Q4Q: Quantum Computation for Quantum Prediction of Materials and Molecular Properties

The driving force for this project was the demonstration that noisy intermediate state quantum computing devices could do some useful tasks in fields of research of interest for the USA government, in particular for the Department of Energy. Thus, we focused our work on the physical chemistry of molecules and materials, with potential impact in the energy industry.

To this goal, we put together a robust team that possesses the skills and curiosity to pursue the proposed objectives: Rosa Di Felice (PI), Anna Krylov (co-PI), Itay Hen (co-PI) and Amir Kalev (senior personnel) at the University of Southern California (USC); Marco Buongiorno Nardelli at the University of North Texas (UNT); Marco Fornari at Central Michigan University (CMU).

PI Rosa Di Felice (USC) has pioneered quantum machine learning by implementing on a D-Wave TwoX (DW2X) adiabatic quantum chip methods to analyze genomic data sets targeting the binding specificity between transcription factors and DNA [1]. She has participated in all objectives, with the main task of executing quantum computations on the existing hardware with quantum software packages, supervising and harmonizing the tasks of all co-PIs. She has initiated an ACS Symposium on "Bridging the gap between quantum computing and high-performance computing in quantum chemistry and materials science", which has gathered the applied quantum computing community in the American Chemical Society National Meetings in the last 4 years. We believe that events like this will greatly help the formation of a new hybrid scientific community that will be able to benefit from and contribute to the progress of quantum computers. Di Felice has also co-led the creation of a new Master's in Quantum Information Science at USC, launched in Fall 2021, within which she has also created a course on "Applications of Quantum Computing".

Co-PI Anna Krylov (USC) is an expert in electronic structure theory of electronically excited and open-shell species [2]. She invented the Spin-Flip (SF) method [3] that enables accurate and robust computations of strongly correlated systems (diradicals, triradicals, etc). Her main task has been the development of algorithms for quantum chemistry, calculating Heisenberg model parameters for single-molecule magnets and including the Heisenberg model and the Q-Chem plugin in quantum software packages [4].

Co-PI Itay Hen and senior personnel Amir Kalev (USC) are expert of adiabatic quantum computation (algorithms, complexity, simulations and implementation) and computational physics (quantum and classical Monte Carlo, optimization, etc.). Recently, they have developed an approach [5] for the computation of molecular Hamiltonian states that goes beyond the VQE. Within the project, they have further developed the algorithm and identified potential areas of application in quantum chemistry.

Co-PIs Marco Buongiorno Nardelli (UNT) and Marco Fornari (CMU) are founding members of the AFLOW Consortium [6, 7]. They developed new tools for high-throughput materials design

and discovery. In particular, their non-iterative projection techniques enable an optimal representation of the electronic structure in terms of tight-binding Hamiltonians. Fornari, in collaboration with Di Felice and Hen, has worked on materials optimization with D-Wave and on interfacing solid-state physics models with gate-model quantum computing (GMQC) approaches [8-10]. Buongiorno Nardelli, in collaboration with Di Felice and Hen, has worked on determining Hubbard parameters for strongly correlated solids to solve the Hubbard models by GMQC. He has developed algorithms and computational tools for the quantum computation of the band structure of solids [11-14].

The PIs have worked together, have published together and are cooperating in follow-up projects that were conceived as a result of this project. The list of references [4-5, 8-15] constitutes the products of this project. There are other publications that acknowledge the project but are not associated to specific project tasks.

We report below concise reports for selected activities. The project products in Refs. [4-5, 8-15] represent extensive reports.

Band structure of solids. [11-14]

In the course of this project, we have developed quantum algorithms for materials structural and electronic properties and on noisy intermediate-state quantum computing devices, and we have design efficient quantum circuits for the calculation of the band structure of solids (Figure 1). A great deal of recent literature explores VQE algorithms in small molecular systems, offering numerous approaches to characterizing energies of physical systems. We have extended these techniques to calculate band structures in solids. We explored the efficacy of numerous existing ansatze, such as UCC, QCC and also develop novel ansatze which guarantee periodic symmetry in the wavefunctions being represented, thus limiting the Hilbert space explored in the classical optimization only to those states which are meaningful in a crystalline structure. To explore excited states, we developed an algorithm to iteratively project out the ground state. We have also explored alternatives in the literature, such as Variational Quantum Deflation. Extending molecular VQE algorithms to calculate band structures in solids poses a particular challenge due to the "global" extent of the system. In general, a crystalline Hamiltonian represented on n qubits will contain the maximum number of 4^n terms, making the usual VQE algorithms much less efficient to implement. This is a "big data" problem, and we have proposed and exploration of dimensional reduction techniques such as Principal Component Analysis (PCA) to reduce the size of our Hamiltonian. We have tackled the rise of barren plateaus in the energy surface - an effect also observed in classical neural networks - which makes the classical optimization step

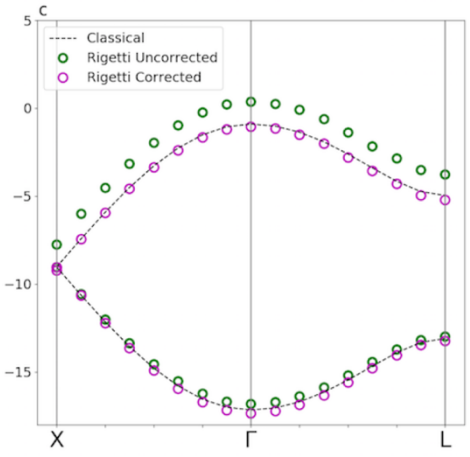

Fig. 1. Lowest 2 bands of bulk silicon, calculated along Brillouin Zone symmetry lines classically and with Rigetti QPUs.

to minimize energy extremely unreliable. We proposed a local pseudo-energy cost function which can be used in place of the true energy to perform the classical optimization. Finally, we implemented a "metadynamic" optimization algorithm such as those employed in molecular dynamics simulations to more efficiently explore our energy surface in spite of the challenges.

Quantum annealing in materials science and chemistry. [8-10 and unpublished]
PI Di Felice and co-PI Fornari have collaborated on the use of a D-Wave device to formulate and solve graph problems that can map optimization schemes in materials science. In an exploratory study, they have derived the quadratic unconstrained binary optimization (QUBO) formula for the Chinese Postman Problem (CPP) [8], with appropriate penalties. It is a prototypical graph problem. Minimization of the CPP QUBO with the classical solver Qbsolv always yields the correct minimum pathway. Minimization of the same QUBO on DW2X yields the correct solution with a probability that depends on problem size and embedding scheme. These results have been published in January 2021 [8]. Applications of color matching combinatorics to materials science problems is underway to study ordering phenomena in highentropy alloys, defected-graphene, and amorphous silicon. We are able to formulate QUBO problems that correctly find the ground state and estimate entropy contributions. In particular, we used a simple model, compatible with the capability of current quantum annealers, to study the relative stability of graphene vacancy defects [9]. By mapping the crucial interactions that dominate carbon-vacancy interchange onto a QUBO problem, our approach exploits the ground state as well as the excited states found by the quantum annealer to extract all the possible arrangements of multiple defects on the graphene sheet together with their relative formation energies (Figure 2). This approach reproduces known results and provides a stepping stone towards applications of quantum annealing to problems of physical–chemical interest. Unpublished results on quantum annealers include the implementation of ground-state energy estimation and the inclusion of solvent effects in molecular ground-state energy estimation.

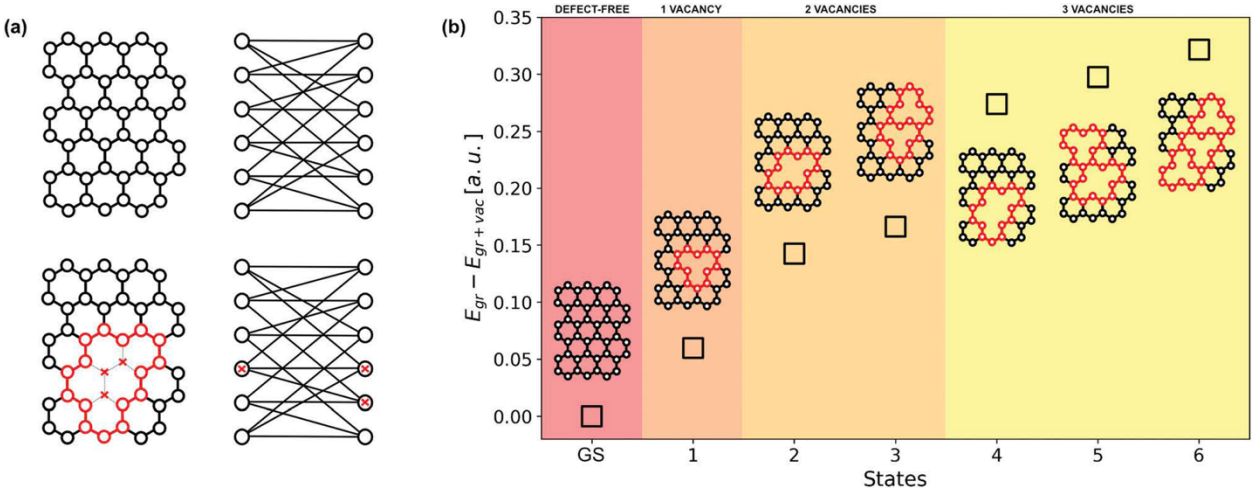

Fig. 2. (a) Left: ball-and-stick (left) and bipartite graph (right) models of free-standing (top) and defective (bottom) graphene. (b) Energy spectrum of the QUBO formulation. A suitable choice for the QUBO parameters permits DW2Q6 to detect many relevant configurations. From Ref. [9].

Quantum chemistry with quantum algorithms on quantum computers: interfacing quantum chemistry software with quantum software. [5, 15]

Near-term quantum devices promise to revolutionize quantum chemistry, but simulations using the current noisy intermediate-scale quantum (NISQ) devices are not practical due to their high susceptibility to errors (Figure 3). This motivated the design of NISQ algorithms leveraging classical and quantum resources. While several developments have shown promising results for ground-state simulations, extending the algorithms to excited states remains challenging. This paper [5] presents two cost-efficient excited-state algorithms inspired by the classical Davidson algorithm. We implemented the Davidson method into the quantum self-consistent equation-of-motion unitary coupled-cluster (q-sc-EOMUCC) excited-state method adapted for quantum hardware. The circuit strategies for generating desired excited states are discussed, implemented, and tested. We demonstrate the performance and accuracy of the proposed algorithms (q-sc-EOM-UCC/Davidson and its variational variant) by simulations of H_2, H_4, LiH, and H_2O molecules. Similar to the classical Davidson scheme, q-sc-EOM-UCC/Davidson algorithms are capable of targeting a small number of excited states of the desired character. The Q-Chem-OpenFermion interface was realized and used for this work. It is available on Github [15].

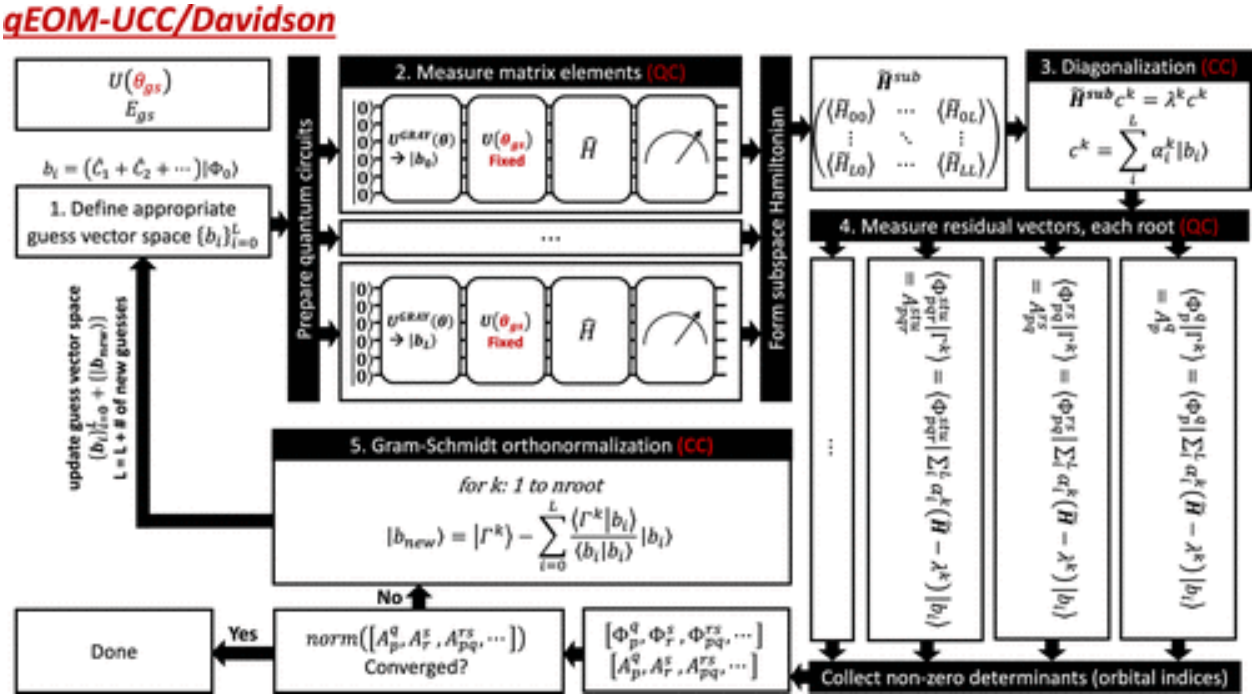

Fig. 3. Sketch of the q-sc-EOM-UCC/Davidson algorithm. "QC" and "CC" refer to the quantum computer and classical computer, respectively. $\{|\Phi_{p\cdots}^{r\cdots}\rangle\}$ and $\{A_{p\cdots}^{r\cdots}\}$ represent excited Slater determinants and their degrees of overlap with residual vector $\{|r^k\rangle\}$. Orbital indices *p*,*q*,*r*,*s*,... refer to spatial orbitals.

Students and postdocs involved in the project.

Kyle Sherbert, PhD 2022, UNT

Anooja Jayaraj, PhD 2022, UNT
Hoa Trinh, USC, PhD not yet received, abandoned work on quantum computing
Samprita Nandi, PhD 2024, USC
Hoda Fatemi Abhari, PhD 2025, USC
Ilaria SIloi, postdoc USC (got a position at the University of Padova, Italy)
Virginia Carnevali, postdoc CMU (got a position at EPFL, Lausanne, Switzerland)
Yonbin Kim, postdoc US (still working at USC)

References, which include project products (the DOI is reported only for project products, highlighted in bold font)

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Other research products.

1. Rosa Di Felice, Gavin O. Jones, James D. Whitfield, Wibe de Jong, Travis Humble, organizers. Symposium "Synergy between quantum computing and high-performance computing in chemistry and materials science", ACS National Meeting Spring 2021, online (previously canceled in Spring 2020 due to COVID-19).
2. Anna I. Krylov, invited speaker: "Quantum chemistry for quantum computing or quantum computing for quantum chemistry?". April 5, 2021. Symposium "Synergy between quantum computing and high-performance computing in chemistry and materials science", ACS National Meeting Spring 2021, online.
3. Ilaria Siloi, Virginia Carnevali, Rosa Di Felice, Marco Fornari, poster presentation: "Modeling order-disorder phase transitions with a quantum annealer", ACS National Meeting Spring 2021, online.
4. Kyle Sherbert, Frank Cerasoli, Marco Buongiorno Nardelli, contributed talk: "Band theory on a quantum computer". APS March Meeting 2021.
5. Frank Cerasoli, Kyle Sherbert, Jagoda Slawinska, Marco Buongiorno Nardelli, contributed talk: "Quantum computation of Silicon electronic band structure". APS March Meeting 2021.
6. Virginia Carnevali, Ilaria SIloi, Rosa Di Felice, Marco Fornari, contributed talk: "D-Wave as a generator of structural models in materials science", APS March Meeting 2021.
7. Ilaria Siloi, Virginia Carnevali, Rosa Di Felice, Marco Fornari, contributed talk: "Modeling order-disorder phase transitions with a quantum annealer", APS March Meeting 2021.
8. Ilaria Siloi, Virginia Carnevali, Rosa Di Felice, Marco Fornari, contributed talk: "Modeling order-disorder phase transitions with a quantum annealer", HPC and Quantum Computing, 3rd Edition, CINECA, Bologna, Italy, December 2020.
9. Virginia Carnevali, Ilaria SIloi, Terry Ethan Stearns, Rosa Di Felice, Marco Fornari, contributed talk: "D-Wave as a generator of structural models in materials science", HPC and Quantum Computing, 3rd Edition, December 2020.
10. Anna I. Krylov, invited speaker: "Q-Chem Open Teamware Project: From Laptops to the Cloud", New Horizons in Scientific Software: From Legacy Codes to Modular Environments Symposium, South Korea (virtual due to COVID-19), November 2020.
11. Anna I. Krylov , invited speaker: "Spin-Forbidden Processes and Molecular Magnetism: New Theoretical Tools for Quantitative Modeling and Insight", Rice University, Houston (virtual due to COVID-19), November 2020.
12. Anna I. Krylov , invited speaker: "Theoretical Treatment of Spin-Forbidden Processes and Molecular Magnetism: Quantitative Tools and Qualitative Analysis", University of St. Andrews, UK (virtual due to COVID-19), September 2020.
13. Ilaria Siloi, Virginia Carnevali, Rosa Di Felice, Marco Fornari, contributed talk: "Modeling order-disorder phase transitions with a quantum annealer", Young Italian conference Quantum Information Science (online), September 2020.
14. Virginia Carnevali, Ilaria Siloi, Terry Ethan Stearns, Rosa Di Felice, Marco Fornari, contributed talk: "D-Wave as a generator of structural models in materials science", Young Italian conference Quantum Information Science (online), September 2020.
15. Frank Cerasoli, Kyle Sherbert, Jagoda Slawinska, Marco Buongiorno Nardelli, contributed talk: "Quantum computation of Silicon electronic band structure". APS March Meeting 2020.

16. Anna I. Krylov , invited speaker : "Theoretical Treatment of Spin-Forbidden Processes: Quantitative Tools and Qualitative Analysis", Photovoltaics workshop, Monash University, Australia, December 2019.
17. Rosa Di Felice, invited speaker: "Materials science and chemistry applications on DWave quantum annealers". December 19, 2019. Workshop on "Quantum Computing and HPC". CINECA, Bologna, Italy.
18. Rosa Di Felice, invited seminar at Calstate Northridge, CA, USA: "Application and performance of adiabatic quantum optimization to a simplified computational biology problem". December 6, 2019. Host: Jussi Eloranta.
19. Rosa Di Felice, invited colloquium at Central Michigan University, MI, USA: "Classical and quantum computation for problems in physical chemistry". October 17, 2019. Host: Marco Fornari.
20. Rosa Di Felice, invited speaker: "Quantum annealing versus classical machine learning applied to a simplified computational biology problem". July 15-19, 2019. Quantum Hiking Conference, Italian Dolomites. Flexible schedule with discussions while hiking and 3 afternoon talks per day.
21. Rosa Di Felice, invited seminar at University of Padova, Italy: "Application and performance of adiabatic quantum optimization to a simplified computational biology problem". June 11, 2019. Host: Stefano Corni.
22. Rosa Di Felice, invited seminar at CalState Long Beach, CA, USA: "Classical and quantum computation for problems in physical chemistry". October 17, 2019. Host: Claudia Ojeda-Aristizabal.
23. Rosa Di Felice, invited colloquium at Center for photochemical sciences, Bowling Green State University, OH, USA: "Classical and quantum computation for problems in physical chemistry". October 17, 2019. Host: Massimo Olivucci.
24. Rosa Di Felice, invited Chemistry colloquium at UC Santa Barbara, CA, USA: "Classical and quantum computation for problems in physical chemistry". October 17, 2019. Host: Lior Sepunaru.
25. Rosa Di Felice, invited speaker: "Quantum annealing versus classical machine learning applied to a simplified computational biology problem". December 18, 2018. Workshop on "Quantum Computing and HPC". CINECA, Bologna, Italy.

Software infrastructure.

Anna I. Krylov, Developed basic software elements for integrating our coupled-cluster codes into Python workflows in preparation to interfacing with OpenFermion [15].