Perspective

Computational Materials Insights Into Solid-State Multiqubit Systems

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The field of materials for quantum information science is rapidly growing with a focus on scaling and integrating new solid-state qubits. However, despite the extraordinary progress, major challenges must still be overcome for scaling. Specifically, there is a critical need for efficient coupling of tens to hundreds of solid-state qubits and multiqubit error correction to mitigate environmental interactions. This Perspective looks forward to the challenges ahead in the realization of multiqubit operations and collective phenomena with a focus on solid-state quantum materials. We provide a theorists' point of view on the modeling and rational design of these multiqubit systems. Our Perspective identifies a path for bridging the gap between the model Hamiltonians used to develop quantum algorithms and control sequences and the *ab initio* calculations used to understand and characterize single solid-state-based qubits.

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I. INTRODUCTION

A new generation of scalable quantum information technologies [1] that utilize the principles of quantum superposition and nonlocal entanglement would unlock applications ranging from quantum computing [2,3] and networks [4–6] to quantum sensing [7,8] and metrology [9,10]. The fundamental building block of these technologies are two-level quantum systems (qubits). A diverse set of qubits have already been identified based on superconducting quantum circuits [11–17], trapped ions [18–20], photons [21–25], molecules [26–28], and, the focus of this Perspective, semiconductor-based solid-state systems [29–35].

Over the past few decades there has been immense progress in the development of semiconductor-based solid-state qubits, and a plethora of qubits have been identified with each having the potential to fill at least one niche in the quantum revolution [1]. For example, atomiclike defects ("artificial atoms") both in three-dimensional (3D) bulk materials such as color centers in diamond [36–39], (di)vacancies in silicon carbide (SiC) [40–45], or defects in ZnO [46–48] and in 2D layered materials [49], such as transition metal dichalcogenides [50,51] or hexagonal boron nitride (hBN) [52–57], have shown promise to fulfill the need for single-photon emitters [58,59]. Single-photon emitters are vital to many quantum technologies (for example, quantum secure communication). Atomlike quantum defects in solids have also been demonstrated as electron spin qubits that can couple to nearby nuclear spins in the material, which are capable of storing quantum information for timescale orders of magnitude longer than the coherence times of the electron spin qubits to act as quantum memories and registers [44,60–70]. A second important class of solid-state qubits are quantum dot-based qubits [30,71–74]. There is a lot of variety in semiconductor-based solid-state qubits even within the subfield of quantum dot-based qubits. For example, spin qubits in electrically gated quantum dots [75–89], which have recently shown much promise for large-scale quantum computation outside of a dilution refrigerator [90,91]. Additionally, epitaxially grown semiconductor quantum dots are a leading candidate source of single photons [92–95], and colloidal quantum dots have recently shown promise as coherent single-photon emitters [96,97].

Essential to these semiconductor-based qubit systems reaching their technological potential is the discovery of improved materials that can coherently store and manipulate qubit states well enough for quantum error correction. Towards this overarching goal, elucidating a detailed understanding of the atomic geometry and electronic structure of the quantum system is a key step. Determination of these properties such as the symmetries (for example, parity and inversion) and associated quantum numbers (energy, orbital angular momentum, spin angular momentum) of the qubit states is critical because quantum algorithms and control sequences rely on this information [43,68,98–104]. Furthermore, the progression from

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fundamental characterization of a single solid-state qubit to applications of multiqubit logical operations relies on robust experimental and theoretical techniques capable of comprehensively characterizing single solid-state qubits. This realization and necessity has spurred the development of a range of experimental and computational methods capable of handling the different length, time, and energy scales inherent to single solid-state qubits. In particular, magneto-optical, electron spin resonance, charge and spin transport, quantum spectroscopy, electron microscopy, and many other optoelectronic measurement techniques have been essential to characterizing the functional behavior of semiconductor-based solid-state qubits [38,51,57,105]. On the theory side, *ab initio* methods have been instrumental in the development of our understanding of the precise atomic geometry, energy levels, and decoherence mechanisms of solid-state qubits [106–109]. Specifically, density functional theory [110–114], correlated and multireference electronic structure methods (e.g., coupled cluster, configuration interaction) [115,116], and methods that include electron-phonon [117–119] and electron-photon [120–124] coupling, and hyperfine interactions [125–129] have been and will continue to be critical in identifying the quantum states and coherence lifetimes of semiconductor-based qubit states [130]. These methods can also be useful in determining parameters that can be utilized as input parameters to open quantum system and dynamical methods based on effective Hamiltonians to study length and time scales that are currently out of reach of *ab initio* methods [131–135].

Given the diversity of semiconductor-based solid-state qubit systems and experimental techniques used to initialize, control, and readout their states, it is no surprise that many computational methods are required and currently used to study these material systems. Moreover, there are many different particle types (electrons, holes, excitons, electron and nuclear spins, phonons, and photons) that need to be modeled and understood in order for semiconductor-based qubits to reach their full potential. Borrowing terminology that originated in the astrophysics community [136] and has recently been discussed in terms of computational many-body physics [137], the terms "multimethod and multimessenger studies" encapsulate the need for diverse methods capable of accurately modeling multiple different particle types, often at the same time and different levels of theory. To this end, we postulate that there will be no single method nor computational formalism that would be able to be universally applied to modeling semiconductor-based qubit systems, but we are confident that intelligently combining and extending current methods will allow for accurate modeling of the inherently large system sizes of semiconductor-based multiqubit systems.

Specifically, as shown in Fig. 1, the modeling of a single atomlike quantum defect in a semiconductor, which can host the electronic qubit states, can often be achieved by considering on the order of tens of atoms. Today, mean-field methods such as density functional theory can readily handle hundreds to upwards of a few thousand atoms on modern supercomputers [138]. In contrast, electronic structure calculations using correlated, multireference electronic structure methods are typically limited to only tens of atoms [115,139–141]. Thus, in terms of atomlike quantum defects, it has become relatively straightforward to perform mean-field computations on most single physical qubits and it is even possible to use high-level correlated methods on some solid-state single physical qubit systems [142,143]. On the other hand, the use of high-level correlated methods on multiple solid-state qubits remains prohibitively computationally expensive due to multiqubit systems containing hundreds to thousands of atoms. Thus, while there are computational methods that are sufficiently accurate for studying a single physical qubit, understanding the interaction between physical qubits remains an additional challenge. One contribution to this challenge is that it is often not valid to assume that a multiqubit system is simply the "product" of identical single qubits. For example, defects and atomic substitutions in a material often lead to strain that, among other things, can rotate the lattice such that the ideal periodicity is disrupted and the identical nature of two different atomic defects within a single material is broken, as depicted in the top panel of Fig. 1. Additionally, the reality for the foreseeable future is that many physical qubits will be needed to form a single logical qubit in order to be compatible with the quantum error correction algorithms used in quantum circuits [3]. This further necessitates the development and application of methods capable of studying system sizes with multiple physical qubits. We envision that this can partly be achieved by increasing the efficiency of current methods used to study single semiconductor-based qubit systems, but it also will involve the development of entirely new algorithms and novel combinations of current methods.

In this Perspective, we discuss the general material and computational challenges that are pervasive in studying any semiconductor-based solid-state multiqubit system. There are other excellent comprehensive reviews on specific solid-state qubits and applications of solid-state qubits that we point the reader to for an understanding of progress in the field over the last decade. Section II provides an overview of the overarching material and computational challenges inherent to semiconductor-based solid-state multiqubit systems along with several important exemplary questions that we surmise *ab initio* studies will help answer in the forthcoming years. Section III then details the opportunities and possible paths forward that we are confident will be instrumental over the coming years in the study of semiconductor-based multiqubit systems. Specifically, Sec. III A examines quantum embedding theories and how they can enable large-scale calculations

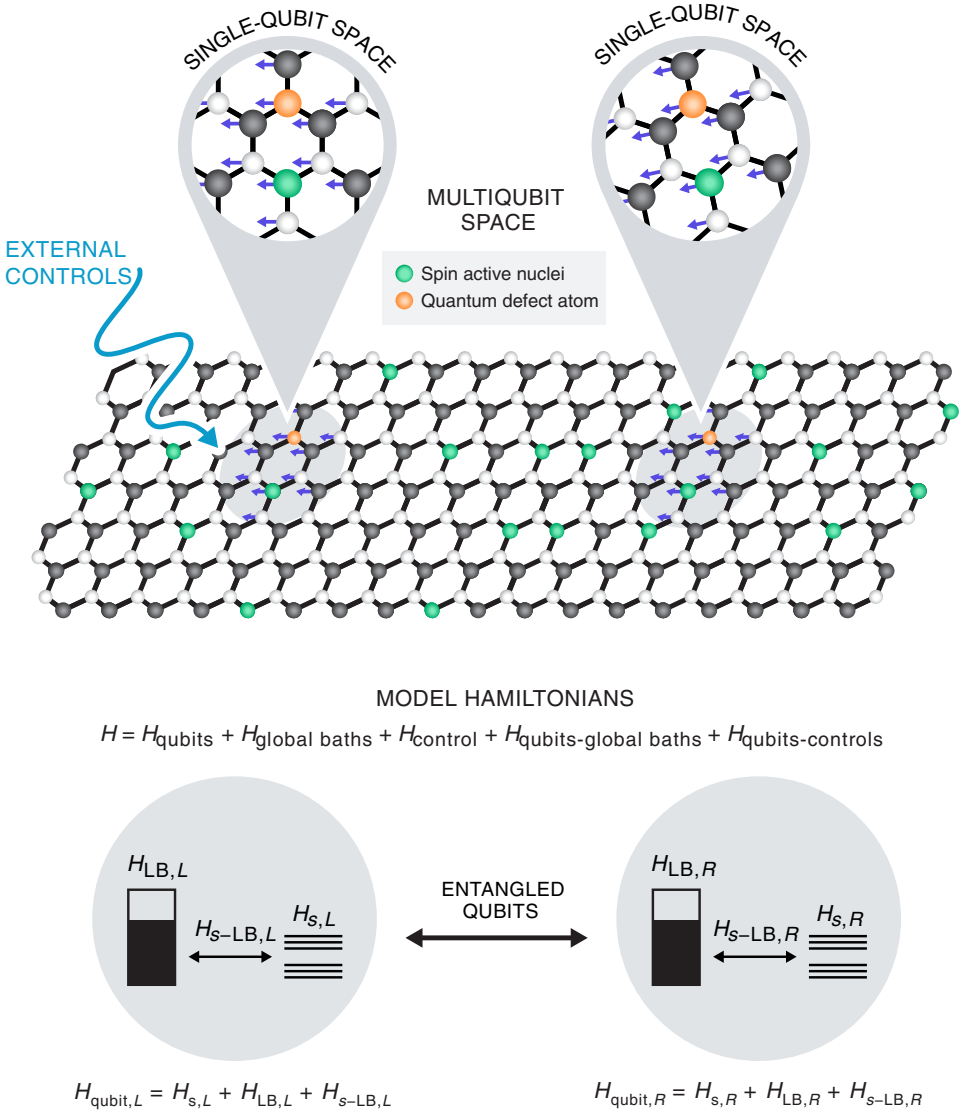

FIG. 1. A schematic of a general 2D semiconductor material containing two quantum defect atoms shown in gold. The defect atoms (gold) host atomiclike electronic states that can act as single physical qubits. The 2D host material also contains spin active nuclei ($I \neq 0$, green atoms) that can be utilized as long-term quantum memories but can also induce decoherence in the form of a spin bath. Atomic motion (for example, via a phonon) is represented by small blue arrows in the top scheme. The bottom scheme shows a hypothetical potential mapping of the top physical system onto two six-level systems denoted by $H_{s,L}$ and $H_{s,R}$, where s stands for the system Hamiltonian and L, R denote the left and right qubits, respectively. These system Hamiltonians are coupled to possibly distinct local baths ($H_{LB,L}$ and $H_{LB,R}$), global baths, and external controls.

of correlated, multireference electronic states of multiqubit systems. In Sec. III B, we discuss recent developments in *ab initio* methods that include electron-phonon and electron-photon interactions while highlighting areas where methodological improvements are still needed. In Sec. III C, we detail how and why the methods described in Secs. III A and III B ought to be combined with state-of-the-art open quantum system methods to study time-dependent phenomena in multiqubit systems such as decoherence and quantum error correction schemes. Section IV concludes this Perspective by providing a brief summary along with our optimistic outlook on the future of semiconductor-based solid-state qubits.

II. CHALLENGES AND OPPORTUNITIES IN MULTIQUBIT STUDIES

Computationally, the challenges inherent to semiconductor-based solid-state multiqubit systems stem from a few fundamental properties. First, the length scales involved in multiqubit systems are typically one or more orders of magnitude larger than that of a single qubit. This is demonstrated in Fig. 1 in which the simulation of a single qubit would require only tens to hundreds of atoms in the unit cell, whereas the simulation of the two-qubit system would involve hundreds to thousands of atoms. This increase of the system size by one or more orders of magnitude significantly impacts the difficulty in accurately describing multiqubit systems using *ab initio* methods, because even the computational cost of "computationally cheap" *ab initio* methods such as density functional theory (DFT) increase with the system size (N) to the third power. Throughout this Perspective, computational scalings will be denoted using the notation $O(N^{\alpha})$, where α is an integer indicating the system size dependence of the computational cost. For example, the computational scaling of standard DFT calculations using the local density approximation (LDA) or the generalized gradient approximation

(GGA) is $O(N^3)$. Furthermore, conventional DFT, in many cases, does not predict reliable electronic energy levels [142,144–148]. And, despite much progress, high-level correlated wavefunction methods that can predict reliable energy levels are still too computationally expensive to be applied to multiqubit systems containing hundreds to thousands of atoms because these methods have computational scalings of at least $O(N^5)$ [149]. The second challenge, in addition to the "raw" increase in the system size, is that the complexity also increases due to the necessity of accurately modeling the external controls involved in multiqubit operations with first principles methods. The types of external controls range from driving via electric and magnetic fields, optical, magneto-optical, optomechanical pulses, and the combination thereof to cavities and device contacts [71–73,150–156]. The addition of the nuclear and light degrees of freedom (i.e., phonon and photon modes) and their coupling to the electronic degrees of freedom to the simulation drastically increases the computational cost and complexity of multiqubit calculations. The third challenge stems from the fact that the external controls, atomic positions, and electronic states of the semiconductor-based qubit materials can vary in time, bringing us to our final fundamental challenge inherent to studying semiconductor-based multiqubit systems: how do we perform quantum dynamics on such large and complex multiqubit systems?

We conjecture that the promising fields of quantum embedding theories utilizing correlated, multireference impurity solvers, [157–162], *ab initio* methods that include electron-phonon [117–119,163] and electron-photon interactions [120–124], and open quantum system techniques [164–176] combined with exascale high-performance computing will be able to bring quantitative accuracy to multiqubit calculations in the forthcoming decade—greatly speeding up the identification, characterization, and optimization of novel solid-state quantum materials.

Prior to delving into the details of recent developments and exciting prospects of these methods and combinations thereof, it is worthwhile to provide a conceptual understanding of the workflow we are imagining and to give a few concrete examples of the types of questions that can be answered by combining quantum embedding theories, electron-phonon and electron-photon calculations, and open quantum system techniques. Conceptually, we envision the workflow beginning with the utilization of quantum embedding methods in order to identify the most important many-body electronic states with quantitative accuracy. Next, electron-phonon and electron-photon computations will be performed to elucidate the specific phonon or photon modes that are best suited to act as control knobs or are most likely to be sources of unwanted decoherence. With the crucial electronic, phononic, and photonic states identified and with their couplings computed, dynamical techniques developed in the open quantum system communities can be rationally chosen to study time-dependent phenomena using model Hamiltonians for the multiqubit system and environmental degrees of freedom based on information gleaned from the first principles calculations.

Let us now break down a few exemplary questions that this workflow can help answer. Three often intertwined categories of questions will be discussed here and throughout Sec. III: formation of stable logical qubits from multiple physical qubits, deterministic coupling and generation of entanglement between qubits, and interqubit decoherence mechanisms in semiconductor-based multiqubit systems. The first prototypical system we discuss is that of electron spin qubits in silicon. In silicon-based quantum computing, the electron spins are typically from either donor atoms or added via electrical contacts in silicon-based quantum dots [82]. Here we focus on electron spins in silicon donated by phosphorus atoms; this system also serves as a prototype for shallow donor spin qubits [33,34]. A great jumping off point to show our vision for using a multimethod and multimessenger approach to multiqubit systems is the recent computation and theoretical study by Swift *et al.* [129] in which first principles calculations of single phosphorus donors in silicon required the use of hybrid functionals [computational scalings of $O(N^4)$] in tandem with traditional, cheaper functionals [computational scalings of $O(N^3)$] to accurately model and converge calculations with large unit cells needed because of the shallow nature of the donor results in rather delocalized electronic states. The use of hybrid functionals was necessitated by the well-known problem of both LDA and GGA functionals to delocalize electronic states too much [129]. Additionally, Swift *et al.* [129] showed that strain has a larger impact on the hyperfine structure than predicted by effective mass theory, highlighting the importance of *ab initio* studies. Quantum embedding theories (Sec. III A) offer a systematic way to combine multiple levels of electronic structure theory in order to correct errors in low-level theories, such as the delocalization error in DFT or complete lack of electron correlation in Hartree-Fock theory, and predict accurate energies and wavefunctions of electronic qubit states. Furthermore, given the recent report of a two-qubit gate between phosphorus donor electrons in silicon [177], accurate descriptions of the electronic wavefunctions will be key to understanding distance-dependent and entanglement properties of shallow donor-based two-qubit states. For example, the extent to which the wavefunction is localized to the donor site will be key to predicting accurate couplings between neighboring donor sites as direct, tunnelinglike coupling typically has an exponential dependence on the overlap of the electronic states. In addition to direct, tunnelinglike coupling, understanding the dipolar coupling and resulting entanglement between semiconductor qubits requires accurate electronic

wavefunctions along with knowledge of their couplings to external controls (for example, photons).

In terms of the deterministic coupling of qubits via dipolar coupling, there is still much to be understood in terms of how two semiconductor-based qubits interact with one another. Two recent experimental findings of room-temperature entanglement between nitrogen-vacancy (NV) defects in diamond [178,179] are worth analyzing as a starting point for where we find computations will be beneficial to this end. In 2013, Dolde *et al.* [178] reported strong coupling between the ground-state spin magnetic dipole moments of NV defects spaced about 25 nm apart in large magnetic fields. In 2017, Bradac *et al.* [179] demonstrated coherent, superradiant emission between multiple NV centers in diamond microcrystals. A central and reoccurring question arises when considering the extensions of these exciting works: are the NV centers and their interactions with their local and global environments truly identical? We surmise that many facets of this question can be, at least in part, answered by the methods discussed in Sec. III. One important facet of this question is whether or not the qubits are subject to the same environmental noise. If, for example, the quantum defect atoms were to have different orientations in the crystal, as those shown in Fig. 1, the qubits would not be equivalent and the noise spectrum for each qubit would be different. Dolde *et al.* [178] postulated that the magnetic field noise was not identical for their two entangled NV centers based on entanglement lifetime measurements of two particular multiqubit states being identical, despite one constituting a decoherence-free subspace if the magnetic field noise were identical for both NV centers. We envision computational and theoretical investigations into the microscopic origin of this non-identical noise spectrum providing much value, and recent computational developments have made it possible to study systems in strong magnetic fields using coupled cluster theory [180] and accurately calculate hyperfine interactions [127–129]. These advances should provide a means to understand how sensitive the qubits are to magnetic field noise on a microscopic level when combined with quantum embedding techniques to enable large system sizes to be studied; see Sec. III A. The superradiant emission reported by Bradac *et al.* [179] was not ideal as a majority of their nanodiamonds did not exhibit superradiant emission, begging the question as to what is the microscopic origin of this finding. Here again, accurate electronic structure techniques that can study the distance and geometry-dependent coupling of the optical transitions of NV centers are needed. Furthermore, a potentially exciting direction would be to analyze the coupling of multiple optically active defects inside of an optical cavity using quantum electrodynamics methods [123,181–185]. And similar to how the impact of magnetic field noise influences the decoherence timescale depending on whether it is local or nonlocal, the impact of local (that is, vibrations concentrated near the quantum defect atom) and nonlocal (that is, global) phonons could have important consequences on the superradiant properties of a collection of quantum defect atoms.

Given that superradiant phenomena have many potential applications in quantum information [130,186–188], such as the generation of entangled photon pairs [189–193], we are also enthused by the prospects of superradiant phenomena in semiconductor colloidal nanomaterials [194]. Furthermore, the recent reports of single colloidal nanomaterials having emission quantum yields of 99.6% [195], achieving single-photon emission [96], and single-photon superfluorescence [196] open the door to realizing colloidal semiconductor nanomaterials as optically addressable qubit platforms for quantum information sciences [97]. We are also excited by the potential of colloidal quantum dot molecules [197–199] as a playground for computational and experimental studies on optically addressable and electron spin qubits in double quantum dot nanostructures, which have proven extremely valuable in other quantum dot communities for studying singlet-triplet qubits in particular [75,83,84,89,100,101,200–208]. However, numerous questions remain about the decoherence mechanisms and role of electron-phonon coupling in these novel colloidal nanomaterials. Thus, we conclude that accurate electronic structure theory via quantum embedding theories, electron-phonon and electron-photon calculations, and dynamical methods will be key to realizing the potential of colloidal nanomaterials for quantum information processing [97].

We now delve into more detail about the recent developments and opportunities with respect to both method development and applications of quantum embedding theories (Sec. III A), electron-photon and electron-nuclear methods (Sec. III B), and utilizing open quantum system techniques (Sec. III C) to study semiconductor multiqubit systems.

III. NEW DIRECTIONS IN MULTIQUBIT STUDIES

A. Quantum embedding theories

The nascent field of quantum embedding theories [157–159,209] will be of great utility to solid-state multiqubit systems, in part because capturing correlated electrons within a single qubit and entanglement between qubits requires computational methods capable of handling large systems of strongly correlated matter. The development of *ab initio* methods that can accurately describe strongly correlated electrons is an old, yet, still very fruitful field in the quantum chemistry and condensed matter physics communities [210]. A few of the methods of particular interest to modeling solid-state qubits are: coupled cluster (CC) [211–218], density matrix renormalization group (DMRG) [219–222], quantum Monte Carlo (QMC) [223,224], many-body perturbation theory [225,226], and

configuration interaction (CI) [116,227]. While these methods have been studied and tested extensively in molecules [140] and bulk solids with small unit cells [149,218,228], their use in semiconductor qubit systems still remains limited. This limited use is a result of the high computational cost of these methods and the large unit cells (or large finite sizes) of solid-state qubits, especially in the case of multiqubit studies (Fig. 1). Specifically, coupled cluster singles and doubles (CCSD) scales as $O(N^6)$, DMRG also scales as $O(N^6)$, and QMC scales as $O(N^5)$. These scalings make using these correlated methods [115] to investigate solid-state single physical qubits very difficult and will continue to make studying multiqubit systems impractical even as computing power reaches the exascale in the forthcoming decade. Fortunately, quantum embedding techniques [158–162,229–240] make it possible to combine (i.e., embed) high-level correlated methods with lower-level *ab initio* methods such as DFT, Hartree-Fock (HF), or Green's functions based methods such as the *GW* approximation [241,242] to calculate accurate ground- and excited-state properties of systems much larger than could be done using only the high-level method [243,244].

The combination of quantum simulation techniques, which formally defines quantum embedding, can be done in a variety of ways. Currently, the three formulations that are front runners for embedding a high-level impurity solver (e.g., CC, DMRG, CI) within a low-level method (e.g., DFT, HF, *GW* approximation) are: density functional embedding, Green's function embedding, and density matrix embedding [159,209]. These formulations utilize the single-particle density [245–247], single-particle Green's function [235,248,249], and single-particle density matrix [158,250,251], respectively, to provide feedback between the embedded system (i.e., impurity or the high-level simulation) and its environment (i.e., the low-level simulation). While we leave the discussion of the details of these methods to the excellent recent reviews [157–159,209], we discuss here a couple aspects of quantum embedding methods that are most relevant to our discussion of solid-state multiqubit systems and provide details for a potential workflow for utilizing these types of calculations in practice to study a multidefect system such as shown in Fig. 1. Common to all quantum embedding methods is the task of dividing the total system into embedded and environment fragments. This partitioning can be done in a variety of ways, including real or energy space. Conceptually, a real-space partitioning is intuitive for defect-based qubits due to the spatial localization of the qubit states. On the other hand, the qubit states in quantum dot-based qubits are delocalized over many atoms, and, thus, an energy-space partitioning seems more natural. However, studies that benchmark the accuracy and convergence of the different partitioning strategies is still an active field of research, especially for semiconductor-based qubit systems [240,252–254].

Along similar lines, because quantum embedding theories and algorithms have been developed primarily within the past few years, they are still a very active field of research and to date, there is no "gold standard" quantum embedding theory. Further we expect that more methodological development, tests, and benchmarking of these methods over the coming years will be key to making quantum embedding theories a reliable and quantitatively accurate computational tool for detailing the electronic structure of solid-state multiqubit systems. Additionally, the extension of the aforementioned *ab initio* methods to include relativistic effects [255–258], such as spin-orbit interactions, is also an important step in making *ab initio* electronic structure calculations of solid-state multiqubit systems generally applicable [103,208,259–262].

We now breakdown the practical steps involved in a quantum embedding theory calculation, using the system shown in Fig. 1 as a generic model for a semiconductor multiqubit system based on quantum defect states. The first step requires performing a low-level (e.g., DFT, HF, second-order MØller-Plesset perturbation theory, etc.) calculation of the entire multiqubit system. The next step involves building the embedded fragments. In the case of real-space embedding, which typically makes logical sense for spatially localized qubit states, the embedded fragments could be chosen based on some spatial criteria [162], such as shown schematically in Fig. 1. Within the localized orbitals in the embedded fragments, a high-level impurity solver (e.g., CC, CI, DMRG, etc.) calculation is performed to capture the correlated, possibly multireference character of the many-body electronic states. Next, self-consistency between the embedded fragments and surrounding system can be imposed [159,209]. In the event that this fragment-surrounding self-consistency is not imposed, quantum embedding methods resemble active space wavefunction methods. Having obtained the correlated, possibly multireference electronic states for the multiqubit system, the analysis of their properties (symmetries, quantum numbers, etc.) begins. In addition to the more common analysis of the energies and symmetries, we are excited by the possibility of utilizing various entanglement measures [263–265] commonly employed in quantum information and the many-body physics communities to probe the interqubit entanglement properties and, specifically, how the entanglement between qubits depends on the microscopic details of the system. In the next section we discuss how to elucidate the impact of the coupling of these many-body electronic states to the electromagnetic and nuclear degrees of freedom using both perturbative and nonperturbative methods. In Sec. III C, we then delve into how the information gleaned from the calculations described in Secs. III A and III B can be

utilized to rationally build simplified model Hamiltonians to investigate the dynamics of multiqubit systems.

B. Electron-phonon and electron-photon interactions

While quantum embedding theories are a promising solution to the multireference, strong correlation, and large system size problems that arise when solving the electronic structure of multiqubit systems with fixed nuclear coordinates and outside of any optical cavity, multiqubit studies will also greatly benefit from methods that include electron-phonon and electron-photon interactions at the quantized, *ab initio* level [163,183,184]. The inclusion of these interactions is especially necessary for understanding the influence of external controls in addition to uncovering the mechanisms and timescales of relaxation and decoherence within and between solid-state qubits [266].

In particular, as shown in Fig. 1, the nuclei surrounding the atomic substitutions that host the physical qubit states (and, generally, the nuclei throughout the multiqubit space) are constantly in motion. This motion can strongly impact the electronic energy levels and dynamics of solid-state qubits through electron-phonon interactions [267]. Typically, the impact of electron-phonon interactions on the coherence time of solid-state qubits is detrimental. This detrimental impact can be dramatically reduced by going to low temperatures; however, a major selling point for solid-state qubits is their potential to operate at room temperature [40,64,66,156,178,268,269]. On the other hand, electron-phonon interactions can provide an avenue for controlling the electronic qubit states via driving of specific phonon modes [270]. Designing phonon-mediated qubit control requires a detailed understanding of the electron-phonon coupling in the material. Thus, first principles methods that include electron-phonon interactions and, ideally, simultaneously capture the multireference character of solid-state qubits are very desirable.

To this end, DFT-based frozen phonon and perturbation theory (DFPT) [271–273] has been the go-to *ab initio* method for the inclusion of electron-phonon interactions in solid-state materials for a while now [214, 274]. However, DFPT inherits the limitations of conventional DFT [275,276] (e.g., fails to capture strong correlations, provides orbital energies of questionable accuracy, etc.) and is also perturbative in the electron-phonon coupling strength by construction. Perturbative methods that are compatible with the high-level methods (e.g., coupled cluster) have been developed, providing a multireference wavefunction-based method for investigating electron-phonon interactions in the ground state of materials. Additionally, *GW* perturbation theory (GWPT) has recently proven a reliable tool for studying electron-phonon coupling in the perturbative coupling regime of charged excitations in materials [118,276–280]. And we envision that the recent extensions of GWPT to include electron-hole interactions via the Bethe-Salpeter equation [281–284] will permit accurate and fruitful studies on the impact of exciton-phonon interactions in optically addressable solid-state qubit systems.

In order to tackle the challenge of strong electron-phonon interactions and phonon driven systems in solid-state multiqubit systems, recent advances that treat electron-phonon interactions on an equal footing as electron-electron interactions will be very valuable [120, 285,286]. Furthermore, we expect that the recent extension of these methods that include electrons and nuclei on equal footings to be compatible with the high-level impurity solvers such as coupled cluster theory [119,163,287–289] makes these methods ready to be combined with the quantum embedding theories discussed in Sec. III A. These combinations could result in accurate computational approaches for studying weak to strong electron-phonon interactions in large, complex multiqubit systems.

Analogous to electron-phonon interactions, electron-photon interactions play a crucial role in many semiconductor-based qubit systems [67,73,290–294]. For example, optical cavities are often utilized to enhance optical transitions [295–301], light pulses are used to initialize qubit states [302], perform gate operations [92,293,303], and readout qubit states [65,304] for the promising category of optically addressable solid-state qubits [292,294,305,306], and electric and magnetic fields are often used to control quantum dot-based spin qubits [78,90,91,307,308]. Therefore, methods capable of accurately calculating transition electric dipole matrix elements along with the impact of light-matter interactions on the electronic structure are desired [184,309].

Similar to DFT-based methods having been the workhouse of electron-phonon interactions in materials, quantum electrodynamics density functional theory (QEDFT) [120,121,181,182,310] has been instrumental in the development of *ab initio* hybrid light-matter methods. However, QEDFT typically does not describe optical excitations accurately. This inaccuracy stems from QEDFT relying on DFT to model the electronic states, which often predicts inaccurate energies and magnitudes of optical transitions [311]. To solve this problem, we are thrilled by the recent development of including electron-photon interactions in a nonperturbative manner within the Bethe-Salpeter equation (BSE) formalism [122]. BSE formalisms for calculating neutral excitations (i.e., excitons) have been tremendously valuable in investigations of the optical properties of materials [225]. Additionally, QED-CC is an exciting example of a method that combines an accurate correlated wavefunction-based approach with quantum electrodynamics [123,124,185]. And, given that equation of motion coupled cluster theories have been very successful at predicting optical absorption energies and transition strengths [215,217,218], we suggest that QED-CC is a

promising method for analyzing interqubit interactions in optical cavities [153,312]. A recent report by Haugland *et al.* [185] highlights the importance of using correlated electronic structure methods when analyzing intermolecular interactions in optical cavities with *ab initio* QED methods. In particular, Haugland *et al.* [185] found that QED-HF and QED-DFT predicted purely repulsive interactions between two H_2 molecules, whereas QED-CCSD was able to capture the attractive interaction. It was also reported that the changes in the strength of dipole-induced dipole and dipole-dipole interactions as a result of the cavity were much larger in QED-CCSD calculations compared to QED-HF and QED-DFT calculations. These findings demonstrate the importance of using correlated electronic structure methods when analyzing intermolecular interactions. Thus, we postulate that these lessons learned from fundamental studies of intermolecular interactions will also apply in studies of interqubit interactions inside cavities. However, unfortunately, QED-CC is computationally expensive. Thus, QED-CC will likely have to be combined with a lower-level method, such as QED-HF, via a quantum embedding scheme in order for studies of multiqubit systems to be both accurate and computationally tractable.

There have also been recent advancements that permit the inclusion of dissipative processes in cavity QED within *ab initio* formalisms [313]. The inclusion of dissipative processes is an important aspect as dissipation arises naturally in cavity-mediated qubit operations and results in energy relaxation—hurting the performance of optically addressable solid-state qubits. Therefore, the ability to study these processes on a first principles level is critical to improving solid-state qubits. Put together, we envision that the methodological advancements on accurately modeling electron-phonon and electron-photon interactions in complex solid-state materials should permit exciting new design principles and quantum control schemes of multiqubit solid-state systems. For example, the dipole-dipole coupling between optically addressable qubits [179,314] could be analyzed and optimized using first principles calculations together with experimental techniques to control the defect-defect distance in materials [315]. In the following section, we delve into how the aforementioned methods for calculating electron-phonon and electron-photon interactions can also be used to provide key input parameters (i.e., matrix elements) to open quantum system techniques that have been developed for studying the central dynamical processes in quantum information processing.

C. Open quantum system techniques

In Sec. III A we discussed the methods that we expect will make it possible to identify the intricate details (e.g., quantum numbers) of the few many-body electronic energy levels of interest in a multiqubit system. Then, in Sec. III B, we outlined a procedure for predicting how the electronic states of interest couple to optical (electron-photon interactions) and mechanical (electron-phonon interactions) degrees of freedom. Two of the goals of the calculation of these couplings are: (1) to understand the intrinsic stability (for example, the energy loss T_1 and dephasing times T_2) of the multiqubit system and (2) to identify specific couplings (e.g., electric dipole transitions, phonon modes) that can be utilized to develop and optimize useful quantum control schemes for the qubits. This brings us to the bottom half of Fig. 1 where we postulate that leveraging the methods of the open quantum systems community [316–318] to study and optimize the performance of solid-state multiqubit systems will be very fruitful.

Specifically, we envision that in order to predict the success and optimize solid-state qubit systems, the static *ab initio* electronic structure methods described in Secs. III A and III B must be combined with open quantum system methods to study the dynamics [317] of these multiqubit systems. While the time-dependent simulation of an entire multiqubit systems (e.g., Fig. 1) along with its environment and time-dependent external controls at a fully quantized first principles level would be ideal, such a treatment is not feasible and it is generally unnecessary to treat all degrees of freedom of the electrons, nuclei, and photon modes at an equal level of theory. Whereas quantum embedding approaches allow for the use of multiple levels of *ab initio* electronic structure theory to be used in a single static electronic structure simulation and possibly over short timescales, open quantum system techniques provide an avenue for studying dynamical processes over much longer timescales than *ab initio* methods (e.g., *ab initio* molecular dynamics) by integrating over specified degrees of freedom. The integration of most of the degrees of freedom permits the dynamics of the remaining degrees of freedom (termed the system) to be scrutinized in detail. The degrees of freedom that are traced over are referred to as the environmental degrees of freedom, and there are many approximations of varying generality used to simulate the dynamics of the environmental and system degrees of freedom [316,319].

In the context of solid-state qubits, the most prevalent baths are: (i) phonon baths, (ii) photon baths, (iii) nuclear spin baths, and (iv) charge noise. Fortunately, decades of work have already gone into developing methods capable of studying the dynamics of an electronic qubit or multiqubit system coupled to these types of baths [316,319–321]. Furthermore, the methods have already been extensively tested against model Hamiltonians (for example, the Anderson-Holstein model for phonon baths [322], Jaynes-Cummings model for photon baths [323], spin star model for spin baths [324], and two-level fluctuator models for charge noise [325–328]).

In what follows, we present a brief discussion of the typical methods that have been developed over the years to study time-dependent phenomena in these model Hamiltonians as well as their extensions. We also discuss recent progress and future opportunities in applying these techniques to *ab initio* simulations. Throughout this discussion, the important question of how to choose both the correct model Hamiltonian and method will be demonstrated by analyzing specific solid-state multiqubit systems.

Most of the common methods used to simulate time-dependent phenomena in open quantum systems (recall that all qubit systems are open quantum systems in practice) are based on: reduced density matrix theories, Green's functions [329–331], semiclassical and quasiclassical treatments [118,332–334], Floquet theory [335–337], and approximate descriptions of the full many-body wavefunction. Recent combinations of these methods also appear to be promising methods for simulating large multiqubit and bath Hilbert spaces while maintaining accuracy and efficiency [338–340]. While the intricacies of these methods are out of the scope of this Perspective, it is worthwhile to elaborate on a few of the techniques and approximations used to make these simulations computationally tractable. For example, formally exact equations of motion for the reduced density matrix (which are generalized quantum master equations) can be greatly simplified by utilizing perturbation theory and timescale separation to obtain Born-Markov master equations. These approximate quantum master equations, such as the Lindblad formalism or Redfield's theory, are computationally much easier to solve. However, as mentioned above and discussed more below, the role of non-Markovian effects on the coherence between qubits is an important open question [130,178,203,266]; we suggest that combining the *ab initio* methods discussed in Secs. III A and III B with non-Markovian open quantum system techniques [341,342] can address these questions. For example, accurate electronic structure calculations can be used to obtain hyperfine parameters [343] in semiconductor qubits that can then be used to build a spin star model Hamiltonians from which non-Markovian dynamics has been studied [324]. Extensions of these to the multiqubit systems will be very interesting and will critically enable the design and understanding of decoherence free subspaces. Another scenario in which non-Markovian effects would be important is understanding the role of electron-phonon interactions on the coherence between two defect qubits. Specifically, we are intrigued by the possibility of elucidating the impact of local and nonlocal phonons on the entanglement and coherence properties between qubits [344,345].

Additional important approximations are incorporated during the separation of system and environmental degrees of freedom. Particularly, the decisions of which degrees of freedom should be included in the system, and how accurately to treat the environmental degrees of freedom are relevant in work trying to model the dynamics of multiple solid-state qubits. To make these approximations physically relevant, using input from both experimental measurements and *ab initio* calculations will be key. In other words, a tight feedback loop between experimental measurements, *ab initio* calculations, and model Hamiltonian predictions will greatly aid the rational design of open quantum system techniques used to model the dynamics of multiqubit systems. As an explicit example of how these pieces can potentially come together, we consider the driving of the two-qubit system shown in Fig. 1 by a laser field at finite temperature. The multimessenger involved in this system is seen immediately, with electronic, nuclear, and electromagnetic field degrees of freedom being involved. One open quantum system direction that may be useful for modeling the dynamics of this driven multiqubit problem is the application of semiclassical and quasiclassical methods in atomistic molecular dynamics simulations. In particular, recent developments of these methods use classical trajectory-based approaches to approximate the quantum dynamics have allowed for nonadiabatic dynamics in a laser field [339], nuclear quantum effects [346], and decoherence and coherence processes [173,347] to be simulated. We envision the role of the methods discussed in Secs. III A and III B to illuminate, with the aid of experimental measurements, which electronic, nuclear, and electromagnetic degrees of freedom need to be included quantum mechanically and which can be modeled classically (for example, can the nuclear motion be modeled well by Ehrenfest dynamics?). Lastly, we note that utilizing similar open quantum system techniques developed to analyze and model semiconductor-metal interfaces [174,175,348] in conjunction with *ab initio* calculations could result in improved multiqubit coherence lifetimes and operation fidelities in quantum dot-based qubits that use gate operations based on quantum transport. In these systems, model Hamiltonians have been instrumental in their development as spin and charge transport through quantum dots and quantum dot arrays using model Hamiltonians has been a staple problem in the open quantum system community for decades [330,349–355]. While it has been challenging to utilize these techniques in *ab initio* settings with large system sizes, recent works provide hope this may be possible for multiqubit studies in the coming years [356–365].

Thus, we envision that open quantum system techniques are primed for applications for studies of real and complex solid-state multiqubit systems. Conceptually, joining the *ab initio* methods discussed in Secs. III A and III B with the abovementioned open quantum system techniques to study a new solid-state multiqubit system would proceed as follows. Experiments and *ab initio* calculations would provide information to guide the modeling and solving of effective system and bath Hamiltonians from which key dynamical properties (e.g., information flow [366]) of the

multiqubit system can be obtained. A recent example of this for a single solid-state qubit was that of analyzing the impact of the nuclear spin bath on the dynamics of the electronic qubit of a NV center in diamond using the open quantum system method of the coupled-correlation expansion method [132,367].

In summary, the comprehensive approach to method development discussed in Sec. III that focuses on the combination of accurate electronic structure methods with sophisticated theories from open quantum systems can address the challenges in characterizing and optimizing the performance of novel multiqubit solid-state systems over the coming decade.

IV. SUMMARY OF THE PERSPECTIVE

Theoretical and experimental studies of multiple physical qubits are essential in the field of quantum information sciences because many physical qubits are often required to produce the single logical qubits that are the building blocks of quantum computing algorithms and circuits [3]. In this Perspective, we describe and provide new directions to overcome the challenges associated with performing accurate *ab initio* computations of solid-state multiqubit systems. Specifically, we outline how quantum embedding theories, *ab initio* methods that include electron-phonon and electron-photon interactions, and open quantum system techniques can all play an important, and often intertwined role, in developing solid-state qubits over the course of the next decades. We confidently conclude that these types of calculations combined with the ever advancing experimental techniques and quantum algorithms will be instrumental to solid-state qubits reaching their full potential.

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