RSC INTEREST GROUP
THEORETICAL CHEMISTRY

Theoretical Chemistry Graduate Student Meeting

4th and 5th September 2025

Book of Abstracts

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About

Welcome from the Chair

On behalf of the Royal Society of Chemistry's Theoretical Chemistry Group, I am delighted to welcome you to this year's online Graduate Student Meeting.

The Theoretical Chemistry Group is one of the RSC's Interest Groups, which are member-led communities supporting both RSC members and the wider chemical sciences. Our aims are to promote and represent the interests of theoretical and computational chemists, to advance knowledge and awareness of the field, to highlight its diversity, to foster collaboration and innovation through events, and to support early-career researchers.

This meeting reflects all of these aims. By being held online, it enables participation from around the globe, reflecting the truly international nature of our community. The breadth of topics represented in the oral and poster sessions showcases the richness and breadth of theoretical and computational chemistry today.

I encourage you to engage actively, take part in stimulating discussions, and use this opportunity to connect with fellow early-career researchers. I hope this meeting not only broadens your scientific horizons, but also helps you build collaborations and friendships that will last throughout your career.

A special word of thanks goes to Patrick Wang and Julia Kaczmarek, our PhD representatives on the committee, who have taken the lead in organising this meeting.

I wish you all an enjoyable and inspiring meeting!

Tanja van Mourik, FRSC, CChem

Chair of the RSC Theoretical Chemistry Group

The Graduate Student Meeting

The Graduate Student Meeting of the Theoretical Chemistry group of the Royal Society of Chemistry has been a proud tradition of the interest group for many years now. The meeting aims to provide early career researchers in the field a platform to showcase their research and connect with their colleagues. This year, we have decided to host the meeting online to increase international participation. We were overwhelmed by the excellent submissions this year from all over the world and had to decline many excellent abstracts. We are indeed in awe of the vibrancy and diversity of current research in the field. We hope you enjoy this meeting!

Organising committee:

Basile Curchod	Julia Kaczmarek	Patrick Wang
University of Bristol	*University of Manchester*	*University of Oxford*

Royal Society of Chemistry Theoretical Chemistry Group

The Theoretical Chemistry Group is one of the RSC's many Interest Groups. The Interest Groups are member driven groups which exist to benefit RSC members, and the wider chemical science community, in line with the RSC's strategy and charter. The group aims:

- To actively promote the interests of theoretical and computational chemists
- To ensure that the interests of its members are adequately represented in the activities of the RSC and other bodies
- To advance knowledge and awareness of computational and theoretical chemistry, highlighting the diversity within the field
- To organise and support conferences, workshops, and other events that foster collaboration and innovation in computational and theoretical chemistry
- To support young researchers in the field of computational and theoretical chemistry

Theoretical Chemistry Group Committee

Tanja Van Mourik
Fernanda Duarte
Marco Sacchi
Samantha Jenkins
Basile Curchod
Marco Sacchi
Timothy Hele
Helen Chappell
Cristina Trujillo
Mark Storr
Patrick Wang
Julia Kaczmarek

Timetable

CT: Contributed Talk, PT: Plenary Talk, IT: Invited Talk

Thursday, 4th of September

Chair: Dr Isabel Creed, Imperial College London, UK

13:15–13:30	**Welcome remarks by Dr Tanja van Mourik**		
13:30–14:30	PT	**Prof Leticia González** University of Vienna, Austria	Excited States at Work: Understanding and Steering Molecular Motion
14:35–14:55	CT	**Edoardo Buttarazzi** Scuola Superiore Meriodionale, Italy	Electronic-Vibrational Fingerprints of Photoexcited States in CoFe Prussian Blue Analogue
15:00–15:20	CT	**Maria Incoronata Sciancalepore** University of Edinburgh, UK	Molecular Simulations for Sustainable Applications: Influence of Molecular Structure and Non-Ideal Interactions on Complex Fluids
15:25–15:45	CT	**Nicholas Lee** University of Oxford, UK	Spin-free Generalised Normal Ordered Coupled Cluster Theory
15:50–16:05	**Break**		
16:05–16:15	IT	**Dr Cate Anstöter** University of Edinburgh, UK	Virtual Winter School on Computational Chemistry *Introduction*
16:15–15:35	CT	**Valerii Chuiko** McMaster University, Canada	Dream of Impossible Calculation
16:40–17:00	CT	**Prateek Rai** Middle Tennessee State University, USA	Computer-aided Discover and Design of Novel Autotaxin-LPA Signalling Axis Inhibitors to Overcome Cancer Therapeutic
17:00 Onwards	**Poster Session #1**		

Friday, 5th of September

Chair: Dr Jessica Jein White, CSIRO Australia

9:30–9:50	CT	**Himanshu Shekhar** Nagoya University, Japan	ReaxFF Reactive MD Study of Lubricant Degradation on DLC in Hard Disk Drives
9:55–10:15	CT	**Lauren Cook** University College London, UK	Towards Conservation of the Quantum Boltzmann Distribution in Mapping Methods: A Normal Mode Approach
10:20–10:40	CT	**Davide Barbiero** EPFL, Switzerland	On the Single-Hessian Thawed Gaussian Wavepacket Dynamics for Electronic Spectroscopy
10:45–11:05	CT	**Anderson Exlonk Gil Peláez** Nicholas Copernicus University in Toruń, Poland	Quantum-Informed Machine Learning for Predicting Fluorescence Quantum Yield
11:10–11:25	**Break**		
11:25–11:45	CT	**Isaac Noble** University of Leeds, UK	Molecular Dynamics Simulations of Pseudomonas Aeruginosa Biofilm Molecules and Structures
11:45–12:45	**Poster Session #2**		
12:45–13:45	PT	**Prof Stefan Grimme** University of Bonn, Germany	g-xTB:DFT Accuracy at Tight-Binding Speed

List of Abstracts – Talks

Thursday 4th September

Excited States at Work: Understanding and Steering Molecular Motion

Prof Leticia González, University of Vienna, Austria

Plenary

Understanding how excited states work lies at the heart of many fascinating processes—from vision and photosynthesis to atmospheric reactions and molecular design. Upon absorbing light, the electronic structure of the molecules is altered, which in turn drives nuclear motion. Excited molecules can relax through various pathways, radiatively or non-radiatively, the latter often in an ultrafast time scale. Despite its omnipresence, the computation of molecular properties in the presence of light is substantially more difficult than calculating electronic ground state reactivity [1]. Particularly, simulating the real-time evolution of the system – the molecular movie – considering all the nuclear and electronic degrees of freedom can be rather challenging [2]. In this talk, I will show examples of excited state photochemistry, with several competing pathways, as well as how excited state dynamical motion can be controlled using laser pulses.

[1] S. Mai, L. González, "Molecular Photochemistry: Recent Developments in Theory", Angew. Chem. Int. Ed. 59, 2-17 (2020)

[2] S. Mai, P. Marquetand, L. González, "The SHARC Approach", Wiley Interdiscip. Rev. Comput. Mol. Sci. 8, e1370 (2018).

Electronic-Vibrational Fingerprints of Photoexcited States in CoFe Prussian Blue Analogue

Edoardo Buttarazzi[1,2], Gerrit N. Christenson[3] , James D. Gaynor[3] and Alessio Petrone[1,2,4] **CT**

[1]Scuola Superiore Meridional, Naples, Italy
[2]Department of Chemical Sciences, University of Napoli Federico II, Naples, I-80126, Italy
[3]Department of Chemistry, Northwestern University, Evanston, Illinois, 60208, USA
[4]Istituto Nazionale di Fisica Nucleare - Sezione di Napoli, Naples, 80126, Italy

Prussian Blue analogues (PBAs) exhibit rich photophysical behavior arising from photoinduced charge-transfer processes between transition-metal centers, which in turn can trigger diverse phenomena such as charge migration, spin crossover, and nonradiative decay. [1, 2, 3, 4, 5, 6]

To elucidate the nature and ultrafast dynamics of these photoinduced states, ligand vibrational modes, particularly CN stretching, can serve as sensitive molecular probes.

In this work [7], we present a detailed theoretical investigation of the electronic and vibrational properties of CoFe PBA, with a focus on dimeric and tetrameric cluster models. Ground- and excited-state electronic structure were described exploiting density functional theory (DFT) and time-dependent DFT (TD-DFT), supported by natural transition orbital and charge-transfer analyses. Computations were performed for both isolated systems and surfactant-solvated models to capture bulk-like and surface-like environments, enabling assessment of surfactant effects on geometry, spectra, and vibrational responses.

Results reproduce experimental UV-Vis trends, highlighting bright Fe-centered transitions in the visible range and spectroscopic changes due to the surface passivation. Energy shifts in the CN vibrations are shown to be crucial to characterize the nature of the photoinduced states, providing a molecular interpretation of time-resolved and energy dependent time-resolved infrared experiments. These trends hold for both isolated and solvated models, unveiling the role of the CN stretching mode as a reliable spectroscopic proxy for of the nature of the excited-state states. The proposed computational protocol thus provides a robust framework for linking electronic structure, vibrational dynamics, and environmental effects in PBAs, offering insights relevant to their photo-functional design.

References

[1] N. Shimamoto, S. Ohkoshi, O. Sato, et. al. Inorg. Chem. 41, 678 (2002)

[2] W.-J. Li, C. Han, G. Cheng, et. al. Small 15, 1900470 (2019)

[3] J. Zimara, H. Stevens, R. Oswald, et. al. Inorg. Chem. 60, 449 (2020)

[4] M. Cammarata, S. Zerdane, L. Balducci et. al. Nat. Chem. 13, 10 (2021)

[5] H. Yi, R. Qin, S. Ding, et. al. Adv. Funct. Mater. 31, 2006970 (2021)

[6] M. Fayaz, W. Lai, J. Li, et. al. Mater. Res. Bull. 170, 112593 (2024)

[7] E. Buttarazzi, G. N. Christenson, A. Petrone and J. D. Gaynor, In preparation (2025)

Molecular Simulations for Sustainable Applications: Influence of Molecular Structure and Non-Ideal Interactions on Complex Fluids

Maria Incoronata Sciancalepore, Nathan Bradley, and Philip J. Camp **CT**

University of Edinburgh, UK

The latest generation of low-emissions electric vehicles implements direct cooling of batteries and recharging stations with thermal management fluids (TMFs). Data centres have also started utilising TMFs for direct cooling of hardware to reduce global greenhouse gases emissions. TMFs require specific physical and chemical properties for efficient heat dissipation, posing significant design challenges. [1] Furthermore, in the context of wind power, efficient lubricant additives are essential for ensuring durability and performance of turbine engines and bearings under pressures of up to 3 GPa. [2] This work uses molecular dynamics (MD) simulations to address these challenges and derive design rules for high-performance fluids from a theoretical understanding of molecular interactions, structures, and dynamics.

The aim of the first project is to establish a reliable methodology for computing the thermal properties of complex fluids and thereby screening candidate TMFs. Classical MD, supplemented with quantum corrections, is used to compute the heat capacity and thermal conductivity of long-chain hydrocarbons with various functional groups. The thermal properties of binary liquid mixtures are also investigated, with a focus on the effects of excess volume of mixing and non-ideal interactions. The methodology is validated against experimental measurements available in the literature or provided by Castrol Ltd.

The second project explores how non-ideal behaviour in lubricant additive solutions influences the surface adsorption of additives at high pressure. This is experimentally challenging, but relevant in the field of lubrication for wind turbines. MD simulations of non-ideal binary mixtures of simple liquids are used to verify the thermodynamic relationship between adsorption and volume of mixing. As a result, a simple protocol is established for predicting the effects of pressure on adsorption from easily obtained volume of mixing data.

These studies demonstrate how molecular structures and non-ideal interactions in complex fluids can be exploited to optimise the performance of TMFs and lubricant solutions. Moreover, the work demonstrates the power of MD simulations in accelerating the design of liquid formulations to meet urgent sustainability challenges.

References

[1] F. Leach, G. Kalghatgi, R. Stone and P. Miles, Transportation Engineering, 2020, 1, 100005.

[2] F. Schwack, F. Halmos, M. Stammler, G. Poll and S. Glavatskih, Wind Energy, 2022, 25, 700–718.

Spin-free Generalised Normal Ordered Coupled Cluster theory

Nicholas Lee, David Tew **CT**

University of Oxford, UK

General open-shell configurations and multi-determinant states arise in many areas of chemistry. However, they have remained challenging systems for modern electronic structure methods. Following Lindgren, we develop a multi-determinantal coupled cluster approach where the wave-operator is normal ordered à la Kutzelnigg and Mukherjee. This generalises previous attempts of coupled cluster methods based on a normal ordered exponential ansatz by allowing the reference state to be an arbitrary wavefunction. Using this ansatz, we were able to generate size-extensive working equations, truncated to second order in cluster amplitudes for computational feasibility. Building on this framework, we discuss several pertinent aspects of our method: (i) Spin-adaptation by adopting a spin-ensemble as the reference state (ii) Dealing with redundancies arising from overdetermination of cluster amplitudes (iii) Addressing potential size inconsistency despite the size extensiveness of the method. The proposed method is further examined on a range of hitherto challenging systems to demonstrate its applicability.

Dream of Impossible Calculation

Valerii Chuiko, Paul W. Ayers **CT**

McMaster University, Canada

The study of chemical systems at the quantum level is crucial for various scientific disciplines, enabling the design of new materials, drugs, and catalysts. While the wave function and Schrödinger equation are central to theoretical chemistry, exact solutions for complex systems are often challenging. Machine learning (ML) and neural networks (NN) have emerged as powerful alternatives for predicting molecular energies, offering efficiency and the ability to capture complex relationships. However, these methods face challenges such as poor generalizability and the need for enormous amounts of relevant, high-quality, training data.

This study presents a novel approach to predicting energies of electronic systems using NN. We demonstrate the effectiveness of our method by training networks on systems of four and six hydrogen atoms, achieving mean absolute errors of 10^{-3} a.u.. Furthermore, we introduce a fine-tuning technique to predict energies of larger systems. By exploiting the size consistency of Full Configuration Interaction energies, we construct artificial training data for a 10-electron system using combinations of smaller hydrogen clusters. This approach allows us to train a neural network using only training data small systems and a very small amount of data for larger systems.

Our results show that this fine-tuning method outperforms other approaches in terms of Mean Absolute Error, demonstrating its potential for accurate energy predictions of larger molecular systems. This work highlights the power of combining artificial data construction, transfer learning, and fine-tuning in electronic structure theory, opening new avenues for efficient and accurate energy predictions of complex systems.

Computer-aided Discovery and Design of Novel Autotaxin-LPA Signalling Axis Inhibitors to Overcome Cancer Therapeutic Resistance.

Prateek Rai, Souvik Banerjee **CT**

Middle Tennessee State University, Murfreesboro, TN, USA.

Therapeutic resistance remains a formidable barrier in effective cancer treatment, contributing to the majority of cancer-related mortalities. The autotaxin (ATX)–lysophosphatidic acid (LPA) signaling axis has emerged as a critical pathway implicated in drug- and radiation-induced resistance by promoting tumor progression, metastasis, and immune evasion. To address this, a comprehensive computational strategy was employed to identify and optimize novel small-molecule inhibitors targeting both ATX and LPA receptor 1 (LPAR-1), with the overarching goal of restoring chemosensitivity in resistant cancers. The study utilized an integrated computer-aided drug discovery (CADD) framework encompassing 3D ligand-based similarity screening, pharmacophore modeling, and consensus docking approaches—including standard, induced-fit, and receptor ensemble docking protocols. Lead candidates were evaluated and refined using all-atom molecular dynamics (MD) simulations and rigorous binding free energy calculations, including MM-GBSA and free energy perturbation (FEP) methods. Long-timescale MD simulations were conducted to assess binding pose stability, conformational flexibility, and protein–ligand interaction dynamics. In addition, advanced simulation methodologies such as steered MD and metadynamics were applied to investigate kinetic and thermodynamic aspects of binding. To further elucidate electronic-level interactions and active site reactivity, hybrid quantum mechanics/molecular mechanics (QM/MM) calculations were performed. This theoretical framework led to the identification of two ATX inhibitors, ATX-1d and MolPort-137, which demonstrated significant synergy with paclitaxel in resistant breast and melanoma cancer cell lines. Simultaneously, a large-scale virtual screening campaign targeting LPAR-1 yielded several promising antagonists now undergoing experimental evaluation. This work exemplifies the power of theoretical and computational chemistry in guiding rational drug design and provides a platform for developing targeted therapies against the ATX–LPA axis to overcome cancer therapeutic resistance.

Friday 5th September

ReaxFF Reactive MD Study of Lubricant Degradation on DLC in Hard Disk Drives

Himanshu Shekhar, Shota Uchiyama, Yuxi Song, Hedong Zhang, Kenji Fukuzawa, Shintaro Itoh and, Naoki Azuma **CT**

Nagoya University, Japan

In heat-assisted magnetic recording (HAMR) hard disk drives, nanometer-thick perfluoropolyether (PFPE) lubricant films operate under extreme conditions of confined shear and elevated temperatures. These harsh environments pose a significant challenge to lubricant stability, directly impacting device reliability and lifespan. In this study, we employ ReaxFF-based reactive molecular dynamics simulations to investigate the mechanochemical degradation pathways of D-4OH, a model PFPE lubricant, confined between diamond-like carbon (DLC) surfaces. Our simulations reveal that PFPE molecules form surface-bound loop and bridge conformations, which degrade through distinct mechanisms triggered by localized stress and bond scission, particularly C–OH cleavage. The findings highlight the conformation-dependent nature of lubricant breakdown under shear and offer insight into molecular design strategies for enhancing lubricant durability in next-generation data storage technologies.

Towards Conservation of the Quantum Boltzmann Distribution in Mapping Methods: A Normal Mode Approach

Lauren E. Cook, Timothy J. H. Hele **CT**

University College London, UK

Simulation of nonadiabatic dynamics is complex due to the coupling between the nuclear and electronic degrees of freedom. Often mapping methods are utilized, where the quantum subsystem is mapped onto classical variables that are propagated with classical mechanics. One can write out a list of desirable criteria for a mapping method, including conservation of the quantum Boltzmann distribution (QBD), reproduction of Rabi oscillations, classical scaling with system size and a derivation from exact quantum dynamics. However, as far we are aware, none of the existing methods satisfy all the criteria.

In single surface dynamics, a method exists that conserves the QBD with classical scaling and a derivation from exact dynamics, known as Matsubara dynamics. The QBD conservation arises due to truncation of the dynamics in the higher nuclear normal modes of the imaginary-time path integral. However, the nonadiabatic counterpart [1], that also uses nuclear normal modes but keeps the electronic degrees of freedom as unchanged Meyer-Miller-Stock-Thoss variables, does not conserve the QBD. We suggest that taking normal modes of the electronic variables may lead to conservation of the QBD for nonadiabatic dynamics.

Here, we will discuss the use of electronic normal modes for an electronic-only system and identify how better understanding electronic normal modes has significantly improved the outlook for developing a method that satisfies all the desirable criteria.

Reference

[1] J. Chem. Phys., 2021, 154, 124124

On the single-Hessian thawed Gaussian wavepacket dynamics for electronic spectroscopy

Davide Barbiero, Jiří J. L. Vaníček **CT**

École polytechnique fédérale de Lausanne (EPFL), Switzerland

We present a study of single-Hessian Gaussian wavepacket dynamics (GWD), a semiclassical method for simulating vibronic spectra of high-dimensional, slightly anharmonic systems. This approach is significantly cheaper than Heller's local harmonic GWD but achieves comparable accuracy in approximating spectra [1,2]. We prove that, like the variational GWD but unlike the local harmonic approach, single-Hessian GWD conserves the effective energy and the non-canonical symplectic structure on the manifold of Gaussian wavepackets [3,4]. To accelerate numerical simulations, while preserving the geometric properties of the method, we implement high-order geometric integrators that are time-reversible and conserve the norm and symplectic structure exactly, regardless of the time step. Using on-the-fly ab initio dynamics on the first excited-state surface of ammonia, we numerically assess the conservation of geometric properties by these integrators, demonstrating that high-order integrators can enhance both accuracy and computational efficiency. Employing the geometric integrators, we compute the photoelectron spectrum of the difluorocarbene anion and the absorption spectrum of methylamine. Single-Hessian GWD reliably reproduces the experimental spectra, outperforming global harmonic models and matching the accuracy of local harmonic GWD. Comparing results across the two systems, we identify which spectral features are sensitive to the choice of reference Hessian [4].

References

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[2] T. Begušić, E. Tapavicza, and J. Vaníček, J. Chem. Theory Comput. 18, 3065 (2022).

[3] J. J. L. Vaníček, J. Chem. Phys. 159, 014114, (2023).

[4] D. Barbiero, J. J. L. Vaníček, In preparation.

Quantum-Informed Machine Learning for Predicting Fluorescence Quantum Yield

Anderson Exlonk Gil Peláez, Saikat Mukherjee, Rachel Crespo-Otero, Anna Kaczmarek-Kędziera **CT**

Nicolaus Copernicus University in Toruń, Poland

Machine learning (ML) provides a data-driven approach for predicting molecular properties at a lower computational cost than first-principles methods, and in some cases even delivers superior accuracy. Here, we present a quantum-informed ML framework to predict fluorescence quantum yields (Φ_{fl}) of neutral boron- and fluorine-containing organic molecules in methanol and chloroform. Starting from a SMILES-encoded database, we generated optimized 3D geometries and computed ground- and excited-state properties using ORCA 6.0 at the ωB97X-D3/def2-TZVP level, including radiative decay rates via the ORCA excited state dynamics module. Solvent effects were treated with a shell model using the SOLVATOR module of ORCA combined with GKS-EDA and its time-dependent extension GKS-EDA(TD). The resulting quantum-chemical descriptors formed the basis of a bespoke dataset. We trained several models such as classification and regression trees, random forests, linear support-vector classifiers, and Lasso regression to predict Φ_{fl} directly. Preliminary results indicate that our leading ML model approaches the accuracy of experimental data while requiring less amount of computational time per molecule than full excited state dynamics—a promising sign of the method's efficiency.

Molecular Dynamics Simulations of Pseudomonas aeruginosa Biofilm Molecules and Structures

Isaac Noble **CT**

University of Leeds, UK

Pseudomonas aeruginosa is a Gram-negative, opportunistic pathogen and a prevalent cause of hospital-acquired infections. These infections are facilitated by the ability of P. aeruginosa to establish long-term colonisation in immunocompromised patients (and on surfaces meant to be kept sterile) by forming biofilms – structured bacterial communities embedded within exopolysaccharide (EPS) matrices. Biofilm EPS matrices prevent P. aeruginosa bacteria from being cleared by antibiotic treatments and disinfectants, hence there is a demand for novel treatments that can target these structures. Compositional analyses have revealed the primary structures and biosynthetic pathways of the three P. aeruginosa biofilm EPS molecules – Pel, Psl and alginate. However, it remains poorly understood how these molecules interact with each other and their chemical environment at the molecular level. To improve understanding in this area, we constructed oligosaccharide models of P. aeruginosa EPS molecules and conducted molecular dynamics (MD) simulations of individual chains in water using GROMACS. The configurational flexibility of each model was quantified by measuring changes to the dihedral angles of their glycosidic bonds. We discovered that each polysaccharide has a unique glycosidic bond dihedral angle profile and global flexibility. Moreover, cation-chain interactions introduced quantifiable changes to chain rotational flexibility.

g-xTB: DFT Accuracy at Tight-Binding Speed

Prof Stefan Grimme, University of Bonn, Germany

Plenary

Recently, we optimized small (vDZP), deeply contracted AO basis sets in molecular DFT calculations using standard ECPs for all elements up to radon[1]. This strategy is further- more applied to a minimal set of AOs which — as a totally new ingredient — is made adaptive, i.e., radially different for symmetry distinct atoms in a molecule. The "breathing" of the AOs in the molecular environment is parameterized efficiently by on-the-fly computed effective atomic charges (obtained by a new EEQ charge model) and coordination numbers. This so-called q-vSZP set[2] provides in typical DFT applications results of about or better than DZ quality. It forms the basis of our third-generation tight-binding model g-xTB (g=general). This includes non-local Fock-exchange as well as other new, many-center Hamiltonian terms (e.g., atomic correction potentials, ACP). It aims at general purpose applicabilipty in chemistry and more closely approaches DFT accuracy (actually ωB97M-V/aTZ[3]) than previous semi-empirical methods at only slightly increased computational cost (factor of 1.5 compared to GFN2 -xTB). It will be consistently available for all elements Z=1-10 with f-electrons included for lanathanides/actinides. The talk describes key improvements of the underlying TB theory as well as extensive benchmarking on a wide range of standard thermochemistry sets.

[1] M. Müller, A. Hansen, S. Grimme, J. Chem. Phys. 158 (2023), 014103

[2] M. Müller, A. Hansen, S. Grimme, J. Chem. Phys. 159 (2023), 164108. Revision: JPC A , doi:10.1021/acs.jpca.4c06989

[3] N. Mardirossian and M. Head-Gordon, J. Chem. Phys. 144 (2016), 214110

List of Posters

Thursday Session

Room/No.	Presenter	Title
Room 1, Poster 1	Akash Hiregange	*Atomistic modelling of CoO_x/TiO_2 interfaces: Deciphering metal-support interactions in Fischer-Tropsch Catalysis.*
Room 1, Poster 2	Alannah Francis	*Accurate and Efficient Modelling of X-ray Spectra for the Nuclear Fuel Cycle*
Room 1, Poster 3	Carmen Coppola	*Novel dye candidates for indoor DSSCs via a combined Machine Learning and Density Functional Theory strategy*
Room 1, Poster 4	Dulce Maria Trejo Zamora	*Study of the mechanism of P. falciparum DHPS using QM/MM simulations*
Room 1, Poster 5	Jaiming Chung	*CHQuant: A Protocol for Quantifying Conformational Sampling with Convex Hulls*
Room 1, Poster 6	James O'Brien	*Computational Investigation on Enantioselective Chalcogen-Bond-Based Quinoline Reduction*
Room 1, Poster 7	Jherome Brylle Woody Santos	*DIVINE: A Deterministic Top-Down Framework for Scalable Clustering of Molecular Dynamics Trajectories*
Room 1, Poster 8	Jinming Liu	*Ultrafast Relaxation of C5-Substituted Uracil Derivatives after UV Excitation*
Room 1, Poster 9	Julian Stetzler	*Quantum Molecular Dynamics with Electronic Excitations Employing exactly Factorized wavefunctions*
Room 1, Poster 10	Abdelazim Mohamed Abdelazim Abdelgawwad	*easyPARM: A Python-Based Framework for Metal Complex Force Field Parameterization*
Room 2, Poster 1	Julien Eng	*VCGen: Setting up Quantum dynamics operators made easy !*
Room 2, Poster 2	K. A. L. Himasha Gunathilaka	*Breaking the Cytokine Storm: a computational study*
Room 2, Poster 3	Krisztina Anna Zsigmond	*CreationCC: A Coupled-Cluster-inspired, particle-number symmetry-breaking wavefunction*
Room 2, Poster 4	Leticia Adao Gomes	*Uncovering the mechanism of photodenitrogenation reactions with NAMD simulations*

Room/No.	**Presenter**	**Title**
Room 2, Poster 5	Montassar Chaabani	*Modeling the Structure, Electronic Properties, and Spectra of PBDB-T:ITIC Interfaces via DFT and TD-DFT Calculations*
Room 2, Poster 6	Philips Kumar Rai	*Effect of Microsolvation on Non-Statistical Behaviour of HO3 Radical*
Room 2, Poster 7	Sujata Nandi	*Semiclassical Dynamic Simulations of Nonadiabatic and Polaritonic Systems*
Room 2, Poster 8	Tuğçe Gökdemir	*Machine Learning of Collective Variables*
Room 2, Poster 9	Cameron Reeve	*Dynamics of Charged, Polarisable Particles in Extreme Atmospheric Environments*
Room 2, Poster 10	Sneha Anna Sunny	*Molecular level characterization of Vadadustat-solvent interactions via noncovalent forces*

Friday Session

Room/No.	Presenter	Title
Room 1, Poster 1	Agustin Morales	*Alkali metal Reduction Mediated by Sn–Sn* π *Bonding*
Room 1, Poster 2	Alexandre de Camargo	*Leveraging Normalizing Flows for Orbital-Free Density Functional Theory*
Room 1, Poster 3	Amina Mouhamed	*Lithium, Quantum Spins, and Radical Chemistry*
Room 1, Poster 4	Arun Ramamurthy	*Exploring the Mechanism of Hydrogen Cyanide Formation on Metal Surfaces in the Interstellar Medium: A Computational Perspective*
Room 1, Poster 5	Asma Jamali Rafsanjani	*Spectral Analysis of Molecular Kernels for Overfitting Mitigation*
Room 2, Poster 6	Cecilia Lanzi	*Innovative semiclassical approaches to IR spectroscopy*
Room 1, Poster 7	Daniel Galano	*Computational Studies of Functionalised Pn7 (Pn = P, As) Clusters for Metal-Free Catalysed Hydroboration of Pyridine; the Role of Entropy*
Room 2, Poster 1	Giridhar Baburao	*Encapsulation of greenhouse gases in clathrate hydrates with insights into structure, energetics, chemical interactions, and environmental implications*
Room 2, Poster 2	Ignacio Pickering	*Accelerating Latent Space Contrastive Organization of Chemical Representations Using N-ary Comparisons*
Room 2, Poster 3	Jacob Guntley	*Reduction of* Na^+ *within a* Mg_2Na_2 *Assembly: A DFT Study*
Room 2, Poster 4	James Merrick	*Ultrafast dynamics of cyclic disulfides*
Room 2, Poster 5	Jessica Jein White	*Enhancing Mineral Carbonation Through Molecular Simulations on Mineral Surfaces*
Room 2, Poster 6	Jingkun Shen	*Learning radicals excited states from sparse data*
Room 2, Poster 7	Leon Ashley	*Photoexcitation of MABN — Quantum Dynamics Analysis*
Room 3, Poster 1	Lesego Mable Mogoane	*Molecular modeling studies to evaluate the bioactivity of H-Thiazine and its derivatives on breast cancer cells.*

Room/No.	**Presenter**	**Title**
Room 3, Poster 2	Lucy Piekarska	*Exploring and explaining vibrational control of electron transfer*
Room 3, Poster 3	Malin Zollner	*Systematic Evaluation of Machine Learning Strategies for Accurate and Generalisable Prediction of Reorganisation Energies in Organic Semiconductors*
Room 3, Poster 4	Markus G. S. Weiss	*Asymmetrically Constrained Adiabatic ALMO-EDA: Designing Catalysts from First Principles*
Room 3, Poster 5	Matthew Ludwig	*Multiwavelet-based prediction of core XPS spectra for thermally-activated delayed fluorescence OLED emitters*
Room 3, Poster 6	Meenakshi Rana	*Electron transfer mediated decay process for the Auger final state of Ne+(1s-1) (H2O)n (n=1,4) cluster*
Room 3, Poster 7	Muskan	*Strong Ferromagnetic Coupling between Co and Co2+ with Odd Electron (Anti)aromatic Radicals via Noncovalent Interaction*
Room 3, Poster 8	Leticia Madureira	*Bond Dissociation of F2, N2 and O2 through the Prism of SCF Stability Analysis*

Accurate and Efficient Modelling of X-ray Spectroscopies for the Nuclear Fuel Cycle

Alannah N. Francis,Timothy G. Burrow, Michael L. Baker, Meagan S. Oakley

The University of Manchester, UK

Deconvolution and assignment of actinide X-ray absorption spectra (XAS) is complex due to strong electron correlation, significant relativistic effects, and the potential for multiple oxidation states present. Quantum chemistry methods are often used to aid in assignment but must accurately capture all of these effects. Here, we investigate the performance of the restricted active space self-consistent field method with spin-orbit coupling (RASSCF-SO), and subsequent approaches for capturing dynamic correlation energy (multiconfigurational pair-density functional theory [RASPDFT-SO] and second-order perturbation theory [RASPT2-SO]) in modelling M4- and M5-edge XAS of U6+ (5f0) systems. This framework was also applied to understand the discrepancy between the experimental M4- and M5-edge XAS spectra for $[UO_2Cl_4]^{2-}$.

Atomistic modelling of CoOx/TiO2 interfaces: Deciphering metal-support interactions in Fischer-Tropsch Catalysis.

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The use of transition metal catalysts in Fischer-Tropsch (FT) reaction, involving the conversion of syngas (CO and H_2 mixture) to liquid hydrocarbons, is crucial in producing sustainable aviation fuels (SAFs).[1] Cobalt-based metal oxides, dispersed on TiO_2 support, have proven to be highly effective precursors of metallic catalysts in FT synthesis, with Mn promoters enhancing the selectivity.[1] The addition of Mn reduces nanoparticle size, enhances cobalt oxide dispersion across the support, and induces a phase transition from spinel Co_3O_4 to rocksalt CoO. Furthermore, at low Mn loadings, a preferential rocksalt CoO interfacial layer is observed between the TiO_2 support and spinel Co_3O_4 phase, indicating metal-support interactions between CoO_x (Co_3O_4 and CoO) and TiO_2. However, the nature of such interactions in the cobalt oxide catalytic precursor remains unknown. In this study, we have used the universal machine-learned force field MACE-MP2 and density functional theory (DFT) to investigate the stability CoO_x/TiO_2 interfaces to have an atomic level understanding of metal-support interactions in cobalt-based FT catalysts. Periodic DFT calculations on the CoO_x/TiO_2 interface models were performed using the rSCAN exchange-correlation (XC) functional and 'light' basis set as implemented in the Fritz Haber Institute ab initio materials simulation (FHI-aims) package.[3] Exhaustive interface model generation was performed using the Zur and McGill algorithm,[4] which were then screened using MACE-MP to identify the most appropriate models. DFT calculations on the pre-screened models show that interfacial stability is higher for CoO/TiO_2 interface compared to Co_3O_4/TiO_2 interface. However, the presence of oxygen vacancy defects on TiO_2 surface enhances the interfacial stability of the Co3O4/TiO2 interface. The oxygen vacancies on TiO_2 surface are stabilized by the movement of oxygen atoms from the Co_3O_4 phase across the interface. Furthermore, our calculations show that interfacial stability increases with increase in oxygen vacancy concentration on the TiO_2 surface. These insights lay the groundwork for future investigations into the effect of Mn-doping on interfacial stabilities. The atomistic understanding gained from this study provides a valuable knowledge for rational catalyst design, enabling targeted modifications to enhance activity, stability, and selectivity in industrial FT processes.

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Novel dye candidates for indoor DSSCs via a combined Machine Learning and Density Functional Theory strategy

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Dye-sensitized solar cells (DSSCs) have become one of the preferred technologies for indoor applications and powering Internet of Things (IoT) devices due to their excellent ability to capture ambient light and convert it into electrical energy [1]. In this context, research has focused on developing new organic dyes that can absorb different artificial light emissions at various intensities [2,3]. However, this process is often costly and time-consuming. In this work, we introduce for the first time a two-stage approach combining Machine Learning (ML) techniques with Density Functional Theory (DFT) methods to speed up the discovery of potentially new efficient organic dyes for indoor DSSCs and to predict their power conversion efficiency (PCE) at different indoor light sources and intensities [4]. Additionally, we evaluated the synthetic accessibility (SAscore) of the novel dye candidates. This approach enabled us to identify three promising dye candidates for indoor DSSCs with PCE > 29% under different artificial lighting conditions. Comparing these data with experimental PCE values from literature dyes under indoor conditions suggests that these three candidates could yield experimental PCE values comparable to or higher than current literature results. Our findings demonstrate that the developed ML-DFT protocol is an effective method to accelerate the discovery of novel, potentially more efficient organic dyes for indoor DSSCs. Ongoing efforts focus on synthesizing the most promising dye candidates, aiming to further enhance the efficiency of indoor DSSCs.

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Study of the mechanism of Plasmodium falciparum dihydropteroate synthase (DHPS) using QM/MM simulations

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Malaria remains a major public health concern in tropical regions, annually claiming over half a million lives worldwide. In Africa, where most cases occur, children under 5 bear about three-quarters (76%) of the mortality rate. This project investigated the mechanistic pathways within the catalytic center of dihydropteroate synthase (DHPS), a key medicinal target for treating and preventing malaria. DHPS catalyzes the nucleophilic substitution of p-aminobenzoic acid (pABA) and 6-hydroxymethyl-7,8- dihydropterin pyrophosphate (DHPP) to form 7,8-dihydropteroate, a crucial intermediate species for microbial DNA synthesis in the folate pathway.

Our efforts to understand this reaction mechanism started with classical molecular dynamics (MD) simulations of the X-ray structure of Plasmodium falciparum DHPS, the parasite responsible for the most severe form of malaria. From this equilibrated structure, we performed QM/MM simulations using the semi-empirical PM6 Hamiltonian, and the resulting structures served as a starting point for free energy calculations.

To explore possible pathways and the identity of the product, we first conducted Steered Molecular Dynamics (SMD), focusing on comparing SN 1 and SN 2 mechanisms, as well as identifying the base deprotonating pABA (the reaction's nucleophile). Subsequently, the most plausible reaction mechanisms were studied using the Adaptative String Method (ASM). Our results show that SN2 mechanism exhibits the lowest energy barrier, both in SMD and ASM; however, the most stable product is found in SMD with the SN1 mechanism, while preliminary ASM results suggest the SN2 mechanism is more favorable. Notably, no surrounding amino acid was involved in any of these cases.

CHQuant: A Protocol for Quantifying Conformational Sampling with Convex Hulls

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Conformational sampling is a key component of any machine learning force field workflow. Existing methods for assessing the coverage of a conformational space can be ambiguous, reference-dependent, and qualitative due to their reliance on visual perception. In this work, we propose CHQuant, a protocol designed to quantify the extent of conformational space coverage. This approach is based on the observation that moving atoms define point clouds that can be characterised by the volumes of the corresponding convex hulls. By applying our method to several small molecules and conformational sampling methods, we show that convex hull volumes behave according to chemical intuition on an atomic, functional group, and molecular level. Convex hull volumes are therefore useful measures that can be used to compare different sampling methods or datasets. Among many applications, the CHQuant approach can be used to monitor the sampling achieved by any molecular dynamics sampler and compare existing conformational databases of the same system.

Computational Investigation on Enantioselective Chalcogen-Bond-Based Quinoline Reduction

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This computational study aims to computationally aid the development of enantioselective chalcogen-bond (ChB) based organocatalysts for the reduction of quinoline. Similar to halogen bonding (XB), ChB has been successfully employed in numerous non-enantioselective organocatalytic reactions, and shows promise for substrate specificity, tunability, and readily forms Lewis-pair adducts with softer Lewis Bases (LB).[1–3]

To gain further insight into experimentally obtained enantioselective ChB-based organocatalysts (vide infra), the structural space occupied by the catalysts under study were investigated through conformational analyses. The results of which identified the importance of structural flexibility of these systems for enantioselective purposes, with a specific focus on the availability of an internal binding pocket for introducing a significant chiral environment to the quinoline salt. Furthermore, potential energy surfaces were calculated for all catalysts investigated. The resulting transition state energies were used to calculate theoretical enantiomeric excess, which was further compared to experimental results.

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DIVINE: A Deterministic Top-Down Framework for Scalable Clustering of Molecular Dynamics Trajectories

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We introduce DIVIsive N-ary Ensembles (DIVINE), a deterministic, top-down clustering framework tailored for molecular dynamics (MD) trajectories. Unlike traditional methods that rely on computationally expensive pairwise distance matrices, DIVINE constructs a complete clustering hierarchy through recursive splits based on n-ary similarity metrics. It supports flexible cluster selection criteria, including a weighted variance metric, and deterministic anchor initialization strategies such as NANI (N-ary Natural Initiation), ensuring consistent and balanced partitions across runs. Applied to a 305 μs trajectory of the folding of the villin headpiece, DIVINE matches or exceeds the clustering quality of bisecting k-means, while reducing computational cost and eliminating variability in results. Its single-pass design makes it easy to explore different clustering resolutions without rerunning the algorithm. With its speed, consistency, and ease of interpretation, DIVINE offers a strong and practical alternative to traditional MD clustering methods. DIVINE is publicly available as part of the MDANCE package: https://github.com/mqcomplab/MDANCE

Ultrafast Relaxation of C5-Substituted Uracil Derivatives after UV Excitation

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Photoexcited 5-fluorouracil (5-FU) exhibits profoundly altered excited-state dynamics, dominated by a near-complete population transfer from the initially excited $\pi\pi*$ state to the $n\pi*$ state, where the population becomes strongly trapped. Although this enhanced $n\pi*$ population is well-documented, the subsequent evolution within this state and the origin of its relaxation pathway diversity remain poorly understood. Here, we combine multilevel electronic structure methods and nonadiabatic dynamics simulations to investigate its excited-state dynamics. Our results reveal that the electron-withdrawing fluorine atom induces near-degeneracy between the $\pi\pi*$ and $n\pi*$ states and enhances their nonadiabatic coupling, facilitating efficient internal conversion (IC). All simulated trajectories exhibit ultrafast $\pi\pi* \rightarrow n\pi*$ transfer. The residual kinetic energy drives further propagation on the excited-state potential energy surfaces, leading to frequent population exchanges between the $\pi\pi*$ and $n\pi*$ states. Crucially, wavepacket localization on the $n\pi*$ surface creates a dynamic bias that suppresses the formation of the pyramidalized C5 configuration essential for accessing the $\pi\pi*$/S0 conical intersection, thereby blocking the direct ground-state relaxation. Consequently, the system evolves into a bidirectional $n\pi* \leftrightarrow \pi\pi*$ trapping regime, characterized by strong interstate coupling and persistent population exchange. Ultimately, relaxation to the ground state proceeds predominantly via internal conversion through the $\pi\pi*$ state. This cooperative trapping mechanism provides a new perspective on the nonadiabatic relaxation of 5-FU.

Quantum Molecular Dynamics with Excitations Employing Exactly Factorized Wavefunctions

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Practical simulation of molecular dynamics – treating both the electron and nuclei as quantum-mechanical particles – in large systems (100s-1000s of atoms) is a major long-standing challenge for theoretical chemistry. Recently, we have developed a new approach based on the exact factorization of the electron-nuclear wavefunctions, with the dynamics of the nuclear part evolving in time under an effective complex potential; the dynamics of electrons and nuclei is formulated without referencing the electronic energy eigenstates. After demonstrating the approach on the analytic vibrationally nonadiabatic model, we will discuss its application to the dynamics of the H2+ molecular ion in a laser field using standard 6-31G and 6-31G** bases to represent the electronic wavefunction. Nuclear derivatives of the electrons and derivatives of the nuclear wavefunction (represented as an ensemble of Quantum Trajectories) are obtained through basis set projections. Analyzing the effect of the electronic basis on the derivative couplings, we find that while the nuclear dynamics change significantly due to the difference in the real potential energy surfaces, the overall factorization structure remains unchanged. We also observe that the wavefunction propagation becomes unstable when the nuclear wavefunction begins to bifurcate and identify step-like features in the basis function 'populations' as the source of the problem. This behavior is further examined within a simplified model of photodissociation to evaluate its stability, particularly in the moving frame or trajectory representation. We show analytically that while the moving frame somewhat reduces the derivative amplitudes, it does not change the problem of the step-function derivatives qualitatively, which means that the current exact factorization is suitable for short laser pulses before the wavefunction bifurcation. We also note that our current analysis is limited to just a few electronic basis functions, which is far from the intended regime of the exact factorization method applied to the electronic-nuclear wavepackets. This regime of the electron-nuclear dynamics will be investigated in future work.

easyPARM: A Python-Based Framework for Metal Complex Force Field Parameterization

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The development of accurate force field parameters for metal-containing molecules remains a significant challenge due to their structural diversity and complex coordination environments. We introduce easyPARM,[1] a Python-based toolkit that automates the generation of reliable bonded parameters for transition-metal complexes. Leveraging quantum mechanical data such as the Cartesian Hessian matrix and atomic charges, easyPARM uses a unique atom typing strategy that differentiates each coordinating atom, thereby improving the fidelity of angle and bond constants, particularly in asymmetric environments. This methodology circumvents the need for manual post-processing or atom-type definition and enables seamless integration into Amber-based molecular dynamics workflows.[2] Validation against density functional theory simulations show excellent agreement in both gas-phase and solvated conditions. The tool supports Gaussian and ORCA input formats and outputs fully compatible Amber parameter files. Overall, easyPARM offers a practical and robust solution for extending classical molecular simulations to a broad range of metal complexes, opening avenues for efficient modeling in materials science, catalysis, and metallodrug design.

VCGen: Setting up Quantum dynamics operators made easy !

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Excited state dynamics play a critical role across a broad range of scientific fields. Importantly, the highly non-equilibrium nature of the states generated by photoexcitation means that excited state simulations should usually include an accurate description of the coupled electronic–nuclear motion, which often requires solving the time-dependent Schrödinger equation (TDSE). Methods to do so are usually cataloged into two categories: (1) on-the fly methods that require an estimation of the potential energy surface (PES) and coupling at each time steps and (2)grid-based methods that require a priori knowledge of the PES and coupling. The methods of the first group allow to capture the full nuclear dimensionality of the system but can be quickly restricted by the amount of single point calculation needed while the methods of the second group allow for a more accurate description of the PES within a restricted dimensionality space.

Multi-Configuration Time-Dependent Hartree (MCTDH) is a method of choice for quantum dynamics as it allows to describes quantum effects such as tunneling or interferences. It requires however the Hamtiltonian to be built prior to the propagation of a nuclear wavepacket. Such Hamiltonian is traditionally built in the diabatic representation as it allows to circumvent singularities in the expression of the non-adiabatic couplings. The constructions of the diabatic potentials and couplings is far from being straightforward and often, the Hamiltonian is modeled. The vibronic coupling approximation is one of those model. The PES are built as a Taylor expansion of which the different terms are either determined from calculations at Franck-Condon, or by fit to the calculated adiabatic energies.

In this poster I present VCGen, a software that aims at making MCTDH calculations accessible for "non-initiates" by providing an – as much as possible ! – automated way for setting up a MCTDH calculation on Quantics from the Cartesian coordinates to the input files. This poster will guide you through the current state of VCGen's development and present you with the program's roadmap for month to come !

In addition to a code developed within the COSMOS program grant, VCGen aspires to provide online resources, such as tutorial and theory videos, showcase examples and exercises.

Breaking the Cytokine Storm: a computational study

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Severe dengue is driven by a TNFα–mediated cytokine storm, yet no approved antiviral therapies are currently available. To explore natural inhibitors, 22 phytochemicals from juvenile Bambusa vulgaris shoots were identified and screened through molecular docking. Sterols exhibited the strongest binding affinities, with ergosterol (–12.9 kcal/mol) outperforming the positive control A7A (–11.6 kcal/mol), followed by βsitosterol (–12.7), stigmasterol (–12.6), campesterol (–12.5), and stigmastanol (–12.4). To further assess conformational stability, 1000 ns molecular dynamics simulations were conducted for unbound TNFα, the TNFα–ergosterol complex, and the TNFα–A7A complex. Principal component analysis revealed that unbound TNFα displayed the highest variance (58.0%), indicative of greater flexibility and reduced stability, whereas ergosterol (43.0%) and A7A (43.7%) reduced variance, consistent with restricted motions and enhanced stabilization. Collectively, these findings suggest that Bambusa vulgaris sterols can offer a promising phytochemical strategy to mitigate cytokine storm in severe dengue.

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CreationCC: A Coupled-Cluster-inspired, particle-number symmetry-breaking wavefunction

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Standard quantum chemistry methods often struggle with either accuracy or the computational cost for strongly correlated systems. Here, we introduce a novel exponential ansatz, termed ‘CreationCC’. It draws inspiration from Coupled Cluster formalism and geminal wavefunctions but breaks the particle-number symmetry. This approach guarantees size-extensivity and size-consistency of the wavefunction without an inherent bias towards any reference state, which makes it particularly interesting. The performance of CreationCC is demonstrated on model systems.

Uncovering the mechanism of photodenitrogenation reactions with NAMD simulations

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Photochemical reactions are examples of green chemistry used to synthesize strained molecules under mild conditions. One example is the light-promoted denitrogenation of bicyclic azoalkanes, which can produce bicyclo[2.1.0]pentane with retained or inverted diastereoselectivity. However, the mechanism behind these reactions has been disputed for over six decades. We employed multireference calculations and non-adiabatic molecular dynamics (NAMD) simulations on a series of diazabicyclo[2.2.1]heptenes to address long standing mechanistic questions. Our simulations provide detailed information on their photophysical properties, reactivities, and mechanistic pathways. We performed complete active space self-consistent field (CASSCF) calculations using an (8,9) active space and the ANO-S-VDZP basis set. We then corrected the CASSCF energies with XMS-CASPT2(8,9)/ANO-S-VDZP. For the parent and two derivatives with methyl substituents, the lowest excitations are nNN(σCN) $\rightarrow \pi*$ and range from 3.94 – 3.97 eV. We created 500 initial conditions using Wigner sampling for each molecule and implemented the fewest switches surface hopping NAMD simulations. Our analysis indicated that the diazoalkanes undergo asynchronous conical intersections, where one 4σCN bond breaks along the S1 and the other σCN breaks after hopping to the S0. We identified two distinct clusters of hopping points from S_1 to S_0: one involving a partially inverted molecular structure and another where the molecule undergoes complete inversion. For each cluster, we identified the corresponding Minimum Energy Conical Intersection (MECI), offering valuable insight into the mechanisms of non-radiative relaxation.

Modeling the Structure, Electronic Properties, and Spectra of PBDB-T:ITIC Interfaces via DFT and TD-DFT Calculations

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Interfaces between PBDB-T and ITIC are modeled through density functional theory (DFT) applied to dimers constructed from various conformers of a PBDB-T segment and the ITIC molecule. These variations represent non-uniformity at the donor–acceptor interface commonly observed in organic solar cells. Based on this model, electronic and optical properties relevant to charge separation are calculated using DFT and time-dependent (TD-) DFT. For nearly half of the dimers, band offsets relative to pristine donor and acceptor materials result in charge trapping, leading to inefficient charge separation and non-geminate recombination. Exciton binding energy is evaluated by TD-DFT using B3LYP and HSE06 hybrid functionals, revealing significant qualitative and quantitative differences.These results are compared with experimental data from selected characterization techniques. Finally, the impact of these findings on key photovoltaic parameters, especially open-circuit voltage and short-circuit current, is discussed, providing insights into optimizing device performance.

The puzzle of high lifetime and low stabilization of HO3● : Rationalization and Prediction

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The puzzle of high lifetime and low stabilization of HO3● : Rationalization and Prediction One of the most important puzzles in atmospheric chemistry is the long-lifetime of HO3● in spite of its low-stabilization energy. In the present work, we have estimated the lifetime of HO3● using classical dynamics simulations by coupling an available neural-network analytical potential energy surface with a chemical dynamics program. The simulation results clearly indicate that at room temperature, the lifetime of HO3● can exceed 1 μs under collision-free conditions. In fact, at 200 K, the lifetime of HO3● can enter the millisecond timescale. This suggests that HO3● is indeed a stable enough intermediate that can affect the outcomes of crucial atmospheric processes.

Semiclassical Dynamic Simulations of Nonadiabatic and Polaritonic Systems

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We have extended the Mixed Quantum-Classical Semiclassical (MQC SC) method to handle nonadiabatic systems using the Meyer-Miller-Stock-Thoss (MMST) Hamiltonian, enabling a consistent dynamic treatment for both electronic and nuclear degrees of freedom. MQC SC can selectively quantize different degrees of freedom, enhancing numerical convergence without compromising accuracy. We have also shown how the recently developed Nonadiabatic Hybrid Wigner dynamics (NHWD) can perform at different model parameter regimes for open quantum system. Semiclassical (SC) approximations strike a balance between high computational cost for accurate quantum methods and near-classical scaling that lacks quantum detail. For complex high-dimensional systems, a hybrid approach is ideal, with high-level quantum descriptions for key modes and lower levels for others. Our work maps out a pathway for finding optimal parameters and SC method choices for different nonadiabatic systems.

We have also studied the TIPS-pentacene polaritonic system in collaboration with the Musser group at Cornell. Our model investigates how the presence of a dark state that interacts with the bright state can modify polariton characteristics. We examine the linewidth, polariton state population, and polariton state energy manifolds when the cavity is in resonance with the bright (S1) state and then systematically detuned towards dark (1TT) state.

Machine Learning of Slow Collective Variables

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Complex systems are characterized by metastable states separated by energy barriers higher than the thermal energy. These energy barriers lead to kinetic bottlenecks that hinder sampling long-timescale processes. This sampling problem severely limits the reachable timescales of molecular dynamics (MD) simulations [1]. Protein folding, crystallization, nucleation, glass transitions, aqueous systems, catalysis, and molecular recognition are just a few of the many long-timescale processes [2].

Many enhanced sampling techniques have been proposed to sample such processes within a reasonable computational timescale [1]. Enhanced sampling techniques usually require a few meaningful degrees of freedom, called collective variables (CVs), before enhancing MD simulations. Choosing CVs is crucial for the accuracy and efficiency of enhanced sampling, and optimal CVs should correspond to the slow degrees of freedom. Conventional methods are widely used, but CVs extracted this way may not capture the slow dynamics of the system. To overcome these issues, machine learning (ML) techniques have been proposed that can reduce the high-dimensional structure to a meaningful low-dimensional structure [2].

Here, we use several ML techniques on an MD simulation of alanine dipeptide to reduce dimensionality and extract CVs from simulation data. As a baseline, we project features onto a lower-dimensional space using principal components analysis (PCA), which is one of the oldest and most well-known dimensional reduction techniques [3]. However, PCA cannot identify significant nonlinear characteristics. We also employ time-lagged independent component analysis (tICA) [4] and spectral map [5]. We demonstrate that these methods are more suitable for capturing the slow dynamics of the system and identifying optimal CVs.

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Dynamics of Charged, Polarizable Particles in Extreme Atmospheric Environments

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Volcanic ash plumes and the Venusian atmosphere are examples of extreme atmospheric environments characterized by scorching temperatures and high densities of charged, polarizable particles. This study investigates the influence of polarization on particle interactions in these environments, employing dynamic simulations built upon a rigorous classical electrostatic formalism.[1,2] Our findings highlight the pivotal role of polarization in driving electrostatic aggregation under extreme conditions, particularly through like-charge attraction at small separations, which would otherwise be neglected under conventional models. By considering complex polarization effects, we reveal significant deviations from traditional Coulomb and hard-sphere models, including the increase in collision efficiency and collisional cross section by up to 25% as well as reduction in the velocity critical for aggregation by up to 30%. These insights have wide-ranging implications for modelling charged particle dynamics beyond extreme environments to broader industrial, atmospheric and astrophysical contexts.

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Molecular level characterization of Vadadustat-solvent interactions via noncovalent forces

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Vadadustat (VD) is a hypoxia-inducible factor prolyl hydroxylase inhibitor under clinical investigation for anaemia associated with chronic kidney disease. Solvent interactions can significantly influence the stability, conformation, and bioavailability of drug molecules. In this study, we employed density functional theory (wb97xd/cc-pVDZ) to investigate the non-covalent interactions between VD and a common pharmaceutical solvent- dimethyl sulfoxide (DMSO). Optimized VD-solvent complexes was analyzed to evaluate binding preference. Binding energy calculations revealed that complex formation is energetically favourable. Non-covalent interaction (NCI) analysis highlighted hydrogen bonding as a dominant stabilizing force, validated by Natural Bond Orbital (NBO) charge transfer assessments and Atoms-in-Molecules (AIM) identification of bond critical points. Energy decomposition analysis (XEDA) further confirmed thermodynamically favourable binding. These findings provide molecular-level insight into VD-solvent stabilization mechanisms, offering a basis for rational solvent selection in drug formulation and potential enhancement of pharmacological performance.

Group 1 Redox Interchange: An Alumanyl Case Study

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Since early 19th Century, group 1 alkali metals have widely served as powerful reducing agents in all fields of chemistry. Notwithstanding Dye and co-workers' startling identification of alkalide (M-; M = Na, K, Rb, Cs) anions,[1] little consideration is applied to the redox behaviour of these group 1 elements in molecular systems. Their use is often guided more by synthetic expedience than by the origin of reducing electrons, with the resulting M^+ typically regarded as an innocent bystander - either incorporated into the target compound or relegated to a by-product salt.

Across the years, a series of isolable aluminium(I) anions ("alumanyls") have been reported.[2] Very recently, the Hill group have introduced a new series of alumanyls incorporating different alkali metals counterions ([SiNDippAlM]2, 1M) and demonstrated that the identity of M+ plays a pivotal role in their stability and reactivity.[3] A combined thermodynamical model is presented that is derived from both theoretical (DFT) and empirical data to accurately evaluate the feasibility of the redox interchange of group 1 elements in alumanyl anions derivatives, through the use of elemental alkali metals (M0) as reducing agents.

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Orbital-Free Density Functional Theory with continuous normalizing flows

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Orbital-free density functional theory (OF-DFT) is a quantum mechanical formulation that provides a way to calculate the molecular electronic energy, relying solely on the electron density. In OF-DFT, the ground state energy is optimized variationally to minimize the total energy functional while enforcing normalization. For real-space systems this formulation has been historically dependent on Lagrange optimization techniques, due to the inability of previously proposed techniques to ensure normalization. In this work, we introduce a Lagrangian-free optimization framework by parametrizing the electron density with normalizing flows (NFs), a class of generative models capable of constructing flexible probability distributions. By integrating NFs with equivariant deep learning and variational inference, we present an innovative approach to the OF-DFT framework in a way that inherently satisfies the normalization constrain. The versatility of our model is illustrated for a diverse range of chemical systems, including a one-dimensional diatomic system, LiH, as well comprehensive simulations of hydrogen, lithium hydride, water, and four hydrocarbon molecules. Notably, the flexibility of NFs allows initialization with promolecular densities, significantly accelerating convergence.

Lithium, Quantum Spins, and Radical Chemistry

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Lithium carbonate is a frontline treatment for bipolar disorder. Yet, despite decades of research, its precise mechanism remains unresolved. Interestingly, its two isotopes, ^{6}Li and ^{7}Li, seem to have different biological effects, pointing to a possible role for nuclear spin (spin 1 for ^{6}Li and spin 3/2 for ^{7}Li). [1] Our inspiration comes from nature's spin experts: birds, whose remarkable magnetoreception is hypothesised to harness the Radical Pair Mechanism (RPM) for detecting Earth's magnetic field. [2]. Since nuclear spin affects how electrons interact in certain chemical reactions, this led us to explore the RPM, where tiny changes in electron spin can influence reaction outcomes and, ultimately, biology. Previous research focused on flavin–superoxide radical pairs, but superoxide's extreme instability limits its relevance in the brain. [1] We therefore chose a more stable, brain-abundant partner: the ascorbyl radical (derived from vitamin C), whose longer lifetime makes it well suited for spin-dependent chemistry. [3] We hypothesise that the nuclear spin of ^{6}Li versus ^{7}Li will differentially affect the flavin–ascorbyl triplet yield.

EXPLORING THE MECHANISM OF HYDROGEN CYANIDE FORMATION ON METAL SURFACE IN THE INTERSTELLAR MEDIUM: A COMPUTATIONAL PERSPECTIVE

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Using quantum chemical calculations, we investigate four distinct mechanistic pathways for the formation of hydrogen cyanide (HCN) under two astrophysically relevant environments: the gas phase and on metal surfaces. The reactivity and feasibility of these reactions are systematically evaluated in both conditions. Furthermore, we explore the adsorption and interaction of HCN with metallic surfaces to elucidate the associated thermodynamic parameters and activation energy barriers governing these processes in the interstellar medium (ISM). Our findings reveal that the presence of metal surfaces provides alternative catalytic pathways that substantially reduce the activation barriers, thereby enhancing the reaction kinetics. This theoretical study demonstrates that the catalytic role of interstellar metal surfaces markedly increases the probability of HCN formation, making surface-mediated reactions the most favorable route compared to gas-phase processes.

Spectral Analysis of Molecular Kernels for Overfitting Mitigation

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Although computational simulations are essential for advancing material science, their high computational cost drives the search for efficient surrogate/regression methods. Machine learning plays a central role in material science by bypassing the need of extensive experimental or computational studies.

Kernel methods are particularly attractive as they are expressive models while being data-efficient. However, the choice of kernel function introduces an important inductive bias that influences the properties of the target function. This choice is inherently linked to the molecular representation employed as the input of the kernel. In general, molecular representations fall into two broad categories: (1) global descriptors that capture whole-system properties and (2) local descriptors that encode atomic environments. Traditionally, the accuracy of the molecular kernel models has been assessed using test-set performance. In this work, we propose an alternative framework to evaluate their robustness by analyzing the molecular kernels alignment with target properties. In addition, recent advancements in spectral analysis of kernel methods have provided insight into the relationship between generalization and truncated representations of the kernel matrix. Motivated by these studies, we explore truncated kernel ridge regression (TKRR) in the context of molecular property prediction. TKRR introduces an additional spectral truncation parameter, offering a framework for improving generalization in kernel-based models for molecular systems. We apply spectral analysis to existing molecular kernels to examine their alignment with the target function in terms of spectral properties. Our results show that in the scenarios where full KRR overfits the training data, TKRR better aligns with the target function, improving generalization. We demonstrated this approach on the QM9 dataset, predicting molecular properties such as HOMO-LUMO gap, internal energies at 0 K and 298 K, Enthalpy and Gibbs energies.

Innovative semiclassical approaches to IR spectroscopy

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Semiclassical vibrational spectroscopy is based on the evolution of classical trajectories and is able to reproduce quantum effects with good accuracy at the cost of a reasonable computational effort. [1-5] Nevertheless, semiclassical vibrational power spectra do not simulate all the features of the experimental IR spectra, since intensities in power spectra are not directly related to IR absorptions. Therefore, we developed a new semiclassical approach to the calculation of molecular IR spectra (SC IR) by employing the time average technique upon symmetrization of the quantum dipole-dipole autocorrelation function. [6-8] The method features excellent accuracy in calculating absorption intensities and frequencies of vibrations, but its need for a Monte-Carlo integration makes it directly applicable only to model systems and small molecules. More recently, we introduced an extended semiclassical approach to IR spectroscopy (e-SC IR), based on the evolution of a single pair of tailored classical trajectories per normal mode. [9] The approach preserves the accuracy of SC-IR spectra when applied to small molecules, and is successfully extended to larger molecular systems, such as ethanol and glycine.

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Computational Studies of Functionalised [Pn7] Clusters for Metal-Free Catalysed Hydroboration of Pyridine; the Role of Entropy

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Catalysts are a staple of the manufacturing sector, with up to 90% of chemical products requiring the use of a catalyst at some point in their preparation. However, these catalysts often being based on expensive, non-renewable precious metals.1,2 Hence, the development of alternative catalysts based on abundant main group elements, such as phosphorus, is becoming an urgent avenue of research.

This work is a computational investigation into a full deduction of the catalysed hydroboration of pyridine mechanism with a metal free, functionalised [P7] Zintl cluster, how the properties of this cycle may change for various analogues of this catalyst, and the key states in the cycle that underpin catalytic activity. We have also investigated how the way free energy is computed affects the overall energetics and subsequent conclusions of this work.

Encapsulation of Greenhouse Gases in Clathrate Hydrates: Structural Insights, Energetics, Chemical Interactions, and Environmental Implications.

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The encapsulation of greenhouse gases (GHGs), such as CCl_4, CF_2Cl_2, CH_3Br, CH_3Cl, CH_4, CO, CO_2, H_2S, CH_3F, N_2O, NF_3, O_3, CF_4, SF_6, and SO_2, within 512, 51262, and 51264 clathrate hydrate cages is being investigated in our study using Density Functional Theory (DFT). The smaller cages introduce steric constraints, leading to bond distortions and significant vibrational blue-shifts, while larger cages offer greater flexibility, resulting in red-shifts or minimal vibrational alterations. Intermediate sized cages provide a nuanced balance between spatial limitations and structural stabilization. In the 512 cage, fewer guests exhibit red-shifts, whereas the larger 51262 and 51264 cages show more frequent red-shifts due to enhanced host–guest interactions.

Accelerating Latent Space Contrastive Organization of Chemical Representations with N-ary and Tanimoto-Weighted Comparisons

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In recent years machine learning autoencoders have been revolutionizing molecular representation learning. However, traditional autoencoder techniques suffer from low data-efficiency and lack semantic organization of the latent spaces. Recently a number of attempts have been made to address these problems. Many of these efforts aim to achieve this through the use of contrastive loss terms that function as a guide to improve chemical soundness. Common approaches use SupCon or NT-Xent loss terms, but these are severely limited by the cuadratic scaling with respect to batch size and their reliance on large batches for hard negative mining. In this work, we leverage the N-ary comparison framework to modify these losses so that they scale linearly with batch size, while maintaining their ability to impose structure in latent space. Our proposed methodology can be integrated with existing methodologies to obtain latent representations of comparable or improved accuracy with a lower computational cost. We demonstrate the effectiveness of our approach on chemical representation learning for large drug-like molecule databases.

Reduction of Na+ within a Mg2Na2 Assembly: A DFT Study

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This project uses DFT to investigate the unusual reactivity of a low-valent Mg(I) dimer upon THF addition. In 2021, Hill and co-workers reported the synthesis of a low-valent Mg(I) dimer $[\mathrm{SiNDippMgNa}]_2$ (1; SiNDipp = $\mathrm{CH_2SiMe_2N(Dipp)_2}$; Dipp=2,6-$\mathrm{iPr_2C_6H_3}$, which is kinetically stabilised by its bulky SiNDipp ligands.[1] In comparison to related systems, for example the nacnac based structures isolated by Jones in 2007,[2] 1 features Na+ counterions that bridge the two monomeric Mg(I) units through cation - - π interactions with the Dipp rings of the ligand framework, causing enhanced stability. Previous quantum theory of atoms in molecules (QTAIM) and natural bond orbital (NBO) studies have shown that there is a degree of cooperativity between the Mg(I)–Mg(I) bond and the Na+ cations. The Mg(I)–Mg(I) bond is also longer (3.21 Å) than the typical Mg(I)–Mg(I) bond length (2.85 Å) and is indeed closer to that observed in metallic magnesium (3.20 Å).

The unique electronic structure of 1 has allowed it to act as a potent reducing agent via electron donation through its Mg(I)–Mg(I) bond.[3] Upon addition of tetrahydrofuran (THF) to 1, the SiNDipp ligands reorganise from seven membered chelates into a 14 membered macrocycle (2), incorporating both magnesium centres into the macrocycle.[4] 2 exhibits a significantly longer Mg–Mg distance (8.81 Å) that is not representative of a bonding interaction. Concurrently both Mg(I) centres are oxidised to Mg(II) whilst extruding a metallic Na(0) mirror, which has been calculated as a exergonic process ΔG= -50.4 kcal mol^{-1}; BP86-D3BJ(SMD=benzene)/Def2-TZVPP//BP86/Def2-SVP) at 298.15 K. This work explores the mechanism for the macrocyclization process to determine an accessible pathway for the transformation of complex 1 to 2 upon THF addition, using density functional theory (DFT).

Triplets in the cradle: ultrafast dynamics in a cyclic disulfide

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Disulfide bonds (S-S) are common chemical moieties which are most often found in the natural world in the structures of various proteins. Crucially, disulfide linkages are widely thought to be responsible for helping to maintain the structural integrity of proteins through cross-linking between nearby cysteine amino acids. This occurs despite the observation that small molecular disulfides in the gas-phase readily dissociate via S-S bond homolysis when exposed to heat and light. In this work, we investigate theoretically the effect of spin-orbit coupling on the nonadiabatic dynamics of 1,2-dithiane, a small model cyclic disulfide system, when dynamics is initiated on the first excited state. From our surface-hopping simulations, we observe that strong spin-orbit coupling in this molecule promotes efficient intersytem crossing between a dense manifold of singlet and triplet states across a wide region of conformational space. In turn, this is predicted to discourage the permanent recombination of the S-termini to reform an intact S-S bond.

Enhancing Mineral Carbonation Through Molecular Simulations on Diopside Surfaces

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Carbon sequestration is a critical component of global efforts to mitigate the effects of anthropogenic climate change. The accumulation of atmospheric carbon dioxide (CO_2), largely due to industrial activities, is the primary driver of global warming. To limit the impacts of climate change, it is essential to not only remove CO_2 from the atmosphere but also store long-term. One promising approach to this is the natural process of carbonate formation, where CO_2 reacts with silicates and other minerals to form stable carbonate compounds [1-4].

In this study, we applied Machine Learning Molecular Dynamics (MLMD) to enable a much longer time scale to investigate the reaction mechanism but at reduced costs within the accuracy of Ab Initio Molecular Dynamics (AIMD) [5]. Molecular modeling focused on the diopside surface to determine optimal parameters—such as temperature and the addition of NaCl or NaF—to facilitate cation dissociation for use in carbonation reactions.

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Learning Absorption and Emission Properties for Radicals

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Emissive organic radicals are nowadays attractive materials in the next-generation highly efficient organic light emitting diode (OLED) devices. However, due to the spin-contamination problem,the multiconfigurational

charachers of the excited states, and the difficulties in optimisation of excited state geometries, the simulation of the optoelectronic properities is very challenging. In this research, we present a new approach that

for the first time the optoelectronic properties of organic radicals are learned from experimental data in the frame of the ExROPPP method, which is based on the Pariser-Parr-Pople theory. In the simulation of

absorption energy and intensity ratio between D1 and bright state, our trained model showed a huge improvement from the literature parameters.1 For emission energy prediction, our current model also has

advantages in accuracy compared with the mathematical model we built based on the stokes shift.

Investigating TICT Structure of MABN via Excited State Dynamics

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The study of charge transfer states is important for a wide array of photochemistry applications. For example, in Thermally Activated Delayed Fluorescence (TADF), charge transfer has a small overlap, which implies there's a small energy gap between the singlet and triplet states. However, in a charge transfer singlet state, the minimum of energy is typically for a configuration such that the donor and acceptor moiety are perpendicular. This means that there would be a low intersystem crossing, low fluorescence and highly probable radiative decay. The presence of a Local Excitation (LE) state in TADF can help promote intersystem crossing.

The study pertains to MABN and its derivatives, isopropyl/tert-butyl substituents in replacement of the hydrogen (DMABN-iPr, DMABN-tBu). There is the phenomena of dual fluorescence in these molecules in which emission can originate from a LE state or an Intramolecular Charge Transfer (ICT) state. The minima in the emissive excited states will determine whether the charge transfer structure is planar or twisted. The analysis of these molecules will be compared to that of the well-studied DMABN.

The dynamics methods used for this study are a classically propagating method, Trajectory Surface Hopping (TSH), and a quantum propagating method, Linear Vibronic Coupling (LVC) + Multi-Configurational Time Dependent Hartree (MCTDH). They differ in the following ways:

TSH:

- Treatment of all nuclear degrees of freedom. - Each time step requires a single point and gradient calculation for each trajectory. - You need many trajectories to reach convergence. - Some quantum effects will not be captured due to the classical propagation.

MCTDH:

- Restricted set of degrees of freedom. - Only need to build the potential energy surface once. - Grid based method, yielding accurate results. - Need to switch to the diabatic representation to avoid computational singularities.

This work aims to study MABN and its derivatives and understand the influence of the substituent groups on the nature of the emissive minima. By using TSH the important vibrational modes can be identified and compared to the results of the LVC model. A LVC Hamiltonian will be constructed and used alongside the MCTDH method. This leads to generalising the Hamiltonian such that the derivatives of MABN can be described.

Molecular modeling studies to evaluate the bioactivity of H-Thiazine and its derivatives on breast cancer cells.

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Breast cancer poses a major global health burden and remains one of the foremost causes of cancer-related mortality among women [1]. In 2022, it was responsible for approximately 2.3 million new cases and over 666,000 deaths worldwide. Within Sub-Saharan Africa, there were 146,130 diagnosed cases and 71,662 reported deaths, with projections suggesting a dramatic 85.7% increase in incidence and an 89% rise in mortality by the year 2040 [2]. Current treatment strategies for breast cancer include surgery, radiotherapy, chemotherapy, hormonal therapy, and targeted therapies. However, patients who survive the disease often face long-term side effects such as chronic pain, fatigue, anxiety, and functional limitations, all of which severely impact their quality of life [3]. As a result, there is a critical need to discover and develop novel anti-breast cancer agents that are both effective and safer.

One promising approach involves the exploration of thiazine-based small molecules, specifically, 4-phenyl-2H-[1,3]thiazino[3,2-a]benzimidazole-2-imine (H-thiazine) and its eight structural derivatives as potential therapeutic agents against breast cancer. Thiazines are heterocyclic compounds that have both sulfur and nitrogen atoms in their structures. These compounds are known to exhibit a range of biological activities, including anticancer and anti-inflammatory effects [4]. This study used quantum mechanical calculations and Structure-Based Drug Design (SBDD) methods, which utilizes computational techniques to identify, model, and optimize potential drug candidates through a detailed understanding of molecular interactions between ligands and biological targets. DFT analysis revealed that the Nitro compound was the most reactive and least stable, suggesting high susceptibility to nucleophilic attack. Following quantum mechanical calculations, SBDD methods, including molecular docking, ADMET profiling, molecular dynamics simulations, and binding free energy analysis were employed. Among the evaluated compounds, the Bromo- and Methyl-substituted thiazine derivatives showed strong potential, outperforming the reference drug Olmutinib in terms of binding affinity, pharmacokinetics, and stability. These findings highlight their promise as EGFR-targeted anticancer agents and warrant further optimization and experimental validation.

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Approximately beyond the Born-Oppenheimer approximation

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Time-dependent density functional theory (TD-DFT) and extended tight-binding (xTB) were used to investigate the ground vibrational and excited state electronic structure of PTZ-CH_2-Ph-CC-**Ni**-CC-NAP. The characters of states T_1-T_4 were assigned qualitativly using electron density difference maps at equilibrium geometry and two points away from equilibrium geometry. After vibrational perturbation, the electronic ordering of said states changed. This indicates behaviour that goes beyond the Born-Oppenheimer approximation.

Prior to this, a benchmarking study of PTZCH_2-Ph-CC-**Pt**-CC-NAP found that GFN2-xTB gave an excellent balance between cost and accuracy for ground-state calculations. A mixed basis approach of dhf-SVP for Pt and def2-SVP for all other atoms, showed the greatest balance between accuracy and cost-effectiveness in excited state calculations. This benchmarking resulted in an efficient workflow for exploring the properties of d8 metal centred trans acetylide complexes.

Balancing Accuracy and Generalisability for Predicting Hole and Electron Reorganisation Energy of Next Generation Organic Semiconductors

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Accurate prediction of reorganisation energies is essential for the rational design of high-performance organic semiconductors, which underpin emerging technologies such as solar cells, organic light-emitting diodes and biosensors. However, limited dataset sizes and poor generalisability hinder current machine learning modelling approaches. In this study, we systematically evaluate how key machine learning strategies – e.g. feature encoding, dataset size and chemical diversity - affect predictive performance. We benchmarked a range of molecular representations (including hybrid descriptor– fingerprint schemes) and algorithms across datasets of varying size and diversity. We find that hybrid high-dimensional encodings consistently outperform simpler descriptors. Accuracy improves with chemically homogeneous datasets, but generalisability suffers. In contrast, systematic augmentation with diverse structures enhances transferability across chemical space. The best performance (R2=0.791, RMSE = 0.106 eV) was achieved using Gradient Boosting Regression Trees trained on hybrid features with a diverse dataset. Our results provide quantitative design rules for constructing scalable and generalisable ML models, supporting data- driven discovery of next-generation organic semiconductors.

A3EDA: Designing Catalysts from First Principles

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Intermolecular interactions are integral to the efficiency and selectivity of catalytic processes. Even subtle stabilization of transition states (TS) by such interactions can lead to significant enhancements in reaction rates, underscoring the importance of understanding the underlying relationships between interaction and activity. Traditional energy decomposition analysis (EDA) methods, including the absolutely localized molecular orbital EDA (ALMO-EDA), offer valuable insights into the nature of ground state interactions, such as electrostatics, Pauli repulsion, dispersion, polarization, and charge transfer. However, while isolating the contributions of individual interactions to catalysis remains a challenge, it is essential for the rational design of efficient catalysts.

In this work, we introduce an asymmetric adiabatic ALMO-EDA framework to switch on individual interactions between the catalyst and the TS structure, providing a direct causal link between catalyst structure and activity. We apply this methodology to pericyclic and nucleophilic addition reactions, to dissect the specific roles of each interaction component. Our findings reveal distinct classes of catalysts that lower activation barriers through common mechanisms, such as strong electrostatic interactions, significant polarization effects, and charge transfer mechanisms. By systematically analyzing these contributions, we offer actionable insights for the rational design of both organocatalysts and Lewis acid catalysts.

By uncovering how specific intermolecular interactions contribute to catalysis, our approach establishes a generalizable framework for studying a wide array of catalytic systems. Our ongoing research aims to extend this methodology to more complex catalytic processes, advancing the field of catalyst design by predicting the impact of individual intermolecular interactions on reaction kinetics.

Multiwavelet-based prediction of core XPS spectra for thermally-activated delayed fluorescence OLEDs

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Thermally-activated delayed fluorescence (TADF) OLED emitters combine mechanical flexibility and high efficiency with metal-free environmentally sustainable production, lending themselves to low-energy lighting and wearable electronics applications. However, device-realistic screening of emitters requires simulations which capture phase/environment effects on bonding and electronic structure due to the disordered local structure of TADF emissive layers. This can be uncovered for such complex materials using core X-ray photoelectron spectroscopy (XPS) but interpreting XPS data for such systems relies upon the ability to predict core electron binding energy (CEBE) shifts between atoms of the same element in different chemical environments.

Here, we demonstrate that integrating the initial maximum overlap method with the multiwavelet DFT code MRChem provides a method for determining CEBEs which shows robust convergence for system sizes up to at least 100 atoms. Combining this with the clustering of Koopmans' CEBEs, to identify a representative subset of carbon atoms for which to run more expensive ΔSCF calculations, enables the prediction of C 1s XPS spectra for TADF molecules in good agreement with experiment. This will provide a platform for future experimental and theoretical investigation into the (electronic) structure of TADF host-guest thin films.

Electron transfer mediated decay process for the Auger final state of Ne+(1s-1) (H2O)n (n=1,4) cluster

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We have investigated the electron transfer-mediated decay (ETMD) process associated with the Auger final state of the Ne+(1s-1) (H2O) n cluster. After core ionization of Ne, a fast Auger decay takes place which produces Ne2+(2s-2), Ne+2 (2s-1 2p-1) and Ne2+(2p-2) dications. Despite the substantial-high double ionization potential for water (≈ 39 eV), the ETMD channel is open for Ne2+(2p-2) state. We have investigated the ETMD process for the 1S and 1D double ionized states of Ne2+(2p-2) in the presence of different numbers of water molecules. Furthermore, we examine the sensitivity of the bond distance on the ETMD decay width of the Ne2+(2p-2) (1S, 1D) state in the Ne (H2O)2 cluster.

Keywords : Auger Decay, electron transfer mediated decay (ETMD), double ionization potential (DIP)

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Strong Ferromagnetic Coupling between Co and Co+2 with Odd Electron (Anti)aromatic Radicals via Non-Covalent Interaction

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We have aimed to understand the interaction between odd electron aromatic or antiaromatic radicals with cobalt and their dipositive ion to understand the magnetic interaction between them. Density functional theory (DFT) along with the complete active space self consistent field (CASSCF) method has been used to calculate the magnetic exchange coupling constant (J) between the radical molecules and Co/Co^{2+}. The DFT-calculated J ranging from 897 to 6060 cm^{-1} and 2534 to 18574 cm^{-1} for the CASSCF method signifies that the odd electron (anti)aromatic-based magnetic molecules could be useful as strong low-dimensional magnetic materials. Frequency analysis reveals that some of the radicals behave as a transition-state structure in the absence of metal but become stabilized upon the addition of Co or Co^{2+}. The noncovalent interaction (NCI) and electron localization function (ELF) analysis indicate that there is no covalent bonding between radicals and the metal. The absence of covalent bonding between the metal and radicals indicates direct ferromagnetic interaction between them. Aromaticity in the studied Co/Co^{2+}–radical complexes has been evaluated using the nucleus independent chemical shift (NICS), harmonic oscillator model of aromaticity (HOMA), and gauge-including magnetically induced currents (GIMIC) analysis, revealing a complex, multidimensional nature of aromaticity. NICS(1) values indicated that the same ring exhibits both aromatic and antiaromatic behavior, depending on the spatial orientation of the metal center. The HOMA value shows a strong correlation with the magnetic exchange coupling constant (J), supporting a link between structural aromaticity and magnetic interaction. The aromaticity index GIMIC is not well correlated with other aromaticity indexes like NICS and HOMA. These observations highlight the need for multiple aromaticity descriptors to fully capture the complex aromatic character of these systems.

Bond Dissociation of F2, N2 and O2 through the Prism of SCF Stability Analysis

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n this presentation we compare bond dissociation potential energy profiles of a series of homonuclear diatomic molecules (N2 and F2) computed using inexpensive Stability Analysis on Density Functional Theory (DFT) and Hartree-Fock(HF) calculations with post-Hartree-Fock and Multireference methods, such as Complete Active Space SCF (CASSCF), which are supposed to be better suited for computational modeling of bond breaking/formation.

The discussion is focused on the insights into the extent to which all these methods are able to address static and dynamic electronic correlation energies at different levels of theory. Progressive unraveling of pi and sigma bonds along the dissociation curve will be illustrated using Natural Bond Orbitals visualizations.