Electron and Exciton Spin Dynamics in Quantum Dots

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Why quantum dots?

- **n-type single quantum dots** and quantum dot arrays
- Carrier localization leads to the spin relaxation slowdown
- Strong Coulomb interaction allows the **trion** formation
- Hyperfine interaction of electron and nuclear spins is effective

<table>
<thead>
<tr>
<th>Element</th>
<th>$^{27}$Al</th>
<th>$^{69(71)}$Ga</th>
<th>$^{75}$As</th>
<th>$^{115}$In</th>
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</thead>
<tbody>
<tr>
<td>$Z$</td>
<td>13</td>
<td>31</td>
<td>33</td>
<td>49</td>
</tr>
<tr>
<td>$I$</td>
<td>$5/2$</td>
<td>$3/2$</td>
<td>$3/2$</td>
<td>$9/2$</td>
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</tbody>
</table>

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Spin dynamics
Pump-probe technique

DICHROISM AND OPTICAL ANISOTROPY OF MEDIA, WITH ORIENTED SPINS OF FREE ELECTRONS

A. G. Aronov and E. L. Ivchenko

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- Faraday Effect (polarization plane rotation)
- Ellipticity Effect (appearance of the circular polarization)
- Kerr Effect (polarization plane rotation of reflected light)

Faraday rotation

Faraday rotation

Sample

B= 3T

M

$\Theta \propto S_z$

Pump: circularly polarized pulse orients carrier spins

$S_z \neq 0 \Rightarrow \begin{cases} n_+ \neq n_- \\ \alpha_+ \neq \alpha_- \end{cases}$

Delay time, $\Delta t$ (ps)

Faraday rotation

Ellipticity

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InAs \( n \)-type quantum dots array

Experiment (Dortmund group):

1. Origin of the Long-living signal at singlet \((S = 0)\) trion excitation?
2. Signals at negative delays, i.e. before the next pump pulse?
3. Growth of Faraday rotation with time?
Outline

1. Introduction. Questions to theory
2. Interaction of light with spins
   - Optical orientation via trions
   - State-of-the-art in electron spin control
   - Supersensitive detection of spin polarization
3. Spin mode-locking effect
   - Phase synchronization condition & passive mode-locking
   - Nuclei-induced active mode-locking
4. Spin dynamics in equilibrium
5. Conclusions
Optical orientation is a transformation of the photon angular momentum to the system of electron spins.

\[
\begin{align*}
| - \frac{1}{2}\rangle_{cb} & \quad | \frac{1}{2}\rangle_{cb} \\
| - \frac{3}{2}\rangle_{vb} & \quad | \frac{3}{2}\rangle_{vb}
\end{align*}
\]

Semiconductor quantum well or self-organized quantum dot:
- at \( \sigma^+ \) pump \(-\frac{3}{2} + 1 = -\frac{1}{2}\) \\
i.e. \((e, hh) = (-1/2, 3/2)\)
- at \( \sigma^- \) pump \(\frac{3}{2} + (-1) = \frac{1}{2}\) \\
i.e. \((e, hh) = (1/2, -3/2)\)

(normal light incidence)

Absorption of circularly polarized light generates spin-polarized electrons and holes.
Long-living electron spin polarization

\[ \sigma^+ \text{ pump} \Rightarrow (-1/2e, 3/2h) \]

The carriers with the spins opposite to those of photocreated electrons are picked out.

Hole spin relaxation/fast spin precession (@ \( B \neq 0 \)) \Rightarrow spin of returning electron is negligible

Resident electrons become spin polarized after recombination.
Spin pumping in quantum dots

Only QDs with a certain spin projection interact with the circularly polarized light.

Technical details:

- Short pump pulse: $\tau_p \ll \tau_{QD}, 2\pi/\Omega_L$
- Four level model: two ground states $\psi_{\pm 1/2}$, two excited states $\psi_{\pm 3/2}$

$$i\hbar \dot{\psi}_{1/2} = V^*_+(t)\psi_{3/2},$$
$$i\hbar \dot{\psi}_{3/2} = \hbar \omega_0 \psi_{3/2} + V_+(t)\psi_{1/2},$$

$$V_\pm(t) = -\int d(r)E_{\sigma\pm}(r, t)d^3r$$

- Returning electron is depolarized

Under $\sigma^+$ pump the wave function transforms as

$$\psi_{1/2}(t \to +\infty) = Qe^{i\Phi} \psi_{1/2}(t \to -\infty)$$
$$\psi_{-1/2}(t \to +\infty) = \psi_{-1/2}(t \to -\infty)$$

Similar to N. Rosen & C. Zener (1932)
Spin pumping in quantum dots

Only QDs with a certain spin projection interact with the circularly polarized light.

Electron spin \( S = \frac{1}{2} \langle \psi | \sigma | \psi \rangle \)

\[
S_z^+ = \frac{Q^2 + 1}{2} S_z^− + \frac{Q^2 - 1}{4} \\
S_y^+ = Q \cos \Phi S_y^− - Q \sin \Phi S_x^− \\
S_x^+ = Q \cos \Phi S_x^− + Q \sin \Phi S_y^−
\]

- Before pump pulse \( S^− \), after \( S^+ \)
- An increase of spin \( z \)-component
- In-plane components rotation

Yugova, MMG, Ivchenko, Efros (2009)
Spin rotation by the optical pulse

- Circular pulse
  - Generates spin coherence (optical orientation)
  - Rotates spin (inverse Faraday effect)

\[ S^+_z = \frac{Q^2 + 1}{2} S^-_z + \frac{Q^2 - 1}{4} \]
\[ S^+_y = Q \cos \Phi S^-_y - Q \sin \Phi S^-_x \]
\[ S^+_x = Q \cos \Phi S^-_x + Q \sin \Phi S^-_y \]

Parameters \( Q \) and \( \Phi \) are determined by the pump pulse intensity, duration and detuning from the resonance (\( \Phi \))


Details in next talk by A. Ramsay
Electron spin coherence generation

Circularly polarized pulses being resonant with the singlet trion transition orient electron spins in quantum dots

Slightly off-resonant pulse rotates spins
Effects of the probe

**QW or ensemble of QDs or QWRs**

**Probe**: transmission/reflection of weak linearly-polarized pulse

weak=does not affect spin coherence

\[ \sigma^+ \]

**Ellipticity and Faraday rotation**

\[ \mathcal{E} + i \mathcal{F} \propto r_+ - r_- \]

**Resonance (trion, exciton, …)**

\[ r_\pm (\omega) = \frac{i \Gamma_{0,\pm}}{\omega_{0,\pm} - \omega - i (\Gamma_{0,\pm} + \Gamma_\pm)} \]

\[ t_\pm = 1 + r_\pm \]

Zhukov, Yakovlev, Bayer, MMG, Ivchenko, et al. (2007)
Probing the electron spins

- **Faraday Effect** (polarization plane rotation)
  \[
  \mathcal{F} = \lim_{z \to +\infty} \int_{-\infty}^{\infty} \left[ |E_x'(z, t)|^2 - |E_y'(z, t)|^2 \right] dt
  \]

- **Ellipticity Effect** (appearance of the circular polarization)
  \[
  \mathcal{E} = \lim_{z \to +\infty} \int_{-\infty}^{\infty} \left[ |E_{\sigma-}^{(t)}(z, t)|^2 - |E_{\sigma+}^{(t)}(z, t)|^2 \right] dt
  \]

**Probe induced field**
\[
\delta E(t) = -4\pi \left( \frac{\omega_{pr}}{c} \right)^2 \frac{ie^{iq|z|}}{2q} N_{QD}^2 \Pi(t)
\]
\[
\Pi_x \propto (n_e - n_{tr}) E_x^{\text{probe}}
\]
\[
\Pi_y \propto (S_z - J_z) E_x^{\text{probe}}
\]

**Spectral function**
\[
\mathcal{E} + i\mathcal{F} \propto \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' f(t)f(t') e^{i\Delta(t-t')}
\]
\[
\Delta = \omega_{pr} - \omega_0; \quad E_{pr} \propto f(t)e^{-i\omega t}
\]

Yugova, MMG, Ivchenko, Efros (2009)
Electron spin coherence detection

Linearly polarized pulse can readout spin polarization

Correct description is based on reflection/transmission rather than on dielectric constant

MMG, Yugova + Dortmund group (2010)
1. Introduction. Questions to theory

2. Interaction of light with spins
   - Optical orientation via trions
   - State-of-the-art in electron spin control
   - Supersensitive detection of spin polarization

3. Spin mode-locking effect
   - Phase synchronization condition & passive mode-locking
   - Nuclei-induced active mode-locking

4. Spin dynamics in equilibrium

5. Conclusions
Electron spin precession mode-locking

Train of pump and probe pulses, $T_R$ is the repetition period

- Large spread of electron $g$-factors $\Rightarrow$ fast dephasing
- Signal reappears before the next pump pulse arrival

\[
\hbar/\tau_p \sim 1 \text{ meV}
\]

\[
\Omega T_R = 2\pi N
\]

Constructive interference

exp.: $N \sim 10^2$
Electron spin precession mode-locking

Train of pump and probe pulses, \( T_R \) is the repetition period

- Large spread of electron \( g \)-factors \(\Rightarrow\) fast dephasing
- Signal reappears before the next pump pulse arrival

\[ \frac{\hbar}{\tau_p} \sim 1 \text{ meV} \]

\[ \Omega T_R = 2\pi N \]

Constructive interference

exp.: \( N \sim 10^2 \)

"Passive" mode-locking:

all synchronized modes have same initial phases owing to the pump
Electron spin precession modelocking – III
Electron spin precession modelocking – III

$S_z$ (arb. units)

Time ($T_R$)

-1.0 -0.5 0.0 0.5 1.0

0.0 0.2 0.4 0.6 0.8 1.0

Electron spin precession modelocking – III

S
S
S
S
H
H
H
H
arb
arb
arb
arb
units
units
units
units
L
L
L
L

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Electron spin precession modelocking – III

\[ S_z (\text{arb. units}) \]

\[ \text{Time (} T_R \text{)} \]

\[ 0.0 \quad 0.2 \quad 0.4 \quad 0.6 \quad 0.8 \quad 1.0 \]

\[ -1.0 \quad -0.5 \quad 0.0 \quad 0.5 \quad 1.0 \]
Electron spin precession modelocking – III

\[ S_z(\text{arb. units}) \]

Time \( (T_R) \)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0
-0.5
0.0
0.5
1.0

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Electron spin precession modelocking – III

$S_z$ (arb. units) vs. Time ($T_R$)
Electron spin precession modelocking – III

$S_z$ (arb. units)

Time ($T_R$)

-1.0 0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

$H$ (arb. units)

-$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)

$S_z$ (arb. units)

Time ($T_R$)

$H$ (arb. units)

$z$ ($z$)

0.0 0.2 0.4 0.6 0.8 1.0

-1.0 -0.5 0.0 0.5 1.0

Time ($T_R$)
Where are other spins with frequencies $\Omega \neq \frac{2\pi N}{T_R}$?
Nuclear effects: experimental evidence

Very high amplitude at negative delays

Ω_{eff} T_R = 2\pi

↑

Nuclei-induced frequency focusing

↑

Ω_{eff} T_R = \pi

Ω_{eff} = \Omega_L + \Omega_{nucl}

Feedback of nuclei is necessary

Nuclei-induced frequency focusing takes place

Classical origin of the focusing

Nuclear spin polarization

\[ m = \sum_i I_i \]

Spin precession between the pulses

\[
\frac{dm}{dt} = [\alpha S(t) \times m(t)] + [\omega \times m(t)],
\]

\[
\frac{dS}{dt} = [\alpha m(t) \times S(t)] + [\Omega \times S(t)],
\]

\((n - 1)T_R < t < nT_R\)

Pump pulse action: \( m^+ = m^- \)

\[
S^+_z = \frac{Q^2 + 1}{2} S^-_z + \frac{Q^2 - 1}{4}
\]

\[
S^+_y = QS^-_y, \quad S^+_x = QS^-_x
\]

Time scales @ \( B = 1 \text{T} \)

1. Electron spin precession
   \[ \frac{2\pi}{\Omega} \lesssim 0.1 \text{ ns} \]
2. Precession in nuclear field
   \[ \frac{2\pi}{\alpha m} \sim 10 \text{ ns} \]
3. Pulse repetition period
   \[ T_R \sim 10 \text{ ns} \]
4. Nuclear spin precession
   \[ \frac{2\pi}{\omega} \lesssim 100 \text{ ns} \]
5. Precession in electron field
   \[ \frac{2\pi}{\alpha S} \gtrsim 10^3 \text{ ns} \]

MMG, Yugova, Efros (2012)
Classical origin of the focusing

Nuclear spin polarization

\[ m = \sum I_i \]

\[ \frac{\, \mathrm{d} m}{\, \mathrm{d} t} = [\alpha S(t) \times m(t)] + [\omega \times m(t)] \]

\[ \frac{\, \mathrm{d} S}{\, \mathrm{d} t} = [\alpha m(t) \times S(t)] + [\Omega \times S(t)] \]

Pump pulse action:

\[ m^+ + S^+ + z = Q_2 S^+ - \frac{1}{4} S^+ + Q_2 S^+ - \frac{1}{4} S^+ \]

Time scales @ \( B = 1 \text{ T} \):

- Electron spin precession: \( \lesssim 0.1 \text{ ns} \)
- Precession in nuclear field: \( \sim 10 \text{ ns} \)
- Pulse repetition period: \( \sim 10 \text{ ns} \)
- Precession in electron field: \( \lesssim 100 \text{ ns} \)

\[ \frac{\Omega T_R}{2\pi} = 150.5 \]

\[ \frac{\Omega T_R}{2\pi} = 100.5 \]

\[ \frac{\Omega T_R}{2\pi} = 50.5 \]

\[ \frac{2\pi}{\alpha S} \gtrsim 10^3 \text{ ns} \]

MMG, Yugova, Efros (2012)
Classical origin of the focusing

Nuclear spin polarization

\[ m = \sum_i I_i \]

Spin precession between the pulses

\[ \frac{dm}{dt} = [\alpha S(t) \times m(t)] + [\omega \times m(t)] \]

“Active” mode-locking:

spin precession frequencies are synchronized in all dots thanks to nuclei

Pump pulse action: \( m^+ = m^- \)

Time scales @ \( B = 1 \) T

1. Electron spin precession
   \[ \frac{2\pi}{\Omega} \lesssim 0.1 \text{ ns} \]

2. Precession in nuclear field
   \[ \frac{2\pi}{\omega} \sim 10 \text{ ns} \]

3. Nuclear spin precession
   \[ \frac{2\pi}{\omega} \lesssim 100 \text{ ns} \]

4. Precession in electron field
   \[ \frac{2\pi}{\alpha S} \gtrsim 10^3 \text{ ns} \]

MMG, Yugova, Efros (2012)
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Magnetic resonance in the Faraday-rotation noise spectrum

E. B. Aleksandrov and V. S. Zapasskii

(Submitted 23 January 1981)

A maximum at the magnetic resonance frequency of sodium atoms in the ground state is observed near the 5896 Å absorption line in the fluctuation spectrum of the azimuth of the polarization plane of light crossing a magnetic field in sodium vapor. The experiment is a demonstration of a new EPR method which does not require in principle magnetic polarization of the investigated medium, nor the use of high-frequency or microwave fields to induce the resonance.

Can spin dynamics be studied without any pump?
Spin noise spectroscopy

Magnetic resonance in the Faraday-rotation noise spectrum

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\[ \langle \vartheta_F(t) \vartheta_F(t') \rangle, \]
\[ \langle \vartheta_K(t) \vartheta_K(t') \rangle \propto \langle S_z(t) S_z(t') \rangle \]

Invited review
Semiconductor spin noise spectroscopy: Fundamentals, accomplishments, and challenges

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Spin noise in electron ensembles

Single spin

\[
\langle s_x \rangle = \langle s_y \rangle = \langle s_z \rangle = 0 \quad \text{but} \quad \langle s_x^2 \rangle = \langle s_y^2 \rangle = \langle s_z^2 \rangle = \frac{1}{3} \times \frac{1}{2} \left( 1 + \frac{1}{2} \right)
\]

Spin ensemble

\[
\sqrt{\langle S_i^2 \rangle} = \sqrt{N} \sqrt{\langle s_i^2 \rangle} = \frac{\sqrt{N}}{2}
\]
Spin noise theory

\[ \frac{\partial \delta s(t)}{\partial t} + \frac{\delta s(t)}{\tau_s} + \delta s(t) \times (\Omega_B + \Omega_N) = \xi(t) \]

Random (Langevin) forces

\[ \langle \xi_\alpha(t')\xi_\beta(t) \rangle = \frac{1}{2\tau_s} \delta_{\alpha\beta} \delta(t' - t) \]

Our task is to calculate

\[ \langle s_\alpha(t)s_\beta(t') \rangle \quad \text{or} \quad (\delta s_\alpha \delta s_\beta)_{\omega} = \int_{-\infty}^{+\infty} \langle s_\alpha(t + \tau)s_\beta(t) \rangle e^{-i\omega \tau} d\tau \]

MMG, Ivchenko (2012)
Single spin in magnetic field

Without field: \[ \dot{s}_j + \frac{s_j}{\tau_s} = \xi_j \]

\[
\langle \delta s_j^2 \rangle_\omega = \frac{2\tau_s\langle \delta s_j^2 \rangle}{1 + (\omega\tau_s)^2} = \frac{1}{2} \left( \frac{\tau_s}{1 + (\omega\tau_s)^2} \right), \quad \langle \xi_j^2 \rangle_\omega = \frac{2\langle \delta s_j^2 \rangle}{\tau_s} = \frac{1}{2\tau_s},
\]

since \( \langle \delta s_j^2 \rangle = \langle s_j^2 \rangle - \langle s_j \rangle^2 = 1/4. \)

In the presence of the field: \[ \left( -i\omega + \frac{1}{\tau_s} \right) s + s \times \Omega = \xi. \]

\( \Omega \parallel 3 \) (axes: 1 \( \perp \) 2 \( \perp \) 3):

\[
\langle \delta s_1^2 \rangle_\omega = \langle \delta s_2^2 \rangle_\omega = \frac{\pi}{4} \left[ \Delta(\omega - \Omega) + \Delta(\omega + \Omega) \right], \quad \langle \delta s_3^2 \rangle_\omega = \frac{\pi}{2} \Delta(\omega),
\]

Lorentian form in strong enough magnetic fields \( (\Omega \tau_s \gg 1) \)

\[
\Delta(\eta) = \frac{1}{\pi} \frac{\tau_s}{1 + (\eta \tau_s)^2}
\]

MMG, Ivchenko (2012)
Singly charged quantum dot ensemble

MMG, Ivchenko (2012)
Outline & Co-authors

Spin noise in quantum dot ensembles, arXiv:1206.3479
Spin coherence generation and detection in spherical nanocrystals, arXiv:1205.2764

Plan
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Co-authors
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A.L. Efros (Naval Research, USA)
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D.R. Yakovlev, M. Bayer (TU-Dortmund, Germany)
Dynamics and fluctuations of electron and nuclear spins can be most conveniently studied by optical means.

Thank you for attention!