Book of Abstracts

Minicolloquium on:

COHERENT DYNAMICS IN QUANTUM MATERIALS

European Physical Society

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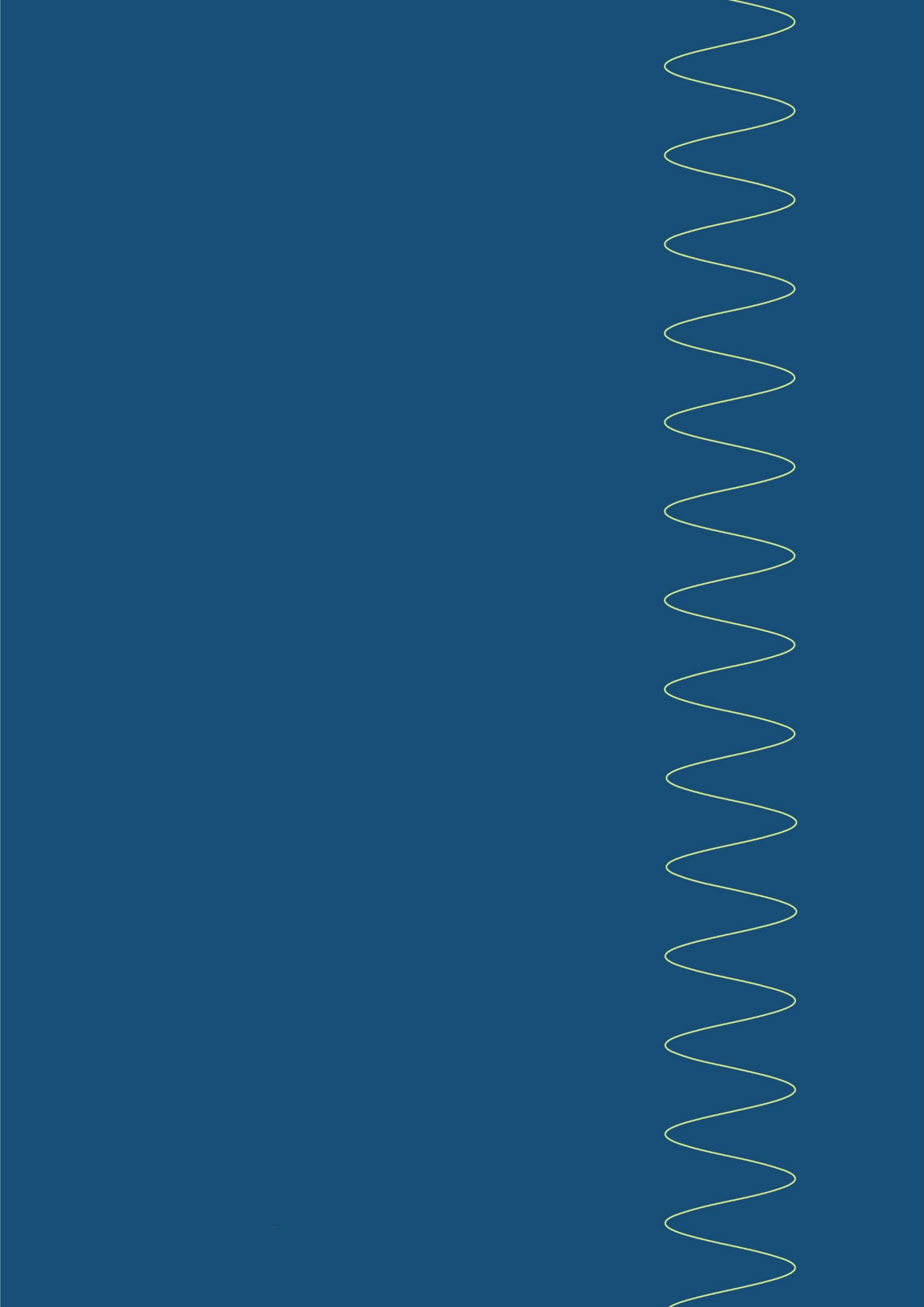

Monday September 4th

CIDiS Building, Room 502

Chair: Gregor Jotzu

Time	Speaker
15:15	Peter Hommelhoff
15:45	Hadas Soifer
16:15	Davide Sangalli
16:45	Anna Galler
17:00	Mattia Udina
17:15	Mariana Gomes
17:30	Angela Montanaro
17:45	Ludwig Mathey

Followed by poster session & general discussion in building 26

Tuesday September 5th

CIDiS Building, Room 502

Chair: Umberto de Giovannini

Time	Speaker
10:45	Gianluca Stefanucci
11:15	Matteo Lucchini
11:45	Netanel Lindner
12:15	Lyudmyla Adamska
12:30	Giacomo Merzoni
12:45	General discussion

The Locations:

ClDis building (Room 502):
https://goo.gl/maps/HqKERWnWpVSWBLxU8

Building 26:
https://goo.gl/maps/zQmJi4oVWjkpLn438

Monday Afternoon Talks

Monday 15:15 - Peter Hommelhoff (Friedrich-Alexander-Universität, Erlangen)

Ultrafast coherent electron dynamics in graphene

We focus intense phase-stable two-cycle laser pulses at graphene and measure the induced currents. Based on a combined experimental-theoretical approach, we find that we can drive electrons in graphene fully coherently. Several years ago, we found that the electrons undergo subsequent coherent Landau-Zener transitions from the valence to the conduction band and back, leading to Landau-Zener-Stückelberg-Majorona (LZSM) interferometry – for the first time observed in a regular room temperature solid [1]. We will report on two current research directions: First, we can now drive the electrons harder, leading to several sign changes of the current as function of peak laser field strength only. We will discuss this observation in terms of LZSM physics, and will show that based on this interferometric method we can sensitively measure material parameters with high accuracy [2]. Second, we will show that we also understand the sub-cycle dynamics at the graphene-gold interface. At this interface, we excite virtual charge carriers, which are out of phase with the real carriers we excite inside of graphene only. Based on these insights, we could demonstrate an ultrafast logic gate potentially allowing petahertz switching bandwidths [3]. References: [1] T. Higuchi, C. Heide, K. Ullmann, H. B. Weber, P. Hommelhoff, Nature 550, 224 (2017); C. Heide, T. Higuchi, H. B. Weber, P. Hommelhoff, Phys. Rev. Lett. 121, 207401 (2018) [2] T. Boolakee, C. Heide et al., manuscript in preparation (2023) [3] T. Boolakee, C. Heide, A. Garzón-Ramírez, H. B. Weber, I. Franco, P. Hommelhoff, Nature 605, 251 (2022)

Monday 15:45 - Hadas Soifer (Tel Aviv University)

Band resolved view on ultrafast photocurrents

In this talk I will discuss how we use time-resolved ARPES to study coupling of light to topological electronic states, and in particular gain better understanding of photocurrent generation in topological insulators. Photocurrents in topological materials have been associated with topological properties of the electronic bands, such as the Berry connection. However, despite some experimental demonstrations, it has remained unclear what part the topology actually plays in the process, and what stems from other contributions. We used time- and angle-resolved photoemission spectroscopy (trARPES) to resolve photocurrents in the excited electronic states of a topological insulator and gained a complete view of the occupied and unoccupied electronic states. Our work provides a microscopic understanding of how to control photocurrents in materials with spin-orbit coupling and broken inversion symmetry, and paves the way to control of currents in topological states.

Monday 16:15 - Davide Sangalli (Istituto di Struttura della Materia (ISM) – CNR

Coherent exciton dynamics from first principles

In this talk I discuss a fully abinitio approach to model the generation of non-equilibrium coherent excitonic states with ultra-short laser pulses. The modelling is achieved via the real-time propagation of the density matrix projected in the Kohn-Sham basis set, within the time–dependent Hartree plus Screened EXchange (TD-HSEX) approaximation [1]. I show how the generated density matrix can be used to model transient spectroscopy signals in the presence of strongly bound excitons. Using LiF as a prototype material, I show that the scheme is able to capture the exciton signature both in time-resolved angle-resolved photoemission spectroscopy and transient absorption experiments [2-4]. The approach is completely general and can become the reference scheme for modeling pump and probe experiment in a wide range of materials. Finally I discuss how the generated coherent excitonic states can be related to non coherent states with a thermal distribution of excitons, and how to compute their lifetimes [5-7]

[1] C. Attaccalite, M. Grüning, and A. Marini, Phys. Rev. B 84, 245110 (2011)
[2] E. Perfetto, D. Sangalli, A. Marini, and G. Stefanucci, Phys. Rev. Materials 3, 124601 (2019)
[3] D. Sangalli, Phys. Rev. Materials 5, 083803 (2021)
[4] D Sangalli, M d'Alessandro, C Attaccalite, arXiv:2211.12241 (2022)
[5] H.-Y. Chen, D. Sangalli, and M. Bernardi, Phys. Rev. Lett. 125, 107401 (2020)
[6] D. Sangalli, E. Perfetto, G. Stefanucci, and A. Marini, The European Physical Journal B 91, 171 (2018)
[7] H.-Y. Chen, D. Sangalli, M. Bernardi, Phys. Rev. Res. 4, 043203 (2022)

Monday 16:45 - Anna Galler (Max Planck Institute for the Structure and Dynamics of Matter, Hamburg)

Mapping light-dressed Floquet bands by highly nonlinear optical excitations

Ultrafast nonlinear optical phenomena in solids have been attracting major interest as novel methodologies for femtosecond spectroscopy of electron dynamics and control of material properties. It is well established that the electronic structure plays a crucial role in these phenomena. However, it is usually assumed that the electronic structure remains unchanged by the laser driving itself, permitting interpretations based on the original field-free bands. Here, we theoretically investigate strong-field nonlinear optical transitions in a prototypical two-dimensional material, hBN, by employing a combination of time-dependent density functional theory and model calculations. We show that the k-resolved conduction band charge occupation patterns induced by an elliptically-polarized laser can be understood in a multi-photon resonant picture; but remarkably, only if using the Floquet light-dressed states. Consequently, our work establishes a direct measurable signature for band-dressing in nonlinear optical processes in solids, and opens new paths for ultrafast spectroscopy and valley manipulation.

Monday 17:00 - Mattia Udina (Sapienza University of Rome)

Terahertz Driven Ionic Kerr effect in SrTiO3

The Kerr effect measures a change in the refractive index proportional to the applied electric field squared, and its ultrafast implementation has been widely used to investigate the nonlinear optical properties of many different systems. More recently, the same mechanism has been exploited with terahertz light pulses to access resonant processes involving lattice vibrations or broken-symmetry collective modes [1-2]. In a recent work [3], we have provided experimental evidence and theoretical description that in insulating SrTiO3 the terahertz Kerr effect admits a sizeable response due to the second-order excitation of an infrared-active phonon. This ionic contribution exploits the ability of strong terahertz pulses to simultaneously excite multiple lattice modes [4] at an intermediate step beside the off-resonant electronic excitations responsible for the conventional electronic Kerr effect. The mechanism is identified thanks to a quantitative theoretical model [5] of the time and polarization dependence of both the electronic and phononic responses. Such a ionic Kerr effect provides a alternative tunable mechanism to modulate the refractive index on ultrashort time-scales and, given the capability of terahertz-driven phonons to couple to order parameters and to drive materials towards metastable states which may not be accessible at thermal equilibrium, it can be used to investigate the electron-phonon coupling across various phase transitions.

[1] *C. L. Johnson, B. E. Knighton, and J. A. Johnson*, Phys. Rev. Lett. 122, 073901 (2019)
[2] K. Katsumi, et al., Phys. Rev. Lett. 120, 117001 (2018)
[3] M. Basini, M. Udina, et al., arVix.2210.14053 (2022)
[4] *D. M. Juraschek and S. F. Maehrlein*, Phys. Rev. B 97, 174302 (2018)
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Monday 17:15 - Mariana Gomes (IFIMUP, Universidade do Porto)

Magnetic-field induced spin transition in NdFeO3

The scientific interest in rare-earth orthoferrites $RFeO_3$ (R=rare-earth; *Pbnm*) is renewing due to their outstanding magnetic properties, including spin-reorientation transitions (SRT), magnetic compensation, and magnetically induced ferroelectricity [1]. $NdFeO_3$ exhibits a SRT, where on cooling, the ferromagnetic moment of the Fe^{3+} spins spontaneously rotates from the c- to the a-direction, gradually changing the magnetic structure from G_xF_z (Γ_4 phase) to G_zF_x (Γ_2 phase) between 170 and 100 K [2,3]. Below 90 K, due to Nd-Fe interactions, the Nd^{3+} spins start aligning antiparallelly to the Fe^{3+} spins, in such a way that the total net magnetization is zero at the compensation temperature of T_{comp} = 7.6 K, becoming even negative at lower temperatures [2,3]. The actual magnetic ordering of the Nd^{3+} spins only occurs at T_N^{Nd} = 1.5 K [3].We will present polarized THz spectroscopy measurements of a $NdFeO_3$ single-crystal cut with an *ab*-plane face, for several fixed temperatures and various applied magnetic fields ($B_{ext}||c$). At zero-field, both at 4 and 8 K, three excitations are observed. According to the magnon selection rules of the Fe^{3+} spins in the Γ_2 phase, two modes are the AFM and FM magnons allowed by symmetry [4]. The additional observed mode can be most likely assigned to a Nd^{3+} crystal field excitation [5]. At 4 K, with increasing magnetic field, the

frequencies of the FM magnon and Nd excitation become closer, almost merging between 6 and 7 T, while the frequencies of the FM and AFM magnons separate. At 8 K, though a similar trend is found, the merging of the FM magnon and Nd excitation is already achieved at 4 T. This result gives clear evidence for a magnetic structure change induced by the applied magnetic field and, consequently, of an unreported magnetic field-induced phase transition for 4 T at 8 K and just above 7 T at 4 K. Furthermore, Raman and infrared (IR) spectra, as well as magnetization measurements as a function of temperature will be shown to ensure the structural and magnetic characterization of the $NdFeO_3$ single crystal sample. We will also present preliminary results of a time-resolved experiment using THz-pump pulses to selectively drive optical phonons to high amplitude, changing the magnetic structure of $NdFeO_3$, which was probed via the Faraday effect, at low temperatures. References:[1] E. Bousquet et al., JPCM 28, 123001 (2016) [2] S. J. Yuan et al., PRB 87, 184405 (2013)[3] W. Sawiski et al., JPCM 17, 4605 (2005) [4] R. M. White et al., PRB 25, 1822 (1982)[5] M. Loewenhaupt et al., JPColloq. 49, C8-921 (1988)

Monday 17:30 - Angela Montanaro (University of Erlangen-Nuremberg)

Clocking superconducting fluctuations in cuprates: a covariance-based approach

In both underdoped and optimally-doped cuprates, superconducting fluctuations survive well above the critical temperature, where either the presence of a competing charge order or the pair phase incoherence yet hinder the formation of a macroscopic superconducting state. Addressing the role of superconducting fluctuations is thus key to elucidate the pairing mechanism itself. However, standard spectroscopies are mostly based on a mean-value approach that, by integrating several stroboscopic repetitions to improve the signal-to-noise ratio, would flatten not only the environmental noise, but also the intrinsic photonic fluctuations (either classical or quantum) that can carry instead genuine information on the material properties. Here, we develop a covariance-based technique to study time-resolved electronic Raman scattering in cuprates. By probing the system with randomized pulses and implementing a single-shot frequency-resolved acquisition, we are able to unveil the spectral correlations imprinted in the pulses by the inelastic scattering from the pump-induced Cooper pair breaking. The momentum selectivity peculiar to Raman scattering, in combination with the sub-picosecond temporal resolution of the technique, allows to measure the correlation dynamics projected onto different regions of the Brillouin zone, thus enabling the isolation of the nodal and antinodal contributions. Our findings reveal gap-size correlations in the pseudogap phase, hinting at the presence of a local pairing that survives even when the superconducting state is macroscopically melted.

Monday 17:45 - Ludwig Mathey (University of Hamburg)

Light-induced dynamics in superconductors and graphene

In this talk, I report our recent studies on light-induced dynamics in superconductors and graphene.
Firstly, I will talk about our study on the Meissner effect in a parametrically driven superconductor using a semiclassical U(1) lattice gauge theory. Specifically, we periodically drive the z-axis tunneling, which leads to an enhancement of the imaginary part of the z-axis conductivity at low frequencies if the driving frequency is blue-detuned from the plasma frequency. In contrast to this enhancement of the conductivity, we find that the screening of magnetic fields is less effective than in equilibrium for blue-detuned driving, while it displays a tendency to be enhanced for red-detuned driving.
Secondly, I will report our observation that the equilibrium state of a bilayer U(1) lattice gauge theory, as a model for bilayer high-temperature superconductors, is a counterflow superfluid state above the critical temperature. This state crosses over into a disordered state at a significantly higher temperature. At this higher temperature, the high-frequency plasmon mode becomes overdamped, while the low-frequency plasmon mode becomes overdamped at the critical temperatures. Experimental signatures and the relation to the pseudogap phase of high-temperature superconductors will be discussed.
Thirdly, I will discuss our proposal to generate Floquet-assisted superradiance in graphene, based on light-induced optical gain, and demonstrate the feasibility of utilizing it for laser operation. We find that the linewidth of the light field in the cavity narrows drastically across the FSP transition, reminiscent of a line narrowing at the laser transition. Next, we demonstrate that the FSP is robust against inhomogeneous broadening, while displaying a reduction of light intensity. We show that the depleted population inversion of near-resonant Floquet states leads to hole burning in the inhomogeneously broadened Floquet spectra. Finally, we show that the FSP is robust against dissipation processes, with coefficients up to values that are experimentally available. We conclude that the FSP presents a robust mechanism that is capable of realistic laser operation.

Monday Evening Posters

Poster #25 Vadim Plastovets (University of Bordeaux)

Coherent dynamics of superconducting energy gap in the presence of a spin-splitting field

In recent years there has been a growing interest in studying the interplay of collective modes in the hybrid superconductor/ferromagnet (S/F) structures. In the present work we address two specific cases of an interaction of uniform time-dependent spin-splitting field with the superconducting order parameter in the presence of spin-orbit coupling (SOC). This can be realized, for example, in a proximized hybrid S/F bilayer where both exchange field and SOC are induced. Using the collisionless approximation we consider the superconducting subsystem in the purely quantum mechanical regime, and describe the coherent temporal evolution of the superconducting condensate within the time-dependent Bogoliubov-de Gennes theory. We show that the

linear response of the superconductor is strongly modified by the Zeeman field and spin-flip processes, which give rise to several resonant frequencies of the so-called superconducting Higgs modes. These modes can be excited directly by the spin-splitting field or by some external laser pulse and can be detected using standard pump-probe technique. In addition to the linear response, the nonadiabatic dynamics of quasi-particle states arising from the intersection of spectral branches from different spin subbands has been considered. Such an intersection can be provoked by a linearly growing Zeeman field whose magnitude exceeds the equilibrium value of the superconducting gap. The dependence of the order parameter on this field and interference effects caused by tunneling of states at the avoided crossing points are studied analytically.

Poster #22 Jonah Messinger (Cavendish Laboratory of Physics, University of Cambridge)

Quantum-Coherent Nuclear Dynamics in the Solid State

In 1954, Dicke developed a formulism for enhanced photon emission by a coherently excited radiating gas.[1] A set of coupled quantum systems, such as the molecular excited states that Dicke describes, cannot be treated as independent, but rather as a single, integrated quantum system. Emission in such a system is accelerated—an effect known as Dicke enhancement—by a factor of up to N^2, where N is the number of interacting molecules. Terhune and Bladwin adapted Dicke enhancement for nuclei in solids.[2] In particular, unlike in a plasma, nuclei in a solid-state lattice, coupled via excited phonon modes and electromagnetic fields, do not radiate independently and are thus subject to collective phenomena that can accelerate their dynamics. The experimental implications of this finding are many. Chumokov et al. demonstrated a 15-fold acceleration of the initial decay of an ensemble of coherently excited ^{57}Fe nuclei.[3] Haber et al. reported quantum beating (Rabi oscillations) between resonantly coupled cavities of ^{57}Fe nuclei.[4] Observations by Metzler suggest delocalized emission from a sample of excited and ground state ^{57}Fe nuclei via the application of mechanical stress, supporting the notion of phonon-mediated nonradiative energy transfer across ^{57}Fe nuclei.[5] Such experimental observations, together with corresponding theoretical work[6,7], has motivated a recent US Dept. of Energy ARPA-E project that seeks to explore the manipulation of nuclear excited states in out-of-equilibrium metal hydrides. Coherence domains on the order of several μm^3 is sought to induce nonradiative nuclear excitation transfer from pairs of hydrogen nuclei to resonant excited states in metal lattice nuclei.[8] This poster serves to present the core thesis of this project and give a status update on the experimental campaign. References: 1.Dicke, R. H. Coherence in Spontaneous Radiation Processes. *Phys. Rev.* **93**, 99–110 (1954).2.Terhune, J. H. & Baldwin, G. C. Nuclear Superradiance in Solids. *Phys. Rev. Lett.* **14**, 589–591 (1965).3.Chumakov, A. I. *et al.* Superradiance of an ensemble of nuclei excited by a free electron laser. *Nature Phys* **14**, 261–264 (2018).4.Haber, J. *et al.* Rabi oscillations of X-ray radiation between two nuclear ensembles. *Nature Photon* **11**, 720–725 (2017).5.Metzler, F. Experiments to investigate phonon-nuclear interactions. (Massachusetts Institute of Technology, 2019).6.Hagelstein, P. L. Recent Progress on Phonon-Nuclear Theoretical Models. (2022).7.Metzler, F., Hunt, C. & Galvanetto, N. Known mechanisms that increase nuclear fusion

rates in the solid state. Preprint at http://arxiv.org/abs/2208.07245 (2022).8.Metzler, F., Hunt, C., Messinger, J. & Galvanetto, N. Probing neutrons and purported fission daughter products from gas-loaded, laser-irradiated metal-hydrogen targets. Preprint at https://papers.ssrn.com/sol3/papers.cfm?abstract_id=4411160 (2023).

Poster #6 Tatiana Bezriadina (University of Hamburg;The Hamburg Centre for Ultrafast Imaging (CUI))

Theoretical description of X-ray absorption by laser-driven electronic system

We theoretically describe the process, in which ultrafast X-ray absorption is applied to measure electron dynamics in a material during the action of a driving optical field. We describe the interaction of an electronic system with the driving electromagnetic field within the Floquet-Bloch formalism and the X-ray absorption probability from a laser-dressed material using the density matrix formalism. Based on this theoretical approach, we created an ab intio computational framework within the LAPW+lo method. We employ Kohn-Sham states obtained by Exciting code as an input for our code. The code calculates laser-induced charge distributions within unit cell and the corresponding time-resolved X-ray absorption spectra. We analyze the connection between laser-induced electron dynamics and changes in X-ray spectra performing calculations for MgO.

Tuesday Morning Talks

Tuesday 10:45 - Gianluca Stefanucci (University of Rome Tor Vergata)

Non-Equilibrium Green's Function methods for real-time simulations of 2D materials

We present a toolbox of Non-Equilibrium Green's Function methods to simulate the correlated dynamics of interacting electrons and phonons in 2D systems. All methods scale linearly in time and they are particle and energy conserving. We discuss the phenomenon of carrier multiplication in photo-excited graphene and the pre-thermalization dynamics of optically excited semiconductors.

Tuesday 11:15 - Matteo Lucchini (Dipartimento di Fisica- Politecnico di Milano)

Validity of the Floquet theory with few-fs pulses

Floquet engineering is a new field of research that investigates the possibility to induce new characteristics and functionalities in advanced materials by using periodic light fields. Being a remarkable example of how optical control of matter enables unforeseen developments, it holds the potential to surpass the current paradigm and foster the opto-electronics and photonics of the

future. For example, the combination of Floquet engineering with short pulses can set the basis for the realization of ultrafast quantum switches of new concept and capable to operate with unprecedented speed. Despite the fascinating and important implications, the applicability of Floquet concepts to short light pulses is still highly debated. Since the Floquet formalism is strictly valid only for perfectly periodic (monochromatic) light, its extension to few-fs light pulses is not trivial. Indeed, the shorter the light pulse, the less periodic it is and the more important the pulse envelope becomes. Therefore, so far, Floquet engineering has been based only on relatively long pulses (hundreds of femtoseconds), thus hindering the investigation of the ultimate speed limit at which the exotic properties of a quantum material can be manipulated with light.Here we tackle this important matter by studying the Floquet state of a prototypical system, the free electron, when illuminated with few-femtosecond pulses of variable time duration. By covering this unexplored regime, our work proves that the Floquet picture holds also for short pulses if properly corrected. We make use of a time-delay compensated monochromator to generate extreme-ultraviolet pulses around 35.1-eV photon energy and use this radiation to ionize a Ne gas target. Few-femtosecond infrared pulses centred around 800 nm and of controlled duration and peak intensity, dress the electron final state creating a Floquet ladder which translates into the appearance of discrete sidebands (SBs) in the photoelectron spectrum. When the pump pulse is longer than 10 cycles traditional Floquet states are observed and the strength of the *n*-th SB scales with the pump intensity following a generalized Bessel function. For durations below 10 optical cycles, the SB amplitude decreases with the pump pulse duration even if the peak intensity is kept constant. By developing an analytical model, we show that the results can be still interpreted in a Floquet-like manner, down to 2 optical cycles. This demonstrates that the Floquet theory can be applied to the short-pulse limit with the opportune corrections, thus extending the Floquet-engineering toolbox.

Tuesday 11:45 - Netanel Lindner (Technion- Israel Institute of Technology)

Dynamical Symmetry Breaking in Optically Driven Two-Dimensional Materials

Floquet engineering of bandstructures via application of coherent time-periodic drives is emerging as a powerful tool for creating new types of topological phases of matter. In this talk, I will show how this tool can also be used to induce non-equilibrium correlated states with dynamical spontaneously broken symmetry. I will present two different manifestations of this phenomenon, which are unique to periodically driven systems, and discuss how they can arise in optically driven two dimensional materials. The first involves spontaneously broken quantum liquid crystalline order, with extreme anisotropy whose directionality rotates as a function of time. The second is realized using amplitude modulated Floquet drives at optical frequencies. I will propose such modulation as a method to parametrically excite low frequency collective modes in an interacting many body system. To illustrate the method, I will discuss the example of terahertz plasmons in a two dimensional electronic system. I will show that using this method, non-dispersive gaps can be opened in the plasmon dispersion. In the presence of a sufficiently strong drive, plasmons in the vicinity of these gaps, which are resonant with the modulation frequency, become unstable and arrange themselves in a crystal-like structure stabilized by interactions and nonlinearities. I will show that the new state breaks the discrete time translational symmetry of the drive as well

as the translational and rotational spatial symmetries of the system and exhibits soft, Goldstone-like phononic excitations. I will show that the phase transition to these correlated steady states is achieved due to the interplay between the coherent external drive, electron-electron interactions, and dissipative processes arising from the coupling to phonons and the electromagnetic environment. Finaly, I will discuss candidate systems for realizing these non-equilbrium phases of matter, and their interplay with the topology of the underlying Floquet bandstructure.

Tuesday 12:15 - Lyudmyla Adamska (University of Modena and Reggio Emilia, and CNR NANO S3)

Analysis of Excitation Channels in Semiconductors under the Influence of Intense Laser Field

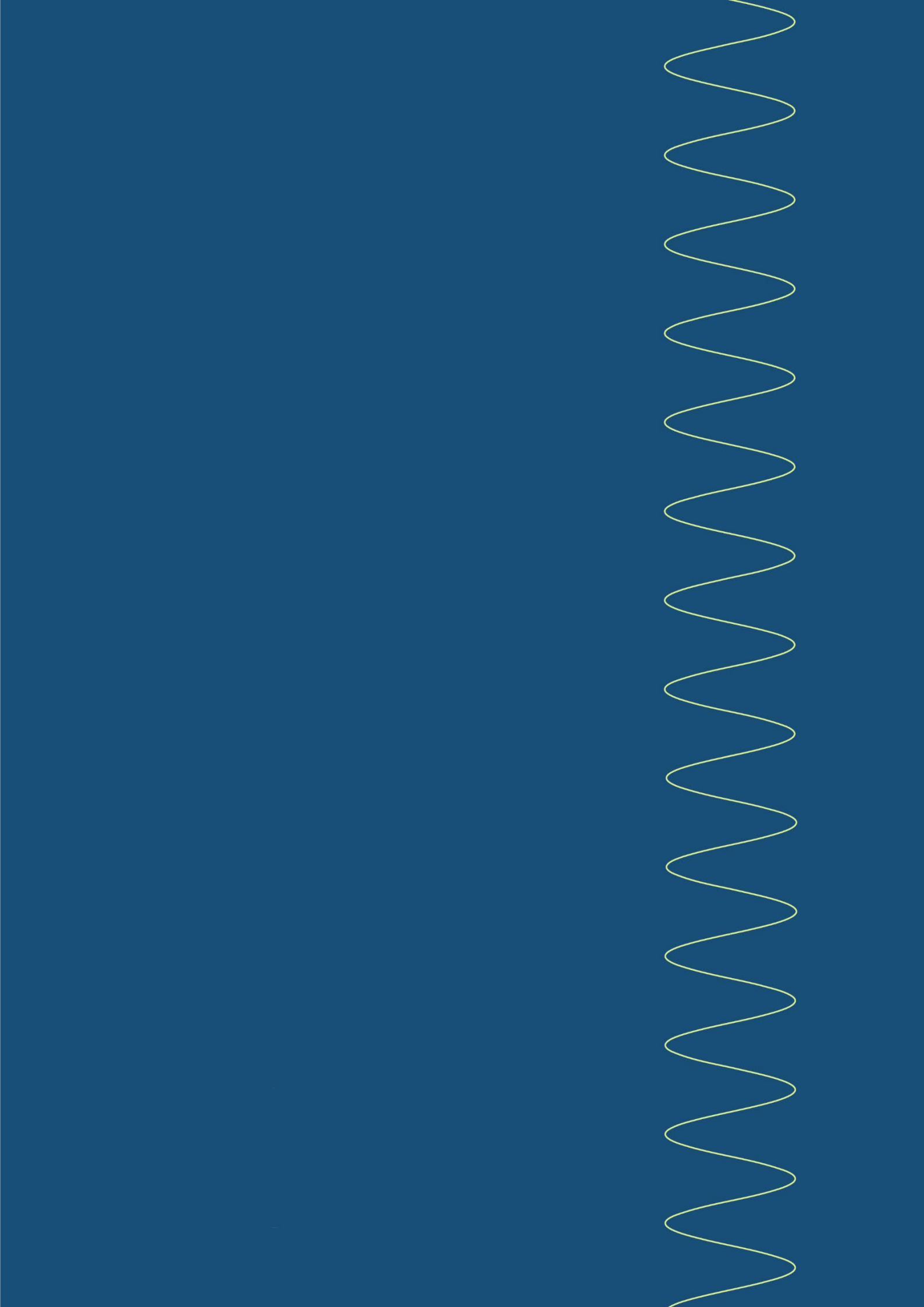
Understanding and manipulation of out-of-equilibrium quantum states is a growing area of scientific and industrial research. Modern experimental techniques are able to probe the dynamics of electrons on attosecond time scales, and time-dependent density functional theory (TDDFT) simulations allow to follow the electronic bands in real time in materials subject to intense laser pulses and assist in interpreting and understanding the experimental data. For intense laser fields, non-linear effects open new intriguing questions and novel opportunities, which are beyond the seminal yet oversimplified analytical Franz-Keldysh model. For this goal, TDDFT may provide us an acceptable trade-off between accuracy and computational cost. In particular, TDDFT has enabled the simulation of transient absorbance and reflectance maps and helped assign their features to the relevant electronic transitions in diamond and silicon. In this work, we apply numerical pump-probe experiments to study the narrow-gap bulk semiconductor germanium, whose semi-core d-levels add extra dynamical features as well as extra computational challenges. We will present our simulations of transient reflectance spectra for moderate and strong laser fields and compare our results to recent experimental findings. Also, we will present the analysis of band occupations in different parts of the Brillouin zone which allows to differentiate between different excitation channels.

Tuesday 12:30 - Giacomo Merzoni (Physics Department, Politecnico di Milano/ SCS instrument, European XFEL)

First high resolution pump probe RIXS on prototypical charge transfer insulators at the EuXFEL

In the last decade, resonant inelastic x-ray scattering (RIXS) has been widely exploited to investigate and understand the physics of various families of quantum materials. Indeed, RIXS spectra exhibit a plethora of excitations that belong to all the intertwined degrees of freedom ruling the physics of these systems. Thanks to the intense efforts in the instrumentation development, best RIXS experiments at synchrotrons have reached 20-30 meV energy resolution allowing the study of excitations in the far-infrared region, like phonons, charge density fluctuations.With the advent of high repetition rate x-ray free electron lasers (XFELs), x-ray spectroscopies became a very promising candidate to perform pump probe (pp) experiments with unique specificity on the probe side. In this context, the development of hRIXS, a high energy and

temporal resolution RIXS instrument at the spectroscopy and coherent scattering (SCS) beamline of the European XFEL, marks a milestone. For the pp-RIXS commissioning we selected two prototypical antiferromagnetic insulators, NiO and La_2CuO_4 (LCO) and pump photon energies larger than the charge transfer gap of the two systems. We measured the L_3-edge RIXS spectra at Ni and Cu, with ≈ 80 meV and ≈ 93 meV resolution respectively. The temporal resolution was ≈ 100 fs. We acquired spectra changing the pump delay between-2 ps and 50 ps, and the laser fluence on the sample between 1 mJ/cm^2 up to 35 mJ/cm^2.The first important result is the consistency of static RIXS spectra taken at XFEL with those measured at the synchrotron, with comparable resolving power. This means that the short and intense FEL pulses are not altering the RIXS process. The major results are those of ppRIXS: in both samples the RIXS spectra are clearly modified by the optical pump in several spectral regions. A long lasting red shift of the dd peaks is detected in both LCO and NiO, accompanied by an increase of the elastic and quasi-elastic (low energy phonons) intensity. The magnon peak is transiently affected by the optical pump too. Moreover, in the case of NiO the optically-induced charge transfer from oxygen to nickel generates a new excitonic state, evident both in the x-ray absorption spectra and in the RIXS, with new spectral features both in the energy loss and energy gain sides. Our results establish pp-RIXS as a superior tool to investigate transient states with unprecedented sensitivity, opening the route to innovative studies on the transient optically excited states in quantum matter.



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