

# **Frontiers in Ultrafast Phenomena in Quantum Materials**

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INFN Frascati National Laboratories

## **Book of Abstracts**



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1

## Attosecond spectroscopy as a window into topological phase structure

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We present a numerical study demonstrating that topological phase transitions can be identified through an attosecond absorption spectroscopy scheme. Focusing on a Chern insulator whose topological character is tuned via second order hopping, we simulate the out of equilibrium electron dynamics driven by a circularly polarized infrared pump and probed by an ultrafast attosecond x ray pulse. Our results reveal a clear laser induced dichroism signal that provides a direct spectral signature of the underlying topological transition. By analyzing these features, we establish a connection between the observed dichroism and the Berry curvature structure of the system. This work demonstrates that attosecond absorption spectroscopy can be extended to detect and characterize non trivial topological phases, opening new avenues for characterizing quantum materials through laser-driven electron dynamics.

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## Pressure-Driven Exciton-to-Trion Conversion in Monolayer WS<sub>2</sub>

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Exciton physics represents a core field in the research on 2D materials from both a fundamental and an application-oriented perspective. In this context, recent years have seen growing interest in understanding the mechanisms governing the conversion of neutral excitons into charged excitons (trions), a process that marks a transition from an optoelectronic response based on bosonic electron-hole pairs to one dominated by fermionic three-body systems. The net charge of trions makes them responsive to the application of an external electric field, in contrast to the insensitivity of neutral excitons, with large implications for the transport properties of the sample. An efficient manipulation of negative trions can thus motivate the design of exciton-based optoelectronic devices with operation principles analogous to those of conventional electronic devices. Furthermore, the fermionic nature of trions fundamentally influences their interaction with spin and valley degrees of freedom. In transition metal dichalcogenides, for instance, the conversion of excitons into trions has been shown to preserve valley polarization, since inter-valley exchange for trions requires spin flips of individual carriers—a process that is energetically unfavourable and spin-forbidden.

To date, efficient exciton-to-trion conversion in 2D semiconductors has been achieved by introducing additional charge carriers via gate voltages or chemical doping [1,2]. Alternatively, highly non-uniform strain gradients have been used to confine excitons and electrons at the nanoscale, enhancing exciton-electron binding and, thus, trion formation [3].

In our work [4], we conduct a deep investigation on the pressure evolution of the photoluminescence spectrum of monolayer WS<sub>2</sub> on different substrates, decoupling exciton and trion contributions based on their different responses to laser power variations. We demonstrate that the dominant mechanism ruling the pressure response of our sample is the competition between the exciton and trion recombination channels, which ultimately drives a full exciton-to-trion conversion above 3 GPa. Compared with approaches based on chemical doping and voltage application, the conversion here observed does not rely on the injection of extra charge carriers in the sample, but it is based on the pressure evolution of the intrinsic doping levels within the electronic band structure. Moreover, in contrast with the conversion processes locally activated in non-uniformly strained flakes, the pressure-induced increase in the trion population occurs at the crystal scale.

The obtained results show that high pressure can serve as a novel, powerful tool for studying exciton-to-trion conversion in two-dimensional semiconductors, driving alternative physical mechanisms in the sample compared to previous experimental approaches. Overall, our work provides deeper insights into the excitonic properties of low-dimensional materials, encouraging further investigations

in this field under high-pressure conditions.

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3

## Tracking photoinduced dynamics in layered semiconductors with ultrafast spectroscopies

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Ultrafast photoexcitation of layered semiconductors leads to the formation of stable excitonic features, making these systems an ideal platform for investigating excitonic many-body interactions and their coupling to lattice and spin degrees of freedom on ultrafast timescales. The goal of this talk is to present experimental protocols based on ultrafast optical and photoemission spectroscopies that enable direct access to excitonic resonances and allow the investigation of their fundamental many-body interactions.

At low temperatures and under quasi-resonant excitation conditions, coherent light–matter interactions can transiently generate a coherent excitonic state, which typically persists for only a few femtoseconds, making its experimental observation particularly challenging. Since quantum coherence is a key ingredient for next-generation technologies, this limitation can be overcome by carefully selecting the layered material and optimizing the experimental parameters in transient optical spectroscopy measurements [1]. Following the coherent-to-incoherent exciton crossover, exciton dynamics evolve on timescales ranging from femtoseconds to several picoseconds, governed by exciton–exciton interactions and coupling to other quasiparticles such as quasi-free carriers and phonons. Time-resolved broadband optical spectroscopy provides direct access to exciton–phonon coupling by identifying spectral fingerprints of the optical response and demonstrating that excitonic resonances can be optically modulated through coupling to coherent atomic vibrations, whose spatial extent can be estimated with sub-picometer resolution [2,3]. In addition, time- and angle-resolved photoemission spectroscopy enables the investigation of exciton formation and subsequent relaxation dynamics in the time, energy, and momentum domains [4].

Extending this framework to layered magnetic semiconductors, polarized and time-resolved broadband optical spectroscopy reveals clear signatures of the coupling between selected optical phonons and the underlying magnetization. These results demonstrate polarization-dependent phonon generation and highlight the interplay among lattice, magnetic, and electronic degrees of freedom.

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[2] S. Mor, V. Gosetti, A. Molina-Sánchez, D. Sangalli, S. Achilli, V. F. Agekyan, P. Franceschini, C. Giannetti, L. Sangaletti, S. Pagliara “Photoinduced modulation of the excitonic resonance via coupling with coherent phonons in a layered semiconductor”, *Phys. Rev. Research* 3, 043175 (2021)

[3] S. Mor, V. Gosetti, V. F. Agekyan, C. Giannetti, L. Sangaletti, S. Pagliara, “Effect of photoinduced screening on the spectroscopic signature of exciton-phonon coupling” *ACS Photonics* 11, 2282-2288 (2024)

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4

## Valley depolarization due to exciton-phonon interactions in monolayer MoS<sub>2</sub>

Author: Yang-hao Chan<sup>None</sup>

Valley degree of freedom in two-dimensional transition metal dichalcogenides was proposed to be a promising qubit candidate for quantum information sciences due to the long coherence time from the coupled spin-valley physics. One of the main decoherence channel was the exchange interactions and the Maialle-Silva-Sham (MSS) type mechanism. Evidences of MSS mechanism were identified in previous experimental and theoretical works. Yet, material specific first-principles investigations demonstrated that exchange interactions may not be required to understand the valley relaxation physics. In this work we revisit the valley depolarization in monolayer MoS<sub>2</sub> by simulating exciton dynamics from first-principles. Intricate competitions between exchange interactions for finite momentum excitons and exciton-phonon couplings lead to rich physics. We found a valley depolarization time of 200 fs at room temperature but do not find clear signatures of MSS mechanism.

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## The real-time ab initio non-equilibrium ultrafast dynamics of excitons in 2D materials

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Pump-probe experiments are essential for probing the nonequilibrium optical properties of 2D materials, providing direct, time-resolved access to exciton formation and thermalization dynamics.

Until very recently, methods for ab initio real-time excitonic dynamics were limited to a coherent-incoherent regime separation, where the Bloch equation/TD-DFT and Boltzmann/RT-BSE formalisms were employed, respectively. This raises a fundamental challenge of treating polarization and carrier degrees of freedom on equal-footing, which has prevented the full-connection thus far.

However, the recently proposed Excitonic Bloch equations not only permit full crossover between regimes, but also explicitly incorporate exciton-phonon coupling while avoiding Hartree overscreening of the ab initio Hamiltonian. Although this theoretical framework exists, a complete, predictive ab initio computational pipeline for real-time exciton dynamics has yet to be established.

A key challenge is the ab initio treatment of exciton-phonon coupling itself—a process critical to energy relaxation, spectral broadening, and phonon-assisted luminescence. Computing the necessary exciton-phonon coupling matrix elements represents a major computational obstacle, which must be overcome before reliable dynamical simulations can proceed.

Equally central is determining the required Q-point grid density to ensure accurate exciton-phonon scattering lifetimes, which directly govern real-time exciton propagation. The convergence of these lifetimes is extremely sensitive and must be handled with care.

This presentation will cover the theory of excitonic pump-probe spectroscopy, detail the challenges in obtaining ab initio exciton-phonon coupling parameters, and discuss the integration of this data into the Excitonic Bloch equations for predictive simulations of exciton dynamics.

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## Light-induced Faraday effect from non-adiabatic breaking of time-reversal symmetry

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Light-induced breaking of time-reversal symmetry in insulating crystals is usually interpreted in terms of the so-called Inverse Faraday Effect (IFE), i.e. the magnetization of media by circularly-polarized light. Early attempts to measure it showed that the strengths of both the direct Faraday effect -i.e. the rotation of the electric-field polarization in the presence of a static applied magnetic field- and the inverse Faraday effects were described by the same parameter - the Verdet constant - that characterized the magneto-optical properties of the medium. In the presence of dipolar lattice modes, a circularly-polarized light pulse can also induce a phonon magnetization, that contributes to the IFE. Following this reasoning, a recent ultrafast THz pump-visible probe measurement of the IFE in insulating SrTiO<sub>3</sub> [1] suggested that the phonon magnetic moment is orders of magnitude larger than what predicted by ab-initio calculations, a result potentially interesting for magneto-optical applications.

Here I will discuss a rather different explanation of the results on Ref. [1], showing that the apparent extra signal attributed to phonons is a consequence of the dynamical nature of the underlying four-wave mixing process involving the THz pump and the visible probe. More specifically, I will show how driving the system under a circularly polarized pump one can access an antisymmetric dynamical component of the rectified non-linear response (Kerr effect) that behaves as a synthetic static magnetic field. Such a component is blind to experiments performed under a linearly-polarized pump [2], that can be indeed quantitatively well understood under the static approximation, which automatically enforces the Kleinmann symmetry. In this view the phonon mode is not necessary, even if it can additionally contribute to the effect. Our results offer a novel perspective on the possibility to tune by circularly-polarized light a large Faraday effect in non-linear media by varying the frequency of the driving field.

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[2] M. Basini, M. Udina, M. Pancaldi, V. Unikandanunni, S. Bonetti, and L. Benfatto, *Phys. Rev. B* 109, 024309 (2024)

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## **EuPRAXIA: Plasma-Based Accelerators for Ultrafast Photon Science**

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Plasma-based accelerators are enabling a new generation of compact ultrafast photon sources for probing matter on femtosecond time scales. EuPRAXIA is a European initiative aimed at developing compact plasma-based accelerators for next-generation light sources and advanced scientific applications. The infrastructure will be implemented at two sites: one at the ELI Beamlines facility, exploiting laser-driven acceleration, and one at the INFN Frascati National Laboratories, based on beam-driven acceleration.

The latter facility, EuPRAXIA@SPARC\_LAB, will host a soft-X-ray SASE FEL beamline, AQUA, delivering few-femtosecond pulses in the 3–10 nm range with high brilliance, together with a VUV beamline, ARIA, providing seeded FEL radiation in the 50–180 nm range. In parallel, the EuAPS betatron source will generate femtosecond X-ray pulses in the 1–10 keV range via laser wakefield acceleration, providing a compact source for ultrafast imaging and X-ray spectroscopy in the hard-X-ray regime.

Together, these sources will form a multi-wavelength platform for investigating ultrafast structural, electronic, and phase-transition dynamics in complex materials.

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## Excitons and Light–Matter Interaction in Emerging Materials for Optoelectronic Applications

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The development of next-generation optoelectronic technologies relies on the identification and detailed understanding of new materials with tailored properties. First-principles simulations of ground and excited states provide a powerful approach to reveal the microscopic mechanisms governing their behavior and to deliver predictive guidance for experimental research.

In this presentation, I will discuss results we obtained in last years from parameter-free atomistic simulations aimed at understanding and controlling the optoelectronic response of novel two-dimensional and layered materials. Using Density Functional Theory (DFT) together with Many-Body Perturbation Theory (GW and the Bethe–Salpeter Equation), I will discuss several key aspects of their electronic and optical properties, including: (i) band-gap renormalization effects, (ii) strong light–matter coupling, (iii) excitonic phenomena and radiative recombination lifetimes, and (iv) the role of doping and chemical substitution as strategies to tune the material response.

Particular attention will be devoted to classes of materials with strong potential for optoelectronic applications, such as transition metal dichalcogenides (TMDs) and layered halide perovskites. In addition, I will present recent results on the excitonic properties of MoSi<sub>2</sub>N<sub>4</sub>-based systems and carbon-based two-dimensional triangulene polymers, highlighting their promise for future technological applications.

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## Excitons in WSe<sub>2</sub> time–resolved ARPES: particle or oscillation?

**Author:** Andrea Marini<sup>1</sup>

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The time–resolved angle–resolved photoemission spectra of WSe<sub>2</sub>, a paradigmatic metal dichalcogenide, is dominated by a peak that, initially observed withing the gap at the *K* valley, scatters, on an ultra–fast time scale of  $\sim 30$  fs to the *S* valley.

In this work we question the commonly used interpretation of the experimental observations in terms of a massive bound electron–hole exciton

scattering with phonon and behaving as a quasi–particle. By using a combined theoretical and experimental investigation we demonstrate that the observed dynamics can be interpreted as the photo–induced transition from a direct to an indirect excitonic insulating phase. The peaks that appear in the experimental spectrum correspond to single–particle levels renormalized by the excitonic spontaneous polarization.

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## Many-Body Simulations of Optical Dichroism and Exciton-Phonon Physics in Quantum Materials

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Understanding the optical response of quantum materials is essential for the development of next-generation photonic and optoelectronic devices. We employ a many-body perturbation theory approach for the study of excited states and optical properties, building on recent methodological advances that have significantly expanded the capabilities for modeling light-matter interactions in complex material systems.

In this work, we present simulations of the optical properties of two representative materials: bulk CrI<sub>3</sub> and bulk MoS<sub>2</sub>. For bulk CrI<sub>3</sub>, which adopts a rhombohedral ( $R\bar{3}$ , BiI<sub>3</sub>-type) crystal structure, the calculations are computationally challenging due to the combined effects of spin-orbit coupling and magnetic ordering.

We investigate the polarization-dependent coupling between light and the electronic structure, revealing distinct responses to left- and right-circularly polarized light associated with the material's magnetic ordering and spin-conserving band-gap transitions within the spin-up channel. For bulk MoS<sub>2</sub>, which crystallizes in hexagonal (P6<sub>3</sub>/mmc) crystal structure, we perform a detailed analysis of exciton-phonon coupling by computing luminescence and Raman spectra, allowing us to investigate how phonon-assisted processes influence the excitonic features that dominate its optical emission.

For CrI<sub>3</sub>, our calculations are performed in close collaboration with experimental groups, using their measurements both to guide the modeling and to benchmark the theoretical predictions. The resulting comparison shows good agreement and highlights the predictive power of the implemented approaches. Together, these case studies demonstrate the versatility of our approach for investigating light-matter interactions across a broad class of quantum materials.

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## Attosecond electron dynamics in doped semiconductors

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Doping is a fundamental element of semiconductor technology, providing a powerful means to tailor electronic properties and underpinning modern microelectronics. As device dimensions continue to shrink toward their fundamental limits, new approaches for ultrafast information processing are required. One promising direction is the use of intense, ultrashort laser pulses to control electronic behavior in solids on sub-cycle timescales, opening the prospect of petahertz-frequency electronics. Although ultrafast light-matter interactions in intrinsic materials have been widely investigated, the influence of doping on electron dynamics occurring on attosecond timescales has not yet been explored.

Here we report the first experimental study of attosecond electron dynamics in doped semiconductors. By employing quasi-isolated attosecond pulses, we measured the ultrafast optical response of germanium in the extreme-ultraviolet (XUV) over a broad range of doping concentrations. For intrinsic samples, comparison with theoretical modeling enabled us to disentangle the roles of intraband motion and virtual interband transitions in the sub-cycle optical response. Extending the same measurements to doped germanium revealed two distinct regimes. At moderate doping levels, where the chemical potential remains inside the band gap, the coherent response closely resembles that of the intrinsic material but is spectrally shifted, enabling controlled tuning of the attosecond response in the XUV spectral range. In contrast, strongly doped degenerate samples exhibit qualitatively different dynamics: interactions between majority carriers and photoexcited carriers modify

the balance between intraband and interband contributions, leading to a substantially altered optical response and interaction with intense ultrashort optical pulses.

These findings demonstrate that the coupling between laser-driven carriers and doping-induced charges provides a means to engineer the field-driven response of semiconductors. Doping therefore emerges as a key parameter for tailoring ultrafast optical properties and designing future optoelectronic technologies operating at petahertz frequencies.

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## Ultrafast Exciton Dynamics in Black Phosphorus tracked by time-resolved ARPES - microscopic insights from quantitative experiment-theory comparison

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**Co-authors:** Aymen Mahmoudi<sup>2</sup>; Claude Monney<sup>2</sup>; Fabian O. von Rohr<sup>3</sup>; Frederic Chassot<sup>2</sup>; Geoffroy Kremer<sup>4</sup>; Joël Morf<sup>2</sup>; Juan Felipe Pulgarin Mosquera<sup>1</sup>; Maxime Rumo<sup>2</sup>; Philipp Werner<sup>2</sup>; Viktor Christiansson<sup>5</sup>

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Accurately modeling the transition from initial Bloch states to final photoelectron states is essential for bridging the gap between intrinsic material properties and experimentally observed intensities in angle-resolved photoemission spectroscopy (ARPES). This is particularly critical for time-resolved studies (trARPES), where the interplay of experimental geometry and many-body dynamics can obscure the underlying physics.

In this work, we present a study of the ultrafast dynamics of excitons in black phosphorus (BP), a single-valley semiconductor with pronounced excitonic anisotropy. By combining trARPES experiments with a quantum-kinetic theoretical framework, we directly access the coherent formation and subsequent redistribution of excitons in momentum space. A central element of our approach is the use of an efficient first-principles framework to compute accurate photoemission matrix elements. This enables a quantitative comparison between our simulations and experimental data, allowing us to disentangle extrinsic final-state effects from intrinsic many-body processes. We reveal that intravalley exciton-phonon scattering plays a decisive role in the rapid dephasing and relaxation of excitons in BP, even in the absence of intervalley scattering channels. Our results establish a robust route for the predictive modeling of nonequilibrium phenomena in materials.

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## Terahertz and Infrared Time-Resolved Spectroscopy in Quantum Materials

**Author:** Stefano Lupi<sup>1</sup>

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The emergence of complex phases in quantum materials—ranging from unconventional superconductivity to topologically protected states is governed by a delicate balance of competing interactions that define their unique low-energy excitations. These excitations are, in most cases, resonant with Terahertz (THz) and Infrared (IR) radiation, making these spectral ranges the ideal playground for investigating quantum dynamics.

In this talk, we will discuss several pivotal experiments performed using both laser-based and Free-Electron Laser (FEL) sources to probe and manipulate different classes of quantum materials. Furthermore, we will present recent advancements in the development of innovative instrumentation and THz/IR sources designed to push the boundaries of spatial and temporal resolution.

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## Shift current in 2D crystals: models and ab-initio calculations

**Authors:** Claudio Attaccalite<sup>1</sup>; Diego Garcia Ovalle<sup>2</sup>; Myrta Gruening<sup>3</sup>; Yuncheng Mao<sup>4</sup>

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In this talk, I will discuss the shift current response in two-dimensional systems. In particular, I will focus on three possibilities: 1) increasing the shift current response by modifying the density of states in twisted materials; 2) using external perturbations, such as magnetic fields, to engineer shift current response; 3) the strong electron-hole interaction in low-dimensional systems. Some of this work has been carried out using simple models, while other parts have been conducted using ab initio methods based on real-time electron dynamics.

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- [2] Magnetic Bulk Photovoltaic Effect in Bernal Bilayer Graphene  
Y. Mao, C. Attaccalite, arXiv preprint arXiv:2511.20498 (2025)
- [3] Moiré amplification of highly tunable shift current response in twisted trilayer graphene, Y. Mao, C. Attaccalite, DG. Ovalle, *PRB* B 111 (19), 195408

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## The EuPRAXIA project: a plasma-based accelerator user facility for the next decade.

**Author:** Massimo Ferrario<sup>None</sup>

The EuPRAXIA@SPARC\_LAB facility is the beam-driven pillar of the EuPRAXIA project, which aims to establish by the end of 2031 the first European Research Infrastructure dedicated to plasma-based accelerators. The facility is expected to demonstrate the usability of such accelerators to deliver high-brightness electron beams in the 1–5 GeV range for a broad user community.

One of the primary scientific objectives of EuPRAXIA@SPARC\_LAB is the development of a short-wavelength Free Electron Laser (FEL) capable of generating ultra-short coherent radiation pulses in the “water window” of the electromagnetic spectrum, enabling in particular advanced biophysical research and material science. Additionally, an X-ray source based on betatron radiation is under development and is expected to be operational by the end of 2026, supported by the Next Generation Eu (PNRR) initiatives.

The generation of high-quality electron beams suitable for FEL operation also represents a crucial milestone towards the long-term vision of a plasma-based Linear Collider (LC). In addition a R&D program on high-repetition-rate plasma accelerator modules is ongoing, in the framework of the PACRI project, to support this objective.

This talk will provide an overview of recent progress within the EuPRAXIA collaboration.

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## Electronic Screening in Two-Dimensional Materials and Metals: Efficient First-Principles Approaches

**Author:** Daniele Varsano<sup>1</sup>

<sup>1</sup> *CNR - Institute of Nanoscience, Italy*

Many-body perturbation theory (MBPT), and in particular the GW approximation, provides a powerful framework to compute quasiparticle (QP) excitations and spectroscopic properties beyond density functional theory. Its application to low-dimensional systems and metals, however, is computationally challenging due to the sharp long-wavelength behavior of the screened Coulomb interaction and the intraband polarizability.

In this talk I present methodological developments aimed at overcoming these limitations. First, I discuss an approach that accelerates the convergence of GW calculations in two-dimensional semiconductors by combining Monte Carlo Brillouin-zone integration with an interpolation scheme for the screened interaction [1,2]. Building on this idea, I introduce an efficient fully ab initio implementation designed for two- and three-dimensional metals, where interpolation and extrapolation techniques allow a correct treatment of intraband screening and dynamical effects. This approach captures the band-gap renormalization in doped two-dimensional semiconductors in excellent agreement with ARPES measurements.[3]

Finally, I show applications to graphene-based systems, where comparisons with high-resolution electron energy-loss spectroscopy highlight the role of quasiparticle and excitonic effects [4], and reveal tunable Dirac plasmon excitations in doped graphene [5].

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[2] A. Guandalini, D. A. Leon, P. D'Amico, C. Cardoso, A. Ferretti, and D. Varsano Efficient GW calculations via the interpolation of the screened interaction in momentum and frequency space: The case of graphene *Phys. Rev. B* 109, 075120 (2024).

[3] G. Sesti, A. Guandalini, A. Ferretti, P. D'Amico, C. Cardoso, M. Rontani, and D. Varsano, A viable route to GW simulations for metals, *arXiv:2508.06930* (2025).

[4] A. Guandalini et al., Excitonic effects in energy-loss spectra of freestanding graphene, *Nano Lett.* 23, 11835–11841 (2023).

[5] A. Tonelli, G. Sesti et al., Dirac plasmon branches in alkali-doped quasi-free-standing graphene, unpublished (2026).

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## Watching polarons form in real time

**Author:** Victor Garcia<sup>1</sup>

**Co-author:** Fabio Caruso

<sup>1</sup> *Christian-Albrechts-Universität zu Kiel*

Polaron formation in pump-probe experiments is a fast, non-equilibrium process arising from the coupled motion of electrons and lattice vibrations, leading to the emergence of a localized quasiparticle. A new first-principles quantum-kinetic approach is introduced to track the real-time dynamics of electrons and the lattice under electron-phonon interactions. We applied this method to the polar insulators MgO and LiF, and determine the characteristic timescales of polaron localization and identify its unique dynamical signatures. The results reveal distinct dynamical signatures of polaron formation and establish clear, practical criteria for its detection in ultrafast pump-probe experiments, providing a direct connection between theory and experiment.

Founded by Marie Skłodowska-Curie Actions (MSCA), TIMES network.

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## Realistic nuclear ensembles for electronic excitation dynamics

**Author:** Hannah Bertschi<sup>1</sup>

**Co-authors:** George Trenins<sup>2</sup>; Mariana Rossi<sup>2</sup>

<sup>1</sup> *Max Planck Institute for the Structure and Dynamics of Matter*

<sup>2</sup> *MPI for the Structure and Dynamics of Matter*

Understanding how large-amplitude anharmonic nuclear motion influences electronic excitations is essential for explaining related phenomena in weakly-bound systems. To model charge transfer and vibronic spectra, we employ real-time time-dependent density functional theory coupled to multitrajectory Ehrenfest dynamics. In this approach, nuclear anharmonicity is incorporated through the sampling of initial conditions. In contrast to more conventional methods, we generate nuclear configurations and momenta using quantum thermostat molecular dynamics [1]. The resulting distributions of nuclear positions and momenta agree well with exact quantum references obtained from path-integral molecular dynamics, even for systems with large-amplitude motion such as the water dimer.

We investigate charge transfer between phenanthrene and a water dimer after excitation with an electric field. We show that the initial nuclear conditions have a strong impact on both the direction and magnitude of charge transfer. Moreover, the nonadiabatic dynamics allows for a back-transfer of electron density to phenanthrene.

[1] M. Ceriotti et al., *Phys. Rev. Lett.* **103**, 030603 (2009).

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## Simulating Nuclear Motion in Light-Driven Systems

**Author:** Mariana Rossi<sup>1</sup>

<sup>1</sup> *MPI for the Structure and Dynamics of Matter*

In this talk, I will discuss recent progress in performing atomistic simulations of solids, liquids and molecules driven by light. I will show examples where light drives nuclear motion and phase transitions by primarily to nuclear degrees of freedom [1], as well as situations where the relaxation of electronic excitations is dramatically changed by nuclear dynamics [2]. These examples are made possible by simulations that join density-functional theory, machine-learning for the nuclear and electronic subspace [3], and advanced molecular dynamics techniques.

[1] E. Stocco, C. Carbogno, M. Rossi, *npj Computational Materials* **11**, 304 (2025); [2] H. Bertschi, G. Trenins, M. Rossi, in preparation (2026); [3] Z. Lou, A. M. Lewis, M. Rossi, arXiv:2602.09938 (2026)

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## Excitonic effects in the photoexcited carrier dynamics of two-dimensional semiconductors.

**Author:** Carlos Betancur<sup>1</sup>

<sup>1</sup> *PhD student, University of Rome Tor Vergata*

This work compares two first-principles frameworks for describing the ultrafast dynamics of photoexcited carriers in semiconductors: the excitonic Bloch equations (XBE), which capture the coupled evolution of coherent, incoherent, and irreducible excitons, and the semiconductor electron-phonon equations (SEPE), derived from the ab initio Kadanoff-Baym formalism and formulated in terms of single-particle occupations and coherences. Using ab initio time-resolved spectroscopy simulations on a monolayer of WSe<sub>2</sub>, it is shown that at low excitation densities, excitonic effects can be effectively described within a two-particle picture, and that, in the absence of bound states, XBE reduces to SEPE and to the Boltzmann equation, providing an accurate description of carrier thermalization toward the band edges.

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## Electronic Dynamics of the Na<sub>5</sub><sup>+</sup> Cluster: A Study Based on the STD LDA Method

**Authors:** Eric SURAUD<sup>1</sup>; Jordan HERAUD<sup>1</sup>; Marc VINCENDON<sup>1</sup>; Paul-Gerhard REINHARD<sup>2</sup>; Phuong Mai DINH<sup>1</sup>; Tingting GU<sup>1</sup>

<sup>1</sup> *Laboratoire de Physique Théorique de Toulouse, Université de Toulouse, Université Paul Sabatier,*

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The interaction of finite electronic systems, such as Na<sub>5</sub><sup>+</sup> clusters, with intense ultrafast laser pulses induces complex electronic dynamics, including ionization and multi-electron excitations. While standard TDDFT captures the laser-driven single-particle dynamics, it is limited in describing electron-electron correlations. To overcome this, we employ the Stochastic Time-Dependent Local Density Approximation (STD LDA), which stochastically samples correlated transitions, including 2p-2h excitations, along TDLDA trajectories using probabilities computed from second-order perturbation theory via the Fermi golden rule. A known issue with STD LDA arises when used in combination with absorbing boundary conditions: particle number is no longer strictly conserved, potentially leading to unphysical effects. To avoid this artifact, the absorbing boundary is switched off during STD LDA propagation. In our procedure, the system is first propagated under the laser field using TDLDA, and STD LDA is subsequently activated at selected switching times to introduce correlated transitions. Furthermore, we systematically investigate the influence of the switching time between TDLDA and STD LDA to evaluate its impact on ionization dynamics and correlation-driven processes.

Na<sub>5</sub><sup>+</sup> clusters are chosen as a model system due to their stable closed-shell configuration, moderate computational cost, and available experimental benchmarks. Their strong surface plasmon resonance enhances energy absorption under resonant laser excitation, promoting multi-electron transitions. Using GPU-accelerated simulations with an extended QDD code, we compute ionization yields and multi-electron transition probabilities, revealing that correlation effects significantly enhance multi-electron excitations and modify ionization dynamics. These results support the use of STD LDA for exploring correlation effects in ultrafast dynamics and point to this methods as a promising strategy for describing high-order electronic excitations in clusters.

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## Real-Time Excitonic Populations and Excitonic Photoemission Spectra Beyond the Dilute Limit

**Author:** Gianluca Stefanucci<sup>1</sup>

<sup>1</sup> *dipartimento di fisica, universita' di roma tor vergata*

We present a first-principles formulation of the excitonic Bloch equations to track, in real time, the populations of coherent, irreducible, and incoherent excitons during and after optical excitation. We then develop a unified framework that links the dynamics of coherent and incoherent excitons to distinct, experimentally accessible excitonic sidebands. Our central result is a practical formula for time-resolved photoemission spectra, applicable over a broad range of temperatures, excitation densities, and pump–probe delays.

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## First-principles description of the dynamical magnetoelectric response in Cr<sub>2</sub>O<sub>3</sub>

**Author:** Torsten Geirsson<sup>1</sup>

**Co-authors:** Alejandro Molina Sanchez<sup>2</sup>; Davide Sangalli<sup>3</sup>

<sup>1</sup> *University of Valencia*

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The magnetoelectric (ME) effect provides a promising pathway for controlling magnetic functionalities using electric fields. While first-principles approaches to the static linear ME response have been rigorously developed and successfully benchmarked, comparable methods for the frequency-dependent linear ME effect remain far less established, despite numerous experimental studies demonstrating pronounced resonance effects associated with the ME response at finite frequencies.

In this work, we investigate the dynamical spin-induced linear ME response from first-principles and provide a systematic comparison of different theoretical frameworks. We implement and assess the independent particle approximation, random-phase approximation, time-dependent density functional theory, and the Bethe–Salpeter equation to assess their performance in describing the frequency-dependent spin ME tensor. These implementations are applied to the prototypical ME material Cr<sub>2</sub>O<sub>3</sub>, and the results are compared to available experimental and theoretical studies.

We show that these approaches capture complementary aspects of the dynamical ME response: low-energy magnonic excitations govern the static and terahertz regime, while electron-hole interactions are essential to reproduce excitonic resonances in the optical regime. Our results establish a direct connection between specific elementary excitations and features in the ME spectrum, providing an interpretation of recent spectroscopic and pump-probe measurements. Additionally, we discuss ongoing efforts to extend this framework to other magnetic materials, such as MnPS<sub>3</sub>.

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## Exciton Transport in Monolayer TMDs

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<sup>1</sup> *University of Valencia*

Atomically thin transition metal dichalcogenides semiconductor emerges as promising candidates for novel optoelectronic application, displaying weak dielectric screening due to its truly two-dimension character. The optical properties are mostly related to inter-band transitions between valence and conduction bands, also called the strongly binding electron-hole pair, exciton. Strain is expected to impact spatiotemporal distribution of excitons, e.g. spatially inhomogeneous strain acts as a driving force for exciton/carrier funneling, similarly to bias fields for charged particles. In this work we demonstrate the capability to manipulate exciton motion via spatially modulated strain fields, where excitonic energy especially its bandgap is largely tunable and the effective mass of electronic valleys is modified resulting in a qualitative change of the excitonic landscape and efficiency of exciton-phonon scattering channels. These transport properties are represented by the coupled two equations, continuity equation and drift-diffusion equation, which derive from zero-order and first-order moment of Boltzmann equation, respectively. In addition, the simulation diffusion coefficient and mobility in latter equation are evaluated by first-principle and experiment qualitatively, as the function of strain fields.

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## Thermal Effects on the Optical Response of Monolayer MoS<sub>2</sub>: A Real-Time Study Integrating Phonon-Induced Dissipation

**Author:** Anna Romani<sup>1</sup>

**Co-authors:** Claudio Attaccalite<sup>2</sup>; Myrta Grüning<sup>1</sup>

<sup>1</sup> *Queen's University Belfast*

<sup>2</sup> *CNRS - CINaM*

The non-equilibrium optical properties of Transition Metal Dichalcogenides (TMDs) are deeply influenced by the interaction between electronic excitations and lattice vibrations. Real-time propagation methods within the Bethe-Salpeter Equation (BSE) or Independent Particle Approximation (IPA) frameworks often rely on a phenomenological constant damping as the dissipation channel to stabilize the dynamics. This approximation fails to capture the temperature-dependent physics essential for describing realistic experimental conditions.

In this work, we implement a temperature-dependent dissipation scheme to study the linear response of monolayer MoS<sub>2</sub>. We account for the optical response by incorporating both thermal expansion effects and temperature-dependent phonon contributions to the self-energy.

Our results highlight how the interplay between lattice expansion and phonon-mediated scattering dictates the broadening and shifting of the MoS<sub>2</sub> excitonic peaks. This approach provides a first-principles foundation for future investigations into non-linear, out-of-equilibrium optical phenomena where energy dissipation via specific phonon modes acts as a critical bottleneck.

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## Ultrafast Formation of a Condensed Carrier Phase in the High-Density Photoexcitation Regime

**Author:** Giuseppe Ammirati<sup>None</sup>

Ultrafast spectroscopy provides a unique window into photoinduced phase transitions, allowing real-time observation of the formation, evolution, and decay of correlated many-body states far from equilibrium. By resolving carrier dynamics on femtosecond timescales, these techniques can disentangle competing electronic phases and reveal the microscopic mechanisms governing collective behavior under extreme photoexcitation.

Photoexcitation in semiconductors gives rise to electron–hole pairs whose character and stability depend critically on carrier density, constituting a well-established physical picture in semiconductor physics.

At low excitation densities, optical pumping generates a dilute gas of free excitons. As carrier density increases, enhanced dielectric screening leads to the formation of an electron–hole plasma (EHP), in agreement with the Mott transition.[1] At higher densities and low temperatures, strong Coulomb interactions stabilize a condensed electron–hole liquid (EHL).[2]

We investigate this phenomenon using thin films of formamidinium lead bromide perovskites (FAPbBr<sub>3</sub>) by means of temperature- and fluence-dependent photoluminescence measurements, which reveal three distinct regimes (free excitons, EHP, and EHL), enabling the determination of critical carrier densities and the construction of a phase diagram.[3]

In light of these results, we further investigated the perovskite film by means of femtosecond transient absorption spectroscopy at 77 K to directly probe the ultrafast dynamics associated with EHL formation. The transient absorption response shows fluence-dependent signatures consistent with many-body effects and phase evolution. At early delays, the spectra exhibit features compatible with a dynamic Burstein–Moss shift and the emergence of low-energy optical gain at high carrier densities. As the system evolves, the transient spectral response converges within tens of picoseconds, indicating a phase transition from the EHL to a regime dominated by free excitons and EHP. Consistently, the photobleaching dynamics show a long-lived decay at low fluence, characteristic of excitonic recombination, while higher fluences introduce an additional ultrafast decay component, reflecting many-body recombination pathways beyond monomolecular processes.

In conclusion, by combining steady-state and time-resolved photoluminescence with femtosecond transient absorption spectroscopy, we provide a comprehensive picture of the formation of the EHL phase in FAPbBr<sub>3</sub> and its dynamics. This approach allows us to directly correlate phase emergence with many-body interactions and their temporal evolution, offering a deeper insight into the mechanisms governing the optical response of metal halide perovskites. These results underscore the relevance of ultrafast spectroscopic methods for disentangling complex phase behavior and highlight the potential of FAPbBr<sub>3</sub> for quantum and optoelectronic applications.

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## **Nonequilibrium physics with ab initio real time propagation schemes. From coherent excitons in the XUV regime to exciton-phonon coupling.**

**Author:** Davide Sangalli<sup>1</sup>

**Co-authors:** Fulvio Paleari<sup>2</sup>; Savio Laricchia<sup>1</sup>

<sup>1</sup> *Istituto di Struttura della Materia (ISM) - CNR*

<sup>2</sup> *CNR - Nanoscience Institute*

I will present recent numerical and theoretical developments, aimed at modelling nonequilibrium dynamics in advanced materials from first-principles, and to capture the generation and dynamics of coherent excitons within the TD-HSEX scheme.

In the first part, I will discuss coherent exciton dynamics in the atto-second regime in LiF, an insulator with the largest known band gap, and hosting strongly bound excitons. The numerical analysis will be focused in the XUV energy range, where a high energy exciton embedded in the continuum appears.

In the second part, I will discuss how exciton-phonon couplings emerge from Ehrenfest dynamics coupled with TD-HSEX simulations, and address the open problem of double counting of screening in the electron-phonon matrix elements that enters into exciton-phonon couplings. Few preliminary numerical results will be also presented in MoS<sub>2</sub>.

Numerical simulations are based on the lumen project ([www.lumen-code.org](http://www.lumen-code.org)), a fork of the yambo code developed within the TIMES doctoral network.

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## Theoretical Modeling of Ultrafast Phase Transitions from the Femtosecond to the Picosecond Scale

**Author:** Matteo Calandra<sup>1</sup>

<sup>1</sup> *Department of Physics, University of Trento*

In this talk, I will introduce a theoretical approach [1,2] to ultrafast phase transitions that captures both electron/hole and phonon dynamics following laser pumping, on time scales ranging from a few femtoseconds to hundreds of picoseconds after the action of the pulse.

At short times, the method relies on solving the manybody Bloch equations coupled to Ehrenfest dynamics in the space of maximally localized Wannier functions. It includes the electric field of the pump explicitly, as well as carrier-carrier, carrier-phonon, and phonon-phonon scattering, treated entirely from first principles [1].

At longer times before recombination, when carrier-carrier interactions generate a photoexcited quasi-equilibrium electron-hole plasma, the approach is based on a constrained density-functional perturbation theory (cDFPT) scheme that accounts for the presence of holes in the valence band and electrons in the conduction band (two-Fermi-level approach) [2]. In this framework, the calculation of forces, phonon dispersion, and carrier-phonon coupling becomes possible, as well as molecular dynamics with machine-learning potentials in the presence of an electron-hole plasma [3].

I will showcase applications of the method to ultrafast ferroelectric switching, light-induced order-disorder phase transitions and other ultrafast phenomena.

This work is funded by the European Union (ERC, DELIGHT, 101052708).

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## Symmetry breaking in TDLDA dynamics of symmetric molecules

**Author:** Phuong Mai Dinh<sup>1</sup>

**Co-authors:** Dale Hugues ; Daniel Dundas ; Eric Suraud ; Paul-Gerhard Reinhard

<sup>1</sup> *Laboratory for Theoretical Physique of Toulouse*

Abstract: In a recent series of papers, we identified an unstable behavior in the electronic response of small molecules to some ultrafast XUV pulses, namely the build-up of a dipole instability at late times. Calculations have been done using real-time Time-Dependent Density Functional Theory in the Local Density Approximation version complemented by a self-interaction correction. In this presentation, I will clarify the mechanism of dipole instability by associating it to a symmetry breaking in small sodium clusters and in the nitrogen dimer [1]. I will focus on the simplified case of direct hole creation at initial time, instead of a full excitation by an ultrafast XUV pulse. In most cases, an oscillation between 2 single particle levels pops up. In an analysis of the energy surfaces associated to such single particle transitions, I will show that these surfaces do indeed display a maximum in the case of direct drilling of a hole state. This characterizes a typical symmetry breaking in mean field models.

[1] P. M. Dinh, P.-G. Reinhard, D. Dundas, D. Hughes, E. Suraud, Eur. Phys. J. D, accepted (2026)

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## Tomography of quantum exciton liquid

**Author:** Luca Perfetti<sup>1</sup>

<sup>1</sup> *Ecole Polytechnique, Palaiseau*

Excitons underpin the operation of optoelectronic devices by governing light–matter interactions in semiconductors. Conventionally, these bound electron–hole quasiparticles are expected to destabilize at high photoexcitation densities owing to screening-induced weakening of the Coulomb interaction. Recent theoretical work, however, has challenged this paradigm by predicting that resonant high-density photoexcitation can generate exceptional points embedded within the electron–hole continuum. Arising at the onset of population inversion, these excitonic states are protected from hybridization with the continuum and persist as long-lived resonances. Here we report time- and angle-resolved photoelectron spectroscopy measurements on two-dimensional hybrid perovskites that provide direct evidence for the existence of such exceptional excitons. Remarkably, these states survive in a regime where the inter-exciton spacing becomes smaller than the spatial extent of the excitonic wavefunction. Our findings establish a new high-density excitonic regime and have important implications for the design and operation of optoelectronic and light-emitting devices under intense excitation.

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## Electronic and Optical Properties of Two-Dimensional Materials: Towards Exciton Dynamics

**Author:** Riccardo Reho<sup>1</sup>

**Co-author:** Ludger Wirtz<sup>1</sup>

<sup>1</sup> *University of Luxembourg*

Understanding and modelling light–matter interaction through spectroscopic techniques is crucial for the development of next-generation technologies, including optoelectronics, quantum devices, and energy conversion [1]. In this context, we investigate the optical properties of two-dimensional materials where reduced screening and strong many-body effects enhance excitonic phenomena. In this talk, I present results on the static electronic and optical properties of van der Waals heterostructures of transition metal dichalcogenides, where stacking and twisting provide a powerful way to tailor excitonic states and optical responses [2]. I will also discuss the case of Bi<sub>2</sub>Se<sub>3</sub>, a prototypical topological insulator, focusing on the role of surface states and their impact on optical

transitions in the two-dimensional limit [3].

From these works, the need for a microscopic modelling of excitonic quantum dynamics naturally emerges. While semiconductor dynamics has long been described within the framework of Maxwell–semiconductor Bloch equations [4], only recently a fully ab initio formalism has been developed through the excitonic Bloch equations (XBE) [5]. In this perspective, I outline future directions beyond the static regime, focusing on the description of ultrafast and non-equilibrium excitonic dynamics. In particular, I will show that the inclusion of exciton-phonon interactions is crucial to capture relaxation and decoherence processes, and I will present preliminary results toward a quantum-dynamical treatment of excitons within the XBE framework.

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## Photoinduced phase transitions in photoexcited semiconductors

**Author:** Giovanni Marini<sup>None</sup>

Ultrafast light pulses provide a unique and powerful handle to control quantum materials far from equilibrium, enabling access to phases and functionalities that are fundamentally inaccessible under thermal conditions. Acting on femtosecond timescales, photoexcitation can selectively perturb electronic, lattice, and spin degrees of freedom, rapidly reshaping the free-energy landscape to access metastable states or, in some cases, to drive the system into long-lived or even permanently altered phases.

In this talk, I present a theoretical perspective on how ultrafast above-gap photoexcitation can be used to induce and control new phases in semiconductors. I discuss representative examples of light-driven transformations, spanning structural, magnetic, and topological phase transitions, and analyze the physical mechanisms that enable their emergence. Particular emphasis is placed on the relationship between photoinduced phase transitions and their thermodynamic equilibrium counterparts, highlighting both fundamental similarities and key differences that arise from non-equilibrium excitation pathways.

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## Electronic and Magnetic Properties of Defected 2D Materials

**Author:** Karina Madela Landivar Zambrana<sup>1</sup>

**Co-author:** Simona Achilli<sup>2</sup>

<sup>1</sup> Phd student-Università degli Studi di Milano

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Over the last decades, two-dimensional (2D) materials have gained significant attention as promising platforms for next-generation quantum technologies [1]. Defect engineering in 2D materials enables control of phenomena relevant for spintronics and quantum information processing [2,3] thanks to their tunable electronic, optical, and mechanical properties. For example, low-energy ion implantation has proven to be an effective route to functionalize graphene [2] and MoS<sub>2</sub> [4].

In this work, we investigate the structural and electronic properties of selected 2D materials, namely graphene, hBN and MoS<sub>2</sub> in the presence of point defects. Our study is based on a Density Functional Theory (DFT) approach, which allows the characterization of the local structure of the defect providing atomic models that can support the interpretation of available Scanning Tunneling Microscopy experiments.

From the point of view of the electronic (and spin) properties, one of the challenges of the theory is the correct description of defect states, in particular in gapped materials. To overcome this limitation of DFT in the estimation of the gap, we employ DFT  $-1/2$  [5,6] technique to the evaluation of selected systems, by comparing them with existing literature. This approach incorporates a self-energy correction directly in the pseudopotential of the atoms constituting the system and allows to accurately describe bandgaps and localized defect states in semiconductors and insulators, while maintaining the computational cost significantly lower than that of many-body methods.

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## Excitonic Enhancement of Shift Current in Janus TMDs: A Real-Time Many-Body Approach

**Authors:** Yuncheng Mao<sup>1</sup>; Ju Zhou<sup>2</sup>; Myrta Gruening<sup>3</sup>; Claudio Attaccalite<sup>4</sup>

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Understanding and controlling nonlinear photocurrents in low-dimensional materials is key to advancing optoelectronics. In this work, we present a first-principles real-time framework to compute the shift-current response (a second-order bulk photovoltaic effect) and apply it to two-dimensional Janus transition-metal dichalcogenides (TMDs). Our approach accurately incorporates excitonic effects and quasiparticle corrections via an effective Hamiltonian derived from many-body perturbation theory.

We show that Janus TMDs exhibit a pronounced enhancement of the shift current at energies associated with C-type excitons. Analysis of the corresponding electron-hole pair distributions reveals that electrons and holes localize on different atoms, leading to a sizeable real-space displacement of charge following optical excitation, thus a sizeable shift-current.

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## Coherent and Incoherent Exciton Dynamics in Two-Dimensional Materials

**Author:** Alejandro Molina Sanchez<sup>1</sup>

<sup>1</sup> *Universidad de Valencia*

Excitons in van der Waals materials exhibit remarkable properties arising from reduced dimensionality, strong Coulomb interactions, and tunable dielectric environments. In this talk, I will discuss recent advances in understanding ultrafast exciton dynamics in these systems, combining state-of-the-art experiments and many-body theoretical simulations. Time-resolved spectroscopies reveal a rich interplay between coherent and incoherent processes, including transient optical responses, exciton population dynamics, and coherent excitonic couplings. Particular emphasis will be placed on the formation and manipulation of coherent excitonic states, as demonstrated in layered materials such as  $\text{BiI}_3$  and monolayer  $\text{WS}_2$ , where sub-10 fs dynamics. Ab initio approaches based on the time-dependent density matrix and Bethe–Salpeter framework provide microscopic insight into these phenomena. These results highlight van der Waals materials as a versatile platform for exploring ultrafast many-body physics and advancing excitonic device concepts.

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## Tunability of exciton landscape in layered materials: effects of electric fields, stacking orders and exciton-phonon coupling

**Author:** Fulvio Paleari<sup>1</sup>

<sup>1</sup> *CNR - Nanoscience Institute*

Low-dimensional layered materials, hosting strongly bound excitons, are highly relevant for optoelectronic applications and constitute an ideal platform for the investigation of nonequilibrium exciton dynamics. In particular, in transition metal dichalcogenide (hetero)bilayers, the landscape of electronic excitations may be engineered either by controlling (i) the spatial localization of electron-hole pairs, leading to the formation of interlayer and intralayer excitons, or (ii) their center-of-mass momentum, as these systems host a complex multivalley exciton dispersion.

In the first part of the talk, I will explore how excitons can be tuned via electric fields, strain and stacking configurations. In TMD heterobilayers featuring a Janus layer (where the resulting intrinsic electric field modifies band alignment), transverse optical phonons can mediate interlayer-intralayer exciton scattering, facilitating efficient interlayer exciton generation after laser excitation of the intralayer state. In bilayer  $\text{WSe}_2$ , certain interlayer excitons exhibit a nonlinear electric-field induced shift that transitions to a linear regime beyond a threshold, with the degree of nonlinearity providing a direct measure of electronic hybridization. While on the one hand our results underscore how excitonic state and valley orderings can be controlled for device applications, on the other hand they also emphasize the fragility of electronic excitations with respect to external conditions.

As most layered materials have an indirect band gap, spectroscopic characterization of the electronic excitations necessarily has to take exciton-phonon scattering into account.

Thus, in the second part of the talk, I will discuss how exciton-phonon first-principles techniques can be used to describe phonon-assisted exciton luminescence. Here I will focus on a new workflow able to exploit lattice symmetries to efficiently rotate both exciton wavefunctions and electron-phonon matrix elements in reciprocal space. In this way, not only calculations on large systems (such as 3D crystals) can be attempted, but at the same time the symmetry analysis offers a way to extract exciton-phonon selection rules. I will discuss applications to the hexagonal and rhombohedral polytypes of boron nitride (BN), where the distinct phonon-mediated emission features revealed by cathodoluminescence measurements can be quantitatively reproduced, and also show examples from challenging 3D indirect systems such as cubic BN and anatase  $\text{TiO}_2$ .

## Ultrafast Exciton Correlations and Valley Dynamics in 2D TMDs

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Two-dimensional (2D) transition metal dichalcogenides (TMDs) have attracted significant attention due to their distinctive optical and electronic properties. These include strong light–matter interactions, tightly bound excitons, excitonic Rydberg states, multiparticle excitonic complexes, many-body effects, and chiral valley-selective optical responses. Such features have enabled the development of prototype optoelectronic devices with enhanced performance and reduced dimensions. Advancing applications in optoelectronics and photonics requires a deep understanding of the physical mechanisms governing the transient optical properties of TMDs.

In this talk, we employ high–temporal resolution optical pump–probe spectroscopy to investigate exciton dynamics immediately following photoexcitation with femtosecond laser pulses. We examine how processes such as exciton population dynamics, exciton energy renormalization, and higher-order Coulomb correlations shape the non-equilibrium optical response of 2D TMDs.

By combining broadband, helicity-resolved pump–probe measurements with theoretical modeling, we disentangle the contributions of two-particle excitons and four-particle biexciton correlations to the coherent optical response. Our results reveal that intervalley four-particle correlations drive the ultrafast formation of bound intervalley biexcitons at energies below the A exciton. At the same time, coupling to intravalley exciton–exciton scattering continua reduces the Hartree–Fock energy renormalization and enhances the broadening of the A excitonic resonance [1].

Extending the probed energy range further allows us to explore the dynamics of valley dichroism for both A and B excitons. We find that the initial decay of the A-exciton circular dichroism signal, along with the emergence of an oppositely signed signal for the B exciton, is primarily driven by rapid intervalley phonon-assisted scattering. On longer timescales, valley depolarization is governed by a complex interplay of phonon-assisted processes, exchange interactions, and Dexter-type energy transfer [2].

Finally, we present a study of the non-equilibrium optical response of TMDs under conditions of strong photoexcitation, with particular emphasis on the photoinduced Mott transition. We demonstrate that, in this regime, excitons dissociate into a metallic electron–hole plasma accompanied by pronounced bandgap renormalization. Notably, this transition occurs in the absence of optical gain. Real-time *ab initio* simulations corroborate these results, showing that the exciton Mott transition is driven by the interplay of nonthermal carrier populations and nonequilibrium dynamical screening [3].

[1] T. Deckert et al. *Phys. Rev. Lett.* 135, 066902 (2025)

[2] O. Dogadov et al. *npj 2D Materials and Applications*, 10, 21 (2026)

[3] O. Dogadov et al. manuscript in preparation

## Detect Topological Phase Transitions using Circular Dichroism Transient Absorption Spectroscopy

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The investigation of topology and topological phase transitions has emerged as a cornerstone in modern condensed matter physics, offering profound implications for the development of fault-tolerant

quantum information technologies. A central challenge in this field lies in identifying robust experimental signatures that can unambiguously characterize the topological nature of electronic states. In this work, we propose Circular Dichroism Transient Absorption Spectroscopy (CD-TAS) as a sensitive tool for detecting topological phase transitions. We first establish a direct analytical correspondence between the Berry curvature, the fundamental geometric quantity encoding a system's topological invariants, and the differential absorption of circularly polarized light. This framework is rigorously validated within the equilibrium Haldane model, where CD-TAS effectively distinguishes between trivial and topological phases. Furthermore, we extend this protocol to non-equilibrium systems by investigating Floquet states in graphene driven by circularly polarized light, demonstrating that the chiroptical response remains a high-fidelity indicator for the effectively induced topological phases under periodic driving. To further assess the experimental feasibility of this detection scheme, we are currently investigating the impact of dissipation effects, specifically accounting for electron-electron and electron-phonon scattering mechanisms to evaluate the potential resilience of the proposed method. These results would provide an optical approach to connect theoretical topological properties with ultrafast spectroscopic measurements.

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**TBA**

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**TBA**

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**Welcome and Opening**

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**Open-System TDDFT: Capturing Decoherence and Relaxation via Markovian Electron-Phonon Dynamics****Author:** Subhojit Pal<sup>1</sup>

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The interplay between coherent electronic motion and dissipative electron-lattice interactions lies at the heart of ultrafast phenomena in quantum materials. Time-resolved spectroscopies have revealed that processes such as carrier relaxation, decoherence etc. can't be understood from a purely unitary evolution of the electronic subsystem. Being a closed system theory, rt-TDDFT can't describe these irreversible processes.

In this talk, I present a first-principles framework that reformulates rt-TDDFT as an open quantum system within Markovian approximation (weak coupling regime) in the density matrix formalism. This unified formalism captures the coherent evolution governed by the time-dependent Kohn-Sham Hamiltonian, as well as the incoherent scattering channels that drive the system toward a stationary thermal state. Our framework opens up the door to the first-principles studies of light-driven quantum materials under the realistic finite-temperature conditions, from Floquet engineering to light-wave electronics.

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## Real-Time TDDFT with Dissipation: Towards First-Principles Ultrafast Dynamics in Solids

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Time-dependent density functional theory (TDDFT) provides a powerful framework to describe electron dynamics in real time, but its standard formulation is inherently unitary and therefore unable to capture dissipative processes that are essential in solids. In this talk, I present recent developments toward a first-principles description of ultrafast dynamics that combines real-time TDDFT with dissipative mechanisms.

The approach is based on a density-matrix formulation of TDDFT, where relaxation and decoherence are introduced through physically motivated scattering terms, enabling a unified treatment of coherent light-matter interaction and energy redistribution. I will discuss how this framework naturally connects to semiclassical Boltzmann approaches while retaining full band-structure resolution and nonperturbative coupling to external fields.

Applications to prototypical materials illustrate the interplay between coherent dynamics and dissipation on ultrafast timescales, highlighting regimes where standard approximations break down. These results open the way to predictive simulations of nonequilibrium phenomena in realistic materials, including pump-probe experiments and ultrafast spectroscopy.

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## Symmetries of excitons and application to resonant Raman spectroscopy

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Excitons play a central role in mediating light-matter interactions in two-dimensional materials and wide-bandgap insulators such as hexagonal boron nitride. While robust *ab initio* methods like the Bethe-Salpeter Equation (BSE) are widely used to compute excitonic energies and wave functions, the symmetry properties of excitons in crystals and their associated selection rules have received comparatively little attention, despite being well established for finite systems in quantum chemistry. In this talk, I will show how excitonic states obtained from BSE transform under crystal symmetry operations. I will also demonstrate how these symmetry properties can be exploited to understand selection rules in Raman scattering and to significantly accelerate state-of-the-art BSE calculations and exciton-phonon computations, which are essential for studying phonon-assisted optical spectroscopies and ultrafast dynamics.