Computational Complexity in Electronic Structure

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In quantum chemistry, the price paid by all known efficient model chemistries is either the truncation of the Hilbert space or uncontrolled approximations. Theoretical computer science suggests that these restrictions are not mere shortcomings of the algorithm designers and programmers but could stem from the inherent difficulty of simulating quantum systems. Extensions of computer science and information processing exploiting quantum mechanics has led to new ways of understanding the ultimate limitations of computational power. Interestingly, this perspective helps us understand widely used model chemistries in a new light. In this article, the fundamentals of computational complexity will be reviewed and motivated from the vantage point of chemistry. Then recent results from the computational complexity literature regarding common model chemistries including Hartree-Fock and density functional theory are discussed.

1 Introduction

Quantum chemistry is often concerned with solving the Schrödinger equation for chemically-relevant systems such as atoms, molecules, or nanoparticles. By solving a differential and/or eigenvalue equation, the properties of the system and the dynamics of the state are obtained. Examples of properties include: equilibrium geometries, the dissociation energy of molecules, and the vibrational frequencies.

The difficulty stems from the accuracy required and the apparent exponential growth of the computational cost with both the number of electrons and the quality of the description of the system. For practical applications, the accuracy required is typically orders of magnitude smaller than the total energy of the system. As a concrete example, the Carbon atom has total electronic energy of about 37.8 Hartrees while the energy of a Carbon-Hydrogen bond is only 0.16 Hartrees. Solving the full eigenvalue equation takes on the order of n^3 operations for an $n \times n$ matrix. However, when describing interacting many-electron systems, the dimension of the matrix increases exponentially with the number of electrons.

Consequently, the computational methods of electronic structure in chemistry are aimed at circumventing exact diagonalization, in the context of electron structure, called the full configuration method. Avoiding exact diagonalization has led to a wide range of computational methods[1–4] for computing properties of chemical interest. These methods have recently[5–9] begun to include *quantum simulation* following Feynman's suggestion[10] to use quantum computers as sim-

ulators. Subsequent development of these ideas in quantum chemistry has led to new proposed methods utilizing quantum computational techniques[8,11–14] and proof-of-principle experiments[15,16]. However, questions about when and where one would expect a quantum computer to be useful[17,18] have not been fully answered. Ideally, computational complexity can provide some answers about when, where, and why quantum computers would be useful for chemistry. At the same time, it could also help formalize intuitive understanding of when we can expect reliable results from traditional computational methods.

Many results in computational complexity have interchanged classical and quantum computers fluidly leading to new results on the complexity of computing properties of quantum systems[19]. We review some recent results appearing in the computational complexity literature that touch on why electronic structure calculations are difficult. Our hope is to encourage future investigations into quantitative understanding of difficult instances of electronic structure calculations.

A similar, but much shorter, discussion of computational complexity in quantum chemistry by Rassolov and Garashchuk[20] appeared in 2008 and provided many conjectures that have since been proven or extended.

This perspective assumes exposure to second quantization and mixed states in quantum mechanics. Standard bra-ket notation is used when referring to quantum states. All the necessary concepts from computational complexity and quantum computation are briefly introduced and motivated to make the article as self-contained as possible. A key omission from the present review is a number of computational complexity results[21,22] concerning quantum–information based wave function ansatzes such matrix product states, density matrix renormalization group and their generalizations[23]. These methods are becoming accepted into the mainstream of computational chemistry[24–26], but do not yet have the widespread availability of the selected methods included in the present work.

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