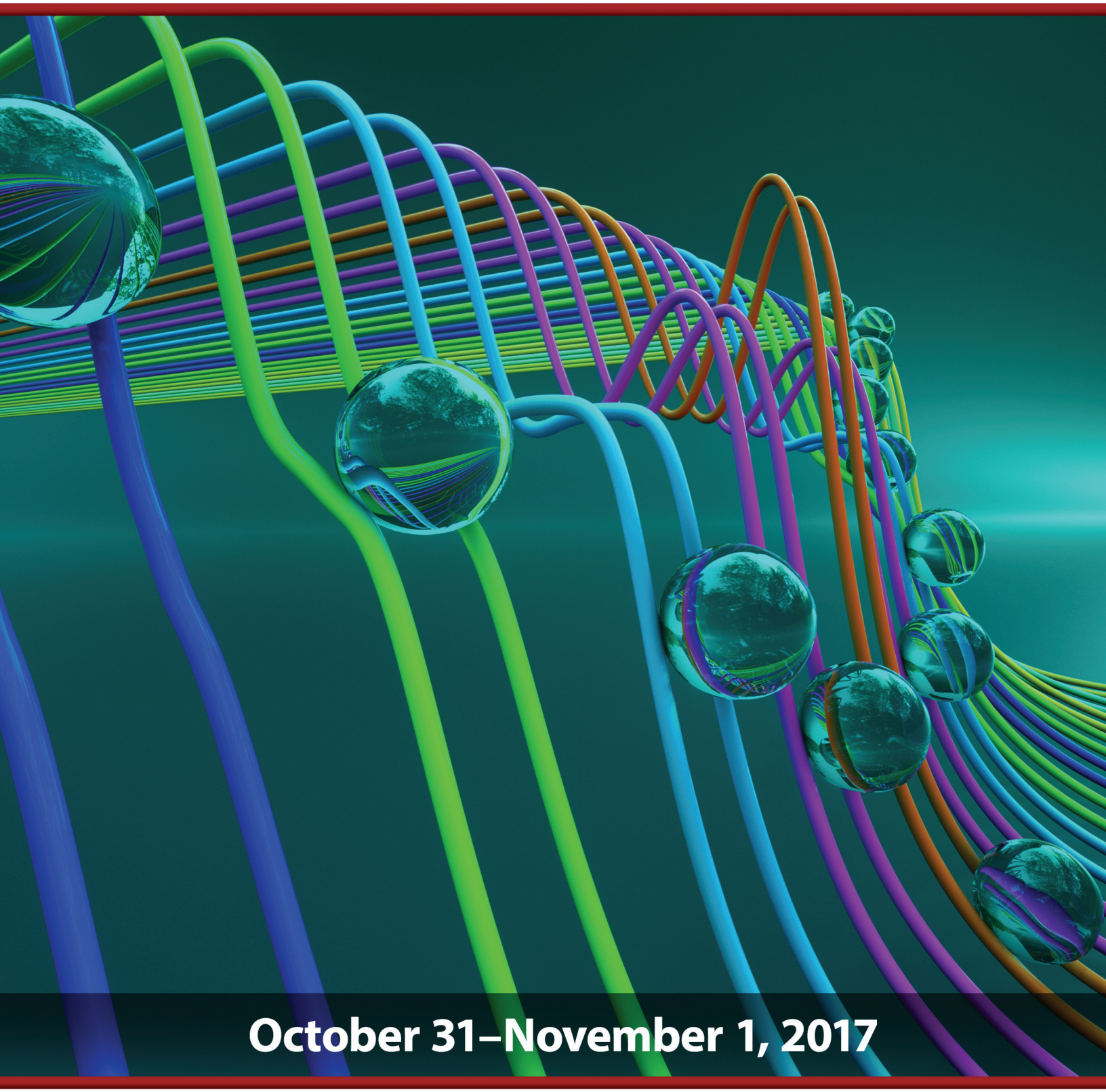
Basic Energy Sciences Roundtable

Opportunities for Quantum Computing in Chemical and Materials Sciences



October 31–November 1, 2017

Cover artwork: Experiments using specially shaped pulses of light demonstrate that quantum states can be guided along "superadiabatic" trajectories, allowing for coherent manipulations with both high speed and robustness.

Image courtesy of David Awschalom, Brian Zhou, and Peter Allen, University of Chicago and Argonne National Laboratory

Report of the Basic Energy Sciences Roundtable

Opportunities for Quantum Computing in Chemical and Materials Sciences

October 31–November 1, 2017
Gaithersburg, Maryland

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Contents

Abbreviations, Acronyms, and Initialisms

1D	one-dimensional
2D	two-dimensional
3D	three-dimensional
BES	Office of Basic Energy Sciences
CASPT2	complete active space perturbation theory second order
DFT	density functional theory
DMFT	dynamical mean-field theory
DMRG	density matrix renormalization group
DOE	Department of Energy
GHZ	Greenberger–Horne–Zeilinger
LHC	light-harvesting complex
NEVPT2	n-electron valence perturbation theory second order
PRO	Priority Research Opportunity
QMC	quantum Monte Carlo
SEET	self-energy embedding theory
VQE	variational quantum eigensolver

Executive Summary

Fundamental transformations in the basic logic of computing are few and far between. Since the invention of digital computers in the early 1940s, the logic underlying computation has remained the same, even as computing hardware evolved from vacuum tubes to silicon transistors. With the advent of quantum computation, a fundamental transformation is near. Quantum computation is based on a different type of logic: rather than being in one of the two states of a classical bit, a quantum bit or qubit can be in a superposition of two states simultaneously. Operations and measurements on these qubits obey the constraints of quantum mechanics. It is now understood that quantum computers have great power in principle to go beyond classical computers, but that not every application is well suited to implementation on quantum computers.

For reasons explained in more detail in the Introduction, scientific problems in chemical and materials sciences are uniquely suited to take advantage of quantum computing in the relatively near future. Indeed, quantum computing offers the best hope to solve many of the most important and difficult problems in this field. For example, quantum materials, such as superconductors and complex magnetic materials, show novel kinds of ordered phases that are natural from the point of view of quantum mechanics but difficult to access via computation on classical computers. Quantum sensors based on solid materials are already widely used but could be greatly improved with insight from quantum computations, as could materials for information technologies. Quantum chemical dynamics is another example of a problem that is intrinsically well suited to studies on quantum computers. Applications of quantum chemical dynamics include catalysis, artificial photosynthesis, and other industrially important processes.

Quantum computers exist in the laboratory and are beginning to exceed 50 qubits, which is roughly the size beyond which their behavior cannot be predicted or emulated on present-day classical supercomputers. While a quantum computer of 50 qubits is almost certainly not powerful enough to tackle the major scientific challenges in chemical and materials sciences, some of these major challenges start to become accessible with a few hundred qubits if error rates can be kept small. This roundtable was convened to ask how emerging quantum computers can be applied to major scientific problems in chemical and materials sciences, in light of Basic Energy Sciences's leading role in these fields and the Department of Energy's leading role in high-performance scientific computation more generally. The main outcome of the roundtable was a consensus that there are scientific problems of great importance on which emerging quantum computers have the potential for disruptive impact, and where comparable progress is unlikely to occur by other means.

These scientific problems are grouped into four Priority Research Opportunities that form the bulk of this report. **Controlling the quantum dynamics of nonequilibrium chemical and materials systems** will enable remarkable progress in major application areas such as catalysis and quantum sensing, and enable an understanding of nonequilibrium systems in which few basic principles are available to guide theory. **Unraveling the physics and chemistry of strongly correlated electron systems** is essential for progress in many basic and applied fields because strong correlations between electrons cause the standard approximations used in current approaches to break down, even when the structure of a material is relatively simple. **Embedding quantum hardware in classical frameworks** requires the development of algorithms that, as the computation is performed, efficiently pass information between classical methods for the "easy" parts of a problem and quantum hardware for the most computationally challenging parts. Such embedding methods could greatly increase the applicability of quantum hardware in the medium term by directing the limited quantum resources to the aspects of a molecule or material where they are most needed. **Bridging the classical–quantum computing divide** relies on the use of quantum algorithms to improve the key inputs of classical algorithms or capture key quantum processes. The latter is particularly important for quantum mechanical processes in open systems when they occur within decoherence-inducing environments such as electromagnetic fields and heat baths (e.g., liquid water).

The report closes with a summary of crosscutting challenges and some comments on how the different communities represented at this roundtable could continue to interact as useful quantum computation becomes a reality. The applications of quantum computing to chemical and materials sciences are still in their infancy, but visionary work has already illuminated the potential, in principle, of quantum computers to revolutionize these areas of science. Identifying specific challenges and the necessary steps to start addressing them, as this roundtable sought to do, is intended to aid progress in this difficult but worthwhile endeavor.

1. Introduction

This introduction seeks to give a brief explanation of why advances in quantum computation can be expected to impact the chemical and materials sciences. Historically, the invention of modern computers transformed theoretical chemical and materials sciences into predictive disciplines for some classes of material and molecular systems. For many important classes of materials and molecules, standard approximations such as density functional theory (DFT) are able, when implemented on high-performance computers, to make usefully accurate predictions about energetics and other key properties. However, other classes of important materials and molecules remain inaccessible to even the most powerful existing computers and are likely to remain so for the foreseeable future. There are also large classes of phenomena, such as those involving dynamics of excited states, for which reliable approximate methods are not available.

As outlined in this introduction, quantum computation promises to make these problems accessible. Additional background and information on the current state of quantum computation, including the various hardware options currently being explored, are available in the Quantum Information Science Factual Status Document available from the Department of Energy (DOE) website.[*] For the purposes of the present report, the essential properties of a quantum computer are just its computational parameters, such as the number of qubits and the speed and fidelity of gate operations. Quantum "gates" or gate operations are the basic quantum logical operations from which more complicated computations can be built, in the standard gate-based model of quantum computation.

Quantum computers operate on a fundamentally different basis from the classical Boolean logic of standard computers. At the lowest level, a standard digital computer applies logical operations (e.g., AND, OR, or NOT) to logical bits, each of which can take two possible values. Of course it is now understood how to formulate many more complicated questions, including the mathematical problems that arise in physics and chemistry, in terms of these classical bits. Although computers have come remarkably far since the early days of electromechanical relays and vacuum tubes, and many mathematical operations are nearly instantaneous, the mathematical problems that arise in systems of strongly interacting quantum particles are in general difficult or impossible because the time required for an accurate computation scales *exponentially* according to the number of particles.

Many years of effort have already gone into understanding how quantum computers—which apply unitary operations to a set of quantum two-level systems, or "qubits"[†]—are in principle enormously more efficient than classical computers in addressing some problems. One representation of a qubit, the "Bloch sphere," is shown in Figure 1. The north and south poles on the sphere correspond to the values of a classical bit, 0 or 1, or up and down in the language of electron spins. The equator of the sphere contains many different quantum states that are equal parts 0 and 1, but their quantum phase is distinguishable by other measurements.

Consequently a single qubit contains more information than a classical bit, at least as long as the qubit's coherence (the details of the superposition of 0 and 1 states) can be maintained. An example of a problem that can be solved rapidly by a quantum computer is the factoring of a very large integer into its prime components, which is currently, and widely expected to remain, exponentially difficult for classical

[*] https://science.energy.gov/bes/community-resources/reports

[†] There are other formulations of a quantum computer that lead to computational power that is roughly equal to the gate-based model mentioned here. For yet other approaches, such as quantum annealing with the transverse Ising model, the computational power is possibly less than the standard gate-based model and in general not as well understood. The roundtable did not discuss these, nor did it cover analog quantum computers or quantum emulators that target individual problems.

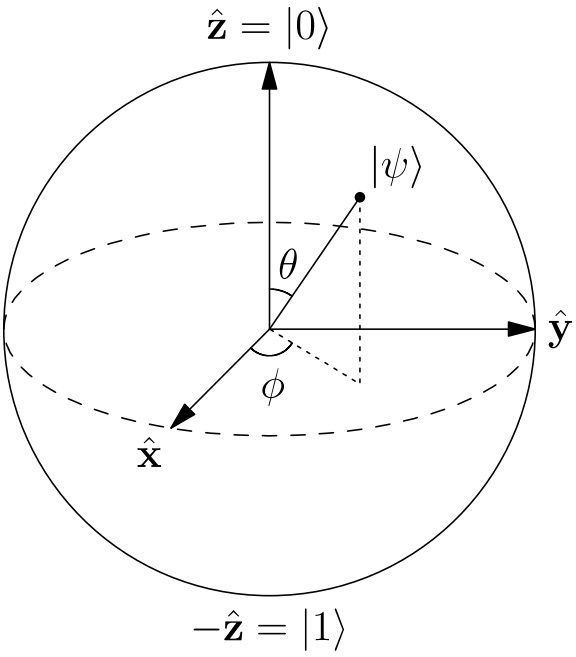


Figure 1. The Bloch sphere, a representation of the possible states of a single qubit (quantum bit). The north and south poles are the states that correspond to classical bit values 0 and 1. Other states on the surface of the sphere are distinguishable quantum states that "superpose" 0 and 1 in different proportions, and even states with the same proportions of 0 and 1 need not be the same quantum state. For example, all states on the sphere's equator are equal-weight superpositions of 0 and 1, but with different relative phases. The coherence in such a superposition leads to many more different states than for a classical bit, and quantum computation uses the ability to make coherent superpositions of the states of many qubits. | Image reproduced under the terms and conditions of the Creative Commons license

computers. Hundreds or possibly thousands of logical qubits are required for many problems originating in computer science, such as factoring. Furthermore, in the presence of errors, each of these logical qubits may require a number of physical qubits for the correction of errors. This difficulty raises the question of whether other kinds of problems might be more promising for the first applications of quantum computers, which are currently limited to fewer than one hundred qubits.

Why is quantum computation relevant to chemical and materials sciences?

As originally explained by the well-known American theoretical physicist Richard Feynman, and justified in more detail by rigorous theorems, quantum computers are particularly well suited to find quantum-mechanical solutions to problems in chemical and materials sciences —at least in principle. The Schrödinger equation that describes time evolution in atomic and molecular systems can be efficiently simulated on a quantum computer; and quantum algorithms have been developed to evaluate properties of physically relevant systems such as energies, excitation gaps, and correlations. The scaling of computational effort with particle number is no longer exponential but only polynomial. For some of these algorithms, current estimates suggest that they will already be able to exceed the performance of the best classical computers when running on as few as 100 qubits *if* the qubits are sufficiently free from error.

The first goal of this roundtable, in informal terms, was to go beyond the "in principle" statement that quantum computers should be good for theoretical chemical and materials sciences by identifying important problems upon which quantum computers might have their first dramatic impact. The second goal was to identify challenges that need to be overcome to realize the potential of quantum computers to solve these science problems. Quantum computers of various types already exist in laboratories around the world. An overview of the current state of the art of the field was given by John Preskill (of California Institute of Technology) at the beginning of the roundtable.

How large do quantum computers need to be to achieve useful results?

Several current or near-term quantum computing machines have more than 50 physical qubits. Given their ability to execute high-fidelity gate operations, this size would place them beyond the current ability to simulate the computation accurately on even a very large classical computer. For comparison, a recent large-scale classical simulation of quantum computing at the National Energy Research Scientific Computing Center was able to model 45 qubits,[1] with a further increase to 49+ qubits, albeit with a small

number (27) of gate operations performed, as reported in a preprint.[2] Although these quantum computing machines do not yet provide the required number of qubits and gate fidelities to solve the major challenge problems identified, they are large enough to conduct detailed studies of how existing and future algorithms scale with the number of qubits (e.g., updated versions of the 2005 estimates[3] shown in Figure 2) and whether they achieve robustness to errors. A sustained effort, closely connected to science questions, to improve quantum algorithms for chemical and materials sciences, as they function on real-world quantum hardware, has the potential to make major progress on problems that otherwise appear to be intractable.

History suggests that a sustained effort coupled to quantum hardware is important: very few of the common algorithms currently used on classical computers existed before algorithms could be tested on physical hardware. Even the programmable von Neumann architecture associated with the word "computer" grew only out of practical experience with the first machines. Although quantum algorithms in a general sense have been considered for many years—mostly in the theoretical computer science community—the effort has been concentrated on standard computer science problems rather than on chemical and materials sciences. In part because of the absence of hardware, the focus has been on algorithms whose speeds can be demonstrated with mathematical rigor. The quantum computing community would like to motivate more scientists to turn their interest to chemical and materials sciences problems and to focus on how well algorithms perform for difficult problems of practical importance, which may be better than mathematical worst-case bounds. Indeed, this is the case for many important classical algorithms, especially in the simulation context. A classic example is the density matrix renormalization group (DMRG). It has been widely used since the 1990s based on empirical evidence for its effectiveness, but a rigorous upper bound on its scaling was only recently established.[4] Perhaps more important than mathematical rigor is a serious effort, respected by the community, to perform unbiased comparisons of algorithms and estimate their speed and robustness.

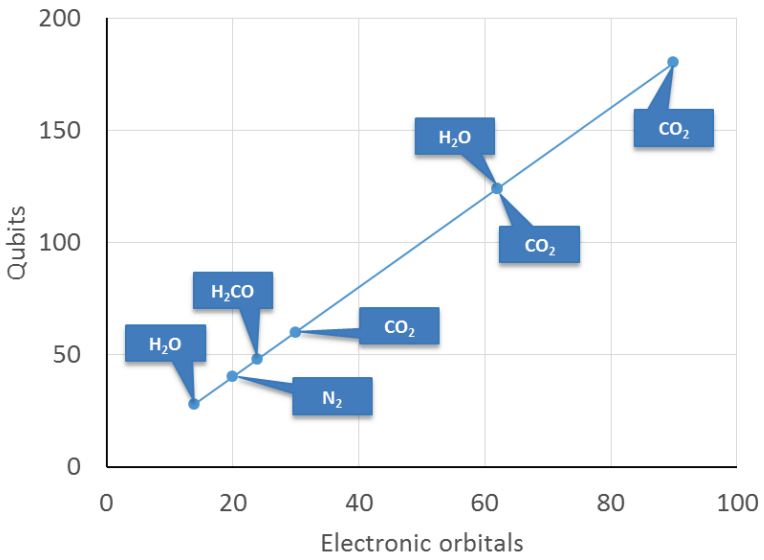


Figure 2. Illustration of linear scaling of the number of qubits with number of electronic orbits used in electronic structure calculations. The molecules corresponding to each calculation are indicated, and some molecules appear more than once because calculations were performed with different numbers of orbitals. | Graph based on information from Rinton Press, *QIC* 15, Improving quantum algorithms for quantum chemistry, M. B. Hastings, D. Wecker, B. Bauer, and M. Troyer, copyright 2015. Used by permission

Fortunately, there is a rapidly growing community of scientists developing quantum hardware and software, first for chemistry and now also for materials problems. This community is not limited to academia and national laboratories; a highlight of the roundtable for many participants was the morning session with presentations from researchers at three major companies (Google, IBM, and Microsoft) and from one of the many startups in this area (IonQ). All three companies have substantial efforts in quantum software and hardware, including quantum chemistry efforts that interface closely with the problems discussed in this roundtable. It is encouraging that quantum chemical and materials sciences are already

recognized by most or all of the major industrial players in quantum computing as viable and important applications for emerging quantum hardware.

How does this area connect to the DOE mission?

The considerable progress in quantum computing hardware to date has been enabled in large part by federal funding via multiple agencies and programs, as well as industrial research and development. Building on this progress to achieve dramatic advances in chemical and materials sciences is a logical goal for DOE's Basic Energy Sciences (BES) office, which is the largest single funder of chemical and materials sciences research in the United States. BES support includes long-standing programs in a wide variety of computational approaches to chemical and materials sciences. Indeed, many of the scientific problems discussed in the following sections were previously identified as BES goals in previous reports, such as the 2016 report *Basic Research Needs Workshop on Quantum Materials for Energy Relevant Technology*. DOE also has considerable experience in integrating high-performance computation with scientific goals. Finally, many of the demanding experimental projects supported by the Office of Science would benefit from the possible advances in chemical and materials sciences discussed in this report.

Roundtable participants identified the following four Priority Research Opportunities (PROs), which are described in detail in the following sections:

1. Controlling the quantum dynamics of nonequilibrium chemical and materials systems

2. Unraveling the physics and chemistry of strongly correlated electron systems

3. Embedding quantum hardware in classical frameworks

4. Bridging the classical–quantum computing divide

The path to substantive applications of quantum computers such as these will not be an easy road. New hardware challenges will emerge as the size of quantum machines grows, and it could well be that the scale of future quantum computers does not show the exponentially rapid growth (i.e., Moore's Law) familiar from the history of classical computing. Fortunately, for some of the scientific problems identified, the hardware size needed for progress is not very much larger than the sizes of quantum computers expected to exist in the next few years—although the number of qubits alone is not very meaningful without a quantification of errors. Decades of experience with classical computers have given the chemistry/materials community some insight into which problems need a truly new approach such as quantum computing. An exciting aspect of the roundtable, noted by several of the experienced computational scientists present, was the glimpse of a pathway by which these truly fundamental and challenging problems might be solved.

2. Priority Research Opportunities

PRO 1: Controlling the Quantum Dynamics of Nonequilibrium Chemical and Materials Systems

Contributors: Giulia Galli, Norman Yao, Wibe (Bert) de Jong, Thomas Devereaux, and Jarrod McClean

Overview

Understanding, predicting, and controlling the quantum dynamics of nonequilibrium systems has the potential to accelerate key science and engineering applications ranging from quantum chemistry and metrology to materials design. Although the past half-century has seen tremendous progress in capturing the low-energy landscape of *equilibrium* quantum systems, it has become increasingly clear that (1) nonequilibrium systems can exhibit fundamentally richer behavior than their static counterparts, and (2) understanding such behavior will be essential to discover novel catalytic pathways, topological materials, quantum sensors, and the like. To solve those questions, one must move beyond a powerful set of classical computational tools that, while ideal for calculating equilibrium low-energy properties, are wholly unsuited for calculating the complex nonequilibrium dynamics of chemical and materials systems.

To this end, a near-term quantum information processor can enable important advances in simulating and predicting the behavior of nonequilibrium systems, well beyond what is achievable via classical computations. The diversity associated with such nonequilibrium systems stems, in part, from the multitude of ways in which such behavior can emerge; for example, out-of-equilibrium dynamics are found in optically driven systems, chemical reactions, frustrated glasses, catalytic pathways, and molecular potentials. The discussion that follows focuses on four scientific challenges at the interface of chemistry, physics, and materials engineering in which a near-term quantum computer can provide transformative insights via simulations of nonequilibrium dynamics.

Scientific Challenges

Quantum simulations of chemical reactions and catalytic pathways

Mastering the guiding principles that underlie catalytic chemical reactions is crucial to our transition to a more sustainable economy. Indeed, catalysis represents an essential component in the production of most chemicals and fuels, with different catalysts functioning to enable diverse chemical tasks ranging from the splitting of water and nitrogen fixation to the selective transformation of complex molecules for pharmaceutical synthesis.[5] Despite its importance and ubiquity, predicting catalytic reactions using computer simulations remains an open challenge, encompassing outstanding problems such as the simulation of chemical reactions in condensed phases, the importance of nonadiabatic phenomena, and the characterization of pathways often involving (multiple) electronic excitations.[6]

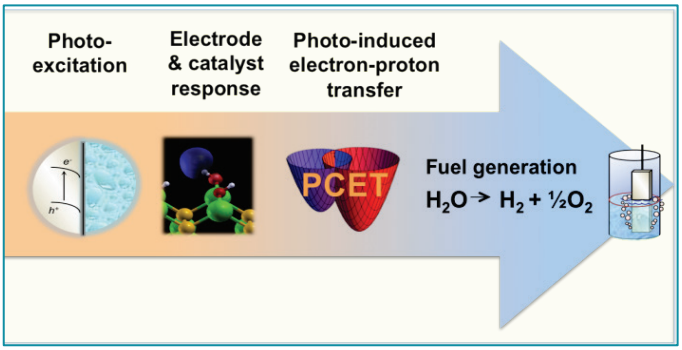
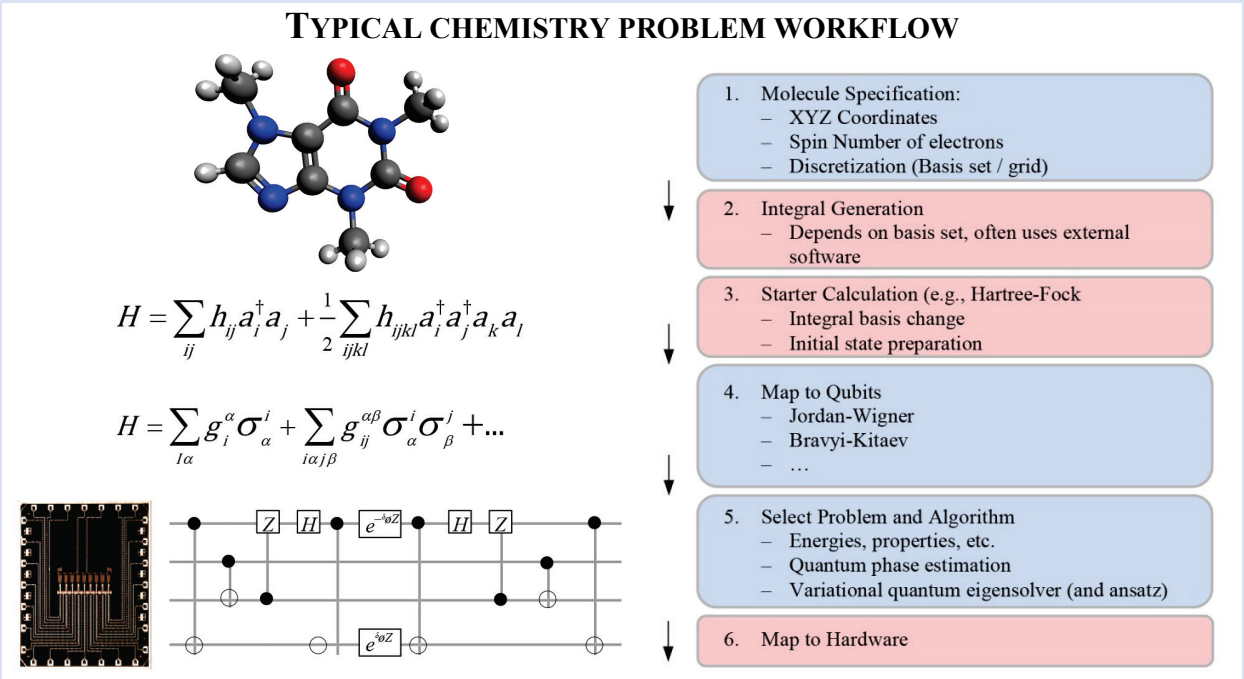


Figure 3. Pictorial representation of a water-splitting reaction occurring on a catalytic surface, which starts with harvesting light energy to form charge carriers and can involve proton-coupled electron transfer (PCET) processes. The full quantum chemical simulation of such a reaction (by solving the time-dependent Schrödinger equation) is expected to be unfeasible with classical computers. | Image courtesy of Giulia Galli, University of Chicago

Although these problems are being pursued with classical computers, with steady progress, quantum computations could provide the necessary leap to solve the time-dependent Schrödinger equation for complex systems, inclusive of surfaces in contact with fluids and eventually in the presence of electric fields (Figure 3). However, a simpler yet ambitious near-term goal is the full quantum simulation of a small-scale chemical reaction, such as OH + CO →H + CO_2, going beyond the Born-Oppenheimer approximation (i.e., including the coupled quantum dynamics of electrons and nuclei). This simulation could proceed in two steps, with the first focusing on the reaction in the gas phase and the second focusing on the reaction in the presence of a solid surface.[7] The sidebar "Mapping a Chemistry Problem onto a Quantum Computer" explains how a chemical reaction is mapped into a set of operations for a quantum computer.

Mapping a Chemistry Problem onto a Quantum Computer



Chemistry simulations have been identified as a leading application for near-term quantum computers and quantum simulators. However, mapping a chemistry problem from the molecular domain to the qubits of a quantum computer is a task requiring a number of important steps[8,9]:

1. Specify a molecule geometry, spin, charge, and basis set.
2. Calculate the integrals within the chosen basis set that define the problem.
3. Perform a Hartree-Fock or correlated calculation to set the reference vacuum state
4. Transform the Hamiltonian into qubits from the second-quantized Hamiltonian.
5. Select the property and algorithm of interest and generate the circuits.
6. Map the quantum circuit to the constraints of the target hardware.

To maximize contributions by scientists in this area, it is key that software tools be built to bridge gaps of knowledge, enabling domain scientists from both chemistry and quantum information to tackle open questions. An open-source example, OpenFermion (http://www.openfermion.org), has been developed to assist users in taking the problem through all of the aforementioned steps.

Image at top courtesy of Jarrod McClean, Google. Quantum circuit at bottom left is based on work reported in and is presented with permission from Nature Publishing Group. *Nature*, State preservation by repetitive error detection in a superconducting quantum circuit, J. Kelly, et al., copyright 2015.

Among the many challenges associated with computing the nonequilibrium dynamics of a chemical reaction, one of the most essential is to establish a rigorous metric assessing the reliability of results. In the case of catalysis, a particularly appealing comparison would be to benchmark these quantum simulation results with the direct measurement of basic pathways. Such measurements have recently been achieved by exploiting the sensitive, broadband, and high-resolution capabilities of time-resolved cavity-enhanced direct frequency comb spectroscopy.[10]

To capture the full reaction dynamics present in catalytic chemistry, it is expected that between ~10^2 and 10^3 qubits would be required, with a gate depth (the number of gate operations carried out by each qubit) of ~10^3 gates per real-time evolution step.[11] To this end, while near-term quantum computers will help provide insight into the short-time behavior of reactions, to access the long-time dynamics may require tailored algorithms that implement simple forms of error correction.

Quantum computing for the discovery of new topological matter

Spurred by recent progress in the melting, enhancement, and induction of ordered states of electrons using out-of-equilibrium techniques, a tantalizing prospect is to use a near-term quantum computer to discover new dynamical phases of matter.[12] In particular, in the presence of strong, broad optical pump pulses, the electronic properties of quantum materials can be dramatically altered, leading to a novel landscape of transient Floquet steady states.[13,14] By manipulating the underlying symmetries governing the material, including for example, inversion and time-reversal, nonequilibrium driving offers the opportunity to create strongly interacting states of matter with no static analogs.

This strategy can be applied to materials to alter their topological properties. For example, frustrated quantum magnets host a delicate interplay of competing orders, as well as putative spin liquid phases, so that weak symmetry-breaking optical perturbations could be expected to have an outsized effect in determining the transient state. Recently, it has been shown that shining circularly polarized light on a Kagome Mott antiferromagnet can transiently induce a Floquet chiral spin liquid (Figure 4).[15] Circularly polarized optical pumping below the charge gap dynamically breaks time reversal and promotes a scalar-spin-chirality term to the spin dynamics. The resulting chiral spin liquid has emergent excitations that obey the "semionic" statistics characteristic of a ν=1/2 Kalmeyer-Laughlin fractional quantum Hall state. Such emergent, fractionalized excitations are also believed to form the basis for quantum computation, which is immune to local noise and decoherence.[16] Therefore, simulating and understanding the dynamics of optically "switchable" interacting topological systems is highly desirable both from a fundamental physics perspective and as a possible new arena for noise-resistant quantum algorithms.

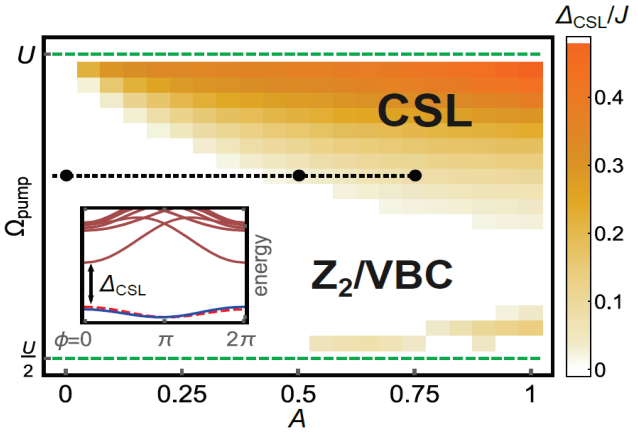


Figure 4. Phase diagram of the driven 2D Kagome antiferromagnet showing a transition into a chiral spin liquid with energy gap Δ_{CSL} in circularly polarized light fields | Image reprinted from M. Claassen, et al., *Nat. Commun.* **8**, 1192, 2017, under the terms and conditions of the Creative Commons CC BY license

However, accessing and predicting the quantum properties of strongly driven, nonequilibrium quantum materials is a tremendous numerical challenge. To date, numerical simulation (even on large-scale computational clusters) has been limited to small 1D and 2D amenable simulations to exact diagonalization and DMRG techniques. To this end, quantum computing has the potential to dramatically

increase the understanding of how novel topological states of matter can be realized and stabilized in realistic materials away from equilibrium. Moreover, once such materials are identified, a quantum processor will be able to simulate the dynamics of fractionalized excitations, suggesting novel strategies for the control and manipulation of these emergent degrees of freedom.[17] Finally, by scaling up the quantum simulation of such driven systems, it may become possible to discern certain blueprints for the engineering of topological matter from the bottom up using a combination of existing materials and optical pulses.

Understanding entangled states in materials for metrology with a quantum computer

At a fundamental level, a quantum computer is powerful because it can prepare and maintain complex superpositions across many quantum degrees of freedom. This allows the design of new sensing materials with improved ability to interrogate chemical systems and quantum materials in order to precisely measure their properties, which is crucial for both fundamental and applied sciences. Current quantum coherent sensors use an ensemble of independent qubits (e.g., nitrogen vacancy centers in diamond), with which one can achieve a significant enhancement in measurement sensitivity, with errors scaling as $\sim 1/\sqrt{N}$, where N is the number of sensors.[18] While the aforementioned scaling suggests that increasing the number of particles always improves the signal-to-noise ratio, crucially, this argument does not capture the effect of inter-qubit interactions. Indeed, above a certain density, interactions among the nitrogen vacancy centers will fundamentally limit the precision of any measurement and thus the maximum achievable sensitivity. More intuitively, once the sensing qubits become too close to one another, their accumulated spectroscopic signal will be dominated by the field of nearby particles rather than the external field being measured.

To overcome this fundamental limit, one must be able to turn the interactions among the sensing particles into a resource and not a hindrance. This can be achieved if the interactions between the particles generate specially entangled quantum states, such as the Greenberger–Horne–Zeilinger (GHZ) state;[19] in this case, the nonclassical properties of the GHZ state enable precision measurements that reduce noise more rapidly than $1/\sqrt{N}$ (the standard statistical reduction from multiple independent sensors). However, an outstanding challenge for materials theory is to understand which strongly entangled states are actually generated in real materials as the density of sensing defects increases, while avoiding enhanced decoherence effects. To this end, a near-term quantum information processor capable of deterministically creating a variety of entangled states would significantly benefit quantum metrology, with enhancements in terms of both measurement sensitivity and bandwidth.[20] In addition to questions about the equilibrium states of sensing materials, quantum computing could test how using nonequilibrium pulsed controls might energetically stabilize these entangled states beyond their normal lifetimes (Figure 5).

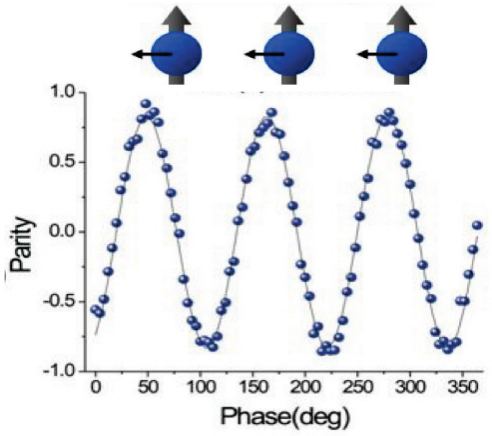


Figure 5. Using a three-ion GHZ state enables a ~1.42× enhancement in spectroscopy over the projection | Image courtesy of Wineland Group, National Institute of Standards and Technology

Simulating frustrated quantum dynamics

In strongly frustrated systems, competing interactions can conspire with fluctuations to prevent simple classical order from emerging.[21] When interactions are short-range, frustration typically relies upon geometry. This is most pertinent in the solid state, in which frustrated exchange interactions have led to the discovery of a number of exciting topological materials in layered 2D Mott insulators. An alternate route to frustration is provided by longer-range interactions, commonplace in chemical systems. For example, understanding the role of long-range

ion-dipole forces (Figure 6) in generating dynamics in aqueous solutions remains a particularly challenging task.[22] By virtue of their intrinsic frustration, the energetic landscape of such systems can be highly complex with multiple nearly degenerate minima. To this end, methods to simulate the dynamics of frustrated quantum systems remain scarce.

By dynamically tuning the energy landscape, a quantum computer may be able to harness tunneling effects to more efficiently sample the landscape of minima in a frustrated system. Moreover, if quantum control techniques can generate finite overlap with a desired state, purification methods can systematically enhance this initial overlap. Finally, nonequilibrium frustrated dynamics can also emerge in disordered systems; in these cases, the disorder can cause localization, which prevents a system from becoming ergodic and sampling all of its allowed phase space.[23] Such systems present a possible alternative to stabilizing quantum coherent behavior in a many-body system without requiring complete isolation from the environment.

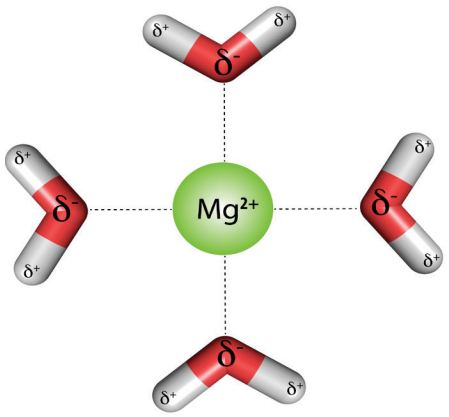


Figure 6. Charge-dipole interactions, where four water molecules are interacting favorably with a magnesium dication. | Image reproduced from Molecular Interactions courtesy of Loren Dean Williams, Georgia Institute of Technology

Computational Challenges

The primary computational challenge for near-term quantum simulations of nonequilibrium quantum dynamics is the gate depth required to achieve interesting simulations.[24,25] At present it is believed that devices will be able to achieve a gate depth of $\sim 10^3$ before error correction is required. It is not yet known if this is sufficient to reach time scales of physical interest for problems in chemical and materials systems. To this end, the further development of novel, testbed-specific algorithms will be essential. While a gate depth of $\sim 10^3$ may be physically feasible, the combination of gate fidelity limitations (currently at 99.9% or less) and qubit lifetimes will put limits on the accuracy of the results of quantum computing–driven simulations.[26] In a post-error correction setting, the difficulties are more likely to be related to initial state preparation, runtime, and analysis.

These considerations lead naturally to the quantification of the impact of different quantum errors on the algorithms of interest. The most common error metric for quantum computations today is the fidelity of the quantum state; however, many alternatives may be preferable.[27] For the simulation of physical systems, it is clear that high fidelity is generally too stringent a requirement and moreover offers little insight into the degree of error expected in practice. This can be seen from a simple example of a collection of quantum spins or particles, where in a computational state of all 0s and 1s, the flip of a single spin will reduce the overlap fidelity with the original state to zero. If the physical system has *local* interactions, the error in a quantity of interest may scale only as a small polynomial, e.g., linear or quadratic function of the number of spins flipped. It is also clear in such a model that as more spins are flipped, the error in the energy increases, while the fidelity remains zero. Therefore, it is important to identify improved metrics that accurately capture the error of a nonequilibrium quantum dynamics simulation. It is likely that the best choices will depend on the specific algorithm, the physical system being studied, and the quantum hardware in use.

The need to quantify errors in quantum simulation naturally leads to the idea of error model–specific algorithms. That is, if there is a reasonable idea of the errors that are expected to occur on a particular quantum hardware setup, are there steps that can be taken to mitigate the impact of these errors on the simulation? For example, if a setup incurs significantly more errors of one type than another, e.g., more

dephasing errors than bit flips, it may be possible to alter the algorithm or encoding to account for this. A number of near-term error mitigation strategies have been suggested along these lines in the context of quantum simulations.[28,29] In these cases, a reasonable error model allows a marked reduction of the impact on the calculation.[30,31]

The additional consideration of the initial state is an important one in the context of dynamics simulations for both practical and theoretical reasons. On the practical side, some of the most efficient schemes for simulating chemical systems allow for both quantum and classical nuclei. However, they introduce the challenge of defining and preparing initial quantum states of both the electrons and the nuclei, which are no longer simple point charges in the system. Although preparing the exact initial state of interest may be difficult, recent work suggests that sampling over an ensemble of initial states can provide approximations to the thermal distributions of interest. Moreover, for ergodic systems with sufficiently long simulation times, this may not be an issue for error-corrected quantum devices. On the theoretical side, the simulation of a quantum system is formally promised to be efficient only if the initial state is efficient to prepare. While we expect this to be true for most natural physical systems, it remains an outstanding question in the field.

Quantum Computing Requirements

The quantum hardware requirements for simulating nonequilibrium dynamics differ greatly, depending on the approach used and the physical system of interest. In fact, in many cases, the exact quantification of the resources required remains an open question. In the case of hybrid analog and digital approaches, it may be possible, using recent sublinear depth algorithms[32,33] for the simulation of a driven lattice or Hubbard model (relevant to quantum materials), to reach dynamics of interest on a system with as few as ~50 qubits and a gate depth of ~10^3.

For early quantum computers, one restricted form of simulation is a hybrid-quantum classical exploration of nuclear dynamics. That is, the potential energy surface is discretized under the Born-Oppenheimer approximation, and the energies of the ground and excited states are calculated using a quantum computer at each nuclear configuration, yielding a more accurate energy surface than by existing means. Dynamics or alternative analyses could then be run on these surfaces using a classical computer. However, as the exact storage of these surfaces grows exponentially with the number of variables, the approach is most suitable for relatively low-dimensional reaction surfaces, such as simple dissociations. For higher-dimensional systems, a treatment with more coupling between electron and nuclear motion is needed, using either surface hopping or eventually full quantum dynamics on a quantum processor.

Finally, the exploration and operation of a quantum information processor by scientific users presents many challenges driven by the experimental nature of quantum hardware and the absence of the essential software needed to program the hardware. Software for quantum computing is in its infancy, and the development of executable code for quantum hardware using current strategies is an arduous and unsustainable enterprise, locking many potential users out of exploring and using this technology. Key for an end-user software platform is the development of a top-to-bottom open-source software platform for quantum computing.

Potential Impact

The control and simulation of nonequilibrium dynamics has the potential to solve a number of outstanding open questions at the interface of the chemical and materials sciences. In the case of catalysis, one such question is the identification of the active site; namely, where the reaction occurs and how many atoms are involved.[5] The ability to simulate chemical reactions in the condensed phase will pave the way to identifying active sites and to mapping the pathways that lead to catalysis, in turn opening the door to controlling and optimizing such pathways. Once pathways are identified and controlled, they may be used

to design catalysts for specific purposes.[34] Through careful engineering, pathway control may eventually enable designer catalysts for important reactions such as nitrogen fixation or carbon capture.[11] On the other hand, the quantum simulation of strongly driven systems also unlocks the ability to characterize entirely new materials and to stabilize nonstandard electronic states, which may not exist in traditional, equilibrium settings. These could include Floquet topological states as well as frustrated long-range systems.[12] Finally, the future use of a near-term quantum computer to prepare and analyze strongly entangled states can enable enhanced metrology, allowing for the direct imaging of nanoscale magnetic, electrical, and thermal properties (of both chemical reactions and quantum materials) while also optimizing the trade-off between sensitivity, spatial resolution, and detection bandwidth.

PRO 2: Unraveling the Physics and Chemistry of Strongly Correlated Electron Systems

Contributors: Thomas Maier, Bela Bauer, Gus Scuseria, Joel Moore, and Thomas Devereaux

Overview

Strong electronic correlations in molecules and solids give rise to complex many-body dynamics wherein electrons are entangled and multiple electronic states are nearly degenerate. This situation presents both opportunities and challenges. The opportunities range from new molecular magnets and nitrogen fixation technologies to novel superconducting, magnetic, and improved quantum information storage materials. The challenge is to understand and provide guidance in the design of this important class of systems.

To address this problem, quantum scientists have designed a myriad of methods to deal with strong correlations. Although much progress has been made in the understanding of correlated systems, classical approaches to the exact solution are fundamentally hampered by the exponential growth of the storage required to represent and manipulate the quantum many-body wavefunction on a classical computer. In many of these systems, the key orbitals and interactions are well understood and there is a simple unit cell or other repeated structure; but computational difficulty grows so rapidly with size that predictions are not possible for the materials or molecules of greatest interest.

By storing the state in entangled qubits, a quantum computer can avoid this problem. Quantum computing therefore promises to reduce the simulation complexity from exponential to polynomial in system size and thus presents a major opportunity in the simulation of strongly correlated molecules and solids. Although the large gate counts and low noise levels required for quantum simulations will likely restrict near-term applications to problem sizes hardly competitive with classical computation, current research on quantum hardware and algorithms promises to make important progress on these issues.

Simulations on quantum hardware thus provide a unique opportunity to advance our understanding of strongly correlated molecules and solids and address a number of questions that are out of the reach of present classical approaches. These include the origin of high-temperature superconductivity, the robustness of spin liquids, the magnetization behavior of molecular magnets, and the catalytic activity of nanoparticles. Progress in our understanding of these issues can lead to revolutionary advances in several important energy-related technologies.

Scientific Challenges

Theoretical modeling and simulation are essential elements in understanding the behavior of molecules and solids and thus guiding the search for systems with optimized properties. The progress made over the past decades allows for quantitative and predictive modeling of weakly correlated molecules and solids from first principles. These systems can be described starting from the standard electronic structure paradigm based on Hartree-Fock or DFT approaches, with many extensions made over the past decades. But systems in which electronic correlations are strong have successfully eluded such mainstream theoretical modeling.

The difficulty of accurately modeling strongly correlated molecules and solids arises from the fact that, in these systems, the residual correlation potential originating from the Coulomb interaction between the electrons is comparable in size to the terms retained in standard approximations. This competition produces complex many-body dynamics, in which particles behave collectively and regularly produce nearly degenerate states.

The family of copper oxide–based high-temperature superconductors provides a clear example of these principles in materials science.[35] The pseudogap phase at weak hole doping[36] from which superconductivity emerges at lower temperatures hosts a number of different states with charge, magnetic, and nematic order, possibly intertwined with a Cooper pair-density wave,[37,38] and their relation to superconductivity remains puzzling.

The families of magnetically frustrated spin systems also have rich phase diagrams in which the frustration of the magnetic interactions prevents the formation of long-range magnetic order.[39,40] This failure leads to many different magnetic states that lie close in energy. As a result, these systems are sensitive to small changes in pressure, strain, or magnetic field, which can switch the system between states.

In molecular systems, the same type of situation arises when correlations are strong. In this case, several states are nearly degenerate, and their correlation effects are critical for accurately describing the formation and breaking of chemical bonds. Conical intersections (regions of potential energy surfaces where electronic states are nearly degenerate) are another example of how even relatively small molecules present considerable challenges for current methods when dynamics are considered.

To address this complexity, researchers need to use methods that treat the relevant degrees of freedom on an equal footing. Techniques based on approximations of a single electron moving in a mean-field of all the other electrons, such as Hartree-Fock or DFT, cannot describe the strong entanglement of the electrons that underlies this behavior. For these cases, quantum chemists and condensed matter physicists have designed a myriad of methods to deal with the strong correlation problem. However, the exponential growth of the many-body Hilbert space with the number of degrees of freedom remains a limiting factor for these approaches; this complexity cannot be overcome with today's petascale and future exascale computing hardware.

Quantum algorithms executed on quantum computing hardware do not face this problem, in principle. They promise to reduce the simulation complexity for quantum many-body systems from exponential to just polynomial,[41] thus presenting a fundamental advantage over classical approaches and creating an exceptional opportunity in the simulation of strongly correlated molecules and solids. This speedup from exponential to polynomial is similar in spirit to the remarkable speedup of factoring by Shor's algorithm, a key theoretical development that helped drive quantum computing research.

Near-term opportunities are in providing high-quality benchmark data for molecules as small as a few atoms. For example, chemical accuracy (4 kJ/mol) has not yet been achieved for a relatively small benchmark system, Be_2,[26] and in simulating simple effective models of solids, such as the Fermi Hubbard model, on small lattice sizes. Such proof-of-principle applications will provide important information regarding the feasibility and utility of quantum computing in addressing grand-challenge scientific problems in the area of correlated molecules and materials. Another opportunity lies in the quantum computation of dynamical quantities of correlated systems, such as the single-particle spectral function or the inelastic magnetic neutron scattering spectrum. Computing these observables remains highly difficult for classical approaches.

Computational Challenges

The main computational challenge of classical approaches to the quantum many-body problem arises from the exponential growth of the Hilbert space. That is, the memory required to store and manipulate the quantum many-body wave-function on a classical computer increases exponentially with the number of degrees of freedom.

The coupled cluster method, the gold standard in quantum chemistry for weakly correlated systems, reduces this complexity to polynomial in system size.[42] But its predictions become very poor for strongly correlated molecules because of the lack of a dominating mean field. Quantum chemists have designed a myriad of methods to deal with this situation. Nevertheless, despite decades of efforts, techniques to deal with strong correlations either lack the polynomial computational cost as a function of size that coupled cluster theory enjoys, thus precluding their use on larger systems, or they are simply not accurate enough to be deemed paradigmatic. The quest for an accurate polynomial cost wave function ansatz for describing molecular strong correlations remains unabated.

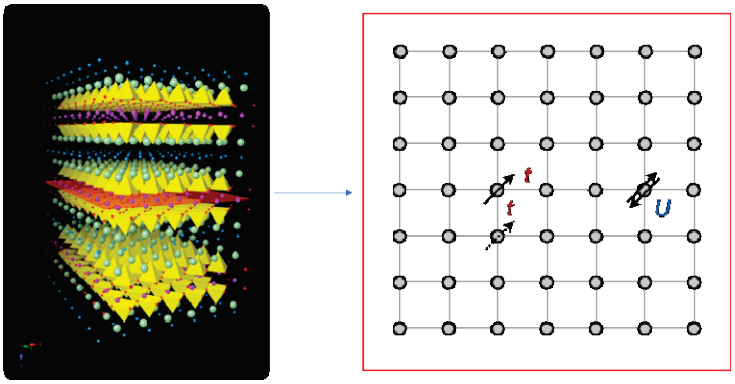


Figure 7. The Fermi Hubbard model provides a low-energy effective description of the copper-oxygen planes in cuprate high-temperature superconductors. It models electrons moving on a 2D square lattice with a nearest neighbor hopping amplitude *t* and a local (on-site) Coulomb repulsion for two electrons with opposite spins on the same site. This model is not integrable, and classical simulations are limited by the exponential growth of the Hilbert space. | Image courtesy of Thomas Maier, Oak Ridge National Laboratory

In condensed matter physics, the full Hamiltonian of a solid is first reduced to a manageable model Hamiltonian that describes the low-energy degrees of freedom. We focus here on cases in which the model Hamiltonian is already known; developing improved ways to extract model Hamiltonians from initial chemical/structural information is discussed later in this document. The Fermi Hubbard model[43] illustrated in Figure 7 is the most prominent example and is particularly relevant to the physics of the high-temperature superconducting cuprates.[44] Over the past decade, there has been significant progress in the development of numerical techniques for studying such models. Techniques ranging from determinantal quantum Monte Carlo (QMC)[45] and exact diagonalization[46] to DMRG,[47] tensor network methods,[48] and dynamical mean-field theory (DMFT)[49,50] provide complementary information on ground states and phase diagrams.

Similar to coupled cluster theory, QMC methods reduce the exponential complexity to polynomial. But this improvement is generally limited by the fermion sign problem, which arises from mapping the *d*-dimensional quantum problem to an equivalent $d + 1$-dimensional classical problem.[51] This problem gives rise to an exponential growth of the statistical error and hence the simulation time, effectively restoring the original exponential scaling for all but a few limiting cases.

The DMRG method is very efficient for 1D systems, but it suffers from a computational effort that scales exponentially with the system width owing to the increasing entanglement content of the wavefunction and the bias of this method toward states with low entanglement. More general tensor network algorithms such as projected entangled pair states or multiscale entanglement renormalization ansatz—which were in fact originally inspired by quantum information science—work directly in 2D; but they scale with a very high power according to the number of quantum correlations in the wavefunction.[48]

In addition, calculating the dynamic properties of a system, even in thermodynamic equilibrium—such as the single-particle spectral function measured in photoemission experiments, or the dynamic magnetic structure factor measured in neutron scattering—remains a challenge for classical approaches. For example, QMC methods do not work directly in real time, but rather in imaginary time; and the analytic continuation required to extract the real-time dynamics remains a key challenge in connecting theory with experiments,[52] a situation that provides additional motivation for PRO 1.

Nevertheless, considerable progress in the development of classical approaches over the last few decades has led to advanced simulation capabilities that will be challenging to surpass with quantum computing in the immediate future. Simulations of bigger or more complex systems on quantum hardware will require large numbers of qubits and gate operations. A quantum simulation of the ground state of the Fermi Hubbard model, for example, for lattice sizes competitive with those of classical computations, will require at least on the order of 100 logical qubits and millions of gates.[53] For materials science problems, quantum algorithms that simulate the system directly in the thermodynamic limit instead of on a finite-size real-space lattice could potentially circumvent this problem.

Another challenge for quantum computations of materials is the efficient preparation of an initial quantum state (the "ansatz") from which a variational quantum eigensolver (VQE)[54] tries to find the ground state of a given Hamiltonian by successively minimizing the expectation value of the Hamiltonian, i.e., the energy. This task is difficult, if not impossible, if the ansatz state does not have sufficient overlap with the true ground state of the system and if there are a number of nearly degenerate states. For example, if the true ground state wave function has superconducting order, but the initial state does not, this procedure will converge to a state that is not the true ground state of the system.

Quantum Computing Requirements

A number of quantum approaches are known for tackling the low-energy behavior of strongly correlated electron systems. For a recent nontechnical overview of the developments in the field of quantum chemistry, see an article by Mueck.[55] Most prominent is a direct simulation approach, akin to full configuration interaction in the chemistry context, or exact diagonalization in condensed matter. This approach was first described for chemistry applications in Aspuru-Guzik[3] and discussed in the condensed-matter context in Wecker et al.[53] Although the minimal number of error-corrected qubits required is relatively small, achieving high accuracy in this approach often comes with the challenge of relatively large gate counts of at least millions of error-corrected gates. Especially for chemistry applications, a large body of literature has attacked this problem, and researchers have reduced the gate counts drastically in many relevant cases compared with early estimates. However, the challenge still remains considerable for near-term quantum computing applications.

In the condensed matter context, a crucial challenge is that the problems exhibit more locality and are thus often more amenable to classical simulation, thus raising the number of qubits at which a quantum computer would exhibit a clear advantage. However, relevant cases become feasible at on the order of 100–200 qubits and 10^6 gates. An additional complication is that in many interesting cases, preparing the correct low-energy states may be difficult, since several competitive low-energy states may be very close to each other in energy. However, quantum computers promise a significant advantage for computing dynamical properties (such as spectral functions or dynamical structure factors), which are largely inaccessible to classical methods.

To go beyond the direct, full simulation of the problem, several alternative approaches have been discussed. A promising direction is the use of variational quantum algorithms.[54] These may considerably reduce the requirements for the number of gates that must be coherently executed in a single run and are naturally resilient to certain types of errors in the gate implementation. However, as these are heuristic approaches, they are chronically difficult to analyze without access to a quantum computer, making it hard to assess the number of gates that need to be coherently executed for a given problem to reach a desired accuracy.

An alternative approach is to embed a quantum computer as a solver in a classical embedding method, such as DMFT or the density matrix embedding theory. This approach has been proposed for complex materials, which defy a direct approach because of the large number of qubits that would be required.[56]

These approaches have the advantage that they work directly in the thermodynamic limit and can solve complex problems with a relatively small number of qubits; however, they require additional approximations that are difficult to control. Furthermore, the gate depth of the relevant circuits is relatively deep, requiring the coherent execution of about 10^8 gate operations.[56]

A central question is whether any relevant problems can be solved with noisy qubits, i.e., with high-quality physical qubits, but without error correction. Current experimental evidence suggests a skeptical outlook: for circuit depths beyond classical simulation capability, noise destroys all quantum advantages. However, as both the hardware and the understanding of the algorithms are likely to see drastic improvements, this conclusion may change. Some recent theoretical work has investigated how to solve these issues,[57,28] but it remains to be seen how these approaches can be brought to bear on practical calculations.

Potential Impact

Prototypical examples of molecular systems in which strong correlations are ubiquitous include the single-molecule magnet Mn_{12} acetate (see Figure 8) and related compounds, a case that exhibits an extremely slow relaxation of its magnetization below a blocking temperature. Another example is the iron-molybdenum cofactor (FeMoco) active site of nitrogenase, the enzyme that breaks the N_2 bond at room temperature. The current process for making ammonia from nitrogen uses high pressure and high temperature and consumes 1% of the world's energy. Accurate quantum calculations of strongly correlated enzyme active sites could unravel their secrets and lead to understanding that would provide the basis for important technological breakthroughs in catalysis and many other relevant areas. Design of novel molecular magnets could have a large impact on low-cost, environmentally friendly materials for storage and a myriad of other technological applications.

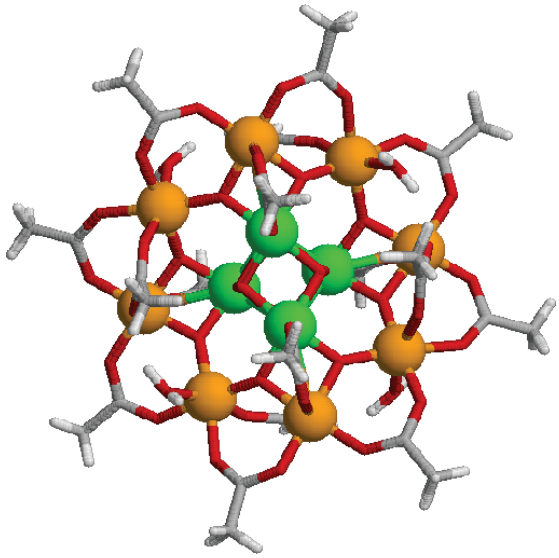


Figure 8. Mn_{12}-acetate single-molecule magnet. Manganese atoms with oxidation states of III and IV are in orange and green, respectively; oxygen is in red and carbon in black. | Image reproduced with permission from Jens Kortus from Molecular Magnetism

A successful quantum computation of the Fermi Hubbard model would have significant impact in condensed matter physics; this model is commonly believed to have the right ingredients to enable an understanding of high-temperature superconductivity in the cuprates. Although it is too simple for a quantitatively accurate description of these systems, it allows researchers to study the essential behavior arising from strong correlations, such as magnetism and superconductivity. Calculating the ground state and finite temperature properties of this model and understanding the mechanism(s) that give rise to these behaviors could lead to a detailed understanding of high-temperature superconductivity in the cuprates. It would allow definition of the minimal set of ingredients necessary to explain these phenomena. Furthermore, the knowledge gained would allow scientists to define a set of principles for designing systems with improved properties, and thus help guide the search for superconductors with higher transition temperatures that could potentially revolutionize US energy technologies.

Moreover, progress in the simulation of frustrated spin systems—such as the Heisenberg model on a Kagome lattice (Figure 9)—could significantly advance our understanding of spin liquids by providing

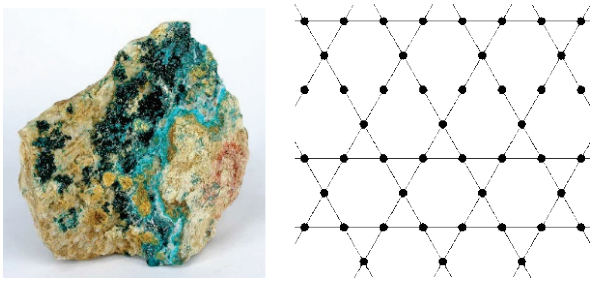


Figure 9. The mineral herbertsmithite (left) is composed of a lattice of copper ions sitting on a kagome lattice (right) that consists of corner-sharing triangles. This lattice structure frustrates the magnetism despite a strong exchange coupling and thus can host a quantum spin liquid state, in which spins do not order down to the lowest temperatures. The exact nature of this state and its competition with other more conventional magnetically ordered states is an active area of current research. | Image by Rob Lavinsky; reproduced from https://commons.wikimedia.org/wiki/File:Herbertsmithite-163165.jpg#filelinks under the terms and conditions of the Creative Commons CC BY license.

new insight into the competition between nearly degenerate low-energy states, including exotic spin liquids and more conventional magnetically ordered states. Elastic and inelastic neutron scattering can measure many properties of the magnetism that are currently beyond what can be accessed by numerical simulation. Such progress is important to understand the robustness of desired spin liquid ground states and thus could open new avenues in the experimental realization of quantum spin liquids. The spin liquid phase has been linked to unconventional superconductivity and may be useful for further advancing quantum computing research.

PRO 3: Embedding Quantum Hardware in Classical Frameworks

Contributors: Peter Love, Sue Coppersmith, John Preskill, Birgitta Whaley, and Marivi Fernandez-Serra

Overview

This section discusses computational approaches that merge quantum and classical pieces by passing information back and forth in real time, with applications to complex molecules and materials and to "open" quantum systems in which quantum orbitals interact with an effectively classical environment, such as a liquid. Applications of interest to BES include the development of complex, bio-inspired molecules with the potential for creating chemical energy from light (artificial photosynthesis) and the creation of new quantum materials, ranging from improving the understanding of defects that limit the scaling of current computers to ever smaller sizes, to new families of magnets and high-temperature superconductors.

Many quantum chemistry methods are efficient because they separate a problem into a relatively easy part that can be treated with computationally inexpensive approaches, and a "hard" or strongly correlated part that can be treated only with more expensive techniques. For example, organic molecules and transition complexes containing only one or two transition metal atoms are well modeled by the current generation of ab initio quantum chemistry methods. These methods—such as complete active space perturbation theory second order (CASPT2)[58] or n-electron valence perturbation theory second order (NEVPT2)[59]—provide an ab initio, systematically improvable framework that describes weakly correlated, delocalized, s- and p- electrons by an approximate and computationally inexpensive method, while the strongly correlated, spatially confined, d-electrons are modeled at a higher level of theory.

Although these methods remain valuable, once the number of transition metal atoms exceeds two, the difficulty of simulating these transition metal–rich compounds quickly becomes insurmountable because of the exponential growth of the Hilbert space necessary to model the strongly correlated electrons.[46] The preceding PRO concentrated on cases in which the strongly correlated problem is known from experiment or an initial one-shot classical calculation, often because there is a simple unit cell or repeated structure. However, in most compounds, there is a need to embed algorithms that could pass information back and forth between quantum and classical pieces in real time and achieve self-consistency.[60]

Such hybrid algorithms would allow major progress on complex transition metal–containing molecules that are of great fundamental and applied interest. In the past decade, the adaptation of the DMRG[61,62,63] to chemical systems allowed accurate calculation of systems with up to three and four transition metal atoms. However, ab initio evaluation of systems with larger numbers of transition metal atoms currently remains completely inaccessible. Thus, active centers of biologically important enzymes, such as FeMoco[‡], which is the primary cofactor of nitrogenase in nitrogen fixation, or the Mn_4O_5Ca core of the oxygen evolving complex[§] (see Figure 10), remain far beyond the current capabilities of ab initio quantum chemical methods.

[‡] See https://en.wikipedia.org/wiki/FeMoco.
[§] See https://en.wikipedia.org/wiki/Oxygen-evolving_complex.

Scientific Challenges

For these strongly correlated large molecules, and for solids for which a simple model Hamiltonian such as the Hubbard model is unavailable, finding a systematically improvable and accurate description of the physical ground state remains far beyond the capabilities of current quantum chemistry methods. Spintronic and battery applications of correlated solids also involve not only the description of the ground state but additionally the description of multiple excited states, particularly to model function at nonzero temperatures. Currently, the description of temperature effects beyond a few lowest-lying excited states cannot be done at the ab initio level. The lack of such a description, beyond the smallest possible cases, inhibits finding new families of high-temperature superconductors and creates problems for above-room-temperature materials applications such as those appearing in nuclear energy applications and warm dense matter.

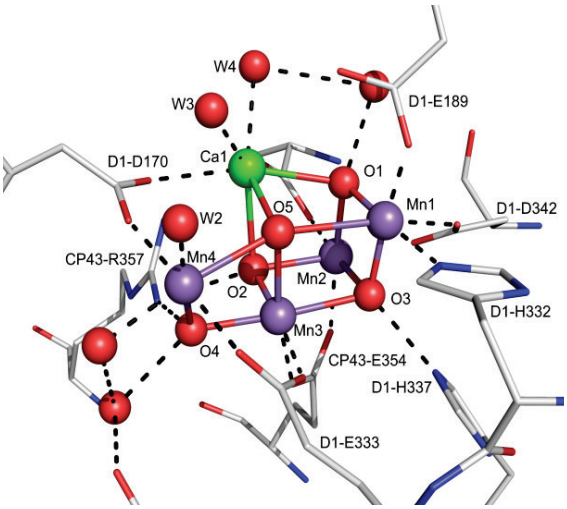


Figure 10. Crystal structure of the oxygen evolving complex of Cytochrome P450 at 1.9 Å resolution. Multiple manganese and oxygen atoms make the description of such an enzymatic center very challenging for traditional methods. | Image reproduced from commons.wikimedia under the terms of the GNU Free Documentation License.

Similarly, simulations of quantum dynamics and open dissipative systems are frequently beyond the reach of traditional methods in quantum chemistry and condensed matter. The quantum simulation of sequential absorption of single photons, subsequent energy transfer, and charge separation that is required to probe these quantum dynamics within a realistic model of photosystem II (see Figure 11) is well beyond the capability of current classical computers and represents a substantial challenge for quantum computers.

In all the aforementioned examples, the common feature that renders the current generation of ab initio methods unsuitable is the growth of the Hilbert space necessary to describe the strongly correlated part of the problem. To overcome this difficulty in both the quantum chemical and condensed matter communities, multiple embedding methods were designed. In an embedding method,[49,64,65] all electrons present in the system are separated into "easy to describe" weakly correlated electrons and "hard to describe" strongly correlated electrons. The weakly correlated electrons are treated by a low-level, computationally inexpensive method with sufficient accuracy. The

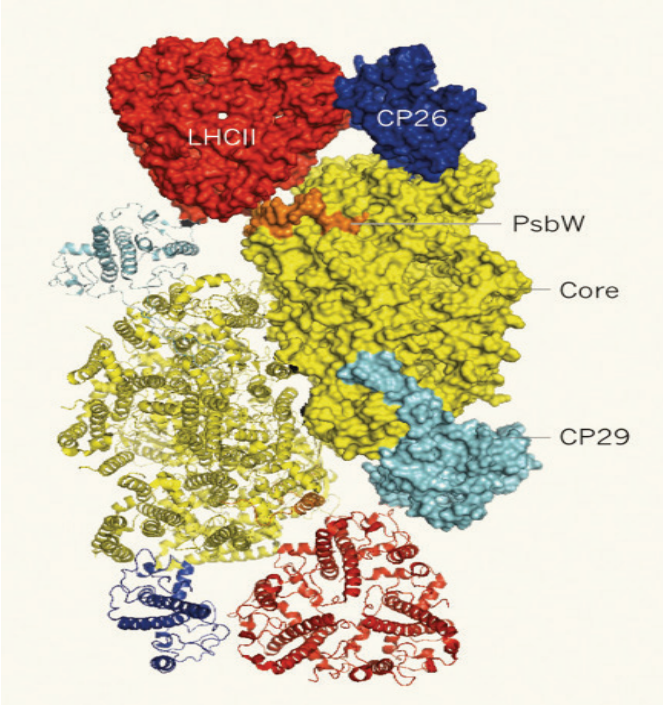


Figure 11. The core complex contains the reaction center (not shown), and the peripheral light-harvesting complexes (LHCII, CP29, and CP26) supply excitation energy to the reaction center. PsbW is a core subunit typical of plants that mediates the association of LHCII with the core | Nature Publishing Group. *Nature*, Structure of spinach photosystem II: LHCII supercomplex at 3.2 Å resolution, P. Wei et al., copyright 2016

description of the strongly correlated electrons requires solving a difficult problem for which the size of the Hilbert space grows exponentially. Furthermore, the behavior of the strongly correlated electrons influences the weakly correlated part of the problem, e.g., by modifying the screening of interactions; thus a self-consistent solution typically requires iteration and the passage of information between weakly correlated and strongly correlated pieces. Consequently, embedding methods that initially were developed for classical computers can be adapted for use in quantum computations, because the part of the calculation involving strongly correlated electrons can be successfully run on a quantum computer. For details, see Figure 12. It is reasonable to envision that in the near future, when machines with around 50–100 qubits become available, embedding calculations could become a dominant technique in the quantum chemistry and condensed matter communities.

Computational Challenges

For the foreseeable future, quantum computers will need to interface intimately with classical computers and software that control the computation and the flow of input data and output results. This is especially true in the short term while quantum computers lack the number of qubits, robustness, and functionality required to run complete simulations. Although rigorous theoretical results underpin our expectations of the benefit of quantum simulation, we anticipate that many practical computations will have to step beyond these proofs and employ algorithms that initially have limited formal proofs.

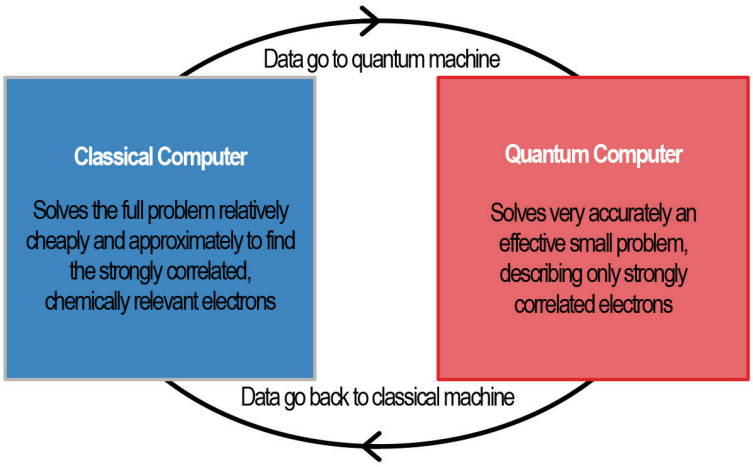


Figure 12. An example workflow between classical and quantum machines. Embedding methods developed for classical computers can be easily adapted to this mixed quantum classical computation scheme. | Image courtesy of Dominika Zgid, University of Michigan

At present, the main computational challenge is thus to design and optimize classical algorithms that are suitable for possible future hybrid quantum classical computations. Currently, there are several classes of classical algorithms that would benefit from the interface with quantum computation/algorithms. Examples include

- Optimizing (e.g., stochastic) of parameters in the quantum simulation, including its initial state
- Searching for (e.g., by combinatorial chemistry) dynamics of molecules and materials driven by results of quantum simulations
- Interfacing with big data analytics, e.g., feeding the results of quantum simulations into classical machine learning, or taking real-world data (e.g., from BES light sources and microscopes) and feeding them into quantum machine learning algorithms, or otherwise leveraging the vast databases of scientific data of modern science
- Physical embedding (e.g., using DMFT,[49] self-energy embedding theory [SEET[64]], and density matrix embedding theory[65]) of quantum simulations of finite systems into models capable of treating extended complex systems at finite temperatures.
- Studying open quantum systems (a quantum system interacting with a classical environment)
- Simulating time evolution in complex molecules, building on PRO 1

Another challenge for current quantum chemical methods on classical computers is creating a series of basis sets that are suitable for quantum computers. Most of the current quantum computations are performed within very small basis sets. It is conceivable, but far from certain, that in the near future, good basis sets will be formed in which the number of orbitals can be kept small while good accuracy is maintained. In the long term, basis sets that can approach completeness in a systematic manner while being sufficiently well conditioned are highly desirable. Such bases are very important for extended systems and in systems with many length scales.

In the current form, the quantum computer is more efficient when Hamiltonians are sparse and the number of required two-body Coulomb integrals is small. Learning how to prepare and optimize Hamiltonians and initial states that are suitable for efficient quantum simulations will certainly constitute a short-term challenge for the current generation of classical quantum chemistry methods.

Finally, although the possibility of interfacing classical and quantum simulation is exciting, further developments are necessary to create a unifying formalism that can first be tested on classical computers and that is compatible for molecules, solids with periodic structure, temperature-dependent problems, and nonequilibrium problems. For example, formalisms based on Green's functions not only are general enough to access a broad class of systems but also could predict both equilibrium and nonequilibrium processes.

Quantum Computing Requirements

Since, in the short term, results from quantum simulation will be inherently noisy, robust schemes must be developed that will couple the classical and quantum computations and give accurate answers without an accumulation of errors from the noise. Existing computational classical embedding methods such as DMFT, density matrix embedding theory, and SEET are already capable in principle of using QMC solvers that yield noisy data.[66] However, there should be additional development of algorithms and a careful evaluation of their noise robustness.

Moreover, current calculations on quantum computers mostly focus on evaluating electronic energy. When a classical–quantum hybrid approach is used, quantum computers should be able to evaluate first-, second-, and higher-order density matrices or Green's functions. A major short-term advantage would be to use quantum computers of a few hundred qubits to evaluate high-level density matrices in quantum chemistry calculations such as CASPT2 or NEVPT2. Similarly, quantum computing algorithms should be optimized to evaluate Green's functions in quantum impurity problems, since the evaluation of these problems is a bottleneck for current classical methods such as DMFT or SEET. The outputs of the quantum simulation, such as density matrices and Green's functions, will then be fed into classical algorithms that might leverage existing implementations of these approaches. The main challenge is to evaluate these quantities on a quantum computer with sufficiently small noise.[57,28]

A separate set of quantum computing requirements is present for quantum dynamics and quantum simulation of open systems. The major computational challenge is to develop efficient propagators for open quantum systems such as LHCII (see Figure 11). Doing so requires simulation of both unitary and dissipative evolution, for both localized "site" excitons and separated charge transfer states. Several components are involved. First these states, which are of variable spatial locality, need to be mapped to a qubit representation of well-defined connectivity.[67] Then either digital (stroboscopic) or continuous Liouvillian propagators need to be developed. For Markovian systems, a number of such schemes exist, some of which have been implemented experimentally. For example, Bell and GHZ states have been prepared as steady states of digital evolution in an ion trap experiment using stroboscopic cycling of sequences of unitary and dissipative terms, where the latter were realized by the addition of ancilla states

and optical pumping.[68] Fewer options are known for propagation in quantum simulation of non-Markovian systems,[69,70] and further development of efficient Liouvillian propagators is desirable.

Another challenge will be handling noise encountered as a result of decoherence of the quantum system, as well as at the classical–quantum interface. Instead of treating quantum decoherence as counter-productive, there is an opportunity to engineer the coupling between the classical and quantum systems. Doing so would allow for controlled dissipation to reach computationally relevant states, as well as for preparing targeted thermal and mixed states.

Finally, the last challenge for classical–quantum hybrid machines will lie in reproducibility, verification, and validation. To advance classical–quantum hybrid machines, a community agreed-upon set of approaches and a consistent set of results/benchmarks is necessary to verify and validate the results of hybrid simulations. Such simulations introduce new types of errors (e.g., model, systematic, statistical, experimental, technological) that are not commonly found in classical simulations. It took a concerted community much effort over decades to develop effective approaches for classical simulation, and there are still many lingering questions about reproducibility. We must address this problem head-on and early in the life of classical–quantum simulations.

Potential Impact

In the long term, quantum computation promises fully predictive simulations of many highly important chemical and material systems, with the same or greater ease with which researchers now use approximate density functional methods that have no systematic basis for controlling error or improving accuracy. The revolutionary advances that quantum computation promises will translate into new materials and technologies that will form the basis of new industries and have a profound impact on our economy and nation. For instance, computational insights into spin-crossover complexes and magnetic ad-atoms on surfaces can be used to optimize future nano-devices. Predictive optimization of strongly correlated active centers of enzymes can be used to advance medical research, in addition to the catalysis and photosynthesis advances mentioned earlier.

It is also expected that entirely ab initio prediction of new classes of high-temperature superconductors will be possible. New superconductors could have a tremendous impact on energy losses in electricity transmission and other applications. Since hybrid classical–quantum simulation will include all orbitals present in the realistic system (not just a model system), we can expect that chemical and physical changes controlling superconductivity will be understood and used in the design of future high-temperature superconductors. The predictive nature of hybrid classical–quantum simulations should also be helpful in simulation of phase diagrams of actinides, a complex materials class of DOE interest.

Finally, the possibility of simulating open dissipative systems as well as time evolution (see PRO 1) should provide an essential theoretical component that can guide many experiments dealing with nonequilibrium processes that are notoriously hard to interpret. In summary, the major impact of classical–quantum computations will lie in delivering a series of modern, systematically improvable tools that can be used in both quantum chemistry and quantum materials. We think that these tools will become essential in understanding the physics phenomena present in new materials, as well as enabling the controlled design of materials that are industrially relevant.

PRO 4: Bridging the Classical–Quantum Computing Divide

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Overview

Quantum computation faces the daunting challenge of outperforming 70 years of exponential growth of classical computational power reflected in Moore's Law. In the near term, by which we mean 50 to 100 physical qubits, quantum computers may pass the threshold of "quantum supremacy"—meaning they themselves cannot be simulated by any existing, or conceivable, classical computer. A more stringent condition is for a quantum computer to solve a useful and interesting computational problem that classical computing cannot solve, and eventually to access grand-challenge science.

However, classical and quantum computing do not exist only in competition. PRO 3 discussed the real-time exchange of information between classical and quantum hardware in hybrid algorithms. This section considers *bridging* between classical and quantum computing—for example, cases in which the strengths of quantum algorithms can be used to improve the key inputs of classical algorithms or to capture key quantum processes in "open systems" in which decoherence by coupling to the environment is present. In other words, PRO 3 discusses hybrid or co-processor computing, while PRO 4 discusses integrating results of separate quantum and classical computations to solve scientific challenges. Open systems appear across chemistry and materials science, including technologically important examples such as solid-liquid interfaces and optical absorption in liquid backgrounds. Such approaches also have the potential to scale up as the ratio of quantum to classical resources used grows over time.

Scientific Challenges

Our PROs are in the areas of quantum chemical and materials sciences. Specifically, this PRO considers the improvement of exact solutions for small instances of the electronic structure problem and the improvement of DFT for larger problems such as clusters of water molecules.

The first example is the determination of exact electronic energies for particularly difficult cases of the electronic structure problem. We call these *electronic structure challenge problems* that are defined by the failure of classical approximate methods. This means that (1) they must exhibit large correlation energy so that self-consistent field approaches fail; (2) they must exhibit a severe fermion sign problem so that QMC fails; and (3) coupled-cluster or similar gold standard correlation approaches must not yield the desired accuracy. In addition, they must present a sufficiently severe multi-reference character to confound full configuration interaction QMC approaches. Clearly, these conditions mean that small example problems will be few in number, and the best classical approaches may require high-accuracy solutions. A bridging approach uses the best classical results as the starting point for a quantum variational method. The quantum resources are used to broaden the space of possible ansatz states available to the variational method and can therefore improve upon the best classical approach.

The second example is the quantum simulation of small clusters of water by VQE. High-accuracy simulations of these systems can inform density functionals that enable DFT to better treat the structure of water in a wide variety of problems. The accuracy requirements are not expected to be as stringent, but the necessity to treat a cluster will require more orbitals for representation of the problem. Such a challenge problem for smaller (50–100 qubit) quantum computers will help investigate whether the best quantum solution on a smaller basis is better than the classical solution on a larger basis. An improvement is possible owing to the higher quality of solution available by quantum means, but is by no means guaranteed. Such problems therefore represent an interesting short-term test case for these methods.

The search for new density functionals that are capable of accurately describing different phases of matter, and simultaneously do not compromise on simulation performance with respect to system size and length scale simulations (in ab initio molecular dynamics approaches), is currently on the rise. The search is motivated by the availability of very accurate experimental data and by the push of machine learning and data science–motivated approaches to fitting complex functions. The parameterization and optimization of complex models is a pervasive problem in many areas, particularly in the development of new density functionals optimized to treat specific systems. It has been shown that—using highly accurate quantum chemical calculations—a standard, generalized gradient approximation and a van der Waals-type of density functional can be optimized to describe liquid water, or general systems with accuracy close to that of the data to which it was fitted (Figure 13).[71,72] This approach would certainly benefit from the availability of even more "exact" data, not only total energies but also density matrices and possibly other observables that can be directly computed using DFT. Such sets of data are directly accessible using quantum computers but are not obtainable with direct experiments; hence they represent the exact numerical experimental results. Combining machine learning with density functional development (including functionals of the density matrix, or other nonstandard approaches) or advancing on the systematic development or optimization of more accurate density functionals[71–73] will enormously benefit from quantum simulation results. The data availability would accelerate and possibly give birth to new ideas for optimization and data fitting, in addition to providing new physical and chemical insight from which new theories or models will emerge.

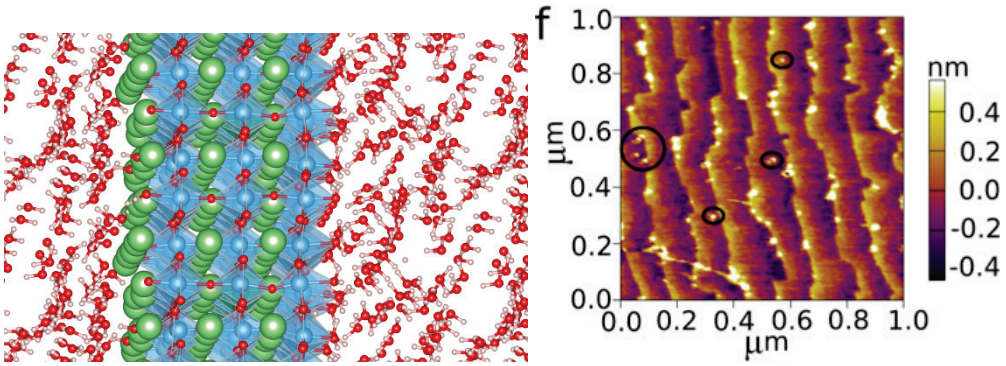

Figure 13. An accurate and fully quantum mechanical description of liquid water will have an impact in many BES priority areas. In the picture, the catalytic properties of $SrTiO_3$ surfaces are strongly coupled to their interactions with ambient water, and a full quantum mechanical atomistic simulation (left) is needed to interpret experimental results (right, atomic force microscope image of $SrTiO_3$ under water after irradiation by ultraviolet light). | Images courtesy of M. V. Fernandez-Serra (left) and M. Dawber (right), both of Stony Brook University.

The techniques to be employed on quantum hardware are VQE with two possible types of states for the initial ansatz that is improved by the quantum computation. Both the coupled-cluster ansatz and tensor network state ansatz will be employed. Both of these can be regarded as gold standard methods for electronic structure methods. Both, fortunately, admit a straightforward route to marry the best classical solutions to quantum algorithms that can improve those solutions.

The use of coupled cluster in VQE has already been investigated theoretically, and it is quite natural to take a coupled-cluster ansatz obtained by classical means and use the parameters of the classical ansatz to create a short quantum circuit producing the corresponding ansatz for the quantum computer. One can therefore use this as a starting point for the VQE solver, and any enhancement of the space of variational wavefunctions will cause the variational estimate of the energy to improve (decrease). In an arms race between classical coupled-cluster and quantum VQE methods, a user is therefore able to transfer any classical improvements immediately to the quantum case. The only possibility of failure is that the

classical method may be able to solve larger problems (with more orbitals) than there are qubits available. As qubit resources grow, the quantum methods must therefore win out.

Similarly, the use of DMRG, matrix-product, and tensor network methods is widespread in condensed matter physics, materials science, and chemistry. These methods suffer from exponential scaling with bond dimension on classical computers, but not on quantum computers. Again, a best-in-class classical tensor network approximation could be used as a starting point for a quantum variational calculation.

Computational Challenges

In the case of variational approaches to electronic structure problems, quantum hardware can access a larger space of variational states than is possible classically. As discussed earlier, the best-case classical variational states can be used as a starting point for such quantum variational calculations. In principle, this means that the use of quantum hardware can only improve upon (or in the worst case, match) classical variational algorithms. In practice, the efficacy of such approaches will be determined by the ability to optimize an ansatz on quantum hardware with more parameters than in the classical case. Therefore, it is worth considering the possibility that quantum approaches to optimization may enable taking a further step forward using VQE-based algorithms.

Optimization is a vast field of numerical analysis that is hugely important in many areas of science and technology. One important application relevant to BES is optimization of time for processing chemical and material transformations. Time-optimal control of processes such as chemical reactions; materials processing; engineering of control of quantum systems; and larger-scale manufacturing processes that rely on exterior features such as workflow, logistics, and transportation requires the optimization of a function that is generally highly nonlinear in the parameters. Minimization of the time to target can mean significant savings in resources and can often increase energy efficiency.[74]

Another key application for BES is the variational optimization of energies for electronic structure calculations. Hybrid classical–quantum computation schemes for electronic structure calculations, such as VQEs, which have been proposed for near-term quantum computers, rely on the ability to perform efficient classical optimization—for example, quantum gate parameters for state initialization, which are potentially off-line from the quantum aspects of the calculation (state preparation and Hamiltonian tomography). However, while the Hamiltonian is 2-local (i.e. has pairwise interactions, but these may be long-range), the complexity of the classical optimization step rises rapidly with the number of qubits on account of the growth in number of gate parameters. Thus, any demonstration of quantum enhancement in optimization algorithms would be greatly impactful. Optimization tasks are also intimately related to questions of coherent control, and control theory provides an additional perspective for physics-inspired approaches to optimization. For example, hybrid schemes such as VQE can be regarded as examples of open loop learning algorithms.

Recent developments in quantum algorithms have provided key components that might enable such an enhancement. First, a quantum algorithm recently proposed for semidefinite programming shows exponential speedup in certain instances, in particular, when the input matrices are mapped to quantum states of low-rank Hamiltonians.[75] This finding has significant implications for all convex optimization problems, which constitute a very large subgroup of optimization instances, as well as for any problem that can be mapped onto sequential convex optimization steps in an iterative procedure. Second, a recently proposed quantum algorithm for gradient calculations[76] shows a significant speedup in efficiency and an improvement in complexity, implying a significant quantum advantage for the performance of optimization calculations in large-scale systems. Additional developments in coherent control theory for quantum systems are showing progress in techniques to search for the global optimal solution via analysis

of the typically high-dimensional control "landscape," i.e., the topographic map of the quantum metrics for successful control as a function of the control parameters.

Together with the developing capability to perform quantum Hamiltonian simulation, these developments suggest that a proven quantum advantage in simulation—combined with quantum speedups in convex optimization and/or quantum gradient calculations—could enable a significant speedup for many-variable optimization problems, particularly those that can be cast in convex form. The time optimal control problem mentioned earlier is one such problem, in which a nonlinear time evolution might be either linearized or made quadratic over small time scales and the local optimization then implemented by a quantum semi-definite programing algorithm.[77] From a fundamental perspective, this problem is highly important because one might thereby find a route to the global optimum of the control landscape.[78] From a practical perspective, an advance of this type would be highly valuable for two application directions of interest to BES: time optimal control of processing and variational hybrid classical–quantum approaches to electronic structure of molecules and materials.

Quantum Computing Requirements

In the case of VQEs, the requirements in terms of qubits can be quite modest—simulations in the quantum supremacy regime of 50–100 qubits are expected to be comparable to the best classical approaches. An example application is given by the proposed simulation of the uniform electron gas—Jellium—by the Google group.[79] In general, for VQEs, the constraint will not be the number of qubits (once the threshold of direct classical simulatability is crossed at around 45–50 qubits). In these algorithms, the number of gate operations or measurements required can easily grow to astronomical numbers.[80] Rapid repetition of measurements or large parallelism of many small independent quantum devices may therefore be necessary.

Potential Impact

In the next 5 years, quantum computers will cross a threshold beyond which classical simulation of the devices will become impossible. Although such quantum supremacy demonstrations are of intrinsic scientific interest, to be truly impactful, this next generation of quantum hardware needs to be applied to addressing problems of grand-challenge science. The bridging approach proposed in this report is one way to reach that goal. Using quantum techniques such as VQE that can improve upon best-in-class classical results should enable researchers to address interesting scientific questions. The electronic structure challenge problems and improvement of water functionals would have significant impact and help establish quantum simulation of materials properties as a reality in the 21st century.

3. Crosscutting Computational Themes and Challenges

Contributor: James Whitfield

The BES Roundtable was successful in identifying four PROs, which are discussed in the preceding chapters. During the roundtable, it became clear that several computational aspects appear in multiple PROs. The crosscutting aspects identified by the roundtable participants are error correction or robustness in the presence of quantum noise, software stack developments, and the validation of quantum simulation results.

Quantum decoherence is responsible for the emergence of classical physics from an underlying quantum system. Since all of the PROs attempt to exploit quantum effects for enhanced computations, it is important to understand and mitigate these decoherence effects. There are two broad ways to fight against the effects of decoherence: fault-tolerant quantum error correction[81] and error mitigation techniques. The first of these two methods has seen continuous development since the 1990s; however, full fault tolerance requires significant qubit overheads that will require significant technological development before such methods become standard. Given the status of current and near-future quantum hardware, requiring full-fledged quantum error correction would greatly hinder applications in the foreseeable future. Consequently, the second option of developing error mitigation will be an important scientific driver in the near future. Error suppression techniques and intermediate levels of quantum error correction in various experimental setups can optimize quantum hardware utilization. In addition, the design of pre-/post-processing filtering is expected to enhance quantum computing noise robustness.

The second crosscutting theme, software stack development, refers to computational tools and software packages needed to assist scientists with minimal backgrounds in quantum computing to set up and execute quantum simulations. Since the technology is still being developed, the concept of the stack is not completely defined. Ideally, the software tools should maintain a user-friendly abstraction level while compiling inputs to experimental pulses/gates/controls of quantum devices. Doing so will require maintaining hardware backend interfaces, useful encapsulations going up and down stack, and the flexibility to integrate new quantum algorithms into software stacks. Good integration of optimal classical and quantum methods will build the quantum computing user base and create the needed infrastructure for the applications outlined in the PROs.

The third theme identified that overlaps with each of the PROs is the scientific challenge of validating the output of quantum simulations. It will be important to establish the correct metrics for quantum simulations/measurements. Examples include local observables, correlation functions, and the fidelity of the computed wave function. The specific metric will ultimately be determined by the specific quantum computing application and the available hardware resources. Even for small quantum systems, full tomography of the wave function is expensive. Hence, near-term quantum validation will likely focus on test cases from solvable models (e.g., the 1D Hubbard model), well characterized molecular test sets, and high-quality benchmarks (for a small representative collection see Lynch and Truhlar[82]). Based on the current pace of development, quantum computation technology will soon possess computational power that exceeds that of direct classical computational simulation. Hence, alternative validation schemes will be needed. Theoretically, a number of works explore the validation of quantum algorithms without requiring a classical numerical reference. Blind quantum computation and interactive proofs[83,84] have been studied in recent years but have not yet been translated to realistic quantum simulations.

Significant progress in these three crosscutting computational themes will be a key enabler of the research identified in the PROs. Another important enabler mentioned by several participants is increased collaboration among quantum computing experts and theoretical chemists and materials scientists. At the moment, these groups tend to attend separate scientific meetings and to supervise distinct groups of junior

researchers. Accomplishing grand-challenge science with quantum computers will require a concerted effort from both computing and applications communities.

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Appendix A: Workshop Participants

BES Roundtable on Opportunities for Basic Research for Quantum Computing in Chemical and Materials Sciences

Chair: Joel Moore, Lawrence Berkeley National Lab/University of California–Berkeley, and Alán Aspuru-Guzik, Harvard University

PRO 1: Enabling and Solving Non-Equilibrium Quantum Dynamics
Giulia Galli, University of Chicago, Argonne National Laboratory: Lead
Norman Yao, Lawrence Berkeley National Laboratory/University of California–Berkeley
Wibe (Bert) de Jong, Lawrence Berkeley National Laboratory
Thomas Devereaux, SLAC National Accelerator Laboratory
Jarrod McClean, Google

PRO 2: Unraveling the Physics and Chemistry of Strongly Correlated Electron Systems
Thomas Maier, Oak Ridge National Laboratory: Lead
Bela Bauer, Microsoft
Gus Scuseria, Rice University
Joel Moore, Lawrence Berkeley National Laboratory/University of California–Berkeley
Thomas Devereaux, SLAC National Accelerator Laboratory

PRO 3: Inercalated Quantum and Classical Approaches
Peter Love, Tufts University: Lead
Sue Coppersmith, University of Wisconsin
John Preskill, CalTech
Birgitta Whaley, University of California–Berkeley
Marivi Fernandez-Serra, Stony Brook University

PRO 4: Hybrid Quantum–Classical Methods
Dominika Zgid, University Michigan: Lead
Robert Harrison, Brookhaven National Laboratory/Stony Brook University
Antonio Mezzacapo, IBM
James Whitfield, Dartmouth University

Crosscutting Themes
James Whitfield: Lead

Invited Participants

Alán Aspuru-Guzik, Harvard University
Bela Bauer, Microsoft
Sue Coppersmith, University of Wisconsin
Wibe (Bert) de Jong, Lawrence Berkeley National Laboratory
Thomas Devereaux, SLAC National Accelerator Laboratory
Marivi Fernandez-Serra, Stony Brook University

Giulia Galli, University of Chicago, Argonne National Laboratory
Robert Harrison, Brookhaven National Laboratory/Stony Brook University
Peter Love, Tufts University
Thomas Maier, Oak Ridge National Laboratory
Antonio Mezzacapo, IBM
Jarrod McClean, Google
Chris Monroe, University of Maryland, IonQ
Joel Moore, Lawrence Berkeley National Laboratory/University of California–Berkeley
John Preskill, CalTech
Gus Scuseria, Rice University
Birgitta Whaley, University of California–Berkeley
James Whitfield, Dartmouth University
Norman Yao, Lawrence Berkeley National Laboratory/University of California–Berkeley
Dominika Zgid, University of Michigan

Invited Observers

Kramer Akli, DOE Office of Science, Fusion Energy Sciences
Dimitri Argyriou, Ames Laboratory
David Awschalom, University of Chicago/Argonne National Laboratory
Paul Bayer, DOE Office of Science, Biological and Environmental Research
Jeff Blackburn, National Renewable Energy Laboratory
Steve Binkley, DOE Office of Science
Tof Carim, DOE Office of Science, High Energy Physics
Lali Chatterjee, DOE Office of Science, High Energy Physics
Hans Christen, Oak Ridge National Laboratory
Claire Cramer, DOE Office of Science, Advanced Scientific Computing Research
David Dean, Oak Ridge National Laboratory
Jim Davenport, DOE Office of Science, Basic Energy Sciences
Jonathan DuBois, Lawrence Livermore National Laboratory
Chris Fecko, DOE Office of Science, Basic Energy Sciences
Bruce Garrett, DOE Office of Science, Basic Energy Sciences
Matthias Graf, DOE Office of Science, Basic Energy Sciences
Barbara Helland, DOE Office of Science, Advanced Scientific Computing Research
Craig Henderson, DOE Office of Science, Basic Energy Sciences
Linda Horton, DOE Office of Science, Basic Energy Sciences
Jim Horwitz, DOE Office of Science, Basic Energy Sciences
Helen Kerch, DOE Office of Science, Basic Energy Sciences
Robert Konik, Brookhaven National Laboratory
Refik Kortan, DOE Office of Science, Basic Energy Sciences
Jeff Krause, DOE Office of Science, Basic Energy Sciences
Harriet Kung, DOE Office of Science, Basic Energy Sciences
Peter Lee, DOE Office of Science, Basic Energy Sciences
Eliane Lessner, DOE Office of Science, Basic Energy Sciences
Ramana Madupu, DOE Office of Science, Biological and Environmental Research
John Mandrekas, DOE Office of Science, Fusion Energy Sciences
George Maracas, DOE Office of Science, Basic Energy Sciences
Gail McLean, DOE Office of Science, Basic Energy Sciences
Raul Miranda, DOE Office of Science, Basic Energy Sciences
Rick Muller, Sandia National Laboratories
Jim Murphy, DOE Office of Science, Basic Energy Sciences
Mick Pechan, DOE Office of Science, Basic Energy Sciences
Mark Pederson, DOE Office of Science, Basic Energy Sciences

Jim Rhyne, DOE Office of Science, Basic Energy Sciences
Tom Russell, DOE Office of Science, Basic Energy Sciences
Jim Rustad, DOE Office of Science, Basic Energy Sciences
John Sarrao, Los Alamos National Laboratory
Andy Schwartz, DOE Office of Science, Basic Energy Sciences
Tom Settersten, DOE Office of Science, Basic Energy Sciences
Jim Siegrist, DOE Office of Science, High Energy Physics
Panagiotis Spentzouris, Fermi National Accelerator Laboratory
Peter Sushko, Pacific Northwest National Laboratory
Ceren Susut-Bennett, DOE Office of Science, Advanced Scientific Computing Research

Appendix B: Workshop Agenda

BES Roundtable on Opportunities for Basic Research for Quantum Computing in Chemical and Materials Sciences

Gaithersburg Marriott Washingtonian Center • October 31 – November 1, 2017

Tuesday, October 31, 2017	
1:00 – 1:30 p.m.	Registration — Lakeside 1 and 2
1:30 – 2:00 p.m.	**Welcome from DOE and Roundtable Charge** Harriet Kung, Associate Director of Science for Basic Energy Sciences
2:00 – 2:15 p.m.	Introduction of Participants
2:15 – 2:30 p.m.	**Opening Remarks** Joel Moore, Lawrence Berkeley National Lab/University of California–Berkeley
2:30 – 3:10 p.m.	**Keynote Presentation** John Preskill, California Institute of Technology (Introduction by Alán Aspuru-Guzik)
3:10 – 3:20 p.m.	**Break — Refreshments Available**
3:20 – 4:00 p.m.	Opportunities for Quantum Computation in Chemistry: Perspective by Alán Aspuru-Guzik (includes overview of homework and time for discussion)
4:00 – 4:40 p.m.	Opportunities for Quantum Computation for Materials: Perspective by Joel Moore (includes overview of homework and time for discussion)
4:40 – 4:50 p.m.	**Break**
4:50 – 6:00 p.m.	**Discussion — Scientific Challenges** 1. What are the long-term chemical and materials sciences challenges that would be opportunities for quantum computing? 2. What are the short-term opportunities to begin demonstrating the value of quantum computing for BES chemical and materials sciences? 3. Why can't these challenges be addressed with today's petascale or tomorrow's exascale classical computers? 4. What advantages will quantum computers provide for these problems? 5. What computational approaches and algorithms are needed to solve these problems on a quantum computer?
6:00 – 7:30 p.m.	**Break for dinner (on your own)**
7:30 – 8:00 p.m.	**Synthesize High-priority Scientific Challenges**
8:00 – 9:00 p.m.	**Discussion: Computational/Algorithmic Challenges** Focus discussion on the priority scientific challenges 1. What computational approaches and algorithms are needed to solve these problems on a quantum computer? 2. How do we evaluate these algorithms for correctness and scaling? 3. How do we test algorithms on testbeds or in classical simulations to know how they are working?

Wednesday, November 1, 2017	
7:00 – 8:00 a.m.	Continental Breakfast — Lakeside 1 and 2
8:00 – 9:00 a.m.	**Quantum Computing Industry Panel Discussion** Alán Aspuru-Guzik, moderator 10 minutes per industry participant + discussion 1. What applications are you targeting in the near term, particularly in chemistry and materials? 2. What are the current challenges in developing quantum computers? How will those challenges impact addressing BES science challenges? 3. What types of architectures (e.g., type and number of qubits) are being pursued by your company? 4. What progress has been made on the software stack?
9:00 – 10:00 a.m.	**Discussion: QC Hardware and Software Tools** Focus discussion on priority scientific directions 1. What software tools, compilers, etc. would accelerate progress in solving chemical/material problems? 2. What are the characteristics of QC hardware (e.g., coherence times, fidelity) needed for these algorithms?
10:00 – 10:15 a.m.	**Break**
10:15 – 11:30 a.m.	**Discussions of Priority Science Challenges** Develop outline from science challenge to algorithms to software and hardware characteristics (can break into groups). Begin with presentation of the report template
11:30 – 1:00 p.m.	Working lunch (brief presentations from groups)
1:00 – 4:00 p.m.	Breakouts for writing assignments
4:00 – 5:00 p.m.	**Presentation of Major Conclusions and Closing Remarks**

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