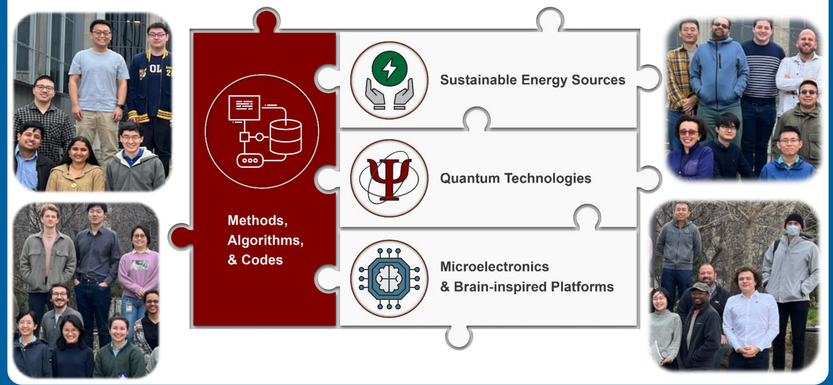
The GALLI GROUP

QUANTUM SIMULATIONS FOR QUANTUM TECHNOLOGIES

THE UNIVERSITY OF CHICAGO
PRITZKER SCHOOL OF MOLECULAR ENGINEERING
THE UNIVERSITY OF CHICAGO | DEPARTMENT OF CHEMISTRY

We develop theoretical and computational methods to predict and engineer the properties of materials and molecules

GROUP OVERVIEW



DEVELOPMENT OF METHODS AND CODES

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

WEST!
pycce
Qbox First-Principles Molecular Dynamics

Time-dependent density functional theory (TDDFT)
Jin et al., J Chem Theory Comput 2023

Many-body perturbation theory (MBPT)
Govoni & Galli, J Chem Theory Comput 2015, 2018; Yu & Govoni, J Chem Theory Comput 2022; Yu et al., J Chem Theory Comput 2024

Quantum defect embedding theory (QDET)
Sheng et al., J Chem Theory Comput 2022; Vorwerk et al., Nat Comput Sci 2022; Chen et al., J Chem Theory Comput 2025

Cluster-correlation expansion (CCE)
Onizhuk & Galli, Rev Mod Phys 2024

Hybrid density functionals
Zhan et al., J Chem Theory Comput 2023, Phys Rev Mater 2025

Stochastic methods for electron-phonon interaction
Kundu & Galli, J Chem Theory Comput 2023, J Phys Chem Lett 2024

We use first principles molecular dynamics

INTEGRATED CAPABILITIES AT WORK

Search for suitable defects
Combinatorial searches with automatic workflows

Finite T properties
Simulations as a function of temperature with first principle MD based on DFT

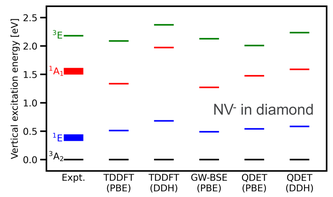
Electronic properties
Calculations of excited states with many body perturb. theory, time-dep. DFT & quantum embedding

Vibronic coupling
Calculations of electron-phonon interaction with dynamical and stochastic methods

Spin dynamics
Dynamical simulations of noisy spin environm. with spin Hamiltonians and cluster-expansions

Scaling of algorithms & Simulations of realistic systems in collaboration with experiments
For all properties, highly scalable, parallel codes, on CPU & GPU
&
Excited states (Fermionic Hamiltonians) on quantum computers
VQE & Shadow tomography-based algorithms + noise mitigation

COMPARISON AND VALIDATION OF METHODS



Unequivocal comparison of **converged** vertical excitation energies computed using **TDDFT**, **GW-BSE**, and **QDET**

All methods qualitatively agree with experiments and each other, though quantitative differences are present

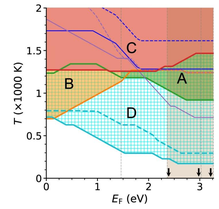
Jin et al., J Chem Theory Comput 2023; Yu et al., J Chem Theory Comput 2024; Chen et al., J Chem Theory Comput 2025

FORMATION OF SPIN DEFECTS

We investigated the formation of a promising spin defect (divacancy, VV) in silicon carbide for quantum technologies, combining **first-principles MD** & **advanced sampling**

We predicted the **optimal annealing temperature** and the **Fermi level** that optimizes the **VV's yield**

Zhang et al., Nat Commun 2023; Kimura et al., Appl. Phys. Lett 2025

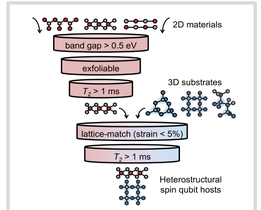


Regions A & D
VV formation

Region B
VV not formed

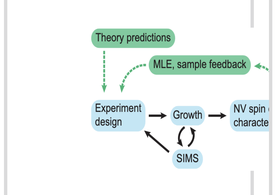
Region C
VV diffusion

COHERENCE TIMES OF SPIN DEFECTS



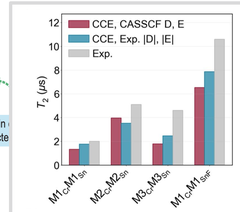
High-throughput search of 2D host materials

Toriyama et al., 2025 (arXiv:2509.00222)



Computationally guided growth processes

Marcks et al., Phys Rev Mater 2024

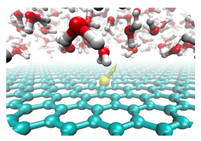


Engineering spin structure of molecular qubits

Baldinelli et al., J Am Chem Soc 2025

QUANTUM SENSING APPLICATIONS

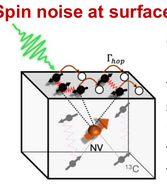
Interfaces between 2D material and water



Coherence time T_2 is sensitive to temperature, density and hydrogen bonding arrangements

Castillo et al., 2025 (arXiv:2507.01220)

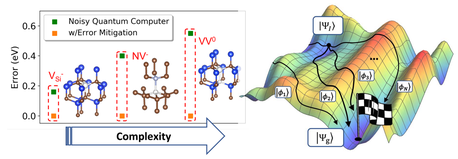
Spin noise at surfaces from first principles



Computational study of NV center coherence times as a function of surface orientation, reconstruction, chemical functionalization, and the density of unpaired (dark) spins.

Nagura et al., 2025 (in preparation)

QUANTUM SIMULATIONS ON QUANTUM COMPUTERS



We solved Fermionic Hamiltonians derived from **quantum embedding** on **quantum computers** (IBM and ion-Q platforms)

- Simulated **realistic spin and developed noise mitigation** techniques

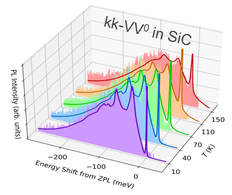
Huang et al., PRX Quantum 2022, J Chem Theory Comput 2023, Phys Rev Research 2024; Ma et al., npj Comp Mat 2020

COMPUTATIONAL SPECTROSCOPY

We have developed methods and software to computationally characterize the complete **optical cycle** of spin qubits to **interpret and guide experiments**

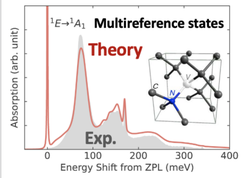
Photoluminescence (PL) spectra (w/ temperature dependence) from generating function approach

Jin et al., Phys Rev Mater 2021



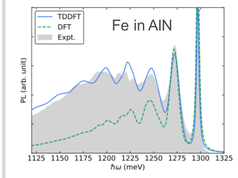
Absorption spectra from spin-flip hybrid TDDFT w/ optimized excited-state geometries

Jin, Govoni & Galli, npj Comput Mater 2022



PL spectra of transition metal impurities in ionic crystals from spin-flip hybrid TDDFT w/ optimized excited-state geometries

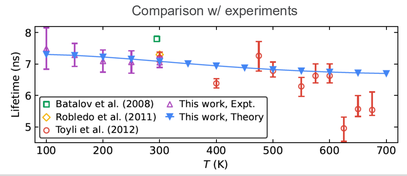
Otis et al., J Phys Chem Lett 2025



Inter-system crossing (ISC) rates

- Excited-state geometries w/ spin-flip hybrid TDDFT
- Spin-orbit coupling matrix elements w/ multireference states from QDET
- Overlap functions w/ ab initio phonons

Jin et al., Phys Rev Lett 2025



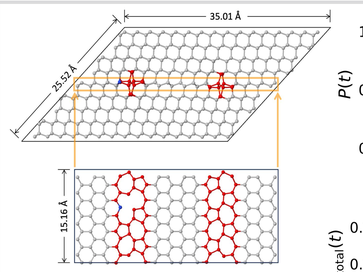
TOWARDS DEVICE APPLICATIONS

We conducted the first **beyond-DFT** computational study of **point defects** in proximity to 1D **mesoscopic defects**:

- Defect formation energy & stability (hybrid DFT)
- Electronic structure (hybrid DFT, GW)
- Vertical excitation energy (hybrid TDDFT, BSE, QDET)
- Optical cycle: ISC, PL, ODMR (hybrid TDDFT, QDET)
- Zero-field splitting (ZFS) & hyperfine parameters
- Coherence time (CCE)

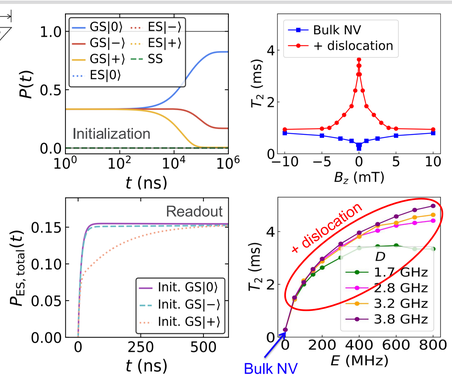
We predicted several NV configurations near a dislocation core retain the desired properties to be **functional as spin qubits**, potentially forming a 1D chain of **coupled qubits**

- **Optical initialization & readout** comparable to NV in bulk diamond
- **Enhanced spin coherence time** (T_2) compared to NV in bulk diamond



Diamond supercell w/ **1727 atoms**
+2 glide dislocations
+1 NV center

Zhang et al., 2025 (arXiv:2507.12387)



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