

50 Years of Optical Orientation in Semiconductors

From the original discovery
to new horizons



Paris, 18-19 June 2018

Scope of the conference:

In 1968, optical spin orientation in semiconductors was discovered by Georges Lampel. This came after the seminal discoveries in gases of Alfred Kastler (optical pumping in the ground state) and of Brossel, Kastler and Bitter (double resonance method of optical orientation in the excited state and its detection by microwave spin resonance) for which Alfred Kastler was awarded the 1966 Nobel prize in Physics.

While the dynamical properties of electron and nuclear spins in gases are simple to grasp as they correspond to localized quantum states, the discovery of the optical spin orientation of itinerant conduction electrons (or sometimes itinerant valence holes) came as a big surprise. It was widely believed that the many scattering processes of conduction electrons in semiconductors (in the 10^{-12} s range) would efficiently depolarize electrons during their metastable lifetime in the 10^{-3} to 10^{-9} s, depending whether the semiconductor has an indirect or direct bandgap respectively). This is why observations of spin effects in solids had only previously been attempted (successfully) on the localized electrons of deep centers in solids as an extension of the localized electrons and nuclei in gases. The discovery of optical spin orientation in semiconductors by Georges Lampel showed both the importance of the spin orbit coupling for band electrons as spin orientation occurs from transfer of orbital orientation by light through the spin orbit coupling and that the spin degree of freedom is however decoupled enough from the orbital momentum to allow for long spin relaxation times, much longer than collision times for the orbital motion.

Optical spin orientation in semiconductors could rely on the large theoretical and experimental foundations of spins in solids, as well exemplified by the monumental books by Anatole Abragam. However, most of the previous work dealt with metals (where one can cite the electron-nuclear interactions with the Overhauser effect in particular), or with impurities in semiconductors (studies of the metal-insulator transitions, the ENDOR techniques). Some of these techniques were key in the development of the optical spin orientation in semiconductors. Both the teams of Ecole Polytechnique and Ecole Normale Supérieure benefited from the environment provided by the Abragam and Kastler (now the Kastler-Brossel) laboratories.

The field opened many areas of investigation over the years. These will be reviewed from both an historical perspective and a forward view to the future. Let's cite:

- The spin in semiconductors: spin orbit interactions- hyperfine interactions-
- Electrons nuclei interactions ... Overhauser effect etc.
- Spin-dependent transport; spintronics ; nuclear effects in the Quantum Hall effect; spin dependent recombination; spin-LED; coupling spin and quantum transport ; valleytronics
- Spin polarized photoemission
- Spin in Diluted Magnetic Semiconductors and Hybrid ferromagnetic/semiconductor structures (Spin-LED, spin photocurrent...)
- polarized electrons sources in atomic and high energy physics....

The Russian School (especially at the Ioffe Institute of the USSR Academy of Sciences in Leningrad [at the time]) and the French School have played a pioneering role in many of these areas.

The time is ripe to have a summary of the past fifty years of polarized electrons (and nuclei) in semiconductors and to have a glimpse of the next (50?) years.

Xavier Marie & Claude Weisbuch

Scientific Committee:

Xavier Marie (chair) - *LPCNO*, Toulouse

Claude Weisbuch (Honorary chair) - *PMC*, Palaiseau

- Thierry Amand - *LPCNO*, Toulouse
- Guillaume Cassabois - *L2C*, Montpellier
- Joel Cibert - *Institut Néel*, Grenoble
- Jean-Michel Gérard - *INAC, CEA-UGA*, Grenoble
- Henri Jaffrès - *CNRS/Thales*, Palaiseau
- Olivier Krebs - *C2N*, Saclay
- Michèle Leduc - *LKB*, Paris
- Henri Mariette - *Institut Néel*, Grenoble
- Daniel Paget - *PMC*, Palaiseau
- Jacques Peretti - *PMC*, Palaiseau
- Denis Scalbert - *L2C*, Montpellier

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- INSA Toulouse
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- Labex NEXT, Toulouse
- SIRTEQ, labellisé Domaine d'Intérêt Majeur par la Région Île-de-France



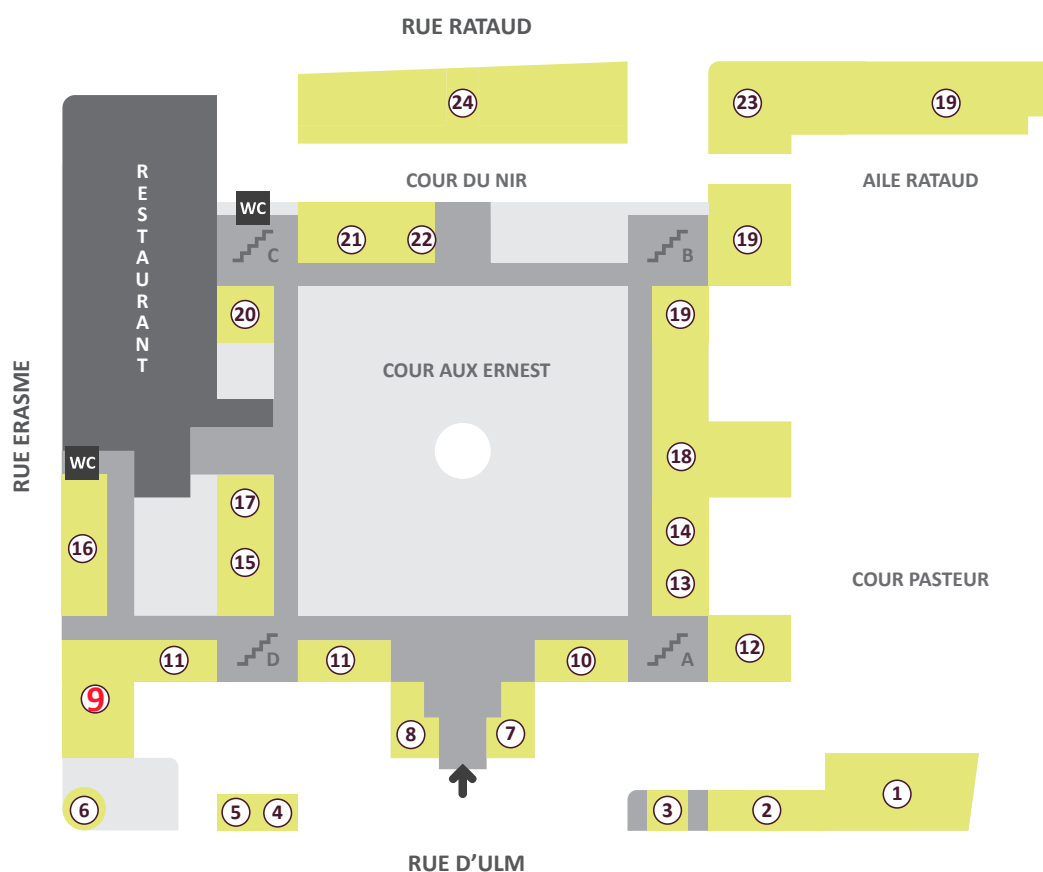
<https://www.50yearsopticalorientation.fr>

Location:

The conference will be held in Ecole Normale Supérieure, 45 rue d'Ulm, 75005 Paris - salle Dussane –

45 RUE D'ULM, 75005 PARIS

RDC



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| ⑫ Département Sciences de l'Antiquité et Études Anciennes | ㉔ NIR |

Program

June 18th

8h20 *Opening, X. Marie & C. Weisbuch*

Chair : M. Leduc

8h30-9h00 Alfred Kastler and the invention of optical pumping :
The advantages of optical pumping
C. Cohen-Tannoudji, *Laboratoire Kastler-Brossel, ENS Paris*
Applications of the method to dense systems
Franck Laloë, *Laboratoire Kastler-Brossel, ENS Paris*

9h00-9h50 Overhauser effect: Birth, Miracle, Present time
G. Lampel, *Physique de la Matière Condensée, Ecole Polytechnique*

9h50-10h20 Early studies of optical spin orientation in France,
C. Hermann, *Physique de la Matière Condensée, Ecole Polytechnique*

10h20-10h45 *Coffee break*

Chair : C. Weisbuch

10h45-11h15 Optical spin orientation in IOFFE Institute in 70-ies,
V. Safarov, *Ecole Polytechnique, Ioffe Institute*

11h15-11h55 Spins and the Quantum Hall Effect.
K. Von Klitzing, *Max Planck Institute for Solid State Research, Stuttgart*

11h55-14h30 *Lunch break and Poster session*

Chair : D. Paget

14h30-15h10 When topology and Spin-Orbit Coupling in Low Dimensions Lead to Novel
Directions in Spintronics
A. Fert, *Unité Mixte de Physique CNRS/Thales*

15h10-15h40 Applying spin polarized electron beams to image chiral magnetization textures
A.K. Schmid, *Lawrence Berkeley National Lab*

15h40-16h10 Exploring condensed matter physics with a single spin microscope,
V. Jacques, *Laboratoire Charles Coulomb, Université de Montpellier*

16h10-16h30 *Coffee break*

Chair : H. Mariette

16h30-17h00 The spin-LED – from optical orientation in II/VI semiconductors to
ferromagnetic spin injectors
G. Schmidt, *Institut für Physik, Martin-Luther-Universität, Halle-Wittenberg*

17h00-17h30 Spin/Valley dynamics in transition metal dichalcogenides,
C. Robert, *LPCNO – Université de Toulouse*

17h30-18h00 Optical orientation, spin and valley dynamics of excitons and biexcitons in
atom-thin transition metal dichalcogenides,
M. Glazov, *Ioffe Institute, St.-Petersburg*

20h00 *Dinner*

June 19th

Chair : J. Cibert

- 8h30-9h00 Spin Hall Effect
M. Dyakonov, *Laboratoire Charles Coulomb, Université Montpellier*
- 9h00-9h30 Photoinduced inverse spin-Hall effect: electrical detection of optically oriented spins in semiconductors
F. Bottegoni, *LNESS-Dipartimento di Fisica, Politecnico di Milano*
- 9h30-10h00 Electrical spin orientation
T. Dietl, *International Research Centre MagTop at the Institute of Physique, Polish Academy of Sciences*
- 10h00-10h30 Spin-dependent photogalvanic effects,
S. Ganichev, *Terahertz Center, University of Regensburg*

10h30-10h50 *Coffee break*

Chair : T. Amand

- 10h50-11h20 Optical orientation of a mesoscopic solid-state spin ensemble,
A. Imamoglu, *Institute of Quantum Electronics, ETH Zurich*
- 11h20-11h50 Cooling, relaxation and spin temperature of nuclear spin system in GaAs.
M. Vladimirova, *Laboratoire Charles Coulomb, Université de Montpellier*

11h50-14h00 *Lunch break and Poster session*

Chair : O. Krebs

- 14h00-14h30 A QD-based spin-photon interface for quantum networks,
C. Le Gall, *Cavendish Laboratory, University of Cambridge*
- 14h30-15h00 From Faraday rotation to spin-noise spectroscopy with a single spin,
L. Lanco, *Centre de Nanosciences et de Nanotechnologies –Marcoussis*
- 15h00-15h30 Optical control of the spin of individual magnetic atoms,
L. Besombes, *Institut Néel, Université Grenoble Alpes, CNRS*
- 16h00-16h30 Challenges and perspectives of optical spin orientation applied to group IV heterostructures,
F. Pezzoli, *LNESS and Università di Milano-Bicocca*

16h30-16h50 *Coffee break*

Chair : D. Scalbert

- 16h50-17h20 “Listening” to the spin noise of electrons and holes in semiconductors,
S. Crooker, *National High Magnetic Field Laboratory, Los Alamos*
- 17h20-17h50 Electron, Hole, and Nuclear Spin Dynamics in Localized-Carrier Systems
J. Hübner, *Institut für Festkörperphysik, Leibniz Universität Hannover*
- 17h50-18h20 Metal-to-insulator transition in n-GaAs revealed in coherent spin dynamics of optically oriented electrons
V. Belykh, *Experimentelle Physik 2, Technische Universität Dortmund*

18h20-18h30 *Closing remarks*

Posters

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Giant resonant ultrashort pulse polarisation rotation in a charged quantum dot – micropillar system

G. Slavcheva, M. Koleva, and A. Rastelli

Institute of Semiconductor and Solid State Physics, Johannes Kepler University Linz, King's College London

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F. Ungar, M. Cygorek, and V. M. Axt

Theoretische Physik III, Universität Bayreuth, Department of Physics, University of Ottawa

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Université de Toulouse, Institut Jean Lamour Nancy, Beijing National Laboratory of Condensed Matter Physics, Key Laboratory of Semiconductor Materials Science Beijing, Unité Mixte de Physique CNRS/Thales and Université Paris-Sud

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Angular Dependence of the Spin Photocurrent in a CoFeB/MgO/n-i-p GaAs Quantum well structure

P. Renucci, L. Zhu, W. Huang, X. Marie, Y. Liu, Y. Li, Q. Wu, Y. Zhang, B. Xu, Y. Lu, Y. Chen
Université de Toulouse, Key Laboratory of Semiconductor Materials Science, Beijing Key Laboratory of Low Dimensional Semiconductor Materials and Devices, College of Materials Science and Opto-Electronic Technology Beijing, Institut Jean Lamour Nancy, Beijing Institute of Nanoenergy and Nanosystems

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Spin precession of holes driven by Rashba spin-orbit coupling confined in the 2D group-IV semiconductor quantum well system

Tatsuki Tojo and Kyozauro Takeda

Quantum Materials Science Lab., Waseda University, Tokyo

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Nuclear polarisation in silicon quasi-one-dimensional rings

Nikolay Bagraev, Wolfgang Gehlhoff, Leonid Klyachkin and Anna Malyarenko

Ioffe Institute, Polytekhnicheskaya St. Petersburg, Technische Universität Berlin

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Olivier Krebs, Emmanuel Baudin, and Aristide Lemaître

Centre de Nanosciences et de Nanotechnologies, Marcoussis, Laboratoire Pierre Aigrain, Paris

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Sergej Markmann, Christian Reichl, Werner Wegscheider and Gian Salis

IBM Research-Zurich, ETH Zurich

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Madalina Furis, N. Rawat, K. Ngan-Hua, S. McGill

University of Vermont, Burlington ; National High Magnetic Field Laboratory, Tallahassee

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S. Yamamoto, R. Matsusaki, R. Kaji, and S. Adachi

Graduate School of Engineering, Hokkaido University

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Quantum control of valley pseudospin in monolayer WSe₂

Baoli Liu, G. Wang, X. Marie, T. Amand, C. Robert, F. Cadiz, P. Renucci, B. Urbaszek

Beijing National Laboratory for Condensed matter Physics, Université de Toulouse

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Laboratoire de Physique des Solides, Université Paris Sud; Cavendish Laboratory, Cambridge ; High Field Magnet Laboratory (HFML-EMFL), Nijmegen ; Laboratoire National des Champs Magnétiques Intenses, Toulouse.

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IPCMS, Université de Strasbourg ; Ecole Polytechnique Fédérale de Lausanne

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D. S. Smirnov

Ioffe Institute, St. Petersburg, Russia

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M.V. Dorokhin, P.B. Demina, A.V. Kudrin, E.I. Malysheva, Yu. A. Danilov, M.V. Ved'

Physical Technical Research Institute, Lobachevsky State University of Nizhni Novgorod

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Technische Universität Dortmund, ; Saint Petersburg State University ; Ioffe Institute Saint Petersburg

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TU Dortmund ; St. Petersburg State University ; Ioffe Institute, St. Petersburg ; Institute of Physics, Warsaw ; Universität Paderborn

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G. Garcia-Arellano, F. Bernardot, , G. Karczewski, C. Testelin, M. Chamarro
Institut des NanoSciences de Paris, Sorbonne Université ; Polish Academy of Science, Institut of Physics, Warsaw

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Early optical detection studies of spin orientation 1967-1977 : My almost continuous failures
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Physique de la matière condensée, Ecole Polytechnique ; Materials Department, University of California at Santa Barbara

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St. Petersburg State University ; Technische Universität Dortmund ; Ioffe Physical-Technical Institute St. Petersburg ; Technische Physik, Universität Würzburg

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Exciton, triplet and phonon interaction in two-dimensional transition metal dichalcogenides

Sabrina Ayari, Aida Hichri, Sihem Jaziri, Thierry Amand

Faculté des Sciences de Bizerte ; Faculté des Sciences de Tunis ; Université de Toulouse

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H. Carrère, A. Balocchi, S. Azaizia, F. Cadiz, S. Mazzucato, D. Lagarde, A. Arnoult, T. Amand, C. Fontaine, and X. Marie

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F. Roux, S. Holzberger, I. Favorskiy, G. Lampel, Y. Lassailly, and J. Peretti

Laboratoire de Physique de la matière condensée, Ecole Polytechnique

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Université Grenoble-Alpes, Institut NEEL ; CEA, INAC Grenoble

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Ultralong spin relaxation time of donor bound electrons in n-doped CdTe measured by spin noise spectroscopy

C. Abbas, S. Cronenberger, H. Boukari, and D. Scalbert

Laboratoire Charles Coulomb, Université de Montpellier ; Université Grenoble Alpes, Institut Neel

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T. Kuroda, T. Mano, N. Ha, K. Sakoda, B. Urbaszek

Université de Toulouse, LPCNO ; LPMC, Ecole Polytechnique ; C2N, CNRS, Université Paris-Sud et Paris-Saclay, ; National Institute for Material Science, Tsukuba

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Optical orientation of exciton states and their complexes in GaAs/AlGaAs [111] quantum dots under magnetic field

T. Amand, M. Manca, M. Vidal, L. Bouet, G. Wang, X. Marie, B. Urbaszek, M. M. Glazov, M. V. Durnev, E. L. Ivchenko, T. Mano, N. Ha, K. Sakoda, T. Kuroda

Université de Toulouse, LPCNO ; Ioffe Physical-Technical Institute, St. Petersburg ; National Institute for Material Science, Tsukuba

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Does the electron spin in a homogenous, non-magnetic semiconductor diffuse like the charge?

F. Cadiz, A. Rowe, L. Martinelli, C. P. Weber, D. Paget

Physique de la matière condensée, Ecole Polytechnique ; Santa Clara University

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Etienne Lorchat , Stéphane Berciaud, Thibault Chervy, Stefano Azzini, James A. Hutchinson, Thomas W. Ebbesen, Cyriaque Genet, Takashi Tanigushi, Kenji Watanabe

Université de Strasbourg, CNRS, IPCMS ; ISIS & icFRC, Université de Strasbourg ; National Institute for Materials Science, Tsukuba

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Kotova L.V., Platonov A.V., Kats V.N., R.André, V.P. Kochereshko

Ioffe Institute, St. Petersburg, ; University ITMO, St. Petersburg, ; Institute Neel CNRS, Grenoble; Saint Petersburg State University

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Dark-Bright exciton coupling in semiconductor Quantum dots

Savvas Germanis , Paola Atkinson and Benoît Eble

Institut des Nanosciences de Paris –CNRS, Sorbonne Université



ABSTRACTS



Overhauser effect: Birth, Miracle, Present time

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The first part of my presentation: **Birth** (early fifties), concerns dynamic nuclear polarization induced by the Overhauser effect [1,2]. I recall the strange circumstances met by A.W. Overhauser, when he gave a theoretical talk as part of his recently defended PhD work—at the 1953 spring APS meeting in Washington [3]. He was less than 28 years old (“a very green, newly minted PhD” from Berkeley,). He predicted that one could obtain a very large enhancement of the nuclear polarization in metals. In the simplest terms, Overhauser was the first to demonstrate that it is possible to line-up, or to polarize, nuclear spins, by a factor 1000 or so larger than one would expect based on then common intuitive notions. The trick that he used was to impose microwave power at the Larmor frequency of the electrons coupled to the nuclear spin system and thereby excite the electronic spins to higher energy, non-thermal equilibrium states, described by a higher electronic spin temperature T_e . Because of the coupling between the electron and the nuclear spins, as the excited electron spins try to return to their lower states to equilibrate with the thermostat temperature T , they reorient the nuclear spins to a situation described by a very low nuclear spin temperature T_n . The nuclear spins then exhibit an enhanced polarization by a factor equal to the ratio of the electronic to the nuclear magnetic moments i.e., by about a factor of $\cong 1000$. When first proposed as a contributed paper at an APS meeting in April 1953, the proposal was met with much scepticism by a formidable array of physics talent. Experimental confirmation of the Overhauser Effect was soon available via the experiments of T.R. Carver and C.P. Slichter [4]. A.W. Overhauser is quoted in a discussion at the end of [5], and a thermodynamic approach was given about one year after the APS meeting by C. Kittel [6]. To end this part, it seems that the idea of double irradiation (electronic and nuclear Larmor frequencies) appears as soon as 1950 in an article by V. Pound [7] quoted by A. Abragam [8].

The second part of my presentation: **Miracle** (1967, 1968), tells about the miracle which happened to me in 1967. At that time, I was measuring the relaxation times T_1 of ^{29}Si nuclei with various doping as a function of the applied magnetic field. This was the continuation of the work we did with D. Gémard who began these experiments in 1964 [9]. In order to increase the initial nuclear polarization, we decided to use the experimental arrangement that was used in [10] and is described in [9]. We started each measurement with an enhanced nuclear polarization obtained by the Overhauser effect [1] in a field of 50 G under r.f. irradiation at 150 MHz. This provided a starting nuclear polarization equivalent to the equilibrium polarization in a field of 100 kG.

After the defence by D. Gémard of his Thèse de 3^{ème} Cycle [9] and his departure from the laboratory B.P. Gregory in Paris, I completed this study at 77K, which became later the first part of my Thèse d’Etat [11], and contains also a spin diffusion theory

with the neutral phosphorous atoms as paramagnetic relaxation centres following and developing the ideas already present in [9].

Ionel Solomon proposed me to study the increase of the nuclear relaxation rate $(T_1)^{-1}$ due to supplementary electrons in the conduction band of a single crystal of silicon, either injected by direct current or by intense illumination of the samples with natural (unpolarised) IR light. These experiments at 300K were the subject of my Thèse de 3ème cycle [12]. They were done without and with intense light illumination, but still with r.f irradiation. While there was no observable signal without light there was a large signal under natural light irradiation This fact did not surprise me, and I attributed it to a drastic decrease of the relaxation time which increases the relative efficiency of the hyperfine relaxation mechanism proportional to the total number of electrons.

When I repeated these experiments at 77K without r.f. irradiation, but with only light illumination the measurement showed the opposite sign of the illuminated signal as compared to the usual one at thermodynamic equilibrium that one measures when the relaxation is done without light illumination. This observation let me stunned for a couple of weeks and I repeated these measurements days after days during one month or so to find possible bugs in the measurements, to convince myself that it was a real physical effect that one had to understand. I remembered then, the experiments I did a few years before, when Ionel Solomon proposed me to do some dynamic nuclear polarisation experiments at 300 K on a silicon crystal irradiated by a 150 MHz r.f. magnetic field [10]. The nuclear polarisation nuclei changes sign [11]. I realised then that the photo-created electrons were out of thermal equilibrium at least during their spin relaxation time, an experimental situation similar to the Overhauser effect [1] when the electronic polarisation is partially saturated and that of the nuclei have the opposite sign when their gyromagnetic factor is negative which is the case for ^{29}Si nuclei. It did not take me long to rush to Ecole Normale and to borrow from my friend F. Laloë a quarter wave plate and a linear polariser to perform measurements under circularly polarized illumination and get the first measurement of optical pumping in semiconductors [13].

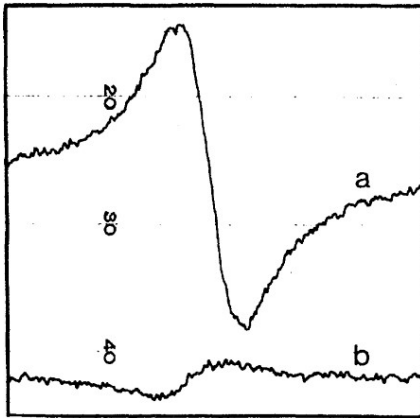


Fig.1a shows the NMR signal of ^{29}Si nuclei when a pure silicon crystal cooled to 77 K in an external field of about 1 G, is illuminated by intense circularly polarized IR light of energy close to the indirect band gap $E_G = 1.09$ eV, creating spin-polarized electrons in the conduction band.

Fig.1b shows, for comparison, the equilibrium NMR signal of the same sample at 300 K in an external field of 8 kG. This measurement shows the opposite sign of the two signals that left me stunned for a couple of weeks.

The whole story and much more can be read in an article mainly written by V.I. Safarov and C. Hermann [14]. More information about the context of the discovery of optical orientation at Ecole polytechnique can be found at [15].

In the last part of my talk: **Present time**, I shall deduce an equation between the nuclear and electronic steady-state polarizations involving only the equilibrium polarizations as

external parameters so that the final relation does not depend explicitly on the values of the Zeeman energies and the temperature and is therefore not limited to high temperatures and low fields. To begin I shall recall the hyperfine interaction between an s-type electron with a magnetic momentum μ_e , and that of a nucleus of magnetic moment μ_n :

$$\mathcal{H}_{\text{hyp}} = -(8\pi/3) \mu_0 \mu_e \mu_n \eta$$

where η is the square of the electron wave function (normalized per unit volume) at the nuclear site. It is well known that the spin magnetic moment is related to the spin angular momentum $\mathbf{S}$ by $\mu_s = \gamma_s \hbar \mathbf{S}$, so that $\gamma_s = \gamma_e$ is the gyromagnetic factor of the electrons and γ_n that of the nuclei. Then,

$$\mathcal{H}_{\text{hyp}} = A(\mathbf{s} \cdot \mathbf{I}) = (A/2)(2s_z I_z + s_+ I_- + s_- I_+)$$

where $A = -(8\pi/3) \mu_0 \hbar^2 \eta$, $\mathbf{s}$ and $\mathbf{I}$ are respectively the spins of the electron and the nucleus, s_z , I_z , are their z-components, Oz being the quantization axis along which the external magnetic field $\mathbf{B}$ is aligned. The two last terms of $\mathcal{H}_{\text{hyp}}$ induce simultaneous flip-flops between the $4I \times (2I+1)$ $(\pm, M)$ eigenstates of the s_z , I_z operators. The lowest state of an electronic spin $\mathbf{s}$ is (+), its energy being: $-\mu_e \mathbf{B} = -\gamma_e \hbar \mathbf{s} \cdot \mathbf{B}$ with $\gamma_e = -\mu_B/\hbar < 0$. (The Bohr magneton μ_B is taken to be positive). The value of the ratio $|\gamma_n/\gamma_e|$, usually smaller than 10^{-3} , imposes that the lower energy is that of the (+, -I) state for all values of the nuclear spin I. The flip flop transitions between the microscopic state occurring from (+, M) to (-, M+1) or from (-, M+1) to (+, M) with the same quantum probability $w(+, M) = w(-, M-1) = w(M)$, whatever the initial and final states.

It should be well known by the students in the audience and is of course evident for the others, that this treatment is not so for the macroscopic situations where we deal with populations of spins, in contact with a thermostat at temperature T. This is the case for the Overhauser effect and one has to deal with populations n_+ , n_- , $N(M)$. For simplicity, we restrict the following treatment to the spins $I = 1/2$ (which is the case for the ^{29}Si nuclei) and the two coupled populations will be noted $(n_+ N_-)$ and $(n_- N_+)$. The thermodynamic steady-state is described by the equation:

$$(n_+ N_-)_0 W_{+- \rightarrow -+} = (n_- N_+)_0 W_{-+ \rightarrow +-}$$

where the subscript 0 states that the populations are in contact with a thermostat at temperature T. This transforms to:

$$(n_+/n_-)_0 \times (N_-/N_+)_0 = (W_{-+ \rightarrow +-}/W_{+- \rightarrow -+})_0 = (W_{\downarrow}/W_{\uparrow})_0 = W_0(T).$$

The arrows indicate that the downward transitions starting from the less populated state are more frequent than the opposite ones, so that $W_0(T)$ is larger than 1. The detailed balance *principle* is an extension of this *theorem* to any steady-state situation indicated by the subscript s . Its validity is quasi universal and it leads to an equation where 0 is replaced by s :

$$(n_+/n_-)_s \times (N_-/N_+)_s = (W_{-+ \rightarrow +-}/W_{+- \rightarrow -+})_s = W_s(T).$$

The detailed balance *principle* expresses simply that $W(T)_s = W(T)_0$. When this equality is used one obtains a relation between the electronic and the nuclear polarization: p_0 , p_s , and P_0 , P_s . They are defined by the generic symbols:

$$\Pi_i = \{(v_+ - v_-)_i / (v_+ + v_-)_i\}$$

where the ν 's stand for the nuclear and electronic populations. Strictly speaking this definition of the polarisation does not apply in metals, due to the Pauli principle [1,2] and from now on, only the case of non-degenerate semiconductors will be discussed. By using the well-known trick and writing $\Pi_i = \text{th}(X_i)$ for the polarizations and by applying the preceding detailed balance *principle* one gets easily:

$$\text{th}(X_s - X_0) = \text{th}(\chi_s - \chi_0)$$

equivalent to:

$$P_s - P_0 = p_s - p_0$$

which is precisely the Overhauser equation, in terms of the polarisations.

One defines the saturation factor $s = (p_0 - p_s)/p_0$, the leakage factor $f = (T_1^{\text{total}})/(T_1^{\text{hyp}})$ and the enhancement of the nuclear polarisation:

$$\alpha = P_s/P_0 = 1 - sf (\gamma_e/\gamma_n).$$

When γ_e/γ_n is positive, which is the case for the ^{29}Si nuclei, the enhancement factor is negative which was essential for the discovery of optical spin orientation [12]. When the electronic orientation is non-resonantly obtained by light illumination with a steady-state polarisation p_s the saturation factor s depends on the band structure of the material, which defines the initial polarisation p_i , on the electronic spin relaxation time τ_s , on the electron lifetime τ and on the light polarisation [14,16,17]. One obtains:

$$P_s - P_0 = p_s - p_0 = (\tau_s/\tau_s + \tau)(p_i - p_0)$$

As a consequence, measuring the nuclear enhancement factor for two opposite circular polarizations should allow for the determination of p_i which, after 50 years, has still not being measured under optical pumping at the indirect band gap energy of silicon.

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Early studies of optical spin orientation in France

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In September 1969, less than two years after Georges Lampel's discovery of Optical Spin Orientation (OSO) by observing the Overhauser effect on ^{29}Si nuclei under light irradiation [1], I joined Lampel's group, Claude Weisbuch was already a member of it. I was privileged to work there for my whole research career. Here I will share my memories on the early studies (1968-1985) of OSO at the Condensed Matter Physics (PMC) laboratory of Ecole Polytechnique and more generally in France. The articles I am quoting are those that seem to me milestones.

Locations have their importance in science development: G. Lampel's discovery was made in the premises of the "historic" Ecole Polytechnique in Paris but in 1969-1974 his group was hosted in the brand-new Jussieu Sciences Faculty, inside Ecole Normale Supérieure (ENS) C. Benoît à la Guillaume's laboratory. Our ENS colleagues were experts in semiconductor optics and in particular luminescence. After R.R. Parsons', a Canadian visitor, observed the GaSb OSO on its luminescence polarization [2], members of G. Lampel's and C. Benoît à la Guillaume's groups realized similar experiments on III-V and II-VI semiconductors and alloys, with cubic zinc-blende and also hexagonal wurtzite crystal structures [3].

The OSO experimental conditions were tricky at that time: sample cooled to He (4.2K) or pumped-He (1.7K) temperature; near band-gap light excitation by a discharge lamp filtered through a monochromator (the multi-lines Kr-laser appeared in the early 1970's and the GaAs-adapted dye lasers, developed by C. Weisbuch and R. Ulbrich, in the late 1970's); lock-in detection using a home-made low-noise amplifier, an expertise of the PMC lab, of the polarization signal modulated either at excitation for zero nuclear orientation or at reception.

Combining optical and radiofrequency techniques, the optical detection of the conduction electron spin resonance on GaSb [4] was observed, providing the first measurement of its conduction electron Landé factor g^* . The strong discrepancy with the predicted value led to a reexamination of the well-known $\mathbf{k}\cdot\mathbf{p}$ perturbation theory [5], used to calculate the effective mass m^* and g^* at the bottom of the Γ_1^c conduction band. It was shown that, whereas g^* can be correctly described only through the interaction between the conduction band and the Γ_5^v valence bands, in m^* the interaction between the Γ_1^c conduction band and the upper Γ_5^c conduction band must be taken into account. Complementary g^* measurements on ternary III-V semiconductor alloys and tight-binding matrix-elements estimations supported this reexamination [6].

OSO experiments on pure III-V semiconductors lead to the observation and interpretation of exciton optical pumping [7,8]. In C. Benoit à la Guillaume's group, II-VI excitons circular and linear OSOs were observed on CdS by A. Bonnot and R. Planel [9].

In the early 1970's, G. Fishman and then D. Paget joined the Lampel group as PhD students; they respectively worked on doped GaAs polarized luminescence [10] and on polarized electrons effect on the nuclei in GaAs in a low external magnetic field [11].

Around 1972 personal relations began between the Ecole Polytechnique scientists and their colleagues working on OSO in semiconductors at Ioffe Institute in Leningrad, the theoreticians M.I. Dyakonov and V.I. Perel, G.E. Pikus and G.L. Bir, and the experimentalists groups of V.I. Safarov and B.P.Zakharchenya. They were soon followed by many scientists' visits from and to these laboratories: the optical detections of polarized electrons-nuclei interaction in GaAs [11] and of the nuclear field shift of the GaSb conduction electron spin resonance [12] were joint studies of V.I. Safarov's and G. Lampel's groups.

C. Weisbuch and G. Lampel opened the way on studies of spin-dependent recombination and OSO in semiconductors [13] by observing a 2.3 times lifetime enhancement on $\text{Ga}_{0.6}\text{Al}_{0.4}\text{As}$ luminescence when photocreated electrons and recombination centers were spin polarized; at that time the nature of the paramagnetic recombination centers had not been clarified yet.

In parallel with the Ecole Polytechnique move to Palaiseau in 1974 a new topic was introduced in the PMC lab: photoemission of electrons polarized by OSO in semiconductors. A motivation was to create highly polarized and convenient electron sources, in particular for High Energy Physics experiments. Proposals to extract intense electron beams with high spin polarization from GaAs activated to Negative Electron Affinity (NEA) were independently published by H.C. Siegmann's group at ETH Zürich [14] and by G. Lampel and C. Weisbuch at Ecole Polytechnique [15]. The first experiment was performed in Zürich; in Palaiseau this technique opened the way to about 20 years of research [16].

Using such a NEA GaAs electron source M. Eminyan and G. Lampel realized an Atomic Physics experiment: polarized electrons collide individual Zn atoms in a vapor and transfer their polarization to the atoms excited in a triplet state, an equivalent to optical pumping in the excited state by using "electronic" pumping [17].

For measuring the electron beam polarization, Lampel's group benefitted from the very efficient and compact Mott detector conceived by M. Campbell, visitor from Edinburgh.

H.-J. Drouhin's work on spin polarized NEA photoemission from GaAs and its energy distribution curves (EDC) and polarized EDCs provided a very detailed spectroscopy of this semiconductor band structure [18, 19], beyond the effective mass approximation. With R. Houdré's study of photoemission from $\text{GaAs}_{1-x}\text{Al}_x\text{As}$ quantum wells [20] began the quest for electron sources with a spin polarization higher than the bulk limit of 50%.

But this is another story, beyond the "early studies of optical orientation"...

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OPTICAL SPIN ORIENTATION IN IOFFE INSTITUT IN 70-IES

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In the 70-ies Ecole Polytechnique and Ioffe Institute groups dominated researches on optical spin orientation in semiconductors. As to Ioffe Institute there were two main reasons for that. The first was a big number of groups working on different aspects of semiconductor physics, as many of them become interested in the new phenomenon discovered by G.Lampel. The second was a very strong implication of theorists, who worked in tight contact and collaboration with experimental groups. Many important results have been obtained on spin relaxation mechanisms, electron-nuclear interactions, orientation and alignment of free and bound excitons, momentum alignment of photoelectrons and spin-dependent transport. These results determined in big part the future directions of research in semiconductor spintronics.

The first Ioffe Institut paper on optical spin orientation was published by A.I.Ekimov and V.I.Safarov in 1970 [1]. The first theoretical paper appeared very soon in 1971 [2]. In that paper M.Dyakonov and V.I.Perel calculated the degree of polarization of electrons as a function of the energy of the exciting photons which create “hot” electrons. They showed that the main mechanism of spin relaxation of these “hot” electrons during their thermalization to the bottom of conduction band is related to k^3 splitting of conduction band. The mechanism (later named Dyakonov-Perel mechanism) can be suppressed by very rapid momentum relaxation in highly doped materials. The experiments on GaAs and GaAlAs by Fleisher’s [3] and Safarov’s [4] groups confirmed these predictions.

The measurements of degree of polarization of luminescence and its depolarization in transverse magnetic field (Hanle effect) offer a means to measure the lifetime and spin relaxation times of photoelectrons [5]. In p-type materials the electron spin relaxation time is dominated by the exchange interaction with holes, theoretically examined by G.Bir, A.Aronov and G.E.Pikus [6] and experimentally studied by A.N.Titkov [7].

A new research direction appeared with the observation by Safarov’s group of quite long (minutes) variations of luminescence polarization when the polarization of exciting light is reversed from σ^+ to σ^- (and vice versa) [8]. They immediately suggested that this is a signature of nuclear polarization effect, all nuclei of host lattice in GaAs and GaAlAs having non-zero magnetic moments. The confirmation came from NMR experiments. They were done in extremely simple conditions: a primitive coil directly related to the RF generator, the frequency being swept by the rotation of the generator’s knob with a simple mechanical motor.

M.Dyakonov and V.Perel [9] indicated that hyperfine electron-nuclear interaction can be represented as interaction of electronic spin with an effective magnetic field $\mathbf{B}_N$ proportional to polarization of all nuclei seen by electrons. The nuclear field $\mathbf{B}_N$ can

reach a very high value ~ 2 Tesla for totally polarized nuclei in GaAs. The electron sees a total field which is a vector sum of external and nuclear field. Then, $\mathbf{B}_N$ can enhance or attenuate the effect of an external magnetic field depending on the orientation of $\mathbf{B}_N$, which is determined by the circular polarization of the light. Under NMR conditions, one can manipulate the mean nuclear spin orientation (i.e. nuclear field) and put it in any orientation relative to the external magnetic field. That explains the shape of optically detected NMR line. The same is valid for nuclear quadrupole resonance, which was optically detected in GaAlAs mixed crystals [10]. The nuclear field strongly modifies the shapes of Hanle curves in oblique magnetic field [11]. This effect represents a general way for the optical observation of nuclear spin polarization.

All experiments have shown that significant nuclear field arises when an external magnetic field exceeds the local field produced at nuclear sites due to the dipole-dipole interaction with other nuclei of the lattice. However we should not neglect an effective magnetic field $\mathbf{B}_e$ (~ 1 Gauss) produced at nuclear sites by polarized electrons (known as the Knight shift in NMR). Thus, even in the absence of external magnetic field a sizeable nuclear polarization and nuclear field can arise. It leads to some peculiar features in the Hanle curves in vicinity of zero external field which was studied in joint work of Ecole Polytechnique and Ioffe Institute groups [12]. Thereby the electron and nuclear spins systems turn out to be tightly coupled: electronic spin system transfers its polarization to nuclear system which in its turn through the nuclear field influences the electronic spin system. It is not surprising that the total electron-nuclear spin system in some conditions become unstable and exhibits hysteresis and even oscillations [13].

The optical orientation of free excitons represented another interesting direction of research. The angular momentum of an exciton is a sum of hole and electron moments and has an integer value. Consequently one can observe not only optical orientation but also optical alignment of exciton angular momentum. In the last case excitation by linearly polarized light induces linear polarization of luminescence. In 1972 G.Bir and E.Pikus developed the theory of these effects for uniaxial crystals [14]. They also predicted transformation of circular polarization into linear one and vice versa in transverse magnetic field. Razbirin's group in Ioffe Institute undertook a detailed study of these effects on free and bound excitons in GaSe crystals at resonant excitation [15, 16]. They observed a very strong orientation and alignment (up to 95%) under resonant excitation. The study in magnetic field confirmed theoretical predictions and gave information on free exciton kinetics.

Mirlin's group studied the "hot" luminescence – recombination of non-thermalized electrons excited high in the conduction band by absorption of photons with an energy much greater than E_g [17]. They observed linear polarization of this "hot" luminescence under excitation with linearly polarized light. M.Dyakonov and V.I.Perel [18] showed that this is due to momentum alignment: absorption of polarized photon produces photoelectrons with anisotropic distribution of mechanical momentum $\mathbf{k}$. In a magnetic field, the Lorentz forces induce the cyclotron motion of electrons and produce some kind of Hanle effect which gives information on "hot" electrons kinetics.

The above-mentioned experiments show that there exists coupling between spins and mechanical momentum of electrons that could lead to spin dependent transport. Direct manifestation of this interaction is so-called the spin-Hall effect, predicted in 1971 by M.Dyakonov and V.Perel [19]. They also predicted an Inverse Spin Hall Effect, which

was experimentally observed in joint work of Rogachev's and Fleisher's groups [20]. E.Ivchenko and G.Pikus showed that in girotoropic crystals the absorption of circularly polarized photons must produce an emf dependent on light polarization [21]. A.Rogachev group observed this polarization dependent photo-galvanic effect in tellurium crystals [22].

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Spins and the Quantum Hall Effect

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The unexpected discovery (more than 35 years ago) of an ESR signal in GaAs/AlGaAs heterostructures connected with an Overhauser shift of more than 0.2 Tesla [1] was the starting point of many investigations of spin phenomena in low dimensional electron systems and the interaction between electron and nuclear spins.

The talk summarizes different aspects of spin phenomena in 2-dimensional systems starting with a historical review and finishing with investigations of spin textures in quantum Hall system observed for the striped phase of quantum Hall systems [2] or at crossings of Landau levels with different spin orientation [3,4].

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When topology and Spin-Orbit Coupling in Low Dimensions Lead to Novel Directions in Spintronics

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The talk will focus on the recent directions in spintronics exploiting topological effects and spin-orbit coupling in low dimensional systems (mainly 2D)¹. I will describe the potential of these new directions for different types of applications.

- 1) Spintronics with surface/interface states of topological insulators (TI) and similar 2D states: the locking between the spin and momentum degrees of freedom at surface/interfaces of TI, Rashba interfaces and some interfaces between oxides lead to very efficient conversions between spin and charge currents by the Edelstein and Inverse Edelstein Effects. These spin/charge conversions are much more efficient than those obtained with 3D materials in today spintronic devices as, for example, SOT-RAMs based on the Spin Hall Effect of 3D heavy metals. Promising similar effects are obtained in 2D materials with large SOC as, for example, ultra-thin films of Transition Metal Dichalcogenides (TMD) or heterostructures associating several types of 2D materials.
- 2) Magnetic skyrmions: the chiral spin interactions (Dzyaloshinskii-Moriya interactions) induced by SOC and inversion symmetry breaking at the interface of a magnetic film with a heavy metal can be used to create skyrmions, nanoscale spin whirls that are stabilized by their topology. These skyrmions can be as small as a few nm and behave as small objects that can be displaced with large velocities by electrical currents. The recent results in several groups on the electrical creation, manipulation and detection of skyrmions at room temperature in magnetic multilayers represent real advances on the route to different types of applications.

¹ Example of recent review: A. Soumyanarayanan, N. Reyren, A. Fert and C. Panagopoulos, *Nature* **539**, 509 (2016).



Applying spin polarized electron beams to image chiral magnetization textures

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Spin polarized electron beams can be applied in low energy electron microscopy to map the orientation of the 3D magnetization vector at high spatial and angular resolution. This application offers uniquely powerful opportunities to investigate the rich physics of chiral magnetism in films and multilayers. This magnetic chirality is fundamentally interesting holds potential for logic and memory applications [1,2]. Using spin-polarized low-energy electron microscopy (SPLEEM), we recently observed chiral domain walls in thin films [3]. We developed a way to tailor and amplify the Dzyaloshinskii-Moriya interaction, which drives the chirality, by interface engineering [4]. We also demonstrate an experimental approach to stabilize skyrmions in magnetic multilayers without external magnetic field, by exchange-coupling homochiral magnetic films to buried layers with perpendicular magnetization [5]; and we demonstrate that chiral spin textures are induced at graphene/ferromagnetic metal interfaces, via a new type of Dzyaloshinskii-Moriya interaction due to a Rashba effect [6]. This work was done in collaboration with T.P. Ma, A.T. N'diaye, S.P. Kang, H.Y. Kwon, C. Won, Z.Q. Qiu, Y.Z. Wu, A. Mascaraque, H. Yang, A.A.C. Cotta, S.A. Nikolaev, E.A. Soares, W.A.A. Macedo, K. Liu, A. Fert and M. Chshiev.

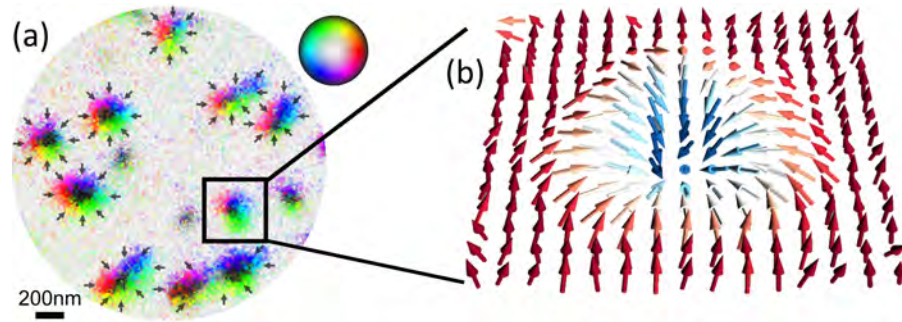


Figure. 1 (a) SPLEEM image of skyrmions in magnetic multilayer designed to stabilize ambient-temperature skyrmion phase [5]. Magnetization vector orientation rendered in color according to color-wheel. (b) Single skyrmion image, 3D vector array shows magnetization at image pixels.

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Exploring condensed matter physics with a single spin microscope

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In the past years, it was realized that the experimental methods allowing for the detection of single spins in the solid-state, which were initially developed for quantum information science, open new avenues for high sensitivity magnetometry at the nanoscale. In that spirit, it was recently proposed to use the electronic spin of a single nitrogen-vacancy (NV) defect in diamond as an atomic-sized magnetic field sensor [1-3]. This approach promises significant advances in magnetic imaging since it provides non-invasive, quantitative and vectorial magnetic field measurements, with an unprecedented combination of spatial resolution and magnetic sensitivity under ambient conditions.

In this talk, I will illustrate how scanning-NV magnetometry can be used as a powerful tool for exploring condensed-matter physics, focusing on chiral spin textures in ultrathin ferromagnets [4] and antiferromagnetic order in multiferroic materials [5].

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The spin-LED – from optical orientation in II/VI semiconductors to ferromagnetic spin injectors

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The electrical injection of spin polarized currents into semiconductors has been the subject of numerous experiments over more than a decade. Among others the idea of a spin transistor similar to the one proposed by Data and Das in 1990 [1] was the driving force to develop contacts suitable to inject spin polarized carriers from ferromagnets into various semiconducting materials. A prototypical device for the detection of spin injection is the so called spin LED in which the injection of spin polarized carriers leads to the emission of circularly polarized light. In contrast to magnetoresistive devices which yield a signal proportional to spin accumulation the spin LED is sensitive to the spin polarization which is a huge advantage in case of low injection efficiencies.

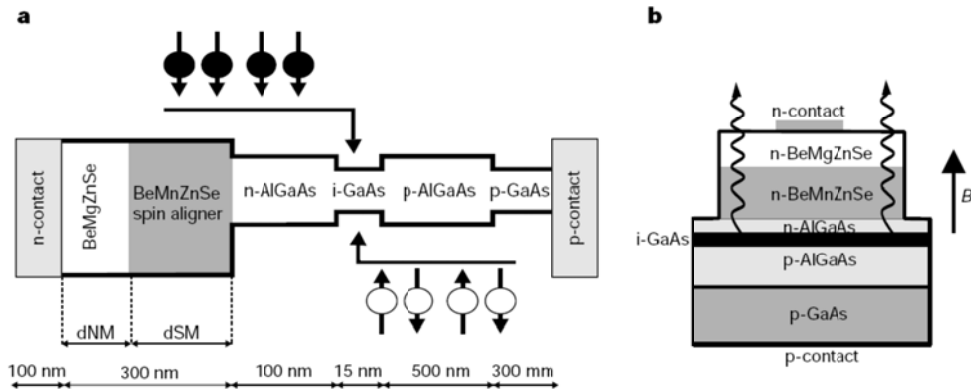


Figure. 1 Device geometry and electric band structure of a spin LED [3]. a, Schematic band structure of the spin-aligner light-emitting diode. Spin-polarized electrons are injected from the left into the active GaAs layer, unpolarized holes from the right. b, Side view of the device showing the direction of the magnetic field and the emitted light.

The development of the spin-LED started with devices in which optically excited carriers diffused from a semimagnetic spin aligner into a semiconductor LED [2]. Later on this was followed by the first real spin-LED in which a dilute magnetic semiconductor was used as an electrode on the n-side of a GaAs based p/n-light emitting diode [3]. Along with these experiments came the description of the spin injection problem using the so-called conductance mismatch [4]. This formalism explained why at that time all attempts to use ferromagnetic metals for spin injection into semiconductors had failed.

Once a better understanding was established soon more experiments followed in which tunnel barriers were used to allow spin injection from metallic ferromagnets into LEDs,

all based on tunnel barriers, either using Schottky contacts or oxide-based tunnel barriers. An important step in this development was the introduction of the so called oblique Hanle effect as a detection scheme for the investigation of spin injection from ferromagnetic contacts magnetized in-plane which is not trivial because of the selection rules in semiconductor LEDs.

Nowadays, spin injection from ferromagnetic metal contacts into LEDs is well established and the efficiencies which can be achieved are quite high.

This presentation will outline the different steps and milestones of the development and show why a very detailed development was necessary although the first injection experiments in 1999 yielded an efficiency close to the possible maximum.

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Spin/Valley dynamics in transition metal dichalcogenides

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Transition Metal Dichalcogenides (TMD) monolayers have emerged as exciting 2D materials as they present strong light-matter interaction governed by robust excitons and interesting spin/valley selective optical selections rules. These rules mandate that the K or K' valleys in momentum space can be selectively populated and probed using polarized light. However, the development of applications based on spin-valley properties of excitons is expected to be very challenging due to the extremely short lifetime of bright states (~ 100 fs - 1 ps) [1].

In this work, we will focus on long-lived dark excitons and discuss about possible valley information encoding. We will present the fine structure of dark excitons and show that short range Coulomb interaction lift the degeneracy between a truly dark state and a gray state which is partially coupled to light in z polarization[2-3] (see Figure 1). We will show that these states can be coupled with longitudinal magnetic field.

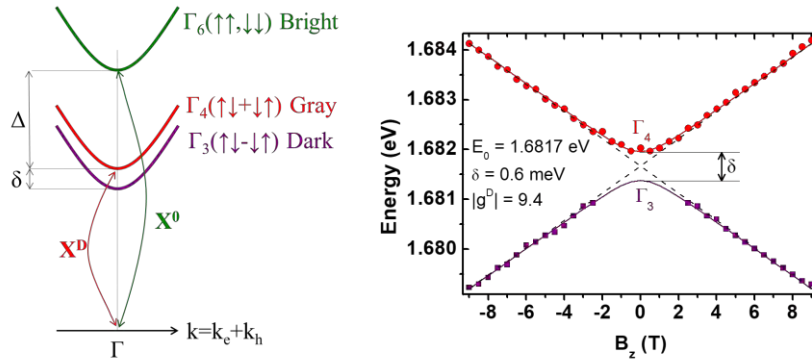


Figure. 1 Sketch of the exciton fine structure at the Γ point of the exciton Brillouin zone. $\uparrow\downarrow + \downarrow\uparrow$ and $\uparrow\downarrow - \downarrow\uparrow$ refer to the coherent superposition of the *intra-valley* conduction-valence pairs. Energy of gray and dark exciton states as a function of longitudinal magnetic field B_z .

Secondly, we measure the spin/valley dynamics of resident electrons and holes in single charge-tunable monolayers of WSe₂ (see Figure 2). In the n-type regime, we observe long (70 ns) polarization relaxation of electrons that is sensitive to transverse magnetic fields B_y , indicating spin relaxation. In marked contrast, extraordinarily long (2 μ s) polarization relaxation of holes is revealed in the p-type regime, that is unaffected by B_y , directly confirming long standing expectations of strong spin-valley locking of holes in the valence band of monolayer TMDs [4].

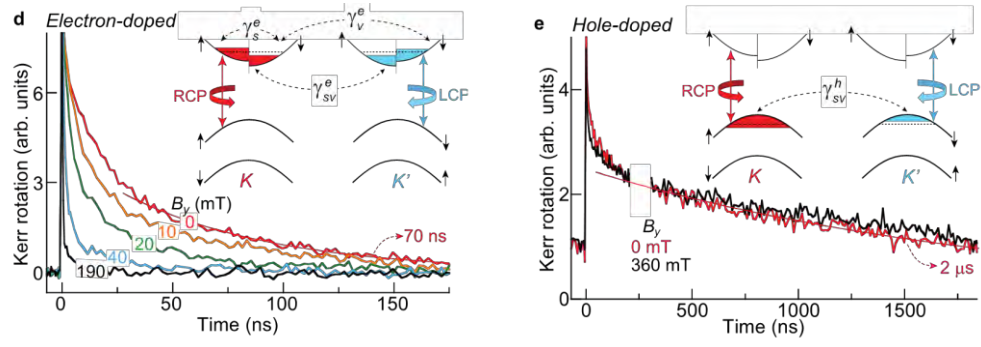


Figure. 2 Time resolved Kerr rotation signal in the electron doped regime and the hole doped regime. Long lifetime of 2 μ s is observed in the hole doped regime due to spin-valley locking in the valence band.

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Optical orientation, spin and valley dynamics of excitons and biexcitons in atom-thin transition metal dichalcogenides

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Monolayers of transition metal dichalcogenides (TMD MLs) attract increasing interest nowadays due to their fascinating optical properties controlled by excitons, Coulomb-correlated electron-hole pairs [1]. Due to relatively large effective masses of the charge carriers and rather weak screening of the Coulomb interaction excitons possess high binding energies up to several hundreds of meV and dominate the optical spectra up to the room temperature. By contrast to conventional III-V and II-VI materials based quantum well heterostructures, the excitons in TMD MLs possess, in addition to spin, the valley degree of freedom, because the direct band gap in transition metal dichalcogenides is realized at the valleys K_+ and K_- related by the time-reversal symmetry.

Here we review recent progress in understanding of spin and valley dynamics of excitons in TMD MLs. First, we discuss the fine structure of excitonic states related to the presence of the valley and spin degrees of freedom. The impact of the conduction and valence band spin splittings at the K-points of the Brillouin zone on the exciton fine structure is analyzed. We identify optically active (bright) and forbidden (dark) transitions. We show that some of spin-forbidden states can be activated in the polarization perpendicular to the monolayer due to the spin-orbit mixing of bands or by an external in-plane magnetic field [2].

We demonstrate that a high degree of optical orientation and exciton alignment can be achieved in TMD MLs under the excitation by circularly and linearly polarized light, respectively. We show that the spin/valley depolarization of excitons is mainly governed by the long-range exchange interaction, which is strongly enhanced in atom-thin crystals compared to the conventional two-dimensional semiconductors based on quantum wells [3,4]. The exciton spin decoherence detected by the time-resolved Kerr rotation is discussed and the temperature dependence of the spin/valley relaxation time is analyzed [5].

We briefly discuss magneto-optical properties of excitons and biexcitons in TMD MLs. The origin of large Lande factor for bright excitons is discussed [6]. We report on the “inverted” circular polarization of biexciton emission, where the sign of magneto-induced circular polarization is incompatible with the order of Zeeman-split levels. We demonstrate that the effect originates from the non-trivial fine structure of biexcitonic states [7].

Finally, we address the optical selection rules for excitonic transitions in the non-linear optical processes of the two- and three-photon absorption. We discuss the giant linear-

circular dichroism at the multiphoton absorption in TMD MLs and the effect of second harmonic generation on excitons [8].

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Spin Hall Effect

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The spin Hall effect (SHE, direct and inverse) was predicted by Dyakonov and Perel [1,2] in 1971. The SHE is due to spin-orbit interaction and consists in the generation of a transverse spin current by a normal electric current. This leads to spin accumulation at the lateral boundaries of a current-carrying sample. The accumulation is limited by spin relaxation processes, and can be influenced by external magnetic field.

The spin current density is described by a *tensor* q_{ij} where the first index shows the direction of flow, and the second one says what component of spin is flowing. Thus q_{xz} is the density of flow in the x -direction of spins polarized along z . Let $\mathbf{q} = \mathbf{j}/e$ be the vector of charge flow density (e is the elementary charge). In the *absence* of spin-orbit interaction one has the drift/diffusion expressions for the two currents:

$$q_i^{(0)} = -\mu n E_i - D \partial n / \partial x_i, \quad (1)$$

$$q_{ij}^{(0)} = -\mu E_i S_j - D \partial S_j / \partial x_i, \quad (2)$$

where $\mathbf{j}$ is the current density, μ and D are the mobility and the diffusion coefficient, $\mathbf{E}$ is the electric field, n is the electron concentration, and $\mathbf{S}$ is the spin density vector.

Eq. (1) is the standard drift-diffusion expression for the electron flow. Eq. (2) describes the spin current of spin-polarized electrons, which may exist even in the absence of spin-orbit interaction, simply because spins are carried by the electron flow. If there are other sources for currents, like for example a temperature gradient, the corresponding terms should be included in Eqs. (1) and (2).

Spin-orbit interaction *couple*s the two currents, and this may be presented in the following simple form [3] (summation on repeating indices is assumed):

$$q_i = q_i^{(0)} + \gamma \epsilon_{ijk} q_{jk}^{(0)}, \quad (3)$$

$$q_{ij} = q_{ij}^{(0)} - \gamma \epsilon_{ijk} q_k^{(0)}. \quad (4)$$

Here γ is the *coupling coefficient* proportional to spin-orbit interaction.

These equations are complemented by the continuity equation for the spin density $\mathbf{S}$:

$$\partial S_i / \partial t + \partial q_{ij} / \partial x_j + S_i / \tau_s = 0, \quad (5)$$

where τ_s is the spin relaxation time. The current-induced spin accumulation at the sample boundaries within the layer defined by the spin diffusion length of $L_s = (D\tau_s)^{1/2}$ directly follow from Eqs. (1-5) in the stationary regime.

A way to observe the so-called "inverse spin Hall effect" (appearance of voltage induced by spin current) in an optical spin orientation setup was proposed by Averkiev and Dyakonov [4]. The corresponding experiment was done in 1984 by Bakun et al [5].

As to the direct spin Hall effect, it was observed only 33 years after the original prediction [6,7] by optical measurements of the current induced spin accumulation at the edges of a semiconductor sample. These experiments have triggered the general interest in this phenomenon.

Since then the spin Hall effect, both in semiconductors and in metals, was studied in numerous work. It was observed not only at cryogenic, but also at room temperature [8]. Many studies were devoted to SHE in heavy metals, where the spin-orbit interaction is strongly enhanced, such as Platinum, and also in structures involving interfaces between normal and ferromagnetic metals, see Hoffmann's review [9]. Such studies are inspired by the hopes that a spin current injected in a ferromagnet will efficiently switch magnetic domains and thus improve magnetic data storage.

Among other related topics are:

Spin Hall magnetoresistance [2, 3]. As seen from Eqs. (3, 4), the spin current induced by the normal electric current in turn provides a correction $\sim \gamma^2$ to the primary electric current. This correction to current is *positive*, i.e. it leads to a slight *decrease* of the sample resistance. By applying a magnetic field, we can destroy spin accumulation and thus *increase* the sample resistance. This effect was clearly observed in thin Pt films by Vélez et al [10]. Earlier, a similar effect was observed in structures involving ferromagnets by Nakayama et al [11].

Swapping spin currents. This effect, related to SHE, was predicted by Lifshits and Dyakonov [12]. It consists in a transformation of spin currents in which the spin direction and the direction of flow are interchanged (swapped): $q_{ij} \rightarrow q_{ji}$. So far, this effect was not observed experimentally. Theoretically, it was further discussed in Refs. [13, 14].

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Photoinduced inverse spin-Hall effect: electrical detection of optically oriented spins in semiconductors

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Optical spin orientation in semiconductors is typically observed by its reversed process, the photoluminescence (PL) due to band-to-band recombination of spin-polarized electrons. PL is effective in characterizing semiconductors featuring a carrier lifetime τ_e shorter than the spin lifetime τ_s but, unfortunately, in the case of the technologically relevant semiconductors Si and Ge, $\tau_e \gg \tau_s$ at room temperature and clear indications of the optical recombination of spin polarized electrons are still missing, even in the case of Si, which is the first semiconductor where optical orientation was indirectly observed by George Lampel back in 1968 [1]. Inspired by the work of Ando. et al [2], we have combined optical spin orientation with electrical spin detection based on the spin-to-charge conversion induced by the inverse spin-Hall effect (ISHE) [3].

Photoinduced ISHE allows for the observation of spin currents with a high sensitivity making it possible to observe, for the first time, optical orientation at the indirect gap of silicon [4]. Moreover, by means of a relatively simple drift-diffusion model the spin lifetime τ_s can be estimated from the photon energy dependence of the ISHE signal in several direct and indirect gap semiconductors such as Si, Ge and GaAs [4, 5]. As compared to previous reports [2], we have also extended the potentiality of the photoinduced ISHE measurements by locally manipulating the light circular polarization by means of metallic micro-antennae. In this respect, we have designed a new opto-spintronic device, which is the spin equivalent of a photovoltaic cell [6]. In this case it is possible to locally generate spin-polarized electrons, with in-plane spin orientation, thus allowing for the accurate determination of the spin diffusion length in Ge [7].

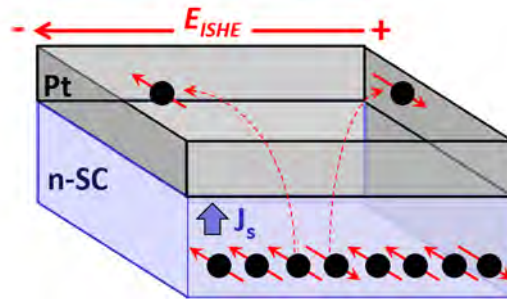


Fig.1. Geometry of the inverse spin-Hall effect process in a Pt/n-doped semiconductor (SC) junction.

The working principle of photoinduced ISHE measurements is sketched in Fig. 1. A circularly polarized laser beam generates spin-polarized electron within the

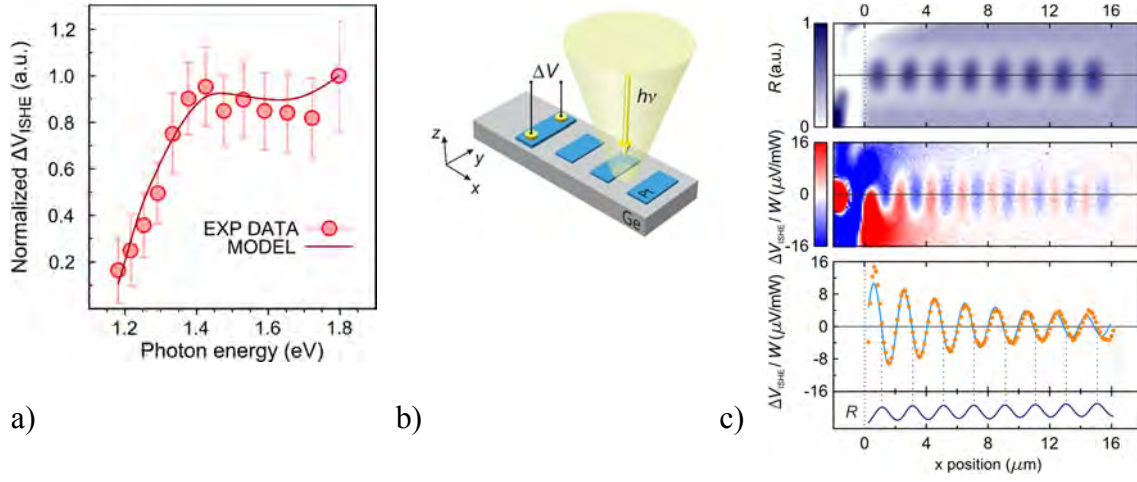


Fig.2. Photon energy dependence of the photoinduced ISHE signal (red dots) in a Pt/Si junction. The continuous line represents a fit of the experimental data based on the Spicer model. b) Geometry of the optical spin injection from metallic micro-antennae. c) ISHE signal profile as a function of the distance between the generation antenna and the Pt detector.

semiconductor (SC). Electrons then diffuse through the Schottky barrier formed by a Pt layer evaporated on the SC. The spin current is converted into a charge current within the Pt layer originating an electromotive field E_{ISHE} which is detected by means of a lock-in amplifier.

Fig. 2a shows the photon-energy dependence of E_{ISHE} in the case of Si. Interestingly the maximum ISHE signal is not obtained at the threshold of the optical absorption, where the initial electron spin polarization is expected to be maximum [8], but at slightly higher energy. However, by considering the interplay between the energy dependence of the optical orientation, optical absorption and the role played by charge and spin diffusion it is possible to nicely fit the experimental data (dashed line in Fig 2a) and estimate the spin lifetime in Si at room temperature [4].

By micro-fabricating a set of metallic antennae (see Fig. 2b) it is possible to inject optically oriented spins with a spatial resolution of $\approx 1 \mu\text{m}$, in a confocal-microscope set it is then possible to generate spins at variable difference from a micro ISHE detector thus spatially visualizing the spin diffusion length in Ge.

In conclusion the combination of optical orientation and ISHE allows for the investigation of spin phenomena in indirect gap semiconductors at temperatures typically outside the reach of more conventional investigation techniques. The application of this approach to Si and Ge opens the way to novel devices combining integrated photonics and spintronics.

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Electrical spin orientation

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Nuclear spin orientation by light pioneered by Lampel [1] has been followed by a series of time-resolved experiments revealing light-induced organization of magnetic moments in dilute magnetic semiconductors [2]. These works have culminated by in a recent demonstration of low-power magnetization manipulation by light at the time scale below 20 ps [3], the achievement paving the way for ultrafast magnetic recording.

In the talk I will review the current status of magnetization orientation by electric fields and currents. In particular, after a brief mentioning of early works on the effects of an electric field on ferromagnetic ordering [4-6], I will present more recent results obtained for a dilute magnetic insulator [7] and an antiferromagnet [8,9].

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Spin-dependent photogalvanic effects

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The paper overviews theoretical and experimental studies on spin-related photogalvanic effects, for recent review see [1]. A short phenomenological introduction is followed by the discussion of the circular photogalvanic effect (CPGE) and the direct (inverse) spin-galvanic effect. Then we consider the pure spin currents and magneto-gyrotropic photocurrents. Finally, we briefly discuss the spin-dependent photogalvanics in topological insulators. Various aspects of these phenomena will be addressed including experimental achievements, phenomenological description, models visualizing physics of nonlinear responses, and microscopic theory of individual effects.

Circular photogalvanic effect and the spin-galvanic have in common, that the current flow is driven by an asymmetric distribution of carriers in k -space in gyrotropic systems with lifted spin degeneracy due to k -linear terms in the Hamiltonian. CPGE requires excitation with circularly polarized radiation which yields optical spin orientation. It is the result of selective photoexcitation of carriers in k -space with circularly polarized light due to optical selection rules. The spin-galvanic effect is caused by asymmetric spin-flip scattering of spin polarized carriers. It is determined by the process of spin relaxation and may be generated by any means of spin injection. The inverse spin-galvanic effect has also been detecting demonstrating that electric current in non-magnetic but gyrotropic materials results in a non-equilibrium spin orientation. In some optical experiments the photocurrent may represent a sum of spin-galvanic and circular photogalvanic effects. Both effects provide methods to determine spin relaxation times. Furthermore, the relative strengths of Rashba and Dresselhaus terms describing the spin-orbit coupling in semiconductor quantum well structures can be extracted from spin photocurrent measurements, opening a way to determine the different contributions to spin-orbit coupling. While for the spin-galvanic and the CPGE photocurrents spin orientation is needed pure spin currents and the magneto-gyrotropic effect can be generated even by unpolarized radiation. Microscopically it is caused by electron gas heating followed by spin-dependent relaxation.

In last several years it has been demonstrated that photogalvanics and magneto-photogalvanics can also be efficiently use for probing topological surface and edge states. It is shown that THz radiation results in spin-polarized dc electric current, which is sensitive to the radiation polarization and may have a component changing the sign by reversing the radiation helicity. An important advantage of photogalvanic effects is that, due to symmetry arguments, they can be used to excite selectively the surface states of most 3D TIs. A possibility to selectively probe the surface states even at room temperature is particularly helpful in the search for novel 3D TIs in which transport experiments are often handicapped by a residual bulk charge carrier density.

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Optical orientation of a mesoscopic solid-state spin ensemble

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Mesoscopic nuclear spin ensemble interacting with a single confined electron spin in a solid-state quantum emitter presents a realization of the central spin model. Dynamic nuclear spin polarization (DNSP) in such quantum emitters, most notably in self assembled InGaAs quantum dots (QD), has been observed both for quasiresonant and nonresonant excitation [1]. Early experiments have demonstrated how excitation light helicity determines the sign of the Overhauser field, in analogy with earlier experiments in atoms or higher dimensional semiconductors. In stark contrast to nonresonant excitation however, bidirectional nuclear spin orientation independent of photon polarization was observed in resonant laser scattering of elementary transitions in neutral and negatively charged QDs. A particularly striking feature of resonant DNSP using the higher energy Zeeman transition at external magnetic fields exceeding 1 Tesla is the flattop absorption spectra, stemming from active locking of the QD resonance to the laser frequency [1].

In this talk, I will present our current understanding of bidirectional DNSP in resonantly driven QD transitions. In particular, I will describe a microscopic model based on the effective noncollinear hyperfine coupling that provides an excellent description of the experimental observations.

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Cooling, relaxation and spin temperature of nuclear spin system in GaAs.

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Cooling of nuclear spin system (NSS) in doped semiconductors via dynamic polarization by optical pumping is a powerful method for harnessing ubiquitous fluctuations of nuclear spin. The idea of NSS cooling is based on the hypothesis of spin temperature, which states that NSS reaches an internal thermal equilibrium long before it comes to equilibrium with the external bath (crystal lattice). Although thermodynamic framework has been successfully employed for the description of a variety of the experimental data, a rigorous check of this concept in semiconductors was impossible until recently, in particular at low magnetic field. The reason for that is the lack of experimental techniques allowing nonperturbative optical control over adiabatic transformation of the NSS.

We have recently developed such methods, based on off-resonant Faraday rotation and spin noise spectroscopy [1,2]. Using these techniques, combined with photoluminescence spectroscopy, we established a comprehensive picture of the nuclear spin relaxation efficiency, its magnetic field, temperature, and carrier concentration dependence in both n- and p-doped GaAs, a model system in the field of nuclear spin physics in semiconductors [3-5].

We also analyzed the interplay between four relevant relaxation mechanisms: hyperfine interaction, quadrupole interaction, spin diffusion towards paramagnetic impurities, and Korringa mechanisms. Figure 1 illustrates these processes in a nuclei-electron coupled system. Understanding of field dependence of NSS dynamics allowed us to obtain a new insight into the NSS thermodynamics, and verify the spin temperature concept in GaAs bulk material and microcavities [6]. We have demonstrated that NSS exactly

follows the predictions of the spin temperature theory, despite the quadrupole interaction that was earlier reported to disrupt nuclear spin thermalization in quantum dots [7]. These results open a way for the deep cooling of nuclear spins in semiconductor structures, with the prospect of realizing nuclear spin-ordered states for high-fidelity spin-photon interfaces.

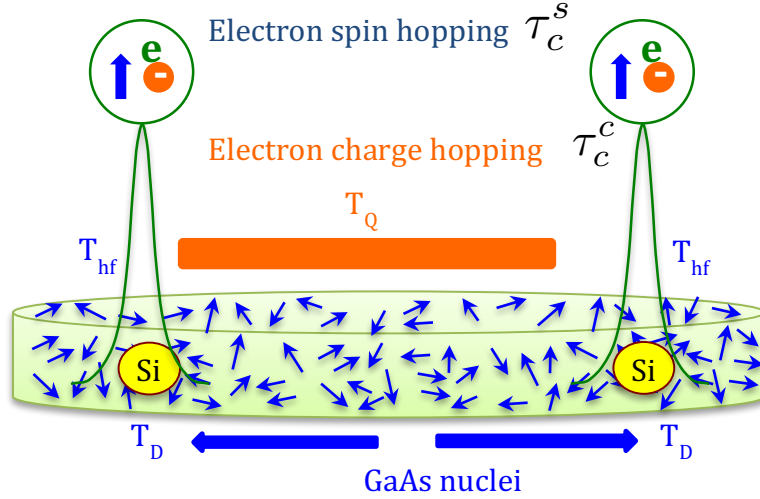


Figure. 1 Relevant interactions in a nuclei-electron coupled system in n-GaAs: fast nuclear spin relaxation under donor orbits via hyperfine interaction (characterized by time T_{hf}), and relaxation of all other nuclei via spin diffusion towards the donor sites and quadrupole interaction.

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A QD-based spin-photon interface for quantum networks

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Advances towards long distance quantum communication and distributed quantum processing require the distribution of entanglement to spatially separated nodes. Using photons to distribute entanglement between individual spins in the solid-state is a promising approach towards building a scalable quantum network [1]. Spins in InGaAs quantum dots (QDs) are one of the leading solid-state systems. They benefit from unrivalled optical properties, allowing single qubit operations on picosecond-timescales and efficient spin-to-photon state transfer. Recently, an important milestone has been reached with the distribution of entanglement between two nodes [2,3].

In this talk, I will give an overview of our recent work on spins in InGaAs QDs. I will present our demonstration of distant entanglement between QD spins at a record-high generation rate of 7.3kHz [3]. I will also examine electron and hole spin coherence in these systems [4,5]. Hyperfine interaction of these central spins with a nuclear ensemble leads to non-trivial features in the decay of spin coherence as shown in Figure 1. While the nuclear spin ensemble currently poses the greatest fundamental hindrance to future developments towards QD-based quantum networks, they are an exciting platform to study many-body physics. As shown in Figure 2, nuclear spins can be cooled to form a low-entropy state [6]. To conclude, I will present outlook towards collective nuclear state engineering and nuclear quantum memory.

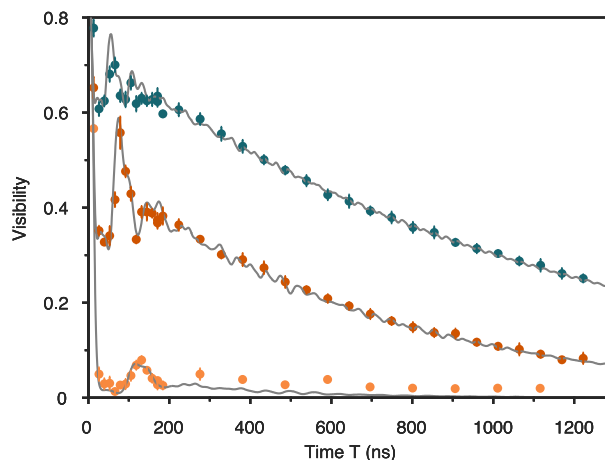


Figure. 1 Hahn Echo visibility of an electron spin in an InGaAs quantum dot at 2T (yellow markers), 3T (orange markers) and 4T (blue markers). The collapse and revivals at short times and the decay at longer times are solely governed by the hyperfine interaction with the strained nuclear ensemble.

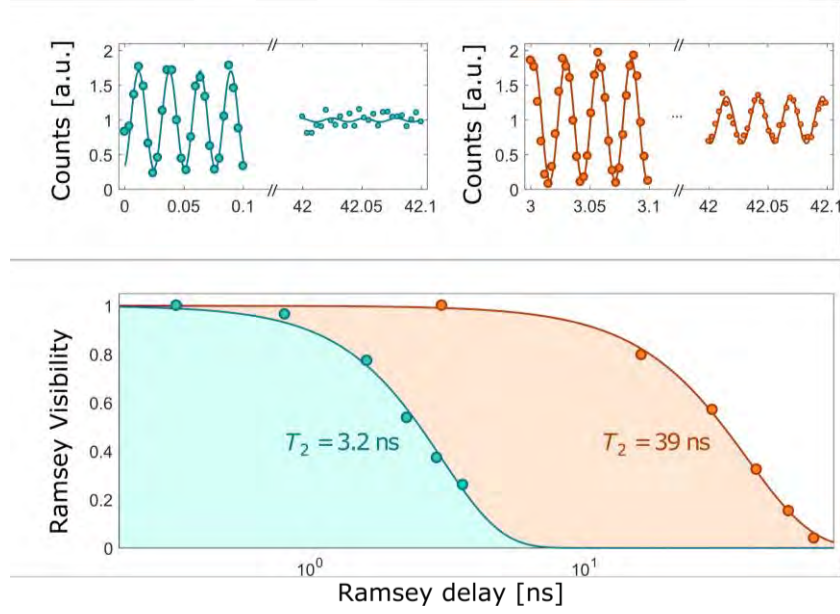


Figure. 2 Visibility of Ramsey fringes for an electron spin in an InGaAs quantum dot with and without electron-mediated optical cooling of the nuclear spin bath. The external field is 6T. The inhomogeneous dephasing time (T_2^*) of the electron is increased by an order of magnitude as a direct consequence of the reduction of nuclear spin fluctuations. If purely achieved through nuclear spin polarisation, such reduction of the nuclear spin fluctuations would require an ensemble polarisation exceeding 99%.

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From Faraday rotation to spin-noise spectroscopy with a single spin

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A linearly-polarized beam transmitted/reflected by a transparent magnetic medium can experience a large rotation of its polarization state; this is the Faraday/Kerr rotation effect. Such an effect is at the heart of spin-noise spectroscopy: this emerging technique consists in registering the rapid fluctuations of a Faraday/Kerr rotation signal, dictated by the fluctuations of magnetization in a material, to extract information on the lifetime and coherence of spin ensembles. Until recently, however, applying such a technique to a single spin was out of reach. In the absence of cavity enhancement, Faraday/Kerr rotation induced by a single spin remains in the millidegree range.

Interfacing a single spin with polarized photons also constitutes a major objective in a very different framework: quantum optics. Single spins can indeed be used as solid-state quantum memories which could communicate by exchanging single photons. All these applications strongly motivate the development of cavity quantum electrodynamics devices embedding a single spin.

We present an overview of our technological and experimental efforts to develop an effective spin-photon interface, using a charged InAs/GaAs quantum dot deterministically coupled to a GaAs/AlGaAs micropillar cavity (Fig. 1). Such a geometry constitutes an excellent light-matter interface into/from which one can efficiently inject/extract photons [1].

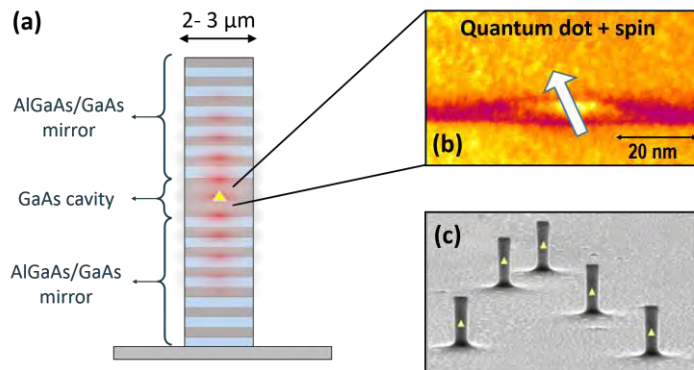


Figure 1 (a) Principle of a micropillar cavity coupled to a single quantum dot. (b) Electron microscope image of a single InAs/GaAs quantum dot (QD). In a charged QD the resident charge carries a spin degree of freedom which can be optically addressed. (c) Electron microscope image of micropillar cavities deterministically coupled to individual quantum dots [1].

In particular, we have reported a $\pm 6^\circ$ polarization rotation induced by a single hole spin (Fig. 2), which has previously been optically pumped in state $|\uparrow\rangle$ or $|\downarrow\rangle$ using resonant, circularly-polarized pulses. Such a macroscopic rotation of polarization corresponds to a three orders of magnitude enhancement, compared to the previous state of the art [2].

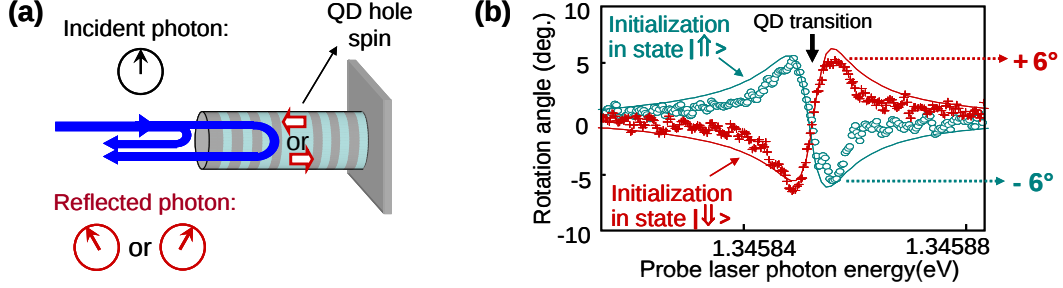


Figure 2. (a) Principle of giant polarization rotation induced by a single spin: the polarization of reflected photons is rotated clockwise or counter-clockwise, depending on the spin orientation. (b) Giant polarization rotation induced by a single spin, initialized either in the $|\uparrow\rangle$ or $|\downarrow\rangle$ state [2].

In addition, we have theoretically described how an ideal rotation of $\pm 45^\circ$ can be reached with very realistic devices: such a configuration would enable a one-to-one correspondance between a single spin state, described in the Bloch sphere, and a single photon polarization state, described in the Poincaré sphere. Recent measurements will be presented, showing the potential of polarization analysis in the Poincaré sphere for the characterization of decoherence processes in solid-state devices [3].

Finally, we will present our recent theoretical study of spin noise spectroscopy at the single spin level. The application of this technique to a spin-photon interface is of particular interest, as each detected photon leads to a quantum measurement back-action on the spin system. We have thus shown that the frequency spectrum of photon correlations displays all the relevant information regarding spin dynamics and photon-polariton dynamics, but also regarding the back-action induced by photon detection events [4]. A perfect back-action can be achieved by detecting a single photon, and the strength of this back-action can be tuned by varying the polarization measurement basis. It thus becomes possible to use the spin-photon interface as a quantum measuring apparatus, which allows investigating quantum measurement and measurement-induced decoherence.

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Optical control of the spin of individual magnetic atoms

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Controlling individual spins in semiconductors is very attracting for fundamental issues and for the potential of such ultimate spin based memories in the field of emerging quantum technologies. Thanks to their expected long coherence time, the spin of individual magnetic atoms embedded in a semiconductor could be an interesting media for data storage and processing. In addition, a variety of magnetic atoms can be incorporated in conventional semiconductor giving a large choice of localized electronic spin, nuclear spin as well as orbital momentum.

It has been shown that the electrical and the optical properties of a semiconductor quantum dot (QD) can be used to probe the spin of individual [1-4] or pairs [5,6] of magnetic atoms. This was first demonstrated in the case of Manganese (Mn) which carries an electronic spin $S=5/2$, a nuclear spin $I=5/2$ and has no orbital momentum. In a II-VI semiconductor QD doped with a Mn atom, the exchange interaction of the Mn spin with the confined carriers shifts the energy of the QD depending on the relative values of the Mn and heavy-hole spins. Six lines, corresponding to the six spins states of the Mn, are observed in the photoluminescence (PL) of the exciton (X). In the presence of the weak magnetic anisotropy induced by the biaxial strain in the plane of the QDs, the electronic spin of a Mn atom can be initialized by the optical injection of spin-polarized carriers (optical orientation and resonant optical pumping) [7]: a spin memory is observed at zero magnetic field. The suppression of the magnetic anisotropy in strain-free QDs [8] or its enhancement in QDs containing a single hole [9] significantly modifies the Mn spin dynamics and its magnetic field dependence.

Under a strong resonant optical field, the energy of any spin state of a Mn atom can also be independently tuned using the optical Stark effect. Under optical excitation resonant with an absorption transition, we can enter the strong coupling regime where hybrid states of matter and light are created. The ground state of the magnetic atoms is "dressed" with light [10]. The spin dependent strong coupling with the laser field modifies the Mn fine structure (interaction with the crystal field responsible for the magnetic anisotropy) and hyperfine structure (interaction with the ^{55}Mn nuclear spin $I=5/2$). We demonstrated that the optically induced modification of the fine structure of the Mn atom significantly affects its spin dynamics and its initialization under resonant optical excitation. In addition to standard optical pumping expected for a resonant excitation, for particular conditions of the laser detuning and excitation intensity, the spin population can be trapped in the state that is resonantly excited [11].

Among the variety of magnetic transition elements that can be incorporated in semiconductors, Chromium (Cr) is also of particular interest: Cr is incorporated in II-VI compounds as a Cr^{2+} ion carrying an electronic spin $S=2$ with an orbital momentum $L=2$ and 90 % of Cr isotopes have no nuclear spin limiting the number of spin states to 5. The non-zero orbital momentum provides a large sensitivity of the electronic spin to local

strain through the modification of the crystal field and the spin orbit coupling. This makes Cr a very promising spin *qubit* for the realization of hybrid spin-mechanical systems.

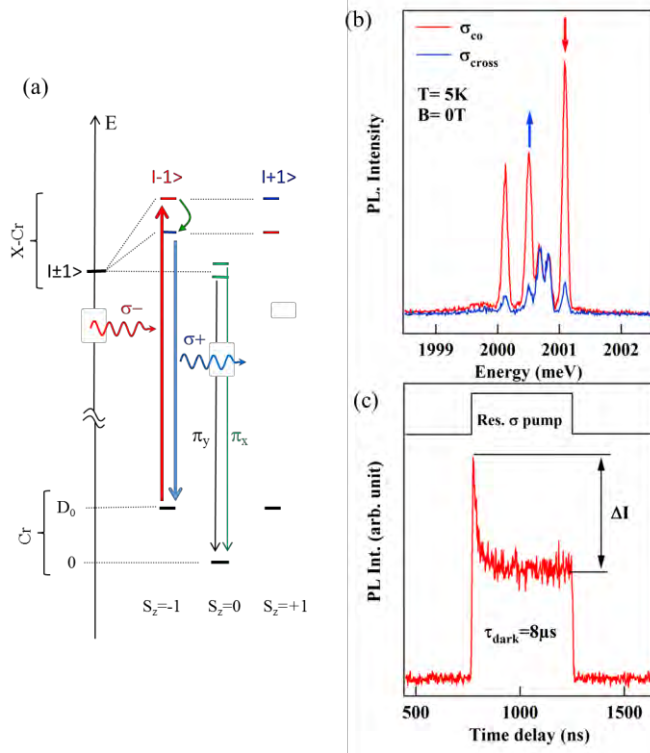


Figure 1: (a) Energy levels in a CdTe/ZnTe Cr-doped QD and configuration of the excitation/detection for resonant optical pumping experiments. The Cr spin states $S_z = 0, \pm 1$ are split by a large magnetic anisotropy $D_0 S_z^2$. For the exciton-Cr complex (X-Cr), the exchange interaction with the bright exciton ($| \pm 1 \rangle$) splits the Cr spin states $S_z = \pm 1$. The higher energy Cr states, $S_z = \pm 2$, and the dark exciton states are not displayed. (b) PL of the exciton in a Cr-doped QD for co- and cross-circularly polarized excitation / detection. (c) Time evolution of the PL intensity of the low energy line of X-Cr under resonant excitation on the high energy line with circularly polarized excitation pulses of 500 ns length separated by a dark time of $\tau_{dark} = 8 \mu s$

QD could be controlled optically. The exchange interaction between the confined carriers and the localized spin is strong enough to allow a mapping of the spin state of the Cr into the QD optical transitions [12]. We have shown that for such magnetic atom with a large magnetic anisotropy, the optical control of the spin is only possible under resonant excitation (resonant optical pumping) [13]. Monitoring the time dependence of the intensity of the resonant PL of the QD during the optical pumping (Figure 1) permits to probe the dynamics of the initialization of the Cr spin. Using this initialization and readout technique we measure a Cr spin relaxation time at $T = 5K$ and in the absence of optical excitation in the microsecond range. The spin relaxation is strongly accelerated when carriers and/or phonons are optically injected in or near the QD [13].

The large spin-strain coupling and the possible optical control of its spin makes Cr a promising platform to study the interaction of the $\{+1; -1\}$ Cr spin *qubit* with surface acoustic waves (phonons propagating at the surface of a sample) which are proposed as efficient quantum bus between different kinds of *qubits*.

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Challenges and perspectives of optical spin orientation applied to group IV heterostructures

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The seminal work of Lampel put forward optical orientation as a prominent technique to provide direct access to the spin properties of semiconductors [1].

Notably, the generation of a non-equilibrium spin population by means of the absorption of circularly-polarized light was especially applied to direct-gap III–V compounds in fruitful conjunction with polarization-resolved photoluminescence (PL) to gain a deeper understanding of spin-dependent phenomena [2, 3].

Optical investigations of the spin physics, however, lagged well behind in group IV semiconductors and remained largely overlooked chiefly because of the genuine indirect bandgap and the concomitant poor light absorption and emission efficiencies [4].

Despite the challenges imposed by the subtleties of the band structure, group IV semiconductors carry unique and advantageous properties. Their centrosymmetric crystal structure guarantees intrinsically long spin lifetimes, as it prevents spin relaxation due to the Dyakonov-Perel mechanism [3, 5]. Moreover, the natural abundance of zero-spin nuclear isotopes suppresses hyperfine interactions and lengthens the spin coherence time [6, 7]. Such features, alongside the key support of advanced technological infrastructures pertaining to the microelectronic industry, recently led group IV materials to gain new momentum. In this context, Ge stands out because of its superior electronic properties. As opposed to Si, the small energy distance between the direct and indirect gap offers indeed straight access to the direct optical transitions.

Here we will discuss how optical spin orientation can be applied to Ge and how this approach is instrumental in disclosing intriguing spin and energy relaxation mechanisms [8, 9]. The emerging physical picture of the Ge spin dynamics stems from the multivalley conduction band and does not have any counterpart in the well-established literature dealing with direct-gap semiconductors [10]. We will also point out how strain and quantum confinement can be leveraged as effective degrees of freedom to tailor the spin-orbit interaction and utilized to further enrich the spin dependent phenomena observable in Ge-based heterostructures [11].

Finally, we will devote our attention to the ongoing efforts to hybridize the spin properties of Ge with the unique electronic states offered by another group IV element, namely α -Sn [12]. Epitaxial GeSn alloy layers grown on Ge-buffered Si substrates offer already exciting prospects in optoelectronics, as elucidated by the demonstration of low-temperature optically pumped laser and by the tunable nature of the gap [13].

In light of these findings, we expect GeSn alloys to play a central role in enabling future convergence of photonics and spintronics concepts on conventional Si substrates. Despite such remarkable properties, however, to date very little is known about the spin-dependent phenomena of Sn-based alloys.

Here we tackle this challenge by using optical spin injection and report the manifestation of spin quantum beatings in the PL under an external magnetic field (see Figure 1) [14].

The demonstrated applicability of light-matter interaction facilitated in this work the anticipation of spin-dependent phenomena pertaining to this notable material. Such findings can open interesting perspectives towards the optical exploitation of a very rich spin physics resulting from the tunable band structure and spin-orbit coupling promised by novel group IV heterostructures.

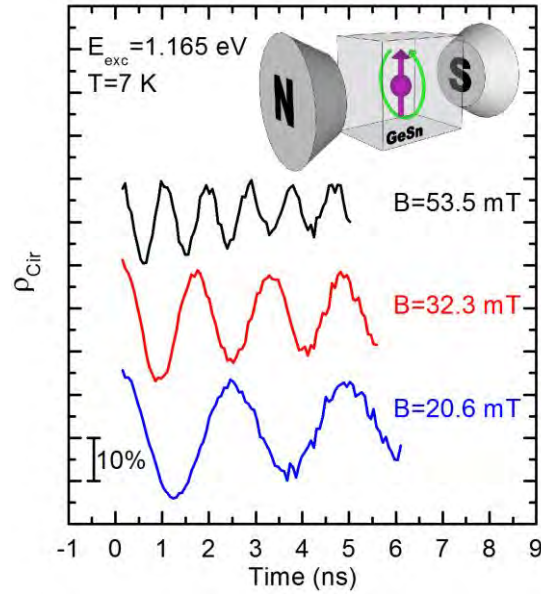


Figure. 1 Spin quantum beats in a fully strained $\text{Ge}_{0.95}\text{Sn}_{0.05}$ layer epitaxially deposited on a Ge buffered Si substrate. Magnetic field dependence of the circular polarization degree (p_{cir}) dynamics measured at a lattice temperature of 7 K and for a laser excitation energy of 1.165 eV. Data obtained by applying a magnetic field strength (B) of 20.6 (blue curve), 32.3 (red curve) and 53.5 mT (black curve) have been shifted for clarity [14].

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“Listening” to the spin noise of electrons and holes in semiconductors

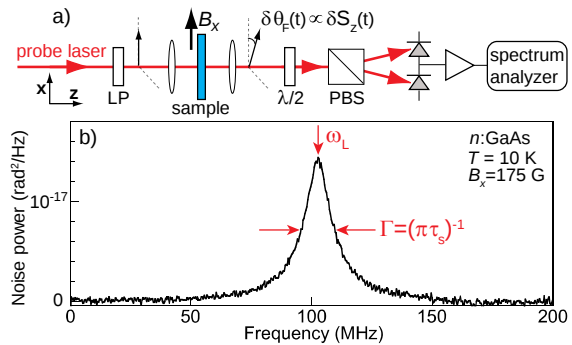
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Not all noise in experiments is unwelcome. Certain types of fundamental noise contain extremely valuable information about the system itself – a classic example being the inherent voltage fluctuations across any resistor (“Johnson noise”), from which temperature can be determined. In magnetic systems, fundamental noise can exist in the form of random spin fluctuations. For example, statistical fluctuations of N paramagnetic spins should generate small noise signals of order $\sqrt{N}$ spins, even in zero magnetic field. In accordance with the Fluctuation-Dissipation Theorem, the spectrum of these fluctuations – if experimentally measurable -- can reveal the important dynamical properties of the spins (such as g-factors and spin decoherence times), but *without* ever driving, exciting, or perturbing the spin ensemble away from thermal equilibrium.

This talk will describe how spin dynamics in semiconductors can be measured by passively “listening” to these random noise signals. We employ a spin noise spectrometer based on sensitive optical Faraday rotation magnetometry. This approach, applied originally to paramagnetic atomic vapors [1], has since been used to measure spin noise from electron Fermi seas in n -type GaAs [2], from electrons and holes localized in InGaAs quantum dots [3], from electron and hole “central” spins coupled to nuclear spin baths [4], and from magnetic dopants in II-VI semiconductors [5]. Interestingly, noise-based approaches also allow to circumvent certain restrictions of linear response theory [6]. Moreover, because fluctuations are recorded directly in the time domain, nontrivial higher-than-second-order spin correlations can also be directly computed from the noise data [7]. This talk will highlight recent developments in semiconductor spin noise spectroscopy, and will discuss how these methods can also be applied to other spintronics-related fields such as magnetization dynamics in ferromagnetic films [8].

Figure: a) Spin noise schematic. b) Example of spin noise from bulk n :GaAs in thermal equilibrium. Random precessing spin fluctuations give measurable noise centered at the Larmor frequency ω_L . The frequency, width, lineshape, and amplitude of the noise encodes valuable information about the underlying spin dynamics.



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Electron, Hole, and Nuclear Spin Dynamics in Localized-Carrier Systems

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We study the effect of localization on the intrinsic spin dynamics in semiconductors starting from a single hole localized in an individually addressable quantum dot (QD) by spin noise spectroscopy (SNS) [1] reaching up to the complex spin dynamics at the intriguing metal-to-insulator transition (MIT) which has been a surprisingly complex and controversially discussed problem in semiconductor physics and which has been finally solved.

First, we present detailed measurements in combination with a full theoretical analysis of the spin noise spectra beyond the fluctuation-dissipation theorem of the heavy-hole spin and the trion spin dynamics of single QD [2]. The data reveal an additional, much weaker noise contribution in the quasi resonant Kerr rotation noise spectra which shows a significantly longer correlation time. Magnetic-field dependent measurements in combination with theoretical modeling prove that this additional noise contribution unveils a charge reoccupation noise which is an intrinsic limitation to naturally charged QDs as optically addressable spin qubits. Nevertheless, the impact of optically evoked losses of spin coherence due to parasitic excitations can be avoided by homodyne detection schemes in future SNS [3], [4].

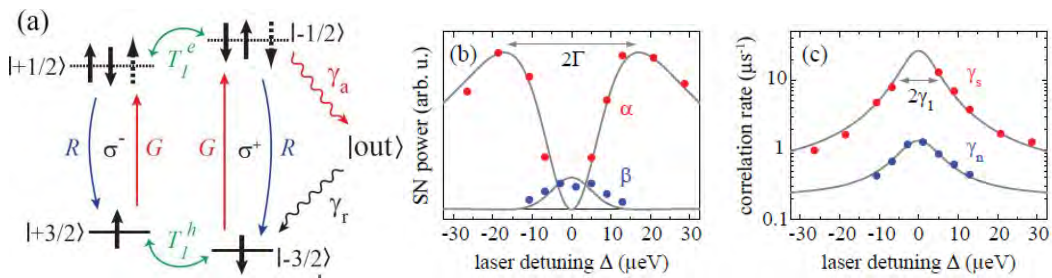


Figure. 1 a) Sketch of the QD states and the relevant transitions under resonant spin noise probing conditions. b) Homogenously broadened SN power spectrum of the main SN contribution α and SN due to (re)charging dynamics β . The gray lines are theoretical calculations (see Ref. [2]). c) Spin correlation rates γ_s (α contribution) and γ_n (β contribution) as a function of detuning.

A complementary system are donor bound electrons in ultrahigh quality, very weakly n-doped GaAs since the widely spaced, quasi-isolated electrons act as an ensemble of identical, individually localized atoms. However, the drawback of this system is the inherent interaction with the nuclear spin bath. Here, we present SNS measurements on an ensemble of donor-bound electrons in ultrapure GaAs:Si covering temporal dynamics over six orders of magnitude ranging from milliseconds to nanoseconds [5].

The SN spectra detected at the donor-bound exciton transition show the multifaceted dynamical regime of the ubiquitous mutual electron and nuclear spin interaction typical for III-V-based semiconductor systems. The data distinctly reveal the finite Overhauser shift of electron spins precessing at zero external magnetic field together with an unmodulated contribution around zero frequency related to the electron spin component parallel to the nuclear spin fluctuations. Moreover, at very low frequencies, features arising from time-dependent nuclear spin fluctuations are clearly resolved which reveals the intricate nuclear spin dynamics at zero and low external magnetic fields. The experimental findings are well described by a microscopic model including both, electron and nuclear spin noise. [5]

In order to unambiguously unravel the intriguing but complex contributions to the spin relaxation at MIT, we combine extensive precision measurements of the optically detected spin dynamics and magneto-transport measurements in a contiguous set of n-doped bulk GaAs structures. Just below the MIT, the interplay between hopping and hyperfine interaction induces loss of spin coherence. At slightly higher doping concentrations, the spin relaxation deviates from the expected Dyakonov-Perel mechanism and is consistently explained by a reduction of the effective motional narrowing close to the MIT attributed to the change of the dominant momentum scattering mechanism in the metallic impurity band, where scattering by local conductivity domain boundaries due to the intrinsic random distribution of donors becomes significant. We fully identify and model all intricate contributions of the relevant microscopic scattering mechanisms, which finally allows the **complete quantitative** modeling of the electron spin relaxation in the entire regime from weakly interacting up to fully delocalized electrons. [6]

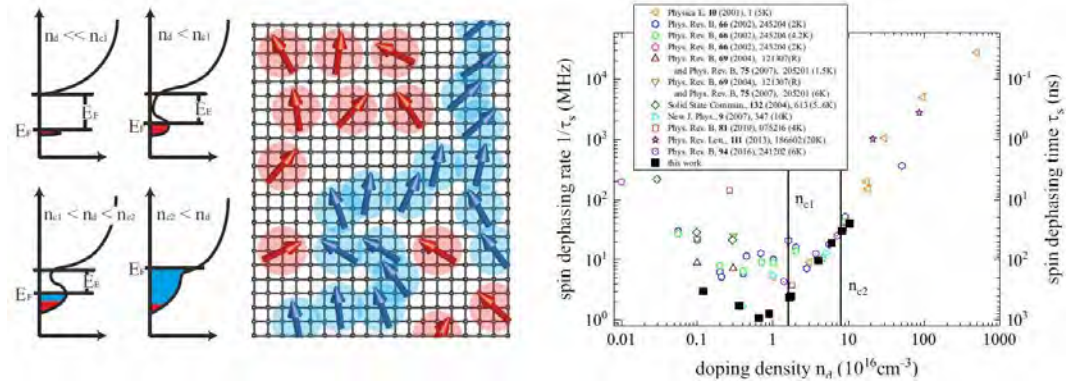


Figure. 2 Left) Effective density of states at different doping concentrations. The density of localized states is depicted in red, delocalized states are shown in blue. middle) a schematic picture of the coexistence of variable range hopping and filamentary electron transport along percolation paths at the MIT. Right) High precision, low temperature measurements (black squares) of the electron spin dephasing time τ_s in n-GaAs as function of donor density n_d compared to literature values of τ_s in the limit of negligible magnetic fields. See Ref. [6]

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Metal-to-insulator transition in n -GaAs revealed in coherent spin dynamics of optically oriented electrons

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The spin dynamics of localized electrons in solids is drastically different from that of mobile electrons. Semiconductors are ideally suited for changing the spin localization in a controlled way either by controlling the donor concentration or temperature. However, because of experimental limitations, a comprehensive study revealing the changes in both longitudinal and transverse spin dynamics when crossing the metal-to-insulator transition (MIT) so far was lacking. The relevant spin relaxation times range from picoseconds to milliseconds depending on doping concentration, temperature, and magnetic field, while common techniques providing access to the spin dynamics, are limited to the maximal time range, time resolution or range of accessible magnetic fields. We overcome these limitations by using the extended pump-probe Faraday/Kerr rotation technique, which allows direct measurement of both the transverse and longitudinal spin dynamics in magnetic fields of any strength with picosecond time resolution over an arbitrary long time range [1].

We investigate the evolution of the electron spin dynamics as consequence of carrier delocalization in n -type GaAs. We find that isolated electrons localized on donors demonstrate a prominent difference of the longitudinal and transverse spin relaxation times T_1 and T_2^* , respectively, which strongly depend on a magnetic field (Fig. 1). While these times were believed to have different nature up to now, we find the surprising relation $T_1 T_2^* \approx \text{const}$ for electrons localized on donors, and explain it by the impact of the magnetic field on the spin diffusion. On the other hand, mobile electrons are characterized by a unique spin lifetime, that is extended by motional narrowing. With rising temperature or donor concentration, we observe a striking 100-fold increase of T_2^* due to the onset of motional narrowing. In the range of donor concentrations just below the MIT, we find a double-exponential longitudinal spin dynamics, which reflects the fast onset of internal equilibrium within the electron spin system, followed by equilibration of the electron spin temperature with the crystal lattice temperature [2].

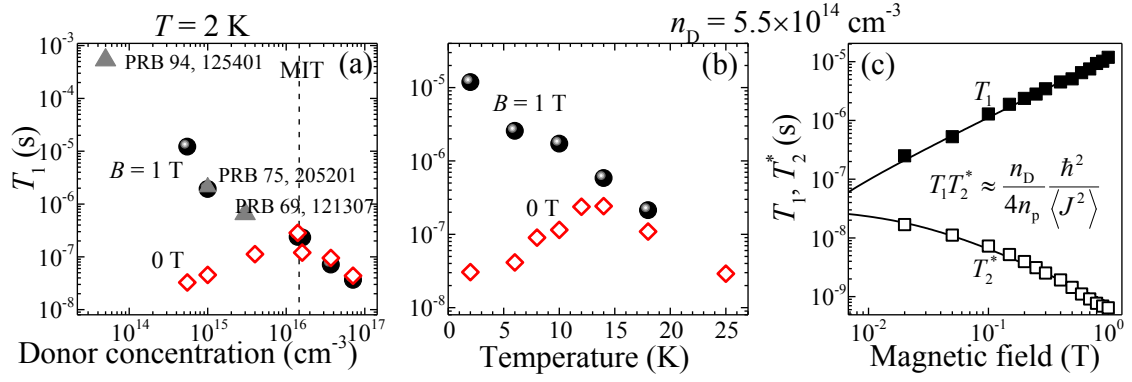


Figure. 1 (a) Dependence of the longitudinal spin relaxation time T_1 on donor concentration for the magnetic field $B = 0$ T (open symbols) and 1 T (solid symbols), temperature $T = 2$ K. Triangles correspond to literature data. (b) Dependence of T_1 on lattice temperature for $B = 0$ T (open symbols) and 1 T (solid symbols) in the sample with donor concentration $n_D = 5.5 \times 10^{14} \text{ cm}^{-3}$. (c) Magnetic field dependencies of T_1 (solid symbols) and T_2^* (open symbols) at different temperatures for $n_D = 5.5 \times 10^{14} \text{ cm}^{-3}$. Lines show linear fits to T_1 and to the reciprocal of T_2^* .

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Giant resonant ultrashort pulse polarisation rotation in a charged quantum dot – micropillar system

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Realisation of a deterministic spin-photon entanglement in semiconductor quantum dots (QDs) is currently one of the major goals towards integrated quantum photonics based on this platform. In general, there are two possible ways of realising a quantum superposition of states: through controlled rotations of either the material spin or the photon polarisation state representing the qubit. One promising method of achieving a quantum superposition of matter states is selectively addressing individual confined charge-carrier spins using circularly polarised photons generated by an external source, and manipulating the spins through optically excited states (charged excitons) by employing the techniques of coherent quantum control and optical orientation [1, 2]. An alternative approach is to make use of the optical polarisation rotation induced by the single spin and thus prepare photon polarisation states with high-fidelity. Coupling a stationary spin qubit to an optical cavity leads to an enhanced efficiency of spin-photon interaction, thereby accumulating polarisation rotation angle by several orders of magnitude due to multiple reflections and round-trips of the pulse between the mirrors. On the other hand, a single QD strongly-coupled to a cavity mode is a solid-state analogue of an atom-cavity system in quantum optics and represents a cavity quantum electrodynamics (cQED) system. Recently, a macroscopic Kerr rotation of photon polarisation ($\sim 6^\circ$) has been observed using a CW excitation in both strong- [3] and weak-coupling [4] regimes in a charged QD-micropillar system. We note, however, that a maximum phase shift of π is predicted by a number of theoretical works in the strong-coupling regime [5, 6].

Theoretical model and numerical results

Resonant coherent interaction of an ultrashort pulse with a discrete-level quantum system results in a pulse resonant absorption/gain (depending on the initial state preparation in ground/excited state) and a phase shift, giving rise to the imaginary/real components of the dielectric susceptibility, respectively. In this work we employ our vector Maxwell-pseudospin master equation formalism [1, 2] to compute the time evolution of the equivalent 4-level quantum system shown in Fig. 1(a), mapping the fundamental trion transitions, under a resonant ultrashort circularly polarised pulse excitation, taking into account the dominant spin relaxation processes. The system is initially prepared in either the spin-down or spin-up state by optical pumping and a circularly polarized π -pulse ($T_p \sim 100$ fs) is injected through an optical fibre from the top DBR mirror.

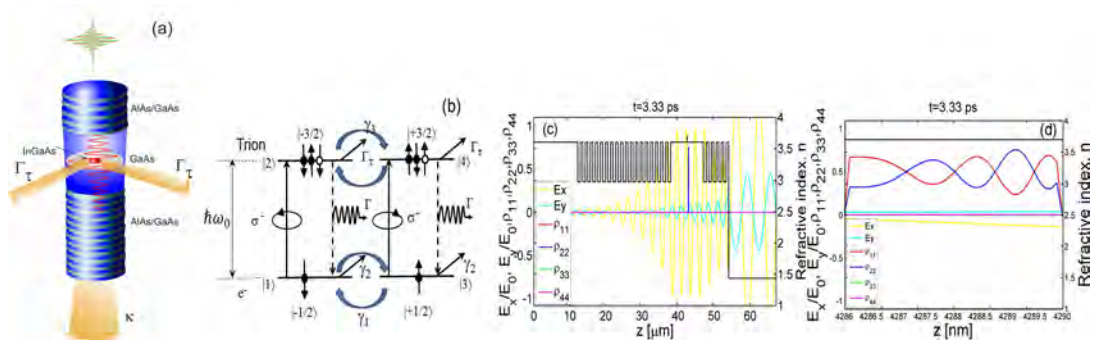


Figure. 1 (a) Sketch of a charged QD-micropillar structure; κ - cavity mode decay rate, Γ_τ - trion decay rate to non-cavity modes; (b) Energy-level diagram of a negatively charged exciton (trion) ground singlet transition driven resonantly by either left- (σ^-) or right- (σ^+) circularly polarised pulses. Electron (γ_1) / hole (γ_3): spin flip relaxation rates; Γ : spontaneous emission rate; γ_2 , Γ_τ : electron and trion spin decoherence rates. (c) Refractive-index profile (black line) of the micropillar cavity from [4] with embedded QD and a snapshot of the electric-field components (yellow and cyan curves) and level populations (red, blue, green and magenta) at $t=3.33$ ps. The pulse is injected at the right boundary through an optical fibre. (d) Zoom-in on (c) of the QD layer.

We calculate the quantum evolution of the four-level system under σ^- circularly Fig. 2(a, b) and x -linearly polarized Fig. 2(c, d) ultrashort pulse excitations [7]. The time-resolved photoluminescence trace (ρ_{22} , blue curve) exhibits fast modulations due to incomplete Rabi cycles. We demonstrate a giant ($\sim \pm\pi/2$) phase shift (Fig. 2(e)) in the circularly polarised pulse transmission spectrum induced by the single spin confined in the QD in a realistic micropillar cavity (Fig. 1(c,d)) operating in the weak-coupling regime. By contrast, when the trion transition is driven by a resonant x -linearly polarized pulse, we show that a y -component of the electric field of the pulse starts to build up, effectively demonstrating polarisation rotation induced by the resonant transition (Fig. 2(d)). This delay in the E_y amplitude build-up results in a red shift in the phase shift spectrum (Fig. 2(f)) with respect to the one for the E_x (Fig. 2(e,f)) leading to a decrease of the rotation angle of the x -polarized component at the detection wavelength at the expense of an increase of the E_y rotation angle at a longer wavelength.

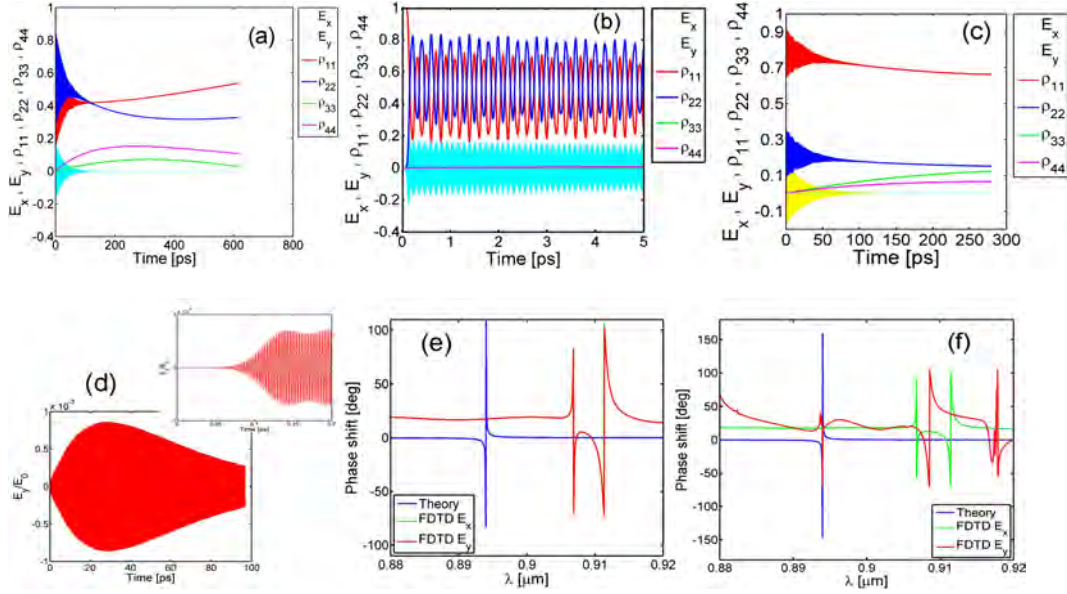


Figure. 2 (a) Time evolution of the electric-field components of a σ^- pulse, and level populations at the right edge of the QD layer (see Fig. 1 (d)) for a spin-down initial state. (b) Ultrashort time dynamics exhibiting Rabi flopping. (c) Time evolution of a resonant x -linearly polarized pulse field components and level populations. (d) Build-up of the E_y pulse component during propagation of a resonant x -linearly polarized pulse (inset: zoom-in of initial build-up). (e) Phase shift (in degrees) of the E_x and E_y pulse components of a circularly polarized σ^- pulse. Blue curve: stationary solution of the density-matrix equations for a 2-level system (no cavity). (f) Phase shift of the E_x and E_y pulse components of a linearly x -polarized pulse.

In addition to the high-fidelity photon polarisation state preparation, such large rotation angles would allow reliable detection of the initial spin state. Enhanced circular dichroism and birefringence under external magnetic field in a Faraday configuration are also expected.

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Mismatch between theoretical and experimental spin-transfer rates in diluted magnetic semiconductors explained by correlation effects

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The spin dynamics of photoexcited carriers in intrinsic II-VI diluted magnetic semiconductors (DMS) is usually dominated by the *sp-d* exchange interaction between carrier spins and localized magnetic moments of impurity ions such as Manganese. Additionally, pronounced excitonic features are often observed in optical experiments performed on DMS heterostructures and it has been shown theoretically that effects beyond Fermi's golden rule become relevant especially for excitations close to sharp structures in the density of states [1], such as for optically excited excitons at the bottom of the exciton parabola. Thus, a meaningful theoretical description of the ultrafast spin dynamics in DMS has to account for excitonic Coulomb correlations as well as carrier-impurity correlations [2,3].

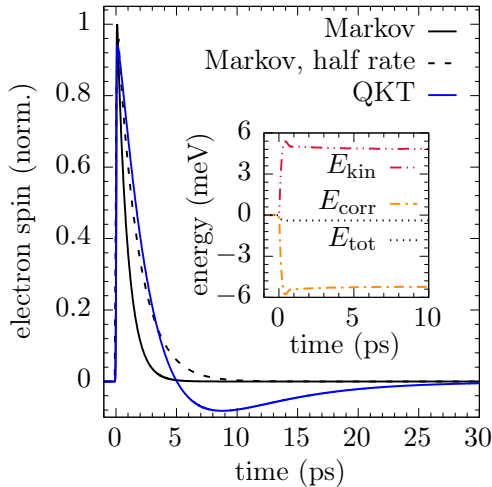


Figure 1: Exciton-bound electron spin dynamics in a 10 nm wide $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Te}$ quantum well after optical excitation at the $1s$ resonance using the quantum kinetic theory (QKT) and Fermi's golden rule (Markov). The inset shows the time-resolved energy contributions (E_{kin} : kinetic energy, E_{corr} : correlation energy, E_{tot} : total energy) during the first 10 ps.

Calculations based on Fermi's golden rule predict systematically different spin-transfer rates compared with experimental data [4]. In particular, when applied to excitons, such calculations consistently overestimate the spin decay. In this contribution we present a theoretical framework which treats the interaction between excitons and magnetic impurities beyond the mean-field level. To achieve this, we use a recently developed quantum kinetic theory (QKT) for excitons in quantum wells [3] that explicitly captures correlations between excitons and Mn ions and thus incorporates many-body effects which are absent in an approach using Fermi's golden rule. Our model includes the magnetic interaction between carriers and impurities, the Coulomb interaction responsible for the formation of excitons, Zeeman terms for carriers and impurities, the light-matter coupling, and scattering due to LA phonons. Furthermore, we account for nonmagnetic scattering of excitons at impurities, an effect which does not contribute to the spin dynamics on a Markovian level but which has a significant impact when treated quantum kinetically [2,3,5]. The Markov limit of our theory in general coincides with Fermi's golden rule and can straightforwardly be derived also for a perpendicular external magnetic field.

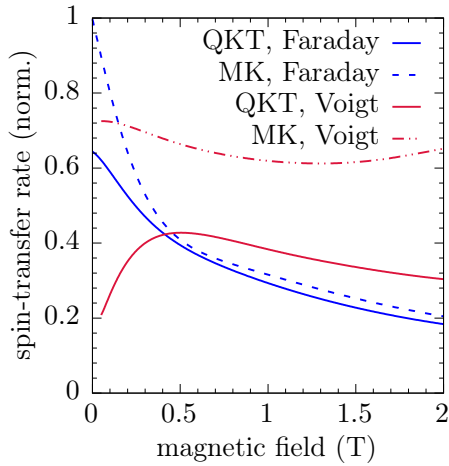


Figure 2: Magnetic field dependence of normalized exciton spin-transfer rates in a 20 nm wide $\text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se}$ quantum well at 4 K after optical excitation (100 fs FWHM) at the $1s$ exciton resonance. We compare quantum kinetic (QKT) with Markovian (MK) calculations.

wave-vector dependent, it is not surprising that a redistribution on the exciton parabola also has consequences for the spin dynamics. All in all, the typical spin-transfer rates obtained in our model are closer to experimental observations since the slower spin decay due to the correlation effects rectifies the overestimation of the rate when using Fermi’s golden rule for excitons [3].

We also consider the magnetic field dependence of exciton spin-transfer rates in Faraday (longitudinal field) and Voigt (transverse field) geometry using the QKT and compare the results with the corresponding Markovian theory for DMS with different doping concentrations [2]. To enable a straightforward comparison, we extract the time where the spin has decayed by a factor of $1/e$, even though the spin dynamics predicted by the QKT is nonexponential in general. Whereas quantitative differences between the predictions of the QKT and the Markovian theory are found for Faraday geometry especially at small magnetic fields, a qualitative trend reversal occurs in the Voigt configuration (cf. Fig. 2). This can again be traced back to the build-up of carrier-impurity correlations which are not captured on a Markovian level and become particularly enhanced due to nonmagnetic scattering at impurities [2]. Similar nonmonotonic features have also been observed in experiments performed on $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ in a transverse magnetic field [4]. Thus, all of our theoretical analysis suggests that correlations beyond Fermi’s golden rule play a key role in the spin dynamics of excitons in DMS nanostructures.

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Considering first the situation without an external magnetic field, our analysis reveals significant memory effects such as a spin overshoot and an initial spin decay which is approximately a factor of two slower compared with the rate obtained via Fermi’s golden rule (cf. Fig. 1). The factor of two can be traced back to a cut-off of the memory kernel induced by the correlations that happens at the bottom of the exciton parabola. Also, significant correlation energies on the order of a few meV are accompanied by a pronounced redistribution of the exciton center-of-mass momenta, leading to an occupation of states with on average higher kinetic energies which can be measured, e.g., by using LO-phonon-assisted photoluminescence. Without phonons, a redistribution towards higher exciton kinetic energies would be completely absent in a Markovian description because of energy-conserving delta functions that only account for single-particle energies while correlation energies are disregarded. Up to moderate temperatures of a few K, the correlation-induced redistribution is found to be much stronger than that resulting from phonons. Since we find that already the Markovian rates are strongly

Electrical initialization of electron and nuclear spins in a single quantum dot at zero magnetic field

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The emission of circularly polarized light from a single quantum dot relies on the injection of carriers with well-defined spin polarization. Here we demonstrate single dot electroluminescence (EL) with a circular polarization degree up to 35% at zero applied magnetic field. This highly efficient electrical injection of spin polarized electrons is achieved by combining ultrathin CoFeB electrodes on top of a spin-LED device with p-type InGaAs quantum dots in the active region [1]. The CoFeB ultrathin injector (with a thickness of about 6 atomic planes [2]) presents a Perpendicular Magnetic Anisotropy (PMA) thanks to Fe(Co)–O bondings, a key property in order to inject electron spin along the growth axis of the structure. In addition, we measure an Overhauser shift of several micro-eV at zero magnetic field for the positively charged exciton (trion X⁺) EL emission, which changes sign as we reverse the injected electron spin orientation. This is a signature of dynamic polarization of the nuclear spins in the quantum dot induced by the hyperfine interaction [3] with the electrically injected electron spin [4]. Both EL circular polarization and Overhauser shift follow the hysteresis cycle of the magnetic electrode. This study paves the way for electrical initialization of electron and nuclear spins in a single quantum dot without any external magnetic field.

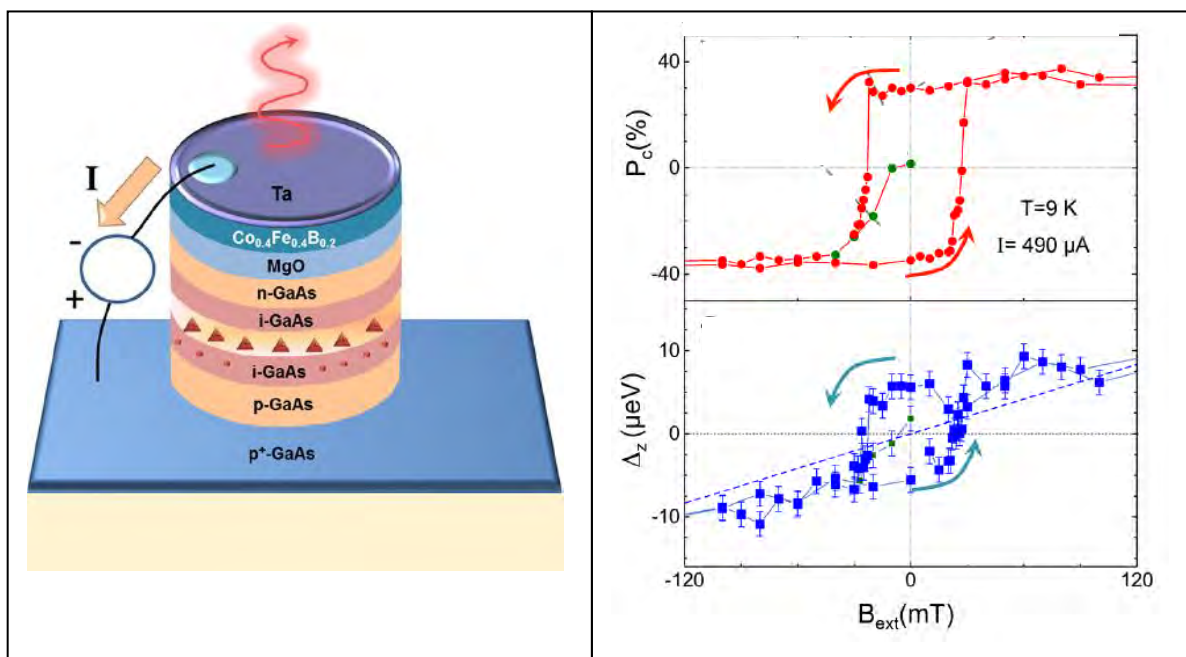


Figure. Left part: scheme of principle of a Spin Light Emitting diode with InAs/GaAs quantum dots. Upper Right part: The circular polarization degree of the electroluminescence (EL) from a single dot is measured as a function of the applied magnetic field B_{ext} . The sweep direction of the field is indicated by arrows. The data points corresponding to the initial magnetization are plotted in green. Lower Right part: the energy difference $\Delta_z = E_{\sigma^+}^{EL} - E_{\sigma^-}^{EL}$ (where $E_{\sigma^+}^{EL}$ and $E_{\sigma^-}^{EL}$ correspond respectively to the spectral position of the EL components right and left circularly polarized) is plotted as a function of B_{ext} . The dashed line corresponds to the linear fit of Δ_z versus magnetic field for larger values of B_{ext} .

From <https://pubs.acs.org/doi/10.1021/acs.nanolett.7b05351>

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- <https://pubs.acs.org/doi/10.1021/acs.nanolett.7b05351>

Angular Dependence of the Spin Photocurrent in a CoFeB/MgO/n-i-p GaAs Quantum well structure

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The ability to convert the information carried by the photon helicity in a spin current that is electrically detectable [1-8] could be of importance for spin-optronic devices. In this work [9], we evidence at room temperature the detection of photogenerated spin currents by using a magnetic electrode without the need of an external magnetic field. The device is based on a semiconductor AlGaAs/GaAs quantum well embedded in a p-i-n junction. The spin filtering is performed owing to a CoFeB/MgO electrode with in-plane magnetization [see Fig upper part]. We observe a helicity-dependent photocurrent when the device is excited under oblique incidence with circularly polarized light. The helicity-dependent photocurrent is explored as a function of the incident and azimuth angles of the incoming light wave vector with respect to the magnetization direction of the magnetic electrode [see Fig lower part]. The results are interpreted as a consequence of the photogenerated average electron spin under oblique incidence in a quantum well governed by optical selection rules involving electron-heavy-hole and electron-light-hole transitions. A systematic study of the helicity asymmetry as a function of the photon energy and applied bias is performed. It demonstrates that this asymmetry is at its maximum close to the GaAs quantum well and (Al,Ga)As bulk optical transitions. The asymmetry can be controlled by an external bias on the structure. Finally, we show that this asymmetry decreases when the temperature increases.

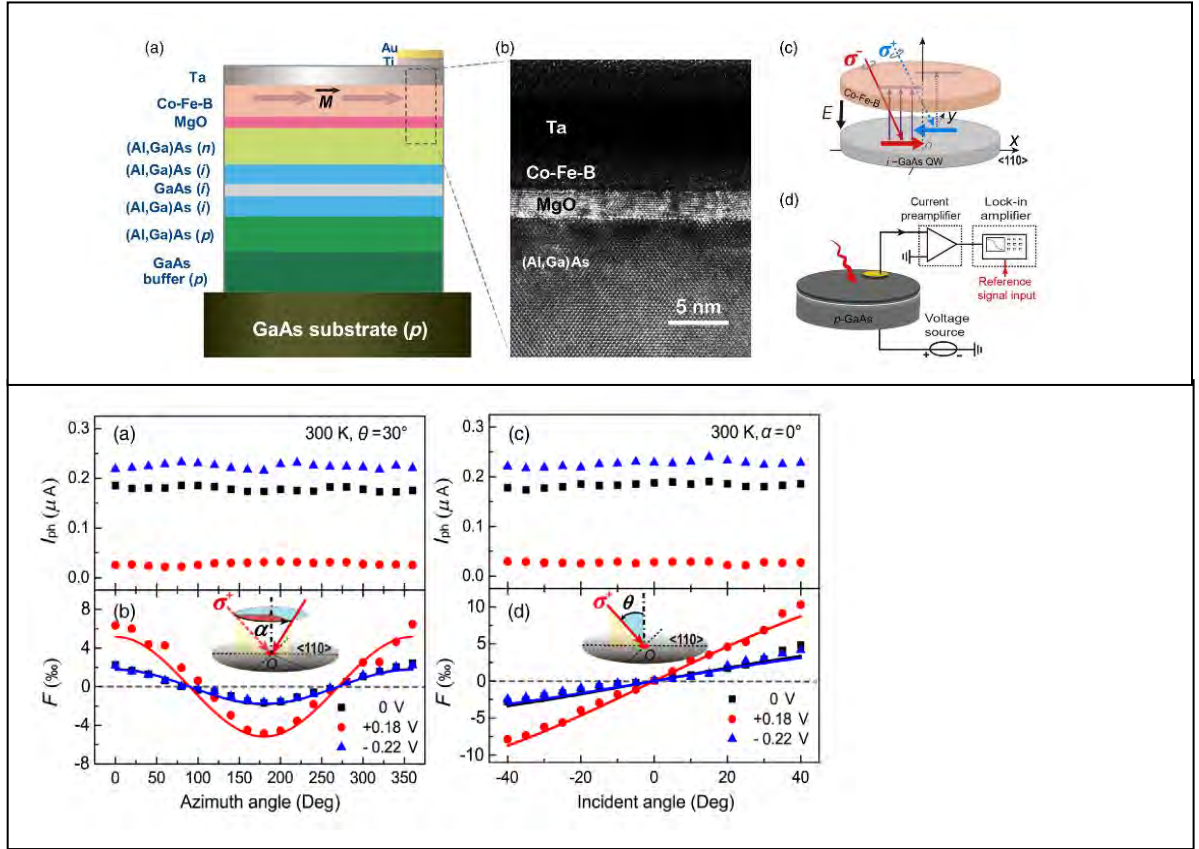


Figure. Upper part: sample structure and photocurrent measurement. (a) The schematic diagram of the cross section of the device. The black bold arrows in Co-Fe-B denote the orientation of the remnant magnetic moment. (b) HRTEM image of the MgO/CoFeB/TaTa spin detector on (Al,Ga)As-based LED structure. (c) Schematic of the difference of photocurrent between right and left circularly polarized light with oblique incidence thanks to a selective spin filtering by the Co-Fe-B/MgO layer. The red and blue horizontal arrows correspond to S_x , the projection along x of the photogenerated average spin S for σ^- and σ^+ , respectively. (d) Schematic diagram of the electrical measurement under bias. Lower part: total photocurrents and helicity asymmetry as a function of the azimuth and incidence angle. (a) The total photo-induced direct currents and (b) the helicity asymmetry of the photocurrents changing with the azimuth angle for the incident angle $\theta = 30^\circ$ under three different bias voltages at 300 K. The solid lines correspond to cosine fitting. (c) The total photo-induced currents and (d) the helicity asymmetry changing with incidence angle with the azimuth angle $\alpha = 0^\circ$ under three different bias voltages at 300 K. The solid lines correspond to the sine fitting. The insets in (b) and (d) illustrate the definition of the azimuth angle and the incident angle, respectively. From Phys. Rev. Applied 8, 064022 (2017)

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Spin precession of holes driven by Rashba spin-orbit coupling confined in the 2D group-IV semiconductor quantum well system

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The spin-orbit interaction (SOI) by Rashba field (RSOI) attracts much interest of physicist, and lots of studies have been carried out for the semiconductor electrons because of the future utilization in spintronics. However, the valence band completeness prevents us to study the RSOI relating phenomena by holes [1]. Here, we discuss the effective magnetic field (EMF) caused by the RSOI (REMF) and study theoretically the dynamics of the spin precession for three types holes confined in the two-dimensional (2D) group-IV semiconductor quantum well (QW) system.

We employ the perturbation approach expanded up to the 2nd-order crossing terms by taking into account both of the particle ($\mathbf{p}$) and crystal ($\mathbf{k}$) momenta. Consequently, two perturbation terms result: H_{SIS} conserves the structure-inversion-symmetry (SIS) and H_{SIA} causes the structure-inversion-asymmetry (SIA). The former includes the perturbations H'_{kp} and H'_{cSOI} (SOI by the crystal potential field), whereas the latter does RSOI (H'_{SIAk} and the cross couplings $H'_{SIAp}-H'_{kp}$ and $H'_{SIAp}-H'_{SIAp}$). The static Rashba field Ξ_0 generates the REMF via the RSOI. However, the REMF driven by the crystal momentum ($\mathbf{k}$ -driven) distributes coaxially in the $\mathbf{k}$ space (Figure. 1(a)), whereas the REMF driven by the particle momentum ($\mathbf{p}$ -driven) distributes anisotropically via the coupling with the $\mathbf{kp}$ term. The generation of the anisotropic REMF is characteristic of semiconductor holes. Moreover, the $\mathbf{p}$ -driven REMF is larger than the $\mathbf{k}$ -driven one, and the REMF works toward the tangential direction of the equi-energy surface (Figure. 1(b)). This REMF further orients the EMF caused by H'_{cSOI} , and resolves the Kramers degeneracy of three types of holes completely. We give the resulting spin polarization of the heavy and light hole along the equi-energy surface in Figure. 1(c) & (d). In the presentation, we report the dynamics of the spin precession by taking into account the inter-hole Coulomb interaction as well as the anisotropic spin polarization focusing on the variation of the 2D quantization and the direction of Rashba field.

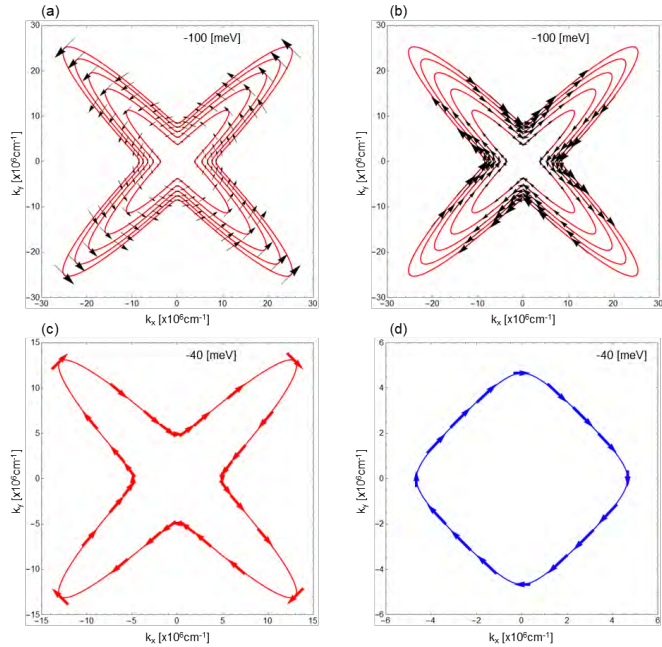


Figure. 1 Distribution of $\mathbf{k}$ -driven (a) & $\mathbf{p}$ -driven (b) REMF and anisotropic spin polarization of heavy (c) and light (d) holes on the equi-energy surfaces.

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Nuclear polarisation in silicon quasi-one-dimensional rings

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We study the first findings of the transmission phase shift (TPS) $\pi/2$ in the $0.7 \cdot (2e^2/h)$ structure of the quantum staircase and in the Kondo-correlated state revealed by the quantum wire which is inserted within one of the arms of the Aharonov-Bohm (AB) ring prepared inside self-assembled silicon quantum well of the p-type. The phase shift in the $0.7 \cdot (2e^2/h)$ structure caused by heavy holes is found to be changed from $\pi/2$ to π by electrically-detected NMR of the ^{29}Si nuclei thereby verifying the spin polarisation in the quantum wire.

The AB double-path interferometer used consists of the self-assembled quantum well (SQW) of the p-type that was formed between δ -barriers by the short-time diffusion of boron from the gas phase into the n-type Si(100) surface. The parameters of the SQW that contains the high-mobility 2D hole gas with hole density $0.8 \cdot 10^{13} \text{ m}^{-2}$ and an elastic free path $l=15 \text{ }\mu\text{m}$ at $T=4.2 \text{ K}$ were defined by the SIMS, STM, cyclotron resonance (CR) and EPR methods. The AB ring was created around the microdefect, $R \approx 2.5 \text{ }\mu\text{m}$, that consists predominantly of the self-interstitials. The source and drain constrictions represent QPC, while the number of the highest occupied mode of the short quantum wire (QW) inserted within one of the arms is controlled by varying the split-gate voltage. The quantum staircase revealed at 77 K is evidence of the heavy hole's tunneling through silicon QW. The experiments are provided by the effective 1D channel length, $0.2 \text{ }\mu\text{m}$, and the QW cross section, $2 \text{ nm} \times 2 \text{ nm}$, which is determined by the SQW width and the lateral confinement due to ferroelectric properties for the δ -barriers heavily doped with boron.

The AB ring of the p-type is prepared on the n-type Si(100) surface to facilitate the Kondo-correlated state created under p^+n junction bias in QW. Since the AB ring's conductance has to be oscillated with a periodicity of a flux quantum h/e when a variable magnetic field threads its inner core, these AB oscillations revealed by the drain-source current seem to be persisted, if transport through the polarised QW is coherent. The phase shift of the AB oscillations measured at specified points on the staircase gives rise to the TPS value of QW. The period mismatch of the AB oscillations from the expected value $\Delta B=0.2 \text{ mT}$ that is observed in weak magnetic fields under the split-gate voltage in the range of the $0.7 \cdot (2e^2/h)$ structure, which causes the extra phase shift equal to $\pi/2$, was found to be suppressed under the saturation of the NMR of the ^{29}Si nuclei. This electrically-detected NMR observed in weak magnetic fields appears to result from the Overhauser shift that is due to the regular magnetic field created by the ^{29}Si nuclei which are polarised by the hyperfine interaction with heavy holes tunneling through QW. The Kondo-correlated state created under reverse p^+n junction bias in QW was observed in studies of Coulomb oscillations. The total phase that results from the AB oscillations is found to be changed by π as two-spin-degenerate levels cross the

Fermi level in leads, whereas the TPS value is equal to $\pi/2$. Thus, the TPS value of the polarised QW, $\pi/2$, that is measured by the NMR of the ^{29}Si nuclei seems to verify a spin polarisation.

Finally, the optical polarisation of the ^{29}Si nuclei induced by circularly polarised light in the n-type substrate was found to effect on the coherent transport through the QW inserted within one of the arms of the AB ring.

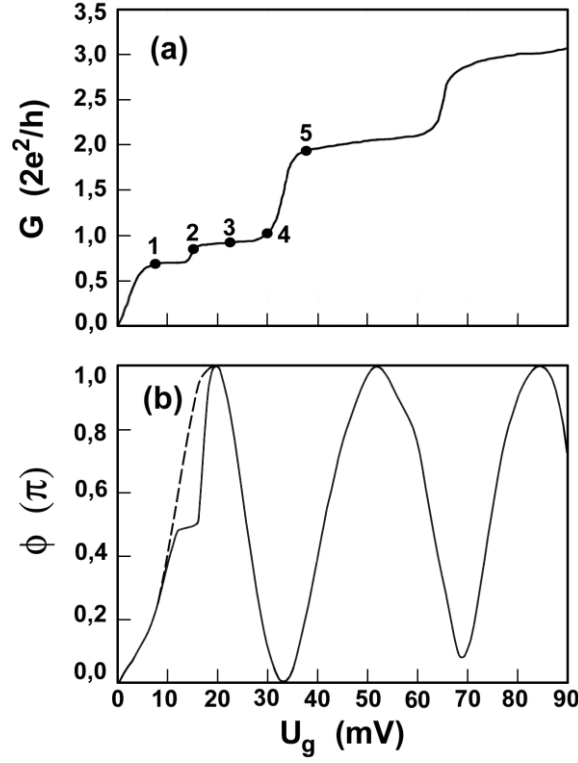


Figure. 1 (a)-The quantum staircase is revealed by heavy holes tunneling through the quantum wire inserted within one of the arms of the Si-based double-path interferometer in zero magnetic field. (b)-The phase of the transmission coefficient of the quantum wire, which is defined by the Aharonov-Bohm (AB) oscillations measured in the range between 0.05 mT and 0.35 mT (solid line) and under the NMR saturation of the ^{29}Si nuclei. $f_0 = 850$ Hz; $H_1 = 5.0 \cdot 10^{-5}$ mT (dash line).

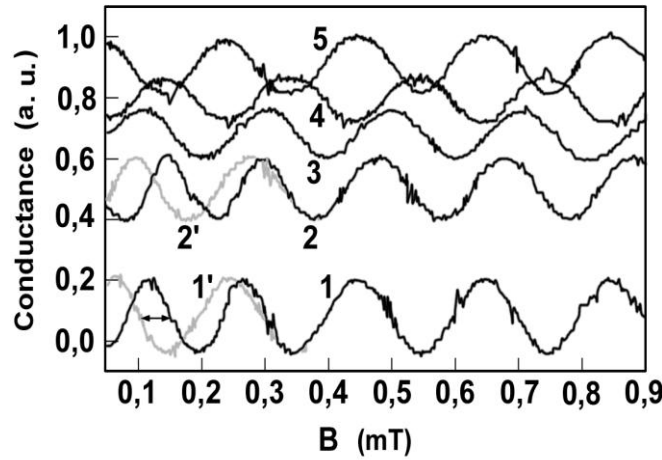


Figure. 2 A series of AB oscillations taken at specified positions on the quantum staircase revealed by the quantum wire (see Figure 1a). The AB oscillations marked as 1 and 2 demonstrate the period mismatch due to the Overhauser shift, which is totally suppressed under the NMR saturation of the ^{29}Si nuclei. $f_0 = 850$ Hz; $H_1 = 5.0 \cdot 10^{-5}$ mT ($1 \rightarrow 1'$; $2 \rightarrow 2'$).

Hyperfine versus exchange interactions in singly Mn-doped InGaAs/GaAs quantum dot

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Semiconductor quantum dots (QDs) doped by a single or few magnetic impurities have been studied in the last decade in order to investigate the exchange interaction between spin carriers in the quantum regime [1-3] and the potential of such system as a solid-state quantum bit [4-8]. In such QDs, the dominant 2-spin interaction is the exchange interaction between the magnetic dopant and the QD-confined hole (up to a few meV). Then comes the electron-hole exchange interaction (~ 0.5 meV) and the exchange interaction between the magnetic dopant and the QD-confined electron (~ 0.05 meV). In this context, the hyperfine interaction of the confined carriers with the few 10^4 nuclear spins of the QD matrix, with typical fluctuations in the μeV range, turns out to be a small perturbation. However, it has been proven to play an essential role for the spin dynamics of a single electron in undoped QDs [9], in particular through the ability, under various experimental conditions, to strongly polarize the nuclear spins. This arises the question whether any such manifestation of the hyperfine interaction between a single electron and the nuclear spin bath can be observed in magnetically doped QDs.

We addressed this issue by focusing on the electron- A^0 system in single InGaAs/GaAs QDs where A^0 represents a neutral magnetic acceptor, with an effective spin $J=1$, provided by a substitutional Mn impurity in the InGaAs matrix [10]. Our investigations of the optical orientation of X^+ trion and of its effective Zeeman splitting reveal that the exchange interaction with the magnetic impurity interferes with the mechanism of dynamical nuclear polarization (DNP) by a spin-polarized electron, leading either to inhibition of DNP or, more surprisingly, to a series of successive DNP sequences when a longitudinal magnetic field is applied, see Figure 1. Thanks to a refined analysis of the Overhauser shift and electron spin orientation, these results can be qualitatively interpreted by invoking two important features of the electron- A^0 exchange interaction: (i) its anisotropy, which induces an effective fast electron spin relaxation preventing any significant DNP, (ii) its longitudinal (Ising-like) strength, which amounts to an effective magnetic field adding or subtracting to the external field depending on the considered ferromagnetic or anti-ferromagnetic electron- A^0 level. Like in the case of undoped InGaAs QDs, a key ingredient is the effective electron spin splitting which determines the energy cost of electron-nuclei flip-flop and subsequently the DNP rate.

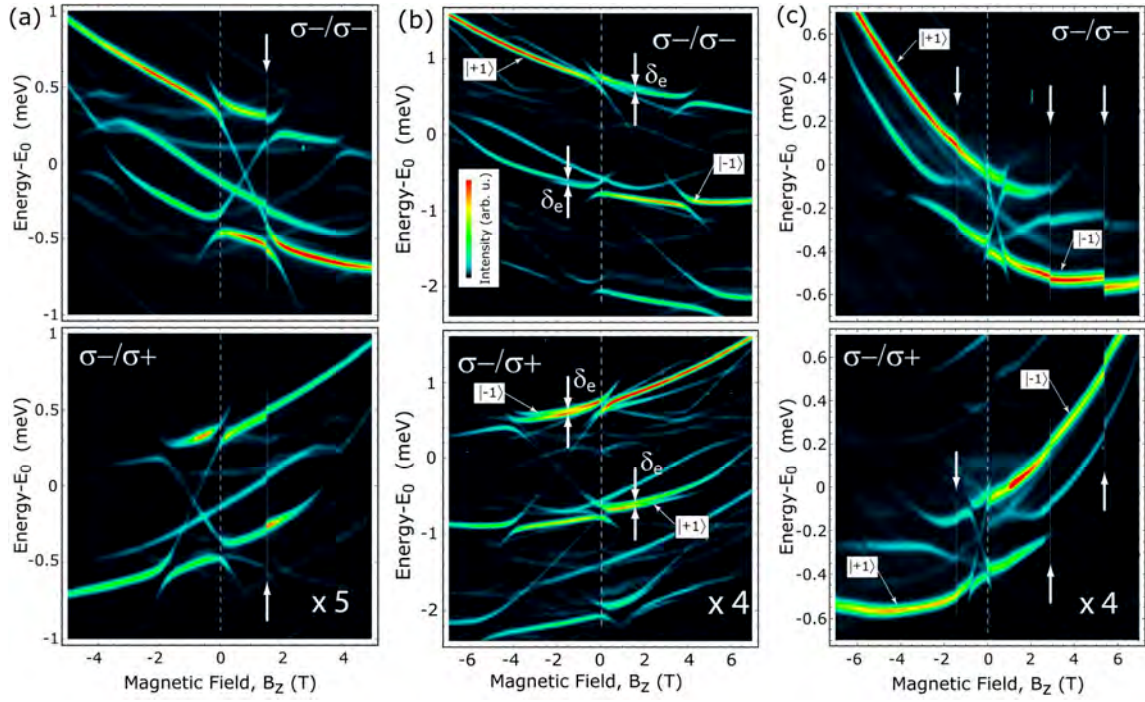


Figure. 1 PL spectra of 3 different Mn-doped InGaAs QDs, under quasi-resonant σ^- excitation as a function of magnetic field B_z , measured in σ^- (top) or σ^+ (bottom) polarization. (a) Moderate exchange with weak anisotropy, 1 DNP sequence (ending by an abrupt shift marked by a vertical arrow), (b) Strong exchange with significant anisotropy giving rise to the spectrally-resolved splitting δ_e responsible for the electron spin relaxation and preventing DNP, (c) Small exchange with weak anisotropy, 3 DNP sequences (marked by 3 arrows).

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Mode-locked electron spin dynamics in many-electron quantum dots

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To perform quantum manipulations with electron spins, a system with a long coherence time is desirable. For electrons in GaAs quantum dots there are two main types of interactions that limit the lifetime and coherence of electron spins. For large quantum dot diameters, the dominant contribution to spin dephasing comes from spin-orbit interaction which scales quadratically with the quantum dot diameter, whereas for a small dot size the hyperfine-interaction is the main mechanism which scales reciprocal with dot diameter. To access an intermediate regime, where the sum of both dephasing contributions has a minimum, lithographically defined quantum dots with variable dot size are fabricated using inductively coupled plasma reactive ion etching of an n-doped GaAs/AlGaAs quantum well (Figure 1). The fabricated dots range from 400 nm up to 1.8 μ m in diameter and contain many electrons each. We measure the electron spin dynamics in an array of nominally identical quantum dots using the time-resolved Kerr-rotation technique and find a spin lifetime that is significantly enhanced in comparison to the two-dimensional electron gas (1 ns) and exceeds the repetition period of the laser ($T_R=12.5$ ns) for the smaller dots, see Figure 2. Remarkably, we find the precession frequency of the electron spins to be locked to the repetition rate of the laser. As can be seen in Fig. 2, the phase of the electron spin precession at a time T_R is independent on external magnetic field, with spin polarization always ending up oriented along the direction of the newly excited spins. This effect is referred to as spin mode-locking and has been observed in singly charged self-assembled quantum dots [1-2]. Explanations of this effect [3-4] rely the trion physics where the spin excitation depends on the orientation of the resident electron. Since our dots contain many electrons we expect this effect to be much less pronounced. Indeed, by depolarizing the nuclear spins of the GaAs host material, we find the spin mode-locking to disappear, indicating that the mechanism for spin locking in our system originates entirely from hyperfine interaction to nuclear spins that focus the electron spin precession frequency to multiples of the laser repetition rate. The dependence of this effect on pump power suggests that a periodic drive of the nuclear spins by the optical excitation is responsible for mode-locking rather than a diffusive fluctuation of the Overhauser field, which asks for alternative explanations to that of nuclear focusing in Ref. [2]. Moreover, we find that under certain conditions, mode-locking can be reversed to an anti-mode-locking where the phase of the precessing spins shows a phase shift of π at the arrival of a new pump pulse (Figure 3).

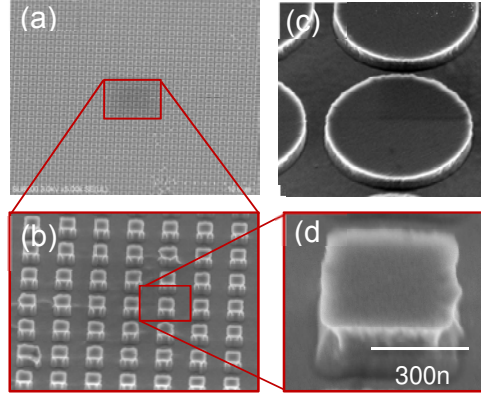


Figure. 1 (a) Scanning electron microscope image of lithographically defined quantum dot array fabricated from a GaAs/AlGaAs quantum well structure. (b) Zoom in of (a). (c)/(d) Fabricated square and round quantum dots.

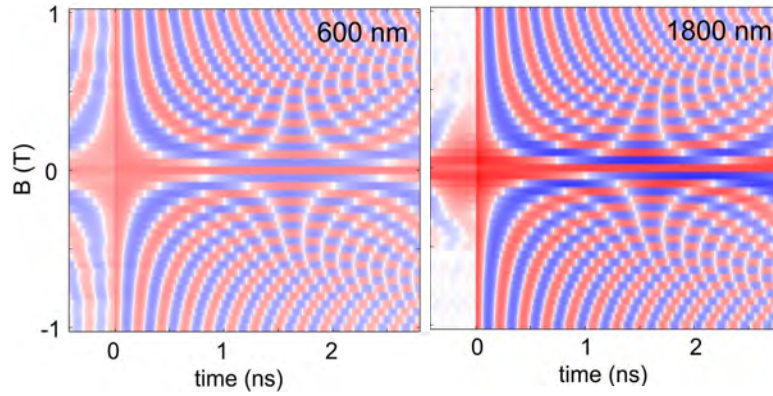


Figure. 2 Time-resolved Kerr-rotation measurements of quantum dot ensemble with dot diameters of 600 and 1800 nm in Voigt geometry. The spin amplitude is color coded and normalized to 1 (red) and -1 (blue).

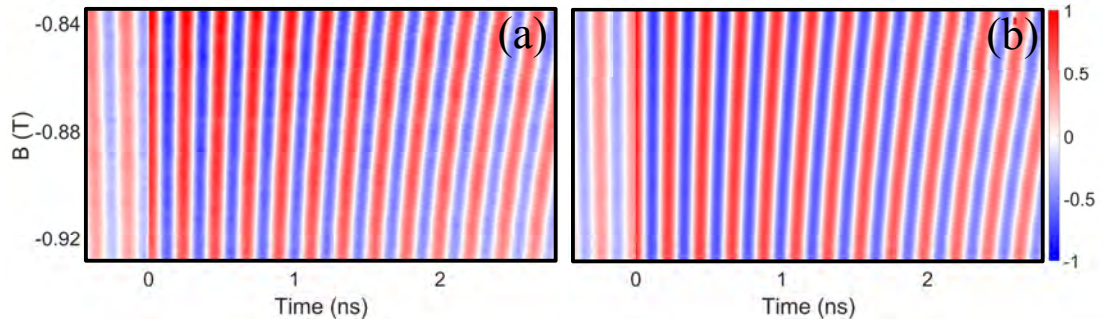


Figure. 3 (a)/(b) Anti-Locking/Locking of electron spin precession in quantum dot ensemble with dot diameter of 400 nm. The spin amplitude is color coded and normalized.

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Electron-Nuclear coherent spin oscillations probed by spin dependent recombination in band-to-band photoluminescence

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Spin-dependent recombination (SDR) via deep paramagnetic centers was uncovered four decades ago by C. Weisbuch and G. Lampel in *AlGaAs* [1] and subsequently studied in *GaAs* by D. Paget [2]. More recently, this effect was the subject of a renewed interest since it has proven to be a quite effective mechanism for obtaining an abnormally high spin polarization degree of free and bound electrons in *GaAs_{1-x}N_x* nonmagnetic semiconductor alloys and *Ga(In)AsN/GaAs* quantum wells up to room temperature [3,4]. The deep center at the origin of this phenomenon in dilute nitrides was identified as an interstitial Ga^{2+} ion [4]. More recently, the role of the hyperfine interaction (HFI) of the centers' electron and nucleus has been the subject of a deeper investigation thanks to the possibility of reaching a strong and efficient nuclear spin polarization of the defect at room temperature by optical orientation experiments [5,6]. Similarly to nitrogen in diamond on one side, and to shallow defects in silicon on the other side, paramagnetic defects in dilute nitrides unite the characteristics of deep and well-isolated paramagnetic centers to an electrically and optically addressable semiconducting system leading *e.g.* to the giant spin-dependent photoconductivity effect [7,8]. In this work we will give a brief survey of the remarkable properties of these defects and discuss the more recent advances on the experimental and theoretical investigations on the HFI in this system, with or without magnetic field [9-13].

Optically or electrically detected magnetic resonance techniques are consistently employed for manipulating and probing the defect spins through the hyperfine interaction, or again to identify the defect chemical nature and related spin dependent recombination pathways. Here, by taking advantage of the remarkable properties of the giant SDR phenomenon, we demonstrate a new scheme for an all-optical investigation of the defect nuclear spins without the need of any magnetic field.

The implementation of a pump-probe photoluminescence experiment leads to the measurement, in the temporal domain, of the hyperfine constant of an electron in strong coupling with a gallium nuclear spin through the HFI by directly tracing the dynamical behavior on the electron-nuclear spin system. The hyperfine constants, defect configuration and the relative abundance of the nuclei involved (Fig. 1) can be determined without the need of electron spin resonance techniques and in the absence of any magnetic field. Information on the nuclear and electron spin relaxation damping

parameters can also be estimated from the oscillations damping and the behavior at long time delay.

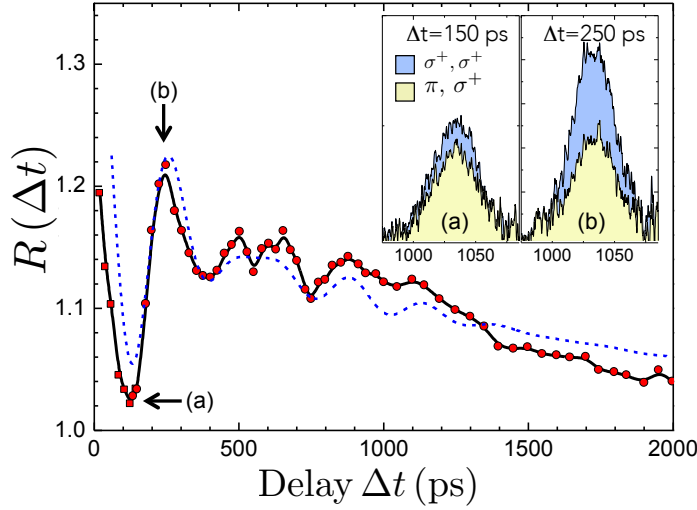


Figure 1. The ratio of the probe pulse photoluminescence intensity under a circularly to linearly polarized pump pulse $R(\Delta t)$ measured as a function of the delay time Δt . The black line is a guide to the eye, while the blue dashed line is a fit to the data according to the modeled developed in [12]. The trace presents an oscillating behavior showing that the PL intensity can be periodically amplified or reduced by adjusting the time delay Δt when the pump pulse is circularly polarized. The oscillation period, beating pattern and decay times extracted from the data allow for the determination of the defect nature, configuration and provide information on the nuclear and electron spin relaxation mechanisms. The inset presents the PL intensities of the circularly polarized probe pulse after a circularly (blue area) or linearly (yellow area) polarized pump pulse at two different time delays (a) and (b).

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Optical Orientation of Excitons in Organic Analogues of Diluted Magnetic Semiconductors

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The selective coupling between polarized photons and electronic states in materials enables polarization-resolved spectroscopy studies of exchange interactions, spin dynamics, and collective magnetic behavior of conduction electrons in semiconductors. At the University of Vermont my research group employs Magnetic Circular Dichroism (MCD) and polarization-resolved photoluminescence (PL) spectroscopy techniques to investigate the excitonic and magnetic properties of electrons in crystalline thin films of small molecule organic semiconductors.

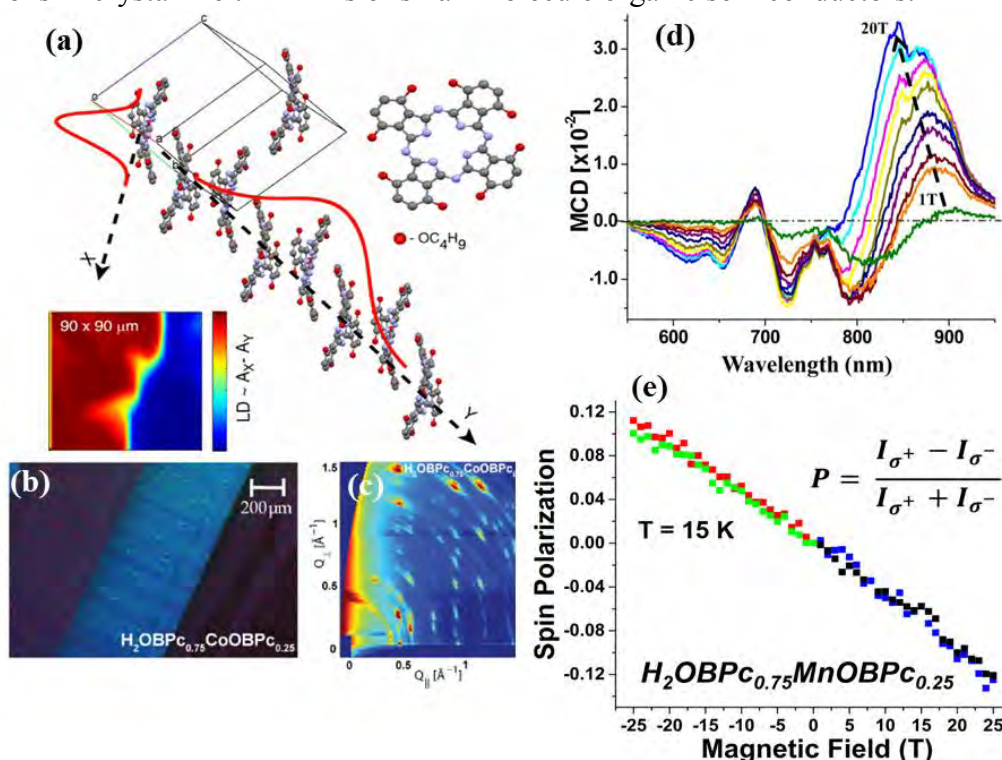


Figure. 1 (a) Schematics of the metal-free octabutoxy phthalocyanine (H₂OBPc) molecule and stacking in crystalline thin films. A delocalized coherent quasi 1D exciton forms along the stacking axis as a result of strong π -orbitals overlap between nearest neighbors. [1] (b) Polarized optical microscope image of crystalline grains in a d-shell ion phthalocyanine alloy : H₂OBPc_{0.75}CoOBPc_{0.25} (c) Grazing incidence X-ray scattering map from the H₂OBPc_{0.75}CoOBPc_{0.25} alloy film reveals the solution-deposited films have superior crystalline quality (d) A series of high field (B < 25T) MCD spectra recorded from a H₂OBPc_{0.75}CoOBPc_{0.25} alloy thin films reveals the bandgap exciton is involved in the magnetic exchange mechanism between Co ions [2,3,4] (e) Spin polarization of bandgap excitons in a H₂OBPc_{0.75}MnOBPc_{0.25} alloy thin film reaches 12% at 25 T and 15K

The focus is on the collective magnetic properties of d-shell ions (Cu^{2+} , Co^{2+} and Mn^{2+}) metal phthalocyanine (Pc) thin films that one may think of as organic analogues of diluted magnetic semiconductors (DMS). These films were deposited in-house using a pen-writing method that results in crystalline films (Figure 1(a), (b) and (c)) with macroscopic long range ordering and improved electronic properties, ideally suited for spectroscopy techniques. [1]

Our MCD experiments reveal that, in analogy to DMS, the extended π -orbitals of the Pc molecule mediate the spin exchange between highly localized d -like unpaired spins. [2] We established that exchange mechanisms involve different electronic states in each species and/or hybridization between d -like orbitals and certain delocalized π -orbitals. More recently, we focused our studies on alloys made of mixtures of Mn^{2+} , Co^{2+} and metal-free (H_2) soluble derivatives of the Pc molecule [3] that offer insight into the possibility of tuning the magnetic interactions in these organic systems. Unprecedented 25T MCD (Figure 1 (d)) and PL experiments (Figure 1(e)) conducted in the unique Florida State Split HELIX magnet at the National High Magnetic Field Laboratory (NHMFL) reveal that we may be able to induce an exchange mechanism that involve different band edge states by varying the mixing ratio. [4, 5] This exchange mechanism involves coherent states that are delocalized over long distances, enabling an RKKY-like behavior for the phthalocyanines. It is very likely that the strong exciton-phonon coupling present in these systems controls the coherence as well as the strength of magnetic exchange. Future work includes molecular species with hydrogen bonding that inhibit this coupling.

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Formation of nuclear field orthogonal to electron spin and nuclear quadrupole effects in self-assembled single quantum dots

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The study of nuclear spin physics in semiconductor quantum dots (QDs) is an active research field currently. This is because the role of hyperfine interaction (HFI), which is the magnetic interaction between a localized electron and the lattice nuclei, is drastically enhanced in QD structures compared with that in bulks and quantum wells due to a strong localization of the electron wave function [1,2]. Actually, a macroscopic nuclear spin polarization (NSP) which is orders of magnitude larger than the value in thermal equilibrium can be generated at cryogenic temperatures, and in turn, the resultant nuclear field (Overhauser field, $\mathbf{B}_n$) up to a few Teslas affects the electron spin dynamics significantly [3]. Because the lattice nuclei act as a reservoir for an optically or electrically injected electron spin, the *engineering* of nuclear spins such as the optical manipulation of the NSP not only leads to the potential applications but also opens up a new spin physics.

The dynamics of NSP is determined by the environment to which the nuclei are exposed. In particular, nuclear quadrupole interaction (QI), which originates from the coupling of a nuclear spin with $I > 1/2$ to the electric field gradients (EFG) has received a lot of attention. Since the lattice strain is used as a driving force for the spontaneous formation process of self-assembled QDs (SA-QDs), the residual strain and the resultant large EFG arise in these QDs. Therefore, the impact of QI is expected to gain considerably and to play key roles for various novel phenomena observed in SA-QDs. Recently, the formation of in-plane nuclear field, $B_{n,x}$ was reported via Hanle effect measurements in single SA-InAs/GaAs QDs [4] where the anomalously broadened curves and hysteretic behaviors were observed, and it seemed to be related also to QI. In addition, the fact that such anomalies in the Hanle curve have not been observed in single droplet GaAs QDs [5] which are free from the internal strain suggests that the strain-induced QI contributes significantly to an anomalous Hanle curve observed in SA-QDs. However, the origin of the anomalies in the Hanle curves has not been revealed entirely. The knowledge of the $B_{n,x}$ formation may lead directly to an optical control of the nuclear field direction, and thus, it is very important.

In this work, we studied the formation mechanism of $B_{n,x}$, which is orthogonal to the photo-injected electron spin $\mathbf{S}$, in the single SA-In_{0.75}Al_{0.25}As/Al_{0.3}Ga_{0.7}As QDs [6]. The PL of a positive trion (X^+) was detected in the (σ^+ , σ^-) basis, and the degree of circular polarization (ρ_c) and the energy splitting between the σ^+ and σ^- PL components (Overhauser shift) were acquired by a single exposure process. In the case of X^+ , the ρ_c is related directly to the z-component of the averaged electron spin $\langle S_z \rangle$ ($= \rho_c/2$).

Figure 1 summarizes the experimental results observed in the individual SA-InAlAs/AlGaAs QDs. The red (blue) curves correspond to the results with increasing (decreasing) transverse magnetic field B_x under σ^- excitation. Fig.1 (a) shows the

observed $\langle S_z \rangle$ (solid lines) and an expected normal Hanle curve (dashed line); the latter is free from the effects of $\mathbf{B}_n$ and has a Lorentzian shape with the full width of ~ 130 mT where the spin lifetime of electron is assumed to be ~ 0.5 ns. The Hanle curves indicate an anomalously large width and hysteretic behavior at the critical magnetic field B_x^c of ± 0.8 T. Such anomalous characters were observed in not only a specific QD but also all the QDs we measured in the same sample. The z -component of $\mathbf{B}_n$ ($B_{n,z}$) deduced from the Overhauser shift decreases gradually with increasing $|B_x|$ as shown in Fig.1 (b). Fig. 1(c) and (d) represent the expected x -components of $\mathbf{S}$ and $\mathbf{B}_n$ which are obtained by substituting the observed $\langle S_z \rangle$ and $B_{n,z}$ to the Bloch equation.

Figure 2 shows the computed results with a proposed model that includes the effects of the nuclear QI: the stabilization of $B_{n,z}$ and the sign inversion between in-plane and out-of-plane components of nuclear g factors. In the calculations, the principal axis of QI is assumed to be the z -axis for simplicity. The model calculations reproduce successfully the characteristics of the observed anomalous Hanle curves, and it indicates that the QI plays key roles in the formation of $B_{n,x}$. While $\langle S_x \rangle$ does not agree with the calculated one, the behavior of expected $B_{n,x}$ agrees well with the computed one. This suggests that another mechanism such as a noncollinear HFI process ($\propto I_x S_z$), which is not included in our model at this stage, might contribute to the $B_{n,x}$ formation in a relatively large $|B_x|$ region. The combination with the standard HFI (i.e., flip-flop process) and noncollinear HFI may improve the qualitative agreement.

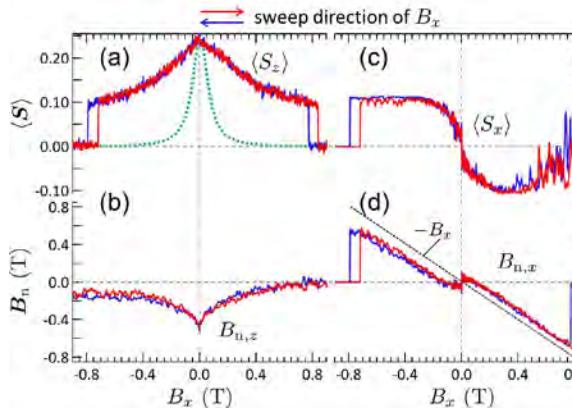


Fig. 1 (a), (b) The observed $\langle S_z \rangle$ and $B_{n,z}$ in a typical InAlAs QD. The dotted line represents the expected normal Hanle curve. (c), (d) The expected $\langle S_x \rangle$ and $B_{n,x}$. The complete cancellation ($B_{n,x} = -B_x$) is indicated by a dotted line.

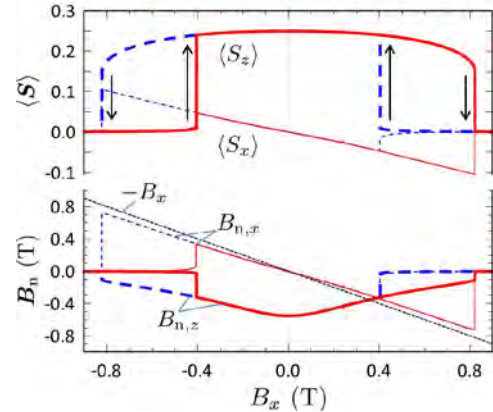


Fig. 2 Model calculations of $\langle \mathbf{S} \rangle$ and $\mathbf{B}_n$ corresponding to the experimental results in Fig. 1. Since $\langle S_y \rangle$ and $B_{n,y}$ are very small, they are not shown here for easy view.

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Quantum control of valley pseudospin in monolayer WSe₂

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2D layered transition-metal dichalcogenide semiconductors [MX₂, M = Mo, W and X = S, Se] have emerged as an exciting platform for valleytronics, a technology that investigates the control over the valley degree of freedom (a local maximum/minimum on the valence/conduction band) of certain semiconductors[1]. A valley degree of freedom termed the “valley pseudospin” is observed in hexagonal MX₂ monolayers (MLs) because of their strong spin-orbit interactions along with inversion symmetry breaking. The manipulation of the superposition states associated with valley pseudospins is one of the important steps toward the application of valley pseudospins in the field of valleytronics.

Here, the ability to control the coherent superposition of the valley pseudospin was demonstrated via the Zeeman effect of the valley neutral exciton states with different valley indices using an external magnetic field applied perpendicular to the ML plane[2], as shown in Figure 1. The coherent superposition rotation of the valley states demonstrates a pronounced manipulation of the valley pseudospins in the applied magnetic field.

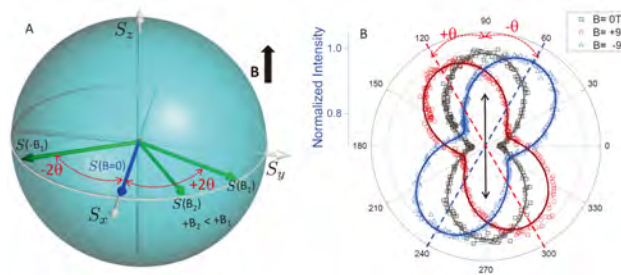


Figure. 1 Control of exciton valley superposition states. (A) Bloch sphere representation of the manipulation of valley superposition states. The valley superposition states can be tuned to different points on the equator by changing the amplitude and direction of the magnetic field in the Z direction. (B) Normalized-angle dependent intensity of neutral exciton emission for $B = 0$ (black), $B = +9$ T (red), and $B = -9$ T (blue) with polar plots. The maximum intensity indicates the direction of the valley superposition states.

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Site-selective measurement of coupled spin pairs in an organic semiconductor

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From organic electronics to biological systems, understanding the role of intermolecular interactions between spin pairs is a key challenge. Here we show how such pairs can be selectively addressed with combined spin and optical sensitivity. We demonstrate this for bound pairs of spin-triplet excitations formed by singlet fission, with direct applicability across a wide range of synthetic and biological systems. We show that the site-sensitivity of exchange coupling allows distinct triplet pairs to be resonantly addressed at different magnetic fields, tuning them between optically bright singlet ($S = 0$) and dark triplet, quintet ($S = 1, 2$) configurations: this induces narrow holes in a broad optical emission spectrum, uncovering exchange-specific luminescence. Using fields up to 60 T, we identify three distinct triplet-pair sites, with exchange couplings varying over an order of magnitude (0.3-5 meV), each with its own luminescence spectrum, coexisting in a single material. Our results reveal how site-selectivity can be achieved for organic spin pairs in a broad range of systems.

<https://arxiv.org/abs/1803.05225> : Sam L. Bayliss, Leah R. Weiss, Anatol Mitioglu, Krzysztof Galkowski, Zhuo Yang, Kamila Yunusova, Alessandro Surrente, Karl J. Thorley, Jan Behrends, Robert Bittl, John E. Anthony, Akshay Rao, Richard H. Friend, Paulina Plochocka, Peter C. M. Christianen, Neil C. Greenham, Alexei D. Chepelianskii

Coherence- and spin-dynamics in high quality GaN/AlGaIn quantum wells

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In the present work, we give an experimental illustration that measuring in parallel both the dephasing and the spin-relaxation rates of excitons gives valuable information to reveal the microscopic mechanisms that govern the spin dynamics in semiconductor nanostructures. We have performed systematic spin- and dephasing-dynamics measurements on narrow linewidth GaN/AlGaIn single quantum wells by means of polarized pump-and-probe[1] and four-wave mixing spectroscopies[2]. Analyzing the decay of the time-integrated four-wave mixing signal at different temperatures and exciton densities enables to determine the homogeneous linewidth broadening γ (also known as dephasing rate) and compare it to the evolution of the spin-relaxation rate with temperature. At low temperature, the homogeneous broadening displays a linear increase when the temperature is raised. This behavior is the typical signature of an exciton interaction with acoustic phonons. In the same temperature range, the spin-relaxation rate remains constant up to 80 K. We conclude that the spin relaxation of excitons, originating from the long-range electron-hole interaction, is affected by motional narrowing. We can even estimate the electron-hole exchange interaction that governs the exciton-spin precession to 0.45 meV at $T = 10$ K, with the frequency of the latter increasing with temperature.

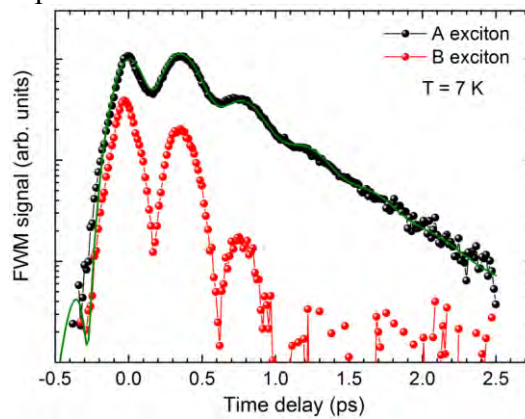


Figure. 1 Time profiles of the four-wave mixing signals for the A- (blackline) and B-exciton (red line) resonances measured at their spectral maxima and plotted as a function of pulse delay. The green line is the best fit to the signal decay of the A exciton.

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Theory of spin inertia in quantum dots

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Over last several decades a number of the new experimental techniques to study the electron spin dynamics, such as pump-probe technique and its extensions, resonant spin amplification and spin noise spectroscopy have been developed [1]. Each of those has certain advantages in specific experimental conditions. However slow electron spin dynamics on the microsecond timescale which is realized in quantum dot structures can be hardly studied by any of these techniques. Only recently, the spin inertia measurement was suggested to resolve this issue [2].

Spin inertia effect is a decrease of electron spin polarization upon modulation of photoexcited spin direction at a frequency ω exceeding an inverse spin relaxation time. Measurement of spin polarization as a function of ω clearly allows one to determine spin relaxation time of resident charge carriers. In this work we develop the theory of spin inertia in self-assembled quantum dots taking into account interplay between the hyperfine interaction with nuclear fields and the Zeeman effect in external longitudinal magnetic field. We considered an ensemble of singly charged quantum dots under pulsed optical excitation with the alternated helicity. The spin polarization of an ensemble is probed by a train of weak linearly polarized probe pulses arriving at a fixed delay with respect to pump pulses. The spin polarization of charge carriers is mapped to the Faraday rotation of the probe light polarization plane or to the ellipticity signal.

Theoretical description of spin inertia in quantum dots is performed using Greens functions formalism for the non-Markovian spin relaxation and accounts for the entanglement between nuclear and electron spins [3, 4]. The microscopic model describes (i) spin precession in external magnetic field and in the Overhauser field of nuclei, (ii) nuclear spin dynamics within the correlation time approximation, and (iii) exponential spin relaxation of resident charge carriers unrelated to the hyperfine interaction. The general expressions for the spin polarization as a function of time and for the spin inertia signal, are obtained.

Resonant optical excitation of charged quantum dots leads to formation of trions. This accordingly to the optical selection rules creates instantaneously spin polarization in the ground state of quantum dots. The trion recombination partially compensates this initial spin polarization. Therefore the total spin generation rate is related to the spin relaxation of trion and resident charge carrier during the trion lifetime. Usually trion spin relaxation give the dominant contribution to the imbalance of photo- and resident-electron spins. In this case the spin inertia signal L can be calculated as

$$L \propto QT_1 \left| \frac{G_{zz}(\omega)}{G_{zz}(0)} \right|, \quad (1)$$

where Q is the trion spin flip probability, $T_1 = G_{zz}(0)$ is an effective spin relaxation time of resident charge carrier and $G_{zz}(\omega)$ is a Fourier transform of the electron spin dynamics Greens function. The first factor in this expression describes the spin polarization efficiency, the second one describes the spin relaxation, and the third multiplier describes the dependence of spin inertia signal on the frequency of polarization modulation ω . This expression shows that the spin inertia directly reveals the absolute value of the spin dynamics Greens function.

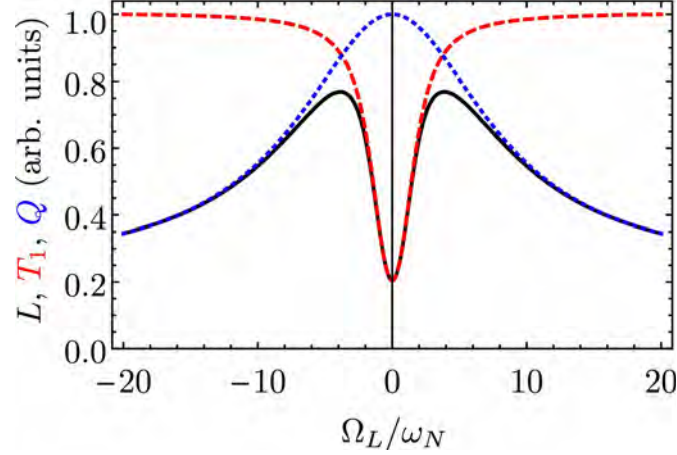


Figure. 1 Spin inertia signal L (black solid line), T_1 (red dashed line) and Q (blue dotted line) as functions of longitudinal magnetic field for zero modulation frequency, $\omega=0$.

Application of an external longitudinal magnetic field along the excitation axis can strongly affect spin dynamics and suppress spin relaxation. Increase of longitudinal magnetic field leads to (i) increase of the average spin relaxation time (T_1) and (ii) suppression of the trion spin relaxation ($\sim Q$). Therefore one can see from Eq. (1), that the spin polarization can have M-like shape as a function of external magnetic field. This effect is illustrated by the black solid curve in Figure 1. The blue dotted curve and red dashed curves in the figure show Q and T_1 , respectively. The M-like shape of spin inertia signal in external magnetic field was indeed observed in the experiments performed in the TU-Dortmund.

Finally we suggest, that the relative phase of spin polarization and pump polarization can be measured, which will provide the complex value of Greens function. This will allow us to obtain directly the time dependence of spin polarization. The developed theory of spin inertia can be applied to any semiconductor system and can serve as a basis for understanding of spin inertia effect in various experimental conditions.

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Spin polarization in $\delta<\text{Mn}>$ -doped InGaAs/GaAs light-emitting diodes

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Magnetic field controlled spin light-emitting diode (SLED) is one of the basic elements for spintronics [1]. Usually, spin-polarized carriers are electrically injected into the SLED active region from a ferromagnetic (FM) layer [1,2], this process is followed by a circularly polarized light emission. The drawback of spin injection is the carriers spin scattering during the transfer from the FM layer to the active region [1]. This is especially crucial for SLED based on hole injection [2]. An alternative is the device where the spin polarization of the carriers is generated directly in the active region due to the interaction with an adjacent ferromagnetic layer [3]. The proximity effect of a ferromagnetic layer on the carriers' spin polarization has been investigated for a number of structures [3-5].

In this work we present a detailed investigation of spin light-emitting diode consisting of an InGaAs/GaAs quantum well (QW) heterostructure with a ferromagnetic $\delta<\text{Mn}>$ layer inserted into the GaAs barrier close to the QW. Circularly-polarized photoluminescence (PL) and electroluminescence (EL) emissions from the diode were studied as a function of temperature and magnetic field.

The structures were fabricated by a two-stage epitaxial growth method. First, a thick n-GaAs buffer layer, 10 nm $\text{In}_x\text{Ga}_{1-x}\text{As}$ QW ($x = 0.16$) and a thin undoped GaAs spacer layer ($d_s = 2 - 10$ nm) were grown subsequently at 650°C on n^+ -GaAs (001) substrates by metal-organic vapor phase epitaxy. At the next stage, the δ -doped $<\text{Mn}>$ layer and the 10-30 nm GaAs cap layer were grown at 380-400°C in the same reactor by laser sputtering of Mn and GaAs targets, respectively. The nominal Mn content in the δ -doped layer (Q_{Mn}) was about 0.1 monolayer. The Au contact was deposited on the top of the sample, and back Ohmic contact was fabricated on the substrate side to form the light-emitting diode structure

PL spectra were measured away from the top Au contacts using a 8 mW He-Ne laser. EL was collected from the back of the samples through the GaAs substrate which is transparent for the QW emission. For EL measurements, the LEDs were forward-biased with an operating current of 3 mA. The external magnetic field was applied along the normal to the QW plane (Faraday geometry). The measurement temperature was 10-100 K.

When a magnetic field is applied, both PL and EL emission become partly circularly polarized. The magnetic field-dependent circular polarization obtained from both photo- and electroluminescence follows the magnetic field dependence of a magnetization of $\delta<\text{Mn}>$ -layer [5]. The example of the magnetic field dependence of the circular polarization degree is shown at Fig.1. The polarization degree (P_{EL}) was calculated as

$$P_{\text{EL}} = \frac{I_+ - I_-}{I_+ + I_-} \cdot 100 \%, \quad (1)$$

where $I_+(I_-)$ are left- (right) circular polarized emission intensity.

Depending on the growth parameters, the EL/PL circular polarization shows an unusual sign inversion that can be explained by an interplay of the Zeeman splitting and Mn-hole interaction effects. Our results can help to understand the origin and to control the spin polarization on Mn doped GaAs structures, a fundamental step for the development of Mn-based spintronic devices.

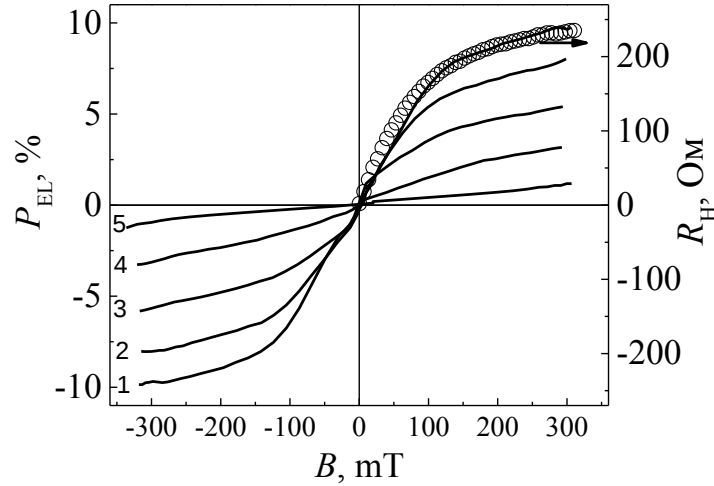


Figure. 1 Magnetic field dependences of the EL circular polarization degree for the InGaAs/GaAs/ δ <Mn> structure measured at a diode current of 3 mA and temperatures of 10 K (curve 1); 20 K (curve 2); 30 K (curve 3); 40 K (curve 4); 50 K (curve 5). The points show the magnetic field dependence of the Hall resistance, measured for a reference single δ <Mn>/i-GaAs structure at a temperature of 10 K.

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Subsecond nuclear spin dynamics in *n*-GaAs

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In semiconductors where lattice nuclei have non-zero spins, the electron-nuclear hyperfine interaction limits the electron spin coherence unless the nuclear spin system is polarized up to a high degree. On the other hand, dynamically polarized nuclear spins can create a strong effective magnetic field, the Overhauser field, B_N , acting upon electron spins. Therefore, control of the nuclear spin polarization by, e.g., time-shaped optical or electric pumping may have application potential [1], and the time scales on which the Overhauser field develops and changes deserve thorough investigation [2].

In this work, we report experimental studies of the nuclear spin relaxation in presence of optical excitation in negatively doped GaAs ($n_D = 4 \times 10^{15}$). Here, we extend results of the previous studies utilizing the synchronous polarization modulation and detection technique, described in Ref. 3. In this case, a cooling of the nuclear spin ensemble was observed under conditions of rapid modulation of the polarization of excitation. The time-resolved photoluminescence study allows us to address a fast subsecond nuclear spin dynamics characterized by time T_N . The nuclear spin relaxation time, T_N , is extracted by the time-of-flight method commonly used for investigation the kinetics of the photoluminescence decays [1,4]. Extrapolation of the subsecond spin relaxation times at a weak power of excitation allows us to conclude on the fast spin nuclear relaxation dynamics of nuclei situated near donor centers, which has not been studied so far.

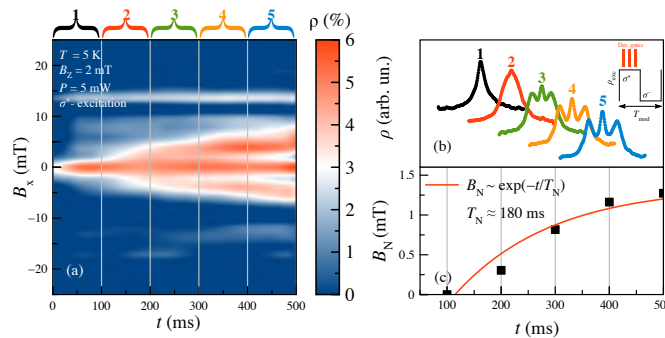


Figure. 1 (a) Colormap of the 2D ToF (Start-Stop experiment) dynamics for modulation of the polarization $f[\sigma^+/\sigma^-] = 1\text{Hz}$ for different B_x and fixed $B_z = 2$ mT. (b) A set of Hanle curves corresponding to different time ranges in the panel (a). (c) Polarization time of the nuclear field B_N .

We show, that the process is governed by the dynamic polarization of nuclear spins subject to an external magnetic field and a time-dependent Knight field [5] of

photo-generated electrons. This encourages also the warm-up of nuclear spin system and relaxation of the non-equilibrium nuclear spin polarization. We find that the optical pumping with the light of alternate helicity induces a fast spin relaxation of donor bound nuclei. This fast spin relaxation time is observed in a subsecond time range of the order of one hundred milliseconds. The experimental results are interpreted within the developed model, which allows to qualitatively describe a drop of the nuclear spin polarization when the light helicity modulation period is of order of one second.

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High-resolution photon echo spectroscopy of optically oriented electrons in CdTe/(Cd,Mg)Te single quantum well

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We demonstrate how the spin-dependent photon echo spectroscopy can be used to evaluate a tiny spin splitting of the ground state. This provides a unique opportunity to distinguish between the various exciton complexes formed from the resident electrons.

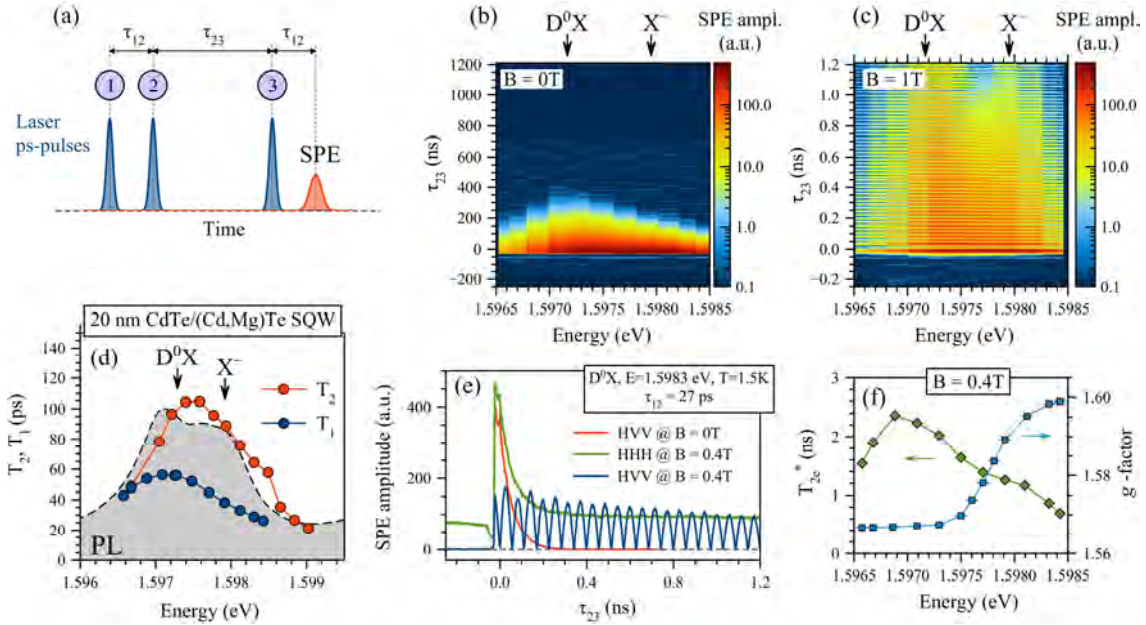


Figure. 1 (a): Timing diagram of photon echo experiment. Stimulated photon echo (SPE) is emitted by QW after excitation by three picosecond laser pulses. (b)-(c): Spectral dependencies of SPE decays measured at B=0T and B=1T, respectively, in HVV excitation geometry. (d): Spectra of PL, optical coherence time T_2 and energy decay time T_1 of studied CdTe/(Cd,Mg)Te SQW. (e): SPE decays measured at energy of 1.5983 eV at B=0T and 0.4T. SPE signal at negative delays τ_{23} actually corresponds to delays $\tau_{23} \approx 12$ ns. (f): Extracted spectral dependencies of g -factor and resident electron spin dephasing time T_{2c}^* . g -Factor experiences a step between optical transitions of D^0X and X^- .

We analyze spectrally resolved dynamics of long-lived magnetic field-induced stimulated (spin-dependent) photon echoes measured from the localized trions (X^-) and donor-bound excitons (D^0X) in a 20-nm *n*-type CdTe/(Cd,Mg)Te single quantum well (SQW) structure subjected to a transverse magnetic field [1]. A sequence of two picosecond laser pulses separated by temporal interval τ_{12} creates a coherent superposition of the ground state doublet which can be retrieved only by optical means due to selective excitation of the same sub-ensemble with a third laser pulse delayed by τ_{23} after the second pulse. In a HVV polarization protocol, when the first exciting pulse is linearly polarized along the magnetic field (H) and two other pulses are orthogonally polarized (V), the oscillations in H-polarized stimulated photon echo (SPE) amplitude are observed when τ_{23} delay is swept on the timescale of a nanosecond [2]. These oscillations correspond to the Larmor precession of the resident electron spin with the frequency proportional to the *g*-factor of the electron spin and magnetic field strength. Variation of the excitation energy in the vicinity of X^- and D^0X optical transitions results in a variation of the Larmor frequency, which manifests a step-like behavior. The tiny sub- μ eV splittings in the Larmor frequency can be resolved at magnetic fields below 1T on X^- and D^0X transitions, which is much less than the homogeneous linewidth of these transitions (about 10 μ eV).

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Electron spin dynamics in an iodine-doped CdTe quantum well

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The dynamics of electrons spins in semiconductors are now becoming increasingly important and researched due to the idea of using spin for processing of information. In particular, single excitons, electrons or holes trapped by individual donors or acceptors in semiconductors at low temperatures are promising spin-qubit candidates due to their combination of advantages of semiconductor and atomic systems [1]. Their semiconductor environment provides a natural localization and ease of fabrication that is absent in atomic and ion qubit systems. Scalability is a common property with other solid-state systems as semiconductor quantum dots (QDs) or nitrogen vacancy centers, but the former ones are rarely identical and the emission efficiency of the latter ones in the zero-phonon line is small (3%).

The distance between electrons localized on donors, R , and the spin exchange interaction $J(R)$ are strongly related to the measured coherence time at low temperature. This exchange interaction, associate either to hyperfine or spin-orbit coupling, will then limit the spin coherence time. In doped bulk (3D) materials, the role of the exchange interaction has been discussed and the spin coherence time as a function of donor concentration has been reproduced, in good agreement with experimental studies in GaAs [2]. Recently, we have considered quasi-2D system, with donors localized in a quantum well, and we have developed a theoretical analysis for this geometry and calculate the exchange interaction $J(R)$, while taking into account confinement and Coulomb interaction.

The studied samples are CdTe/CdMgTe heterostructures, grown by molecular-beam epitaxy on a (100)-oriented GaAs substrate. This kind of systems present some advantages as: (i) the increase of the electron wave function localization, with respect to 3D system; (ii) purified optical selection rules for the circularly-polarized transitions creating excitons bound to donors, D^0X . [3].

We have determined the spin dephasing times as a function of temperature and concentration of donors using a degenerated pump-probe technique, the photo-induced Faraday rotation, which is well-adapted to study spin relaxation and decoherence times of both excited states and resident electrons. We present the theoretical results for this geometry and we compare them with experimental results at low temperature, 2K. At fixed concentration, we observed that as the temperature is increased, the bound electron is thermally ionized and become itinerant; the electronic spins in conduction band interact with localized states by the isotropic term of the exchange interaction and, at the same time, the spin relaxes according to the characteristic processes of itinerant and localized states. [4]

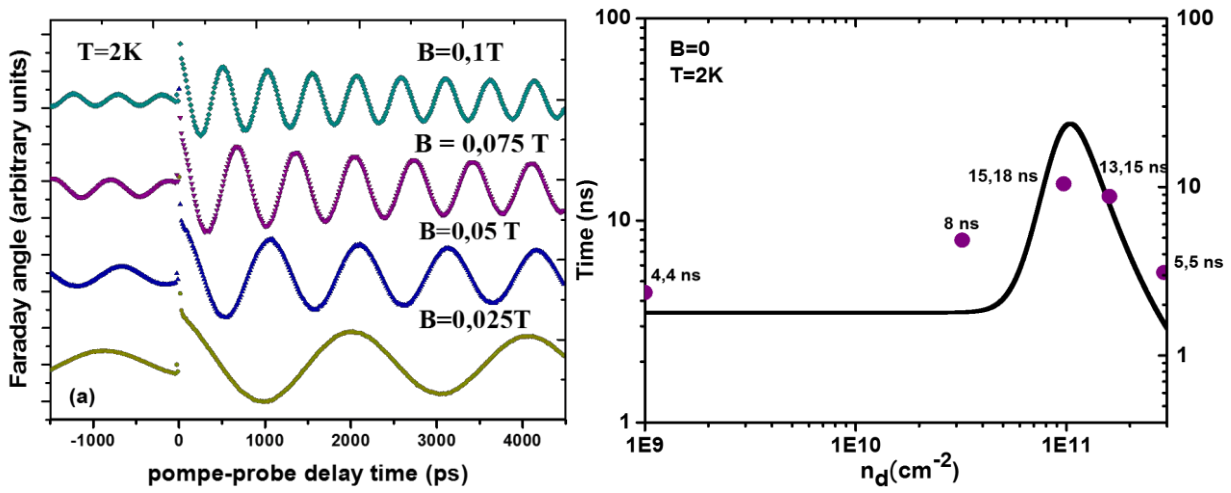


Figure 1 (a) PFR decay obtained at 2K for different magnetic fields at a fixed doping concentration. (b) Decoherence time for different doping concentrations and the theoretical fit.

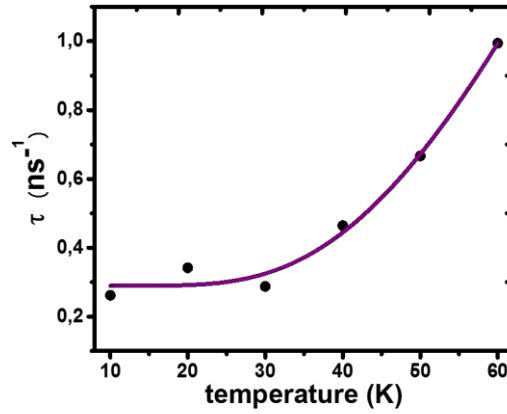


Figure 2. Spin relaxation rate as a function of the temperature. The results are fitted according with the model presented in reference [4]

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Early optical detection studies of spin orientation 1967-1977

My almost continuous failures

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Just after Georges Lampel discovered optical spin orientation in Si by the detection of the Overhauser polarization on nuclei, he gave me the task to optically detect the spin polarization of conduction electrons in Si. I will describe the early attempts, marred by the insufficient understanding of Si PL at low temperatures (spectrum dominated by electron-hole droplet recombination [Haynes1966]) and limited light sources.

The conclusion of this MSc thesis part was: *“The results obtained so far in silicon seem rather thin compared with the many results that seem to make polarized emission through optical pumping or through thermodynamic equilibrium polarization feasible: lifetime, relaxation time, symmetry of the conduction band. However, we have already obtained an interesting result: by the negative result of the experiment of light emission by spins at thermodynamic equilibrium, we can estimate that the relaxation time is of the order of microseconds which corresponds to a linewidth of 50 mG.*

We plan to continue experiments in silicon by improving the experimental conditions: the excitation spectrum for optical pumping could be better defined by placing a monochromator on excitation. The thermodynamic equilibrium signal will be increased by using a large static magnetic field (1kG) and an immersion dewar which allows a temperature of 1.5 °K. We also propose to use soon a Neodyne laser (YAG: Nd) emitting a few watts at 1,06 μm whose coefficient of spectral efficiency is 1 for optical pumping measurements by dynamic polarization or radiative recombination.”

Although these conditions were later met (G. Lampel), attempts to observe polarized light emission from Silicon, even doped, are still not successful, fifty years later!

In an attempt to salvage my MSc thesis [Weisbuch1970], Ionel Solomon led me to perform the detection of optically induced nuclear resonance in the rotating frame (the optical realization of Redfield’s rotary saturation [Redfield1955]). This experiment succeeded quickly but the calibration of the nuclear field gave a number ten times larger than expected for the photo created carrier density. Thus, this result was not published.

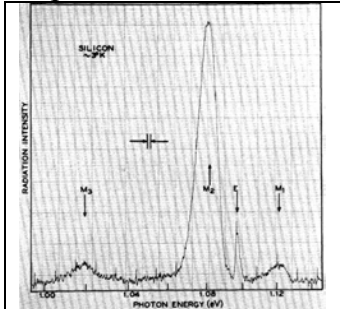
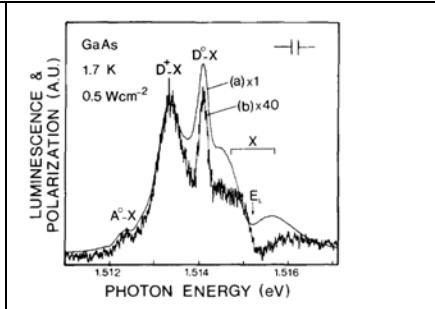
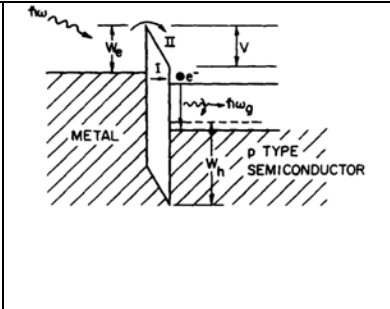
I then undertook the study of optical detection of electronic polarization in the best possible materials system, ultra-pure GaAs. This 8 years long study culminated with an amazing result: for ultra-pure material, at low temperatures, excited resonantly, the observed polarization was exactly zero. In the meanwhile, other obtained large, polarizations in doped materials near the theoretical limit. My studies had however a number of interesting “by-products”: long spin relaxation times at low excitation (~30ns) [Weisbuch 1976], spin dependent recombination [Weisbuch 1974], development of tunable infrared dye lasers, resonant Brillouin scattering of excitonic polaritons, their resonant fluorescence [Weisbuch 1977], determination of the sign of the g-factor of

conduction electrons in GaAs (and the first measurement of g in InP and CdTe), hot electron spectroscopy, the reexamination (with C. Hermann and G. Lampel) of the k.p theory in III-V semiconductor and their alloys...

Other unsuccessful experiments attempted included the optical detection of spin polarized electrons injected from a ferromagnet into a semiconductor [Scifres1973].

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Figure 1: Emission spectrum of electron-hole droplets (M_2 line) in high purity Si ([Haynes1966]; at the time, he attributed the line to biexcitons). This line dominated the early attempts to observe electron orientation through light emission. No spin memory, thus no polarization is expected under such circumstances.	Figure 2: Photoluminescence and polarization spectrum in GaAs, i.e. difference between the two circular components of luminescence. X: exciton-polariton line; D^0X, D^+-X, A^0-X: excitons bound to neutral donors, ionized donors and neutral acceptors respectively. Excitation: 0.5 W cm^{-2} at 1.65 eV. Note the very small degree of polarization at the center of the polariton luminescence. I thought that if excited resonantly, polariton polarization would be large (avoiding the cascade of relaxation of e-h pairs towards the polariton recombining state), but it was then exactly zero! [Weisbuch 1977a]. This was explained as due to the relaxation of the exciton-polariton through its spin-orbit mixed hole wavefunction [Weisbuch1977b].	Detection of tunneling of spin-polarized (ferromagnetic metal) or optically polarized electrons into p-type semiconductor. Such electrons will emit circularly polarized photons when recombining [Scifres 1973]. This is a very early attempt to spintronics. The experiment was attempted in 1975 by myself during a 1 month internship at XEROX park: the metal was MnBi, to have the magnetization perpendicular to the layer, but its spin polarization is only 13%. The GaAs was too lightly doped, with an optically detected spin polarization of only 10-15%. Not surprisingly, no clear spin orientation of electrically injected electrons was optically detected.

The study of excitons and trions in semiconductor nanostructures by means of photon echo techniques.

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The interaction between material excitations (excitons and trions) in semiconductor nanostructures and light can be very effective due to high oscillator strength of the excitations. It was shown [1] that using the photon echo (PE) technique, one can transfer an information contained in the optical field into a spin system of quantum wells (QWs) where it is decoupled from the optical field and may persists for a long time. Semiconductor quantum dots (QDs) are promising for storing an information about the optical excitation since spin relaxation of electrons and holes in these nanostructures is characterized by significantly long times [2]. Recently the difference of excitons and trions behavior in PE studies has been demonstrated experimentally and theoretically [3].

It is interesting to follow the manifestations of exciton and trion spin dynamics in PE studies and its possible applications.

Since the excitons and trions have a different energy level structure and states mixing in external magnetic field (Fig. 1), they contribute to PE signal in a different way.

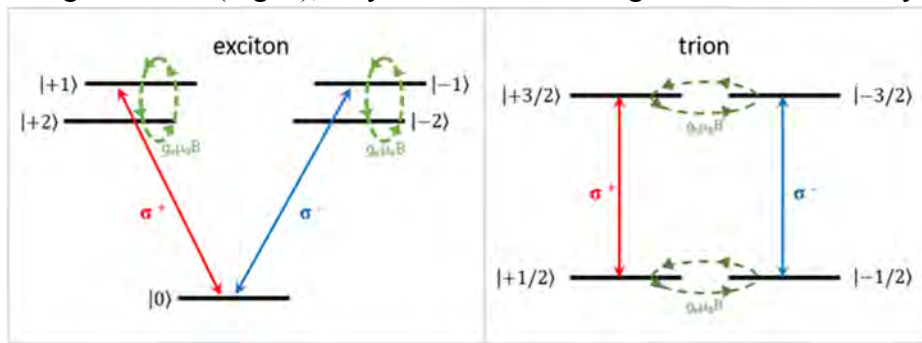


Figure. 1 the schemes of the exciton and trion energy structures and selection rules for excitation with circular polarized light in external magnetic field.

To describe theoretically the ensembles of excitons and trions one should use density matrices and solve the Lindblad equations for the corresponding Hamiltonian. In the experimental study [4] several polarization protocols have been used: the linear polarization of excitation pulses parallel to QD axes x or y (z-axis is the structure grows axis) denoted hereafter as H and V, respectively, and the linear polarization parallel to

the tilted on 45o axes x and y (denoted D and X, correspondingly). The PE signal has been detected in the same technique.

Here are calculated PE signals for exciton ensemble in direct (H and V) polarizations of exciting pulses:

$$P_{HHH}^{exc} \propto e^{-\frac{2\tau_{12}}{T_2}} \left[\cos^2(\Omega_p \tau_1) + \frac{\delta_0^2 \sin^2(\Omega_p \tau_1)}{4\Omega_p^2} \right], \quad (1)$$

$$P_{VVV}^{exc} \propto e^{-\frac{2\tau_{12}}{T_2}} \left[\cos^2(\Omega_m \tau_1) + \frac{\delta_0^2 \sin^2(\Omega_m \tau_1)}{4\Omega_m^2} \right], \quad (2)$$

$$P_{HVV}^{exc} = P_{VHV}^{exc} = P_{HHV}^{exc} = P_{VVH}^{exc} = P_{HVV}^{exc} = P_{VHH}^{exc} = 0, \quad (3)$$

Here

$$\Omega_p \equiv \sqrt{(\omega_L^e + \omega_L^h)^2 + (\delta_0/\hbar)^2},$$

$$\Omega_m \equiv \sqrt{(\omega_L^e - \omega_L^h)^2 + (\delta_0/\hbar)^2}.$$

ω_L and ω_L^h are electron and hole Larmor precession frequencies respectively, τ_1 is the delay time between pulses, δ_0 is an isotropic exchange interaction constant, the exponential factor describes relaxation processes after the pulses action. Indices HHH etc. denote polarization of the first and second pulses and of the detected PE signal, respectively.

Calculated PE signals for trion ensemble in direct polarizations are [1]:

$$P_{HHH}^{tr} = P_{VVV}^{tr} \propto \frac{1}{4} e^{-\frac{2\tau_{12}}{T_2}}, \quad (4)$$

$$P_{HVV}^{tr} \propto e^{-\frac{2\tau_{12}}{T_2}} \cos((\omega_L^e - \omega_L^h)\tau_1), \quad (5)$$

$$P_{VHV}^{tr} \propto e^{-\frac{2\tau_{12}}{T_2}} \cos((\omega_L^e + \omega_L^h)\tau_1), \quad (6)$$

Analysis of the expressions for excitons (1-3) and for trions (4-6) have been made in Ref. [3], and allows us to propose the polarization protocols of optical excitation to separate experimentally the contributions of the exciton and the trion PE signals.

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Exciton, triplet and phonon interaction in a two-dimensional transition metal dichalcogenides

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In monolayer transition metal dichalcogenides, tightly bound excitons have been discovered with a valley pseudospin optically addressable through polarization selection rules [1]. This valley pseudospin is strongly coupled to the exciton center-of-mass motion through electron-hole exchange. Hence, the pronounced electron-hole Coulomb exchange in exciton realizes a strong valley-orbital coupling that can be orders of magnitude larger than the radiative recombination and momentum scattering rates [1]. In such, a strong coupling regime, the pseudospin splitting from the valley-orbit coupling becomes spectrally resolvable. As both the conduction and valence band edges are at the degenerate K and -K valleys, the lowest energy exciton states can be classified by the valley configurations as well as the spin configurations [1-2]. Therefore, the peculiar band structure with its nonequivalent valleys has direct consequences for the trions, which may form optically bright states using carriers located within one valley (in which a hole and two electrons with opposite spins in the same valley form a bound trion state: intra-valley trions T^S) or in different valleys (where the two constituent electrons with same spins are located in different valleys : inter-valley trions T^T) [2,3]. Indeed, because of the different spin configurations, the diagonal exchange interaction present in the inter-valley trions lifts the energy degeneracy and leads to a shift of ~ 7 meV relative to the intra-valley trions [2,3]. Recently, using polarization resolved two-color pump-probe spectroscopy, results [3] show distinct valley polarization dynamics as the resonant pump/probe energy is tuned across the trion resonance and attributed to the two types of trions. They proved that the inter-valley trion polarization exhibits a fast decay of ~ 4 ps. In contrast, the valley polarization associated with the intra-valley trion persists much longer, and decays with a time constant much larger (> 25 ps) than the experimental observation range. A valley depolarization time longer than the population relaxation

time associated with the intra-valley trions means that these trions recombine before valley scattering occurs, leaving the residual electron valley/spin polarized.

In our work motivated by the ref [3] we :i) give a theoretical model to study the optical properties of the inter and intra-valley trion ii) calculate the time decay of the two type of the trion.

Furthermore, in TMDs, the exciton-phonon interaction plays an important role in optoelectronic devices performances. The efficiency of these processes can be enhanced by large optical transition strength in the light matter interaction, or by realizing doubly resonant conditions, meaning both incident and emitted photons are in resonance with electronic transitions whose energy spacing matches an optical phonon. For instance, in monolayer WSe₂, the trion has a binding energy of 30meV, that is, trion (T)- plus a phonon is nearly energy degenerate with the neutral exciton (X₀), resulting in a unique excitonic doubly resonant condition. In fact, since the phonon energy matches the exciton and triplet trion T^T energy separation, while the singlet trion T^s lies at a detuning of 7 meV, rendering phonon scattering inefficient. Moreover, the key source of relaxation for electron spin in TMDs are interactions with phonons. While in-plane phonons do not direct couple to an electron's spin degree of freedom, they may couple indirectly via a spin-orbit induced mixing of the eigenstates. In contrast, out-of plane phonons arising due to the TMDs two-dimensional nature, couple directly to the electrons spin via the creation of ripples and curvature with the TMD sheet. Together these electron-phonon coupling mechanisms provide a number of differing relaxation channels which are necessary to be understood to ascertain the suitability of TMDs as a platform for future spin-qubits. This will form the main focus of our work in which we will calculate the electron spin relaxation time in these materials.

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Electron Spin Relaxation Time in GaAsBi Epilayers and Quantum Well Structures

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The incorporation of small concentrations of bismuth (Bi) into GaAs yields a significant reduction of the band gap energy [1]. As a consequence the dilute bismide alloys, GaAs_{1-x}Bi_x, are interesting for potential optical telecommunication or photovoltaic applications [2,3]. As Bismuth is a heavy atom, the dilute bismides are also characterized by a much larger spin-orbit interaction compared to GaAs [4]. Indeed an increase of the valence band spin-orbit (SO) split-off energy was clearly evidenced in GaAsBi with values reaching ~700 meV and above (*i.e.* twice the GaAs value) for a bismuth composition of about 10% [5]. The role of the SO interaction on the measured and calculated electron *g*-factor in bulk GaAsBi has also been demonstrated [6]. Similar observations were made on exciton *g*-factors in quantum wells in the low temperature range [7]. Moreover, it was shown theoretically that the electron spin relaxation time in GaAsBi decreases drastically when the Bi content increases [8]. The experimental investigation of the electron spin relaxation rate in a bulk sample with a bismuth fraction of 2.2% confirmed these predictions [9]. However a measurement of the bismuth dependence of the electron spin relaxation time is still lacking.

In this work we have measured the electron spin dynamics by time and polarized resolved photoluminescence spectroscopy both in GaAsBi bulk and quantum well (QW) structures with various Bi compositions. The thin bulk layers are 150 nm thick. The investigated quantum well structures consist of 7-10 nm quantum wells capped with a 100nm GaAs barrier [10].

It is well established that the electron spin relaxation in non-intentionally doped III-V quantum wells is dominated by the Dyakonov-Perel spin relaxation mechanism [11-13]. In figure 1 the time-evolution of the PL circular polarization at T=150 K for the three QW structures is displayed. We observe clearly the decrease of the electron spin relaxation time when the Bi content increases: it drops from $\tau_s \sim 280$ ps for $x=2.4$ % down to 40 ps for $x=7$ %. This result is perfectly consistent with the Bi-induced increase of the spin-orbit interaction yielding a more efficient Dyakonov-Perel spin relaxation. However a more quantitative comparison with theory is not possible at this stage since this would require a measurement of the electron momentum relaxation rate in each sample. The PL circular polarization dynamics in GaAsBi samples with x varying from 1.16 % to 3.83 % are displayed in Figure 2 - a (T= 100 K). The drop of the electron spin

relaxation time with the Bismuth content is again clearly measured. In bulk III-V semiconductor the dominant electron spin relaxation mechanism may depend on the temperature and doping concentrations [14]. However it was shown theoretically that for undoped bulk GaAs or GaAsBi, the electron spin relaxation time is dominated by the Dyakonov-Perel mechanism. In particular, for a Bismuth content of 2%, the calculated Dyakonov-Perel electron spin relaxation time is typically two orders of magnitude shorter than the Bir-Aronov-Pikus spin relaxation time, whatever the lattice temperature is in the range 50-300K [8]. Therefore we interpret the measured bismuth dependence of the spin relaxation time in bulk GaAsBi as a consequence of the bismuth-induced increased efficiency of the Dyakonov-Perel spin relaxation mechanism, a similar effect as the one observed for quantum wells.

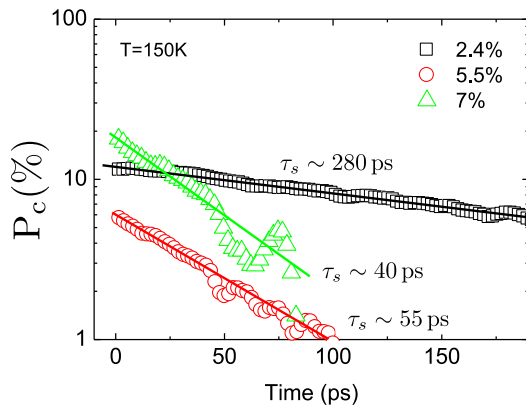


Figure. 1 Photoluminescence circular polarization dynamics in three quantum well structures with different Bi contents ($x = 2.4\%$, 5.5% , 7%). $T = 150$ K. The grey dashed lines are guides to the eye.

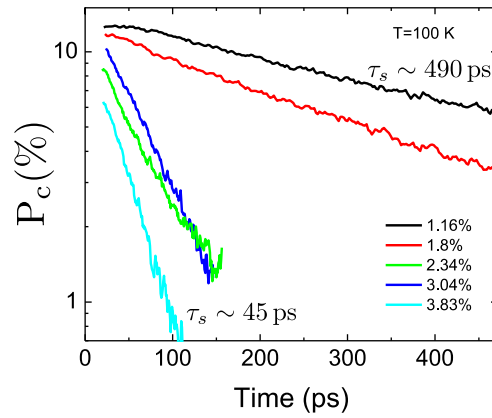


Figure. 2 Time evolution of the photoluminescence circular polarization in bulk $\text{GaAs}_{1-x}\text{Bi}_x$ for $x = 1.16\%$, 1.8% , 2.34% , 3.04% and 3.83% .

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Optical pumping in bulk silicon studied by spin-resolved low-energy photoemission spectroscopy

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Optical orientation of electron spin in semiconductors has been evidenced for the first time in silicon 50 years ago by observing the enhancement of ^{29}Si nuclei polarization under circularly polarized light illumination [1]. Since that time, optical pumping and related techniques have produced a great number of results on conduction electron spin dynamics and on spin injection in semiconductor-based devices. But most of these results have been obtained in direct bandgap semi-conductors. In silicon, although the electron spin relaxation time is known to be large, the situation is not so favourable because of the indirect nature of the band gap, of the weak spin-orbit interaction and, above all, of the large value of the carrier lifetime relative to the spin relaxation time. Thus, very few experimental results have been obtained [2]. From the theoretical point of view the situation is also not very clear and the different approaches developed up to now provide contradictory results [3,4].

Here, we report on the study of optical orientation of electron spin in bulk silicon by spin-resolved low-energy photo-emission spectroscopy. In these experiments, a p-type silicon crystal was activated to negative electron affinity by co-adsorption of caesium and oxygen. In this situation, conduction electrons excited with near-band-gap light can be emitted into vacuum [5]. We then measured their energy distribution and spin polarization under circularly polarized light excitation for photon energies $h\nu$ ranging from 1.17 eV, which gives access to optical transitions at the indirect gap $E_G = 1.12$ eV, up to 3.53 eV, which is slightly larger than the direct gap $E_{DG} \approx 3.30$ eV. We show that, for $h\nu$ very close to the direct gap E_{DG} , the spin polarization of electrons emitted at the maximum excitation threshold is positive (while it is negative in III-V compounds) and reaches values close to theoretical predictions. At the same time, electrons directly excited in the L minimum of the conduction band exhibit a negative spin polarization. In contrast, for excitation energies very close to the indirect bandgap, the spin polarization vanishes even for SOI layers of thickness smaller than the expected spin diffusion length. These results show that, although spin-resolved low-energy photo-emission spectroscopy provides direct information on the physics of conduction electron spin in semiconductors, the optical pumping process in silicon is still not fully elucidate.

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Light-hole exciton in a nanowire quantum dot

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Achieving a good control of the light-hole heavy-hole configuration of the ground state in semiconductor quantum dots is a main challenge: it governs the characteristics of light emission, it conditions their use in quantum information processing, and it determines the spin anisotropy including the magnetic anisotropy if the dot contains a dilute magnetic semiconductor. Usual dots grown by the Stranski-Krastanov method host heavy-holes; one possibility is to apply a well-designed strain, for instance to a dot grown by droplet epitaxy [1]. A proper strain configuration can also be built in if the dot is included in a nanowire.

We have grown such CdTe and (Cd,Mn)Te quantum dots embedded in ZnTe or (Zn,Mg)Te nano-wires, using gold-catalyzed molecular beam epitaxy. The shape, size, and composition of the dots have been determined by x-ray dispersive spectroscopy and by transmission electron microscopy associated to geometrical phase analysis [2].

In an elongated (Cd,Mn)Te quantum dots in ZnTe, the light-hole character is unambiguously demonstrated by a micro-photo-luminescence analysis of the single nanowire, by comparing the polarization and the far-field radiation diagram of the dot (Fig. 1b) and that of the base of the ZnTe nanowire which is HH (Fig. 1a) [3, 4]. This character is confirmed by the value of the giant Zeeman splitting measured on the same single dot under applied magnetic field [4]. The electronic properties of the quantum dots are calculated by solving numerically a 6-band k.p Hamiltonian incorporating the distribution of elastic strain and piezoelectric field

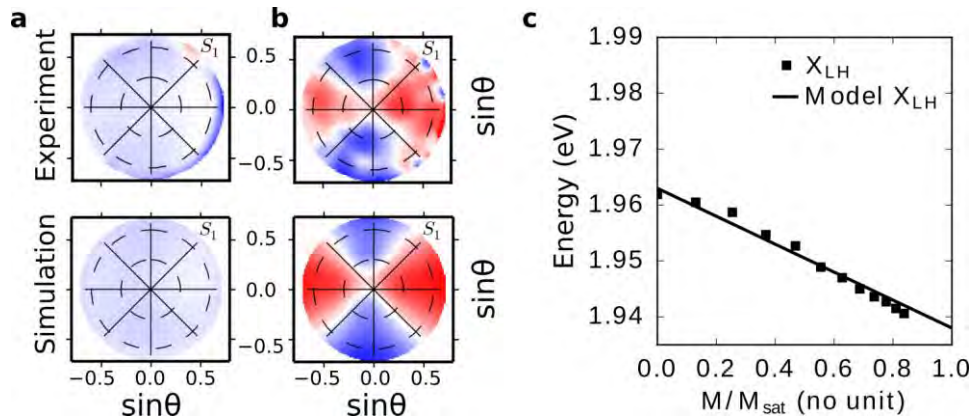


Figure. 1 Fourier plane image of the Stokes parameter of the PL from the ZnTe (a) and (Cd,Mn)Te dot (b). (c) Giant Zeeman shift of the PL emission from the light hole exciton under applied magnetic field.

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Ultralong spin relaxation time of donor bound electrons in n-doped CdTe measured by spin noise spectroscopy

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In the recent years the spectroscopy of spin noise has won its spurs for spin dynamics related studies in semiconductors, largely because it exhibits several quite attractive features. Noticeably it enables almost perturbation-free detection of spontaneous electron spin fluctuations. In addition as the amplitude of spin noise grows when the size of the probed region is reduced, it is well adapted for spatially resolved studies. Also the spin noise spectrum is quite sensitive to internal effective fields, which allows to probe locally the existence of nuclear fields. Finally, the combination of spin noise spectroscopy and optical heterodyning has been demonstrated, which permits enhanced sensitivity and broadband detection, while keeping a high spectral resolution [1].

In this poster we will present results obtained by heterodyne detection of spin noise in an n-doped CdTe epilayer with donor density $n \sim 3 \times 10^{17} \text{ cm}^{-3}$. Thanks to the enhanced sensitivity gained by heterodyne detection we could detect the spin noise of electrons bound to neutral donors for probe powers as low as $5 \mu\text{W}$. In these conditions we observe an extremely slow hopping rate W_0 of electron spin between neighbouring donors, and extremely slow electron spin relaxation rate ν_s (see Figure 1). The observed noise spectrum at zero field is characteristic of electron spin precession in the frozen nuclear field acting on the electrons bound to the donors and exhibits two components. A central lorentzian line corresponding to the electron spin component along the nuclear field, and two satellites gaussian lines corresponding to the spin precession in the nuclear field.

We analyze our results in the framework of a theoretical model, which takes into account both the electron spin precession, and the hopping between donors, but slightly modified to take into account an eventual non-zero nuclear spin polarization [2]. In agreement with the theory we can see that the satellites merge with the central line as the hopping rate increases (at the highest probe power). Surprisingly W_0 and ν_s become quite small at the lowest probe power. We find that the electron spin relaxation time becomes longer than $1 \mu\text{s}$ in this regime.

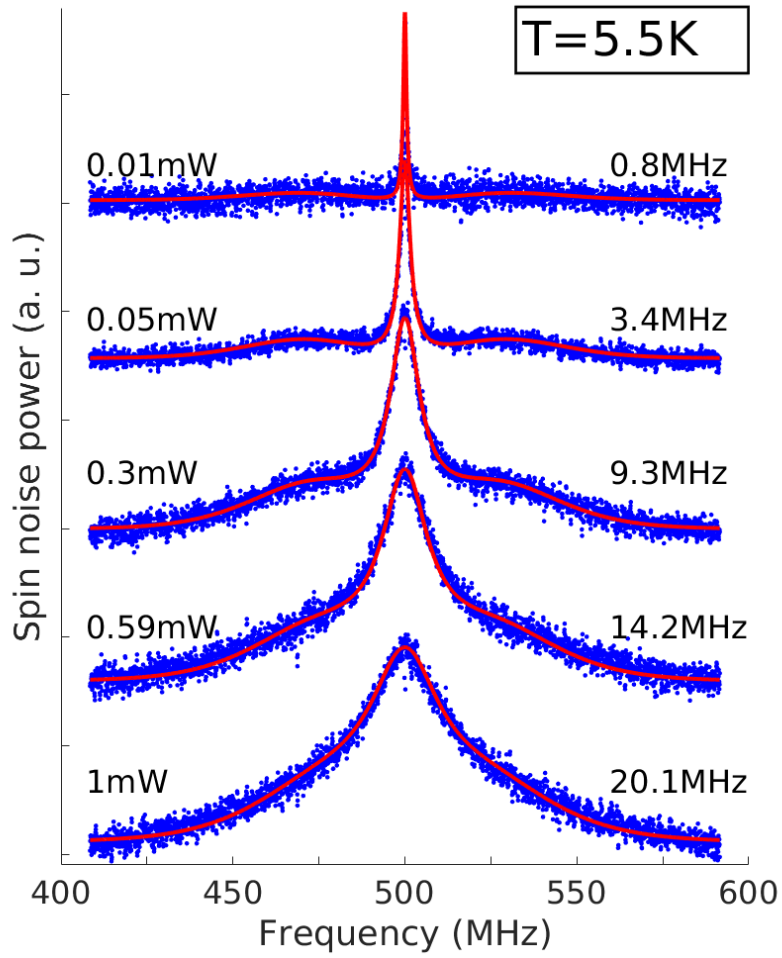


Figure. 1 Series of spin noise spectra measured by the heterodyne technique at zero magnetic field and for different probe powers as shown in the Figure. The frequency difference between the probe and the local oscillator is 500 MHz, and the probe is redshifted from the exciton resonance by 3 meV. The best fits (red curves) are obtained for fixed values of $\delta_e=31$ MHz and $\nu_s=50$ kHz. Only the hopping rate W_0 is varied across the series (values given in MHz in the Figure).

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Nuclear magnetization in gallium arsenide quantum dots: Knight field, nuclear Hanle effect

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Optical and electrical control of the nuclear spin system allows enhancing the sensitivity of NMR applications and spin-based information storage and processing. The nuclear spin dynamics has been formerly widely studied for donors in GaAs, where many effects have been uncovered as *e.g.* dynamic nuclear spin polarization, nuclear Hanle effect, nuclear spin diffusion, quadrupole effects, ...[1,2]. In III-V semiconductor nanostructures, due to enhanced confinement, the electron and nuclear spin dynamics are more strongly coupled [2,3]. This effect is even more pronounced in semiconductor quantum dots (QD), where both spin systems can be controlled optically or electrically in charge tuneable devices [4,5].

In this communication we investigate optical pumping of carrier and nuclear spins in single strain-free GaAs QDs grown on [111]A substrates. Dynamic nuclear polarization (DNP) in semiconductors is commonly achieved in the presence of a stabilizing external magnetic field [1]. We show here that efficient DNP at zero magnetic field occurs through the optical pumping of positively charged exciton X^+ with circularly polarized light (Fig1a). The strong interaction of a single, optically injected electron spin with the nuclear spins acts as a stabilizing effective magnetic field (Knight Field) on the nuclei, whose amplitude is much stronger than the local field, in contrast to the case of donors in GaAs for which the opposite is true. We show that it is possible here to optically tune the Knight Field in amplitude and direction (Fig1b). In nuclear Hanle experiments, using a small transverse magnetic field, we are able to control the longitudinal and transverse components of the nuclear spin polarization in the absence of lattice strain – that is, in dots with strongly reduced static nuclear quadrupole effects – as reproduced by our model calculations (Fig1c). In charge tuneable QDs under quasi resonant excitation, it is possible to tune the DNP both in amplitude (as in InAs QDs [5]), and sign by changing the bias on the device, although the X^+ PL polarization surprisingly does not change sign over this voltage range (Fig. 1d). Voltage control of DNP is further confirmed in Hanle experiments under oblique external magnetic field.

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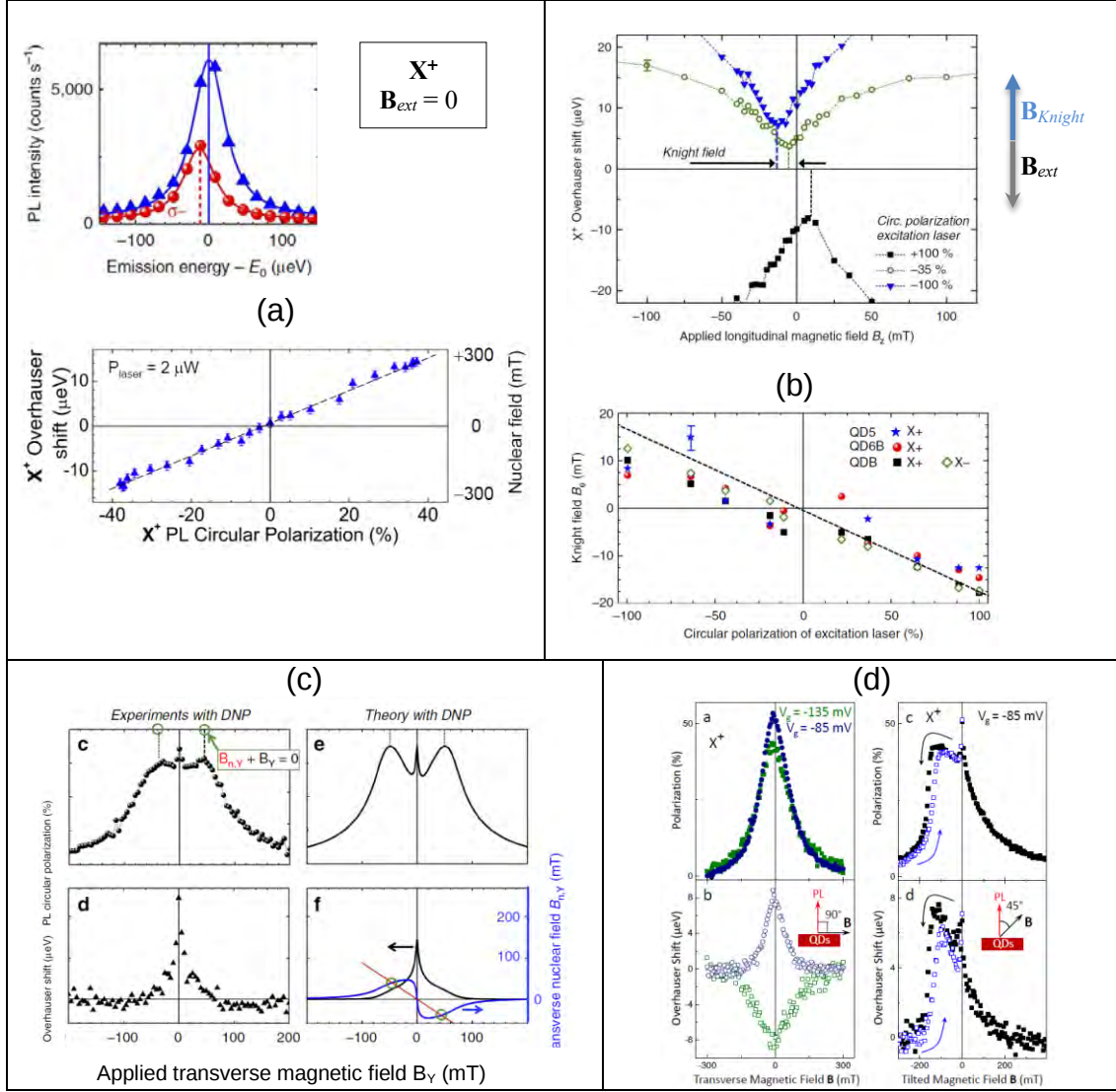


Figure 1 : (a) X⁺ PL spectra σ⁺ (triangles) and σ⁻ (circles) polarized at maximum laser power P = 4 μW (top); Overhauser shift as a function of the measured X⁺ PL polarization with varying laser excitation polarization degree P_{circ} at constant power of 2 mW; the QD is excited non-resonantly. (b) Average Knight Field (B_{Knight}) determination and tuning: the external magnetic field for which a dip occurs is a measure of its amplitude (top); B_{Knight} extracted by Gaussian fitting for three typical dots, demonstrating Knight field tuning. The data show a roughly linear dependence as B_{Knight} ∝ <S_e> ∝ -(P_{circ}/2). (c) Nuclear effects in Hanle experiment with transverse external field (B_{ext,y} ⊥ Oz): when laser power is strong (1.2 μW), X⁺ PL circular polarization presents a non-monotonic decay when increasing B_{ext}, (top left); a strong Overhauser shift is measured, decreasing rapidly as B_{ext,y} increases (down left). Model calculation of the electron spin polarization in X⁺ (top right) and the longitudinal (B_{n,z}, black line) and transverse (B_{n,y}, black line) nuclear field when B_{ext,y} increases (down right). The compensation points where B_{ext,y} + B_{n,y} = 0 are marked by green circles; red line: -B_{ext,y}. (d) Tuning the DNP both in amplitude and sign by changing the bias on the QD device: from -30 μeV (P_n = -22%) to +10 μeV (+7 %) (left); nuclear Hanle Effect under oblique external magnetic field, showing the asymmetry of the electron spin polarization as well as DNP bistability effect.

Optical orientation of exciton states and their complexes in GaAs/AlGaAs [111] quantum dots under magnetic field

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Electron spin orientation and subsequent relaxation in semiconductor nanostructures strongly depend on crystalline symmetry [1,2]. Here we show how in charge tuneable [3] GaAs/AlGaAs trigonal quantum dots (QD) grown on [111]A-oriented GaAs substrates (Fig. 1a), optical selection rules are modified with respect to the usual ones observed for QD grown on a [001] substrate (nanocrystals of C_{2v} symmetry). When a magnetic field is applied along the [111] growth axis, we observe in photoluminescence spectra recorded in Faraday configuration, in addition to the expected bright states, also nominally dark transitions for both neutral (X^0) and charged excitons $X^\pm$ (Fig. 1b). We uncover a strongly non-monotonic, sign-changing field dependence of the bright neutral exciton splitting resulting from the interplay between exchange and Zeeman effects Fig1(c). Our theory shows quantitatively that these surprising experimental results are due to magnetic-field-induced $\pm 3/2$ heavy-hole mixing, an inherent property of systems with C_{3v} point-group symmetry [2,4,5]. The description of the hole-Zeeman effect requires here two independent g -factors: g_{h1} , g_{h2} which can be determined experimentally.

Other exciton complexes were also unambiguously identified, such as negatively or positively “hot” trions states X^{-*} and X^{+*} , thanks to their specific magneto-optics signature. The latter is determined by an interplay between heavy-hole mixing and Coulomb exchange, characteristic for these quantum dots with trigonal symmetry [6]. Our magneto-photoluminescence experiments performed on single quantum dots in the Faraday geometry uncover characteristic emission patterns for each excited electron-hole complex, which are very different from the photoluminescence spectra observed in (001)-grown quantum dots [6,7]. Comparison between experiment and a detailed theory allows for precise charge state identification, as well as extraction of electron-hole exchange interaction constants and g -factors for the charge carriers occupying excited states [6].

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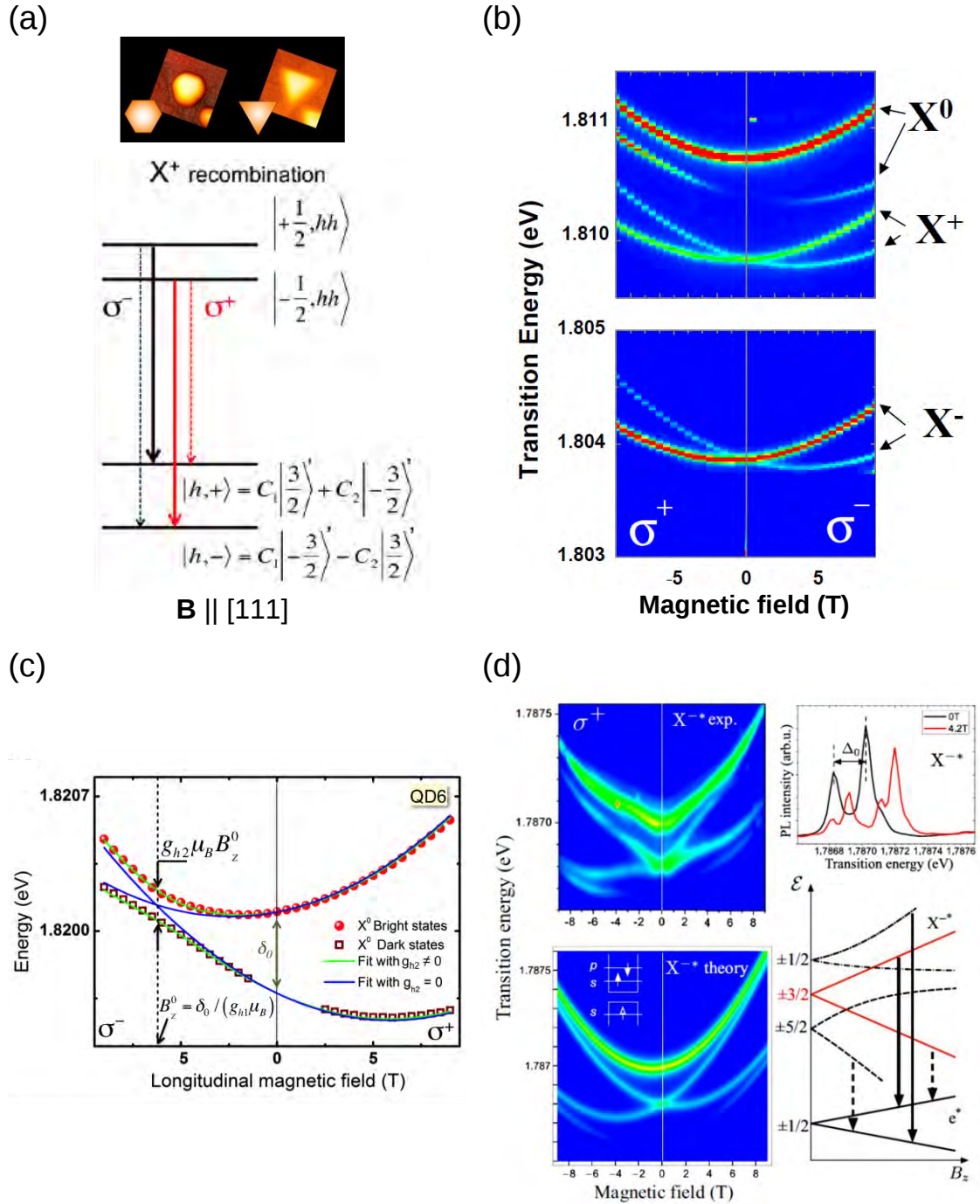


Figure. 1 (a) *Top*: AFM images of GaAs quantum dots grown on (111)A substrates reveal the trigonal symmetry of the dots, with shapes varying depending on crystallization temperature. *Bottom*: Bright and dark states for a typical QD vs. longitudinal magnetic field, showing neutral (X^0) and charged ($X^\pm$) exciton pattern. (b) Scheme of recombination of X^+ with hole states mixed by magnetic field; $C_{1,2}$ are coefficients determining the hole eigenstates under $\mathbf{B} \parallel [111]$. (c) Transition energy of the nominally dark (squares) and bright (circles) X^0 state as a function of $\mathbf{B}$. Left- and right-hand sides represent the states with $s_z = +1/2$ and $s_z = -1/2$, respectively. The green curve is a theoretical fit, taking into account electron-hole exchange, hole g-factors as well as the exciton diamagnetic shift. (d) Theoretical and experimental results for magneto-PL of the X^{*-} complex; *top*: Measured PL pattern in σ^+ polarization (*left*) and measured PL spectra in σ^+ polarization for two values of magnetic field (*right*); *bottom*: theoretical calculation of the PL pattern (*left*); *right*: energy levels and schematic illustration of selection rules in σ^+ polarization. The lower part of the panel shows the energy levels of the p -electron (e^*) which remains after X^{*-} recombination; dashed arrows depict the transitions that become active due to heavy-hole mixing by magnetic field (i.e., $g_{h2}^s \neq 0$).

Does the electron spin in a homogenous, non-magnetic semiconductor diffuse like the charge?

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Since the electron carries both charge and spin, it is natural to think that the two quantities should be transported identically, except perhaps for the tendency of the spin polarization to disappear by spin relaxation and for effects such as the spin Hall effect [1] and the spin helix [2] effects caused by the spin-orbit coupling. The purpose of this work is to show that spin-spin couplings due to the Fermionic nature of electrons in the degenerate limit, or hole-mediated couplings can also result in differentiated spin and charge transport.

For a bulk p-doped ($N_A = 10^{17} \text{ cm}^{-3}$) GaAs sample, we measure the values of charge (D) and spin (D_s) diffusion constants at 15 K [3]. For a photoelectron concentration larger than $3 \times 10^{16} \text{ cm}^{-3}$, the signal is sufficient to use the transient charge and spin grating technique, which consists in creating a transient interference pattern of photoelectron concentration and spin orientation, respectively, at the sample surface. This pattern is probed optically and the dependence of the decay of this pattern as a function of delay gives the charge and spin diffusion constants, respectively [4]. For lower photoelectron concentrations, the sample is excited by a diffraction-limited laser spot, and one monitors the spatial profile of the resulting luminescence intensity and polarization. The former yields information about the charge diffusion while the latter does so for the spin diffusion. With prior knowledge of the minority carrier and spin lifetimes it is then possible to obtain the corresponding diffusion constants. The results are summarized in Fig. 1.

The increase of charge and spin diffusion constants above 10^{16} cm^{-3} is due to degeneracy of the photoelectron gas in which the Fermionic nature of the electrons modifies the usual Einstein relation such that the mobility must be multiplied by a numerical factor larger than unity, related to the spin stiffness. Another consequence of

degeneracy is to cause a spin-dependent diffusion, due to which electrons of majority spin diffuse faster than electrons of minority spin.

The decrease of charge diffusion constant at intermediate electron concentrations is caused by ambipolar effects, due to the electrostatic interaction with more massive holes. Note that ambipolar coupling affects the spin diffusion less than the charge diffusion, so that $D_s > D$ in the whole concentration range covered by Fig. 1. This can be understood by invoking a spin coupling between majority and minority spin electrons which is mediated by the electrostatic coupling with the holes. Consequently, over a large range of photo-electron densities the spin diffusion constant is larger than that of the charge.

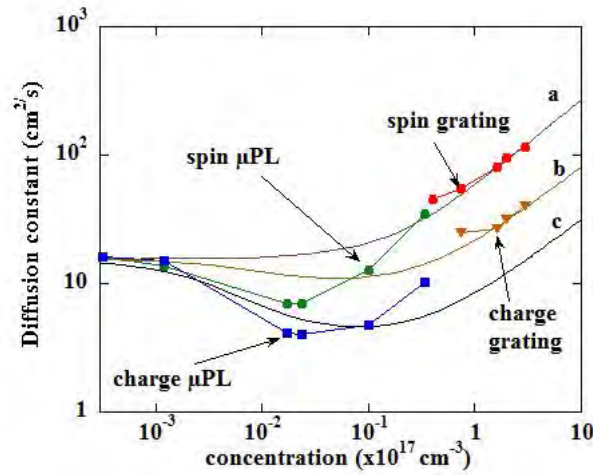


Figure. 1 Values of the charge and spin diffusion constants at 15K on a p-doped ($N_A=10^{17} \text{ cm}^{-3}$) GaAs sample as a function of photoelectron concentration. For $n < 3 \times 10^{16} \text{ cm}^{-3}$, these values are measured using a polarized μPL technique [5], while for higher concentrations, they are obtained using charge and spin grating techniques [4]. In the whole concentration range, the spin diffusion constant is larger than the charge one. The curves show the results of a theoretical model.

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Room-temperature valley contrasting properties in hybrid systems based on atomically-thin semiconductors

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Two-dimensional materials, such as transition metal dichalcogenides (TMDs, here we shall consider semiconducting TMDs with chemical formula MX_2 , with $\text{M}=\text{Mo}$, W , and $\text{X}=\text{S}$, Se , Te), graphene or boron nitride compose a unique toolkit of atomically-thin crystals with remarkable electronic, optical, vibrational, mechanical and topological properties. Monolayer TMDs feature bright direct bandgap emission, unusually strong excitonic effects (even at room temperature) and, crucially, unique spin-valley locked properties. The latter arise from the combination of strong spin-orbit coupling and inversion symmetry breaking. As a result, using circularly polarized light, so-called valley excitons can be selectively formed at well-defined locations in reciprocal space (namely the K and K' valleys, Fig. 1a). Conversely, linearly polarized light makes it possible to form coherent superpositions of valley excitons. Controlling the valley pseudospin (i.e., valley population and coherence) is the essence of opto-valleytronics, a very active field of research [Xu14,Wan18].

The observation of significant valley polarization and coherence in bare TMD monolayers requires stringent experimental conditions such as low temperatures and quasi-resonant optical excitation (Fig. 4c). Indeed exciton-exchange and other depolarization and dephasing mechanisms are prone to suppress valley-contrasting properties on short (ps and below) timescales [Xu14,Wan18]. Several efforts are currently being made to find new schemes, where the valley pseudospin remains addressable under less demanding conditions.

In this paper, we present two complementary approaches to preserve valley contrasting properties up to room temperature. Our general strategy is to take advantage of the 2D nature of TMD monolayers and optimally couple them to either i) other partner 2D materials (including, e.g., graphene and boron nitride) in the form of van der Waals heterostructures or ii) 2D plasmonic fields.

First, we will consider van der Waals heterostructures made from TMD monolayers interfaced to graphene monolayers (Fig. 1b). Owing to highly efficient interlayer energy transfer from the TMD monolayer to graphene [Fro18], the TMD

exciton lifetime becomes comparable to the intervalley scattering time. As a result, large room temperature degrees of valley polarization up to 40% and valley coherence up to 20% are reported in WS₂/Graphene heterostructures. These valley contrasts are assessed through a comprehensive polarimetry analysis based on the Mueller formalism. The TMD/graphene system has recently been hailed as a useful building block for spin-polarized carrier injection in a graphene channel and our results sheds new light on the microscopic mechanisms underlying novel opto-spintronic and opto-valleytronic functionalities.

Second, TMD monolayers can be optimally coupled to plasmonic resonators, where (optical) spin-orbit interactions between the polarization of photons and surface plasmon wavevectors can be induced. This coupling provides a unique capability to map the valley pseudospin onto directional near-field modes, thereby achieving valley-momentum locking. Along this line, we will discuss our recent demonstration of room temperature chiral coupling of valley excitons in a TMD monolayer with (optical) spin-momentum locked surface plasmons [Che18]. At the onset of the strong coupling regime, we measure spin-selective excitation of directional flows of polaritons. Operating under such conditions, our platform yields surprisingly robust valley contrasts and coherence as opposed to their total absence for the uncoupled valley excitons in bare TMD monolayers at room temperature. These results open rich possibilities, easy to implement, in the context of chiral optical networks.

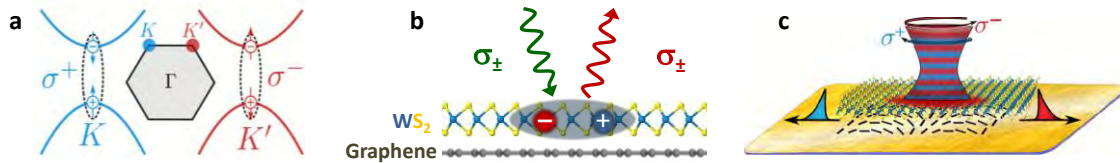


Figure. 1 **a** – Illustration, in the momentum-energy space of spin-valley locking and chiral selection rules in a TMD monolayer. **b** - Sketch of a TMD/graphene heterostructure. **c** Simplified sketch of chiral coupling between valley excitons and spin-momentum locked surface plasmons [Che18].

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Spintronics and exciton optics

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Normally, spintronics studies phenomena that associated with electron spin. In the optical orientation the nonequilibrium electron spin population is created by polarization of the incident photons, and the relaxation of the spin to the equilibrium state is studied. An important stage of these processes is spin relaxation. The classical mechanism of the spin relaxation is the precession mechanism of Dyakonov and Perel. The reason for this relaxation are bulk and interface spin-orbital interactions. However, excitons and photons can also have an angular momentum. Therefore, for excitons and photons, the same spin phenomena can occur as for electrons. In this research we investigate the manifestations of the bulk and interface contribution to the spin-orbit interaction on exciton resonances in quantum wells.

Reflectivity spectra at normal and oblique light incidence are studied in structures with symmetric and asymmetric ZnSe, GaAs and CdTe quantum wells in the range of light and heavy hole excitons. It is found that when the linearly polarized light falls obliquely on a structure with a symmetric quantum well in *p* or *s* polarization (the polarization vector lies or is perpendicular to the plane of incidence, respectively), the reflected light in the exciton range acquires a circularly polarized component. This phenomenon contradicts the Fresnel reflection laws, according to which, when the linearly polarized light falls in *p* or *s* polarization, the reflected light must remain in the same polarizations. It is found that the effect is observed only if the light wave vector **K** has a component directed along the crystallographic axes [100] and [010] and is absent if **K** is directed along [110] and [1-10]. The value of the polarization conversion is very small of the order of 1%. For the exciton with heavy hole, this effect was much weaker, and was observed only due to the interaction between light and heavy hole states.

The observed effect of the polarization conversion in the symmetric quantum well is explained by the manifestation of the spin-orbit interaction due to the bulk mechanism (Dresselhaus contribution), which give contributes proportional to $\alpha(\sigma_x K_x + \sigma_y K_y)$ into the exciton energy in quantum wells.

The manifestation of the interface mechanism of the spin-orbit interaction, (Rashba mechanism), was studied in asymmetric quantum well structures. This mechanism gives contributes proportional to $\beta(\sigma_x K_y - \sigma_y K_x)$ into the exciton energy. To reveal the Rashba mechanism in an asymmetric quantum well, a magnetic field lying in the plane of the structure was applied. In this case, the effect of polarization conversion from a linear to a circular changing sign under inversion of the magnetic field was studied. A comparison of these effects on light and heavy hole excitons made it possible to measure the contributions of Rashba and Dresselhaus on excitons in quantum wells.

Dark-Bright exciton coupling in semiconductor Quantum dots.

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Dark exciton (DE) localized in a semiconductor quantum dot (QD), made of an electron-hole pair with parallel spins, has been demonstrated to reach extremely long coherence lifetimes in the order of $\sim 100\text{ns}$ [1]. As a consequence, DE can be used for developing solid-state quantum storage devices. In principle, DE are not able to emit or absorb photon, however symmetry breaking of the QD confinement potential makes the DE optically active and thus addressable.

The QD symmetry lowering induces a dark-bright coupling responsible of the brightening of the DE. We will present two different experimental strategies for quantifying the dark-bright coupling.

The first one is particularly well adapted for the case of very weak dark-bright, and consequently unobserved DE luminescence under no-applied magnetic field. It is based on the change in the degree of linear polarization of the bright and dark exciton in an In(Ga)As/GaAs quantum dot as a function of a magnetic field slightly tilted from the Voigt geometry. In contrast, the second one is used on highly asymmetric Quantum dot where the DE states luminescence is observable without magnetic field. The amplitude of the anti-crossing between the dark and bright branches developing in Faraday magnetic field, directly reads the Dark-bright coupling.

Furthermore, we developed a model based on the valence band mixing effect and electron/hole exchange interaction to describe the fundamental origin of the dark-bright coupling. This theoretical description is consistent with previous calculation based on sophisticated tight-binding approach [2], which remains unintuitive. We show then that the dark-bright coupling depends on the valence band mixing parameters, offering new experimental way to control the DE optical activity through novel techniques as strain tuning experiments [3]. This experimental work can pave the way to control and enhance the optical activity of DE, making available long-lived coherent states for quantum optics.

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