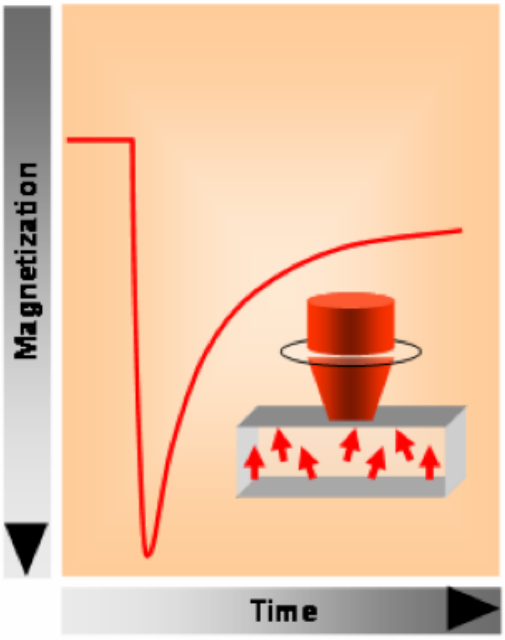
Magneto dynamical studies at Fermi@Elettra. A White paper.



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II. PREFACE

"It's a piece of good luck that nature offered mankind an immediate sensuous experience of what magnetism is about in the form of a mineral that you can pick up from the ground" [1]. To really appreciate this sentence, it's necessary to recall that it is commonly held that the world of tangible objects, which we directly feel and see, is classical and non-relativistic. On the other hand, magnetism turns out to be essentially quantum-mechanical and relativistic.

Progress in the understanding of magnetism was initially as slow and unsatisfactory as for electricity and this lasted well into XVIII century, although the magnetic compass had been in current use for more than 2 millennia. It was not until Oersted's discovery in 1820 that magnetic experiments became easy to perform, thus giving magnetism a much needed boost. Thereafter and within a few years, scientific observations and applications blossomed , opening the way to Maxwell's synthesis.

In the modern times, "we truly live in an electromagnetic world and electromagnetic phenomena form the basis of the modern industrialized society. This fact alone gives the old topic of magnetism a modern day vitality" [2]. In fact, the most advanced applications of magnetism today are closely related to the technology underlying magnetic storage, memory and electrical generators. These applications have fueled a renaissance in magnetism research based on artificially engineered magnetic materials.

From a scientific point of view, these developments and technologies, were made possible thanks to a series of challenging experiments based on the measurements of forces exerted on samples place in magnetic fields. Later experiments involved measurements on magneto-optical Faraday and Kerr effects.

With the development of neutron diffraction [3–6] and spectroscopy techniques (x-ray magnetic absorption dichroism, X-ray magnetic scattering, etc) it was finally possible to determine the spin structure on atomic level. Today, it is hard to imagine modern research into magnetism without polarized X-ray probes.

A central problem in modern magnetism research is the understanding of the magnetization in excited states. Such states, typically initiated by changes in temperature, optical excitations, fields (charge currents) or spin currents, define the operational processes and timescales in magnetic devices. Their understanding is therefore of interest from both a scientific and a technological perspective.

The most fundamental excitations lie in the femtosecond time regime. In practical devices, the time scale is limited today by charge propagation through wires to about hundreds of picoseconds. Excitations which give access to the fundamental femtosecond time scale of electrons in metals can only be triggered by lasers [7, 8] or relativistic electron bunches [9]. In particular, laser-based experiments, where excitation energy is pumped into the electronic system, have shown fascinating effects like ultrafast demagnetization [8, 10–12] and all optical switching [13, 14].

The strongly non-equilibrium conditions developed in magnetic materials following excitation by intense femtosecond laser pulses represent a subject that has attracted continuous growing interest over the last two decades [2, 8, 10, 13–30]. However, due to this strong non-equilibrium, the conventional description of magnetic phenomena in terms of thermodynamics is no longer valid. As a result, the ultrafast channels for transferring energy and angular momentum between photons, electrons, spins, and phonons remain elusive and a subject of debate [2].

Surprisingly, it has been reported that laser excitation of a magnetic metallic film may lead to a demagnetization of the material within 50 fs, thus much faster than the typical spin-spin and spin-phonon characteristic times [28]. Furthermore, it has recently been demonstrated that light can excite the metallic spin system also before electron thermalization, via excitation of coherent optical phonons [18]. Such intriguing observations raise fundamental questions about the mechanisms re-

sponsible for the ultrafast optical excitation of spins in metals. In this context it has been suggested that in a metallic magnet, photons may also excite spins non-thermally via an ultrafast interaction between the angular momentum of the photons and the spin system [15]

Magnetization in solids reacts either to external temperature, pressure, magnetic field, and changes on various time scales and their relation the the size of magnetic structures and domains has been a prominent subject in magnetism because of the numerous applications.

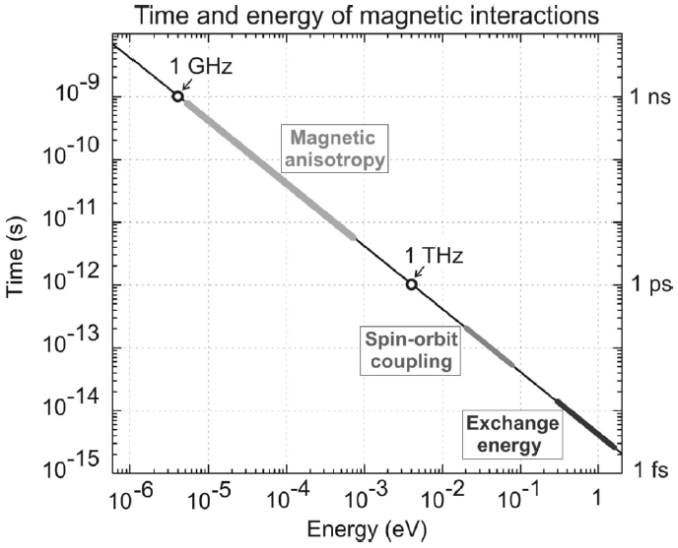
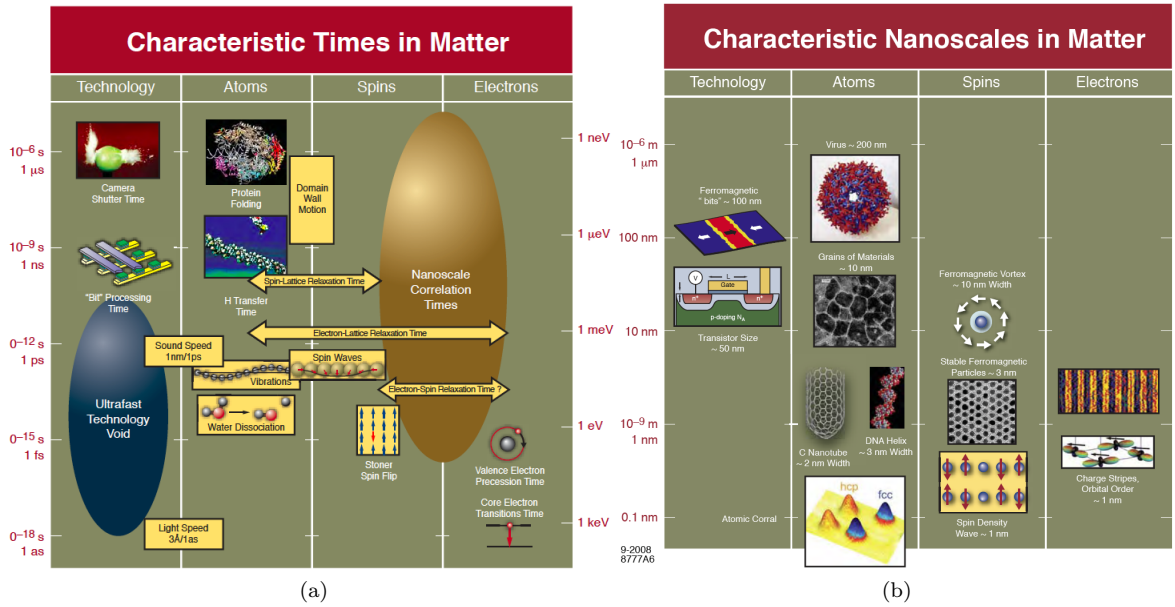


FIG. 1: General time-energy correlation plot given by t = h/E which allows comparison of results in the frequency and time domains. On the correlation line we have indicated typical ranges for three important magnetic energies and the associated time scales found in the 3d transition metal systems [2]

Over the last 15 years, the development of magneto-optics with ultra short and intense pulsed lasers has opened a new field, *ultra fast magnetization dynamics*. The studies of ultrafast magnetic dynamics in nanoscopic magnetic bodies have become one of the most exciting topics in contemporary magnetism. They break new ground scientifically and explore length and times scales for tomorrows magnetic technologies. More generally, they support the demand for smaller and faster of our high-tech society. The future development of magnetic recording technology where the quest is for smaller magnetic bits and faster magnetic switching depends to a large extent on the outcome of this research. The natural time scales of the physical processes in such systems span an enormous range of about fifteen orders of magnitude, depending on the atomic, electronic, and spin structure and parameters such as temperature.

Typically, ultrafast studies employ pulsed lasers for both excitation of the sample and probing the ensuing changes of the magnetization, in so-called pumpprobe experiments. Such experiments now have a time resolution below one picosecond (10^{-12} s), well into the femtosecond (10^{-15} s) range. The timescales in the picosecond and femtosecond range are of great interest because they naturally correspond to important magnetic energies, as illustrated in Figs. 1 and 2a.

Ultra fast techniques have been applied to look for an answer to many fundamental aspects of magnetism. For example, for more than two decades, researchers have been attracted to the question: what happens in a ferromagnet after suddenly exciting it by a short laser pulse, rapidly heating up the electron gas? In the early 1990s, Vaterlaus and co-workers carried out time- and spin-resolved photoemission (TSPE) experiments on gadolinium, yielding a rough estimate of the demagnetization



(a) (b)

FIG. 2: Illustration of our present knowledge about typical times involved in the interactions of atoms, electrons, and spins. On the right, the corresponding quantum-mechanical interaction energies as estimated from the energytime correlation $\Delta E \cdot \Delta t = h \sim 4$ fs·eV. [31]

time, τ_M ≈100±80 ps [32]. This value was soon thereafter reproduced by theoretical estimates of spinlattice relaxation [33]. In view of the above, new results in 1996 by Beaurepaire et al. exploiting time-resolved magneto-optical Kerr effect (TRMOKE) studies on nickel thin films came as quite a surprise [8, 28]. It was found that demagnetization after sub-100 fs pulsed-laser excitation proceeds well within a picosecond.

Among these techniques, the time resolved MOKE (TRMOKE) enjoys the greatest popularity beacause it allows to measure the fast magnetization dynamics with the magneto-optic Kerr effect (see Fig 3).

While lasers and pump probe magneto-optics have paved the way for the studies of the ultra fast dynamics, the ultimate goal relies on the understanding of the intimate nature of the magnetization: the angular momentum. In fact the essence of magnetization is angular momentum and thus the key issue in magnetization dynamics are the processes underlying the change of angular momentum.

It is clear that the sample evolves through a laser-excited electronic state where the modifications of the magnetization take place. The key question is: what is angular momentum doing when the system is in its excited state? How the different components of the total angular momentum 1 are competing the one respect to the other?

$$J = S_{elec} + L_{elec} + L_{phonons} + L_{exciting\ photon} \tag{1}$$

Simply replacing the femtosecond laser probe pulse with ultrashort probe pulses supplied by a soft x-ray FEL would allow the understanding of this transition state. Time-dependent XMCD measurements would reveal the separate evolution of the spin and orbital components of the magnetization (time resolved S_{elec} and L_{elec}, see equ. 1), providing key information on the transfer of angular momentum out of the spin system that has to accompany any magnetization change. Time dependet XAS spectra would follow the time resolved electronic structure ($L_{exciting\ photon}$, see equ. 1). Time and spin resolved photoemission spectra would directly measure changes in the spin resolved band structure in the laser-excited state. As illustrated in Fig. 4, we do not know at present whether

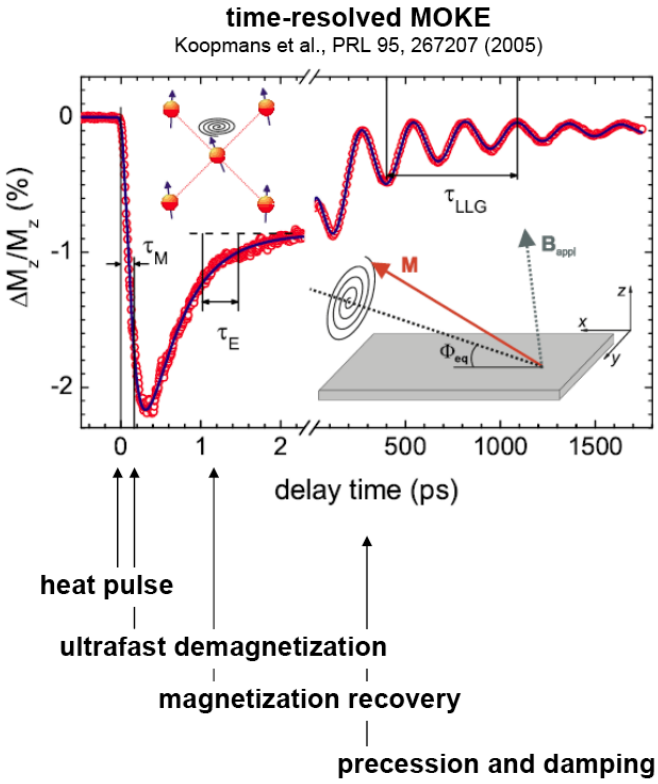


FIG. 3: Reduction of the perpendicular component of the magnetization M after laser heating a Ni thin film at t=0. The sub-picoseconds quenching of M is followed by a recovery via electronphonon relaxation, and at t > 100 ps by a damped precession of M about the applied field. [10, 11]

and how the electronic bands, the exchange splitting, and the spin-orbit coupling change in the laser-excited state.

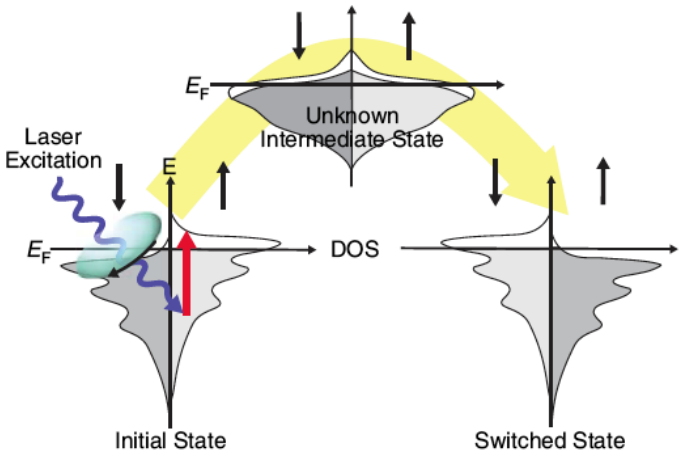


FIG. 4: The evolution of the spin-dependent electronic structure through excited states after laser excitation remains an unsolved science problem. [31]

III. X-RAY TECHNOLOGY

Key developments in x-ray science over the last thirty years are based on the unique properties of modern synchrotron radiation sources: high flux, high brightness and coherence, energy tunability and high energy resolution, polarization control, and pulsed time structure and associated instrumentation (imaging detectors with high spatial resolution and high efficiency, high-speed photon counting detectors, high accuracy optics for preserving source brightness and for focusing and analyzing the energy of the photons).

Soft x-ray spectroscopic studies include near-edge x-ray absorption fine structure (NEXAFS), x-ray emission spectroscopy (XES), and photoemission (PES). The longer wavelength ($\sim$ 1 Å) of lower-energy soft x-rays ($\sim$ 1 keV) enables the investigation of the electronic structure of matter.These techniques yield information on the electron distribution in occupied and empty orbitals in molecules and the electronic energy bands in solids, providing information on how atoms are bound together in solids, liquids, and molecules. Linear-polarization-dependent NEXAFS and XES studies have provided detailed information of directional bonding and orientational order at surfaces. Finally, the angular momentum of circularly polarized x-rays has allowed access to the angular momentum of electrons and consequently the ability to quantitatively separate the spin and orbital parts. X-ray magnetic circular dichroism (XMCD) studies preferentially use soft X-rays, where the magnetic effects are largest, for either x-ray absorption spectroscopy or resonant scattering.

Key future development areas of soft X-ray spectroscopic techniques are their extension to the multi-dimensional phase space of length scales down to nanometers, time scales down to femtoseconds, or even atto-seconds, and energy scales to tens of micro-eV (Fig 2a, 2b).

More recently, interest has turned to short-wavelength, free electron lasers (FELs) [34][35], which can produce light pulses with peak brilliance up to ten orders of magnitude higher than the pulses generated in the present third generation synchrotron light sources and with photon energies spanning from the vacuum ultraviolet to the hard x-ray, i.e. from about 10 eV (120 nm) to 10 keV (0.12 nm).

FEL sources can operate in several ways. To date, most of the existing and planned very- short-wavelength FELs (e.g. FLASH, LCLS, SCSS, XFEL and SPARX) have employed the self amplification of spontaneous emission (SASE) [36] mode of operation. While it is possible to get extremely high brilliance, the temporal structure of a SASE output pulse is normally composed of a series of micro-pulses that individually have random phase and highly fluctuating peak intensity and time duration. For SASE devices, synchronization to external sources is normally limited by the temporal jitter of the accelerator. This jitter can be tens of femtoseconds or greater, especially for accelerators based upon non-superconducting cavities. As an alternative, short- wavelength FEL sources based upon seeding techniques, in which the FEL pulse is initiated by a coherent radiation pulse generated by a conventional laser, can produce output pulses with a well-defined temporal shape and intensity stability [37][38] that permits relatively straightforward synchronization with external pump or probe lasers. In principle, seeded FELs can obtain output radiation bandwidths close to the Fourier transform limit.

The FERMI@Elettra FEL facility, [39], comprising two FELs, will use a combination of coherent seeding and harmonic upshifting [40–44] to provide coherent emission over a photon energy range of 12-300 eV (and up to 900 eV at the third harmonic)

Parameter	FEL-1	FEL-2
HGHG Stages	1	2 (“fresh bunch” in 2nd stage)
Fundamental Wavelength range [nm]	100 to 20	20 to 3 (1 at 3rd harm.)
Output pulse length (rms) [fs]	< 100	20 – 100 (< 10 future goal)
Bandwidth (rms) [meV]	17 (at 40 nm)	100 (at 4.2 nm)
Polarization	Fully Variable	Fully Variable
Repetition rate [Hz]	50	50
Peak power [GW]	1 to >5	0.5 to 2
Harmonic peak power (% of fundamental)	~2	~0.2 (at 4.2 nm)
Photons per pulse	10^{14} (at 40 nm)	$2x10^{12}$ (at 4.2 nm)
Pulse-to-pulse stability	≤ 30 %	~40 %
Pointing stability [μrad]	< 20	< 20
Virtual waist size [μm]	250 (at 40 nm)	120
Divergence (rms, intensity) [μrad]	50 (at 40 nm)	10 (at 4.2 nm)

FIG. 5: Nominal parameter values for the FERMI light beam and pulse structure.

IV. ”EARLY” XAS PUMP-PROBE EXPERIMENTS

At contemporary synchrotron radiation (SR) sources, powerful X-ray techniques to study the structure and function of matter are limited in time resolution by the electron bunch length of 30-100 ps [full width at half-maximum (FWHM)].
In the quest for short pulse duration and short wavelength, one route is to convert femtosecond (fs) laser pulses to X-rays by high-order harmonic generation (HHG) [45–47](where the flux drops rapidly with increasing photon energy) or by slicing electron bunches by fs laser pulses at insertion devices of a SR source.

A. High-order harmonic generation (HHG) sources

Significant efforts have been devoted to perform table top x-ray experiments using (HHG) from commercial femtosecond laser systems [48–50]. In these experiments the $M-$edges of the ferromagnetic $3d$ transition metals can be probed. From a phenomenological point of view, the HHG processes is a third steps process triggered by the electric field of a very intense laser pulse interacting with an atomic or molecular system. The ionized electron is accelerated into the continuum by the same laser field and and subsequently re-collides with the parent ion as the laser field reverse. During the collision, the kinetic energy is released as high energy photons.
The HHG process is a promising optical techniques for generating coherent radiation with temporal duration approaching the attosecond timescale and photon energies in the Extreme Ultra-Violet (EUV) and the Soft X-Rays (SXR) spectral regions.
An HHG source has been recently used to perform a challenging experiment resulting from an international collaboration between the T-ReX Laboratory in Trieste (Italy) and the Center for X-Ray Optics (CXRO) of the Lawrence Berkeley National Laboratory (LBNL), located in Berkeley (USA).

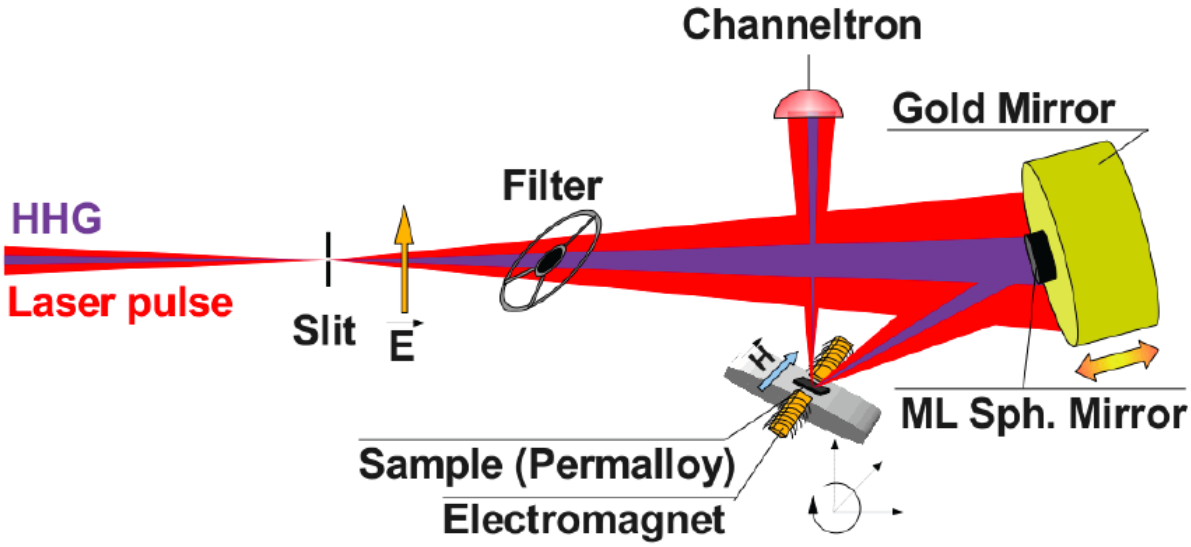


FIG. 6

Experimental chamber setup. With linearly polarized radiation, a magnetic dichroism is obtained with the magnetic field transverse to the rejection plane and with p polarized radiation (as shown) [49, 50]

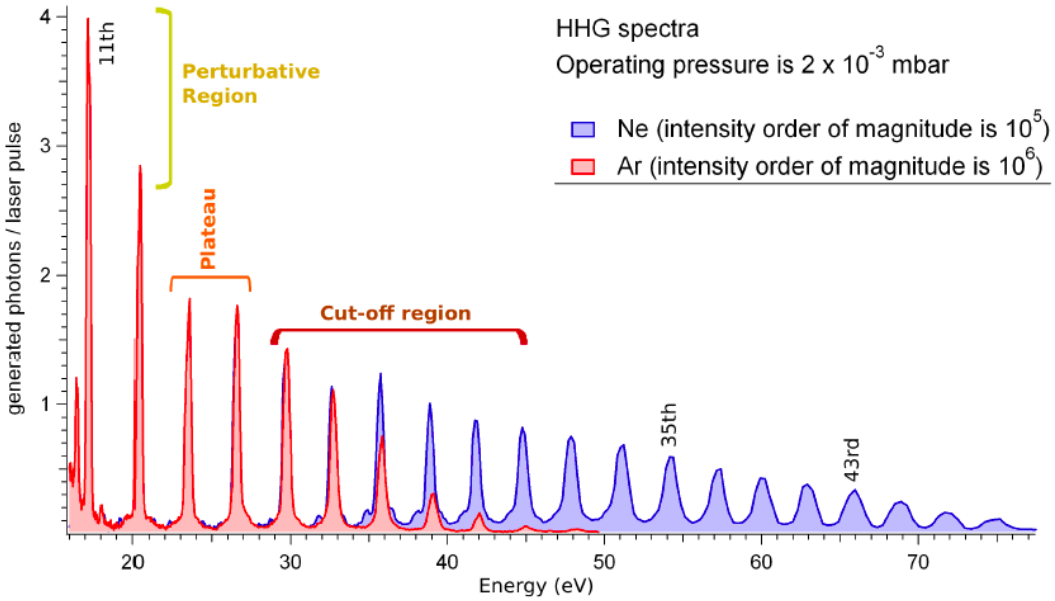


FIG. 7: HHG spectra generated in Argon and Neon using 2:2 mJ, 50 fs, near-IR (0 800 nm) laser pulses. For Argon the perturbative, plateau and cut-o regions are highlighted. Due to the higher ionization potential Ip of Neon, its cut-o region extends to higher energy but the total number of photons is lower (this spectrum is multiplied by a factor of 10 in order to make a visual comparison with Argon). The 35th and the 43rd harmonics, employed in this thesis work, are resonant with respectively the Fe and Ni M-absorption edges. [49, 50]

B. femtoslicing

However, it would be desirable to perform soft X-ray pump-probe experiments with high photon flux also at the $L-$edges since the application of the XMCD sum rules at these absorption edges is much more straightforward (see fig. 9). Another possibility to generate high energy X-ray femto-pulses is to manipulate the time structure of relativistic electron bunches to produce shorter SR pulses [51–53].

In electron bunch slicing [54] the interaction in an undulator of a femtosecond laser pulse with the several picosecond long electron bunch delivered by the synchrotron forms two femtosecond electron sub-pulses in the main bunch (two slices) of lower and higher energy. These slices can be extracted from the bunch in the passage through a bending magnet and production of highly coherent femtosecond X-rays occurs in another undulator device (the radiator). Since only a small portion of the bunch is used, these sources are mainly limited by the low photon flux (close to 10 photons / laser pulse on the sample). Besides, a pump beam must be provided externally. Even so, the slicing technique has some advantages: the beam has a good spatial coherence and by means of a helical undulator a complete polarization control can be achieved, an appealing feature for magnetic structure studies. The low flux issue could be solved with the electron bunch compression [54], which is obtained during its passage in a magnetic chicane. In this device slower electrons travel shorter paths with respect to faster electrons and bunch compression down to subpicoseconds can be achieved.

This approach, slicing electron bunches by fs laser pulses, was experimentally demonstrated [55] and applied to x-ray absorption spectroscopy [56] at the Advanced Light Source in Berkeley with radiation from a bend magnet. A first facility to produce 100 fs undulator radiation with linear and circular polarization and photon energies up to 1400 eV was constructed in 2004 at the 1.7 GeV electron storage ring BESSY II in Berlin and is now in routine operation for pump-probe experiments. Some recent experiments have also been performed using x-rays from large-scale synchrotron X-ray facilities [12, 57].

However, X-ray femtoslicing experiments have so far been limited due to the low photon flux.

To overcome the limitation of photon flux (in X-ray slicing experiments) and low photon energy (in table to HHG experiments) a FEL source with variable (circular, linear) polarization would be of utmost importance. The limited repetition rate in the FEL (typically 50 Hz compared to 1 kHz in X-ray slicing experiments) will be more than compensated by the significantly higher photon flux per pulse.

A particularly interesting and so far unique feature of the proposed set-up at Fermi is the possibility to perform laser pump/X-ray probe experiments as a function of sample temperature (down to LHe) and in high magnetic fields of several Tesla. This opens up a completely new avenue for research in ultrafast magnetization or demagnetization dynamics.

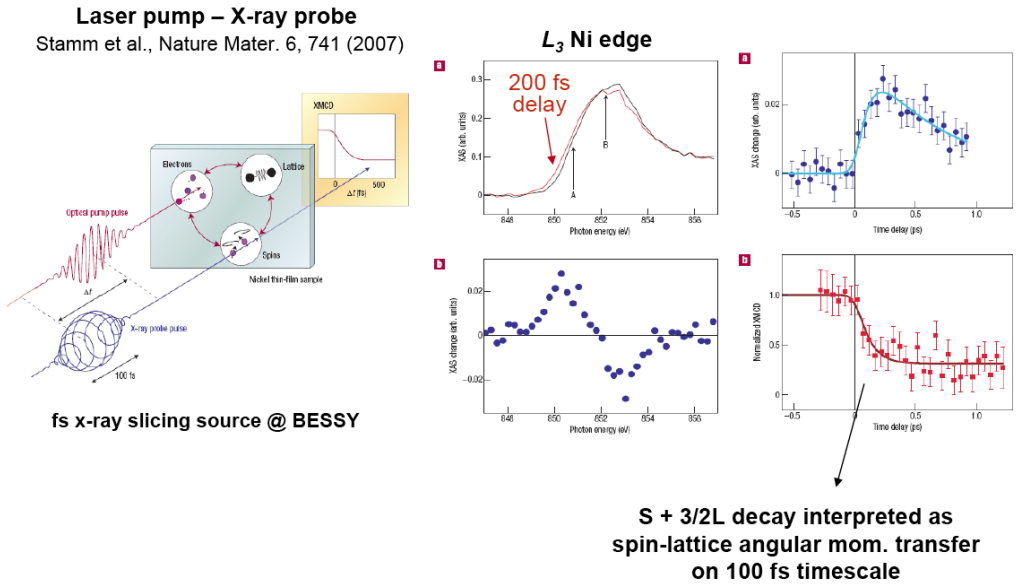


FIG. 8: Slicing source experiment: XAS spectra obtained with linearly polarized femtosecond X-ray pulses at normal incidence. The absorption at the L_3 edge is shown for a 15-nm-thick Ni film 200 fs after (red line) and without (black line) laser excitation. [12, 57]

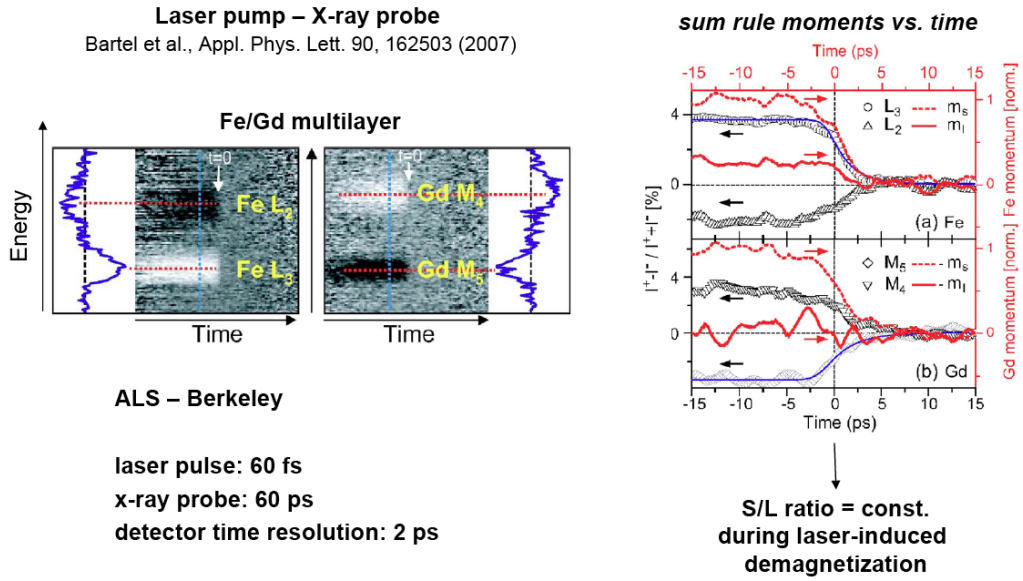


FIG. 9: Slicing source experiment: (left) Time-energy maps of the transient XMCD at the Fe $L_{2,3}$ (left) and Gd $M_{4,5}$ (right) edges demonstrate a complete loss of the dichroism across the absorption spectrum after laser irradiation. Shown on the sides are XMCD spectra of Fe and Gd averaged over 1.5 ps. (right) Transient dichroism (left ordinate) at the Fe $L_{2,3}$ (a) and Gd $M_{4,5}$ edges (b) and transient spin and orbital momenta ms, ml (right ordinate) of Fe (a) and Gd (b). The demagnetization of both materials is synchronous. For an easier comparison the antiferromagnetically coupled Gd momenta are inverted in sign [58]

v. TIME RESOLVED XAS/XMCD AT BACH@ELETTRA BEAMLINE

VI. TIME RESOLVED XAS/XMCD AT MAGNEDYN@FERMI BEAMLINE

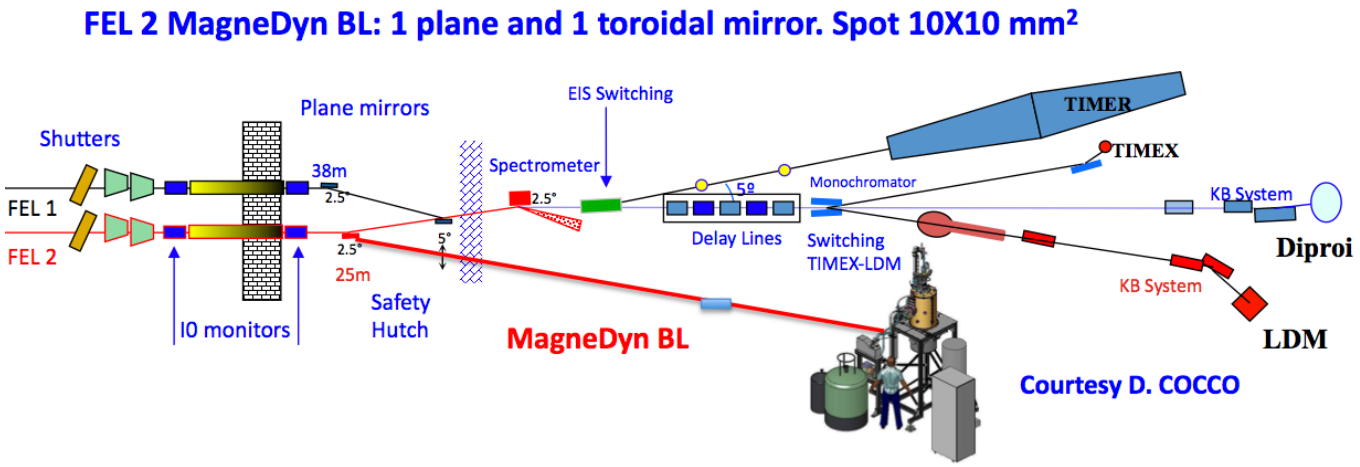


FIG. 10: Layout of the beamline for magneto-dynamical studies at Fermi FEL source. From left to right, there are the isolation vacuum valves, beam-defining apertures, beam position monitors, radiation intensity monitors, a gas absorption cell, three plane mirrors inside the safety hutch, a photon energy spectrometer, and the other currently approved beamlines with their switching and focusing systems.

TABLE I: Magnedyn beamline parameters

time resolution	Tunable photon energy ranges	Polarizations	# photons/pulse
<100 fs	1^{st} harm: 3-20nm [60 - 410 eV] 3^{rd} harm: 1-7 nm [177-1200 eV]	linear; circular left/right	$2 * 10^{11}$ $2 * 10^{4}$

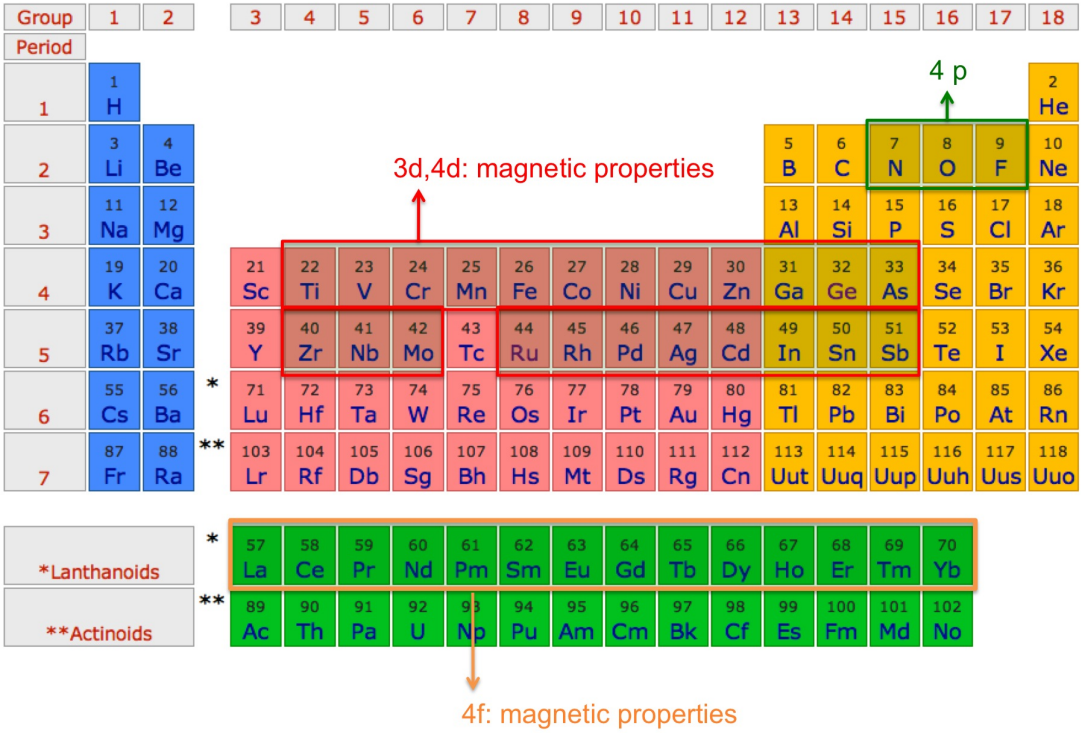


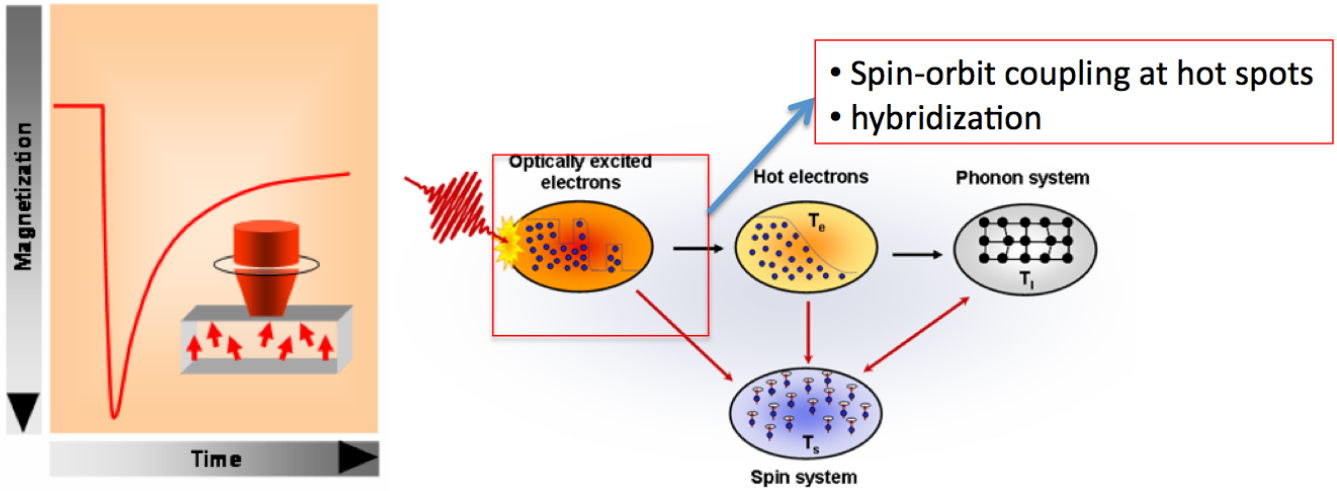
FIG. 11: The third harmonic emission from FEL-2 will be exploited to reach photon energies in the 300-1000 eV range corresponding to the L-edges of magnetically active elements (e.g. Fe and Co)

VII. EXAMPLES OF SCIENTIFIC CASES

A. From fundamentals to materials aspects in ultrafast magnetism

Femtomagnetism is concerned with the exchange of angular momentum and energy between the spin, electron and phonon subsystems in solids with long range magnetic order on picosecond and femtosecond timescales. Initial work has focused on an understanding of the fundamental principles underlying the phenomena of ultrafast demagnetization [insert ref] and all-optical switching. In the last 15 years, this work has been carried out predominantly with optical fs-lasers. Only recently, the use of fs-pulsed x-rays from synchrotron slicing sources has been able to add important insight via studies separating spin and orbital moments [insert ref]. As a result, a microscopic three temperature model has emerged very recently [insert ref], that seems to describe the phenomena encountered in ultrafast demagnetization on both the fs and ps timescale. We are thus now at the point where (a) this theory can be tested and (b) the insight gained so far can be used to design more complex materials systems with a tailored dynamic response. As the ultrafast response of a magnetic system to a fs optical excitation of the electronic system seems dependent on details of the electronic structure occupied by the hot electrons (amount of spin-orbit coupling at hot spots; hybridization) as well as on the atomic element under study (μ_B/atom) it will in the future be possible to generate more sophisticated systems such as alloys, thin film multilayers and nanostructured samples in order to modify the ultrafast magnetic response to an optical pump pulse. This approach will enable a more rigorous test of the theories of ultrafast demagnetization which were recently put forward [insert ref] and may lead to a materials science in ultrafast magnetism which in the long run

enables applications e.g. in opto-spintronics. To make progress in this emerging area, fs temporal resolution, atomic species sensitivity, spin/orbital-momentum discrimination and spatial resolution on a nm length-scale have to be combined in pump-probe experiments. These requirements make this program ideally suited for a seeded soft x-ray laser which can provide spectrally pure and reproducible x-ray pulses with <100 fs pulse duration, intrinsic jitter-free synchronization to a pump laser and sufficient coherent flux per pulse at variable polarization [insert ref] to collect scattering patterns and this obtain spatial resolution. The latter can be established in reciprocal space or via holography and/or coherent diffraction imaging in real space.[insert ref] We propose to study alloys, thin film multilayers and nanostructured samples systematically tailored to exhibit a fs to ps magnetic response to an optical excitation. The key approach is to generate hot electrons in sample via an IR laser pulse and to interrogate the magnetic state of the system via XMCD and XMLD in coherent scattering experiments. A unique opportunity to carry out this research program can be created at MAGNEDYN@FERMI where the L- and M-edges of $3d$ metals and the M- and N-edges of $4f$ metals within a composite sample structure can be accessed.



(a) (b)

FIG. 12: 3 T model used to describe the interaction of an ultra-short laser pulse with the electron, phonon and spin subsystem of a ferromagnet. The microscopic mechanism leading to an ultrafast demagnetization are stil not clear

B. Ultrafast demagnetization dynamics in metallic ferromagnets after core hole excitation

Here the transient ferromagnetic order would be probed by x-ray magnetic circular dichroism after an UV to XUV optical excitation. There has been a lot of work on these ultrafast magnetization

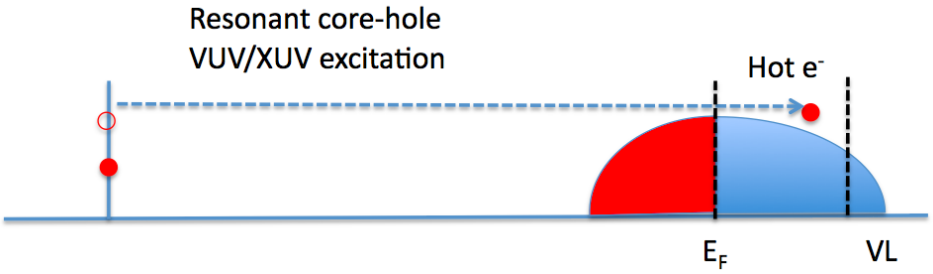


FIG. 13: Resonant core-hole VUV/XUV excitation.

dynamics in recent years and understanding of the phenomenon is currently becoming more consistent. However, what remains controversial is the direct interaction of the optical field and the electronic excitation vs. phonon-mediated demagnetization. Hence it would be interesting to change the pump photon energy in the VUV / XUV range and study systematically the demagnetization after excitation at different photon energy while e.g. keeping the absorbed energy constant. In particular pronounced effects can be expected in the vicinity of resonant core hole excitations. This would lead to better insight into energy conversion through secondary electrons in the ferromagnetic system and their effect on the magnetic order.

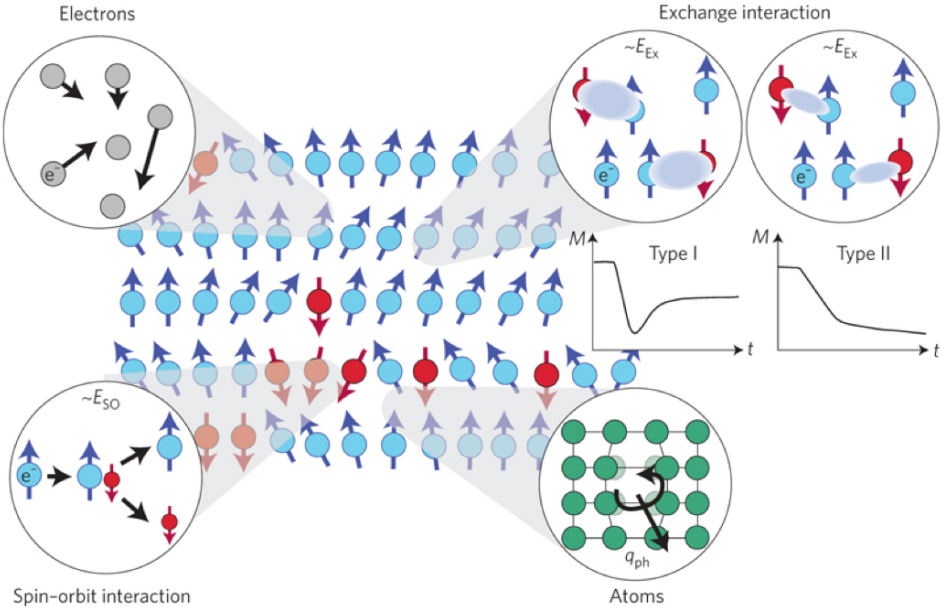


FIG. 14

C. Interaction of different degrees of freedom in the optically excited state of ferromagnetically ordered correlated materials.

Considering manganites like $RMnO_3$ it is interesting to study the coupling of lattice, charge, and spin degrees of freedom. While this has been done for charge and lattice, the spin degree requires the circular polarization. Actually, such an experiment is difficult to perform at the Berlin slicing source because it would require transmission of x-rays through the sample. Such samples are challenging to prepare because of the required crystalline order. In a FEL experiment the photon flux might be high enough to actually do a study in reflection. Here, it migt be interesting to access the M edge of the lanthanide R beyond 1 keV photon energy in addition to the O and Mn edges.

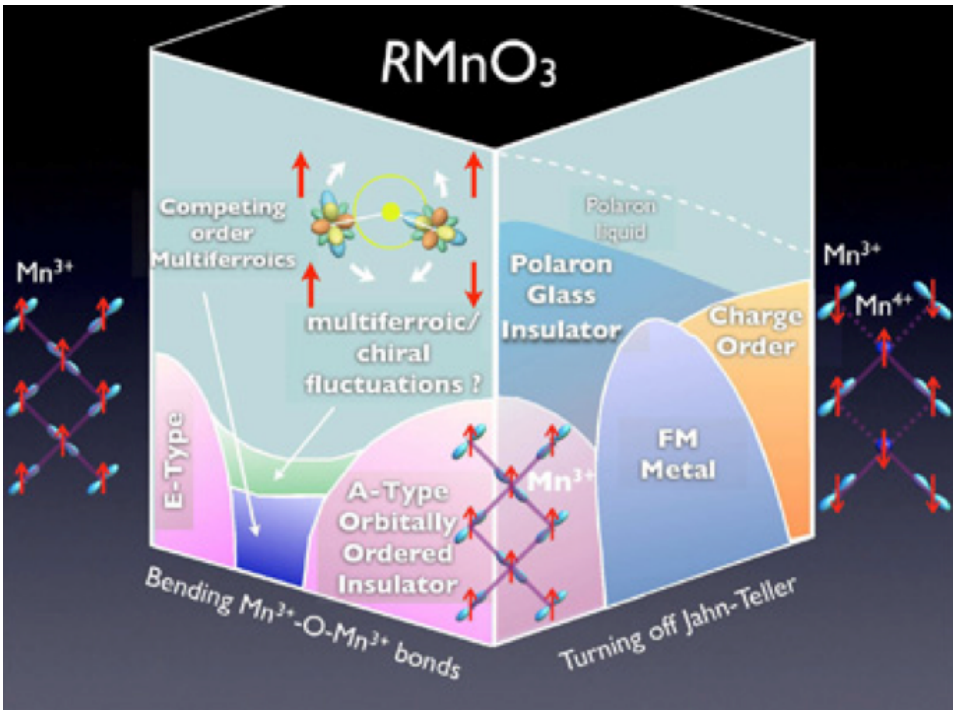


FIG. 15: Complex oxides display an extremely rich interplay of charge, spin, and lattice interactions. Nature has already shown how powerful this interplay is: superconductivity with the highest critical temperatures, ferroelectricity with exceptional dielectric response, colossal magnetoresistance, novel magneto-electric coupling and negative thermal expansion from 2 to 1400 K. Of the complex oxides, perovskites are likely the most versatile and engaging materials simply due to the enormous variety of physical phenomena they exhibit. The physical behavior of the perovskite manganites can be varied through this large array of phenomena by simply tuning the Mn-O-Mn bond-angles and the strength of Jahn-Teller interactions. The strong coupling between spin, charge and lattice that is found in these materials makes them the ideal playground to investigate strong effects using new state of the art facilities such as XFEL.[59–62]

D. Dynamics of magnetic fluctuations and magnetization relaxation processes

In a ferromagnet, excitation energy can be stored in the electron-, phonon- and spin-reservoirs. For the equilibration of a magnetic system both energy and angular momentum have to be transferred between these reservoirs with an overall conservation of both quantities. The underlying mechanisms

to effectively transfer angular momentum in the solid on the sub-picosecond time scales are still unclear. While optical pump-probe techniques allow for ultrafast excitations and the study of their evolution (probe) on the macroscopic scale by use of the magneto-optical Kerr effect, little is known about the processes on the atomic to nanometer length scale because of the lack of real or momentum space resolution of optical techniques.

Synchrotron-based resonant magnetic scattering [63] and coherent diffractive imaging [64, 65], [66], can address these issues if performed with fs time resolution. Unfortunately, x-ray femtosecond-slicing techniques at storage rings [12, 55] can not be employed for such experiments due to severe flux limitations preventing (high resolution) imaging. The length and the time scales of spin-lattice excitations by an optical pump can be uniquely probed by <100 femtosecond soft X-FEL probe pulses. By combining resonant magnetic diffraction, Fourier transform holography (FTH) [64, 65] [66], Gnther2008] and high-resolution HERALDO [67, 68] with an FELs high brightness, short pulse structure, and fully transverse coherence, the dynamics of magnetic fluctuations and magnetization relaxation processes can be studied at extremely fast time scales on the nanoscale.

E. Towards femtosecond control of magnetic anisotropy: Observing the temporal evolution of the orbital moment anisotropy following excitation with a strong laser pulse

Studies of ultrafast magnetization dynamics are currently mostly performed using visible or near infrared light from tabletop laser sources [7, 8, 24, 69, 70]. Some recent experiments have also been performed using x-rays from large-scale synchrotron X-ray facilities [12, 57]. Ultra fast lasers produce short pulses (< 30 fs), simplifying femtosecond time resolution experiments. X-rays on the other hand allow for element specific experiments in combination with high spatial resolution due to the high magnetic contrast at absorption edges. However, X-ray femtoslicing experiments - where a certain number of electrons are sliced out of a regular electron bunch of the standard filling pattern using a powerful femtosecond laser - have so far been limited due to the low photon flux. Significant efforts have thus also been devoted to perform table top x-ray experiments using high harmonic generation (HHG) from commercial femtosecond laser systems [48]. In these experiments the M-edges of the ferromagnetic $3d$ transition metals can be probed. However, it would be desirable to perform soft X-ray pump-probe experiments with high photon flux also at the L-edges since the application of the XMCD sum rules at these absorption edges is much more straightforward. The application of the sum rules in turn would allow us to extract information on the temporal evolution of the spin and orbital moments (or their ratio) after excitation with a short pump laser pulse. To overcome the limitation of photon flux (in X-ray slicing experiments) and low photon energy (in table to HHG experiments) a FEL source with variable (circular, linear) polarization would be of utmost importance. The limited repetition rate in the FEL (typically 50 Hz compared to 1 kHz in X-ray slicing experiments) will be more than compensated by the significantly higher photon flux per pulse. A particularly interesting and so far unique feature of the proposed set-up at Fermi is the possibility to perform laser pump X-ray probe experiments as a function of sample temperature (down to LHe) and in high magnetic fields of several Tesla. This opens up a completely new avenue for research in ultrafast magnetization or demagnetization dynamics. Of particular interest for ultrafast demagnetization dynamics experiments are materials with large magneto-crystalline anisotropy as used for example in media for future magnetic recording (e.g. FePt based alloys in the L10 phase). Here it is of interest to study the temporal evolution not only of the spin to orbital moment ratio, but also of the anisotropy of the orbital moment [71–73]. The materials of interest require large magnetic fields to saturate the sample magnetic moment along the hard magnetization axis. In the proposed set-up 7 Tesla magnetic fields are available to align the magnetization along

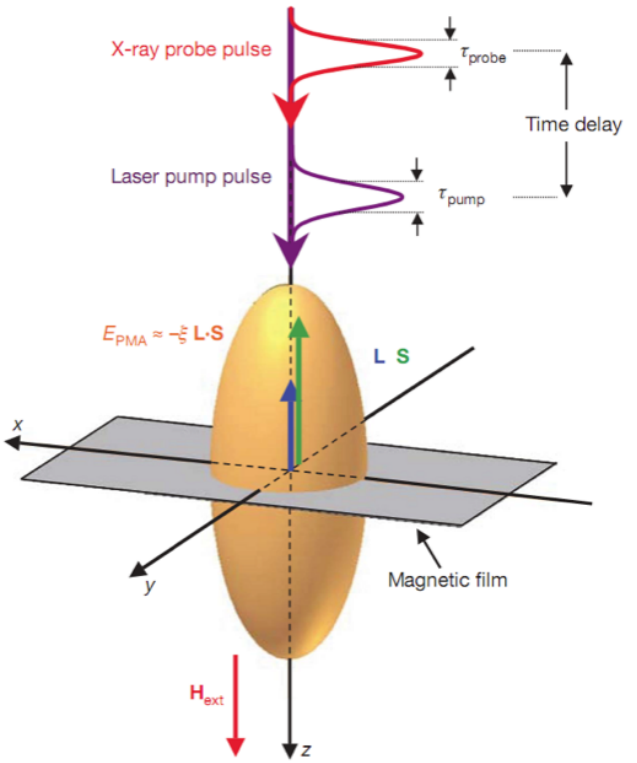


FIG. 16: Sketch of the geometry of the pumpprobe experiment at the femtoslicing synchrotron beam line at BESSY. Time-resolved XMCD allows measurement of the ultrafast dynamics of spin and orbital momenta along the quantification axis z parallel to the applied magnetic field. Optical pulses with a central wavelength of $\lambda_{pump} = 790$ nm and a duration of $\tau_{pump} = 60 \pm 20$ fs excite the ferromagnetic films perpendicularly, aligning the electric vector E in the film plane. The density of absorbed laser energy is $E_{abs} = 12$ mJ cm^{-2}. The ellipsoidal shape of -E_{PMA} illustrates the perpendicular anisotropy of the film. The easy magnetization direction is defined by the largest value of the z-axis projected value of L, (L_z). On applying the external magnetic field H_{ext}, the spin magnetic moment S aligns parallel to the orbital magnetic moment L along the z axis. A variable delay can be set between the near-infrared pulse and the X-ray probe pulse.[57]

any direction in the half plane, see Fig 16 [57]. The experiments will allow to gain insight in the temporal evolution of the orbital moment anisotropy in technologically relevant materials.

F. Spatial and temporal scales relevant for spin motion.

Spin motion - or in other words spin excitation - occurs at characteristic spatial scales and with characteristic times which are defined by the actual fundamental interactions governing magnetic behavior. The aim of this short section is to give an idea of the various scales.

Spatial scales. Magnetic behavior in matter is established by a number of fundamental interactions which might be in competition (see O. Portmann et al., in Encyclopedia of Condensed matter Physics, Elsevier Academic Press, 191 (2005), ISBN: 0-12-227610-8). Essentially, the intra-atomic exchange interaction, established by the Pauli principle and the atomic Hund rules, is the strongest one, being of the order of few eV per atom. This interaction is responsible for establishing the magnetic moment in atoms and solids. Its understanding and knowledge originates with the birth of quantum mechanics

and is established by atomic spectroscopy in great details. Any excitation involving this interaction occurs on atomic scales. On the next level of the energy scale (the range 10 to 100 meV per atom) is the inter-atomic exchange interaction, which is also of quantum mechanical origin. It is short ranged and favors, in some particular circumstances, parallel alignment of neighboring magnetic moments in solids against the Pauli principle which would prefer anti-parallel alignment in the ground state. Spin excitations involving this interaction – which means "rotating" the spins of neighboring atoms – cannot appear at nearest neighbor spatial scales, because of the existence of further fundamental interactions relevant for determining the magnetic state of matter: the spin-orbit coupling induced magnetic anisotropy also of quantum mechanical origin favoring some spatial direction of the spin to point at - and the dipolar interaction, which originates within the classical Maxwell equations of magnetostatic, and essentially favors antiparallel spin alignment over large distances. These last two interactions are much weaker – of the order of the 0.1 meV per atom – but by no means irrelevant: they are in fact in competition with the interatomic exchange interaction and therefore establishes minimum characteristic spatial scales for spin excitations, given essentially by the ratio of the interatomic exchange interaction and these small interactions. Typically, the domain walls - resulting from the competition between interatomic exchange interaction and magnetic anisotropy - are some few 10 nm wide and domains - resulting from the competition between the exchange interaction and the dipolar interaction - are at least some 100 nm wide. Any spin excitation (except the atomic sized one) cannot appear at spatial scales smaller than the one given here. **These are exactly the spatial scales accessible by state of the art microscopes based on X-rays**. Notice that these spatial scales are not accessible by conventional optical microscopes.

Temporal scales. Any interaction in magnetism can be transformed into a characteristic effective magnetic field by dividing it by the Bohr magneton. Any effective magnetic field exerts a torque onto the local spin by means of the Landau Lifshitz equation, so that one can associate to any effective magnetic field a characteristic time for spin excitation, by multiplying the effective field with the gyromagnetic factor of the electron and taking the inverse of the resulting product. In this way, we arrive at the result that at atomic scales the spin excitation leading to the destruction of the local magnetic moment occurs at the femto-second time scale and is the shortest process possible in spin dynamics. The characteristic time associated with the interatomic exchange is larger than this - some few 100 femtoseconds typically - and affects the dynamics at spatial scales of few 10 nm. This means that resolving spin dynamics driven by the interatomic exchange interactions requires short pulses **and** high spatial resolution (or nanometer-sized magnetic elements). At large spatial scales and larger times the spin dynamics is determined by the weakest magnetic interactions. While the spin motion at larger scales and larger times have been elucidated in the recent years by a string of spectacular pump-probe experiments, mainly performed with the new generation of Synchrotron radiation sources such as ALS, ELETTRA or SLS, our knowledge of magnetism in the spatial and temporal regime where quantum mechanics becomes relevant is very scarce, in particular due to the limitation of the spatial resolution of pump probe experiments based on table top laser experiments. The new generation of X-ray sources should allow extending our understanding in a new spatial and temporal regime which is bound to replace the current state of the art science and technology.

Advanced experiments: Magnetism in the ranges of space and time where quantum mechanics dominates is not known not even roughly and its study represents the most important challenge of future experiments with the new class of Soft X-ray Light Sources. Let us be more specific: We know e.g. that electrons passing through a magnetic materials change their spin polarization in analogy to the Faraday effect of optics (see W. Weber, S. Riesen H.C. Siegmann, "Magnetization precession by hot spin injection, Science **291**, 1015 (2001))). We suspect that this is made possible by transferring some spin to the target electrons, via the exchange interaction. Similar experiments where spin torque is relevant must work with the same rules. But we do not know at all how this

angular momentum is transferred and it evolution in time, which must be related to the interatomic exchange time scale. At this point, we also must point out that probing the inter-atomic time scale means probing the time evolution of the exchange process itself – recall that the exchange interaction literally involve the actual exchange of particles, which must take some time to happen (the exchange time) – and that one could think of being able to follow the actual particle exchange process itself. In other words: identical particles are indistinguishable in Quantum Mechanics, but while this is certainly true at large time scales, particles might become, for a very short moment, related to the inter-atomic exchange time, distinguishable.

G. Single shot experiments.

The detailed picture of magnetism acquired at the sub-micrometer spatial scales and sub-nanosecond time scales is restricted to such processes which can be accessed by pump-probe experiments, where the initial state is repeated many times to acquire statistics and the subsequent process follows always the same time evolution. However, on these spatial and temporal scales, there is a further class of phenomena which do not proceed in a deterministic way, such as the motion of a domain walls (see e.g. [74]) and the dynamics of other types of topological excitations and magnetic singularities, such as the vortex core motion. Non-deterministic processes also appears when the magnetic probe is subject to extremely strong magnetic fields required to reduce the precessional switching time [9]. Yet non-repetitive processes are exactly the kind of phenomena really happening in practical situations related to magnetic recording or to magnetic logical applications. The understanding of this kind of processes on 10 picoseconds time scale and with 100 nm resolution is potentially crucial for real applications and require the single shot or near single shot experiments which should be envisages within the new class of X-ray Light Sources.
A further yet unresolved problem, directly connected to the possibility of their motion, is the evolution of internal spin structure of domain walls or similar topological singularities during their motion. This is an intricate problem that depends on the actual driving force (Oersted field versus spin torque) and would clarify in details the relevance of both mechanisms during the motion. Imaging of these processes requires some ps temporal resolution and some 10 nm spatial resolution, again in a single shot mode.

Beyond the interatomic exchange time
As discussed above, the magnetic moment is defined at time scales of the order of the intra-atomic exchange interaction, i.e. femto-seconds. This means that at these scales we expect that the atomic magnetic moment can be manipulated by suitably exciting the magnetic system, i.e. with an intense laser pulse heating the sample well above the Curie temperature. Let us describe the physical situation for the simple case of a triplet and a singlet state. For a short time, the singlet atomic states (which are not occupied in the ground state, where the Hund-rules favor the triplet state) become occupied, leading to a strong decrease of the magnetic moment. There are experiments indicating that this rapid precessional motion between triplet and singlet state occurs (see e.g. [75]), but the process is very poorly understood and one hopes that resorting to X-ray **spin polarized** spectroscopy will allow sorting out in details the various channels along which the magnetization relaxes. Notice the technical aspects related to this kind of spectroscopy: when the excitation is provided by very short pulses, the formation of the spin polarization during the photo-excitation and photo-ejection process is a very interesting, yet non-understood aspect of the photo-emission process which is of course very relevant, as photo-emission (a very short process itself) is the method of choice for interrogating spin systems for future practical applications.

General problems in physics
A more general question in quantum mechanics relates to the tunneling process. While the tunneling process e.g. of the spin state in nanoparticles - is well studied both experimentally and theoretically in the Fermi-golden rule limit (see e.g. Gatteschi and Sessoli, Angew. Chem. Int. Ed. 2003, 42, 268, 2003), the details of its temporal evolution, like the tunneling times and the spin state during the tunneling process remain unknown and relates to the very essence of a quantum mechanical process. The spin must spend some time in the tunneling barrier, and is not clear how long it stays there and what is it doing there (see e.g. A. Steinberg, "How much time does a tunneling particle spend in the barrier region?", [76]. In other words: at short time scales quantum fluctuations must

be realized and the new generation of X-rays sources might be ideal to image them in real time and real space. We point out that small nano-objects - like the core of a spin vortex configuration in small disks suitably tuned to the quantum tunneling limit, might offer a unique opportunity to study the details of the quantum mechanical tunneling process with yet not existing insights and beyond the Fermi Golden rule time scale.

A further general question which might be tackled by the new class of X-ray sources relates to the very interpretation of quantum mechanics. According to the Dirac postulate of projection of states, any attempt to measure the state of a quantum mechanical system projects its state into one of the possible eigenstates of the relevant Hamiltonian. If very short times are available, one might start asking questions like How long is the time required for this projection process and how does it evolve in time (see e.g. [77, 78]). Because of the intrinsic non-deterministic nature of this process, one would rely on single shot experiments. The new generations of X-ray sources offers the real perspective of entering this realm of physics which has been established as postulates during the birth of Quantum mechanics and that actually still awaits a solid experimental validation or falsification. Associated to this type of questions is the question about the time it takes for a photon to be absorbed. The characteristic time scale of optical excitations is the inverse of the Rabi frequency. This time scale is typically, for atomic transitions, in the few picoseconds range (or larger). This means that a pulse of the order of few femtoseconds like the one delivered by X-ray free electron lasers will be probing a system where the optical excitation is – from the point of view of the Schrödinger type quantum mechanical evolution – not yet completed. Accordingly, excitation and decay are, on the time scales allowed by the new generation of pulsed light sources, not exactly separated in time. One must therefore start to ask whether the observations performed in this regime are also carrying some trace of the "collapsing" of the wave function which must have occurred on order that some excited states are populated during the short excitation pulse. Therefore the very experiments which are designed for detecting the microscopic details of the dynamics of an ensemble of interacting atoms actually are simultaneously asking questions related to the very foundations of quantum mechanics and to the actual measurement process.

H. Magnetization dynamics with time-resolved XMCD

Many central topics in condensed-matter physics are related to strongly correlated systems. A wide variety of experimental results and theoretical investigations in recent years have convincingly demonstrated that several transition metal oxides have dominant states that are not spatially homogeneous. This occurs when several physical interactions spin, charge, lattice, and/or orbital are simultaneously active and is at the origin of interesting effects, such as colossal magnetoresistance or high-temperature superconductivity. The spontaneous emergence of electronic nanometer-scale structures in transition-metal oxides and the existence of many competing states are properties often associated with complex matter where nonlinearities dominate. This electronic complexity could have potential consequences for applications of materials with strongly correlated electrons, because not only charge (semiconductor electronics), or charge and spin (spintronics), but in addition lattice and orbital degrees of freedom, are of relevance, leading in the most interesting cases to giant responses as a consequence of small perturbations. Moreover, several metallic and insulating phases compete, increasing the potential for novel behaviors (see for instance: [Tokura 2000, Dagotto 2005]). The field of spintronics is concerned with the search for highly spin-polarized materials, such as half-metals, with the aim of enhancing for instance the tunneling magnetoresistance of magnetic tunnel junctions, which are active members of magnetic random access memory elements. Half-metallic ferromagnets are solids that are metals with a Fermi surface in one spin channel, and insulators or semiconductors in the other spin channel. They give rise to 100% spin polarization at the Fermi energy, which makes them ideal materials for spin-dependent devices and spin injection. Recently, transition metal oxides (Fe_3O_4, $La_{0.7}Sr_{0.3}MnO_3$, CrO_2) have become the most actively studied half-metals. The electronic structure of transition metal oxides is not well understood yet because 3d electrons are strongly correlated and cannot be adequately described within a standard band theory framework. We are interested in the following aspects of the electronic structure: i) many-body effects (polaron or spin-polaron formation) and ii) microscopic processes governing magnetization dynamics. In this context, dissipation of spin angular momentum to other degrees of freedom on an ultrafast timescale is crucial. An efficient novel channel for angular momentum dissipation to the lattice can be opened by femtosecond laser excitation of a ferromagnet. With X-ray magnetic circular dichroism it has been shown [12, 57] that the quenching of spin angular momentum and its transfer to the lattice occurs with a time constant of about 100 fs. Spinorbit coupling together with coherent electronic excitations was invoked as a source of femtosecond demagnetization, although neglecting spinlattice relaxation. We propose to study magnetic transition metal oxides to time-resolved XMCD technique in which the probe femtosecond pulses from FEL FERMI@Elettra will be coupled to pump laser light.

I. Towards a comprehensive understanding of magnetization dynamics in complex materials.

In many materials with particularly intriguing properties, magnetism is a consequence of a coupling between spin, orbital and charge degrees of freedom. For example in transition-metal oxides the sign of the superexchange is determined by the relative orientation of the orbitals on neighboring metal sites [79], which effectively couples spins and orbitals and - via the orbitals - also spins and structure. A comprehensive understanding of magnetism and magnetic dynamics in such systems hence requires probing the interplay of all relevant degrees of freedom. This can readily be achieved very selectively via resonant soft x-ray diffraction [80, 81]. For instance in the doped layered manganites, which are a particularly well suited class of model systems, orbital order and spin order lead to different diffraction features, which are well separated in momentum space [82]. Similarly in stripe-ordered nickelates charge and spin order lead to separated superstructure reflections [80]. All these superstructure reflections can be studied at the respective $2p \rightarrow 3d$ resonance in the soft x-ray range, accessible at FERMI. From the temporal response of the different degrees of freedom to a perturbation induced by a pump-laser pulse the coupling mechanisms and the processes of energy and angular momentum transfer can be accessed. By following the dynamics of all degrees of freedom involved in magnetism a comprehensive understanding of magnetic dynamics in some of the most intriguing magnetic materials will be achieved.

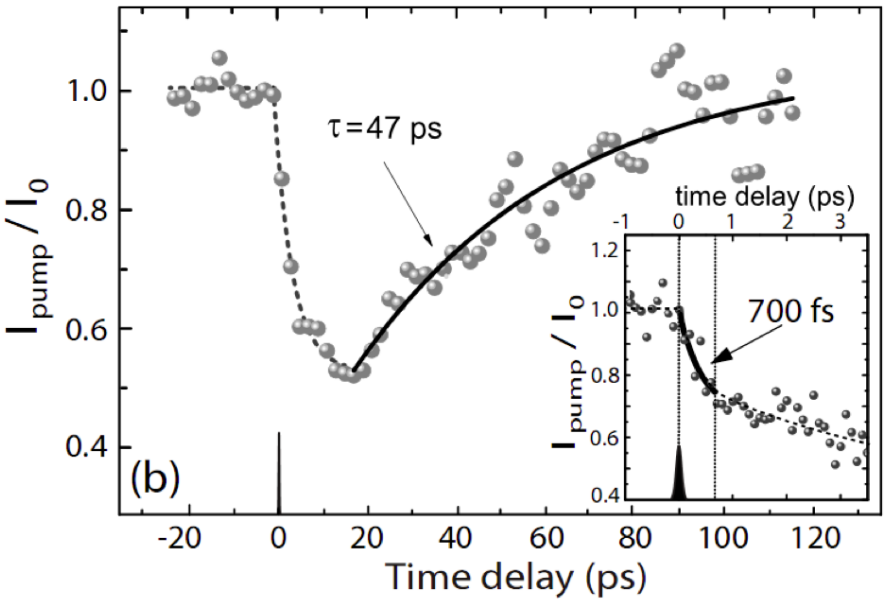


FIG. 17: Temporal response of the (1/2,1/2,1/2) antiferromagnetic diffraction peak in EuTe [[81]]. In order to understand the mechanism of the loss and recovery of magnetic order complementary information about other degrees of freedom would be helpful.

J. Magnetic dynamics in dilute systems.

For magnetic constituents in devices properties like magnetic anisotropy and band structure have to be carefully tuned to achieve the desired properties. This is typically done via alloying or by magnetically doping semiconductors. In order to understand magnetism in such materials XMCD as an element selective probe has proven to be most useful and magnetization dynamics has been

successfully studied at slicing sources like the femto-slicing facility at BESSY II [12, 57, 83–85]. One example is the complex behavior found in ferrimagnetically coupled GdFeCo system [I. Radu et al., Nature accepted (2011)]. The small flux available at slicing sources limits such studies so far to those constituents with fairly high concentration in the probed volume. For ultrathin films and laterally patterned systems this requirement is not necessarily fulfilled. In terms of physics and possible applications also dilute magnetic systems like dilute magnetic semiconductors [12], systems with magnetic impurities [86, 87], or Kondo-like systems are of highest interests. With the availability of high-flux circularly polarized soft x-rays at FERMI such systems can now be addressed. The results will considerably advance our understanding of magnetic coupling mechanism and how to tune them.

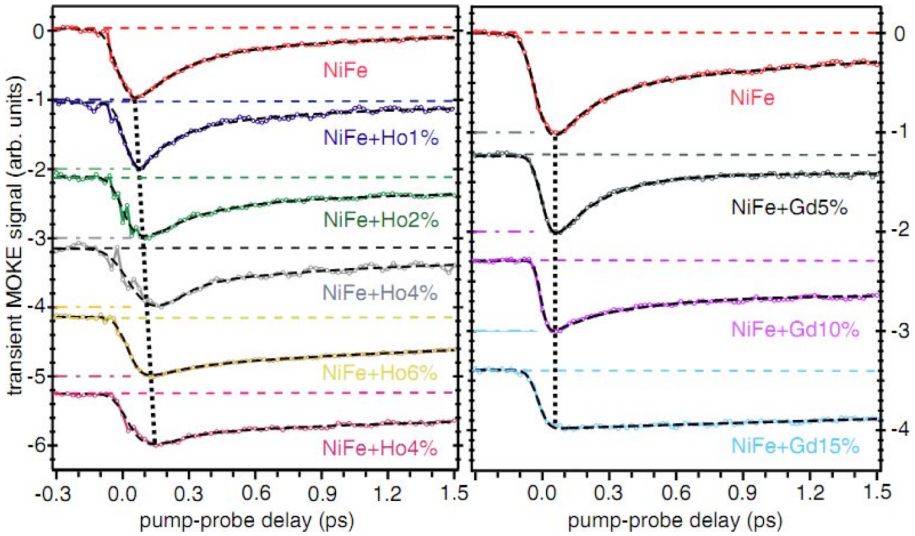


FIG. 18: Different lanthanide impurities ions in permalloy have very different effects on the magnetization dynamics [[69]]. An element-selective probe could clarify the interplay of the different constituents.

K. Transient phases in complex materials.

Complex energy landscapes in materials with different coupled degrees of freedom generally exhibit several local minima, out of which not all are reachable under equilibrium conditions. Far away from equilibrium the situation can be different, because the electronic system can be driven into an excited state too rapidly for the lattice to follow. From there the system may relax to a new transient state. Characterizing these states does not only help to understand the materials energy landscape itself, but may also reveal meta-stable state with intriguing new properties. An example is the recently discovered transient phase formed in magnetite under the influence of an 800-nm laser pulse. This phase persists for about one nanosecond and is characterized by the existence of orbital order and a drastically reduced band gap [N. Pontius et al. submitted]. It is hence completely different from all known equilibrium phases of this material. We expect to find such phases in various bulk and molecular systems and propose to characterize them with time-resolved spectroscopy at FERMI. Best suited for the pump-probe experiments are photon-in-photon-out techniques that are not affected by space charge effects induced by the pump laser. In order to characterize the transient phases we propose a combination of x-ray absorption spectroscopy and resonant inelastic x-ray scattering (RIXS), which allows to address the unoccupied density of states and the fundamental excitations. In particular for the comparably low RIXS-cross section a free-electron laser source is necessarily

required.

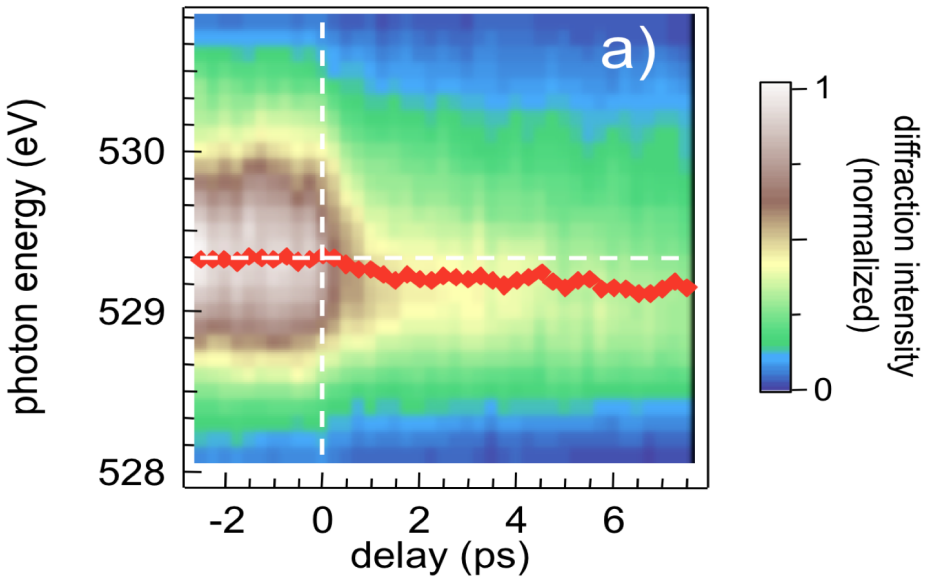


FIG. 19: After irridiation with an 800-nm laser pulse the resonance energy of the (00?) orbital order reflection in magnetite shifts down in energy, indicating a drastic reduction of the band gap and the formation of a transient phase [N. Pontius et al. submitted].

L. Ultrafast Spin Dynamics of Nanoscale Phase Transitions in Advanced Magnetic Materials.

A particular aspect of ultrafast spin dynamics, which is both scientifically very appealing and technologically highly relevant can be observed during magnetic phase transitions in advanced nanoscale magnetic materials. Observing and understanding for example the impact of spin fluctuations will shed light into the coupling between spin-charge-lattice parameters and associated relaxation channels in complex materials systems. Time resolved XMCD spectroscopy at FEL2@FERMI in an ultrafast optical pump - X-ray probe scheme will provide in a unique way element-specific information on the fsec temporal evolution of local spin and orbital moments during such phase transitions.

Furthermore, the short wavelength of soft X-rays enables in principle magnetic imaging with nanoscale spatial resolution, which seems to be feasible with novel (CDI) but also with conventional x-ray optics based techniques. Recent magnetic x-ray microscopy studies have revealed repeatedly strong stochastic components in various nanoscale magnetic processes, such as the nucleation and Barkhausen avalanche processes in nanogranular systems [88, 89]]. So far, the underlying principles of this non-deterministic behavior on a fast time scale is not clear. Following the relaxation of the spin system upon excitation with a fast optical pulse into a remanent state throughout the hysteresis loop, which mimics snapshots of the onset of the spin reconfiguration will help to elucidate these principles. These studies will not only have significant impact both for a deep and fundamental understanding of spin systems, but they will also support the quest to develop new functionalities in technologically relevant materials.

The overarching topic in the realm of spintronics deals with the exploitation of the spin as the major degree of freedom. Research on spintronic combines scientific beauty due to the quantum mechanical character of the spin, but opens also the opportunity to decrease power consumption in future devices by exploiting both the spin and charge degrees of freedom of an electron individually. Concomitantly, there is an ongoing quest to discover advanced magnetic materials with enhanced performance, such as having a high spin polarization to act e.g. as spin injectors. It has been shown, that the interface in a heterostructure of thin magnetic films is largely responsible for spin injection and transport e.g. in spin torque devices [90].

Heusler compounds offer a plethora of spintronic and multiferroic applications including magnetic tunnel junctions and spin momentum transfer devices. For example, in Ni_2MnGa there is a martensitic transition between a cubic and a tetragonal phase where the axis of tetragonal distortion is coupled to the magnetocrystalline easy axis, allowing ferromagnetically driven, ferroelastic actuation. Time resolved XMCD experiments will help to understand the dynamics and the magnetic character of this phase transition [91].

The compatibility with current Si technologies and given their high T_c makes Fe_xSi_{1-x} alloys highly desired and promising candidates as spintronic materials. In thin film form, varying growth parameters can stabilize different degrees of chemical and structural order, and these variations allow the electronic and magnetic properties of the alloy to be specifically tuned. Recent advances in thin film preparation allow to grow crystalline bcc-like Fe_xSi_{1-x} ($0.55 < x < 0.78$) thin films with varying chemical order (A2, B2 and $D0_3$ structures), and also amorphous films ($0.45 < x < 0.71$). These systems exhibit significant differences in both magnetization and anomalous Hall Effect (AHE) between crystalline and amorphous films of the same concentration. Amorphous films exhibit a magnetization larger than a crystalline film with the same composition and a very large AHE. These variations could be due to differences in the spin dynamics between the two systems. Hence, ultrafast time-resolved XMCD measurements on amorphous and crystalline FeSi samples will tremendously support the investigation of these peculiarities.

In the rush towards higher storage density media, the concept of a (Heat) Thermally-Assisted

Magnetic Recording ((H)TAMR) appears as an interesting path to follow. The idea is to overcome the superparamagnetic limit by lowering the switching field (H_{sw}) of the ferromagnetic storage medium at the writing temperature ($\sim 100C$), while H_{sw} remains high at room temperature (reading temperature) to preserve the stored information. This has been achieved by using exchange-coupled FeRh/FePt bilayers, where FePt is the highly anisotropic ferromagnetic storage layer [92–94]. The FeRh alloy in nearly equiatomic composition undergoes an antiferromagnetic (AF) to ferromagnetic (FM) first-order phase transition just above room temperature [95–97]. When in the AF phase (room temperature), it does not affect the magnetization reversal of FePt, whereas in the FM phase (above 100 C), it successfully reduces H_{sw} by exchange-spring coupling with FePt. The AF-FM transition can occur on a time scale of the order of a few picoseconds, which makes it an interesting specimen for ultrafast timing experiments. Tuning the Rh composition can be used to alter the AF-FM transition temperature. Fe-rich alloys can suppress the transition entirely, leading to a pure ferromagnetic ground state.

Because of the nature of the AF-FM transition, the metamagnetic transition is accompanied by a structural lattice distortion. The $Fe_{1-y}Rh_y$, system, with y lower than 0.52, has a bcc structure at room temperature, and an ordered phase (CsCl-type structure) is obtained above $y = 0.2$. It is well known that the ordered $Fe_{1-y}Rh_y$ alloy with $y = 0.48 - 0.52$ is antiferromagnetic up to about 350 K and that a first-order transition to a ferromagnetic takes place at that temperature. At the antiferromagnetic- ferromagnetic (AF-F) transition, the bcc phase retains but is accompanied by a volume expansion of the unit cell by about 1%. The transition temperature, which is sensitive to the degree of ordering, increases with increasing y. On the other hand, the fcc $Fe_{1-y}Rh_y$ is obtained in the range of y from 0.25 to 0.40 by quenching from high temperature to room temperature, since the martensic transformation temperature is lowered below room temperature. This means that extrinsic effects such as strain and grain size have a strong effect on the temperature of the transition as well as its breadth in both applied magnetic field and temperature. Previous work suggests that the AFM and FM states coexist during the phase transition but that while AFM domains nucleate uniformly, FM domains nucleate at defects and grain boundaries. Studies of single-crystal nanoparticles, these extrinsic effects could be examined in detail. Recently, the growth of epitaxial FeRh films either on regular MgO substrates or on transparent devices allowing for measurements in x-ray transmission and sustaining temperatures up to 300 C has been demonstrated. Although a lot of investigations have already been carried out on FeRh in a quasistatic field-regime (magnetization, heat capacity, Mossbauer spectrometry, photoelectron emission spectroscopy, XMCD), very few studies have been done on the time-dependence of the AF-FM phase transition [98], and nothing has been reported so far regarding time-dependent magnetization reversal in the FM phase.

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