Book Of Abstracts

14th OpenMolcas Developers' Workshop

5 – 7 October 2026

Valencia, Spain

Contents

- David Picconi (Heinrich Heine University Düsseldorf, Germany). *Photodynamics of Multi-Mode Multi-State Molecular Quantum Systems in a Dissipative Environment*.
- Igor Schapiro (TU Dortmund University, Germany). *Hybrid QM/MM NAMD Simulations*.
- Irene Conti (Università di Bologna, Italy). *Light-Activated Mechanisms in DNA: The Origin of Multiple Rare Mutations?*
- Luis Manuel Frutos (Universidad de Alcalá, Spain). *Nonadiabatic Molecular Dynamics under External Forces in OpenMolcas*.

2.2. Tuesday · 6th October

- Francesco Aquilante (EPFL, Switzerland). *Electron Correlation: The Roads Less Traveled By*.
- Alessandro Nardi (Nantes Université, CNRS, CEISAM, France). *Extending Automated Selection of Nuclear Coordinates for Reduced Dimensionality Non-Adiabatic Dynamics: Sparse PCA and Alternative Coordinate Systems*.
- Giacomo Bassi (Technical University of Munich). *Density Matrix Renormalization Group to Target Core-Excited States of Transition-Metal Complexes*.
- Sergey Bokarev (Technical University of Munich, Germany). *An Ultrafast Dynamics Kaleidoscope with OpenMolcas*.
- Jesús Lucia-Tamudo (University of Vienna, Austria). *Anharmonic Vibronic Coupling Model in Trajectory Surface Hopping Simulations at the MS-CASPT2 Level of Theory*.
- Leticia González (University of Vienna, Austria). *One, Two, Three Irons: From Multireference Photochemistry to Magnetic Frustration*.

2.3. Wednesday · 7th October

- Javier Cerezo (Universidad de Murcia, Spain). *Vibrationally Resolved Spectroscopy at Multireference Perturbative Level: An OpenMolcas–FCClasses3 Interface*.
- Marek Krośnicki (University of Gdańsk, Poland). *Can We Describe Electron Charge Transfer in Magnetite in Embedded Cluster Calculations?*

- Jesús Cerdá (Universitat de València – Instituto de Ciencia Molecular [ICMol], Spain). *DRAGON@OpenMolcas: A New Interface for Diabatization of Excited States in Molecular Aggregates*.
- Valera Veryazov (Lund University, Sweden). *Stop Programming? AI Agents for Scientific Software Development*.
- Marco Garavelli (Università di Bologna, Italy). *First Baby Steps Towards a GPU-Based CASSCF–CASPT2 (OpenMolcas?) Software*.
- Coen de Graaf (Universitat Rovira i Virgili, Spain). *GronOR, a Massively Parallel and GPU-Accelerated Code for Non-Orthogonal Configuration Interaction*.

3. Flash Communication

OpenMolcas after COVID-19

Ignacio Fdez. Galván[1]

[1] Department of Physics, TU Dortmund University, D-44227 Dortmund, Germany, email: *ignacio.fernandez@tu-dortmund.de*

The Molcas project has a long history, dating back at least to 1989. Some 30 years later the transition of most of its code base to an open-source model was finalized and the first OpenMolcas article was published in late 2019.[1] Shortly after that, the world was struck by a pandemic that kept most researchers isolated and substantially altered existing habits and workflows. This was taken as an opportunity to initiate a much needed effort of modernization and restructuring of the OpenMolcas source code to better take advantage of current compiler features and facilitate future maintenance and further developments. A milestone was reached this year, when the conversion to free-format (a.k.a. Fortran-90) code was completed. It is therefore timely to report on the motivations, goals, and details of the code modernization, with examples of the most significant changes and an outlook to the future.

In the meantime, development in OpenMolcas has not stalled and new methods and features have continued to be added to the package, especially in the form of interfaces with external software, as evidenced in the latest publication.[2] Some useful tools for setting up and analyzing typical calculations are often overlooked and will be reminded.

Figure 1. OpenMolcas wears a face mask.

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Multireference Wave Functions in Polarizable Solvent Environments: The Fluctuating-Charge Approach

Chiara Cappelli[1]

[1] Scuola Normale Superiore, Piazza dei Cavalieri 7 I-56126 Pisa (IT), *chiara.cappelli@sns.it*

The accurate description of molecular systems in solution requires a balanced treatment of the electronic structure of the solute and its interaction with the surrounding environment. This becomes particularly challenging when the solute exhibits a pronounced multireference character and, at the same time, mutual solute–solvent polarization plays a significant role.

In this talk, I will present our polarizable embedding approach for solvated systems, in which the environment is described through the atomistic fluctuating-charge (FQ) force field.[1-3] The FQ charges respond self-consistently to the quantum-mechanical electronic density, providing an efficient description of mutual polarization while retaining an explicit molecular representation of the solvent.

Particular emphasis will be placed on recent extensions of this framework to multireference wave functions within multiconfigurational self-consistent field methods (MCSCF/FQ)[4,5] and the density matrix renormalization group (DMRG/FQ).[6]

Selected applications to solvated molecules illustrate the role of environmental polarization and the potential of these developments for multireference simulations of complex molecular systems within OpenMolcas.

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From Multiconfigurational Wave Functions to Single/Oligo-CSF Descriptions: The Quantum Anamorphosis Journey

G. Li Manni[1], M. Song[1], L. Bonfirraro[1],, K. Abdulhaq, C. Calzado[2], V. Robert[3], I. Fdez. Galván[4], R. Lindh[5]

[1] Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart (Germany), email: *giovannilimanni@gmail.com*
[2] Universidad de Sevilla, c/Profesor García González, s/n., 41012 Sevilla (Spain),
[3] CNRS/Université de Strasbourg, 4 rue Blaise Pascal, 67000 Strasbourg (France),
[4] TU Dortmund University, Otto-Hahn-Straße 4, 44227 Dortmund (Germany),
[5] Uppsala University, P.O. Box 576, Uppsala 75123 (Sweden)

First introduced in 2020, Quantum Anamorphosis (QA)[1,2] is a state-preparation strategy that, through tailored molecular orbital transformations, enables a dramatic compression of ground- and excited-state[3] electronic wave functions in spin-adapted many-body bases. This is particularly advantageous for systems with many unpaired electrons, which remain a major challenge in modern electronic structure theory. QA has been successfully applied across a range of problems, from low-dimensional spin models to polynuclear transition metal (PNTM) complexes, key active sites in biological and biomimetic catalysis.[4-6] For large PNTM clusters, such as the P-cluster and FeMo-cofactor, a genetic-algorithm-driven QA protocol has also been developed.[7] The resulting wave function compactification is highly beneficial for methods that approximate full-CI solutions, such as FCIQMC, GAS or selected-CI methodologies. QA has enabled the development of efficient quantum Monte Carlo multi-reference perturbation theory approaches[8] and a Stochastic Difference-Dedicated Configuration Interaction algorithm.[9] In addition, QA has led to a perturbatively corrected, spin-adapted restricted open-shell Hartree-Fock method, retaining the computational cost of standard ROHF while allowing access to low-spin states with correct spin symmetry.[10] In this talk, I will present the key principles of QA, recent methodological developments, and applications to magnetic interactions in representative biomimetic clusters[4-6] ($Fe(III)_4S_4$, $Mn(IV)_3O_4$, $Co(II)_3Er(III)O_4$) and larger architectures such as the P-cluster.[7] Magnetic exchange couplings, spin ladders, and temperature-dependent susceptibilities provide stringent benchmarks of the accuracy achievable with QA.

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Three Views on Nonadiabatic Dynamics

Sandra Gómez[1,2]

[1] Departamento de Química, Módulo 13, Universidad Autónoma de Madrid, Madrid, Spain, email: sandra.gomezr@uam.es
[2] IADCHEM, Institute for Advanced Research in Chemistry, Universidad Autónoma de Madrid, Madrid, Spain

The seminar Three Views on Nonadiabatic Dynamics will explore three complementary approaches for simulating nuclear motion in photoexcited molecular systems: Multiconfiguration Time-Dependent Hartree (MCTDH)[1], Direct Dynamics Variational Multiconfigurational Gaussian (DD-vMCG)[2], and Trajectory Surface Hopping (TSH)[3].

Using three representative systems, the talk will show how each method offers a different perspective on nonadiabatic dynamics, and how the nature of the problem influences the choice of approach. The comparison will show how the choice of method can affect the results and understanding of photoinduced processes, and why it is important to compare different approaches in a clear and consistent way.

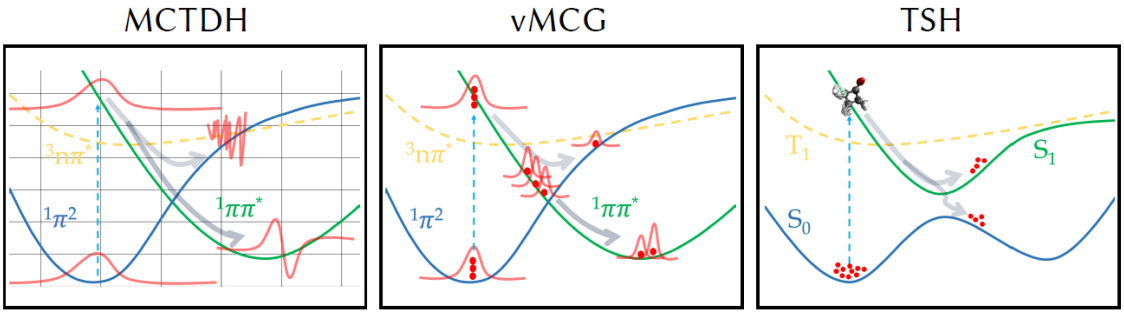

Figure 1. Nonadiabatic dynamics methods that will be presented, each assigned to a representative system.

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OPAL – Orbitals, Plotting, Analysis, and Layout

Luca De Vico

Dipartimento di Biotecnologie, Chimica e Farmacia, Università degli Studi di Siena, Via Aldo Moro 2, 53100, Siena (Italy),
email: *Luca.DeVico@unisi.it*

In this presentation, I will introduce OPAL, a suite of self-contained browser tools for computational chemistry.[1] OPAL tools were built so you don't have to install anything on a computer: they run in your browser. They are based on JavaScript and are self-contained, which means they do not require an active internet connection to work. The tools are still being developed (at the time of writing) but will soon be part of the OpenMolcas/Tools folder and distributed under the same license. A separate clone will also be distributed as a standalone progressive web app (PWA).

OPAL tools are, for the most part, usable with any computational chemistry program that produces a Molden output file, directly or indirectly. However, they are specifically geared toward reading and exploiting OpenMolcas HDF5 output files, which unlocks advanced features. Their intended use is to simplify the life of any OpenMolcas user when, *e.g.*, planning a multiconfigurational calculation, choosing orbitals for an active space, analyzing the results, and presenting them (Figure 1). OPAL tools can also draw and visualize molecules, compare geometries, analyze geometry optimizations, and run simple OpenMolcas calculations remotely.

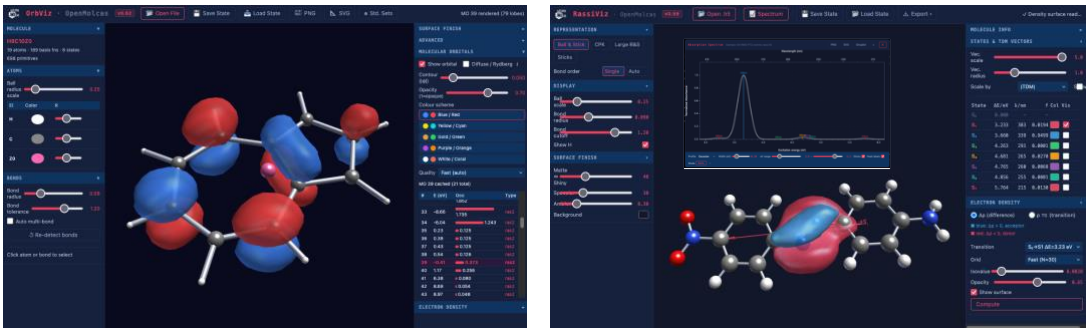

Figure 1: Screenshots of two OPAL tools. Left: OrbViz quickly visualizes orbitals, already divided between RAS spaces, if present. Right: RassiViz analyzes an HDF5 Rassi output and visualizes the electron density difference or the transition between two electronic states. It can also produce a simulated absorption spectrum (inset).

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From Quantum Algorithms to Better Therapies and Materials: Transitioning from Near-Term to Early Fault-Tolerant Impact in Quantum Computing for Molecular Simulations

Stefan Knecht[1]

[1] Algorithmiq S.r.l., Via della Chiusa 15, 20123 Milano, Italy, email: *stefan@algorithmiq.fi*

Quantum computing is at the threshold of transforming molecular simulation, offering solutions to some of the most intractable challenges in multiconfigurational chemistry.

In this talk, I will outline our project, awarded as the sole winner of the Wellcome Leap Quantum4Bio Global Challenge[1]. I will discuss a series of groundbreaking advancements, enabling large-scale quantum chemistry simulations that were previously infeasible and setting the stage for the first credible demonstration of useful quantum advantage in molecular modelling. With these innovations at hand, I will present an end-to-end procedure — from defining a molecular structure to computing error-mitigated and error-corrected ground- and excited-state energies as well as molecular properties for molecules and materials using contemporary quantum hardware at a 50+ qubit scale. This marks a significant step towards practical, early-fault tolerant hardware-compatible quantum chemistry simulations for molecular systems as well as materials. Likewise, benchmarking remains crucial. I will highlight briefly our novel high-performance DMRG implementation for large-scale quantum-chemistry simulations. The code leverages multi-GPU and multi-node architectures to enable large active spaces and bond dimensions, with strong scaling and competitive performance against established DMRG implementations.

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14th OpenMolcas Developers' Workshop
5 – 7 October 2026 | Valencia, Spain

CI algorithms in OpenMolcas

Roland Lindh [1]

[1]The Quantum Chemistry Group
Uppsala University
Sweden

The presentation deals with the implementation of a new library of CI algorithms adapted to the OpenMOLCAS open-source codes. This introduce the Algebraic Spin-Tensor Approach, ASTA, as implemented in the Split-Graph UGA formalism of Malmquist in the basis of Configuration State Functions, CSFs. This has been co-developed with the Slater-Determinant code of Vancoille -- the Faroald routines. The target is to develop general stand-alone codes for the underlying CI algorithms of methods as CASSCF, RASSCF, GASSCF, LASSCF, and all forms of selected CI approaches.

Tuning photochemistry with static electric fields

Nicolas Ferré[1] (presenting author), Isabel Eder[1], Roland Lindh[2], Miquel Huix-Rotllant[1]

[1] Aix Marseille Univ, CNRS, ICR, Marseille, France, email: *nicolas.ferre@univ-amu.fr*
[2] Department of Chemistry for Life Sciences, Uppsala University, Uppsala 75123, Sweden

When a chromophore features electronic states with significantly different dipole moments, the application of an external electric field shifts their relative energies, possibly modifying its photochemistry. In this talk, I will present a simple alternative method to OpenMolcas &FFPT implementation of a static electric field, using the ElectroStatic Potential Fitted (ESPF) charge and dipole operators.[1] Following the validation of this new approach, basic applications using ethylene and penta-2,4-dieniminium cation (PSB3) highlight its capabilities, especially regarding their minimum-energy conical intersection structures.

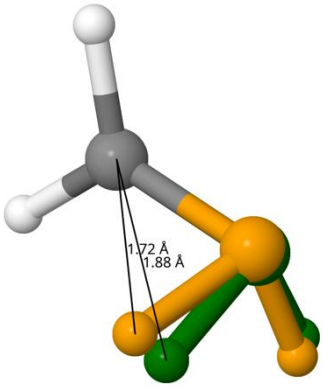

Figure 1. Comparison of ethylene MECI structures in the presence (green) or absence (orange) of a 100 MV/cm external static electric field parallel to its ground state dipole moment.

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Photodynamics of multi-mode multi-state molecular quantum systems in a dissipative environment

David Picconi[1]

[1] Heinrich Heine University Düsseldorf, email: *david.picconi@hhu.de*

Ultrafast processes in condensed phase photoexcited molecular systems involve the transition from a coherent dynamical regime – where a precise phase relation exists between different wave packet components – to incoherent "classical-like" dynamics. The decoherence process is driven the dissipation due to the surrounding molecular environment.

From the computational viewpoint, modelling the dynamics of nonadiabatic vibronic quantum systems interacting with fluctuating environments becomes especially challenging when the system's high dimensionality precludes the calculation of its eigenstates. To overcome this limitation, a novel eigenstate-free formalism is introduced. This approach represents the open quantum system as a mixture of high-dimensional, time-dependent wave packets, governed by coupled Schrödinger equations,[1] while the environment is modeled using a multi-component quantum master equation.[2]

A computationally efficient implementation of this formalism employs a variational Gaussian/ multiconfigurational time-dependent Hartree (G-MCTDH) ansatz for the wave packets and propagates the environment dynamics using hierarchical equations truncated at the first or second level. The methodology is validated through simulations of the dynamics in vibrationally excited adsorbates,[2] multichromophoric aggregates, and symmetry-breaking donor-acceptor systems,[3,4] explicitly incorporating multiple vibrational modes and accounting for the effects of the residual bath in full dimensionality. These results demonstrate the potential of the approach as a powerful quantum dynamical method for modeling complex system–bath interactions involving numerous degrees of freedom across multiple time scales.

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Light-Activated Mechanisms in DNA: the Origin of Multiple Rare Mutations?

Irene Conti

Industrial Chemistry Department "Toso Montanari", Università di Bologna, Italy; email: *irene.conti@unubo.it*

DNA contains all the information of any living being, but unfortunately, we are still unclear about the effect of light (which fuels life itself) on this pivotal element of our existence and its replication.

Over billions of years, DNA has developed sophisticated photo-protection ultra-fast decay processes, preventing genetic code damages caused by a prolonged permanence on top of reactive excited states. Nevertheless, it is of primary preventive importance to characterize these secondary processes that lead to harmful mutations. A computational theoretical approach could be a useful tool to unveil these rare decay mechanisms, as it is capable of simulating the corresponding transient spectra, which can then be directly compared with experimental signals. As these are electronic processes within a highly complex system, the hybrid QM/MM 'COBRAMM' method[1] (developed in our group) has been employed, adopting the open-source OpenMolcas software[2] for the high level PT2 calculations for the QM part and Amber for the MM environment. The computing effort enabled us to extend the QM part to four interacting bases, corresponding to two pairs of stacked bases, allowing us to deciphering both base-pairing and π-stacking interactions.

The figure below shows how this QM/MM model enabled to explore very different degenerative processes[3], combining the high level accuracy of the QM method with the flexibility of the MM environment.

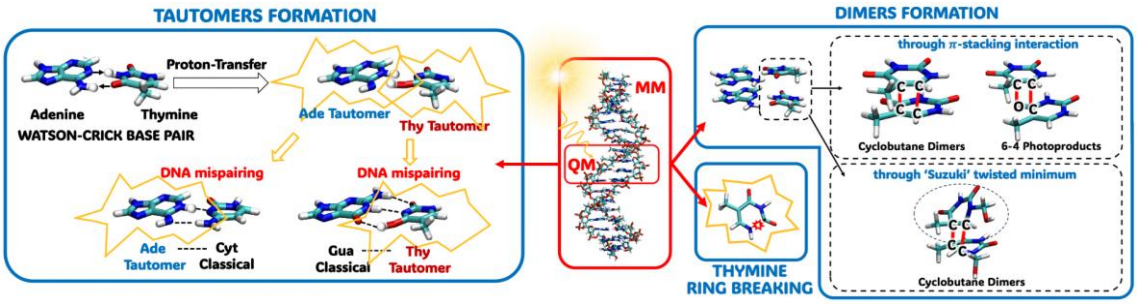

Figure. Simulated rare DNA mutations[3]: Tautomer formation induced by base-paired proton transfer process, leading to DNA mispairing (left-side); Cyclobutane and 6-4 Thymine dimers formation (right-side, above). (c) Thymine ring-opening (right-side, below).

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14th OpenMolcas Developers' Workshop
5 – 7 October 2026 | Valencia, Spain

Nonadiabatic Molecular Dynamics under External Forces in OpenMolcas

Luis Manuel Frutos[1], Ignacio Fdez. Galván,[2] Igor Schapiro[2]

[1]University of Alcalá, Department of Analytical Chemistry, Chemical Physics and Chemical Engineering, Alcalá de Henares, Madrid, Spain. [2]Department of Physics, Technical University Dortmund, Germany

We present recent developments in the external-force module (EXTF) of OpenMolcas permitting to perform molecular dynamics simulations under the effect of mechanical forces. Particular emphasis is placed on wall-type external forces, in which a molecular system is compressed between two virtual walls.[1] This model provides a simple representation of directional molecular deformation and of impact-like conditions relevant to ball-milling simulations.

The EXTF implementation allows mechanical forces to be applied along selected internal coordinates and using different time profiles, making it possible to describe both stationary and time-dependent mechanical perturbations during molecular dynamics. Wall-type forces can therefore be incorporated directly into molecular trajectories to investigate how mechanical compression modifies potential energy surfaces, reaction pathways.

These external-force models are mainly intended to be combined with excited-state nonadiabatic molecular dynamics, using recent improvements in the OpenMolcas nonadiabatic dynamics framework. These developments provide a general computational framework for studying mechanically controlled excited-state processes and for exploring the connection between molecular deformation, electronic-state transitions, and chemical reactivity in mechanophotochemistry and mechanophotocatalysis.[2]

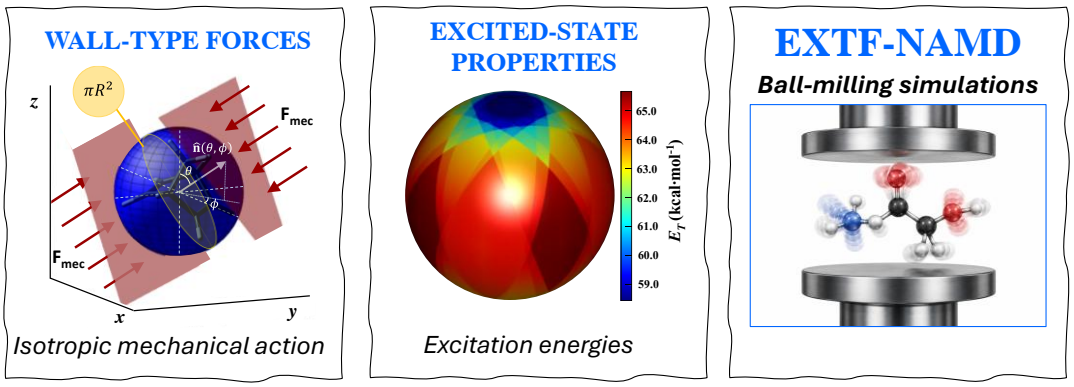

Figure 1. Schematic overview of wall-type external forces (left), their effect on excited-state energies, e.g. excitation energies with impact direction (center), and nonadiabatic molecular dynamics under wall-type compression representative of ball-milling conditions (right).

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Extending Automated Selection of Nuclear Coordinates for Reduced-Dimensionality Non-adiabatic Dynamics: Sparse PCA and Alternative Coordinate Systems

Alessandro Nicola Nardi[1], Morgane Vacher[1]

[1] Nantes Université, CNRS, CEISAM, UMR6230, F-44000 Nantes, France, email: *alessandro.nardi@univ-nantes.fr*

Simulating non-adiabatic excited-state dynamics is central to understanding light-driven processes, but the cost of accurate fully-quantum simulations scales poorly with the number of nuclear degrees of freedom, so reduced-dimensionality models are typically built using chemical intuition. This work introduces two extensions to an automated protocol,[1] implemented in OpenMolcas, for selecting the most relevant nuclear coordinates for reduced-dimensionality non-adiabatic dynamics: (i) a new subspace-extraction methods, Sparse PCA, complementing the original PCA-based approach, and (ii) their application across three coordinate representations: Cartesian coordinates, dimensionless normal mode displacements, and non-redundant internal coordinates, with the latter allowing for non-linearity in the subspace. The protocol involves two steps: full-dimensional low-level dynamics are performed and analysed to extract a reduced coordinate subspace using the above methods and representations, and the dynamics are then rerun within the reduced subspace and benchmarked against the full-dimensional reference using electronic and geometric descriptors. Tested on three reactions - *trans*-to-*cis* isomerization of azomethane, butyrolactone ring-opening, and excited-state intramolecular proton transfer in 3-hydroxychromone - the protocol successfully identifies reduced coordinate spaces in each case, offering a systematic, low-bias way to build reduced-dimensionality models for fully-quantum dynamics methods.

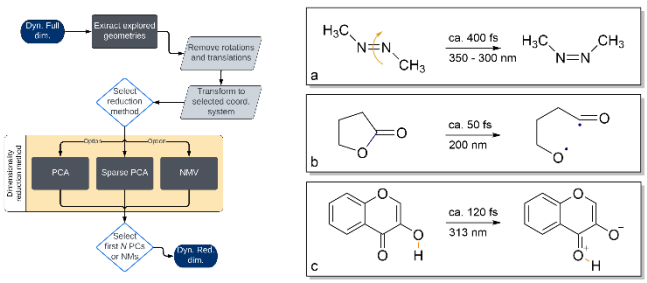

Figure 1. Schematic workflow of the protocol and benchmark reactions considered in this work.

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14th OpenMolcas Developers' Workshop
5 – 7 October 2026 | Valencia, Spain

DENSITY MATRIX RENORMALIZATION GROUP TO TARGET CORE EXCITED STATES OF TRANSITION METAL COMPLEXES

Giacomo Bassi[a], Mihael Erakovic[b,c], and Sergey I. Bokarev[a,d]

[a] Department of Theoretical Chemistry, Technical University of Munich, Germany, *g.bassi@tum.de*
[b] Department of Chemistry and Applied Biosciences, ETH Zurich, Switzerland
[c] Department of Physical Chemistry, Ruđer Bošković Institute, Zagreb, Croatia
[d] Physics Institute, University of Rostock, Germany

Core-level spectroscopy offers a powerful tool to probe the electronic structure of molecules, but the accurate simulation of core-excited states remains challenging in systems with strong electron correlation. In this work, we combine core-valence separation (CVS) with the density matrix renormalization group (DMRG) to create an efficient and scalable framework for targeting core-excited states with large active spaces. The method avoids the need to resolve the full manifold of lower-lying valence states, and it is therefore well suited for difficult excited-state problems in multireference chemistry.

The technique, in its CVS-DMRGSCF flavor, fully relies on the Open Molcas infrastructure and well interfaces with all the existing modules. We tested our approach on different systems, including two different transition metal complexes. The results show that the method is robust for different types of core orbitals and remains effective even in cases where standard multireference treatments become impractical. Furthermore, the CVS framework is extended to a multi-core-valence separation scheme to describe hole-screening effects in doubly core-excited states. We also analyze the core-valence separation using quantum-information diagnostics, which provide a clearer picture of how the approximation acts on the wavefunction. Finally, a second-order perturbative correction is introduced to improve the accuracy of the excitation energies while preserving the computational advantages of the approach.

Together, these results establish core-valence separated DMRG as a practical tool for modeling X-ray spectra in strongly correlated molecular systems. The multi-core generalization could also be applied to systems with multiple metallic centers, where several localized core regions interact with a shared valence manifold. This opens a route toward more flexible and physically realistic simulations of complex core-level spectra in extended or multi-center transition metal systems, and it offers a way to core excited state specific targeting in the multi reference context.

AN ULTRAFAST DYNAMICS KALEIDOSCOPE WITH OPENMOLCAS

Sergey Bokarev[a,b]

[a] Department of Theoretical Chemistry, Technical University of Munich, Germany, *sergey.bokarev@tum.de*
[b] Institute of Physics, University of Rostock, Germany

OpenMolcas has become one of the standard tools for studying core-excited states, owing to its efficient multireference methods and capabilities for treating spin–orbit coupling (SOC), projection techniques, and CVS-DMRG. It is widely used by groups across the globe to predict various types of static X-ray spectra. Moreover, OpenMolcas frequently serves as an electronic-structure engine for theoretical studies of time-resolved X-ray spectroscopy, with a major focus on nonadiabatic nuclear dynamics. Recent advances in X-ray sources and instrumentation, however, increasingly enable the observation of processes occurring on the electronic timescale.

In my talk, I will give an update on our recent developments connected to OpenMolcas aimed at describing such ultrafast electronic processes. Their successful simulation requires not only reliable electronic-structure methods, as available in OpenMolcas and further developed in our group, but also an accurate treatment of ionization, autoionization, and coupled electron–nuclear dynamics. Each of these components presents substantial challenges of its own and requires rather different theoretical and computational approaches. I will discuss how these different pieces can be brought together into a methodological kaleidoscope, combining different patches of electronic structure and dynamical methods to simulate ultrafast electronic and coupled electron–nuclear dynamics.

Anharmonic Vibronic Coupling Model in Trajectory Surface Hopping Simulations at the MS-CASPT2 level of theory

Jesús Lucia-Tamudo[1], Leticia González[2]

[1] Institute of Theoretical Chemistry, Faculty of Chemistry, University of Vienna, Währinger Straβe 17, 1090, Vienna, Austria, email: *jesus.lucia.tamudo@univie.ac.at*

In this contribution, we present anharmonic vibronic coupling (AVC) Hamiltonians in combination with trajectory surface hopping (TSH) dynamics, providing a more flexible description of potential energy surfaces (PESs) than standard linear vibronic coupling (LVC) models. Furthermore, the reparametrization procedure has been automated, considerably reducing the manual effort traditionally associated with the reconstruction and fitting of multiple vibrational modes.

LVC models constitute a widely employed framework for nonadiabatic quantum dynamics simulations due to their efficient parametrization of the PESs involved in excited-state relaxation processes. However, standard LVC models rely on the harmonic approximation and on a local expansion around a reference geometry, limiting their applicability in systems displaying large-amplitude motions, conformational flexibility, or strongly anharmonic vibrational modes governing the nonadiabatic dynamics. To overcome these limitations, some quantum dynamics studies have previously explored reparametrized vibronic Hamiltonians capable of reconstructing problematic regions of the PES beyond the harmonic approximation[1].

TSH provides a computationally cheaper alternative to fully quantum dynamical propagation. Nevertheless, standard on-the-fly TSH simulations remain computationally demanding due to the need to evaluate electronic energies, gradients, and couplings at every time step of the dynamics. For this reason, previous studies introduced the combination of standard LVC Hamiltonians with TSH dynamics as an efficient alternative to computationally demanding on-the-fly simulations[2-5]. However, these approaches still relied on the harmonic LVC approximation, thus inheriting the same limitations associated with anharmonic and flexible systems. Combining AVC Hamiltonians with TSH therefore preserves the computational efficiency characteristic of the standard LVC/TSH framework while considerably extending its applicability.

Our results show that the proposed methodology is capable of reproducing the main features of photoinduced relaxation dynamics, including intersystem crossing, in systems where the standard LVC approach fails, while maintaining a very low computational cost.

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One, Two, Three Irons: From Multireference Photochemistry to Magnetic Frustration

Leticia González [1]

[1] Institute of Theoretical Chemistry, Faculty of Chemistry, University of Vienna, Währingerstrasse 17, 1090 Vienna, Austria
email: *leticia.gonzalez@univie.ac.at*

Iron-based chemistry can provide many electronic-structure challenges, from photoinduced spin changes to multimetallic photochemistry and magnetic frustration. In this presentation, I will discuss three recent examples spanning one, two, and three iron centers. For an Fe(III) spin-crossover complex, multistate RASSCF calculations with OpenMolcas were used to construct a vibronic Hamiltonian for nonadiabatic dynamics across multiple spin manifolds.[1] In bimetallic fulvalene complexes, multireference calculations were employed to rationalize the markedly different photochemical behavior of Fe- versus Ru-containing systems.[2] Finally, I will briefly discuss triangular $Fe(III)_3O$ motifs in metal–organic frameworks, where magnetic frustration poses a different electronic-structure challenge and requires more approximate treatments.[3]

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Vibrationally Resolved Spectroscopy at Multireference Perturvative Level: An OpenMolcas–FCClasses3 Interface

Javier Cerezo[1], Juliana Cuellar-Zuquin[2] , Fabrizio Santoro[3] and Javier Segarra-Martí[2]

[1] Departamento de Química Física, Universidad de Murcia, 30100 Murcia Spain, email: *javier.cerezo@um.es*
[2] Instituto de Ciencia Molecular, Universitat de Valencia, P.O. Box 22085, ES-46071 Valencia, Spain
[3]Consiglio Nazionale delle Ricerche, Istituto di Chimica dei Composti Organo Metallici, Via G. Moruzzi 1, I-56124 Pisa, Italy

We present an interface between the open-source electronic structure packages OpenMolcas[1] and FCClasses3[2] for the simulation of vibrationally resolved spectra within the harmonic approximation at the CASPT2/RASPT2 level. The interface parses OpenMolcas outputs, automates FCClasses3 input generation, and streamlines the evaluation of transition dipole derivatives required for Herzberg–Teller (HT) contributions.

Applications to electronic absorption, resonance Raman and electronic circular dichroism span nucleobases, semi-rigid organic dyes, and chiral organic chromophores. Comparison with experiment and, where appropriate, TD-DFT shows that suitable Franck–Condon (FC) or Franck–Condon/Herzberg–Teller (FCHT) models yield realistic vibrationally resolved spectra, demonstrating the accuracy achievable by combining vibronic models with a CASPT2 electronic-structure description (Figure 1). Overall, the OpenMolcas–FCClasses3 interface offers an automated and flexible route from multiconfigurational calculations to the quantitative simulation of experimental spectroscopic band shapes.

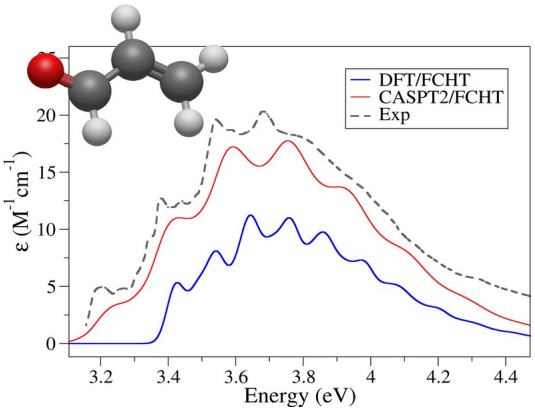

Figure 1. Experimental and simulated UV/vis spectra of acrolein using CASPT2 and DFT.

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Can we describe electron charge Transfer in Magnetite in embedded cluster calculations

Marek Krośnicki[1], Kamil Nalikowski[1], Jakub Kłos[1], Wit Krośnicki[1], Valera Veryazov[2]

[1] Institute of Theoretical Physics and Astrophysics, University of Gdansk,
ul. Wita Stwosza 57, 80-308 Gdansk, Poland, email: *marek.krosnicki@ug.edu.pl*
[2] Division of Computational Chemistry, Chemical Center, Lund University, P.O. Box 124, SE-221 00 Lund.

The Ab Initio Model Potential Embedded Cluster (AIMP) method allows for the description of defects in solids, particularly in wide-band-gap ionic crystals [1]. The AIMP model potentials are optimized in a self-consistent procedure for each atom of the host lattice [2]. In this contribution, we report an application of the AIMP embedded-cluster method to the description of the electronic structure of magnetite. Fe_3O_4 is a mixed-valence oxide. At low temperatures, below the Verwey transition temperature, $T_V = 122$ K, Fe_3O_4 exhibits a charge ordered structure in which the octahedrally coordinated iron atoms located at the B1 and B2 sites have formal charges of 2+ and 3+, respectively [3]. Above this temperature, magnetite undergoes a structural transition, after which the long-range charge ordering of the B1 and B2 sites is removed, and the B sites acquire an average charge of +2.5. The Verwey transition is associated with an approximately two-order-of-magnitude increase in electrical conductivity, without a significant change in the band gap between the low- and high-temperature phases. In this talk, we construct embedded-cluster models of the Trimeron structures of magnetite (B sites:Fe^{3+}-Fe^{2+}-Fe^{3+}). We consider Fe_2O_{10} embedded clusters and model both the polaron hopping process and lattice vibrations that drive changes in the geometry of the B1–Fe^{2+} and B2–Fe^{3+} sites. We demonstrate using embedded clusters that long-range charge ordering creates a barrier to electron hopping, including the blocking of some Trimerons under the assumption of a static structure. In contrast, in the high-temperature structure, assuming an average valence of +2.5 creates a symmetric environment in which electron hopping is almost costless. Therefore, as suggested in the literature, there must be a mechanism that generates short-range charge ordering, which slows down the movement of charge carriers. We discuss the high- and low-spin multiplicities of the many-electron states of the cluster structures within the RASSCF/RASPT2/RASSI methodology [4].

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DRAGON@OpenMolcas: A New Interface for Diabatization of Excited States in Molecular Aggregates

Jesús Cerdá[1], Pau Armada[1], Juan Aragó[1]

[1] Instituto de ciencia molecular, c/ Catedrático José Beltrán 2, Paterna, 46980, Spain., email: *jesus.cerda@uv.es*

The accurate description of excited states in multifragment molecular systems requires diabatic representations that cleanly separate local excitations (LE) from charge-transfer (CT) states. Most existing diabatization tools, however, are either limited to a single type of state or rely on single-reference electronic structure methods, severely restricting their applicability to the complex, strongly correlated systems encountered in photochemistry and molecular aggregates.

We present DRAGON, a software package for the diabatization of multiple excited states in multifragment systems. DRAGON is based on the Fragment Particle-Hole Diabatization (FPHD) scheme.[1,2] FPHD exploits the particle-hole character of excitations projected onto molecular fragments to construct a well-defined diabatic basis that simultaneously captures both local and charge-transfer states — a combination that most alternative approaches cannot achieve. Beyond providing a clean diabatic representation, DRAGON enables the characterization of adiabatic states in terms of their LE and CT contributions, and the direct computation of electronic and excitonic couplings between fragments.

DRAGON can be interfaced with several quantum chemistry codes, though existing interfaces are limited to single-reference methods. The interface with OpenMolcas, currently under active development, is the first to bring full multireference capability to DRAGON, enabling the construction of diabatic Hamiltonians from CASSCF and RASSCF wavefunctions — essential for systems where static correlation and multi-configurational character play a decisive role.

These capabilities have been put to work in applications of direct relevance to molecular photophysics and materials science. DRAGON has been used to simulate the absorption spectra of molecular aggregates, revealing how intermolecular interactions redistribute oscillator strength and reshape spectral line shapes. It has also been employed to build effective model Hamiltonians that capture the essential physics of exciton transport in organic materials,[3,4] providing a rigorous first-principles foundation for subsequent semiclassical dynamics simulations. We will describe the theoretical foundations of FPHD, the current status of the OpenMolcas interface, and discuss some applications in detail.

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Stop Programming? AI Agents for Scientific Software Development

Valera Veryazov[1]

[1] Division of Computational Chemistry, Department of Chemistry, Lund University, P.O. Box 124, SE-221 00 Lund, Sweden; *valera.veryazov@teokem.lu.se*

Scientific software often outlives the technologies used to build it. More than twenty years ago I developed GV, the first graphical user interface for Molcas. It later evolved into LUSCUS, but its C code remained dependent on external graphics libraries such as GLUT. Maintaining such software requires continuous adaptation to changing APIs, operating systems, compilers, and libraries - work that is necessary but rarely scientific.

I recently used Claude as a coding agent to create an Emscripten/WebAssembly version that runs entirely in a browser. A working version appeared within a few days. The same agent-assisted workflow now supports development of lgv[1], the current descendant of GV and LUSCUS. lgv reads and visualizes many molecular and volumetric formats, including LUSCUS grids, Molden, XYZ, CIF, and OpenMolcas input, and supports structure editing, orbitals, densities, and electrostatic potentials. The important result is not that an agent can translate C. It can navigate a mature code base, diagnose defects, and implement substantial new functionality, including molecular-mechanics optimization and construction of electrostatic potentials.

A second example comes from OpenMolcas itself. Given a scientific description of a new property derived from a density matrix, an agent can search the repository, determine where the relevant quantities are produced and stored, find an appropriate integration point, and implement new code using the conventions of the existing program. The scientist can concentrate on defining the property and validating its mathematical, numerical, and physical meaning rather than tracing every data path manually.

A third example concerns infrastructure. Nested include dependencies make dependency discovery and recompilation expensive in the existing CMake workflow. An agent generated and maintains a set of Makefiles that captures these dependencies and makes ordinary incremental builds nearly instantaneous. Porting, refactoring, interface adaptation, documentation, and routine maintenance are similarly suitable for delegation.

The same idea extends beyond programming. I use ChatGPT to revise and develop lecture material for OpenMolcas-related summerschools, including WFS[2] and QCES[3]. It can help restructure explanations, suggest examples, improve slides and exercises, and expose places where an explanation is incomplete. Again, the useful division of labor is not to delegate scientific judgement, but to delegate much of the routine work between an idea and its usable presentation.

The deliberately provocative conclusion is that scientists should stop programming - at least in the traditional sense of writing and debugging every routine themselves. Scientific responsibility cannot be delegated: agent-generated code and teaching material must still be reviewed, tested, and validated. But much of the translation from a well-specified idea to executable code, documentation, or teaching material can be. Our time is better spent on algorithms, observables, approximations, explanations, and new questions.

In keeping with this conclusion, the present abstract was written by an AI agent from the author's unedited notes.

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First baby steps towards a GPU-based CASSCF-CASPT2 (OpenMolcas ?) software

Marco Garavelli[1] (presenting author), Vishal Kumar Jaiswal[1], Piero Altoè[1]

[1] Dipartimento di Chimica Industriale "Toso Montanari", Università di Bologna, Viale del Risorgimento 4, 40136 Bologna, Italy, *email:* marco.garavelli@unibo.it
[2] NVIDIA, Developer Relations - Computational Chemistry, *email: paltoe@nvidia.com*

This contribute will illustrate the first AI-assisted steps in implementing a CASSCF&CASPT2 software running on GPU-NVIDIA architectures: can non-expert developers of electronic structures methods start playing a role? And which role?

GronOR, a massively parallel and GPU-accelerated code for non-orthogonal configuration interaction

Coen de Graaf[1], Tjerk Straatsma[2]

[1] Universitat Rovira i Virgili, Tarragona, Spain, email: *coen.degraaf@urv.cat*
[2] University of Alabama, Tuscaloosa, USA

Electronic coupling is a rather elusive concept from an experimental point of view, but can be studied in great detail with quantum chemical approaches. Theoretical studies often rely on models or TD-DFT approaches, which either depend on input of experimental data or have limited accuracy. Our non-orthogonal configuration interaction with fragments (NOCI-F) implemented in GronOR[1] as open-source code offers a thorough and reliable way to evaluate energy and electron transfer without losing the beauty of the intuitive interpretation of the results inherent to the phenomenological models.

GronOR is interface to OpenMolcas for the calculation of the fragment wave functions and the one- and two-electron integrals. I will overview the design principles and the implementation strategy to make GronOR scale linearly on several large HPC facilities and take full profit of the GPUs on these installations. Furthermore, I will illustrate the versatility of the NOCI-F approach with a case study of the singlet fission process in a self-assembled stack of indolonaphthyridines[2].

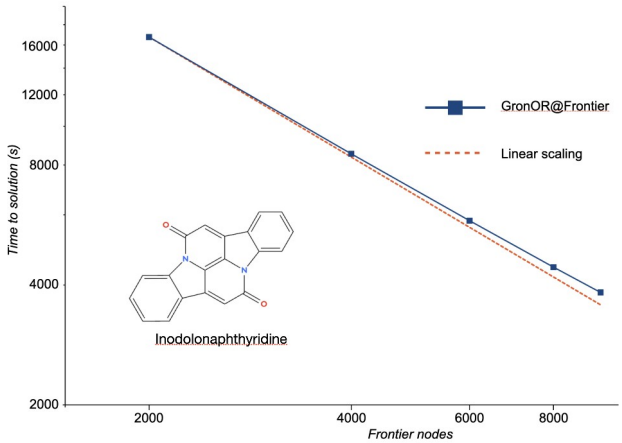

Figure 1. Scaling behaviour of GronOR on Frontier at Oak Ridge National Laboratory for a trimer of indolonaphthyridines with a CAS(8,8) on each monomer.

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