# Quantum Materials from First Principles

## Galli Group

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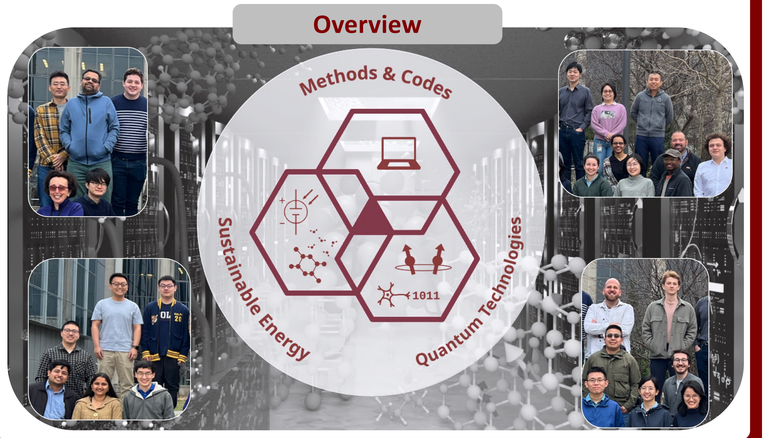
**We develop theoretical and computational methods to predict and engineer material and molecular properties from first principles.**

## Overview




## Methods and Code Development

**We develop methods based on quantum mechanics, and open-source software** to understand and predict the properties of solids and molecules at the microscopic scale.

**First Principles Molecular Dynamics (FPMD)**

**Many Body Perturbation Theory (MBPT)**
M. Govoni and G. Galli, JCTC 2015, 2018; V. Yu et al. 2024 (submitted).

**Time dependent DFT (TDDFT)**
Y. Jin et al. JCTC 2023.

**Quantum Embedding Theory (QDET)**
M. He, M. Govoni, and G. Galli, npj Comput. Mater. 2020; N. Sheng, C. Vorwerk, M. Govoni, and G. Galli, JCTC 2022.

**Cluster-Correlation Expansion (CCE)**
M. Onizhuk and G. Galli 2024 (submitted).

**Hybrid Density Functionals**
J. Zhan, M. Govoni, and G. Galli JCTC 2023.

**Path Integral Molecular Dynamics (PIMD)**
A. Kundu et al. Phys. Rev. Mater. Lett. 2021; PNAS 2022; JPCL 2024.

Qbox First-Principles Molecular Dynamics

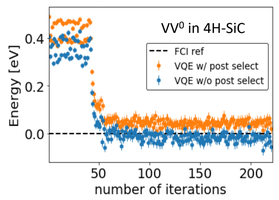
WEST!

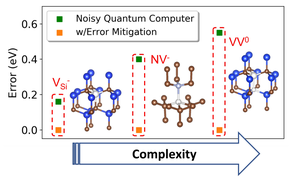
pycce

Codes are developed within the Midwest Integrated Center for Computational Materials: MICCoM

## Quantum Simulations on Quantum Computers

We use QDET to simulate the eigenstates of **spin-defect** systems with variational quantum eigensolver and shadow tomography-based algorithms.



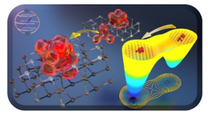




B. Huang, N. Sheng, M. Govoni, and G. Galli, JCTC 2023; B. Huang, M. Govoni, and G. Galli, PRX Quantum 2022; B. Huang et al. Phys. Rev. Research 2024.

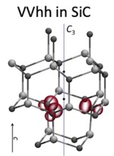
We reduce the noise effects with the **zero-noise extrapolation** technique.

## Spin Qubits for Quantum Information Technologies

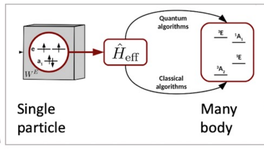
We aim to understand the atomic and electronic structure, and coherence properties of novel **spin defects in semiconductors** as platforms for **quantum information processing**.



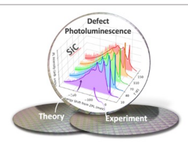
**Free energy surfaces**
Advanced sampling coupled *w/quantum forces*



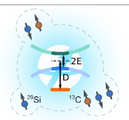

**Electronic properties**
Density functional theory (DFT) and quantum chemistry methods




**Many body states**
Quantum Embedding Theories




**Optical properties**
Time Dependent DFT or the Bethe Salpeter Equation

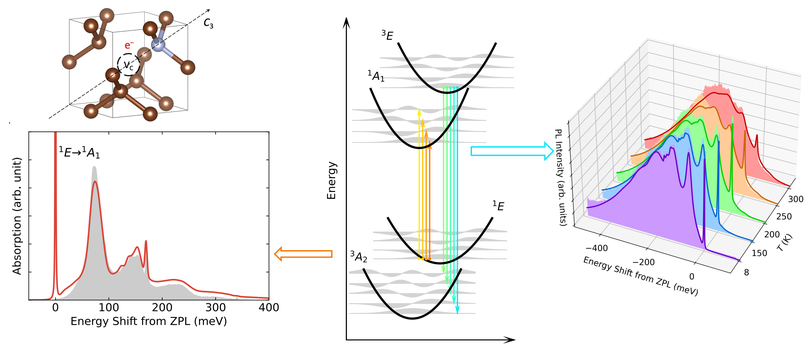



**Spin and coherence properties**
Spin Hamiltonians with ab initio parameters

## Computational Spectroscopy of Spin Defects

We develop *ab initio* approaches to **compute optical spectra of point defects in semiconductors and insulators.**

The TDDFT calculation of vibrationally resolved photoluminescence spectra, with excited state analytical forces, including spin-flip, is implemented in the **WEST** code.

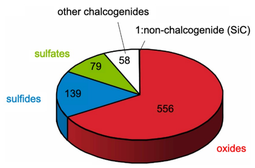



Yu Jin, Victor Wen-zhe Yu, Marco Govoni, Andrew C Xu, and Giulia Galli, JCTC 2023; Yu Jin, Marco Govoni, and Giulia Galli, npj Comput. Mater. 2022; C. P. Anderson*, E. O. Glen*, C. Zeledon, A. Bourassa, Y. Jin, Y. Zhu, C. Vorwerk, A. L. Crook, H. Abe, J. Ul-Hassan, T. Ohshima, N. T. Son, G. Galli, and D. D. Awschalom, Sci. Adv. 2022; Y. Zhu, B. Kovos, M. Onizhuk, D. Awschalom, and G. Galli, Phys. Rev. Mater. 2021; Y. Jin, M. Govoni, G. Wolfowicz, S. E. Sullivan, F. J. Heremans, D. D. Awschalom, and G. Galli, Phys. Rev. Mater. 2021.

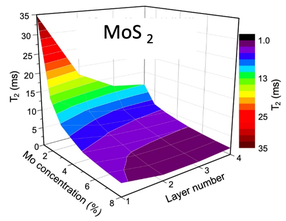
## Coherence Properties of Spin Qubits

We use CCE-based methods **to understand decoherence dynamics in spin qubits:**

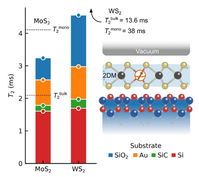
High-throughput predictions: oxides



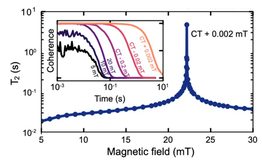

2D Systems



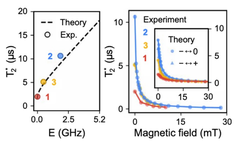

van der Waals heterostructures



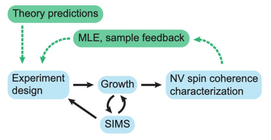

Clock transitions in oxides




Molecular qubits




Guiding growth experiments



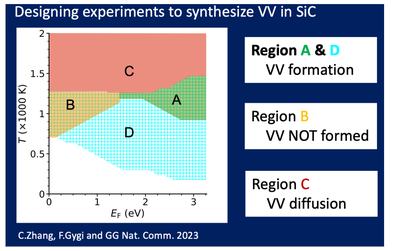

M. Onizhuk, Y.-X. Wang, J. Nagura, A. A. Clerk, and G. Galli, PRL 2024; M. Onizhuk and G. Galli, PRB 2023; J. C. Marcks*, M. Onizhuk*, N. Delegan, Y.-X. Wang, M. Fukami, M. Watts, A. A. Clerk, F. J. Heremans, G. Galli, and D. D. Awschalom, Phys. Rev. Mater. 2024; S. L. Bayliss, P. Deb, D. W. Laorenza, M. Onizhuk, G. Galli, D. E. Freedman, and D. D. Awschalom, PRX 2022; M. Onizhuk and G. Galli, Adv. Theory Simul. 2021; Y. Zhu, B. Kovos, M. Onizhuk, D. Awschalom, and G. Galli, Phys. Rev. Mater. 2021; M. Onizhuk and Giulia Galli, Appl. Phys. Lett. 2021; M. Onizhuk, K. C. Miao, J. P. Blanton, H. Ma, C. P. Anderson, A. Bourassa, D. D. Awschalom, and G. Galli, PRX Quantum 2021; A. Bourassa, C. P. Anderson, K. C. Miao, M. Onizhuk, H. Ma, A. L. Crook, H. Abe, J. Ul-Hassan, T. Ohshima, N. T. Son, G. Galli, and D. D. Awschalom, Nat. Mater. 2020.

## Formation of Spin Defects

We investigated the formation of a promising defect in SiC (double vacancy-VV) for quantum technology applications.

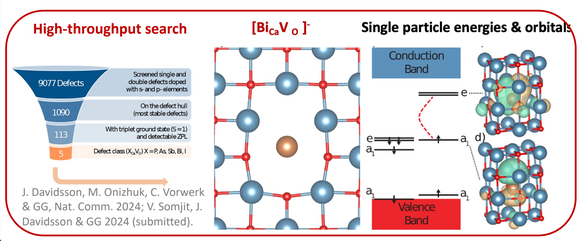
We combined **first principles MD & advanced sampling.**

We predicted the **optimal annealing temperature** and how to engineer the **Fermi level** to optimize the VV's yield in silicon carbide.

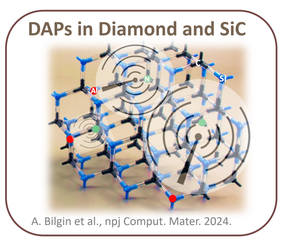



## Prediction of New Platforms

We search for new platforms by carrying out **high-throughput searches and by exploring defect long** range interactions, e.g., dipolar interaction between **donor-acceptor pairs (DAPs).**




J. Davidsson, M. Onizhuk, C. Vorwerk & GG, Nat. Comm. 2024; V. Somjit, J. Davidsson & GG 2024 (submitted).




A. Bilgin et al., npj Comput. Mater. 2024.

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