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# Stochastic Algorithms in Sparse Antisymmetrized Hilbert Spaces

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**14. ABSTRACT**

The final report of grant FA9550-16-1-0256 details the developments and successes made in the exponentially-complex computational challenge posed by quantum many-body problems throughout chemistry, materials science and condensed matter fields of research. The work focused on the development of fast randomized iteration approaches for computing thermal and response properties of systems in these fields, via novel stochastic sampling methods, which enabled beyond state-of-the-art calculations on a number of key correlated electron systems. These included the polarizability of molecular systems, spin correlations of graphene sheets, and thermal calculations on strongly correlated lattice models. In addition to thermal and response properties, the ability to stochastically project into appropriate subspaces was considered, and the limitations on such an approach detailed. As an alternate stochastic compression, the work enabled by this grant has also investigated a novel approach to stochastically sampling a compressed, non-linear parameterization of the solution vector. This work draws on inspiration from the optimization of neural networks in order to hugely increase the complexity of states which can be considered in this way. Key successes of the work include the development of a new algorithm to stochastically sample the response of a quantum system to perturbations, the demonstration of this algorithm to resolve a discrepancy in the literature based on alternative (inexact) methods, the development of a new computational approach to sample and optimize highly flexible and complex non-linear wave function ansatze, and the thorough analysis of the limitation of stochastic subspace sampling, which has found use in some proposed quantum computing algorithms.

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# Final Report:
## *Stochastic Algorithms in Sparse Antisymmetrized Hilbert Spaces*

### Outputs

This 2-year grant (covering Sept. 2016 – Sept. 2018) has seen a research output of **4 published papers** derived from the work directly enabled by this grant, along with **14 invited talks** by the PI which were (wholly or partially) based on work achieved during the grant. The four research papers, including one in the high-profile 'general-physics' journal '*Physical Review Letters*', were:

[1] "Projector Quantum Monte Carlo for Nonlinear Wavefunctions"
Schwarz, Alavi, **Booth**, *Physical Review Letters*, **118**, 176403 (2017).
[2] "Density Matrices in Full Configuration Interaction Quantum Monte Carlo: Excited States, transition dipole moments and parallel distribution",
Blunt, **Booth**, Alavi, *J. Chem. Phys.*, **146**, 244105 (2017).
[3] "Response Formalism within Full Configuration Interaction Quantum Monte Carlo: Static Properties and Electrical Response",
Samanta, Blunt, **Booth**, *J. Chem. Theory Comput.* **14**, 3532 (2018).
[4] "Non-linear Biases, Stochastically Sampled Effective Hamiltonians, and spectral functions in quantum Monte Carlo methods",
Blunt, **Booth**, *Physical Review B*, **98**, 085118 (2018).

High-profile invited presentations over this period included a workshop at the Schrödinger Institute in Vienna (titled: Synergies between mathematical and computational approaches to quantum many-body physics), and a presentation at the opening of the Simons' Institute in computational quantum physics in the Flatiron, New York. Below, we discuss the work and advances made from the grant in the context of the original objectives of the project; where success was achieved, the avenues of further research which have opened, as well as in which areas of the initial project proposal we were not as entirely successful. This will also cover the content and highlights of the published papers acknowledging the grant above.

### Successes

The project proposal focused on developing fast randomized iteration methods for quantum many-body problems. Quantum many-body problems arise in a number of areas, from nuclear physics to condensed matter and quantum chemistry, however it is mainly these last two areas that were focused on. Quantum problems suffer from an exponential scaling bottleneck, whereby any state is represented as a linear superposition of an exponentially large Hilbert space. Furthermore, when dealing with Fermions (such as electrons, the objects of primary interest for chemical, materials or condensed matter problems), the overall state has to obey antisymmetry with respect to exchange of any two particle coordinates.

The setting for the work was then to exploit the duality between fast randomized iteration methods for linear algebra, and the solution to quantum many-body problems in exponentially large, antisymmetric Hilbert spaces. Previous work (by the PI and others) had focused on finding the ground state solution to a given problem, which maps down to finding an extremal eigenvalue to a matrix. The progress in this grant was to develop alternate descriptions of these systems, which exploited the mapping to other linear algebra operations, which could similarly be solved by fast randomized iteration methods, building on the success in exploiting this duality for the ground state. For instance, the solution to a system of linear equations can be mapped to the response of a system to an external perturbation – the fundamentals of spectroscopy. Similarly, the exponentiation of an exponentially large matrix is required to compute thermal (finite-temperature) properties of an interacting electron system, can this be treated in a similar fashion? Coupled to these challenges, it was necessary to also investigate transformations in the underlying representation of the problem, to aid efficiency, and to allow for these quantities to be easily extracted. The four main aims of the original proposal, along with the outputs that particularly pertain to them, were therefore as follows:

- Development of stochastic methods for solution of **linear equations**, giving the response of an interacting system to an external perturbation [Refs. 2, 3, 4]
- Development of stochastic methods for a **matrix exponential**, allowing for the extraction of thermal and temperature-dependent observables [Refs. 4]
- **Stochastic transformations** in representation of problems for the above thermal and response states [Refs. 1, 3, 4]
- **Applications** to problems in materials science and quantum chemistry [Refs. 1, 2, 3, 4]

These represented ambitious aims for the project, but were all at least partially successful, leading to development of new methods, which are now finding application areas in diverse areas of quantum simulation. The challenge of solving for linear equations to calculate response properties of systems (**Objective 1**) was taken up primarily in Ref. 3, where a stochastic algorithm was developed which sampled the full, exponential Hilbert space of classical electron solutions, which allowed for a stationary and stable sampling of the *response* of a system to a static perturbation. The application of this newly developed method was to the area of quantum chemistry, and next-level accuracy in quantities such as the polarizability of a molecule, i.e. the response of a molecule to an external electric field. In the method, we demonstrated the ability of the developed stochastic method to sample in spaces of dimension up to $10^{14}$, in order to solve these linear equations, far beyond the capability of existing deterministic methods. This allowed for the resolution of a long-standing disagreement in the literature about the polarizability of the nitric oxide molecule, where existing methods diverged from one another.

The development of this method required a further technical development, which was detailed in Ref. 2. While sampling the response of the wave function

was possible with the stochastic dynamic, an additional complication came from the fact that the polarizability is computed as the contraction of this wave function with the ground state. This quadratic form required a new sampling to avoid errors resulting from the non-linear function of stochastic variables. This technical challenge was overcome in Ref. 2, with the computation of *transition density matrices*. In addition to computing these, a demonstration of large-scale parallelism on distributed memory architecture was also detailed and benchmarked, showing impressive scalability to larger systems.

Future research directions building on this objective that were not completed include the extension to *dynamic* perturbations. For instance, while the polarizability is the response of a system to an applied, static electric field, a key further quantity will be the dynamic polarizability, which is the response to a time-varying electric field, namely light. We believe that the method can be further extended in this direction to compute spectral properties which will further increase the scope of the developed method.

**Objective 2** involved the sampling of finite-temperature quantities via the matrix exponentiation in these antisymmetric Hilbert spaces. This was achieved via a stochastic transformation (**Objective 3**), which allowed for the stochastic projection of the exponentially large Hilbert space into a space that could then be readily manipulated in a standard deterministic fashion. In order to compute these thermal quantities, a set of statistical snapshots from the thermal state distributions at different temperatures was used to form a stochastically generated Krylov subspace into which the Hamiltonian of the system was projected. While this allowed finite temperature results to be extracted, Ref. 4 also highlights and examines the difficulty in controlling the biases that are inherent in this type of stochastic projection. Overall, while the algorithm was shown able to go beyond the state-of-the-art in computing these quantities for application to condensed matter systems, it also found a scaling for introduction in sampling bias which rapidly limited the application of the approach to ever-larger systems. Analysis of these types of limitation are also finding application now in quantum computing algorithms, where stochastic sampling is also employed. Therefore, even though the developed method was only partially successful, a thorough analysis of the deficiencies of the approach has been illuminating for a range of methods in other areas with similar characteristics of stochastically derived subspace projection.

**Objective 3** was concerned with stochastically transforming the representation of the problem. In combination with objective 2 (described above), this was performed in the context of thermal and spectral states in solid-state systems. However, in an alternative approach detailed in Ref. 1, this stochastic sampling was instead used to transform the solution vector into a non-linear parameterization of the space. This new sampling of *non-linear* wave function forms in these spaces allowed for a huge compression in the wave function, such that the parameters could now be stored exactly, but that the sampling of the space was now performed using the stochastic dynamics previously developed for ground state quantities. This entered the realm of 'variational Quantum Monte Carlo' methods, and the approach formulated a new approach within a now restricted Ansatze class. The new methodology was shown able to optimize many millions of parameters, by drawing on algorithms developed for the optimization of deep neural networks. This represented a significant jump in the

complexity of states which could be considered, and was applied to both strongly-correlated lattice models, as well as *ab initio* materials science problems such as the spin correlations in graphene. This approach has been subsequently picked up in a number of other research groups internationally, which now are looking into these techniques for the stochastic optimization of many-variable quantum states.

**Objective 4** concerned the application to a broad range of systems, especially those in predictive quantum chemistry. All publications contained application to systems from a diverse range of fields, in order to try to disseminate the information to as broad an audience as possible. Particular highlights include the significant step in accuracy achieved in demonstrating the polarizability of nitric oxide [Ref. 3], as well as in the spin correlations of Graphene [Ref. 1].