



### Science Meeting – Scientific Report

The scientific report (WORD or PDF file - maximum of seven A4 pages) should be submitted online within two months of the event. It will be published on the ESF website.

***Proposal Title:** Exploration of ultra-fast timescales using time dependent density functional theory and quantum optimal control theory*

***Application Reference N°:** 5702*

#### 1) Summary (up to one page)

Manipulation of electrons by ultra-short laser pulses opens up the vast and largely unexplored physical landscape of ultra-short time scales. One possibility in this landscape is to use electronic spins, which can be optically manipulated (flipped) using lasers to store data as binary bits. The advantage of such a technique is obvious: reduction in the speed and size of the storage device by orders of magnitude. Recently, great advances have been made in the production of short-time laser pulses and this has led to very fast flipping of spins and modulation of charge in materials. However, we are still far from optimal manipulation and control of spins, needed for device production. Two major reasons for this are (a) the lack of theoretical understanding of the phenomena behind this spin-light interaction induced spin-flip and (b) the lack of unambiguous experimental access to the fundamental spin-dependent processes on the ultrafast timescales of few femto- to attoseconds

The CECAM-Psik workshop was aimed at getting together scientists exploring this field of "ultra-short time-scales" to discuss recent advances and areas of lack of understanding.

## 2) Description of the scientific content of and discussions at the event (up to four pages)

Ultra-fast light-induced demagnetization was demonstrated in 1996, where demagnetization times (in Ni) faster than a few pico-seconds were achieved using intense electromagnetic pulses. Recently, these spin-manipulation times have been reduced to a few femtoseconds, owing to great advances made in the production of short-time laser pulses. However, we are still far from achieving sufficiently controlled manipulation of spins required for production of useful devices. Two main reasons behind this are the lack of full understanding of the phenomena leading to demagnetization and limited theoretical as well as experimental tools to investigate spin dynamics on such ultra-fast time scales.

This field started with two pioneering pump-probe experiments which reported magnetization changes on specific characteristic time scales. The experiments by Beaurepaire et al. investigated Ni and found that the magnetization changes much faster than previously predicted by spin-lattice coupling. This opened a new path of manipulating spins, under non-equilibrium conditions, order of magnitudes faster than at thermal equilibrium. Since then several dedicated pump-probe experiments (including magneto-optical, x-ra, and photo-electron techniques) have demonstrated the same.

However, the underlying fundamental physics still remains a matter of debate. One reason behind this is that the current experimental approaches are essentially limited to response of the material in a few 10s to 100s of fs. In this regime a complex interplay of spin, charge, and lattice vibrations implies a difficulty in unraveling the mystery behind each process separately, which is essential to achieve desired microscopic understanding of the process.

Theoretically, there have been a number of attempts at explaining this optically induced spin-dynamics-- combined action of spin-orbit coupling and interaction between spins and laser photons. Super-diffusive spin transport where excited electrons carry spin with them from one part of the sample to another. Elliott-Yafet mechanism where electron-phonon or electron-impurity mediated spin-flip is the major contributor. However, all these attempts to understand the physics of spin modulations by light have one thing in common-- they are all based on model Hamiltonians. The problem with this is that models based on totally different underlying physics are seemingly able to explain the same experimental data, rendering it impossible to separate fact from fiction. What one requires in such a situation is a fully *ab-initio* theoretical description. Time-dependent density functional theory (TD-DFT), which extends density functional theory into the time domain, is a formally exact method for describing real-time dynamics of charge and spin under the influence of an external field like an applied laser pulse.

Given this situation following questions needed a detailed exploration and were discussed during the workshop:

1. how TD-DFT can be used to understand the phenomenon of demagnetization,
2. how various models can be designed based on highly accurate TD-DFT calculations,
3. how these models can then be used to study large systems,
4. what kind of information can experiments provide in this endeavor,
5. how can optimal control theory be used to control and manipulate spins,
6. how can the time resolution in experiments be pushed to even shorter times scales
7. what are the suitable materials to investigate in order to facilitate microscopic insight.

### 3) **Assessment of the results and impact of the event on the future directions of the field (up to two pages)**

All talks were given 1 hour, however, the speakers were allowed to talk only for 35 mins. A 25 mins long discussion time provided at the end of each talk to discuss problems associated with the field covered by the talk led to many important points being identified.

speakers were specifically requested requested to present not just the fantastic new results, but also problems and brick-walls encountered in their specific area. This lead to many future collaborative developments in this field viable.

The main topic discussed during the workshop were:

1. An overview of the experimental and theoretical developments in manipulation of charge and spins in ultra-short time scales
2. Model Hamiltonians and established experimental approaches
  - 2a What can we learn about the physics at ultra-short times scales from model Hamiltonians
  - 2b What are the crucial open questions considering the experimental information gained so far
3. ab-initio theory and recent experimental developments
  - 3a The ab-initio theory for study of laser induced charge and spin-dynamics, namely time-dependent density functional theory
  - 3b What is the underlying physics described by TDDFT
  - 3c What are the limitations of TDDFT and what is required to overcome these limitations
  - 3d What are the recent experimental developments and how theory can meet these experiments
4. Optimal control of charge and spins
  - 4a How can TDDFT be combined together with optimal control theory to manipulate spins using specially designed laser pulses

4b Can ab-initio calculations provide parameters for model Hamiltonian for studying systems at large length scales.

4c What are promising materials or material combinations and what are the suitable experimental methods to reach controlled manipulation of spins.

**4) Annexes 4a) and 4b): Programme of the meeting and full list of speakers and participants**

## Annex 4a: Programme of the meeting

### Day 1 - September, 28th 2015

- 13:10 to 13:30 - Welcome
- 13:30 to 13:55 - **Martin Aeschlimann** [All-optical control of magnetic order](#)
- 13:55 to 14:20 - Discussion
- 14:20 to 14:55 - **Angel Rubio** [Extending time-dependent density functional theory to account for many body photon-electron quantum phenomena: towards QED-chemistry](#)
- 14:55 to 15:20 - Discussion
- 15:20 to 15:45 - **Andreas Scherz** [Nucleation and transient ferromagnetism in a laser-driven first-order phase transition](#)
- 15:45 to 16:30 - Discussion

Special Lecture by Prof. E. K. U. Gross at EPFL

- 17:00 to 18:00 - Presentation
- 18:00 to 19:00 - Get together

### Day 2 - September, 29th 2015

- 09:00 to 09:35 - **B. Koopmans** [Laser-induced spin-currents versus local dissipation of angular momentum at the femtosecond time scale](#)
- 09:35 to 10:00 - Discussion
- 10:00 to 10:30 - Coffee Break
- 10:30 to 11:05 - **Peter Oppeneer** [Developing ab initio theory for ultrafast laser-induced magnetization dynamics](#)
- 11:05 to 11:30 - Discussion
- 11:30 to 12:05 - **Kazuhiro Yabana** [Time-dependent Density Functional Theory for Strong Laser Pulses in Dielectrics](#)
- 12:05 to 12:30 - Discussion
- 12:30 to 14:00 - Lunch
- 14:00 to 14:35 - **Weinelt Martin** [Different time scales of ultrafast demagnetization - the Gd puzzle](#)
- 14:35 to 15:00 - Discussion
- 15:00 to 15:35 - **Olle Eriksson** [Spin dynamics and density functional theory. formulations and examples](#)
- 15:35 to 16:00 - Discussion
- 16:00 to 16:30 - Coffee Break
- 16:30 to 17:05 - **Alexey Melnikov** [Ultrafast Charge- and Spin-Dynamics Induced by the Transport of Laser-Excited Spin-Polarized Hot Carriers in Metallic Multi-Layers](#)
- 17:05 to 17:30 - Discussion
- 17:30 to 18:05 - **Johan Mentink** [Ultrafast control of the exchange interaction](#)
- 18:05 to 18:30 - Discussion
- 18:30 to 19:45 - Poster Session

*Day 3 - September, 30th 2015*

- 09:00 to 09:35 - **Alexey Kimel** [All-optical magnetization reversal with femtosecond laser pulses: the role of interfaces](#)
- 09:35 to 10:00 - Discussion
- 10:00 to 10:30 - Coffee Break
- 10:30 to 11:05 - **Andrea Eschenlohr** [The Importance of Femtosecond Charge and Spin Transport for Ultrafast Magnetization Dynamics](#)
- 11:05 to 11:30 - Discussion
- 11:30 to 12:05 - **Sangeeta Sharma** [Laser induced ultrafast demagnetization in solids: a time-dependent density functional theory perspective](#)
- 12:05 to 12:30 - Discussion
- 12:30 to 14:00 - Lunch
- 14:00 to 14:35 - **Tobias Kampfrath** [Controlling spin dynamics with terahertz pulses](#)
- 14:35 to 15:00 - Discussion
- 15:00 to 15:35 - **Samir Lounis** [Dynamical Spin Excitations in Itinerant Atoms and Nanostructures](#)
- 15:35 to 16:00 - Discussion
- 16:00 to 16:30 - Coffee Break
- 16:30 to 17:05 - **John Kay Dewhurst** [Kohn-Sham equations for ground state and time-dependent density functional theory of quantum electrodynamics](#)
- 17:05 to 17:30 - Discussion
- 17:30 to 18:05 - **Stefano Sanvito** [Real time time-dependent DFT for spin dynamics of real materials](#)
- 18:05 to 19:30 - Discussion
- 19:30 to 22:00 - Social Dinner

*Day 4 - October, 1st 2015*

- 09:00 to 09:35 - **Uwe Bovensiepen** [Bosonic excitations in high temperature superconductors analysed by femtosecond time- and angle-resolved photoemission](#)
- 09:35 to 10:00 - Discussion
- 10:00 to 10:30 - Coffee Break
- 10:30 to 11:05 - **Esa Rasanen** [Quantum optimal control of ultrafast charge dynamics](#)
- 11:05 to 11:30 - Discussion
- 11:30 to 12:05 - **Alberto Castro** [Controlled ultrafast manipulation of electronic spin and charge in quantum dots](#)
- 12:05 to 12:30 - Discussion
- 12:30 to 14:00 - Lunch
- 14:00 to 14:35 - **Stefan Mathias** [Ultrafast laser-induced charge- and spin-dynamics in alloys and multilayer structures](#)
- 14:35 to 15:00 - Discussion
- 15:00 to 15:35 - **Carsten Ullrich** [The Time-Dependent Particle-Hole Map](#)
- 15:35 to 16:00 - Discussion

- 16:00 to 16:30 - Coffee Break
- 16:30 to 17:05 - **Carlo Andrea Rozzi** [Ultra-fast charge dynamics in photovoltaic materials](#)
- 17:05 to 17:30 - Discussion
- 17:30 to 18:05 - **Martin Eckstein** [Theoretical description of photo-induced dynamics in correlated systems using dynamical mean-field theory](#)
- 18:05 to 18:30 - Discussion
- 18:30 to 19:30 - General Discussion and Conclusion

## Annex 4b: Full list of speakers and participants

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### Organizers

<b>Uwe BOVENSIEPEN</b>	Germany,	Speaker
<b>E.K.U. GROSS</b>	Germany,	Speaker
<b>Sangeeta SHARMA</b>	Germany,	Speaker

### participants

Martin AESCHLIMANN	Germany,	Speaker
Moritz BARKOWSKI	Germany,	poster
Alberto CASTRO	Spain,	Speaker
John Kay DEWHURST	Germany,	Speaker
Martin ECKSTEIN	Germany,	Speaker
Peter ELLIOTT	Germany,	poster
Olle ERIKSSON	Sweden,	Speaker
Andrea ESCHENLOHR	Germany,	Speaker
Isabella FLOSS	Austria,	poster
Reena GUPTA	Germany,	participant
Johan HELLSVIK	Sweden,	poster
Tobias KAMPFRATH	Germany,	Speaker
Alexey KIMEL	The Netherlands	Speaker
B. KOOPMANS	The Netherlands	Speaker
Hyungjun LEE	Switzerland,.	participant
Samir LOUNIS	Germany,	Speaker
Weinelt MARTIN	Germany,	Speaker
Stefan MATHIAS	Germany,	Speaker
Yu-ichiro MATSUSHITA	Germany,	participant
Alexey MELNIKOV	Germany	Speaker
Johan MENTINK	The Netherlands	Speaker
Wolfgang MOSER	Austria,	Poster
Markus MÜNZENBERG	Germany,	Poster
Peter OPPENEER	Sweden,	Speaker
Esa RASANEN	Finland,	Speaker
Ilya RAZDOLSKI	Germany,	Poster
Carlo Andrea ROZZI	Italy,	Speaker
Angel RUBIO	Spain,	Speaker
Leonid SANDRATSKII	Germany,	Poster
Stefano SANVITO	Ireland,	Speaker
Andreas SCHERZ	Germany,	Speaker
Axel SCHILD	Germany,	Poster
Hans Christian SCHNEIDER	Germany,	Poster
Yasushi SHINOHARA	Germany,	Poster
Jacopo SIMONI	Ireland,	Poster
Maria STAMENOVA	Ireland,	Poster
Daniel STEIL	Germany,	Poster
Carsten ULLRICH	USA,	Speaker
Kazuhiro YABANA	Japan,	Speaker