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2024
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2024

20th ETSF-YRM 2024
Young Researchers' Meeting

Toulouse, 27-31 May 2024

PROGRAM AND BOOK OF ABSTRACTS

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Website and contact

https://lcpq.github.io/yrm2024/
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Venue

FeRMI seminar room @ Université Paul Sabatier
Université Paul Sabatier
1st stage of the building 3R4
76 Rue Sébastienne Guyot
31400 Toulouse

GPS Coordinates:
Latitude: 43.56075
Longitude: 1.46774

Direction from the City Center
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Organizers

Abdallah Ammar *LCPQ, CNRS, Toulouse, France*
Leonardo Biancorosso *University of Trieste, Italy*
Sofia Canola *Institute of Physics of the Czech Academy of Sciences, Czech Republic*
Karthikeyan Saravanabavan *CEA-LITEN, Grenoble, France*
Amandine Laurent (administrative support) *LCPQ, Toulouse, France*

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Special acknowledgement to **LCPQ laboratory**, especially to the **director Dr. Thierry Leininger**.

Overview program

	Monday 27/05	Tuesday 28/05	Wednesday 29/05	Thursday 30/05	Friday 31/05
	Electronic Structure and methods development	***Optical Properties of Materials***	***Vibrational Properties of Materials and Transport***	***Strongly Correlated Systems and Magnetism***	***Machine Learning in Materials Science***
9.20 - 9.50	Registration				
9.50 - 10.00	Introductory remarks				
	chair : Sofia Canola	*chair : Leonardo Biancorosso*	*chair : Laura Urquiza*	*chair : Karthikeyan Saravanabavan*	*chair : Aloïs Castellano*
10.00 - 10.55	**Gianluca Levi**	**Mirko Vanzan**	**Svenja M. Janke**	**Carlos Mejuto-Zaera**	**Jannes Nys**
10.55 - 11.20	*Coffee*	*Coffee*	*Coffee*	*Coffee*	*Coffee*
11.20 - 11.45	Gabriele Fabbro	June Groothuizen	Felana Noeliarinala Andriambelaza	Coraline Letouzé	Henry Fried
11.45 - 12.10	Fileto Rodríguez	Pierpaolo D'Antoni	Raveena Gupta	Fatema Yahya Mohamed	Prashant Kumar
12.10 - 12.35	Simone Brozzesi	Line Jelver	Mahé Lezoualc'h	Marco Marino	José Batista
12.35 - 13.00	*Lunch*	*Lunch*	*Lunch*	*Lunch*	***Industry session*** **Julia M. Schaefer**
13.00 - 14.00					*Conclusion*
	chair : Sara Giarrusso	*chair : Svenja M. Janke*	*chair : Leonardo Biancorosso*	*chair : Maria Andreea Filip*	
14.00 - 14.55	**Maria Andreea Filip**	**Laura Urquiza**	**Aloïs Castellano**	**Sara Giarrusso**	
14.55 - 15.20	*Coffee*	Jozef Janovec	*Coffee*	*Coffee*	
15.20 - 15.45	Stefano Paggi	**Poster session** *with Coffee*	Maram Ali Ahmed Musa	Yiming Zhang	
15.45 - 16.10	Amer Alrakik		Maxime Mignolet	Emil Östberg	
16.10 - 16.35	Domenico Corona			Daniele Veraldi	
16.35 - 17.00	Ian-Evan Michel				
		Social dinner			

Detailed program

<u>**Day 1: Monday May 27th 2024**</u> *Electronic Structure and methods development*

9.20 - 9.50 Registration
9.50 - 10.00 Introductory remarks

Invited Talk

10.00 - 10.55 **Gianluca Levi** (University of Iceland)
Calculating Challenging Electronic Excitations in Molecules and Solids by Converging on Saddle Points of the Electronic Energy Surface

10.55 - 11.20 *Coffee break*

Contributed Talks

11.20 - 11.45 **Gabriele Fabbro** (University of Toulouse III)
Coupled cluster method for molecular properties

11.45 - 12.10 **Fileto Rodríguez** (Vrije Universiteit Brussel)
Unraveling the Raman Spectra of Complex Materials: Autocorrelation Function Formalism as a State-of-the-Art technique

12.10 - 12.35 **Simone Brozzesi** (University of Rome Tor Vergata)
Moiré-induced Dirac cones replicas and minigaps opening in graphene/hBN superlattices

12.35 - 14.00 *Lunch break*

Invited Talk

14.00 - 14.55 **Maria Andreea Filip** (Max Planck Institute for Solid State Research, Stuttgart)
Transcorrelated Hamiltonians for Second Row Elements

14.55 - 15.20 *Coffee break*

Contributed Talks

15.20 - 15.45 **Stefano Paggi** (University of Toulouse III)
Multi-channel Dyson equation: application to neutral excitations

15.45 - 16.10 **Amer Alrakik** (LCPQ, University of Toulouse III)
Solution to the Thomson Problem for Clifford Tori with an Application to Wigner Crystals

16.10 - 16.35 **Domenico Corona** (University of Rome Tor Vergata and INFN)

Self-consistent Hubbard parameters in doped layered transition-metal oxides for sodium-ion batteries

16.35 - 17.00 **Ian-Evan Michel** (CEA Paris-Saclay)
DFT Simulation Study of Hexagonal Phosphorus Monolayer on Au(111) and Adsorbate Structure Exploration using Genetic Algorithm

Day 2: Tuesday May 28th 2024 *Optical Properties of Materials*

Invited Talk

10.00 - 10.55 **Mirko Vanzan** (University of Milano)
Design of Au-Based Nanoalloys as Plasmocatalyst

10.55 - 11.20 *Coffee break*

Contributed Talks

11.20 - 11.45 **June Groothuizen** (University of Amsterdam)
Electronic structure and optical properties of exciton condensate candidate TiSe2 using many-body perturbation theory

11.45 - 12.10 **Pierpaolo D'Antoni** (Università degli Studi di Trieste)
Dichroism of plasmonic chiral nanoalloys by rational design

12.10 - 12.35 **Line Jelver** (University of Southern Denmark)
Nonlinear Plasmonics in Nanostructured Phosphorene

12.35 - 14.00 *Lunch break*

Invited Talk

14.00 - 14.55 **Laura Urquiza** (École Polytechnique de Paris)
Excitonic effects revealed through resonant inelastic X-ray scattering and complementary X-ray spectroscopies

Contributed Talks

14.55 - 15.20 **Jozef Janovec** (University of the Basque Country/ Materials Physics Centre, Spain) *Exploiting Optical Properties of Cementitious Oxides for Photonic Metaconcrete Design from first principles*

15.20 - 18.00 **Poster session (and coffee)**

19.30 **Social dinner @ Bambèe Bistro**

Day 3: Wednesday May 29th 2024 *Vibrational Properties of Materials and Transport*

Invited Talk

10.00 - 10.55 **Svenja M. Janke** (University of Warwick)
Influence of the Organic Cation Orientation on the Absorption Spectra of 2D Hybrid Organic-Inorganic Perovskites

10.55 - 11.20 *Coffee break*

Contributed Talks

11.20 - 11.45 **Felana Noeliarinala Andriambelaza** (LSI, CNRS, CEA/DRF/IRAMIS, Ecole Polytechnique de Paris)
Electronic properties of 2D Transition Metal Dichalcogenides ternary alloys: an ab initio study

11.45 - 12.10 **Raveena Gupta** (University of Liège)
Exploring electron-phonon interactions in diamond and silicon: insights from spectral analysis

12.10 - 12.35 **Mahé Lezoualc'h** (CEA Paris-Saclay)
Electronic transport calculations in different CDW phases of 1T-VTe2 and 1T-VSe2

12.35 - 14.00 *Lunch break*

Invited Talk

14.00 - 14.55 **Aloïs Castellano** (University of Liège)
From thermodynamics to vibrational spectra and transport: a theory of the anharmonic crystal

14.55 - 15.20 *Coffee break*

Contributed Talks

15.20 - 15.45 **Maram Ali Ahmed Musa** (Ecole Polytechnique de Paris)
Why Metavalent materials as so special? The cubic Antimony

15.45 - 16.10 **Maxime Mignolet** (University of Liège)
Phonon angular momentum in magnetic systems

Day 4: Thursday May 30th 2024 *Strongly Correlated Systems and Magnetism*

Invited Talk

10.00 - 10.55 **Carlos Mejuto-Zaera** (International School for Advanced Studies, SISSA)
Local and Non-Local Interactions in Strongly Correlated Materials: Embedding Through Auxiliary Particles

10.55 - 11.20 *Coffee break*

Contributed Talks

11.20 - 11.45 **Coraline Letouzé** (IMPMC, Sorbonne Université)
Computing X-ray absorption in strongly correlated materials beyond multiplet theory

11.45 - 12.10 **Fatema Yahya Mohamed** (Ecole polytechnique de Paris)
Excitons in Helium Under Pressure

12.10 - 12.35 **Marco Marino** (University of Milan)
Ab initio study of the Fe-phthalocyanine/Cr2O3(001) spinterface

12.35 - 14.00 *Lunch break*

Invited Talk

14.00 - 14.55 **Sara Giarrusso** (University of Paris-Saclay)
Applications of the exact factorization formalism in density-functional theory

14.55 - 15.20 *Coffee break*

Contributed Talks

15.20 - 15.45 **Yiming Zhang** (UCLouvain)
In search of new phonon-limited superconductors

15.45 - 16.10 **Emil Östberg** (University of Lund)
Microscopic theory of current-induced skyrmion transport and its application in disordered spin textures

16.10 - 16.35 **Daniele Veraldi** (Sapienza University of Roma)
Fully Programmable Spatial Photonic Ising Machine

Day 5: Friday May 31th 2024 *Machine Learning in Materials Science*

Invited Talk

09.00 - 09.55 **Jannes Nys** (École Polytechnique Fédérale de Lausanne)
Ab-initio variational wave functions for the time-(in)dependent many-electron Schrödinger equation

Contributed Talks

09.55 - 10.20 **Henry Fried** (University of Luxembourg)
A Machine Learning Approach to Predict Tight-binding Parameters for Point Defects via the Projected Density of States

10.20 - 10.45 *Coffee break*

10.45 - 11.10 **Prashant Kumar** (LCPQ, University of Toulouse III)
In Silico study of chemical degradation of host molecular materials in OLEDs

11.10 - 11.35 **José Batista** (Université de Liège)
Temperature Dependent Dielectric Properties of Bulk Hexagonal Boron Nitride

Industry session

11.35 - 12.30 **Julia M. Schaefer** (Heliatek GmbH)
Transitioning from Academia to Corporate R&D as a Computational Chemist

12.35 - 14.00 Lunch

14:00 - 16:00 *Scientific discussion and conclusion*

List of posters

Sarbajit Dutta (Ecole Polytechnique)
Plasmon satellites in Cesium

Stephanie Egome Nana (University of Lorraine)
Bound states of H_2^+ *using a multicenter approach based on complex Gaussian functions*

Mohammed Gunes (Ecole Polytechnique)
Charge density as a functional of the potential: Connector Theory approach

Daniele Lagasco (LCPQ, Université Toulouse III)
Ab initio modelling of electronic and magnetic properties of small Wigner molecules

Alessia Muroni (University of Rome Tor Vergata)
Unveiling the optical secrets of ice surfaces: insights from advanced computational approaches

Emma Nogué (King's College London)
Advancing Photoemission Spectra Analysis with Three-Body Green's Function Formalism

Mexx Regout (University of Liège)
Magnon lifetimes with frustration of exchange

Vasil Saroka (University of Rome Tor Vergata)
Electronic properties of nanopatterned hydrogenated graphene superlattices

Marie Tardieux (University of Amsterdam)
Engineering site mixing to understand magnetic topological insulators through DFT calculations

Abstracts of oral presentations

Solution to the Thomson Problem for Clifford Tori with an Application to Wigner Crystals

A. Alrakik , M. Escobar Azor, V. Brumas, G. L. Bendazzoli, S. Evangelisti, J. A. Berger

In its original version, the Thomson problem consists of the search for the minimum-energy configuration of a set of point-like electrons that are confined to the surface of a two-dimensional sphere (S2) that repel each other according to Coulomb's law, in which the distance is the Euclidean distance in the embedding space of the sphere, i.e., R3. In this work, we consider the analogous problem where the electrons are confined to an n-dimensional flat Clifford torus Tn with n = 1, 2, 3. Since the torus Tn can be embedded in the complex

manifold Cn, we define the distance in the Coulomb law as the Euclidean distance in Cn, in analogy to what is done for the Thomson problem on the sphere. The Thomson problem on a Clifford torus is of interest because supercells with the topology of a Clifford torus can be used to describe periodic systems such as Wigner crystals. In this work, we numerically solve the Thomson problem on a square Clifford torus. To illustrate the usefulness of our approach, we apply it to Wigner crystals. We demonstrate that the equilibrium configurations we obtain for large numbers of electrons are consistent with the predicted structures of Wigner crystals. Finally, in the one-dimensional case, we analytically obtain the energy spectrum and the phonon dispersion law.

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

N.F. Andriambelaza,[1] *C. Giorgetti*[1]

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First principles calculations are performed to investigate the properties of monolayer transition metal dichalcogenides (TMDs) ternary alloys of the form MXY, with M = Mo, W and $X \neq Y$ = 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 band structure analysis, we saw that the introduction of the Y chalcogen fine tune the band gap of a MX_2 monolayer keeping the direct band gap at *k* point. The partial density of states analysis reveals that the band gap is mainly due to 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.

References

[1] Zollner, K., Junior, P. E. F., & Fabian, J. (2019). Strain-tunable orbital, spin-orbit, and optical properties of monolayer transition-metal dichalcogenides. Physical Review B, 100(19), 195126.
[2] Tancogne-Dejean N., Giorgetti C., Véniard, V. (2015). Optical properties of surfaces with supercell *ab initio* calculations: Local-field effects. Physical Review B, 92(24), 245308.
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Temperature Dependent Dielectric Properties of Bulk Hexagonal Boron Nitride

J. Batista,[1] *A. Castellano,*[1] *M. Verstraete*[1,2]

[1] Nanomat group, QMAT center, CESAM research unit and European Theoretical Spectroscopy Facility, Université de Liège, allée du 6 août, 19, B-4000 Liège, Belgium
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Hyperbolic materials are structures that have opposite-sign dielectric constants when light is made to propagate along different directions. The fact that these two characteristics coexist in a single material can be exploited in novel optical applications. This is particularly interesting as these kinds of materials allow, contrary to usual metals and dielectrics, for light with large magnitude wave vectors to propagate inside them with no exponential decay, thus making it possible to make optical devices much smaller than what is currently allowed. More generally, these materials are the gateway for unique bulk electromagnetic states which can tailor light-matter interaction at the nanoscale, offering a wide range of new nanophotonic applications. Hyperbolicity is usually constructed artificially with meta-materials combining metallic and insulating segments, but can also be found in anisotropic bulk materials, such as hexagonal Boron Nitride, in certain frequency ranges.

From an optical perspective materials are characterised by their dielectric tensor ε, which tells us how easily light propagates in a material, affecting both the speed of light and its absorption. In this work we show our approach to calculate the temperature dependency of ε, based on Machine Learning and Molecular Dynamics sampling and Density Functional Perturbation Theory. In particular, we focus on the case of bulk hBN and shed light on future improvements and applications to predict temperature-dependent Raman spectra.

Moiré-induced Dirac cones replicas and minigaps opening in graphene/hBN superlattices

S. Brozzesi,[1] A. Zobelli,[2] M. Palummo,[1] O. Pulci[1]

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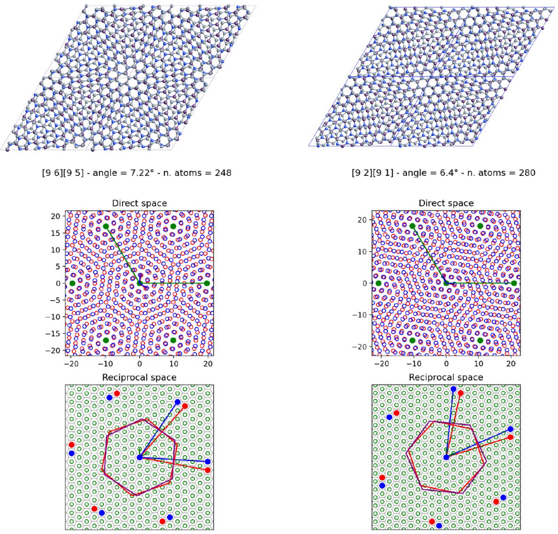

Moiré pattern is a novel and extensive structure that emerges from the interference of multiple periodic templates. In condensed-matter physics, moiré patterns, also known as moiré superlattices, can be created by stacking two or more two-dimensional (2D) layered materials with a small twist angle and/or a slight lattice mismatch. The presence of moiré pattern results in the formation of a long range moiré potential, which interacts with the electrons of the system and affects the dispersion of the energy levels. In recent years, moiré superlattices have garnered significant attention due to their remarkable manifestation of previously unexplored phenomena and unique functionalities [1], like strongly correlated phases [2] and subsequent observation of superconductivity and topological states [3], and exotic excitonic states [4] arising from the interaction between excitons and moiré potential. The aim of this study is to demonstrate that the existence of a moiré pattern in a graphene/hBN heterostructure has a substantial influence on the energy levels dispersion of the system. In particular, there is the intention to show that this influence goes beyond the coupling between the planes and the rehybridization of levels that can be expected in van der Waals heterostructures, but significantly affects the energy levels dispersion of graphene within an energy range inside the hBN gap, where only contributions from C atoms are expected to be present. By means of DFT calculation based on a localized basis set approach, this study demonstrates that the presence of the long range moiré potential results in the formation of six replicas [5] of the graphene Dirac cones around the K-K' point of the 1BZ of graphene, which are separated by a reciprocal space superlattice vector Gm from the main cone. The intersection between the main cone and its replicas result in the opening of minigaps in the energy levels of graphene, without the direct interplay of the interaction with the hBN states. The relative positions of these minigaps is dependent on the moiré reconstruction of the system and can be tailored by tuning the rotation angle between the layers.

References

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[2] Yuan Cao et al., Nature 556.7699 (2018), pp. 80–84
[3] Yuan Cao et al., Nature 556.7699 (2018), pp. 43–50
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[5] Ivo Pletikosić et al., Physical review letters 102.5 (2009), p. 056808

From thermodynamics to vibrational spectra and transport: a theory of the anharmonic crystal

A. Castellano[1]

[1] University of Liège, Belgium

In the solid state of matter, atoms vibrate around equilibrium positions. Using the harmonic approximation and perturbation theory, these vibrations can be described as quantized quasiparticles called phonons, which provides a complete framework to describe a large variety of phenomena occurring in crystals[1], from the thermodynamic properties to vibrational spectral or the way heat flows. However, the harmonic approximation is founded on the assumption that the contributions to the potential energy beyond the second order Taylor expansion can be treated as small perturbations. When these anharmonic contributions are not negligible, the harmonic approximation and its perturbation theory often becomes quantitatively or even qualitatively wrong. Nevertheless, the phonon picture is still useful in strongly anharmonic materials, so that theories to improve on the harmonic approximation have emerged in the literature[2-4]. All of them are constructed on the idea that phonons are renormalized from interactions with a bath made of all other phonons, making them dependent on the temperature. In this talk, I will present a complete framework to predict properties of anharmonic crystals, using the mode-coupling theory of anharmonic lattice dynamics[4] that we introduced recently. I will describe in particular how the method improves the harmonic prediction of thermodynamic properties, vibrational spectra or heat transport by incorporating directly the anharmonicity in the description of the phonons.

References

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Self-consistent Hubbard parameters in doped layered transition-metal oxides for sodium-ion batteries

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Sodium-ion batteries share similarities with the well-established lithium-ion batteries currently in widespread use [1]; in fact, analogously to lithium-ion positive electrodes, sodium-ion cathodes store sodium through an intercalation mechanism. Cathodes based on P2-Na_xTMO_2 crystals, i.e., sodium-based layered transition-metal oxides (LTMOs), have attracted growing interest owing to their high potential and capacity [2]. However, the geometric structure of LTMOs may undergo a Jahn-Teller distortion during charge/discharge cycles. Doping has proven effective in stabilizing the P2 geometric structure improving the cycling stability by limiting structural transitions as well as delaying them at higher voltages. Ab-initio calculations within the *Quantum ESPRESSO* suite [3] are employed to determine the optimal dopants, the total energy of the system, and estimate the sodium-ion intercalation potential. The structural and electronic properties of doped LTMOs are investigated at a GGA + *U* level of theory, where the on-site Hubbard *U* parameters are evaluated self-consistently using linear-response theory [4], so to realistically include the screening from the environment which is crucial for quantitatively predictive calculations.

References

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Dichroism of plasmonic chiral nanoalloys by rational design

P. D'Antoni[1], *D. Toffoli*[1], *G. Fronzoni*[1], *M. Stener*[1], *L. Sementa*[2], *A. Fortunelli*[2]

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[2] CNR-ICCOM, Consiglio Nazionale delle Ricerche, Italy

Understanding the physical nature of the plasmon phenomena for finite nano-sized systems could expand the reach of enhanced spectroscopy techniques, indeed plasmons can potentially amplify the local field by many orders of magnitude allowing the study of even single molecules1. In a recently published paper2 we study, via TDDFT simulations, a series of chiral gold/silver alloy nanowires to explore whether silver doping can produce an enhancement of circular dichroism at the plasmon resonance in these systems, and to identify the quantum-mechanical origin of the observed effects. We find a strong plasmonic dichroism when one or two helixes of gold atoms are substituted by silver in a linear chiral nanotube, whose pure gold counterpart does not display any plasmonic dichroism, and we rationalize this finding in terms of "decoupling" the destructive interference of excitations in the pure gold nanotube via alloying. However, further attempts to increase the plasmonic dichroism by considering multi-shell gold nanowires in which one entire shell is doped with silver did not produce the desired effect, but rather a decrease in circular dichroism. We show that this latter result is due to a more severe destructive interference in the dipole excitation contributions and suggest that further amplification should be possible in principle by properly tuning simultaneously the nanowire structure and chemical ordering.

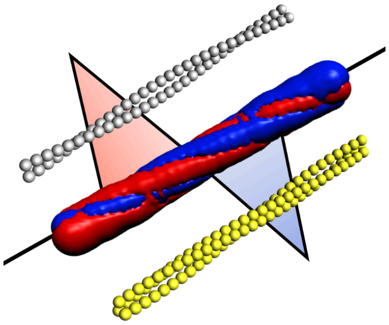

Figure 1: In grey are the silver rows of atoms used to alloyed the gold (in yellow) chiral nanotube. At the center of the panel is the perturbed electron density calculated at plasmon energy

References
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Coupled cluster method for molecular properties

G. Fabbro[1], T. Saue[1]

[1] Laboratoire de Chimie et Physique Quantiques , Toulouse

In theoretical chemistry, we recognize that the energy of a system ranks among the most crucial pieces of information, and its calculation stands as a fundamental aspect for method validation. However, within experimental chemistry, the spotlight shifts away from energy, recognizing the paramount importance of other quantities. Among these, equilibrium geometry, dipole moment, NMR shielding, vibrational frequencies, and more stand out [1]. Bridging the gap between experimental and theoretical chemistry is imperative, and this link is forged through the exploration of molecular properties. These properties shed light on how a molecule interacts with external perturbations, rendering them highly sensitive to the electronic structure under scrutiny. It is thus clear that the accurate computation of these properties hinges on the precise calculation of electronic structure. Among the arsenal of theoretical chemists, the Coupled Cluster (CC) method stands out as one of the most accurate. Renowned for its capability to systematically describe electronic correlation and its size extensivity [2], CC is a powerful and widely-utilized technique in computational chemistry for predicting energies and molecular properties. This talk will delve into the essential concepts concerning the computation of time-independent molecular properties, with a keen focus on the current methodology employed using the coupled–cluster method, as implemented in the DIRAC program [3].

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Transcorrelated Hamiltonians for Second Row Elements

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The transcorrelated (TC) method is based on a non-unitary transformation of a system Hamiltonian, using a Jastrow factor to explicitly account for required cusp behaviour of the wavefunction at particle coalescence. This transformation leads to a non-Hermitian Hamiltonian with additional three-body terms, the complexity of which has long precluded extensive use of this technique. Recently, with the development of efficient treatment methods for the three-body terms,[1] as well as the design of highly flexible parametrised Jastrow factors that can be optimised effectively by Variational Monte Carlo (VMC),[2] the TC method has been shown to generate highly accurate total and relative energies for atomic and molecular systems of first row elements[2,3] using Full Configuration Interaction Quantum Monte Carlo (FCIQMC) or coupled cluster approaches to obtain the remaining correlation energy. TC energies exhibit fast convergence with basis set size,[3] as well as increased sparsity of the ground state wavefunction,[2] which is crucial for the efficiency of solvers like FCIQMC and for the ease of representation of such wavefunctions on emerging quantum computers.[4]

In this talk, we present the TC formalism and expand the methods described above to investigate properties of second row elements, for which the presence of core electrons significantly increases the variance of the energy to be minimised during the Jastrow optimisation. Basis sets which account for core correlation therefore significantly improve the quality of the obtained energies, with similar trends to those seen with first row systems observed. Convergence with basis set size is fast, with TC total atomic energies achieving sub-milliHartree errors relative to the basis set limit where conventional approaches still yield errors on the order of 50 milliHartree. For ionisation potentials, we find that the moderately sized cc-pCVTZ basis set is sufficient to achieve near chemical accuracy.

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Unraveling the Raman Spectra of Complex Materials: Autocorrelation Function Formalism as a State-of-the-Art technique

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Raman spectroscopy has gained increasing attention for the last decades due to the popularization of laser sources and its capabilities. Owing to its sensibility, differentiating between materials with similar structures (due to polymorphism, dopants, defects…), ease of implementation, and sustainability, it has found a place in the characterization toolbox of many scientists. Nevertheless, Raman spectroscopy is susceptible to numerous parameters, primarily concerning the spectrometer configuration and physical conditions during the obtention of the spectrum. To tackle this pressing problem, CHARISMA2020 arises as a collaborative pioneering project between scientists in academia and industrial partners whose objective is to harmonize and standardize this technique. Within the modeling work package, our primary objective has been the development of robust theoretical protocols and spectra through the utilization of cutting-edge computational techniques. The resultant ab initio theoretical spectra serve as pristine references for experimentalists, facilitating the harmonization of the technique.

For this purpose, two methods have been used: the diagonalization of the dynamic matrix followed by intensity computation (DM), and the lesser-known autocorrelation function formalism (ACF) method.[1] ACF is a powerful method which has been available for the past two decades but is underutilized by most of the theoretical spectroscopy community.

Among the materials targeted, the first focus was placed on molecules in solution. We show how the ACF method retrieves accurate spectra for these species, and how commonly used DM-based methods fail to properly capture effects created by the presence of a solvent. This is attributed to the presence of hydrogen bonds with the solute, requiring explicit solvent, and to the conformations that the system can adopt. Additionally, several calcium carbonate polymorphs were studied. We show how one of the most common protocols to compute the electromagnetic properties when using the ACF method in this family of materials is unphysical and biased. Additionally, we demonstrate how the most employed methodology for computing Raman spectra in these structures—i.e. DM-based method—fails to accurately reproduce several features in the spectra of these systems. We therefore establish an alternative protocol verifying that it provides better accuracy when computing the spectrum of these materials compared to the prevailing methodologies.

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A Machine Learning Approach to Predict Tight-binding Parameters for Point Defects via the Projected Density of States

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Understanding the electronic and optical properties of defects in materials is required for the development of technological applications. A well established method to simulate defects is the use of density functional theory on supercells. But this method is restricted to relatively small supercells. Semi-empirical tight-binding models can overcome this problem. Hence, devising a reliable method to obtain good parameterizations is an ongoing research effort. We present a novel approach which reduces the number of fitting parameters to a minimum. We start with a fit of the pristine material, enlarged by a distance dependence of the hopping parameters. Upon introducing a defect, we add parameters for hoppings from the defect to neighboring atoms. Furthermore, the defect adds a distance-dependent perturbation to the onsite-energies. The fit is performed on the local (i.e., atom projected) density of states with the help of a machine learning algorithm. We test our method on the localised defect states associated with a carbon substitution in hBN, a material for which standard fitting tools do not produce reliable results.

Applications of the exact factorization formalism in density-functional theory

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The exact factorization is an overarching strategy in quantum mechanics, which stems from probability theory and has been applied to several fields, including molecular dynamics calculations and density-functional theory (DFT).

I will focus my talk on the applications of the exact factorization formalism in this latter case. Within Kohn-Sham (KS) DFT, the exact factorization provides a means to break down the exchange-correlation potential, revealing the individual components responsible for features of the exact potential like “peaks” and “steps” [1]. These features are extremely challenging to reproduce with current approximations. Nonetheless, they are fundamental to describe correlation effects in open-shell systems and in processes like molecular dissociation. I will illustrate how these features are related to the underlying correlated wavefunction in a simple model system for a stretched heteronuclear molecule. Moreover, this formalism has been considered mostly for real-valued wavefunctions. However, in the case of complex current-carrying states, an effective vector potential associated to the paramagnetic electronic current density emerges [2], which has so far received little attention. I will present an example of such electronic vector potential in a simple model of an atom in a triplet state.

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Electronic structure and optical properties of exciton condensate candidate TiSe2 using many-body perturbation theory

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In this talk I will present ab-initio calculations of the electronic structure of transition-metal dichalcogenide TiSe2, a layered van der Waals compound. Although this material has been investigated for decades, the electronic structure of its high-temperature “normal” phase is still under debate, with the distinction between semimetallic or semiconductive structure only recently beginning to be firmly established by bulk probes. Also, the nature and driving mechanism of the charge density wave (CDW) phase that is formed when it is cooled down below TCDW ≈ 200 K is not yet understood. The CDW phase shows signs of exciton condensation, the condensation of electron-hole pairs. To aid the interpretation of the various existing experimental results and to provide accurate predictions for the optical response, reliable first-principles calculations are required that provide a good description of the small electron- and hole-pockets at the L and Γ points of the Brillouin zone and the changes to the band structure in the distorted CDW lattice. Previous ab-initio literature suggests that correcting density functional theory (DFT) results with the one-shot GW approximation to many-body perturbation theory for the self-energy, is not sufficient to describe the semimetallic character of TiSe2 and that a quasiparticle self-consistent GW (QSGW) treatment is necessary. However, previous results with this method included spin-orbit coupling (SOC) corrections only a posteriori, instead of in the self-consistent cycle. Here, I will present results of the DFT, GW and QSGW calculations done with the code SPEX (spex.readthedocs.org) which allows for full treatment of spin-orbit coupling. Additionally, a further characterization of the optical properties of the material, based on the calculation of the dielectric function, will be addressed.

Exploring electron-phonon interactions in diamond and silicon: insights from spectral analysis

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This study delves into the intriguing phenomena of electron-phonon interactions in diamond and silicon, two nonpolar crystals. Despite their nonpolar nature, these materials exhibit noteworthy spectroscopic and optical features attributed to the formation of polarons—quasi-particles arising from interaction of electrons with vibrations in crystal lattices. By analyzing the many-body spectral function, we uncover distinct characteristics in each material. In diamond, a plateau structure emerges in the spectral function, reminiscent of but distinguishable from the satellites observed in the polar Frohlich case. Conversely, the satellites observed in silicon more closely resemble the Frohlich polaron case. Our investigation extends to temperature-dependent spectral functions, employing both the standard Dyson–Migdal approach and the cumulant expansion method. The latter, accounting for higher-order electron–phonon interactions, yields refined spectral functions, offering insights into both conduction and valence band edges. Through detailed examination, we elucidate the nuanced behaviors of electron-vibration interactions in these materials.

Influence of the Organic Cation Orientation on the Absorption Spectra of 2D Hybrid Organic-Inorganic Perovskites

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Hybrid organic-inorganic perovskites (HOIPs) allow combining organic and inorganic materials at the nanoscale and hence open up a wide area of tunability. In two-dimensional HOIPs, both organic and inorganic components can contribute to the electronic frontier levels. To predict new photovoltaic materials from theory, a fundamental understanding of the electronic excitations, their photophysical signature and their relationship with the underlying structure is essential. I use a Frenkel-Holstein Hamiltonian to model the organic exciton contribution and its coupling to underlying vibrations. The 2D HOIP inorganic exciton binding energy depends approximately linearly on the inorganic band gap. To estimate the inorganic exciton contribution to the absorption spectrum, I benchmark the amount of Hartree Fock exchange in hybrid density functional theory calculations including spin-orbit coupling.

For the quaterthiophene-based lead-halide HOIPs (AE4T)PbX_4 (X = Cl, Br, I), variation of the halide anion leads to structural changes in the organic layer, causing changes in the vibronic progression in the absorption spectrum.

The bithiophene-based 2D HOIP (AE2T)PbI_4 shows disorder along the stacking direction in X-ray scattering experiments, resulting in several possible atomic structural models for the organic component.

Here, I demonstrate that we can use a Frenkel-Holstein Hamiltonian-based model to investigate how different orientations of the organic cation can alter the organic vibronic progression and the inorganic exciton contribution to the absorption spectrum.

Exploiting Optical Properties of Cementitious Oxides for Photonic Metaconcrete Design from first principles

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Cement is the single most produced material by humans. Being the main ingredient of concrete extensively used in construction, there is a constant demand for its improvement and development. The downside of the large scale cement production is a massive CO2 footprint, responsible for roughly 10% of the global emission. Consequently, there is a pressing need to mitigate the environmental impact of concrete. A potential solution involves reducing energy consumption of buildings related to the heat management, by embedding phase change materials in concrete [1] or by utilizing the radiative cooling mechanism [2]. Projects like MIRACLE (Photonic Metaconcrete with Infrared RAdiative Cooling capacity for Large Energy savings) aim to pioneer a new generation of environmentally friendly concrete capable of passive cooling, thereby reducing energy consumption in buildings. The proposed solution takes advantage of the radiative cooling phenomenon, exploiting the heat release within the atmospheric transparency window (ATW). The ATW represents a range of infrared frequencies for which the atmosphere is permeable. If an object emits heat within the ATW, it escapes the atmosphere and dissipates into outer space, resulting in a formation of thermal equilibrium with a significantly colder environment than the ambient temperature. This permits the object to reach temperatures lower than its surrounding. In practice the research aims to develop photonic concrete by employing structured metamaterials [4] to manipulate the absorption and emission spectrum of concrete. Alternatively, modifying the electronic structure through doping or incorporating nanocomposites like graphene oxide layers into the cement paste represents another approach. The radiative cooling is a phenomenon based on the interaction of the matter with light. Whatever the form, the design of radiative cooling systems relies on an understanding of the optical properties of the materials involved. To engineer effective daytime radiative coolers, we require materials whose absorbance and radiance match the ATW. Additionally, to minimize heat absorption from the Sun, the metaconcrete should exhibit high reflectivity at frequencies outside the ATW range. Both absorbance and reflectance are inherent material characteristics linked to their optical dielectric properties. These properties originate at the atomic and electronic levels and can be accurately predicted by first principles calculations. To advance the ideas of photonic concrete, we provide an overview of the optical properties and excitonic spectra of relevant oxides. In our research, we compute the frequency-dependent dielectric function using various levels of approximation [5,6]. The electronic structure based on the Kohn-Sham (KS) approach often suffers from significant inaccuracies in band gaps and bands structures. A potential solution lies in the GW approximation, which corrects KS energy states by treating electrons and holes as screened quasiparticles [6]. Nevertheless, to accurately simulate experimental spectra, we must consider the two-body electron-hole interaction [7], which significantly influences spectral features around the band gap energy. This interaction can be accounted for by solving the Bethe-Salpeter equation (BSE) [7].

Our specific focus is on understanding the excitonic features of fundamental oxides such as CaO, SiO2, Al2O3 and Fe2O3 which serve as building blocks for complex cement oxides like alite (Ca3SiO5) and belite (Ca2SiO4). Additionally, we investigate other clinker phases, including gypsum (CaSO4.2H2O) and calcium sulphate (CaSO4), as well as hydrated phases like portlandite (Ca(OH)2) and tobermorite (Ca5Si6O16(OH)2.4H2O/ Ca5Si6O16(OH)2.7H2O), among others. The dielectric spectra obtained from these studies are utilized to examine the light scattering properties and serve as input for machine learning software aimed at predicting the ideal composition, shape, and distribution of the studied metamaterial. From a computational standpoint, we employ GGA [8] and SCAN meta-GGA [9] exchange-correlation ground state calculations, combined with G0W0 correction and the BSE approach as implemented in the VASP package [10].

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Nonlinear Plasmonics in Nanostructured Phosphorene

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Phosphorene has emerged as a candidate material for optoelectronics as well as extreme-ultraviolet and attosecond nanophotonics due to the excellent nonlinear optical properties as well as the possibility of actively tuning the light-matter interaction through chemical doping and patterning of the two-dimensional crystal. [1–4] In this work, [5] we introduce a second-principles framework based on density functional theory (DFT) and maximally localized Wannier functions (MLWFs) which enables the calculation of the plasmonic resonances as well as their related nonlinearities in the optical response of phosphorene nanoribbons (PNRs). We present highly tunable plasmonic resonances which due to the anisotropy as the crystal can be tuned both in terms of the edge termination, doping level, and width of the ribbons and which results an excellent nonlinear response and high-harmonic generation.

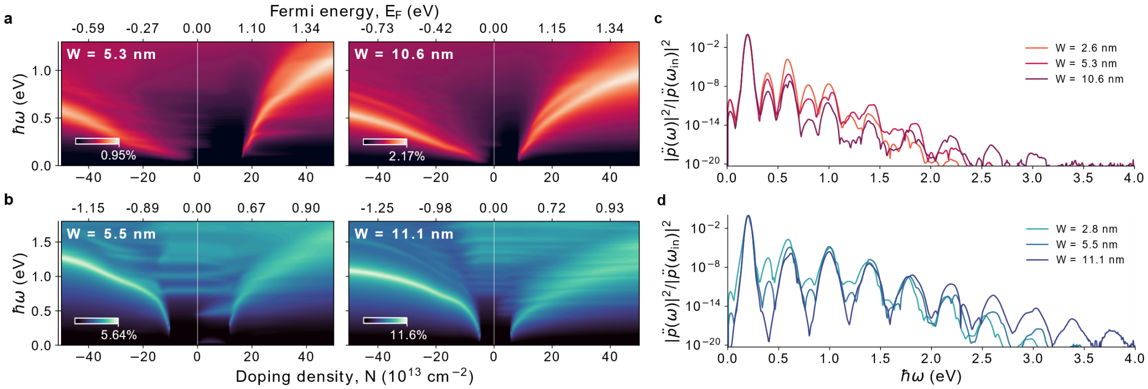

Figure 1: a-b Absorption cross section illustrating the resonances of the phosphorene nanoribbons with a armchair and b zigzag terminated edges when tuning the charge density and increasing the nanoribbon width. c-d High-harmonic generation at input frequency 0.2 eV. c shows the results for armchair terminated ribbons and d shows the results for zigzag terminated ribbons.

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In Silico study of chemical degradation of host molecular materials in OLEDs

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In this work, we present a computational study on host materials in OLEDs such as e.g., carbazole, triphenyl and sulphonyl types of host molecules. The nature of our study is two-fold; firstly, we investigate the degradation pathways of the above-mentioned molecules from a static point of view using state-of-the-art computational methods, secondly, we use semiclassical non-adiabatic dynamics to give more detailed insights into the degradation pathways of the host molecules from a dynamics point of view.

In the static analysis, we have calculated bond dissociation energies (BDEs) in charged and neutral ground states; to ensure the stability of the host molecules as charge carriers in the emissive layer of OLEDs. Double-hybrid DFT calculations were performed to estimate the BDEs. Further, we have also examined the stability in the excited-state, charged-excited state (arising from e.g., an exciton-polaron annihilation process) and from highly excited states (arising from e.g., an exciton-exciton annihilation process). [1] The excitation energies were computed with the algebraic diagrammatic construction theory of second order ADC (2) method.

In the semiclassical non-adiabatic dynamics study, we have used the fewest-switches surface hopping (FSSH) technique coupled to TD-DFT calculations to run dynamics for 1 ps. In the neutral-singlet state we have modelled two scenarios. In the first one, we are modeling the reactivity of the lowest excited state (i.e., starting the dynamics simulations from S1) while in the second scenario we mimic an exciton-exciton annihilation process, and thus we are starting our dynamics from a higher lying Sn state.[2]

To further push the limits of dynamics simulation, we have implemented a feedforward fully connected neural network (NN) that takes as input the atom coordinates of the molecule and makes predictions for energies and forces with a combined loss function. The model consists of 6 layers with 10,000 neurons per layer and can predict energies with mean absolute error (MAE) of 0.0376 - 0.0379 eV that meets “chemical accuracy” threshold (1 kcal mol^{-1} = 0.043 eV), and coefficient of determination (R2) as 0.999. For the force prediction, the MAE and R2 are 0.0449 - 0.0450 eV A^{-1} and 0.998 respectively. Both two results suggest almost linear correlations between NN predictions and QC references. [3]

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Computing X-ray absorption in strongly correlated materials beyond multiplet theory

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X-ray absorption spectroscopy (XAS) is routinely used to probe the electronic state and local environment of selected elements in solids. As the incident photon is absorbed, a core electron is excited to the open valence shell of the absorbing atom, and the measured XAS cross-section reflects the initial and final electronic states. In presence of strong electronic repulsion, the electrons tend to localize on the ions. Therefore the main features of X-ray absorption spectra on strongly correlated materials can be described in a local, ionic picture (multiplet theory [1]). However when the valence electrons still propagate or couple to excitations such as magnons or phonons, one needs to go beyond multiplet theory and consider larger models. This raises a computational issue, as diagonalising large entangled quantum systems is undoable in practice. The workaround consists in systematically truncating the computational Hilbert space according to some criteria. The choice of the truncation criterion distinguishes the few methods of "truncated wavefunctions", like Matrix Product States (MPS) or truncated Configuration Interaction (CI). In this talk I will present the recent advances in the calculation of beyond-multiplet XAS response [2-4], via such truncated wavefunction representations, focussing in particular on our work with an MPS-based impurity solver [5].

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Calculating Challenging Electronic Excitations in Molecules and Solids by Converging on Saddle Points of the Electronic Energy Surface

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Modelling electronic excitations of molecular and solid-state systems relevant for natural photochemical processes and energy-conversion applications poses a significant challenge. The size of the systems is usually too large for accurate, but computationally costly wave function methods, while the excitation typically involves charge transfer or other significant rearrangements of the electron density, which are problematic for cheaper linear-response time dependent density functional theory (TDDFT) calculations. Variational, orbital-optimized density functional calculations of excited states offer a computationally efficient alternative, yielding in many cases better results than TDDFT. However, widespread application of this time-independent approach faces the challenge that the excited state solutions are n-th order saddle points on a high dimensional electronic energy surface. I will present recent methodological advancements for orbital optimized calculations of excited states. An effective strategy involves estimating the saddle point order of the target excited state and identifying the degrees of freedom corresponding to negative curvature in a first step of constrained optimization, where the orbitals involved in the excitation are held fixed [1]. Thereby, approximate second-order direct optimization algorithms, such as a generalized mode following approach [2], can be used to converge on the target saddle point. We have applied this method in density functional calculations of large sets of charge transfer and Rydberg excited states of molecules, showing that significant improvements can be achieved compared to TDDFT calculations, even at the level of generalized gradient approximation (GGA) functionals [1,3]. The method can also be used in combination with explicit solvation models to elucidate the photoinduced dynamics of solvated metal complexes [4]. Calculations of the long-debate excitations in the charged nitrogen-vacancy center in diamond using meta-GGA functionals yield excitation energies in close agreement with high-level many-body calculations [5].

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Electronic transport calculations in different CDW phases of 1T-VTe2 and 1T-VSe2

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Charge density waves (CDW) occur in many solids, especially in materials of low dimensionality like transition metal dichalcogenides (TMDs). In this order, materials exhibit a spatial modulation of the charge and a periodic lattice distortion due to the coupling of the electrons to the lattice vibrations. Recently, it has been reported the possibility to switch CDW nanoscale domain in 1T-VTe2 monolayer by scanning tunneling bias pulses [1]. Motivated by these results we studied the electronic transport properties of the different CDW phases of 1T-VTe2 and 1T-VSe2 monolayers. First, we calculated the harmonic phonons of the system within the Density Functional Perturbation Theory (DFPT) as implemented in QUANTUM ESPRESSO [2],[3],[4]. We found phonons with imaginary frequencies corresponding to (4x1), (3x√3) and (√3x√21) lattice distortion for 1T-VTe2 and phonons with imaginary frequencies corresponding to (4x1) and (√3x√7) lattice distortion for 1T-VSe2. The eigenvectors of these phonon modes allowed us to set up the atomic structure for the different CDW phases. Then, we combined the DFT code Fireball [5] with a Keldysh-Green's formalism to calculate the electronic transmission through different CDW phases. Finally, with an applied bias on the system we calculated the electric current flowing in the structures. We found an order of magnitude of difference in the electric current with CDW phases oriented in an orthogonal direction compared to the current flow. These results combined to the manipulation of CDW phases are promising to build new nanodevices.

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Why Metavalent materials are so special? The case of cubic Antimony

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Higher chalcogenides such as GeTe, GeSe and Sb_2Te_3, have been recently proposed as a new class of materials, called metavalent, which has an intermediate character between metallic and covalent compounds. [1] Metavalent materials attract significant interest due to their remarkable properties such as moderate electronic conductivity increased coordination numbers incompatible with the (8–N) rule for semiconductors; large optical dielectric constants, $\varepsilon\infty$; large bond polarizability, as measured by Born effective charges, Z*; and large lattice anharmonicity, as measured by the Grüneisen parameter, $|\gamma TO|$.[2] In order to shed light on the peculiar properties of metavalent materials and better understand their origin, we have compared cubic antimony, the simplest compound of the family, with lead telluride in the rocksalt structure. While cubic antimony is unstable and metallic, lead telluride is stable and a small gap semiconductor. Still, their band structures reveal strong similarities. We have further analyses the origin of the instability of cubic antimony by calculating its static response function using time-dependent density-functional theory and making the link to the nesting properties of its Fermi surface [3]. Ongoing research is dealing with the dynamical response function (which can be measured by electron energy loss spectroscopy) and phonon properties.

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Ab initio study of the Fe-phthalocyanine/Cr2O3(001) spinterface

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In organic spintronics, the problem of understanding the interfaces of organic-inorganic heterostructures is acquiring major interest, so to coin the term spinterface[1]. In our research, we attempt to understand how the spinterfaces can be used in antiferromagnetic spintronics devices. In particular, the adsorption of organic molecules on antiferromagnetic substrates [2] offers a channel to modify the magnetic properties of the substrates through light, without the need of a strong spin-orbit coupling and consequent effect on the spin transport properties.

Transition-Metal (TM) oxides with an antiferromagnetic ground-state and low spin-orbit coupling are optimal candidates. Among them, Cr_2O_3 has the appealing feature to be in between a charge-transfer and a Mott-Hubbard insulator. Among the organic molecules used in spintronic devices, porphyrins and phthalocyanines (Pc) are quite known as versatile species. Thus, we consider here FePc, with a magnetic center increasing the magnetic coupling with the substrate. Therefore, through ab-initio DFT+U+VdW methods we study the Fe-phthalocyanine/Cr2O3(0001) system, determining the mechanisms of adsorption and its magneto-optical properties (at the independent particle level).

In particular, we point out the existence of a structurally meta-stable ferromagnetic state of the surface (meta-stable-FM), in addition to the stable antiferromagnetic one (stable-AF), and how these two different states of the surface imply a different magneto-electronic coupling with the molecule. From the magnetic perspective, the meta-stable-FM state favors an antiferromagnetic coupling between the substrate and the molecule, while the stable-AF favors a ferromagnetic coupling. From the electronic perspective, the former has no preferential adsorption site, while the latter strongly bonds to the molecular N atoms through the surface Cr atoms.

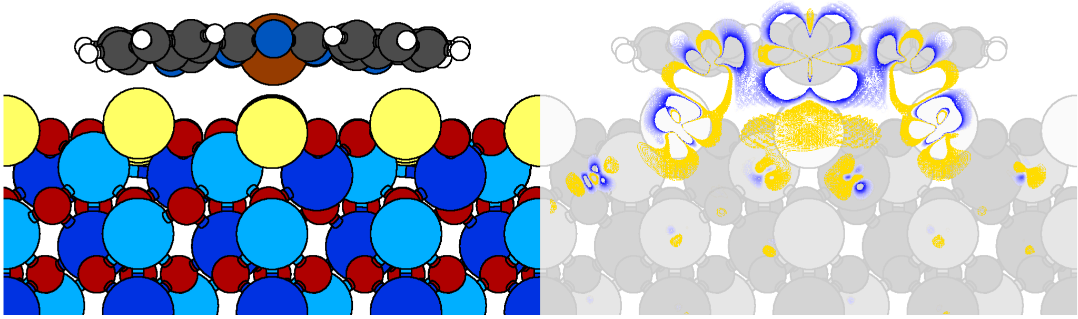

Fig. On the left, side view of the minimum adsorption configuration of the FePc/Cr2O3(0001) in the stable-AF termination of the Cr2O3(0001) substrate; on the right, its charge-density variation (yellow: increase; blue: decrease ; isolines in the range from −0.001 to 0.001 Å-3).

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Local and Non-Local Interactions in Strongly Correlated Materials: Embedding Through Auxiliary Particles

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Strongly correlated materials and molecules display unconventional electronic phases which can be readily accessed by tuning external parameters, making them remarkably promising for technological applications. In the road towards leveraging these systems for device design, an important role is played by theoretical models providing phenomenological descriptions of their electronic structure in a computationally affordable manner. In this talk, I will discuss how to capture the main spectral features of multi-orbital models relevant for quantum materials, such as iron pnictides or transition-metal perovskites, using a quasiparticle-based description. Moreover, I will show how to introduce the effect of non-local correlations in such descriptions, enabling the *ab initio* simulation of molecular systems. This is done within the recently introduced ghost Gutzwiller (gGut) framework, which can be seen as an embedding model deriving from the traditional Gutzwiller variational Ansatz. gGut captures strong electronic correlation in terms of an effective, quasiparticle Hamiltonian, allowing e.g., computationally inexpensive access to spectral functions. The key here is the introduction of auxiliary states, the eponymous ghosts, which model correlation in terms of one-body fluctuations, and which are ultimately projected out when computing observables. Crucially, despite this comparatively simple structure, the spectra obtained from gGut for correlated systems are qualitatively accurate in both low and high energy regimes. I will discuss how the reliability of the gGut framework and its comparatively modest computational cost make it a promising addition to the theoretical tool-set for material exploration.

DFT Simulation Study of Hexagonal Phosphorus Monolayer on Au(111) and Adsorbate Structure Exploration using Genetic Algorithm

I.-E. Michel, A. Karn, M. Lezoualc'h, C. Chacon, A. Smogunov, Y. J. Dappe and J. Lagoute

Understanding the electronic properties of surface adsorbates is crucial for interpreting experimental data from STM and LDOS measurements. A computational investigation is presented to elucidate the origin of a peculiar spike observed in experimental dI/dV spectra of Hexagonal phosphorus monolayer on a Au(111) surface. Using Density Functional Theory (DFT) calculations within the Quantum ESPRESSO framework, a comprehensive analysis is conducted, including band structure, projected band structure calculations, density of states (DOS), partial density of states (PDOS), local density of states (LDOS) in filtered Brillouin Zone, projected spatial charge densities and iso-DOS calculations. Results reveal that the spike originates from a flat band in the unoccupied states arising from the coupling between s and p_z orbitals of phosphorus atoms and the gold surface state. This insight provides a valuable understanding of the underlying electronic properties and mechanisms responsible for the observed experimental features.

Furthermore, a general simulation workflow is introduced, consisting in the joint utilization of Genetic Algorithm and DFT to explore adsorbate structures on surfaces, with the aim of predicting experimentally observable adsorbate structures. First results on the P/Au(111) system demonstrate the capability of this approach to identify common adsorbate structures such as pentamers, chains and clusters. However, further development is needed to allow for simulations of larger surface areas. Techniques such as Machine Learning and Cluster Expansion are potential candidates. This work lays the groundwork for my thesis, enabling the discovery and investigation of the growth and electronic properties of new strain-induced phosphorene allotropes.

Phonon angular momentum in magnetic systems

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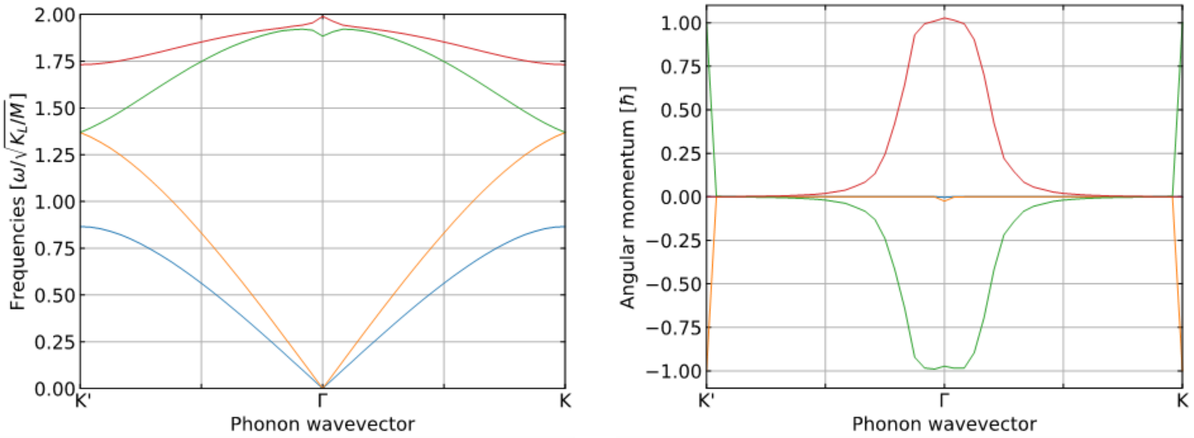
Phonons are quanta of vibrations in crystals, and carry energy and linear momentum. They can also carry angular momentum which originates from symmetry breaking. One can either consider a system with broken spatial inversion or broken time reversal symmetry. Depending on the broken symmetry, the phonon angular momentum exhibits different behaviors and characteristics.
In this talk, the focus will be put on phonon angular momentum originating from time reversal symmetry breaking (TRSB). In presence of TRSB, an extra term appears in the equation of motion of the ions. This extra term is the molecular Berry curvature, which acts as an effective magnetic field on the ions. An introduction to the molecular Berry curvature will be given through the Saparov model, which is based on the Haldane model. The impact of the molecular Berry curvature is two-fold: it will induce a splitting of the optical bands at Γ, and it also results in a non-zero net phonon angular momentum.



Phonon band structure and phonon angular momentum in the Saparov model.

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 Γ-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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Ab-initio variational wave functions for the time-(in)dependent many-electron Schrödinger equation

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Describing the ground states, excited states, and dynamics of many-electron quantum systems is crucial for applications such as predicting electronic structures in quantum chemistry, the properties of condensed matter systems, and the behaviors of complex materials. However, the real-time evolution of non-equilibrium quantum electronic systems poses a significant challenge for theoretical and computational approaches, due to the system's exploration of a vast configuration space. This work introduces a variational approach for fermionic time-(in)dependent wave functions, far surpassing mean-field approximations by capturing many-body correlations. The proposed methodology involves parameterizing the time-evolving quantum state using powerful neural networks, enabling us to capture close to all many body correlations. The time-dependent variational Monte Carlo technique is employed to efficiently compute the optimal time-dependent parameters. The results showcase the ability of our variational approach to accurately capture the time evolution of quantum states, providing insight into the quantum dynamics of interacting electronic systems, beyond the capabilities of mean-field. At the same time, the scaling behavior of the method is much more favorable than the commonly applied methods designed to capture correlations in quantum chemistry, such as FCI.

Microscopic theory of current-induced skyrmion transport and its application in disordered spin textures

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Magnetic skyrmions hold great promise for realizing compact and stable memory devices that can be manipulated at very low energy costs via electronic current densities. In this work, we extend a recently introduced method to describe classical skyrmion textures coupled to dynamical itinerant electrons. In this scheme, the electron dynamics is described via nonequilibrium Green's functions (NEGF) within the generalized Kadanoff-Baym ansatz, and the classical spins are treated via the Landau-Lifshitz-Gilbert equation. The framework is here extended to open systems, by the introduction of a non-interacting approximation to the collision integral of NEGF. This, in turn, allows us to perform computations of the real-time response of skyrmions to electronic currents in large quantum systems coupled to electronic reservoirs, which exhibit a linear scaling in the number of time steps. We use this approach to investigate how electronic spin currents and dilute spin disorder affects skyrmion transport and the skyrmion Hall drift. Our results show that the skyrmion dynamics is sensitive to the specific form of spin disorder, such that different disorder configurations lead to qualitatively different skyrmion trajectories for the same applied bias. This sensitivity arises from the local spin dynamics around the magnetic impurities, a feature that is expected not to be well captured by phenomenological or spin-only descriptions.

At the same time, our findings illustrate the potential of engineering microscopic impurity patterns to steer skyrmion trajectories.

Multi-channel Dyson equation: application to neutral excitations

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In this talk I will discuss a new method to calculate neutral excitation energies, which is based on the use of the four-body Green's function instead of the two-body one employed in standard approaches. More specifically, I will use a multi-channel Dyson equation [1], which couples the two-body and four-body Green's functions through a multi-channel self-energy that has the advantage of being static and containing only the bare Coulomb interaction. I will illustrate our approach by applying it to a two-level model which has single and double excitations [2]. Our method can correctly describe both kind of excitations, unlike standard approaches (e.g. Bethe-Salpeter with a static kernel), in good agreement with the exact results.

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Excitonic effects 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. This photon-in photon-out spectroscopy, with energy and momentum resolution, allows for element and orbital specificity, as well as access to a wide range of energies [1,2]. Despite its importance, simulating RIXS spectra remains a challenge for theory and often relies on multiplet approaches that approximate solid-state effects and are not parameter-free.

In my presentation, I will provide an introduction to the calculation of neutral excitations within the framework of many-body perturbation theory (MBPT) by solving the Bethe-Salpeter equation (BSE), which is a state-of-the-art technique for describing electron-hole interactions.

I will focus on corundum α-Al_2O_3, a prototypical example of a wide bandgap material with strong excitonic effects. I will discuss different levels of approximation for valence and core neutral excitations in Al_2O_3, and how their spectra, calculated within the BSE, accurately capture strong excitonic effects present in experiments [3,4]. Finally, I will analyze RIXS spectra in terms of excitation pathways [5], highlighting two different regimes: Raman and Fluorescence, that can be connected with complementary X-ray spectroscopies, such as inelastic X-ray scattering and X-ray emission spectroscopy.

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Design of Au-Based Nanoalloys as Plasmocatalyst

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The growing interest in gold-based nanoalloys arises from the possibility of combining the optical features of gold with the catalytic capabilities of other transition metals, leading to a new generation of highly-effective photocatalysts.[1] Among the many alloys, nanosystems where gold is mixed with other transition metals such as Pt, Pd or Rh are extremely promising since they can successfully catalyze several essential reactions such as hydrogen generation from water, oxygen evolution, and hydrogenation of double bonds.[2] Notwithstanding their relevance to date it is still not clear how do nanoalloys structural properties, such as shape and chemical ordering, affect the optical and photocatalytic features.[3] Our study provides a systematic investigation on the optical properties of Au-based nanoalloys, obtained upon coalescence of Au-icosahedra against an icosahedral metallic M-seed (with M=Pt, Pd, and Rh) at 600K. We simulated the alloys synthesis by means of classical Molecular Dynamics simulations4 and then applied Atomistic Classical Electromagnetic calculations to retrieve the nanoalloys optical absorption spectra.[5] Our calculations and further analysis with the Sapphire package[6], show a general tendency of Au atoms to diffuse above the M-seed, with the kinematics of the sintering process i.e., the time requested to get an almost spherical versus oblate shapes and the stabilization of one morphology versus another, strongly depending on the metal M. We find some clear correlation between the nanoalloys plasmonic peak intensities and structural descriptors of the systems shapes and chemical ordering. Such correlations have been further validated with calculations performed on model systems, which structures emphasize a specific descriptor over the others. Although there is room for significant improvements in the accuracy of the species interactions and in the modelling of the optical response, our work represents a fundamental step toward the comprehension of the interplay between chemical, structural and optical features in metallic nanoalloys, and will serve as a guide for future studies on their photocatalytic activity.

Keywords: nanoalloys, molecular dynamics, optical spectra, photocatalysis

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Fully Programmable Spatial Photonic Ising Machine

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Many developments in science and engineering depend on tackling complex optimization problems, which are difficult to solve on a large scale with standard computers. The challenge motivates intense search for new specific computing hardware. Recently, Optical Ising Machines have been demonstrated for computing the minima of spin Hamiltonians, paving the way to new ultra-fast hardware for optimization tasks formulable as Ising models and machine learning. However, the proposed systems lack scalability and are limited to solving a restricted class of problems. In this talk, we introduce a scalable and fully programmable optical spin-glass simulator based on spatial light modulation. Our design takes advantage of the Gauge method to encode both spins and couplings in the phase of the electric field, while exploiting the inherently parallelism of coherent light to compute multiple problems simultaneously. Using only one spatial light modulator, our design is simple to implement and scale up, eliminating the impact of pixel alignment and reducing the use of bulk optical components. Our results point out Spatial Photonics Ising Machines (SPIM) as a tool for spin-glass problems and provide a general route towards large-scale computing that exploits speed, parallelism and coherence of light.

In search of new phonon-limited superconductors.

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Since the observation of superconductivity a century ago, superconductors with excellent fabricability, low cost, and high transition temperature have remained rare. The search for new superconducting materials could benefit from a high-scale, automatic, computational framework. Here, we use the AiiDA computational infrastructure to automatically perform first-principles calculations using the Quantum ESPRESSO and EPW software, starting from the Materials Cloud 3-dimensional structure database (MC3D) to find novel BCS phonon-limited superconductors. From an initial screen, we performed high-accuracy calculation for over 300 dynamically stable structures and we found several categories of novel BCS superconductors, suggesting promising avenues for further experimental studies and practical applications.

Abstracts of poster presentations

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 exhibits 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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Bound states of H_2^+ using a multicenter approach based on complex Gaussian functions

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The behavior of molecules in a variety of chemical and physical environments is strongly determined by their quantum states. Therefore, an accurate theoretical description is essential for the understanding of a variety of phenomena. Moreover, in diverse fields such as astrophysics, radiobiology, and plasma physics, certain collision processes involve both bound and continuum molecular states. In this study, we aim to represent the bound and continuum states of the molecular ion H_2^+ using a set of complex Gaussian-Type Orbitals (cGTOs), that is to say GTO characterized by complex exponents. For continuum states, these functions have been employed so far only within a monocentric approach [1,2]. For H_2^+, we tackle the theoretical challenge of a multi-center system. To start with a simple model of a bound state, we express the molecular orbital ($1\Sigma_g^+$) as a linear combination of atomic orbitals (1*s*) given by a single Slater type function centered on both nuclei. Each Slater function is represented by an expansion of optimized cGTOs. The calculation of the molecular bound state energy, given by the expectation of the system's Hamiltonian, requires the evaluation of several integrals (Coulomb, exchange, kinetic...). With the cGTOs expansion we have been able to perform them analytically thanks to mathematical properties of Gaussian functions [3]. In a second step, we develop a computational code for a variational approach using a fixed cGTOs basis, and where only linear coefficients are optimized. This allows us to recompute the eigen-energies (refer to Figure 1) and generate the wave functions (see Figure 2) of the diatomic H_2^+ system, thus providing a clear improvement in the results. For the future, we plan to apply our cGTOs methodological approach to address the more challenging problem of the multicentric continuum.

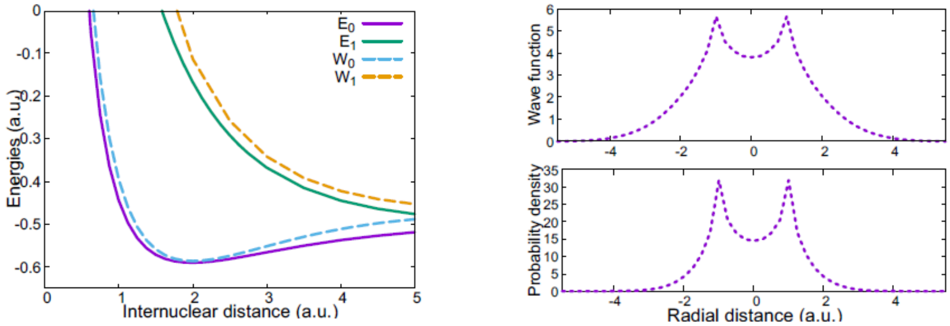

Figure1. Left: The energy curves of the bound state (E_0, W_0) and first excited state respectively (E_1,W_1), as a function of the internuclear distance. The solid lines correspond to the calculation using a fixed cGTO basis within the variational method, while the dashed lines correspond to the use of the initial Slater functions. Right: Representation of the ground state wavefunction as well as the probability density using cGTOs. The equilibrium internuclear distance is 2 a.u.

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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.

Ab initio modelling of electronic and magnetic properties of small Wigner molecules

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In this poster I will present my ongoing PhD research activity related to the study of electronic and magnetic properties of Wigner molecules in low dimensions. Wigner molecules are realized by means of the electron localization which takes place at low density (rS>>1, with rS the Wigner Seitz radius [1]); in this case the Coulomb potential dominates with respect to the kinetic energy of each particle. The theoretical study of these electronic molecules is motivated by the recent observation of their charge distribution in experiments [2,3,4], which allows the study of their magnetic ordering. In this presentation I will show my studies related to the electronic density profile at different values of rS for electrons confined to a one-dimensional (1D) box of length L at zero temperature. In particular I will present the crossover in the density profile between the low and strong interacting limit for different number of electrons.

These preliminary studies pave the way for the investigation of the magnetic properties of Wigner molecules and their excitations in 1D and 2D. I will also show that border effects could play an important role. This could lead to an effective Hamiltonian different from the Heisenberg model which describes the low energy excitations of a long electronic chain [5].

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Unveiling the optical secrets of ice surfaces: insights from advanced computational approaches

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Despite its ubiquitous presence, fundamental questions persist regarding the structure and properties of ice. Several research groups have investigated the proton order and proton disorder of different ice surfaces to better understand the behavior of ice at low temperatures [1-4]. Understanding how ice surfaces interact with light is pivotal for various applications, ranging from climate modeling to material science.

This study employs advanced computational methods, including Density Functional Theory (DFT) and beyond-DFT approaches such as GW and BSE, to investigate the equilibrium geometry, electronic structure, optical properties, and excitonic wave functions of ice surfaces. Additionally, Surface Anisotropic Reflectivity (RAS) is analyzed to uncover how variations in surface proton distribution affect their optical properties. Our primary objective is to elucidate the intricate relationship between surface structure and optical behavior, particularly focusing on how variations in surface proton distribution impact optical spectra. By examining different surfaces characterized by varying order parameters SOH, which quantify the distribution of dangling OH bonds, we aim to understand how alterations in surface proton distribution influence optical properties. Our findings promise to deepen our understanding of ice's optical characteristics, with potential implications for diverse scientific disciplines.

Keywords: Ice surfaces, optical properties, DFT, GW calculations, BSE, excitonic wavefunctions, RAS

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Advancing Photoemission Spectra Analysis with Three-Body Green’s Function Formalism

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Green’s Function formalism is one of the main methods used in materials science and condensed matter physics, particularly for predictive modelling of technologically significant materials. This study focuses on refining photoemission spectra analysis, crucial for understanding electronic structure and excitations in solids.

Photoemission spectra can be straightforwardly obtained from the one-body Green’s function, which is calculated solving the Dyson equation $G_1=G_0+G_0\Sigma G$, where G_0 is the noninteracting G and Sigma the self-energy, which contains all the many-body effects of the system and needs to be approximated. Currently used approximations struggle to describe satellite peaks, which are very important in strongly correlated systems, even in simple systems like Si [1].

To address this problem, some of us have recently proposed a three-body Green’s Function (3-GF) formalism, which shows promising results in accurately describing correlation effects in model systems. [2,3]

In this work I will present my preliminary results on the extension of the 3-GF formalism to finite temperature using the Matsubara Green’s Functions [4].

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Magnon lifetimes with frustration of exchange

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My research goal is to study and predict the behavior of magnons (quantized spin waves) and heat transport in topological materials (like $Co_3Sn_2S_2$). This is done with the use of density functional theory, Wannier functions and spin dynamics calculations. As a stepping stone to this I study the influence of long- and short-range Heisenberg exchange on magnon lifetimes. With these lifetimes it is possible to determine transport coefficients for phenomenological and DFT based Heisenberg Hamiltonians.

In my poster I will show Heisenberg exchange parameters for hcp Cobalt and $Co_3Sn_2S_2$, from DFT and TB2J, together with magnon lifetime simulations for phenomenological Hamiltonian constructions with frustration of exchange.

Electronic properties of nanopatterned hydrogenated graphene superlattices

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Contemporary mobile networks and emerging wireless technologies (internet of things etc.) require broadband channels to exchange rapidly a huge amount of data [1,2]. Harnessing frequencies from 0.1 to 10 THz has potential to increase the state-of-the-art high speeds by 2 orders of magnitude [3]. However, despite the latest advancements in THz technology [4], we still lack proper materials to realize suitable THz photonic components. The route to compact THz devices is seen in the usage of carbon nanostructures as building blocks for detectors, emitters and passive components. Such carbon nanostructures, as nanotubes and graphene nanoribbons, exhibit unique electronic and optical properties that make them very promising candidates for THz components [5]. However, carbon nanotube and nanoribbon monolithic on-chip integration is challenging because it may result in significant change of their intrinsic properties after an embedment into a substrate. We investigate with first principles methods the successful integration of nanoribbons into a single-layer graphene and show that their electronic properties persist in the integrated structures forming a 2D graphene superlattice.

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Engineering site mixing to understand magnetic topological insulators through DFT calculations

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The discovery of magnetic topological matter over the past decade has heralded an era of breakthrough proposals that offer great potential for next-generation technologies. This field has demonstrated that combining nontrivial band topology with magnetic order can lead to the emergence of exotic phenomena such as the Quantum Anomalous Hall Effect (QAHE) [1]. The main purpose of my research project is to combine experiment and theory to design best performance candidates for the QAHE e.g. materials that are stable up to technologically relevant temperatures. The van der Waals compound MnBi2Te4 was the first intrinsic magnetic topological insulator used as a material platform to explore the QAHE [2,3,4]. Its structure, comprising septuple layers of Te-Bi-Te-Mn-Te-Bi-Te stacked along the crystallographic c-axis, provides an expansive playground for the introduction of antisite defects. Recently it has been shown by our group that tuning the stoichiometry of the MnSb2Te4 material by Mn/Sb antisite mixing (Mn in the Sb site and vice versa) is an effective way to increase the magnetic ordering temperature and to obtain net spin-polarized states [5]. This poster addresses the theoretical aspects of my work, in which first-principles calculations using Density Functional Theory (DFT) are performed by means of the VASP code. Based on experimental data, we aim to build supercell models to simulate realistic fractional occupancies and different Mn distribution patterns of the potential QAHE candidate MnSb2Te4. The calculations not only enable the analysis of the bulk band topology and magnetic ground states of our intermixed models but also provide a deeper understanding of the impact of Mn/Sb antisite defects on the magnetic anisotropy of the compounds. Eventually, we aim to extend the investigation by correlating the theoretical results with experimental data through Curie temperature calculations and by constructing fully ab initio tight-binding Hamiltonians via the Wannier90 code. These Hamiltonians are used to reproduce surface band structures to compare to ARPES measurements and to compute the Z2 invariant, a crucial determinant of nontrivial topological states.

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