Electron and Exciton Spin Dynamics in Quantum Dots

M.M. Glazov

Ioffe Physical-Technical Institute, St-Petersburg, Russia

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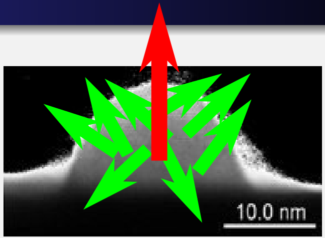
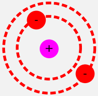
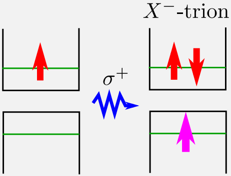
Why quantum dots?

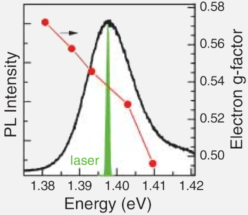
n-type **single quantum dots** and quantum dot arrays

- Carrier localization leads to the spin relaxation slowdown
- Strong Coulomb interaction allows the trion formation
- Hyperfine interaction of electron and nuclear spins is effective









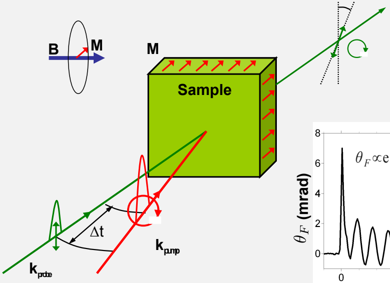
Element	^{27}Al	$^{69(71)}$Ga	^{75}As	^{115}In
Z	13	31	33	49
I	$5/2$	$3/2$	$3/2$	$9/2$

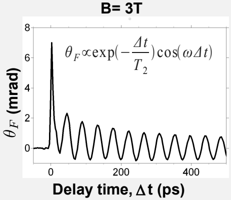
DICHROISM AND OPTICAL ANISOTROPY OF MEDIA WITH ORIENTED SPINS OF FREE ELECTRONS

A. G. Aronov and E. L. Ivchenko

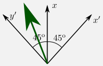
A. F. Ioffe Physicotechnical Institute,
Academy of Sciences of the USSR, Leningrad

Sov. Phys. Solid State (1973)

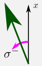




Faraday rotation



Ellipticity



Pump: circularly polarized pulse orients carrier spins

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

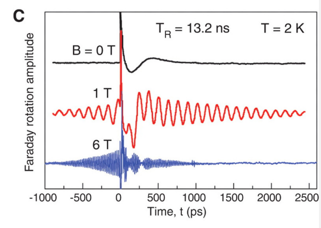
Faraday rotation

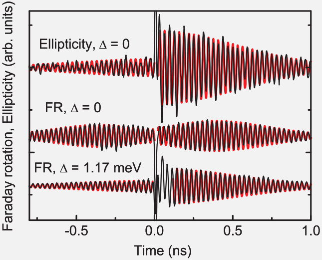
$$\Theta \propto S_z$$

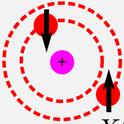
- $\mathcal{F}$araday Effect (polarization plane rotation)
- $\mathcal{E}$llipticity Effect (appearance of the circular polarization)
- $\mathcal{K}$err Effect (polarization plane rotation of reflected light)

InAs *n*-type quantum dots array

Experiment
(Dortmund group):







X^--trion

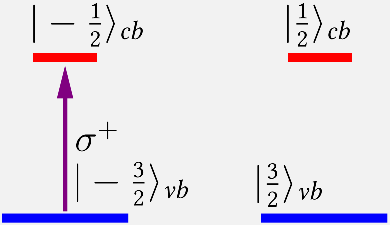
1. Origin of the Long-living signal at singlet ($S = 0$) trion excitation?
2. Signals at negative delays, i.e. before the next pump pulse?
3. Growth of Faraday rotation with time?

Outline

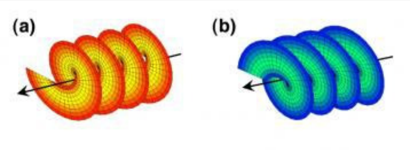
1. Introduction. Questions to theory
2. Interaction of light with spins
   - Optical orientation via trions
   - State-of-the-art in electron spin control
   - Supersensitive detection of spin polarization
3. Spin mode-locking effect
   - Phase synchronization condition & passive mode-locking
   - Nuclei-induced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

Optical orientation in nanosystems

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



(normal light incidence)



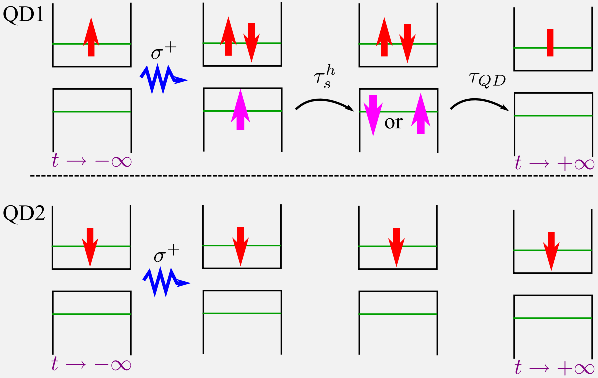
Semiconductor quantum well or self-organized quantum dot:

- at σ^+ pump $-\frac{3}{2} + 1 = -\frac{1}{2}$
  i.e. $(e, hh) = (-1/2, 3/2)$
- at σ^- 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



σ^+ 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 (@ $\boldsymbol{B} \neq 0$) $\Rightarrow$ 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 $\psi_{\pm 3/2}$

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

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

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

- Returning electron is depolarized

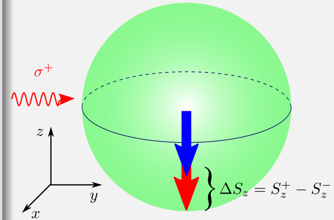
Under σ^+ pump the wave function transforms as

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

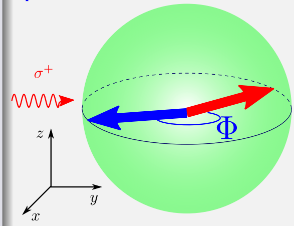
$$\psi_{-1/2}(t\to+\infty) = \psi_{-1/2}(t\to-\infty)$$

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

Spin orientation

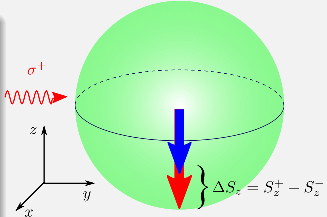


Spin rotation

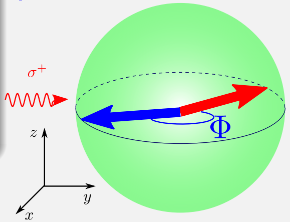


Only QDs with a **certain spin projection** interact with the **circularly polarized light**

Spin orientation $\sim \frac{Q^2-1}{4}$



Spin rotation $\sim \Phi$



Electron spin $S = \frac{1}{2}\langle\psi|\boldsymbol{\sigma}|\psi\rangle$

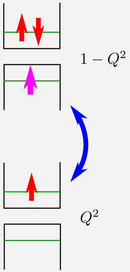
$$S_z^+ = \frac{Q^2+1}{2} S_z^- + \frac{Q^2-1}{4}$$

$$S_y^+ = Q\cos\Phi S_y^- - Q\sin\Phi S_x^-$$

$$S_x^+ = Q\cos\Phi S_x^- + Q\sin\Phi S_y^-$$

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

Yugova, MMG, Ivchenko, Efros (2009)



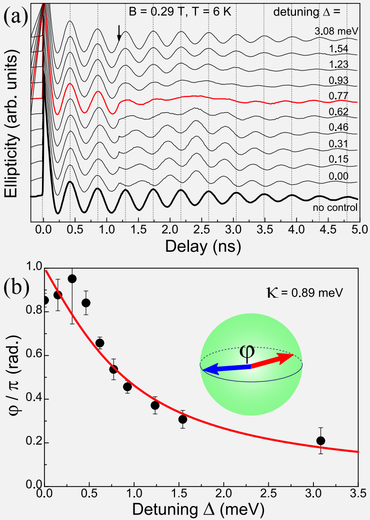
Circular pulse

- generates spin coherence (optical orientation)
- rotates spin (inverse Faraday effect)

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

Phys. Rev. B **80**, 104436 (2009)

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



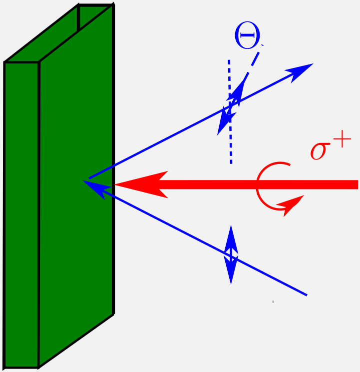
A. Greilich et al. (2009): InGaAs QDs
C. Phelps et al. (2009): CdTe QWs

Details in next talk by A. Ramsay

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

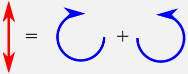
Slightly off-resonant pulse rotates spins

QW or ensemble of QDs or QWRs



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

Ellipticity and Faraday rotation

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

Resonance (trion, exciton, …)

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

$$t_\pm = 1 + r_\pm$$

Zhukov, Yakovlev, Bayer, MMG, Ivchenko, et al. (2007)

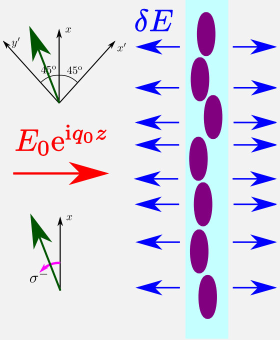
Probing the electron spins

- Faraday Effect (polarization plane rotation)

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

- 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

$$\delta \boldsymbol{E}(t) = -4\pi \left(\frac{\omega_{\mathrm{pr}}}{c}\right)^2 \frac{\mathrm{i}e^{\mathrm{i}q|z|}}{2q} N_{QD}^{2d} \boldsymbol{\Pi}(t)$$

$$\Pi_x \propto (n_e - n_{tr}) E_x^{probe}$$

$$\Pi_y \propto (S_z - J_z) E_x^{probe}$$

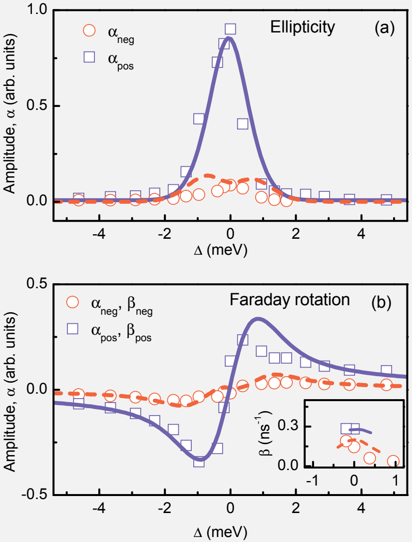
Spectral function

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

$$\int_{-\infty}^{\infty} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t' f(t) f(t') e^{\mathrm{i}\Delta(t-t')}$$

$$\Delta = \omega_{\mathrm{pr}} - \omega_0;\ \boldsymbol{E}_{\mathrm{pr}} \propto f(t) e^{-\mathrm{i}\omega t}$$

Yugova, MMG, Ivchenko, Efros (2009)



MMG, Yugova + Dortmund group (2010)

Linearly polarized pulse can readout spin polarization

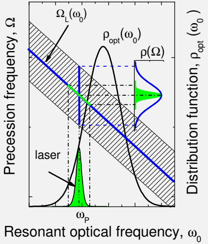
Correct description is based on reflection/transmission rather than on dielectric constant

Outline

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

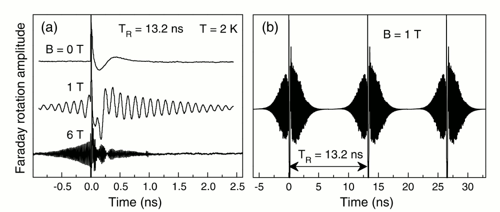


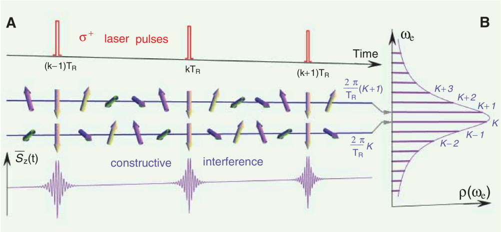
$\hbar/\tau_p \sim 1$ meV

$\Omega T_R = 2\pi N$

Constructive interference

exp.: $N \sim 10^2$





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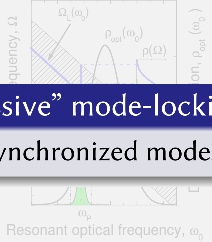
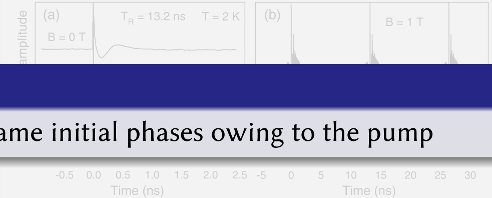




“Passive” mode-locking:

all synchronized modes have same initial phases owing to the pump

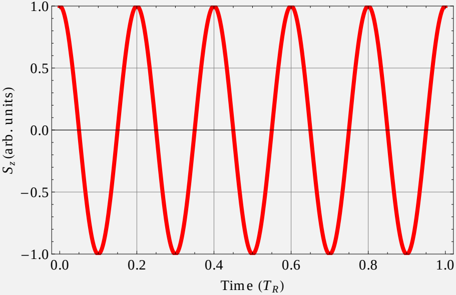
$\hbar/\tau_p \sim 1$ meV

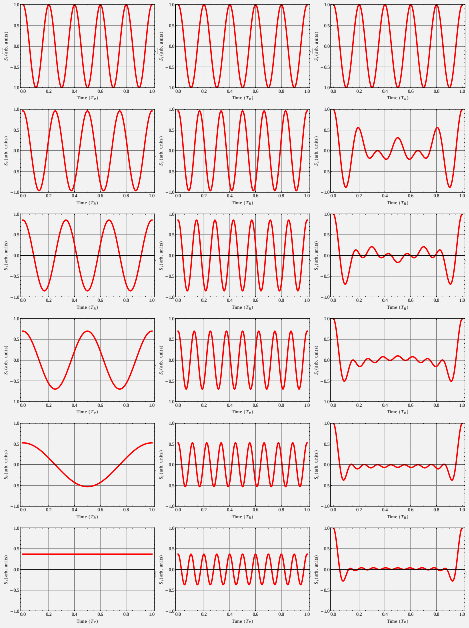
$\Omega T_R = 2\pi N$

Constructive interference

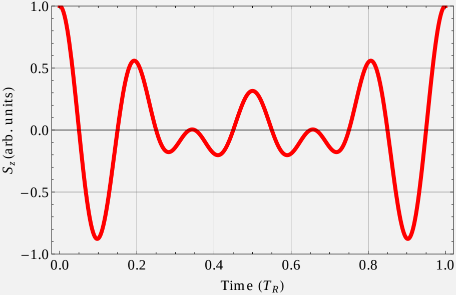
exp.: $N \sim 10^2$

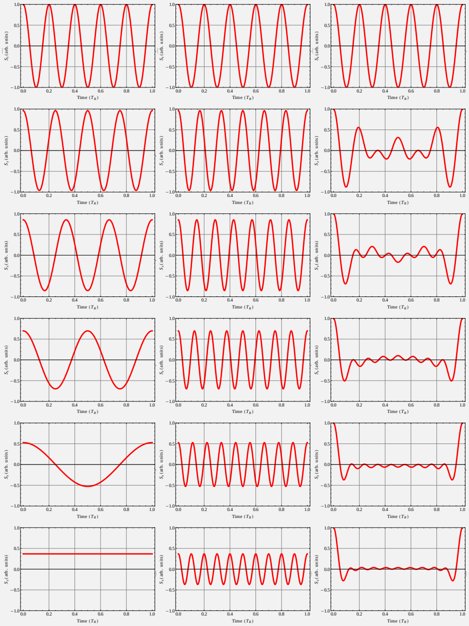
Electron spin precession modelocking – III





Electron spin precession modelocking – III





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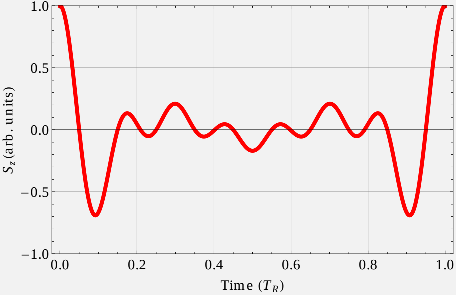

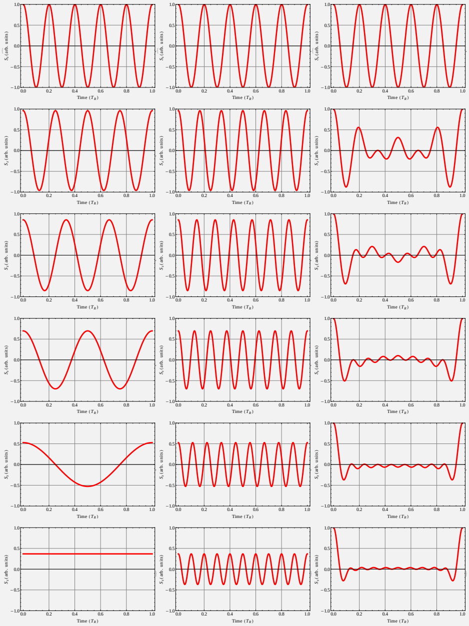

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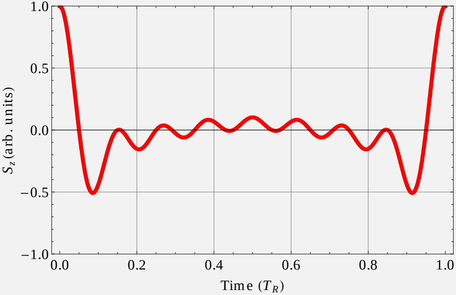
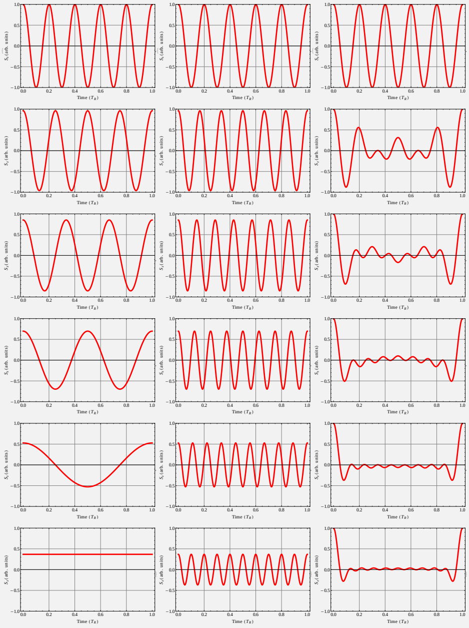
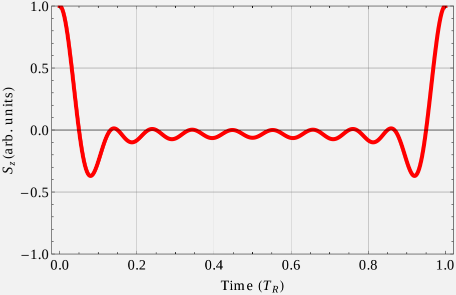
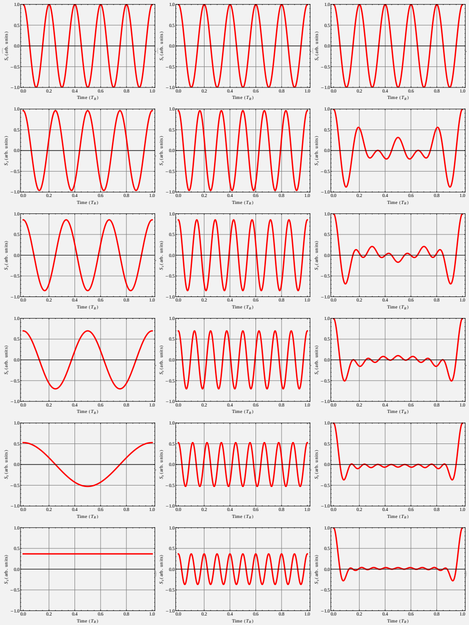





1.0
0.5
0.0
−0.5
−1.0
0.0
0.2
0.4
0.6
0.8
1.0
S_z (arb. units)
Time (T_R)



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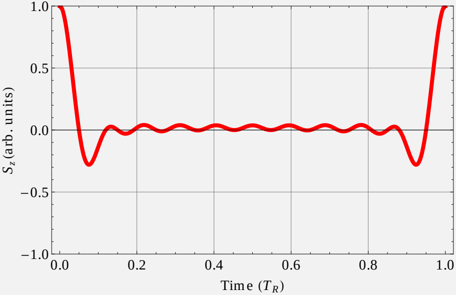
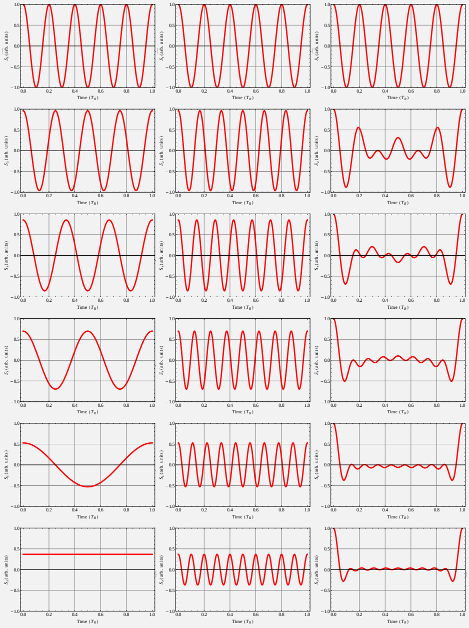
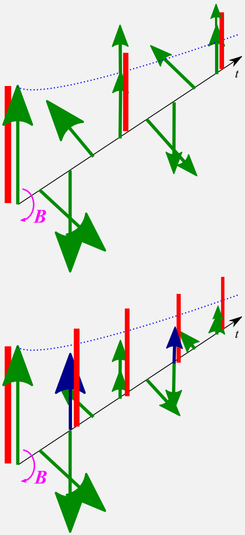




Problem

Where are other spins with frequecies $\Omega \neq \frac{2\pi N}{T_R}$?

Very high amplitude at negative delays



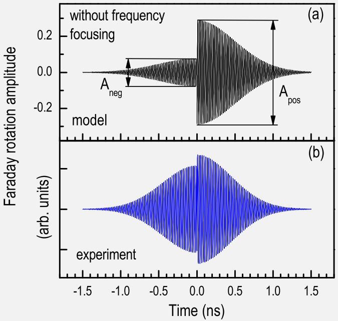
$\Omega_{\text{eff}} T_R = 2\pi$

$\Uparrow$

Nuclei-induced frequency focusing

$\Uparrow$

$\Omega_{\text{eff}} T_R = \pi$

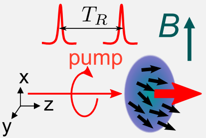


$$\boldsymbol{\Omega}_{\text{eff}} = \boldsymbol{\Omega}_{\text{L}} + \boldsymbol{\Omega}_{\text{nucl}}$$

Feedback of nuclei is necessary

Nuclei-induced frequency focusing takes place

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



Nuclear spin polarization

$$\boldsymbol{m} = \sum_i \boldsymbol{I}_i$$

Spin precession between the pulses

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

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

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

Pump pulse action: $\boldsymbol{m}^+ = \boldsymbol{m}^-$

$$S_z^+ = \frac{Q^2+1}{2} S_z^- + \frac{Q^2-1}{4}$$

$$S_y^+ = Q S_y^-, \quad S_x^+ = Q S_x^-$$

Time scales @ $B = 1$ T

1. Electron spin precession
   $$\frac{2\pi}{\Omega} \lesssim 0.1 \text{ ns}$$
2. Precession in nuclear field
   $$\frac{2\pi}{\alpha m} \sim 10 \text{ ns}$$
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)

Nuclear spin polarization

$$m = \sum I_i$$

Time scales @ $B = 1$ T

1. Electron spin precession

$\lesssim$ 0.1 ns

n nuclear field

$\sim$ 10 ns

tion period

$\sim$ 10 ns

n precession

$\lesssim$ 100 ns

n electron field

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

Spin precessi

$$\frac{dm}{dt} = [\alpha S($$

$$\frac{dS}{dt} = [\alpha m$$

$(n$

Pump pulse a

$$S_z^+ = \frac{Q^2+1}{2} S_z^- + \frac{Q^2-1}{4}$$

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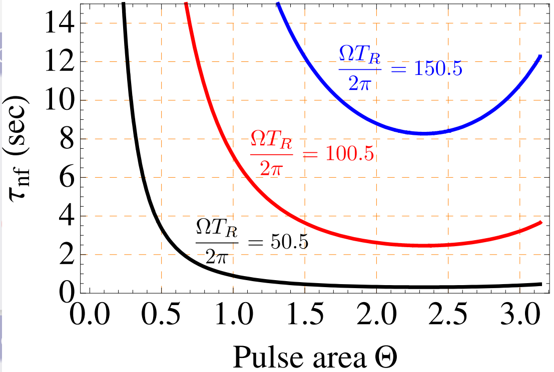

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"Active" mode-locking:

spin precession frequencies are synchronized in all dots thanks to nuclei

Pump pulse action: $\boldsymbol{m}^+ = \boldsymbol{m}^-$

$$S_z^+ = \frac{Q^2+1}{2} S_z^- + \frac{Q^2-1}{4}$$

$$S_y^+ = Q S_y^-, \quad S_x^+ = Q S_x^-$$

Time scales @ $B = 1$ T

1. Electron spin precession $\frac{2\pi}{\Omega} \lesssim 0.1$ ns
2. Precession in nuclear field 2π … ~ 10 ns
4. Nuclear spin precession $\frac{2\pi}{\omega} \lesssim 100$ ns
5. Precession in electron field $\frac{2\pi}{\alpha S} \gtrsim 10^3$ ns

MMG, Yugova, Efros (2012)

Outline

Magnetic resonance in the Faraday-rotation noise spectrum

E. B. Aleksandrov and V. S. Zapasskiĭ

(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 microwave fields to induce the resonance.

Contents lists available at ScienceDirect

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

Can spin dynamics be studied without any pump?

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

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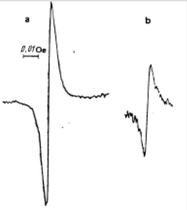

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Invited review

Semiconductor spin noise spectroscopy: Fundamentals, accomplishments, and challenges

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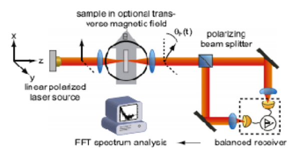
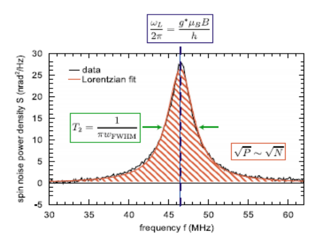
Institut für Festkörperphysik, Leibniz Universität Hannover, Appelstraße 2, D-30167 Hannover, Germany

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

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



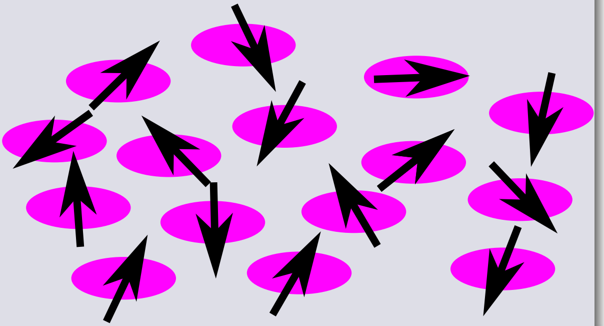


Single spin

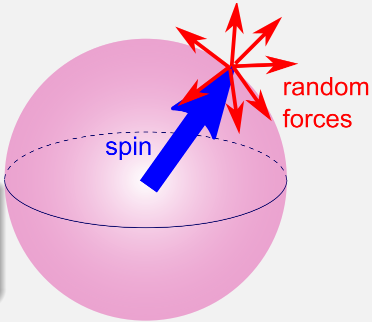
$$\langle s_x \rangle = \langle s_y \rangle = \langle s_z \rangle = 0 \quad \text{but} \quad \langle s_x^2 \rangle = \langle s_y^2 \rangle = \langle s_z^2 \rangle = \frac{1}{3} \times \frac{1}{2}\left(1 + \frac{1}{2}\right)$$

Spin ensemble

$$\sqrt{\langle S_i^2 \rangle} = \sqrt{N}\sqrt{\langle s_i^2 \rangle} = \frac{\sqrt{N}}{2}$$



$$\frac{\partial \delta \mathbf{s}(t)}{\partial t} + \frac{\delta \mathbf{s}(t)}{\tau_s} + \delta \mathbf{s}(t) \times (\mathbf{\Omega}_B + \mathbf{\Omega}_N) = \boldsymbol{\xi}(t)$$



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

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

MMG, Ivchenko (2012)

Single spin in magnetic field

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

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

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: $\left(-\mathrm{i}\omega + \frac{1}{\tau_s}\right)\boldsymbol{s} + \boldsymbol{s}\times\boldsymbol{\Omega} = \boldsymbol{\xi}\,.$

$\boldsymbol{\Omega} \parallel 3$ (axes: $1 \perp 2 \perp 3$):

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

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

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

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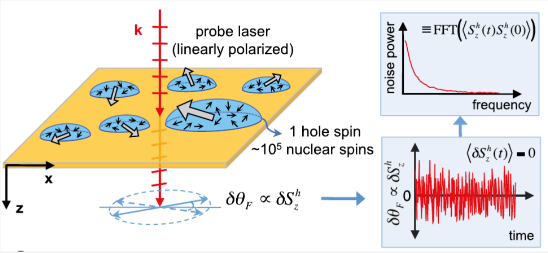
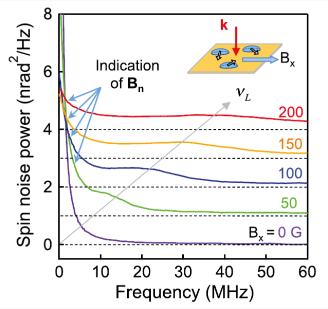
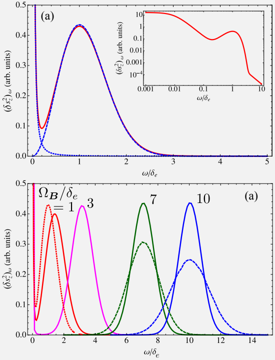
Singly charged quantum dot ensemble

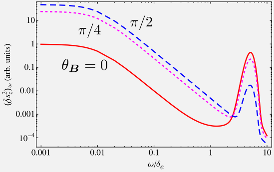




Yan Li, Sinitsyn, Smith, Reuter, Wieck, Yakovlev, Bayer, Crooker



Theory



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Outline & Co-authors

Coherent spin dynamics of electrons and excitons in nanostructures (a review), Phys. Solid State 54, 1 (2012)

Spin noise in quantum dot ensembles, arXiv:1206.3479

Spin coherence generation and detection in spherical nanocrystals, arXiv:1205.2764

Plan

1. Introduction. Questions to theory
2. Interaction of light with spins
3. Spin mode-locking effect
4. Spin dynamics in equilibrium
5. Conclusions

Co-authors

E.L. Ivchenko, D.S. Smirnov (Ioffe Institute, Russia)
I.A. Yugova (St.-Petersburg State University, Russia)
Al.L. Efros (Naval Research, USA)
E.A. Zhukov (Moscow State University, Russia)
D.R. Yakovlev, M. Bayer (TU-Dortmund, Germany)

Take home message:

Dynamics and fluctuations of electron and nuclear spins can be most conveniently studied by optical means

Thank you for attention!