Optics

Ultrashort and Ultraintense Laser Science

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Edited by **Kristie Ames**

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Preface

Optics is generally defined as the scientific study of sight and the behavior of light, or the properties of transmission and deflection of other forms of radiation. The power of laser pulse is growing gradually every year due to development in ultra-short and ultra-intense laser technologies. Due to the easy accessibility of these new machines to researchers, experiments have witnessed total remodeling in non-linear optics. This is because an advanced physical picture of light matter correlation comes to fore whenever the laser pulse intensity approaches or exceeds intra-atomic field power. A very broad band of electromagnetic beam is ascertained when the laser rays are adeptly changed into fluxes of charged or neutral particles. The conservative phenomena of non-linear optics like harmonic generation, self-focusing, ionization, etc. exhibit widely contrasting reliance on laser pulse intensity in contrast to the established rules. This branch of research is developing at a rapid pace. The book imparts knowledge on the latest progress and testified results acquired by writers in some selected areas of this widespread domain of science. This book will provide information to readers who specialize in the subject of laser matter interactions.

The information contained in this book is the result of intensive hard work done by researchers in this field. All due efforts have been made to make this book serve as a complete guiding source for students and researchers. The topics in this book have been comprehensively explained to help readers understand the growing trends in the field.

I would like to thank the entire group of writers who made sincere efforts in this book and my family who supported me in my efforts of working on this book. I take this opportunity to thank all those who have been a guiding force throughout my life.

Editor

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Part 1

Femtosecond-Time-Scale Physics

Magnetization Dynamic with Pulsed X Rays

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1. Introduction

Lasers have become more and more useful and a large field of application is nowadays reached including medicine, biology but also fundamental research as physics for instance. It is also in the fundamental research area that recently a fast developing new field is growing: Ultra-short high-energy pulsed X rays. Compared with the lasers community where first technological developments were recently achieved [Spi1997, Dre2001 Schn1999, Kra2009] in order to reach higher energies (5-100 eV), the X-ray community is using high energy X rays from large facilities, for instance the synchrotron storage ring facilities were a large UV and X-ray energy range is produced but were time resolved spectroscopy is only starting since a few years [Sch2000, Scho2000, Hol2005]. It is my aim here to describe the actual state of the art in the field of X rays and especially concerning the different X-ray pulse length and intensities. In the second part I will develop the application in the field of magnetism of the time resolved X-ray spectroscopy and microscopy.

The description of the High-energy X-ray pulse section (2.) will include technical details about the energy range of the X rays, the different time resolution and density of photons produced in the facilities as Synchrotron and X-ray Free electron lasers (X-FEL). The f-slicing possibilities at BESSY (Germany) and also the X-FEL facilities in Europe and in USA will be developed. The recently launched free-electron laser at the FLASH facility in Hamburg and LCLS in Stanford are the two first free electron sources in the world.

Description and discussion of applications using the pulsed X-ray sources are given in section (3.) and will introduce some of the actual motivations in the field of ultrafast magnetization dynamics using ultrafast X-ray pulses. It is divide into two sub-sections; one concerning the spectroscopies performed using the time structures of X rays and the second the time resolved imaging techniques actually developed in the world.

2. Time resolved spectroscopy's using the temporal structure of X rays

In recent years, magnetism at ultrafast time scales has been a growing topic of interest. A thorough understanding of femtosecond magnetism will address the important questions of how fast the magnetization can be reoriented in a material and what physical processes is behind and limits to this speed. In the spatial domain, magnetism at nanometer length is a topic directly relevant to data storage, since future advances in this technology will require a further reduction in device dimensions to increase the storage density. These considerations have motivated a variety of studies using magnetooptic effects in conjunction with ultrafast light pulses to explore these fundamental limits. These studies currently make use of visible-wavelength light from ultrafast lasers, or X-rays from large-scale synchrotron x-ray facilities. Ultrafast lasers produce short pulses (~30 fs), making possible femtosecond time resolution [Beau1996, Cin2006], but with a spatial resolution that is generally limited by the wavelength of the probe light. X rays, on the other hand, allow for high spatial resolution and high contrast imaging at the elemental absorption edges of ferromagnetic materials. However, the available time resolution to date is too slow to resolve the fastest dynamics. Because of this, significant efforts have been devoted to using short or isolated electron bunches of X rays pulses at synchrotron to perform time resolved microscopy with X rays. More recently femtosecond strong laser pulses are used to slice short burst (100 fs) of X rays from synchrotron radiation [Stam2007, Boe2010] Magnetic imaging techniques as for instance X-ray PhotoEmission Electron microscopy (X-PEEM), Scanning Transmission X-Ray Microscopy (STXM) or X-ray Resonant Elastic Scattering (XRES), are currently using the short X-ray pulses in order to accede to time resolved imaging in the picosecond time range. Unfortunately, the f-slicing technique in synchrotrons produces a strongly reduced photon flux hindering the f-second magnetic imaging at facilities as synchrotrons.

2.1 Magnetic imaging using the ps time structure of the synchrotron

2.1.1 Magnetic domains and Vortices under magnetic field pulse excitations

In order to move magnetic domains one of the simplest way one can think of is to apply short magnetic field pulse perpendicular to the magnetization. In this way the field will exert a torque on the sample magnetization vector and induce a rotation of the spins. In a second step the out of equilibrium spins will start to relax in order to transfer the energy from the external field to the lattice, by characteristic precession and damping mechanism. Many experimental description of this process in soft and hard magnetic materials were performed aiming to model the dynamic of relaxation mechanisms in the pico and nanosecond time ranges. Even if the simple idea of a magnetic field pulse excitation is strike forward compared with electronic excitations, in practice this method suffers from the difficulties to produce strong and short magnetic pulses as well as sharp on and off sets (rise times) of the magnetic pulses. Several methods for the generation of magnetic field pulses have been used. Electrical pulse generators for instance (limited by the self-inductance of the electric circuit) with rise times of more than 100 ps and further lithography "stripe lines" were developed in order to reduce the rise times [Ele1996]. Further improvements of the rise-time was archived using optical switches, which can be optically controlled and which are based on lithography fabricated photoconductive " Austin" switches (based on metal-GaAs-metal junctions) [Ger2002] or alternatively "Schottky diodes" switches (based on metal-semiconductor junctions) [Acre2001)]. Beside the large ~50 ps rise times a second limitation is the low induced magnetic fields (~0.1 T) produced by the set-up at the sample location. This often limits the experiments to soft material as permalloy and soft CoFe alloy films (Fig1). Such systems where extensively studied in the past 10 years focusing on reduced dimensions in nanostructures and lithography designed vortices structures.

[Cho2004, Schne2004, Raa2005, Weg2007, Kras2005, Kuc2004, Vog2005, Vogel 2005, Fuk2006, Vog2008, Hey2010, Uhl2011]

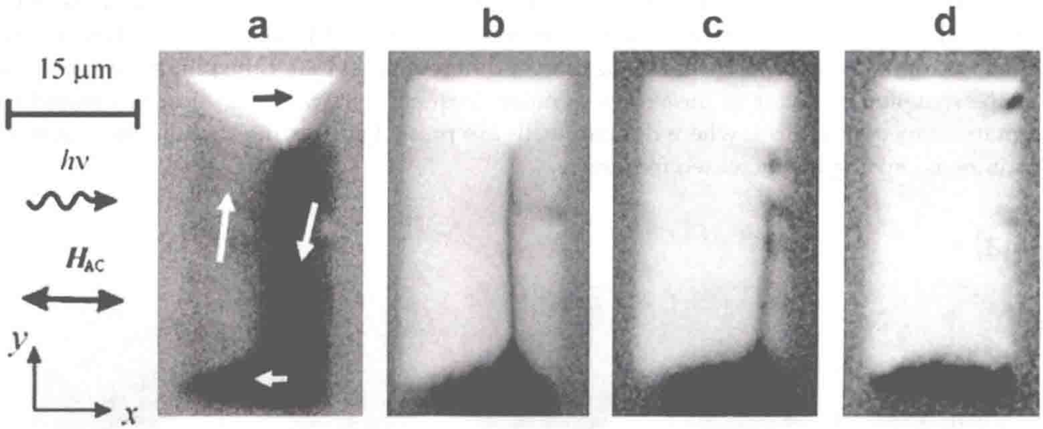

Fig. 1. Magnetic response of the x-component of the magnetization (bright areas are magnetized to the right, dark areas to the left) in a permalloy platelet of 16 · 32 µm2 size and 10 nm thickness for three different field amplitudes I (1.5 Oe), II (2.0 Oe) and III (2.5 Oe). (a) XMCD-PEEM snapshot of the domain pattern in dynamic mode at excitation amplitude I; arrows denote the local magnetization direction. (b)-(d) Snapshots of magnetic domain patterns at maximum magnetic response excited with increasing amplitudes. [Weg2007]

Furthermore, extremely large effective magnetic field pulses can be produced by femtosecond laser pulses combined with the heating of an exchange-biased system. Recently it was suggested that ultrafast switching could be induced via laser-induced reorientation of an exchange coupled antiferromagnet such as $TmFeO_3$ [Kim2005]. A strong magnetic field pulse has also be generated by a relativistic electron bunch combining short duration of 1ps and high field strength ~100 Tesla [Stam2005]. The counterpart of such experiments is that it is accompanied by a strong electric field. Up to now, no time resolved study using a pump-probe set-up has been archived using these high magnetic field pulses.

Time-resolved scanning transmission x-ray microscopy (STXM) in NiFe thin films was studied in order to define the role of domain wall pinning on the dynamic behavior of magnetic vortex structures [Van 2008]. The X-ray magnetic circular dichroism (XMCD) effect, was used as contrast mechanism for the imaging of the structures (Fig 2). In contrast with the X-PEEM, the STXM geometry is sensitive to the projection of the magnetization along the photon propagation direction; therefore, the in-plane magnetized sample was tilted over 60° with respect to the incoming photon beam in order to observe the magnetization. A full image can be constructed by scanning the sample along both in-plane directions. The lateral resolution is determined by the zone plate of the beam line and is about 30 nm. Time-resolved measurements were performed in order to investigate the dynamic behavior in magnetic vortex structures. The natural time structure in the storage ring of the synchrotron delivers photon flashes every 2 ns in the so-called multibunch mode.

This allows the experiment to follow a typical pump-and-probe scheme, with the incoming photon flashes as probe and the externally applied in-plane magnetic field pulses as pump. The magnetic structures were repeatedly excited every 82 ns by sending an electric current in the stripline underneath the structures. The current pulses induce magnetic field pulses with amplitudes of about 10 mT and a full width at half maximum of about 1 ns (500 ps of rise and falling time). The excitation was synchronized with the x-ray flashes of the synchrotron, which probe the magnetization at different times *t* after the pump. The analysis of the dynamic behavior of the vortex gyration frequency show that they are increased in square-shaped structures, where domain walls are present suggesting that the domain wall pinning is causing the increased frequency.

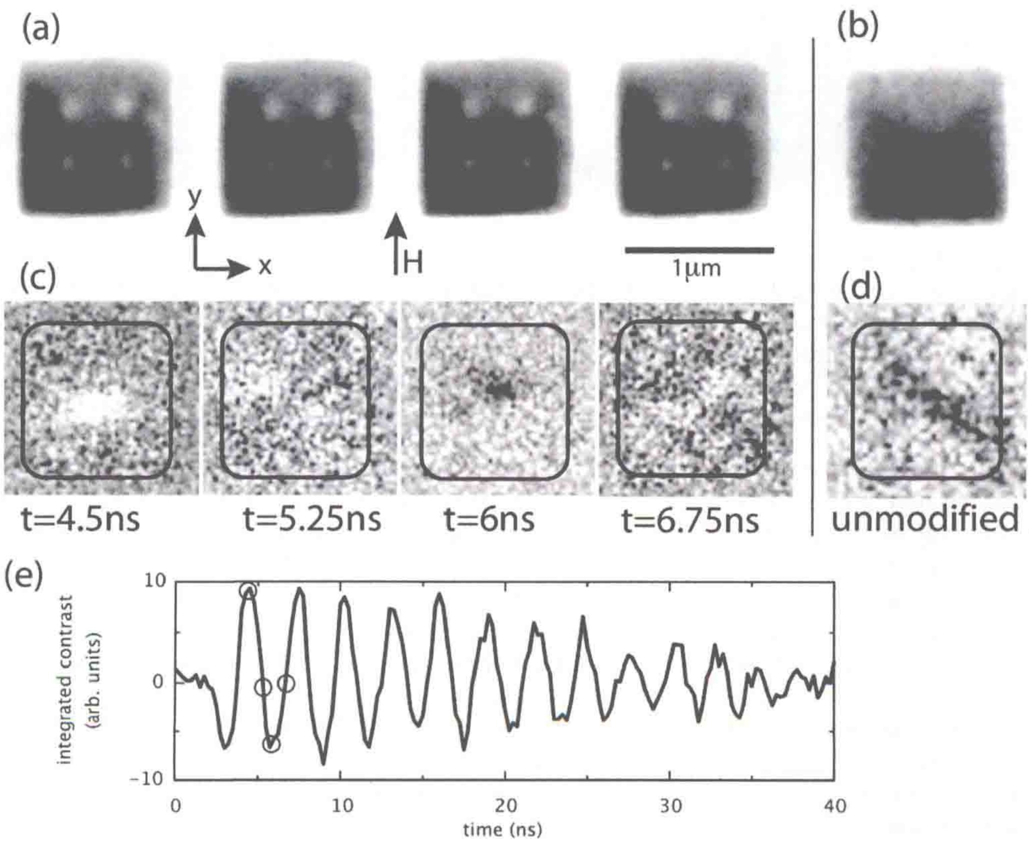

Fig. 2. (a) Sequence of STXM images for a 1 μm x 1 μm x 50 nm modified square during one period of the oscillation. (b) image of an unmodified structure and shows that the domain wall motion can span a larger area of the structure when no defects are created. The intensity is proportional to the *x* component of the magnetization, revealing the Landau configuration and the small gyrotropic motion of the vortex structure. The total contrast in this sequence oscillates with the resonance frequency, as shown in (e). The four STXM images in (a) and (c) correspond to the four markers in (e). The magnetic pulse *H* starts at *t*=2 ns. [Van 2008].

2.1.2 Magnetic domains under femtosecond laser excitation

In order to study the magnetization dynamics in oriented ferromagnetic domains, after a femtosecond pump laser excitation, a precise nanometer scale characterization of the magnetic domain contrast and domain configurations is of great importance. This more recent studied aspect of the space resolved dynamics aims to discribe the influence of a laser excitation on the magnetic domains in different time ranges (nanosecond, picosecond and femtosecond scales). The characterization of the dynamic of the magnetic domain configurations helps to understand the demagnetization process because it provides a description of the magnetization in space. Using X rays these studies benefit from the chemical sensitivity of the circular polarized X rays and from the high spatial resolution (30 nm) of the magnetic imaging mode of X-PEEM instruments that are nowadays currently working at synchrotron storage rings. Appropriate femtosecond pump laser can easily be implemented one such instruments in order to address thermal effects of the laser pump in the ps range. The ultrafast modifications induced by a infra-red laser pump on the magnetic domain configurations is still unknown. Questions concerning the induced changes in the magnetic contrast in a magnetic domain and the size and shape of the domains are still pending. The typical time resolution of the actuel experiment is ~60 ps using the multibunch mode and 10 ps using the low alpha operation modes currently provided in synchrotron storage rings. The time resolution limitation is strongly related with the limited X-ray flux and with the imaging technique by them self where high flux is mandatory.

One of the interesting subjects today is the study of the dynamics in the picoseconde time range of domain sizes and of the magnetic contrast provided using the X-ray circular magnetic dichroism (XMCD) as a function of the pump probe delay. This can be studied either in in-plane oriented magnetic domains or in perpendicular oriented domains.

Following the excitation of ferromagnetic materials with ultra-short laser pulses, a sequence of relaxation mechanisms takes place. The first one is related to the ultra-fast demagnetization. The second mechanism is related to electron - spin and lattice energy transfer, most important within a few picoseconds after the excitation. This mechanism depends on several parameters: the electron-phonon coupling, the material's specific heat, the magneto-crystalline anisotropies and specific interactions like the ferromagnetic or anti-ferromagnetic coupling. One of the goal is to correlate the results with the laterally averaged spectroscopic information obtained using XMCD time resolved spectroscopy.

The experimental method consists in measuring the FM domain contrast in ferromagnetic materials set into a remanent state. Magnetic imaging in the pump probe configuration is setup using the triggered imaging detection mode to obtain a XMCD contrast image at the Fe, Co, or Ni L_3 edges. Moreover the laser fluence necessary to de-magnetize the films is typically in the order of a few mJ/cm^2 and, in the best cases, this allows achieving complete demagnetization of the films. Using focalization of the laser this can easily be achieved by focusing the laser spot onto e few 10 micronmeter on the sample surface. The XMCD signals are probed in a gated mode at different time delays between the laser pump pulse and the probe pulse of circularly polarized synchrotron radiation. The time resolved magnetic signal is extracted from a time-delay sequence of XMCD images and will allow extracting the magnetic components in a semi-quantitative way as a function of time delay. Intensive research in this field is developing using the X-PEEM imaging technique and extension toward other techniques as time-resolved scanning transmission x-ray microscopy (STXM) is expected soon.

2.2 Spectroscopy using slicing techniques or X-FEL pulses

2.2.1 Pump probe with lasers using f -slicing

In order to perform experiments using ultrashort X-ray pulses of only ~100 fs in synchrotrons storage rings one had to modify the large electron bunch time structure of 60-80 ps. This can be performed by using a femtosecond laser pulse to slice the electron bunch. The first generation of fs X-ray pulses in third generation synchrotron radiation sources was proposed [Zho1996] and experimentally demonstrated at the Advanced Light Source (ALS) in Berkeley [Sch2000, Scho2000] using x-ray radiation from a bend magnet. The first undulator-based facility was constructed and successfully commissioned at BESSY [Holl2005].

Such an installation has been set up at BESSY (Berlin) and also at SLS (Villigen) and consists on a slicing of the electron bunches using a femtosecond infra-red laser [Kah2005]. The source at BESSY is based on laser-induced energy modulation ("femtoslicing") and subsequent angular separation of the short-pulse x-rays emitted by an elliptical undulator. The femtosecond X-ray source is thus delivering X-ray pulses of 100 fs (fwhm) duration with tuneable polarization.

The electronic synchronization between the laser pulse and the electron bunches is adjusted so that the electric field of the laser interacts with the bunches at the maximum of the intensity (Fig 3). A specific insertion device names Modulator hosts the laser-electron bunch interaction where the femtosecond laser pulse copropagates with an electron bunch, causing an oscillatory energy modulation of the electrons in the short overlap region. The off-energy electrons are transversely displaced by dispersive elements in order to extract the short component of radiation emitted in a subsequent device (the "radiator"). The second device (Radiator) deviates the two electron bunches with a different angle, so that the angular separation allows extracting only the short radiation component.

The THz signal is the prime diagnostics tool for optimizing the femtoslicing source, when starting an experiment. In addition to being crucial for diagnostics of the laser-electron interaction, the THz radiation itself is useful for experiments where intense ultrashort THz pulses of well-defined temporal and spectral characteristics are required [Holl2006].

The ultrashort X-ray pulses produced by slicing thus provides a strongly reduced flux of 10^4 photons s^{-1} mrad-2mm-2per 0.1% BW, compared to 10^6 photons s^{-1} mrad-2mm-2per 0.1% BW using the single electron bunch. The static measurements using all the bunches we can typically expect at 700 eV a flux of 10^{13} photons s^{-1} mrad-2mm-2per 0.1% BW. The reduction of the flux us thus extremely important when performing time resolved experiments and is in the limit of any experimental set up possibilities when using the sliced beam. This motivates to develop a Bragg-Fresnel zone plate beam line were a photon flux of more than a factor 10 is provided. The energy range of the X rays produced at the beam line at BESSY II ranges from 600 eV to 1400 eV.

The pump probe experiment using such slicing set up are done in a specific pump-probe geometry using the transmitted X-rays at the element core level threshold (ex: Fe, Co, Ni L_2 and L_3), as a probe and a femtosecond laser as a pump. They were carried out using the