Nanoscale Transport Phenomena

Thursday, November 30, 2023 - Friday, December 1, 2023

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Book of Abstracts

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Opening / 1

Fonda-Fasella Award: Profiling metal organic/metal hybrid interfaces: photoemission orbital tomography as an identikit tool

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Photoemission tomography originates from the conjunction of experiment and theory and finds use in obtaining information about the spatial distribution of individual molecular orbitals. Experimentally, it is based on momentum-resolved photoemission spectroscopy to obtain constant binding energy 2D momentum maps, revealing information about the electron probability distribution in molecular orbitals. Theoretically, these tomograms are visualized as hemispherical cuts through the molecular orbital in momentum space. Up to now, photoemission tomography (PT) has been applied to various organic molecules forming well-oriented monolayers on single crystal surfaces or to two-dimensional materials, allowing the assignment of molecular orbitals and the extraction of geometric information, among others. Still, many other new developments have been achieved recently, allowing to follow chemical and physical modifications taking place at molecular/metal interfaces.

From following possible on-surface reactions, whether inter- or intramolecular [1], to tracking the occupancy of unoccupied layers via charge transfer from the surface or by alkaline doping [2] and defining the energy level alignment and orientation of the different layers forming p/n junctions [3] - these are just a few cases where PT has recently expressed its potential.

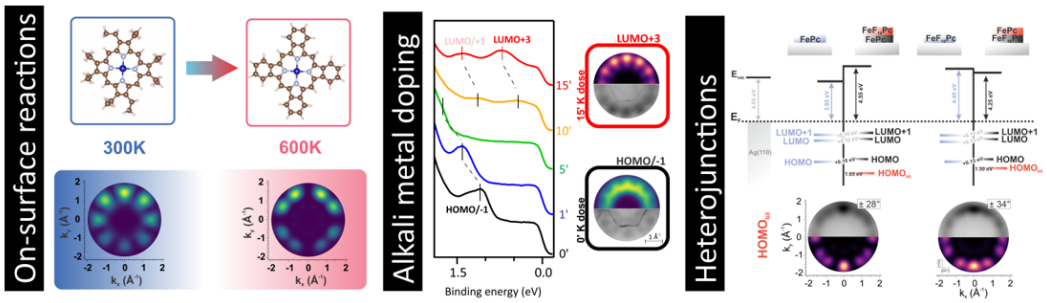

Figure 1: Applications of photoemission tomography on different molecular/metal systems

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Opening / **2**

Experimental facilities at FERMI

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The interest in studying dynamic phenomena at femtosecond-nanometer (fs-nm) length-time scale has motivated technical advances, such as the development of free electron lasers (FELs), and resulted in relevant achievements in many research fields, ranging from physics and material science to chemistry and biology. Reaching the frontier of the fs-nm scale and the energy scale of chemical bonds, holds the promise of getting the very essence of dynamics in molecules and nanostructures, that are the building blocks of all types of matter.

In this short introduction we present the unique opportunities opened by the FERMI seeded FEL [1], including the special operation modes (multi-colors, polarization and phase control, etc) that have enabled innovative experimental approaches, such as coherent control [2] and extreme ultraviolet four wave mixing (EUV FWM) [3]. We will provide an overview of the FERMI experimental facilities, i.e.: five FEL beamlines, a THz beamline and two laser laboratories, as well as the envisioned upgrade of the FEL source and instruments, with the outlook of reaching the photon energy range typical of L-edges of 3d transition metals.

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Spin transport / **1**

Exploring the role of ultrafast energy transport in nanoscale magnetization switching

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In order to establish all-optical techniques in future data processing technology, it is crucial to understand the processes that define the fundamental spatial limits of ultrafast laser induced magnetization reversal. The key prerequisite to achieve all-optical switching is to keep sufficient energy within the electronic system for a characteristic time scale before it is transferred locally to the lattice or –in the case of nanoscale excitation gradients –is lost by ultrafast, lateral energy transport.

To explore all-optical switching as well as lateral energy transport on the nanometer spatial scale, we performed transient magnetic grating experiments at the free electron laser FERMI in Trieste, Italy. We imprinted excitation gratings in a GdFe alloy with periodicities of 87 nm by interference of two coherent femtosecond light pulses in the extreme ultraviolet spectral range. The subsequent ultrafast evolution of the magnetization pattern was probed by diffraction of a third, time-delayed pulse tuned to the Gd N-edge at a wavelength of 8.3 nm. By examining the simultaneously recorded first and second order diffraction and by performing reference real-space measurements with a wide-field magneto-optical microscope, we conclusively demonstrated the ultrafast emergence of all-optical switching on the nanometer length scale [1].

In a follow up experiment, we managed to follow the ultrafast dynamics of magnetization gratings with systematically decreasing periodicities down to only 17 nm, i.e. with excitation gradients approaching the mean free path of excited electrons. By combining atomistic spin simulations with simple two-temperature diffusion models, we could extract spatial-temporal maps of the electron temperature to determine the nanometer spatial limit for optical control of magnetization [2].

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Spin transport / 2

First-principles-based investigations of optically induced ultrafast demagnetization

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The origin of laser-induced demagnetization has been much debated since its discovery [1]. To understand the mechanism of light-matter interaction on ultrafast timescales, we use the time-dependent density functional theory (TDDFT) [2] and compute the optically-induced demagnetization of L1 ferromagnetic FePt. Our calculations show that the fs demagnetization in TDDFT is a longitudinal magnetization reduction and results from a nonlinear optomagnetic effect, akin to the inverse Faraday effect. The demagnetization scales linearly with the intensity of the pump pulse in the perturbative limit, i.e., $\Delta M_z \propto E^2$. Moreover, the magnetization dynamics happens dominantly at even multiples $n\omega_0$, (n = 0, 2, ...) of the pump-laser frequency ω_0, whereas odd multiples of ω_0 do not contribute.

We investigate the importance of coherence in the electron density matrix, given by off-diagonal elements, in contrast to changes in the spin occupation numbers, given by the diagonal elements. We show that electronic coherence plays a dominant role in the demagnetization process, whereas interpretations such as optically-induced spin transfer, based on occupation numbers, poorly describe the ultrafast demagnetization. Dynamical correlations within the density-functional framework are shown to have an appreciable yet distinct effect on the amount of demagnetization depending on the laser intensity [3].

Secondly, we investigate the importance of ultrafast magnon degeneration for the demagnetization process. We develop quantum kinetic equations [4] to treat the full nonequilibrium and mode dependent electron-magnon scattering process, assuming an s-d exchange interaction. Our first-principles-based calculations for Fe predict the excitation of nonthermal magnons and a demagnetization consistent with experiments.

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Spin transport / 3

Picosecond spin emission from antiferromagnets

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Interfaces in heavy metal–antiferromagnet heterostructures have recently become highly investigated and debated systems in the effort to create spintronic devices that function at terahertz frequencies. Such heterostructures have great technological potential because antiferromagnets can generate sub-picosecond spin currents which the heavy metal can convert into charge signals. In this talk I will present our recent work on the optically induced picosecond spin transfer from antiferromagnets to Pt using time-domain THz emission spectroscopy. We will focus on two studies in antiferromagnetic insulators $KCoF_3$ and $KNiF_3$, and in antiferromagnetic metal FeRh. Through our studies, we are able to shine light on the microscopy of spin transfer at picosecond timescales and identify key figures of merit for its efficiency. Our results are important for progressing in the fundamental understanding of the highly discussed physics of the heavy metal/antiferromagnet interfaces, which is the necessary cornerstone for the designing of femtosecond antiferromagnetic spintronics devices with optimized characteristics.

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Phonon transport / 1

The effect of echoes interference on phonon dynamics in a nanophononic SiN membrane

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Nanophononic materials are at the forefront of current research for thermal management applications. These are periodic nanostructures, able to generate constructive and destructive phonon interferences that profoundly change phonon dynamics. The periodicity imposes a new Brillouin zone, whose range in wavevector is much shorter than the one "intrinsic"to the material and related to interatomic distances.

As such, phonon dispersions and velocities are strongly modified, leading to an effective thermal conductivity reduction [1-2]. However, the role of periodicity in phonon attenuation remains unclear. In this work, we have used the extreme ultraviolet transient grating technique (EUV-TG) [3] to measure phonon frequencies and lifetimes in a nanoporous phononic membrane at wavelengths between 50 and 100 nm, comparable to the nanostructure lengthscale. Phonon lifetime is found to be strongly reduced in the nanostructure. Supported by finite element calculations, we show that this is due to an increasing contribution of coherent phonon interference, which becomes dominant for wavelengths between about half and twice the inter-pores distance. Despite this, vibrational energy transport is enhanced through an energy flow among the coherent modes created by reflections. This interference of phonon echos from periodic interfaces is likely another aspect of the mutual coherence effects recently highlighted in amorphous and complex crystalline materials.

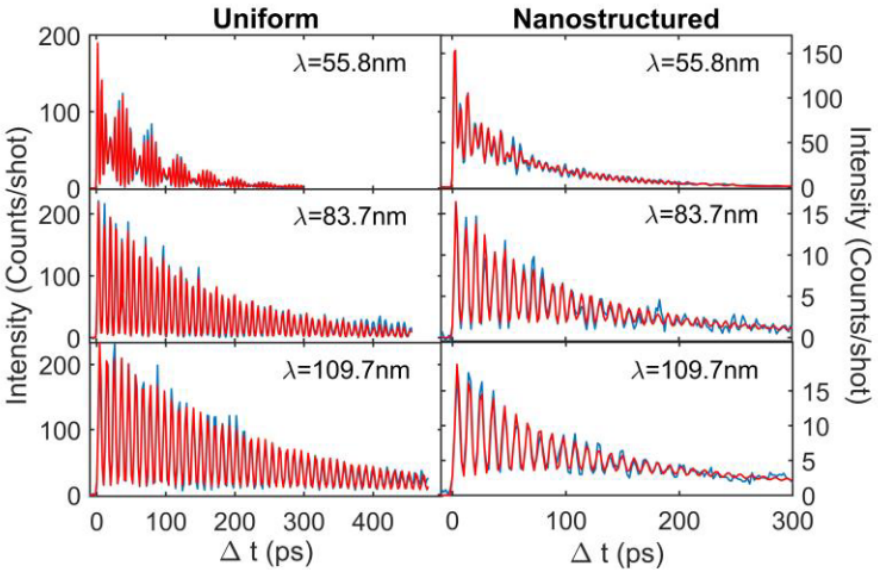

Figure 1: EUV TG spectra collected on a uniform (left column) and a nanostructured (right column) SiN membrane (blue). the individual panels. Red lines are best fit. The phono wavelength (equal to the TG period) is indicated.

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Phonon transport / 2

Ultrafast X-ray diffraction studies of energy transport at the nanoscale

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Ultrafast X-ray diffraction (UXRD) experiments provide a unique access to coherent vibrations and energy transport at the nanoscale.[1] Bragg-peak shifts are especially useful experimental observables in nano-layered systems, and contemporary laser-based sources of hard x-rays with femtosecond pulse duration have sufficient x-ray flux and stability to analyze the dynamics of films with single-digit nanometer thickness [2,3], although XFEL studies excel [4]. We shall discuss recent experiments on nanoscale metals and metal-insulator structures with examples for materials that are relevant for ultrafast magnetism and plasmon assisted chemistry.

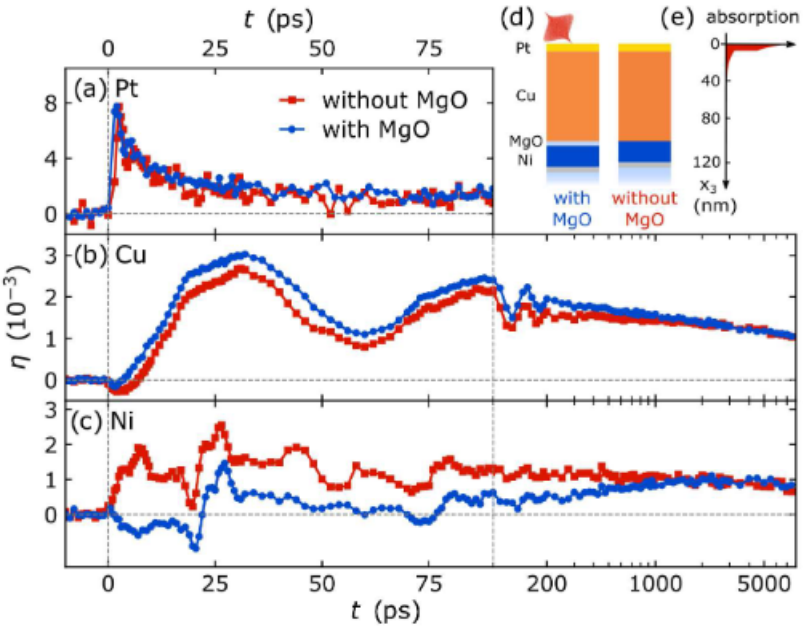

Figure 1: Comparison of the picosecond strain response in Pt–Cu–(MgO–)Ni heterostructures measured by UXRD: We measure the transient strain in the (a) Pt, (b) Cu and (c) Ni layers of the heterostructures with and without MgO interlayer as depicted in (d) with absorption (e) in the Pt layers. The suppression of the electronic heat transport from Pt to Ni by the MgO interlayer changes the strain response of the Ni detection layer from expansion to compression and delays the rise of the quasi-static expansion associated with the Ni heating. (reproduced from ref. [1])

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Phonon transport / 3

Hypersound and heat transport in functional nanomembranes

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Continuous miniaturization of electronics, increasing computing power and data transmission speed, an alternative to silicon-based electronics, and obtaining new sources of clean energy are among the most vital challenges in physics, chemistry, and material engineering. The search for new materials, structures, and composites that can meet these requirements has contributed to the spectacular development of na-noscience and nanotechnology in the last two decades. What is essential, the search for new nanomateri-als with application potential is often a compromise between excellent properties and incurred energy and environmental costs.

In this talk, I will present the results of experimental research focused on quasi-2D structures such as few-nanometer thick freestanding membranes based on organic and inorganic materials. The mains

fo-cus will be on (i) the energy harvesting in nature-inspired membranes for ultra-fast light-to-motion con-version [1], (ii) consolable flow of heat in silicon thermal diode [2], (iii) elastic size effect in $MoSe_2$ [3], (iv) GHz signal filtering in 2D hybrid colloidal crystals [4]. The core part of the talk will be dedicated to mechanical and thermal evaluation of ultra-thin materials utilizing all-optical methods.

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Phonon transport / **4**

Investigating phonons and thermal transport by pump-probe spectroscopy

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Controlling nanoscale heat transfer is crucial to design next-generation electronic devices as heat management becomes a bottleneck to increase their efficiency. However, the governing mechanisms for heat transfer at these length scales are completely different from bulk-counterparts and are not well understood. Therefore, as electronic devices continue to shrink in size, the demand for precise and efficient characterization tools becomes increasingly crucial to overcome or complement the limitations of conventional measurement approaches.

We implemented a novel and versatile experimental setup based on pump-probe spectroscopy, to explore electron and phonon dynamics, capable of performing transient reflectivity and time-resolved Raman spectroscopy measurements under the same conditions. I will therefore introduce the principles and applications of transient reflectivity, which serves as a valuable tool in capturing changes of reflectivity after ultrafast excitation, offering unique insights into the dynamic behavior of electron, and phonon processes. Moreover, I will introduce time-resolved Raman spectroscopy as a powerful method, that provides a distinct perspective by probing vibrational modes associated with lattice vibrations. This technique enables direct observations of phonon lifetime given by anharmonic effects, shedding light on the intricate interplay between lattice dynamics and thermal transport phenomena.

We applied these techniques to classic semiconductors, such as silicon and germanium, showing diverse mechanism responsible for the energy flow after ultrafast excitation, critical to understand heat dissipation in miniaturized devices. These results prove that the combination of these techniques is an effective approach to investigate materials for efficient next-generation electronic devices.

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Spin transport / 4

Single femto-second laser pulse to switch magnetization in a spintronic device

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In 1996, Bigot et al. made a major discovery that launched the new research area of ultra-fast magnetism by demonstrating that a femtosecond laser pulse excitation could generate a sub-picosecond demagnetization of a thin Ni film [1]. However, a complete deterministic all-optical switching (AOS) was only demonstrated ten years later using circularly-polarized laser pulses on a GdFeCo ferrimagnetic alloy [2]. This phenomenon was later named all-optical helicity-dependent switching (AO-HDS) [3]. AO-HDS can be observed for a large variety of samples [4,5]. The main inconvenience of AO-HDS is the necessity of a large number of pulses, which makes AO-HDS a slow process [6]. Another type of AOS, single pulse all-optical helicity-independent switching (AO-HIS), is more promising for applications because the reversal is much faster [7].

Here we will only consider deterministic magnetization switching induced by a single femtosecond or picosecond laser pulse, we will consider magnetization reversal resulting from the direct interaction between the ultra-short laser pulse and the magnetization reviewed recently in [8]. We will also discuss how light can generate spin-polarized femtosecond current pulses to reverse the magnetization of thin ferromagnetic films in magnetic heterostructure reviewed recently in [9]. Finally our last results demonstrating ultra-fast magnetisation reversal (in less than one pico-second) [10] in various perpendicularly magnetized ferrimagnetic and ferromagnetic spin-valve structure [11-14]. Those results open the field of ultra-fast spintronics.

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Electron transport / 1

On the element-selectivity of core-level transient grating spectroscopy

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Transient grating (TG) spectroscopy is one of the most established four-wave mixing methods. The decay of the excitation grating (heat, charges, magnetic, etc.) is usually due to energy relaxation and transport phenomena. Extreme ultraviolet (EUV) TG spectroscopy is by now well-established and has been used to explore dynamical effects and transport phenomena, mostly in solids. [1–5] Its extension to the hard X-ray regime was recently demonstrated, promising to access nanoscale transport using the Talbot effect in order to generate two identical incident beams that cross on the sample. [6] I will present recent results demonstrating hard X-ray TG spectroscopy of liquid solutions that were recently obtained at SwissFEL.

In the optical domain, TG spectroscopy has been the forerunner of multidimensional (MD) spectroscopies. The ultimate goal of current efforts in short wavelength non-linear methods is to achieve MD EUV/X-ray spectroscopies. These would allow interrogating specific atoms probe their cross-talk in the course of a photoinduced process. Crucial to reach this goal is the ability to demonstrate that the excitation triggered by the incident beams is specific to the element one tunes to and is not due to a cascade of Auger electrons triggered by the core-excitation. So far attempts to demonstrate it have pointed to the latter rather than the former. [3]

I will present our recent results on the EUV TG spectroscopy of spinel Co_3O_4, a material that consists of both Co^{2+} and Co^{3+} centres with tetrahedral and octahedral coordination, respectively. These results point to an element-selectivity of the EUV induced excitation. I will discuss the potential for future developments in core-level MD spectroscopies.

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Electron transport / **2**

Lattice dynamics in 2D materials probed by time resolved soft X-Ray spectroscopies at Elettra synchrotron

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In the fields of optoelectronics there is growing interest in studying the response to optical excitation of 2D materials as these systems are of fundamental relevance for the development of the next generation of environmentally sustainable optoelectronic devices and catalysts. In order to probe the lattice dynamics in such materials down to the sub-nanosecond timescale we developed a setup at the ALOISA beamline of the Elettra synchrotron that exploits the chemical selectivity of X-ray photoemission (XPS) in an optical pump/X-ray probe experiment. In this talk, I will present our results on the study of the early times in the 2H–1T′ phase transition induced by optical excitation 2H-$MoTe_2$ [1]. I will discuss hydrogen dehydrogenation of nanoporous graphene obtained by laser heating [2]. Finally, I will present the recent results on the CDW in $TbTe_2$ as seen by core level photoemission.

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Electron transport / **3**

Imaging heterogeneity in light induced phase transition on the nano-femto scale

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Heterogeneity in phase transitions is common when they occur near equilibrium, but what happens far from equilibrium? One of the most studied light-induced non-equilibrium phase transitions is the insulator-metal phase transition in VO2. THz spectroscopy has suggested that the photo-induced phase transition occurs via nucleation and growth, while time resolved diffraction has suggested that new, non-thermal phases are generated. However, the process has not been directly observed.

Here, I will present our recent results using the PAL XFEL to image the phase transition with holography with 25 nm spatial resolution and 150 fs time resolution. I will show that in our conditions, neither proposed scenario occurs, in stead, we can attribute the domain dynamics to a phase transition at constant volume, followed by a volume expansion.

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Poster Session / 1

2D Materials Intrinsic property and OER reaction mechanism: Evidence from in-situ XAS studies

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Efficient water oxidation is a pivotal process in the production of clean hydrogen fuel through water splitting, hinging on the effectiveness of catalysts. 2D materials, celebrated for their distinctive attributes like high surface-to-volume ratios and customizable electronic structures, hold immense potential for advancing this critical reaction. Nonetheless, a series of challenges persists in harnessing their full potential. First and foremost, optimizing the activity, stability, and scalability of 2D nanostructures remains imperative [1].

To address this, researchers employ innovative techniques such as surface reconstruction and heterointerface engineering, which enhance key properties like electrical conductivity, stability, charge transfer, and the activation of catalytically active edge sites, all of which significantly impact the performance of these materials in water oxidation. Furthermore, the surface properties of 2D materials play a pivotal role in their catalytic performance, necessitating meticulous attention to surface chemistry and morphology for achieving superior catalytic activity and long-term stability. Electrochemical performance, another critical facet, demands meticulous optimization to ensure the efficient use of 2D materials in water oxidation. Additionally, the integration of catalysts poses its own set of challenges, with strategies ranging from anchoring catalysts onto 2D material surfaces to encapsulating them within nanostructures to bolster water oxidation efficiency. Beyond these technical hurdles, the synthesis and characterization of 2D materials for water oxidation require scrupulous consideration. Recent advancements in the field, including insights derived from our group, underscore the pivotal role of 2D composites in elevating the efficiency of water oxidation processes. The combination electrochemical measurements with in-situ XAS to unveil the catalytically active phases, pinpoint the reaction centers, and elucidate the intricate mechanisms underlying the OER [2].

A noteworthy discovery is the dual participation of confinement, working in tandem to deliver exceptional OER performance. Moreover, the confinement of serves as an ingenious safeguard, averting aggregation or losses that might occur in free space, thereby ensuring improved cycling stability of the catalyst. Importantly, this study emphasizes the instrumental role played by confined materials as pre-catalysts in the water-splitting process, potentially paving the way for groundbreaking advances in various energy conversion and storage technologies.

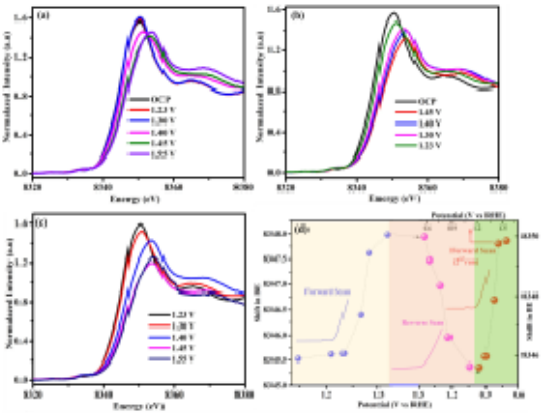

Figure 1: Electrochemically controlled in-situ Ni K-edge XAS.

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Poster Session / 2

Low-dimensional liquid-crystalline halide perovskites

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Hybrid Organic-Inorganic Halide Perovskites (HOIHPs) represent an emerging class of semiconducting materials, widely employed in a variety of optoelectronic applications. Despite their skyrocket growth in the last decade, a deep understanding on the structure-properties relationship is still under investigation. Moreover, other properties may still be overlooked. In this communication we report two novel intrinsic liquid crystalline halide perovskites by utilizing fluorinated ionic liquid crystalline (FILC) mesogens based on polyfluorinated alkylimidazolium cations [1]. We explored the solid-to-liquid crystalline transi-tion and the nature of the emerging stable liquid crystalline phases. The multiple intermolecular fluorous-fluorous interactions were found to be substantial for the solid- and liquid- crystalline order of the liquid crystalline perovskites. Moreover, the structure of the incorporated FILC was found to tune the proper-ties of the liquid crystalline phase. Collectively, these results may pave the way for a new class of halide perovskite-based soft materials.

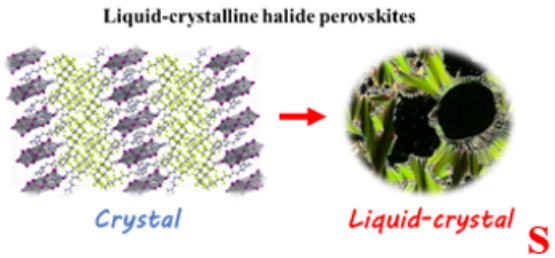

Figure 1: Schematic illustration of the solid-crystalline structure of the developed perovskites and the revealed ther-mally triggered liquid-crystalline phase imaged under a polarized optical microscope (POM).

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Poster Session / 3

Ultrafast X-ray induced magnetization dynamics in Co and Ni

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We investigated the role of electronic excitation, relaxation and transport processes in X-ray induced ultrafast demagnetization of magnetic single- and multilayer systems. In what follows, we report on the results obtained with the newly developed modeling tool, XSPIN, which enables nanoscopic description of electronic processes occurring in X-ray irradiated ferromagnetic materials [1,2]. With this tool, we have studied the specific response of cobalt/platinum (Co/Pt) multilayer system irradiated by an ultrafast XUV pulse at the M-edge of Co (photon energy around 60 eV) and the response of cobalt/palladium (Co/Pd) multilayer system at the L-edge of Co (photon energy around 778 eV) [1,2]. Those were previously studied experimentally at the FERMI and LCLS free-electron facilities respectively [3,4]. The XSPIN simulations show that the magnetic scattering signal from cobalt decreases on the femtosecond timescales considered due to electronic excitation, relaxation and transport processes both in the cobalt and in the non-magnetic layers. The signal decrease scales with the increasing fluence of incoming radiation, following the trend observed in the experimental data. Confirmation of the predominant role of electronic processes for X-ray induced demagnetization in the regime below the structural damage threshold, achieved with our theoretical study, is a step towards quantitative control and manipulation of X-ray induced magnetic processes on femtosecond timescales.

Additionally, we describe with XSPIN simulations the demagnetization observed in a single layer of nickel (Ni) [5], and present a more comprehensive extension of the XSPIN code, XSPIN+, which takes into account atomic movement and changing band structure predicted with density functional tight binding (DFTB) formalism

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Poster Session / 4

Ultrafast dynamics of photoexcited states in Cerium Oxide

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Transition metal oxides (TMO) have interesting catalytic properties that make them promising candidates for efficient photoelectrochemical catalysts. Carrier lifetime and mobility are crucial properties that determine the catalytic efficiency of these materials. It is then essential to investigate the dynamics of the electronic and structural modifications induced by photoexcitation, to understand and possibly tune the functionality of the material. For some TMO (an exemplary case is Fe2O3), the formation of small polarons affects the lifetime mobility of both charge carriers, hampering their use in photocatalysis. The possible formation of photoinduced polarons is detrimental for the oxide-based photocatalysts in which the photoactivity is based on the interaction between adsorbed molecules and photoexcited electrons. However, for some materials like CeO2, the functionality is based on the reversible exchange of oxygen ions with the reactants. For such oxides, the formation of photoinduced polarons can enhance reducibility and photocatalytic activity, while the structural deformations correlated with the charge localization, may also play a role in further decreasing the oxygen vacancy formation energy and enhance the redox functionality. In a recent study performed with pump-probe optical spectroscopy, an ultrafast blue shift of photoinduced absorption by 0.4 eV was identified and tentatively ascribed to the formation of photoinduced small-polaron states within ultrashort times (330 fs) from photoexcitation [1].

The experiment presented here exploits pump-probe X-ray spectroscopy to correlate the dynamics of the element-specific electronic modifications with the structural changes, to investigate the out-of-equilibrium processes which take place in the material after photoexcitation. The experiment has been carried out at the FXE instrument of the European X-FEL on a 50 nm polycrystalline CeO2 film. We performed pump-probe Ce L3 edge X-ray absorption spectroscopy in the near (XANES) and extended (EXAFS) energy range and Ce L(alpha) X-ray emission spectroscopy (XES) experiments, exploiting the ultra-short and ultra-intense free electron laser (FEL) pulses to probe the dynamics of photoinduced modifications of the local electronic and atomic structure around cerium ions with a time resolution

below 100 fs. We excited the film using a laser pump with energy above the CeO2 band gap (4.6 eV) and we probed the dynamics of the excited states within a delay time range extending up to 250 ps. The pump-probe XANES spectra indicate a Ce^{3+}-like excited phase at short delay ($\leq$ 300 fs), which decays in 300 - 400 fs into a metastable excited state, tentatively ascribed to the formation of a photoinduced small polaron. This result is confirmed by the pump-probe Ce L XES spectra measurements, that show a compatible dynamics. The analysis of EXAFS spectra demonstrates the presence of structural distortions in the photoexcited state, compatible with small polaron formation. The formation of a photoinduced polaronic state is possibly responsible for the observed stabilization of the excited state for a lifetime above 250 ps.

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Poster Session / 5

Nanoscale structural dynamics by EUV transient gratings

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The study of collective dynamics of condensed matter at the nanoscale, such as heat transport processes, vibrational modes or magnetization dynamics, is important both for advancing fundamental science and modern technology. Experimental tools for probing such dynamics in the sub-100 nm length-scales and on the relevant timescale (i.e. picosecond and sub-ps) mainly rely on combinations between ultrafast lasers and ad hoc sample's nanostructuration.

In this contribution we present an alternative approach, developed at the FERMI free electron laser (FEL) facility (Triste, Italy), where the sensitivity to the sub-100 nm length-scale is obtained by using the extreme ultraviolet transient gratings (EUV TG) approach [1,2], which is a specific case of so-called four-wave-mixing (FWM) methods. The EUV TG is obtained by crossing two ultrafast EUV FEL pulses, while the dynamics stimulated by the EUV TG is probed by monitoring the transient diffraction of a third, time-delayed FEL pulse. We will discuss the application of this new experimental tool for studying nanoscale thermal transport in thin membranes of crystalline silicon and amorphous silicon nitride. In the amorphous sample the wavelength dependence of the thermal transport timescale is consistent with a diffusive behaviour [2], while in the crystalline sample the heat transport timescale shows a marked deviation with respect to the diffusive regime [2,3]. We will also show the possibility to generate and detect the dynamics of bulk and surface phonons on a previously inaccessible wavelength range and in ultra-thin samples [4].

Finally, we will briefly present the potential of EUV TG in other contexts, such as ultrafast magnetic dynamics at the nanoscale [5].

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Poster Session / **6**

A Novel free-electron laser single-pulse Wollaston polarimeter for magneto dynamical studies

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Here we report on the conceptual design, the hardware realisation and the first experimental results of a novel and compact x-ray polarimeter capable of single-pulse linear polarization angle detection in the extreme ultraviolet (EUV) photon energy range. The polarimeter is tested by performing time resolved pump-probe experiments on a $Ni_{80}Fe_{20}$ permalloy film at the $M_{2,3}$ Ni edge at an externally seeded free-electron laser (FEL) source. Comparison with similar experiments reported in the literature shows the advantages of our approach also in view of future experiments.

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Poster Session / **7**

Time-resolved chemically-selective spectroscopic investigation of the Fe_2O_3/Al redox reaction

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the hematite (α-Fe_2O_3) / aluminium (Al) thermite redox reaction at the femtosecond timescale by using ultrafast extreme ultraviolet (EUV) transient absorption spectroscopy. After an ultrafast ignition of the reaction by an optical pulse the electronic dynamics was monitored by EUV pulses as a chemically selective probe, tuned to the Fe M and to the Al $L_{2,3}$ absorption edges. Our observations reveal a blue shift in the zero-absorbance point, emerging within the first few hundred femtoseconds. This shift is attributed to the formation of a small polaron in hematite, indicating a two-photon absorption mechanism as the excitation process. Additionally, the differences observed in the transient absorption traces for hematite deposited on aluminium versus parylene suggest the occurrence of an electron transfer between aluminium and hematite during the first phases of the redox.

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Poster Session / 8

Manipulating soft x-ray matter interaction by collision-induced changes of electronic populations

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The irradiation of a solid with focused soft x-ray FEL pulses transforms its constituent atoms into an electronically highly excited, transient state on an ultrashort time scale. In this regime, the solid is brought into the warm dense matter state and new bound-bound electron transitions may open –thus dramatically modifying the absorption as well as scattering properties of the solid. These phenomena were demonstrated for copper in two experiments at the European XFEL SCS instrument.

A strong absorption peak in the pre L-edge region appeared in the absorption spectrum of a thin Cu foil upon irradiation by focused XFEL pulse with photon energy around the Cu L-edge and intensities above 10^{13} W/cm^2. This absorption peak appears due to $2p_{3/2} \rightarrow$ 3d transitions opened due to holes in the Cu 3d shell that are produced by electron-impact ionization. This process takes place on a 10-100 fs timescale: L-shell photoionization and subsequent Auger-decay create an out-of-equilibrium population of continuum electronic states. Electron-electron and electron-ion/atom collisions then drive the system to a state of warm dense matter. At higher fluence, the pre-edge peak broadens, drops, and the spectral absorption profile becomes smooth without a pronounced L–absorption edge. To describe the observed absorption profile we combined the modeling based on plasma description and solid-state description. Namely, we used the kinetic Boltzmann approach to get the populations of atomic configurations, and real-space multiple-scattering code FEFF10 to calculate the x-ray near-edge absorption spectra. This enabled a qualitative explanation of the observed features in the absorption profile.

The fundamental relation between scattering and absorption suggests that the opening of absorption channels on $2p_{3/2} \rightarrow$ 3dd transitions should result in additional resonant elastic scattering contribution. This was demonstrated by spectrally resolving the reflected x-ray radiation around a superlattice peak of a $[B_4C$ (2nm) / Cu (2nm) / SiC (2nm)$]_{15}$ multilayer sample irradiated by focused XFEL pulse with a photon energy around the Cu L-edge. In line with the absorption experiment, at XFEL intensities above 10^{13} W/cm^2 we observed a strong enhancement of the diffracted intensity in the pre-edge spectral region. This demonstration paves the way towards control of atomic scattering properties.

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Poster Session / 9

X-ray resonant magnetic scattering setup for ultrafast magnetization dynamic studies at the CITIUS facility

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Studies of laser-induced magnetization dynamics in complex magnetic materials have been regularly carried out ever since it was first observed by Beaurapaire et al. [1]. With the advent of femtosecond light sources, these studies have drastically advanced, allowing us to investigate the nature of the processes at work, when the system is driven out of equilibrium. A setup for X-ray magnetic scattering studies has been developed at the CITIUS facility hosted by the University of Nova Gorica, Slovenia [2] to probe the magnetization dynamics at ultra-short timescales. The source beam is a commercial Coherent double-stage amplifier, seeded by a VITARA oscillator, producing mode-locked, Q-switched pulses of 35-fs pulse duration, with a central wavelength of around 800 nm and a bandwidth of about 70 nm. A High harmonic generation process is employed to produce photons with the energies required to probe the M edge of magnetic transition metals. The magnetic system is pumped with a fraction of the direct infrared. The setup has the flexibility of operating in both transmission and reflection methods. The entire spectral range of high harmonics produced is detected after the interaction of the sample and this provides the added advantage of comparing the dynamics of multiple elements in a material, simultaneously. Here, we report the results of the experiments that were carried out to commission the setup and provide some hints about possible future studies.

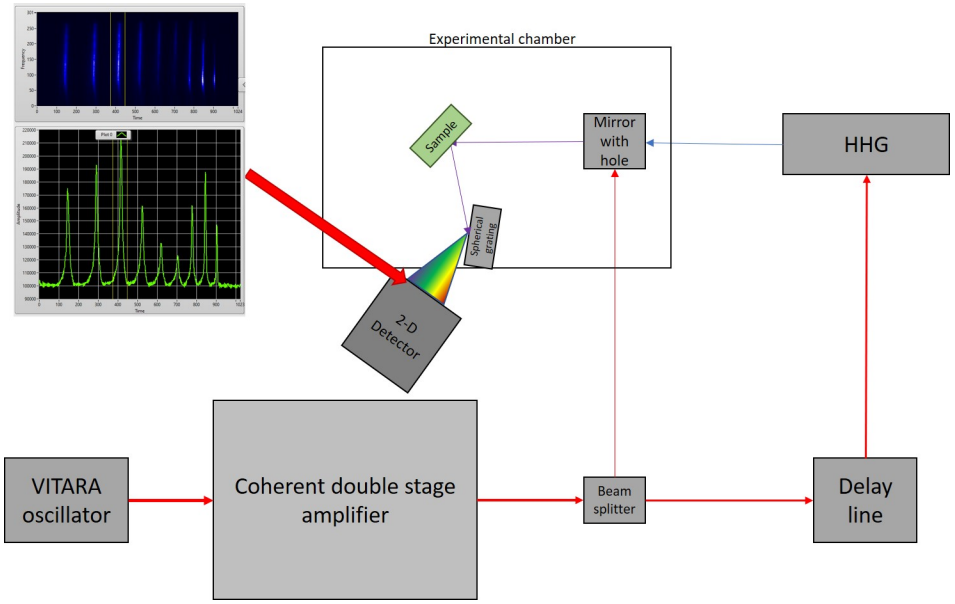

Figure 1: Scheme of the setup installed at the CITIUS beamline with a typical spectrum recorded during experiments.

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Poster Session / 10

Selective nanoscale thermoelastic response due to MoS_2 monolayer deposition in silicon-based heterostructures

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The attractiveness of 2D materials such as molybdenum disulfide (MoS_2) comes from their tunable physical properties and application in thin nanodevices [1]. An important aspect is the response to thermal perturbations, i.e., how heat is dissipated, thermoelastic effects leading to the generation of acoustic waves and substrate interactions [2,3]. In our work, we focus on studying the dynamics of a MoS_2 monolayer on top of a silicon substrate using 84-nm-period transient gratings generated with the FERMI free electron laser. The results present a marked difference in the thermoelastic response due to the monolayer coverage (Figure 1(a), (b)). While two acoustic modes, Rayleigh and longitudinal, are detected for the blank membrane, here only the longitudinal is clearly observed. We relate this to the different motion involved in the modes and discuss the implication for devices. Underneath the acoustic modulations, the thermal response results to be on average slower for the heterostructures rather than the filmless mem-brane. However, such decay constant is shown to be critically dependent on the excitation energy (Fig-ure 1(c)).

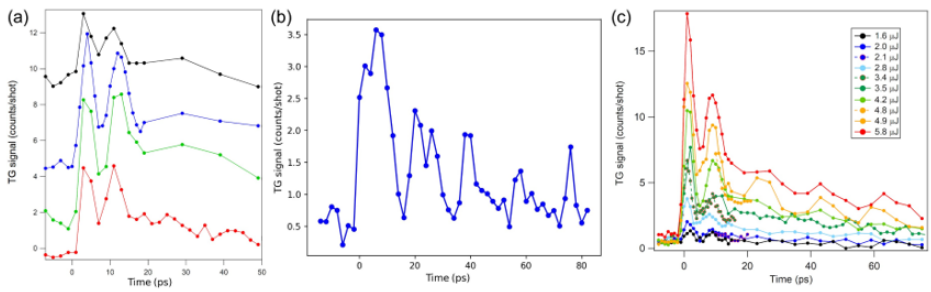

Figure 1: TG signals from (a) MoS_2/Si membrane at different sample positions (b) blank Si membrane sample (c) pulse energy dependence for the MoS_2/Si wafer sample.

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Poster Session / 11

Angular Momentum Relaxation Dynamics studied with Transient Gratings

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The most debated question in the field of ultrafast magnetism, still not fully answered after almost 30 years of research is: how is angular momentum dissipated after ultrafast demagnetization? Several explanations have been put forward, and it is believed that a full picture requires multiple considerations which include the electronic, lattice and spin degrees of freedom in the materials. However, in many cases, the explanation is rather qualitative. In order to answer this question, we use a novel technique, called the transient grating (TG) technique, which has been proven to be a sensible tool for studying ultrafast magnetization dynamics [1].

The time-dependent response induced by nanoscale extreme ultraviolet gratings in a thin film of CoGd alloy shows clearly different relaxation dynamics depending on the beam polarization and the detection geometry. Moreover, all-optical TG measurements exhibit similar behavior at a longer timescale due to the microscale gratings, suggesting a yet undisclosed intermediate relaxation mechanism for the angular momentum. These effects will be investigated further by performing resonant TG experiments at different absorption edges of the magnetic materials and at different wavevector excitations. We can only speculate at the moment, but new measurements comparing transient polarization and intensity grating employed with the polarization analysis of the TG signal may offer us a completely novel view of the physics at play. This will definitely help uncover, quantitatively, the role of the two most prominent scattering channels, i.e. the phonon and magnon systems.

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Poster Session / 12

Ultrafast magnetization enhancement in exchange-coupled composite magnets

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Tailoring all-optical magnetization in exchange-coupled composite magnets is of scientific interest for ultrafast magneto-optic devices. In this study, we investigate the femtosecond laser-activated magnetization dynamics in FeNi/FePt composite magnet using time-resolved magneto-optical Kerr effect (tr-MOKE) spectroscopy across the visible and extreme ultraviolet spectral ranges. The visible tr-MOKE reveals the ultrafast demagnetization and transient magnetization enhancement occurring on timescales shorter than 1 ps followed by slower recovery. Resonant tr-MOKE at Ni $M_{2,3}$ and Pt $N_{6,7}$ edges provides insights into the individual element contribution to the magnetic dynamics. The resonant tr-MOKE was carried out at the MagneDyn end-station, located at the externally seeded Extreme Ultra-violet Free-Electron Laser (EUV FEL) FERMI at Elettra Sincrotrone Trieste. We infer that the observed ultrafast enhancement in visible tr-MOKE is a result of the interplay between the spin disordering and dynamic nature of exchange coupling at the FeNi/FePt interface upon laser excitation. The study yields insights into the ultrafast magnetic manipulation in soft/hard magnetic composites which paves the way for the realization of high-speed, ultra-responsive magneto-optic devices for cutting-edge technology.

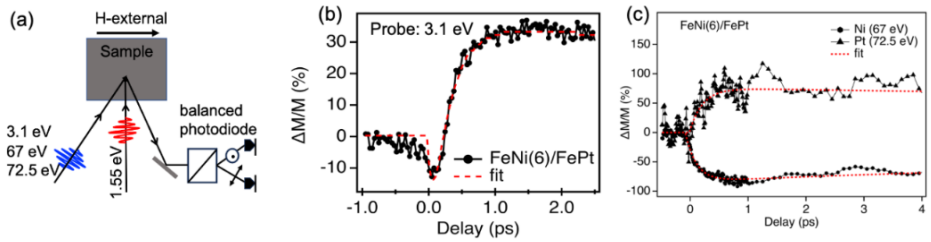

Figure 1: Schematic of longitudinal tr-MOKE: the sample is excited with a 1.55 eV pulse and probed at different energies i.e., 3.1 eV in the visible and 67 eV and 72.5 eV in the extreme ultraviolet region. The magnetization is mea- sured from the reflected probe pulse from the sample, which is collected and detected by a balanced photodetection scheme. (b) Magnetization dynamics in the visible range, FeNi(6)/FePt show ultrafast demagnetization and tran- sient magnetization enhancement on timescales of femtoseconds before recovery. (c) Relative change in magnetization dynamics sensitive to Ni $M_{2,3}$ and Pt $N_{6,7}$ edges.

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Poster Session / 13

Characterization of variable polarization non-linear high order harmonic emission of a seeded free-electron laser and its application to time resolved spin dynamics at the $L_{2,3}$ edge of magnetic transition metal

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Variable polarization is a required feature of light sources employed to investigate with x-rays the chirality of magnetic textures [1] and more general any symmetry in matter. The possibility to select light polarization is, in particular, attractive for those experiments, which aim at exploring the local symmetry of the sample under scrutiny, e.g., the lattice geometry of a crystal, the chirality of a molecule, or the presence of a net atomic magnetic moment. Moreover, several spectroscopic methods rely on the opportunity to choose a well-defined adsorption edge in combination with a peculiar light polarization. Non-linear high order harmonic emission from an undulator can be successfully used to extend the tuning range of a free electron laser (FEL) beyond the fundamental wavelength range supported by the available electron beam energy and undulator parameters. In a recent publication [2], characterization of FERMI source to reach $L_{2,3}$ edge of transition metal has been presented, demonstrating good performances for time resolved spectroscopic studies of spin dynamic, with an estimated flux at Co L-edge (about 780 eV) of about 2.5x107 photons/pulse at the sample plane. However, for planar undulators, on-axis harmonic emission is known to be possible only for odd harmonics, limiting the applications of high order harmonic to linear polarization. Instead, for circularly polarized undulators odd high order harmonic of FEL emission occurs off-axis, preventing to extend the polarization control toward shorter wavelengths. For variable polarization (e.g., APPLE-II type) undulators, it has been demonstrated that a special magnetic field configuration can be found, allowing to produce on-axis harmonics with a substantial degree of elliptical polarization [3]. In this poster, we will report on recent experiments performed at DiProI and Magnedyn end-stations of FERMI free-electron laser aimed to exploit such a scheme of generate partially circular polarized light to perform dichroic experiments on magnetic specimens. The feasibility of time resolved experiments at $L_{2,3}$ edge of magnetic transition metal with variable polarization control pave the way either to extend Fourier Transform Holography to shorter wavelengths, improving the imaging resolution [4], as well to perform spectroscopic studies of spin dynamics and topology involving combination of electronic levels with different excitation lifetime, i.e. the energetically shallow, M-edge, and deeper, L-edge.

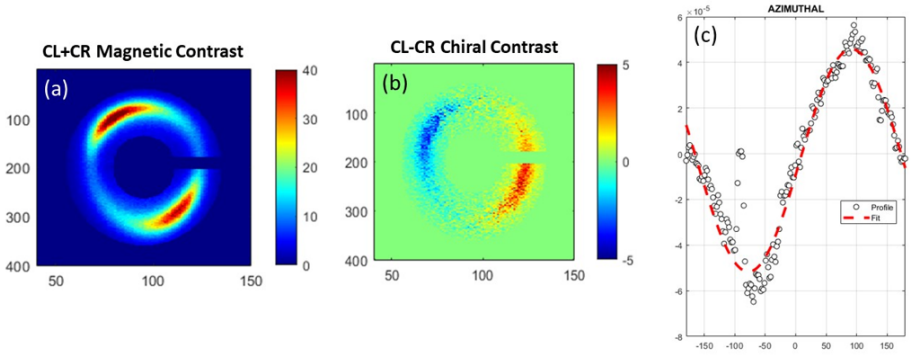

Figure 1: (a) Magnetic resonant scattering recorded of a Synthetic Antiferromagnetic (SAFs) Pt/CoFeB/Ru multilayers system recorded on a CCD detector at the Fe L-edge. (b) Chiral contrast, obtained as the unbalance of magnetic scattered photons with different ellipticity, due to a coherent magnetic spin spiral texture induced in the SAF sample by the energetic balance of different spin interactions such as perpendicular magnetic anisotropy (PMA), the Dzyaloshinskii-Moriya (DM) interaction and the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interlayer coupling. (c) Azimuthal profile of (b) highlighting a Neel type preferential spin ordering of the spin spiral in agreement with synchrotron measurement [5].

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