

Jean-Yves Bigot
Wolfgang Hübner
Theo Rasing
Roy Chantrell *Editors*

Ultrafast Magnetism I

Proceedings of the International
Conference UMC 2013 Strasbourg,
France, October 28th - November 1st,
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Editors

Jean-Yves Bigot
Institut de Physique et Chimie des
Matériaux de Strasbourg (IPCMS)
Université de Strasbourg and CNRS
Strasbourg
France

Wolfgang Hübner
Department of Physics
University of Kaiserslautern
Kaiserslautern
Germany

Theo Rasing
Institute for Molecules and Materials (IMM)
Radboud University
Nijmegen
The Netherlands

Roy Chantrell
Department of Physics
The University of York
York
UK

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Preface

This volume on Ultrafast Magnetism is a collection of articles presented at the international “Ultrafast Magnetization Conference” held at the Congress Center in Strasbourg, France, from October 28th to November 1st, 2013. This first conference, which is intended to be held every two years, received a wonderful attendance and gathered scientists from 27 countries in the field of Femtomagnetism, encompassing many theoretical and experimental research subjects related to the spins dynamics in bulk or nanostructured materials. The participants appreciated this unique opportunity for discussing new ideas and debating on various physical interpretations of the reported phenomena. The format of a single session with many oral contributions as well as extensive time for poster presentations allowed researchers to have a detailed overview of the field.

Importantly, one could sense that, in addition to studying fundamental magnetic phenomena, ultrafast magnetism has entered in a phase where applied physics and engineering are playing an important role. Several devices are being proposed with exciting R&D perspectives in the near future, in particular for magnetic recording, time resolved magnetic imaging and spin polarized transport, therefore establishing connections between various aspects of modern magnetism. Simultaneously, the diversity of techniques and experimental configurations has flourished during the past years, employing in particular Xrays, visible, infra-red and terahertz radiations. It was also obvious that an important effort is being made for tracking the dynamics of spins and magnetic domains at the nanometer scale, opening the pathway to exciting future developments. The concerted efforts between theoretical and experimental approaches for explaining the dynamical behaviors of angular momentum and energy levels, on different classes of magnetic materials, are worth pointing out. Finally it was unanimously recognized that the quality of the scientific oral and poster presentations contributed to bring the conference to a very high international standard.

The organization of this conference has greatly benefitted from dedicated people. First we thank our sponsors for making the event possible. Let us mention institutions like the European Research Agency (Research Advanced Grant ATOMAG),

the French Ministry of Education and Research (Equipex UNION and Labex NIE), the National Scientific Research Center in France (CNRS), the University of Strasbourg and the University of Kaiserslautern. We acknowledge our friendly industrial partners who also contributed significantly to the success of the conference: Coherent, Amplitude Technologies, Newport, Fastlite, Zhinst, Femtolasers, Acalbf. The local and international committee had to review 115 contributions which appear to be an excellent up-to-date condensate of the actual research and development in ultrafast magnetism. Our special thanks go in particular to Drs. Nabila Kadiri and Mircea Vomir (CNRS, University of Strasbourg) the organization, the web site as well as the preparation of this volume.

Key words: Coherent magnetic processes · Femtosecond · Magnetic recording · Magnetism · Magneto-Optics · Optics · Spin photo-emission · Spins · Ultrafast Dynamics

Contents

Part I Ultrafast Magnetism Dynamics in Semiconductors

Femtosecond Laser Pulses Switch Magnetic States via Strongly-Correlated Spin-Charge Quantum Excitations	2
Ilias E. Perakis	
Investigation of Non-Thermal Process in the Dynamics of Photo-Induced FMR in (Ga, Mn)As	5
T. Matsuda and H. Munekata	
Time Resolved Spectroscopy in Narrow Gap MOVPE Grown Ferromagnetic Semiconductors	8
G. A. Khodaparast, M. Bhowmick, C. Feeser, B. W. Wessels, D. Saha, G. D. Sanders and C. J. Stanton	
Magnetization Evolution in Semiconductor Heterostructures After Laser Excitation	11
O. Morandi, G. Manfredi and P.-A. Hervieux	
Phase and Spin Relaxation Dynamics in High-Quality Single GaN/AlGaN Quantum Well	14
M. Gallart, M. Ziegler, B. Hönerlage, P. Gilliot, E. Feltin, J.-F. Carlin, R. Butté and N. Grandjean	
Experimental Observations of Optical Spin Transfer and Spin-Orbit Torques in Magnetic Semiconductors	16
P. Němec, E. Rozkotová, N. Tesařová, T. Janda, D. Butkovičová, F. Trojánek, P. Malý, V. Novák, J. Zemen, K. Olejník and T. Jungwirth	

Laser-Induced Spin Dynamics in Ferromagnetic (In,Mn) As at Magnetic Fields up to 7 T	19
R. R. Subkhangulov, H. Munekata, Th. Rasing and A. V. Kimel	
Evolving Magnetization Dynamics in Mn_{3-x}Ga	23
J. M. Wikberg, I. Razzdolski, A. Kirilyuk, Th. Rasing, J. Sadowski, M. Ottoson, Y. Wei and P. Svedlindh	
Part II Ultrafast Magnetism Dynamics in Metals	
Electronic Scattering Dynamics and Ultrafast Magnetization Dynamics	27
M. Aeschlimann, D. Steil, M. Cinchetti and H. C. Schneider	
Influence of the Magnetization Compensation Point on the All-Optical Magnetization Switching	30
L. Le Guyader, I. Radu, A. Eschenlohr, S. El Moussaoui, M. Buzzi, I. Razzdolski, R. Medapalli, M. Savoini, Ch. Stamm, R. Mitzner, K. Holldack, T. Kachel, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, F. Nolting and A. V. Kimel	
Element-Specific Probing of Ultrafast Magnetization Dynamics in the Visible Spectral Range	32
M. Savoini, A. R. Khorsand, A. Kirilyuk, A. V. Kimel, A. Tsukamoto, A. Itoh and Th. Rasing	
Ultrafast Non-local Spin Dynamics in Metallic Bi-Layers by Linear and Non-linear Magneto-Optics	34
A. Melnikov, A. Alekhin, D. Bürstel, D. Diesing, T. O. Wehling, I. Rungger, M. Stamenova, S. Sanvito and U. Bovensiepen	
Balance of Angular Momentum and Magnetization Switching in Ferrimagnetic Alloys	37
Andrei Kirilyuk	
Disentangling Spin and Charge Dynamics with Magneto-Optics	40
E. Carpene, F. Boschini, H. Hedayat, C. Piovera, C. Dallera, E. Puppini, M. Mansurova, M. Münzenberg, X. Zhang and A. Gupta	
Laser-Induced Spin Dynamics in Amorphous NdFeCo	44
J. Becker, I. Razzdolski, A. Tsukamoto, A. Itoh, A. Kirilyuk, A. V. Kimel and Th. Rasing	

Probing Ultrafast Spin Moment Change of Bcc Iron in Crystal-Momentum Space: A Proposal	47
M. S. Si, J. Y. Li, D. S. Xue and G. P. Zhang	
Angular Dependence of Gilbert Damping in Ferromagnetic Metallic Systems	50
E. Barati, M. Cinal, D. M. Edwards and A. Umerski	
Novel Dual-Colour Architecture for Ultrafast Spin Dynamics Measurements in the Sub-10 Fs Regime	53
C. S. Gonçalves, A. S. Silva, M. Miranda, F. Silva, P. Oliveira, H. Crespo and D. S. Schmool	
Spin Dynamics in Rare Earth Doped Cobalt Ferromagnetic Films	56
L. H. F. Andrade, M. Vomir, J. Kim, M. Sanches Piaia, A. D. Santos and J.-Y. Bigot	
Ultrafast Ferrofluids Magnetization Frameworks	59
A. Larionescu, C. Buzduga and C. Ciufudean	
Magnetization Reversal in a Cobalt Nanoparticle	62
G. Klughertz, P.-A. Hervieux and G. Manfredi	
Ultrafast Magnetization Dynamics Driven by Equilibration of Temperatures and Chemical Potentials	65
B. Y. Mueller and B. Rethfeld	
Layer-Specific Probing of Ultrafast Spin Dynamics in Multilayered Magnets with Visible Light	69
Yu Tsema, M. Savoini, A. Kirilyuk, A. Tsukamoto and Th Rasing	
Precession of the Magnetization and Breathing Motion of Assemblies of Co-Pt Nanoparticles	72
Hasan Kesserwan, Valérie Halté and Jean-Yves Bigot	
Laser Heated Ferromagnetic Simulations	76
Raghuveer Chimata, Jonathan Chico, Anders Bergman, Lars Berqvist, Biplab Sanyal and Olle Eriksson	

Part III Spin Waves Dynamics

Excitation and Control of Spin Wave by Light Pulses	80
Takuya Satoh, Yuki Terui, Rai Moriya, Boris A. Ivanov, Kazuya Ando, Eiji Saitoh, Tsutomu Shimura and Kazuo Kuroda	
k-Vector Distribution of Magneto-Static Spin Waves Excited by Micro-Fabricated Antenna Structures	83
H. G. Bauer, J.-Y. Chauleau, G. Woltersdorf and C. H. Back	
Spin-Wave and Spin-Current Dynamics in Ultrafast Demagnetization Experiments	86
M. Münzenberg	
Novel Optical Properties of Spin-Wave Excitations in Non-Centrosymmetric Oxides: The Case of Ba₂CoGe₂O₇	89
Sándor Bordács and Yoshinori Tokura	
Nano-Orbitronics in Silicon	92
B. N. Murdin, K. Litvinenko, Juerong Li, E. Bowyer, M. Pang, P. T. Greenland, B. Villis, G. Aeppli, A. F. G. van der Meer, B. Redlich, H. Engelkamp and C. R. Pidgeon	
Evanescence Exchange Magnons in a 1D Magnonic Crystal	94
M. Pereiro, C. Etz, L. Bergqvist, A. Bergman and O. Eriksson	
Magneto-Optic Study of Picosecond Magnetization Dynamics in Garnet Films	98
M. V. Logunov, S. A. Nikitov, M. V. Gerasimov, A. V. Spirin and A. V. Balyasov	
Spin-Polarized Electron Scattering in Permalloy Films: A Spin-Wave Study	100
Mohammad Haidar and Matthieu Bailleul	
Spin-Wave Modes in a CoFeB Magnonic Crystal Waveguide	103
M. Mansurova and M. Münzenberg	
Laser-Induced Giant Skyrmions and Skyrmion-Compounds in a Thin Magnetic Film of TbFeCo	106
M. Savoini, M. Finazzi, A. R. Khorsand, A. Tsukamoto, A. Itoh, L. Duò, M. Ezawa, A. Kirilyuk and Th. Rasing	

Part IV Theory of Spin Dynamics

Theory of Femtosecond Laser-Induced Demagnetization 111
 Karel Carva, Marco Battiato, Dominik Legut and Peter M. Oppeneer

**Relaxation Dynamics of Majority and Minority Electrons
 After Ultrashort Laser Excitation** 116
 B. Y. Mueller, M. Cinchetti, M. Aeschlimann, H. C. Schneider
 and B. Rethfeld

**A Local Approach to Ultrafast Magnetization Dynamics
 in Ferromagnetic Transition Metals** 120
 W. Töws and G. M. Pastor

**Ultrafast Quenching of the Exchange Interaction
 in a Mott-Insulator** 123
 Johan H. Mentink and Martin Eckstein

**Spin Dynamics and Exchange Interactions from
 the First- and Second-Principles Calculations** 126
 Mikhail I. Katsnelson

Λ -Processes Induced by Chirped Lasers 128
 G. Lefkidis and W. Hübner

**Ultrafast Demagnetization After Laser Pulse Irradiation
 in Ni: Ab Initio Electron-Phonon Scattering and Phase
 Space Calculations.** 131
 Christian Illg, Michael Haag and Manfred Fähnle

Ultrafast Spin Flip on Homodinuclear Clusters 134
 W. Jin, C. Li, G. Lefkidis and W. Hübner

Switching Dynamics of Two Sub-lattice Magnets. 137
 Sönke Wienholdt and Ulrich Nowak

The Landau-Lifshitz-Bloch Equation for Quantum Spin 140
 P. Nieves, D. Serantes and O. Chubykalo-Fesenko

**Inertial Regime of the Magnetization: Nutation resonance
 Beyond Precession Resonance** 143
 J.-E. Wegrowe, M. Meyer, M. Hayoun and E. Olive

Multiscale Modeling of Ultrafast Magnetization Dynamics	146
T. A. Ostler, J. Barker, R. F. L. Evans, U. Atxitia, R. W. Chantrell, O. Hovorka and O. Chubykalo-Fesenko	
What Can We Learn About Magnetization Dynamics from First-Principles Calculations?	150
Paul J. Kelly	
Theoretical Modeling of Coherent Ultrafast Spin-Light Interactions: From One to Many-Electron Systems	152
P.-A. Hervieux, G. Manfredi, O. Morandi, J. Zemanian, Y. Hirschberger and A. Dixit	
Localization of Magnetic Normal Modes on Topological Defects	156
F. J. Buijnsters, A. Fasolino and M. I. Katsnelson	
Effect of the Variation of the Bond Length on Laser-Induced Spin-Flip Scenarios at Ni₂	159
D. Chaudhuri, G. Lefkidis, A. Kubas, K. Fink and W. Hübner	
Coarse-Graining Approach to Atomistic Spin Dynamics	162
T. Nystrand, J. Venemalm, J. Werperts, O. Eriksson, J. Chico and A. Bergman	
Coherent Ultrafast Spin-Light Interactions in One- and Two-Electron Systems	166
Y. Hirschberger and P.-A. Hervieux	
Noncollinear Ballistic and Diffusive Spin Transport: Magnetic-Field Dependence	169
Steffen Kaltenborn and Hans Christian Schneider	
Semi-relativistic Quantum Electron Dynamics—A Lagrangian Approach	172
A. Dixit, Y. Hirschberger, J. Zamanian, G. Manfredi and P.-A. Hervieux	
Electron Lifetimes in a 2D Electron-Gas with Rashba SO-Coupling: Screening Properties	175
S. Vollmar, A. Ruffing, S. Jakobs, A. Baral, S. Kaltenborn, M. Cinchetti, M. Aeschlimann, S. Mathias and H. C. Schneider	
Non-equilibrium Spin-Spin Interactions in Strongly Correlated Systems	179
A. Secchi, S. Brener, A. I. Lichtenstein and M. I. Katsnelson	

Study of the X-ray-Plasma Interaction for High Intensity Laser Pulses 183
 O. Morandi, J. Zamanian, G. Manfredi and P.-A. Hervieux

Part V Ultrafast Coherent Magnetism

Femtosecond Opto-Magnetism 187
 Alexey V. Kimel

Optical Magnetization Control in EuO Films 190
 Manfred Fiebig

Non-thermal Light-Induced Spin Dynamics in YIG: Co Films via the Photomagnetic Effect 194
 A. Stupakiewicz, M. Pashkevich and A. Maziewski

Ultrafast Charge Contribution to Magneto-optics in Strong Correlated Magnetic Oxides 197
 C. Piovera, F. Boschini, H. Hedayat, C. Dallera, M. Münzenberg, A. Gupta and E. Carpena

Heat Assisted Magnetic Recording 200
 Tim Rausch, Ed Gage and John Dykes

Photo-induced Ferromagnetic Resonance in Systems Incorporating Magnetic Junctions 203
 H. Munekata

Nonlinear Spin Waves in Two-Dimensional Arrays of Magnetic Nanodots 206
 Konstantin Guslienko, Yuri Kobljanskyj, Gennady Melkov, Valentyn Novosad, Samuel D. Bader, Michael Kostylev and Andrei Slavin

Ultrafast Photoinduced Linear and Circular Anisotropy in Multiferroic Manganite $YMnO_3$ 210
 M. Pohl, V. V. Pavlov, I. A. Akimov, V. N. Gridnev, R. V. Pisarev, D. R. Yakovlev and M. Bayer

Magneto-optical Wave Mixing in Garnets 214
 M. Barthelemy, M. Sanches Piaia, H. Vonesh, M. Vomir, P. Molho, B. Barbara and J.-Y. Bigot

Quantum Femtosecond Magnetism in a Strongly Correlated Manganese Oxide	218
Tianqi Li, Aaron Patz, Leonidas Mouchliadis, Jiaqiang Yan, Thomas A. Lograsso, Ilias E. Perakis and Jigang Wang	
Ultrafast Opto-magnetism in KNiF₃	221
D. Bossini, A. M. Kalashnikova, R. V. Pisarev, Th. Rasing and A. V. Kimel	
Classical Modeling of Coherent Ultrafast Demagnetization Experiments	224
Y. Hirschberger and P.-A. Hervieux	
 Part VI Ultrafast Magnetism Control	
Sub-nanosecond Heat Assisted Magnetic Recording of FePt Media	228
D. Weller, O. Mosendz, H. J. Richter, G. Parker, S. Pisana, T. S. Santos, J. Reiner, O. Hellwig, B. Stipe and B. Terris	
Controlling Ultrafast Transport in Magnetic Heterostructures	232
A. J. Schellekens and B. Koopmans	
Ultrafast Magnetoacoustics in Nickel	235
Ji-Wan Kim, Mircea Vomir and Jean-Yves Bigot	
Thermally Assisted All-Optical Helicity Dependent Switching of Ferrimagnetic Amorphous Fe_{100-x}Tb_x Thin Films	238
A. Hassdenteufel, B. Hebler, C. Schubert, A. Liebig, M. Teich, J. Schmidt, M. Helm, M. Aeschlimann, M. Albrecht and R. Bratschitsch	
Ultrafast Laser-Excited Spin Transport in Au/Fe/MgO(001): Relevance of the Fe Layer Thickness	241
A. Alekhin, D. Bürstel, A. Melnikov, D. Diesing and U. Bovensiepen	
All-Optical Switching in CoTb Alloys: Composition and Thickness Dependent Studies	244
Ute Bierbrauer, Sabine Alebrand, Michel Hehn, Matthias Gottwald, Daniel Steil, Daniel Lacour, Eric E. Fullerton, Stéphane Mangin, Mirko Cinchetti and Martin Aeschlimann	

Picosecond Strain Pulses for Ultrafast Magnetoacoustics 248
 O. Kovalenko, V. Shalagatskiy, T. Pezeril, V. Gusev,
 D. Makarov and V. V. Temnov

**Ultrafast Demagnetization Rates in Two-Component
 Magnetic Materials** 251
 O. Chubykalo-Fesenko, U. Atxitia, P. Nieves, J. Barker
 and R. W. Chantrell

**Lattice-Mediated Optical Control of Magnetic
 Anisotropy in FeBO₃** 255
 D. Afanasiev, I. Razdolski, D. Bolotin, S. V. Yagupov,
 M. B. Strugatsky, A. Kirilyuk, Th. Rasing and A. V. Kimel

**Dual-Pump Manipulation of Ultrafast Demagnetization
 in TbFeCo** 258
 T. Y. Cheng, J. Wu, R. W. Chantrell, X. Zou, T. Liu,
 J. W. Cai and Y. B. Xu

**Terahertz Response and Ultrafast Laser-Induced
 Dynamics of Spins and Charges in CoFe/Al₂O₃ Multilayers** 261
 J. D. Costa, T. Huisman, R. Mikhaylovskiy, J. Ventura,
 J. M. Teixeira, D. Schmool, G. Kakazei, S. Cardoso,
 P. Freitas, Th. Rasing and A. V. Kimel

**Nonthermal Magnetization Switching by Ultrashort
 Acoustic Pulses** 264
 O. Kovalenko, T. Pezeril and V. V. Temnov

**Improving the Efficiency of Ultrafast Optical Control
 of Magnetism in GdFeCo Continuous Films
 and Submicron Structures** 267
 R. Medapalli, M. Savoini, I. Razdolski, S. Khorsand,
 A. M. Kalashnikova, A. Tsukamoto, A. Itoh, A. Kirilyuk,
 Th. Rasing and A. V. Kimel

**Magneto-Optical Resistance Induced and Controlled
 by Laser Pulses** 270
 Michèle Albrecht, Mircea Vomir and Jean-Yves Bigot

Part VII Spin Photo-Emission Dynamics

The Valence Band Structure of Gadolinium Studied with Time-Resolved Photoemission 274

B. Frietsch, J. Bowlan, R. Carley, M. Teichmann,
J. Wolter and M. Weinelt

Mechanisms of Multiphoton Photoemission from Metal Surfaces 278

Xuefeng Cui, Cong Wang, Adam Argondizzo and Hrvoje Petek

Time-Resolved Photo-Emission Electron Microscopy of Nanomagnetic Logic Chains 281

Z. Gu, R. Storz, M. Marcus, A. Doran, A. Young, A. Scholl,
W. Chao, D. Carlton, B. Lambson, M. Nowakowski and J. Bokor

Spin-Selective Excitation Pathways in Nonlinear Photoemission from Metal Surfaces 284

A. Winkelmann, C.-T. Chiang, M. Pazgan, T. R. F. Peixoto
and J. Kirschner

Part VIII X-Ray and Far UV-Spin Dynamics

Ultrafast Demagnetization Dynamics in the Presence of Nanometer Sized Magnetic Domains 288

Jan Lüning

Catching the Moment — Magnetization Dynamics Studied with X-ray Photoemission Electron Microscopy 291

L. Le Guyader, S. El. Moussaoui, M. Buzzi and F. Nolting

Accessing the Magnetic Susceptibility of FeRh on a Sub-nanosecond Time Scale 294

Federico Pressacco, E. Mancini, V. Uhler, E. E. Fullerton
and C. H. Back

Engineering Ultrafast Magnetism 297

I. Radu, C. Stamm, A. Eschenlohr, F. Radu, R. Abrudan,
K. Vahaplar, T. Kachel, N. Pontius, R. Mitzner, K. Holldack,
A. Föhlisch, R. F. L. Evans, T. A. Ostler, J. Mentink,
R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk,
A. V. Kimel and Th. Rasing

Ultrafast, Element-Specific Magnetization Dynamics of Multi-constituent Magnetic Materials by Use of High-Harmonic Generation 300
 T. J. Silva, E. Turgut, S. Mathias, C. La-o-vorakiat, P. Grychtol, R. Adam, D. Rudolf, H. T. Nembach, M. Aeschlimann, C. M. Schneider, H. C. Kapteyn, M. M. Murnane and J. M. Shaw

Ultrafast Spin Dynamics on the Nanoscale 303
 C. E. Graves, A. H. Reid and H. A. Dürr

Element Selective Investigation of Spin Dynamics in Magnetic Multilayers 307
 Dennis Rudolf, Chan La-O-Vorakiat, Marco Battiato, Roman Adam, Patrik Grychtol, Justin M. Shaw, Emrah Turgut, Pablo Maldonado, Stefan Mathias, Hans T. Nembach, Thomas J. Silva, Martin Aeschlimann, Henry C. Kapteyn, Margaret M. Murnane, Peter M. Oppeneer and Claus M. Schneider

Element- and Time-resolved Dynamics in Rare-Earth/Transition Metals Alloys 310
 N. Bergeard, V. López-Flores, V. Halté, M. Hehn, C. Stamm, N. Pontius, E. Beaurepaire and C. Boeglin

Space Charge Effects Occurring During Fast Demagnetization Processes 313
 Nathan Beaulieu, Gregory Malinowski, Azzedine Bendounan, Mathieu G. Silly, Christian Chauvet, Damjan Krizmancic and Fausto Sirotti

Ultrafast Spectroscopy with Spin Polarization 317
 V. Lollobrigida, R. Ciprian, F. Offi and G. Panaccione

Magnetic-Field-Dependent Fraunhofer Diffraction Pattern by 4f Imaging System in Transparent Magneto-optic Thin Film 320
 Djati Handoko, Je-Ho Shim, Dong-Hyun Kim, Tae Kyu Kim and Jaehun Park

Part IX Terahertz Spin Dynamics

Ultrafast Spin Precession and Transport Controlled and Probed with Terahertz Radiation	324
T. Kampfrath, M. Battiato, A. Sell, F. Freimuth, A. Leitenstorfer, M. Wolf, R. Huber, P. M. Oppeneer and M. Münzenberg	
THz Spin Dynamics: Phonon-Induced Spin Order	327
K. W. Kim, M. Porer, A. Pashkin, A. Sell, T. Kampfrath, A. Leitenstorfer and R. Huber	
Terahertz Spectroscopy of Femtosecond Spin Dynamics in Orthoferrites	331
R. V. Mikhaylovskiy, E. Hendry, V. V. Kruglyak, A. Wu, R. V. Pisarev, Th. Rasing and A. V. Kimel	
Contribution of Magnetic Circular Dichroism in All-Optical Light Helicity-Dependent Magnetic Switching	334
A. Tsukamoto, S. Kogure, H. Yoshikawa, T. Sato and A. Itoh	
Author Index	337

Part I
Ultrafast Magnetism Dynamics
in Semiconductors

Femtosecond Laser Pulses Switch Magnetic States via Strongly-Correlated Spin–Charge Quantum Excitations

Ilias E. Perakis

Department of Physics, University of Crete, 71003 Heraklion, Crete, Greece, and Institute of Electronic Structure & Laser, Foundation for Research and Technology-Hellas, 71110 Heraklion, Crete, Greece

ilias@physics.uoc.gr

Abstract The technological demand to bump the Gigahertz switching speed-limit of today's magnetic memory and logic devices into the Terahertz regime underlies the entire field of spin-electronics and integrated multi-functional nano-devices. In this talk, I show how all-optical switching based on the ultrafast quantum-mechanical manipulation of spins could meet this challenge.

Introduction

The technological demand to push the gigahertz switching speed limit of magnetic devices into the terahertz regime is met by all-optical magnetic switching based on coherent spin manipulation in strongly correlated systems, where the strength of local spin-spin, Coulomb, and electron-lattice interactions exceeds the kinetic energy and width of the electronic energy bands. Strong correlations between electrons in neighboring lattice sites determine the optically-induced coherent nonlinear dynamics and lead to time-dependence absent in weakly-correlated magnetic systems¹. By analogy to femto-chemistry and photosynthetic dynamics, where photo-products of chemical/biochemical reactions can be influenced by creating suitable superpositions of molecular states, we show that femtosecond (fs) laser-excited coherence between spin/orbital/charge quantum states in neighboring sites can switch magnetic orders² by “suddenly” breaking the delicate balance between competing phases of correlated materials such as the colossal magneto-resistive (CMR) manganites³. We predict theoretically and observe in the experiment² fs photoinduced switching from antiferromagnetic to ferromagnetic ordering. Ferromagnetic spin correlation is driven by the establishment, within the ~100 fs time interval of the pulse duration, of laser-induced coherent superpositions of electronic quantum states in neighboring lattice sites. The development of ferromagnetic correlations *during a single fs laser pulse* reveals an initial quantum coherent regime of magnetism, clearly distinguished from the pre-

viously studied picosecond lattice-heating regime characterized by phase separation without laser intensity threshold behavior. Note that, in the studied material, a transition from antiferromagnetic to ferromagnetic ordering *cannot be induced by increasing the temperature*, which excludes laser-induced heating as a possible mechanism during the 100fs pulse duration, where the observed magnetic phase transition occurs². Our theory is based on equations of motion for *composite fermion density matrices*, which treat the strong Jahn-Teller, Coulomb, and Hund's rule magnetic interactions, as well as the nonlinear coherent fs optical photoexcitation. Our results, summarized in Fig. 1, show fs local spin nonlinear dynamics and underpin fast quantum spin-flip fluctuations correlated with coherent laser-induced superpositions of electronic states to initiate local ferromagnetic correlations via quantum kinetic processes beyond the statistical approach².

In the present work, we show fs photoinduced switching from antiferromagnetic to ferromagnetic ordering in $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$, by observing the establishment, within 120 fs, of a huge temperature-dependent magnetization with *photoexcitation threshold* behavior absent in the optical reflectivity (see Fig. 2). Our theory is based on equations of motion for the density matrix of composite fermion operators that describe the electronic excitations of the strongly correlated system, including local self-energy time-dependent effects and quantum spin fluctuations. Our simulations predict, in particular, fs *quantum spin-flip fluctuations* correlated with photoexcited coherent superpositions of electronic states to initiate local ferromagnetic correlations between neighboring chains that are antiferromagnetically coupled in the ground state, as shown schematically in Fig.1.

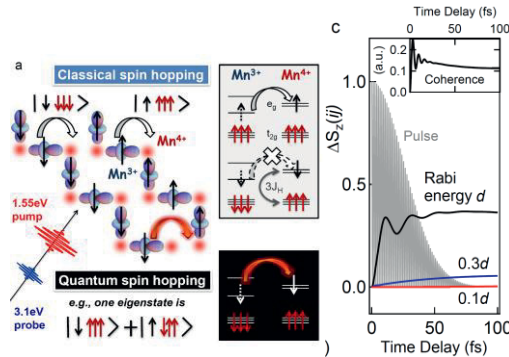


Figure 1. (a) Schematics of ultrafast excitations of a CE-type AFM/CO/OO order. Mn^{4+} configurations, with empty doubly-degenerate e_g orbitals and t_{2g} orbitals filled by three spin-aligned electrons, mostly populate the one-dimensional (1D) zig-zag chain corner sites. The 1D chain bridge sites are mostly populated by Mn^{3+} configurations, with an additional e_g -orbital electron populating alternating orbitals (OO). For classical spins, electron conduction and optical transitions are restricted within the same 1D ferromagnetic chain (white arrow). Quantum spin-flip fluctuations, however, allow e_g electrons to hop on sites with opposite local t_{2g} spin, by forming non-equilibrium total spin eigenstates (red arrow). (b) Calculated photoinduced total spin and inter-atomic coherence (inset) for three Rabi energy values. The laser pulse time-dependence is superimposed, demonstrating that FM local correlations transiently build-up from the AFM ground state (total spin zero) during the nonlinear photoexcitation.

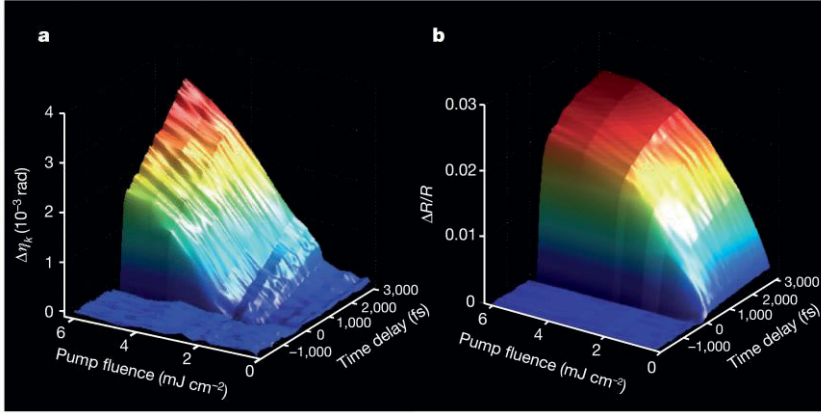


Figure 2. A three-dimensional view demonstrating the distinct femtosecond spin and charge dynamics as well as photo-excitation threshold behavior observed experimentally in Ref. [2]. (a) Time-resolved ellipticity change and (b) differential optical reflectivity as function of pump fluence. Measurements were taken under the same experimental conditions. A photo-excitation threshold is seen for emergence of femtosecond magnetization, whereas the charge dynamics exhibits no threshold. Magnetic field $B=0.5\text{T}$ and temperature $T=30\text{ K}$.

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References

- [1] J. Y. Bigot, M. Vomir and E. Beaurepaire, “Coherent ultrafast magnetism induced by femtosecond laser pulses” *Nature Physics* **5**, 515 (2009).
- [2] T. Li, A. Patz, L. Mouchliadis, J. Yan, T. A. Lograsso, I. E. Perakis, and J. Wang, “Femtosecond switching of magnetism via strongly correlated spin-charge quantum excitations” *Nature* **496**, 69 (2013).
- [3] E. Dagotto, T. Hotta and A. Moreo, “Colossal magnetoresistant materials: the key role of phase separation”, *Physics Reports* **344**, 1 (2001).

Investigation of non-thermal process in the dynamics of photo-induced FMR in (Ga,Mn)As

T. Matsuda and H. Munekata

Imaging science and engineering laboratory, Tokyo Institute of Technology

4259-J3-15 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan

mazda@isl.titech.ac.jp

Abstract We report experimental results which indicate the contribution of non-thermal process in photo-induced ferromagnetic resonance (FMR) in (Ga,Mn)As excited by weak, fs laser pulses.

Introduction

In metallic systems, it has been recognized that randomization of ordered spin systems by the intense laser irradiation (mJ/cm^2) is the fundamental process to realize ultra-fast magnetic excitation^[1,2]. In (Ga,Mn)As, photo-induced FMR (ph-FMR) could be triggered by a few $\mu\text{J}/\text{cm}^2$ laser pulse without an external magnetic field^[3,4]. There have been two different arguments to account for the basic mechanism: the ultra-fast spin randomization (thermal process)^[4,5], and excitation of electronic states associated with ordered spins (non-thermal process)^[3]. We report here that, in the time-resolved magneto-optical (TRMO) measurements for the weak excitation regime ($\leq 10 \mu\text{J}/\text{cm}^2$), signals due to the ultra-fast randomization have been hardly observed, and the onset of ph-FMR strongly depends on excitation wavelength. These results suggest significant contribution of non-thermal effect.

Experimental method

TRMO measurements were carried out at 10 K with one-color pump and probe technique. The light source and the sample were a mode-locked Ti:sapphire laser (a pulse width of 150 fs with repetition rate 76 MHz) and a 100 nm-thick $\text{Ga}_{0.98}\text{Mn}_{0.02}\text{As}$ grown on GaAs(001), respectively. Experimental setup has been detailed in Ref.3. The wavelength has been varied in the range $\lambda = 750 - 900 \text{ nm}$, whereas the polarization plane was fixed at the [010] GaAs axis for both pump

and probe beams. The fluence of the probe beam was fixed at 84 nJ/cm^2 , whereas that of the pump (P^*) was varied between $0.34 \text{ } \mu\text{J/cm}^2$ and $10 \text{ } \mu\text{J/cm}^2$.

Results and discussion

Characteristic oscillatory signals due to ph-FMR have been observed for the wavelengths of $\lambda = 880 \text{ nm}$ or shorter. We have found a striking difference concerning *the onset of oscillation* between the data obtained with $\lambda < 820 \text{ nm}$, the region S, and those with $\lambda > 820 \text{ nm}$, the region L. TRMO data obtained with $P^* = 1.7 \text{ } \mu\text{J/cm}^2$ are shown in Figs.1(a) and (b). Oscillation starts immediately after the excitation in the region L, whereas it is accompanied with noticeable time delay in the region S. Note that, in the ultrafast time regime ($\leq 2 \text{ ps}$), signals due to ultrafast demagnetization have hardly been observed (Fig.1(b)).

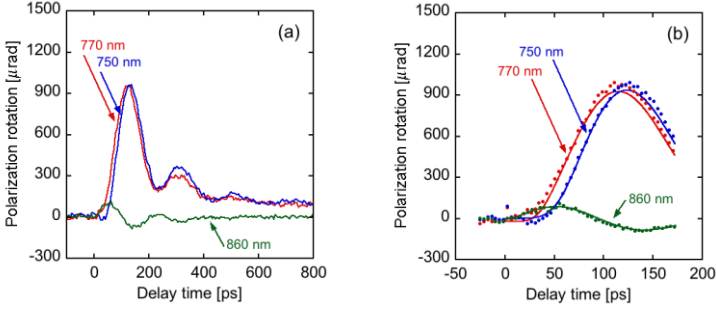


Fig. 1. Photo-induced FMR in (Ga,Mn)As measured at long time scale (a), and short time scale (b). Lines and dots in (b) are experimental and calculated TRMO data, respectively.

Magnetization dynamics was calculated numerically by solving LLG equation. We have added a new *delay term* into the dynamics of an effective field that was developed earlier^[3]. Before excitation, the effective field \mathbf{H} and the magnetization \mathbf{M} both lie along the in-plane [100] direction; with certain time delay τ_g after the excitation at $t = 0 \text{ ps}$, \mathbf{H} rotates toward the out-of-plane [001] direction and relax back to the [100] direction, with its time constant τ_1 and τ_2 , respectively. During this event, \mathbf{M} precesses around \mathbf{H} with natural damping. With this scenario, the angle $\theta(t)$ of \mathbf{H} with respect to the [100] axis was formulated as follows:

$$\theta(t) = \text{erf}(t - \tau_g) \exp(-t/\tau_1) \{1 - \exp(-t/\tau_2)\} \quad 1$$

Here, the delay term is expressed by the error function $\text{erf}(t)$ with the retardation time τ_g . Our approach has reproduced successfully the experimental data, from which τ_g is extracted and summarized in Fig. 2 as a function of excitation photon energy and wavelength. Reflecting the instantaneous oscillations, $\tau_g = 0$ in the region L. This fact indicates the presence of electronic states which allow di-

rect access to the spin subsystem (Fig.3). In the region S, the τ_g value increases with increasing photon energy. This fact suggests that we enter into the region in which another states, presumably the states associated with a host semiconductor, is first excited, and the excess energy is then transferred to the spin subsystem (Fig.3). A dip observed at 790 nm is presumably due to enhanced LO phonon scattering [6].

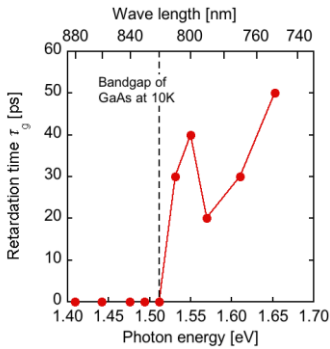


Fig. 2. Photon energy and wavelength dependence of retardation time τ_g .

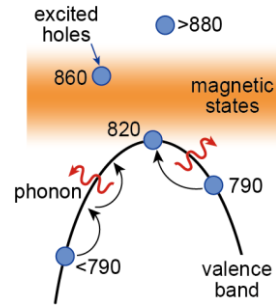


Fig. 3. Schematic illustration of the excitation process for the ph-FMR. Values in the figure indicate the excitation wavelength.

Conclusions

We have found the presence of two different excitation channels for the photo-induced ferromagnetic resonance in (Ga,Mn)As. In the region L ($820 < \lambda \leq 880$ nm), magnetization precession occurs instantaneously with pulsed laser excitation. In the region S ($\lambda < 820$ nm), the precession is accompanied with retardation whose retardation time varies with the wavelength.

References

- [1] E. Beaurepaire, J. -C. Merle, A. Daunois and J.-Y. Bigot, “Ultrafast Spin Dynamics in Ferromagnetic Ni” *Phys. Rev. Lett.* **76**, 4250 - 4253 (1996).
- [2] I. Radu, *et al.*, “Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins”, *Nature* **472**, 205 - 208 (2011).
- [3] Y. Hashimoto, S. Kobayashi and H. Munekata, “Photoinduced Precession of Magnetization in Ferromagnetic (Ga,Mn)As” *Phys. Rev. Lett.* **100**, 067202 (2008).
- [4] D. M. Wang, *et al.*, “Light-induced magnetic precession in (Ga,Mn)As slabs: Hybrid standing-wave Damon-Eshbach modes”, *Phys. Rev. B* **75**, 233308 (2007).
- [5] E. Rozkotova *et al.*, “Light-induced magnetization precession in GaMnAs” *Appl. Phys. Lett.* **92**, 122507 (2008).
- [6] J. S. Blakemore, “Semiconducting and other major properties of gallium arsenide”, *J. Appl. Phys.* **53**, R123 - R180 (1982).

Time Resolved Spectroscopy in Narrow Gap MOVPE Grown Ferromagnetic Semiconductors

G.A. Khodaparast^{a*}, M. Bhowmick^a, C. Feeser^b, B.W. Wessels^b, D. Saha^c, G.D. Sanders^c, C.J. Stanton^c

^a Department of Physics, Virginia Tech, Blacksburg, VA 24061 USA

^b Department of Materials Science and Engineering, Northwestern University, Evanston, IL 60208 USA

^c Department of Physics, University of Florida, Gainesville, FL 32611 USA

* khoda@vt.edu

Abstract We report on time resolved experiments that provide insight into the time scales and the nature of the interactions in ferromagnetic InMnAs and InMnSb. Theoretical calculations are performed using an 8 band $\mathbf{k}\cdot\mathbf{p}$ model including non-parabolicity, band-mixing, and the interaction of magnetic Mn impurities with itinerant electrons and holes.

Introduction

Carrier-induced ferromagnetism in magnetic III-V semiconductors has opened up new opportunities for device applications, as well as fundamental studies in material systems in which itinerant carriers interact with the localized spins of magnetic impurities. A low temperature MBE technique is nearly always used to prepare thin ferromagnetic films, although MOVPE, an alternative technique, allows single phase magnetic InMnAs and InMnSb compounds to be deposited at 500° C, much higher than that used in MBE. Films with hole densities of 10¹⁸ cm⁻³ can have T_c above room temperature [1].

In this work, we perform time resolved differential transmission (TRDT) studies to obtain insight into the dynamics in MOVPE grown ferromagnetic films on the picosecond time scale. To understand the effects of ferromagnetic order on the electronic structure and subsequent relaxation dynamics, we calculate the electronic structure for bulk InMnAs and InMnSb. By calculating the electronic band structure, we can determine where photoexcited carriers are generated by the pump pulse and which regions of the electronic structure are sampled by the probe pulse. The calculations are based on an 8 band $\mathbf{k}\cdot\mathbf{p}$ model which includes the conduction and valence band mixing [2]. We use $\mathbf{k} = 0$ Bloch basis states for the conduction bands, heavy-holes, light holes and split-off holes for a total of 8 states in-

cluding spin. In our model, we include the effects of the spontaneous magnetization of the Mn ions and the sp-d coupling of this magnetization to the electrons and holes [2]. Figure 1 shows an example of the calculations for ferromagnetic InMnSb at 77 K in the absence of an externally applied magnetic field.

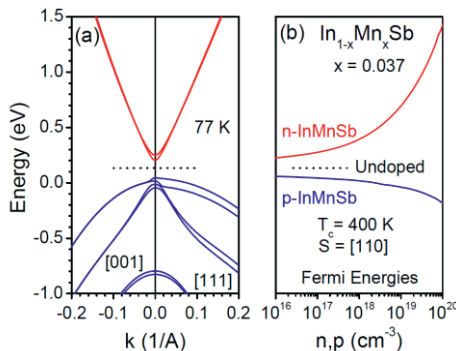


Fig. 1. The band structure and the Fermi energies as a function of carrier density for InMnSb at 77 K as a function of n and p carrier concentrations. The Fermi energy for the undoped semiconductor is indicated by the dotted line. The Curie temperature is taken to be 400 K.

Experimental Approach

By probing the dynamical behavior of the nonequilibrium carriers created by intense laser pulses, we gain valuable information about the band structure and different scattering mechanisms. Using TRDT in the MID-IR, we achieved tunability of carrier dynamics and relaxation times with characteristics unobtainable in MBE grown ferromagnetic structures [2]. The MOVPE grown InMnAs structure is an 800 nm thick film with a Mn content of 4%, and the InMnSb film is a 200 nm film with a Mn content of 3.7%. Both samples, demonstrated T_c above room temperature. The laser pulses were tuned in NIR and MIR using different sources with repetition rates of 1 KHz and pulse durations of ~ 100 fs.

As shown in Fig. 2 for InMnSb, we observe the sensitivity and tunability of the carrier dynamics to the initial excitation. The initial increase in the differential transmission is due to free carrier Drude absorption where the fast component of the temporal evolution is attributed to the relaxation of hot electrons and the slower component is due to electron-hole recombination. Exploiting the selection rules for interband transitions, spin-polarized carriers are created using circularly polarized pump beams. By monitoring the transmission of a weak delayed probe pulse that has the same circular polarization (SCP) or opposite circular polarization (OCP) as the pump pulse, as shown in Fig. 3, the optical polarization can be extracted. Figure 3b shows the exponential fit to the SCP-OCP in the TRDT signal which gives a spin relaxation time of ~ 6.0 ps, which is much higher than the re-

ported time scale for other narrow gap ferromagnetic semiconductors. This fact is due to much higher hole mobilities in these material systems.

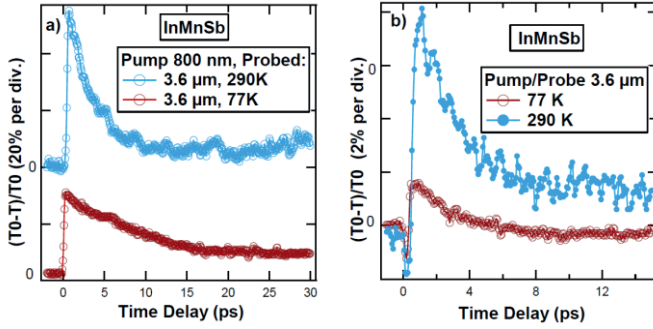


Fig. 2. TRDT a) in a non-degenerate scheme, b) when the pump and probe are the same wavelength. Both the magnitude of the TRDT and the time scale of the relaxations vary by tuning the initial excitation wavelength.

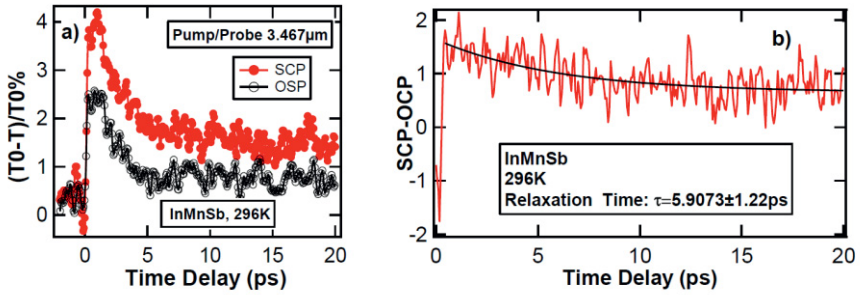


Fig. 3. a) Spin Polarized Time Resolved Differential Transmission b) Exponential fit to the SCP-OCF, to extract the spin relaxation time.

References

- [1] C. E. Feeser, L. Lari, V. K. Lazarov, J. A. Peters, and B. W. Wessels “Structural and magnetic properties of epitaxial $\text{In}_{1-x}\text{Mn}_x\text{Sb}$ semiconductor alloys with $x > 0.08$ ” *J. Vac. Sci. Technol. B* **30**, 032801 (2012).
- [2] M. Bhowmick, T. R. Merritt, and G. A. Khodaparast, Bruce W. Wessels, Stephen A. McGill, D. Saha, X. Pan, G. D. Sanders, and C. J. Stanton , “Time-resolved differential transmission in MOVPE-grown ferromagnetic InMnAs ” *Phys. Rev. B* **85**, 125313 (2012).