Coherent spin dynamics in semiconductor low-dimensional systems

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Spin-related phenomena in mesoscopic transport 17th of September, 2012



Electron spin plays major role in various phenomena. Most importantly, the spin of electrons results in the Pauli exclusion principle, which in turn underlies the periodic table of chemical elements and makes our existence possible.

It is needless to explain in this audience the manifestations of spin in the electron transport. Hence, the aim of my talk is to demonstrate alternative, in particular, optical means to access electron spin in low-dimensional systems.

Pump-probe technique





- *F*araday Effect (polarization plane rotation)
- *Ellipticity Effect* (appearance of the circular polarization)
- Kerr Effect (polarization plane rotation of reflected light)

Awschalom et al. (1985); Zheludev et al. (1994)

Motivation



Quantum wells, free electrons



T. Korn et al. (2009) M.M. Glazov

Quantum dot ensembles



A. Greilich et al. (2006)

Single quantum dots



M.H. Mikkelsen et al. (2007)



A. Brunetti et al. (2006)

Pump-probe is a widespread technique to measure

relaxation times, precession frequencies

- What can we learn from "microscopics" of signals?
- What about signal amplitudes?

We focus here on quantum dots:



n-type **single quantum dots** and quantum dot arrays

- Carrier localization leads to the spin relaxation slowdown
- Possibility to address resonantly singlet trion
- Hyperfine interaction of electron and nuclear spins is effective
- Similar physics for quantum wires, wells and bulk materials with low, $n_d a_B^d \ll 1$, density electrons/holes

Other systems: Phys. Solid State 54, 1 (2012)





Element	²⁷ Al	⁶⁹⁽⁷¹⁾ Ga	⁷⁵ As	¹¹⁵ In
Z	13	31	33	49
Ι	5/2	3/2	3/2	9/2

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Spin pump-probe signals for InAs *n*-type quantum dots array

Experiments:









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

Outline



Introduction. Questions to theory

2 Interaction of light with spins

- Optical orientation via trions
- Detection of spin polarization.

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

- Emergence of Faraday rotation
- Spin dynamics in equilibrium
- Conclusions

Optical orientation in nanosystems



Optical orientation is a transformation of the photon angular momentum to **the system of electron spins**

$$\begin{vmatrix} -\frac{1}{2} \rangle_{cb} & |\frac{1}{2} \rangle_{cb} \\ \sigma^{+} \\ |-\frac{3}{2} \rangle_{vb} & |\frac{3}{2} \rangle_{vb} \end{vmatrix}$$

(normal light incidence)



Semiconductor quantum well or self-organized quantum dot:

• at σ^+ 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. $(e, hh) = (1/2, -3/2)$

Absorption of circularly polarized light

generates spin-polarized electrons and holes

Long-living electron spin polarization





 $\sigma^+ \text{ pump} \Rightarrow (-1/2_e, 3/2_h)$

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

Shabaev, Efros, Gammon, Merkulov (2003); Greilich et al (2006); Zhukov, Yakovlev, Bayer, MMG, Ivchenko, Karczewski, Kossut (2007)

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Spin pumping in quantum dots



Only QDs with a certain spin projection interact with

the circularly polarized light

Technical details:

- Short pump pulse: $au_p \ll au_{QD}, 2\pi/\Omega_{
 m L}$
- Four level model: two ground states $\psi_{\pm 1/2}$, two excited states $\psi_{\pm 3/2}$
- Returning electron is depolarized Under σ^+ pump the wave function transforms as

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

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

Transformation of the wave function means transformation of the spin

Similar to N. Rosen & C. Zener (1932)



Parameters Q and Φ are determined by the pump pulse shape, area, and detuning from resonance

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Coherent Spin Dynamics in Semiconductors

Spin pumping in quantum dots



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



Spin rotation by optical pulse





Phys. Rev. B 80, 104436 (2009)

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





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

Slightly off-resonant pulse rotates spins

Spin coherence detection



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QW or ensemble of QDs, ...



Ellipticity and Faraday rotation

$$\mathcal{E}+\mathrm{i}\mathcal{F}\propto r_+-r_+$$

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Probe: transmission/reflection of weak linearly-polarized pulse weak=does not affect spin coherence

= + +

 $r_+ \neq r_- \Rightarrow$ rotation of the linear polarization plane of the probe pulse and appearance of its ellipticity

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)

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Probing the electron spins

• Faraday Effect (polarization plane rotation)

$$\mathcal{F} = \lim_{z
ightarrow +\infty} \int_{-\infty}^{\infty} \left[|E_{x'}^{(t)}(z,t)|^2 - |E_{y'}^{(t)}(z,t)|^2
ight] \mathrm{d}t$$

• Ellipticity Effect (appearance of the circular polarization)

$$\mathcal{E} = \lim_{z \to +\infty} \int_{-\infty}^{\infty} \left[|E_{\sigma^-}^{(t)}(z,t)|^2 - |E_{\sigma^+}^{(t)}(z,t)|^2 \right] \mathrm{d}t$$

Probe induced field

$$\begin{split} \delta \boldsymbol{E}(t) &= -4\pi \left(\frac{\omega_{\mathrm{pr}}}{c}\right)^2 \frac{\mathrm{i} \mathrm{e}^{\mathrm{i} q |\boldsymbol{z}|}}{2q} N_{QD}^{2d} \boldsymbol{\Pi}(t) \\ \Pi_x \propto (n_e - n_{tr}) E_x^{probe} \\ \Pi_y \propto (\boldsymbol{S}_z - \boldsymbol{\mathcal{I}}_z) E_x^{probe} \end{split}$$



Yugova, MMG, Ivchenko, Efros (2009)

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 $\Delta = \omega_{\rm pr} - \omega_0; E_{\rm pr} \propto f(t) {\rm e}^{-{\rm i}\omega t}$



Electron spin coherence detection







Linearly polarized pulse can readout spin polarization

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

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Outline





- 2 Interaction of light with spins
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- Spin dynamics in equilibrium
- **Conclusions**

Resonant spin amplification and mode-locking





Signal dependence on \boldsymbol{B} at a fixed delay Δt

Resonant spin amplification is

determined by commensurability of spin precession and pump repetition periods

$$\begin{array}{c} \text{Hind Watch
\text{Hind Watch
Hind Watch
\text{Hind Watch
Hind Watch
Hind$$

Kikkawa, Awschalom (1998)



In inhomogeneous systems

certain electrons become polarized stronger than others (mode-locking)

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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





Greilich, Yakovlev, Shabaev, Efros, Yugova, Oulton, Stavarache, Reuter, Wieck, Bayer (2006)

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Coherent Spin Dynamics in Semiconductors

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
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Where are other spins with precession frequecies $\Omega \neq \frac{2\pi K}{T_P}$?



Nuclear effects: experimental evidence







Greilich, Shabaev, Yakovlev, Efros, Yugova, Reuter, Wieck, Bayer (2007)

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Classical origin of the focusing





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Classical origin of the focusing





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Classical origin of the focusing



Random nuclear spin dynamics

Random nuclear spin flips driven by precessing electron spin:

$$\frac{1}{\tau_{\rm n}} = \frac{\alpha^2 \tau_{\rm c}}{1 + (\Omega \tau_{\rm c})^2} \sim \frac{\alpha^2}{\Omega^2 \tau_{\rm c}}$$

M.I. Dyakonov and V.I. Perel' (1973)

V. Korenev (2010)

A. Greilich et al (2006)

Phenomenological electron correlation time $\tau_c \propto 1/W_{tr}$, where W_{tr} is the trion creation probability. At PSC nuclear spin flips stop.

✓ Our approach shows that the nuclear dynamics is **directed**!

Dynamical nuclear polarization

Equilibrium approach/detuned pump:

$$\frac{\mathrm{l}m_x}{\mathrm{d}t} = -\frac{m_x - qS_x(m_x)}{T_{1e}} - \frac{m_x}{T_1}$$
A.W. Overhauser (1953)

PSC is generally not stable!?

✓ Nonequilibrium regime for the pump-probe conditions!
 ✓ Tuning is possible for resonant pump!

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Nuclei induce effective tuning of electron spin precession frequencies to the synchronous with pump repetition period values



Why does spin Faraday signal amplitude grow with time?

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Emergence of Faraday rotation



Inhomogenous array:

- Spread of resonant frequencies
- Related spread of g-factors
- Random nuclear fields



Pump-probe signal calculation

QDs array, correlated distribution of resonant frequencies, ω_0 , and *g*-factors, $p(\omega_0, g)$:

$$\mathcal{S}(\Delta t) \propto \int \mathrm{d}\omega_0 \mathrm{d}g \, p(\omega_0,g) S_z(\omega_0,g,\Delta t) imes$$

$$\left\{ egin{array}{c} \mathcal{F}(\omega_0-\omega_{
m pr}) \ \mathcal{E}(\omega_0-\omega_{
m pr}) \end{array}
ight\}$$



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Emergence of Faraday rotation



Inhomogenous array:

- Spread of resonant frequencies
- Related spread of g-factors
- Random nuclear fields





No Faraday rotation signal at t = 0

Faraday rotation signal appears at $t \neq 0$

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Emergence of Faraday rotation



Inhomogenous array:

- Spread of resonant frequencies
- Related spread of g-factors
- Random nuclear fields



Simple model $\mathcal{F}(t) = \frac{1}{2} \sqrt{\frac{\pi}{2\tau_p^2}} \exp\left[\frac{-\Delta^2 \tau_p^2 / 2 - (\Omega' t)^2}{8\tau_p^2}\right] >$ $\left[2\Delta\tau_p\cos\left(\tilde{\Omega}_0 t\right) + \frac{\Omega' t}{\tau_p}\sin\left(\tilde{\Omega}_0 t\right)\right]$



Faraday rotation does not reflect "averaged" spin dynamics of electron ensemble!

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Inhomogenous array:

• Spread of

Faraday and ellipticity are formed by different ensembles of spins

- Different temporal behavior
- Faraday rotation does not reflect "averaged" spin dynamics of electron ensemble

• панионт пистса

fields

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Spin fluctuations





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Spin fluctuations





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Spin fluctuations





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Spin noise spectroscopy

Magnetic resonance in the Faraday-rotation noise spectrum

E. B. Aleksandrov and V. S. Zapasskii

(Submitted 23 January 1981) Zh. Eksp. Teor. Fiz. 81, 132-138 (July 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 microware fields to induce the resonance.



Invited review

Semiconductor spin noise spectroscopy: Fundamentals, accomplishments, and challenges

Georg M. Müller, Michael Oestreich, Michael Römer, Jens Hübner*

Institut für Festkörperphysik, Leibniz Universität Hannover, Appelstraße 2, D-30167 Hannover, Germany





$\langle \vartheta_{\mathcal{F}}(t) \vartheta_{\mathcal{F}}(t') \rangle,$

 $\langle \vartheta_{\mathcal{K}}(t) \vartheta_{\mathcal{K}}(t') \rangle \propto \langle S_z(t) S_z(t') \rangle$



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Autocorrelations





Electron velocity noise:

$$\langle v_x(t)v_x(0)
angle = rac{v_F^2}{2}e^{-t/ au_p}$$

Diffusion coefficient

$$D = \int_0^\infty \langle \mathbf{v}_x(t) \mathbf{v}_x(0) \rangle \mathrm{d}t = \frac{\mathbf{v}_F^2 \tau_p}{2}$$



Electron spin noise:

$$\langle s_z(t)s_z(0)\rangle = \langle s_z^2\rangle e^{-t/\tau_s}$$

Magnetic susceptibility

$$\mu_{zz}(\omega) \propto \int_0^\infty \langle s_z(t) s_z(0)
angle e^{\mathrm{i}\omega t} \mathrm{d}t$$

Autocorrelations





Electron velocity noise:

$$\langle v_x(t)v_x(0)
angle = rac{v_F^2}{2}e^{-t/ au_p}$$

Diffusion coefficient

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Electron spin noise:

$$\langle s_z(t)s_z(0)\rangle = \langle s_z^2\rangle e^{-t/\tau_s}$$

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$$\mu_{zz}(\omega) \propto \int_0^\infty \langle s_z(t) s_z(0)
angle e^{\mathrm{i}\omega t} \mathrm{d}t$$



Single spin

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

Spin ensemble



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Spin noise theory

Spin fluctuation δs in a dot

$$\frac{\partial \delta \boldsymbol{s}(t)}{\partial t} + \frac{\delta \boldsymbol{s}(t)}{\tau_{s}} + \delta \boldsymbol{s}(t) \times (\boldsymbol{\Omega}_{\boldsymbol{B}} + \boldsymbol{\Omega}_{N}) = \boldsymbol{\xi}(t)$$

Zero field

0.5 (a)

Nuclei fluctuations

$$(\delta s_{\alpha}^2)_{\omega} = \frac{\pi}{6} \left\{ \Delta(\omega) \right\}$$

$$+\int_0^\infty d\Omega_N F(\Omega_N)$$

$$[\Delta(\omega - \Omega_N) + \Delta(\omega + \Omega_N)]\}$$

(90) (art 0.0 (000)

 ω/δ_s

Random (Langevin) forces

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

$$(\delta s_{\alpha} \delta s_{\beta})_{\omega} = \begin{pmatrix} g_{\beta} & g_{\alpha} \\ g_{\beta} & g_{\alpha} \\ g_{\beta} \\ g_{\alpha} \\ g_{\alpha} \\ g_{\beta} \\ g_{\alpha} \\ g_{\alpha} \\ g_{\beta} \\ g_{\alpha} \\ g_$$

MMG, lvchenko (2012)

• Zero field peak • Peak at $\Omega \sim \langle \sqrt{\Omega_N^2} \rangle$

 $\omega | \delta_r$

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Spin fluctuation δs in a dot

$$\frac{\partial \delta \boldsymbol{s}(t)}{\partial t} + \frac{\delta \boldsymbol{s}(t)}{\tau_s} + \delta \boldsymbol{s}(t) \times (\boldsymbol{\Omega}_{\boldsymbol{B}} + \boldsymbol{\Omega}_{N}) = \boldsymbol{\xi}(t)$$

Transverse field

- Zero field peak is reduced
- Main peak shifts $\sim \Omega_B$



Spin noise theory & experiment







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Spin noise spectroscopy

provides information about spin precession and dephasing in close-to-equilibrium conditions

Prospects & Challenges



Spins in cavities entanglement via spin



Smirnov, MMG (2012)

Conclusions



Faraday effect and ellipticity provide complementary information about dynamics of electron and nuclear spins in QDs

Coherent spin dynamics of electrons and excitons in nenostructures (a review)

Plan

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