Saint-Petersburg State University

SOLAB

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Spin phenomena in quantum dots revealed by charged exciton (trion) photoluminescence

Sergei VERBIN

Spin Optics Laboratory, St. Petersburg State University, 198504 Peterhof, Russia

E-mail: syuv54@mail.ru

Outline

Introduction.

I. *Subject of study* –

- trion photoluminescence (PL) of quantum dots (QDs) ensemble

II. Negative circular polarization (NCP)

of InP and InAs QDs trion photoluminescence itself

and as a *method of spin polarization study*

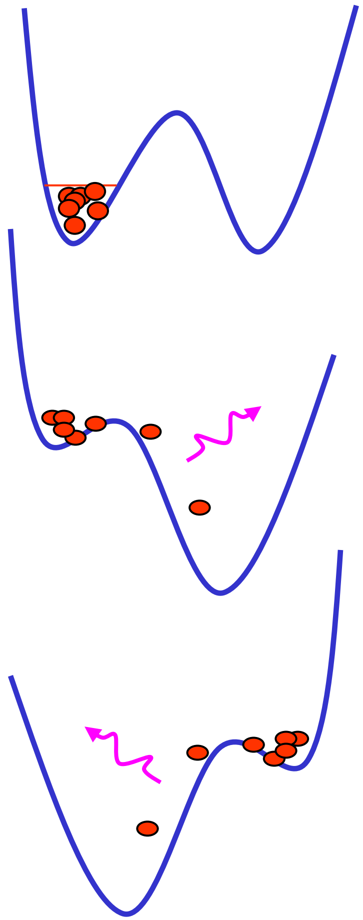
III. Long-lived spin polarization of resident electron in QDs

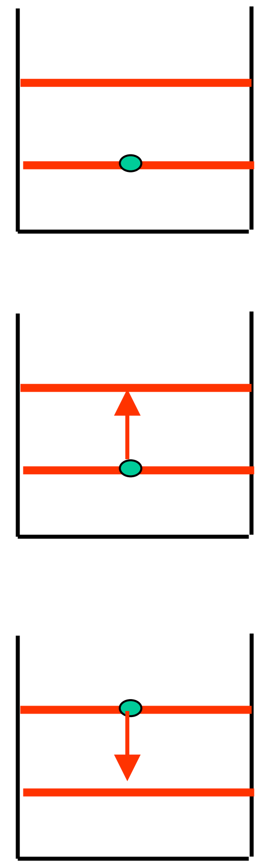
IV. Hyperfine interaction of electron and nuclear spins in QD

V. Time-resolved Hanle effect in QDs ensemble

Ad: Optical Detection of Nuclear Magnetic Resonance (ODNMR) at the QDs ensemble

Memory cell in electronics and spintronics





«1» is storing

«0» is storing

Spatial transfer of charge -

- **needs the time**
- **Joule heating**

No spatial transfer of charge –

1) **higher work frequency**
2) **no Joule heating**

The carrier scattering in QDs

The **study of carrier spin dynamics**

of quantum confined objects in heterostructures,

in particular, **in quantum dots** -

- one of the main tasks of **spintronics**

Main mechanisms of carrier scattering

"working" in the bulk semiconductors

are eliminated in QDs

due to the carrier localization.

The role of hyperfine interaction

From other side, due to the same carrier localization
the **electron spin dynamic** in QDs
is dependent on the **hyperfine interaction**
of electron and nuclear spins
more than in bulk semiconductors.

Results of our recent study of the effect
of hyperfine interaction on spin dynamic in QDs
are briefly reviewed in this report.

List of main reviewed papers

I.Ya.Gerlovin et al., *Phys. Rev. B*, **69**, 035329 (2004).

M.Ikezawa et al., *Phys. Rev. B*, **72**, 153302 (2005).

R.Oulton et al., *phys. stat. sol. (B)*, **243**, 3922 (2006).

B.Pal et al., *Phys. Rev. B*, **75**, 125322 (2007)

R.Oulton et al., *Phys. Rev. Lett.*, **98**, 107401 (2007).

R.V.Cherbunin et al., *Phys. Rev. B*, **80**, 035326 (2009).

T.Auer et al., *Phys. Rev. B*, **80**, 205303 (2009).

I.V.Ignatiev et al., *Opt.&Spectr.*, **106**, 375 (2009).

K.Flisinski et al., *Phys. Rev. B*, **82**, 081308 (2010).

S.Yu.Verbin et al., *JETP*, **114**, 681 (2012).

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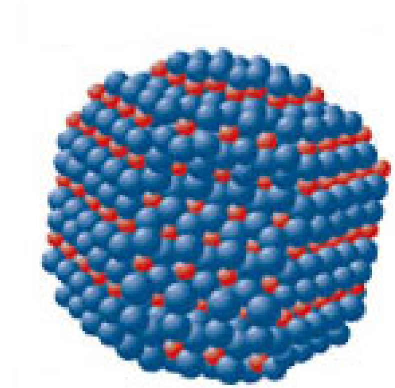
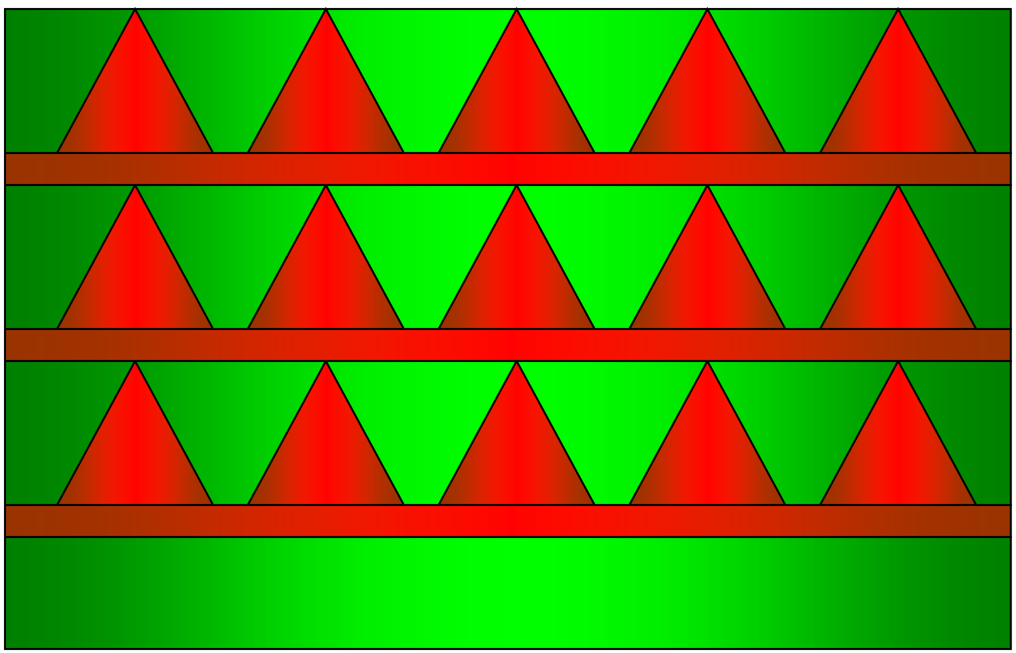
Quantum Dots = “artificial atoms”

Three-dimensional (3D) potential well with the size ~ de Broglie wavelength =>

=> electronic levels in quantum dots are well resolved in energy (!)

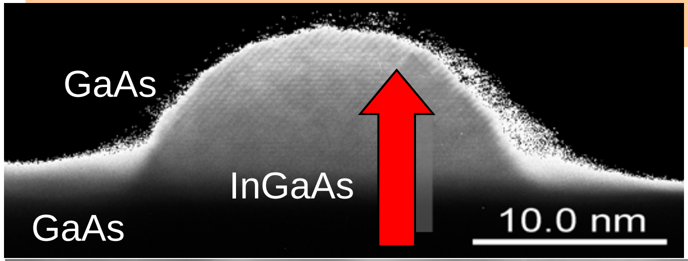
Single ZnSe quantum dot in broader-band environment (semiconductor, glass, liquid etc) -->

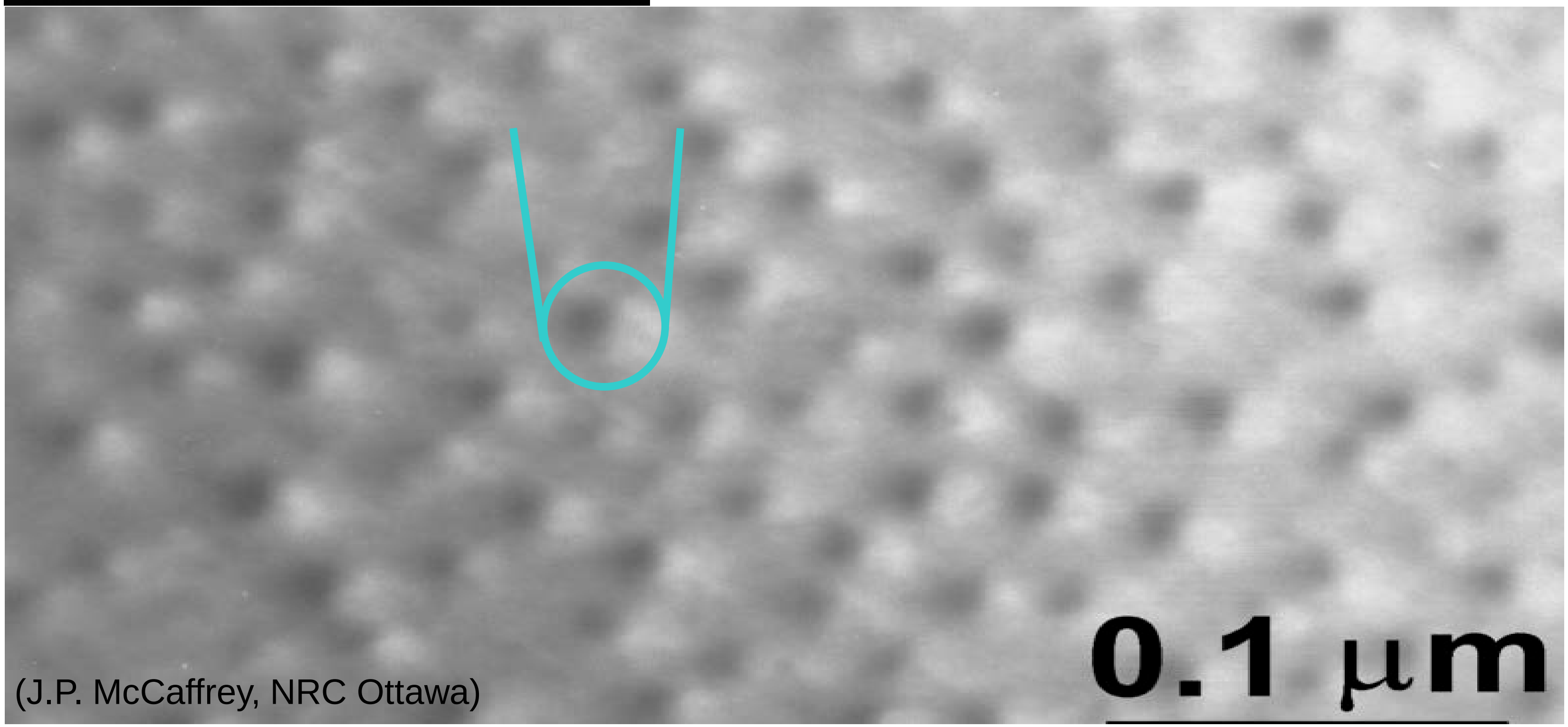




← Semiconductor heterostructure with quantum dots <u>ensemble</u> – nanocrystals in the bulk “barrier” semiconductor with broader forbidden gap E_g

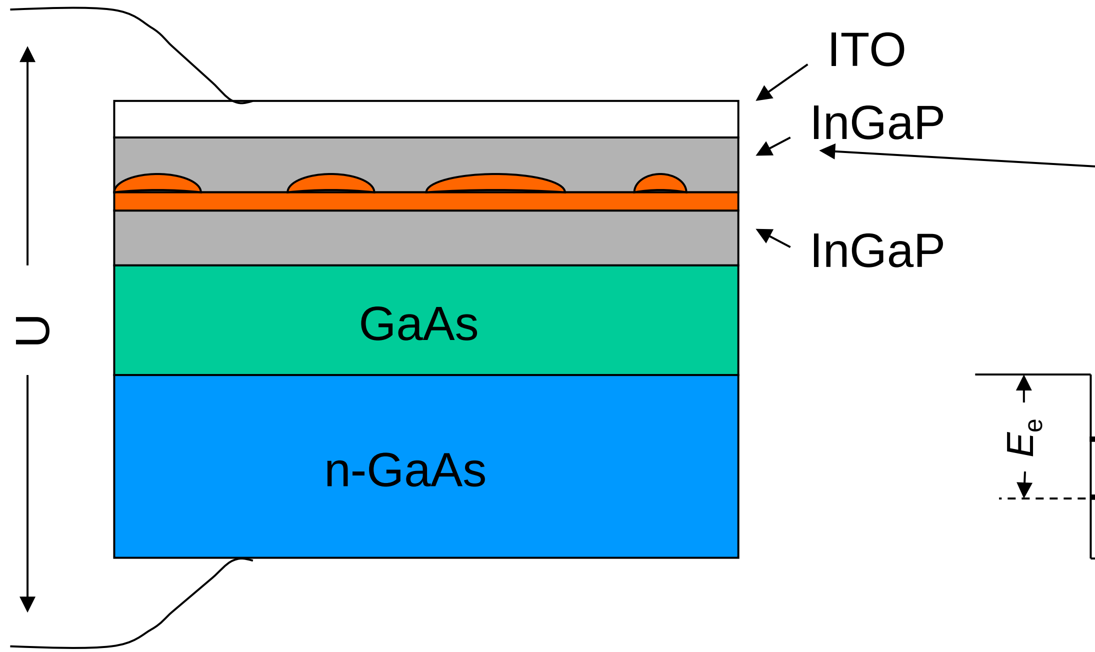
Single InGaAs/GaAs quantum dot

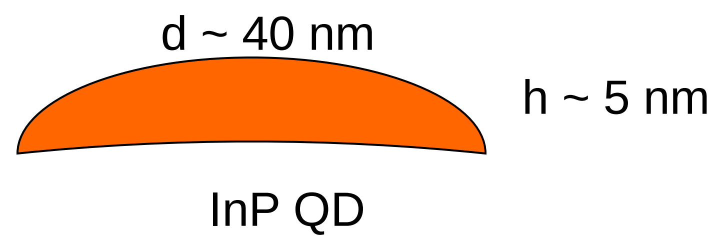


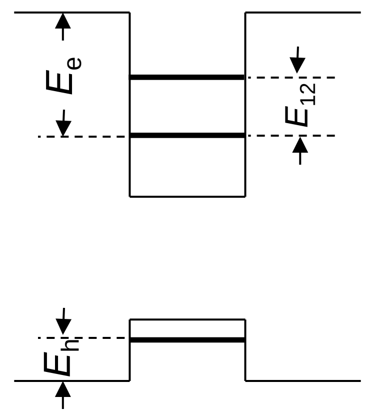


Single quantum dot spectroscopy – the particular field of research

Subject of the study: InP/InGaP QD ensemble

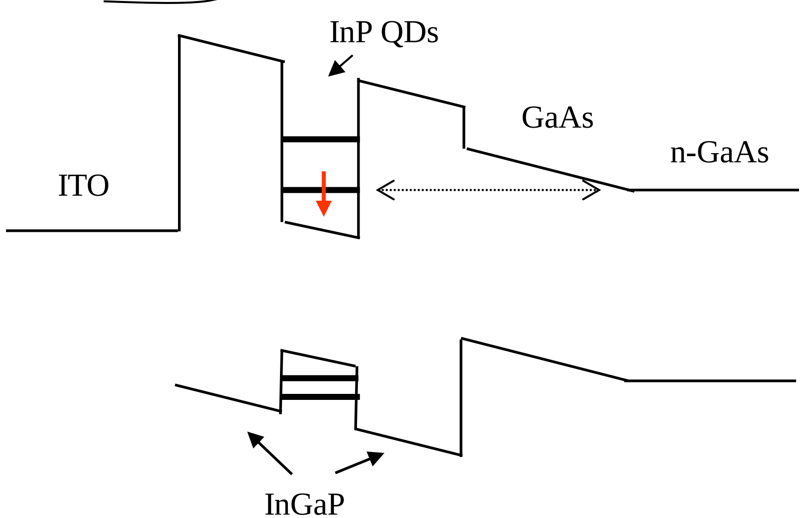






E_e ~ 230 meV;
E_{12} ~ 10 – 20 meV
E_h ~ 15 meV;

QDs density ~ 10^{10} cm^{-2} (AFM);

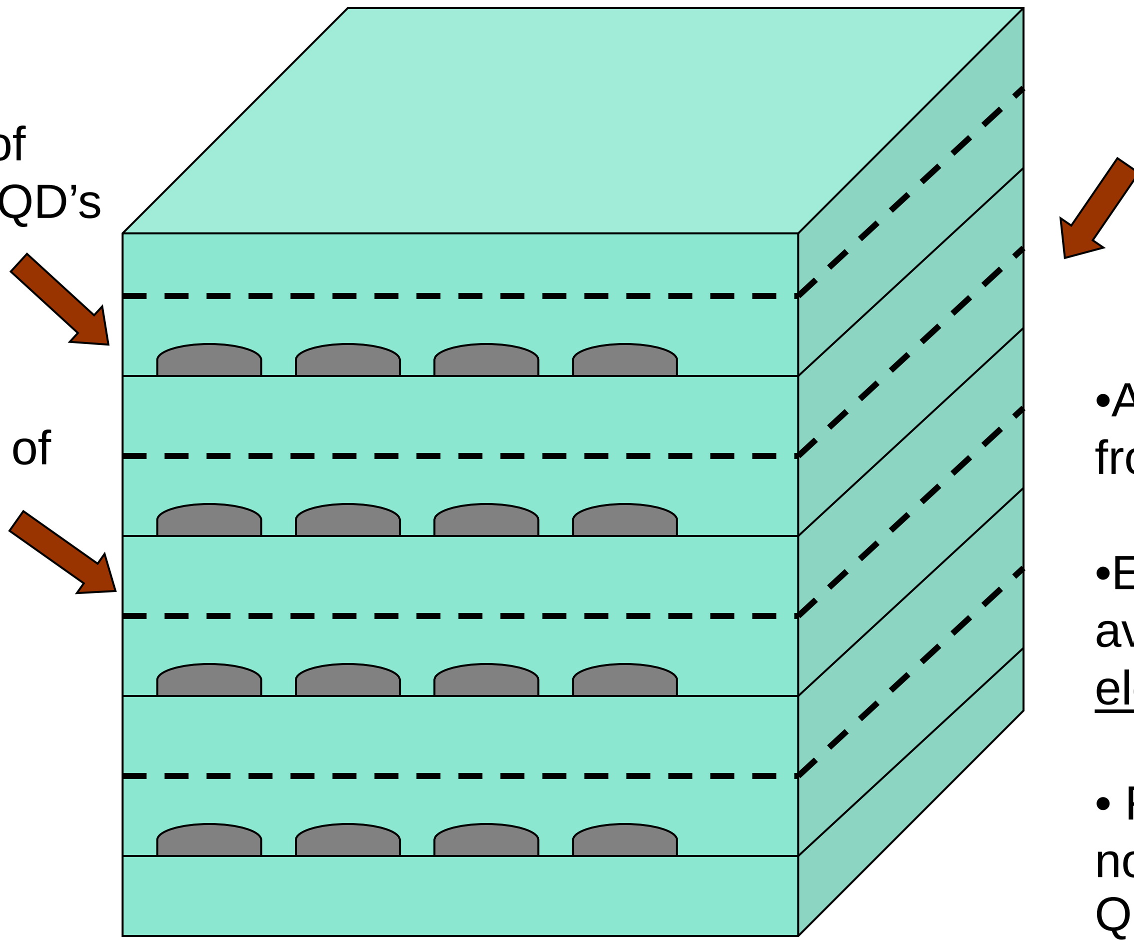


InP QD in external electric field

Subject of study:
(In,Ga)As/GaAs QDs ensemble

20 layers of
(In,Ga)As QD's

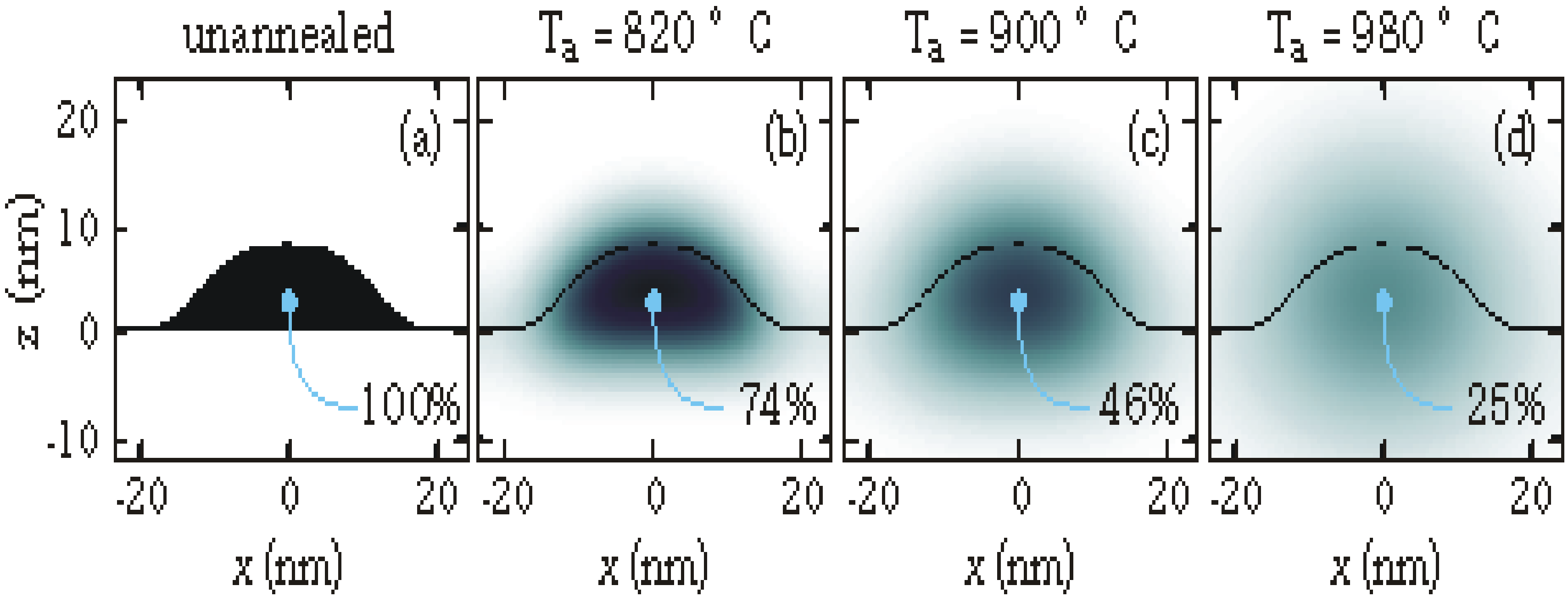
Barriers of
GaAs



n-δ-modulation
doped sheets of Si

- Annealing at T from 900 °C to 980°C
- Every QD with **one** on average a single resident electron
- Reference samples have no δ-doped sheets => QDs don't contain resident electrons

Sample annealing

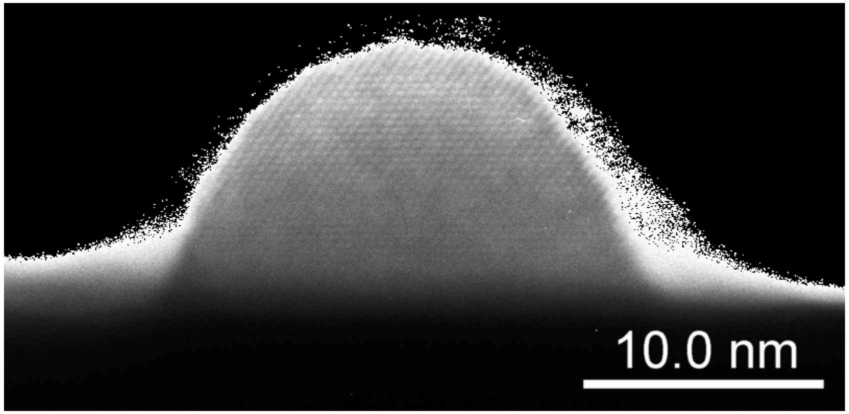


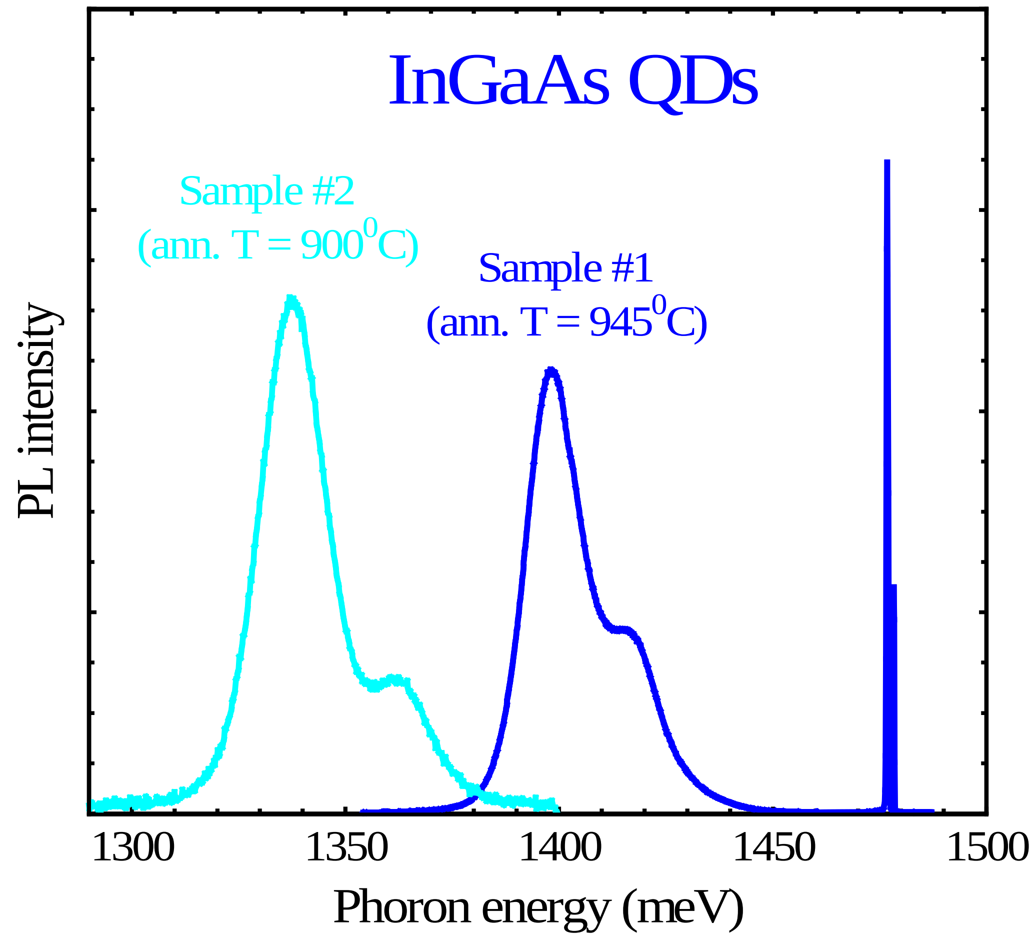
M.Yu. Petrov et al. PRB (2008)

As grown:
- 20 layers of InAs QDs in GaAs

Post growth annealing:
- InAs → InGaAs

PhotoLuminescence (PL) characterization





20 layers of InGaAs QDs with areal density $\rho \sim 10^{10}$ cm^{-2}

Statistic distribution of QDs size and composition in the ensemble under study leads to the inhomogeneous broadening of QD emission bands in PL spectra

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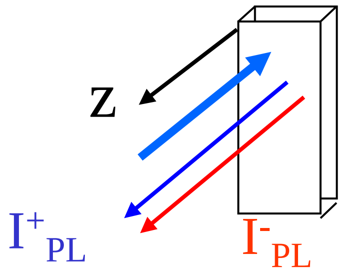
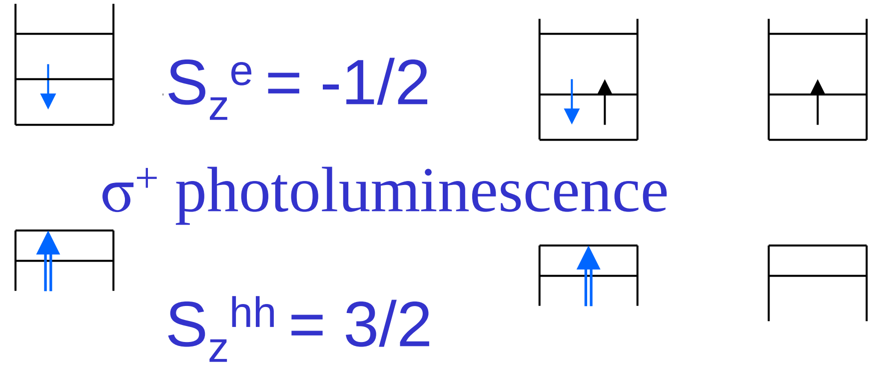
σ^+ excitation by circularly polarized light



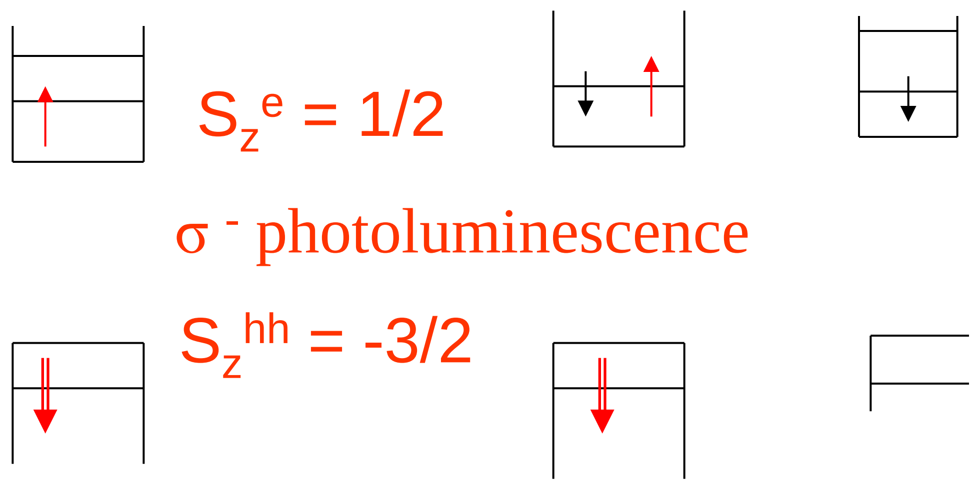
Sample

PhotoLuminescence (PL)

Neutral QDs Negatively charged QDs



Exciton Trion (exciton + resident electron)



We studied ρ^c_{PL} - degree of the PL circular polarization:

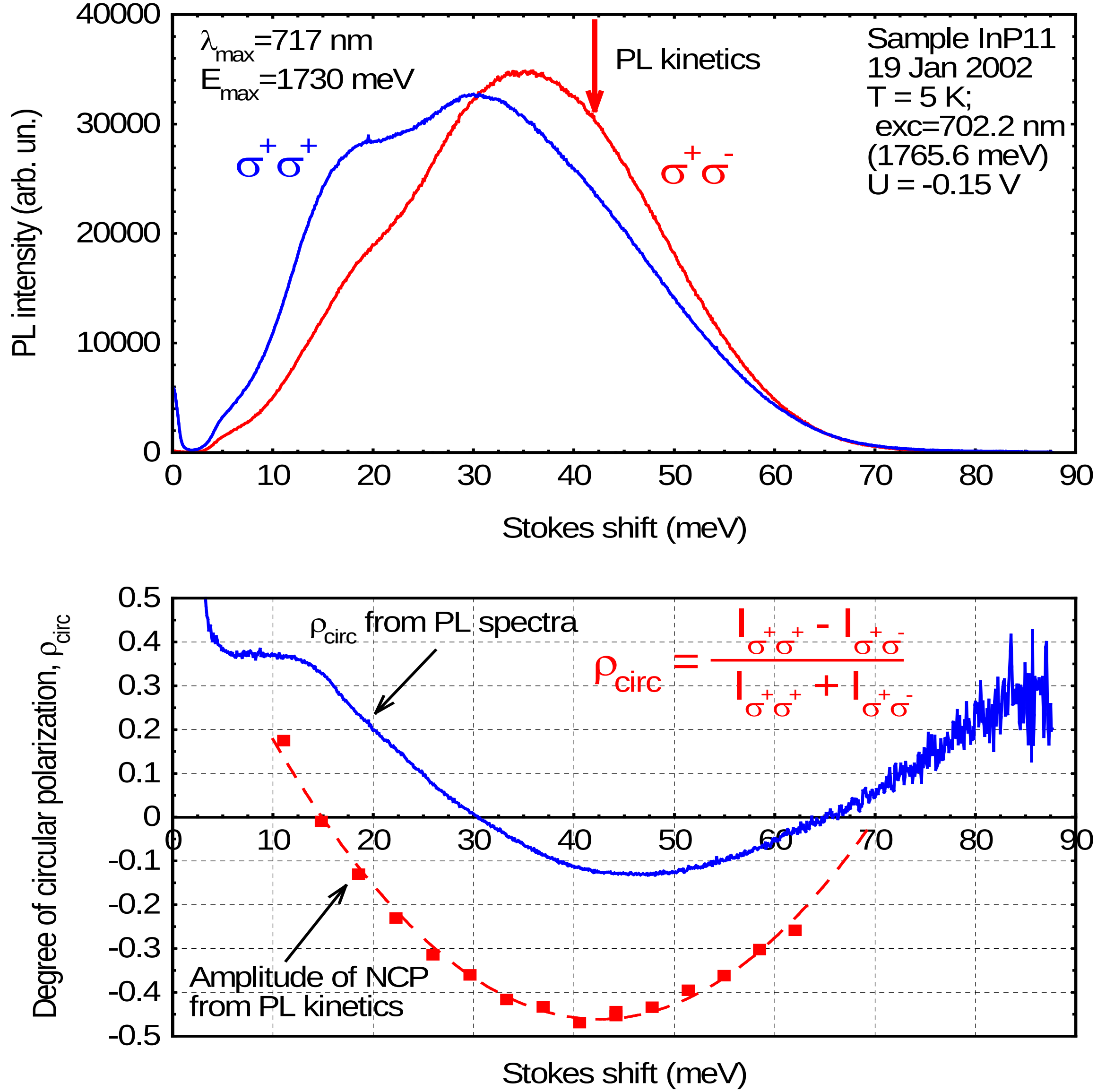
$$\rho^c_{PL} = \frac{I^s_{PL} - I^o_{PL}}{I^s_{PL} + I^o_{PL}}$$

in dependence on $h\nu_{det}$ (spectra) and on time delay after exciting light pulse (kinetics).

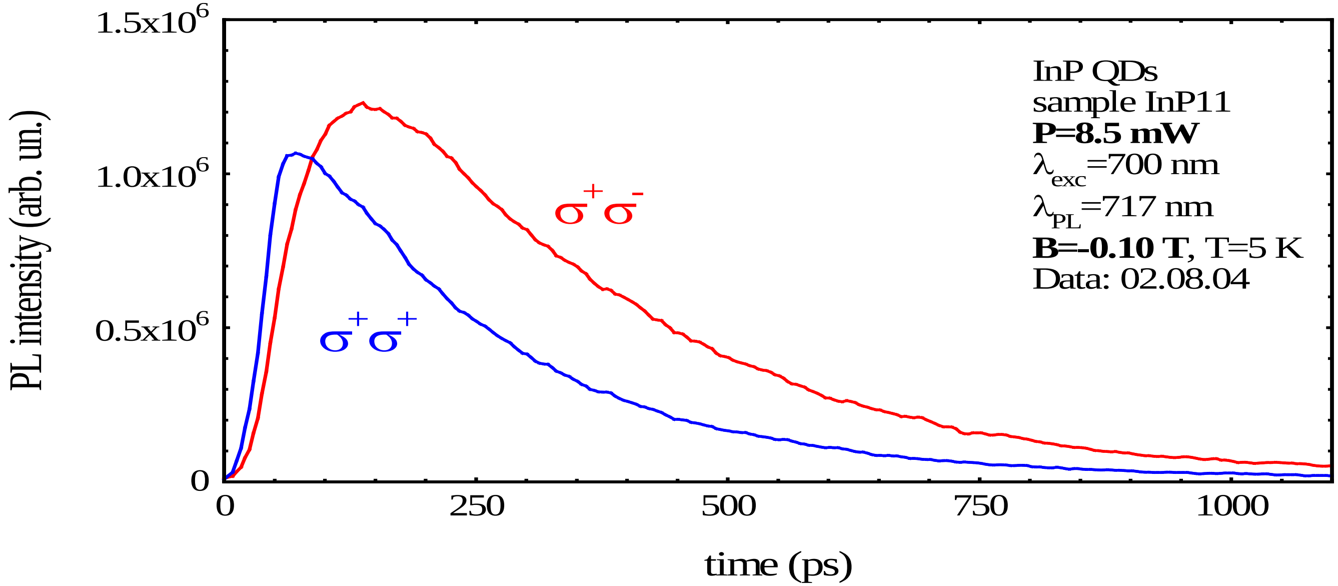
$I^{s(o)}_{PL}$ - the intensity of the PL component with the same (opposite) helicity* as that of the excitation beam

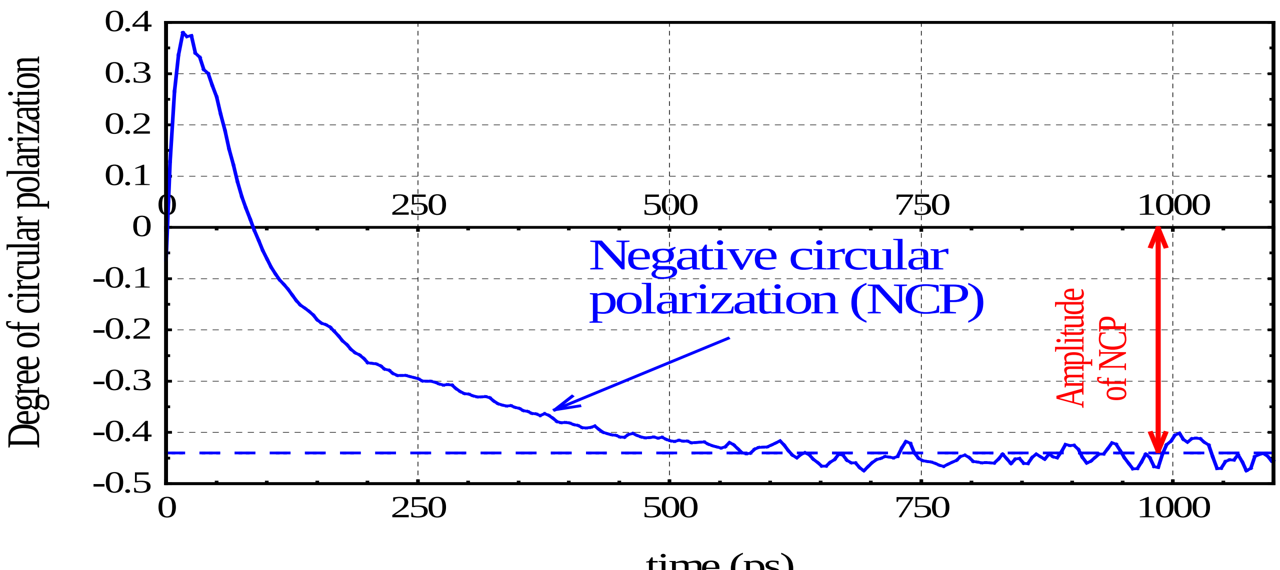
*Helicity – sign of circular polarization degree

Circularly polarized PL of InP QDs

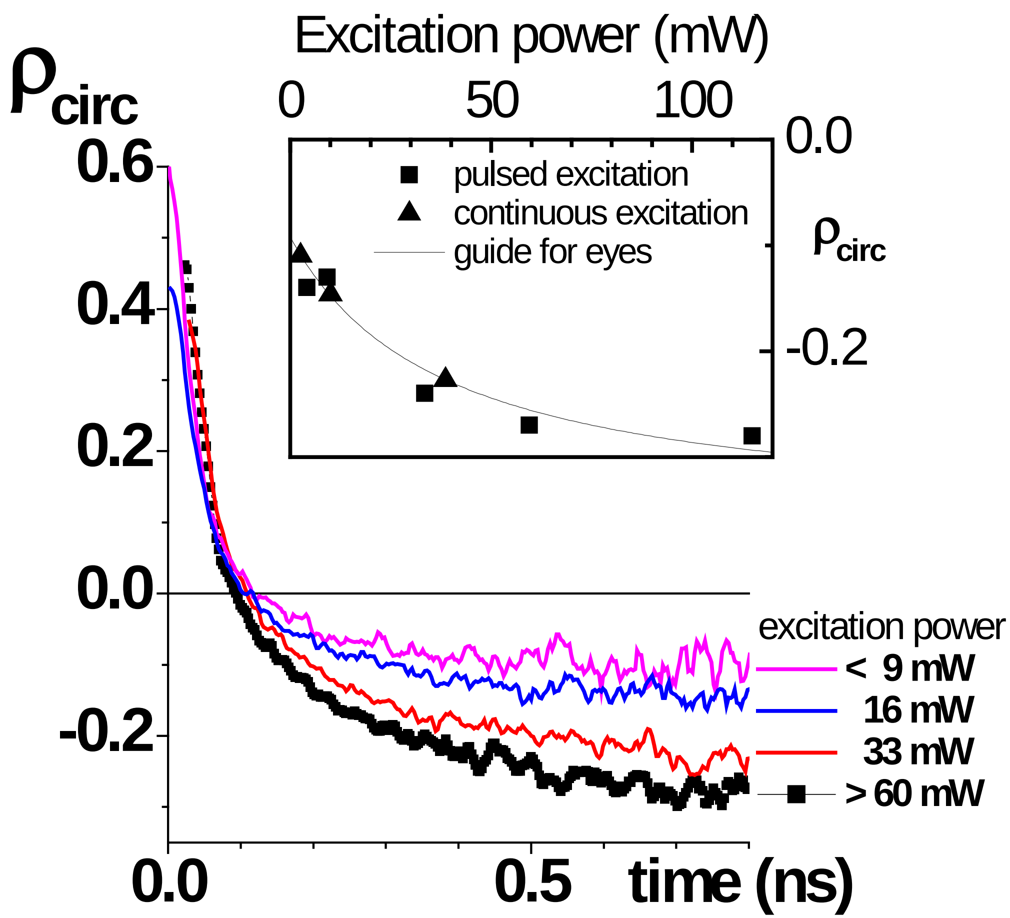


Kinetics of circularly polarized PL of InP QDs



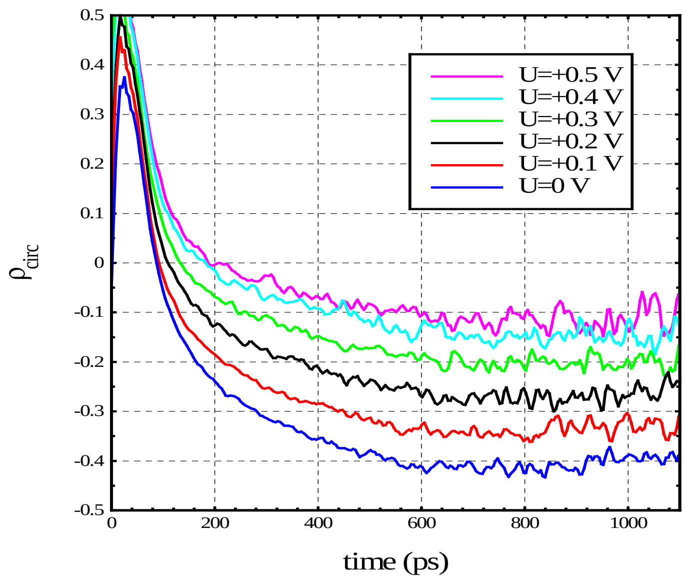


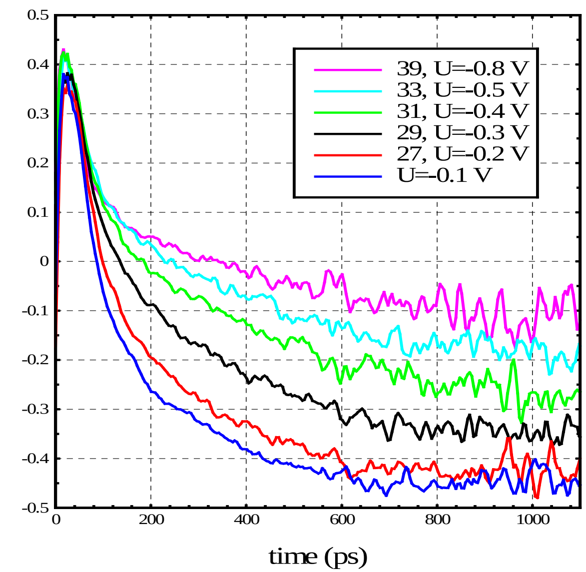
Power dependence of NCP kinetics

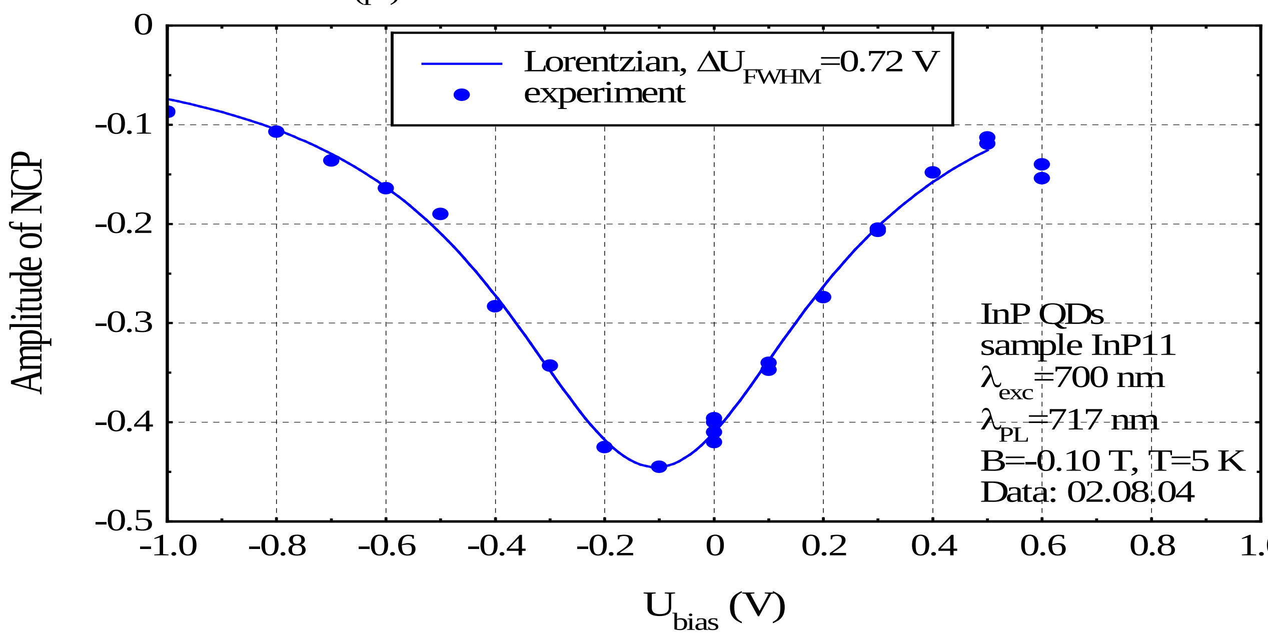


Power dependence of NCP value demonstrates the rise of orientation of **resident** electron spins.

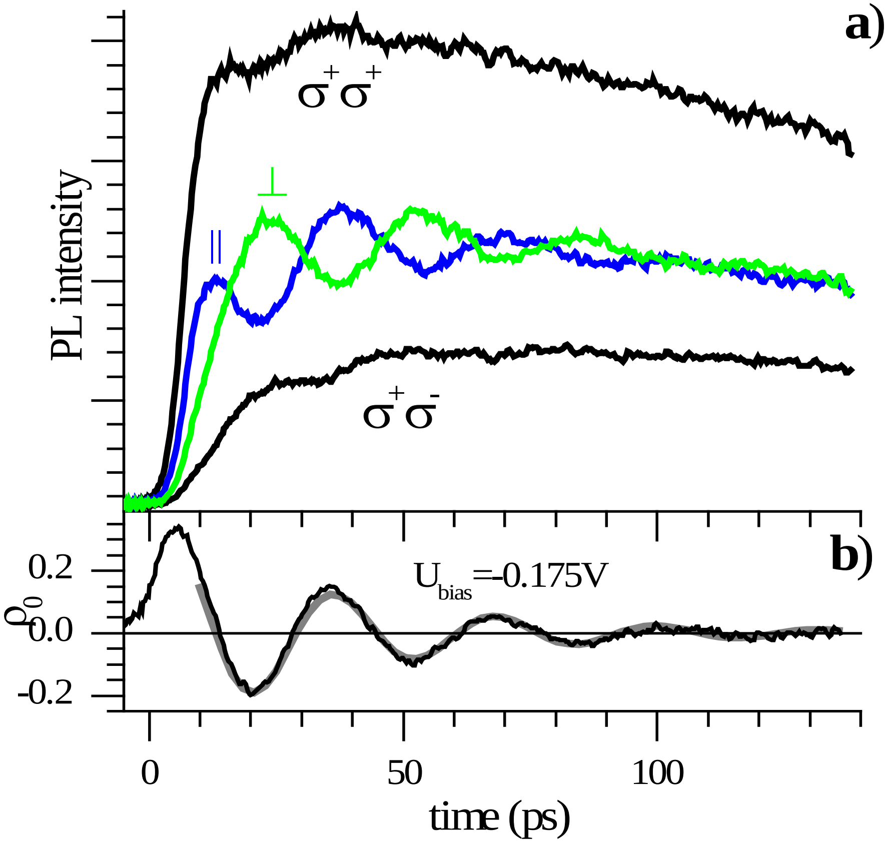
Dependence of NCP on applied bias

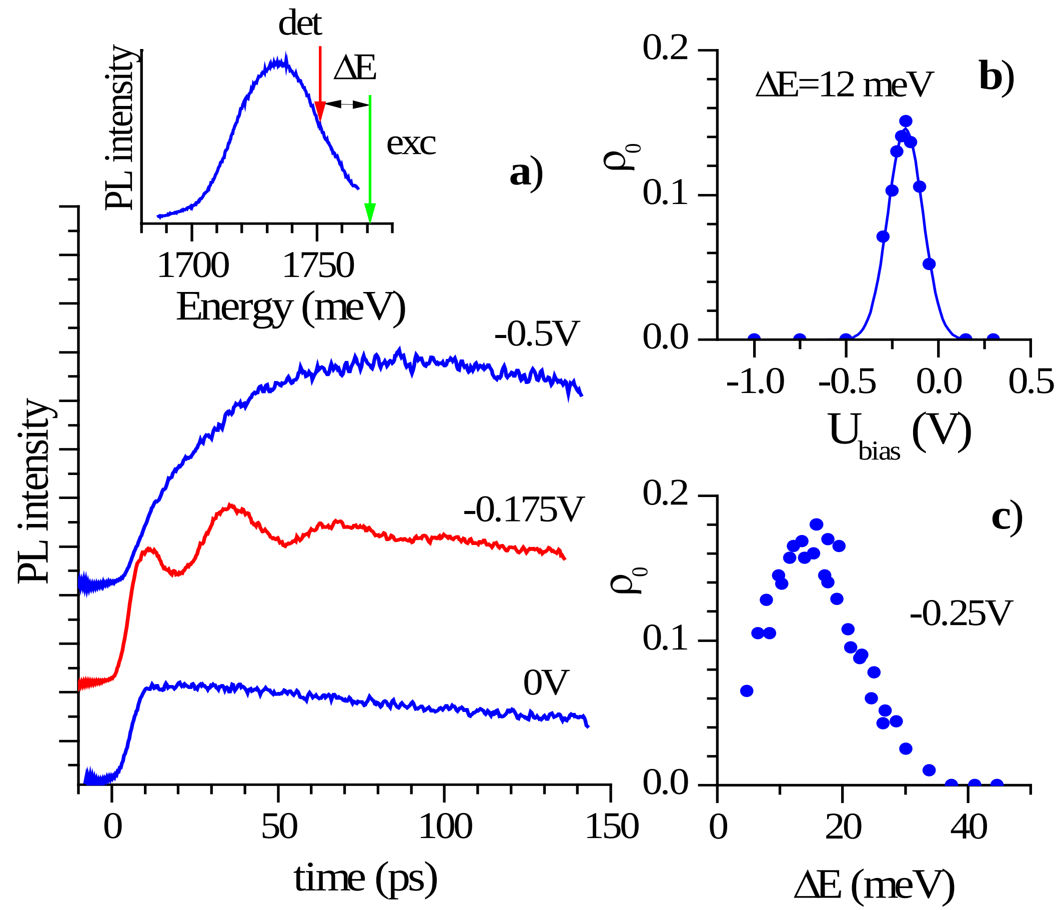






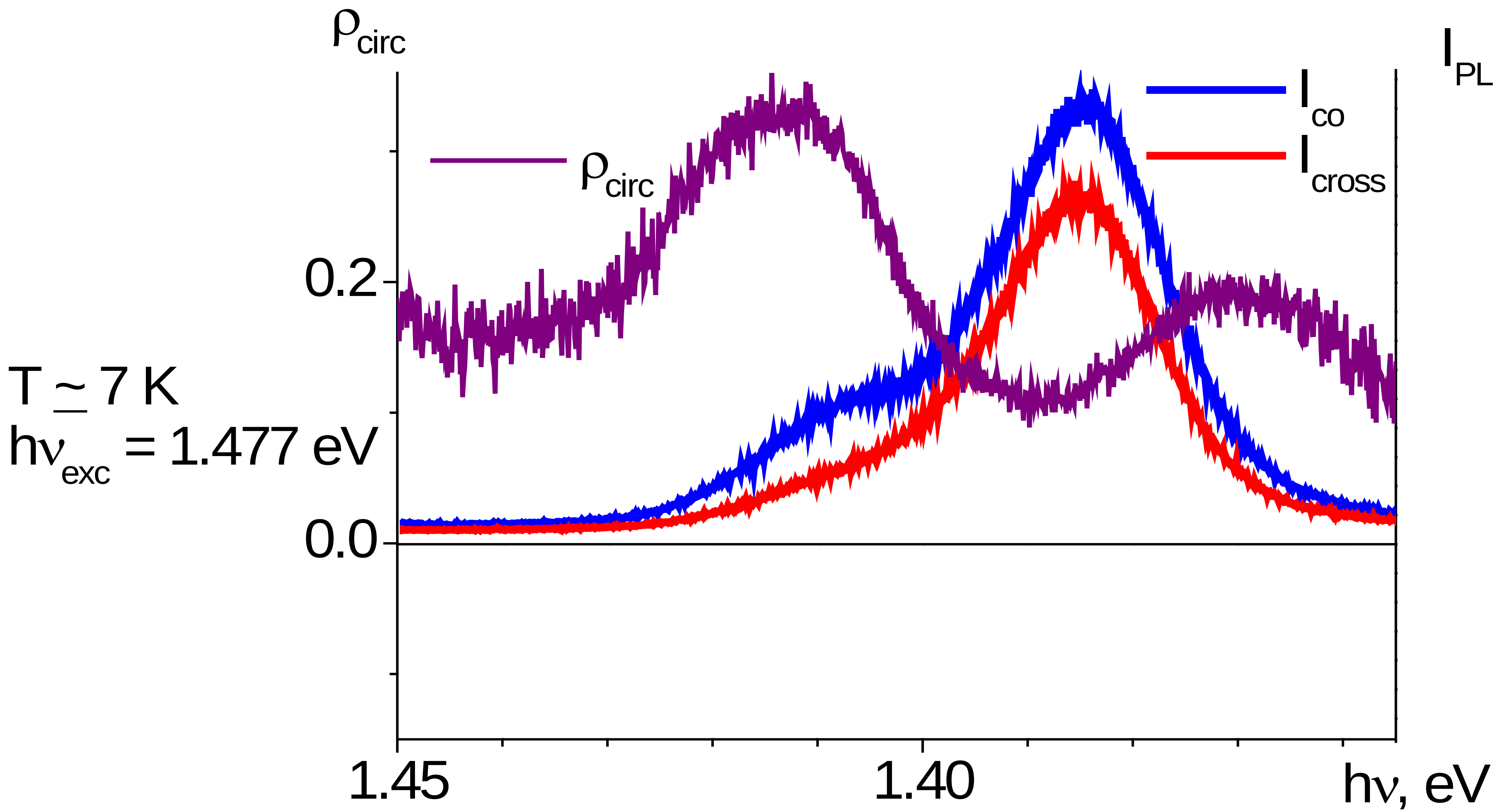
Bias dependence of <u>trionic</u> quantum beats



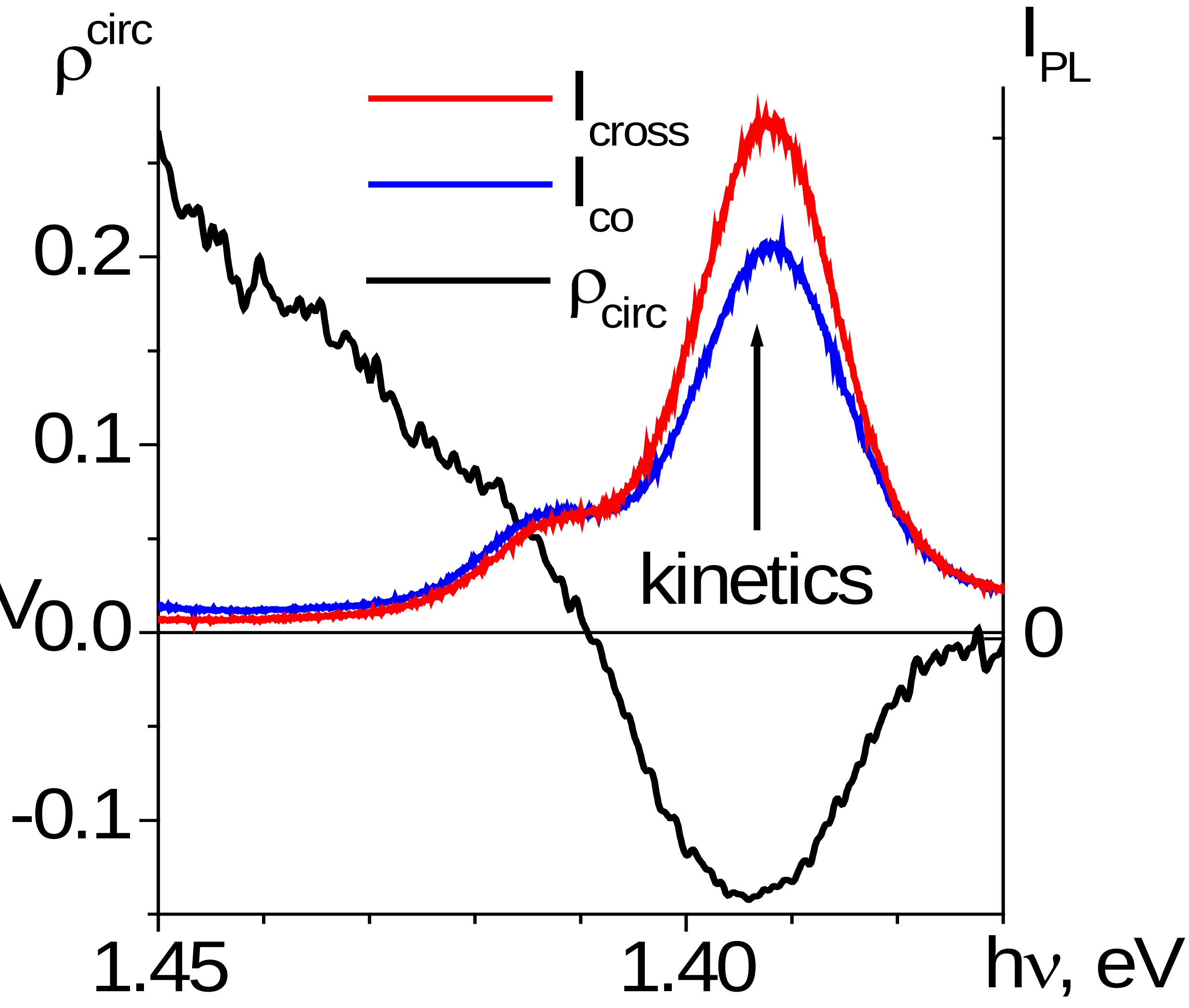


[Kozin *et al.*, PRB**65**, 241312(R) (2002)]

PL spectra polarization of uncharged InGaAs QDs (non-doped heterostructure)



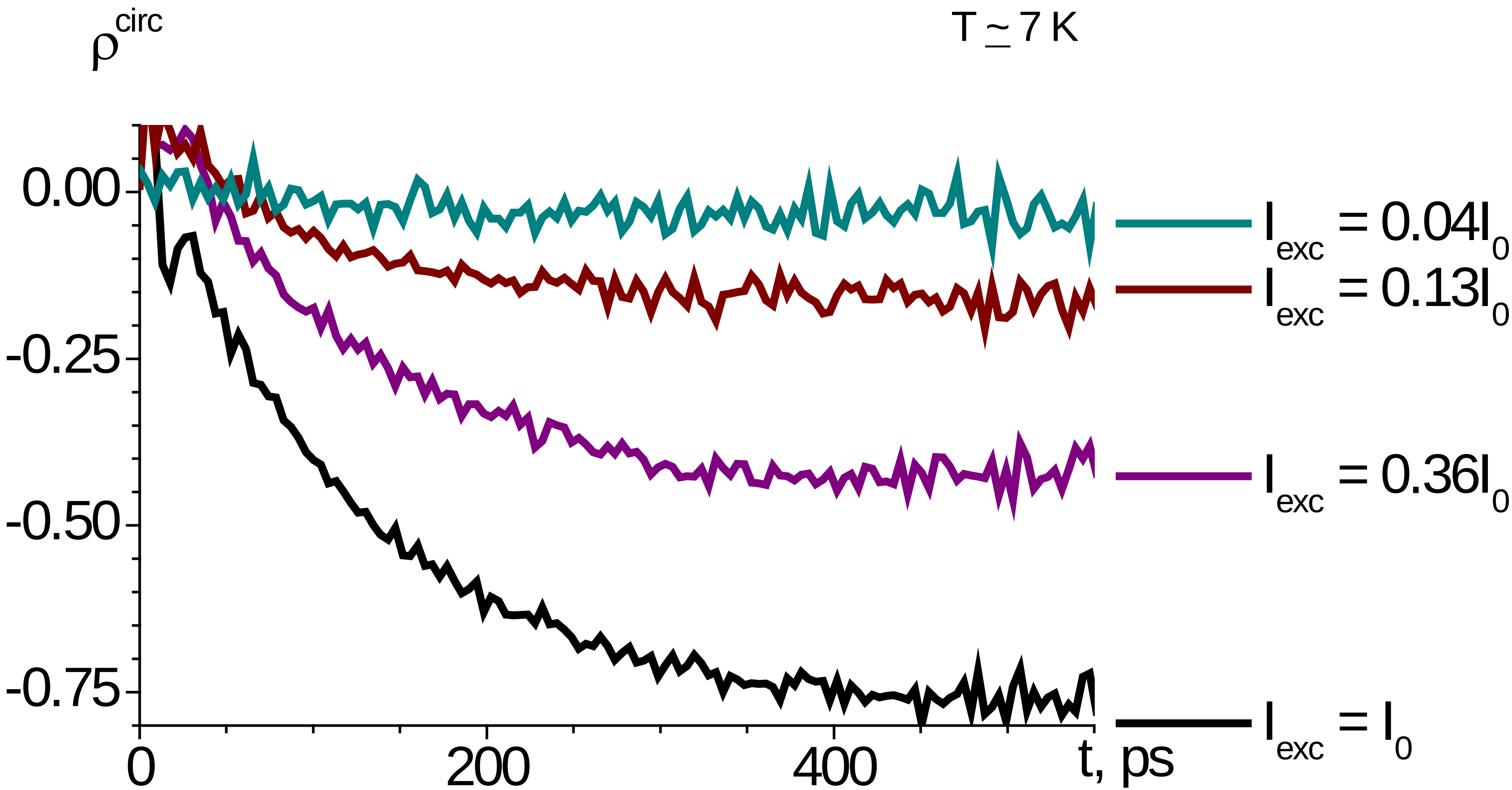
NCP of PL spectra of charged InGaAs QDs



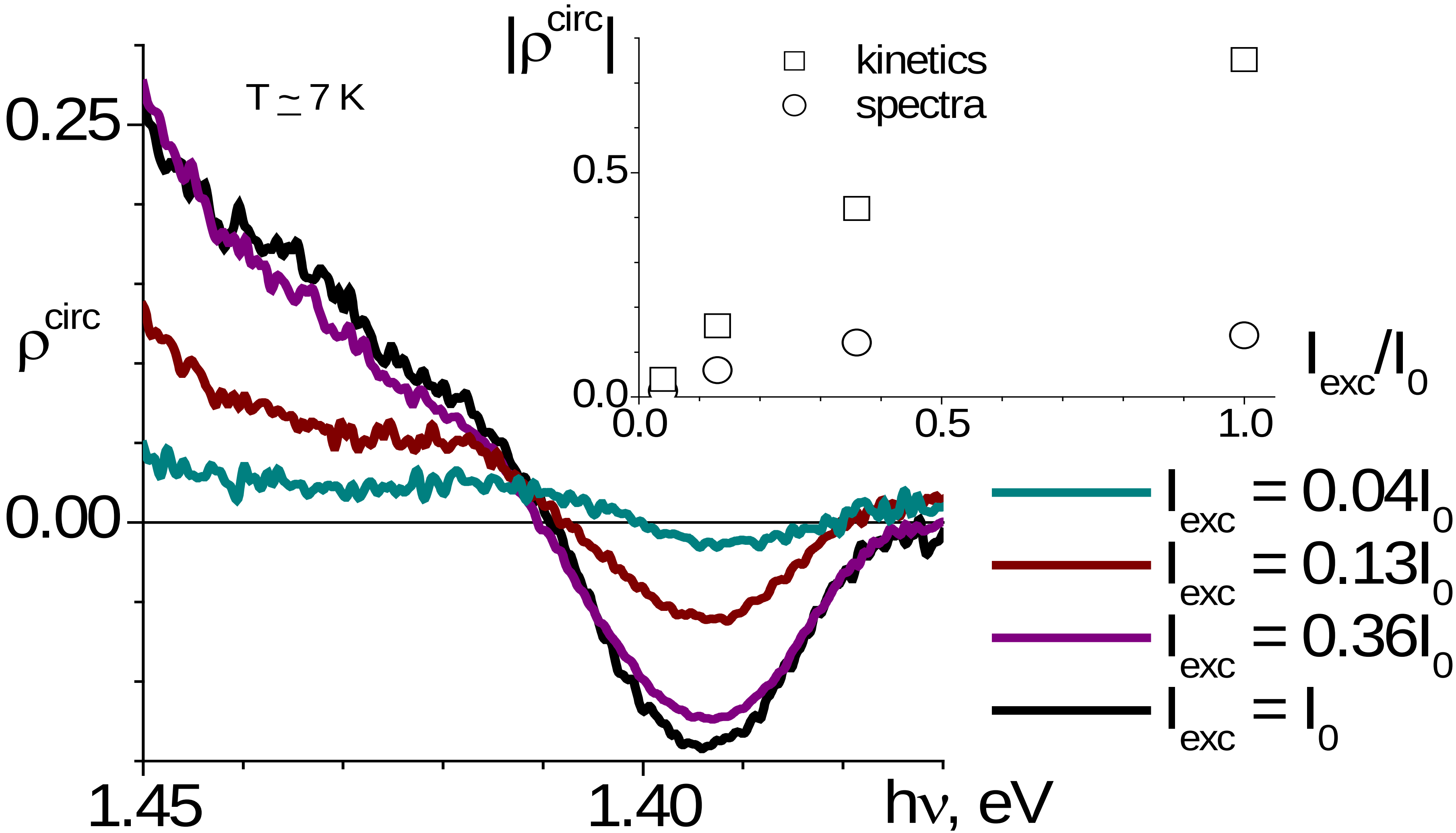
T ≃ 7 K

$h\nu_{exc}$=1.476 eV

Power dependence of NCP kinetics of InGaAs QDs PL



Power dependence of NCP of InGaAs QDs PL



Polarization properties of InGaAs and InP QD PL

I. Degree of circular polarization of PL:

a) is positive for

a1) the emission of neutral QDs and

a2) the emission from excited states of singly negative charged QDs

b) is negative for emission from ground states of singly negative charged QDs at the excitation to the excited states or to wetting layer

II. Absolute value of NCP degree increases with the **excitation power**

(up to 75-80 % for PL kinetics of InGaAs QDs)

Observation of NCP

Negative circular polarization at similar conditions has been found earlier in InP quantum dots (*Dzhioev et al., Phys. Solid State, **40**, 1587 (1998)*) but the model of its appearing proposed there does not explain experimental results mentioned above:

dependences on excitation energy and power

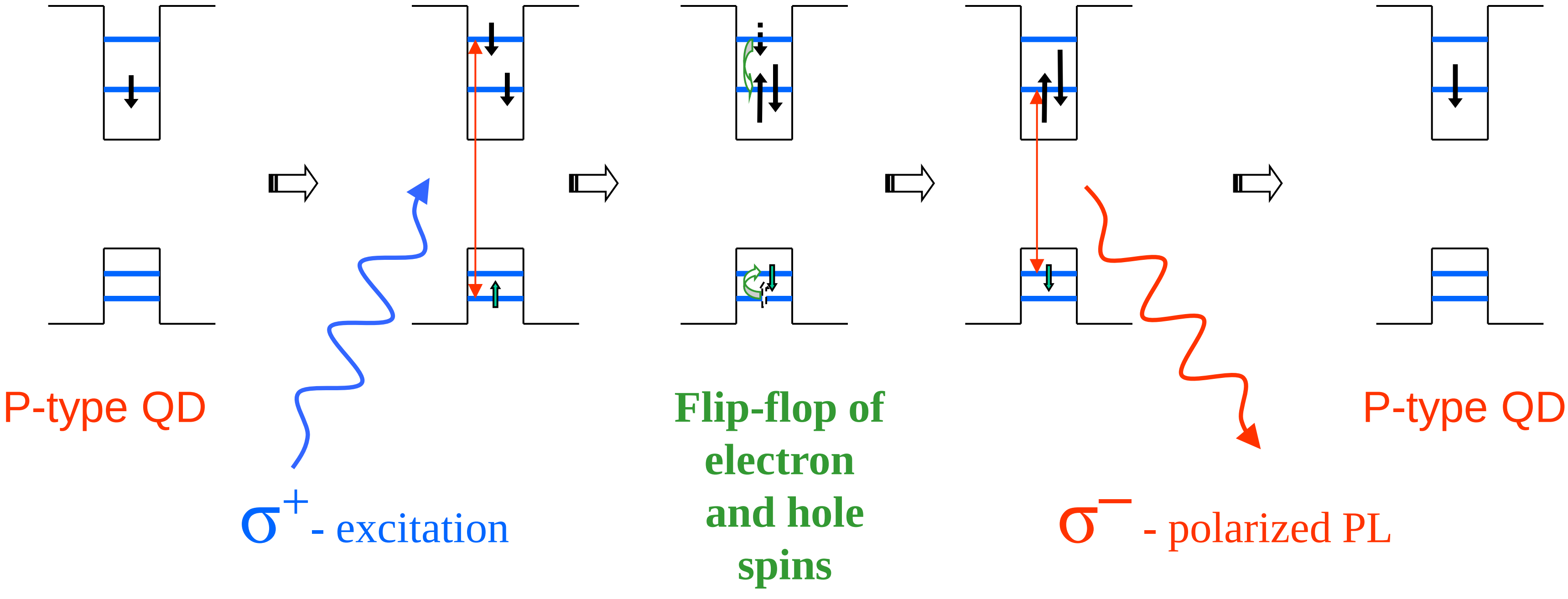
InAs quantum dots: *S.Cortez et al., Phys. Rev. Lett., **89**, 207401 (2002)*

GaAs quantum dots : *S.Bracker et al., PRL **94,** 047402 (2005)*

A. Shabaev et al., Phys. Rev. B 79, 035322 (2009)

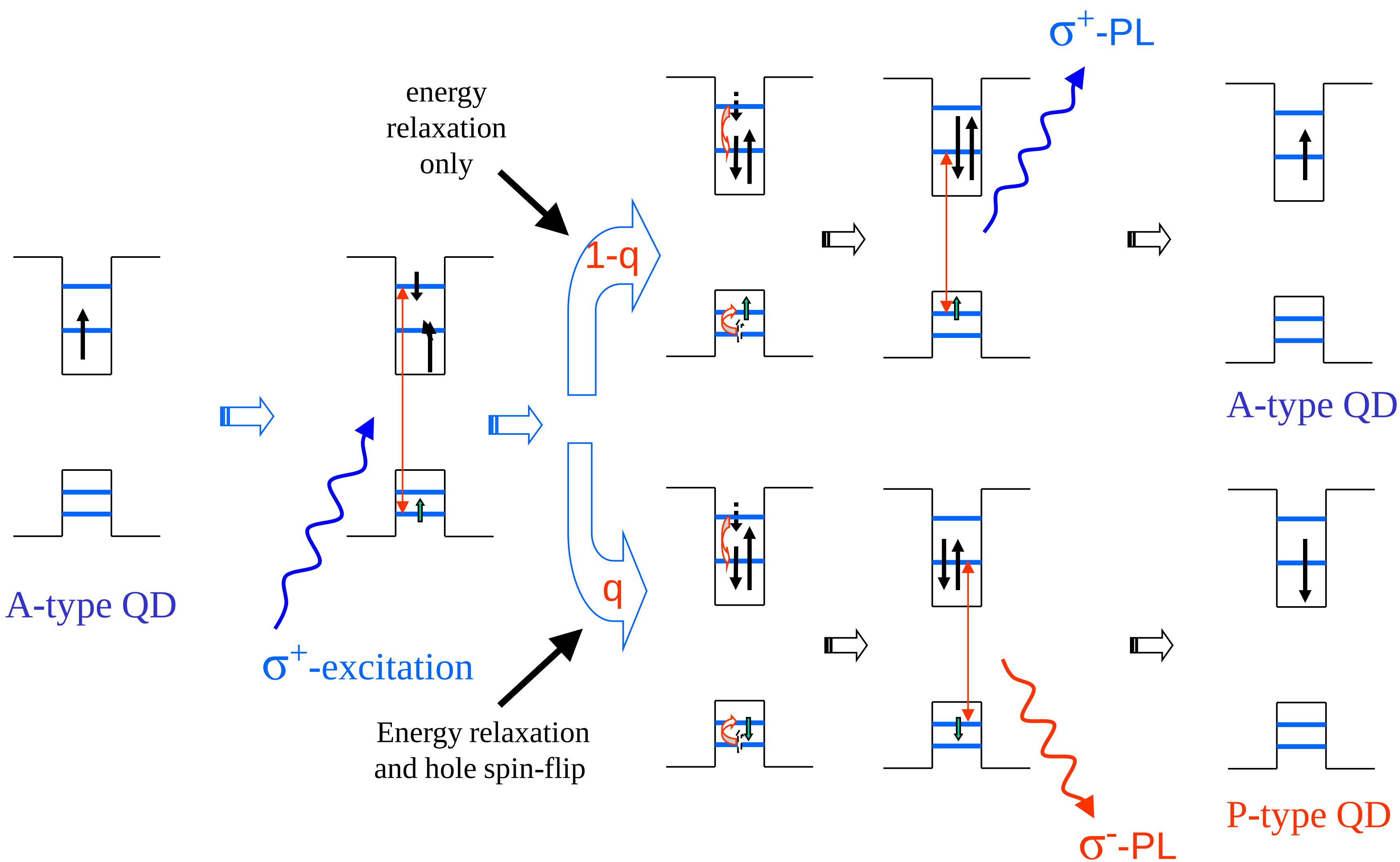
Model of NCP appearing

Our model is based on the mechanism proposed by K.V.Kavokin and published in *phys.stat.sol.(a)* ***195****, 592 (2003)*



• if resident electron spin is parallel to photogenerated electron spin (P-type QD), PL polarization is negative
• reversal of polarization sign is the result of flip-flop of spins of electron and hole due to their exchange interaction

Spin polarization mechanism



• If probability of spin-flip of photogenerated hole is equal q, then the probability of A-type QDs conversion to P-type QD is equal q too

Role of conservation of resident electron spin orientation

Conditions when NCP appears

1) Photogeneration of electron and hole at excited states of A-type and P-type QDs by circularly polarized light

2) After carrier relaxation to QD ground states and their recombination the relative number of P-type QDs **increases**

3) The rise of absolute value of NCP degree with excitation power means the accumulation of P-type QDs

4) Such accumulation is possible only at the conservation of resident electron spin orientation at least to the next pulse of exciting light

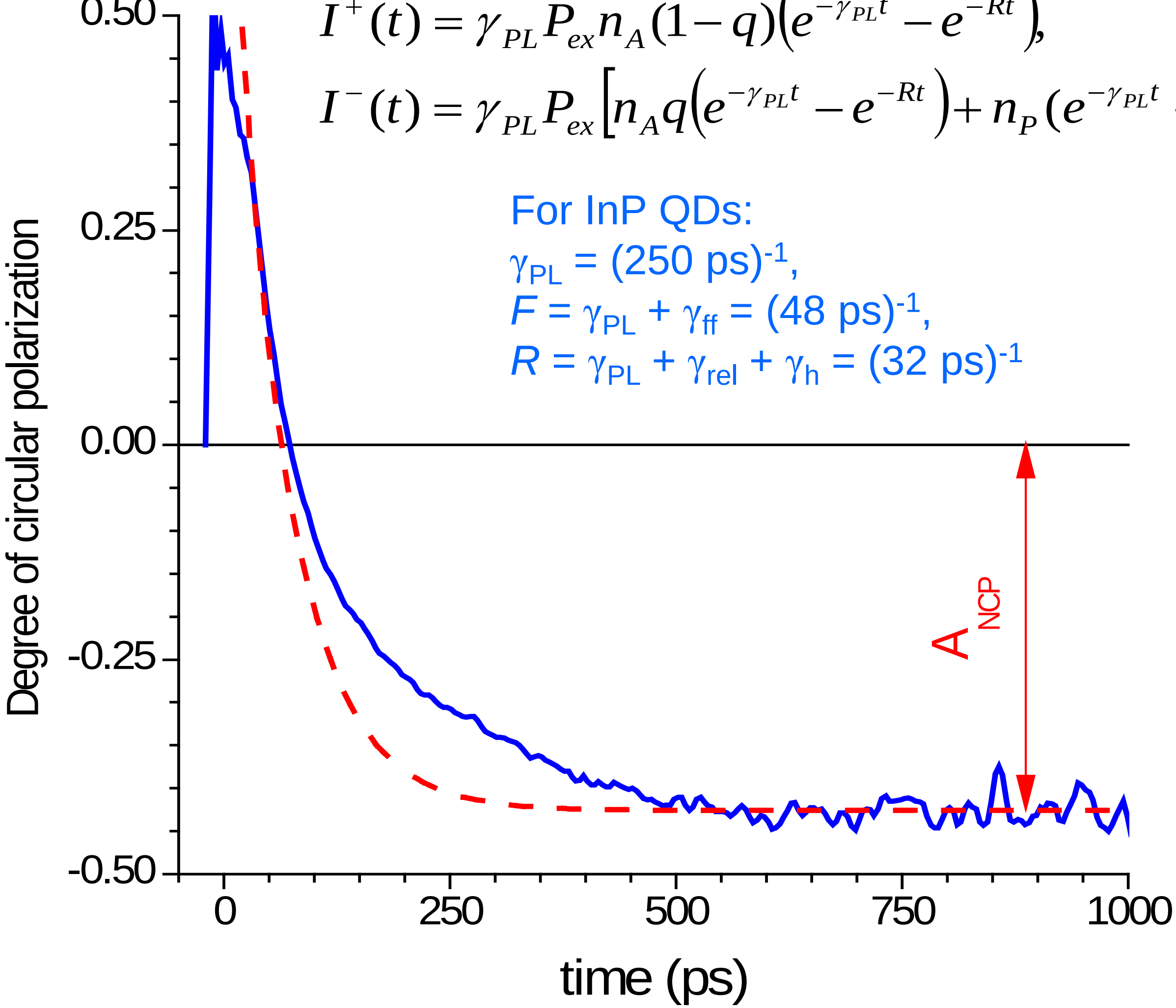
Use of NCP for evaluation of resident spin polarization

$$I^{+}(t) = \gamma_{PL} P_{ex} n_A (1-q)\left(e^{-\gamma_{PL} t} - e^{-Rt}\right),$$

$$I^{-}(t) = \gamma_{PL} P_{ex}\left[n_A q\left(e^{-\gamma_{PL} t} - e^{-Rt}\right) + n_P\left(e^{-\gamma_{PL} t} - e^{-Ft}\right)\right].$$

$$\rho_c(t) = \frac{I^{+}(t) - I^{-}(t)}{I^{+}(t) + I^{-}(t)}$$

For InP QDs:
γ_{PL} = (250 ps)$^{-1}$,
$F = \gamma_{PL} + \gamma_{ff}$ = (48 ps)$^{-1}$,
$R = \gamma_{PL} + \gamma_{rel} + \gamma_h$ = (32 ps)$^{-1}$



$A_{NCP} = n_A(1-2q) - n_P$
n_A – part of A-type QDs;
n_P – part of P-type QDs;
($n_A + n_P = 1$).

In our experiments:
q ~ 0.05 – 0.10
At low temperatures
and excitation powers
A_{NCP} ~ - ($n_P - n_A$) = -S

I.V.Ignatiev et al., Opt. Spec. ***106****, 375–387 (2009)*

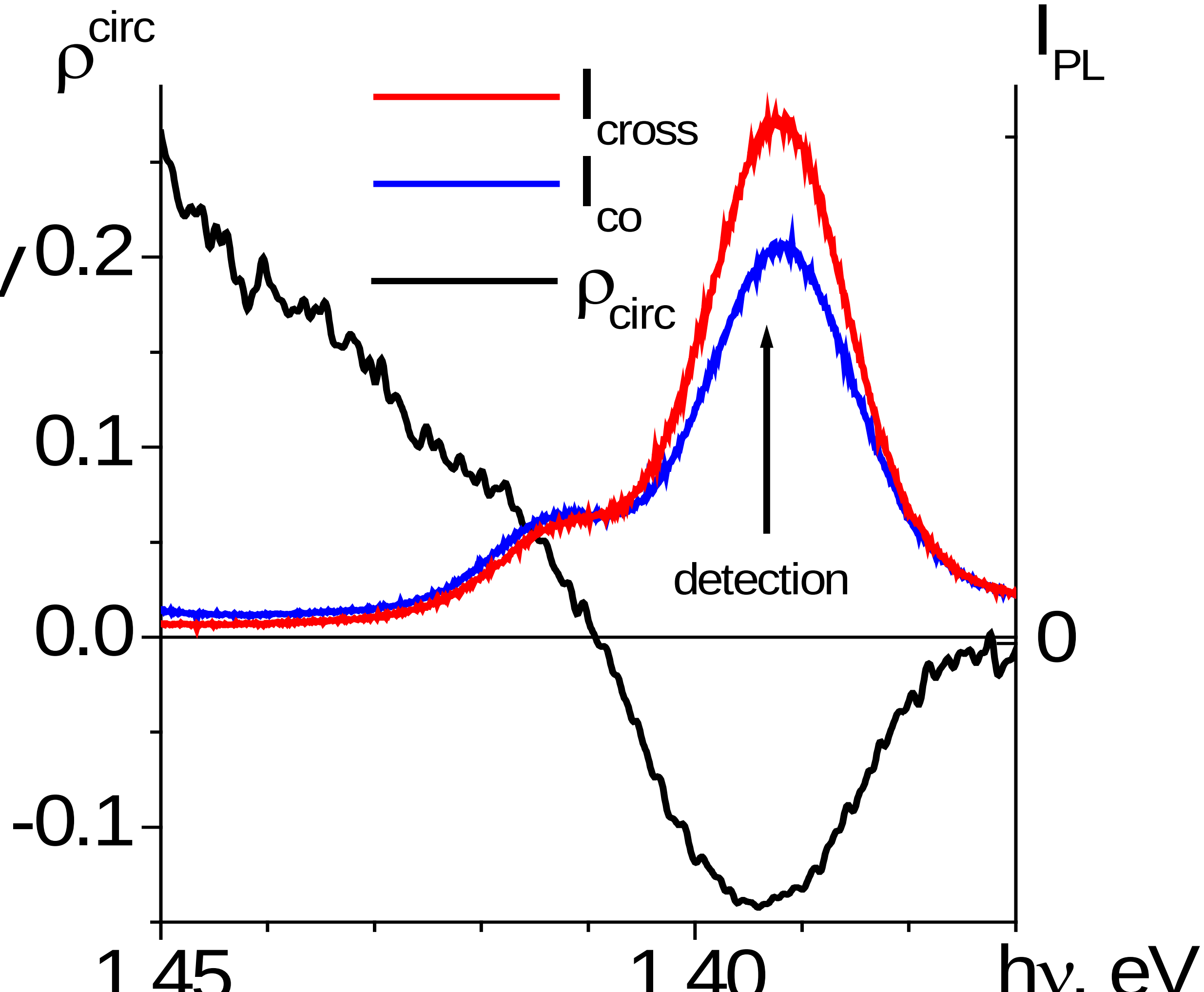
Negative Circular Polarization (NCP) of negative singly-charged InGaAs QDs PL

is a **measure of spin polarization of resident electron** in QD

$$\rho = \frac{I^{\sigma+} - I^{\sigma-}}{I^{\sigma+} + I^{\sigma-}}$$

$T \simeq 2$ K

$h\nu_{exc}$=1.476 eV



I.V.Ignatiev et al.,
Opt. Spec. ***106****, 375–387 (2009)*

$$\langle S_Z \rangle \propto \rho_{max} \equiv NCP$$

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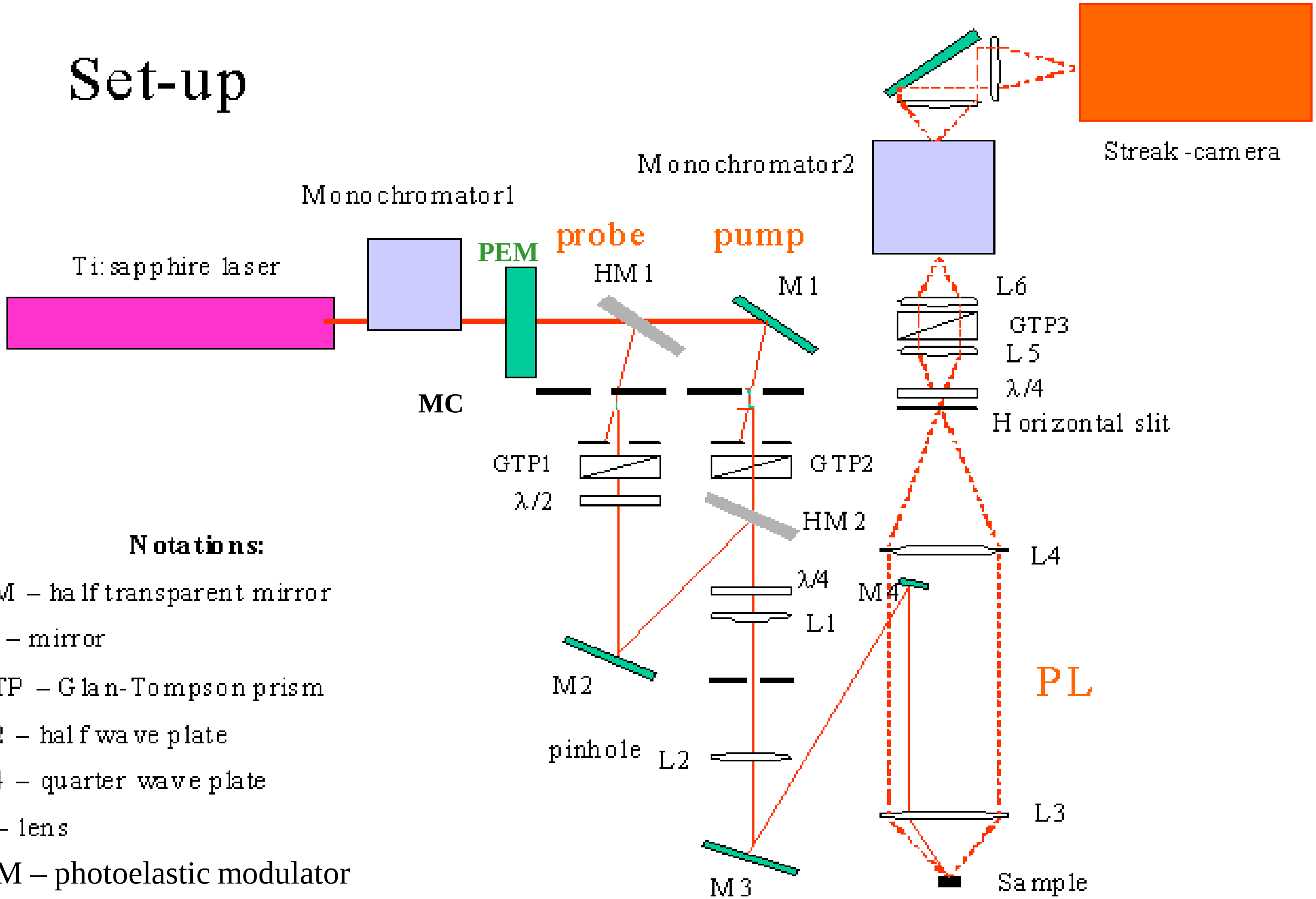
Two-beam set-up

Set-up



Notations:

HM – half transparent mirror

M – mirror

GTP – Glan-Tompson prism

λ/2 – half wave plate

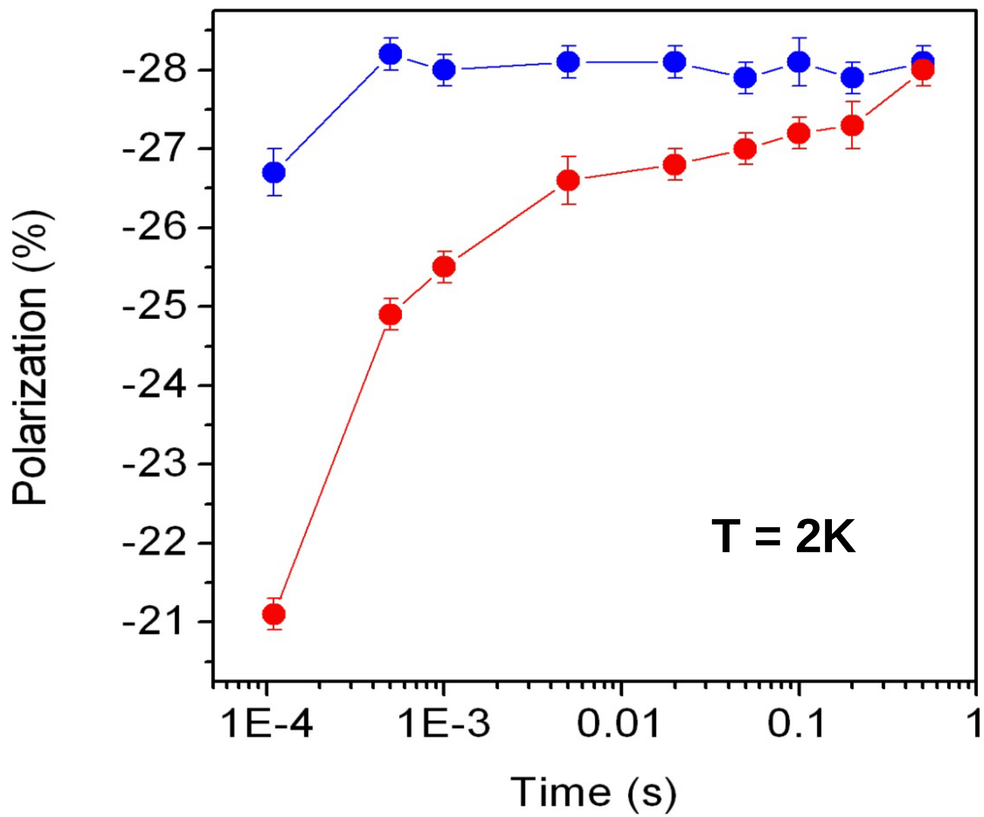
λ/4 – quarter wave plate

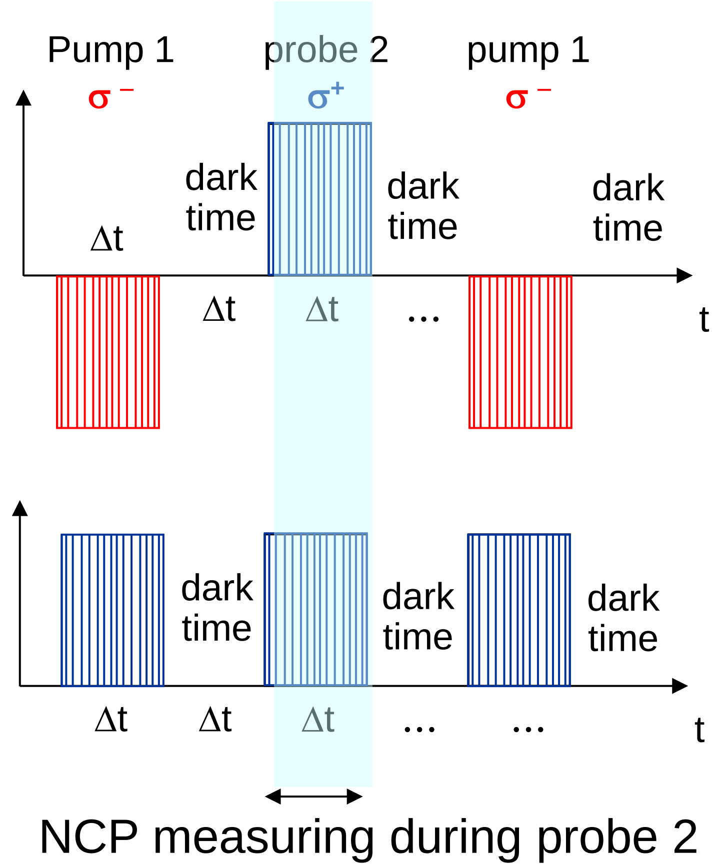
L – lens

PEM – photoelastic modulator

MC – alternate chopper

Spin memory of InGaAs QDS





- Long-time (up to ~10^2 ms) spin memory of resident electrons at the absence of external magnetic field

- **What is the role of hyperfine interaction with nuclear spins in QDs?**

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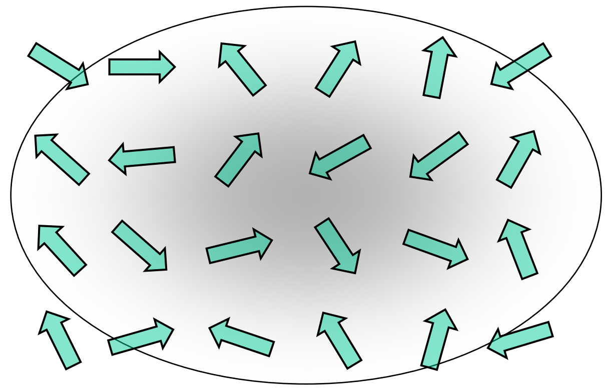
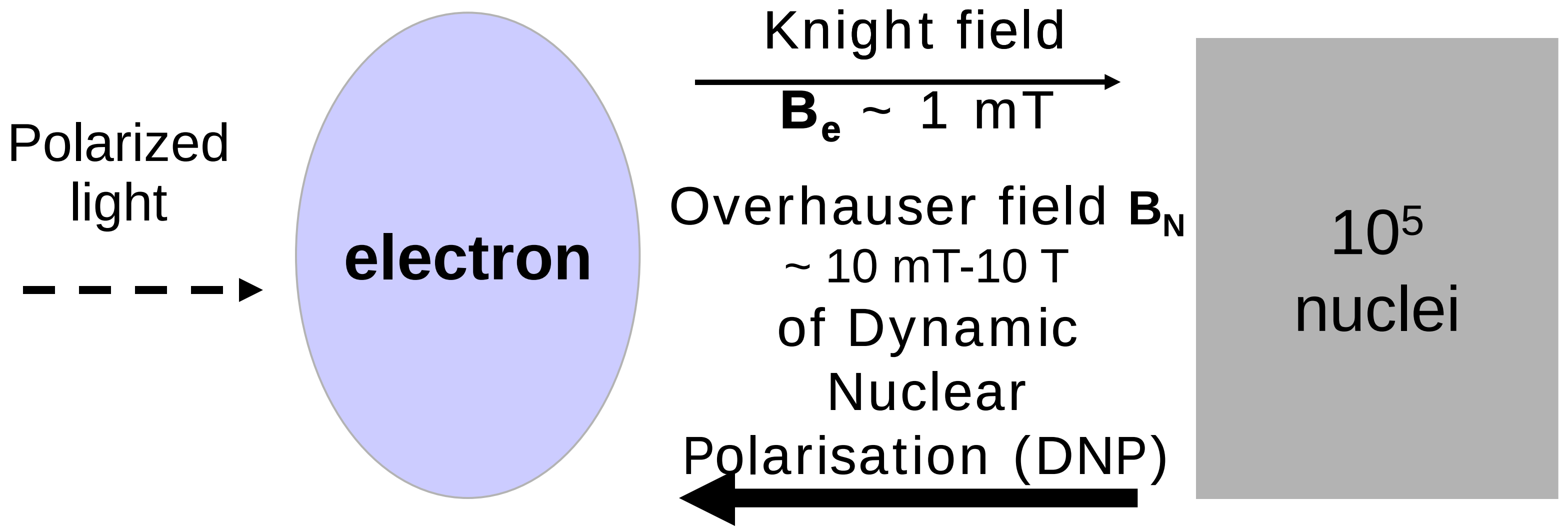
Hyperfine interaction of electron and nuclear spins

~10^5 nuclei



Electrons have s-type wave function in ground state



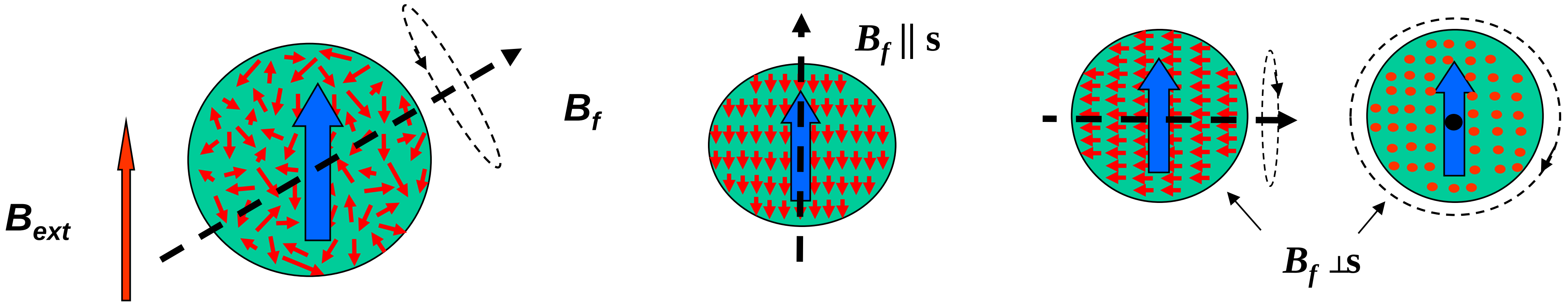
DNP is only one of the possible mechanisms ruling the Nuclear Spin Polarization (NSP)

Fluctuations ΔI_N of total nuclear spin I_N in QDs - - other type of possible NSP

[Theory: *Merkulov et al. PRB, (2002)]*.

$$\Delta I_N / I_N \propto \sqrt{n} \ (n \sim 10^5 \text{ in QDs under study})$$

Fluctuations ΔI_N influence on the electron spin as effective magnetic field B_f with incidental value and direction through the QDs ensemble.



Fluctuations of ΔI_N influence on the electron spin in KT, when external field $B_{ext} < B_f$

Theoretical estimations of periods:

electron spin precession in the field of "freezed" nuclear spins fluctuation ~ 1 нс

nuclear spin precession in the Knight field created by the electron spin ~ 1 мкс

nuclear spin relaxation at their dipole-dipole interaction ~ 100 мкс

Measurement of NCP of QDs PL as an instrument to study hyperfine interaction in QDs

- Electron spin optically oriented

by circularly polarized light polarises nuclear spins.

- The orientation of latter ones (NSP)

may support or destroy electron spin polarisation.

- The NSP may be researched *via* its influence

on electron spin polarisation

studied by measurement of NCP of QDs PL

Two configurations of external magnetic field

Faraday configuration

Magnetic field B_{ext} is

parallel to optical axis (Z)

(and to electron spin oriented by circularly polarized light;

to direction of QD structure growth;

to *grad*(*F*) of electrical field;

to DNP (NSP$^{||}$))

Voigt configuration

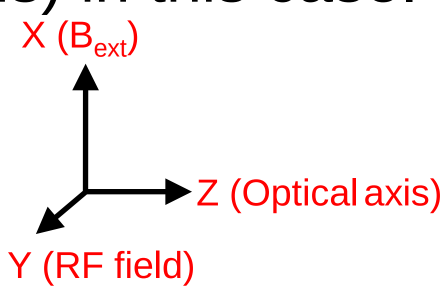
Magnetic field B_{ext} (X-axis) is *perpendicular* to optical axis (Z) (and to electron spin oriented by circularly polarized light;

to direction of QD structure growth;

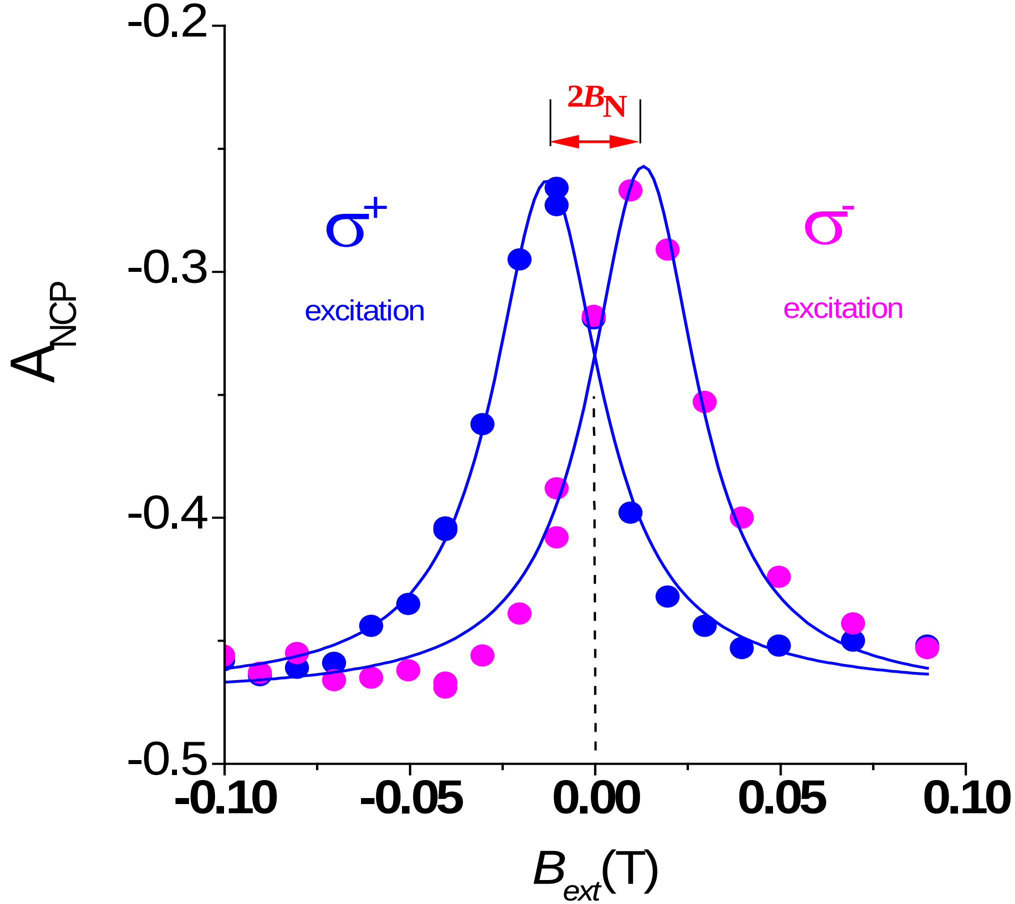
to *grad*(*F*) of electrical field;

to NSP$^{\perp}$)

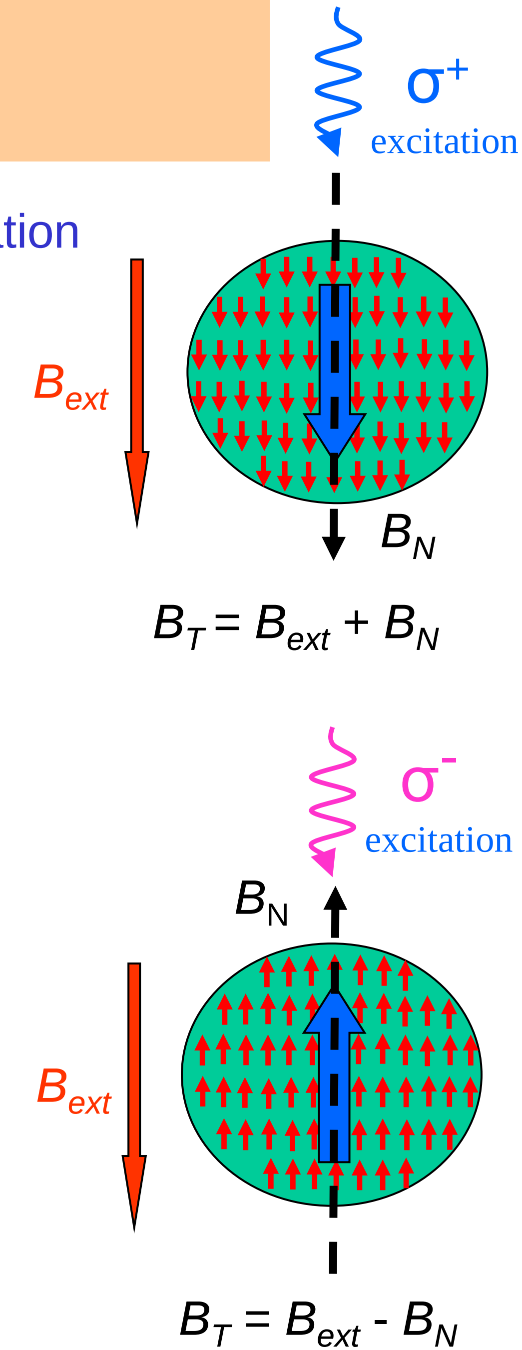
The NSP$^{||}$ component is parallel to B_{ext} (X-axis) in this case.



Influence of two components of NSP on resident electron spin polarization



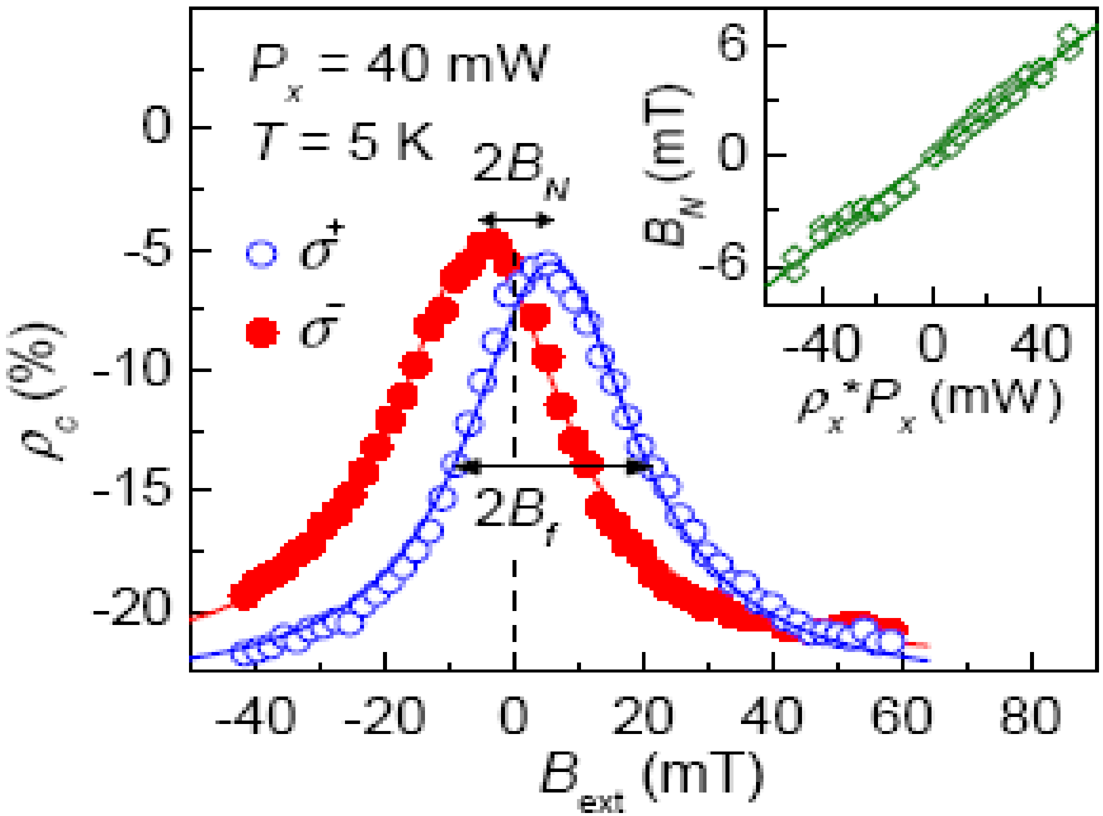
Faraday configuration



$< \boldsymbol{B}_f > = 0$

Influence of two components of NSP on resident electron spin polarization

(the helicity of PL polarization is marked here relatively to exciting light helicity)



$(<B_f^2>)^{1/2} \approx 15$ mT is practically independent on excitation power.

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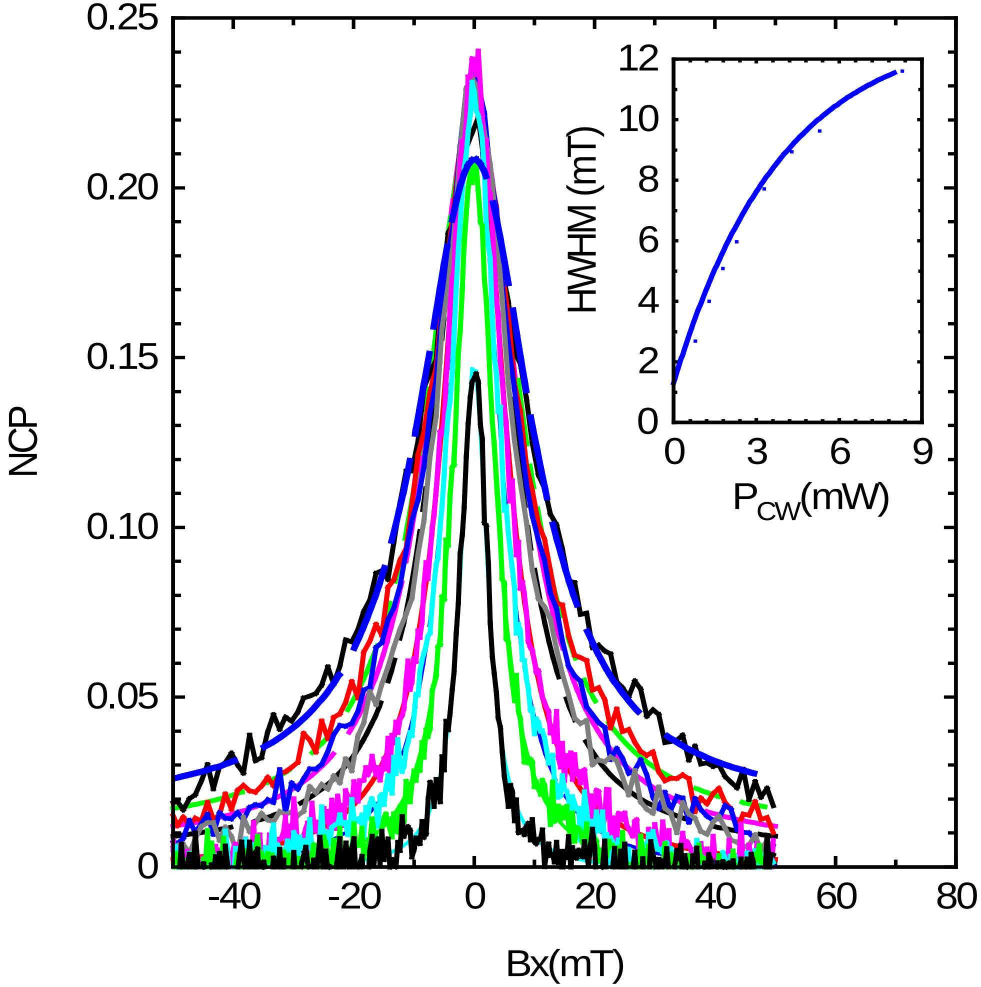
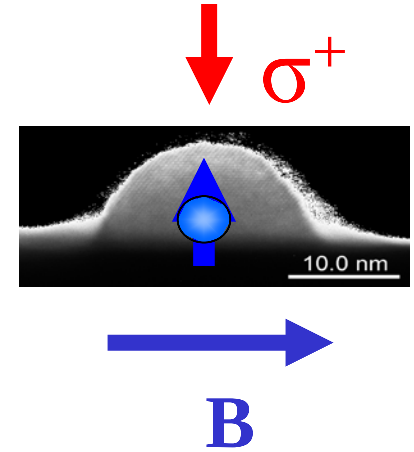
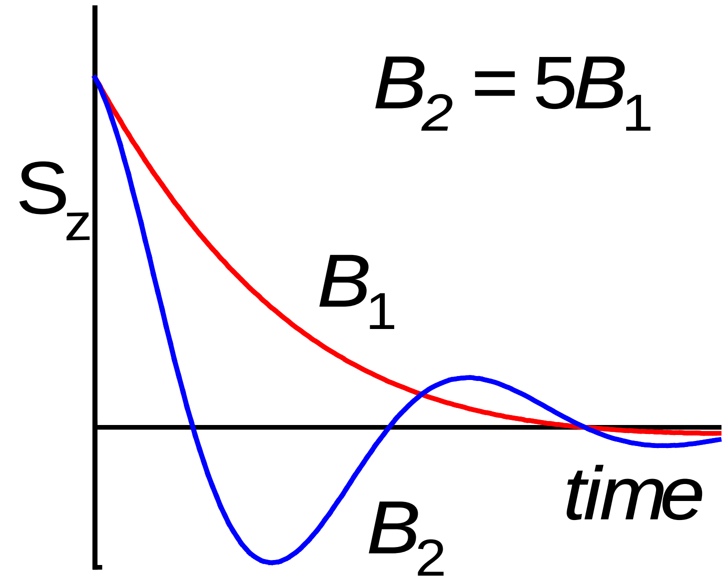
«Usual» Hanle effect (Voigt configuration)



Hanle curves measured at various excitation power at suppression of nuclear polarisation

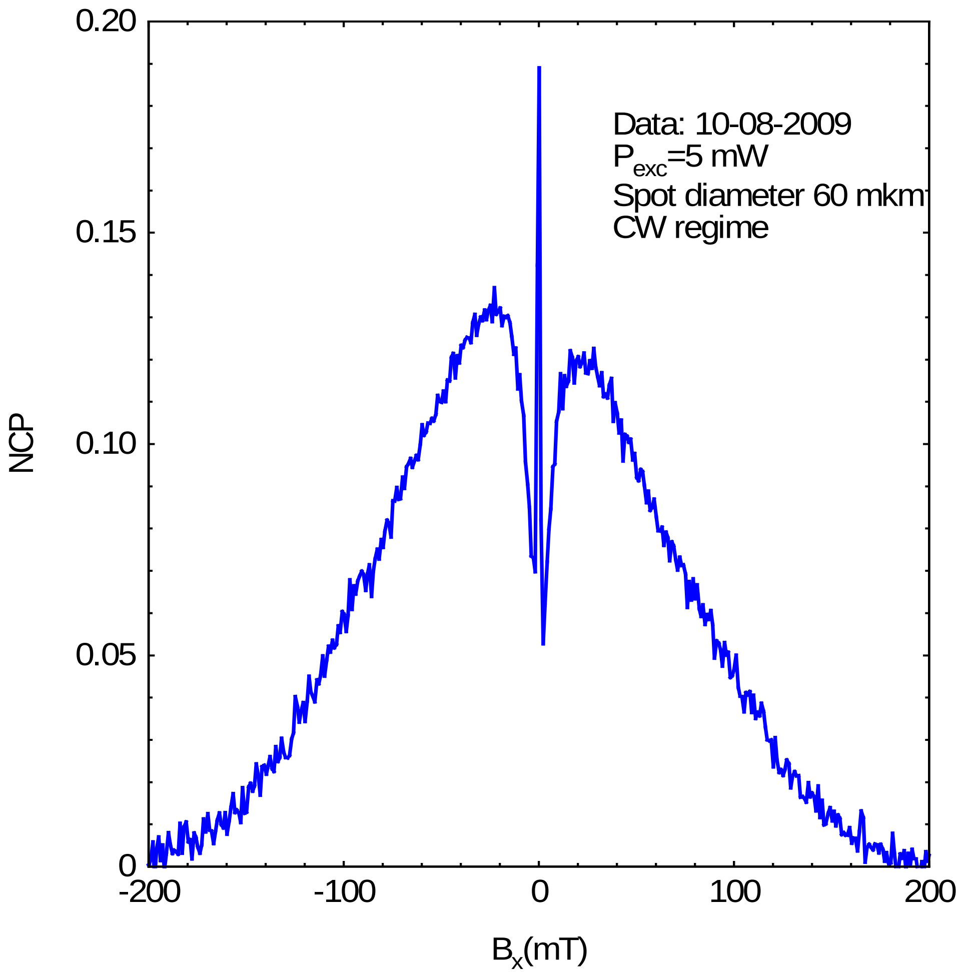




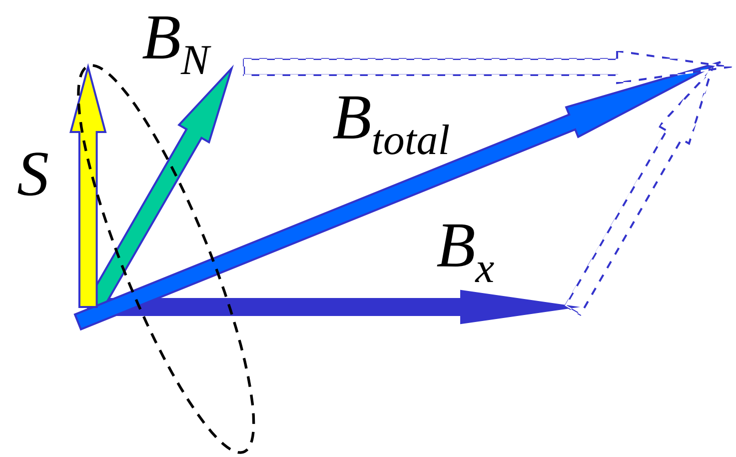
$S = S_0/(1+(B/B_{1/2})^2)$,

$B_{1/2} = \hbar/(|g_e|\mu_B T_2)$.

Hanle effect in NCP

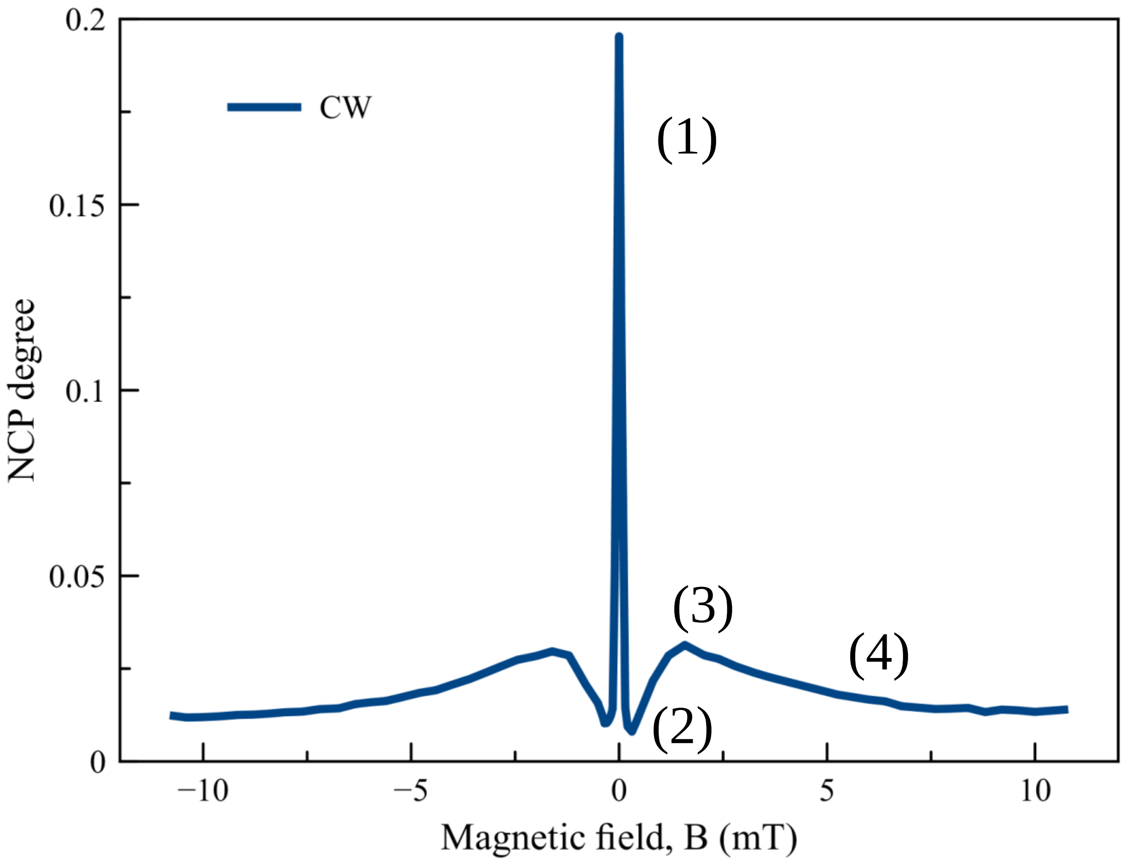


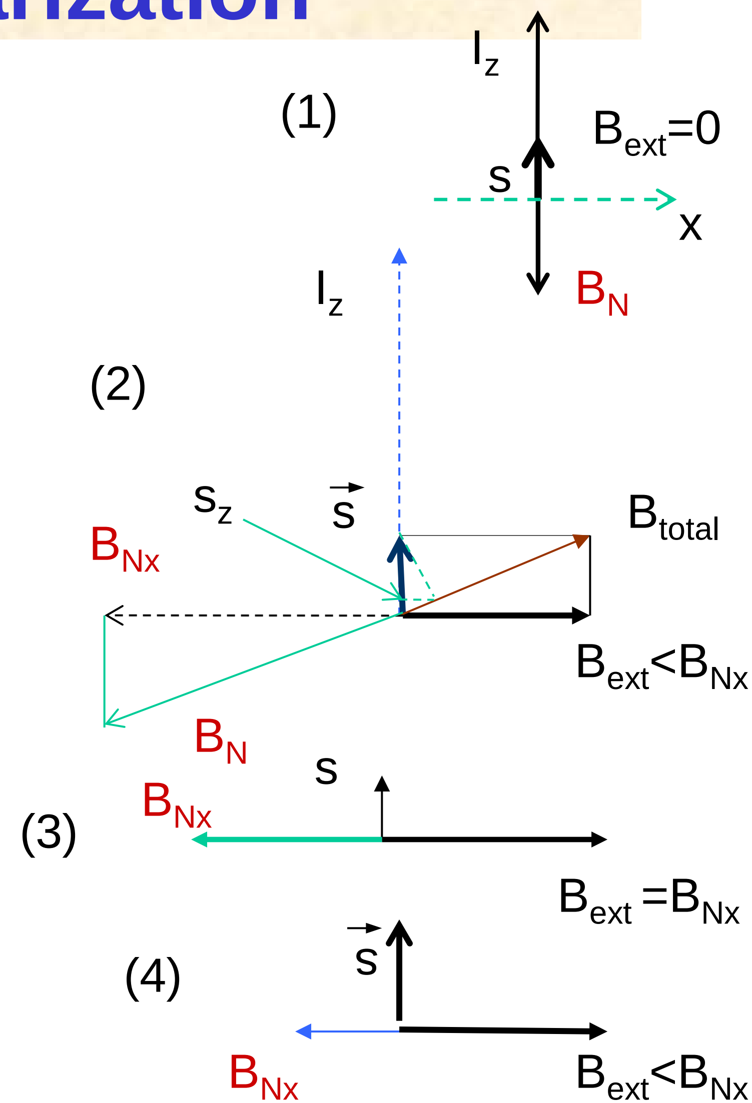
W-like dependence in small magnetic fields is explained by Paget et al., PRB (1977) as an amplifying the external magnetic field by the nuclear field.



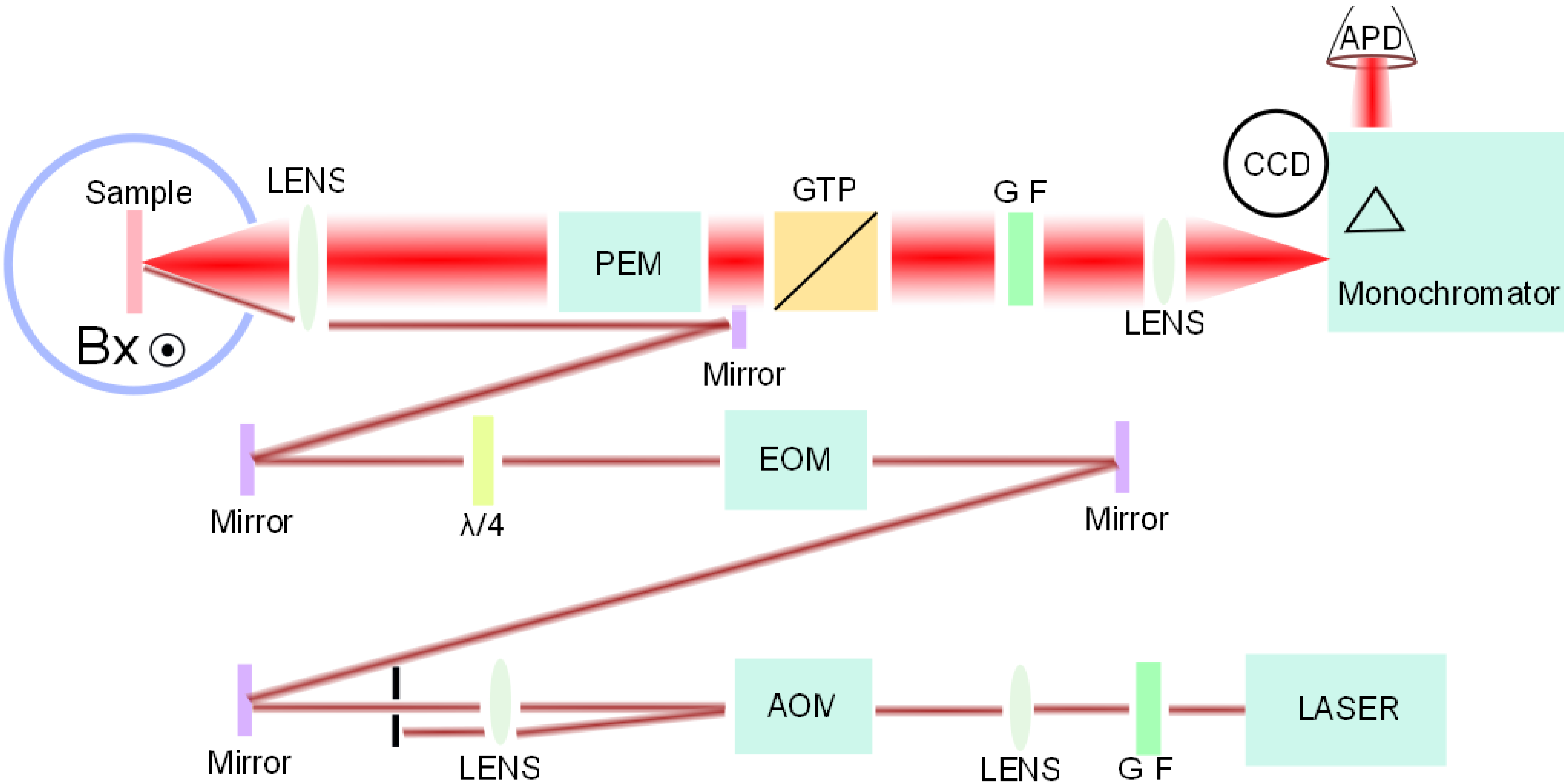
- Tilting the nuclear field B_N
- Positive feedback for tilting
- Reducing the pumping rate of B_N with tilting

Hanle effect influenced by nuclear spin polarization

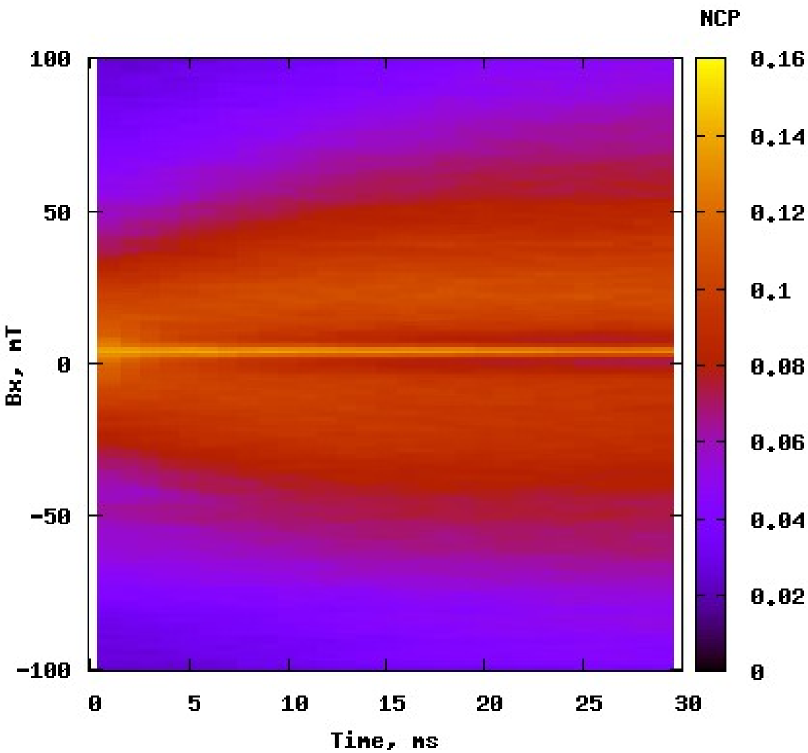




Set-up for the time-resolved Hanle effect study



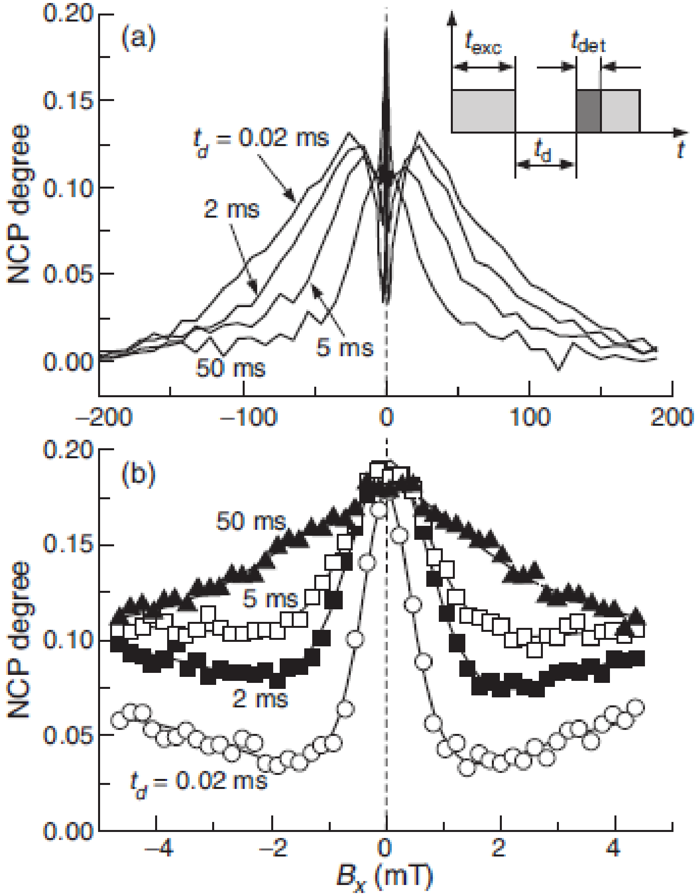
Rise of nuclear polarisation measured by time-resolved Hanle effect



Time evolution of NCP degree measured at external field strength equal to 2 mT and to 50 mT allows us to analyze **rise**-times of nuclear polarization components parallel ($NSP^{||}$) and perpendicular ($NSP^{\perp}$) to external magnetic field direction.

$NSP^{||}$ and $NSP^{\perp}$ are $\perp$ and || to the optically oriented electron spin.

Relaxation of nuclear polarisation measured by time-resolved Hanle effect



Time evolution of NCP degree measured at external field strength equal to 2 mT and to 50 mT allows us to analyze times of **relaxation** of nuclear polarization components parallel (NSP$^{||}$) and perpendicular (NSP$^{\perp}$) to external magnetic field direction.

NSP$^{||}$ and NSP$^{\perp}$ are $\perp$ and || to the optically oriented electron spin.

Nuclear Spin Polarization (NSP) influence on electron spin polarization – two opposite results:

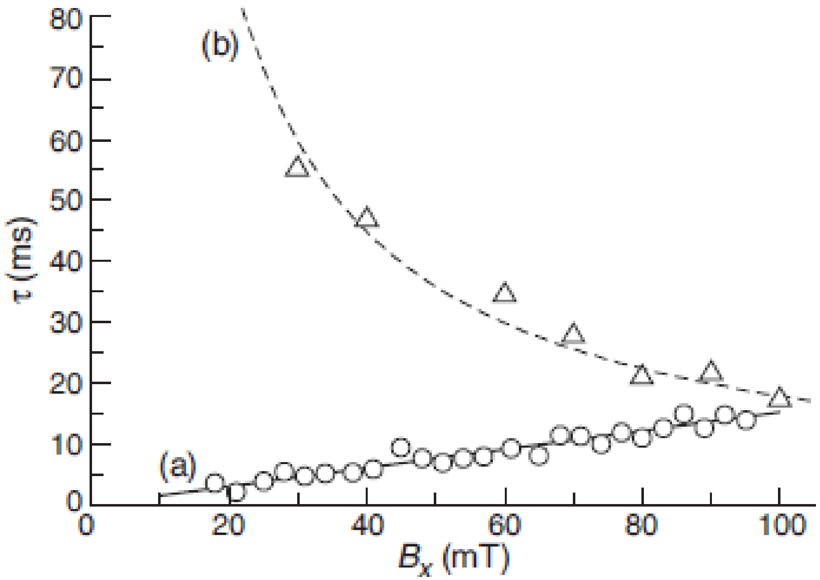
W-range of Hanle curves (B_{ext} < 50 mT) –
- the electron spin polarization is *destroyed* by NSP =>
=> nuclear spins are polarized parallel to B_{ext}
and are *perpendicular* to the electron spin so.

Wings of Hanle curves (B_{ext} > 50 mT) –
- the electron spin polarization is *stabilized* by NSP =>
=> nuclear spin polarization has the component
parallel to the electron spin (and perpendicular to B_{ext})

It has allowed us to define firstly
the time behaviour of these two components of NSP separately.

Time behavior of two components of dynamic nuclear polarization

Times of rise and of relaxation of nuclear polarization component parallel to external magnetic field direction are nearly of 5 ms and independent from external magnetic field strength



Field dependences of times
a) of rise
b) of relaxation
of nuclear polarization component perpendicular to external magnetic field direction

Such behavior of nuclear polarization could not be explained in the frame of existing phenomenological models and demands to develop new theoretical approach

Conclusion

Spin carrier dynamics in QDs is dependent
from hyperfine interaction
between electron and nuclear spins.

At the analysis of dynamics of hyperfine interaction
it is necessary to concern dynamics not only of parallel
but also of perpendicular component
of Nuclear Spin Polarisation.

It is a challenge for developing
of new theoretical approaches.

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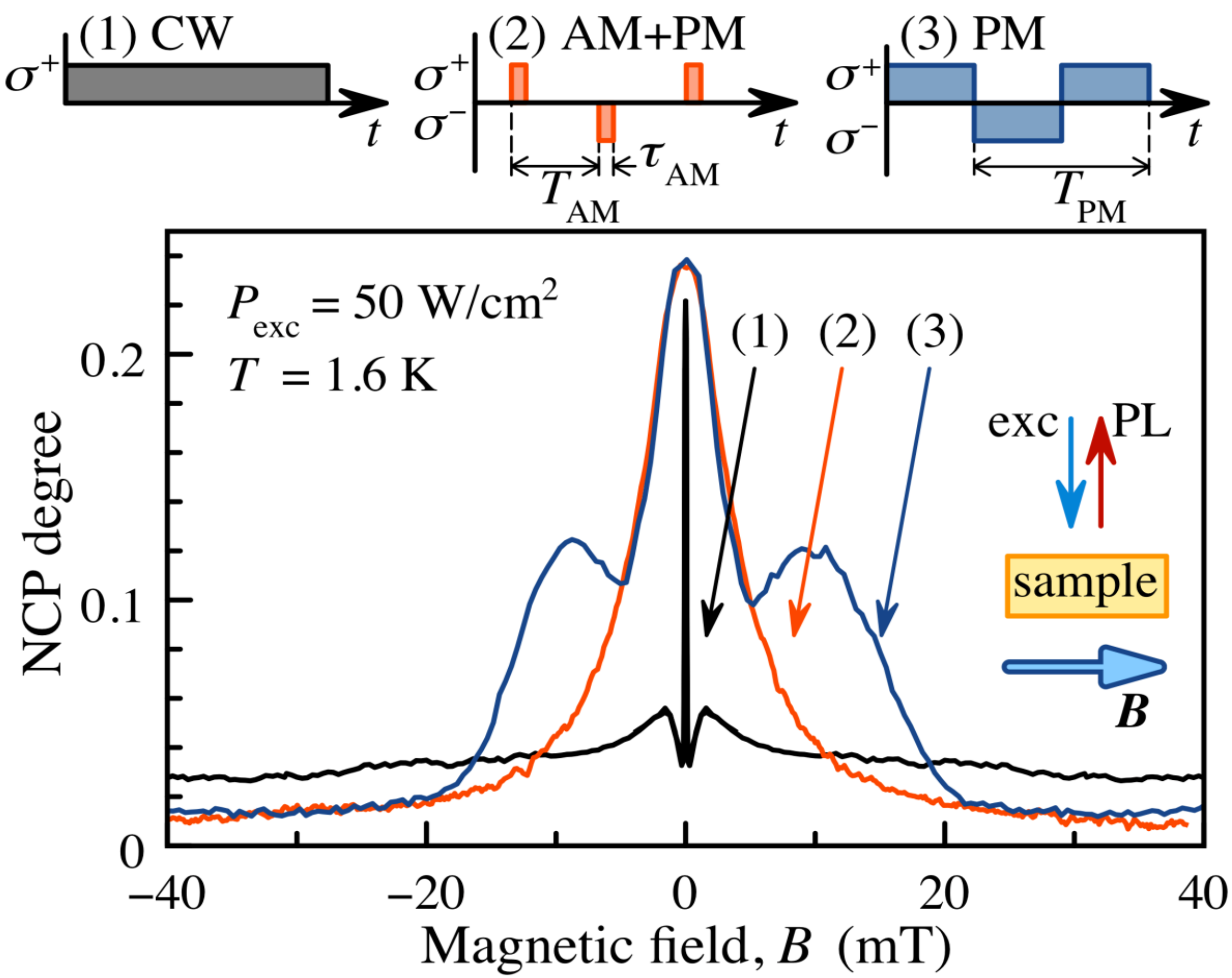
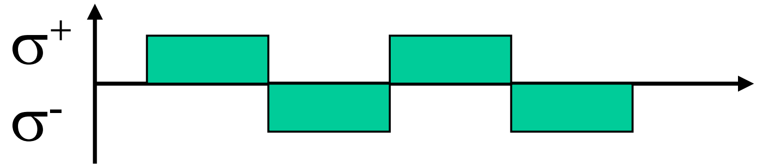
Hanle curves at the modulation of exciting light polarization

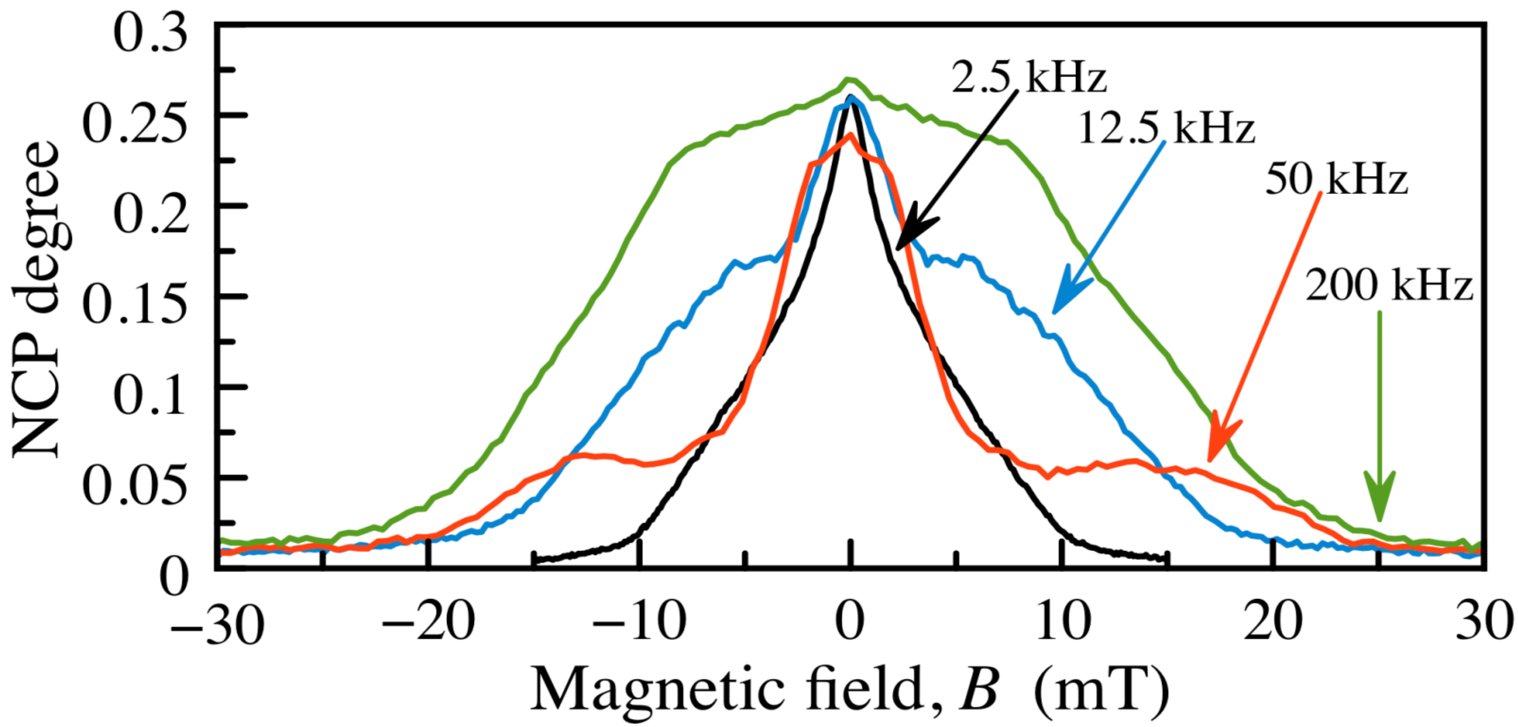


Moreover weak magnetic field *along* the direction of excitation has been additionally applied:

fresh results are presented at *M. S. Kuznetsova et al., http://arxiv.org/abs/1303.4192*

Resonances





Resonances shift to the larger field strength at the rise of modulation frequency of exciting light polarisation!

Measurements at modulation of optical excitation and at application of radiofrequency (RF)

- CW optical orientation of the electron spins influences on the nuclear spin orientation too

- External magnetic field also influences on both the electron and nuclear spins

- Exciting light with modulated polarization or application of radiofrequency field (RF) influence **on nuclear spins only**

ODNMR experiments in heterostructures with QDs

The ODNMR has been observed by single QD spectroscopy
of unstrained GaAs/AlGaAs heterostructures
(D.Gammon *et al*., Science 277, 85 (1997), M.N.Makhonin *et al*., arXiv:1002.0523v2 (unpublished).)

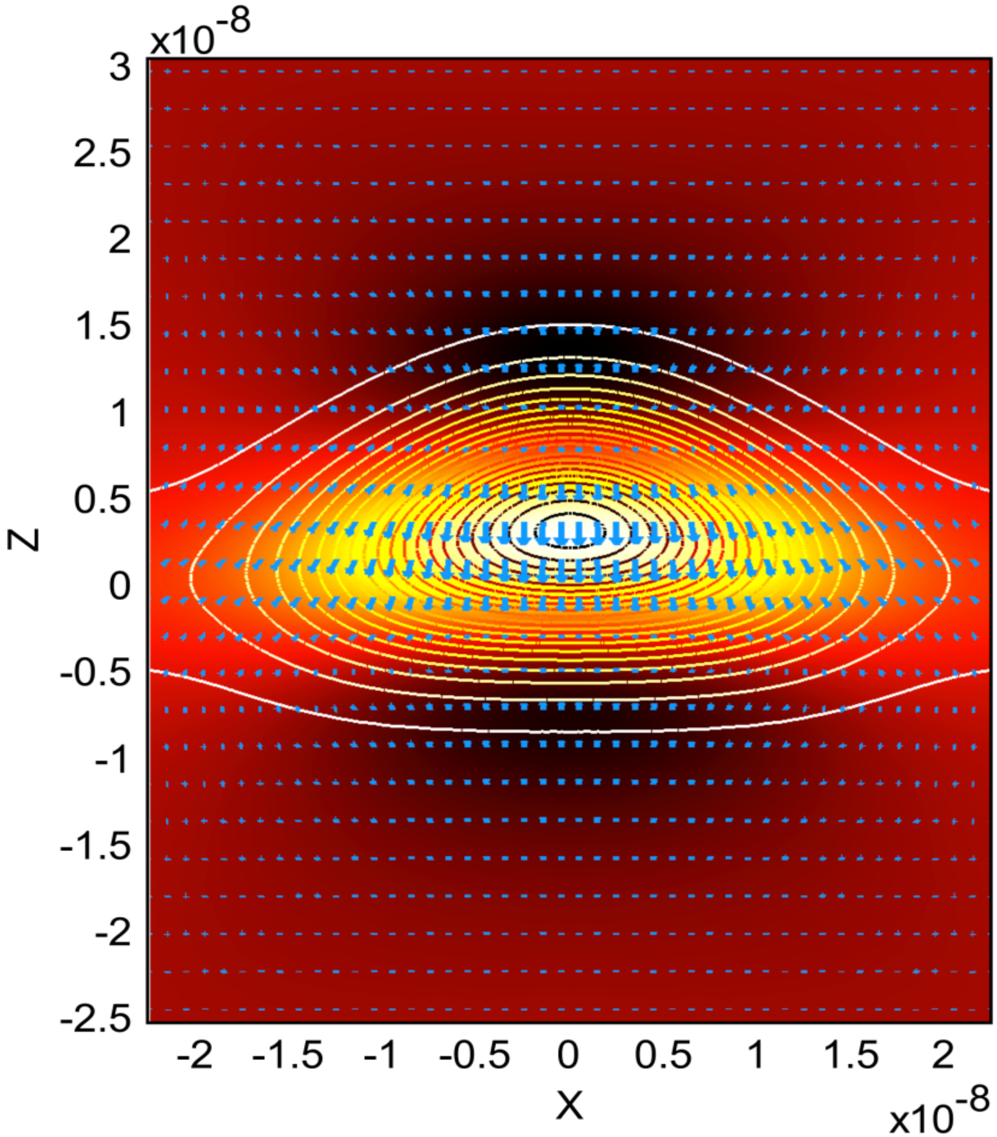
The heterostructure with InGaAs/GaAs QDs under study:
1) has much more (~10^{10} cm^{-2}) density of QDs
2) is strained due to the crystal lattice mismatch between InGaAs and GaAs

The both properties are more real for the future applications but
it is impossible to study single QD at such high density.

In result we have studied the <u>ensemble</u> of <u>strained</u> InGaAs/GaAs QDs
where effect of the **<u>inhomogeneous broadening</u>** is considerable

The disordered strain leads to the gradient ∇F of electric field
who splits the nuclear spin states into Kramers doublets $|\pm m/2>$
(nuclear quadrupole splitting)

Influence of uniaxial deformation of QD

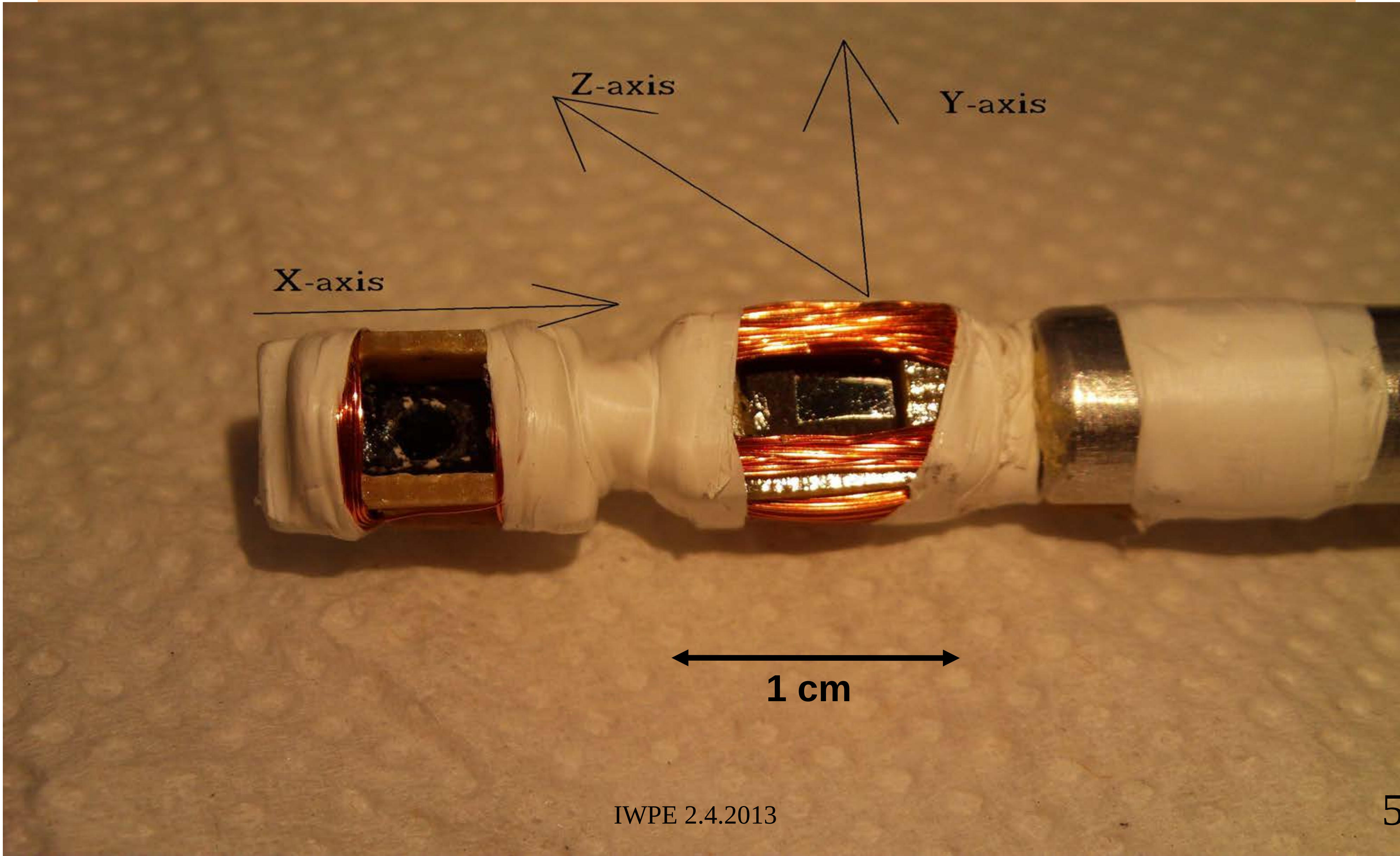


Direction of main axis
of deformation tensor
is shown by arrows.

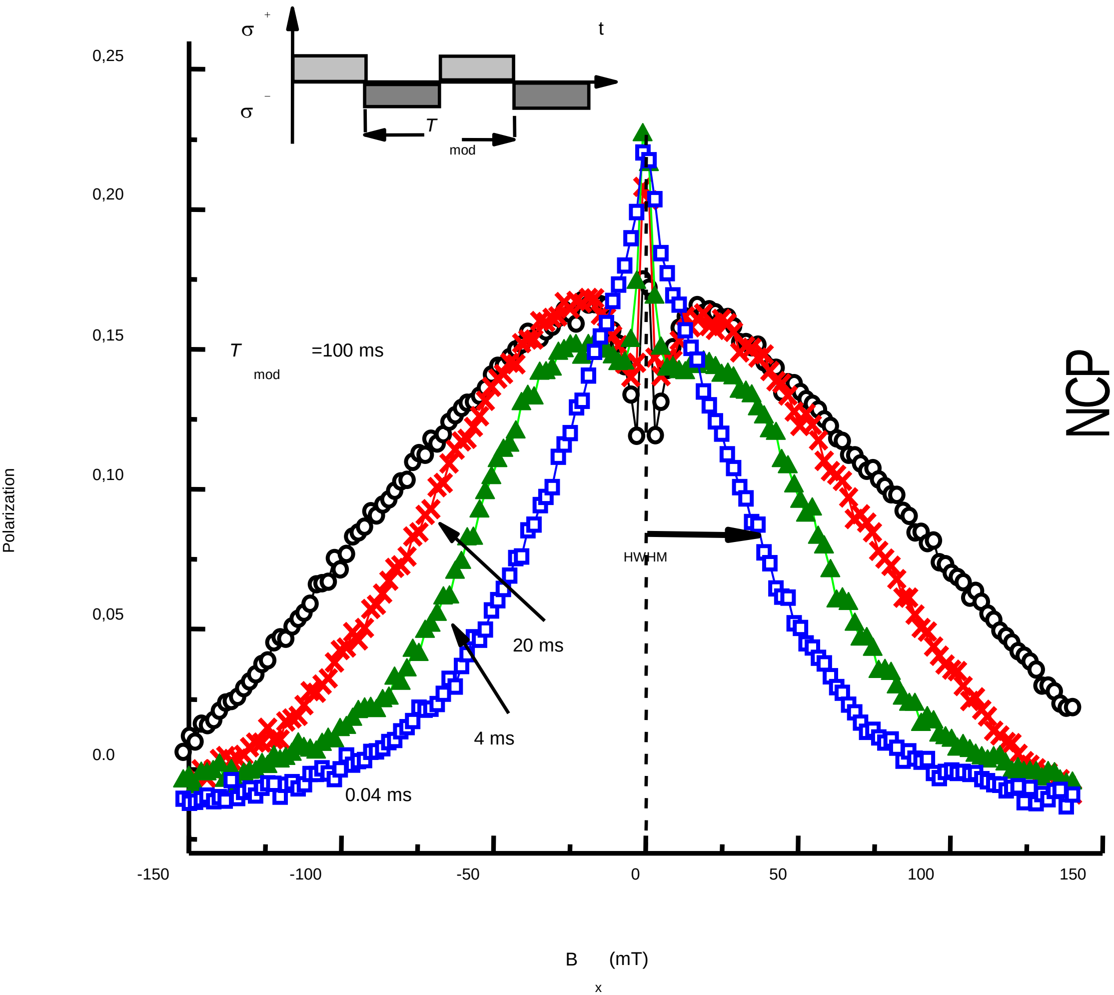
Points wth equal concentration
of In atoms are shown by lines.

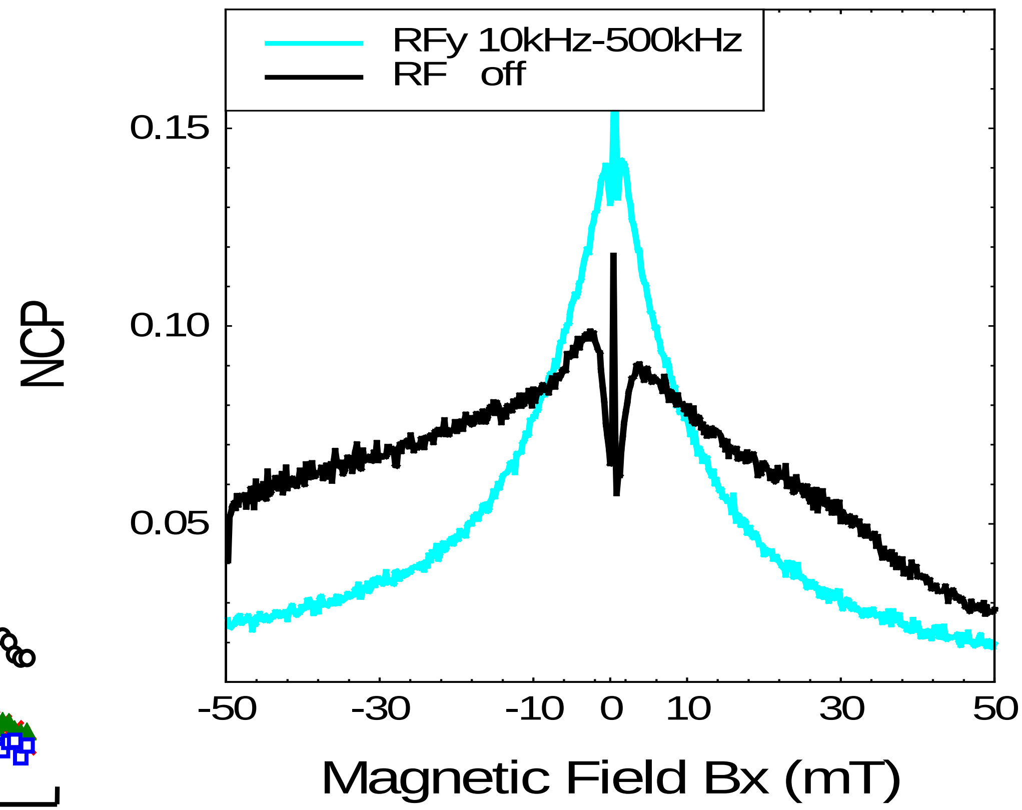
ε_{zz}**(max) = 0.0117**

The sample holder with RF coils



Overhauser field destroying





⇒ by fast modulation of excitation polarization

⇒ by radio frequency

(Field B_x is swept very slowly (7 s/point))

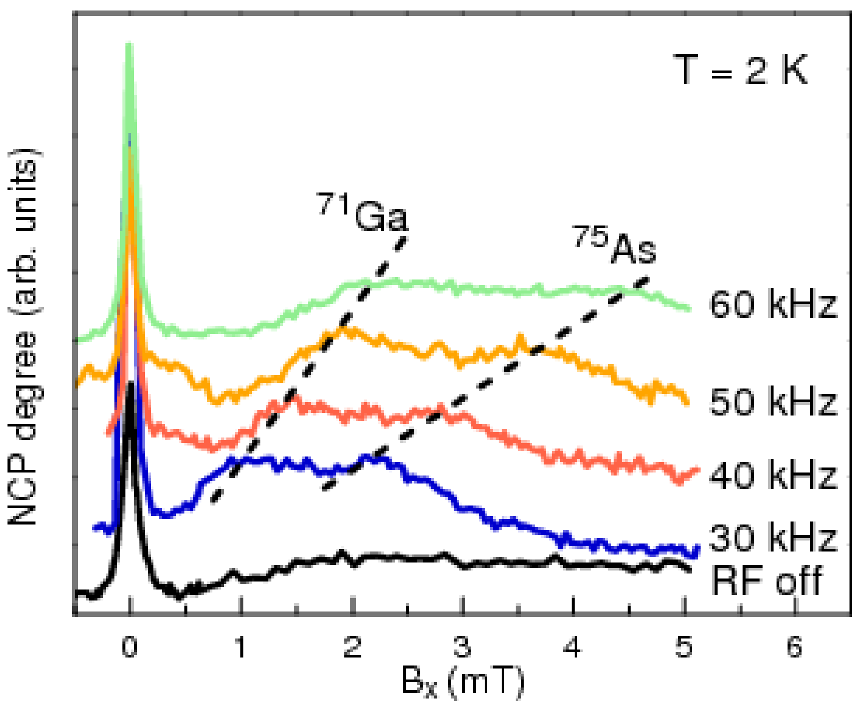
RF influence on Nuclear Spin Polarization (NSP)

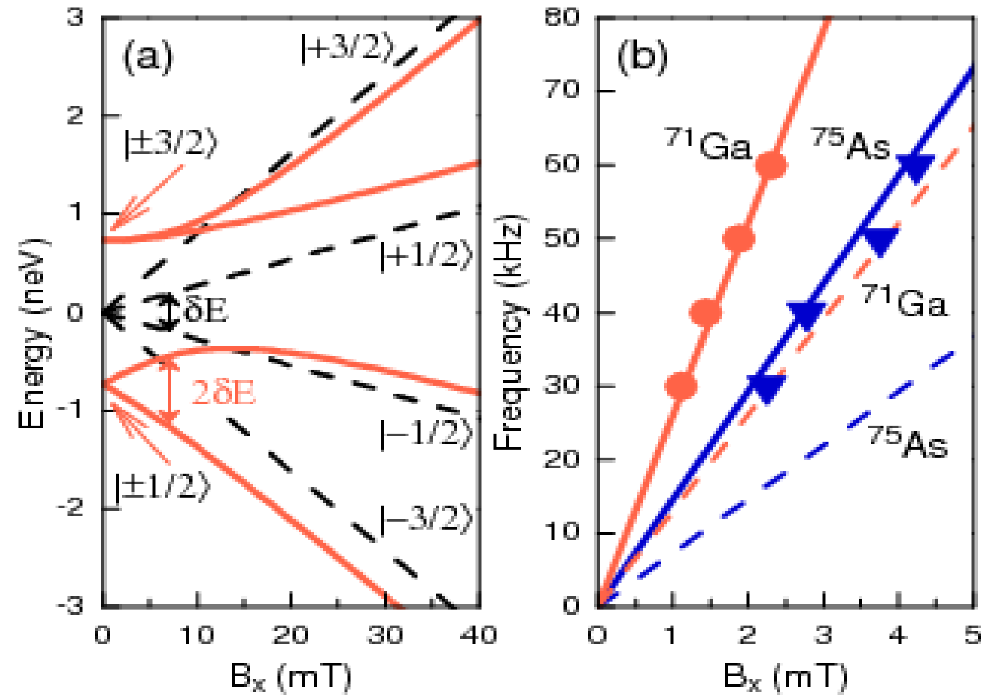
W-range of Hanle curves (B_{ext} < 50 mT) –
- the electron spin polarization is *destroyed* by NSP =>
=> nuclear spins are polarized *<u>perpendicular</u>* to the electron spin and parallel to B_{ext} => destroying of $NSP^{||}$ by rf application increases the electron spin polarization

Wings of Hanle curves (B_{ext} > 50 mT) –
- the electron spin polarization is *stabilized* by NSP =>
=> nuclear spin polarization has the component $NSP^{\perp}$ *<u>parallel</u>* to the electron spin and *<u>perpendicular</u>* to B_{ext} => destroying of $NSP^{\perp}$ by rf application decreases the electron spin polarization

Resonances in the W–range of Hanle curves

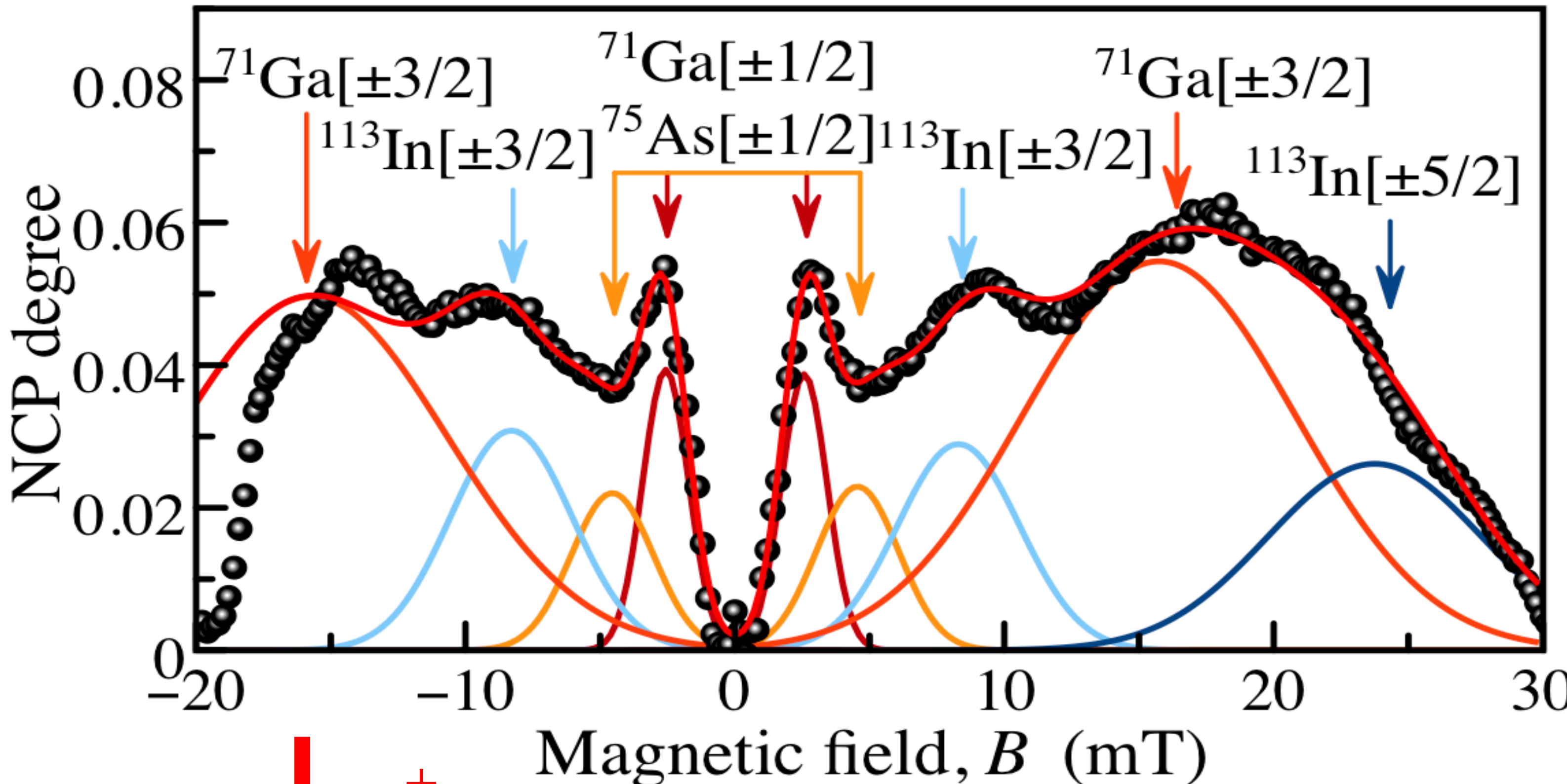
Solid and dashed lines – calculation with and without the influence of quadrupole interaction



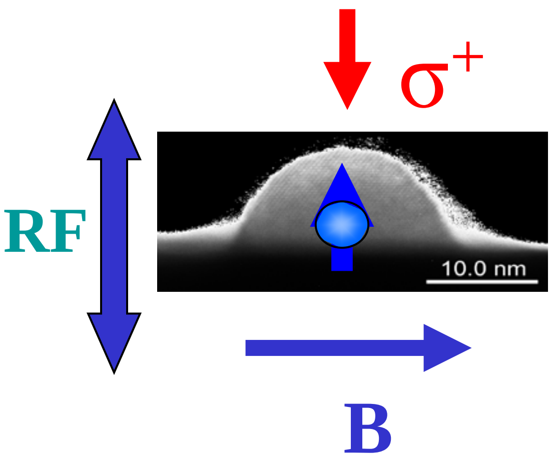


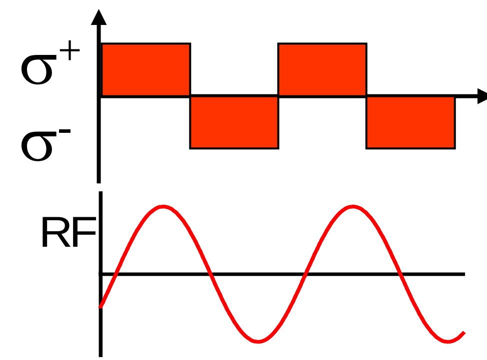
•Resonances in small B_x are due to the transitions between |±1/2> states of ^{71}Ga and ^{75}As
The applied radio frequencies are much smaller than quadropole ones, ν_Q (hundreds of kHz)

Effect of synchronization of RF pump and of polarisation modulation



f_{PM}=67 kHz





R.V.Cherbunin et al.,
*Phys. Rev. B, **84**, 041304 (2011).*

Nuclear quadrupole splitting

• The appearance of the nuclear spin component parallel to the electron spin is the result of nuclear quadrupole splitting

(*R. I. Dzhioev and V. L. Korenev, Phys. Rev. Lett.* ***99****, 037401 (2007)*)

for nuclear spins with $I = |\pm m/2>$ ($m \geq 3$) at electric field gradient ∇F.

Isotope	^{69}Ga	^{71}Ga	^{75}As	^{113}In	^{115}In
I	3/2	3/2	3/2	9/2	9/2
ν_Q, kHz (for ε_{zz}=0.01)	564	353	1490	388	383

•Zeeman splitting becomes comparable with $h\nu_Q$ in the range from 27 mT (^{71}Ga) to 200 mT (^{75}As)

• The main reason for the gradient is the disordered strains of the interface between QD and barrier

• The strain is the result of the difference between QD and barrier lattice constants.

Acknowledgments

This report is the review of joint research fulfilled in cooperation with my colleagues from Saint-Petersburg State University (R.Cherbunin, I.Gerlovin, I.Ignatiev, M.Kuznetsova, M.Petrov)

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and by Prof. Dr. Manfred Bayer (T.Auer, K.Flisinski, A.Greilich, R.Oulton, D.Yakovlev).

I would like to thank all my coauthors for the fruitful and pleasant cooperation.

Thanks for your attention