



**27<sup>th</sup> ETSF Workshop on Electronic Excitations**

**Shining Light on Matter  
through Interdisciplinary  
Theoretical Approaches**



Marseille, France

3 –7 June 2024



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## Welcome

The workshop series of the European Theoretical Spectroscopy Facility (ETSF) provides a forum for excited states and spectroscopy in condensed-matter physics, chemistry, nanoscience, materials science, and molecular physics attracting theoreticians, code developers, and experimentalists alike.

The 2024 edition of the ETSF workshop is hosted by Aix Marseille University (AMU), whose development strategy is dedicated to interdisciplinary research. AMU welcomes international experts in physics, quantum chemistry, material science, and code development, with the aim of fostering fruitful exchanges between participants and the local community.

The workshop serves as an open forum to discuss various emerging topics like ultrafast phenomena via non-equilibrium dynamics, time-resolved vibrational and electronic spectroscopy at the attosecond scale, and charge and energy transfer in solids, hetero- and nano-structures, including the role of point and extended defects and thermal effects. The development and application of the most advanced methods in the description of electronic excitations, such as multiscale and embedding methods as well as theoretical and numerical developments, will enrich the discussion.

Workshop topics include:

- Non-equilibrium dynamics
- Charge and energy transfer in solids, in hetero- and nano- structures
- Advanced methods in the description of electronic excitations
- Theoretical and numerical developments

We are glad to welcome you in Marseille, wishing you an inspiring workshop with lots of interesting science and fruitful discussions.

The organizers

## General information

### ETSF workshop team (Organizing and program committee)

- Elena Cannuccia (Aix Marseille Université, Marseille, France)
- Guido Fratesi (Università degli Studi di Milano, Milan, Italy)
- Vitaly Gorelov (LSI, CNRS, Ecole Polytechnique, Palaiseau, France)
- Roberta Poloni (SIMaP, CNRS, Grenoble, France)
- Rajarshi Sinha Roy (Université Claude Bernard Lyon 1, ILM, CNRS, Lyon, France)
- Maryam Azizi, (Université Catholique de Louvain, Louvain-la-Neuve, Belgium)

## Local committee

- Elena Cannuccia (Aix Marseille Université, Marseille, France)
- Nathalie Bonifay (Aix Marseille Université, Marseille, France)
- Marie-Pierre Sergent (Aix Marseille Université, Marseille, France)
- Guillaume Vinconneau (Aix Marseille Université, Marseille, France)

## ETSF steering committee

The ETSF steering committee (SC) consists of nine elected research team leaders (RTLs). The current SC members are:

- Matthieu Verstraete (Université de Liège, Liège, Belgium) - Chair
- Davide Sangalli (CNR, Istituto di Struttura della Materia, Rome, Italy)
- Vitaly Gorelov (CNRS, Ecole Polytechnique, Palaiseau, France)
- Letizia Chiodo (Università Campus Bio-Medico di Roma, Roma, Italy)
- Giorgia Fugallo (Nantes University, CNRS, Nantes, France)
- Andre Schleife (University of Illinois, Urbana Champaign, Urbana, USA)
- Daniele Varsano (CNR - Istituto Nanoscienze, Centro S3, Modena, Italy)
- Hans-Christian Weissker (CNRS Marseille, Marseille, France)
- Zeila Zanolli (University of Utrecht, Utrecht, the Netherlands)

## Support

The organizers acknowledge financial support from the  $\Psi_k$  Network, CNRS, Société Française de Physique - Provence, Ville de Marseille, AMUTECH and Aix Marseille Université and Laboratoire Physique des Interactions Ioniques et Moléculaires.

## Workshop venue

The 27th workshop of the ETSF will be hosted by Aix-Marseille University (AMU) and CNRS, at the auditorium of **Campus Joseph Aiguier, 31 Chemin Joseph Aiguier 13009 Marseille**. In case of workshop-related problems you cannot solve, please feel free to contact Elena Cannuccia by phone +33 6 51 58 85 15 or mail [elena.cannuccia@univ-amu.fr](mailto:elena.cannuccia@univ-amu.fr).

## How to get to the Campus Joseph Aiguier by public transportation

### From Marseille train station Saint Charles in about 25-30 minutes

- The metro line 2 (M2, red line), directed to Sainte Marguerite-Dromel stops at Rond Point du Prado. Once there, cross the Boulevard Michelet and take the bus B1, directed to Campus Luminy, Parc National de Calanques, that stops at Michelet-Bonnaude. Walk 6 minutes.

Alternatively go ahead to Sainte Marguerite-Dromel terminus by metro line 2, walk 4 minutes, than take bus 48

### From Marseille old harbour Vieux Port in about 30-35 minutes.

- The metro line 1 (M1, blue line), directed to La Fourragère stops at Castellane. Once there, take line 2 (M2, red line) directed to Sainte Marguerite-Dromel. Either you stop at Rond Point du Prado and take the bus B1 or go ahead to Sainte Marguerite-Dromel, and take bus 48 directed to Hôpital Clairval, stop at Aiguier CNRS, and walk 1 minute.

Please use the smartphone app "RTM" to check timetables. Physical tickets can be bought at any metro stations or on the bus. Dematerialised tickets can be purchased by credit card at the metro gates and on the buses.

**Pay attention:** During this period due to track renewal works, the metro closes at 21:30 from Monday to Thursday! Two replacement buses, called bus M1 and bus M2, run from 21:30 to all the metro stops.

## How to access to the Campus Joseph Aiguier

Once you have arrived at the Campus you will need to go to the guard's lodge (see map of the Campus). They have the list of the participants and they will direct you to the registration desk, which is located in the building behind the lodge (purple building on the map). On the following days, all you need to do is to show to the guard the conference badge that you received when you registered.

## Internet access

In Campus Joseph Aiguier, an **eduroam** is available. If eduroam does not work with you, we have 30 WIFI accounts to distribute at the time of registration.

## Poster session

The poster session organised on June 4 will take place in the entrance hall of the auditorium.

## Coffee breaks and lunches

Coffee breaks will be organised in the entrance hall of the auditorium. Lunches will be served in the canteen (see map of the Campus) starting from 13h00 - 13h15.

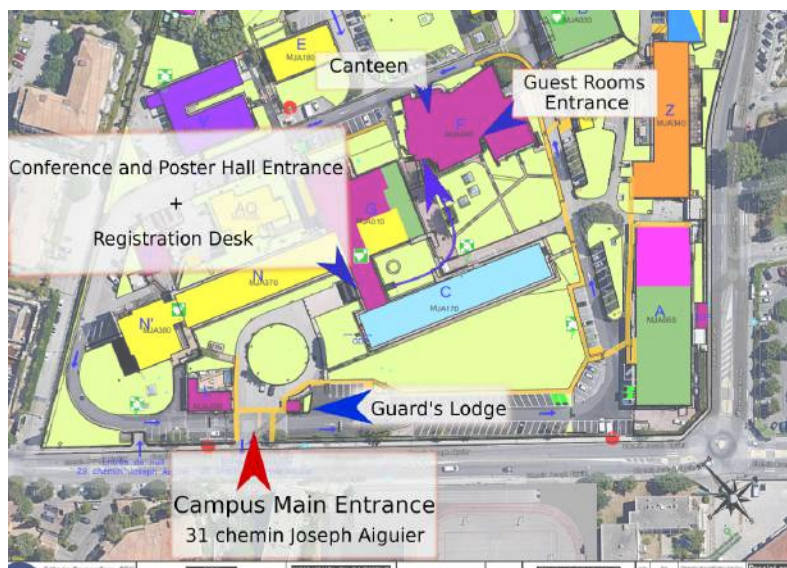
## Social program

We organized a **crui**se to the Calanques which is scheduled for 6 June with departure at 17h30 from Quai de Belge, Vieux Port. The service will be operated by the company Icard Maritime. We recommend that each participant brings a full bottle of water on board! After that we will walk directly to the social dinner location.

## Social dinner

At 19h30 the restaurant "Rowing Club" will host the social dinner. The address is **34 Bd Charles Livon, 13007 Marseille**. We will enjoy a stunning view of the entrance to the Marseille View Port.

## Map of Campus Joseph Aiguier



## Program

	<b>Monday, 3 June</b>
14:00	ETSF steering committee meeting
15:30	<i>Registration &amp; coffee</i>
16:00	Opening
	Introductory lectures
16:15	<b>Claudia Draxl</b> – Theoretical spectroscopy: State of the art and challenges
17:00	<b>Nicolas Ferré</b> – pH effects on the photochemistry of retinal proteins
18:00	Welcome evening

	<b>Tuesday, 4 June</b>
	Spectroscopy and point defects
09:00	<b>Ádám Gali</b> – Theoretical spectroscopy of defect qubits
09:40	Martino Silveti – Ab initio approach to point defect in wurtzite boron nitride
10:00	Alexander Kirchoff – Excited-state geometry relaxation of point defects in mono-layer hexagonal boron nitride
10:20	<i>Coffee break</i>
	Multiscale, embedding and low dimensional materials
10:50	<b>Xavier Blase</b> – Embedding many-body perturbation theory in large scale environments
11:30	Gabriele Iuzzolino – Solvation dynamics influences the photophysics of a nucleic acid-protein interaction model
11:50	Joachim Galiana – Quantum dynamics around PPEs’ conical intersections for both spectroscopic and real-time studies
	Low dimensional materials and heterostructures
12:10	Claudia Cardoso – Efficient many-body perturbation theory calculations for 2D materials
13:00	<i>Lunch</i>
15:00	<b>Alberto Zobelli</b> – Atomic scale mapping of electric field and charge density by 4D-STEM: a study on perfect and defective h-BN
15:40	Fulvio Paleari – Distinguishing different stackings in layered materials via luminescence spectroscopy
16:00	<i>Coffee break</i>
16:30	Riccardo Reho – Excitonic responses in MoS <sub>2</sub> /WS <sub>2</sub> and MoSe <sub>2</sub> /WSe <sub>2</sub> heterostructures from first-principles: impact of stacking order and interlayer distance
16:50	Elisa Serrano Richaud – Modelling electronic and optical properties of graphene and boron nitride lateral heterostructures
17:10	<b>Poster session</b>



<b>Wednesday, 5 June</b>	
	Ultrafast spectroscopy
09:00	<b>Alicia Palacios</b> – Theoretical methods for the description of correlated electron and nuclear dynamics at the attosecond time scale
09:40	Tatiana Bezriadina – Theoretical description of X-ray absorption of laser-driven materials
10:00	Andreas Windischbacher – Imaging Excitons in C60 Thin Films with Photoemission Orbital Tomography
10:20	<i>Coffee break</i>
	Exciton dynamics
10:50	<b>Ivan Maliyov</b> – Ultrafast spectroscopy of semiconductors: probing the nonequilibrium electron-phonon dynamics
11:30	Claudio Attaccalite – Dynamics of coherent excitons in resonantly driven semiconductors
11:50	Fabienne Michelini – Discussing protection in FMO complex from NEGF-based exciton transport modelling and simulation
12:10	ETSF general meeting
13:00	<i>Lunch</i>
	Non equilibrium dynamics
15:00	<b>Lou Barreau</b> – Ultrafast electron dynamics observed with attosecond pulses
15:40	Brice Arnaud – Non-linear phononics in THz excited Bi <sub>2</sub> Te <sub>3</sub> nanofilms
16:00	<i>Coffee break</i>
16:30	Kevin Lévêque-Simon – TDDFT simulations applied to pump-probe experiments with sapphire
16:50	Lu Qiao – X-ray transient absorption spectrum at Zn K-edge: Pauli blocking and core-hole screening modification
17:10	<b>Isabelle Régner</b> - General Public Seminar on "Influence of gender stereotypes on cognitive performance and hiring decisions"

<b>Thursday, 6 June</b>	
	Theoretical and numerical developments
09:00	<b>Julien Toulouse</b> – Relativistic electronic-structure methods based on effective quantum electrodynamics
09:40	Frank Ortmann – Linear scaling approach for optical excitations using maximally localized Wannier functions
10:00	Jesus Villalobos – MZ-vector approach to BSE analytic gradients
10:20	<i>Coffee break</i>
	Advanced methods for electronic excitations
10:50	<b>Pierluigi Cudazzo</b> – Dynamical effects in the spectra of neutral excitations
11:30	Pina Romaniello – The multi-channel Dyson equation : coupling many-body Green's functions
11:50	Davide Sangalli – Symmetries and sum rules in the Bethe-Salpeter equations: theory and implications
12:10	Laura Urquiza – Excitonic Effects in $\alpha$ -Al <sub>2</sub> O <sub>3</sub> revealed through resonant inelastic x-ray scattering and complementary x-ray spectroscopies
12:30	Jan-Hauke Graalmann – Pressure Dependence of Intra- and Interlayer Excitons in 2H-MoS <sub>2</sub> Bilayers
13:00	<i>Lunch</i>
	Non linear optics
14:30	<b>Michele Celebrano</b> – Free-space interferometric routing of upconverted light by dielectric metasurfaces
15:10	Simone Grillo – Tunable Second Harmonic Generation in 2D Materials from First-Principles
15:50	Social program
19:30	Workshop dinner

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<b>Friday, 7 June</b>	
	Theoretical developments for total energy and spectroscopy I
09:00	<b>Maria Hellgren</b> – Correlation effects in solids with RPA and beyond
09:40	Abdallah El-Sahili – From spectra to total energy: beyond the GW approximation designing effective interactions
10:00	Iskander Mukatayev – XPS core-level electron removal energies and chemical shifts by ab initio theory
10:20	<i>Coffee break</i>
	Theoretical developments for total energy and spectroscopy II
10:50	<b>Bo Peng</b> – Photoinduced electronic and spin topological phases in monolayer single-element ferroelectrics
11:30	Nasrin Farahani – Revealing fingerprints of valence excitons in x-ray absorption spectra with the Bethe-Salpeter equation
11:50	Martin Unzog – Modeling and investigating K-edge spectra of single-wall carbon nanotubes
12:10	Closing remarks

## Oral contributions

## Theoretical spectroscopy: State of the art and challenges

**Claudia Draxl**

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Many-body approaches to electronic excitations have become an indispensable tool for an in-depth understanding of the subtle processes that take place in a wide variety of materials. In particular, many-body perturbation theory (MBPT) allows us to treat the interplay between competing interactions of similar strength and on the same energy scale, which can give rise to exciting phenomena. Elucidating the underlying mechanisms and keeping up with the recent developments in experimental techniques, requires the development of advanced methodologies. In this talk, I will first give an overview of the state-of-the-art methods in describing the response of matter to light on different length scales. Selected examples of optical absorption, core-level spectroscopy, and resonant inelastic x-rays scattering (RIXS) will highlight the interplay of electron-electron interaction, electron-vibrational coupling, electron-hole correlation, and exciton-exciton coupling. I will then report on our recent progress in developing methods to tackle exciton-phonon coupling and exciton dynamics, and discuss ways to keep such calculations within affordable computational times, even for complex materials.

### References

[1] G. Onida, L. Reining, and A. Rubio, "Electronic excitations: density-functional versus many-body Green's function approaches, *Rev. Mod. Phys.* 74, 601 (2002).

## **pH effects on the photochemistry of retinal proteins**

**N. Ferré**

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Most microbial rhodopsin proteins, ie photoactive biomolecules featuring the all-trans retinal chromophore, exhibit pH-dependent absorption spectra and/or retinal photoisomerization quantum yield. In this talk, I will present two hybrid quantum mechanics/molecular mechanics (QM/MM)-based approaches, aiming at investigating how pH changes can be effectively considered thanks to a careful selection of the most important protonation microstates. Specifically, I will first show how constant-pH molecular dynamics simulations provide effective initial conditions for studying the photochemistry of Anabaena Sensory Rhodopsin using non-adiabatic excited state molecular dynamics.[1] Then, I will introduce an extension of Olivucci's Automatic Rhodopsin Modeling approach, based on a simple electrostatic approach which can identify the most important protonation microstates at a given pH.[2] Its application to Archaeorhodopsin-3 will illustrate how this approach allows to compute pH-dependent absorption spectra.

### **References**

- [1] Pieri E, Weingart O, Huix-Rotllant M, Ledentu V, Garavelli M, Ferré N. Modeling pH-dependent biomolecular photochemistry, *J. Chem. Theory Comput.* 20, 842 (2024).
- [2] Cardenas G, Ledentu V, Huix-Rotllan M, Olivucci, M, Ferré N. Automatic Rhodopsin Modeling with Multiple Protonation Microstates, *J. Phys. Chem. A* 127, 9365 (2023).

## Theoretical spectroscopy of defect qubits

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Solid-state defects acting as single photon sources and quantum bits are leading contenders in quantum technologies. Despite great efforts, not all the properties and behaviours of the presently known solid-state defect quantum bits are understood. Furthermore, various quantum technologies require novel solutions, thus new solid-state defect quantum bits should be explored to this end. These issues call to develop *ab initio* methods which accurately yield the key parameters of solid-state defect quantum bits and vastly accelerate the identification of novel ones for a target quantum technology application. I give a brief overview about recent developments with emphasizing the importance of the electron-phonon interaction in the manifestation of the key magneto-optical properties and spin relaxation and coherence times.

### References

- [1] *Ab initio* theory of the nitrogen-vacancy center in diamond, Adam Gali, *Nanophotonics* 8, 1907-1943 (2019).
- [2] Recent advances in the *ab initio* theory of solid-state defect qubits, Ádám Gali, *Nanophotonics* 12, 359-397 (2023).

## Ab initio approach to point defect in wurtzite boron nitride

Martino Silveti<sup>a,b</sup>, Fulvio Paleari<sup>c</sup>, Ádám Gal<sup>d,e</sup> and Elena Cannuccia<sup>a,b</sup>

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The wurtzite polymorph of boron nitride (wBN) plays the role of intermediary in the hexagonal (hBN) to cubic (cBN) phase transition with the hexagonal to wurtzite phase transformation occurring at around 13 GPa [1]. Millimeter-size samples of wBN have been successfully produced and been shown to be stable at ambient pressure [2]. The wide band gaps of hBN, cBN and wBN make these materials interesting as potential defect hosts as they provide a large range of energies available for defect-induced levels. Moreover, like cBN, wBN has been shown to be a super-hard material [3], ideal for optoelectronic devices under harsh conditions. Nonetheless, while a pretty rich literature of studies on color centers in hBN and cBN is available, the optical properties of wBN have not been deeply investigated yet. In this study we use Many-Body Perturbation Theory on top of Density Functional Theory (DFT) to explore the electronic and optical properties of pristine wBN in the range of pressures involved in the hexagonal to wurtzite phase transition [4]. The  $G_0W_0$  approximation [5] provided the quasiparticle-corrected band structure while the Bethe-Salpeter equation with screening effects was used to obtain the absorbance spectrum. The supercell approach is then used to study the thermodynamics for some simple point defects in wBN (vacancies and carbon substitutionals) by mean of hybrid functional DFT. The formation energies and Charge Transition Levels (CTL) [6] were derived using a correction scheme [7] to account for the electrostatic interaction between the defect periodic images. A simple Franck-Condon model is then used to study the multiphonon emission for such defects. Finally, we explore the optical properties of defective wBN by solving the Bethe-Salpeter equation for such systems. Since evaluating the screening for the supercell may be computationally prohibitive we circumvent this practical obstacle by using the one of the pristine unit cell. The Bethe-Salpeter equation is then solved including this approximated screening in the kernel while still retaining the supercell wavefunctions. A comparison between the absorbance spectra of the defective and the pristine materials is presented.

### References

- [1] A. Segura et al., *Journal of Physical Chemistry C* 2019 123 (33), 20167–20173, DOI: 10.1021/acs.jpcc.9b06163
- [2] Chunlin Chen et al., *PNAS* 116 (23) 11181–11186, <https://doi.org/10.1073/pnas.1902820116>
- [3] Liu, Y. et al. *Sci Rep* 9, 10215 (2019). <https://doi.org/10.1038/s41598-019-46709-4>
- [4] Martino Silveti, Claudio Attaccalite and Elena Cannuccia, Pressure dependence of electronic, vibrational and optical properties of wurtzite-boron nitride, *Phys. Rev. Materials* 7, 055201 (2023), <https://doi.org/10.1103/PhysRevMaterials.7.055201>
- [5] G. Onida, L. Reining, and A. Rubio, “Electronic excitations: density-functional versus many-body Green’s-function approaches, *Rev. Mod. Phys.* 74, 601 (2002).
- [6] Christoph Freysoldt, Blazej Grabowski, Tilmann Hickel, Jörg Neugebauer, Georg Kresse, Anderson Janotti, and Chris G. Van de Walle, *Rev. Mod. Phys.* 86, 253 – Published 28 March 2014
- [7] Christoph Freysoldt, Jörg Neugebauer, and Chris G. Van de Walle, *Phys. Rev. Lett.* 102, 016402 – Published 5 January 2009



## Excited-state geometry relaxation of point defects in monolayer hexagonal boron nitride

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Point defects in hexagonal boron nitride (hBN) are often discussed as single-photon emitters for quantum technologies. Understanding the dependence of electronic and optical properties on the geometry might help to identify the atomic structure of the defects and is also crucial in order to make these emitters applicable. Here, we study three defects in a monolayer of hBN, namely  $C_B C_N$ ,  $C_B O_N$ , and  $C_N V_N$ , from an ab initio approach. We use (constrained) density functional theory to obtain optimal geometries of the electronic ground state and the first excited state, and then refine quasi-particle energies and optical excitation energies using a  $GW$  and BSE based approach. All three defect systems host transitions between deep lying defect states. We find the lowest defect excitons between 4 eV ( $C_B C_N$ ) and 2 eV ( $C_B O_N$ ) and significant Stokes shifts of 0.2 eV and 0.7 eV, respectively [1]. For  $C_N V_N$ , we additionally observe a rotation of the polarization of the defect exciton between absorption and emission.

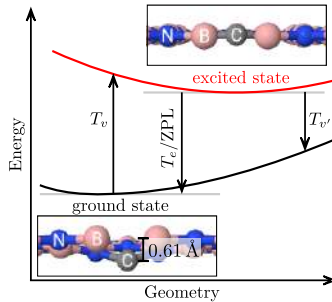


Figure 1: The configuration coordinate diagram visualizes the different vertical transition energies, e.g., in absorption ( $T_v$ ) and emission ( $T_{v'}$ ).

### References

- [1] A. Kirchhoff, T. Deilmann, and M. Rohlfing, “Excited-state geometry relaxation of point defects in monolayer hexagonal boron nitride”, *Phys. Rev. B* 109, 085127 (2024).

## Embedding many-body perturbation theory in large scale environments

**Xavier Blase<sup>a</sup>**

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In many situations pertaining to physics, chemistry or biology, an “active” subsystem is embedded in a large scale, potentially disordered, environment that strongly renormalizes its charged and neutral excitations. We present our efforts towards the development of embedded *GW* and Bethe-Salpeter many-body perturbation theory for the study of the electronic properties of subsystems immersed in large scale polarizable environments. Starting with semi-empirical descriptions of the environment, within the so-called polarizable continuous model (PCM) and QM/MM partitioning, [1-3] we will present fully *ab initio* QM/QM (*GW*/RPA) formalisms allowing to treat polarizable environments containing hundred thousand atoms. [4] Our approach is made possible by partitioning the susceptibility within a fragment, or subsystem-based, approximation, together with an automatic compression of the susceptibility operator using an optimal polarization basis. Application to organic systems is ideally suited to the fragment approximation, but we show that insulating covalent systems can be partitioned, facilitating e.g. the study of defects in multilayer hexagonal-boron-nitride in the true dilute limit. [5] This work is a collaborative effort involving mainly D. Amblard (PhD student) and Gabriele D’Avino at CNRS/Grenoble, together with J. Li and I. Duchemin at CEA/Grenoble that all contributed to the development of the FIESTA and BEDEFIT packages, implementing many-body perturbation theory with Gaussian basis sets.

### References

- [1] I. Duchemin, D. Jacquemin, and X. Blase, “Combining the *GW* formalism with the polarizable continuum model: A state-specific non-equilibrium approach”, *J. Chem. Phys.* 144, 164106 (2016).
- [2] I. Duchemin, C.A. Guido, D. Jacquemin, and X. Blase, “The Bethe-Salpeter formalism with polarisable continuum embedding: reconciling linear-response and state-specific features”, *Chem. Sci.* 9, 4430 (2018).
- [3] J. Li, G. D’Avino, I. Duchemin, D. Beljonne, and X. Blase, “Combining the Many-Body *GW* Formalism with Classical Polarizable Models: Insights on the Electronic Structure of Molecular Solids”, *J. Phys. Chem. Lett.* 7, 2814 (2016).
- [4] D. Amblard, X. Blase, I. Duchemin, “Many-body *GW* calculations with very large scale polarizable environments made affordable: A fully *ab initio* QM/QM approach”, *J. Chem. Phys.* 159, 164107 (2023).
- [5] D. Amblard, G. D’Avino, I. Duchemin, X. Blase, “Universal polarization energies for defects in monolayer, surface, and bulk hexagonal boron nitride: A finite-size fragments *GW* approach”, *Phys. Rev. Materials* 6, 064008 (2022).

## Solvation dynamics influences the photophysics of a nucleic acid-protein interaction model

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A molecular description of the interplay between nucleic acids (NA) and proteins is vital to understand the most basic processes of life, from DNA replication to protein expression and synthesis. Isolation of transient NA-protein complexes in their biological conformation is a challenging task: a promising technique for this goal is to induce *in vivo* NA-protein crosslinking (NPCL) by femtosecond-stimulated UV laser pulses.<sup>1</sup> The photo-addition of phenylalanine to thymine results an exemplificatory reaction of such a class of crosslinking and the photocyclization of 5-benzyluracil (5BU) to 1,2-indaneuracil has been proposed as a model reaction for studying the mechanism of NPCL.<sup>2</sup> In this context, we present a combined experimental and theoretical study of the ground-state conformational equilibrium and the photophysics of 5BU in methanol,<sup>3</sup> exploiting the framework of the DFT and TD-DFT.<sup>4</sup> We performed an *ab initio* molecular dynamics (AIMD), adopting the ADMP<sup>5</sup> method and an hybrid QM/MM ONIOM<sup>6</sup> partition scheme enforcing non-periodic boundary conditions<sup>7</sup>. We recorded the UV absorption spectrum in methanol solution and simulated the band shape from TD-DFT analysis of the AIMD trajectory, finding a good agreement between experiments and simulation. The simulated absorption spectrum, calculated on such ensemble, allowed to give a molecular interpretation of the experimental UV-Vis lowest energy band, involved also in the induced photo-reactivity upon irradiation. In particular, the first two excited states both contribute to 5BU lowest energy absorption. Moreover, as main result, the nature and brightness of such electronic transitions is strongly influenced by 5BU conformation and the microsolvation of its heteroatoms. Our study suggests a dependence of NPCL on the surrounding environment and remarks the importance of AIMD and an accurate treatment of the solvent to understand the photophysics of this model system; it also may serve as a starting point for a non-adiabatic treatment of the photoinduced evolution of the system in the excited state.

Figure 1 : The photocyclization of 5BU (left) and A snapshot from the AIMD of 5BU in methanol (right).



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## Quantum dynamics around PPEs' conical intersections for both spectroscopic and real-time studies

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In this presentation, we discuss the use of nonadiabatic quantum dynamics for the study of different poly(phenylene)ethynylenes (PPEs, fig. 1 left), building-blocks of light-harvesting dendrimers. PPEs exhibit strongly nonadiabatically coupled electronic excited states. In this context, the Born-Oppenheimer approximation breaks down, which requires molecular dynamics to explicitly take into account the coupling between the electronic excited states. We propose linear and quadratic vibronic coupling diabatic model Hamiltonians for reproducing the PESs of the simplest PPEs. We use these diabatic potentials and inter-state couplings to run quantum dynamics calculations using the Multi-Configuration Time-Dependent Hartree (MCTDH) formalism for explicit wavepacket propagation.

The first building block is the symmetrically meta-substituted phenylene (m22, fig. 1 left). The absorption spectra of m22 and its localized 2-ring fragment (p2, fig. 1 left) are similar.[1–2] However, their emission spectra are different, with m22 exhibiting an unusual Stokes shift of about  $2000\text{ cm}^{-1}$ . In a recent work[3], we identified this red-shifted spectrum as a contribution resulting from the strong inter-state coupling between the first two electronic excited states (fig. 1 right), mostly due to acetylenic vibrations.

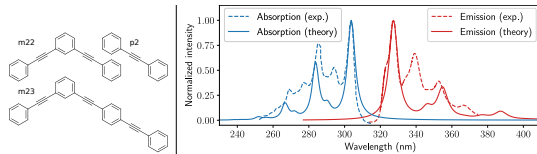


Figure 1: Left: poly(phenylene)ethylenes studied in this work. Right: Absorption and emission spectra of m22 (normalized intensity), experimental measurement[1] and theoretical simulation[3].

The second building block of interest is an asymmetrically meta-substituted PPE (m23, fig. 1 left) for which excitation-energy transfer (EET) occurs thanks to different branch lengths hence a significant energy gradient. Using analogous Hamiltonian models and wavepacket propagation, we are able to reproduce the expected ultrafast ( $< 25\text{ fs}$ ) EET from an excitation on the shortest branch toward the second branch.[4] We further characterize the EET phenomenon *via* electronic-vibrational relations, vibrational energy analysis and evaluation of electronic coherence.

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## Efficient many-body perturbation theory calculations for 2D materials

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Many-body perturbation theory methods, such as the GW approximation, are the state of the art for the calculation of quasiparticle (QP) properties of a large range of materials. However, evaluating the GW self-energy is often computationally challenging due to the frequency and momentum convolutions. These difficulties were recently addressed by the developments of the multipole approximation (MPA) [1,2] and the W-av [3,4] methods. On the one hand, MPA, with its optimized sampling of the polarizability in the frequency complex plane and a multi-pole interpolation, leads to an accuracy comparable with full-frequency methods at much lower computational cost. On the other hand, W-av drastically improves the convergence of the QP corrections of 2D semiconductors with respect to the Brillouin zone sampling, by combining a Monte Carlo integration with an interpolation scheme able to represent the screened potential between the calculated grid points. In this work, we present these two theoretical schemes and show examples of the accuracy and computational gains when applied to prototype 2D systems.

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## Atomic scale mapping of electric field and charge density by 4D-STEM: a study on perfect and defective h-BN

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The charge density distribution, which fully defines the ground-state properties of a material system, can be accurately measured in single crystals using X-ray diffraction. However, there is still a lack of experimental techniques capable of measuring charge density redistribution in defective or heterogeneous crystals at the relevant atomic scale with the required precision. While this information can be effectively evaluated by modern numerical methods such as DFT, these theoretical predictions still require experimental validations. In recent years, four-dimensional scanning transmission electron microscopy (4D-STEM), in which a 2D convergent beam electron diffraction pattern is acquired at each probe position, has emerged as an attractive alternative approach. In principle, the technique can simultaneously provide precise structural determinations and capture details of local electric fields and charge densities. However, accurate extraction of quantitative data at the atomic scale is challenging, mainly due to probe propagation and size-related effects, which may even lead to misinterpretations of qualitative effects.

Here we discuss the ultimate capabilities of 4D-STEM through a comprehensive study of pristine and defective h-BN flakes. We demonstrate that while precise charge quantification at individual atomic sites is hindered by probe effects, 4D-STEM can directly measure charge transfer phenomena with sensitivity down to a few tenths of an electron and a spatial resolution on the order of a few angstroms.

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## Distinguishing different stackings in layered materials via luminescence spectroscopy

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Despite its simple crystal structure, layered boron nitride features a surprisingly complex variety of phonon-assisted luminescence peaks. We present a combined experimental and theoretical study on ultraviolet-light emission in hexagonal and rhombohedral bulk boron nitride crystals. Emission spectra of high-quality samples are measured via cathodoluminescence spectroscopy, displaying characteristic differences between the two polytypes. These differences are explained using a fully first-principles computational technique that takes into account radiative emission from “indirect”, finite-momentum, excitons via coupling to finite-momentum phonons. We show that the differences in peak positions, number of peaks and relative intensities can be qualitatively and quantitatively explained, once a full integration over all relevant momenta of excitons and phonons is performed.[1]

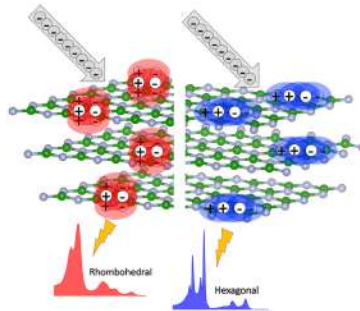


Figure 1: Scheme of phonon-assisted luminescence in rBN (left) and hBN (right). The light-emitting recombination of excitons – bound electron-hole (e-h) pairs – can be mediated by out-of-plane lattice vibrations only in rBN, resulting in a different emission spectrum with respect to the hBN case, when only in-plane phonons contribute.

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**Excitonic responses in MoS<sub>2</sub>/WS<sub>2</sub> and MoSe<sub>2</sub>/WSe<sub>2</sub>  
heterostructures from first-principles:  
impact of stacking order and interlayer distance**

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We present *first-principles* calculations of the optical properties of MoS<sub>2</sub>/WS<sub>2</sub> and MoSe<sub>2</sub>/WSe<sub>2</sub> van der Waals (vdW) heterostructures of Transition Metal Dichalcogenides (TMDs). Using the Many-Body approach implemented in the YAMBO [1,2] code, we compute GW corrections to the electronic structure, absorption and PhotoLuminescence (PL) spectra. We detail the electronic valley splitting and the origin of each optical transition, discussing the enhancement and quenching of the inter/intra layer exciton states due to different stacking and polarization directions. Furthermore, we demonstrate a substantial variation of the exciton peaks (of the order of  $\sim 100$  meV) and of the exciton radiative lifetimes (from picosecond to nano/micro-seconds) due to the influence of stacking alignment and interlayer distance. The ability to control exciton energy and lifetimes offers promising prospects for applications in quantum information and optical sensing.

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## Modelling electronic and optical properties of graphene and boron nitride lateral heterostructures

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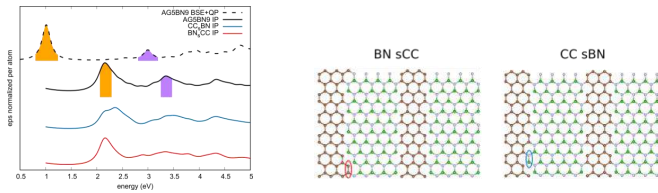
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Graphene (Gr) and hexagonal boron nitride (hBN) have a similar lattice parameter ( $\sim 1.5\%$  mismatch) and different properties, Gr is a metal known by its high conductivity and hBN is a large gap insulator ( $\sim 6\text{eV}$ ) with a strong UV emission. Due to these two remarks, they are perfect candidates to be stacked side-by-side [1] in a lateral heterostructures instead of one of the top of the other in a more common vertical heterostructure.

The properties of a lateral heterostructure are sensitive to the presence of defects, interface shape and roughness, for instance an heterostructure composed of Gr and hBN ribbons alternatively with armchair interface (AGBN-LHS) have a direct gap tunable with the size of Gr and hBN part [2]. We present first a study of AGBN-LHS electronic structure as a function of Gr and hBN width carry out with density functional theory and optical spectrum calculated by GW and BSE.

However, during the synthesis of this kind of heterostructures defects can appear at the interface [3]. Using a parametrised tight-binding (TB) model we look at the effects on the absorption spectra due to the presence of different defects as roughness interface, Stone-Wales and vacancy defects.



**Figure 1:** In the left panel, the absorption spectra normalized per atom is shown for different structures. The spectra of an ideal heterostructures composed of armchair graphene ribbon of 5 rows and armchair hBN ribbon of 9 rows, AG5BN9 (black lines). The dashed spectra has been calculated with GW and BSE and the continuum spectra within the parametrised TB model. The color areas set the correspondence between both techniques. The red line (blue line) spectrum correspond to the same structure with a substitutional defect of a BN (CC) dimer by a CC (BN) dimer, represented in middle (right) panel.

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## Theoretical methods for the description of correlated electron and nuclear dynamics at the attosecond time scale

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Monitoring electron dynamics in matter requires the use of coherent light sources that can offer attosecond resolution. For more than two decades, attosecond experiments have been mostly performed using table-top experimental set ups where trains of attosecond pulses are produced through high-harmonic generation (HHG). The success of HHG techniques has been recently recognized with the Nobel Prize in Physics, in 2023. As revolutionary as this attosecond technology, free electron laser (FEL) facilities have sprout over the world in the last years, generating high brilliance and high intense pulses with a high frequency tunability. FEL and X-Ray FEL (XFEL) applications range from material characterization, biology, engineering or chemical physics. And, nowadays, it is also possible to generate attosecond pulses at XFEL upon the introduction of self-amplified spontaneous emission (SASE) schemes. Although operating at different frequency ranges, HHG and XFEL now offer the necessary time resolution to access and steer electronic motion in matter. This talk will present our most recent theoretical advances to describe novel applications and experiments using attosecond pump-probe spectroscopic techniques in gas phase targets [1].

This talk will briefly review recently developed theoretical methods to interpret and predict ultrafast charge migration phenomena in biomolecules [2], using HHG and FEL technique, as well as our progresses on accurate, nearly-exact, methodologies to describe attosecond pump-probe experiments in small molecules and atoms [3,4]. XUV-IR pump-probe schemes will be discussed, since they have been widely employed to retrieve electronic dynamics information, for instance, in applications exploring photoionization time delays employing attosecond electron streaking or RABBIT (reconstruction of attosecond beating by interference of two-photon transitions) techniques [5,6]. Despite the success of these applications, several laboratories are pursuing the “ideal” alternative, i.e., the use of attosecond XUV pump – attosecond XUV probe schemes, where the shorter wavelengths ensure a negligible distortion of the atomic or molecular potential. Recent applications obtaining time-resolved images of the ultrafast dynamics triggered in excited and ionized atomic and molecular targets will be presented, with particular focus on the role of the electron-electron correlation and electron-nuclear coupled motions.

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## Theoretical description of X-ray absorption of laser-driven materials

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We developed an ab initio approach to describe a 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 obtain laser-dressed electronic states within the Floquet-Bloch formalism combined with the Density Functional Theory [1] within the LAPW+lo method [2]. We describe the interaction of a laser-dressed electronic system with an X-ray pulse using the density matrix formalism and calculate the X-ray absorption probability. We show that the developed technique gives information about dynamical changes in the electronic population of valence and conduction bands in site- and orbital-resolved manner. We calculate the modification of partial density of states (PDOS) in the presence of external driving, analyze the connection between laser-induced electron dynamics and changes in X-ray absorption spectra by performing calculations for wurtzite ZnO.

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## Imaging Excitons in C60 Thin Films with Photoemission Orbital Tomography

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Angle-resolved photoemission spectroscopy is an established technique to characterize the electronic and structural properties of well-ordered molecular films. In combination with femtosecond laser pulses, the method also gives access to the excited state landscape of these films. In particular, using such a setup, an ultrafast energy conversion process has been imaged experimentally after excitation of C60 thin films [1]. In our contribution, we aim to understand these femtosecond-resolved photoemission measurements on a theoretical level. For this, we approach the excited states of C60 clusters within the many-body perturbation framework of the Bethe-Salpeter equation. As a first step, we compare and analyse the calculated exciton wavefunctions and their single-particle contributions (Figure). Then, building on our calculations, we employ a recently introduced formalism [2] to disentangle the experimental photoemission data and identify the momentum fingerprints of four different excitonic states during the energy relaxation cascade.

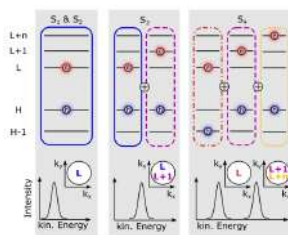


Figure: Single particle contributions to the exciton wavefunctions of four excited states in C60 dimers (top, S1-S4), and their expected photoemission signatures (bottom).

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**Ultrafast spectroscopy of semiconductors:  
probing the nonequilibrium electron-phonon dynamics**

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Nonequilibrium electron dynamics governed by electron-phonon interactions plays a central role in the operation of various electronic devices and is essential for understanding many phenomena in condensed matter. The real-time Boltzmann transport equation (rt-BTE) using scattering integrals computed from first principles can describe various dynamic phenomena involving electrons and phonons. In the first part of the talk, I will present a joint experimental and theoretical investigation of the excited-state electron and hole cooling dynamics. Combining the transient absorption (TA) measurements with unprecedented temporal resolution and the *ab initio* ultrafast electron-phonon dynamics, we show conclusive evidence in TA spectra for the asymmetric hot electron and hole cooling in nitrides and zinc-blende materials.

The second part of the talk will focus on the structural dynamics, in particular, the phonon-phonon interactions incorporated into the rt-BTE. For realistic systems and dense momentum grids, these calculations become extremely challenging because of the high computational cost of the scattering integrals. Here, we employ multirate infinitesimal methods, implemented in the Sundials library [1], to split different time scales in coupled electron and phonon rt-BTE. Additionally, I will present a recently developed data-driven approach, dynamic mode decomposition (DMD), to speed up rt-BTE and learn the dominant patterns from dynamics [2]. I will conclude with an overview of our Perturbo code [3], an open-source framework to study electron interactions and dynamics in materials, highlighting open problems and future directions.

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***Ab initio* study of exciton-exciton transitions in pump and probe experiments****C.Attaccalite<sup>a</sup>, M. D'Alessandro<sup>b</sup>, D. Sangalli**<sup>a</sup>*CINaM Campus de Luminy 163 Avenue de Luminy, Case 913, 13288 Marseille Cedex 9**E-mail: [claudio.attaccalite@univ-amu.fr](mailto:claudio.attaccalite@univ-amu.fr)*<sup>b</sup>*Istituto di Struttura della Materia-CNR (ISM-CNR), Area della Ricerca di Roma 1, Monterotondo Scalo, Italy*<sup>c</sup>*Istituto di Struttura della Materia-CNR (ISM-CNR), Via del Fosso del Cavaliere 100, 00133 Roma, Italia*

In pump-probe spectroscopy, two laser pulses are employed to garner dynamical information from the sample of interest. The pump initiates the optical process by exciting a portion of the sample from the electronic ground state to an accessible electronic excited state, an exciton. Thereafter, the probe interacts with the already excited sample. The change in the absorbance after the pump provides information on transitions between the excited states and their dynamics. In this work, we study these exciton-exciton transitions by means of an *ab initio* real-time propagation scheme based on dynamical Berry phase formulation. The results are then analyzed taking advantage of a Fermi golden rule approach formulated in the excitonic basis-set and in terms of the symmetries of the excitonic states. Using bulk LiF and two-dimensional hBN as two prototype materials, we discuss the selection rules for transitions involving strongly bound Frenkel excitons, for which the hydrogen model cannot be used.

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## Discussing protection in FMO complex from NEGF-based exciton transport modelling and simulation

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Natural devices, such as those involved in photosynthesis, can display intriguing characteristics linked to the complexity of their architecture and operation. They have therefore emerged as avenues towards a renewed understanding of biological functions and bioinspired technological alternatives to semiconductor-based nanoelectronics [1]. We put the focus on the Fenna-Matthews-Olson (FMO) complex, renowned for its high efficiency of photon-to-electron conversion [2]. This natural light-harvesting system operates in bacteria in between a large antenna, the chlorosome, and the reaction center where electrochemical conversion takes place. With the aim of deepening the present understanding of this natural device, and reproducing artificial FMO counterparts, we have developed a framework suitable for exciton quantum transport using the non-equilibrium Green's function formalism, starting from the pioneering work of Pelzer and co-workers [3]. We first confirm that the coupling to local vibrations is crucial to assist exciton extraction from FMO. We further develop the study regarding the impact of reservoir on the functioning of the FMO architecture. Interestingly, we find that for a small increase of the exciton number in the reaction centre, the output reservoir, the exciton flow is reversed, and energy is released by recombination in the present modelling. Finally, we examine the role of the particular FMO architecture on such a protection mechanism.

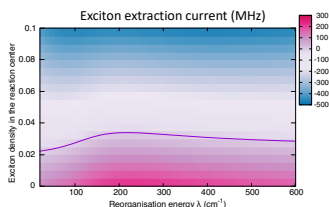


Figure: Exciton current mapping as a function of the exciton number in the reaction center and the coupling to vibrations in terms of reorganisation energy (in  $\text{cm}^{-1}$ ).

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## Ultrafast electron dynamics observed with attosecond pulses

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The recent years have seen spectacular progress in the generation of attosecond ( $1 \text{ as} = 10^{-18} \text{ s}$ ) light pulses, rewarded by the 2023 Physics Nobel Prize [1]. Their ultrashort duration allows the observation of electron dynamics on their natural timescale. In particular, with electron interferometry techniques, the photoionization process can be characterized in the time domain. So-called photoionization time delays [2] can be measured, which are very sensitive to the atomic/molecular potential.

In this talk, the generation of attosecond pulses will be introduced, and illustrative measurements of photoionization dynamics will be presented: in the vicinity of the Cooper minima in Argon [3] and in the presence of autoionizing resonances [4,5].

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## Non-linear phononics in THz excited Bi<sub>2</sub>Te<sub>3</sub> nanofilms

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Density Functional Theory (DFT) calculations not only allow to predict the vibrational and optical properties of solids[1] but also to understand and disentangle the mechanisms playing a key role in the generation of acoustic and optical coherent phonons[2-5]. Recent experiments performed on a Bi<sub>2</sub>Te<sub>3</sub> nanofilm have shown that a THz pulse launches a coherent  $A_{1g}$  phonon[5]. Such an observation can be explained by invoking either a sum frequency process[6] or non linear phonon-phonon couplings[7]. By resorting to group theory and calculating energy surfaces from first-principles, the main phonon-phonon couplings can be identified. Furthermore, a minimal model can be built to explain qualitatively pump-probe experiments. This model is validated by simulating the detection process as the modification of the dielectric function arising from the generation of optical or acoustic phonons can easily be evaluated at the random phase (RPA) approximation level[8,9]. Therefore, ab-initio calculations when combined with models are invaluable tools to shed lights on the complex mechanisms at the heart of ultrafast physics.

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**TDDFT simulations applied to pump-probe experiments with sapphire****K. Lévêque-Simon<sup>a</sup>, R. Sinha-Roy<sup>b</sup>, V. Véniard<sup>a</sup>, P. Lample<sup>c</sup>, M. Weis<sup>d</sup> and S. Guizard<sup>c</sup>**<sup>a</sup> *Laboratoire des Solides Irradiés (UMR 7642), CEA, CNRS, Ecole Polytechnique,*  
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The aim of this theoretical research work is to investigate the dynamics of solids in order to better understand the excitation and relaxation processes at ultrafast time scales. In particular, we will focus on wide band-gap materials like quartz ( $\text{SiO}_2$ ) and sapphire ( $\text{Al}_2\text{O}_3$ ) with optical cycle temporal resolution and beyond, with sub-femtosecond resolution. Thus we will address the physical mechanisms of laser-matter interaction and initial electronic relaxation in solids (multiphoton ionization, band gap modulation...) in connection with time resolved pump-probe experiments which currently operates at LIDYL/LOA [1]. We were first interested in the absorption spectrum for  $\text{Al}_2\text{O}_3$  in the linear regime using the versatile *Octopus* DFT/TDDFT open-source software [2,3] based on a space/time resolution of the Kohn-Sham equations. In this approach, the dielectric function is obtained as the Fourier transform of the total electric current. We then carried out benchmarking of this spectra by considering as a reference those generated from the TDDFT linear response (DP) [4] and from the Bethe-Salpeter equation (EXC) [5] codes, which operate in reciprocal space and frequency domain. From this analysis, the *Octopus* input parameters (XCFunctional) suitable for the study of sapphire have been obtained. The latter, leading to an optical bandgap in agreement to the one measured experimentally (around 9 eV), enable to simulate appreciably the three-photon absorption process in the non-linear regime. In this regime, we will present results for the change in reflectivity of the material after excitation by the pump. These results will be presented for several delays between the pump and the probe.

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## X-ray transient absorption spectrum at Zn K-edge: Pauli blocking and core-hole screening modification

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X-ray transient absorption (XTA) spectroscopy can provide valuable insight into the atomic structure, the electronic properties, and the dynamics of materials. Zinc oxide (ZnO) is a prototypical wide-bandgap material with many applications in optoelectronics and photovoltaics. Combining state-of-the-art first principle calculations and picosecond X-ray transient absorption spectroscopy at the Zn K-edge of ZnO thin films, we reveal XTA spectra are composed of overlapping contributions from lattice heating, core-hole screening modification, and Pauli blocking. Specifically, we analyze the computed results obtained from the Bethe-Salpeter equation combined with time-dependent density functional theory and constrained density functional theory to disentangle the effect of Pauli blocking and core-hole screening modification. Our combined experimental and theoretical approach reveals that core-hole screening modification intensifies with increasing excitation densities; its contribution to the XTA spectra dominates over Pauli blocking.

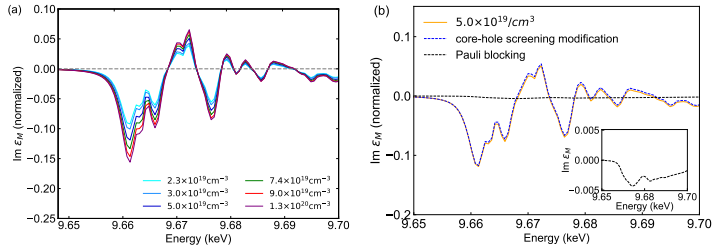


Figure 1: (a) Computational non-thermal spectral contributions to the XTA in the XANES at the Zn K-edge of ZnO with rising carrier densities. (b) Decomposition of the non-thermal XTA at the Zn K-edge of ZnO into contributions from Pauli blocking and core-hole screening modification. Inset shows the effect of the Pauli blocking only.

## Relativistic electronic-structure methods based on effective quantum electrodynamics

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It is important to take into account the effects of special relativity in the quantum description of electronic systems with heavy elements. Relativistic electronic-structure computational methods based on the no-pair Dirac–Coulomb or Dirac–Coulomb–Breit Hamiltonian have thus been developed and are now routinely applied to molecular systems (see, e.g., Refs. 1–3). The next challenge for relativistic quantum chemistry is to go beyond the no-pair approximation, i.e., including the quantum-electrodynamics (QED) effect of virtual electron–positron pairs. This is desirable not only for an increased accuracy but also in order to put relativistic quantum chemistry on deeper theoretical grounds.

An attractive approach to performing ab initio relativistic calculations beyond the no-pair approximation is to use an effective QED Hamiltonian with the Coulomb or Coulomb–Breit two-particle interaction (see, e.g., Refs. 2 and 4–7). This effective QED theory properly includes the effects of vacuum polarization through the creation of electron–positron pairs but does not explicitly include the photon degrees of freedom. It is, thus, a more tractable alternative to full QED for electronic-structure calculations.

In this presentation, I will review this effective QED theory, I will discuss the possibility to formulate a relativistic density-functional theory based on it [7], and I will show results on a one-dimensional hydrogen-like model system [8].

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## Linear scaling approach for optical excitations using maximally localized Wannier functions

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We present a theoretical method for calculating optical absorption spectra based on maximally localized Wannier functions, which is suitable for large periodic systems. For this purpose, we calculate the exciton Hamiltonian, which determines the Bethe-Salpeter equation for the macroscopic polarization function and optical absorption characteristics. [1] The Wannier functions are specific to each material and provide a minimal and therefore computationally convenient basis. Furthermore, their strong localization greatly improves the computational performance in two ways: first, the resulting Hamiltonian becomes sparse and, second, the electron-hole interaction terms can be evaluated efficiently in real space, where large electron-hole distances are handled by a multipole expansion. For the calculation of optical spectra, we employ the sparse exciton Hamiltonian in a time-domain approach, which scales linearly with system size. [2]

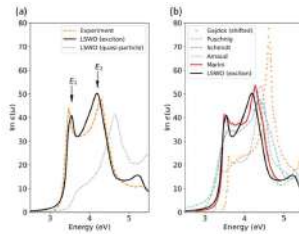


Figure: Imaginary part of the dielectric function for bulk silicon.

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## Z-vector approach to BSE analytic gradients

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We present an implementation of excited-state analytic gradients within the Bethe-Salpeter equation formalism using an adapted Lagrangian Z-vector approach with a cost independent of the number of perturbations when excited-state electronic dipole moments are required. In this framework, we assess the accuracy of neglecting the screened Coulomb potential derivatives, as well as the impact of replacing the GW quasiparticle energy gradients by their Kohn-Sham analogs. The pros and cons of these approaches are benchmarked using both a set of small molecules for which very accurate reference data are available and the challenging case of increasingly extended push-pull oligomer chains. Finally, we present the BSE analytic gradient equations with respect to atomic coordinate displacements and using atom-centered localized basis functions, useful for optimizations and quantum photochemistry analysis, for example, structural reorganizations inside a photo-induced chemical reaction.

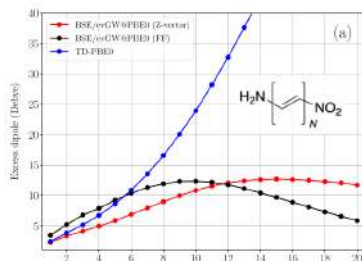


Figure 1: Excess dipole moments for increasing push-pull oligomer chain for BSE@PBE0 (analytical and numerical) and TD-PBE0.

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## Dynamical effects in the spectra of neutral excitations

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One of the big challenges of theoretical condensed matter physics is the description, understanding, and prediction of the correlation effects induced by the mutual interaction between particles on materials properties. In both electronic and optical spectra the Coulomb interaction and the electron-phonon coupling cause a renormalization of the energies and change of spectral weight. Most importantly, they induce a finite lifetime on the quasi-particle (QP) excitations and can lead to new structures, often called satellites. The latter are pure many body effects and can be linked to the coupling of excitations, also termed dynamical effects. Standard methods developed in the framework of many body perturbation theory namely GW and the Bethe-Salpeter equation (BSE)[1] are often not able to capture this complex physics. Instead, approaches based on a picture of electron-boson coupling such as the cumulant expansion are promising for the description of plasmon and phonon satellites.

Motivated by the recent success of the cumulant expansion of the one particle Green's function in the description of photoemission spectra[2,3,4], we generalized the cumulant approach to the evaluation of the electronic polarizability[5,6,7]. In this way we provide a new full ab-initio tool to include dynamical effects beyond the standard BSE in the description of neutral excitations as measured in optical absorption, photoluminescence, electron energy loss and inelastic X-rays scattering spectroscopies. The developed approach is applied to the study of phonon sidebands in the absorption and photoluminescence spectra of monolayer BN[8].

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## The multi-channel Dyson equation : coupling many-body Green's functions

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We present the multi-channel Dyson equation that combines two or more many-body Green's functions to describe the electronic structure of materials. In this talk we use it to model photoemission spectra by coupling the one-body Green's function with the three-body Green's function. We demonstrate that, unlike methods using only the one-body Green's function, our approach puts the description of quasiparticles and satellites on an equal footing.[1] We propose a multi-channel self-energy that is static and only contains the bare Coulomb interaction, making frequency convolutions and self-consistency unnecessary. Despite its simplicity, we demonstrate with a diagrammatic analysis that the physics it describes is extremely rich. Finally, we present a framework based on an effective Hamiltonian that can be solved for any many-body system using standard numerical tools. We illustrate our approach by applying it to the Hubbard dimer and show that it is exact both at 1/4 and 1/2 filling.[2]

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## Symmetries and sum rules in the Bethe-Salpeter equations: theory and implications

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The Bethe-Salpeter Equation (BSE) describes the physics of bound electron-hole pairs, e.g. excitons, in complex materials. Within the most common approximation, which involves a static kernel, it is a rather simple and intuitive equation in which electrons and holes interact via a bare exchange interaction plus a direct and statically screened term. However, despite its simplicity, it hides a number of subtleties and issues. I consider here two of such issues. The first is connected to the popular GW+BSE scheme, e.g. the inconsistency between the ground state approximation, for the self-energy, and the approximation entering the BSE kernel. The second is connected to the spin structure of the BSE in magnetic materials or in presence of spin-orbit coupling when using a static kernel.

In the talk I discuss how these issues have implications in different physical problems:

- (i) the search for excitonic-insulators in extended systems [1];
- (ii) the definition of the total spin of excited states and excitons [2-3];
- (iii) the calculation of magnons dispersions [4].

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**Excitonic Effects in  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> revealed through resonant inelastic x-ray scattering and complementary x-ray spectroscopies**

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Resonant inelastic X-ray scattering (RIXS) is a powerful technique for studying elementary excitations due to its element and orbital specificity, as well as its access to a wide range of energies[1, 2]. Despite its importance, simulating RIXS spectra remains a challenge for theory, often relying on multiplet approaches that approximate solid-state effects and are not parameter-free.

In my presentation, I will focus on RIXS in  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> using a many-body approach [5, 6]. By employing the Bethe-Salpeter equation (BSE), the state-of-the-art technique for describing electron-hole interactions, we accurately capture excitonic effects crucial for reproducing experimental optical and core absorption spectra [3, 4]. This allows for a proper description of the intermediate and final states of RIXS. I will discuss RIXS spectra within two different regimes, Raman and fluorescence, and establish connections with complementary spectroscopic techniques, such as inelastic x-ray scattering and x-ray emission spectroscopy. Finally, for shallow core excitations, I will compare and discuss spectra calculated with all-electron and pseudopotential plane-wave methods.

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## Free-space interferometric routing of upconverted light by dielectric metasurfaces

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Frequency upconversion of near-infrared photons to the visible range is strategic for information technology, as it can provide an alternative for the read out of telecom signals using efficient silicon-based detectors. Light upconversion is a nonlinear process mediated by matter that consists in the interaction of either energy-degenerate photons, such as in second-harmonic and third-harmonic generation (THG), or photons with different energies, such as in sum-frequency generation (SFG). We recently investigated frequency upconversion in both dielectric [1] and plasmonic nanoantennas [2]. Thanks to the adopted dual-beam pump scheme, where an ultrashort pulse ( $\omega$ ) at telecom wavelength ( $\lambda = 1551$  nm) impinges on the sample along with its frequency-doubled replica ( $2\omega$ ), THG and SFG are degenerate ( $\lambda = 517$  nm). This, along with coherence, enables the interference between the processes. Yet, we found that in individual nanoantennas symmetry plays a major role in enhancing/suppressing the interference between SFG and THG. By tuning the relative phase between the two impinging pulses, we performed all-optical switching of upconverted light with efficiency  $> 50\%$  in asymmetric nanoantennas [2].

Optical metasurfaces are rapidly emerging as flexible, ultrathin and multi-functional platforms to manipulate light [3]. Recently, they were also applied to efficient nonlinear light conversion and steering [4]. Here, by applying the above dual-beam pump scheme to a periodic AlGaAs metasurfaces, we attain all-optical routing of the upconverted telecom photons in the visible range as in [1,2]. This is attained by tuning the metasurface diffraction to sample the meta-atom nonlinear emission in the Fourier plane, hence breaking the detection symmetry. Using the relative phase between the pump pulses as a tuning knob, we routed the upconverted radiation among different metasurface diffraction orders with an efficiency up to 90% [5]. This is enabled by the maximization of constructive/destructive interference between SFG and THG in specific k-space directions. We also demonstrate that the linear polarization state of both pump and emission allows to reconfigure the routing between different sets of diffraction orders. Although the current demonstration is conducted by leveraging the linear polarization state of both the excitation and the emitted upconversion, given the complex polarization pattern of the involved processes, more intriguing polarization states can be obtained in specific orders. This might translate in the encoding of well-defined polarization states among the diffraction orders. The proposed approach can be envisioned as an all-optical method to route upconverted telecom photons into various detection channels. The combination of the interferometric and nonlinear character of the emitted light could be also extremely appealing for applications to nonlinear sensing.

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## Tunable Second Harmonic Generation in 2D Materials from First-Principles

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Non-linear optical frequency conversion, where optical fields interact with a non-linear medium to generate new field frequencies, is widespread in modern photonic systems. However the challenging aspect often lies in tuning the non-linear electrical susceptibilities responsible for such phenomena in a specific material. Consequently, the dynamic control of optical non-linearities, utilized as a spectroscopic tool, has, until now, mainly been confined to research laboratories. With the idea of being able to create devices with a tunable non-linear response, in this work we explore a mechanism for electrically manipulating second-order optical non-linearity in two-dimensional materials. In particular, we will consider a 2 layers system (2H-MoS<sub>2</sub>), which presents a zero second harmonic generation (SHG) due to spatial inversion symmetry. By using an ab initio real-time approach [1], developed within the Yambo code [2], we show how the effect of an external vertical electric field can induce and modify an SHG response and the role of bound excitons in this non-linear response. From an experimental point of view, different experiments have revealed the possibility of using external fields to induce and modify SHG in 2D materials [3-6].

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## Pressure Dependence of Intra- and Interlayer Excitons in 2H-MoS<sub>2</sub> Bilayers

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The optical spectrum of the MoS<sub>2</sub> bilayer changes under pressure as theoretical and experimental studies have shown [1].

Our computational investigations of the structural, electronic and optical properties are based on elasticity theory, DFT, *GdW* and the Bethe-Salpeter equation. The stress conditions in our diamond anvil cell-experiment result in an effective shift of the excitation energy of the A exciton towards lower energies with increasing pressure. This shift corresponds strongly to the behavior of the direct band gap at the K point. Due to a growing valence band splitting for increasing pressures, the interlayer exciton shows a smaller shift. In total, the A-IL energy splitting decreases under pressure.

Furthermore, the theoretical reproduction of the experimental results shows a suppression of the transfer of hydrostatic pressure due to the interaction between the bilayer and the substrate.

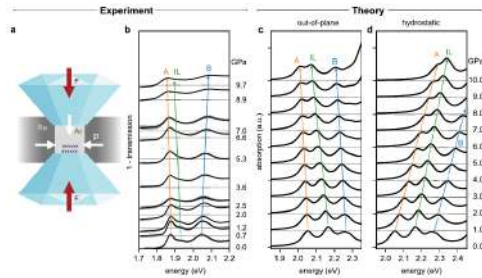


Figure 1: (a) Scheme of the experiment. By pushing the diamond anvils together, the MoS<sub>2</sub> bilayer inside the cell gets compressed. (b) Measured spectra for different pressure values. (c,d) Theoretical results for out-of-plane and hydrostatic compression. The two extreme cases show a blue- and red-shift, respectively. The shifts in the experiment lie between these cases.

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## Correlation effects in solids with RPA and beyond

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I will introduce a set of beyond-RPA approximations based on the exact-exchange vertex function [1,2]. These have recently been implemented for solids within the Quantum ESPRESSO ACFDT package, using the techniques of density functional perturbation theory [3,4]. I will demonstrate that the inclusion of exact exchange solves most of the known deficiencies of the RPA and that high accuracy can be achieved on a wide range of systems. Furthermore, self-consistency can be reached via the optimized effective potential (OEP) approach giving access to structural and vibrational properties, as well as an improved starting point for describing charged excitations within the MBPT formalism. In addition, the OEP can be used to variationally optimize the fraction of exchange of low-cost hybrid functionals. Finally, I will present some recent applications of these methods to excitonic insulators [5] and the phase diagram of high-pressure solid hydrogen [6].

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## From spectra to total energy: beyond the GW approximation designing effective interactions

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The total energy and electron addition and removal spectra can in principle be calculated exactly from the one-body Green's function (GF). In practice, the GF is most often obtained from an approximate self-energy. For the band structure of solids and energy levels of molecules, the GW approximation [1] has become the state-of-the-art approach. This approximation is a first-order expression of the self-energy in terms of the screened Coulomb interaction. By the way of contrast, it is not clear what is the best framework to access the ground state total energy. Indeed, most total energy calculations are today performed using density-functional theory (DFT), not Green's functions. This is due both to the fact that GF calculations have usually a higher computational cost than DFT calculations, and to the fact that there is today no well established approximation for the total energy in the GF framework. Still, there are good reasons to investigate ways to use Green's functions to calculate the total energy [2, 3]. First, exact expressions for the total energy as functional of GF and/or the self-energy are known in principle. Second, the GF framework suggests powerful and systematic approximations. However, the GW approximation, while suitable for spectra, is in practice not satisfactory for the total energy, where usually a high precision is required. Moreover, the validity of the GW approximation is limited to weakly to moderately correlated systems. In this talk, I will present general theoretical developments and suggest new approximations based on the use of effective interactions in order to improve total energy calculations based on Green's functions. The theoretical developments and the quality of the approximations are illustrated through applications to an exactly solvable model, the Hubbard dimer. The results confirm our conjectures and motivate future applications to real materials [4].

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XPS core-level electron removal energies and chemical shifts by *ab initio* theoryV. Olevano<sup>a,b,c</sup>, I. Mukatayev<sup>a,c,d</sup>, F. Moevus<sup>c</sup>, G. D'Avino<sup>a,b,c</sup>, B. Sklénard<sup>a,c,d</sup> and J. Li<sup>a,c,d</sup><sup>a</sup> ETSF, European Theoretical Spectroscopy Facility, France  
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X-ray photoelectron spectroscopy (XPS) measures electron removal energies, providing direct access to core and valence electron binding energies, hence probing the electronic structure. In addition, it also provides informations on the chemical composition and type of bonding, which could be inferred from the shift of the binding energy (also known as chemical shift). In this talk we will present a benchmark of *ab initio* many-body COHSEX and *GW* approximations, with respect to Hartree-Fock (HF) and density-functional theory (DFT), on the complete electron binding energies of noble gas atoms (He-Rn) which spans 100 keV. We will also present a study of the chemical shift of the carbon 1s electron in a set of molecules, providing insights into the physical origin of the chemical shift. Our results demonstrate that *GW* achieves an accuracy within 1.2% in XPS binding energies, by systematically restoring the underestimation from density-functional theory (DFT, error of 14%) or the overestimation from Hartree-Fock (HF, error of 4.7%). Such results also imply the correlations of *d* electrons are very well described by *GW*.

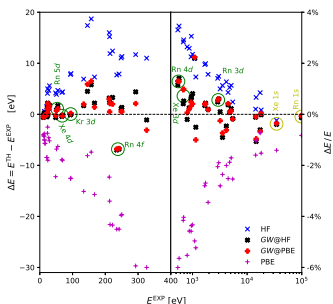


Figure 1: Noble gas atoms ZORA PBE, HF, and *GW* electronic energy absolute (left, 0 to 400 eV) and relative (right, from 400 to  $10^5$  eV) errors with respect to the experiment.

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## Photoinduced electronic and spin topological phases in monolayer single-element ferroelectrics

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Monolayer group-V materials exhibit rich physics such as single-element ferroelectricity, nontrivial topology, varied spin-orbit coupling, and light-induced structural dynamics. We show with *ab initio* calculations that light can induce hidden transient phases in both the ferroelectric and paraelectric monolayers. We illustrate the nontrivial topological character of these system by using the recently introduced idea of spin bands and spin-resolved Wilson loops, and show that the topology changes via the closing of the respective band gaps in the presence or absence of photoinduced structural phase transitions. The evolution of topological edge states is also discussed. Our study provides multiple strategies to tailor electronic and spin topology via ultrafast control of photoexcited carrier and structural dynamics.

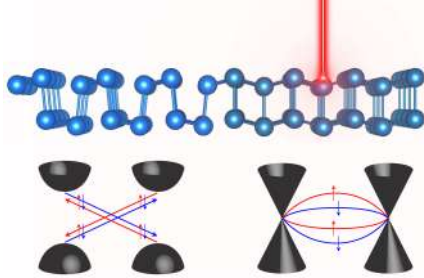


Figure 1: Photoinduced structural and topological phase transitions in monolayer bismuth.

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## Revealing fingerprints of valence excitons in x-ray absorption spectra with the Bethe-Salpeter equation

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The Bethe-Salpeter equation (BSE) is a powerful theoretical approach that is capable to accurately take electron-hole interactions into account. It significantly improves the optical absorption spectra of insulators and semiconductors. At the same time, the BSE can accurately treat x-ray absorption spectra (XAS), since excitonic effects can be also crucial for the description of core-excited states. We developed an ab initio framework to describe a pump-probe experiment, in which an x-ray pulse probes solid-state valence excitons by means of x-ray absorption spectroscopy. In this process that illustrated in Figure. 1, a pump pulse creates a valence exciton and an x-ray probe pulse induces a transition from a core orbital to a formed vacant electronic state in valence bands. We apply the BSE on top of GW approximation within a full-electron framework to calculate both valence-excited states and core-excited states. Our framework provides an accurate description of features in the x-ray absorption spectra related to an excited state. We illustrate our results with a calculation for a 4H-SiC semiconductor and demonstrate that x-ray absorption spectra carry orbital-specific information about valence excitons [1]. Our theoretical framework is of relevance for an accurate modeling of pump-probe experiments of photo-excited materials that utilize novel capabilities offered by x-ray science for probing matter.

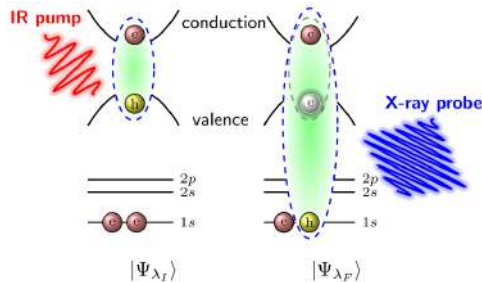


Figure 1: Scheme of the considered pump-probe experiment: first, a pump pulse creates a "valence exciton", where a red circle bounded by a yellow circle represents the created exciton. Second, an x-ray probe pulse creates a "core exciton" by inducing a transition of an electron from a core-shell to a vacancy in a valence band.

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## Modeling and investigating $K$ -edge spectra of single-wall carbon nanotubes

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Recently, Senga *et al.* measured  $K$ -edge electron energy loss spectroscopy spectra of individual single-wall carbon nanotubes (SWCNTs) [1]. In these experiments, each chirality is distinguished by a unique fine structure visible at the absorption edge. This fine structure was already observed in the diameter cumulative experiments of Kramberger *et al.* [2], where it was suggested that the fine structure is closely related to the Van Hove singularities (VHSs) seen in the ground state density of states (DOS). This relationship was used in Ref. [1] to explain the results for the (6,5) SWCNT by first superimposing an appropriately shifted DOS on the (6,5) spectrum and then identifying a peak in the fine structure with a particular VHS. In this work, we investigate this suggested relationship in detail. We model X-ray absorption spectra of SWCNTs using the state-of-the-art Bethe-Salpeter equation method, as recently implemented in VASP for X-ray spectra [3]. In addition, we use an implementation of the final-state approximation [4]. In all spectra we use a hybrid functional to correctly describe the orbitals. We first compare our modeled spectra with the experiments of Senga *et al.* [1] and show that we can reproduce the experimental fine structure in all cases. In particular, the results using the Bethe-Salpeter equation are in excellent agreement with the experimental results. We then contrast the modeled  $K$ -edge spectra with the initial state spectra of SWCNTs and show that there is no simple correlation between a VHS and a peak in the absorption edge. We study the excitonic properties of a graphene supercell and suggest that this discrepancy arises due to periodicity effects perpendicular to the nanotube axis.

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## Poster contributions

## Assessing Exchange-Correlation Functionals for Solids: Energy vs. Density Accuracy

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The accuracy of Kohn-Sham density functional theory (1) calculations relies on the performance of the exchange-correlation (XC) functional approximations. A recent critique (2) suggests that some of these approximations prioritize accurate energies over densities, contrary to the spirit of density functional theory. To investigate this issue for solid-state systems, we evaluate the performance of XC functionals from various rungs of Jacob's ladder in reproducing highly accurate energies, densities and XC potentials (3,4) for three prototypical solids: Si (semiconductor), NaCl (insulator), and Cu (metal).

By comparing the errors in predicted energies and densities, we identify several hybrid and semilocal functionals that consistently perform well across all material classes. Notably, functionals designed to satisfy exact constraints tend to be among the top performers, highlighting the importance of these constraints in functional development.

Our analysis reveals that, on average, functionals published up to the early 2000s simultaneously improve the prediction of both densities and energies. However, this trend is often not observed for more recent functionals, although errors in energy and density continue to evolve in a correlated manner.

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## DFT Prediction of quantum materials for spintronic applications

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### Abstract

It is well known that technological advances in spintronics are strongly linked to dilute magnetic semiconductor materials (DMS) and recently discovered semiconductor compounds. The main objective is to improve the Curie temperature (TC) above room temperature to extend the scope of spintronic applications. Theoretically, understanding the magnetic exchange mechanism can provide suggestions for exploring high-Tc materials. In all prototypical DMS materials of groups IIIIV and II-VI, such as Mn-doped GaAs or ZnO, the maximum TC remains limited to 180-185 K (1). To date, the magnetic mechanism is not fully understood. These difficulties could be overcome in the recently discovered I-II-V (2) and I-III-IV (3) family of semiconductors (SCs). In these SC families, charge and spin manipulation are independent.

In contrast, the Tc record reported for iron-doped III-V ferromagnetic semiconductors iron (Ga, Fe)-doped Sb semiconductors (4), reaching 340 K, has raised hopes for DMS projects. These materials have certain advantages over Mn-doped materials. In addition, the similarity between SC III-V GaSb and the I-II-V (NaZnAs) and I-III-IV (NaGaSi) semiconductors (3) gives us the idea of looking for an Iron-based DMS that can present ease of characterization with a relatively high Curie temperature. To achieve this goal, we will use DFT theory (5) to describe the electronic structure of the proposed DMSs.

**Keywords:** Semiconductors, DMS, Curie temperature, DFT

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## First principles approach to non-collinear magnetization dynamics

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The investigation of magnetization dynamics is a highly active field of research on both the experimental and the theoretical side. In our group, we focus on the computational description of condensed matter properties using the Korringa-Kohn-Rostoker (KKR) multiple scattering approach to Density Functional Theory (DFT)[1]. Central quantity of this technique is the KKR- Green's function, which is determined from a combination of single (atomic) site scattering quantities and expressions related to the finite or infinite structure. This clear separation, for instance, allows for the convenient investigation of disordered materials. Furthermore, the Green's function being this approach's central quantity, the framework can conveniently be extended to Linear Response Time Dependent DFT (LRTDDFT).

In the recent years, our group's main aim was the description of magnons, i.e. collective magnetic excitations, often referred to as spin waves, in collinear systems [2]. Our first-principles approach allows for the investigation of magnons and their lifetimes in collinear magnets, e.g. ferro or anti-ferromagnets on an equal footing. This capability is central for the interpretation of results from experimental techniques such as Spin Polarized Electron Energy Loss Spectroscopy (SPEELS) [3].

On my poster, I would like to present my current effort to extend our methods to non-collinear magnetic systems, i.e. those without a common spin quantization axis. The latter can either arise from competing exchange interactions or from relativistic effects. Investigating the dynamics of non-collinear magnets is of great interest as, in these systems, several magnetic degrees of freedom and charge density fluctuations can be coupled. This opens up new decay channels for the spin dynamics, which are absent in the collinear case. Additionally, the coupling of the spin and charge degrees of freedom is a step to paving the way to construct efficient interfaces between conventional electronics and spintronic devices.

So far, I generalized the our existing method to be able to self-consistently describe the ground state of non collinear systems. Furthermore, we are now able to treat Spin-Orbit Coupling in those systems. Currently, my focus lies on the generalization of the LRTDDFT scheme described above. My poster will consequently include methodological details, preliminary results and an overview over future tasks yet to be tackled.

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## Do we know the spectra of small aluminum clusters? Pitfalls and solutions in Real-Time TDDFT calculations.

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Aluminum has been studied as an unconventional material for plasmonics since some time [1]. The advantages of Al are its low cost as well as the fact that the surface plasmon for nanoparticles occurs far in the UV, unlike in the more traditional plasmonic materials like Ag and Au. A disadvantage is certainly the high sensitivity to oxidation which poses enormous problems for the experimental fabrication of small Al nanoparticles and clusters. In addition, optical measurements high in the UV are likewise very difficult.

However, also for theory the situation is difficult. Taking the presumably simple cluster Al<sub>13</sub> as an example, a brief review of the literature shows that while there are several optical spectra published in different contexts, they are entirely contradictory and do have no similarity between them, in some cases.

The reason can be traced back to the high energy of the plasmon, above the ionization energy, and to the fact that the transitions that contribute involve states which are largely part of the continuum, in particular for the anion.

In the present work we show how we can describe the system properly. To this end, we combine jellium calculations using the octopus code [2], where the calculation domain is finite, with calculations using the spherical jellium model where no box is imposed and, in addition, the polarizability can be expressed in terms of only the occupied states. This comparison allows us to determine the proper parameters for absorbing boundaries [3] which in turn produce correct spectra for all charge states, +1, neutral, and -1, of the Al<sub>13</sub> clusters. In turn, this allows us to calculate the size-dependence of the surface-plasmon resonances.

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## Quantum-mechanical investigation of photoinitiators for high resolution 3D printing

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The efficiency of 3D laser nanoprinting, specifically its resolution and speed, is intricately tied to the sensitivity of photoinitiators (PIs) utilised<sup>[1]</sup>. This investigation employs quantum-mechanical calculations, including density functional theory, time-dependent density functional theory and GW-BSE, to delve into the mechanisms of multiphoton absorption active PIs. Key examined PIs are Irgacure 651 and Irgacure 369 as Norrish type I, DETC and BBK as Norrish type II, benzil, biacetyl and tetramethylbenzil as “reluctant” Norrish type I. Norrish type I initiators produce radicals in the first triplet states, while Norrish type II initiators do so through a bimolecular reaction with a co-initiator, inducing hydrogen atom transfer. DETC and BBK in addition exhibit a mysterious three-photon radical generation mechanism when combined with pentaerythritol triacrylate, based on a three-photon process instead of the expected two-photon initiated pathway<sup>[1]</sup>. Conversely, two-step absorption PIs generate radicals akin to Norrish type I but from high triplet states. The research aims to uncover distinctions and similarities in efficiency and radical formation mechanisms triggered by multiphoton absorption, aligning theoretical insights with experimental data. The ultimate goal is to develop more efficient PIs, enhancing the performance of 3D laser nanoprinting in terms of speed and resolution.

Irgacure 651: 2,2-dimethoxy-2-phenylacetophenone; Irgacure 369: 2-benzyl-2-N,N-dimethyl- amino-1-(4-morpholinophenyl)-1-butanone  
 DETC: 7-diethylamino-3-thenoylcoumarin; BBK: ((2E,6E)-2,6-bis(4-(dimethylamino)benzylidene)-4-methylcyclohexanone

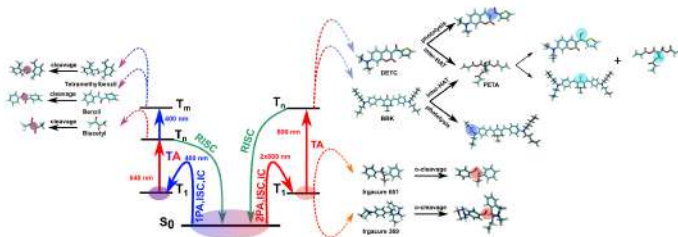


Figure: Radical formation mechanism of Norrish type I, type II and “reluctant” type I photoinitiators.

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## Exciton transport in 2D TMD

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Excitonic transport offers interesting features for logical gate [1], optoelectronic [1], quantum computing [2] applications. Recently, experiments have been carried out to demonstrate how to control this transport and in particular, 2D materials have emerged as a versatile platform to make this new type of devices possible using various means (electric field, strain, coupling with other particles). Promising results have been obtained: diffusion length up to  $60\mu\text{m}$ , diffusion speed up to  $1.8 \times 10^7\text{ms}^{-1}$  and unidirectional propagation [1]. On the other hand, exciton structure bands have been calculated in the literature using many-body Bethe-Salpeter approach [3]. These data have thus allowed empirical models of exciton properties.

In a first part, I will propose an overview of different techniques employed to control exciton transport in 2D layers, and then introduce the non-equilibrium Green's function we develop to unravel aspects of this controllability.

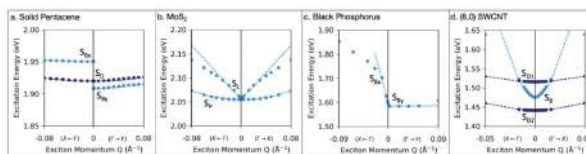


Figure: Exciton band structure extracted from [3].

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## A Parameter-Free Approach for Modeling Valence and Core-Level Photoelectron Spectroscopy

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Core-level spectroscopy provides valuable information about the local chemical environment of atoms in molecules by probing core-electronic structure whereas valence-level spectroscopy offers valuable insight into hybridization and bonding via valence-electronic structure. Despite their similarity, modeling core-electronic structure is challenging owing to large orbital-relaxation effects and relativistic corrections. We overcome these challenges by combining the generalized Kohn–Sham semicanonical projected random phase approximation (GKS-spRPA) method with the spin-free exact two-component theory in its one-electron variant (SFX2C-1e) followed by a perturbative treatment of spin-orbit coupling (SOC) to model the K- and L-edge X-ray photoelectron spectroscopy (XPS), valence-level PES and non-resonant X-ray emission spectroscopy (XES) of molecular systems. The core and valence-electron one-particle states, required for the computation of the XES spectra, are obtained directly in a single calculation of the neutral system without any use of core-hole reference states. A comprehensive analysis demonstrates that the X2C-GKS-spRPA method achieves an accuracy of approximately 0.2 eV for valence-level PES and XES, while mean absolute errors (MAEs) of less than 1 eV are observed for core K-edge and L-edge XPS of third-period elements. We also show that an analytic continuation technique, with a  $\mathcal{O}(N^4)$  computational cost, can be used to obtain highly accurate X-ray emission spectra of molecules such as  $C_{60}$  and  $S_8$  with multiple core-hole states.

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## Plasmon satellites in Cesium

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Photoemission spectroscopy is one of the most direct ways to access the electronic structure of materials. The main peaks usually correspond to the quasiparticle band structure. Replicas of these peaks, called satellites, are entirely due to interactions [1]. Here, we address satellites that can be interpreted in an electron-boson coupling picture, where the bosons are charge excitations such as plasmons and electron-hole pairs that are induced by the removal of an electron from the sample. In the homogeneous electron gas (HEG) at a density corresponding to a simple metal like sodium, plasmons are the prominent collective excitations [2]. At lower densities, instead, the HEG exhibit exotic features: the plasmon dispersion switches sign becoming negative and additional excitonic collective modes have been recently predicted [3,4]. It is, therefore, very natural to ask whether such signatures can be seen in real materials and ponder about their impact on the shape of photoemission satellites. Cesium is a metal with low electron density that shows a negative plasmon dispersion [5]. In this study, we investigate the effect of such dispersive nature on the shape of the satellites in the photoemission spectra of cesium using Time-Dependent Density Functional Theory and Many-Body Perturbation Theory, where we combine the GW approximation for the self-energy with the cumulant expansion for the Green's function [1]. Our calculations provide guidance for ongoing experiments.

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## Quantum Topology Study for Unveiling the Charge Transfer Excitation in the Complex of Ethylene and Tetrafluoroethylene

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In this poster, we report the findings of a study conducted on the complex of ethylene and tetrafluoroethylene which serves as a model system for studying features of charge transfer (CT) excited states. The CT excited state is described well as a single-electron excitation between specific orbitals of donor and acceptor. Using this pilot study, we try to create a systematic application of Quantum Chemistry Topology (QTP) and its adaptation to CT excited states. Understanding of QTP of excited states in ethylene and tetrafluoroethylene shows how CT occurs within this complex and can shed light on the fundamental principles of electron movement in molecules after excitation. In general, by unraveling the behavior of electrons in excited states, we could unlock new possibilities for the development of functional materials and advance our knowledge of electronic properties. In particular, CT excitation is a cardinal phenomenon in many areas of science like photochemistry, photovoltaics, and molecular conductance.

- The calculations are done using the Gaussian 16 package[1]. We first optimized the structures of ethylene and tetrafluoroethylene. We took the framework of hybrid **Density Functional Theory** by using CAM-B3LYP[2] functional and the aug-cc-pVTZ basis set[3].
- For calculating excited states we used Time-Dependent (TD) - DFT with CAM-B3LYP functional. Over the past few years, TDDFT has advanced to one of the most popular theoretical approaches to calculating excited state properties of medium-sized and large molecules up to about 200 atoms.
- The natural transition orbital (NTO) is calculated, and the transition density matrix (TDM) is plotted using Theodore [4] by processing Gaussian outputs for excited states. For calculation, Theodore considers a fragment-based excited state analysis within a correlated electron-hole picture.
- To evaluate the electron density trends between ground and excited states, we consider the one-electron charge density[5]. We produce the contour by calculating the laplacian of these densities obtained from quantum calculations. The visualization of the charge density is done by Aimall[6].

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## Excitons in Helium Under Pressure

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The absorption and the electron energy loss spectra of solid helium at different pressures are calculated using a first principles approach based on the Bethe-Salpeter equation of many-body perturbation theory. The results are used to explain the effect of pressure on HCP helium that has been measured by spectroscopy experiments (1,2). We found a well defined exciton peak, showing linear pressure dependence, both in the peak position and intensity. We calculated the excitonic dispersion of the loss function along the  $\Gamma$ -M direction for momentum transfers beyond the first Brillouin zone. We found strong parabolic-like dispersion of the spectra, dominated by the screened Coulomb interaction. Near Bragg reflection points we found an anomalous angular dependence of the exciton peak, due to the crystal local field effects, analogously to the plasmon in graphite (3).

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## Orbital Kerr Effect and Terahertz Detection via the Second Order Hall Effect

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Quantum transport in periodic crystals offers a wide variety of applications in nanoelectronics and paves the way for the characterization of quantum materials. At the first order in the electric field, the anomalous Hall effect is forbidden in non-magnetic materials, but it is allowed when the system has a non-centrosymmetric structure. The leading order contribution is proportional to the Berry curvature dipole [1], and the corresponding current is a companion phenomenon of an electric-induced magnetization driven by the orbital moment [2]. Since both effects do not require spin-orbit coupling, the nonlinear Hall effect and the orbital Edelstein effect are alternatives to using lighter materials that are also cheaper and more abundant.

Motivated by a previous experiment on MoS<sub>2</sub> monolayer under strain [3], we show that a direct connection between the Berry curvature dipole and the orbital Edelstein effect is only possible in gapped Dirac materials. In addition, we reveal the connection between the nonlinear current and the current-induced orbital magnetization through the magneto-optical Kerr effect at the second order in the electric field [4], which we tagged as the orbital Kerr effect [5]. Remarkably, the Kerr angle obtained is proportional to the terahertz responsivity, which accounts for the efficiency of a rectifier to detect the incoming light radiation. We compare the Kerr efficiency of several tight-binding models and DFT simulations in this regime with previous experiments reported in the literature, and we conclude that the orbital Kerr effect can be considered as a hallmark of the orbital accumulation in light materials.

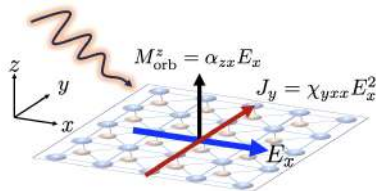


Figure 1: Orbital Kerr effect in a 2D non-magnetic material with inversion symmetry breaking.

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## Metal-TetraPhenyl-Porphyrins Adsorption on Atom-Thick and Bulk Magnetic Oxides

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Metal tetraphenyl porphyrins (M-TPP) can host different metal ions within their tetra-pyrrole ring, providing exceptional flexibility in the molecular properties and functions. Molecular layers of M-TPP typically self-assemble on surfaces forming square-like superlattices of planar molecules with a high degree of structural order. That makes them an ideal benchmark for the study of their properties by surface science techniques and facilitates applications where well-defined hybrid interfaces are requested. Additionally, these ordered layers can pave the ground to the subsequent growth of 3D molecular architectures. Among supporting surfaces, oxides and oxidized layers raise interest because of their intrinsic properties (e.g., as antiferromagnetic substrates for organic spintronics [1]), or as protective layers.

We have combined experimental techniques and *ab initio* methods based on density functional theory (DFT) to investigate Co-, Ni-, Zn-, and VO-TPP molecules adsorbed on an atom-thick oxide [namely Fe(001)-p(1x1)O] where the oxide layer is able to preserve the characteristic features of quasi-free molecules as we evidence by UV-photoemission/inverse photoemission spectroscopy and calculations. [2] Adsorption structures are characterized by well ordered 5x5 superlattices that can attain different orientations depending on the central atom, despite a moderately-dependent computed energetics points to a subtle influence.[3] DFT analysis also points to a dominant antiferromagnetic coupling between the molecular spin and the substrate one. VO-TPP, which further offer an additional degree of freedom through the V=O bond orientation, also form square ordered superstructures with both upwards and downwards cases possible.[4] The interaction of a selected case (Co-TPP) at the surface of antiferromagnetic NiO substrate is then studied as a prototypical case for perspective antiferromagnetic spintronics applications.

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### Alkyl group-induced enhanced ultrafast charge migration in pyrrole-based structures

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Pyrrole and derivatives are essential constituents of organic molecules. By investigating their ultrafast response to sudden ionization, we gain insights into their photoreactions and photoresponsivity [1-3]. We examine the effect of alkyl substitution on the orbital characteristics, ionization spectrum, and charge migration dynamics preceding the onset of nuclear motion in pyrrole, N-methylpyrrole, and N-ethylpyrrole by non-Dyson ADC(3) approach [3-4]. Our results demonstrate a significant charge transfer between the pyrrolyl ring and the alkyl group upon ionization from the HOMO-4, HOMO-2 and HOMO-3 orbitals respectively. The characteristics of the involved orbitals remain consistent after alkylation. The alkylated molecules exhibit significant ultrafast charge transfer between the alkyl group and the pyrrolyl ring upon ionization, and the extension of the alkyl chain increases the exchanged integrated charge between the two parts from about 0.3e to 0.5e. These results can find significant applications in the form of chemically tuned ultrafast charge migration through alkyl-group effects.

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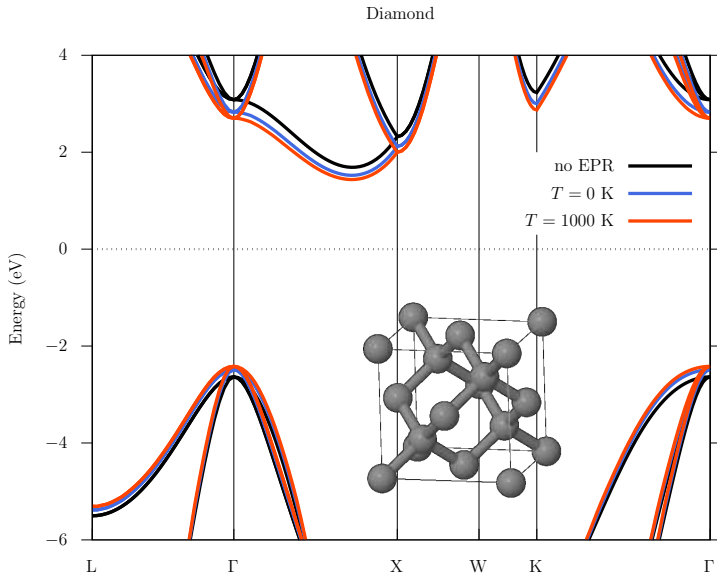
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## Electron-phonon interaction using Gaussian orbitals: Silicon and Diamond

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Electron-phonon interaction is a crucial mechanism in solid state physics that is responsible for a multitude of phenomena. However, in electronic structure calculations it is often neglected. We developed an ab-initio implementation on top of density-functional theory that combines finite differences calculations with the perturbative Allen-Heine-Cardona framework in order to calculate the temperature-dependent electron-phonon renormalization (EPR) of the electronic bandstructure using a basis set of localized Gaussian orbitals.



Our implementation circumvents the limiting problems of previous implementations while maintaining a good agreement with the literature. For the direct band gaps of silicon and diamond we find a zero-point renormalization of  $-45$  meV and  $-407$  meV, respectively. In addition, the approach allows to evaluate Debye-Waller contributions beyond the rigid-ion approximation.

## How to calculate the optical properties of a non-infinite material within time-dependent density functional theory in reciprocal space ?

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For 3D crystals, the longitudinal formalism of the time-dependent density functional theory can be used to calculate the absorption spectrum, which is the response to a transverse field. It relies on (i) the Ehrenreich result that the transverse contraction of the macroscopic dielectric tensor is equal to the longitudinal one in the optical limit [1], and (ii) the Adler and Wiser formula [2] that relates the longitudinal contraction of the macroscopic dielectric tensor to the macroscopic average of the inverse dielectric function.

For an isolated 2D object, we use of a supercell to isolate the object from its replicas, and allow the reorganisation of the electronic states at the interface. The 3D formalism fails. The macroscopic average leads to the effective medium theory with vacuum [3]. Cutoff potentials [4] have been proposed to reduce the size of the calculation, but they still present a vacuum dependence and prevent to access to the perpendicular component. The Selected-G method [3] cures these problems, by removing the vacuum when solving the Dyson equation.

One of the key point of this framework is the thickness of the matter  $L_z^{mat}$ . Its influence on the out-of-plane component, calculated from the response function to the macroscopic total potential with the slab potential, reveals a complete change in the nature of the object. By considering a value deduced from the atomic positions, we get for the macroscopic dielectric function the properties that we expected from a semi-infinite object. For a 2D object, the extension in real space of the Kohn-Sham response function is much larger than the atomic positions leading to include the interfaces in the definition of the thickness of the quasi-2D object. It results that the longitudinal contraction of the macroscopic dielectric tensor is no more equal to the transverse one. The response function to the macroscopic total potential is a linear combination of the absorption and the plasmon spectra, exhibiting the same features as the reflectance or the transmittance of an electromagnetic field impinging an ultra-thin layer [5-6].

Concerning the case of the surface, we expect a clear distinction between the response functions to the macroscopic total potential and to the external potential. Nevertheless, the object introduced in the supercell is very thin compared to what it is necessary to depict a semi-infinite system, and far from the limit  $L_z^{mat} \rightarrow \infty$ , where the slab potential tends to the 3D Coulomb potential. Moreover, the interaction between replicas still occurs. We explain the reasons why one must couple the 3D potential and the no-interface limit within the Selected-G formalism to mimic the semi-infinite object.

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## Exploring Two-Photon Absorption in Metal-Organic Frameworks: A Computational Study

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A metal-organic framework (MOF) is a material comprised of metal atoms or metallic aggregates, e.g. metal-oxo nodes, with organic linkers that form crystalline networks. Owing to the diversity of the components, a large number of MOFs has been synthesized (> 70.000) and even a higher number has been reported computationally [1]. So far, due to their big pore size and their ability to interact with other molecules, MOFs have been used mainly for gas adsorption, separation, catalysis, gas storage, etc. [2,3]. Recently, MOFs have emerged in new application areas exploiting their electronic and optical properties [4] including multiphoton absorption (MPA). The photoactive linkers in MOFs can exhibit optical nonlinearities when interacting with an intense laser field [5–8]. Materials demonstrating high nonlinear optical effects are important in numerous advanced applications, analytical and medical techniques, high-capacity telecommunications, micromachining, and super-resolution microscopy [9]. In this project, the Two-Photon Absorption (TPA) response of different MOF structures are studied computationally in order to understand the mechanisms behind the increase in the 2PA response. A deeper understanding of these mechanisms is essential for designing new MOFs with enhanced 2PA performance.

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## Excited state photoemission momentum mapping of organic molecules from time-dependent density functional theory

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Angle-resolved photoemission spectroscopy is a powerful experimental technique to reveal the electronic structure of thin films and interfaces. In particular for organic molecular layers, constant binding energy angular distributions, so-called momentum maps, have been shown to be related to the Fourier transform of the electron-emitting initial state molecular orbital. This connection bridges experiment and theory in photoemission orbital tomography (POT). The experimental extension of POT to optically excited states has, of late, been demonstrated via femtosecond pump-probe spectroscopy [1], paving the way for tracing the momentum distribution of electrons on ultrafast time scales. However, linking the measured momentum maps to the spatial structure of the exciton wave functions has yet to be achieved. Recently, we have proposed a simple procedure which involves a coherent sum of the ground-state Kohn-Sham orbitals that construct the exciton wave function [2].

Here, we validate this approach by solving Casida's equation for a series of organic molecules in gas phase leading to optical absorption spectra and exciton compositions. The so-obtained momentum maps are then cross-checked by directly simulating angle-resolved photoemission spectra within the framework of real-time TDDFT. To this end, we record the flux of the emitted electrons through a detector surface using the so-called t-SURFF method [3]. The comparison of the resulting momentum map features confirms, that the excited molecular states correspond to a superposition of several single-particle excitations.

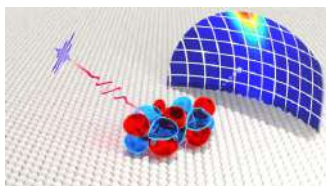


Figure 1: Scheme of the simulated angle-resolved photoemission spectroscopy experiment (© Markus A. Huber, University of Regensburg).

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## Momentum-space signatures of charge-transfer excitations in donor-acceptor dimers from GW/BSE calculations

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This contribution is part of the ongoing “Orbital Cinema” project [1], in which the time- and momentum-resolved analysis of charge-transfer (CT) excitations as measured by time- and angle-resolved photoemission spectroscopy (trARPES) shall be investigated. A particular system of interest is a bilayer of the donor/acceptor molecules H2TPP and PTCDA adsorbed on Cu[1x2](100)-2O. This system is a promising candidate for trARPES measurements of CT excitations, because i) the passivated surface allows for a longer exciton lifetime, making the measurement of excitonic properties more feasible, ii) PTCDA orders well on the Cu[1x2](100)-2O surface, iii) H2TPP is known to order well on many substrates and iv) the energy alignment of H2TPP and PTCDA (gas-phase) KS-energies makes them a promising candidate for an interlayer CT exciton (with electron transfer from the H2TPP to the PTCDA layer).

On the theoretical side, long-range electron-hole interactions in charge-transfer excitations pose a challenge to the ab-initio treatment of such systems, where local or semi-local TDDFT simulations have often turned out to be problematic. Here, the GW/BSE formalism has been shown successful in calculating electronic properties of CT excitations and is therefore the method of choice in this study.

Using the Fiesta code [2], in this contribution we present gas-phase calculations of PTCDA-H2TPP dimers. As expected, the first exciton is found to be a CT exciton (Figure 1), however, it has a very low oscillator strength. A possible excitation-pathway suggested by our results could be the excitation of a Frenkel exciton within the PTCDA layer, which then cascades down to the CT exciton as the exciton’s hole “hops” from the PTCDA to the H2TPP. Using a formalism developed by us [3], we make predictions how this process shows up in trARPES experiments. In particular, we expect the PTCDA Frenkel-exciton and the PTCDA-H2TPP CT-exciton to show very similar photoelectron momentum-patterns (Figure 2), but appearing at different kinetic energies. This shift in kinetic energies is expected to be observable in a time-resolved measurement, as the system cascades from the higher-lying excited state to the low-lying CT excited state.

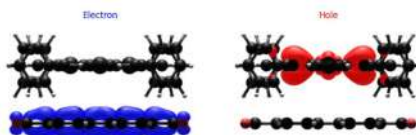


Figure 1: Electron- and hole-density of the CT exciton.

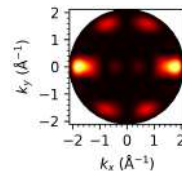


Figure 2: Dominant momentum pattern of the CT exciton.

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## Predicting Novel 2D AsBiX<sub>3</sub> (X = S, Se, Te) Auxetic Monolayers with Favorable Optical and Photocatalytic Water Splitting Properties.

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### ABSTRACT:

The design of two-dimensional multifunctional materials is highly desirable for nanoscale device applications. In this study, we report the structural, electronic, mechanical, and photocatalytic properties of chalcogenide-based monolayers AsBiX<sub>3</sub> (X = S, Se, and Te) using first-principles calculations. The stability of these monolayers is confirmed through energetic and mechanical analyses, as well as ab initio molecular dynamics simulations. The analysis of mechanical properties reveals significant mechanical anisotropy and a bidirectional in-plane negative Poisson ratio in the monolayers. Additionally, the computed electronic band structures, obtained with and without spin-orbit coupling, indicate that these monolayers are indirect-gap semiconductors. At the Heyd-Scuseria-Ernzerhof level, the values of the band gap are determined to be 1.91 eV for AsBiS<sub>3</sub>, 1.66 eV for AsBiSe<sub>3</sub>, and 1.32 eV for AsBiTe<sub>3</sub>. These monolayers have a very high absorbance on the order of  $\sim 5 \times 10^5 \text{ cm}^{-1}$  in the visible and ultraviolet regions with considerable anisotropy. We also found that monolayers hold a high mobility anisotropy. The predicted solar-to-hydrogen efficiency of all monolayers surpasses the critical value (>10%) for the economical production of hydrogen from photocatalytic water splitting. Notably, AsBiS<sub>3</sub> and AsBiSe<sub>3</sub> monolayers have appropriate band-edge positions that perfectly match the conditions for photocatalytic water splitting at pH = 0, and the band gap and band-edge positions can be adjusted through strain engineering. With these outstanding properties, AsBiX<sub>3</sub> (X = S, Se, and Te) monolayers present themselves as promising candidates for applications in optoelectronics, mechanics, and photocatalytic water splitting.

**KEYWORDS:** first-principles calculations, 2D materials, optical absorption, carrier mobility, auxetic materials, photocatalytic water splitting



## Spin spirals from linear-response theory: the case of NiBr<sub>2</sub>

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Low-symmetric magnetic ground states are among many of the exotic properties of low-dimensional materials. In the case of NiBr<sub>2</sub>, a two dimensional transition metal halide that shows a spin-spiral as its magnetic ground state [1], first principles calculations within the GW+BSE have been applied to study its excitonic properties in the spin-conserving channel [2]. In this work, we extend the exploration to finite-momentum excitonic properties in the spin-flip channel and compare results at three levels of theory: GW+BSE, TDDFT and the generalised Bloch theorem.

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## Controlling High Harmonic Generation by Strain Engineering in Tunable Black Phosphorene

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Two-dimensional (2D) phosphorene attracted a lot of attention due to its unique properties such as widely tunable bandgap, high carrier mobility, and anisotropic photoelectronic response [1]. We show that strong field processes in such systems can be optimized and controlled by strain engineering. Using ab-initio time-dependent density functional theory (TDDFT), we investigate ultrafast electron dynamics and high harmonic generation (HHG) subject to band structures engineering under controlled biaxial tensile (+ve) and compressive (-ve) strains  $\epsilon \in [-10\%, 2\%]$ . While  $\epsilon = -10\%$  resulted in closing of band gap,  $\epsilon = 2\%$  increased the gap by 22% (w.r.t. 0.9 eV in pristine phosphorene).

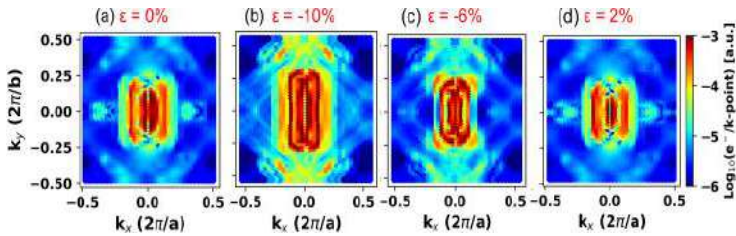


Figure: Sub-cycle dynamics of momentum-resolved excited electrons near the minimum of vector potential for (a) pristine, (b) -10%, (c) -6%, and (d) 2% strained phosphorene.

Due to its intrinsic in-plane anisotropy, harmonic yield with laser polarization along the armchair (AC) direction or  $x$ -direction is higher than that of the zigzag (ZZ) or  $y$ -direction. With reduction of band-gap, from 2% to -10% strain, the valence band near  $\Gamma$ -point becomes more flat and discreet, resulting in large electronic density of states (DOS) and enhanced electronic excitation, which reflects in their ultrafast sub-cycle dynamics under laser excitation. The areas near the  $\Gamma$ -point contains highest excited electrons density due to high DOS near the  $\Gamma$ -point.  $\epsilon = -10\%$  induces larger DOS and thus yields more excited electrons compared to other cases which eventually contributes to the HHG. Nearly, an order of magnitude enhancement of Harmonic intensity is achieved for  $\epsilon = -10\%$  along AC direction.

The current study opens up the possibility of strain induced HHG control with applications in attosecond nanophotonics and strong-field driven ultrafast electronics in 2D materials [2].

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**Phonons, quasi-phonons and *semi-classical swings*:  
insight from exactly solvable models**

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Despite the phonon concept is one of the most widely used, studied, reviewed[1] concept in modern condensed matter, some, even basic, properties are still debated. In this talk I will discuss, trying to shed light on, some of the main open questions.

Density-Functional Perturbation theory (DFPT) is routinely[2,3] used to calculate non-adiabatic properties, including phonon widths. Even if DFPT is a semi-classical theory it is widely accepted that can provide the same kind of information of a fully quantistic theory. I will start my talk by demonstrating that this well fact is, actually, based on a faulty solution of the linearized Ehrenfest dynamics[4]. In particular I will demonstrate how a decaying semi-classical phonon oscillation corresponds to an energy violating and, thus nonphysical, solution of the linearized atomic real-time dynamics.

I will, then, show that the exact time-dependent DFPT phonon energy is real and always over-estimates the real frequency, obtained as solution of the full Many-Body problem. This difference induces a gap between the quantistic and classical phonon energies that increases with the energy indetermination.

The conclusion will be that non-adiabatic effects lead to the breakdown of the DFPT description and imposing the use of a fully quantistic approach.

After this crucial premise I will move to the discussion of the screening of the electron-phonon vertex. This is, indeed, commonly assumed to be well described by a static screening. This basic property has made possible to link the *Ab-Initio* approach to the physic of model Hamiltonians, like Fröhlich's.

I will approach the screening of the electron-phonon vertex by using both the equilibrium[5] and out-of-equilibrium (via the Langreth theorem). By using the second I will show how, the equilibrium limit, a fully symmetric, doubly *dynamically* screened, form of the phonon self-energy can be, indeed, introduced[6].

I will then consider an exactly solvable three-dimensional homogeneous electron gas. I will show that by approximating the dynamical with a static screening large deviations from the exact solution appear. I will conclude the talk by discussing the variational properties of the self-energy and by introducing an effective local-field correction  $\Lambda_q(\omega)$  that can, approximatively, amend a statically screened phonon self-energy[6].

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## Theoretical investigation of interband and plasmonic excitations in mono-metallic In and bi-metallic AgIn nanoclusters

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Nanomaterials exhibit unique optical properties with applications in various fields such as nonlinear and quantum optics, solar cells, and photocatalysis. In particular, the localized surface-plasmon resonance (LSPR) in the UV range plays a central role in understanding how these materials respond to external electromagnetic perturbations [1]. Nevertheless, the complex nature of these materials, which is characterized by their high reactivity, aggregation, and instability, can pose challenges for both their efficient characterization and utilization [2]. In this regard, classical (electrodynamics Mie model) and *ab initio* simulations offer a powerful framework for exploring the properties of these materials. By evaluating the interplay of size, composition, and environment, which are reflected in spectral features such as the LSPR, these simulations can shed light on the physical-chemical properties of these systems [3].

Current advancements in nanoplasmonics are focused on exploring cost-effective alternatives to traditional plasmonic materials such as silver and gold. In a recent study [2], our collaborators demonstrated the potential of combining noble metals with trivalent metals, such as indium, to achieve a blue shift in the plasmonic feature. This shift is desirable for the development of plasmonics in the UV range. Theoretical simulations are expected to provide further insights into the optical properties of such bimetallic nanoparticles.

In this work, we use the Yabana-Bertsch time-evolution formalism (RT-TD-DFT) implemented in the real-space code *Octopus* [4] to investigate the electronic structure of model systems comprising mono-metallic In and bi-metallic AgIn nanoparticles with varying sizes and compositions, as well as their optical properties. We are specifically interested in evaluating how shifts in their plasmonic features in the UV region are related to the metallic composition.

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## Precise optical absorption spectra of Ag clusters and nanoparticles from efficient RT-TDDFT+U calculation

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The modeling of the Localized Surface-Plasmon Resonance (LSPR) in silver clusters and nanoparticles depends on exchange-correlation functionals (xc-functionals) within Density Functional Theory (DFT) and Time-Dependent DFT (TDDFT). While optical absorption spectra calculated using hybrid and range-separated hybrid functionals are known to have excellent agreement with experiments [1,2] their computational cost restricts their use to smaller clusters. Simple functionals such as LDA or GGA fail to describe the 4d electrons accurately, generally placing them too close to the Fermi energy. Consequently, the inter-band transitions start at lower energies than expected, leading to too low LSPR energies and over-broadening of the LSPR peak. RT-TDDFT+U emerges as a promising alternative [3,4], which improves the description of the 4d electrons by adding an empirical Hubbard-like effective U correction to the Kohn-Sham Hamiltonian. In our present work, we carried out RT-TDDFT+U calculations using the same value of the effective U parameter that has been found to provide good results in bulk silver [5]. Both the electron-hole-type discrete spectra of the smallest clusters and the broad plasmon resonances of the larger sizes ( $\approx 1000$  atoms) are precisely obtained. The excellent agreement with experiment for all sizes indicates that the U parameter is surprisingly transferable.

Our calculations successfully capture the oscillations in the LSPR position caused by the shell-closing effect for intermediate-sized clusters. However, calculations performed using highly symmetric icosahedral clusters, spherical fcc-based clusters, and geometrically optimized lowest-energy clusters of similar sizes reveal that the geometry and the overall shape of the particle are important for the energetic position of the LSPR. Furthermore, we qualitatively and quantitatively confirm the increased localization of the 4d electrons when the U correction is applied. The average occupation of the 4d orbitals reveals a stronger Coulombic screening effect in larger particles compared to smaller ones, as well as reduced screening on the surface compared to inside the particle.

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## Application of strain on non-trivial topological transitions in organic materials

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In this work, we present the effect of strain on the non-trivial topological transition of one-dimensional polymers with two bridges. As shown in Ref. [1], the controllable topological phase transitions in polymers allow for unconventional optical selection rules and exciton series. In the present study, we consider this effect as a function of strain and take one step further by investigating the impact of mechanical strain on optical absorption. Our work opens up a new way for the design of novel optoelectronic devices by applying a minimum value of strain.

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## Charge density as a functional of the potential: Connector Theory approach

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The first widely successful formulation of the exchange-correlation effects within Density Functional Theory in real materials was based on the Local Density Approximation (LDA) which uses the results of the homogeneous electron gas model to describe the real material at hand. Motivated by this idea, we present results of a general and in principle exact theory called the connector theory (COT) which formulates a systematic way to simulate a real system for a given model by making use of the “connector” which is a mathematical object bridging the model and real system. We show that by using a connector we are able to create accurate functionals for describing certain observables, without diagonalizing the Hamiltonian. Further, we recall that the connector theory provides a spectrum between locality and non-locality in which the connector acts as a tuning parameter, and LDA is just one special case of this theory (Vanzini et al., 2022). We focus our study on calculating the charge density as a functional of a given mean-field potential within the framework of COT. Our results successfully capture the correct behavior of the charge density in cubic He with almost no computational cost. We further compare local and bilocal connector results, and propose new systematic ways to implement these ideas in other materials.

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PS Core level chemical shift by *ab initio* methods: from mean-field to many-body theoI. Mukatayev<sup>a,b,c</sup>, F. Moevus<sup>b</sup>, B. Sklénard<sup>a,b,c</sup>, V. Olevano<sup>a,b,d</sup> and J. Li<sup>a,b,c</sup><sup>a</sup> ETSF, European Theoretical Spectroscopy Facility, France  
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The binding energy of core electrons may not only provide information on the chemical composition, but also some additional information, such as the type of bonding, which could be inferred from the shift of the binding energy (also known as chemical shift). Here we will present the study of the chemical shift using different theories, from mean-field (DFT, HF) Hartree-Fock and density-functional theory, to many-body perturbation theory (COHSEX, *GW*). We benchmarked the accuracy of the chemical shift of carbon 1s electron in a set of molecules against experiments. Besides, our study reveals the physical origin of the chemical shift.

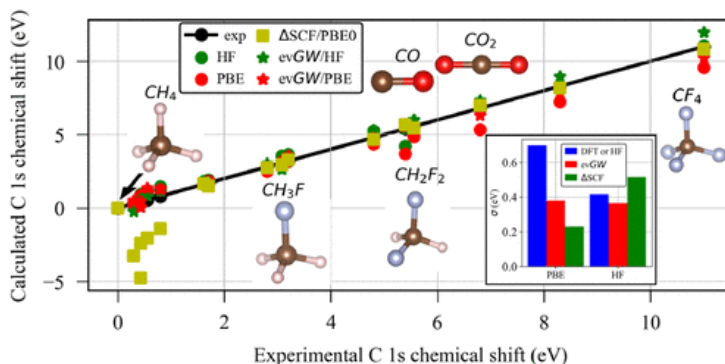


Figure 1: Chemical shift of the carbon 1s level calculated by HF, DFT-PBE, ΔSCF in hybrid PBE0, and *GW* on top of HF and DFT, as compared to the experiment.

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## Optical Properties of Squaraine Molecules and Aggregates from Density Functional Approaches and Many-Body Perturbation Theory

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Squaraine dyes (SQs) are known for their unique molecular structure, characterized by a donor-acceptor-donor arrangement, which enables strong intramolecular charge transfer and extended  $\pi$ -electron systems. These properties result in remarkable optical and electronic characteristics, including high absorption and emission coefficients in the visible-near infrared (NIR) range, as well as notable thermal and chemical stability. SQs have found diverse applications in fields such as photovoltaics, photosensors, fluorescence probes, and biomedical agents. However, understanding their optical behavior at the molecular level is essential for maximizing their potential in various technologies. Despite their importance, SQs pose challenges for theoretical description due to their zwitterionic electronic ground state. This complexity makes density functional based methods insufficient for accurately predicting some of their optically excited state properties<sup>1</sup>.

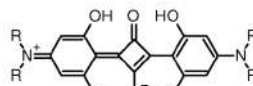


Figure : Molecular structure of anilino squaraines, where R denotes the variable terminal groups

In this contribution, we present a careful study of the ground state and excited state properties of single SQ molecules as well as cluster of SQ aggregates in the gas phase. For this, we employ ground state and time-dependent density functional theory using the ORCA code<sup>2</sup>. In particular, we put special emphasis on the influence of the exchange-correlation (xc) functional on the prediction of optical absorption spectra of SQs. Different classes of xc-functionals are compared, ranging from the generalized gradient approximation (GGA) and popular hybrid functionals to long-range corrected range-separated functionals such as the CAM-B3LYP<sup>3</sup>. Moreover, we also explore the performance of optimally-tuned range-separated hybrid functionals. As an alternative route to the excited state properties, we perform GW/BSE calculations within the framework of many-body perturbation theory using the FIESTA code<sup>4</sup>. We assess the role of the DFT starting point and also present tests on how the Tamm-Dancoff approximation affects the results, both, on the TDDFT as well as on the BSE level of theories. Impressively, our high-level theoretical approach (CAM-B3LYP  $\rightarrow$  GW/BSE) yields reasonable agreement with experimental absorption data for tetramer SQs molecules. These analysis using GW-BSE, standard TDDFT (B3LYP), and CAM-B3LYP offers insights into SQ molecules' optical and electronic properties, clarifying computational method strengths and limitations for enhanced SQ-based material design and utilization.

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## High-throughput DFT prediction of new possible halide superconductors

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### Abstract

Hydrides in numerous superconductors, this study focuses on experimental validation, specifically confirming that compressed sulfur hydride H<sub>3</sub>S exhibits a TC of 203 K at pressures approximately 150 GPa. The overarching objectives are twofold: firstly, to unravel the microscopic mechanisms governing superconducting hydride materials; and secondly, to contribute to the exploration of novel high-pressure superconducting hydrides. To achieve these goals, we employ a first-principles method based on Density Functional Theory (DFT), enabling the prediction of electronic, chemical, and phononic properties of hydride materials. The quest for high-temperature superconductors has led to the investigation of halides, especially those combining light elements like hydrogen and fluorine with heavier elements such as La. The discovery of superconductivity in hydrogen sulfide (H<sub>2</sub>S) under high pressure sparked interest in exploring halides for their potential as high-T<sub>c</sub> superconductors. Notably, LaH<sub>10</sub> stands out as the highest T<sub>c</sub> superconductor in this category, reigniting the exploration of halides for new high-T<sub>c</sub> superconductors. Despite the excitement surrounding LaH<sub>10</sub>, understanding the superconductivity mechanism in LaH<sub>10</sub> and other halide-based superconductors remains an active and open area of research, with numerous unanswered questions. The discovery of LaH<sub>10</sub>, however, has paved the way for exciting avenues in investigating halides and their potential as high-T<sub>c</sub> superconductors.

In this study, we employ electronic structure prediction techniques to explore new halide superconductors. Additionally, machine learning serves as a valuable tool for validating and discovering new predicted materials, enhancing the efficiency of material discovery in the pursuit of advancing high-temperature superconductors.

**Keywords:** Electronic structure, superconductivity, Electron-phonon coupling, DFT, GGA

## Electronic properties of 2D Transition Metal Dichalcogenides ternary alloys: an ab initio study

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First principles calculations are performed to investigate the properties of two-dimensional (2D) transition metal dichalcogenides (TMDs) ternary alloys of the form MXY, with M = Mo, W and X ≠ Y = S, Se. We are particularly investigating the influence of the composition of these alloys on the evolution of the band gap, and the role of the internal electric field orientation on the electronic properties of these atomically thin 2D materials. To have a better understanding of the composition dependent, the thermodynamic stability, structural and electronic properties of these alloy configurations at various concentrations have been comparatively studied using the density functional theory (DFT). We found that the introduction of the foreign Y atom affects the lattice constants of the 2D systems, in agreement with literature [1]. From bandstructure analysis, we saw that the introduction of the Y chalcogen fine tune the band gap of a MX<sub>2</sub> monolayer keeping the direct band gap at K point. The partial density of states analysis reveals that the band gap is mainly due the *d* orbital of M atom and the *p* orbitals of X atoms. Beyond these ground-state properties, the electronic density, and the Kohn-Sham structure of studied systems are used to calculate absorption and electron energy loss spectra, within time-dependent density functional theory (TD-DFT) using the Selected-G method [2]. The spatial dependence of the response function as well as of the induced density are also analysed [3]. Our results show clearly specific signature of the electronic density for each chalcogen.

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Electron removal energies in noble gas atoms up to 100 keV: *ab initio* GW vs XPS

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X-ray photoelectron spectroscopy (XPS) measures electron removal energies, providing direct access to core and valence electron binding energies, hence probing the electronic structure. Here, we will present the benchmark of the *ab initio* many-body GW approximation on the complete electron binding energies of noble gas atoms (He-Rn), which spans 100 keV. Our results demonstrate that GW achieves an accuracy within 1.2% in XPS binding energies, by systematically restoring the underestimation from density-functional theory (DFT, error of 14%) or the overestimation from Hartree-Fock (HF, error of 4.7%). Such results also imply that the correlations of *d* electrons are very well described by GW

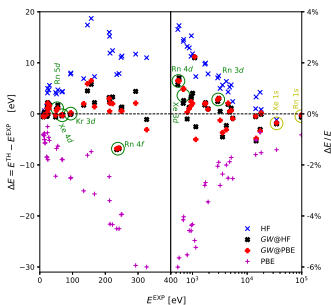


Figure 1: Noble gas atoms ZORA PBE, HF, and GW electronic energy absolute (left, 0 to 400 eV) and relative (right, from 400 to  $10^5$  eV) errors with respect to the experiment.

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## Characterization of point defects in silicon obtained via deterministic Ge ion implantation

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Point defects in semiconductors have been envisioned as suitable components for quantum technology applications. In this perspective achieving a control on their placement, stoichiometry and related properties is a stepping stone in view of a practical exploitation in devices. We present a combined experimental and theoretical study that allowed us to characterize defects formed by the implantation of Ge ions in silicon. The experimental protocol guarantees a deterministic placement with reasonable precision. Our theoretical model, based on a multiscale approach supports the experimental findings leading to a complete characterization of the chemical nature of the defect and its properties in the silicon matrix, including the explanation of the peculiar temperature-activated many-body transport observed for a nm-long array of Ge-vacancy defects in a silicon-based transistor.

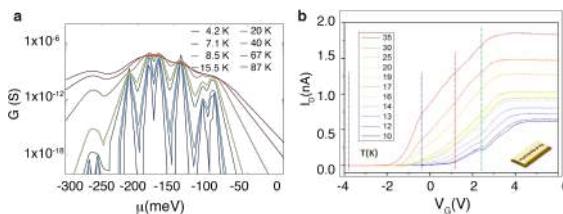


Figure 1: a) theoretical temperature-dependent conductance of an array of eight defects as a function of the chemical potential. b) experimental trans-characteristic for an array with eight pairs of Ge ions implanted in a Si channel as a function of the gate voltage

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## Theoretical study of internal conversion: complementary static and dynamic analysis of a bare and functionalized porphyrin

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We address the internal conversion (IC) within the Q band of a bare porphyrin (BP) and one functionalized version (FP). We employed a static TDDFT approach to identify the active normal modes based on the Reorganization Energy (RE) between the  $Q_x$  and  $Q_y$  states and, based on a method that we recently proposed<sup>1,2</sup>, we predict the existence of crossing points of their potential energy surfaces (PES). Additionally, we have performed non-adiabatic molecular dynamics (NAMD) simulations to assess the predictions of the static analysis. Essential dynamics and normal mode analysis of a statistical significant set of simulated trajectories shows that the static, less computationally intensive method, is indeed useful and reliable as an heuristic tool and as a guide for more complex and CPU-time intensive dynamical analysis of internal conversion close to conical intersections.

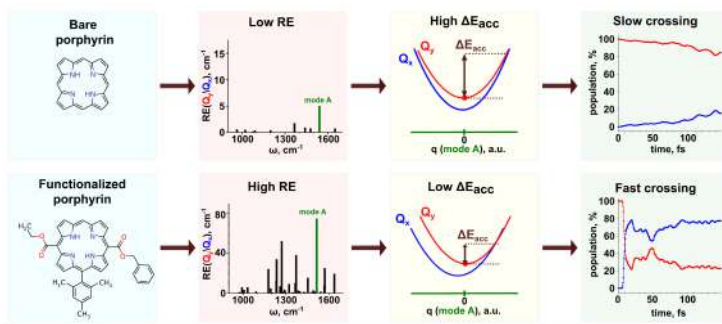


Figure 1: The prediction of internal conversion for bare and functionalized porphyrins by: 1) utilizing Reorganization Energy (RE) between  $Q_y$  and  $Q_x$ , 2) scanning potential energy surfaces (PES) along the active (high RE) normal mode 3) performing excited state population dynamics.  $\Delta E_{acc}$  represents the access energy from the local minimum of the  $Q_y$  to the crossing with the  $Q_x$  PES along the active normal mode.

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## Combining ab initio theories with machine learning to study the vibrational properties of molybdenum sulfides

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Transition metal chalcogenides represent a topical family of materials as they hold promise in the field of energy generation and storage. Catalytic activity for the hydrogen evolution reaction (HER) is one of the most exciting properties of molybdenum sulfides, which opened to an intense research effort. This project focuses on the theoretical study of their vibrational behaviour, especially their infrared and Raman modes, in order to find the structure-property relations that can help to better understand the catalytic mechanisms. The state-of-the-art methods to address such study are based on DFT, whose implementation can be computationally expensive, especially when working at finite temperature. To overcome this difficulty, we resort to machine learning (ML). In particular, we use the ML-Assisted Canonical Sampling approach (MLACS), which exploits molecular dynamics to explore phase space and optimize the ML parameters for a model to compute the energy and forces for a given system. This Machine Learning Interatomic Potential can replace the DFT potential in the calculation of vibrational spectroscopy or transport.

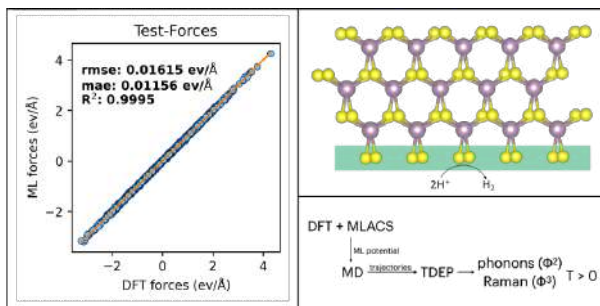


Figure 1: **Left:** Correlation graph between DFT- and ML forces computed for a 96-atom supercell of multilayer hexagonal MoS<sub>2</sub>. **Top-right:** sketch of the HER on hexagonal MoS<sub>2</sub> [1] (only one layer is shown for simplicity); S and Mo are represented respectively in yellow and purple. **Bottom-right:** scheme of the workflow (the ML interatomic potential is obtained from DFT calculations and MLACS and is used to run MD simulations; the resulting trajectories are then used as input for the Temperature Dependent Effective Potential method, TDEP, to obtain the second and third derivatives of the total energy at finite T).

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## Van der Waals Electrodynamics: plasmons and phonons in BN-capped graphene

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Plasmons and polar phonons are elementary electrodynamic excitations of matter capable of scattering electrons. Van der Waals heterostructures offer the opportunity to couple excitations from different layers via long-range Coulomb interactions, modifying the excitations themselves and their scattering of electrons. We study the complex dynamical interactions between electrons, plasmons and polar phonons with an efficient *ab initio* model to solve the dynamical electric response of Van der Waals heterostructures, accompanied by a formalism to extract relevant spectroscopic and transport quantities [1]. We apply those developments to BN-capped graphene, in which polar phonons from BN couple to plasmons in graphene. We study the nature of the coupled excitations and their coupling to graphene's electrons. Regimes driven by either phonons or plasmons are identified, as well as a truly hybrid regime corresponding to the plasmon-phonon-polariton at long wavelengths. Those are studied as a function of the graphene's Fermi level and the number of BN layers. In contrast with standard descriptions in terms of surface-optical phonons, we find that the electron-phonon interaction stems from several different modes. Moreover, the dynamical screening of the coupling between BN's LO phonons and graphene's electrons crosses over from inefficient to metal-like depending on the relative value of the phonons' frequency and the energetic onset of interband transitions. While the coupling is significant in general, the associated scattering of graphene's carriers is found to be negligible in the context of electronic transport.

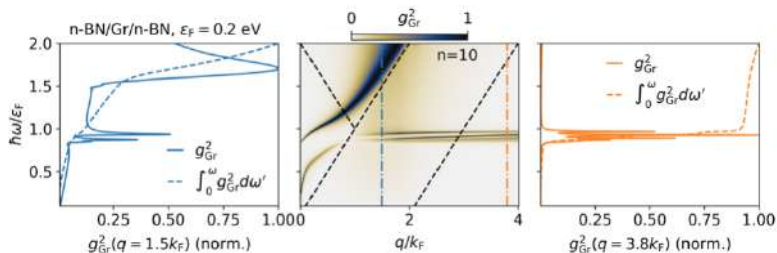


Figure 1: (Center) The colorscale represents the strength of the coupling of graphene's electrons with the electrodynamic modes of a 10-BN/Gr/10-BN heterostructure, normalized at each value of momenta  $q$ . We can see that both graphene's plasmon (dispersive excitation on the left) and BN's phonons (flatter excitation on the right) can scatter graphene's electrons. Coupled plasmon-phonon excitations (anti-crossing in the middle) also scatter. Curves on the left and right panels show the coupling for two selected values of momenta, corresponding to the vertical cuts in the central panel.

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## Emergence of flat bands in the quasicrystal limit of boron nitride twisted bilayers

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We investigate the electronic structure and the optical absorption onset of hexagonal boron nitride bilayers at small twist angles [1] and for twists in the vicinity of  $30^\circ$  [2]. Our study is carried out with a tight-binding model that we developed on purpose and validated against DFT simulations. We demonstrate that approaching  $30^\circ$  (quasicrystal limit), all bilayers sharing the same moiré supercell develop identical band structures, irrespective of their stacking sequence. This band structure features a bundle of flat bands laying slightly above the bottom conduction state which is responsible for an intense peak at the onset of independent-particle absorption spectra. These results reveal the presence of strong, stable and stacking-independent optical properties in boron nitride  $30^\circ$ -twisted bilayers. By carefully analyzing the electronic spatial distribution, we elucidate the origin of these states as due to interlayer B-B coupling. We take advantage of the physical transparency of the tight-binding parameters to derive a simple triangular model based on the B sublattice that accurately describes the emergence of the bundle. Being our conclusions very general, we predict that a similar bundle should emerge in other close-to- $30^\circ$  bilayers, like transition metal dichalcogenides, shedding new light on the unique potential of 2D materials.

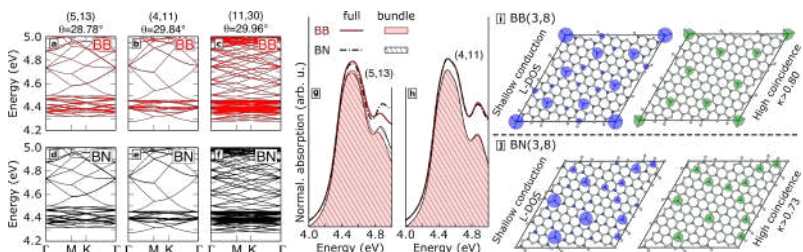


Figure 1: (a–f): Conduction bands of different bilayer stackings at different twist angles close to  $30^\circ$ . (g, h): IP absorption spectra including all empty states or only those in range 4.34 eV to 4.48 eV. (i, j): Local DOS (blue circles) and B-on-top-of-B map (green circles).

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## Optical properties of hBN point defects as Qubits

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Over the last two decades, interest in Two-Dimensional (2D) materials has increased due to their potential applications in quantum information technologies [1]. In particular, defect centers in wide-bandgap materials, such as hexagonal Boron Nitride (hBN), have been proposed as quantum bits (qubits) [2]. Photoluminescence (PL) of defect centers is used to readout the information stored in the qubit. Recently, it has been shown via first-principles calculations that explicitly including exciton-phonon coupling in hBN with defects is essential to reproduce the experimentally observed PL [3]. To date, phonon-assisted PL has only been computed for hBN with B-vacancy. However, other defects and the coupling between them have not been investigated yet.

In this work, we systematically study common defects in hBN (among which, the Nitrogen vacancy) and defect networks via Density Functional Theory (DFT) and Many-Body Perturbation Theory (MBPT), leveraging a recent implementation of the YAMBO code [4, 5]. We quantify the impact of defects configurations on the optical characteristics of hBN and compute phonon-assisted PL, offering a quantitative description of the many-body interplay between defect states and phonons. Defect networks are studied in the Wannier basis on top of the above MBPT description to account for large system sizes.

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## Insights into $\text{Ag}_2\text{Mo}_3\text{SeO}_{12}$ : A Theoretical Exploration of its Structural, Electronic, and Thermoelectric Behavior

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The structural, electronic, optical and thermoelectric properties of the new quadruple perovskite  $\text{Ag}_2\text{Mo}_3\text{SeO}_{12}$  were investigated using generalized gradient approximation (GGA-PBE) via density functional theory under Wien2k software. The Tran Blaha modified Becke-Johnson exchange potential (TB-mBJ) is employed to give an accurate result of the band gap value of the studied material. The band structure, total and partial densities of states, optical properties, and thermoelectric properties are discussed in detail. It is found that this new compound exhibits a band gap of around 1.45 eV with the indirect transition between valence and conduction band using TB-mBJ approximation. Moreover, the dielectric function, absorption coefficient as well as reflectivity calculations was obtained. A remarkable evolution of the absorption is noted in the visible range which proves the potential of quadruple perovskite  $\text{Ag}_2\text{Mo}_3\text{SeO}_{12}$  as an absorber solar cell and optoelectronic applications. The presented results of the studied material in this work open a new road for photovoltaic material design.

## How does temperature affect the internal conversion of photoexcited adenine and adenosine?

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It is well known that canonical DNA nucleobases and nucleosides undergo ultrafast internal conversion to the ground state after UV photoexcitation, either in the gas phase or solution. These ultrafast processes are the key to the remarkable photostability of DNA. However, one aspect that has received little attention is how the internal conversion depends on the ambient temperature. In this work,<sup>1</sup> we simulate the nonadiabatic dynamics of adenine and adenosine to elucidate how the energy excess provided by increasing the temperature affects their lifetimes. Our surface hopping simulations confirm that the excited state lifetime of adenine does not depend on temperature, whereas the lifetime of adenosine does, in agreement with experimental work. At 0 K, this transfer reduces the mean kinetic energy of the adenine moiety in such an extension that internal conversion is inhibited, and the lifetime is extended by a factor of 2.3 compared to that at 400 K. The crucial difference is the intramolecular vibrational energy transfer from adenine to the ribose group, which only occurs in adenosine. The modeling also allowed to discard the influence of viscosity, a previously proposed alternative explanation.

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## Theoretical insight into terahertz spectroscopy of the dye atacamite

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Atacamite is relatively well known for being formed on the surface of bronze statue due to corrosion. Atacamite is an unusual and attractive halide mineral. It is polymorphous with three other minerals; clinoatacamite, botallackite, and metastable paratacamite by sharing the same chemistry of  $Cu_2Cl(OH)_3$ . They crystallize in different structures with atacamite in orthorhombic, clinoatacamite in monoclinic, botallackite in monoclinic, paratacamite in rhombohedral (replacement of  $Cu$  by small amounts of another cation is necessary to stabilize the paratacamite structure). The  $Cu_2Cl(OH)_3$  are grouped as natural cuprates. In the meaning that they contain copper and oxygen, they are like the well-known high-Tc superconducting cuprates. However, the magnetic properties of the  $Cu_2Cl(OH)_3$  family are not clear. It is widely accepted that the magnetic interaction between the  $Cu^{2+}$  spins in the high-Tc superconducting cuprates is an essential factor in the occurrence of the high-Tc superconductivity. Therefore we think it is of interest to investigate the behaviors of the  $Cu^{2+}$  spin behaviors in the natural cuprates. First, we investigated the most known compound atacamite. Few papers have been published on the vibrational spectroscopy of these minerals. This may be because many of the bands relating to the  $CuCl$  and  $CuO$  vibrations occur below  $400\text{ cm}^{-1}$  and are therefore not easily measured. Further techniques such as infrared reflectance spectroscopy are also limited by cut-off at even higher wavenumbers. Raman spectroscopy has been used to probe the molecular structure of atacamite and paratacamite. The Raman spectra of atacamite and paratacamite are different. Even at first principles level there are not many articles on atacamite, in general only in the last decade first principles calculations for the condensed matter have been published precisely since phononic calculations have become routine. The calculated total and partial electronic densities of states of  $Cu_2Cl(OH)_3$  are shown in Fig. 1 and we want to obtain phonons using the *ph.x* program from the QUANTUM ESPRESSO suite.

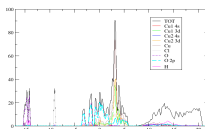


Figure 1: Typical for undoped cuprates, the valence band is dominated by Cu 3d and O 2p states.

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## Theoretical study of bound excitons in the $T$ -phase $SnS_2$ monolayer

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$SnS_2$  is a layered material with absorption in the visible spectrum and a suitable energy level alignment for application as an electrode in visible-light hydrogen fuel cells [1]. It is also a paradigmatic case of the group IV chalcogenides, which have recently aroused much interest in applications involving second harmonic generation response (SHG) [2]. Here, we present a comprehensive theoretical study of bound excitons in the  $T$ -phase  $SnS_2$  monolayer to aid in possible technological applications. We employ many-body perturbation theory methods [3] on top of the DFT-PBE electronic structure [4]. We have found a richer structure of bound excitons than reported previously [5], probably due to an overscreening, in that work, caused by the interlayer interaction. The first bright-exciton energy of 2.3 eV is also in better agreement with the experimental optical gap of 2.23 eV [1]. We present a detailed discussion on the composition and nature of the excitonic states. As a next step, we will extend this study to other interesting group IV chalcogenide systems, which are non-centrosymmetric, as the  $H$ -phase  $SnS_2$  monolayer, and  $SnS$  and  $SnSe$  layered systems, for which we also will investigate the SHG response [6].

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## How To Automatically Find The Symmetry Of Defect Orbitals

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Quantum technologies like single photon emitters and qubits can be enabled by point defects in semiconductors, with the NV-center in diamond being the most prominent example. There are many different semiconductors, each potentially hosting interesting defects. The symmetry properties of the point defect orbitals can yield useful information about the behavior of the system, such as the interaction with polarized light. We have developed a tool to perform symmetry analysis of point defect orbitals obtained by plane-wave density functional theory simulations. The software tool, named ADAQ-SYM [1], calculates the overlap between the original and symmetry-transformed orbital (see Fig. (a)), maps these overlaps to characters, identifies the irreducible representations, and finds allowed optical transitions and polarizations (see Fig. (b)).

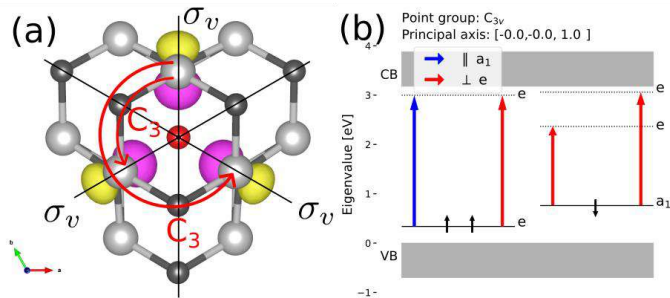


Figure: (a) A defect orbital from a divacancy in SiC (C<sub>3v</sub> point group), with reflection planes marked with black lines, and the two 120-degree rotations marked with red arrows. (b) Diagram showing irreducible representations and allowed transitions with polarization between defect orbitals in the band gap of SiC with a divacancy.

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Sun, Jun 2nd		Mon, Jun 3rd	
Young researchers activity	Time		
Young researchers round table	Morning	Collaboration Teams Discussion	

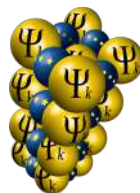
Young Researchers Social Program	14:00	ETSF SCM	Intro Lectures
	15:30	Reg. coffee	
	16:00	Opening	
	16:15	Claudia Draxl	
	17:00	Nicolas Ferré	
18:00	Welcome drink + Surprise		

ETSF SCM = ETSF Steering Committee meeting

Tue, Jun 4th			Wed, Jun 5th		
	Session	Time		Session	
Adam Gali	Spectroscopy & point defects	09:00	Alicia Palacios	Ultrafast Spectroscopy	
Martino Silveti		09:40	Tatiana Bezriadina		
Alexander Kirchhoff		10:00	Andreas Windischbacher		
Coffee break		10:20	Coffee break		
Xavier Blase	Multiscale, embedding & low-dim. materials	10:50	Ivan Maliyov	Exciton dynamics	
Gabriele Iuzzolino		11:30	Claudio Attaccalite		
Joachim Galiana		11:50	Fabienne Michelini		
Claudia Cardoso		12:10	ETSF GM		
Lunch		13:00	Lunch		
Alberto Zobelli	Low dim. materials and hetero-structures	15:00	Lou Barreau	Non equilibrium dynamics	
Fulvio Paleari		15:40	Brice Arnaud		
Coffee break		16:00	Coffee break		
Riccardo Reho		16:30	Kevin Lévêque-Simon		
Elisa Serrano		16:50	Lu Qiao		
Poster session		17:10	Seminar Isabelle Régner		

Thu, Jun 6th			Fri, Jun 7th		
	Session	Time		Session	
Julien Toulouse	Theoretical & numerical devs.	09:00	Maria Hellgren	Total energy & spectroscopy I	
Frank Ortman		09:40	Abdallah El-Sahili		
Jesus Villalobos		10:00	Iskander Mukatayev		
Coffee break		10:20	Coffee break		
Pierluigi Cudazzo	Advanced methods for electronic excitations	10:50	Bo Peng	Total energy & spectroscopy II	
Pina Romaniello		11:30	Nasrin Farahani		
Davide Sangalli		11:50	Martin Unzog		
Laura Urquiza		12:10	Closing remarks		
Jan-Hauke Graalmann	Non lin Opt	12:30			
Lunch		13:00			
Michele Celebrano	Non linear Optics	14:30			
Simone Grillo		15:10			
Social program		15:30			
Workshop dinner		19:30			

ETSF GM = ETSF General meeting



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