

2023 INCITE Proposal Submission

Proposal

Title: Large-scale simulations of materials for energy and quantum information science

Principal Investigator: Marco Govoni

Organization: Argonne National Laboratory

Date/Time Generated: 6/17/2022 6:40:23 PM

Section 1: PI and Co-PI Information

Question #1

Principal Investigator: The PI is responsible for the project and managing any resources awarded to the project. If your project has multiple investigators, list the PI in this section and add any Co-PIs in the following section.

Principal Investigator

First Name

Marco

Last Name

Govoni

Organization

Argonne National Laboratory

Email

mgovoni@anl.gov

Work Phone

5302200081

Address Line 1

9700 Cass Ave

Address Line 2

(No answer given.)

City

Lemont

State

Illinois

Zip Code

60439

Question #2

Co-PI (s)

First Name

Giulia

Last Name

Galli

Organization

University of Chicago & Argonne National Laboratory

Email

gagalli@uchicago.edu

First Name

Francois

Last Name

Gygi

Organization

University of California Davis

Email

fgygi@ucdavis.edu

Question #3

Institutional Contact: For the PI's institution on the proposal, identify the agent who has the authority to review, negotiate, and sign the user agreement on behalf of that institution. The person who can commit an organization may be someone in the contracts or procurement department, legal, or if a university, the department head or Sponsored Research Office or Grants Department.

Institutional Contact

Institutional Contact Name

Diane Hart

Institutional Contact Phone

(630) 252-7677

Institutional Contact Email

dhart@anl.gov

Section 2: Project Information

Question #1

Select the category that best describes your project.

Research Category

Materials Science: Condensed Matter and Materials

Question #2

Please provide a project summary in two sentences that can be used to describe the impact of your project to the public (50 words maximum)

Project Summary

Using large scale simulations based on quantum mechanics, we tackle two classes of problems: designing (i) sustainable materials to efficiently capture and convert solar energy, and (ii) materials to build novel, optically addressable quantum platforms, including quantum sensors.

Section 3: Early Career Track

Question #1

Early Career

Starting in the INCITE 2022 year, INCITE is committing 10% of allocatable time to an [Early Career Track](#) in INCITE. The goal of the early career track is to encourage the next generation of high-performance computing researchers. Researchers within 10 years from earning their PhD (after December 31st 2012) may choose to apply. Projects will go through the regular INCITE Computational Readiness and Peer Review process, but the INCITE Management Committee will consider meritorious projects in the Early Career Track separately.

Who Can Apply: Researchers less than 10 years out from their PhD that need LCF-level capabilities to advance their overall research plan and who have not been a previous INCITE PI.

How to Apply:

In the regular application process, there will be a check-box to self-identify as early career.

- *The required CV should make eligibility clear.*
- *If awarded, how will this allocation fit into your overall research plan for the next 5 years?*

Projects will go through the regular INCITE review process. The INCITE Program is targeting at least 10% of allocatable time. When selecting the INCITE Career Track, PIs are not restricted to just competing in that track.

- *What is the Early Career Track?*
 - *The INCITE Program created the Early Career Track to encourage researchers establishing their research careers. INCITE will award at least 10% of allocatable time to meritorious projects.*
- *Will this increase my chances of receiving an award?*
 - *Potentially, this could increase chances of an award. Projects must still be deemed scientifically meritorious through the review process INCITE uses each year.*
- *What do I need to do to be considered on the Early Career Track?*
 - *In the application process, select 'Yes' at 'If you are within 10 years of your PhD, would you like to be considered in the Early Career Track?' You will need to write a paragraph*

- about how the INCITE proposal fits into your 5-year research and career goals.*
- *What review criteria will be used for the Early Career Track?*
 - *The same criteria for computational readiness and scientific merit will be applied to projects in the Early Career Track as will be applied to projects in the traditional track. The different will be manifest in awards decisions by the INCITE management committee.*
-

Early Career Track

If you are within 10 years of your PhD, would you like to be considered in the Early Career Track? Choosing this does not reduce your chances of receiving an award.

No

If 'yes', what year was your PhD? If 'no' enter N/A

N/A

If 'yes', how will this allocation fit into your overall research plan for the next 5 years? If 'no' enter N/A.

N/A

Section 4: INCITE Allocation Request & Other Project Funding/Computing Resources

Question #1

OLCF Summit (IBM / AC922) Resource Request - 2023

Node Hours

150000

Storage (TB)

10

Off-Line Storage (TB)

Question #2

OLCF Frontier (Cray Shasta) Resource Request – 2023

Node Hours

25000

Storage (TB)

5

Off-Line Storage (TB)

10

Question #3

OLCF Frontier (Cray Shasta) Resource Request – 2024

Question #4

OLCF Frontier (Cray Shasta) Resource Request – 2025

Question #5

ALCF Theta (Cray XC40) Resource Request - 2023

Node Hours

500000

Storage (TB)

25

Off-Line Storage (TB)

100

Question #6

ALCF Polaris Resource Request - 2023

Node Hours

150000

Storage (TB)

10

Off-Line Storage (TB)

20

Question #7

ALCF Polaris Resource Request - 2024

Question #8

ALCF Polaris Resource Request - 2025

Question #9

ALCF Aurora (Intel Xe) Resource Request – 2023

Node Hours

25000

Storage (TB)

5

Off-Line Storage (TB)

10

Question #10

ALCF Aurora (Intel X^e) Resource Request – 2024

Question #11

ALCF Aurora (Intel X^e) Resource Request – 2025

Question #12

List any funding this project receives from other funding agencies.

Funding Sources

Funding Source

DOE/BES

Grant Number

MICCoM, Early Career Research Program

Question #13

List any other high-performance computing allocations being received in support of this project.

Other High Performance Computing Resource Allocations

Section 5: Project Narrative and Supplemental Materials

Question #1

Using the templates provided here, please follow the [INCITE Proposal Preparation Instructions](#) to prepare your proposal. Elements needed include (1) Project Executive Summary, (2) Project Narrative, (3) Personnel Justification and Management Plan, (4) Milestone Table, (5) Publications Resulting from prior INCITE Awards (if appropriate), and (6) Biographical Sketches for the PI and all co-PI's. Concatenate all materials into a single PDF file. Prior to submission, it is strongly recommended that proposers review their proposals to ensure they comply with the proposal preparation instructions.

Concatenate all materials below into a single PDF file.

- 1. Project Executive Summary (One Page Max)**
- 2. Project Narrative (15 Pages Max)**
- 3. Personnel Justification and Management Plan (1 Page Max)**
- 4. Milestone Table**
- 5. Publications resulting from prior INCITE Awards (if appropriate)**
- 6. Biographical Sketches for the PI and all co-PI's.**

2023_INCITE_combined.pdf

The attachment is on the following page.

PROJECT EXECUTIVE SUMMARY

Title: Large-scale simulations of materials for energy and quantum information science

PI and Co-PI(s): M. Govoni (ANL & UChicago), G. Galli (UChicago & ANL), F. Gygi (UC Davis)

Applying Institution/Organization: Argonne National Laboratory

Resource Name(s) and Number of Node Hours Requested: OLCF/Summit 150k, OLCF/Frontier 25k, ALCF/Theta 500k, ALCF/Polaris 150k, ALCF/Aurora 25k

Amount of Storage Requested: 25 TB

This proposal requests allocations to carry out large-scale quantum simulations of nanoparticles for energy and of point-defects for quantum information science. Our objective is to tackle two classes of problems: designing (i) sustainable materials to efficiently capture and convert solar energy, and (ii) materials to build novel, optically addressable quantum platforms, including quantum sensors. We plan to simulate electronic excited state properties of heterogeneous materials, inclusive of defects and interfaces, using coupled first principles molecular dynamics and electronic structure methods beyond density functional theory (DFT), as implemented in the Qbox (<http://qboxcode.org/>) and WEST (<http://west-code.org/>) open-source codes. Both Qbox and WEST are optimized and have been used on high performance DOE architectures. Qbox is also coupled with a suite of codes (SSAGES) for advanced sampling and WEST is coupled to both Qbox and Quantum Espresso (<https://www.quantum-espresso.org/>). The proposed simulations are planned within the Midwest Integrated Center for Computational Materials (MICCOM; <http://miccom-center.org/>), one of the computational materials science centers funded by DOE, and planned within the Early Research Career Program. Within this program we have planned the coupled use of exascale and near-term quantum computing resources.

We will simulate point defects in wide band gap semiconductors for the realization of qubit and quantum sensors, as well as assemblies of nanostructured building-blocks that are present, e.g., in nanostructured solar cell devices. The main deliverables are: (i) predictions of the structural and electronic properties of heterogeneous systems, to be compared with experiments in order to obtain an integrated mechanistic understanding of the interaction of defective and nanostructured materials with light; (ii) validated data for systems of interest for sustainability and quantum technologies, computed on large-scale DOE platforms, which will serve as reference results.

The functional materials and in general the problems chosen within this project are well aligned with several of the research priorities of DOE, addressing at least three DOE grand challenges (including *design synthesis of tailored properties of materials, correlation of electronic and atomic constituents, quantum information science*). In particular, the study of materials for quantum information technologies will have a significant impact on the science underpinning the National Quantum Initiative and the DOE quantum hubs. We note that the lack of theoretical generalizable models that can guide and help understand a wealth of experimental observations on quantum emitters, has limited the development of candidates for solid-state quantum information technologies to only a few. First principles simulations are key to improving predictions and interpretation of experiments, thus broadening the class of materials that can host defects relevant for QIS. We emphasize that because of the generality of the algorithms and codes used in this project, the validated, reference results obtained here will have an impact on several research fields relevant to the DOE mission, e.g., nanophotonics, neuromorphic computation as well as liquid solar fuels.

All codes used here and developed by the MICCoM team are open-source codes. By making software and research data freely available to the community of users of DOE leadership computing resources, we will broaden DOE's user base of pre-exascale architectures. The findings and results of this project will be published in peer-reviewed journals, and data will be disseminated using data collections, as well as using data curation and exploration tools (Qresp: <http://www.qresp.org/>) developed within MICCoM.

PROJECT NARRATIVE

1 SIGNIFICANCE OF RESEARCH

This proposal requests allocations to carry out large-scale simulations of materials for energy and quantum information science planned within the Midwest Integrated Center for Computational Materials (MICCoM; <http://miccom-center.org/>), and planned within the Early Career Research Program led by Govoni.

MICCoM (PI G. Galli) develops and disseminates interoperable computational tools—open-source software, data, simulation methods, and validation procedures—enabling the simulation and prediction of functional materials for energy conversion (e.g., solar energy) and properties of optically driven solid-state materials for quantum information sciences (QIS). One of the goals of MICCoM is to use quantum simulation codes on high-performance architectures for both validation and material prediction purposes.

The DOE Early Career Research Program (ECRP) funded project “Optical Control of Spin-polarization in Quantum Materials” (PI M. Govoni) aims to understand and control the use of light to manipulate spin-defects in solids for the design of new computing technologies and new nanoscale sensors with enhanced sensitivity.

The simulations proposed in this INCITE project focus on heterogeneous systems composed of atomistic and nanoscale building blocks, and the emphasis is placed on the understanding and characterization of defects and interfaces as well as the prediction of spectroscopic properties.

We will focus here on two specific categories of materials and properties:

- I. Optical and coherence properties of spin-defects in wide band-gap semiconductors for qubits and quantum sensors: simulations will focus on potential and free-energy surfaces of point defects in three-dimensional (3D) solids and electronic properties of spin defects, as well as defective materials under strain fields.
- II. Assembly of nanoscale building blocks and structural and electronic properties of nanostructured materials for energy conversion: simulations will focus on nanoparticle interactions and electronic properties of individual and interacting building blocks.

Results of the simulations will be used both for prediction of material properties as well as to interpret measurements carried out at Argonne National Laboratory (primarily in the experimental group of Dr. Joseph Heremans) and at the University of Chicago (primarily in the experimental groups of Profs. David Awschalom and Dmitri Talapin). Data will be made available in peer-reviewed publications and they will be disseminated using data collections (see, e.g. <http://www.west-code.org/database/index.php>; <http://www.quantum-simulation.org/>) as well as using data curation and exploration tools (Qresp: <http://www.qresp.org/>) developed within MICCoM.

1.1 Adherence to DOE mission

The choice of functional materials and problems, and the simulations planned within this INCITE proposal, are well aligned with several of the research priorities of DOE, addressing at least three DOE grand challenges: (i) design synthesis of tailored properties, (ii) correlation of electronic and atomic constituents, and (ii) quantum information science and quantum technologies.

In particular, the proposed study of novel approaches to solid-state quantum information and engineering for next-generation computation, communication, and sensing has implications for the National Quantum

Initiative. Quantum information science addresses how the fundamental laws of quantum mechanics can be exploited to achieve dramatic improvements in how information is acquired, transmitted, and processed. It is a rapidly developing interdisciplinary field of science and technology, drawing from physical science, computer science, mathematics, and engineering, with investments from several funding agencies and DOE, including the Q-NEXT hub located at Argonne National Laboratory. In this endeavor, theoretical approaches based on numerical simulations are key to designing new solid-state qubits for quantum information, computation, cryptography and sensing. Hence, the activity planned here is well aligned with the DOE focus on exploring new frontiers in quantum technologies. Ultimately, this project will pave the way for and integrate computational and experimental strategies to achieve desired functionalities in solid-state quantum information science by providing robust modeling techniques of materials imperfections. Because of the generality of the algorithms and codes used within this project, we will have impact on several other research fields relevant to the DOE mission, e.g. nanophotonics and neuromorphic computation.

The activity on materials for solar energy and sustainability is well aligned with several DOE energy grand challenges and in particular with several of the questions outlined in the recent round table on solar fuels (https://science.osti.gov/-/media/bes/pdf/reports/2020/Solar_Fuels_Brochure.pdf?la=en&hash=8F16DD447672592D60FB2C35DC6B39911D9BAF02), in particular the following questions: *How can we enable and exploit the direct interaction of solar excitation with chemical change to achieve high selectivity and high efficiency of solar-to-chemical energy conversion?* And: *How can the fundamental science of integration advance the predictive design and control of interfaces and processes to enhance the performance, including durability, of solar fuels systems?* Understanding solar energy absorption in heterogeneous materials is a prerequisite to answering both of these questions.

1.2 Advances enabled by this INCITE award

The results of the simulations performed in this INCITE project will increase our understanding of the atomistic properties of heterogeneous materials, including defective systems relevant to quantum technologies and nanostructured materials relevant to solar energy conversion. The successful completion of the scientific and computational challenges presented in this proposal will strongly benefit from the research activity of the assembled team, that has a long track record of experience in: (i) understanding the equations that govern the fundamental microscopic mechanisms of energy-matter interaction; (ii) developing numerical methods that apply the fundamental equations of quantum mechanics to large scale simulations, with numerous publications in high profile peer-reviewed journals. In addition, the team is collaborating with the Quantum Hub Q-NEXT, and several Energy Frontier Research Centers (EFRCs), including NPQC, Q-MEEN-C, CHOISE and AMEWS.

The realization of this project requires a sizeable use of high-performance supercomputers: INCITE is the most suitable resource for the realization of our project. The total time required for the calculations proposed here cannot be acquired through any other allocation program and the parallelization of the algorithms and codes used in this project could not be exploited on any small-scale computing resource. At present several of our production runs typically use 1,024 nodes. Finally, the massively parallel codes developed by the PI (WEST) and by one of the co-PIs (Qbox) which will be used in this project are accessible to the materials science community via open-source licenses, serving the purpose of broadening the user base of DOE leadership computing facilities.

1.3 Previous INCITE awards

In the following we list previous INCITE allocations which have extensively used the codes Qbox & WEST, which we propose to use within this proposal.

2006. “Computational Spectroscopy of Aqueous Solutions”. G. Galli (UC Davis), D. Prendergast (UC Berkeley), E. Schwegler (LLNL), F. Gygi (UC Davis), J. Grossman (UC Berkeley). 2,500,000 processor hours.

2007-2009. “Water in confined states”. G. Galli (UC Davis), J. Grossman (UC Berkeley), F. Gygi (UC Davis), E. Schwegler (LLNL). 1,500,000, 6,000,000, and 2,000,000 processor hours for each year, respectively.

2010. “Quantum Simulations of Nanostructural Materials for Renewable Energy Applications”. G. Galli (UC Davis), F. Gygi (UC Davis). 1,000,000 processor hours.

2011-2013. “Vibrational Spectroscopy of Liquid Mixtures and Solid/Liquid Interfaces”. G. Galli (UC Davis), F. Gygi (UC Davis). 15,000,000, 25,000,000, and 100,000,000 processor hours for each year, respectively.

2014. “Vibrational and Optical Spectroscopy of Electrolyte/Solid Interfaces”. G. Galli (UChicago), F. Gygi (UC Davis). 70,000,000 processor hours.

2015-2017. “Computational Spectroscopy of Heterogeneous Interfaces”. G. Galli (UChicago), M. Govoni (ANL), F. Gygi (UC Davis). 180,000,000, 150,000,000, and 200,000,000 processor hours for each year, respectively.

2020-2021. “Large-Scale Simulations of Light-Activated Matter”. G. Galli (UChicago), M. Govoni (ANL), F. Gygi (UC Davis). In the first year, 1,200,000 node hours on Theta, in the second year 600,000 node hours on Theta, 290,000 node-hours on Summit.

2 RESEARCH OBJECTIVES AND MILESTONES

As mentioned earlier, this project is focused on light-activated systems: materials for QIS and materials for solar energy conversion. In particular, we focus on **point defects in wide band-gap semiconductors for the realization of qubit or quantum sensors** (Thrust I, described in section 2.1), and **assemblies of building blocks in complex media that are present, e.g., in nanostructured solar cells** and photo-electrodes (Thrust II, described in section 2.2). Our calculations are carried using first principles methods based on Density Functional Theory (DFT) coupled with molecular dynamics (MD) (first principles molecular dynamics (FPMD)), and on many body perturbation theory (MBPT). The Qbox code [Gygi2008] is used for FPMD simulations; both Qbox and Quantum Espresso (QE) [Giannozzi2009] are used for electronic structure calculations at the DFT level (the choice of codes depends on the size of the system, level of DFT, whether semi-local or hybrid DFT [Brawand2016, Brawand2017, Skone2014, Skone2016], is used and on the required k-point sampling). WEST is used for MBPT and quantum embedding theory calculations. Absorption spectra are accelerated with the combined use of WEST and TensorFlow [Dong2021]. Quantum embedding theory calculations are done by coupling Qbox and WEST [Sheng2022]. The resulting effective Hamiltonians, with parameters derived from first principles calculations, are diagonalized with Pyscf on classical computers and with Qiskit on near-term quantum computers. The latter step (diagonalization on classical or quantum computers) will not be performed at ALCF or OLCF, but will be carried out on mid-size clusters at Argonne (LCRC, RCC) or using the IBM Quantum Experience. A description of the methodologies and codes used in the project is given in section 3.

2.1 Optical and coherence properties of spin defects

A grand challenge in quantum information science and engineering is represented by **the design of materials that can host qubits that are both robust and easily controllable**. A few physical platforms have been studied including superconducting circuits, trapped atoms and ions, semiconductors, and ion-doped insulators. Here we focus on color centers in semiconductors and insulators.

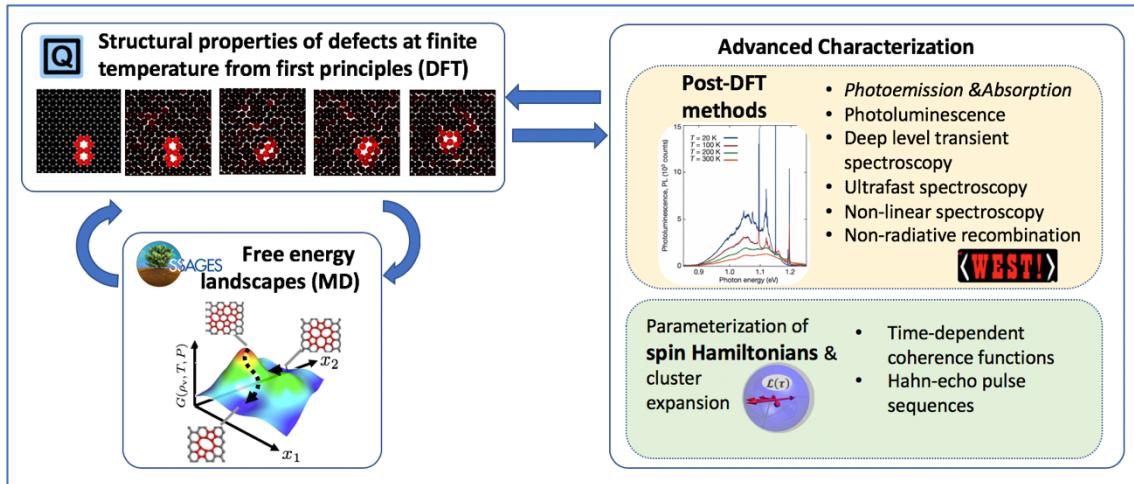


Figure 1 - List of current (italics) and future spectroscopic characterization techniques (right panel) and their coupling with first principles molecular dynamics (FPMD) simulations and advanced sampling (left panels). FPMD are carried out at the density functional and hybrid density functional theory (DFT) level.

Qubits encoded by electronic states of defects in semiconductors are promising candidates for room temperature quantum information processing and for quantum sensors because they exhibit spin states with coherence times long enough for measurements to be carried out before the quantum information is destroyed. Initialization, manipulation, and readout operations have been demonstrated experimentally using optical absorption, ground state microwave pulses, and photoluminescence, respectively [Castelletto2013, Christle2015, Fuchs2015, He2015, Koehl2015, Lienhard2016, Seo2016a, Seo2016b, Weber2010, Whiteley2019, Widmann2015].

Predictive modeling of defects based on first-principles numerical simulations (see Fig.1) will provide a detailed microscopic understanding of the physical processes occurring in quantum light emitters following excitation [see, e.g. Seo2016b, Castelletto2013], which is crucial to then develop new classes of color centers for quantum technology applications.

We describe below the main milestones of this thrust.

2.1.1 Formation energy and electronic properties of point defects in wide band gap semiconductors

Qbox and Quantum Espresso (see section 3) will be used to compute defect formation energies at zero and finite T, respectively, and Qbox will be employed for large-scale calculations using hybrid functionals for which good scaling on the ALCF Theta platform has been demonstrated (see section 3). WEST will be used to compute electronic properties of point defects beyond DFT, including zero-phonon lines, absorption and photoluminescence spectra (the capability to efficiently compute photoluminescence spectra is being built and tested within MICCoM; all timings given below include only calculations of absorption spectra). In some cases, we will use Qbox coupled with the SSAGES suite of codes [Sidky2018, Cunzhi2020] to

compute free energy surfaces of defects. SSAGES is a suite of codes for phase space sampling developed within MICCoM; it includes (1) standard, untempered metadynamics, (2) the nudged elastic band method, (3) forward flux sampling, (4) Basis Function Sampling, (5) Adaptive Bias Force, (6) Finite Temperature String, (7) Swarm of Trajectories and (8) static bias/umbrella sampling. In the coupling of Qbox with SSAGES, the latter occupies a fraction of a node, and the performance is entirely dominated by the efficiency of Qbox.

We will investigate the formation mechanism and the formation energy of defects in diamond thin films and interfaces of diamond and SiC polytypes (with focus on double vacancies VV^0 and vacancies coupled with transition metals (e.g., Cr and V) in SiC and N, Si and Ge vacancy pairs in diamond films). Each of the four systems has ~ 1000 atoms, and will be simulated using an energy cutoff of 60 Ry. Formation energies of defects in bulks will first be validated against literature results. These systems and properties have been chosen due to active collaborations of the team with experimentalists at ANL and UChicago dedicated to the study of these systems for quantum technologies. While electronic property and formation energy validation will be carried out for a wide variety of defect charge states and combinations of vacancies and impurities, free energy calculations will be investigated only for a subset of those identified as promising. Based on the computer time recorded for DFT-PBE calculations of SiC defects up to ~ 700 atoms and hybrid DFT calculations of SiC defects up to 432 atoms with Qbox, and considering that we plan to study 5 samples at the DFT-PBE level at zero temperature and ~ 8 samples at the hybrid-DFT level, we estimate the cost of this part of the project to be **300,000 node hours on ALCF/Theta**. We emphasize that calculations with these samples have been thoroughly tested for convergence in terms of numerical parameters (including pseudopotentials, basis sets, k-point sampling, level of bisection) on Theta. These calculations were used for estimates, considering that runs no longer than 50 ps will be carried out for about 2 samples at the DFT-PBE level of theory. It is reasonable to assume that we will carry out simulations both at the PBE and hybrid level. The exact number of hours (determined in particular by the balance between PBE and hybrid-DFT calculations) will depend on the results obtained. We will determine the thermodynamical stability of defects under various conditions, e.g., at the proximity of heteropolytypic interfaces.

2.1.2 Spectroscopic signatures of point defects in wide band gap semiconductors

We will use calculations carried out with WEST, on snapshots generated by Qbox, to identify spectroscopic signatures of promising defects and we will compare our results with measurements carried out at Argonne National Laboratory and the University of Chicago. We will focus on photoluminescence spectroscopy, optically detected magnetic resonance, and spin coherence time measurements. In particular, we will use WEST to compute optical signatures of defects: zero-phonon line energy and linewidth, phonon sideband, Debye-Waller factor, including weak and strong correlation as implemented in WEST's quantum defect embedding theory (see Fig. 2). Spin coherence times will be simulated using the pyCCE software. Several pyCCE runs will be collected and submitted using ensemble job submission scripts. We will determine the sensitivity of quantum characteristics to materials properties. All validating measurements will be performed at ANL for defects in diamond (NV^- , SiV^- , GeV^-) and in SiC (VV^0 , VSi^- , NV , Cr_{Si}^-). The estimate of times for this part is based on the same sample sizes used in section 2.1.1 for both DFT-PBE and hybrid DFT, and for each sample we considered zero-phonon line and absorption calculations. Based on the samples utilized for our tests, we estimate the budget of this part of the project to be **150,000 node hours on OLCF/Summit and 150,000 node hours on ALCF/Polaris**. We will run on Polaris the simulations of point defects in interfaces which, due to their size, will benefit from the advanced memory and bandwidth of NVIDIA A100s (note that the WEST code has been demonstrated at full scale on both Summit and Perlmutter phase 1).

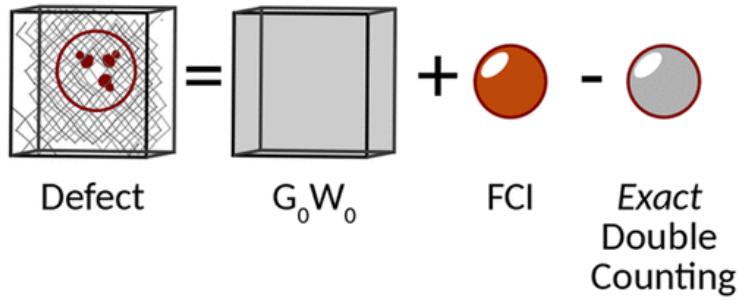


Figure 2 A Green's function formulation of the quantum defect embedding theory (QDET) allows us to study strongly correlated states within a periodic crystal. A double counting scheme is rigorously derived within the GoWo approximation and implemented in the WEST code. Adapted from Ref. [Sheng2022].

2.2 Assembly of nanoscale building blocks

We will address the dynamical and electronic properties of nanostructured materials for solar energy conversion (see Fig. 3), with focus on III-V compounds, which are being synthesized at the University of Chicago in the group of Prof. Talapin. Nanoparticles incorporating Cd and Pb have achieved the best performance in terms of emission colors spanning the visible spectrum, luminescence quantum yield, and narrow emission line widths. However, they contain toxic elements. InP quantum dots and other III-V compounds are non-toxic and retain the spectral range of size-tunable emission while expanding the scope of applicability of soluble semiconductor emitters to lighting and display technologies, and to imaging applications. We aim to simulate and predict the electronic properties (WEST) of these dots, the structural properties (at the atomistic level, Qbox) of their surfaces and interfaces, building on our work on InAs [Scalise2018].

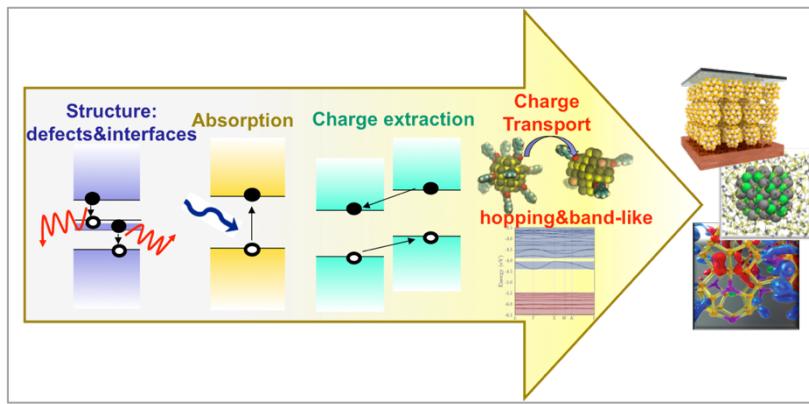


Figure 3 Schematic representation of the physical processes investigated in thrust II of this project (assembly of nanostructured materials for solar energy conversion). We will investigate charge (electron and hole) transport in ensembles of nanoparticles (NPs). Structural and electronic properties (absorption, photoemission and band offsets) will be obtained from calculations based on density functional and many body perturbation theory.

2.2.1 Structural and electronic properties of single dots and of assemblies of dots

We will simulate assemblies of III-V semiconductor dots, following the strategy adopted for InAs [Scalise2018] and illustrated in Fig.3. We will compute FPMD trajectories for specific ligands used experimentally and we will compare our results with Raman and XPS experiments and we will identify promising motifs for the assembled structures by building ab-initio stability diagrams. The validation strategies and results obtained here will impact application ranging from solar to thermal energy conversion (although here we focus only on solar energy conversion). Based on the calculations carried out on samples of the same size as those considered here for InAs [Scalise2018] and based on our plans on using the same functional for structural properties (PBE), we estimate **200,000 node hours on ALCF-Theta** for the structural properties reported in the blue panel of Fig. 4 (FPMD and ab initio stability diagrams). Timing are based on runs obtained on Theta with a previous INCITE allocation. We considered the $In_{38}Sn_{28}Sn_{106}S_{121}$ sample for testing purposes, although the number of Sn and S atoms (composing the

matrix where the nanoparticles are immersed) may slightly vary. The experiments for the validation (red panels in Fig. 4) are already available for InP and data are being collected for InP alloyed with Ga.

Electronic properties will be computed both at the DFT and MBPT theory level. Different exchange and correlation functionals will be tested, and possibly, for some configurations, GW calculations (WEST) will be carried out for comparison. Optical spectra will be computed at the BSE level starting either from GW or from hybrid DFT results, depending on the quality of the comparison with experiments obtained in the two cases. Qbox electronic structure calculations coupled with WEST capabilities for the calculation of lifetimes will also provide access to modeling electronic mobilities that, using suitable models, may be qualitatively compared with those observed experimentally. We estimate the cost of this part of the project to be **25,000 node hours on Frontier and 25,000 node hours on Aurora**. All estimates are based on the same system sizes as for InAs and on spectra evaluated starting from single particle wavefunctions obtained at the hybrid DFT level. **Electronic properties will be computed in the second half of 2023.**

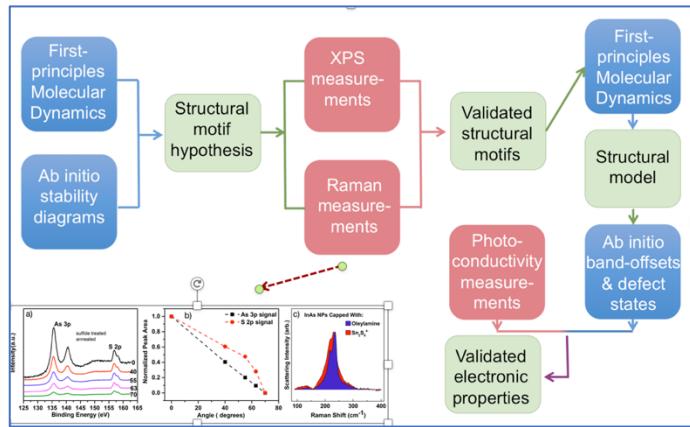


Figure 4 Schematic representation of the calculations, blue panels, including experimental validation, red panel, that are planned for assembly of nanoparticles (see text).

3 COMPUTATIONAL READINESS AND PARALLEL PERFORMANCE

The scalability of our codes has been tested on ALCF-Theta and on OLCF/Summit.

The **Qbox** code has undergone specific optimizations targeting the Intel KNL architecture thanks to Early Science Programs both at NERSC (NESAP – targeting Cori) and at ALCF (Theta ESP – targeting Theta). Fig. 5 shows excellent scaling of Qbox for the Hartree-Fock exact exchange calculation in hybrid-DFT calculation 2,048 KNL nodes (131,072 cores) which are a key part of this proposal. An example is shown for a typical insulator (SiC) that will be used in this project to study defects for quantum information science.

As mentioned above, **WEST** is interfaced with **Quantum Espresso** and **Qbox**. WEST has undergone specific optimizations targeting the Intel KNL architecture thanks to Early Science Programs both at NERSC (NESAP – targeting Cori) and at ALCF (Theta ESP – targeting Theta). The strong and weak scaling of WEST up to 2,048 KNL nodes (131,072 cores) is shown in Fig. 6 for a simulation of a (CdSe)₃₄ nanoparticle (884 electrons), a representative building block of nanostructured materials studied here. WEST has also been used to carry out G₀W₀ full-frequency simulations for systems of ~10,000 electrons using the entire Summit computer at OLCF. WEST has been ported to work at full scale on NERSC/Perlmutter (NVIDIA A100 GPUs) thanks to a Tier 1 NERSC NESAP program.

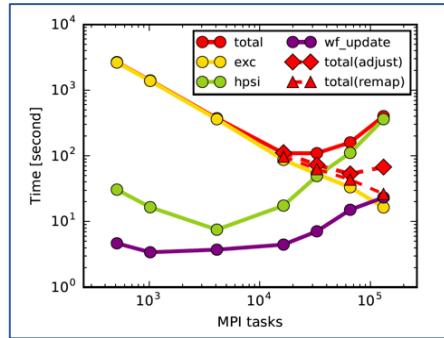


Figure 5 - The strong scaling of Qbox hybrid-DFT calculations was extended on Theta by optimal data remapping methods (red triangles). An example is shown for solid (SiC_{256}) using the PBE0 functional.

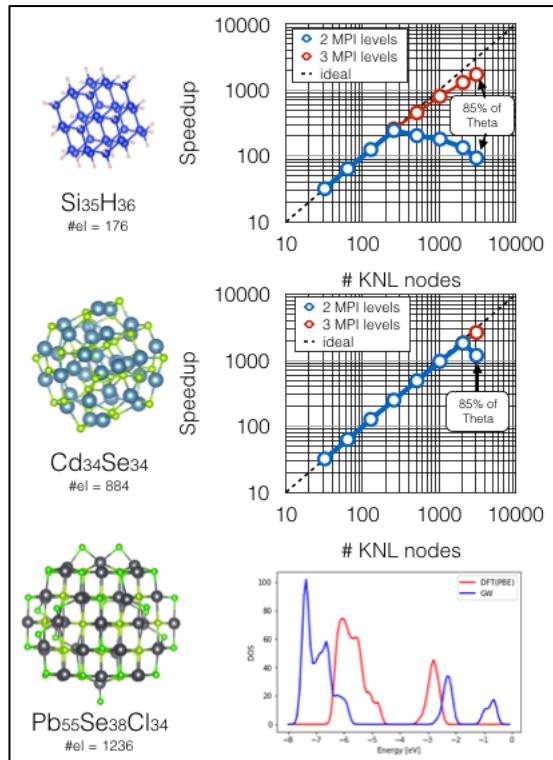


Figure 6 WEST strong scaling on Theta. The original layout of the code includes two MPI layers of parallelization: (1) parallelization of fast Fourier transforms (FFTs); (2) parallelization over eigenvectors of the dielectric matrix (also called PDEP). The product of the total number of grid points over which FFTs are performed, and the total number of eigenvectors $N_{FFT} \times N_{pert}$ determines the code strong scaling limit of WEST (blue dots). A third MPI layer over electronic states was recently introduced. With the new 3 MPI layers implementation the new strong scaling limit of WEST is $N_{FFT} \times N_{pert} \times N_{band}$, allowing users to scale both small and medium size systems to the full machine (red dots). Examples on nanoparticles studied in this INCITE project are shown (left column), where calculations are performed both at the DFT and GW level.

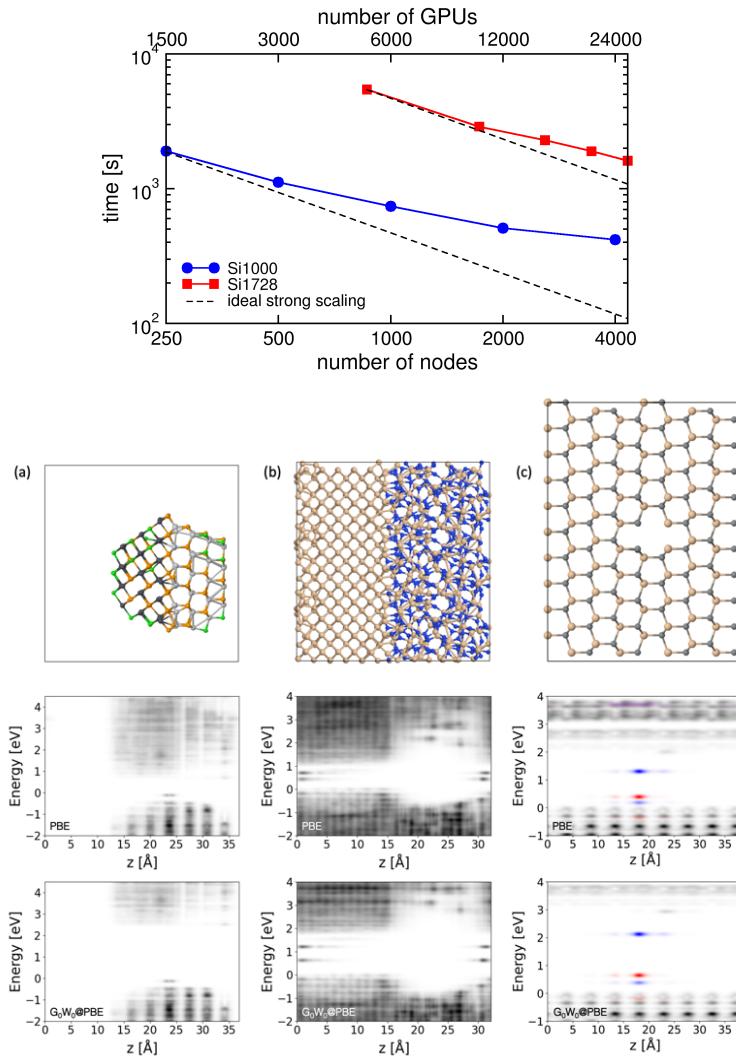


Figure 7 – Top sub-figure. Strong scaling of the GPU version of WEST on the Summit supercomputer for two silicon supercells containing 1,000 atoms (blue circles) and 1,728 atoms (red squares), respectively. The black dashed lines indicate the slope of ideal scaling. 80 quasiparticle energies (around the Fermi level, 40 below and 40 above) were calculated for each system. Timing results correspond to the total wall clock time, including the time spent on I/O operations and CPU-GPU communications. **Bottom sub-figure.** Large-scale full-frequency G_0W_0 calculations using the GPU version of WEST: (a) a Janus-like heterostructure formed by a chlorine-terminated nanoparticle made of cadmium sulfide and lead sulfide (2,816 electrons), (b) an interface of silicon and silicon nitride (10,368 electrons), and (c) a neutral hh divacancy in 4H silicon carbide (16,392 electrons, spin-polarized). The top panels report a side view of the simulation cells. For the ball-and-stick atomic structures the Cl, Cd, S, Pb, Si, N, C atoms are colored in green, black, orange, light gray, beige, blue, and dark gray, respectively. The bottom and middle panels report the local density of states obtained using $G_0W_0@PBE$ or DFT energies, respectively. A color scale that ranges from white to black is used to plot the LDOS; white areas indicate energy gaps. For the hh divacancy in SiC, the defect states in the up (down) spin channel are shown in red (blue). Adapted from Ref. [Yu2022].

The simulations performed here will be executed by the PI and co-PIs together with graduate students and post-doctoral research associates. To facilitate code deployment and fast turn around and the efficient use of Qbox-WEST coupling capabilities we have recently created containers running both on HPC clusters and ANL-Theta and insured that no loss of performance was observed, as illustrated in Fig. 8.

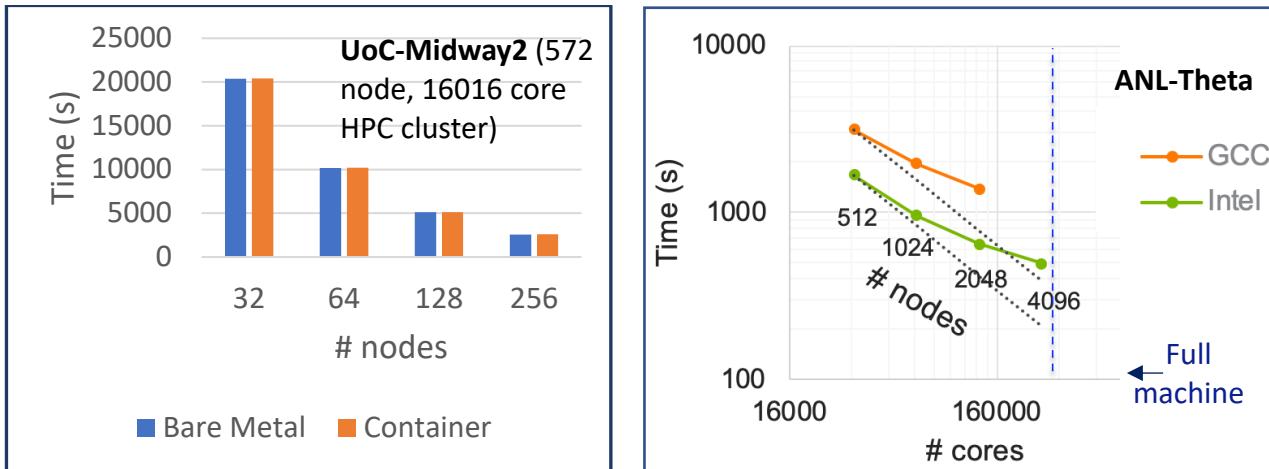


Figure 8 - Performance of a container with the following software stack on a HPC cluster (left panel) and on ANL-Theta (right panel): Intel compiler (2019.up4); Intel MKL (2019.up4); MPICH (3.3) Python 3.7.6 (PyPi); West (4.0.1); Qbox (1.68.2). The timings are for a G_0W_0 calculation of a nanoparticle with 884 electrons (representative of the sizes which will be considered here (see Thrust II), carried out compiling the codes with either GCC or the Intel compiler, as a function of the number of nodes.

Finally, we note that scientific discussions are expected to take place with all team members through weekly meeting. Should any computational setbacks arise, they will be solved by the developers of the codes present in the team (Gygi for Qbox and Govoni for West) and an efficient management of the allocated resources is assured by the assistance of ALCF/OLCF personnel. Several graduate students and postdoctoral researchers will benefit from the proposal through mentorship in use of high-performance computing facilities for large-scale simulations. The team will make timely and effective use of the INCITE allocation either with single jobs that require >30% of the machines or with ensemble jobs that bundle multiple calculations together into single job submissions (improving throughput in the queues). The team expects to use less than 10% of the current allocation for jobs below the capability size, dedicated to analysis and post-processing.

3.1 Use of Resources Requested

The simulations described in the previous sections amount to the following requested computer time:

Thrust I.1 ~300,000 node hours on ALCF/Theta
Thrust I.2 ~150,000 node hours on OLCF/Summit
Thrust I.2 ~150,000 node hours on ALCF/Polaris

Thrust II.1 ~200,000 node hours on ALCF/Theta
Thrust II.1 ~25,000 node hours on ALCF/Aurora
Thrust II.1 ~25,000 node hours on OLCF/Frontier

The total time requested is 500,000 node hours on ALCF/Theta, 150,000 node hours on ALCF/Polaris, 150,000 node hours on OLCF/Summit, 25,000 node hours on ALCF/Aurora, 25,000 node hours on OLCF/Frontier. All estimated times on Theta and Summit were based on timings acquired during access to the machines. Time on Polaris is inferred based on time acquired on NERSC/Perlmutter phase 1 through NESAP Tier 1. Times on Aurora and Frontier are inferred based on the single node comparisons with ThetaGPU and current development done at ANL/JLSE (<https://www.jlse.anl.gov/>). Detailed milestones are reported in the corresponding, separate table, and number of atoms and functional chosen for all tests are specified in the sections where each thrust is described.

3.2 Data strategy

Total data storage requirements are estimated at 25 TB (maximum). For each system of the size planned here studied with WEST, one needs $\frac{1}{2}$ TB for the generation of dielectric screening (WSTAT) and $\frac{1}{2}$ TB for full GW calculations. These files are necessary to restart the calculations. When carrying out BSE calculations, an additional 1TB is necessary for the concurrent use of Qbox and WEST. A Qbox restart file for a 512-atom SiC sample (60 Ry) takes \sim 6.2 GB. A simulation that saves restart files at regular intervals for later analysis would require approximately 1000 restart files, i.e. a total of \sim 6 TB. If we consider three samples (MD and spectroscopic properties running at a time), we get to an estimate of 25 TB. Several of these files must be available for archival, yielding an estimated value of $25 \times 4 = 100$ TB.

The dissemination plan of this proposal includes making data of simulations available at <http://www.quantum-simulation.org/reference/index.htm>, where FPMD simulations data are already collected, and electronic structure data at <http://www.west-code.org/database/index.php>, where several collections are already online [LaCount2019, Dawson2018].

As mentioned earlier, all codes used in this project and developed by the team are open-source, freely available to the community of users capable of using leadership computing resources. By awarding this project, DOE will enable the team to conduct high-risk, high-payoff large scale simulations on quantum information and energy problems. A growing interest of the community in our open-source software, developed specifically to take advantage of the DOE massively parallel computing facilities, is therefore expected, which will increase and **broaden the community of researchers capable of using next generation leadership computing resources**. The scientific findings of this project will be published in peer-review journals, and the results of the simulations will be exported in the form of digital contents to be disseminated in national and international conferences (in particular APS, ACS and MRS), press articles and web pages. Data obtained in this project will be collected and integrated within the MICCoM data task and disseminated using Qresp (<http://www.qresp.org/>) [Govoni2019]. The scientific outcomes and computational advances achieved in this proposal will also be used as didactic material in advanced schools and workshop that will be coordinated with the MICCoM DOE center.

3.3 Computational Approach

The computations presented in this project will be performed in the framework of DFT and MBPT, carrying first principles molecular dynamics simulations (FPMD) and GW/BSE/quantum embedding calculations described below in 3.2.1 and 3.2.2, respectively.

3.3.1 First principles molecular dynamics

The cornerstone of all our calculations is the FPMD method, which will be used to derive finite-temperature structural and electronic properties of solid-state defects and nanostructured materials. Within FPMD, the Newton equations of motion (Ordinary Differential Equations) of a set of atoms are solved by finite differences, and the interaction between atoms is computed at each step within DFT, by solving the **Kohn-Sham (KS) equations**. The latter are non-linear partial differential equations (PDE) solved self-consistently using iterative techniques. We plan to employ both **semi-local (GGA) and hybrid functionals**, depending on the system and the property under investigation. Hybrid functionals require the evaluation of the Hartree-Fock exchange for which we will adopt a **space decomposition technique** developed by team member F. Gygi [Gygi2009, Dawson2015]. This method allows one to carry out large scale FPMD simulations with hybrid functionals, with controlled accuracy in spite of approximations used in computing the overlap between electronic single-particle wave functions.

Recently the team has developed **sc-hybrid functionals**, where the fraction of exact exchange depends on the material dielectric properties [Brawand2016, Brawand2017, Skone2014, Skone2016, Zheng2019]. These functionals have already been successfully used for color centers in semiconductors and insulators and for heterogeneous materials (e.g. solid-liquid interfaces) and will be used in this project for both defects and nanostructured systems.

3.3.2 Many Body Perturbation Theory: *GW*, *BSE* and quantum embedding calculations

Band gaps, band edges, defect levels and spectra will be computed using MBPT for snapshots extracted from FPMD simulations or for optimized zero-temperature structures. The **Dyson and Bethe-Salpeter equations** (PDE) will be solved using both (i) Density Functional Perturbation Theory (DFPT) [Baroni2001] using iterative techniques developed by the PI's group [Wilson2008, Pham2013] and applied to large scale calculations [Govoni2015, Govoni2018], and (ii) by performing calculations in finite field [Nguyen2019, Ma2019]. The details of the theory based on DFPT are given in a review paper [Ping2013] and the details of the computational procedure can be found in Refs. [Govoni2015, Govoni2018]. Details of the basic principles of MBPT can be found in Ref. [Onida2002].

In sum, using strategy (i) a perturbative solution of the Dyson equation is obtained in two steps: an iterative diagonalization of the dielectric matrix is performed, via a Davidson diagonalization. At each step several linear systems are solved using the conjugate gradient method to obtain the linear variation of the charge density, in the DFPT frameworkT [Baroni2001]. A subsequent expansion of the Green's function and of the screened Coulomb potential in a separable way is carried out, followed by a frequency convolution via a Lanczos iterative algorithm [Rocca2008]. The iterative diagonalization of the dielectric matrix uses a deflation technique that does not require the calculation of already-converged eigenvectors.

Using strategy (ii) and computations in finite fields, we not only avoid the calculation of virtual electronic states (as done already within DFPT), but we also bypass altogether the calculation of dielectric matrices. In addition, our formulation uses linear combinations of Bloch orbitals that are localized in appropriate regions of real space [Gygi2009], leading to substantial computational savings [Nguyen2019, Ma2019]. Several advantages were demonstrated: calculations beyond the random-phase approximation become straightforward, and the complexity and scaling of solving the BSE [Rocca2012] is the same when using local or hybrid-DFT calculations as starting points. When using GW, we were able to carry out calculations with vertex corrections. The finite field method led to an **improvement in both accuracy and efficiency in the calculations of optical spectra of large molecular and condensed systems, and to their coupling with FPMD**. Using this coupling, we computed exciton binding energies of several molecules and absorption spectra of condensed systems of unprecedented size, including water samples with hundreds of atoms [Nguyen2019, Ma2019]. In addition, using the coupling we developed a quantum embedding technique [Ma2020] that is applicable, for example, to defects in semiconductors and to the study of qubits.

We reduced the scaling of BSE calculations and have computed exciton binding energies of several molecules and absorption spectra of condensed systems, including water samples with hundreds of atoms. The formulation implemented in Ref. [Nguyen2019] uses linear combinations of Bloch orbitals that are localized in appropriate regions of real space using the recursive bisection method, thus leading to substantial computational savings. We note that calculations beyond the random-phase approximation become straightforward in finite field, and the complexity and scaling of solving the BSE is the same when using local or hybrid-DFT calculations as starting points. The same finite field approach can also be used to perform GW calculations and obtain quasi-particle orbitals, through the evaluation of the eigenvectors of the irreducible density-density response function. Recently, a method based on **machine learning was introduced to speed-up MBPT calculations** [Dong2021]. In particular, the data sets exchanged by WEST and Qbox for the calculation of absorption spectra were used to train a convolutional neural network surrogate model of the dielectric screening computed in finite field. Interoperability between WEST and

Tensorflow (<https://www.tensorflow.org/>) enabled computational gains of up to two orders of magnitude for the calculation of absorption spectra of molecules and materials at finite temperature.

The ability to compute the screened Coulomb interaction efficiently and at-scale on DOE facilities has made possible quantum embedding calculations [Sheng2022]. In the presence of specific localized electronic states, e.g., those associated with spin defects in semiconductors or insulators, one may obtain geometries and energies of excited states by performing DFT calculations with constrained occupation numbers. In addition, the effect of the environment around the defect may be incorporated in the calculation through the definition of an effective Hamiltonian including a screened Coulomb interaction. Using these two concepts, we have built a quantum embedding theory that goes beyond the constrained random phase approximation [Ma2020, Ma2021, Sheng2022]. The accuracy and effectiveness of this approach was demonstrated by investigating several spin defects in semiconductors that are of interest for quantum information technologies. Importantly, the proposed quantum embedding theory is able to capture strongly-correlated electronic states of active regions (defined by the localized electronic states of the defects), with the rest of the system described within DFT. An important consequence of the quantum embedding theory, as demonstrated in Ref. [Ma2020, Huang2022] is that an effective Hamiltonian describing strong correlations within the active site may be defined and solved on classical as well as on **quantum computers** using interoperability schemes with quantum chemistry and quantum computing software packages, e.g., PySCF and Qiskit (<https://qiskit.org/>). Quantum computers promise, in principle, a much more favorable scaling as a function of system size than classical architectures although noise and quantum correction issues are still affecting the accuracy of results obtained on near-term quantum computers and substantial progress is needed before complex materials science simulations may be carried out on quantum computers.

We report below the details of the codes used in this project.

3.3.3 Codes used: WEST, Qbox, Quantum Espresso and pyCCE

In this project we will use the WEST, Qbox, PyCCE and Quantum Espresso codes. They are briefly described below.

WEST [Govoni2015, Govoni2018, Yu2022] **performs large-scale many-body perturbation theory** (MBPT) calculations providing electronic and optical spectroscopic characterization of complex materials. The distinctive features of WEST (<http://www.west-code.org>) are as follows: use of DFPT [Baroni2001] to avoid explicit calculation of virtual electronic states; use of spectral decomposition of dielectric matrices (no explicit diagonalization and storage of the full matrix are required); calculation of GW self-energy with full frequency integration; and perturbative calculations starting from semi-local and hybrid DFT wave functions. WEST achieves optimal scaling on both ALCF/Theta and OLCF/Summit (full machine runs demonstrated, [Yu2022]). WEST implements MBPT inclusive of spin-orbit coupling [Scherpelz2016], k-point sampling for solids, and electron-phonon interactions without empty states [McAvoy2018]. WEST and Qbox are coupled to enable GW [Nguyen2019] and Bethe-Salpeter-Equation (BSE) [Ma2019] calculations in finite fields. Input/output (I/O) are structured in JavaScript Object Notation format, thus enabling seamless integration with pre-/post-processing tools and compatibility with Jupyter notebooks. Pre- and post-processing is carried out with Python scripts, including analysis tools, an interface with Qbox and QuantumEspresso. The documentation is generated using markup language and the popular Sphinx tool (<http://www.sphinx-doc.org>). WEST is available on GitHub at <https://github.com/west-code-development>. A private GitLab server is also available (<http://greatfire.uchicago.edu>) to allow for flexible management of internal development. Continuous integration is in place to automatically test the integrity of the code at every addition. Qbox-WEST coupling is described in detail in the next sections. WEST is also coupled to TensorFlow for speeding up the calculation of absorption spectra using machine learning [Dong2021]. The WEST code features an efficient implementation of the quantum embedding defect theory (QDET) [Sheng2022], which has enabled the study of strongly correlated electrons in point-defects.

Qbox [Gygi2008] is a first-principles molecular dynamics (FPMD) code. Notable features of the Qbox code include DFT and hybrid-DFT MD simulations (using the Recursive Subspace Bisection [RSB] algorithm [Gygi2009, Dawson2015] that reduces the cost of Hartree-Fock exchange calculations, van der Waals density functionals [Wu2012] and the SCAN functional [Sun2015, LaCount2019], and on-the-fly calculation [Gygi2003] of maximally localized Wannier functions (MLWFs) for the computation of vibrational spectra. A client-server feature allows for the use of Qbox as a *quantum engine* controlled by an external driver program. This offers the possibility of controlling multiple FPMD simulations from a single driver program. This client-server feature has been used to implement coupling with the SSAGES suite of codes for free energy calculations [Sidky2018, Sevgen2018] with LAMMPS [Plimpton1995, Puli2017], and with WEST. In addition, Qbox has been interfaced with the code BoltzTrap [Madsen2006, Puli2019] and compressive sensing lattice dynamics calculations. We also implemented user-friendly modules for infrared (IR), Raman, and sum-frequency generation spectroscopies, and ionic conductivity [Rozsa2018] and hybrid functionals for interfaces [Zheng2019]. Qbox makes extensive use of the XML markup language in input and output, thus facilitating post-processing by automatic tools and the curation of simulation datasets. A number of such data collections have been made available at www.quantum-simulation.org. Qbox is available on GitHub at <https://github.com/qboxcode>. A private GitLab server has also been installed (<http://scherzo.ucdavis.edu>) to allow for flexible management of internal development.

Qbox and WEST are coupled within a client-server strategy that has permitted the reformulation of GW and BSE calculations in finite field (instead of using density functional perturbation theory). In particular, we recast, the calculation of the screened Coulomb interaction in finite field, which is performed in Qbox with both semi-local and hybrid functionals. Messages between the two codes are exchanged through the file system using appropriate synchronization system calls. We managed to reduce the latency of the client-server messages, without compromising the parallel performance of the codes. We have also successfully run it on ANL-Theta. Using this coupling, we computed exciton binding energies of several molecules and absorption spectra of condensed systems of unprecedented size, including water samples with hundreds of atoms [Nguyen2019]. In our implementation, WEST is the driver program of multiple Qbox instances, which initially perform a wavefunction optimization step to compute the electronic ground state of the system. WEST generates the perturbation δV , and the self-consistent calculations are solved by Qbox, which returns the linear variation of the charge density δn . Data for both δV and δn are exchanged by the two codes using an XML file that conforms to the annotated function3d.xsd XML schema. The coupling between WEST and Qbox facilitates the execution of response commands by keeping in memory the electronic structure of the system [Govoni2021].

In addition to computational savings for GW and BSE, the coupling of Qbox and WEST to carry out molecular dynamics in finite field has allowed us to compute IR and Raman spectra and ionic conductivities of molecular materials and to develop new hybrid functionals for interfaces and in general heterogeneous systems [Zheng2019].

Quantum ESPRESSO (QE) is a DFT electronic structure code [Giannozzi2009, Giannozzi2020] used here for some zero-T calculations, especially in the case of those defects for which fine k-point grids are required. The code uses a hybrid OpenMP-MPI parallelization scheme and, although self-contained, can take advantages of highly optimized libraries like BLAS, LAPACK, SCALAPACK, FFTW3, ESSL, PESSL, GDLib and the GSL library. QE is parallelized on different levels: k-points (linear scaling with the number of processors), bands and plane waves (high cpu scaling and RAM distribution), FFTs are parallelized over tasks (improves load balancing) and the lowest level parallelizes linear algebra (improves scalability and distribute RAM). The porting of QE to NVIDIA GPUs is available [Giannozzi2021]. Both computation and data structures are distributed in order to fully exploit massively parallel architectures. Co-PI Marco Govoni is in contact with the developers of the QE project and contributes regularly to the source.

PyCCE is an open-source Python library to simulate the dynamics of spin qubits in a spin bath, using the cluster-correlation expansion (CCE) method. PyCCE includes modules to generate realistic spin baths, employing coupling parameters computed from first principles with electronic structure codes, and enables the user to run simulations with either the conventional or generalized CCE method. PyCCE enabled the calculation of the calculation of the Hahn-echo coherence time of the nitrogen vacancy in diamond; the calculation of the coherence time of the basal divacancy in silicon carbide at avoided crossings; and the magnetic field orientation-dependent dynamics of a shallow donor in silicon. The complete documentation and installation instructions are available at this URL (<https://pycce.readthedocs.io/en/latest/>).

3.4 Development Work

The WEST code is already ported to work on NVIDIA GPUs (including A100s on Perlmutter). The PI has demonstrated simulations with the WEST code to scale on the entire Summit machine reaching ~60PF (including time spent on I/O). The porting of WEST is described in a manuscript submitted for peer-review in J. Chem. Theory Comput [Yu2022]. A version of Qbox that works on NVIDIA GPUs has recently been developed with support of semilocal functionals. The porting to NVIDIA GPUs of pyCCE is underway, and will be completed by the end of 2022. The PI has access to JLSE and nodes that are representative of Exascale machines powered by AMD and Intel GPUs. The porting of WEST to such architectures is expected to be completed in spring 2023.

4 REFERENCES

- [Baroni2001] S. Baroni, S. de Gironcoli, A. Dal Corso, P. Giannozzi, Phonons and related crystal properties from density-functional perturbation theory, *Reviews of Modern Physics*, 73 (2001) 515-562.
- [Brawand2016] N. P. Brawand, M. Vörös, M. Govoni, and G. Galli, Generalization of dielectric dependent hybrid functionals to finite systems, *Phys. Rev. X* 6, 041002 (2016).
- [Brawand2017] N. P. Brawand, M. Govoni, M. Vörös, and G. Galli, Performance and self-consistency of the generalized dielectric dependent hybrid functional, *J. Chem. Theory Comput.* 13, 3318 (2017).
- [Castelletto2013] S. Castelletto, B. C. Johnson, V. Ivády, N. Stavrias, T. Umeda, A. Gali, and T. Ohshima, A silicon carbide room-temperature single-photon source, *Nature Materials* 13, 151 (2014).
- [Christle2015] D. J. Christle, A. L. Falk, P. Andrich, P. V. Klimov, J. Ul Hassan, N. T. Son, E. Janzén, T. Ohshima, and D. D. Awschalom, Isolated electron spins in silicon carbide with millisecond coherence times, *Nature Materials* 14, 160 (2015).
- [Cunzhi2020] Cunzhi Zhang, Federico Giberti, Emre Sevgen, Juan J. de Pablo, Francois Gygi & Giulia Galli, Dissociation of salts in water under pressure, *Nature Comm.* 11, 3037 (2020)
- [Dawson2015] W. Dawson, and F. Gygi, Performance and Accuracy of Recursive Subspace Bisection for Hybrid DFT Calculations in Inhomogeneous Systems, *J. Chem. Theory Comput.* 11, 4655 (2015).
- [Dawson2018] William Dawson, and Francois Gygi, Equilibration and analysis of first-principles molecular dynamics simulations of water, *J. Chem. Phys.* 148, 124501 (2018).
- [Dong2021] S. Dong, M. Govoni, and G. Galli, Machine Learning Dielectric Screening for the Simulation of Excited State Properties of Molecules and Materials, *Chem. Sci.* 12, 4970 (2021).

[Freysoldt2014] C. Freysoldt, B. Grabowski, T. Hickel, J. Neugebauer, G. Kresse, A. Janotti, and C. G. Van de Walle, First-principles calculations for point defects in solids, Rev. Mod. Phys. 86, 253 (2014).

[Fuchs2015] F. Fuchs, B. Stender, M. Trupke, D. Simin, J. Pflaum, V. Dyakonov, and G. V. Astakhov, Engineering near-infrared single-photon emitters with optically active spins in ultrapure silicon carbide, Nature Commun. 6, 7578 (2015).

[Giannozzi2009] P. Giannozzi et al., J. Phys.: Condens. Matter **21**, 395502 (2009). <http://www.quantum-espresso.org>

[Giannozzi2020] P. Giannozzi et al., J. Chem. Phys. 152, 154105 (2020).

[Govoni2015] M. Govoni, and G. Galli, Large scale GW calculations, J. Chem. Theory Comput. 11, 2680 (2015). <http://www.west-code.org>

[Govoni2018] M. Govoni, and G. Galli, GW100: Comparison of Methods and Accuracy of Results Obtained with the WEST Code, J. Chem. Theory Comput. Article ASAP. (2018) DOI: 10.1021/acs.jctc.7b00952

[Govoni2019] M. Govoni, M. Munakami, A. Tanikanti, J. H. Skone, H. B. Runesha, F. Giberti, J. de Pablo, and G. Galli, Qresp, a tool for curating, discovering and exploring reproducible scientific papers. Sci. Data 6, 190002 (2019)

[Govoni2021] M. Govoni, J. Whitmer, J. de Pablo, F. Gygi, and G. Galli, Code interoperability extends the scope of quantum simulations, npj Comput. Mater. 7, 32 (2021)

[Gygi2003] F. Gygi, J.-L. Fattebert, E. Schwegler, Computation of Maximally Localized Wannier Functions using a simultaneous diagonalization algorithm, Computer Physics Communications, 155 (2003) 1-6.

[Gygi2008] F. Gygi, IBM J. Res. & Dev. 52, 1 (2008). <http://qboxcode.org>

[Gygi2009] F. Gygi, Compact Representations of Kohn-Sham Invariant Subspaces, Phys. Rev. Lett. 102, 166406 (2009).

[He2015] Y.-M. He, G. Clark, J. R. Schaibley, Y. He, M.-C. Chen, Y.-J. Wei, X. Ding, Q. Zhang, W. Yao, X. Xu, C.-Y. Lu, and J.-W. Pan, Single quantum emitters in monolayer semiconductors, Nature Nanotechnology 10, 497 (2015).

[Huang2022] B. Huang, M. Govoni, and G. Galli, Simulating the electronic structure of spin defects on quantum computers, PRX Quantum 3, 010339 (2022).

[Koehl2015] W. F. Koehl, H. Seo, G. Galli, and D. D. Awschalom, Designing defect spins for wafer-scale quantum technologies, MRS Bulletin 40, 1146 (2015).

[LaCount2019] Michael D. LaCount and François Gygi, Ensemble first-principles molecular dynamics simulations of water using the SCAN meta-GGA, J. Chem. Phys. 151, 164101 (2019)

[Lienhard2016] B. Lienhard, T. Schröder, S. Mouradian, F. Dolde, T. T. Tran, I. Aharonovich, and D. Englund, Bright and photostable single-photon emitter in silicon carbide, Optica 3, 768 (2016).

[Nguyen2019] N. L. Nguyen, H. Ma, M. Govoni, F. Gygi, and G. Galli A finite field approach to solve the Bethe Salpeter equation, Physical Review Letters, 122 (2019) 237402.

[Ma2019] He Ma, Marco Govoni, Francois Gygi and Giulia Galli. A Finite-field Approach for GW Calculations Beyond the Random Phase Approximation. J. Chem. Theory. Comp., 15 (1), 154-164 (2019). 10.1021/acs.jctc.8b00864

[Ma2020] H. Ma, M. Govoni and G. Galli, Quantum simulations of materials on near-term quantum computers, npj Comput. Mat. 7, 32 (2020).

[Ma2021] H. Ma, M. Govoni and G. Galli, Quantum Embedding Theory for Strongly-correlated States in Materials, J. Chem. Theory Comput. 17, 2116 (2021).

[Madsen2006] G.K.H. Madsen, D.J. Singh, BoltzTraP. A code for calculating band-structure dependent quantities, Computer Physics Communications, 175 (2006) 67-71.

[McAvoy2018] R. L. McAvoy, M. Govoni, and G. Galli, Coupling First-Principles Calculations of Electron-Electron and Electron-Phonon Scattering, and Applications to Carbon-Based Nanostructures, J. Chem. Theory. Comput. 14, 6269 (2018).

[Onida2002] G. Onida, L. Reining, and A. Rubio, Electronic excitations: density-functional versus many-body Green's-function approaches, Rev. Mod. Phys. 74, 601 (2002).

[Pham2013] Tuan Anh Pham, Huy-Viet Nguyen, Dario Rocca and Giulia Galli , GW calculations using the spectral decomposition of the dielectric matrix: Verification, validation, and comparison of methods Phys. Rev. B 87, 155148 (2013).

[Ping2013] Y. Ping, D. Rocca, and G. Galli, Electronic excitations in light absorbers for photoelectrochemical energy conversion: first principles calculations based on many body perturbation theory, Chem Soc. Rev. 42, 2437 (2013).

[Plimpton1995] S. Plimpton, Fast Parallel Algorithms for Short-Range Molecular Dynamics, Journal of Computational Physics, 117 (1995) 1-19.

[Puli2017] Marcello Puligheddu, Francois Gygi, and Giulia Galli, First-principles simulations of heat transport, Phys. Rev. Materials 1, 060802(R) (2017).

[Puli2019] Marcello Puligheddu, Yi Xia, Maria Chan and Giulia Galli, Computational prediction of lattice thermal conductivity: A comparison of molecular dynamics and Boltzmann transport approaches, Phys. Rev. Mat. 3, 085401 (2019).

[Rocca2008] D. Rocca, R. Gebauer, Y. Saad, and S. Baroni, Turbo charging time-dependent density-functional theory with Lanczos chains, J. Chem. Phys. 128, 154105 (2008).

[Rocca2012] D. Rocca, Y. Ping, R. Gebauer, and G. Galli, Solution of the Bethe-Salpeter equation without empty electronic states: Application to the absorption spectra of bulk systems, Phys. Rev. B 85, 045116 (2012).

[Rozsa2018] Viktor Rozsa, Ding Pan, Federico Giberti, and Giulia Galli, Ab Initio Spectroscopy and Ionic Conductivity of Water under Earth Mantle Conditions, PNAS, 115(27), 6952-6957 (2018).

- [Scalise2018] E. Scalise, V. Srivastava, E. Janke, D. Talapin, G. Galli, S. Wippermann, Surface chemistry and buried interfaces in all-inorganic nanocrystalline solids, *Nature Nanotechnology*, 13 (2018) 841-848.
- [Scherpelz2016] P. Scherpelz, M. Govoni, I. Hamada, and G. Galli, Implementation and validation of fully relativistic GW calculations: spin-orbit coupling in molecules, nanocrystals, and solids, *J. Chem. Theory Comput.* 12, 3523 (2016).
- [Seo2016a] H. Seo, M. Govoni, and G. Galli, Design of defect spins in piezoelectric aluminum nitride for solid-state hybrid quantum technologies, *Sci. Rep.* 6, 20803 (2016).
- [Seo2016b] H. Seo, A. L. Falk, P. V. Klimov, K. C. Miao, G. Galli, and D. D. Awschalom, Quantum decoherence dynamics of divacancy spins in silicon carbide, *Nat. Commun.* 7, 12935 (2016).
- [Seo2017] H. Seo, H. Ma, M. Govoni, and G. Galli, Designing defect-based qubit candidates in wide-gap binary semiconductors for solid-state quantum technologies, *Phys. Rev. Mat* 1, 075002 (2017).
- [Sevgen2018] E. Sevgen, F. Giberti, H. Sidky, J.K. Whitmer, G. Galli, F. Gygi, J.J. de Pablo, Hierarchical Coupling of First-Principles Molecular Dynamics with Advanced Sampling Methods, *Journal of Chemical Theory and Computation*, 14 (2018) 2881-2888.
- [Sheng2022] N. Sheng, C. Vorwerk, M. Govoni, and G. Galli, Green's function formulation of quantum defect embedding theory, *J. Chem. Theory Comput.* 18, 3512 (2022).
- [Sidky2018] H. Sidky, Y.J. Colón, J. Helfferich, B.J. Sikora, C. Bezirk, W. Chu, F. Giberti, A.Z. Guo, X. Jiang, J. Lequieu, J. Li, J. Moller, M.J. Quevillon, M. Rahimi, H. Ramezani-Dakhel, V.S. Rathee, D.R. Reid, E. Sevgen, V. Thapar, M.A. Webb, J.K. Whitmer, J.J. de Pablo, SSAGES: Software Suite for Advanced General Ensemble Simulations, *The Journal of Chemical Physics*, 148 (2018) 044104.
- [Skone2014] J. H. Skone, M. Govoni, and G. Galli, Self-consistent hybrid functional for condensed systems, *Phys. Rev. B* 89, 195112 (2014).
- [Skone2016] J. H. Skone, M. Govoni, and G. Galli, Nonempirical range-separated hybrid functionals for solids and molecules, *Phys. Rev. B* 93, 235106 (2016).
- [Sun2015] J. Sun, A. Ruzsinszky, J.P. Perdew, Strongly Constrained and Appropriately Normed Semilocal Density Functional, *Physical Review Letters*, 115 (2015) 036402.
- [Weber2010] J. R. Weber, W. F. Koehl, J. B. Varley, A. Janotti, B. B. Buckley, C. G. Van de Walle, and D. D. Awschalom, Quantum computing with defects, *Proc. Nat. Acc. Sci.* (2010). DOI: doi.org/10.1073/pnas.1003052107
- [Whiteley2019] Samuel J. Whiteley, Gary Wolfowicz, Christopher P. Anderson, Alexandre Bourassa, He Ma, Meng Ye, Gerwin Koolstra, Kevin J. Satzinger, Martin V. Holt, F. Joseph Heremans, Andrew N. Cleland, David I. Schuster, Giulia Galli, David D. Awschalom, Probing spin-phonon interactions in silicon carbide with Gaussian acoustics, *Nature Physics*, 15, 490-495 (2019)
- [Widmann2015] M. Widmann, S.-Y. Lee, T. Rendler, N. T. Son, H. Fedder, S. Paik, L.-P. Yang, N. Zhao, S. Yang, I. Booker, A. Denisenko, M. Jamali, S. A. Momenzadeh, I. Gerhardt, T. Ohshima, A. Gali, E. Janzén, and J. Wrachtrup, Coherent control of single spins in silicon carbide at room temperature, *Nature Materials* 14, 164 (2015).

[Wilson2008] H.F. Wilson, F. Gygi, G. Galli, Efficient iterative method for calculations of dielectric matrices, *Physical Review B*, 78 (2008) 113303; and H.F. Wilson, D. Lu, F. Gygi, G. Galli, Iterative calculations of dielectric eigenvalue spectra, *Physical Review B*, 79 (2009) 245106.

[Wu2012] J. Wu, F. Gygi, A simplified implementation of van der Waals density functionals for first-principles molecular dynamics applications, *The Journal of Chemical Physics*, 136 (2012) 224107.

[Yu2022] V. Yu, M. Govoni, GPU Acceleration of Large-Scale Full-Frequency GW Calculations, *J. Chem. Theory Comput.* Under review. arXiv: <https://arxiv.org/abs/2203.05623>

[Zheng2019] H. Zheng, M. Govoni, G. Galli, Dielectric dependent hybrid functionals for heterogeneous materials, *Phys. Rev. Mat.* (2019).

PERSONNEL JUSTIFICATION AND MANAGEMENT PLAN

PERSONNEL JUSTIFICATION

This proposal is based on a well-established collaboration between researchers of three institutions: Argonne National Laboratory, the University of Chicago, and the University of California Davis. Proponents of the present project combine long-term expertise in coding, optimized software use, and command of massively parallel high-performance computing resources for material science.

PI, co-PI, post-doctoral research associates (PD), graduate students (GS) and senior collaborators (SC) participating in the project are:

- Marco Govoni (PI, ANL & UChicago) – software lead for the WEST code (<http://west-code.org>)
- Giulia Galli (co-PI, UChicago & ANL) – director of the Midwest Integrated Center for Computational Materials (<http://miccom-center.org>)
- Fran ois Gygi (co-PI, UCDavis) – software lead for the Qbox code (<http://qboxcode.org>)
- Elizabeth Lee (PD, UChicago) – simulations of point-defect formation energies
- Arpan Kundu (PD, UChicago) – path-integral molecular dynamics simulations
- Giacomo Melani (PD, UChicago) – first principles molecular dynamics simulations
- Christian Vorwerk (PD, UChicago) – quantum embedding calculations
- Wenzhe Yu (PD, Argonne National Laboratory) – WEST code developer, porting to GPUs, and expert of electronic structure calculations
- John McFarland (PD, Argonne National Laboratory) – simulations of excited states
- Shenli Zhang (PD, UChicago) – simulations of defective and nanostructured materials
- Cunzhi Zhang (PD, UChicago) – simulations of point-defect formation energies
- Yu Jing (GS, UChicago) – simulations of light-matter interaction in solid-state materials for quantum technologies
- Nan Sheng (GS, UChicago) – quantum embedding calculations
- Benchen Huang (GS, UChicago) – hybrid classical-quantum computing for electronic structure calculations
- Jonah Nagura (GS, UChicago) – calculations of open quantum systems
- Mykyta Onizhuk (GS, UChicago) – lead developer of pyCCE
- Abigail Potoshman (GS, UChicago) – machine learning for electronic structure calculations
- Andrew Xu (GS, UChicago) – calculations of absorption spectra with the WEST code
- Jiawei Zhang (GS, UChicago) – calculations of excited state properties
- Yizhi Zhu (GS, UChicago) – calculations of point-defects in diamond and SiC
- Matthew Pham (GS, ANL & UChicago) – High-throughput calculations for electronic structure calculations

The experiences of team members are complementary and make the team very well positioned to solve the combined set of problems proposed here.

MANAGEMENT PLAN

Allocations will be made monthly or quarterly, depending on specific needs, by the PI and the co-PIs. The PI is the point of contact of the project and will provide, as needed, updates on the status of the work including publications, awards, and highlights of accomplishments.

The PI has successfully managed or participated in other INCITE, ALCC and other projects for several years. The personnel is funded by DOE/BES: MICCoM center (<http://miccom-center.org>), and DOE Early Career Research Program.

Proposal Title: Large-scale simulations of materials for energy and quantum information science

Year1			
I.1: Formation energy and electronic properties of point defects in wide band gap semiconductors	Resource: ALCF-Theta Filesystem storage (TB and dates): 25 TB Archival storage (TB and dates): 100 TB Software Application: Qbox, WEST, QE, SSAGES Tasks: Total energies, potential energies and electronic structure calculations Dependencies: WEST calculations will be run on snapshots of FPMD trajectories obtained with Qbox	Node-hours: ~ 300,000 Q1-2	
I.2: Spectroscopic signatures of point defects in wide band gap semiconductors	Resource: ALCF-Polaris & OLCF/Summit Filesystem storage (TB and dates): 10 TB Archival storage (TB and dates): 20 TB Software Application: WEST, pyCCE Tasks: Spectra (including absorption and photoemission) calculations with quantum embedding, decoherence times Dependencies: WEST calculations will be run on snapshots of FPMD trajectories obtained with Qbox and/or on optimized structures obtained with Qbox or QE	Node-hours: ~ 150,000 in each machine Q2-4	
II.1: Structural properties of single dots and of assemblies of dots	Resource: ALCF-Theta Filesystem storage (TB and dates): 25TB Archival storage (TB and dates): 100 TB Software Application: Qbox and QE Tasks: First principles MD and structural minimizations with semilocal and hybrid DFT Dependencies: None	Node-hours: ~ 200,000 Q1-2	
II.1: Electronic properties of single dots and of assemblies of dots	Resource: ALCF/Aurora & OLCF/Frontier Filesystem storage (TB and dates): 5TB Archival storage (TB and dates): 10 TB Software Application: WEST Tasks: WEST calculations will be run on snapshots of FPMD trajectories obtained with Qbox and/or on optimized structures obtained with Qbox or QE Dependencies: Structural properties simulated on Theta, and validated with experiments.	Node-hours: ~ 25,000 in each machine Q3-4	

PUBLICATIONS RESULTING FROM INCITE AWARDS

1. Victor Wen-zhe Yu and Marco Govoni, GPU Acceleration of Large-Scale Full-Frequency GW Calculations, arXiv:2203.05623v2.
2. Christian Vorwerk, Nan Sheng, Marco Govoni, Benchen Huang, and Giulia Galli, Quantum Embedding Theories to Simulate Condensed Systems on Quantum Computers, arXiv:2105.04736v5.
3. Han Yang, Marco Govoni, Arpan Kundu, and Giulia Galli, Computational Protocol to Evaluate Electron-Phonon Interactions within Density Matrix Perturbation Theory, arXiv:2206.00891.
4. Nan Sheng, Christian Vorwerk, Marco Govoni, and Giulia Galli, Green's Function Formulation of Quantum Defect Embedding Theory, *J. Chem. Theory Comput.*, 18 (6), 3512–3522 (2022).
5. Benchen Huang, Marco Govoni, and Giulia Galli, Simulating the Electronic Structure of Spin Defects on Quantum Computers, *PRX Quantum*, 3 (1), 010339 (2022).
6. Sijia S. Dong, Marco Govoni, and Giulia Galli, Machine Learning Dielectric Screening for the Simulation of Excited State Properties of Molecules and Materials, *Chem. Sci.*, 12 (13), 4970–4980 (2021).
7. Marco Govoni, Jonathan Whitmer, Juan de Pablo, Francois Gygi, and Giulia Galli, Code Interoperability Extends the Scope of Quantum Simulations, *npj Comput. Mater.*, 7 (1), 32 (2021).
8. Yu Jin, Marco Govoni, Gary Wolfowicz, Sean E. Sullivan, F. Joseph Heremans, David D. Awschalom, and Giulia Galli, Photoluminescence Spectra of Point Defects in Semiconductors: Validation of First-Principles Calculations, *Phys. Rev. Materials*, 5 (8), 084603 (2021).
9. Elizabeth M. Y. Lee, Alvin Yu, Juan J. de Pablo, and Giulia Galli, Stability and Molecular Pathways to the Formation of Spin Defects in Silicon Carbide, *Nat. Commun.*, 12 (1), 6325 (2021).
10. Han Yang, Marco Govoni, Arpan Kundu, and Giulia Galli, Combined First-Principles Calculations of Electron-Electron and Electron-Phonon Self-Energies in Condensed Systems, *J. Chem. Theory Comput.*, 17 (12), 7468–7476 (2021).
11. Katherine J. Harmon, Kendra Letchworth-Weaver, Alex P. Gaiduk, Federico Giberti, Francois Gygi, Maria K. Y. Chan, Paul Fenter, and Giulia Galli, Validating First-Principles Molecular Dynamics Calculations of Oxide/Water Interfaces with X-Ray Reflectivity Data, *Phys. Rev. Materials*, 4 (11), 113805 (2020).
12. He Ma, Marco Govoni, and Giulia Galli, Quantum Simulations of Materials on Near-Term Quantum Computers, *npj Comput. Mater.*, 6 (1), 85 (2020).
13. Ryan L. McAvoy, Marco Govoni, and Giulia Galli, Coupling First Principles Calculations of Electron-Electron and Electron-Phonon Scattering, and Applications to Carbon-based Nanostructures, *J. Chem. Theory Comp.*, 14 (12), 6269-6275 (2018).
14. Alex Gaiduk, Jeffrey Gustafson, Francois Gygi, and Giulia Galli, First-Principles Simulations of Liquid Water Using a Dielectric-Dependent Hybrid Functional, *J. Phys. Chem. Lett.*, 9, 3068-3073 (2018).

15. Alex Gaiduk, T. Anh Pham, Marco Govoni, Francesco Paesani and Giulia Galli, Electron affinity of liquid water, *Nature Comm.*, 9, 247 (2018).
16. Peter Scherpelz and Giulia Galli, Optimizing surface defects for atomic-scale electronics: Si dangling bonds, *Phys. Rev. Materials* 1, 021602(R) (2017)
17. Nicholas P. Bawand, Marco Govoni, Márton Vörös, and Giulia Galli, Performance and self-consistency of the generalized dielectric dependent hybrid functional, *J. Chem. Theory Comput.* 13, 3318 (2017).
18. Matthew Goldey, Nicholas Bawand, Márton Vörös, and Giulia Galli, Charge Transport in Nanostructured Materials: Implementation and Verification of Constrained Density Functional Theory, *J. Chem. Theory Comput.* 13 (6), 2581-2590 (2017).
19. Alex P. Gaiduk and Giulia Galli, Local and Global Effects of Dissolved Sodium Chloride on the Structure of Water, *J. Phys. Chem. Lett.* 8, 1496-1502 (2017).
20. Nicholas Bawand, Matthew Goldey, Márton Vörös, and Giulia Galli, Defect states and charge transport in quantum dot solids, *Chem. Mater.* 29 (3), 1255-1262 (2017)
21. Matthew B. Goldey, Daniel Reid, Juan de Pablo and Giulia Galli, Planarity and multiple components promote organic photovoltaic efficiency by improving electronic transport, *Phys. Chem. Chem. Phys.* 18, 31388-31399 (2016)
22. Nicholas P. Bawand, Márton Vörös, Marco Govoni, and Giulia Galli, Generalization of Dielectric-Dependent Hybrid Functionals to Finite Systems, *Phys. Rev. X* 6, 041002 (2016)
23. Peter Scherpelz, Marco Govoni, Ikutaro Hamada, and Giulia Galli, Implementation and Validation of Fully-Relativistic GW Calculations: Spin-Orbit Coupling in Molecules, Nanocrystals and Solids, *J. Chem. Theory Comput.* 12 (8), 3523-3544 (2016).
24. Alex P. Gaiduk, Marco Govoni, Robert Seidel, Jonathan Skone, Bernd Winter, and Giulia Galli, J Photoelectron spectra of aqueous solutions from first principles, *J. Am. Chem. Soc. Comm.* 138, 6912 (2016).
25. Ding Pan and Giulia Galli, The fate of carbon dioxide in water-rich fluids at extreme conditions, *Science Advances* 2, e1601278 (2016).
26. Performance and Accuracy of Recursive Subspace Bisection for Hybrid DFT Calculations in Inhomogeneous Systems, William Dawson and Francois Gygi, *JCTC* 11, 4655 (2015).
27. First-principles framework to compute sum-frequency generation vibrational spectra of semiconductors and insulators, Quan Wan, and Giulia Galli, *Phys. Rev. Lett.* 115, 246404 (2015).
28. Marco Govoni and Giulia Galli, “Large scale GW calculations”, *J. Chem. Theory Comput.* 11, 2680 (2015).
29. Alex P. Gaiduk, Francois Gygi and Giulia Galli, “Density and Compressibility of Liquid Water and Ice from First-Principles Simulations with Hybrid Functionals”, *J. Phys. Chem. Lett.* 6, 2902 (2015).

30. Martin Schlipf and Francois Gygi, “Optimization algorithm for the generation of ONCV pseudopotentials”, *Comp. Phys. Comm.* 2015 (ASAP article).
31. Alex P. Gaiduk, Cui Zhang, François Gygi and Giulia Galli, “Structural and electronic properties of aqueous NaCl solutions from ab initio molecular dynamics simulations with hybrid density functionals,” *Chemical Physics Letters* [Frontier Article] 604(3): 89, (2014).
32. Wan, Quan, Leonardo Spanu, François Gygi and Giulia Galli, “Electronic structure of aqueous sulfuric acid from first principles simulations with hybrid functionals,” *The Journal of Physical Chemistry Letters* submitted, (2014).
33. Wan, Quan, Leonardo Spanu, Giulia Galli and François Gygi, “Raman spectra of liquid water from ab initio molecular dynamics: vibrational signatures of charge fluctuations in the hydrogen bond network,” *Journal of Chemical Theory and Computation* 9(9): 4124–30, (2014).
34. Zhang, Cui, Tuan Anh Pham, François Gygi and Giulia Galli, “Communication: electronic structure of the solvated chloride anion from first principles molecular dynamics,” *The Journal of Chemical Physics* 138(18): 181102, (2014).
35. Quan Wan, Leonardo Spanu and Giulia Galli, “Solvation Properties of Microhydrated Sulfate Anion Clusters: Insights From Ab Initio Calculations”, *J. Phys. Chem. B* 116, 9460 (2012).
36. Cui Zhang, Leonardo Spanu and Giulia Galli “Entropy of Liquid Water from Ab-initio Molecular Dynamics”, *J. Phys. Chem. B* 115, 14190–14195 (2011).
37. Cui Zhang, Jun Wu, Giulia Galli and Francois Gygi , “Structural and Vibrational Properties of Liquid Water from van der Waals Density Functionals”, *J. Chem. Theory Comput.* 7, 3054-3061 (2011).
38. Cui Zhang, Davide Donadio, Francois Gygi and Giulia Galli “First Principles Simulation of the Infrared Spectrum of Liquid Water using Hybrid Density Functionals” *J. Chem. Theory Comput.* 7, 1443 (2011).
39. François Gygi and Ivan Duchemin, “Efficient Computation of Hartree–Fock Exchange Using Recursive Subspace Bisection”, *J. Chem. Theory Comput.* 9, 582–587 (2013).
40. Duchemin and F. Gygi, “A scalable and accurate algorithm for the computation of Hartree–Fock exchange”, *Comput. Phys. Comm.* 181, 855 (2010).

Marco Govoni
Email: mgovoni@anl.gov

Professional Preparation

University of Modena and Reggio Emilia, Nanoscience and Nanotechnology, Ph.D.	2012
University of Modena and Reggio Emilia, Physics, M.S.	2008
University of Modena and Reggio Emilia, Physics, B.S.	2006

Appointments

Scientist, Argonne National Laboratory, Materials Science Division & Center for Molecular Engineering	2021 – present
Scientist, University of Chicago, Pritzker School of Molecular Engineering	2018 – present
Assistant Scientist, Argonne National Laboratory, Materials Science Division & Center for Molecular Engineering	2016 – 2021
Argonne National Laboratory, Materials Science Division	2015 – 2016
Postdoctoral Researcher University of Chicago, Pritzker School of Molecular Engineering	2014
Postdoctoral Researcher University of California Davis, Dept. of Chemistry	2012 – 2014
Postdoctoral Researcher	

Five Publications Most Relevant to This Proposal

1. N. Sheng, C. Vorwerk, M. Govoni, and G. Galli, “Green’s Function Formulation of Quantum Defect Embedding Theory”, *J. Chem. Theory Comput.* 18, 3512 (2022), <https://doi.org/10.1021/acs.jctc.2c00240>
2. M. Gerosa, F. Gygi, M. Govoni, and G. Galli, “The role of defects and excess surface charges at finite temperature for optimizing oxide photoabsorbers,” *Nature Materials*, 17, 1122, (2018), <https://www.nature.com/articles/s41563-018-0192-4>
3. T.J. Smart, F. Wu, M. Govoni, and Y. Ping, “Fundamental principles for calculating charged defect ionization energies in ultrathin two-dimensional materials,” *Phys. Rev. Mat.* 2, 124002 (2018), <https://journals.aps.org/prmaterials/abstract/10.1103/PhysRevMaterials.2.124002>
4. M. Govoni, and G. Galli, “Large scale GW calculations,” *J. Chem. Theory Comput.*, 11, 2680, (2015), <https://pubs.acs.org/doi/10.1021/ct500958p>
5. J.H. Skone, M. Govoni, and G. Galli, “Self-consistent hybrid functional for condensed systems,” *Phys. Rev. B* 89, 195112, (2014), <https://journals.aps.org/prb/abstract/10.1103/PhysRevB.89.195112>

Research Interests and Expertise

Development and use of theoretical and computational methods to study materials for energy and quantum information science. Numerical methods. Software development. Exascale and quantum computing.

Synergistic Activities

1. Member of the management team of the Midwest Integrated Center for Computational Materials (<http://miccom-center.org>)
2. PI of the Early Career Research Program entitled “Optical Control of Spin-polarization in Quantum Materials”
3. Software development: project lead of the WEST code (<http://west-code.org>)

4. 23 invited talks, including invitations: to major international conferences
 - a. the annual march meeting of the American Physical Society in 2022, 2018, and 2016
 - b. the annual fall meeting of the American Chemical Society in 2021 and 2014
 - c. the annual meeting of the Electrochemical Society in 2017
 - d. the annual SIAM conference on computer science and engineering in 2017
- to workshops:
 - e. GW goes Large Scale organized by Aalto University, Finland in 2020
 - f. Materials Genome Initiative at Exascale organized by U S California, Spetses, Greece in 2018
 - g. the 18th International Workshop on Computational Physics and Materials Science: Total Energy and Force Methods, organized by ICTP, Italy in 2017
 - h. the OPTIMADE Workshop on Open Databases Integration for Materials Design, organized by the Lorentz Center, Leiden, Netherlands in 2016
 - i. the workshop on Recent Progress in Numerical Green's Functions Methods in Physics and Chemistry, organized by the Telluride Science Research Center USA in 2016
- at leading research institutions:
 - j. Condensed Matter seminar at Case Western Reserve University in 2021
 - k. Physics colloquium at University of Missouri-Columbia in 2021
 - l. Seminar at Central Michigan University in 2017
5. Reviewer for: Science Advances, Nature Light, Physical Review Letters, Physical Review Materials, Physical Review B, IOP Nanotechnology, AIP Advances, ACS Journal of Chemical Theory and Computation, AIP Journal of Chemical Physics, MDPI Materials, npj Computational Materials, International Journal of Quantum Chemistry, Chem Phys Chem, Carbon.

Collaborators (past 5 years including name and current institution)

Awschalom, David, University of Chicago
 Blum, Volker, Duke University
 Bradforth, Stephen, University of Southern California
 Ceriotti, Michele, EPFL
 Cheng, Man-Hin, University of Chicago
 Clayton, Connor, Carnegie Mellon University
 de Pablo, Juan, University of Chicago
 Dong, Sijia, Northeastern University
 Du, Xiaochen, Duke University
 Gaiduk, Alex, Facebook
 Galli, Giulia, University of Chicago
 Gerosa, Matteo, Shell
 Giberti, Federico, University of Chicago
 Gygi, Francois, University of California, Davis
 Heremans, Joe, Argonne National Lab
 Hamada, Ikutaro, Osaka University
 Kim, Siyoung, University of Chicago
 Ma, He, Google
 Marri, Ivan, University of Modena and Reggio Emilia
 McAvoy, Ryan, IBM
 Munakami, Milson, University of Chicago

Nguyen, Ngoc Linh, EPFL
 Ossicini, Stefano, University of Modena and Reggio Emilia
 Paesani, Francesco, University of California San Diego
 Pan, Ding, Hong Kong University
 Pham, Tuan Anh, LLNL
 Ping, Yuan, University of California Santa Cruz
 Ropo, Matti, University of Turku
 Runesha, Hakizumwami, University of Chicago
 Scherpelz, Peter, Modern Electron
 Schwegler, Eric, LLNL
 Seidel, Robert, Helmholtz-Zentrum Berlin
 Seo, Hosung, Ajou University, Korea
 Skone, Jonathan, LBNL
 Smart, Tyler, University of California Santa Cruz
 Tanikanti, Aditya, Argonne National Laboratory
 Vörös, Marton, Samsung
 Wang, Wennie, University of Chicago
 Winter, Bernd, Fritz-Haber-Institut der Max-Planck-Gesellschaft
 Wu, Feng, University of California Santa Cruz
 Yang, Han, University of Chicago
 Zheng, Huihuo, Argonne National Laboratory

FRANCOIS GYGI

Professor of Computer Science, University of California Davis
1 Shields Ave, Davis, CA 95616, fgygi@ucdavis.edu
<http://faculty.engineering.ucdavis.edu/gygi>

Professional Preparation

Ph.D. Physics, Ecole Polytechnique Federale (EPFL), Lausanne, Switzerland, 1988
M.S. Physics, Ecole Polytechnique Federale (EPFL), Lausanne, Switzerland, 1983

Appointments

Professor, Department of Computer Science, University of California Davis	2009-present
Professor, Department of Applied Science, University of California, Davis	2005-present
Project leader, Center for Applied Scientific Computing, Lawrence Livermore National Laboratory (LLNL)	1999-2005
Computer Scientist, Center for Applied Scientific Computing, Lawrence Livermore National Laboratory (LLNL)	1998-1999
Senior Researcher, IRRMA, EPFL, Lausanne, Switzerland	1993-1998
Post-Doctoral Researcher, IBM Research Division, Zurich, Switzerland	1991-1992
Post-Doctoral Researcher, AT&T Bell Laboratories, Murray Hill, NJ2010–present	1988-1990

Five Publications Most Relevant to This Proposal

1. A Kundu, M Govoni, H Yang, M Ceriotti, F Gygi, G Galli, “Quantum vibronic effects on the electronic properties of solid and molecular carbon”, Phys. Rev. Mat. 5, L070801 (2022).
<https://doi.org/10.1103/PhysRevMaterials.5.L070801>
2. N. L. Nguyen, H. Ma, M. Govoni, F. Gygi, G. Galli, “A finite-field approach to solving the Bethe-Salpeter equation”, Phys. Rev. Lett. 122, 237402 (2019).
3. H. Ma, M. Govoni, F. Gygi, G. Galli, “A finite-field approach for GW calculations beyond the random phase approximation”, J. Chem. Theory Comput. 15, 154 (2018).
4. M. Gerosa, F. Gygi, M. Govoni, G. Galli, “The role of defects and excess surface charges at finite temperature for optimizing oxide photoabsorbers”, Nature Materials 17, 1122-1127 (2018).
<https://doi.org/10.1038/s41563-018-0192-4>
5. E. Sevgen, F. Giberti, H. Sidky, J.K.Whitmer, G. Galli, F. Gygi and J.J.de Pablo, “Hierarchical Coupling of First-Principles Molecular Dynamics with Advanced Sampling Methods”, J. Chem. Theory Comput. 14, 2881 (2018) DOI:10.1021/acs.jctc.8b00192

Research Interests and Expertise

Numerical methods and algorithms for electronic structure calculations. Electronic structure of solids, liquids and nanostructures. Large-scale parallel algorithms. Molecular dynamics. Multiscale modeling.

Synergistic Activities

1. Distribution of open-source high-performance, parallel, first-principles molecular dynamics software (<http://qboxcode.org>).
2. Dissemination of web standards for electronic structure calculation data (<http://www.quantum-simulation.org>).

Collaborators and Co-editors:

Collaborators (past 5 years including name and current institution)

M. LaCount, UC Davis
G. Galli, U Chicago
J. de Pablo, U Chicago
J. Whitmer, U Notre Dame
A. Gaiduk, U Chicago
V. Rozsa, U Chicago
E. Sevgen, U Chicago
F. Giberti, U Chicago
H. Ma, U Chicago
M. Govoni, ANL
H. Zheng, ANL
W. Dawson, RIKEN, Japan
K. Lejaeghere, Ghent, Belgium
M. Schlipf, UC Davis
W. Pickett, UC Davis
R. Faller, UC Davis
A. Wexler, UC Davis

Giulia Galli
Liew Family Professor of Molecular Engineering & Professor of Chemistry
The University of Chicago, 5640 South Ellis, ERC 233, Chicago, IL 60637

Professional Preparation

PhD, Int'l School for Advanced Studies, Trieste, Italy, PhD in Physics	1986
MS, Int'l School for Advanced Studies, Trieste, Italy, Master in Physics	1984
BS, University of Modena, Italy, 'Laurea' in Physics, summa cum laude	1982

Appointments

Professor of Chemistry, The University of Chicago	2017 – Present
Director of the Midwest Integrated Center for Computational Materials	2015 – Present
Argonne National Laboratory Senior Scientist, Argonne National Laboratory	2014 – Present
The Liew Family Professor of Electronic Structure and Simulations, Pritzker School of Molecular Engineering, The University of Chicago	2013 – Present
Professor of Physics, University of California, Davis	2009 – 2013
Professor of Chemistry, University of California, Davis	2005 – 2013
Quantum Simulations Group, Leader, Lawrence Livermore National Laboratory	2000 – 2005
Staff Physicist, Lawrence Livermore National Laboratory	1998 – 2000
Senior Scientist, Swiss Federal Institute of Technology Lausanne, Switzerland	1995 – 1998
Senior Researcher, Swiss Federal Institute of Technology Lausanne	1991 – 1995

Five Publications Most Relevant to This Proposal

1. *Stability and molecular pathways to the formation of spin defects in silicon carbide*, Elizabeth. M. Y. Lee, Alvin Yu, Juan J. de Pablo, and Giulia Galli, Nat. Commun. 12, 6325 (2021). arXiv:2109.0642
2. *Green's function formulation of quantum defect embedding theory*, N. Sheng*, C. Vorwerk*, M. Govoni, and G. Galli (*equal contribution), J. Chem. Theory Comput. accepted (2022). arXiv:2203.05493
3. *Simulating the electronic structure of spin defects on quantum computers*, B. Huang, M. Govoni, and G. Galli, PRX Quantum 3, 010339 (2022).
4. *PyCCE: A Python Package for Cluster Correlation Expansion Simulations of Spin Qubit Dynamic*, M.Onizhuk, G.Galli, Adv. Theory Simul., 2100254 (2021).
5. *Probing the Coherence of Solid-State Qubits at Avoided Crossings*, M.Onizhuk, K.C.Miao, J.P.Blanter, H.Ma, C.P.Anderson, A.Bourassa, D.D.Awschalom, and G.Galli, PRX Quantum, 2, 010311 (2021). 10.1103/PRXQuantum.2.010311

Research Interests and Expertise

Prof. Galli's research activity is focused on the development and use of theoretical and computational methods to understand and predict the properties and behavior of materials (solids, liquids, and nanostructures) from first principles.

Synergistic Activities

1. Member of the technical and scientific committee of the Italian Institute of Technology (2020-2023)
2. Committee on Condensed Matter and Materials Research, National Academy of Science (2019-)
3. Editorial Boards: Nature Partner Journals (2017-2018); Physical Review Materials (2017-); Nature Scientific Data (2016-); ACS Central Science (2015-); Journal of Physical Chemistry (2013-); Journal of Chemical Physics (2013-); Journal of Theoretical and Computational Chemistry (2014-); ACS Open Access (2015-); SIAM "Multi-scale Modeling and Simulation" (2004-2010), Computer Physics Communication (Specialist Editor, 2003- 2013).
4. Scientific Advisory Boards: Chemical Science Center, Princeton (2019-); SIMES (Stanford for Materials and Energy Sciences) SAC (2016-); The NoMaD Laboratory, European Center of excellence, headquarters in Berlin, Germany, Chair (2016-); National Center of Competence in Research on New Materials, Marvel, EPLF, Switzerland, Chair (2014-); Thomas Young Center for Theory and Simulation of Materials, London, UK, Chair (2010-); Effree Energy Frontier Research Center, Member (2011-2018); CNGMD Energy Frontier Research Center (2014-2018); Max-Planck-Institut für Festkörperforschung Stuttgart, Germany, Member (2012-); NSF Center for Chemical Innovation “Powering the Planet”, Member (2009-2014); Molecular Foundry, LBNL, Member (2001-2011); The Deutsches Elektronen-Synchrotron (DESY) German national research, SAB member (2018); Center for dynamics and control of materials, UT Austin MRSEC, SAB member (2017-2019); Joint Center for Artificial Photosynthesis (JCAP), SAB member (2016-2019); C2SEPEM (LBNL computational materials science center), SAB member (2017-).
5. APS Counselor elect (2016-2020); APS DCOMP, Chair elect and chair (2006-2008).

Collaborators and Co-editors:

Collaborator Name	Organization Affiliation
Abe, Hiroshi	Nagoya University
Amassian, Aram	North Carolina State University
Anderson, Christopher P.	University of Chicago
Anderson, John S.	University of Chicago
Angyan, Janos G.	Université de Lorraine, France
Anikeeva, Polina	Massachusetts Institute of Technology
Asbury, John B.	Penn State
Awschalom, David D.	University of Chicago
Bao, Zhenan	Stanford University
Barry, Edward	Argonne National Laboratory
Benfenati, Fabio	Istituto Italiano di Tecnologia
Bezanilla, Francisco	University of Chicago
Bhang, Jooyoung	Ajou University
Blanton, Joseph P.	University of Chicago
Blum, Volker	Duke University
Bourassa, Alexandre	University of Chicago
Bradforth, Stephen E.	University of Southern California
Brawand, Nicholas P.	Institute for Molecular Engineering
Bronstein, Noah D.	Unknown
Burns, Raelyn	Argonne National Laboratory
Cai, Zhengxu	University of Chicago
Cao, Yuhong	National Center of Nanoscience and Technology CAS

Carlson, Robert M. K.	Stanford Instit. for Mater. & Ener. Sciences
Carvalho-de-Souza, Joao L.	University of Arizona
Ceriotti, Michele	Swiss Federal Institute of Technology in Lausanne
Cestellos-Blanco, Stefano	University of California, Berkeley
Chan, Maria K.Y.	Massachusetts Institute of Technology
Chanana, Ashish	NIST
Chen, Wei	National University of Singapore
Cheng, Man-Hin	Hong Kong University of Science and Technology
Cheng, Shaobo	Brookhaven National Laboratory
Chiu, I-Ting	University of California, Davis
Cho, Wooje	University of Chicago
Choi, Kyoung-Shin	University of Wisconsin-Madison
Cifra, Michal	Institute of Photonics and Electronics of the Czech Academy of Sciences
Clayton, Connor	University of Maryland
Cleland, Andrew N.	University of Chicago
Colon, Yamil	University of Notre Dame
Coropceanu, Igor	University of Chicago
Crisp, Ryan	Friedrich-Alexander University Erlangen–Nürnberg
Crook, Alexander L.	University of Chicago
Dahl, Jeremy E. P.	Stanford Instit. for Mater. & Ener. Sciences
Dahlberg, Peter D.	The Institute for Biophysical Dynamics
Darling, Seth B.	Argonne National Laboratory
Dauphas, Nicolas	University of Chicago
De Hoe, Guilhem X.	University of Chicago
De Oca, Joan Manuel Montes	University of Chicago
de Pablo, Juan	University of Chicago
DeLongchamp, Dean M.	National Institute of Standards and Technology
Diroll, Benjamin	Argonne National Laboratory
Dombrowski, James	Worcester Polytechnic Institute
Dong, Sijia	Northeastern University
Du, Xiaochen	Duke University
Dudenas, Peter J.	National Institute of Standards and Technology
Elam, Jeffery W.	Argonne National Laboratory
Engel, Gregory S.	Institute for Biophysical Dynamics
Fedin, Igor	University of Alabama
Felts, Alanna M.	Northwestern University
Feng, Mingzhen	University of California, Davis
Fenter, Paul	Argonne National Laboratory
Filatov, Alexander	University of Chicago
Fokin, Audrey A..	Justus-Liebig University, Germany
Francese, Tommaso	University of Chicago
Frano, Alex	University of California San Diego
Gaiduk, Alex P.	Institute for Molecular Engineering
Gali, Adam	Budapest U. of Technology & Economics
Gallagher, Audrey T.	Northwestern University

Gavini, Vikram	University of Michigan
Gerken, James	University of Wisconsin-Madison
Gerosa, Matteo	University of Chicago
Ghosh, Krishnendu	College of Charleston
Giberti, Federico	Institute for Molecular Engineering
Glen, Elena O.	University of Chicago
Goldey, Matthew	Institute for Molecular Engineering
Goldsmith, Zachary	University of Illinois, Urbana-Champaign
Gorodetsky, Alon A.	University of California, Irvine
Govindaraju, Gokul	University of Wisconsin-Madison
Govoni, Marco	Argonne National Laboratory
Greenwood, Arin R.	Institute for Molecular Engineering
Gu, Jing	National Renewable Energy Laboratory
Gunn, Brandon	University of California, San Diego
Gustafson, Jeffery	San Diego State University
Gygi, Francois	University of California, Davis
Hack, John	University of Chicago
Hamada, Ikutaro	Nat'l Instit. For Mater. Science, Japan
Hammes-Schiffer, Sharon	University of Illinois, Urbana-Champaign
Harmon, Katherine J.	Argonne National Laboratory
Harshan, Aparna	University of Illinois, Urbana-Champaign
Hautzinger, Matthew P.	University of Wisconsin-Madison
Hazarika, Abhijit	CSIR-IICT
He, Qiming	University of Chicago
He, Xiang	Argonne National Laboratory
Heki, Larry	University of California, Santa Barbara
Heremans, F. Joseph	Argonne National Laboratory
Hirao, Kimihiko	RIKEN Advanced Institute, Japan
Hjort, Martin	Lund University
Hoenig, Eli	University of Chicago
Hohman, J. Nathan	Lawrence Berkeley National Laboratory
Holt, Martin V.	Argonne National Laboratory
Huang, Zhengjie	North Carolina State University
Iscen, Aysenur	Max Planck Institute for Polymer Research
Janke, Eric M.	University of Chicago
Jaramillo, Thomas	University of California, San Diego
Jin, Yu	University of Chicago
Johnson, Justin	NREL
Kanai, Shun	Tohoku University
Kash, Benjamin	University of Chicago
Kautz, Rylan	University of California, Irvine
Kawamura, Airi	University of Chicago
Kawasaki, Jason Ken	University of Wisconsin-Madison
Kim, Siyoung	University of Chicago
Koolstra, Gerwin	University of California, Berkeley
Kovos, Berk	University of Chicago
Krivosudsky, Ondrej	Institute of Photonics and Electronics (IPE)
Kroupa, Daniel M.	University of Washington
Kundu, Arpan	University of Chicago

Kung, Harold H.	Northwestern University
Laasner, Raul	SRON Netherlands Institute for Space Research
Lanzani, Guglielmo	Istituto Italiano di Tecnologia e Dipartimento di Fisica Politecnico di Milano
Lapa, Pavel N.	Argonne National Laboratory
Lee, Dongho	University of Michigan
Lee, Elizabeth M. Y.	University of Chicago
Lee, Min-Han	Singapore University of Technology and Design
Lei, Yusheng	Stanford University
Letchworth-Weaver, Kendra	James Madison University
Lewis, Nicholas H. C.	University of Chicago
Li, Fei Hua	Stanford Instit. for Mater. & Ener. Sciences
Lin, Yu	Stanford Instit. for Mater. & Ener. Sciences
Lindberg, Ann	University of Wisconsin-Madison
Lipke, Mark C.	Rutgers University
Liu, Chong	University of California, Los Angeles
Liu, Jia	Harvard University
Liu, Mingzhao	Brookhaven National Laboratory
Liu, Xiaojie	Shanghai Institute of Materia Medica CAS
Liu, Zhenxian	Carnegie Institution of Washington
Liyanage, Luckshitha	Stanford University
Lu, Haipeng	Hong Kong University of Science and Technology
Lu, Timothy K.	Massachusetts Institute of Technology
Ma, He	Institute for Molecular Engineering
Ma, Xinyou	University of Chicago
Mane, Anil	Argonne National Laboratory
Mansoor, Iram F.	Rutgers University
Mao, Wendy L.	Stanford University
Martinez, Marissa	University of Colorado Boulder
Martinson, Alex B. F.	Argonne National Laboratory
Mazzotti, Sergio	University of Chicago
McAvoy, Ryan L.	University of Chicago
McNichols, Brett W.	Colorado School of Mines, Golden, CO
Mehta, Apurva	Stanford University
Melosh, Nicholas A.	Stanford Instit. for Mater. & Ener. Sciences
Miao, Kevin C.	University of Chicago
Miller, Elisa M.	National Renewable Energy Laboratory
Mohtashami, Yahya	University of California, Santa Barbara
Molhave, Kristian	National Centre for Nano Fabrication and Characterization
Morbec, Juliana	University of Chicago
Mulfort, Karen L.	Argonne National Laboratory
Munakami, Milson	University of Chicago
Murphy, Julia	University of Chicago
Narkeviciute, Ieva	Stanford University
N'Diaye, Alpha T.	Lawrence Berkeley National Laboratory
Nealey, Paul	University of Chicago
Nguyen, Ngoc Linh	University of Chicago

Nie, Nicole Xike	Carnegie Institution for Science
Norris, David	ETH Zürich
Ogitsu, Tadashi	Lawrence Livermore National Laboratory
Ohno, Hideo	Tohoku University
Ohshima, Takeshi	National Institutes for Quantum and Radiological Science and Technology
Onizhuk, Mykyta	University of Chicago
Opalka, Daniel	Max Planck Institute, Solid State Research
Otto, John P.	Institute for Biophysical Dynamics
Pach, Gregory	US National Renewable Energy Laboratory
Paesani, Francesco	University of California, San Diego
Pan, Ding	University of Chicago
Park, Tae Joon	Purdue University
Peverati, Roberto	University of Minnesota
Pham, Tuan Anh	Lawrence Livermore National Laboratory
Pickett, Warren E.	University of California, Davis
Ping, Yuan	University of California, Santa Cruz
Podeszwa, Rafal	University of Silesia, Katowice, Poland
Prominski, Aleksander	The Univeristy of Chicago
Puligheddu, Marcello	University of Chicago
Qiao, Yijun	Argonne National Laboratory
Radmilovic, Andjela	Carleton College
Rahul, Kar	RIKEN Advanced Institute, Japan
Ramanathan, Shriram	Purdue University
Rodolakis, Fanny M.	Argonne National Laboratory
Rogers, John A.	Northwestern University
Ropo, Matti	University of Turku
Rozsa, Viktor	Univeristy of Chicago
Rozyyev, Vepa	University of Chicago
Runesha, Hakizumwami B.	University of Chicago
Rusishvili, Mariami	University of Chicago
Satzinger, Kevin J.	Pacific Northwest National Laboratory
Scalise, Emilio	University of Milano-Bicocca
Schaller, Richard	Northwestern University
Schatz, George C.	Northwestern University
Scheidt, Rebecca A.	US National Renewable Energy Laboratory
Scherpelz, Peter	University of Chicago
Schneider, Sebastian	Stanford University
Schreiner, Peter R.	Justus-Liebig University, Germany
Schuller, Ivan K.	University of California, San Diego
Schuller, Jon A.	University of California, Santa Barbara
Schuster, David I.	University of Chicago
Schwiegler, Eric	Lawrence Livermore National Laboratory
Scuseria, Gustavo	Rice University
Seidel, Robert	University of Southern California
Sellinger, Alan	Colorado School of Mines, Golden, CO
Seo, Hosung	University of Chicago
Sevgen, Emre	University of Chicago
Shafer, Padraic	Lawrence Berkeley National Laboratory

Shen, Zhi-Xun	Stanford Institut. for Mater. & Ener. Sciences
Sheng, Nan	University of Chicago
Sibener, Steven J.	University of Chicago
Sidky, Hythem	National Institutes of Health
Singh, Rohan	IISER Bhopal
Skone, Jonathan H.	University of Chicago
Sohoni, Siddhartha	University of Chicago
Solis-Ibarra, Diego	Universidad Nacional Autónoma, Mexico
Somogyi, Balint	Budapest Univ. of Technology & Economics
Son, Nguyen T.	Temple University
Song, Jong Won	RIKEN Advanced Institute, Japan
Sprik, Michiel	University of Cambridge, UK
Srivastava, Vishwas	University of Chicago
Stahl, Shannon	University of Wisconsin-Madison
Strohbeen, Patrick	University of Wisconsin-Madison
Sui, Fan	University of California, Davis
Sullivan, Sean E.	California Baptist University
Sun, Dali	North Carolina State University
Szalewicz, Krzysztof	University of Delaware
Takamura, Yayoi	University of California, Davis
Talapin, Dmitri	University of Chicago
Tanikanti, Aditya	Argonne National Laboratory
Taylor, DeCarlos E.	U.S. Army Research Laboratory
Tian, Bozhi	University of Chicago
Tiede, David	Argonne National Laboratory
Tiede, David M.	Argonne National Laboratory
Tirrell, Matthew V.	University of Chicago
Tkachenko, Boryslav A.	Justus-Liebig University, Germany
Tokmakoff, Andrei	University of Chicago
Toney, Michael F.	University of Colorado
Tong, Xiao	Northeastern University, China
Toulouse, Julien	Université Pierre et Marie Curie, France
Truhlar, Donald G.	University of Minnesota
Ul-Hassan, Jawad	Linköping University
Vardeny, Shai R.	Los Alamos National Laboratory
Vardeny, Z. Valy	University of Utah
Vetter, Eric	North Carolina State University
Vo, Hien	University of Chicago
Voros, Marton	Argonne National Laboratory
Vorwerk, Christian	University of Chicago
Voth, Gregory A.	University of Chicago
Wan, Quan	University of Chicago
Wang, Lili	The Institute for Biophysical Dynamics
Wang, Tonghui	New Mexico State University
Wang, Wennie	University of Chicago
Wang, Zhongyang	University of Chicago
Wasielewski, Michael R.	Northwestern University
Whiteley, Samuel J.	HRL Laboratories, LLC
Whitmer, Jonathan	University of Notre Dame

Williams, Nicholas E.	The Institute for Biophysical Dynamics
Winter, Bernd	Helmholtz-Zentrum Berlin
Wippermann, Stefan	Max Planck Institute, Germany
Wolfowicz, Gary	Argonne National Laboratory
Wu, Yilei	Stanford University
Xia, Yi	Northwestern University
Xu, Shaui	Case Western Reserve University
Yan, Hao	Stanford Instit. for Mater. & Ener. Sciences
Yang, Fan	Stanford Instit. for Mater. & Ener. Sciences
Yang, Han	University of Chicago
Yang, Peidong	University of California, Berkeley
Yang, Shijia	North Carolina State University
Ye, Meng	Tsinghua University
Ye, Zifan	University of Chicago
Yesibolati, Murat	Technical University of Denmark
Yim, Donggyu	SK Hynix Inc.
Young, Ryan M	National Cancer Institute
Yu, Alvin	University of Chicago
Yu, Jin	University of Chicago
Yu, Luping	University of Chicago
Yuan, Yue	University College London
Zaluzec, Nestor J.	Argonne National Laboratory
Zeledon, Cyrus	University of Chicago
Zeng, Hao	University of Chicago
Zhang, Cunzhi	University of Chicago
Zhang, Shenli	University of Chicago
Zhang, Zhe	University of New Mexico
Zhao, Donglin	London South Bank University
Zheng, Huihuo	Argonne National Laboratory
Zhou, Chenyu	Brookhaven National Laboratory
Zhu, Kai	University of California, Santa Cruz
Zhu, Yimei	Brookhaven National Laboratory
Zhu, Yizhi	University of Chicago
Beard, Matthew C.	National Renewable Energy Laboratory
Draxl, Claudia	Humboldt University of Berlin
Ferguson, Andrew	The Univeristy of Chicago
Freedman, Danna	Northwestern University
Gardel, Margaret	University of Chicago
Geiger, Franz	Northwestern University
Gomez, Enrique	Penn State
Gray, Harry B.	California Institute of Technology
Hemley, Russ	George Washington University
Huang, Benchen	University of Chicago
Kagan, Cherie	University of Pennsylvania
Kim, Jeongnim	Intel Corporation
Klimov, Viktor	Los Alamos National Laboratory
Kozinsky, Boris	Harvard University
Louie, Steven G.	University of California, Berkeley
Marzari, Nicola	EPFL, Lausanne, Switzerland

Moore, Joel	University of California, Berkeley
Nozik, Arthur J.	National Renewable Energy Laboratory
Panagiotopoulos, Athanassios Z.	Princeton University
Rubio, Angel	Max Planck Institute, Germany
Scheffler, Matthias	Max Planck Institute, Germany
Shirts, Michael	University of Colorado, Boulder
Vorhees, Peter	Northwestern University
Windus, Theresa	Iowa State University
Co-editor Name	Organization Affiliation
Bertozzi, Carolyn	Stanford University
Quin-Chen, Long	Pennsylvania State University
Scuseria, Gustavo	Rice University
Skinner, Jim	University of Chicago

Graduate and Postdoctoral Advisors and advisees:

Name	Organization Affiliation
Roberto Car	Princeton University
Michele Parrinello	ETHZ, Zurich, Switzerland
GS & PA - Alumni	Organization Affiliation
Aliano, Antonio	Politecnico di Torino, Italy
Wilson, Hugh	CSIRO, Melbourne, Australia
Ahmad, Zeeshan	University of Chicago
Alvarez, Jesus	University of Chicago
Brawand, Nicholas	University of Chicago
Donadio, Davide	University of California
Gaiduk, Alex	University of Chicago
Gerosa, Matteo	University of Chicago
Giberti, Federico	University of Chicago
Gustafson, Jeffrey	University of Chicago
Handlin, Mzuri	University of Chicago
He, Yuping	Sandia National Laboratory
Kaur, Amandeep	University of California Davis
Li, Tianshu	The George Washington University
Li, Yan	Physical Review B
Lu, Deyu	Brookhaven National Laboratory
Murray, Éamonn D.	Imperial College, London, UK
Nguyen, Ngoc Linh	University of Chicago
Pham, Tuan Anh	Lawrence Livermore National Laboratory
Ping, Yuan	University of California, Santa Cruz
Rocca, Dario	University of Lorraine
Savic, Ivana	University College Cork, Ireland
Spanu, Leonardo	Shell Corporation
Wan, Quan	University of Chicago
Ye, Meng	University of Chicago
Zhang, Cui	Princeton University
Zheng, Huihuo	University of Chicago
GS & PA - Current	Organization Affiliation

Bilgin, Anil	University of Chicago
Bousquet, Matthew	University of Chicago
Castillo, Alfonso	University of Chicago
Huang, Benchen	University of Chicago
Jin, Yu	University of Chicago
Le, Lien T.	University of Chicago
Nagura, Jonah	University of Chicago
Onizhuk, Mykyta	University of Chicago
Paul, Arghadwip	University of Chicago
Poteshman, Abigail	University of Chicago
Sheng, Nan	University of Chicago
Xu, Andrew	University of Chicago
Ye, Zifan	University of Chicago
Zhan, Jiawei	University of Chicago
Zhu, Yizhi	University of Chicago
Chatteraj, Swarnabha	University of Chicago
Kundu, Arpan	University of Chicago
Lee, Elizabeth M. Y.	University of Chicago
Melani, Giacomo	University of Chicago
Shin, Yongjin	University of Chicago
Vorwerk, Christian	University of Chicago
Zhang, Cunzhi	University of Chicago
Zhang, Shenli	University of Chicago

Section 6: Software Applications and Packages

Question #1

Please list any software packages used by the project, and indicate if they are open source or export controlled.

Application Packages

Package Name

Quantum Espresso

Indicate whether Open Source or Export Controlled.

Open Source

Package Name

WEST

Indicate whether Open Source or Export Controlled.

Open Source

Package Name

Qbox

Indicate whether Open Source or Export Controlled.

Open Source

Package Name

PyCCE

Indicate whether Open Source or Export Controlled.

Open Source

Section 7: Wrap-Up Questions

Question #1

National Security Decision Directive (NSDD) 189 defines Fundamental Research as "basic and applied research in science and engineering, the results of which ordinarily are published and shared broadly within the scientific community, as distinguished from proprietary research and from industrial development, design, production, and product utilization, the results of which ordinarily are restricted for proprietary or national security reasons." Publicly Available Information is defined as information obtainable free of charge (other than minor shipping or copying fees) and without restriction, which is available via the internet, journal publications, textbooks, articles, newspapers, magazines, etc.

The INCITE program distinguishes between the generation of proprietary information (deemed a proprietary project) and the use of proprietary information as input. In the latter, the project may be considered as Fundamental Research or nonproprietary under the terms of the nonproprietary user agreement. Proprietary information, including computer codes and data, brought into the LCF for use by the project - but not for generation of new intellectual property, etc., using the facility resources - may be protected under a nonproprietary user agreement.

Proprietary Information

Are the proposed project and its intended outcome considered Fundamental Research or Publicly Available Information?

Yes

Will the proposed project use proprietary information, intellectual property, or licensing?

No

Will the proposed project generate proprietary information, intellectual property, or licensing as the result of the work being proposed?

If the response is Yes, please contact the INCITE manager, INCITE@doeleadershipcomputing.org, prior to submittal to discuss the INCITE policy on proprietary work.

No

Question #2

The following questions are provided to determine whether research associated with an INCITE proposal may be export controlled. Responding to these questions can facilitate - but not substitute for - any export control review required for this proposal.

PIs are responsible for knowing whether their project uses or generates sensitive or restricted information. Department of Energy systems contain only data related to scientific research and do not contain personally identifiable information. Therefore, you should answer "Yes" if your project uses or generates data that fall under the Privacy Act of 1974 U.S.C. 552a. Use of high-performance computing resources to store, manipulate, or remotely access any national security information is prohibited. This includes, but is not limited to, classified information, unclassified controlled nuclear information (UCNI); naval nuclear propulsion information (NNPI); and the design or development of nuclear, biological, or chemical weapons or of any weapons of mass destruction. For more information contact the Office of Domestic and International Energy Policy, Department of Energy, Washington DC 20585, 202-586-9211.

Export Control

Does this project use or generate sensitive or restricted information?

No

Does the proposed project involve any of the following areas?

- i. Military, space craft, satellites, missiles, and associated hardware, software or technical data
- ii. Nuclear reactors and components, nuclear material enrichment equipment, components (Trigger List) and associated hardware, software or technical data
- iii. Encryption above 128 bit software (source and object code)
- iv. Weapons of mass destruction or their precursors (nuclear, chemical and biological)

No

Does the proposed project involve International Traffic in Arms Regulations (ITAR)?

No

Question #3

The following questions deal with health data. PIs are responsible for knowing if their project uses any health data and if that data is protected. Note that certain health data may fall both within these questions as well as be considered sensitive as per question #2. Questions regarding these answers to these questions should be directed to the centers or program manager prior to submission.

Health Data

Will this project use health data?

No

Will this project use human health data?

No

Will this project use Protected Health Information (PHI)?

No

Question #4

The PI and designated Project Manager agree to the following:

Monitor Agreement

I certify that the information provided herein contains no proprietary or export control material and is correct to the best of my knowledge.

Yes

I agree to provide periodic updates of research accomplishments and to acknowledge INCITE and the LCF in publications resulting from an INCITE award.

Yes

I agree to monitor the usage associated with an INCITE award to ensure that usage is only for the project being described herein and that all U. S. Export Controls are complied with.

Yes

I understand that the INCITE program reserves the right to periodically redistribute allocations from underutilized projects.

Yes

Section 8: Outreach and Suggested Reviewers

Question #1

By what sources (colleagues, web sites, email notices, other) have you heard about the INCITE program? This information will help refine our outreach efforts.

Outreach

By what sources (colleagues, web sites, email notices, other) have you heard about the INCITE program? This information will help refine our outreach efforts.

DOE program managers

Question #2

Suggested Reviewers

Suggest names of individuals who would be particularly suited to assess the proposed research.

Tuan Anh Pham (pham16@lbl.gov), LLNL

Section 9: Testbed Resources

Question #1

The ALCF and OLCF have test bed resources for new technologies, details below. If you would like access to these resources to support the work in this proposal, please provide the information below. (1 Page Limit)

The OLCF Quantum Computing User Program is designed to enable research by providing a broad spectrum of user access to the best available quantum computing systems, evaluate technology by monitoring the breadth and performance of early quantum computing applications, and Engage the quantum computing community and support the growth of the quantum information science ecosystems. More information can be found here: <https://www.olcf.ornl.gov/olcf-resources/compute-systems/quantum-computing-user-program/quantum-computing-user-support-documentation>.

The ALCF AI Testbed provides access to next-generation of AI-accelerator machines to enable evaluation of both hardware and workflows. Current hardware available includes Cerebras C-2, Graphcore MK1, Groq, Habana Gaudi, and SambaNova Dataflow. New hardware is regularly acquired as it becomes available. Up to date information can be found here: <https://www.alcf.anl.gov/alcf-ai-testbed>.

Describe the experiments you would be interested in performing, resources required, and their relationship to the current proposal. Please note, these are smaller experimental resources and a large amount of resources are not available. Instead, these resources are to explore the possibilities for these technologies might innovate future work. This request does not contribute to the 15-page proposal limit.

2023_INCITE_Quantum_final.pdf

The attachment is on the following page.

QUANTUM COMPUTING RESOURCES

Within this program we will use a quantum embedding theory to predict and understand the electronic structure of correlated electronic states of defects in materials, of relevance to quantum sensing and metrology. **The parameters of effective Hamiltonians, computed from first-principles within this project, will form the input of quantum simulations on near-term quantum computers.**

The main deliverables are: (i) predictions of the structural and electronic properties of heterogeneous systems, to be compared with experiments in order to obtain an integrated mechanistic understanding of the interaction of point defects in materials with light; (ii) validated data for systems of interest for quantum information science on large scale DOE platform, which will serve as reference results and will be used to **carry out simulations of quantum materials on near-term quantum computers.**

We request access to the IBM Q Hub, which will be achieved using the Qiskit software. The team has recently published a work that discusses the use of quantum error mitigation strategies to simulate effective Hamiltonians derived from quantum embedding theories [Huang2022].

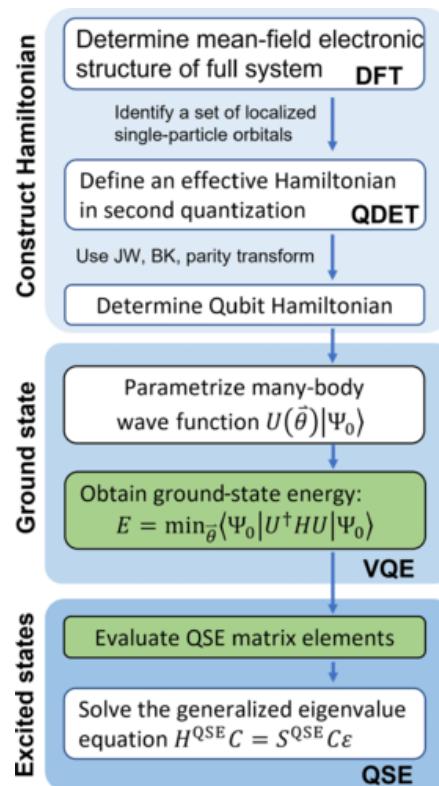


Figure: The work flow used to simulate the ground- and excited-state energies of spin defects, with operations executed on a quantum computer indicated in green. The transformation from a second quantized to a qubit Hamiltonian may be obtained with a Jordan-Wigner (JW), Bravyi-Kitaev (BK), or parity transformation. DFT and QDET denote calculations carried out using density functional theory and the quantum defect embedding theory, respectively. VQE and QSE denote the variational quantum eigensolver and quantum subspace expansion algorithms used for ground- and excited-state calculations, respectively. See Ref. [Huang2022] for definition of the equations.

Reference:

[Huang2022] B. Huang, M. Govoni, G. Galli, *Simulating the Electronic Structure of Spin Defects on Quantum Computers*, PRX Quantum 3, 010339 (2022). DOI: 10.1103/PRXQuantum.3.010339