Multimessenger Approach to out-of-equilibrium DYnamics in Complex Systems (MADYCS)

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Book of Abstracts

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The rich physics of bilayer kagome metals

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Long-range electronic order descending from a metallic parent state constitutes a rich playground to study the intricate interplay of structural and electronic degrees of freedom. Kagome materials appeared as the perfect stage for such explorations. Specifically, RV6Sn6 (R = rare earth atom) bilayer kagome metals are topological systems with Dirac-like itinerant states, van Hove singularities and correlated flat bands [1,2]. Interestingly, within this family, ScV6Sn6 also features a charge density wave (CDW) phase that affects the susceptibility, the neutron scattering, and the specific heat, and whose motif differs significantly from that of the siblings AV3Sb5 (A = K, Rb, Cs) and FeGe. As such, in this presentation, I will give an overview about the rich physics recently unveiled in bilayer kagome metals, and will also discuss about the dynamics of that CDW phase [3]. Specifically, while there is no consensus yet on its microscopic origin, with contradicting affirmations supporting both electron-electron and electron-phonon interaction, we reveal a prominent role played by the structural degrees of freedom in the stabilization of the charge order.

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Spin texture and electron dynamics in chiral crystals.

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Chirality has emerged as a trend topic in condensed matter, as it enforces special symmetries in collective excitations [1, 2] and magnetic ordering [3] and it is responsible for novel quasiparticles beyond Dirac, Weyl and Majorana fermions [4]. The physics of chiral crystals is further enriched by the unique spin arrangements in momentum space [5], by non-linear optical and transport properties that can support large photocurrent and photogalvanic effects [6].

In my talk, I will briefly summarize the results of our research activity on chiral crystals. I will discuss the radial spin texture observed in trigonal tellurium (Te), one of the simplest chiral crystals available [7], and I will explain how the spin texture evolves in the momentum space under the influence of the local point group symmetry [8]. By using time and angle resolved photoemission spectroscopy, we have tracked the dynamical change in the band structure of Te upon excitation of coherent phonons that are capable to alter the crystal symmetry [9]. The same experimental approach has been applied to (TaSe4)2I, a chiral crystal in which charge ordering and strong electron phonon interaction are responsible for opening gap at the Weyl points [10]. Finally, I will give an outlook of the perspectives and potentialities of chiral crystals for opto-electronics.

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Terahertz electric-field-driven dynamical multiferroicity in strontium titanate

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The emergence of collective order in matter is among the most fundamental and intriguing phenomena in physics. In recent years, the ultrafast dynamical control and creation of novel ordered states of matter, not accessible in thermodynamic equilibrium, is receiving much attention. Among those, the theoretical concept of dynamical multiferroicity has been introduced to describe the emergence of magnetization by means of a time-dependent electric polarization in non-ferromagnetic materials [1,2]. In simple terms, a large amplitude coherent rotating motion of the ions in a crystal induces a magnetic moment along the axis of rotation. However, the experimental verification of this effect is still lacking.

With our work [3], we provide the first evidence of room temperature magnetization in the archetypal paraelectric perovskite SrTiO3 due to dynamical multiferroicity. To achieve it, we resonantly drive the infrared-active soft phonon mode with intense circularly polarized terahertz electric field, and detect a large magneto-optical Kerr effect. A simple model, which includes two coupled nonlinear oscillators whose forces and couplings are derived from ab-initio calculations using self-consistent phonon theory at finite temperature [4], qualitatively reproduces our experimental observations in the time and frequency domains. A quantitatively correct magnitude of the effect is obtained when one also considers the phonon analogue of the reciprocal of the Einstein - de Haas effect, also called the Barnett effect, where the total angular momentum is transferred from the coherent phonon motion to the electrons. Our findings show a new path for designing ultrafast magnetic switches by means of coherent control of lattice vibrations with light.

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Ultrafast band structure dynamics in bulk 1T-VSe2

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Complex materials encompassing different phases of matter can display new photoinduced metastable states differing from those attainable under equilibrium conditions.
These states can be realized when energy is injected in the material following a non-equilibrium pathway, unbalancing the unperturbed energy landscape of the material.
Guided by the fact that photoemission experiments allow for detailed insights in the electronic band structure of ordered systems, here we study bulk 1T-VSe2 in its metallic and charge-density-wave phase by time- and angle-resolved photoelectron spectroscopy.
After near-infrared optical excitation, the system shows a net increase of the density of states in the energy range of the valence bands, in the vicinity of the Fermi level, lasting for several picoseconds.
We discuss possible origins as band shifts or correlation effects on the basis of a band structure analysis.
Our results uncover the possibility of altering the electronic band structure of bulk 1T-VSe2 for low excitation fluences, contributing to the understanding of light-induced electronic states.

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Ultrafast quench of the electronic order in the strongly coupled charge-density-wave system VTe2

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The coexistence of strong electron-phonon coupling and non-trivial topological physics in quantum materials can lead to new exciting phenomena, and the study of the interplay of these multiple quantum orders constitute a new paradigm in condensed matter. Here, by combining time and angle-resolved photoemission spectroscopy (TR-ARPES) and broadband time resolved optical spectroscopy (TR-OS), we investigate the effect of an optical excitation on the electronic and structural properties of the strong-coupling charge-density-wave (CDW) system VTe2. Using TR-OS measurements we unveil the presence of two independent amplitude modes (AM) of the CDW phase [1]. Moreover, by performing TR-ARPES experiments, we show that at high excitation fluences the closure of the gap

is not controlled by the excitation of the CDW amplitude modes, but it takes place on a much slower time scale. This time scale could suggest that the gap dynamics is mainly governed by the excitation of high-frequency strongly-coupled optical phonons which result in loss of long range order of the CDW phase [2,3].

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Ultrafast dynamics and coherent excitations of 4f-orbitals derived electronic states in the Kondo semi metal CeSb

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The CeSb magnetic phase diagram contains at least 16 different magnetic phases in the H-T plane [1] comprising different sequences of ferromagnetic and paramagnetic (001) planes stacked along the c-axis. The complexity is thought to arise from the interplay of Kondo, spin-orbit and crystal-field effects. [1, 2] Lattice modulation in the magnetic phases was also observed [3].
The phase diagram [4, 5] and the magnetic excitations [1] were thoroughly studied by neutron scattering. Recently the sensitivity of the electronic structure to the magnetic phase has been demonstrated [6] and additional magnetic excitations were found in the ordered phases [7].
While the main features of the magnetic behavior are understood and successfully modeled using effective interaction approach [8] the microscopic origin of the interactions is still puzzling [2, 7]. Here we present and discuss our investigation of the ultrafast non-equilibrium dynamics in different magnetic phases in CeSb with focus on the magnetic excitations. We confirm the presence of the recently reported [7] additional modes in the ground-state antiferromagnetic phase and show that their frequencies are magnetic-field independent. In the high-magnetic field ferromagnetic phase we identify a previously unobserved $^2E_{2g}$, Ce^{3+} crystal-field excited state. The associated coherent oscillatory optical response can be linked to the real-time quantum evolution of the superposition state involving the ground state and the $^2E_{2g}$ crystal field excited state.

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Coherent and dissipative coupling in magnon-phonon hybridization

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Hybrid quasi-particles in condensed matter systems are observed at the degeneracy points between different degrees of freedom exhibiting non-zero coupling. These solid-state chimeras could be exploited in a wealth of applications, from transducers to sensors, to memory and logic units 1. In particular, magneto-elastic coupling allows for efficient magnon-phonon hybridization in the few gigahertz and inverse micron range [2], resulting in so-called magnon-phonon polarons.
The physics of such hybrids is typically modelled in the framework of coherent coupling (CC), which results in degeneracy lifting and gapping of the dispersion (avoided crossing, a.k.a. Wigner-Von Neumann hybridization gap). Recently, in the context of magnon-photon hybridization, a different coupling mechanism was observed, stemming from correlation in the dissipation of the parent modes into a common incoherent bath: this further channel was dubbed dissipative coupling (DC). DC is raising interest for its relevance in the ever-present competition between coherence and dissipation in coupled systems [3].

We report on magnon-phonon hybridization as observed in a magnonic-phononic crystal: the sample is composed of a regular array of thin (20 nm) ferromagnetic stripes, with sub-micron periodicity [4]. Via ultrashort optical pulses we trigger magnons and acoustic phonons, and we observe their intertwined dynamics via time-resolved Magneto-Optical Kerr Effect [5]. Our results provide evidence of the coexistence of CC and DC in the coupled magnon-phonon polaron dynamics [6]. Moreover, we obtain preliminary results hinting at the possibility to suppress DC by tuning the direction of an external static magnetic field with respect to the sample anisotropies. This observation suggests that suitable fields allow for shaping the dissipation rates of the parent modes, resulting in choking of the dissipation-based DC channel.

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Ultrafast anisotropic electronic properties of ZnAs2 semiconductor

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Comprehension of anisotropic materials is crucial for developing polarization-sensitive photodetectors and polarizers 1. Binary II-V semiconductors are among the most promising candidates to reach this goal, exhibiting anisotropic optical and electronic responses [2]. As a part of this family, ZnAs2 has an energy absorption edge that varies by more than 30 meV when light polarization is varied

from parallel to orthogonal to the crystallographic c-axis. As a result of the optical anisotropy, reflectivity is 1.5 times larger when the electric field is parallel to the c-axis, both in the region of transparency [1.2 mm - 20 mm] and at energies larger than the optical gap (around 1.1 eV in the near IR close to the visible part of the spectrum) [2], [3], [4].
Despite this, what gives rise to these properties in these materials is still an open question. To address this inquiry, we have performed a detailed investigation of the band structure of this material using angle-resolved photoemission spectroscopy (ARPES), which unveils different effective masses along ΓX and ΓY directions in the Brillouin zone. Furthermore, we have carried out time-resolved ARPES, that reveals a unique splitting in the transiently populated conduction band that is not reproduced by ab initio calculations. The goal of our activity is to clarify the origin of this behavior in the conduction band which may pave the way to the use of ZnAs2, and II-V semiconductors, in optoelectronic devices leveraging their large anisotropic properties, and the long-lasting charge populations left by light absorption in the visible spectrum.

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Charge Density Waves in ZrTe3: the fate of nesting in real 3D materials

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Among the materials hosting Charge Density Wave (CDW) phases, transition metal tri-chalcogenides have attracted considerable attention thanks to their quasi-one-dimensional (1D) nature. ZrTe3 is of particular interest because its Fermi surface comprises both a 3D hole like pocket centered at Γ and quasi 1D bands at the zone edges [1,2]. Extensive ARPES studies have shown that the CDW transition, setting in at 63 K [3], is mainly driven by the quasi 1D states, with the opening of a pseudo gap at the D point of the Brillouin zone. For this reason, Fermi Surface Nesting (FSN) between these states was proposed as the driving force of the transition. However, FSN alone does not properly explain the observed changes in the gap and in the 3D band with temperature [4]. We performed time and angle resolved photoemission spectroscopy (trARPES) measurements on ZrTe3 single crystals, probed with 6 eV and 20.9 eV photon energy, revealing a transient photoinduced energy shift of both the 3D and 1D states and subsequent coherent oscillation of the band structure compatible with the excitation of an Ag phonon mode. Hence, our experiment indicates the presence of a strong electron phonon coupling, that could be involved in the CDW formation, in agreement with complementary observations [5] and providing an alternative scenario to the nesting mechanism.

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Time-resolved Raman spectroscopy on bulk and monolayer MoS2

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MoS2 belongs to the class of graphene-like, layered materials called transition metal dichalcogenides (TMDs). Semiconducting TMDs, like MoS2, show an indirect to direct bandgap transition when the system is exfoliated down to the monolayer structure, which makes them promising materials for future applications in optoelectronics and related fields 1. Unravelling the relaxation pathways of photoexcited electrons in such systems is of great applicative interest. Remarkably, the structural and dimensional tunability of electronic properties makes TMDs benchmark systems to study the interplay between electronic and vibrational degrees of freedom.
Time-resolved Raman spectroscopy (TRRS) can provide direct access to the electron-phonon coupling dynamics, as well as the incoherent phonon relaxation [2]. The aim of our experiment is studying the deexcitation dynamics of MoS2 after impulsive optical pumping of the excitonic bandgap. At the SPRINT-NFFA facility, we have carried out a TRRS study on the system, monitoring the temporal evolution of two specific Raman active features, namely the out-of-plane A1g mode and the in-plane E12g mode, after the photoexcitation. Equilibrium Raman spectroscopy studies have linked the modification of such phonon lineshapes to doping effects, providing the background for studying the optically induced, transient doping of the system [3,4,5]. To explore the effect of the indirect-to-direct bandgap transition on the system relaxation dynamics, the experiment was performed on both bulk and monolayer MoS2, revealing significant differences in the phenomenology and relaxation timescales. The role of the substrate on the monolayer deexcitation dynamics was explored by selecting substrates with different electrical and thermal properties.
After presenting the results of the TRRS study, I will discuss their perspective integration in a multimessenger, time-resolved spectroscopy study.

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Triggering and detecting coherent magnons via Transient Grating Spectroscopy

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Magnons, which are quanta of magnetic excitations, have been extensively studied due to their importance in both fundamental research and technological applications. Specifically, magnons with frequencies in the GHz range are of particular interest as they are utilized in modern communication systems. Here, we introduce a novel all-optical method for exciting magnons in the GHz range with precise control over their wavevector. This approach involves utilizing the Transient Grating (TG) technique on a CoGd thin film with perpendicular magnetic anisotropy (PMA). Via interference of two pump pulses, we create a spatially periodic intensity pattern known as a TG, where areas of maximum (or minimum) intensity correspond to constructive (or destructive) interference. When an external magnetic field of appropriate intensity and orientation is applied, spin precession can be induced at the TG maxima 1. However, due to the periodic nature of the excitation, this results in a standing spin wave with the same wavevector as the TG. As the spin wave modulates the optical properties of the material, a probe beam is diffracted; separation of magnetic and non-magnetic contributions to the diffracted intensity is obtained through polarization analysis.
Here, we present experimental results conducted at the NFFA-SPRINT lab on a CoGd thin film with PMA, demonstrating the effectiveness of the methodology and supporting the proposed excitation mechanism. Additionally, we showcase the potential extension of this technique into the extreme ultraviolet (EUV) regime, where larger TG wavevectors can be achieved. Measurements performed at the TIMER-FERMI beamline on FeGd multilayers, also featuring PMA, highlight the capability of this novel approach to measure magnon dispersion, showcasing a potential application in magnon spectroscopy.

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Nanoscale structural dynamics by EUV transient gratings

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Collective dynamics at the nanoscale in condensed matter is important for advancing both fundamental science and modern technology. The study of heat transport processes, vibrational modes or magnetization dynamics in the sub-100 nm length-scales can greatly benefit from the development of experimental tools for probing such dynamics and on the relevant timescale (i.e. ps and sub-ps) without relying on ad hoc sample's nanostructuration.
In this contribution we present a "contact-less"approach, where the sensitivity to the sub-100 nm length-scale is granted by the use of extreme ultraviolet transient gratings (EUV TG) 1. We will discuss the application of this new experimental tool (available at the FERMI free electron laser facility in Triste; Italy) for the study of non-diffusive nanoscale thermal transport in thin membranes of crystalline silicon and amorphous silicon nitride [1,2]. We will also show the possibility to use EUV TG for generating and detecting the dynamics of bulk and surface phonons on a previously inaccessible wavelength range [3], as well as nanoscale magnetization gratings [4] and coherent magnons [5].

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Direct observation of excitonic dephasing by time- and angle-resolved photoelectron spectroscopy

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Coherent light matter interaction plays a pivotal role in future quantum technologies. The dephasing of electronic excitations in condensed matter system proceed on ultrashort time scales due to microscopy many body interactions and can only be indirectly measured by linear spectroscopies, whereas nonlinear optical methods do no provide momentum resolution. In this talk I will discuss ARPES experiments employing a double coherent pulse excitation, in a prototypical bulk transition metal dichalcogenide semiconductor (bulk 2H:Wse2). This material exhibits a very fast excitonic dephasing due to its indirect bandgap (inderectly estimated to be <20 fs on the surface 1). Theoretical analysis demonstrate that the temporal envelope of ARPES interferograms collected with our methos is directly related to the microscopic dephasing time. We experimentally collect interferograms at the K point, the location in reciprocal space of A excitons. Results are fitted by a microscopic theory based on Bloch equations, leading to a dephasing time of 15 fs.
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Experiments with seeded FEL and detection of Photoelectron Angular Distribution (PAD)

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A well-known problem of FELs is the shot-to-shot instability of several parameters of the XUV pulses, which is usually mitigated with single-shot diagnostics and data sorting in post-processing, based on the available metadata. When such data sorting is crucial, the verification on the outcome of the experiment can be sensitively delayed with respect to the data acquisition, so that potential issues in the experiment may not be promptly diagnosed. Considering the high costs and limited availability of beamtime at FEL facilities, such delay in the data evaluation can be considered a relevant issue.

The seeded FEL FERMI provides, on top of the longitudinal coherence, also good repeatability for most pulse parameters, but it is still subject to considerable oscillations in the pulse intensity, which indeed is the primary parameter to be monitored for the purpose of data sorting. Depending on the specific design of each experiment, some observable quantities can exhibit more robust behaviours than others against FEL pulse oscillations or S/N ratio in the detection. PADs in general do not depend on the pulse intensity (except for the special case of contributions from processes with different non-linearity), but they can still be affected by the non-uniform response of the detector, while the required Abel inversion is sensitive to the S/N ratio. Under proper circumstances though, specific

parameters of PADs can be monitored to evaluate almost in real time the outcome of the experiment, and to intervene promptly to correct any faults.

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Ultrafast dynamics of electronic and structural modifications induced by pho-toexcitation in cerium oxide

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The electronic and structural modifications occurring in photocatalysts after photoexcitation largely determine the light-induced functionality of such materials. Cerium oxide is a relevant catalyst with enhanced redox properties, due to its ability to easily and reversibly release oxygen atoms from its lattice.

In a pump-probe optical spectroscopy experiment on cerium oxide films we recently identified aphotoinduced blueshift of the band-gap absorption feature ascribed to photoinduced absorption, that was tentatively ascribed to the coupling between photo-excited charge and the structural distortions into a so-called photoinduced small-polaron state 1.
The availability of ultra-short and ultra-intense X-ray pulses from a free electron laser (FEL), tunable in energy, has allowed us to obtain a detailed picture of the modifications occurring in cerium oxide upon photoexcitation. In particular pump-probe X-ray absorption spectroscopy (XAS) has provided element-specific information on the ultrafast dynamics of the local electronic and atomic structure induced by photoexcitation above the band gap.

We report here the results of pump-probe XAS experiments performed at the FXE instrument of the European XFEL on cerium oxide films. The measurements were performed at 10 degrees incidence, to minimize sample damage and maximize the sensitivity of the method to the thin film investigated. The sample was excited using a laser pump pulse with energy above the band gap and probed by XFEL pulses at the Ce L3 edge. We successfully measured pump-probe Ce L3-edge XAS spectra in the total fluorescence yield acquisition mode by acquiring X-ray absorption near-edge spectroscopy (XANES) and extended X-ray absorption fine structure (EXAFS) spectra with time resolution of approximately 100 fs and up to 10 ps delay time. In the XANES region we observed a dynamic evolution of both the intensity and the shape of the pump-probe signal, reflecting the changes in the 5d states from which the various features originate. The analysis of the pump-probe EXAFS spectra has demonstrated the presence of photoinduced structural distortions with a comparable lifetime as the electronic modifications. The obtained results allow to have previously unavailable details on photoinduced small-polaron states, that possibly determine a long-living transient increase of the redox functionality of the material.

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Ultrashort pulses with (ultra)short wavelengths: novel schemes for few-femtosecond/attosecond pulse generation in the ultraviolet to soft X-Ray region

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In the attosecond molecular dynamics community, there is great interest in the development of light sources capable of producing tunable ultraviolet (UV) pulses with few femtoseconds duration. Indeed, the ability to excite resonantly (usually in the UV range) allows one to study the electronic processes of molecules in their neutral state, i.e., without ionizing it. Big strides in this direction have been made recently thanks to the resonant dispersive wave emission technique1. This technique however yields pulse energies lower than 1 μJ (after removing the fundamental), which could be limiting in experiments, especially after considering transport losses to the target.
In this talk we will present a novel scheme[2], based on cross-phase modulation in a gas-filled hollow-core fiber, that allows the generation of self-compressing, tunable, few-fs pulses in the UV, with energies >10 μJ.
In attosecond experiments, the data quality is often also limited by the poor photon flux of soft XRay attosecond probe pulses. This is attributable to multiple factors including the low single atom efficiency of the high-harmonic generation process, the difficulty in optimizing phase-matching and the limited spectral tunability of attosecond pulses.
We will discuss recent progresses in efficient attosecond pulse generation and in spectral tunability, achieved through the use of a parametric waveform synthesizer[3] based on the coherent synthesis of different pulses obtained via optical parametric amplifiers.

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Scattering and imaging with x-ray beams carrying orbital angular momentum

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The interaction of light beams with magnetic materials defines the rich set of analytical tools in magneto-optics, covering photon energies from infra-red to hard x-rays. In addition to the spin angular momentum (SAM) associated to the light polarization, Laguerre-Gaussian (LG) beams carry also an orbital angular momentum (OAM) of $\ell\hbar$/photon 1 associated to an azimuthal dependence $\exp(i\ell\phi)$ of the electric field phase. Over the last thirty years, OAM beams at vis-IR wavelengths found applications in biology, telecommunication, imaging and quantum technologies [2]. Their capability to exert a mechanical torque was exploited to create optical spanners for manipulating small particles. The azimuthal phase dependence introduces a singularity on the propagation axis and a radial modulation of the intensity (ring-shaped), properties that have been used to modify magnetic ordering, to improve the spatial resolution in microscopy, and to enhance the edge sharpness

in phase-contrast imaging. Over the last decade, the generation of OAM beams at shorter wavelengths, from XUV to hard x-rays, is also finding an increasing number of applications, often based on extrapolations of previous work carried out in the visible range. For instance, as it happened for the SAM, the handedness imposed by the OAM has been exploited to perform x-ray spectroscopic studies of magnetic materials [3] and of chiral molecules [4], and a recent ptychography study [5] showed that the attainable spatial resolution in the reconstructed XUV images increases with ℓ. All these extensions of the use of OAM beams from the visible to the x-rays open new perspectives for element selective spectroscopy and imaging of magnetic structures.

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Halide Perovskite Artificial Solids as a New Platform to Simulate Collective Phenomena in Doped Mott Insulators

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The development of quantum simulators, artificial platforms where the predictions of many-body theories of correlated quantum materials can be tested in a controllable and tunable way, is one of the main challenges of condensed matter physics. Here we introduce artificial lattices made of lead halide perovskite nanocubes as a new platform to simulate and investigate the physics of correlated quantum materials. We demonstrate that optical injection of quantum confined excitons in this system realizes the two main features that ubiquitously pervade the phase diagram of many quantum materials: collective phenomena, in which long-range orders emerge from incoherent fluctuations, and the excitonic Mott transition, which has one-to-one correspondence with the insulator-to-metal transition described by the repulsive Hubbard model in a magnetic field. Our results demonstrate that time-resolved experiments provide a quantum simulator that is able to span a parameter range relevant for a broad class of phenomena, such as superconductivity and charge-density waves.

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Ultrafast Energy Delocalization Between Optically Hybridized J-Aggregates in a Strongly Coupled Microcavity probed by two-dimensional electronic spectroscopy

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Hybrid-polaritons are energy states obtained from the strong coupling between matter excitons and optically confined photons 1. In case of donor-acceptor microcavity system, when both excitons are coupled with the same cavity mode three polariton states are formed. Such states are named: Upper Polariton Branch (UPB) energetically higher with respect to the donor exciton, Lower Polariton Branch (LPB) energetically lower with respect to the acceptor exciton and Middle Polariton Branch (MBP) energetically placed between the excitons. Recently, the use of organic semiconductors as donor and acceptor molecules come into the spotlight due to their large binding energy allowing to observe polariton effects at room temperature [2]. Such effects can impact to the energy transport properties of the system by promoting an energy flow between molecules spatially separated over mesoscopic distances as it is observed in recent photoluminescence experiments [3]. It is theoretically predicted that the more efficient connection between molecules can be only provided when the excitonic and photonic component of the polaritons are balanced. This will produce a polariton wavefunction that is spatially delocalized among the entire system [4]. However, the ultrashort lifetime of the photons in the cavity and uncertainty of the spectral assignment of polariton states have hindered the direct experimental observation of such effect so far.

In this view, two-dimensional electronic spectroscopy (2DES) is a perfect tool to detect ultrafast processes and disentangle spectral features since it provides time resolved excitation/detection energy 2D map with sub-20fs temporal resolution. Specifically, this technique collects the information from the third order nonlinear signal generated from the system after the interaction with three delayed broadband laser pulses. The first and the second pulses act as a pump and the third one as a probe. The delay between the first two pulses is labeled as t1 (coherence time) and the delay between the second pump and probe is labeled t2 (waiting time). The response function of the system is then collected with a spectrometer which provides the detection energy resolution of the experiments. The 2D maps are obtained by scanning both delays (t1, t2) and by Fourier transforming the t1 time traces [5]. Lately, 2DES has been exploited to study polariton dynamics in various systems, even to resolve energy transfer (ET) processes in hybrid architectures where the nature of the polaritons was strongly excitonic [6].

Here, we apply 2DES with ultra-broadband visible pulses to study ET in a microcavity comprised J-aggregates spatially distanced by 2μm, where the photonic and the excitonic contribution of the polaritons are similar. The results show a spectral signature of the coupling between the UPB, MPB and LPB states. Specifically, by exciting the UPB, we observe a quasi-instantaneous signal of all the other polaritons by providing a direct connection between the donor and acceptor molecules. As a proof of concept, we also performed 2DES experiment on the out-of-cavity system where a direct excitation of the donor does not provide an ET to the acceptor. Supported also by 2DES simulations, we rationalize our data by proposing a direct energy delocalization among the polaritons responsible of wiring the energy flow between the two molecules. Our findings can be exploited to improve energy transport properties in light harvesting devices.

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Femtosecond spin dynamics: from demagnetization to spintronics

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The manipulation of the magnetization is essential for our society as it is applied in transformers, radio frequency components and data storage media. Magnetic random access memory (MRAM) devices may combine the advantages of static RAM and flash memory as it is fast and non-volatile. In MRAM a spin current is used to read and write the state of the memory cells. Therefore, it is one of the few real-world applications of spin-based electronics or "spintronics".

This talk focuses on the possibility to directly detect spintronic quantities (the spin voltage, spin current and spin transport) by femtosecond spin-resolved photoelectron spectroscopy. In a laser-excited iron film we can detect the spin-split chemical potentials, which form the "spin voltage", the driving force for a spin current [1,2]. If we deposit a thin gold film onto the iron sample and excite the iron film through the transparent substrate, we can study spin injection and -accumulation. The dynamics of the spin polarization in the gold film [2] can be described by a "spin capacitance", which is similar to the capacitance in charge-based electronics.

An outlook into future opportunities provided by momentum microscopes at free electron lasers will be given. These instruments allow for time, spin and angel-resolved photoelectron spectroscopy at a high repetition rate using an efficient detection scheme.

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Terahertz saturable absorption in black phosphorus

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Black phosphorus is a unique two-dimensional (2D) material with a tunable infrared band gap and anisotropic conduction properties. We investigate the ambient pressure nonlinear terahertz (THz) electrodynamics of black phosphorus and found that its THz saturable-absorption properties can be understood within a thermodynamic model by assuming a fast thermalization of the electron bath. While black phosphorus does not display the presence of massless fermions at ambient pressure and temperature the material's anomalous THz nonlinear properties can be accounted for by a relativistic massive Dirac dispersion, provided that the Fermi temperature is low enough. This suggests that an optimal tuning of the Fermi level could be a strategy to engineer a strong THz nonlinear response

in other massive Dirac materials, such as transition-metal dichalcogenides or high-temperature superconductors

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Ultrafast Coherent THz Lattice Dynamics Coupled to Spins in the van der Waals Antiferromagnet $FePS_3$

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We employed the time-resolved magneto-optical setup described in 1 to study the optically driven lattice and spin dynamics of a 380 nm thick exfoliated flake of the antiferromagnetic van der Waals semiconductor $FePS_3$ as a function of excitation photon energy, sample temperature and external magnetic field [2]. We found evidence of a coherent optical lattice mode with a frequency of 3.2 THz.
The amplitude of the coherent signal vanishes as the phase transition to the paramagnetic phase occurs, revealing its close connection to the long-range magnetic order. The observed phonon mode is known to hybridize with a magnon mode in the presence of an external magnetic field [3], which we utilize to excite the hybridized phonon-magnon mode optically. These findings open a pathway towards the generation of coherent THz photomagnonic dynamics in a van der Waals antiferromagnet, possibly scalable down to thinner flakes.
The talk will discuss the properties of the tabletop setup as well as the investigation of the phonon and phonon-magnon dynamics in $FePS_3$.

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Influence of Metal/Organic-Molecules Interface on Magnetic Anisotropy in Co Thin Films: A Time-Resolved MOKE Investigation

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We investigated by means of the ultrafast time-resolved magneto-optical Kerr effect (MOKE) spectroscopy 1 the effect of the interface between organic molecular semiconductors and cobalt on the magnetic anisotropy of polycrystalline Co thin films. Comparison of the effect was measured on interfaces of Co with: nonmagnetic metal (Al), metalorganic complexes tris(8-hydroxyquinoline)gallium (Gaq_3) and M-phthalocyanines (M=Cu, Co) as well as Buckminster-fullerene (C_{60}) molecules.

In general, the transient MOKE signals were found to exhibit damped coherent spin wave oscillations (CSWO) with frequencies up to several tens of GHz. Detailed analysis of the spin-wave temperature and magnetic field dependences allowed us to compare the influence of different molecular interfaces.

We found that the thin Co films interfaced with molecular layers display strong hardening of the CSWO frequency at low T with a rather sharp transition in the 150 K - 170 K range. Interfaces with different molecules show qualitatively and quantitatively similar behavior despite different molecular shapes. The hardening is attributed to increase of the interface induced anisotropy due to the hybridization between the molecular-orbitals and the Co d-orbital derived interface states [2]. We also found that the CSWO are more damped at low T, which we attribute to an increase of magnetic anisotropy inhomegeneity.

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Ultrafast dynamics of excitons and many-body excitonic states in 2D heterostructures based on transition metal dichalcogenides

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Two dimensional (2D) transition metal dichalcogenides (TMDs) have received increasing attention because of their optical and electronic properties, including enhanced light-matter interaction, strongly bound excitons, exciton Rydberg states, multiparticle excitonic complexes, and valley-selective circular dichroism 1. Some of these properties are exploited in the realization of prototypical optoelectronic devices with improved performances and decreased size. Multiple layers of TMDs can also be stacked to form heterostructures (HS) with tailored electronic and optical properties. Most of TMD-based heterobilayers have type II band alignment leading to fast charge separation and formation of interlayer excitons (ILX) characterized by ultra-long population recombination and twist angle dependence [2].
In my talk, I will report on the non-equilibrium optical response of 2D TMDs and their related HS measured by pump-probe optical spectroscopy techniques. In the first part of the talk, I will focus on the dynamics of Rydberg excitons and trions in high quality TMD monolayers encapsulated with hBN [3]. The second part of the talk will be dedicated to the study of charge transfer dynamics in TMD HS. I will show that it is possible to simultaneously detect interlayer hole and electron transfer processes on a 100 fs timescale. The formation dynamics of ILX bleaching signal shows a distinct picosecond delayed growth dynamics significantly longer than that of intralayer excitons. Theoretical calculations based on microscopic Heisenberg equations of motion find that the delayed formation is mainly related to phonon-assisted interlayer scattering of photo-excited carriers that give rise to finite-momentum (i.e. optically dark) hot ILX which quickly exchange energy and momentum with phonon population and become bright [4].

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Probing and characterization of hot-phonons in nonequilibrium setups

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The appearance of hot-phonons states in pump-probe experiments as a powerful tool for characterizing the time-dynamics of the energy flows, as well in perspective of engineering on/off switchs based on the lattice degrees of freedom and for heat transport. While these features were initially thought to be specific of semiconductors, hot-phonons has been recently predicted and observed as well in metal systems under particular conditions.
In this contribution, taking MgB2 as example, we discuss the different possible ways for revealing and characterizing hot-phonons, spanning from time-resolved optics, spectroscopy and probes of the lattice dynamics.

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Ultrafast core level photoemission spectroscopy and diffraction

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The wealth of information that is encoded in the spectra of core level photoelectrons, like binding energies, line shapes, and diffraction-induced intensity modulations, makes this type of spectroscopy an attractive tool for the study of out-of-equilibrium quantum materials.

Ultrafast structural manipulation can for example manifest hidden or metastable phases. Time-resolved core-level photoelectron spectroscopy (trXPS) and the resulting multiple scattering-induced photoelectron diffraction (trXPD) seem ideal for supporting the characterization of such out-of-equilibrium systems, and for establishing what are the implications for electronic band structure manipulation.

This talk will illustrate two experiments that demonstrate some new possibilities of time-resolved core level spectroscopy.

A detailed explanation of the observed changes in line shape for trXPS results in an effective and straightforward electronic thermometer encoded in the core level shape that can universally be applied to out-of-equilibrium metals.

Then, time-resolved x-ray photoelectron diffraction is applied to the study of ultrafast surface structural manipulation for the topological insulator Bi2Se3. The movement of the topmost surface atoms following a coherent phonon excitation is revealed with sub pm precision, and is explained with a minimalistic ball-and-spring model.

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High-order harmonic generation for the investigation of ultrafast dynamics in semiconductors: attosecond soft-X spectroscopy and HHG in solids

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The interaction of very short and intense laser pulses with noble gases generates high-order harmonics, which creates coherent Extreme UltraViolet (EUV) and Soft-X Ray radiation on a tabletop scale. This allows ultrafast spectroscopy to achieve extreme temporal resolutions, reaching the attosecond level, and site and chemical selectivity. These unique features enable the observation of electronic dynamics in molecules and solids, triggered by ultrafast laser pulses, and the investigation of fundamental aspects of light-matter interaction. A compelling all-optical method to perform these investigations is transient absorption in the XUV. However, the use of ultrafast spectroscopy in this spectral range is still somewhat limited, even now, by the technical difficulty of the needed setups and the poor generation efficiency of the high-order harmonic generation (HHG) sources, especially when going toward higher photon energies. Here I will present the effective XUV generation inside a microfluidic device made by femtosecond laser exposure followed by chemical etching. This microfluidic method allows for controlling and manipulating the harmonic generation conditions in gas at the micrometre scale with remarkable versatility, enabling a high photon-flux and phase matching on broadband harmonics up to 200 eV. This XUV source is coupled with a beamline for transient absorption and reflectivity measurements in molecules and solids recently built at CNR-IFN and equipped with a flexible XUV spectrometer for high-resolution and high dynamic range measurements, with a polarimeter for the characterization of the HHG polarization.
Moreover, HHG spectroscopy is a versatile technique for probing condensed matter, as it can reveal optical band structure and berry phase with light. The polarization state of the harmonic field can help us learn more about the sample's symmetry and the physics of HHG. We used this method to study how semiconductors react to a strong mid-IR linearly polarised driving field at 3.2 μm. Specifically, to demonstrate how HHG can be used as a spectroscopic tool in solids, we used a polarization and time-resolved scheme, to investigate ultrafast processes in condensed media. Because the process is highly nonlinear, this could enable us to study out-of-equilibrium properties of solids with higher sensitivity than typical femtosecond pump-probe techniques.

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Probing condensates' coherence by time-resolved ARPES: From superconductors to excitonic insulators

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With its direct correspondence to the electronic structure, angle-resolved photoemission spectroscopy (ARPES) is a ubiquitous tool for the study of quantum materials. When ex-tended to the temporal domain, time-resolved ARPES offers the potential to move beyond equilibrium properties, exploring both the unoccupied electronic structure as well as its dy-namical response under ultrafast perturbation 1. In this talk, I will discuss how time-resolved ARPES can probe the coherence of many-body condensates, from high-temperature superconductivity [2,3], to spin-correlation-driven pseudogap [4], and ex-citonic insulating behavior [5].

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Femtosecond time-resolved polarimetry at the free-electron laser FERMI

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The analysis of the polarization status of electromagnetic waves is fundamental for a large number of fields of research and technological applications. Intense magnetic fields modify the polarization of the emitted thermal radiation in white dwarf stars. Atomic displacements in solids -phonons- can be detected by Raman scattering with the use of light polarization analysis. Magnetic layers, due to the magneto-optical Kerr effect (MOKE), modify the polarization of transmitted and reflected photons. Extending these techniques both to the x-ray regime and to the femtosecond time-scale might impact our knowledge and technology.
We present here a polarimeter designed for femtosecond MOKE experiments in the EUV range. The polarimeter, combined with the high degree of polarization control and the femtosecond time-resolution of the free-electron laser FERMI, can capture the evolution of the magnetization of single atomic species. We show the design of the device and several experiments performed in the latest years.

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2D metal/semiconductor interfaces: excitonic and charge dynamics

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Transition metal dichalcogenides offer unparalleled opportunities to create new functional heterostructures. Their strongly bound excitons enable precise manipulation of the coupling between light and electronic excitations, while van der Waals coupling allows interfacing of defect-free nanostructures without the constraints of chemical bonds and lattice parameters.
In this presentation, I will showcase the combination of momentum resolved techniques, in particular time-resolved ARPES and femtosecond electron diffraction to explore excitons in TMDs, their dynamics, and details of the electronic wavefunctions involved. I will then show the effects of interfacing WSe_2 with nanostructured Au, resolving a multi-directional energy exchange on timescales shorter than the electronic thermalization of the nanometal. This is followed by non-radiative exciton recombination, electron-phonon coupling, and diffusive charge-transfer that determine the subsequent energy backflow. Stepping fully into the 2D world, I will discuss WSe_2/Graphene interfaces, where we identified a new hot-carrier energy transfer mechanism, the Meitner-Auger interfacial energy transfer. This mechanism arises from dipole-monopole interactions and dominates the energy flow, giving rise to anomalous hot hole dynamics signatures.

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Multi-messenger femtosecond soft X-ray spectroscopy on solids at the European XFEL

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X-ray Free electron lasers (XFELs) produce intense coherent femtosecond pulses in a large energy range, extending the capabilities of laser-based sources. The soft X-ray regime is of paramount importance since it allows resonant studies of many materials with prospective technological applications. Photon-in/photon-out techniques at XFELs have been successfully implemented to study ultrafast dynamical processes in solids. In some cases, the entanglement between the relevant degrees of freedom in the material has been understood in a single experiment. Generalizing these observations to any class of material requires the use of a multi-messenger techniques like photoelectron spectroscopy. The technique has been essential to understand the properties of complex materials, surfaces and interfaces. It provides simultaneous understanding of the electronic, spin, chemical and structural properties. The measured spectra generally contain information on the excitation spectrum and the excitation probabilities thanks to the many-body nature of the photoemission process. Furthermore, the intrinsic time scale of the photoelectric effect enables the study of the fundamental material dynamics. The technique has been successfully implemented down to the pico-second time resolution at synchrotrons. Femtosecond experiments are routinely done using high harmonic generation up to around 100 eV. The extension of the technique to higher photon energies is only possible using superconducting acceleration technology that produces femtosecond pulses at MHz repetition rates. In this contribution, some examples of femtosecond soft X-ray spectroscopy on solids at the European XFEL will be presented. Moreover, the implementation of time resolved experiments at the SXP instrument will be described.