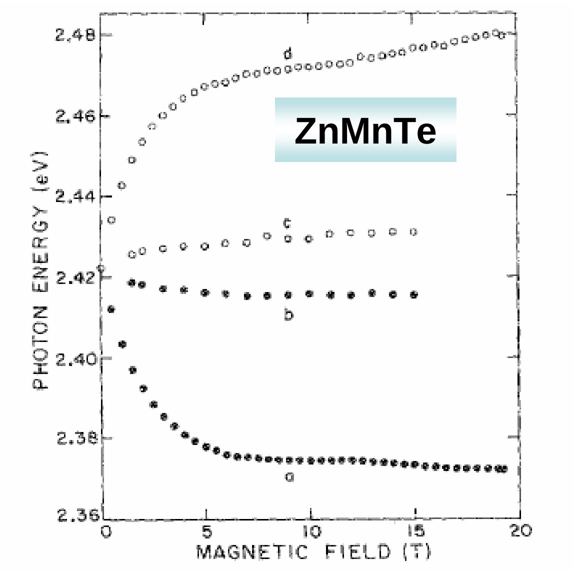
Spin-carrier interaction: local exchange

interaction between one localized spin and one carrier:
(Kondo Hamiltonian)

$$-\beta\,\vec{s}.\vec{S}_i\;\delta\left(\vec{r}-\vec{R}_i\right)$$

giant Zeeman effect

$$g_c\mu_B\mu_0\vec{s}.\left(\cancel{\vec{H}}+\lambda\vec{M}_{\mathrm{Mn}}\right)$$



$$M_{\mathrm{Mn}}=\chi_{\mathrm{Mn}}H$$

$$\chi_{Mn}=\frac{C_0x_{eff}}{T+T_{AF}}$$

Electronic and spectroscopic properties of semiconductors

Spin-carrier interaction: local exchange

interaction between one localized spin and one carrier:
(Kondo Hamiltonian)

$$-\beta\,\vec{s}.\vec{S}_i\;\delta\left(\vec{r}-\vec{R}_i\right)$$

giant Zeeman effect

$$g_c\mu_{\mathrm{B}}\mu_0\vec{s}.\left(\cancel{\vec{H}}+\lambda\vec{M}_{\mathrm{Mn}}\right)$$

$$m_c=\chi_P\,\lambda\,M_{\mathrm{Mn}}$$

χ_P = Pauli susceptibility

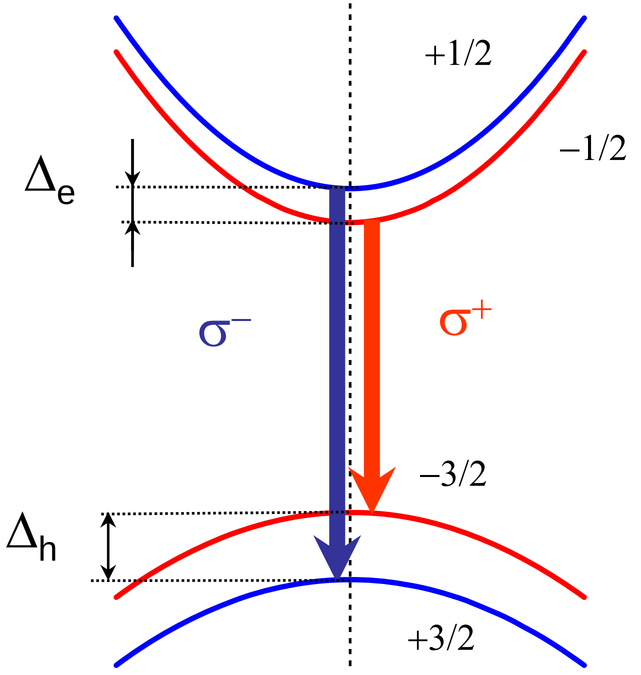
carrier induced ferromagnetism

$$M_{\mathrm{Mn}}=\chi_{\mathrm{Mn}}\left[H+\lambda\;m_c\right]$$

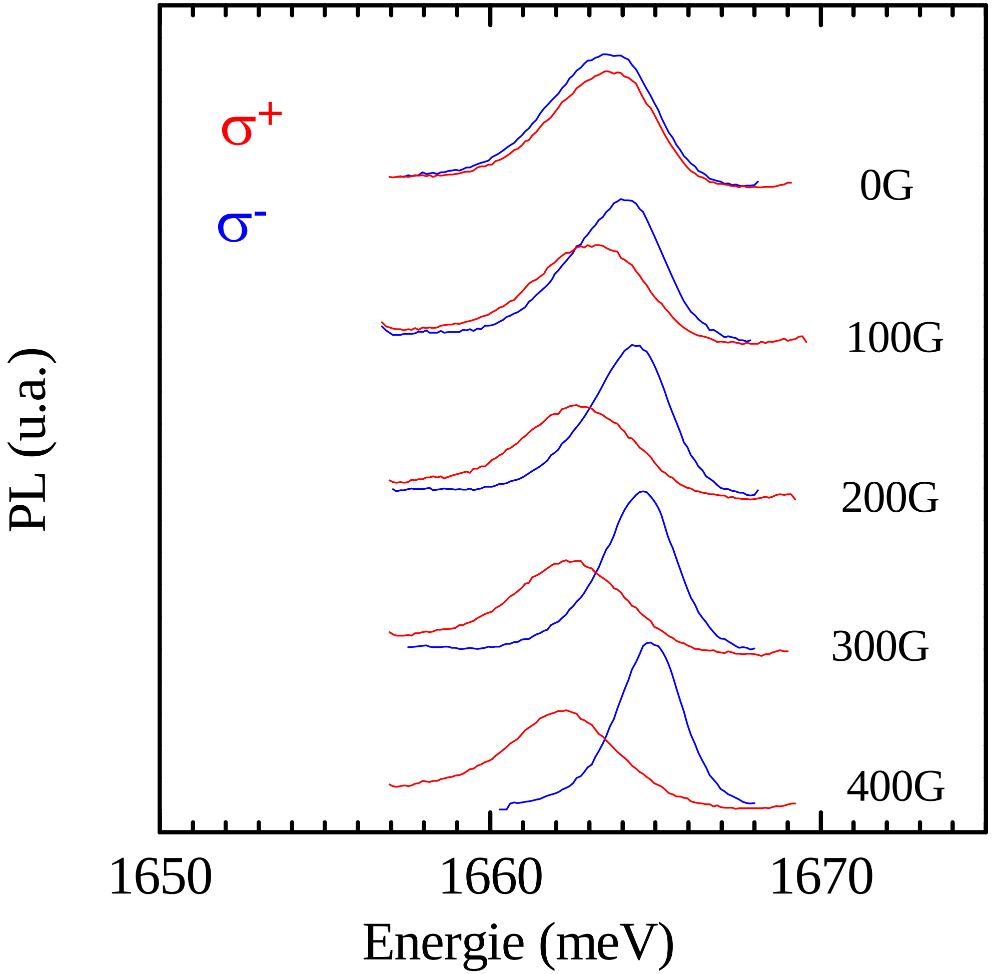
$$\chi_{Mn}=\frac{C_0x_{eff}}{T+T_{AF}}$$

$$M_{\mathrm{Mn}}=\frac{\chi_{Mn}}{1-\chi_{Mn}\chi_P\lambda^2}H=\frac{C_0x_{eff}}{T+T_{AF}-T_F}H,\quad T_F=C_0x_{eff}\;\lambda^2\;\chi_P$$

Diluted magnetic semiconductors: mean field model in tellurides



CdMnTe QW
magneto-optical spectroscopy



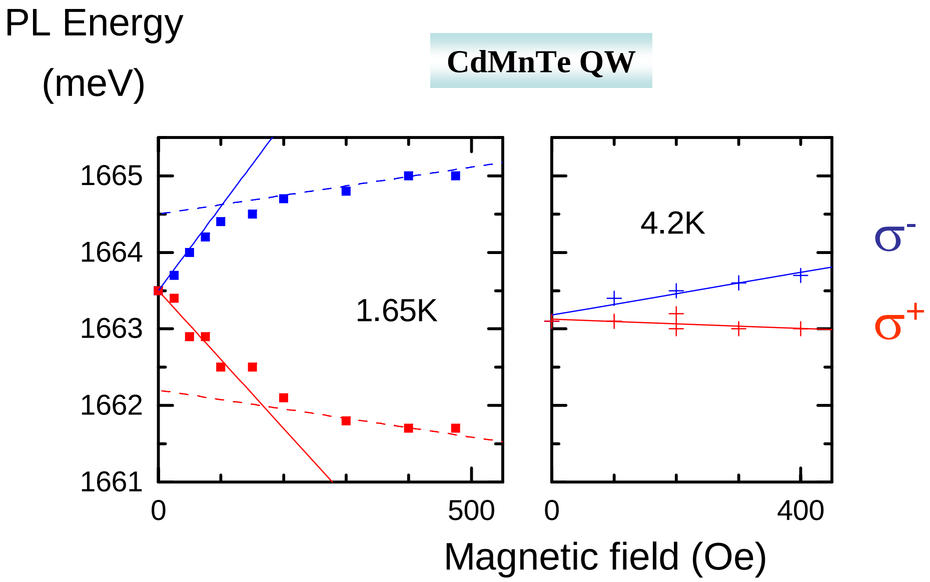
PL at 2.1K
2.4% Mn
$1.6{\times}10^{11}$ cm^{-2}

Haury 1997

Diluted magnetic semiconductors: mean field model in tellurides

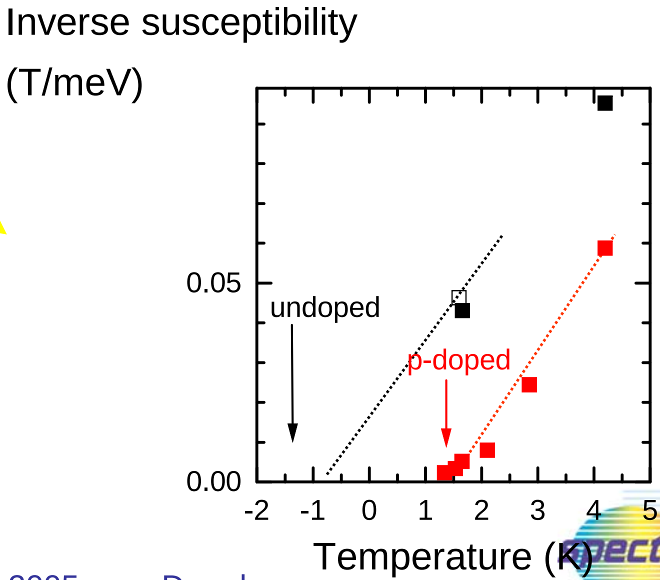
At weak applied magnetic field
in the paramagnetic phase:
enhanced giant Zeeman effect

(2.4% Mn, p=1.6 10^{11} cm^{-2})



susceptibility

$$\chi \approx \frac{d\Delta}{dB}$$



non doped

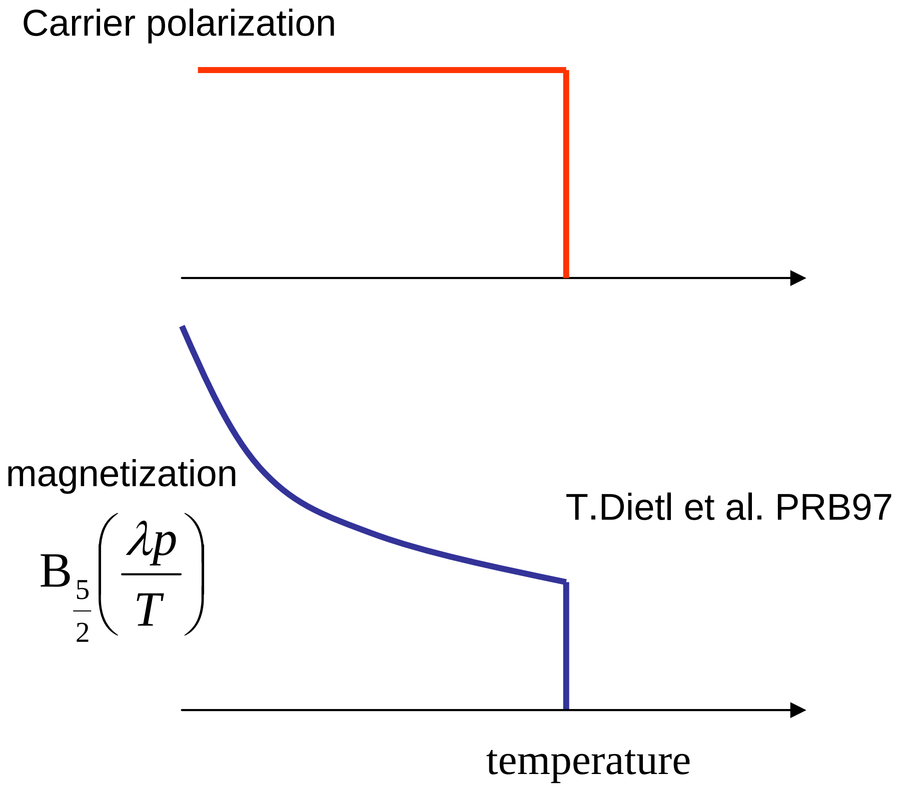
$$\boxed{\chi = \frac{C}{T + T_{AF}}}$$

doped

$$\boxed{\chi = \frac{C}{T + T_{AF} - T_F}}$$

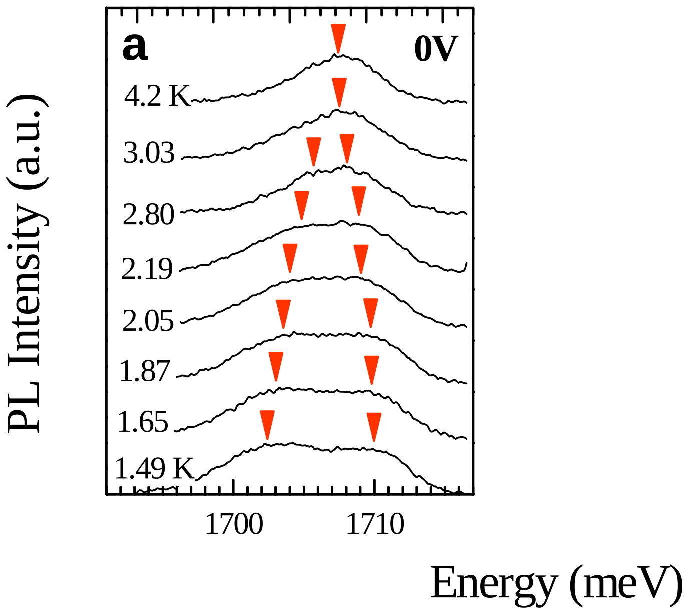
Haury 1997

Diluted magnetic semiconductors: mean field model in tellurides



$$B_{\frac{5}{2}}\left(\frac{\lambda p}{T}\right)$$

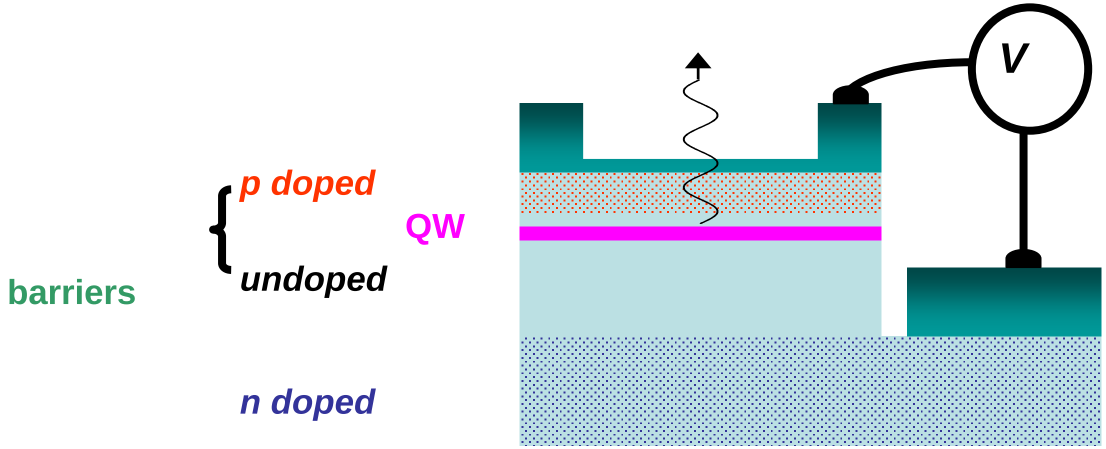
T.Dietl et al. PRB97

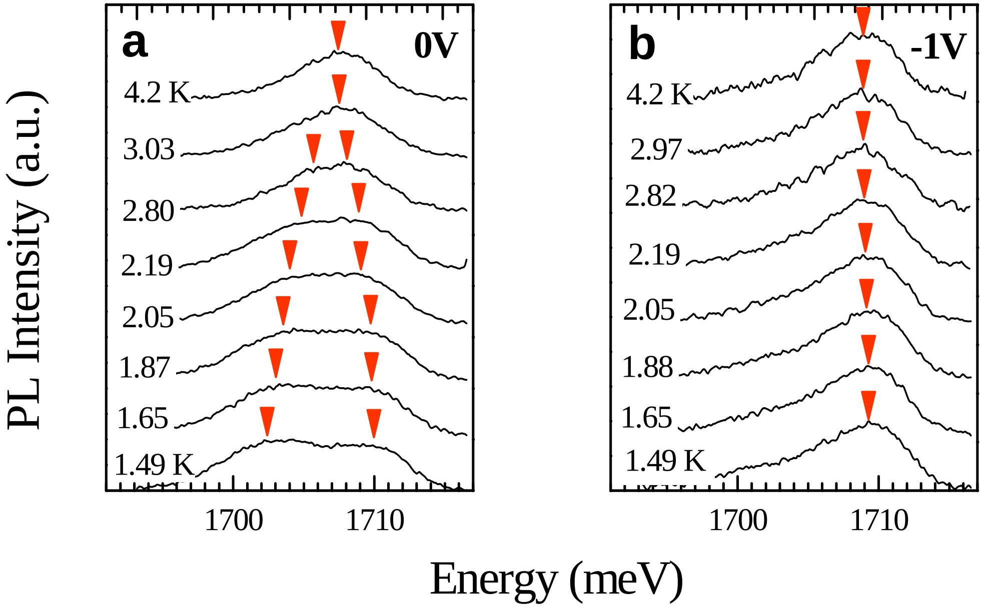


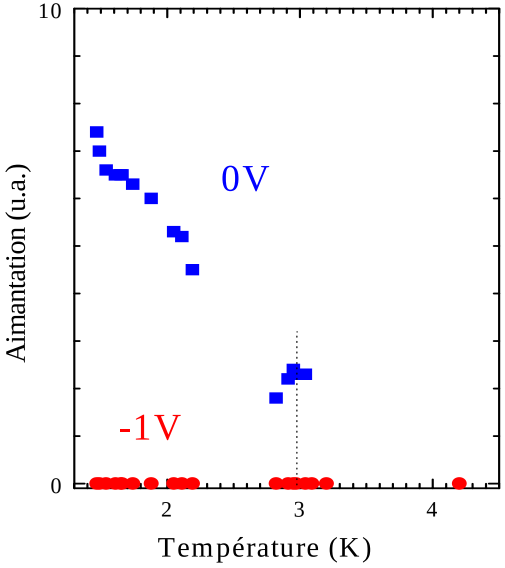
Boukari 2002

Diluted magnetic semiconductors: mean field model in tellurides

Control through an electrostatic gate in a *pin* diode: II-VI







Hole gas

depleted

Boukari PRL'02

Experimental manifestations of disorder?

The spontaneous magnetization
is **proportional** to the carrier density

Spontaneous magnetization vs. carrier density?

Complete hole polarization

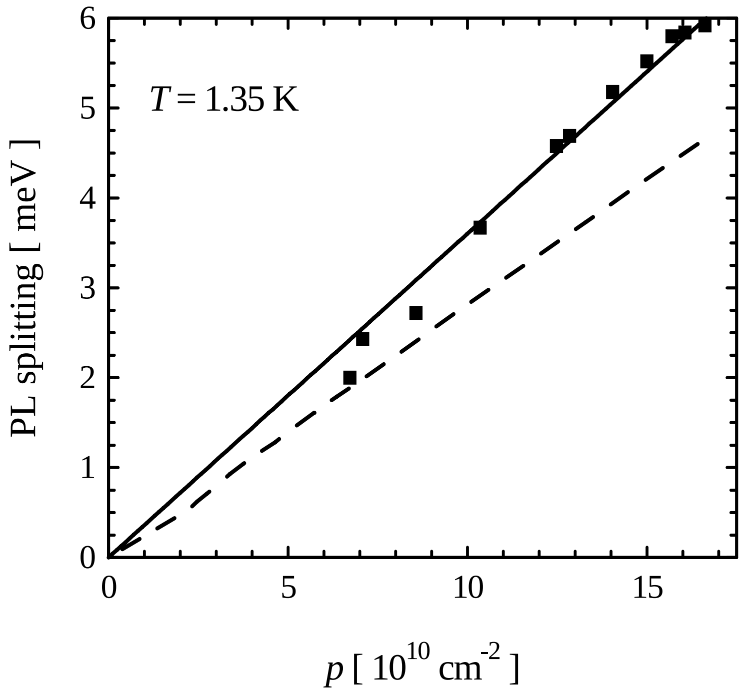
$$\langle s_z \rangle = \tfrac{1}{2}\left(p_\uparrow - p_\downarrow\right) = \tfrac{1}{2}\,p$$

effect on the Mn spins

$$\langle S_z \rangle = \chi_{Mn}\,\beta\,\langle s_z \rangle$$

Giant Zeeman splitting

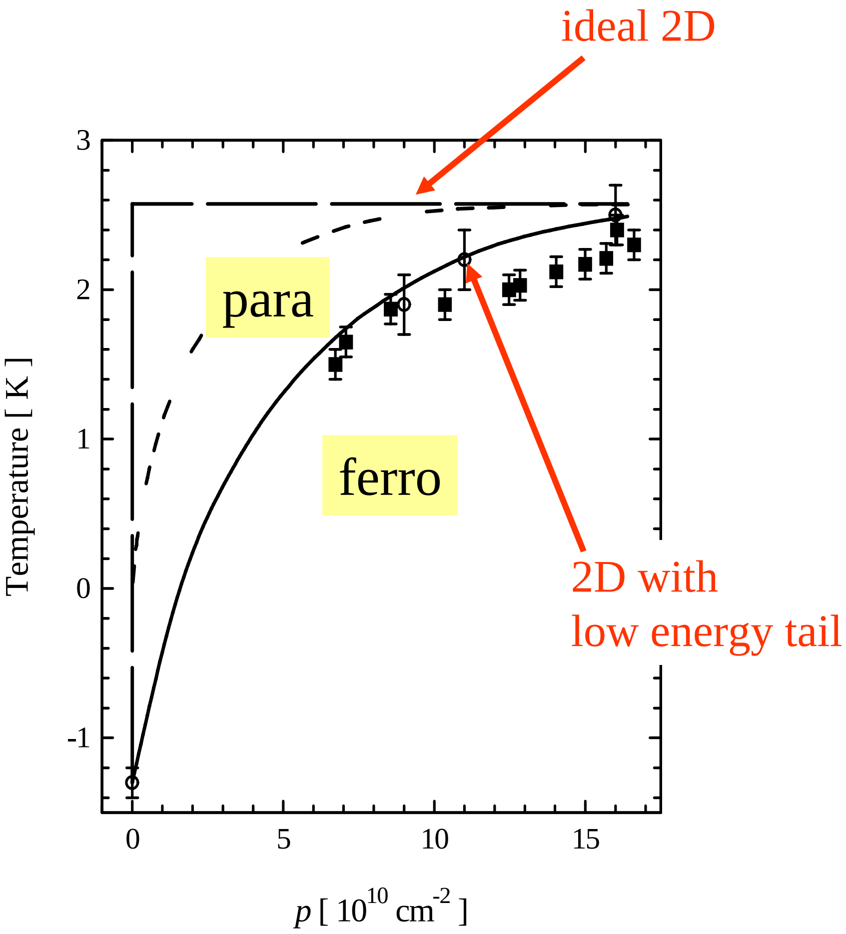
$$\Delta = \beta\,\langle S_z \rangle$$



Kossaki '01, Boukari'02

Experimental manifestations of disorder?

dependence of the critical temperature on the carrier density



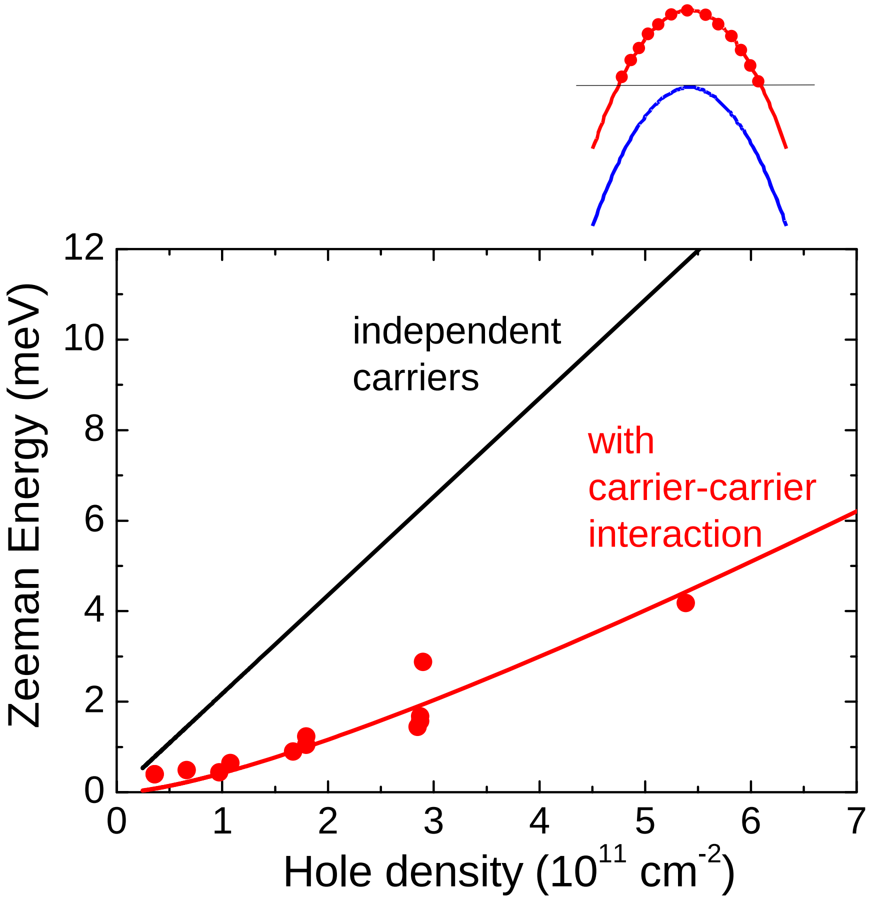
$$T_F = C_0 x_{eff}\ \lambda^2\ \chi_P - T_{AF}$$

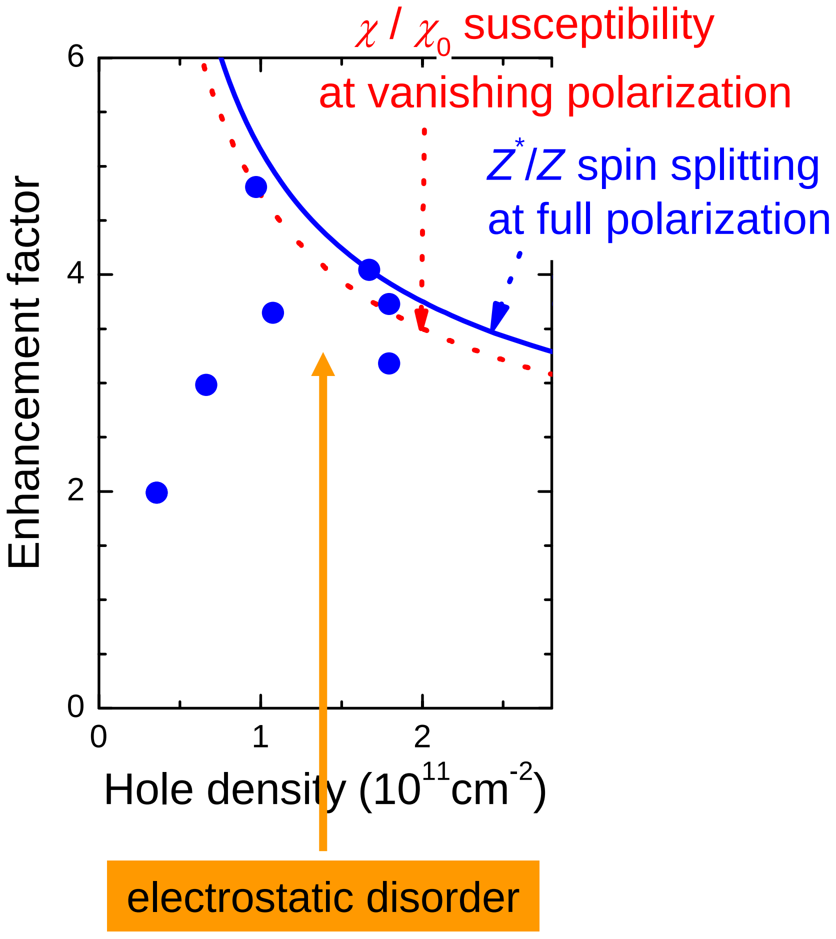
Kossacki '01

Experimental manifestations of disorder?

the hole susceptibility is enhanced by carrier-carrier interactions

onset of full polarization of the hole gas
in a very diluted sample





χ / χ_0 susceptibility
at vanishing polarization

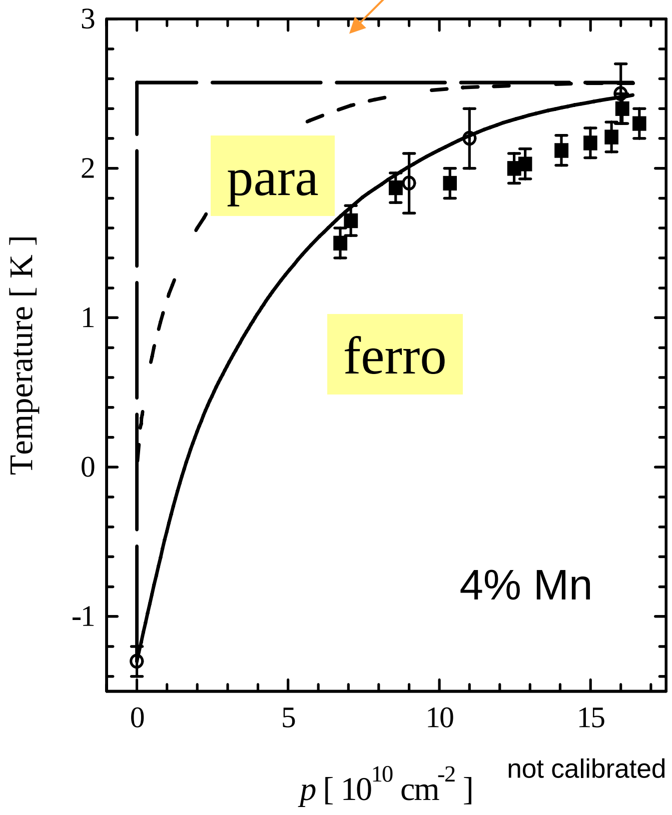
Z^*/Z spin splitting
at full polarization

electrostatic disorder

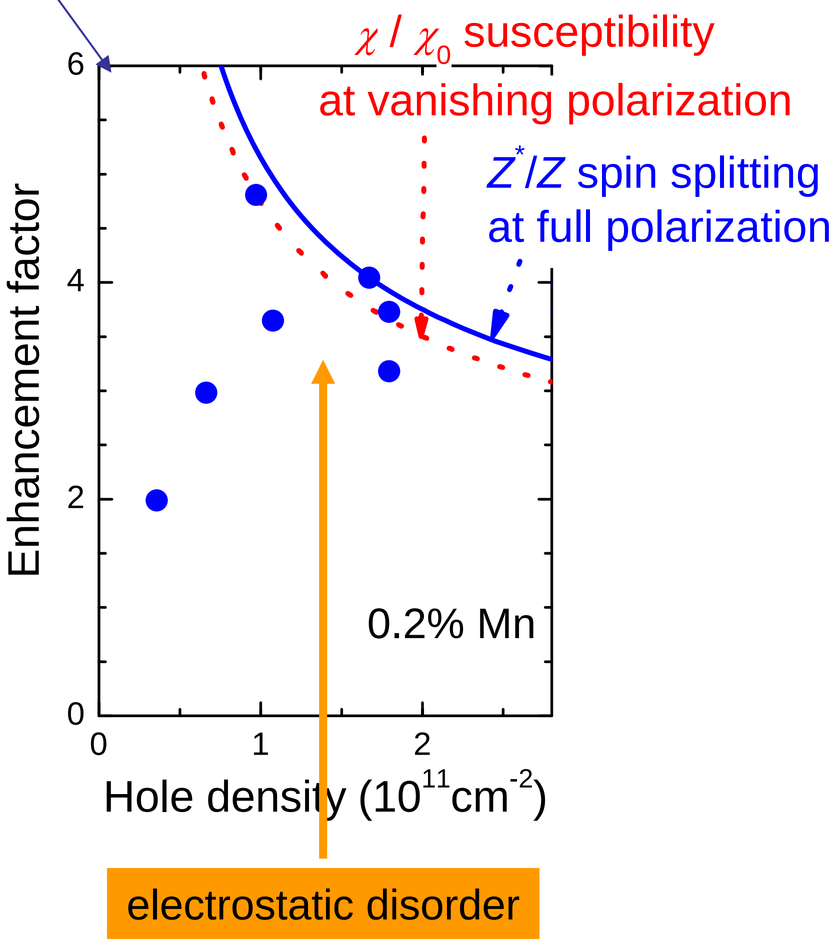
Boukari '05, calculation by F.Perez, LPN

Experimental manifestations of disorder?

$$T_F = C_0 x_{eff} \lambda^2 \chi_P - T_{AF}$$



not calibrated



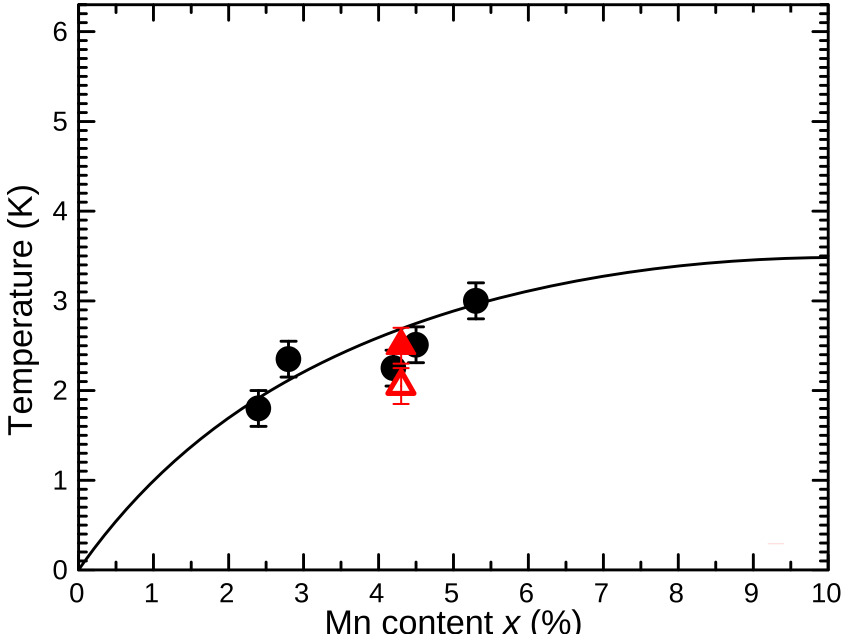
χ / χ_0 susceptibility at vanishing polarization

Z^*/Z spin splitting at full polarization

electrostatic disorder

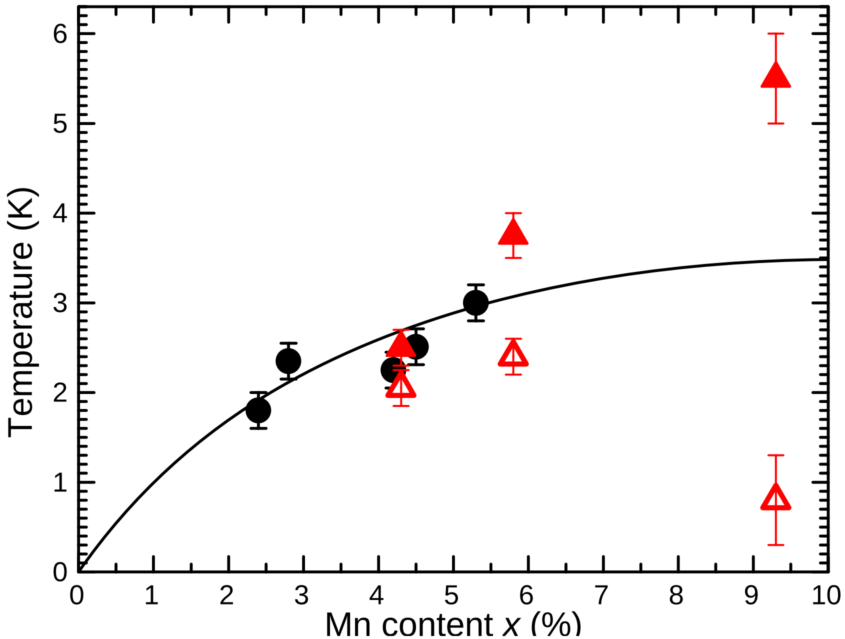
Experimental manifestations of disorder?

Curie-Weiss / critical temperature in CdMnTe quantum wells



Experimental manifestations of disorder?

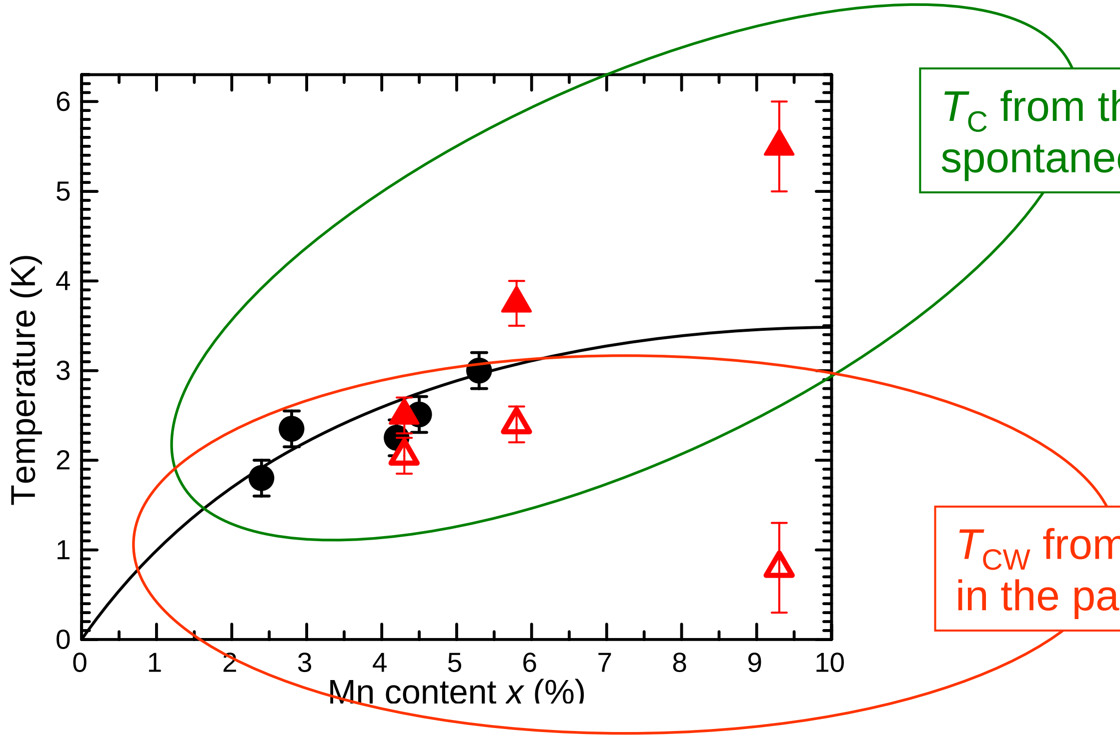
Curie-Weiss / critical temperature in CdMnTe quantum wells



Maslana '02

Experimental manifestations of disorder?

Curie-Weiss / critical temperature in CdMnTe quantum wells



T_C from the onset of spontaneous local magnetization

T_{CW} from the susceptibility in the paramagnetic phase

$T_C > T_{CW}$

qualitative experimental deviations from the mean field model:

at high carrier density, moderate spin density:

magnetization loops (superexchange) *see T.Dietl talk*

at low carrier density:

decrease of T_c (electrostatic disorder)

at large Mn content:

Curie-Weiss vs critical temperature

1. II-VI diluted magnetic semiconductors
   - why II-VI's
   - spectroscopy

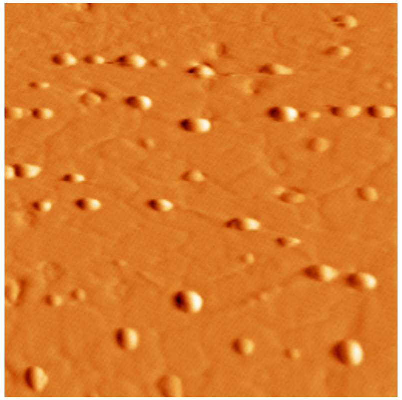
2. 2D carrier induced ferromagnetism:
   - experimental evidences of disorder?

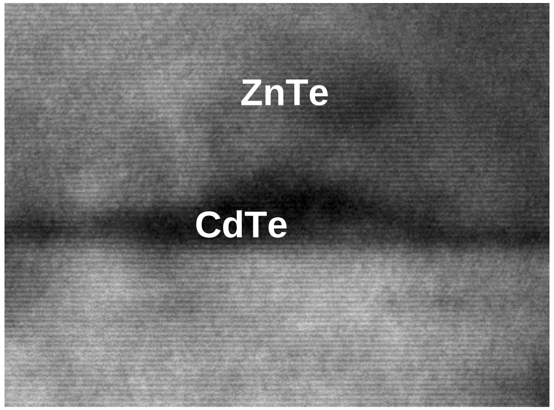
3. **Quantum dots with Mn**
   - **one single spin in a quantum dot**

Observation of a single quantum dot

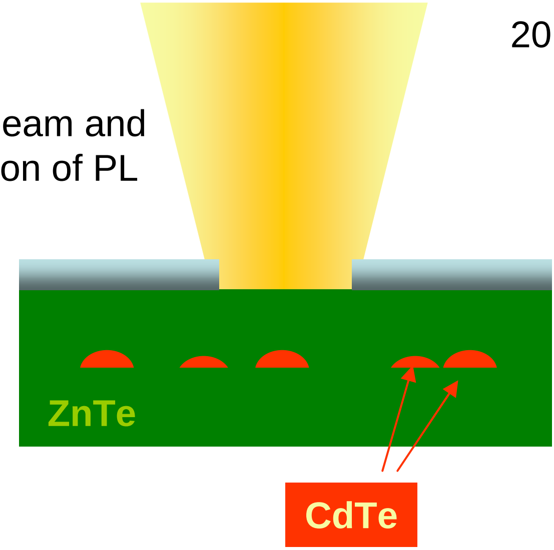
CdTe/ZnTe at 1.5 K

single quantum dot for a single photon emission





typical QD size
20nm x 20nm x 3nm

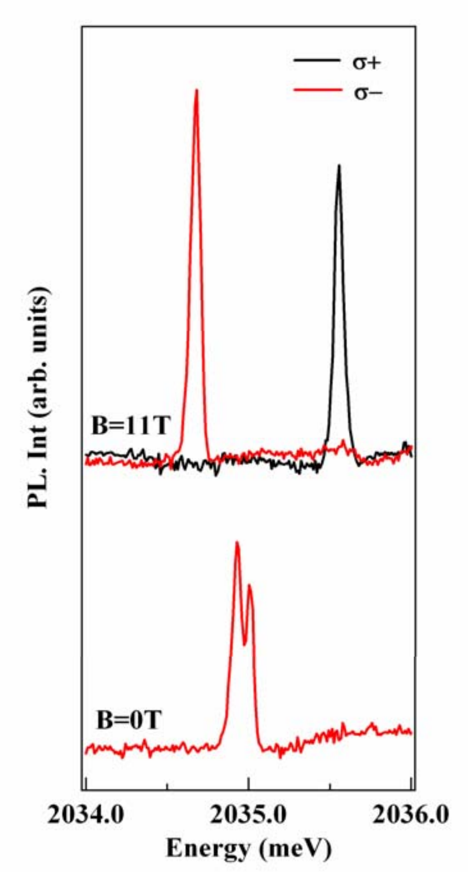


laser beam and
detection of PL

mask

ZnTe

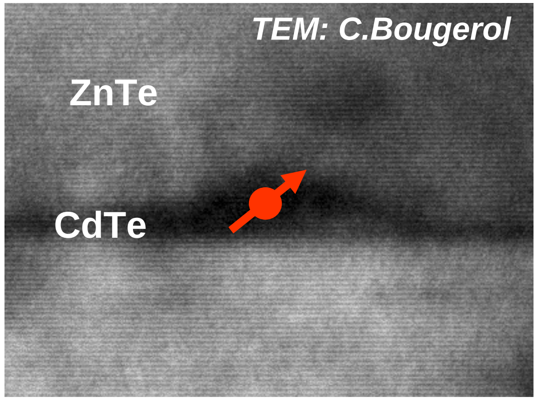
CdTe



L. Besombes *et al.*, Phys. Rev. B **65**, 121314 (2002)

DMS: a single Mn spin in a single quantum dot.

↗ = one Mn impurity
with spin 5/2

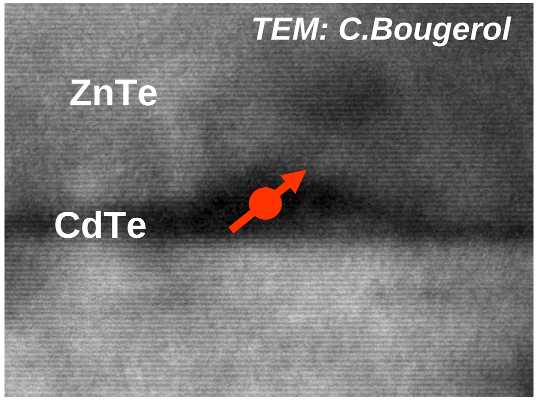


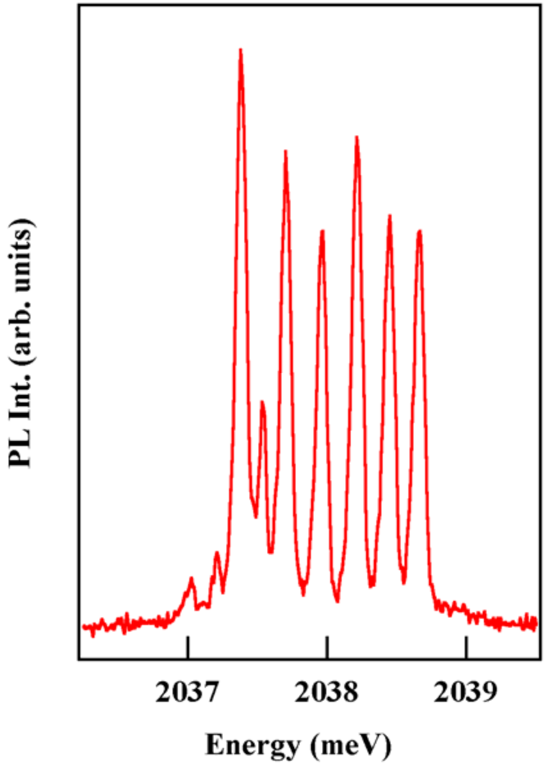
?

DMS: a single Mn spin in a single quantum dot.

L. Besombes *et al.*, Phys. Rev. Lett. 93, 207403 (2004)

↗ = one Mn impurity
with spin 5/2



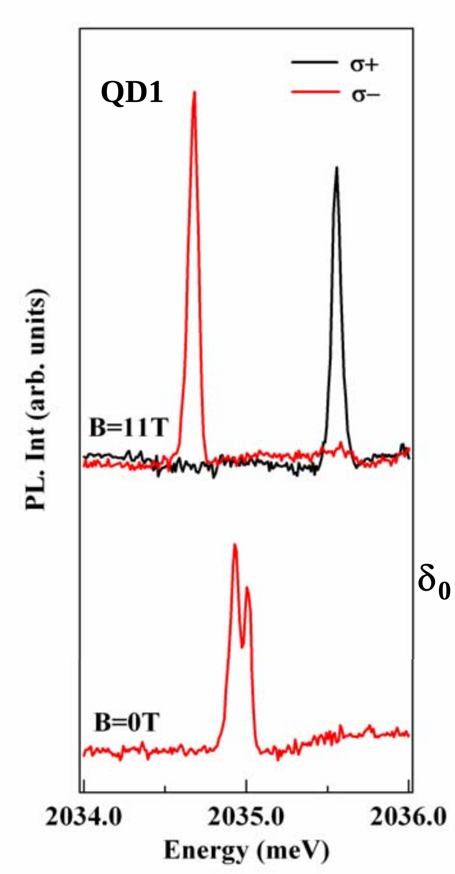


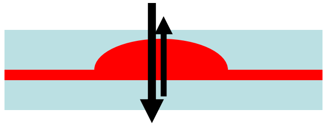
DMS: a single Mn spin in a single quantum dot.

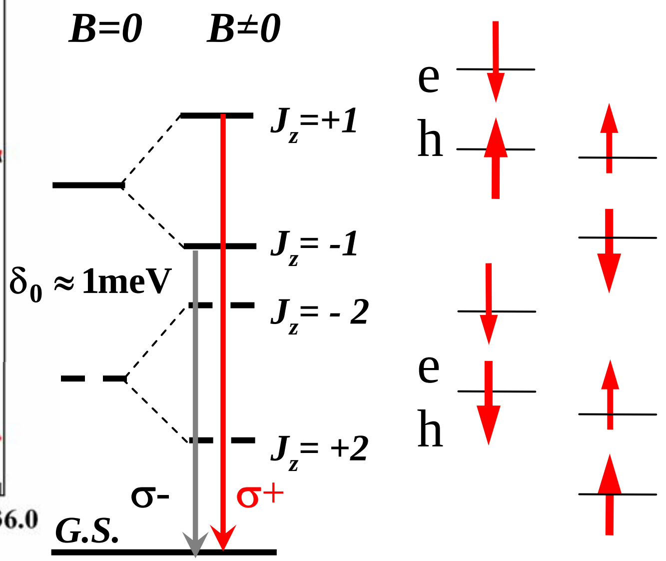
◆ **Reference CdTe/ZnTe QD sample:**

electron: spin 1/2

hole: anisotropic, J_z=±3/2







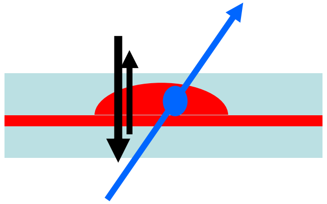
bright exciton

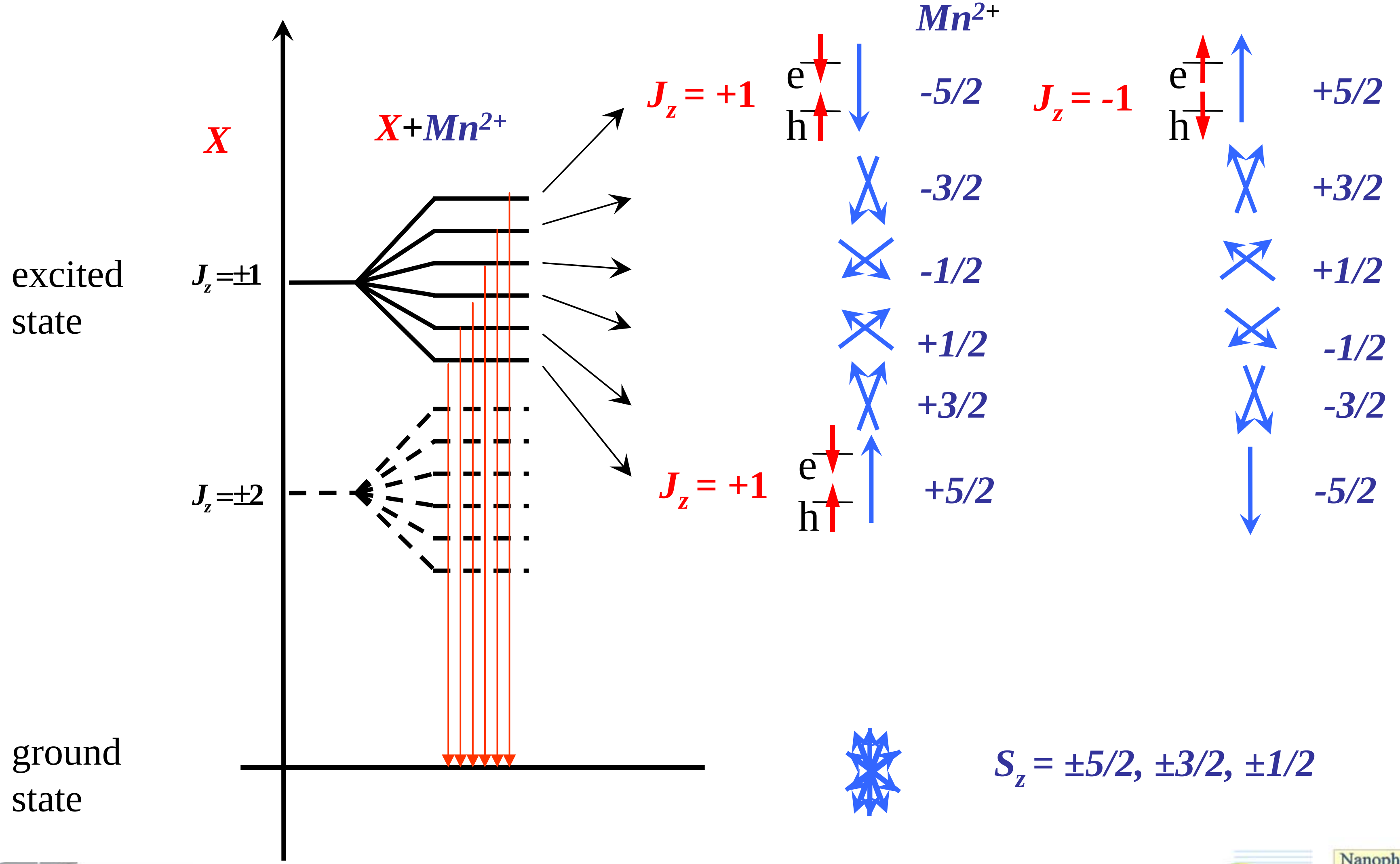
dark exciton

DMS: a single Mn spin in a single quantum dot.

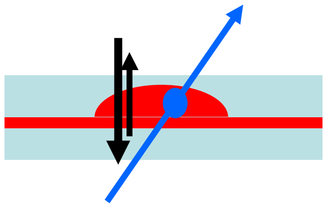
Exciton-Mn Exchange Coupling

$$-\beta\, \vec{s}.\vec{S}_i\; \delta\left(\vec{r}-\vec{R}_i\right)$$





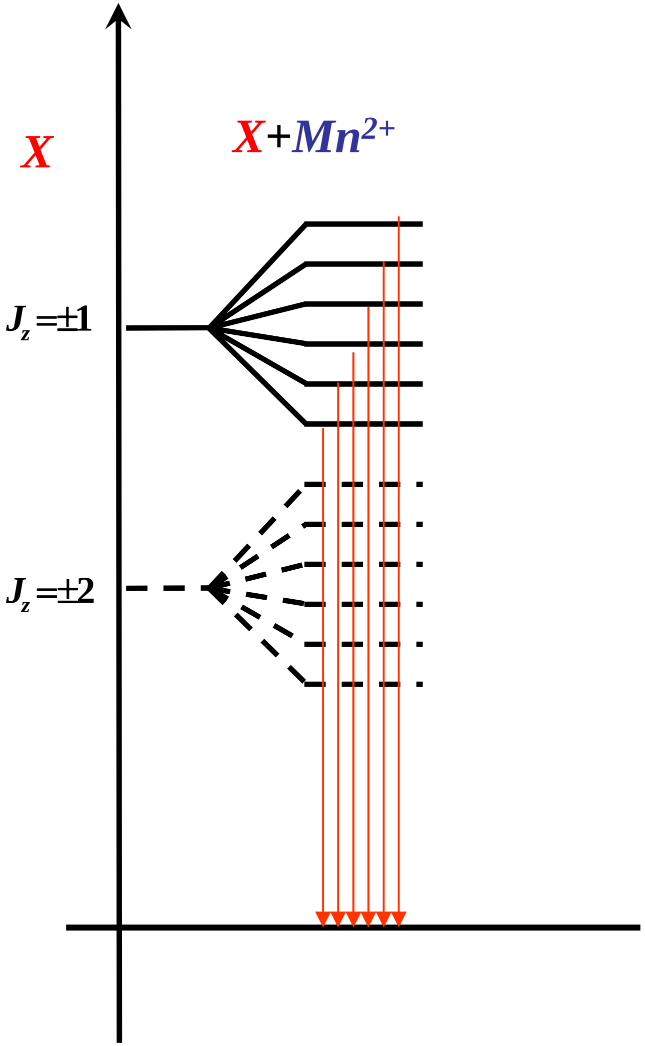
DMS: a single Mn spin in a single quantum dot.

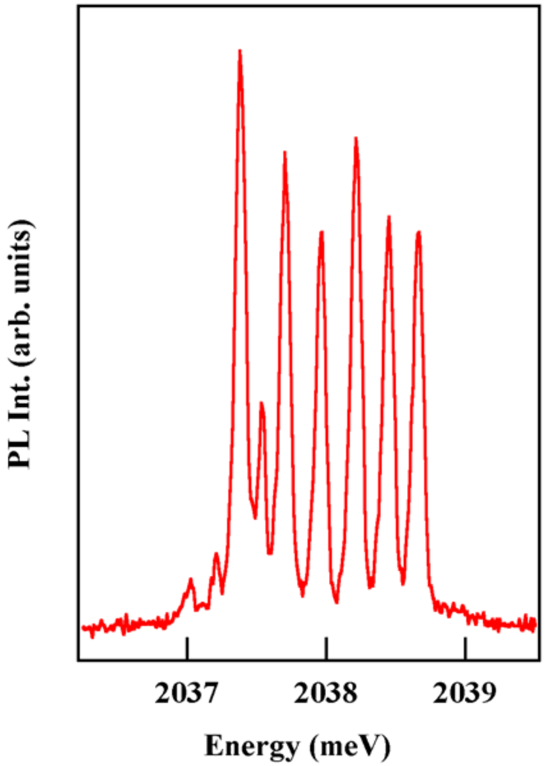


Exciton-Mn Exchange Coupling

excited
state

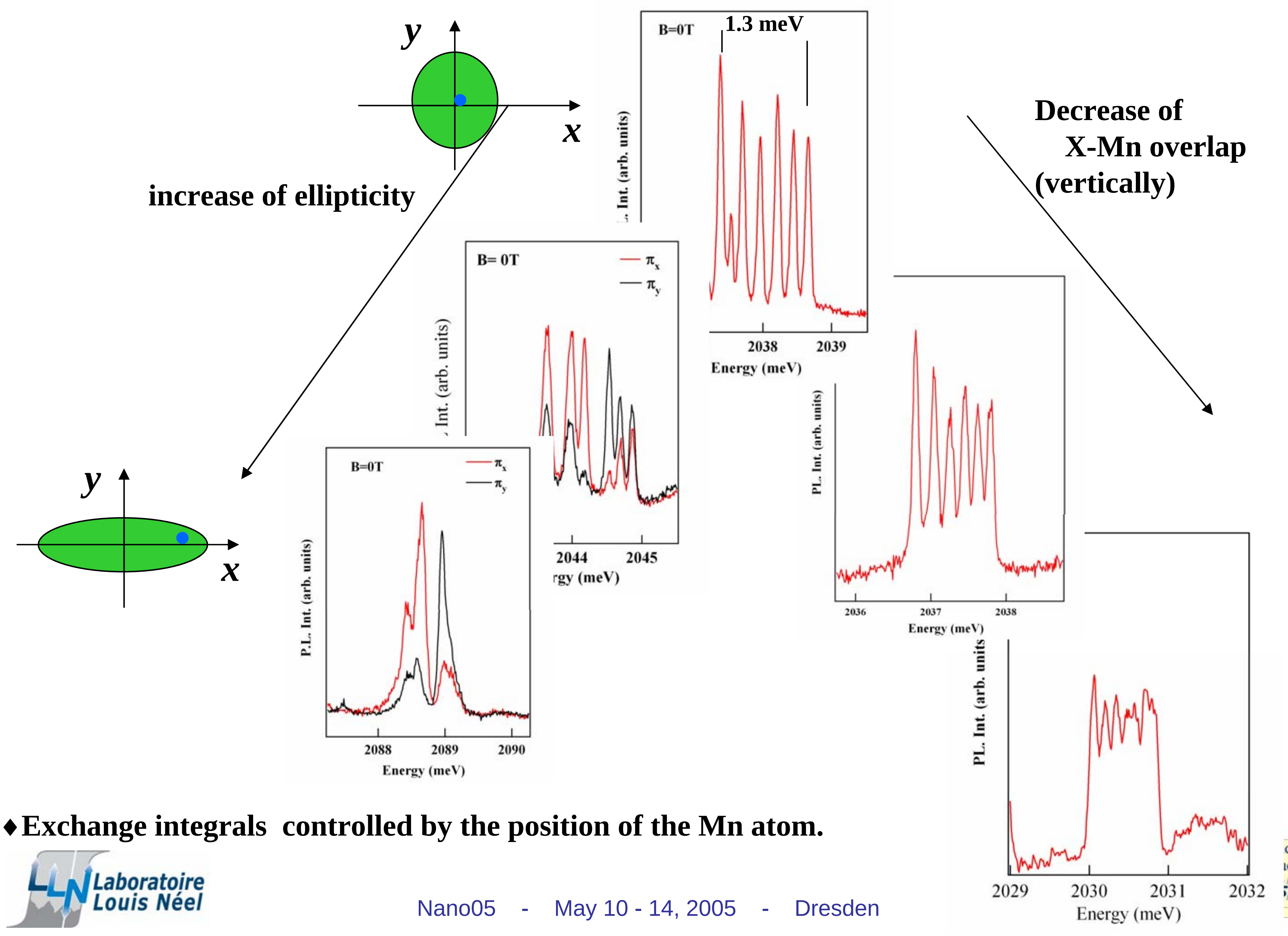
ground
state





DMS: a single Mn spin in a single quantum dot.

Exciton-Mn Overlap



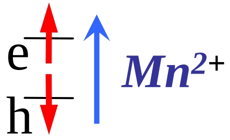
increase of ellipticity

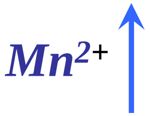
Decrease of
X-Mn overlap
(vertically)

♦ Exchange integrals controlled by the position of the Mn atom.

DMS: a single Mn spin in a single quantum dot.

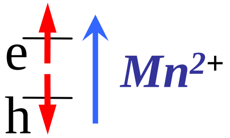
applying a magnetic field

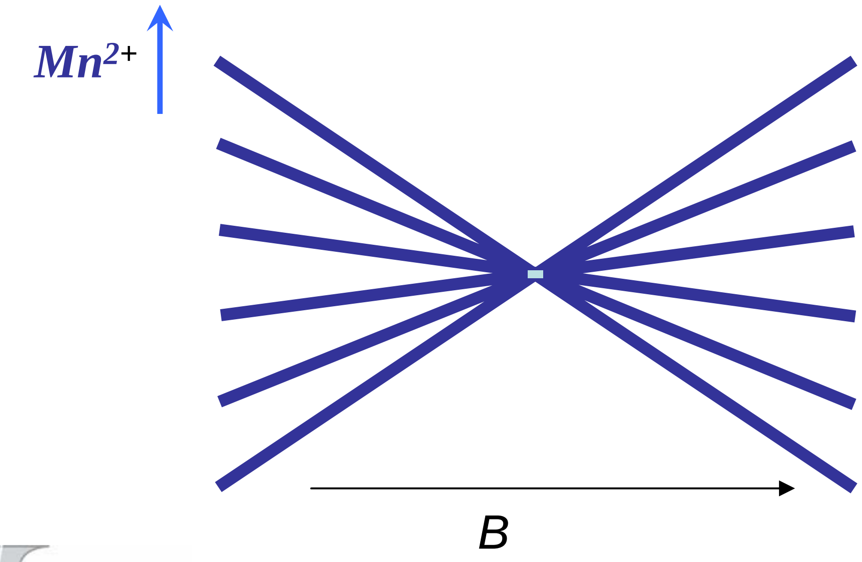




DMS: a single Mn spin in a single quantum dot.

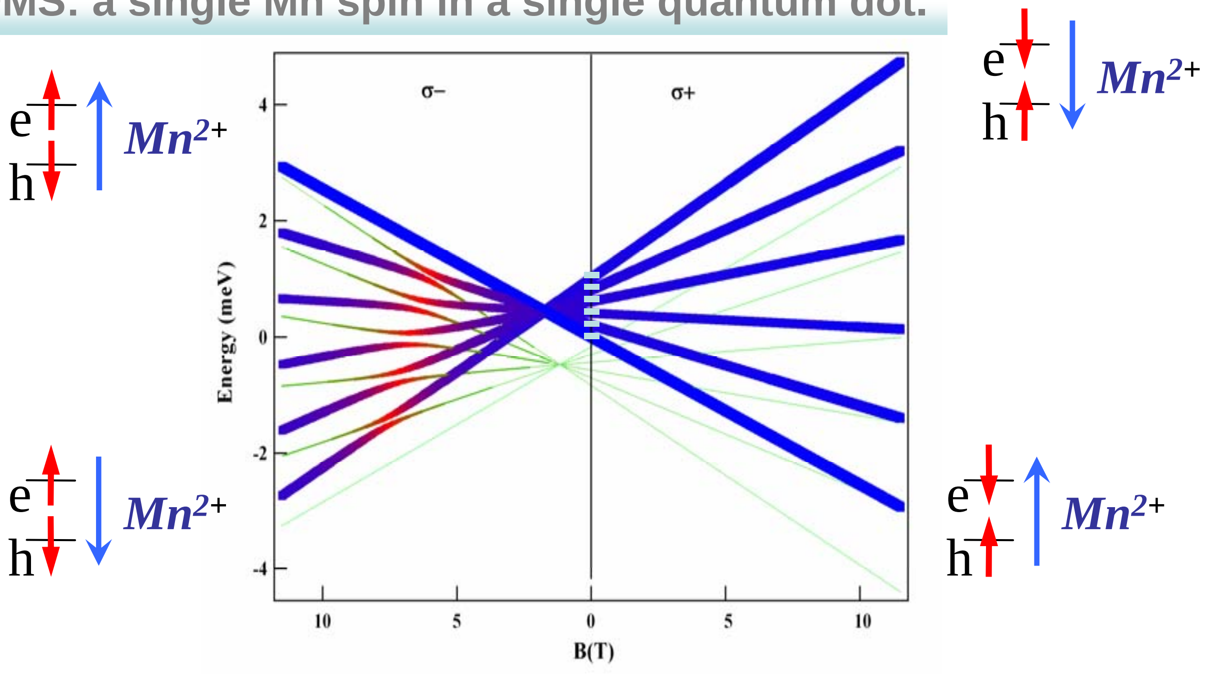
applying a magnetic field

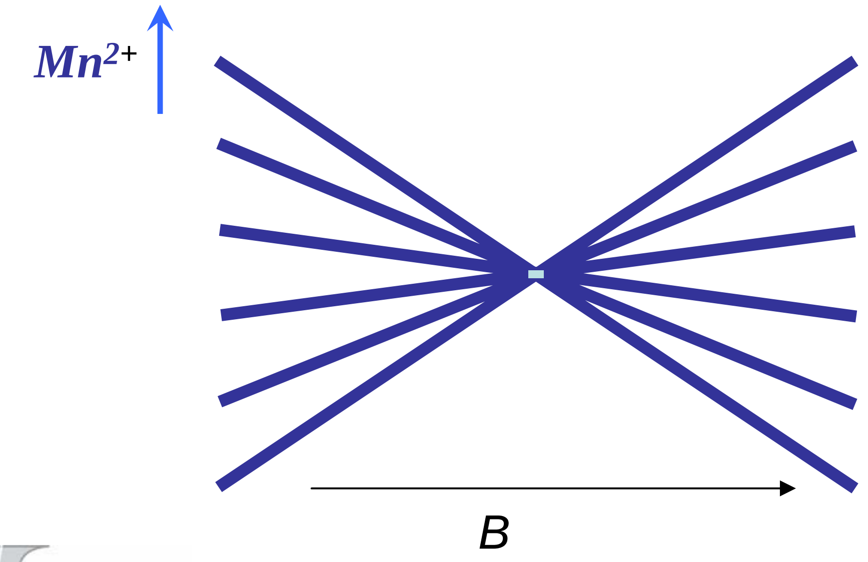




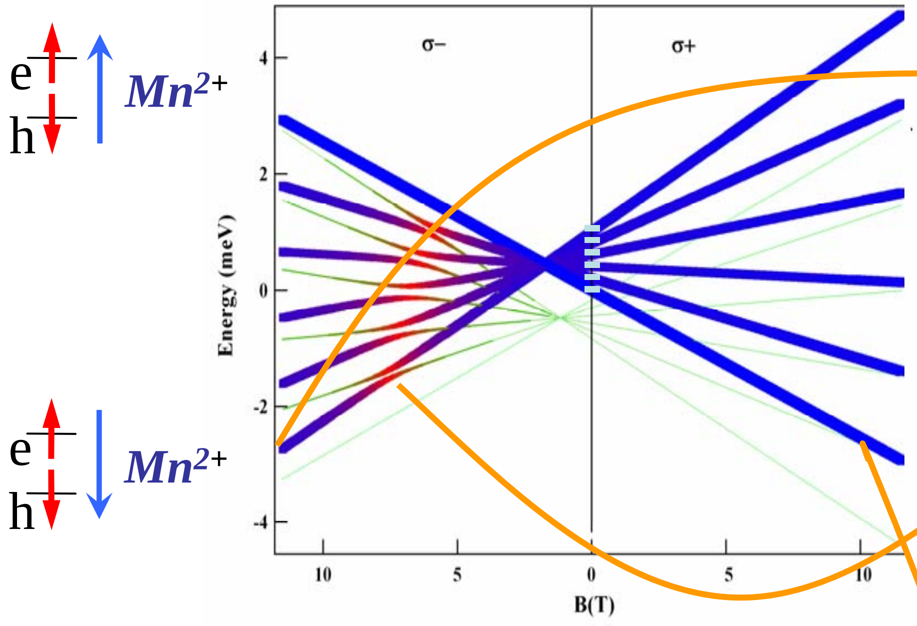
DMS: a single Mn spin in a single quantum dot.

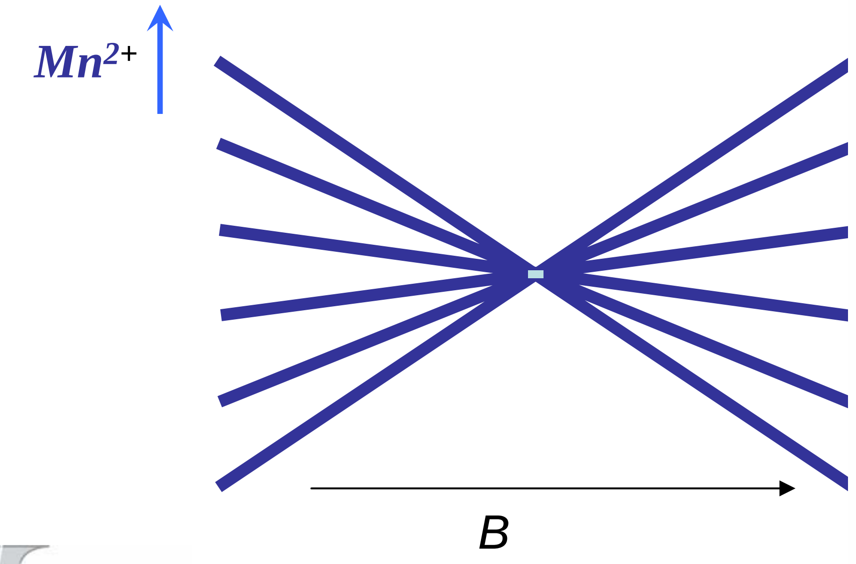
applying a magnetic field

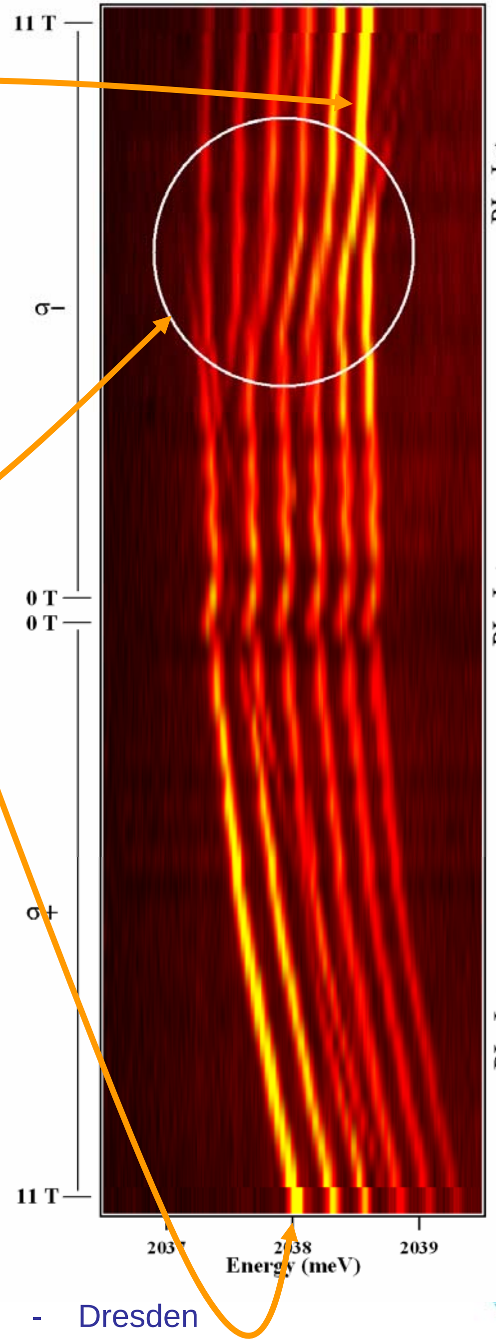


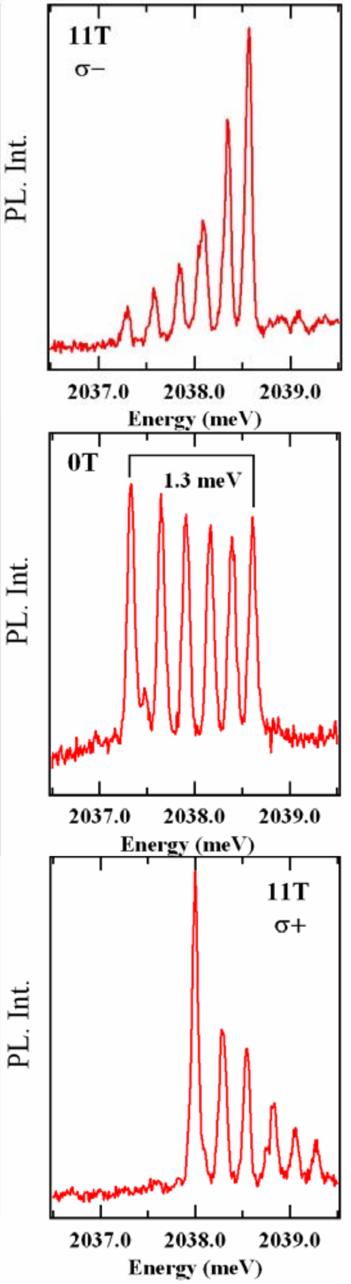


DMS: a single Mn spin in a single quantum dot.

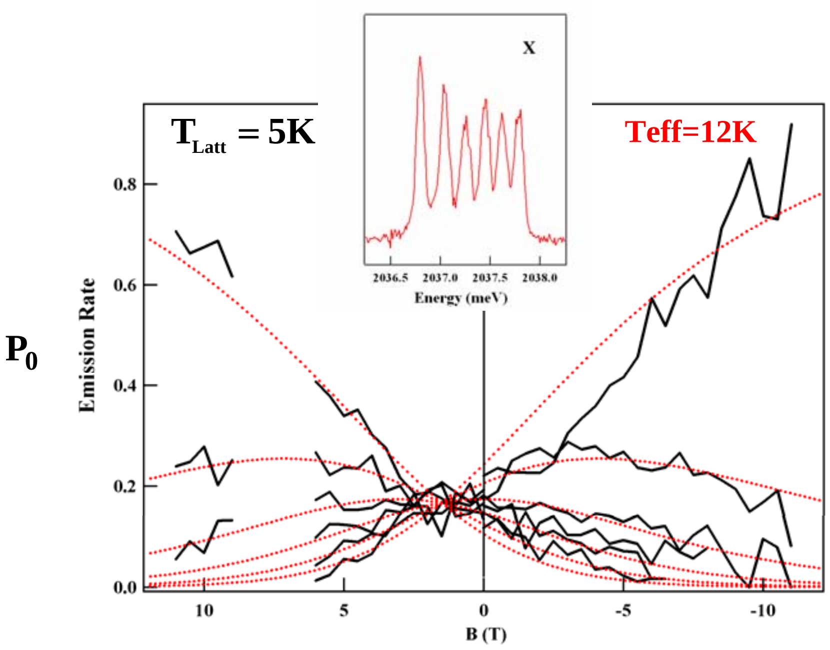






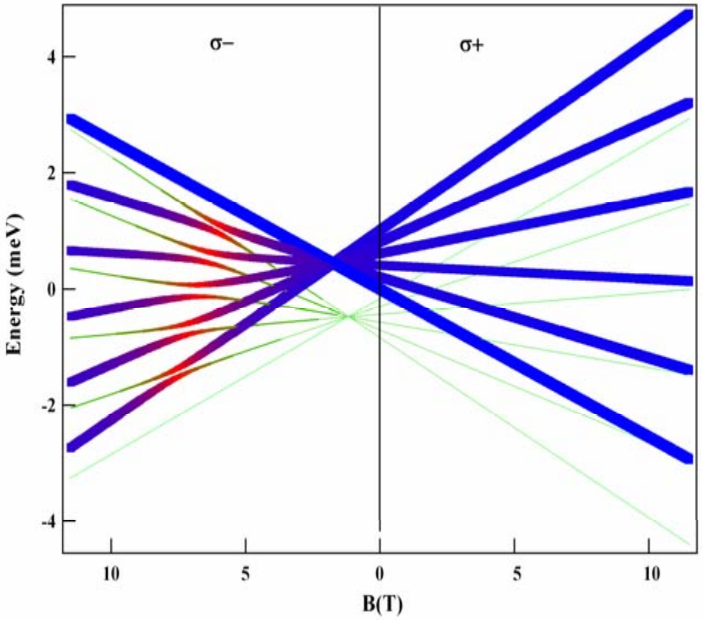


Polarization of the Mn Spin Distribution

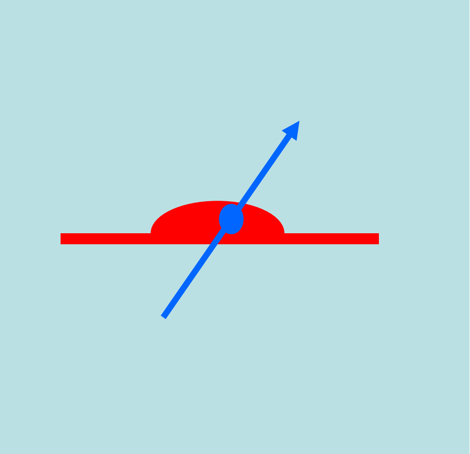


- ♦**Boltzmann distribution of the Mn-Exciton system:** $T_{eff} \neq T_{lattice}$

- ♦**Calculated Mn-Exciton energy levels:**

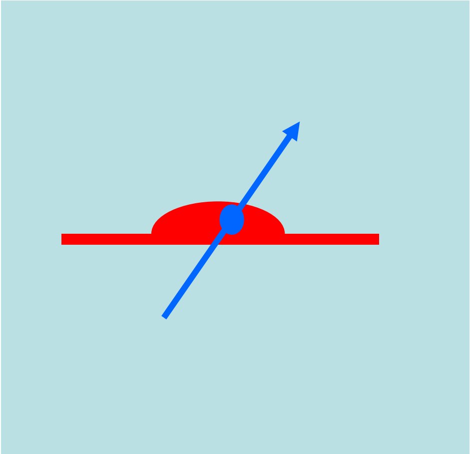


⇒ ***T_1* << exciton lifetime (ns)**

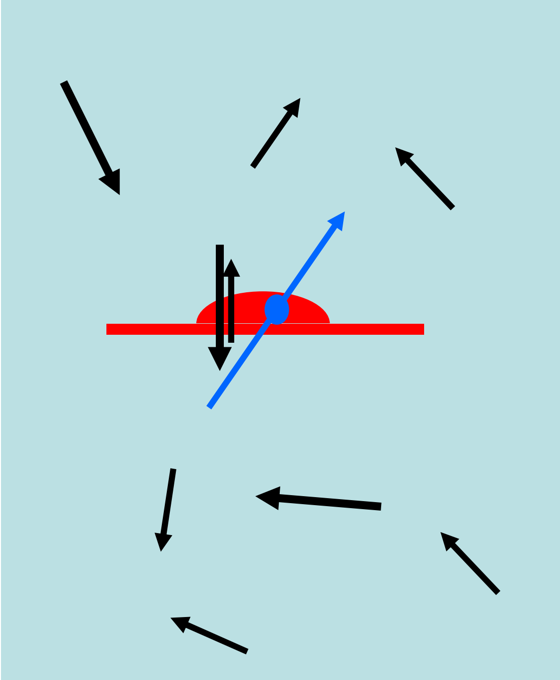


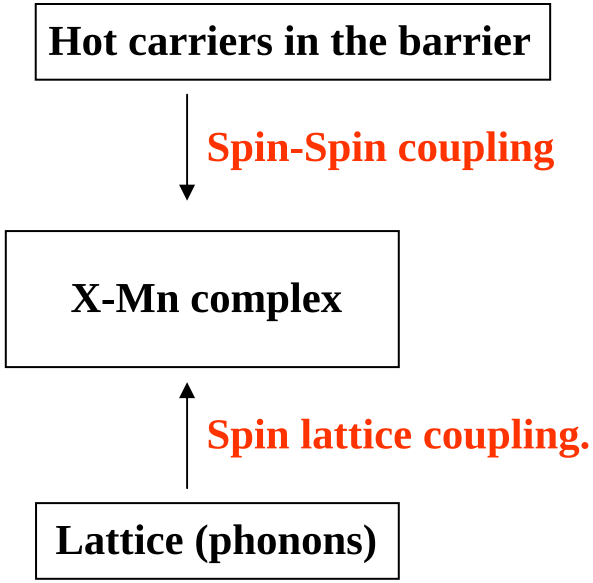
one Mn spin:
T_1, T_2 >> ms

T_1 << ns



one Mn spin:
$T_1, T_2 >> ms$





Hot carriers in the barrier

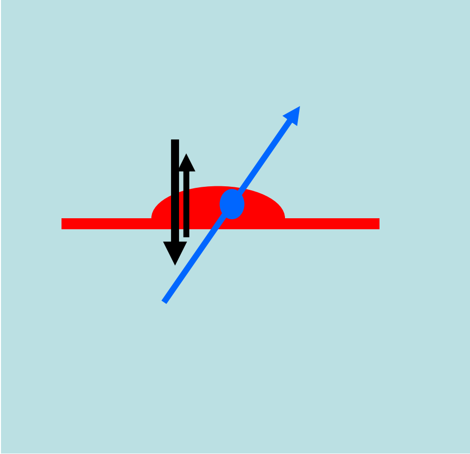
Spin-Spin coupling

X-Mn complex

Spin lattice coupling.

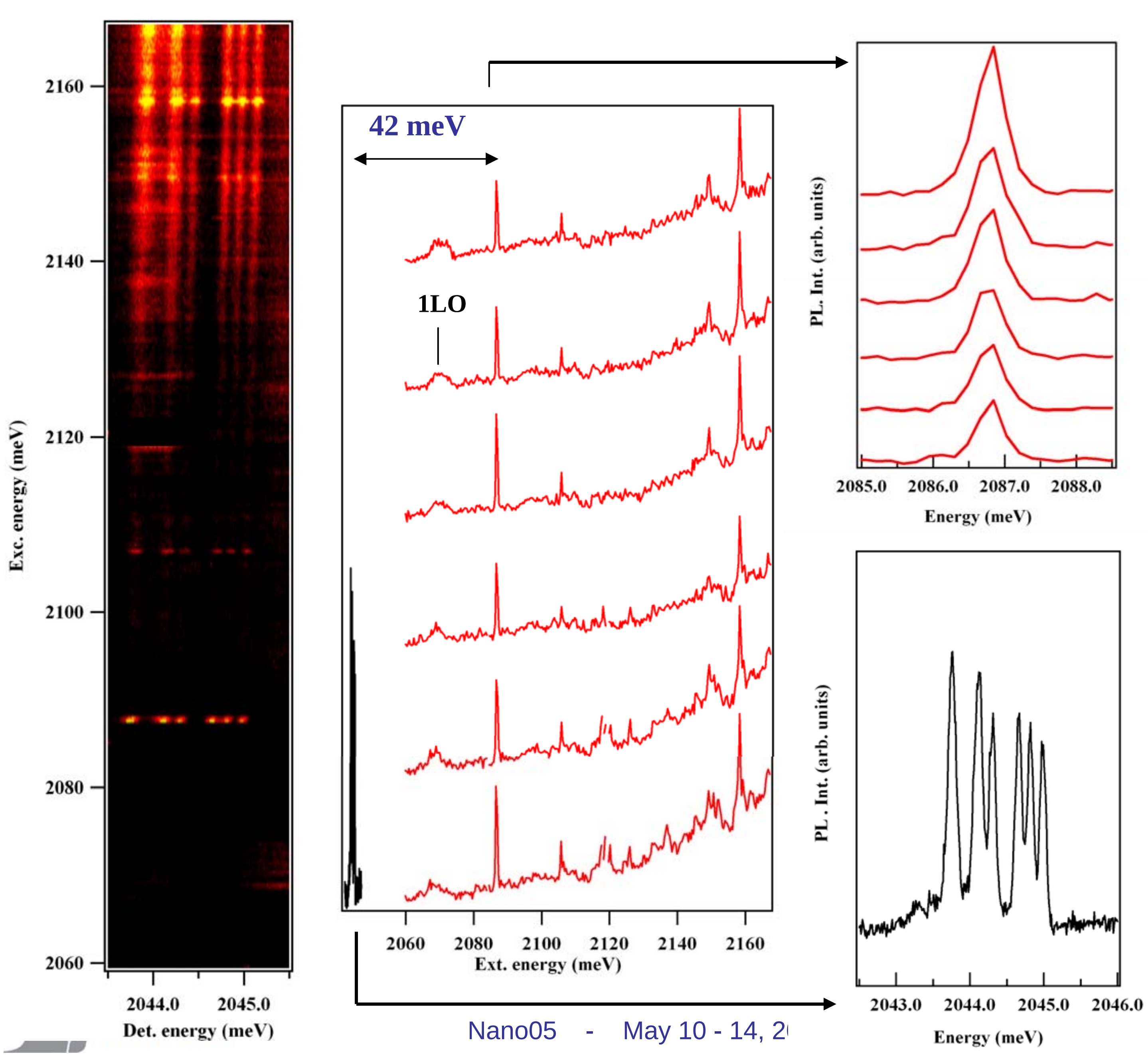
Lattice (phonons)

$T_1 << ns$



?

Excited States in Mn Doped QDs



♦Complex excited states Structure:
- -lh
- -phonons
- -p states
- -Indirect transitions

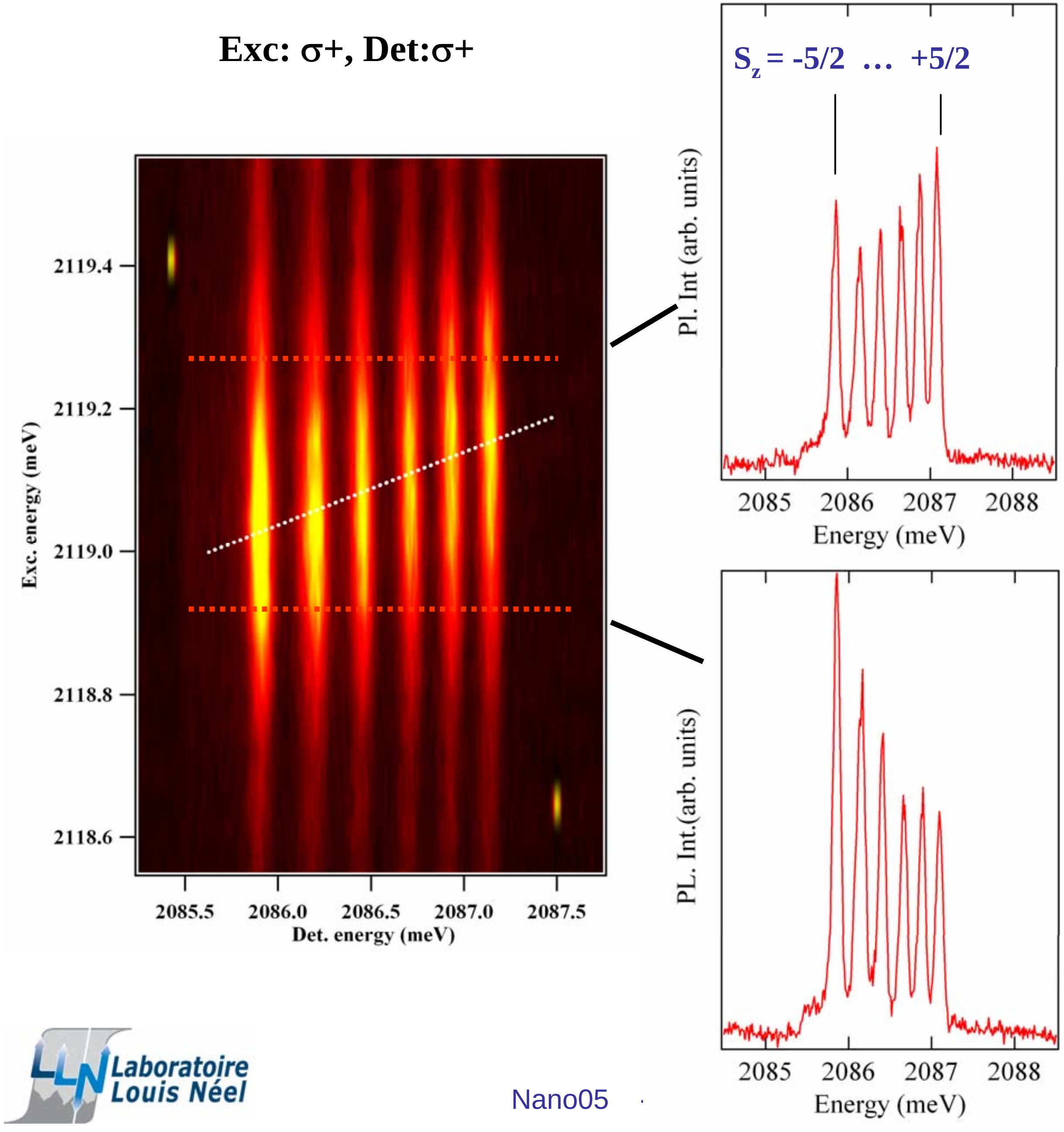
♦Broad excited state: Weak exchange coupling with the excited states.

⇕

Weaker exciton-Mn overlap.

Spin Selective Excitation of a Single Magnetic Atom

Exc: σ+, Det:σ+



- ♦**Residual exchange coupling with the Mn atom.**

⇩

Select the spin state of the Mn atom with the excitation energy.

- ♦**Non-equilibrium distribution**

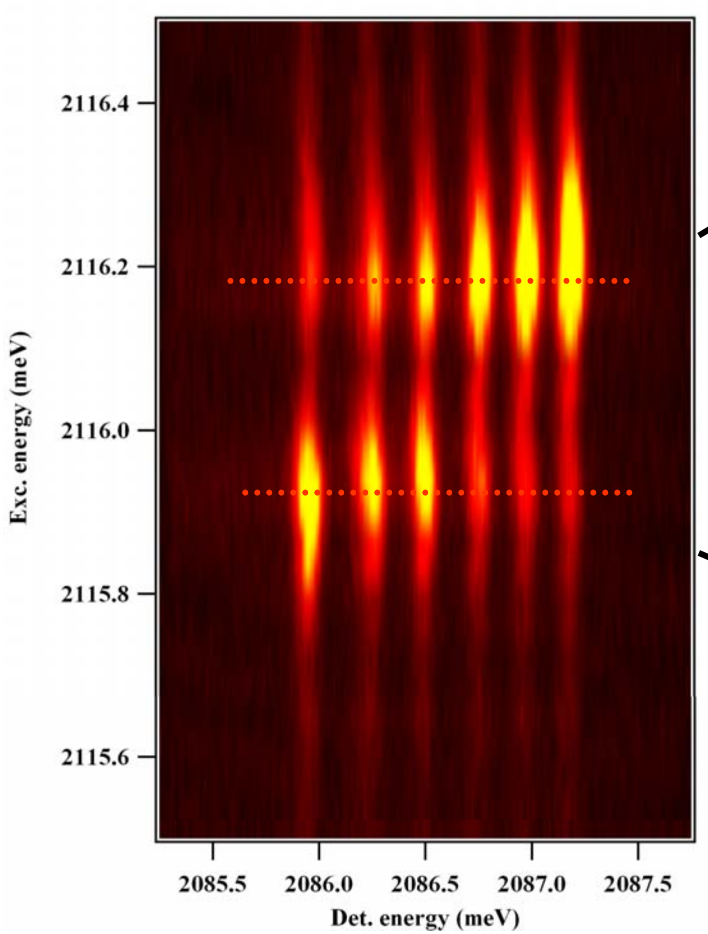
⇩

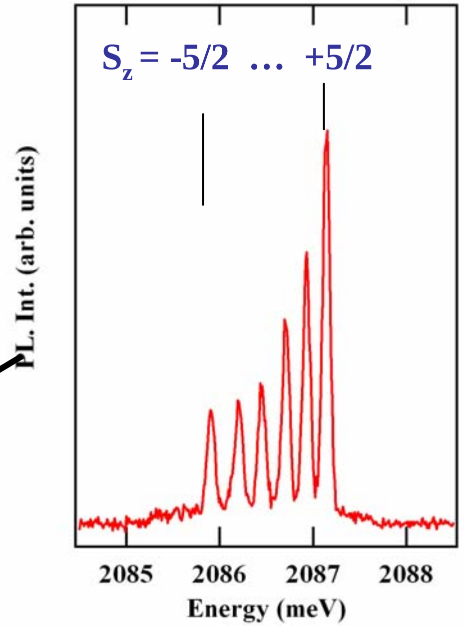
Weak relaxation of the X-Mn during the lifetime of the exciton.

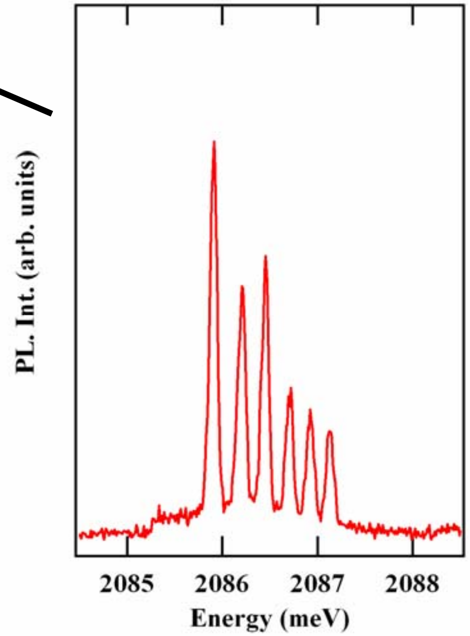
The exciton can probe the spin state of the Mn atom.

Spin Selective Excitation of a Single Magnetic Atom

Exc: σ+, Det:σ+







- ♦Residual exchange coupling with the Mn atom.

⇩

Select the spin state of the Mn atom with the excitation energy.

- ♦Non-equilibrium distribution

⇩

Weak relaxation of the X-Mn during the lifetime of the exciton.

The exciton can probe the spin state of the Mn atom.

DMS: a single Mn spin in a single quantum dot.

one single quantum dot, with a single Mn spin inside
with one electron-hole pair

slow dynamics if resonantly excited
spin relaxation

how slow?
spin manipulation?