FUTURE ENERGY

The Promise and Challenges of Quantum Computing for Energy Storage

Alan Ho,[1,*] Jarrod McClean,[1,*] and Shyue Ping Ong[2,*]

Jarrod McClean is a research scientist in Google's Quantum Artificial Intelligence Lab working on the development of practical quantum algorithms for quantum simulation and other problems. He received his PhD in Chemical Physics from Harvard University specializing in quantum chemistry and quantum computation supported by the US Department of Energy as a Computational Science Graduate Fellow. His research interests broadly include quantum computation, quantum chemistry, numerical methods, and information sparsity.

Alan Ho is a product manager in Google's Quantum Artificial Intelligence Lab working on identifying applications of quantum computing. He has spent his career in a number of engineering and entrepreneurial roles. His engineering interests broadly include quantum computation, quantum chemistry, and productization of new technologies.

Shyue Ping Ong is an Associate Professor in the Department of NanoEngineering at the University of California, San Diego. He received his PhD in Materials Science and Engineering from the Massachusetts Institute of Technology in 2011. His research interests are in the inter-disciplinary application of *ab initio* calculations and informatics to the discovery and study of novel materials for energy and other applications.

With recent advances by industry, the emergence of quantum computing at a capability that surpasses the limits of classical computing is fast approaching. An example of these advancements is the superconducting qubit technology developed at Google (as seen in Figure 1). A key area where quantum computing has been predicted to offer dramatic advances is in applications to materials science and quantum chemistry. Here there is a close link between the natural system and engineering quantum devices, allowing for dramatic advances in what can be simulated and how these systems can be understood. This subfield has evolved rapidly over the past few years with experimental demonstrations and theoretical advances alike. A broad perspective on this relationship is laid out in a recent review article.[1] Our natural instinct is to harness the newly found and unprecedented problem-solving capabilities of quantum computing and direct them toward the defining challenges of our time. In this Future Energy, we frame and explore the opportunity of applying quantum computing to energy storage. Here we focus on computational materials design of batteries as a specific example.

State of Current Methods for Battery Modeling

The full promise of intermittent renewable energy technologies to displace significant greenhouse gas emissions requires large quantities of reliable and cheap energy storage capacity to come online at an accelerated pace. While the recent advances in battery technology and its scale of deployment are encouraging, there is always increasing demand to build safer and lighter batteries, or find replacements for materials that are in high demand (e.g., cobalt).

At the same time, the performance and cost of a battery are intimately tied to the properties and integration of its component materials (e.g., anode, electrolyte, and cathode). Given these design constraints, there has been considerable interest in the use of computational modeling to predict better materials and designs for batteries and energy storage, many of which are based on the solution of Schrödinger's equation for the constituent electrons.

Indeed, there are many research efforts using quantum mechanical calculations performed on classical computers to inform solutions to the grand challenges at the forefront of sustainable energy.[2] Among the many approaches to solving the Schrödinger equation, density functional theory or "DFT"[3] calculations are the most common. DFT calculations have an impressive and growing track record of accurate, insightful predictions that have helped guide the development of energy materials in lithium-ion batteries, hydrogen production and storage materials, superconductors, photovoltaics, and thermoelectrics, to name a few.[4]

For lithium-ion batteries,[5] expert practitioners have developed DFT-based methods to calculate and predict several properties critical to battery operation with good resolution: equilibrium cell voltages, voltage curves (0 K and finite temperature), ionic mobility, and thermal and electrochemical stability. Knowing these properties is often sufficient to evaluate and screen for key battery performance metrics, namely energy density, charging and discharging time, safety, and cycle life.

For all its success, however, the accuracy afforded by modern DFT methods builds on a foundation of sophisticated physical approximations and assumptions regarding the interaction of electrons, most notably the choice of the exchange-correlation functionals. Naturally certain approximations prove more accurate than others for
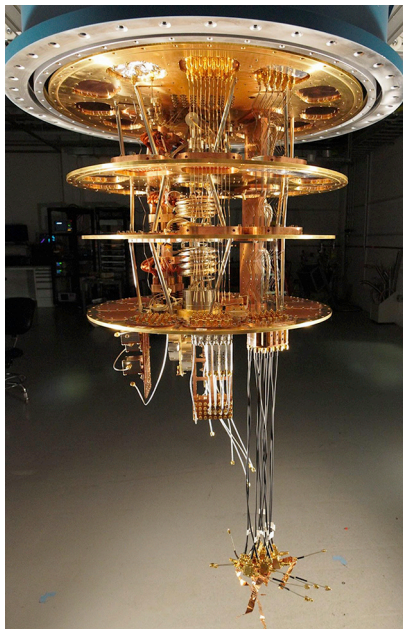
Figure 1. Google's Dilution Refrigerator
Superconducting qubits operate at near absolute zero temperatures (10 mK), and hence require multiple cooling stages. From the top down, the different cooling stages are 4 K, 1 K, 100 mK, 10 mK. The quantum chip is mounted on the bottom and is controlled via microwave signals sent through superconducting wires.

calculating certain properties or specific classes of materials or materials systems, and there is already a healthy debate stirring within the theoretical chemistry community as to the overall philosophical approach of developing new functionals to ensure they are physically sound and widely applicable.

As it stands, DFT's effective implementation as a tool to develop battery technology increasingly requires intersecting expertise in both computational electronic structure methods (especially knowing their limitations) as well as battery electrochemistry. While DFT has performed admirably and is an indispensable tool in any battery researcher's arsenal, several areas of investigation critical to the advancement of battery technology has pushed beyond DFT's limit. By navigating around the methodological obstacles

inherent to DFT using a quantum computing approach, there is the opportunity to open a window and shed light upon previously unexplorable materials systems that are critical to the operation of batteries.

We have focused here on DFT so far, but do not wish to neglect mention of other methods that have the potential to overcome some of the challenges of DFT. Alternative approaches such as quantum Monte Carlo[6] or tensor network methods[7] have an advantage of systematic refinability over DFT, and have been applied to material systems. However, the computational cost of mitigating the sign problem or storing explicitly the wave function or a compressed version of it on a classical device can make these methods prohibitively expensive for material systems of interest. In contrast, the most advanced quantum algorithms are expected to scale similarly to DFT with accuracy reflecting exact diagonalization. Recent developments in classical machine learning attempt to circumvent direct wave function simulation,[8,9] but these methods are still in their early stages.

Promise of Quantum Computing for Materials Simulation

Starting in 2005, researchers began to speculate that quantum computers could offer solutions to chemistry and material problems that were otherwise inconceivable on classical computers.[10] In particular, they offered the opportunity to provide essentially exact solutions to chemistry problems for a cost similar to that of simpler approximate methods on classical computers. These methods would be free of the errors and ambiguities present in current methods, and perhaps more importantly would negate the need for domain knowledge required to effectively utilize current methods such as DFT. While quantum methods do utilize some domain knowledge in the preparation of initial states, this can be more readily baked in as it depends only on

the Hamiltonian and not on the folklore surrounding one functional or another. Said in another way, it would help to democratize the use of computational simulations of materials, especially when quantum computing is made broadly available as a cloud service assisted by open-source, domain-specific software packages such as OpenFermion.[11]

Moreover, these computers open possibilities that have few to no current analogs in classical simulation. They are able to efficiently simulate the complex dynamics of strongly correlated, excited state, and complex interfacial systems that are necessary to achieve an accurate simulation of modern devices such as solar cells, batteries, and energy networks. Dynamics in these regimes offer a microscopic view of these systems that experiments cannot presently dream of, at an accuracy not currently possible with today's simulation techniques. These insights have the potential to change the design landscape for many of these materials.

Challenges of Quantum Computing

While the promise of quantum computing is immense, so are the challenges faced by those developing these devices. In particular, today's systems are limited, in terms of both number of entangled qubits and fidelity, which in turn dictate the number of quantum operations one can perform on the device. Progress is being made toward truly "fault-tolerant" quantum computers, which yield an unlimited number of quantum operations, bound only by one's patience; however, these machines may not emerge for a number of years. In the meantime, researchers are working hard to develop better ways to utilize the pre-fault-tolerant machines expected to be present in the near term.

Many expect that in a parallel to classical computing, the best algorithms will likely be developed with the aid of quantum computers themselves. For
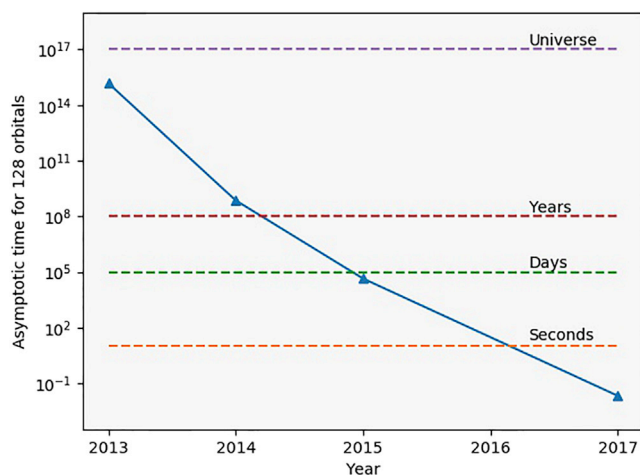
Figure 2. Cartoon Schematic of the Progress of Quantum Algorithms as a Function of the Year
The asymptotic time required on a 100-MHz quantum processor to measure the energy of a state to a fixed precision for a 128 orbital system, constructed loosely on the bounds from Table I in Babbush et al.,[17] is shown. Asymptotic time is a caricature designed to illustrate algorithmic progress in coarse terms only, and assumes a number of factors to make its depiction, such as unit pre-factors, target precision, and overlaps with target states. It is designed to show the rapid trend of progress and not to provide accurate estimates of the gate counts or physical time.

this reason, many early devices will be made available through remote or cloud services designed to allow algorithm designers to access these early devices, and bridge the gap to fault-tolerant machines.

While fault-tolerant machines are still some years away, there have still been many advances in algorithms for these machines. Through careful mathematical analysis, one can precisely bound the costs of running these machines for problems within chemistry and optimization. In 2013, the best algorithm bounds had a scaling factor of around $O(n^{11})$ for gates as a function of orbitals.[12] However, algorithmic triumphs reduced that number to closer to $O(n^6)$ within a year, and more recently reduced the expected asymptotic cost of running exact chemistry simulations on a quantum computer to costs comparable with running a hybrid DFT computation on a classical computer, or a depth closer to $O(n^3)$.[13]

However, due to the advanced nature of current classical simulation methods, the current lower end of a "useful"

quantum computer for quantum chemistry in a system that can simulate is estimated to be ~128 orbitals by the authors. This orbital number is loosely based on a comfortable margin from one of the largest numerically exact treatments of non-trivial active spaces in chemical systems, which successfully treated 72 spin orbitals using density matrix renormalization group in both ground and low-lying excited states[14] and estimates for the FeMoco complex, which predict a useful active space at 114 spin orbitals.[15] This means that to simulate interesting compounds, if one made very coarse estimates based only on rough bounds, only 3 years ago one might have expected to execute more than 10^{12} gate layers (as seen in Figure 2); however state-of-the-art algorithms have reduced these estimates to closer to 10^6. While current hardware is expected to admit on the order of 10^2–10^3 layers of gates, assuming the progression of algorithms and hardware continue to advance at the current pace we may see algorithms that work on pre-fault-tolerant devices for certain "useful" quantum simulations. Indeed it has been speculated

that hybrid quantum-classical algorithms, such as the variational quantum eigensolver,[16] and their inherent robustness to errors may help pre-fault-tolerant devices reach interesting regimes earlier than expected.

Addressing Selected Challenges in Battery Modeling
As a tool to characterize and design battery systems, one can envision quantum computing approaches would first be used to complement DFT's strengths and shortcomings. DFT can run into difficulty in modeling systems and processes involving large variations in electronic structure. For example, computing the properties of molecules, metals, or oxides with accuracy each requires different exchange-correlation functionals or DFT extensions. In contrast, a quantum computer addresses the basic physics without approximation, apart from minor discretizations that need not be tailored on a system-by-system basis.

At the same time, the underlying physical processes at the heart of battery operation occur at the electrochemically active interfaces of materials with very different electronic structure. In Li-ion batteries, a defining example is the solid electrolyte interphase (SEI) that forms between the anode (a conductive solid, Li metal or graphite) and non-aqueous liquid electrolyte during cycling. The SEI itself is a complex assortment of inorganic compounds and organic and polymeric species, which plays a crucial role in providing electrochemical stability during battery operation, but is notoriously difficult to characterize. The complex reaction paths involved in its formation remain only coarsely resolved, and expert DFT practitioners readily acknowledge the challenge and accuracy limitations of their approach.[18] Studying this problem on a quantum computer, one could simulate the exact dynamics of the system, which need not suffer from limitations related to accurate

representations of electronic excited states and/or classical nuclei that might hamper descriptions of proton transport.

A particularly egregious failure of current DFT exchange-correlation functionals is in reactions involving significant electronic structure changes in a molecular species, which are especially relevant in cutting-edge Li-O_2 and Li-S batteries that hold the potential for achieving much higher energy densities compared with Li-ion. In Li-O_2 batteries, for example, O undergoes a change from a covalent double bond in molecular O=O, to O_2^{2-} in Li_2O_2 and finally to O^{2-} in Li_2O. While simple empirical approaches such as fitting a constant correction to account for the O_2 binding energy error and self-interaction error have been developed for main group oxide formation energies,[19] such approaches become much less reliable for peroxide and superoxide species for which reliable experimental data are much more difficult to come by. These have limited progress on fundamental issues such as whether an intermediate superoxide (O_2^-) species can be stabilized relative to molecular O_2 and Li_2O_2 (similar to the Na-O_2 system), and the likely candidate crystal structures for LiO_2. Although these are but a few of the many technical impediments (including control of surface reactivity, suppression of dendrite formation, etc.) to the commercial viability of Li-O_2 batteries, they are nevertheless important questions that highly accurate quantum computer simulations may potentially help address.

Conclusion

Although quantum computing is in its early days, the potential impact that it can have on energy storage warrants further investigation and research. Due to the known limitations of current computational chemistry techniques, quantum-based computational chemistry techniques may help better elucidate material properties more accurately and

ultimately help the industry to design better batteries.

DECLARATION OF INTERESTS

A.H. and J.M. are part of Google's Quantum AI Lab and are working toward developing and commercializing quantum computing. S.P.O. an associate professor at the University of San Diego who is developing new informatics approaches to design novel energy materials.

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[1]Google, 340 Main Street, Venice, CA 90291, USA

[2]Department of Nanoengineering, University of California, San Diego, 9500 Gilman Drive, La Jolla, CA 92093, USA

*Correspondence: alankho@google.com (A.H.), jmcclean@google.com (J.M.), ongsp@eng.ucsd.edu (S.P.O.)

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