Simulating the generation, evolution and fate of electronic excitations in molecular and nanoscale materials with first principles methods

1. Introduction and overview

2. Highlights of progress and interactions
Simulating electronic excitations in molecular and nanoscale materials with first principles methods.
Our team: Principal investigators

Daniel Haxton (Chemical Sciences, LBNL).

**Martin Head-Gordon** (Chemical Sciences, LBNL).

Anna Krylov (Chemistry, USC).

Xiaoye (Sherry) Li (Computational Res., LBNL).

C. William McCurdy (Chemical Sciences, LBNL).

Esmond Ng (Computational Research, LBNL).

Sam Williams (Computational Res., LBNL).

Chao Yang (Computational Research, LBNL).
Our team: Postdocs and students.

Jiri Brabec (EN).
Evgeny Epifanovsky (AIK, MHG).
Khaled Ibrahim (SW).
Jeremiah Jones (DJH).
Ilya Kaliman (AIK).
Nicholas Mayhall (MHG).
Natalie Orms (AIK).
Francois-Henry Rouet (XL).
Eugene Vecharynski (CY).
Alec White (CWM, MHG).
Dmitry Zuev (AIK).
Outline of highlights.

LibTensor developments
   Anna Krylov, Evgeny Epifanovsky, Ilya Kaliman,
   Khaled Ibrahim, Sam Williams, Francois-Henry Rouet,
   Sherry Li, Eugene Vecharynski, Dmitri Zuev, Natalie Orms,
   Chao Yang, Jiri Brabec, Esmond Ng

   Treating resonances using complex scaling
   Alec White, Martin Head-Gordon, Bill McCurdy

   Active space spin-flip methods for excited states
   Nick Mayhall, Martin Head-Gordon
Efficient tensor contraction algorithms and codes for wave-function methods

Development of reduced scaling implementations of existing coupled cluster methods for electronically excited states (both bound and resonances).

Approach: Explore matrix and tensor decompositions that reduce data size; exploit locality; combine with efficient parallelization.

Benefits:
• Extend applicability of reliable and proven methods as CC/EOM-CC to larger molecules,
• Apply the same underlying ideas and libraries for reduced scaling to other methods, such as those developed by Head-Gordon and McCurdy.
• Better exploit high-performance computing resources.
1. Cholesky decomposition (CD) and RI of ERI is implemented in Q-Chem 4.1 (released).
2. Libtensor (tensor library) is extended to handle CD/RI representations (published & released).
3. RI/CD implementation of CCSD and all EOM-CCSD methods (EE, IP, EA, SF) completed and validated (published). RI/CD combined with FNO.
4. New infrastructure in libtensor (expressions eval-n).
5. Shared-memory parallel performance of libtensor is improved following findings by Khaled Ibrahim/Sam Williams (code released, paper submitted).
6. Extension of Davidson solver for interior eigenvalues is implemented.
7. GPLMR is implemented in Q-Chem using libtensor tools (collaboration with Vecharynski & Yang, paper submitted).
New Implementation of High-Level Correlated Methods Using a General Block Tensor Library for High-Performance Electronic Structure Calculations

Evgeny Epifanovsky,1,2,3 Michael Wormit,1,2 Tomasz Kus,1,2 Arie Landau,1,2
Dmitry Zuev,1,2 Kirill Khistaev,1,2 Prashant Manohar,1,2 Ilya Kaliman,1,2
Andreas Dreuw,1,2 and Anna I. Krylov1,2

THE JOURNAL OF CHEMICAL PHYSICS 139, 134105 (2013)

General implementation of the resolution-of-the-identity and Cholesky representations of electron repulsion integrals within coupled-cluster and equation-of-motion methods: Theory and benchmarks

Evgeny Epifanovsky,1,2,3 Dmitry Zuev,1 Xintian Feng,1 Kirill Khistaev,1 Yihan Shao,3 and Anna I. Krylov1
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New algorithms for iterative matrix-free eigensolvers in quantum chemistry

Dmitry Zuev*, Eugene Vecharynski*, Chao Yang*, Natalie Orms*, Anna Krylov*
* Department of Chemistry, University of Southern California, Los Angeles, California 90089-0482
* Computational Research Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

Analysis and Tuning of Libtensor Framework on Multicore Architectures

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Wednesday, July 30, 2014
Libtensor developments:

2. Extension to massively parallel (>1000 nodes): via fusion with CYCLOPS (Epifanovsky, Solomonik): Pilot code is ready, performance studies in progress.
3. Extension to intermediate regime (up to 100 nodes): Pilot code is ready, performance studies in progress (Kaliman, see poster!). Strategy:
   - Replicated memory;
   - Known to scale well up to 100s of nodes;
   - MPI-1 only (very portable);
   - Simple code and algorithms;
   - Can reuse existing infrastructure.
Improving Locality and Parallelization in Libtensor

- **Existing Challenges:**
  - No inherent management of locality
  - Potential for contention and serialization

- **We developed a new memory management scheme:**
  - Restrict thread migration
  - Explicit memory affinity allocation
  - Parallelize the allocator by creating private heaps
  - Parallelize deallocator using garbage collection
Improving Locality and Parallelization in Libtensor

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  - Explicit memory affinity allocation
  - Parallelize the allocator by creating private heaps
  - Parallelize deallocator using garbage collection
Improvement to Libtensor Scaling

- Previous Scalability:
  - 9.5x speedup using 24 cores
- Scalability after Optimization:
  - 15.3x speedup with 24 cores
  - Parallel Efficiency improved from 39.5% to 64%.
- Ideal scalability is limited by libtensor’s tasking model:
  - Amdhal Effect from master-worker model with barriers
- Further Improvement requires managing task dependencies.
  - Alternatively, concede optimizations for an elegant distributed implementation.
• Analysis and tuning of Libtensor stack of libraries
  – Identified bottlenecks (locality, serialization, load balancing)
  – Implemented a new memory management system for optimal locality management and to avoid serialization.
  – Searched tuning space for optimal configuration.
  – attained a 1.8x improvement on Edison, more than 2x on large AMD SMPs.

• Submitted Publication (under review)
Benchmarking CD-CCSD
Example: (mU)$_2$-H$_2$O: 158 electrons; C$_1$ symmetry
6-31+G(d,p) basis: 489 b.f.
cc-pVTZ basis: 882 b.f.
Calculations on a single Xeon-Dell node (16 cores; node28)

<table>
<thead>
<tr>
<th>Method/Basis</th>
<th>Memory</th>
<th>CCSD wall (one iter-n), hours</th>
</tr>
</thead>
<tbody>
<tr>
<td>CD2/6-31+G(d,p)</td>
<td>100 GB</td>
<td>5.1</td>
</tr>
<tr>
<td>CD2/FNO/6-31+G(d,p)</td>
<td>100 GB</td>
<td>1.4</td>
</tr>
<tr>
<td>CD2/FNO/cc-pVTZ</td>
<td>300 GB</td>
<td>12.2</td>
</tr>
</tbody>
</table>

Parallel scaling on node 31
“New algorithms for iterative matrix-free eigensolvers in quantum chemistry”, by D. Zuev, E. Vecharynski, C. Yang, N. Orms, A. Krylov

- Different algorithms (modified Davidson procedure and GPLMR by Vecharynski) for interior eigen-states are developed and implemented in production-level code in EOM-CC (released in QChem 4.2);
- Robust performance for high-lying eigenstates (such as highly excited states, core ionized states, etc);
- Significant computational savings for valence states.

GLMR converging G2 roots at specified energy shift

<table>
<thead>
<tr>
<th>Shift, a.u.</th>
<th>N iter</th>
<th>Stored vecs</th>
<th># of matvec</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.6</td>
<td>4</td>
<td>16</td>
<td>18</td>
</tr>
<tr>
<td>9.6</td>
<td>4</td>
<td>16</td>
<td>18</td>
</tr>
<tr>
<td>10.1</td>
<td>4</td>
<td>16</td>
<td>18</td>
</tr>
<tr>
<td>11.1</td>
<td>4</td>
<td>16</td>
<td>18</td>
</tr>
</tbody>
</table>

Davidson converging 1 EOM-IP root of a given character

<table>
<thead>
<tr>
<th>MO</th>
<th>N iter</th>
<th>Max vec</th>
<th># of matvec</th>
<th>Cost relative to canonical algorithm</th>
</tr>
</thead>
<tbody>
<tr>
<td>46 (2nd IP)</td>
<td>10</td>
<td>22</td>
<td>11</td>
<td>78%</td>
</tr>
<tr>
<td>45 (3rd IP)</td>
<td>11</td>
<td>24</td>
<td>12</td>
<td>50%</td>
</tr>
</tbody>
</table>
• RI gradient is nearly completed (coll-n w/Head-Gordon).
• CD gradient is in progress.
• Extension of libtensor to distributed parallel architecture via fusion with CYCLOPS is nearly completed, benchmarks are (collaboration with Solomonik).
• Performance benchmarking and improvements (exploiting data locality) are ongoing (coll-n with Ibrahim and Williams).
• Sparsity/rank reduction explorations are ongoing.
LibTensor developments
   Anna Krylov, Evgeny Epifanovsky, Khaled Ibrahim,
   Sam Williams, Francois-Henry Rouet, Sherry Li,
   Eugene Vecharynski, Chao Yang,
   Jiri Brabec, Esmond Ng

Treating resonances using complex scaling
   Alec White, Martin Head-Gordon, Bill McCurdy

Active space spin-flip methods for excited states
   Nick Mayhall, Martin Head-Gordon
The problem: Compute the energies and lifetimes of metastable states of molecules as a function of molecular geometry – complex potential energy surfaces

\[ E(\vec{R}) = E_r(\vec{R}) - i \Gamma(\vec{R}) \]
\[ \text{lifetime} = \hbar/\Gamma(\vec{R}) \]

Metastable states have complex energies: solutions of the Schrödinger equation with pure outgoing wave scattering boundary conditions

State of the art: No general method exists for applying the tools of modern quantum chemistry to metastable states of polyatomic molecules
Why is this an important problem?

- Electrons attach to molecules via metastable resonance states to initiate “Electron-Driven Chemistry”
- Significant for radiation damage
Formal solution is complex scaling of electronic coordinates

- Under complex scaling, wave functions of metastable states are square integrable.
- Hence can be treated with quantum chemistry methods

\[ \theta \]

\[ \text{Re}(r) \]

\[ \text{Im}(r) \]

Complex r-Plane

Complex Energy-Plane

Bound states

Resonances

Continua

But both formal and practical problems prevent application of this idea to polyatomic molecules
Gaussian basis functions with complex exponents accomplish complex scaling asymptotically

**Approach:** Implement complex exponents for Gaussian basis functions in the Q-Chem suite of codes to accomplish the effects of *Exterior Complex Scaling*

\[ \phi(\mathbf{r}) = N (x - A_x) \left( y - A_y \right)^m (z - A_z)^n \exp \left[ -\alpha e^{-2i\eta} (\mathbf{r} - \mathbf{A})^2 \right] \]

**Benefit:** Renders complex-energy resonance wave functions square-integrable. Allows treatment of polyatomic molecules by SCF, CCSD, CI, and other quantum chemistry methods

\[ \langle ij | kl \rangle = \int d\mathbf{r}_1 \int d\mathbf{r}_2 \phi_i(\mathbf{r}_1)\phi_j(\mathbf{r}_2) r_{12}^{-1} \phi_k(\mathbf{r}_1) \phi_l(\mathbf{r}_2) \]

**Central Challenge:** *Accomplished this year!* Alec White adapted Q-Chem to calculate two-electron integrals over Gaussian basis with mixed real and complex exponents
1\textsuperscript{st} Results: Resonances via static exchange (Alec White)

- Static Exchange Approximation: Finding the complex eigenvalues of an effective one-electron Hamiltonian

$$\hat{H} = \hat{T} + \hat{V}_{nuc} + 2\hat{J} - \hat{K}$$

Procedure:

1. Determine electron density of the closed shell target in a real basis
2. Diagonalize $H$ in a basis including complex Gaussian basis functions
3. Repeat for many values of $\eta$ and find stationary value
Designing Complex Gaussian Basis Sets: add complex functions to correlation consistent sets (Alec White)

- cc-pvXZ basis sets used for the real calculation.
  - Augmented with even-tempered set of 7 complex diffuse functions
  - For main second row elements, we used spd diffuse functions for dz bases, spdf for tz, and so on.
  - The convergence properties of these basis sets appear to be adequate for small molecules. Results below in eV.

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>caug-ccpvDZ</td>
<td>3.98980</td>
<td>-0.65830</td>
<td>2.64813</td>
<td>-0.75697</td>
<td>5.80248</td>
<td>-0.00071</td>
</tr>
<tr>
<td>caug-ccpvTZ</td>
<td>3.87613</td>
<td>-0.61104</td>
<td>2.64539</td>
<td>-0.80621</td>
<td>5.53680</td>
<td>-0.37414</td>
</tr>
<tr>
<td>caug-ccpvQZ</td>
<td>3.85113</td>
<td>-0.61782</td>
<td>2.60813</td>
<td>-0.79131</td>
<td>5.47779</td>
<td>-0.34736</td>
</tr>
</tbody>
</table>
Application to anions of nitrogen-containing aromatics: Only Metastable or Some Bound?

- Nenner and Schulz (1975) reported that pyrazine, pyrimidine, and s-triazine have bound anions, but more recent studies have suggested otherwise.

- CCSD(T) calculations of adiabatic and vertical electron affinities show no bound anions in any of these molecules.
Conventional quantum chemistry vs Complex basis function calculations (Alec White)

- CCSD(T) calculations show no bound anion states mol)

<table>
<thead>
<tr>
<th></th>
<th>Benzene</th>
<th>Pyridine</th>
<th>Pyrimidine</th>
<th>Pyrazine</th>
<th>s-Triazine</th>
</tr>
</thead>
<tbody>
<tr>
<td>vertical EA, kcal/mol</td>
<td>23.959</td>
<td>15.602</td>
<td>9.194</td>
<td>10.804</td>
<td></td>
</tr>
<tr>
<td>adiabatic</td>
<td>18.167</td>
<td>19.016</td>
<td>9.646</td>
<td>3.269</td>
<td>4.647</td>
</tr>
</tbody>
</table>

- Static Exchange calculations give results for lifetimes

<table>
<thead>
<tr>
<th></th>
<th>Benzene</th>
<th>Pyridine</th>
<th>Pyrimidine</th>
<th>Pyrazine</th>
<th>s-Triazine</th>
</tr>
</thead>
<tbody>
<tr>
<td>Re[E]</td>
<td>3.1288</td>
<td>2.6459</td>
<td>2.3480</td>
<td>2.0809</td>
<td>2.2580</td>
</tr>
<tr>
<td>Im[E]</td>
<td>-0.2646</td>
<td>-0.1897</td>
<td>-0.1303</td>
<td>-0.1590</td>
<td>-0.0803</td>
</tr>
<tr>
<td></td>
<td>3.0219</td>
<td>2.7481</td>
<td>2.8258</td>
<td>8.8740</td>
<td>8.6586</td>
</tr>
<tr>
<td></td>
<td>-0.2291</td>
<td>-0.1706</td>
<td>-0.1660</td>
<td>8.8740</td>
<td>8.6586</td>
</tr>
<tr>
<td></td>
<td>9.4297</td>
<td>9.1366</td>
<td>8.8765</td>
<td>8.8740</td>
<td>8.6586</td>
</tr>
<tr>
<td></td>
<td>-1.2191</td>
<td>-1.1151</td>
<td>-0.9884</td>
<td>-1.2838</td>
<td>-0.9917</td>
</tr>
</tbody>
</table>

- Differences between molecules agree with experiment to within 0.2 eV. See Alec’s poster for more details!
Current Development: Complex Self Consistent Field Calculations on Metastable Anion States

- Trial function is a determinant of spin orbitals
  \[ \Phi = |\varphi_1 \bar{\varphi}_2 \cdots \varphi_n \bar{\varphi}_n \varphi_s| \]
  \[ \frac{\delta}{\delta \varphi_i} \frac{\langle \Phi | H | \Phi \rangle}{\langle \Phi | \Phi \rangle} = 0 \]

- Working SCF equations from complex variational (stationary) principle

- Complex Restricted Open-Shell Hartree-Fock (ROHF) demonstrated by McCurdy et al. in 1980s

- Q-Chem implementation of Complex Unrestricted Hartree-Fock (UHF) with different orbitals for different spins will dissociate polyatomic metastable anions correctly
Outline of highlights.

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Active space spin-flip methods for excited states
  Nick Mayhall, Martin Head-Gordon
Spin-flip method
How it works for breaking a single bond
Spin-flip method
How it works for breaking a single bond

1. RHF configuration is poor at stretched bond-lengths
Spin-flip method
How it works for breaking a single bond

1. RHF configuration is poor at stretched bond-lengths

2. Triplet is a spin-pure single configuration at all bond-lengths
Spin-flip method
How it works for breaking a single bond

1. RHF configuration is poor at stretched bond-lengths

2. Triplet is a spin-pure single configuration at all bond-lengths

3. 4 configurations describe H$_2$ at all bond-lengths: 1 spin flip from $M_s=1$ triplet
Spin-flipping

- Find the highest multiplicity low-energy configuration...
- ... which becomes the single reference...
- ... whose half-occupied orbitals define the active space

- e.g. \((\text{H}_3\text{N})_5\text{Cr}^{\text{III}}\text{OCr}^{\text{III}}(\text{NH}_3)_5^{4+}\)
  - \(\text{Cr(III)} \Rightarrow d^3\)
  - \(\Rightarrow 6\) unpaired e’s
  - \(\Rightarrow\) heptet single reference

- How do we do the spin-flip electronic structure?
  1. By excitation level (e.g. SF-CCSD etc). Spin impure.
  2. As a complete active space (CAS). Spin pure.
SF-CAS methods

• Simplest: full CI in SF active space: SF-CAS
  • Poor performance: orbitals for high spin state
  • Orbital optimization (SF-CASSCF) would help
  • ... but is state-specific.... so we look elsewhere

• Add extra configurations: variationally or perturbatively
  • 1X: variationally
    • RAS-SF
    • SF-CAS(1x)
  • 2X: perturbatively
    • SF-CAS(2x)

• 1X: var -- P. Zimmerman, D. Casanova, F. Bell, MHG,
  • JCP 137, 164110 (2012); PCCP 15, 358 (2013)
• 1X: PT -- N. Mayhall, M. Goldey, MHG, JCTC 10, 589 (2014).
• S: PT -- N. Mayhall, MHG, JCP 141, 044112 (2014).
• Quintet reference, cc-pVTZ
• SF-CAS does not even match Hartree-Fock.
• Including 1x (hole, particle) surpasses HF
• Including all singles (partial 2x) recovers significant dynamic correlation
Efficiency of perturbative approach (Nick Mayhall).

Timings via perturbation theory vs variational approach

SF-CAS(h,p)₁ solves a dressed non-Hermitian eigenvalue problem in the SF-CAS space.

For small numbers of spin flips, this can be done by explicit diagonalization giving large speedups for large molecules.

Example of 6 states (S, T, Q) of a polyradical porphyrin verdazyl derivative.

<table>
<thead>
<tr>
<th></th>
<th>RAS-SF</th>
<th>SF-CAS(h,p)₁</th>
</tr>
</thead>
<tbody>
<tr>
<td>time (s)</td>
<td>7479.392</td>
<td>61.028</td>
</tr>
<tr>
<td>relative</td>
<td>100.00%</td>
<td>0.82%</td>
</tr>
</tbody>
</table>
SF-NOCI: non-orthogonal configuration interaction
(Nick Mayhall, Eric Sundstrom, Paul Horn)

• Starting from SF-CAS
  1. Localize active-space orbitals & freeze them
  2. Relax each SF-CAS configuration individually
  3. Diagonalize H in this new “relaxed” determinant basis

• A very special form of non-orthogonal CI !!
  • No 2 configurations can coalesce
  • Each relaxation is well-conditioned (large gap)
  • Building the NOCI Hamiltonian is highly efficient
  • Rate-determining relaxations are embarrassingly parallel

• N. Mayhall, P. Horn, E. Sundstrom, MHG, PCCP (submitted)
• For NOCI, see: E. Sundstrom, MHG, JCP 140, 114103 (2014).
Ionic / covalent competition in LiF (Nick Mayhall)

- SF-CAS uses triplet orbitals -- neglects ionic relaxation
- SF-NOCI strongly relaxes the ionic configuration
  - gives a qualitatively correct curve: \( \Delta H_f(\text{expt}) = 82 \text{ kcal/mol} \)
Low-lying states in Cu$^{II}$(Fsbqdi)$_2$ (eV) (Nick Mayhall)

- 3 unpaired e’s in 3 orbitals
  - Use quartet ROHF single reference
  - 2 low-lying doublets, 1 quartet
  - Higher ionic CT configurations....

<table>
<thead>
<tr>
<th>type</th>
<th>config</th>
<th>multiplicity</th>
<th>SF-CAS (eV)</th>
<th>SF-NOCI (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>local</td>
<td>$\alpha\beta\alpha$</td>
<td>doublet</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>local</td>
<td>$\alpha\alpha\beta + \beta\alpha\alpha$</td>
<td>doublet</td>
<td>0.05</td>
<td>0.12</td>
</tr>
<tr>
<td>local</td>
<td>$\alpha\alpha\alpha$</td>
<td>quartet</td>
<td>0.07</td>
<td>0.18</td>
</tr>
<tr>
<td>LLCT</td>
<td>$+\alpha-$</td>
<td>doublet</td>
<td>4.6</td>
<td>2.6</td>
</tr>
<tr>
<td>LMCT</td>
<td>$\alpha-+$</td>
<td>doublet</td>
<td>11.4</td>
<td>5.0</td>
</tr>
<tr>
<td>MLCT</td>
<td>$\alpha+-$</td>
<td>doublet</td>
<td>13.8</td>
<td>7.8</td>
</tr>
</tbody>
</table>
Magnetic couplings in Cu$^{II}$($^5$Fsbdqi)$_2$ (Nick Mayhall)

- Even for the low-lying states, SF-NOCI makes a big difference in the magnetic couplings...
  - Expt: $J(\text{ML}) \gg 200 \text{ cm}^{-1}$
  - $J(\text{LL}) \ll J(\text{ML})$

<table>
<thead>
<tr>
<th></th>
<th>SF-CAS</th>
<th>SF-NOCI</th>
</tr>
</thead>
<tbody>
<tr>
<td>$J(\text{CuL})$ / cm$^{-1}$</td>
<td>-231</td>
<td>-548</td>
</tr>
<tr>
<td>$J(\text{LCu})$ / cm$^{-1}$</td>
<td>-165</td>
<td>-434</td>
</tr>
<tr>
<td>$J(\text{LL})$ / cm$^{-1}$</td>
<td>-9</td>
<td>-36</td>
</tr>
</tbody>
</table>

Wednesday, July 30, 2014
On-going work and future plans

• We are working hard to add dynamic correlation to the new SF-NOCI approach:
  • Density functional theory and perturbation theory are both under investigation

• See Nick Mayhall’s poster for exciting progress on how to turn a class of strong correlation problems that involve $n$ spin flips into 1-spin flip problems!

• Gradients for the spin flip CAS methods are underway

• Use of large-scale eigensolvers (Yang, Li) will be needed for large numbers of spin flips.
Summary and conclusions
3 selected highlights

• Developments in the LibTensor framework
  • Enables advanced electronic structure methods
  • Work cuts across physical sciences, math, comp. sci.
  • Major progress in scaling, solvers, new capabilities

• Advances in the treatment of molecular resonances
  • Completed complex 2-electron integral code
  • Preliminary results on basis set convergence
  • First pilot applications to medium-sized aromatic molecules

• New active space spin-flip methods for excited states
  • Development of highly efficient perturbation theory
  • New non-orthogonal framework is highly promising