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

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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

Spatial transfer of charge -
• needs the time
• Joule heating

«1» is storing

«0» is storing

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

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at the QDs ensemble

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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 ensemble – 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

InP QD in external electric field

InGaP

GaAs

n-GaAs

ITO

InP QD

d ~ 40 nm

h ~ 5 nm

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

$E_e$ ~ 230 meV;
$E_{12}$ ~ 10 – 20 meV
$E_h$ ~ 15 meV;
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
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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Ad: Optical Detection of Nuclear Magnetic Resonance (ODNMR) at the QDs ensemble
σ⁺ excitation by circularly polarized light

Neutral QDs

\[ S_{Ze} = -1/2 \]

\[ S_{Zhh} = 3/2 \]

Negatively charged QDs

\sigma⁺ photoluminescence

Exciton

\[ S_{Ze} = 1/2 \]

\[ S_{Zhh} = -3/2 \]

Trion (exciton + resident electron)

\[ \rho_{cPL} = \frac{I_{sPL} - I_{oPL}}{I_{sPL} + I_{oPL}} \]

We studied \( \rho_{cPL} \) - degree of the PL circular polarization:

\( I_{sPL} \) - the intensity of the PL component with the same (opposite) helicity* as that of the excitation beam

*Helicity – sign of circular polarization degree

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Circularly polarized PL of InP QDs

$\lambda_{\text{max}} = 717$ nm
$E_{\text{max}} = 1730$ meV

Sample InP11
19 Jan 2002
$T = 5$ K;
exc $= 702.2$ nm
(1765.6 meV)
$U = -0.15$ V

Degree of circular polarization, $\rho_{\text{circ}}$

$\rho_{\text{circ}}$ from PL spectra

$\rho_{\text{circ}} = \frac{|\sigma^+ - \sigma^-|}{\sigma^+ + \sigma^-}$

Amplitude of NCP from PL kinetics

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Kinetics of circularly polarized PL of InP QDs

- InP QDs sample InP11
- P=8.5 mW
- $\lambda_{\text{exc}}=700$ nm
- $\lambda_{\text{PL}}=717$ nm
- B=0.10 T, T=5 K
- Data: 02.08.04

The graph shows the PL intensity (arb. un.) and the degree of circular polarization over time (ps). The red and blue lines represent positive and negative circular polarization, respectively. The amplitude of NCP is indicated on the graph.
Power dependence of NCP value demonstrates the rise of orientation of resident electron spins.
Dependence of NCP on applied bias

Amplitude of NCP vs. $U_{\text{bias}}$ (V)

- Lorentzian, $\Delta U_{\text{FWHM}}=0.72$ V
- Experiment

InP QDs, sample InP1
$\lambda_{\text{exc}}=700$ nm
$\lambda_{\text{PL}}=717$ nm
$B=0.10$ T, $T=5$ K
Data: 02.08.04

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Bias dependence of trionic quantum beats

[Kozin et al., PRB65, 241312(R) (2002)]
PL spectra polarization of uncharged InGaAs QDs (non-doped heterostructure)

\[
\begin{align*}
T & \sim 7 \text{ K} \\
\hbar \nu_{\text{exc}} & = 1.477 \text{ eV}
\end{align*}
\]
NCP of PL spectra of charged InGaAs QDs

\[ T \sim 7 \text{ K} \]

\[ h\nu_{\text{exc}} = 1.476 \text{ eV} \]
Power dependence of NCP of InGaAs QDs PL

$|\rho^\text{circ}|$

$I_{\text{exc}}/I_0$

$T \approx 7 \text{ K}$

$I_{\text{exc}} = 0.04I_0$

$I_{\text{exc}} = 0.13I_0$

$I_{\text{exc}} = 0.36I_0$

$I_{\text{exc}} = I_0$

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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)
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


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

A-type QD

$\sigma^+$-excitation

Energy relaxation and hole spin-flip

$\sigma^+$-PL

$\sigma^-$-PL

A-type QD

P-type QD

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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

\[ I^+(t) = \gamma_{PL} P_{ex} n_A (1 - q) \left( e^{-\gamma_{PL} t} - e^{-R t} \right) \]
\[ I^-(t) = \gamma_{PL} P_{ex} n_A q \left( e^{-\gamma_{PL} t} - e^{-R t} \right) + n_P \left( e^{-\gamma_{PL} t} - e^{-F t} \right) \]

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

For InP QDs:
\[ \gamma_{PL} = (250 \text{ ps})^{-1}, \]
\[ F = \gamma_{PL} + \gamma_f = (48 \text{ ps})^{-1}, \]
\[ R = \gamma_{PL} + \gamma_{rel} + \gamma_h = (32 \text{ ps})^{-1} \]

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

In our experiments:
\[ q \sim 0.05 - 0.10 \]
At low temperatures and excitation powers
\[ A_{\text{NCP}} \sim -(n_P - n_A) = -S \]

I.V. Ignatiev et al.,
Negative Circular Polarization (NCP) of negative singly-charged InGaAs QDs PL is a measure of spin polarization of resident electron in QD.

\[
\rho = \frac{I^{\sigma^+} - I^{\sigma^-}}{I^{\sigma^+} + I^{\sigma^-}}
\]

\(T \sim 2\ \text{K}\)

\(h\nu_{\text{exc}} = 1.476\ \text{eV}\)


\[\langle S_z \rangle \propto \rho_{\text{max}} \equiv NCP\]
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Ad: Optical Detection of Nuclear Magnetic Resonance (ODNMR)
at the QDs ensemble
Two-beam set-up

Set-up

Notations:
HM – half transparent mirror
M – mirror
GTP – Glan-Tompson prism
$\lambda/2$ – half wave plate
$\lambda/4$ – quarter wave plate
L – lens
PEM – photoelastic modulator
MC – alternate chopper
Spin memory of InGaAs QDS

- Long-time (up to $\sim 10^2$ 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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Ad: Optical Detection of Nuclear Magnetic Resonance (ODNMR) at the QDs ensemble
Hyperfine interaction of electron and nuclear spins

Electrons have s-type wave function in ground state

$\sim 10^5$ nuclei

Polarized light

Electron

Knight field $B_e \sim 1 \text{ mT}$

Overhauser field $B_N \sim 10 \text{ mT-10 T}$ of Dynamic Nuclear Polarisation (DNP)

$10^5$ nuclei

DNP is only one of the possible mechanisms ruling the Nuclear Spin Polarization (NSP)
Fluctuations $\Delta I_N$ of total nuclear spin $I_N$ in QDs - other type of possible NSP

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

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

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

Fluctuations of $\Delta I_N$ 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 $\sim 1$ нс
- nuclear spin precession in the Knight field created by the electron spin $\sim 1$ мкс
- nuclear spin relaxation at their dipole-dipole interaction $\sim 100$ мкс
• 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 $B_{\text{ext}}$ is **parallel** to optical axis ($Z$)
(and to electron spin oriented by circularly polarized light;
to direction of QD structure growth;
to $\text{grad}(F)$ of electrical field;
to DNP (NSP$^\parallel$))

Voigt configuration

Magnetic field $B_{\text{ext}}$ (X-axis) is **perpendicular** to optical axis ($Z$)
(and to electron spin oriented by circularly polarized light;
to direction of QD structure growth;
to $\text{grad}(F)$ of electrical field;
to DNP (NSP$^\perp$))
The NSP$^\parallel$ component is parallel to $B_{\text{ext}}$ (X-axis) in this case.
Influence of two components of NSP on resident electron spin polarization

$$A_{NCP}$$

$$B_{ext}(T)$$

$$\sigma^+$$

$$\sigma^-$$

Excitation

Faraday configuration

$$B_T = B_{ext} + B_N$$

$$B_T = B_{ext} - B_N$$

$$\langle B_f \rangle = 0$$

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Influence of two components of NSP on resident electron spin polarization

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

\[ \langle B_f^2 \rangle^{1/2} \approx 15 \text{ mT is practically independent on excitation power.} \]
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Ad: Optical Detection of Nuclear Magnetic Resonance (ODNMR)
at the QDs ensemble
«Usual» Hanle effect (Voigt configuration)

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

\[ S = \frac{S_0}{1 + (B/B_{1/2})^2}, \]

\[ B_{1/2} = \frac{\hbar}{|g_e|\mu_B T_2}. \]
**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

**Graphical Data**

Data: 10-08-2009
$P_{exc}=5$ mW
Spot diameter 60 mkm
CW regime

**NCP** vs. $B_x$ (mT)

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Hanle effect influenced by nuclear spin polarization

(1) \( B_{ext} = 0 \)

(2) \( B_{ext} < B_{Nx} \)

(3) \( B_{ext} = B_{Nx} \)

(4) \( B_{ext} < B_{Nx} \)
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⊥) to external magnetic field direction.

NSP|| and NSP⊥ are ⊥ 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⊥) to external magnetic field direction.

NSP∥ and NSP⊥ are ⊥ and || to the optically oriented electron spin.
Nuclear Spin Polarization (NSP) influence on electron spin polarization – two opposite results:

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

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

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

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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
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 ensemble of strained InGaAs/GaAs QDs where effect of the inhomogeneous broadening is considerable

The disordered strain leads to the gradient $\nabla F$ of electric field who splits the nuclear spin states into Kramers doublets $|\pm m/2\rangle$ (nuclear quadrupole splitting)
Influence of uniaxial deformation of QD

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

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

\[ \varepsilon_{zz}(\text{max}) = 0.0117 \]
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))

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RF influence on Nuclear Spin Polarization (NSP)

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

Wings of Hanle curves ($B_{\text{ext}} > 50$ mT ) –
- the electron spin polarization is stabilized by NSP =>
=> nuclear spin polarization has the component NSP┴ parallel to the electron spin and perpendicular to $B_{\text{ext}}$ => destroying of NSP ┴ by rf application decreases the electron spin polarization
Resonances in the W–range of Hanle curves

- Resonances in small $B_x$ are due to the transitions between $|\pm 1/2>$ states of $^{71}$Ga and $^{75}$As.
- The applied radio frequencies are much smaller than quadrupole ones, $\nu_Q$ (hundreds of kHz).

Solid and dashed lines – calculation with and without the influence of quadrupole interaction.
Effect of synchronization of RF pump and of polarization modulation

\[ \sigma^+ \]

\[ \sigma^- \]


\( f_{\text{PM}} = 67 \text{ kHz} \)
The appearance of the nuclear spin component parallel to the electron spin is the result of nuclear quadrupole splitting


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

<table>
<thead>
<tr>
<th>Isotope</th>
<th>( ^{69}\text{Ga} )</th>
<th>( ^{71}\text{Ga} )</th>
<th>( ^{75}\text{As} )</th>
<th>( ^{113}\text{In} )</th>
<th>( ^{115}\text{In} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( I )</td>
<td>3/2</td>
<td>3/2</td>
<td>3/2</td>
<td>9/2</td>
<td>9/2</td>
</tr>
<tr>
<td>( v_Q ), kHz (for ( \varepsilon_{zz}=0.01 ))</td>
<td>564</td>
<td>353</td>
<td>1490</td>
<td>388</td>
<td>383</td>
</tr>
</tbody>
</table>

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

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.
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) and from laboratories headed by Prof. Dr. Yasuaki Masumoto (M.Ikezawa, B.Pal) and by Prof. Dr. Manfred Bayer (T.Auer, K.Flisinski, A.Greilich, R.Oulton, D.Yakovlev).

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