Tensor Methods in Quantum Chemistry

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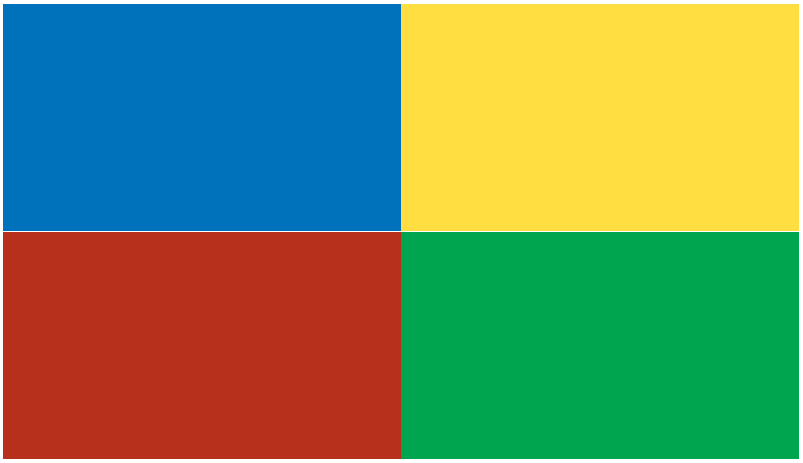

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Part I.

Context

Chapter 1.

Scope

The broader context of this working group is to gain a broad understanding of the use (and abuse) of tensor methods in quantum chemistry. There is a lack of coherent material accessible to members of the community. In particular, though there has been a proliferation of user-friendly black box methods, there are few, if any strategies to bring researchers to a point where they are able to meaningfully contribute in a sustainable way to a software ecosystem.

Computational simulations have gained prominence as a third pillar of scientific exploration, supplementing theory and experiment [1, 52]. For any given problem, the interplay between forming a theoretical model consistent with experimental evidence is now augmented by numerical methods. Numerical methods have risen to greater prominence due to their ability to provide efficient models from large datasets (big-data and machine learning models) however, in the physical sciences, they are of greater importance than facilitating glorified fitting procedures. In quantum chemistry, computational methods have been able to provide quantitative values (occasionally) at par with experimental methods, heralding the "third age of quantum chemistry" [43, 46, 47].

An often overlooked aspect of the modeling process, involves the numerical approximations and software implementation approaches to computational quantum chemistry. Unlike theoretical results, or even experiments, more often than not computational studies are not held to the same level of rigorous reproducibility. Computational studies are often nigh impossible to replicate due to the large amount of computational resources and time required. This, coupled with the proliferation of commercial or simply closed source codes are key drivers [40]. The reproducibility crisis in computational quantum chemistry is slowly being acknowledged [29, 42] and can only be exacerbated by the unfocused implementation of further numerical methods.

Concurrently, the advent of computational methods has highlighted the lack of communication between various scientific disciplines. Code in this sense functions as an equalizer, allowing for a unified understanding of the nature in which algorithms used in widely differing contexts can be in fact, similar in implementation. As a relevant example, alternating least squares, used for recommendation systems in the data-science literature shares much of its core (numerical) concepts with the density matrix renormalization group method used for obtaining ground state energies and observables from quantum systems.

1.1. Second Quantization

Most of the quantum chemistry literature [6, 21, 54] ([16] is an exception) is formulated in terms of wavefunctions, and with little reference to the discretized second quantization formulation. Tensor networks and their studies pertaining to the physical sciences are typically described in terms of the second quantization formalism. This has bleed into the code structures as well, which will have consequences discussed in a later section.

1. Density Matrix Renormalization Group The most well known applications of tensor methods in the context of quantum chemistry have traditionally been in the context of replacing full CI calculations with DMRG [4].
   This is a variant of Wilson's NRG method with a non-energy optimal approach to truncating the resultant space.

1.2. Goals

Primarily, the working group focused on the following:

- Reconciliation of concepts between disciplines
  - Cross pollination of gainful concepts
- Understanding code structures and implementation methods
  - Scalability, interoperability with mathematical models, and documentation
  - Performance bottlenecks
  - Programming language considerations
- Identification of future directions and current challenges
- and the unique (virtual) circumstances under which the IPAM program involved

workflows towards the mitigation of the same.
It was hoped that a standard library could begin to be established, similar to the HPX committee [22].

Chapter 2.

Quantum Chemistry

Quantum chemistry as will be used in this report can be understood as the formulation of electronic structure problems for systems corresponding to real materials. To keep the report to a manageable length we will neglect any discussion of density functional theory in this section. The starting point for such a discussion can be completely pragmatic, taking a classical Hamiltonian and "quantizing" it with the transform of the momentum. A slightly more rational explanation (following [55]) begins with writing the molecular non-relativistic Schrödinger equation.

Equation 1 — Molecular Non-relativistic Schrödinger.

$$\mathbf{H}\Psi = E\Psi \tag{2.1}$$

With the normal notation of **H** being the Hamiltonian operatior, Ψ being the total wave-function, and E as the energy eigenvalue. For most calculations of interest in quantum chemistry we denote the time independent Hamiltonian operator in atomic units by:

Equation 2 — Non-relativistic time independent Hamiltonian.

$$\mathbf{H} = -\frac{1}{2}\sum_{i}^{n}\nabla_i^2 - \frac{1}{2}\sum_{A}^{N}\frac{1}{M_A}\nabla_A^2 - \sum_{i}^{n}\sum_{A}^{N}\frac{Z_A}{r_{iA}} + \sum_{i>j}^{n}\frac{1}{r_{ij}} + \sum_{A>B}^{N}\frac{Z_A Z_B}{R_{AB}} \tag{2.2}$$

which is written out for n electrons and N nuclei with ∇^2 representing the Laplacian operator. Electrons have indices i and j while each nucleus is denoted by A or B with the second index deployed to prevent double counting an interaction. M_A is the mass ratio of the nucleus to the electron, Z_A is the atomic number of nucleus A and r represents the distance, where the lower indices clarify the objects between which the distance is being measured.

This is often understood as being the sum of terms, namely (in order) the kinetic energy of the electrons, the kinetic energy of the nuclei and the potential energy contributions of nuclear-electronic, electronic, and nuclear interactions.

While details may be found in the standard literature [6, 21, 32, 54], typical approximations involve being able to consider the nuclei to be "clamped" (the Born-Oppenheimer

assumption) and the adiabatic assumption (electronic motions follow the nuclei – timescale separation). This allows us separate the nuclear (constant at a fixed geometry) and the electronic parts of the Hamiltonian as [47]:

Equation 3 — Total energy after BO and adiabatic approximations.

$$\begin{aligned} \mathbf{H}_{elec} &= \sum_{i}^{n} \mathbf{h}(i) + \sum_{i>j}^{n} \frac{1}{r_{ij}} \\ H_{elec}\Psi_{elec} &= E_{elec}\Psi_{elec} \\ E_{total} &= E_{elec} + \sum_{A>B}^{N} \frac{Z_A Z_B}{R_{AB}} \end{aligned} \tag{2.3}$$

Where we have used the one electron operator $\mathbf{h}(i)$:

Equation 4 — One electron operator.

$$\mathbf{h}(i) = \frac{1}{2}\nabla_i^2 - \sum_{A}^{N} \frac{Z_A}{r_{iA}} \tag{2.4}$$

The second term in $\mathbf{H}_{elec}$ is often written (under the approximations considered) in terms of the coulomb (J) and exchange (K) integrals as $\sum_i^N \sum_{j>i}^N (J_{ij} - K_{ij})$ [21]. Relativistic effects and the Dirac equation are not considered here, though they are known to account for anomalous observations in systems with heavy elements (e.g. group 12 dimethyl metal-carbon bond lengths, p block ionization energy trends etc.) [9].

The essential feature of quantum chemistry is the potential energy surface, a hypersurface which is defined by the potential energy of atoms over all possible arrangements [6]. Describing this surface efficiently (a sampling problem) and subsequently forming (minimum energy) paths (a global optimization problem) are the bedrock of a large segment of quantum chemistry applications.

Methods of solving the equations described can be divided into the standard two forms of treating partial differential equations numerically, that is by considering a real space grid and mesh-less methods. In quantum chemistry, the mesh-less methods are more common compared to the gird methods, however, for tensor applications Khoromskaia and Khoromskij [24] demonstrate the superiority of using grids.

2.1. Linear Combination of Atomic Orbitals (LCAO)

For an understanding of the mesh-free methods the LCAO framework (Dewar and Kelemen [7] demonstrates this pedagogically for H_2). The LCAO asserts that a molecular wave function can be written in terms of a linear combination of a atomic orbitals, and each electron in the system can be described by a single one electron wave function (the orbital approximation). This neglects both static and dynamic correlations. The spin orbitals (one-electron wave functions) are composed of three continuous variables for

space, and one spin parameter which is discrete and can take only two values (often written as α and β). However, the consideration of spin orbitals and their product (known as the Hartree Product [54]) does not account for Pauli's principle, that is, there are no restrictions on regarding the manner in which the spin orbitals combine to form a molecular orbital and thus it fails for any system of more than one electron. With the neglect of correlations, we are able to separate the spin orbitals into spatial and spin segments. For the Hydrogen molecule, we have two electrons and two protons which implies twelve coordinates for the four particles, notwithstanding spin. In this situation, we can write the spin orbital as $\psi_i\sigma_s$ where ψ is the spatial portion of the orbital x, y, z and σ is the spin portion. A trial form of the wave-function which satisfies the Pauli principle can be constructed by expressing the molecular wavefunction as a normalized determinant composed of spin orbitals, and this is the Slater determinant. In a multi-particle system it is written as:

Equation 5 — Multi-particle Slater determinant.

$$\begin{aligned}\Psi(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N) &= \frac{1}{\sqrt{N!}} \begin{vmatrix} \psi_1(\mathbf{x}_1) & \psi_2(\mathbf{x}_1) & \cdots & \psi_N(\mathbf{x}_1) \\ \psi_1(\mathbf{x}_2) & \psi_2(\mathbf{x}_2) & \cdots & \psi_N(\mathbf{x}_2) \\ \vdots & \vdots & \ddots & \vdots \\ \psi_1(\mathbf{x}_N) & \psi_2(\mathbf{x}_N) & \cdots & \psi_N(\mathbf{x}_N) \end{vmatrix} \\ &\equiv |\psi_1, \psi_2, \cdots, \psi_N\rangle \\ &\equiv |1, 2, \dots, N\rangle \end{aligned} \tag{2.5}$$

Although this form of the wavefunction is often taken to be the starting point from which further corrections are derived, it should be emphasized that this is still an ansatz, and other valid guesses for the wavefunction are equally valid.
The final approximation before obtaining the molecular wavefunction is to expand the spatial orbitals as linear combinations of basis set functions which in turn are often considered to be the self consistent field atomic orbitals of individual atoms.

2.2. Configuration Interaction

We can obtain the effects of electron correlation in some sense by requiring a solution in the form of a combination of slater determinants (distributing electrons across various molecular orbitals) instead of a single slater determinant. This is the underlying concept behind the configuration interaction methods and these are based on the variational principle; aside from which they are formulated in a manner analogous to the Hartree-Fock equations [21]. We can write the wavefunction as a linear combination of determinants, and the molecular orbitals used for the exited Slater determinants are from a Hartree-Fock calculation. These orbitals are then fixed, which makes this a constrained optimization problem. Formally, we have [21]:

Equation 6 — Configuration interaction wavefunction.

$$\Psi_{CI} = a_0\Phi_{HF} + \sum_S a_S\Phi_S + \sum_D a_D\Phi_D + \sum_T a_T\Phi_T + \cdots = \sum_{i=0} a_i\Phi_i \tag{2.6}$$

Where we have denoted the relative excitation states w.r.t to HF configuration with S, D, T for singly excited, doubly excited, and triply excited.
Perhaps more pertinently, though truncated CI methods are not in general size extensive, the Quadratic CISD (QCISD) class of methods have enough high order terms to ensure the results appear to be size extensive.
With that in mind, and given that the full CI methods can be thought to be limited only by the basis set (as the formulation is rooted in the variational principle), we **do not** expect that going to the full CI limit will change the results substantially for smaller. However, since the full CI limit at a large enough basis set might account for even more of the electron correlation, we might expect the results to be even closer to experimentally observed values, barring relativistic effects (see Fig. 2.1).

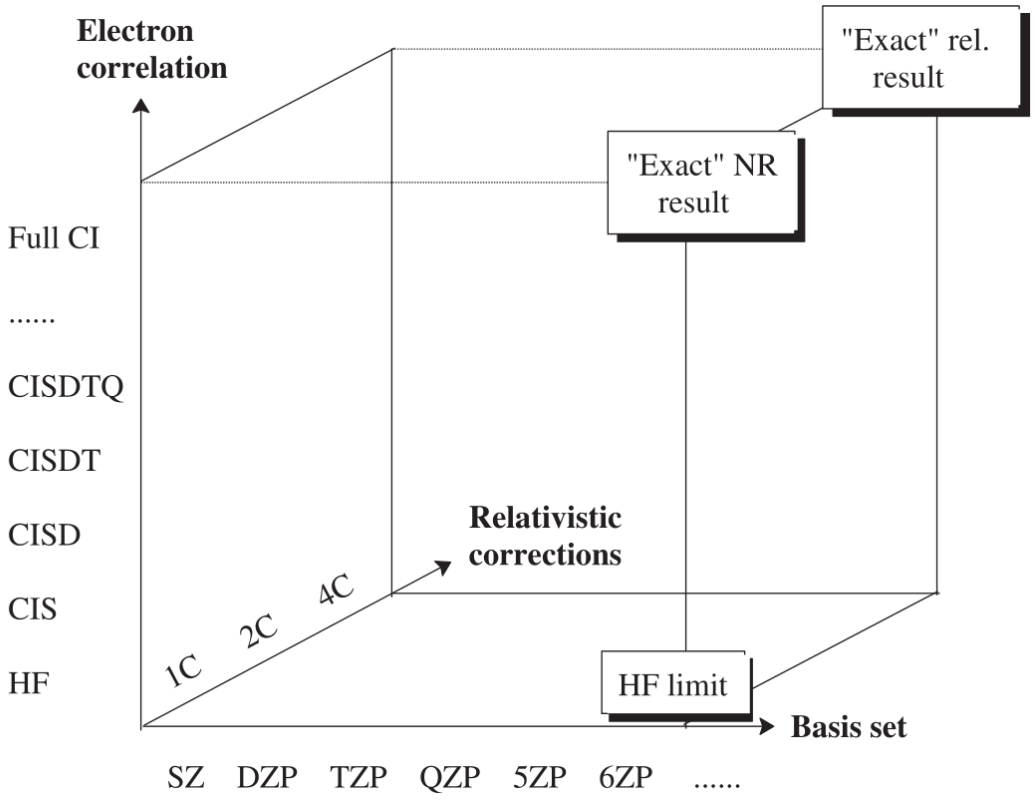

Figure 2.1.: Conceptual mapping of the full CI method in the QC context from Jensen [21]

Similar to the variational approach, other approaches to systematically include corrections to the basic wavefunction exist in the literature (coupled cluster methods [32, 54], cluster perturbation series [3]), and readers are referred to the literature [21] for details and strict equivalence relations with CI methods.

Part II.

Implementations

Chapter 3.

Languages and DSLs

The idea of forming programming languages which maximally combine expert knowledge with performance oriented low level code is the basis of rationalizing the existence of Domain Specific Languages (DSLs). It is not really that these ideas are new [50], but they suffer from bitrot over time. BLOCK is now meant to be used as a black box, and the source is not distributed on Github; similarly, the Quantum Chemistry version of the cyclops tensor framework (CTF) [51], Aquarius [51] has also suffered from a lack of development. CTF [31] is reasonably well developed, but it has fallen behind in terms of speed compared to the later stage libraries opt-einsum which can take advantage of dispatching efficiently to specialized hardware.

3.1. Quantum Chemistry Software

Quantum chemistry has suffered for many years as a retarded child of computer science in terms of the communal adoption of closed source tools which can be traced back to the original sin of Gaussian becoming closed source [13]. Lately, there has been a slight re calibration of the community's moral compass in a positive sense, and several previously closed source packages have become open-source (e.g. the GAP potentials). The veritable zoo of software [48] developed cannot be enumerated in any reasonable treatise and will be left to the discretion of the reader.
We note in passing that the difficulty in obtaining meaningful information from the outputs of a standard quantum chemistry code has given the community many new avenues of exploration including the entire field of computational material sciences. For issues concerning insight the community tools focus on visualization (OVITO [53], VMD) or structural analysis (d-SEAMS [14]) for molecular dynamics , and workflow flexibility for quantum chemistry. Flexibility and reproducibility is ensured by meta-analysis methods such as those implemented by AiiDA [18, 41] or PyIron [20], or the set of tools which build off of ASE (atomic simulation environment) [28] like GPAW [8, 10].
The crux of these tools is to maximally enable user-interactivity. However, these tools do not typically work in an interchangeable manner at the level of theory. Most quantum chemistry codes work in a pidgin syntax which can only be understood in the context of their own manuals.

Chapter 4.

High Performance Computing Considerations

Tensor methods are popular in quantum chemistry as we have aforementioned, where one of the most important computational kernels is tensor contraction. Tensor contraction, a high-order product between two tensors along with their common dimensions, which is the foundation for a spectrum of applications, such as quantum chemistry, quantum physics and deep learning [2, 12, 27, 33, 44, 45]. Usually, a sequence (could be hundreds) of tensor contractions are used in these applications. Tensor contraction, a.k.a. tensor-times-tensor or mode-$(\{n\},\{m\})$ product [5], is an extension of matrix multiplication, denoted by

Equation 7 — Tensor product.

$$\mathcal{Z} = \mathcal{X} \times_{\{n\}}^{\{m\}} \mathcal{Y}, \tag{4.1}$$

where $\{n\}$ and $\{m\}$ are tensor modes to do contraction.
Multiple challenges exist to enhance the performance of tensor contractions even on the most powerful supercomputers.

4.1. Challenges

1. **High computing power needed to compute these contractions**
   A tensor contraction could easily goes to very high computation complexity. Given two N^{th} -order cubic tensors to be contracted on M dimensions with dimension size I needs $\mathcal{O}(I^{2N-M})$ floating-point operations (Flops). When N is in a high order, with even I is mild, then the floating-point operations could be reach to a very large number. For example, 10^18 Flops for two fifth-order tensors contracted on 1 dimension, with all dimension sizes as 100. This high computation is challenging to solve on a single computer. Besides, it is only one tensor contraction, where in quantum chemistry, we could easily use tens or hundreds of contractions together. The traditional solution is deploying the problem on a supercomputer or cluster to parallelize the computation.
2. **Huge space is required to run these contractions in memory**
   Beyond the high computation power, tensor contractions are also expensive in memory usage. The storage usage of two input tensors plus the output tensors all grows

exponentially with tensor order N. Say an NA^{th} tensor $\mathcal{X} \in \mathbb{R}^{I \times I \cdots \times I}$, then the storage needed is $\mathcal{O}(I^N)$. Though the storage value is smaller than the number of Flops, it still consumes non-trivial space. For the same fifth-order tensor with all dimension sizes as 100, its storage size is $\mathcal{O}(10^{10})$, easily larger than the memory size of a single computer. A traditional solution is distributed computing using supercomputers or clusters. While recent research develops techniques [34, 35] to leverage new memory technologies, such as the new Intel Optane DC persistent memory [19].

3. **Parallelized on distributed memory systems meet the scalability issue**
However, the traditional distributed computing solution for the high computation and memory demands does not work ideally. Deploying an algorithm on distributed computers generally need communications among computing nodes to update shared data, synchronize the execution, etc. Thus, the communication overhead could be a big portion of the total parallel execution time especially on a large supercomputers with hundreds or thousands of computing nodes. There is a tradeoff between how much memory could be locally used on each single computing node and how many nodes to deploy/parallelize the contraction algorithm. Some recent research put their effort on using GPUs [25, 26].
4. **Sparsity, adopted in tensor contractions to reduce computing requirements, needs good algorithms and data structures to achieve high performance**
To overcome the first two challenges, another approach to reduce computation and communication challenges is exploring sparsity in tensors. There have been research on exploiting dense block-pairs of the two input tensors, and then do multiplication by calling dense BLAS linear algebra and have the output tensor pre-allocated using domain knowledge or a symbolic phase [17, 23, 38, 39, 49], such as libtensor [11, 36], TiledArray [39], and Cyclops Tensor Framework [30]. Despite the element/pair-wise sparsity could compress the tensor effectively, while the two sparse tensor contraction (SpTC) algorithm, emerged from recent quantum chemistry research [15, 37], still lacks sufficient research to be efficient. Some recent efforts have been put in this direction as initial efforts [34, 35], but the challenges still need to be better resolved.

Part III.

Conclusions

Chapter 5.

Outlook

The purpose of outlining a grand unifying plan is to encourage the community to partake in its eventual success. We emphasize that the time has passed for disparate groups to struggle in closed source developments. Inclusive authorship like the NWChem paper is a laudable and achievable goal as a community. The recent successes of for-profit companies like Google and their ilk in academic arenas should further serve as a wake-up call; it is only in their ability to connect working scientists of various disciplines have their successes been assured. GPAW holds monthly developer meetings, Stan likewise has developer hours, as do DeepChem and some other; however, these are still currently few and far apart. Contrary to some beliefs, these have not diluted the ability of the steering groups nor swamped the communities with frivolous demands. Recently, even the Fortran steering committee has opened a public forum (in the form of the J3 proposals repository) to bring more of the community into the decision making process. Computational chemistry, once one of the early adopters of the net has fallen behind, and it can only be hoped that programs like those held at IPAM will be able to stem the flow of bad practices and secrecy.

Tensor algorithms are well defined within their niches and have seen successes on data which is traditionally part of quantum chemistry. However, they need to be merged with the nebulous lexicon of quantum chemistry. Many avenues are unexplored, relativistic qc, electrostatics. Outlook, they're powerful, expressive tools which need to be coerced into the qc lexicon for efficient use.

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