Title

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Journal

The Journal of Physical Chemistry B, 125(44)

ISSN

1520-6106

Authors

Lavroff, Robert H
Pennington, Doran L
Hua, Ash Sueh
et al.

Publication Date

2021-11-11

DOI

10.1021/acs.jpcb.1c08679

Peer reviewed

Introduction to ACS Virtual Issue

Recent Innovations in Solid-State and Molecular Qubits for Quantum Information Applications

Robert H. Lavroff, Doran L. Pennington, Ash Sueh Hua, Barry Yangtao Li, Jillian A. Williams, and Anastassia N. Alexandrova*
Department of Chemistry and Biochemistry, University of California, Los Angeles, Los Angeles, CA 90095-1569, USA
E-mail: ana@chem.ucla.edu

The field of quantum information science (QIS) has gained significant interest in the chemistry community over recent years. Through use of the quantum superposition principle, two-level quantum systems (qubits) can hold more information than the classical bits which comprise conventional computers, enabling a massive increase in the memory that machines can store and the speed at which certain types of calculations can be performed. Chemical systems well-suited for this purpose fall into a variety of material classifications and qubit-formation mechanisms, from altering the spin of an electron to leveraging the polarization of photons. The major stumbling block all of these systems share, however, is quantum decoherence: the loss of a definite phase between the two quantum states as the system relaxes to an impure state over time. Significant increases in the coherence time of qubits, and in the number of qubits that can be simultaneously entangled, will greatly enhance the utility of quantum computing and push us closer to achieving computational goals not currently possible with classical computers, such as factoring large numbers for cryptography applications and the efficient simulation of quantum systems such as large molecules. In this Virtual Special Issue, we present articles from three different arenas in qubit research: discovery of new qubit candidates and devices, elucidation of decoherence mechanisms to increase qubit lifetime, and computational applications of qubit-based quantum computers. The figure shown above highlights four such articles, showcasing cutting edge qubit technology from quantum error correction to sustainable hydrogen production.

In the **presentation of new qubits**, we survey a variety of qubit systems. Gao et al., and Beke et al. report newly-prepared defect qubits, which exploit defect sites within extended material lattices to access optically-addressable electron-spin qubits.[1,2] Chen et al. employs this system in the construction of a new device that uses mechanically-driven vibrations to stimulate electronic transitions within a defect qubit,[3] and Wang et al. computationally studies the interaction between neighboring defect sites to create “artificial molecules,” introducing a new degree of freedom for defect qubit design.[4] Another type of defect qubit based on nuclear spin rather than electron spin within a P:Si system is studied computationally by Chibisov and Chibisova.[5] Peng and Levine performed computational analysis on Si-based molecular dynamics of the interaction between defects during nonradiative recombination.[6] Qubits based on magnetic molecules are attractive owing to their synthetic tunability. These systems employ molecules with unpaired electrons that can be characterized by Electron paramagnetic resonance (EPR) and measured for quantum information by optical means, such as photoluminescence. Freedman and

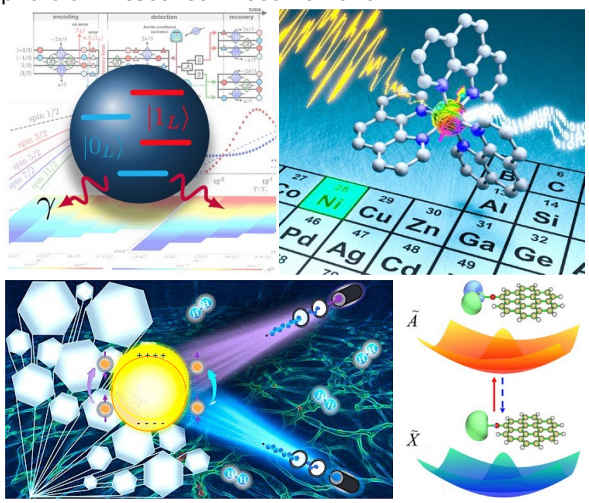

Figure. (a) Logical qubits with embedded quantum error correction in the form of molecular nanomagnets. (b) Optical readout in octahedral Ni(II) complexes for quantum sensing applications. (c) Two-qubit spintronic-photoelectrochemical hydrogen production using surface plasmon resonance excitations. (d) Franck-Condon tuning of Ca optical cycling centers using arene ligands.

colleagues introduce a new single molecule magnet qubit based on a Ni^{2+} complex, a concept that is further explored by Wasielewski, Han, Sherwin, Awschalom and Freedman using a V^{3+} system.[7, 8] Christensen et al. also present a novel qubit found in the spin-polarized triplet state of benzoisophosphinoline, characterized by the hyperfine coupling of phosphorous.[9] By controlling the chemical

environment, Xu et al. study the anisotropic behavior of a single-molecule magnet within an oriented single crystal,[10] and Garcia-Garibay and colleagues embed paramagnetic organic spin centers within a metal-organic framework.[11] Najafi et al. study superconducting resonators as a means of achieving coupling between single molecule magnet qubits, which is necessary for the implementation of quantum logic gates.[12] Similarly, Lino et al. studied a coupling strategy for nuclear-spin qubits through placing NMR-active nuclei in close proximity to each other within a single molecule or in a dimer.[13] Kung et al. describe a spintronic qubit in which circularly-polarized light excites different electron transitions depending on the polarization direction.[14] Lee and colleagues report a new tunnel barrier material for a transmon qubit that operates by measuring the voltage across a superconductor junction to enable novel quantum circuit elements.[15] Dickerson et al. attack optical cycling calcium qubits by functionalizing bound arene ligands, while Krylov discovers similar dual cycling centers in one molecule through addition of strontium.[16,17] Finally, Kothe et. al. leverage a hyperpolarized, pentacene-based molecular solid to achieve a 14-qubit entangled system.[18] These recent advances highlight the diverse nature and burgeoning study of qubit systems and devices.

Lifetime studies of existing solid-state and molecular qubits include observation and tuning of vibrational decoherence, as well as a variety of decoherence time optimization strategies. Wasielewski and colleagues demonstrate enhancement of spin-polarization lifetime in core/shell quantum dots through functionalization with organic radicals.[19] In another study, spin-selectivity in the photophysics of diradical molecules is shown.[20] Spin-phonon coupling, as a phenomenon strongly involved in coherence lifetimes, is a frequent topic of research. In molecular qubits, Albino et al. use first-principles to investigate spin-phonon coupling[21] and Santanni et al. delve into the role of vibrational symmetry in spin-phonon decoherence.[22] Wasielewski & Freedman and colleagues study spin-phonon coupling in modular qubit arrays.[23] In solid-state spin qubits, Lunghi studied the limiting effects of vibrations on spin lifetime.[24] Chang and Hyeon-Deuk investigate quantum resonance in defect qubits and how it affects orbital depolarization and phonon dynamics.[25] For electron spin decoherence induced by neighboring nuclear spins, Chen et al. utilize many-body simulations of molecular qubits to relate spin-spin distance to decoherence.[26] Canarie et al. correlates this decoherence in organic radicals from the structure and molecular geometry.[27] Wang et al. measure coherence in “Schrödinger-Cat states” by Landau–Zener–Stückelberg–Majorana interferometry.[28] Most recently, Kais et. al. develop an efficient method for quantum state characterization via the maximal entropy approach applied to the density matrix.[29]

At the **interface between chemistry and computer engineering**, theorists and experimentalists alike explore the assembly of quantum hardware, implement quantum computing algorithms, and find strategies to reduce the complexity of calculations on quantum computers. Wang & Yuan and colleagues discuss the use of zirconia’s molybdenum vacancy qubit in computing applications.[30] Electronic structures are affected largely by the change of molecular symmetries, and Setia et al. tune these molecular symmetries to reduce computational qubit requirements.[31] Ryabinkin, Genin, and Izmaylov develop computational chemistry methods in quantum computing algorithms, such as the iterative qubit coupled cluster and unitary partitioning approaches are introduced to quantum computing, as well as the measurement of all compatible operators in a series of single-qubits.[32, 33, 34] Carretta show that molecular nanomagnets have a potential advantage in the crucial rush toward quantum computers, and even leverages EPR and other magnetic resonance techniques to assess the lifetime of qubits via chiral-induced spin selectivity.[35, 36] Kottman et al. build on these newfound efficiencies by approaching the quantum eigensolver algorithm in a basis-set free approach, thereby reducing the number of qubits needed for accurate calculation.[37] Gao e al. put variational quantum eigensolvers to the test in the presence of noise by computing the energy profile of the biradical lithium superoxide dimer.[38] Sugisaki et al. propose a quantum algorithm for directly calculating vertical ionization energies on a quantum computer.[39]

A variety of breakthroughs in quantum computing algorithms for solid-state and materials chemistry are also presented. Fan et al. demonstrate the use of equation of motion theory for swift and accurate band gap calculations on a quantum computer, and Pavošević and Flick use the same for energetic calculations of polariton systems, many of which have applications to qubits themselves.[40,41] Finally, Becerra et. al. quantify uncertainty for microkinetic models via a quantum algorithm for matrix algebra, with applications to heterogeneous catalysis and other condensed-matter phenomena.[42]

The articles presented in this Virtual Issue highlight advances in solid-state and molecular quibits throughout the last two years. As breakthroughs continue in the field of quantum information science, we step closer and closer to the age of quantum computing, where we may reap the benefits of exponential increases in memory and performance, drastic improvements in cybersecurity and other cryptography applications, and most important to the chemistry community, affordable and accurate computational modeling of quantum systems. We hope this Virtual Issue helps advance the field and inspire future scientific discoveries.

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