SI2-SSI: Chronus Quantum (ChronusQ) - Sustainable Open-Source Quantum Dynamics and Spectroscopy Software

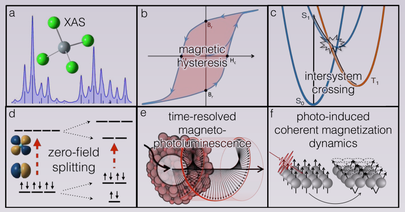
Xiaosong Li, Eugene DePrince III, Anne McCoy, Eitan Geva

Functionality Overview

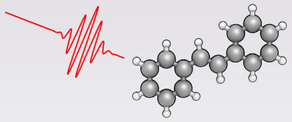
Interaction between matter and electromagnetic radiation can be modeled from first-principles using time-dependent quantum mechanics. Properties can be computed to provide molecular insights to experiment.

Spectroscopic Properties

ChronusQ provides convenient modules to evaluate a number of properties which are important to the field of spectroscopy.



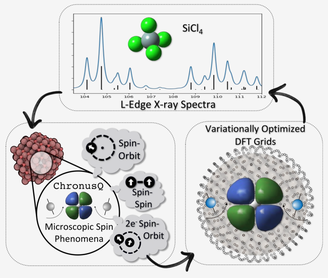
Time-Dependent Electronic Structure



A primary component of ChronusQ is a high-performance and robust integrator for the time-dependent Schrödinger and Dirac equations. Time integration in the presence of an arbitrary number of fields of various character and amplitude envelopes is supported. For a given perturbation, evaluation of a full suite of electronic properties allows for simplified access to spectroscopic properties in the time and frequency domains. Prebuilt modules for the linear absorption and electric circular dichroism are provided.

Relativistic Electronic Structure Theory

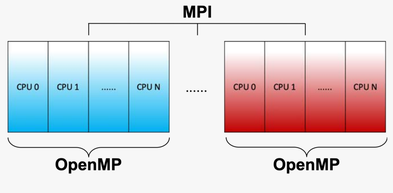
Relativistic electronic structure theory is required to accurately treat heavy-elements and simulating many important phenomena such a spin-orbit coupling, the inert pair-effect, and spin-forbidden reactivity. ChronusQ provides an implementation of the exact two-component (X2C) relativistic method using both Hartree-Fock and Kohn-Sham wave functions.



Technical Highlights

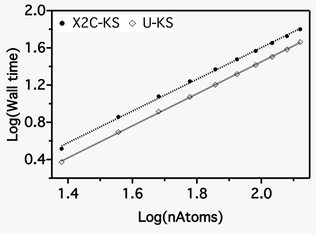
Hybrid OpenMP + MPI Parallelism

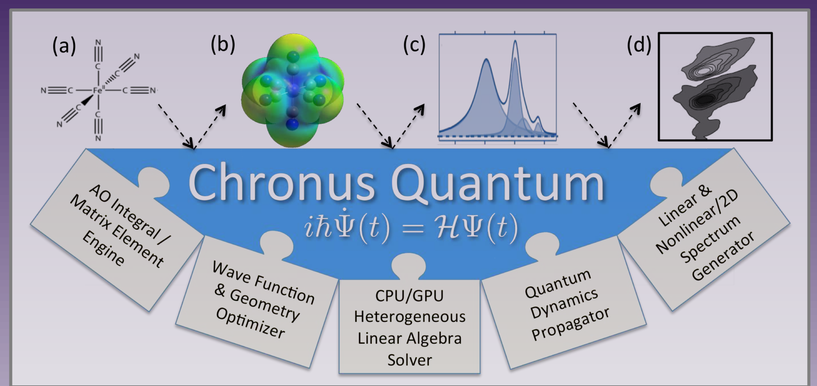
ChronusQ utilizes a hybrid parallelism scheme which exploits shared memory parallelism (OpenMP) on a single compute node and MPI across compute nodes to minimize data communication.



Scalable Relativistic DFT

ChronusQ implements a scalable algorithm for the treatment of non-collinear relativistic density functional theory using torque free auxiliary variables. [1]

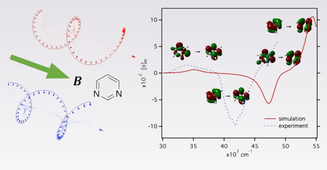




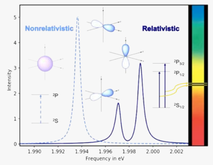
Highlighted Applications

Magnetic Circular Dichroism

Magnetic circular dichroism (MCD) spectra are able to provide unique insights into properties of chemical systems, but their modelling requires accurate and efficient treatment of magnetic fields. ChronusQ provides a variational approach to capturing this computationally challenging spectroscopy. [2]



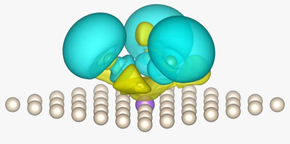
Relativistic RT-EOM-CCSD



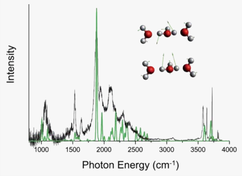
Quantitative agreement with experimental spectra often requires high levels of theory. Combined with a relativistic Hamiltonian, these high levels of theory can be prohibitively expensive, especially for explicitly time dependent methods. ChronusQ circumvents this by using a dipole autocorrelation based formulation of real-time equation-of-motion coupled cluster, resulting in accurate, efficient high-level spectra. [3]

Non-Colinear Quantum Embedding

ChronusQ can efficiently model localized spin phenomena, such as spin transitions and spin-orbit coupling with a novel, two-component embedding methodology. Two-component calculations can be sped up by orders of magnitude while still capturing the essential physics of the system. [4]



Vibrational Perturbation Theory



Second-order Vibrational Perturbation Theory allows for calculation of vibrational energies and intensities beyond the Harmonic approximation. It is able to better account for large amplitude motions and provides insights into the couplings between modes, such as bending overtones and stretching fundamentals.

Obtaining ChronusQ

ChronusQ is open source and released under GPLv3

- http://www.chronusquantum.org
- https://urania.chem.washington.edu/chronusq/chronusq_public

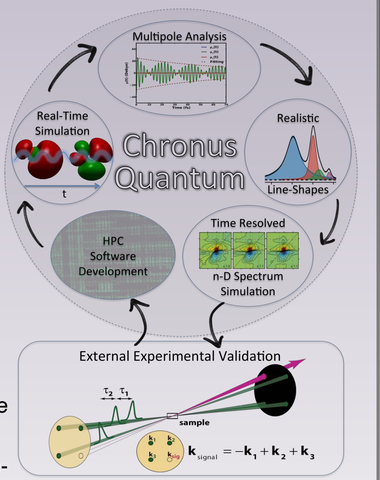
Future Development

Time-Resolved Spectroscopy

- ❑ 2D Infrared Spectroscopy
- ❑ Transient UV/Vis Spectroscopy

Furthering Relativistic Electronic Structure Theory

- ❑ Relativistic Multireference Configuration Interaction Theory
- ❑ Relativistic Unitary Coupled Cluster Theory
- ❑ 4-component wavefunctions and effective QED corrections.
- ❑ Projection-based relativistic in non-relativistic quantum embedding
- ❑ Extension of two-component relativistic methods to include two-electronic spin-couplings



Nuclear Nonadiabatic Dynamics

- ❑ Mixed Quantum-Classical Ehrenfest Dynamics
- ❑ Fully quantum Nuclear Electronic Orbital Dynamics

ChronusQ Citation

Williams-Young, D.B.; Petrone, A.; Sun, S.; Stetina, T. F.; Lestrange, P.; Hoyer, C. E.; Nascimento, D. R.; Koulias, L.; Wildman, A.; Kasper, J.; Goings, J. J.; Ding, F.; DePrince III, A. E.; Valeev, E. F.; Li, X.; *WIREs Comput. Mol. Sci.*, **2019,** e1436**.**

Select ChronusQ Publications

[1] Petrone, A.; Williams-Young, D.B.; Sun, S.; Stetina, T.; Li, X.; *Eur. J. Phys. B.* **2018**, 91, 169.

[2] Sun, S.; Beck, R. A .; Williams-Young, D. B.; Li, X.; *J Chem. Theory Comput.* **2019**, 15(12), 6824-6831

[3] Koulias, L.; Williams-Young, D. B.; Nascimento, D. R.; DePrince III, A. E.; Li, X.; *J. Chem. Theory Comput.*, **2019**, 15(12), 6617–6624.

[4] Hoyer, C.E.; Williams-Young, D.B.; Huang, C.; Li, X.; *J. Chem. Phys.* **2019**, 150, 174114

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