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Ultrafast Magnetism I

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

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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, Acalbfi. 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

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Part I
Ultrafast Magnetism Dynamics in Semiconductors

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

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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 systems[1]. 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 orders[2] by "suddenly" breaking the delicate balance between competing phases of correlated materials such as the colossal magneto–resistive (CMR) manganites[3]. We predict theoretically and observe in the experiment[2] 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-

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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 occurs[2]. 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 approach[2].

In the present work, we show fs photoinduced switching from antiferromagnetic to ferromagnetic ordering in $Pr_{0.7}Ca_{0.3}MnO_3$, 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.

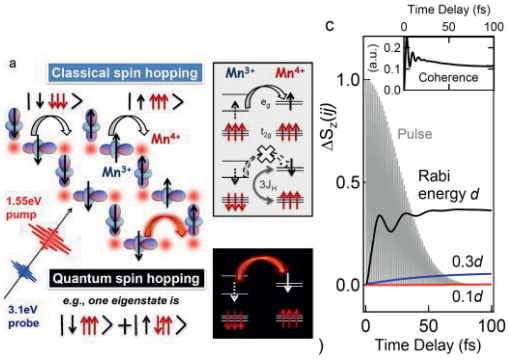

Figure 1. (a) Schematics of ultrafast excitations of a CE–type AFM/CO/OO order. Mn4+ configurations, with empty doubly–degenerate eg orbitals and t2g orbitals filled by three spin–aligned electrons, mostly populate the one–dimensional (1D) zig–zag chain corner sites. The 1D chain bridge sites are mostly populated by Mn3+ configurations, with an additional eg–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 eg electrons to hop on sites with opposite local t2g 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.

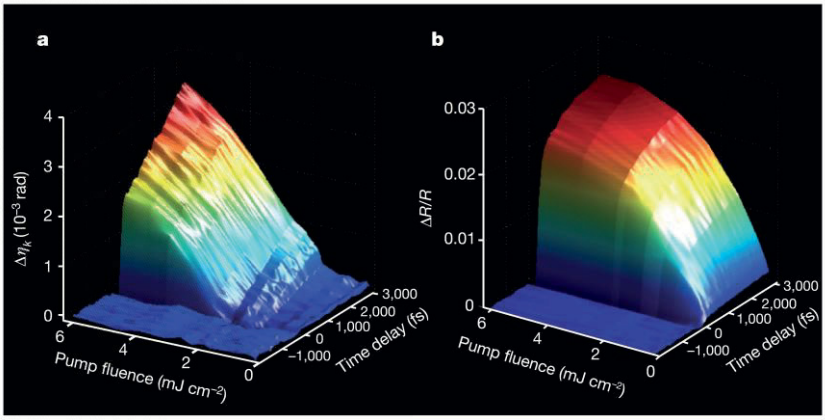

Figure 2. A three-dimensional view demonstrating the distinct femtosecond spin and charge dynamics as well as photo-excitation threshold behavior observed experimentally in Ref. [2]. (a) Time-resolved ellipticity change and (b) differential optical reflectivity as function of pump fluence. Measurements 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=0.5T and temperature T=30 K.

Acknowledgments The authors would like to thank the THALES program NANOPHOS for financial support.

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

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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 μJ/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 μJ/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 $Ga_{0.98}Mn_{0.02}As$ grown on GaAs(001), respectively. Experimental setup has been detailed in Ref.3. The wavelength has been varied in the range λ = 750 - 900 nm, whereas the polarization plane was fixed at the [010] GaAs axis for both pump

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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 μJ/cm^2 and 10 μJ/cm^2.

Results and discussion

Characteristic oscillatory signals due to ph-FMR have been observed for the wavelengths of λ = 880 nm or shorter. We have found a striking difference concerning *the onset of oscillation* between the data obtained with λ < 820 nm, the region S, and those with λ > 820 nm, the region L. TRMO data obtained with P* = 1.7 μJ/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 (≦ 2 ps), signals due to ultrafast demagnetization have hardly been observed (Fig.1(b)).

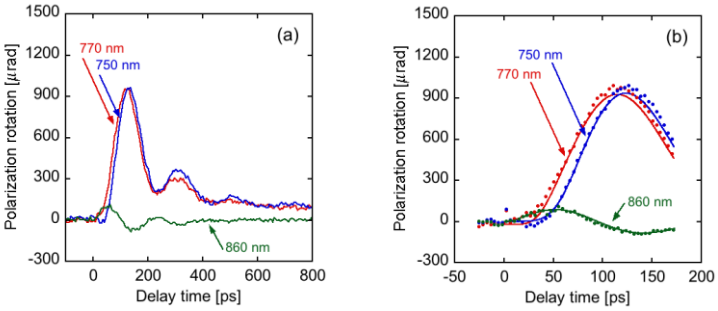

Fig. 1. Photo-induced FMR in (Ga,Mn)As measured at long time scale (a), and short time scale (b). Lines and dots in (b) are experimental and calculated TRMO data, respectively.

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 $\boldsymbol{H}$ and the magnetization $\boldsymbol{M}$ both lie along the in-plane [100] direction; with certain time delay τ_g after the excitation at t = 0 ps, $\boldsymbol{H}$ 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, $\boldsymbol{M}$ precesses around $\boldsymbol{H}$ with natural damping. With this scenario, the angle $\theta(t)$ of $\boldsymbol{H}$ with respect to the [100] axis was formulated as follows:

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

Here, the delay term is expressed by the error function 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, τ_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].

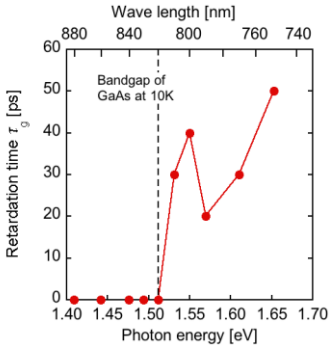

Fig. 2. Photon energy and wavelength dependence of retardation time τ_g.

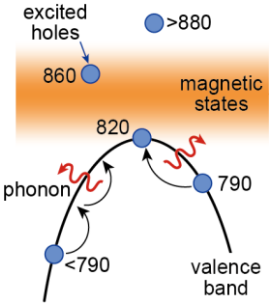

Fig. 3. Schematic illustration of the excitation process for the ph-FMR. Values in the figure indicate the excitation wavelength.

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.

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Time Resolved Spectroscopy in Narrow Gap MOVPE Grown Ferromagnetic Semiconductors

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Abstract a 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 **k·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 **k·p** model which includes the conduction and valence band mixing [2]. We use **k** = 0 Bloch basis states for the conduction bands, heavy-holes, light holes and split-off holes for a total of 8 states in-

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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.

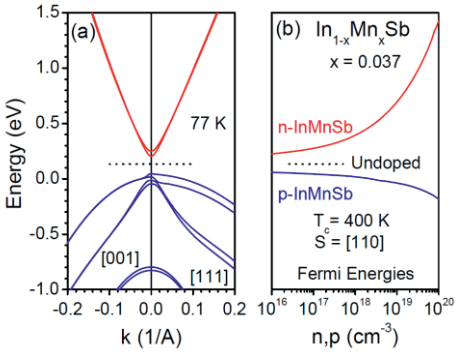

Fig. 1. The band structure and the Fermi energies as a function of carrier density for InMnSb at 77 K as a function of n and p carrier concentrations. The Fermi energy for the undoped semiconductors is indicated by the dotted line. The Curie temperature is taken to be 400 K.

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.

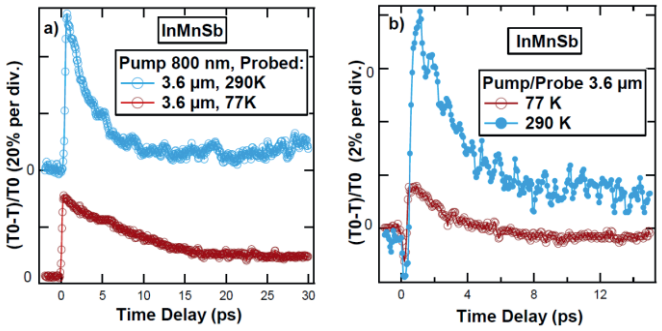

Fig. 2. TRDT a) in a non-degenerate scheme, b) when the pump and probe are the same wavelength. Both the magnitude of the TRDT and the time scale of the relaxations vary by tuning the initial excitation wavelength.

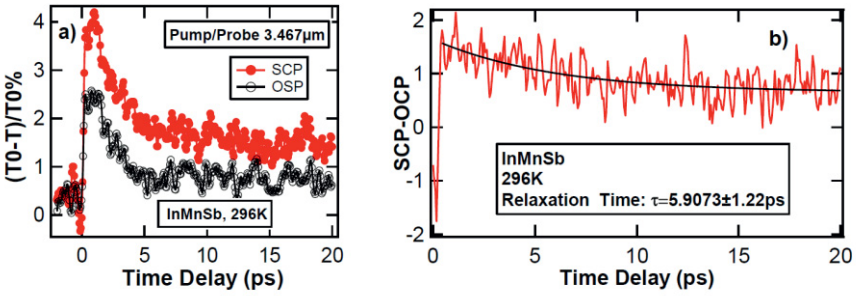

Fig. 3. a) Spin Polarized Time Resolved Differential Transmission b) Exponential fit to the SCP-OCP, to extract the spin relaxation time.

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