XI Workshop on Novel Methods for Electronic Structure Calculation

XI WNMESC

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La Plata, Buenos Aires
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Dear friends and colleagues, again this year, as we have done every odd year since 2005 we are organizing the Workshop on Novel Methods for Electronic Structure Calculations, this time will be their 10th edition.
The workshop will be dedicated to providing an up-to-date with the different mechanisms of calculation on materials, as well as the study of different materials. In general, during the workshop an intense exchange is targeted between speakers and participants.

Always the relaxed atmosphere that has been generated at the meeting has allowed communication between speakers and participants, generating ideas for new lines of work, participation in projects, etc. As in the past it will be a great pleasure that you can participate in the workshop and a great joy that we can meet again or visit us first time.

The workshop will be held in the city of La Plata (Argentina), in the National University of La Plata, between 16 and 17 December 2024.

Previous editions

- 2005: Seminar on ab-initio solid state calculations, La Plata – Argentina.
- 2007: Workshop on novel methods for electronic structure calculations and 6th FPLO hand-on, La Plata – Argentina.
- 2009: Workshop on novel methods for electronic structure calculations, La Plata – Argentina.
- 2011: 4th Workshop on novel methods for electronic structure calculations, and first southamerican congress on materials, La Plata – Argentina.
- 2013: V Workshop on novel methods for electronic structure calculations, Montevideo – Uruguay.
- 2015: VI Workshop on novel methods for electronic structure calculations, La Plata – Argentina.
- 2017: VII Workshop on novel methods for electronic structure calculations, Santiago de Chile – Chile.
- 2019: VIII Workshop on novel methods for electronic structure calculations, La Plata – Argentina.
- 2021: IX Workshop on novel methods for electronic structure calculations, La Plata – Argentina.
- 2023: X Workshop on novel methods for electronic structure calculations, La Plata – Argentina.

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EXACTAS

	Monday 16	Tuesday 17
09:00 – 09:15	Opening talk	
09:15 – 10:00	María C. Asensio	Silvana Tumminello
10:00 – 10:45	Pablo Esquinazi	Emilio Artacho
	BREAK	
11:00 – 11:45	Valeria Ferrari	Jorge Facio
11:45 – 12:15	Harry Saltos Sánchez	Helena Ramírez
	LUNCH	
14:15 – 15:00	Perla Balbuena	Michelle Johannes
15:00 – 15:45	Robert Guzmán Arellano	Manuel Richter
	BREAK	
16:00 – 16:45	Félix Requejo	Martín Esteves
16:45 – 17:30	Leandro Socolovsky	Verónica Sánchez

LIST OF SPEAKERS

- **María C. Asensio**
  Materials Science Institute of Madrid (ICMM), Spanish National Research Council (CSIC), Spain.
- **Manuel Richter**
  Leibniz Institute for Solid State and Materials Research IFW Dresden, Germany.
- **Jorge Facio**
  CONICET, Instituto Balseiro, Centro Atómico Bariloche, Argentina.
- **Harry Saltos Sánchez**
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- **Silvana Tumminello**
  German Aerospace Center DRL, Institute of Materials Research Metals and Hybrid Materials, Germany.
- **Emilio Artacho**
  Theory Group Nanogune, Spain – Theory of Condensed Matter, Cavendish Laboratory, University of Cambridge, UK.
- **Valeria Ferrari**
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- **Martín Esteves**
  Facultad de Química, Universidad de la República, Uruguay.
- **Verónica Sánchez**
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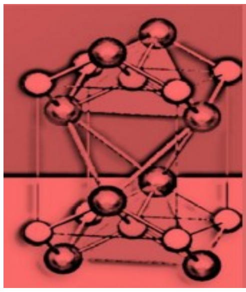
ORAL TALKS



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NanoARPES and Electronic Imaging: A Cutting-Edge tool to Direct Electronic Structure determination of nanomaterials

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Angle-Resolved Photoemission Spectroscopy (ARPES) has emerged as a foundational technique in the study of advanced materials due to its unique capability to directly probe electronic structures within momentum space. The complex physics underlying these materials is profoundly influenced by many-body interactions and electronic confinement, shaped by concepts such as symmetry and topology that have revolutionized our understanding of matter. ARPES has played a pivotal role in the discovery and characterization of materials exhibiting strongly correlated electronic states and nontrivial topological properties. Over the past two decades, ARPES has seen remarkable advancements in angular, spatial, temporal, and spin resolution. These technical innovations, coupled with novel material synthesis methods, have paved new avenues for exploring and manipulating nanomaterials. This presentation reviews the latest developments in ARPES, with a particular focus on NanoARPES, highlighting scientific breakthroughs1-6 enabled by these advancements and emphasizing the critical role of this technique in modern methods for electronic structure calculations.

[1]. Nano-ARPES, Wikipedia. Website: https://en.wikipedia.org/wiki/Nano-ARPES

[2]. Z. Gao, et al., Large-area epitaxial growth of curvature-stabilized ABC trilayer graphene, Nat. Commun. 11 (2020) 546.

[3]. V.L. Nguyen, et al., DOI: doi.org/10.1038/s41565-020-0743-0. This Nat. Nanotechnol. (2020) article is the results of an outstanding international collaboration with Prof. Dr. Young. LEE (Seoul, Korea).

[4]. Y. Ma et al., this Nature Communications paper (2017) demonstrates for the first time the charge spin separation in unidimensional ground boundaries in DOI://doi.org/10.1038/ncomms14231.

[5]. V.L. Nguyen, et all., Author Correction: Layer-controlled single-crystalline graphene film with stacking order via Cu–Si alloy formation, Nat. Nanotechnol. 16 (2021) 114–116. https://doi.org/10.1038 /s41565-020-00821-z.

[6]. C. Bao, et all., Spatially-resolved electronic structure of stripe domains in $IrTe_2$ through electronic structure microscopy, Commun. Phys. 4 (2021) 229. https://doi.org /10.1038/s42005-021-00733-x.

Environmental stability of $Cr_2Ge_2Te_6$

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Some layered magnetic materials like $Cr_2Ge_2Te_6$ sustain magnetic long range order even in the atomically thin limit and have sparked intense research efforts to implement them in nano-spintronic devices. On the other hand, there are serious practical obstacles to such plans due to their poor environmental stability. In order to actively prevent rapid degradation, a microscopic understanding of the involved reactions and energy scales is needed. Here we perform an in-depth investigation of the $Cr_2Ge_2Te_6$ surface by x-ray photoemission spectroscopy in combination with density functional theory. We find that the surface degradation is driven by germanium oxidation. Tellurium oxidation proceeds in two steps involving an intermediate state of partial oxidation. Oxidation starts rapidly at the fresh surface but slows down afterwards continously. Our results single out germanium as the main driver of the initial steps of surface degradation of $Cr_2Ge_2Te_6$.

Oxide-Water Interfaces Under Radiation: Initial Findings and Future Challenges

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The interaction between ionizing radiation and oxide-water interfaces is a key topic in material science research in the quest for materials with technological applications, such as nuclear waste management, metal corrosion in aqueous environments, and biological systems exposed to radiation. This talk will explore atomic-level processes occurring at these interfaces, with particular emphasis on hydrogen generation in oxides such as zirconia and copper oxide, both of which are used in nuclear facilities.

The current challenges in understanding the mechanisms behind these processes through electronic structure methodologies will be discussed, including density functional theory (DFT) and many-body perturbation theory (MBPT) approaches, such as the GW approximation and the Bethe-Salpeter equation (BSE). Our preliminary results in these materials, focusing on excitonic properties and radiation-induced processes, will also be presented.

This work is part of the Horizon project titled ”Materials Radiation: From Basics to Applications” (MAMBA), which aims to deepen the understanding of material responses to irradiation and apply this knowledge to tailor and control the properties of materials exposed—either intentionally or unintentionally—to intense radiation environments. Finally, the talk will outline the future steps in this research, including modeling different oxide facets, analyzing excitonic wave functions, and using machine learning to describe the corresponding oxide-water interfaces.

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Computational Approaches to the Design of Lead-Free Perovskite Solar Cells

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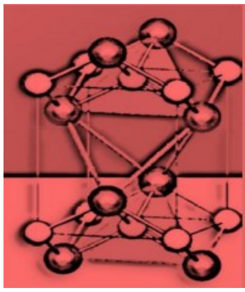
Perovskite solar cells (PSCs) have shown significant promise as an efficient alternative in the renewable energy sector, demonstrating impressive results in capturing solar energy. A key component of these materials is inorganic perovskites, such as $CsPbX_3$ (X = Cl, Br, I), which have band gap values ranging from 1.72 eV for $CsPbI_3$ to 2.31 eV for $CsPbBr_3$.

However, the presence of lead (Pb) in these compounds raises environmental concerns, particularly because it can become a contaminant at the end of the cells'; lifespan.

This study aims to explore lead-free alternatives or those with reduced Pb content by focusing on the theoretical and computational investigation of the physical properties of the compounds $CsGe_nPb_{1-n}X_3$ and $CsSn_nPb_{1-n}X_3$ (X = Cl, Br, I; $0 \leq n \leq 1$). We look at key aspects such as formation energy, electronic structure and band gap. To accomplish this, we conducted a series of ab initio calculations based on quantum mechanics using Density Functional Theory (DFT), along with some machine learning techniques for added insight.

Additionally, this research assesses the structural and electronic properties of these compounds, examining their energetic stability, photovoltaic efficiency, and other essential characteristics. We also considered the Jahn-Teller effect and its impact on the structural stability of the cells, exploring different configurations of the octahedral structures in the materials.

Our ab initio calculations were carried out using the Pseudopotential plus Plane Wave method implemented in the Vienna Ab initio Simulation Package (VASP). These calculations were complemented by numerical simulations of solar cell efficiency using SCAPS-1D software to identify the optimal configuration for maximum power conversion efficiency by testing ZnO for the electron transporting layer and Cu_2O hole transporting layer.



Understanding mechanisms of ion and electron transport in reconfigurable electrochemical neuromorphic components

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New materials are emerging as promising components of reconfigurable electrochemical random access memory devices. Among them, transition metal oxides such as those containing vanadium, tantalum, titanium, have shown interesting behaviors regarding their abilities to emulate neuronal behavior, including synapses and other neuronal conductance characteristics, which make them potential candidate components for neuromorphic analog computing devices. In parallel, materials based on molecular complexes able to display switchable redox behavior appear as another set of promising components that suitably arranged as molecular films may yield artificial neuronal behavior. Here we analyze the evaluation of ion and electron transport characteristics in these two sets of materials using first principles computational tools, and discuss the fundamental physical and chemical aspects that may drive targeted conductance and memory retention properties. Using first principles theory and simulations we highlight similarities and differences between these two sets of materials regarding their conductance properties and neuromorphic behavior.

Electronic and magnetic properties of graphene-fluorographene nanoribbons: Controllable semiconductor-metal transition

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We investigate the electronic and magnetic properties of graphene channels (2–4 nm wide) embedded within fluorographene, focusing on two distinct interfaces: the fully fluorinated α 'alpha' interface and the half-fluorinated β 'beta' interface. Density functional theory (DFT) calculations reveal that $\alpha\alpha$ systems exhibit semiconducting behavior with antiferromagnetic ordering, closely resembling pristine zigzag graphene nanoribbons. In contrast, $\alpha\beta$ systems display ferromagnetism and a width-dependent semiconductor-to-metal transition. To enable the study of larger systems, we develop and validate effective Hubbard models for both $\alpha\alpha$ and $\alpha\beta$ channels.

In order to determine the similarities and differences between graphene channels and graphene nanoribbons, where both of which have similar crystalline structure, we studied the graphene channel dependence with the degree of fluorination at the channel edges. A good observable is the localization of the states near the Fermi level. The states localized near the α interfase have almost all their weight on the graphene channel and these penetrate evanescently on the fully fluorinated graphene regions. On other hand the states localized near the β interfase, which are similar to the edge states of zigzag or Klein graphene nanoribbons, are less localized than those of graphene zigzag nanoribbons.

The effective Hubbard models were build upon DFT results and a Wannier function analysis, these models accurately reproduce the electronic structure and magnetic ordering observed in DFT calculations. Crucially, our $\alpha\beta$ model successfully captures the semiconductor-to-metal transition.

Application of this model to larger systems reveals the persistence of a ferromagnetic state with spin polarization localized at the α edge. Our results demonstrate the potential of fluorination for targeted property engineering and provide a basis for exploring graphene-fluorographene systems in device applications ranging from microelectronics to spintronics.

Stability and Properties of Small Atomic Clusters: Complementarity Between In Situ X-ray Absorption Experiments and Simulations.

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The geometry and electronic structure of transition-metal nanoparticles change drastically when their size is reduced below 1-1.5 nm (i.e., fewer than 100 - 150 atoms) due to quantum confinement effects.[1][2] In this size regime, the metallic band structure transforms into a series of discrete electronic levels, giving rise to novel properties distinct from those of larger nanomaterials or bulk metals. The discretization of energy levels causes sub-nanometer-sized metal clusters to behave like atomic-scale semiconductors, preventing collective phenomena such as localized surface plasmon resonance (LSPR), which is typically observed in larger metallic nanoparticles.[3] When clusters consist of only a few atoms, a network of d-orbitals interconnects the metal atoms, with inter-atomic distances comparable to chemical bonds ($1–2\AA$). The ”floppy”; nature of these structures results in structural fluxionality,[4] a characteristic that has the potential to enhance catalytic activity. Recently, these sub-nanometer-scale materials have garnered significant interest in the field of catalysis, particularly as catalysts composed of only a small number of atoms, providing unique properties and new catalytic mechanisms.[5]

For characterization, X-ray absorption near-edge structure (XANES) spectroscopy and near-ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) are employed as powerful, noninvasive methods to directly determine the structural and electronic state of each chemical element in the clusters. When used in ”in situ”; or ”operando”; modes, these techniques offer direct visualization of the system under realistic conditions, i.e., under thermodynamic equilibrium with its environment.[6]

In this work, we demonstrate the sensitivity of these experimental techniques in determining both structural and chemical aspects of Cu atomic clusters, even under sub-nanometric conditions. These measurements also provide a foundation for subsequent simulations, enabling us to describe the origin of the stability and properties of these systems, particularly in relation to oxygen, as a function of concentration and temperature when supported on HOPG.

[1]. P. Jena, Q. Sun, Chem. Rev. 2018, 118, 5755–5870..

[2]. M. Zhou, C. Zeng, Y. Chen, S. Zhao, M. Y. Sfeir, M. Zhu, R. Jin, Nat Commun 2016, 7, 13240.

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[4]. P. López-Caballero, R. Garsed, M. P. de Lara-Castells, ACS Omega 2021, 6, 16165–16175.

[5]. V. Porto, D. Buceta, B. Domínguez, C. Carneiro, E. Borrajo, M. Fraile, N. Davila-Ferreira, I. R. Arias, J. M. Blanco, M. C. Blanco, J. M. Devida, L. J. Giovanetti, F. G. Requejo, J. C. Hernández- Garrido, J. J. Calvino, M. López-Haro, G. Barone, A. M. James, T. García-Caballero, D. M. González-Castaño, M. Treder, W. Huber, A. Vidal, M. P. Murphy, M. A. López-Quintela, F. Domínguez, Advanced Functional Materials 2022, 32, 2113028.

[6]. D. Buceta, S. Huseyinova, M. Cuerva, H. Lozano, L. J. Giovanetti, J. M. Ramallo-López, P. López-Caballero, A. Zanchet, A. O. Mitrushchenkov, A. W. Hauser, G. Barone, C. Huck-Iriart, C. Escudero, J. C. Hernández-Garrido, J. J. Calvino, M. López-Haro, M. P. de Lara-Castells, F. G. Requejo, M. A. López-Quintela, Chem. - Eur. J. 2023, 29, e202301517.

Some questions that appears when doing experiments with magnetic nanostructures

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In this talk we will present some topics of our experimental research on magnetic nanostructures. We will present the quest for understanding interacting cluster glass or superferromagnetism states; and giant magnetoresistance and electric transport in graphene-based nanoheterostructures. The first situation happens when a dispersion of magnetic nanoparticles is immersed in a non-magnetic medium. When concentration of these nanoparticles is low, meaning that each one are far from the others, the phenomenon called superparamagnetism takes place. If nanoparticle concentration increases, dipolar interaction takes place. Some models where developed to address the issue, but they fail when nanoparticle concentration is higher. For that situation, the observed magnetic behavior is called interacting cluster glass or superferromagnetism. There are not reliable models to explain the observed features.

The second situation that requires a theoretical explanation is observed in nanoheterostructures consisting of magnetic nanoparticles attached to reduced graphene oxide layers. At first, such a structure should display negative magnetoresistance at low temperatures and electric conduction that should follow a variable range hopping model. These behaviors where not observed in several measurements performed on this system.

How the Effective Bond Energy Formalism (EBEF) benefits from first-principles results

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Materials development and implementation relies nowadays on multiscale computational simulations to efficiently design the experimental steps of materials/components from their production and life-cycle to their reuse or recycle [1,2].

In the mesoscale range, materials simulations make intensive use of calculated phase diagrams and thermodynamic properties with commercial and open-source thermodynamic software and encoded thermodynamic information developed with the CALPHAD method (Calculation of Phase Diagrams and Thermochemistry) [3]. The strength of this method is the excellent ability to make reliable extrapolations in temperatures and compositions within the accuracy of experimental measurements. Several models are used to describe all the stable and metastable phases in a given material system, most of them are based on the Compound Energy Formalism that can greatly benefit from electronic methods, such as DFT (Density Functional Theory), incorporating energies of hypothetical compounds (end-members). The Effective Bond Energy Formalism is a recently developed method [4,5,6] which benefits from DFT calculated end-members to obtain effective bonding energies, and in this way, improving the extrapolation behavior in high-order material systems and reducing model parameters achieving lower computational effort and time.

We present examples of significant technological interest, sigma and mu phase in steel, Ni-based and Co-based superalloys, where EBEF has been tested and validated against experimental measurements, software where this method is already implemented and potential application cases.

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[4]. Dupin N, Kattner UR, Sundman B, Palumbo M, Fries SG, "Implementation of an Effective Bond Energy Formalism in the Multicomponent Calphad Approach". J Res Natl Inst Stan, 23 (2018) 123020. https://doi.org/10.6028/jres.123.020.

[5]. J. Pereira dos Santos et al. "Applying the Effective Bond Energy Formalism (EBEF) to Describe (σ) the Sigma Phase in the Co-Cr-Ni-Re System", J. Phase Equilib. Diffus. (2023). https://doi.org/10.1007//s11669-023-01079-3

[6]. Dupin N, "The Effective Bond Energy Formalism", TofA Meeting 2024.

Electronic stopping power for nuclei traversing matter from real-time time-dependent density-functional theory

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The study of particles (mostly nuclei) shooting through matter has a long history and is of relevance to understand radiation damage in important applied contexts, such as nuclear engineering, space, and radiotherapy. From the basic viewpoint radiation-damage processes are important as paradigmatic problems in the physics far from equilibrium. For projectiles with initial velocities around 0.2 a.u. or more the projectile slows down mostly by exciting the electrons of the target. Among the several models, theories and first-principles techniques to study electronic stopping processes, in 2007 we started a line of research based on the computational replication of those processes from first principles: In a simulation box of around 100-1000 atoms we put a projectile somewhere and start moving it across the box while the wave-functions evolve, within the framework of real-time time-dependent density-functional theory. The method involves many approximations, from technical to fundamental, but it has shown to be usefully accurate in its prediction of electronic stopping power at least in some cases. In this talk I will introduce the method, illustrate it with prototypical results, discuss some theoretical considerations around it, and present the latest results in the context of better understanding low-velocity threshold and Barkas effects in insulators and our still insufficient progress in addressing electron projectiles, of great relevance in radiotherapy.

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Symmetry-obstructed Peierls transition in a topological semimetal

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In many quantum materials, the properties of interest emerge from the interplay between electronic and structural instabilities. A textbook example is the Peierls transition in one- dimensional systems, where a reduction in translational symmetry—driven by the energy gain from associated electronic reconstruction—yields a metal-to-insulator transition. In this talk, we will consider the case of $PtBi_2$, a polar compound whose structure derives from a centrosymmetric phase with larger translational symmetry. Through first-principles calculations, we will show that these two phases are connected by a metal-to-semimetal transition that, similar to the Peierls transition, is driven by an attempt to open a gap in the electronic structure.

In $PtBi_2$, however, this gap fails to fully develop due to constraints imposed by the symmetry of the underlying crystal distortion. Our results not only help us understand the origin of the nontrivial topology in this compound but also illustrate how the Peierls transition can be enriched in higher dimensions through the preservation of additional symmetries.

Impact of the Poole-Frenkel effect on the electric transport of hydrogenated amorphous silicon solar cells

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The Poole-Frenkel effect (PFE) on the electrical transport of a-Si:H based p-i-n solar cells in the initial state was studied using computer simulations. The PFE has been implemented in our computer code by modifying the occupancy function and recombination rate expressions in the Shockley-Read-Hall formalism. The capture cross sections of charged states were modified by including a dependence directly proportional to the square root of the electric field, while the capture cross sections of neutral states were left unchanged. We analyze the impact of the PFE on the dark and light current-voltage characteristic curves when the effect is adopted in each layer separately and throughout the device. We found that the enhanced recombination and lower trapping through charge localized states near the interfaces can explain the changes in the current-voltage curves. Under AM1.5 illumination, the current density at low forward voltages and short circuit conditions decreased mainly when the PFE was implemented inside the intrinsic layer, while the open circuit voltage increased depending on the density of defects adopted inside the p-layer. Our results can be used to explore if the inclusion of the PFE enables a better design of a-Si:H and μc-Si:H solar cells.

Design and optimization of energy materials through computational analysis

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Although batteries and fuel cells are generally considered electrochemical systems, a surprising amount of their performance stems from the physics of the materials that make up their basic components: anode, cathode and electrolyte. Ionic conduction, electronic conductivity, chemical stability and voltage can all be traced back to intrinsic materials properties which are governed by fundamental physics, and specifically by the quantum mechanical properties of their electronic structure. In this talk, I will discuss how computational simulation (mainly DFT) can be used to gauge how these microscopic and atomistic properties of materials enhance or detract from the macroscopic performance in an electrochemical environment.

First, I will contrast oxide and phosphate batteries to show how the energy nearness of the transition metal d-states to the oxygen p-states introduces a bonding-antibonding pair that determines both the voltage and the stability against outgassing which potentially leads to flammability. This necessarily induces a trade off between stability and performance, and I will suggest some methods of mitigating this intrinsic contradiction. I will show how computational analysis of oxide coatings pointed towards a method of stabilizing inherently unstable materials against degradation that is currently being used as industry standard. Finally, I will relate the concepts learned from Li$^+$ diffusion in batteries to p$^+$ diffusion in solid acid fuel cells and introduce a new family of phosphide materials that were developed using computational prediction and analysis.

Room temperature superconductivity: State of the Art

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In recent years, a significant number of publications in high-citation journals have reported that hydrogen compounds under pressure may exhibit superconductivity at room temperature; however, these findings and their interpretations remain highly controversial. Conversely, for the last 50 years, several independent studies from transport, magnetization, and magnetic force microscopy—published in less prominent journals—have indicated the presence of superconductivity in pure graphite samples, with critical temperatures surpassing 300K. Experimental and theoretical studies suggest that superconductivity in graphite occurs at specific two-dimensional stacking faults or interfaces, which constitute a negligible mass compared to the surrounding non-superconducting graphite matrix [1]. The limited amount of superconducting material, the unexpectedly high critical temperatures, added to the considerable skepticism within the scientific community have contributed to this phenomenon remaining largely "hidden" [2]. Recent research in Grenoble, France, employing simple methods to isolate superconducting graphite particles from their non-superconducting counterparts, has obtained an increased superconducting yield, revealing superconducting onset temperatures around 700K and zero resistance up to 500K [3].
This presentation will summarize recent findings, highlighting the potential impact of these results on nano- and microelectronics.

[1]. Evidence for room temperature superconductivity at graphite interfaces. P.D. Esquinazi, C.E. Precker, M. Stiller et al., Quantum Studies: Mathematics and Foundations 5, 41-53 (2018).

[2]. Invited review: Graphite and its hidden superconductivity. P. Esquinazi, Papers in Physics 5, 1-15 (2013).

[3]. Magnetic field sorting of superconducting graphite particles with T_c>400K, Manuel Núñez-Regueiro et al., arXiv:2410.18020 (2024).

Structural, electronic and optical properties of TiO_2 nanoparticles and derived sodium titanate nanotubes $Na_2Ti_2O_5.H_2O$ doped with transition metals (Fe, Cu, Ni). A theoretical-experimental study

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This work presents the synthesis, structural characterization, and optical properties of anatase nanoparticles, both pure and doped with transition metals (Fe, Cu, Ni), obtained via modified sol-gel methods [1]. Additionally, it compares these nanoparticles with doped Na_2 $Ti_2O_5.H_2O$ titanate nanotubes derived from the precursor nanoparticles. A structural model for onedimensional nanostructures is proposed, based on first principles, which successfully accounts for the complex diffraction patterns observed in the system [2].
The optical properties, particularly the bandgap energies and Urbach tails, were studied to enhance the model describing the absorption edge of the materials. This study aims to understand the impact of doping atoms on the electronic properties and, consequently, the optical behavior of the materials. The main findings indicate that the anatase nanoparticles exhibit no significant changes in bandgap energies. However, a moderate red shift is observed in the indirect bandgap energies of the nanotubes. All doped samples show higher Urbach energies compared to the pristine ones, suggesting the introduction of impurity electronic states within the bandgap. Furthermore, greater optical activation is noted, evidenced by higher Urbach energies in sodium dititanate systems compared to anatase nanoparticles. These observations are consistent with our electronic and optical DFT [3,4] calculations. The phenomena can be interpreted in terms of the hybridization characteristics of the impurity states introduced into the electronic gap.

[1]. Esteves, M., Amy, L., Fernández-Werner, L., Marotti, R. E., Faccio, R., & Mombrú, A. W. (2023). The Journal of Physical Chemistry C.

[2]. Esteves, M., Fernandez-Werner, L., Pignanelli, F., Romero, M., Chialanza, M. R., Faccio, R., & Mombru, A. W. (2020) Ceramics International, 46(3), 2877-2886.

[3]. G. Kresse, J. Furthmüller, Physical Review B, 54 (1996)

[4]. G. Kresse, J. Furthmüller, Computational Materials Science, 6 (1996).

Unraveling Iron Oxide Nanoparticle Reactivity via Integrated Experimental and Computational Approaches

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With the increasing urgency to mitigate environmental degradation, the promotion of sustainable, eco-friendly industrial processes is imperative. In this context, enzyme-mimicking nanomaterials, or nanozymes, have emerged as promising alternatives, offering high stability and reusability. Among these, iron oxide nanoparticles (NPs) exhibit intrinsic peroxidase-like catalytic activity, efficiently decomposing H_2O_2 and oxidizing organic compounds. Despite extensive research on their catalytic properties, the precise reaction mechanisms governing their activity remain unresolved, particularly with respect to their structural characteristics and the influence of the surrounding medium.
This study focuses on nanozymes composed of Fe_3O_4 (magnetite) and Fe_2O_3 (hematite) nanoparticles. Using density functional theory (DFT) and classical molecular dynamics simulations, we explore the chemical reactivity of these materials in both vacuum and under microsolvation by water. The climbing image nudged elastic band (CI-NEB) method is employed to locate transition state structures, enabling the determination of thermodynamic feasibility and kinetic barriers of the H_2O_2 decomposition reaction. We analyze the (001) surface of both materials and relate our findings to experimental data, including electron paramagnetic resonance (EPR) and transmission electron microscopy (TEM) results.

Our simulations reveal key insights into the differences in reactivity between Fe_3O_4 and Fe_2O_3, particularly in the production of OH and OOH radicals, as observed experimentally. While both surfaces predominantly expose Fe^{3+} ions, Fe_3O_4 nanozymes exhibit significantly higher OH radical production. We have analyzed in the first place possible H_2O_2 decomposition mechanisms in the surfaces. We then performed classical MD simulations for understanding water solvation patterns. This allowed us to extract the most populated solvent conformations, and include a few water molecules directly interacting with the surface in the electronic structure calculations. By examining the electronic structure and charge transfer mechanisms, we elucidate the influence of water microsolvation and provide a comprehensive explanation for the experimentally observed catalytic differences between these materials.