Tutorial: Quantum Defect Embedding Theory

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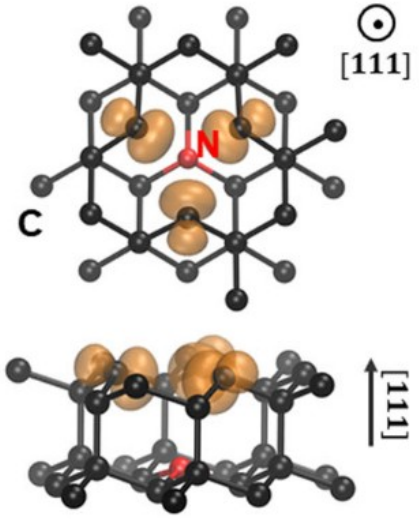
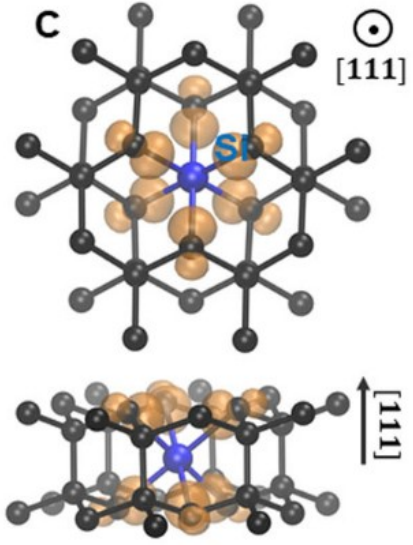
Spin Defects in Semiconductors

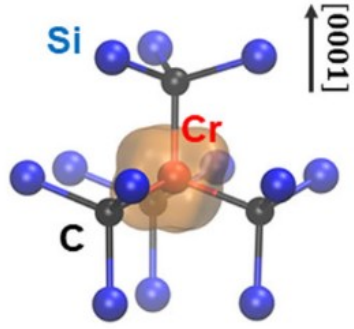
(a) **NV in diamond**



(b) **SiV in diamond**

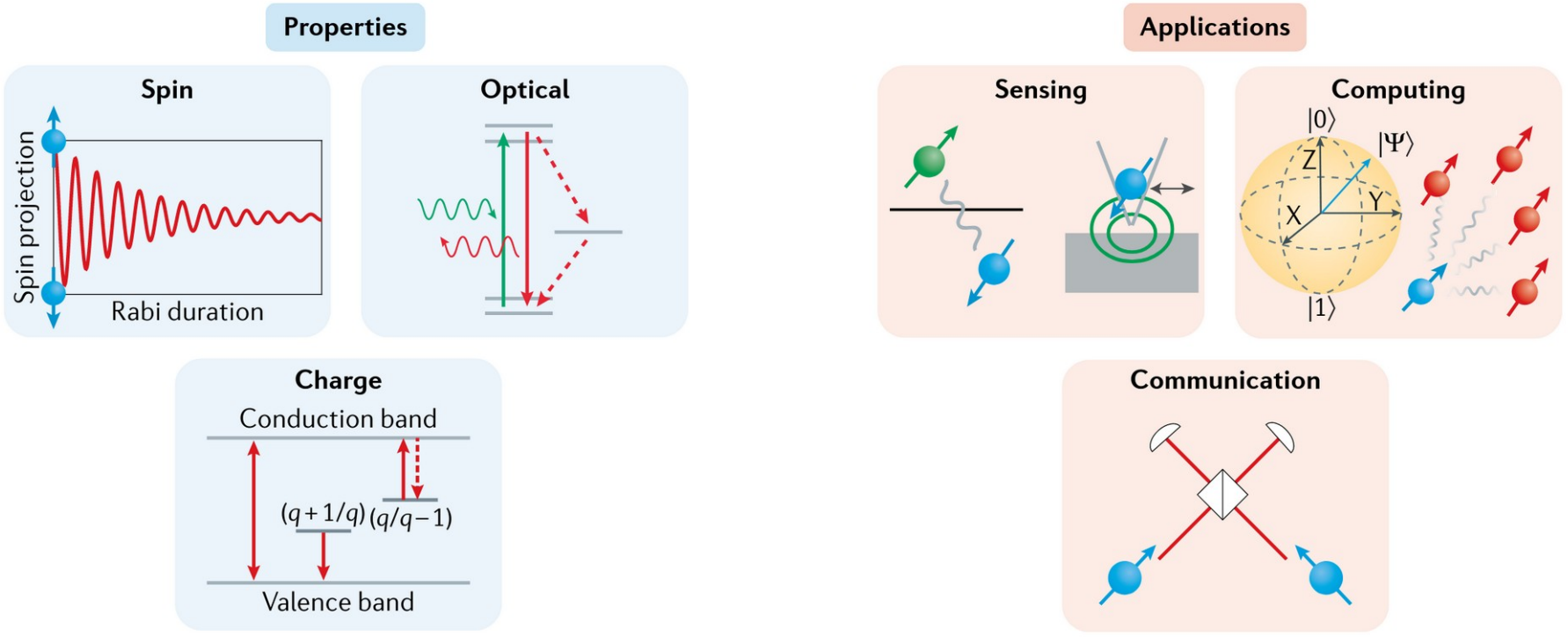


(c) **Cr in 4H-SiC**



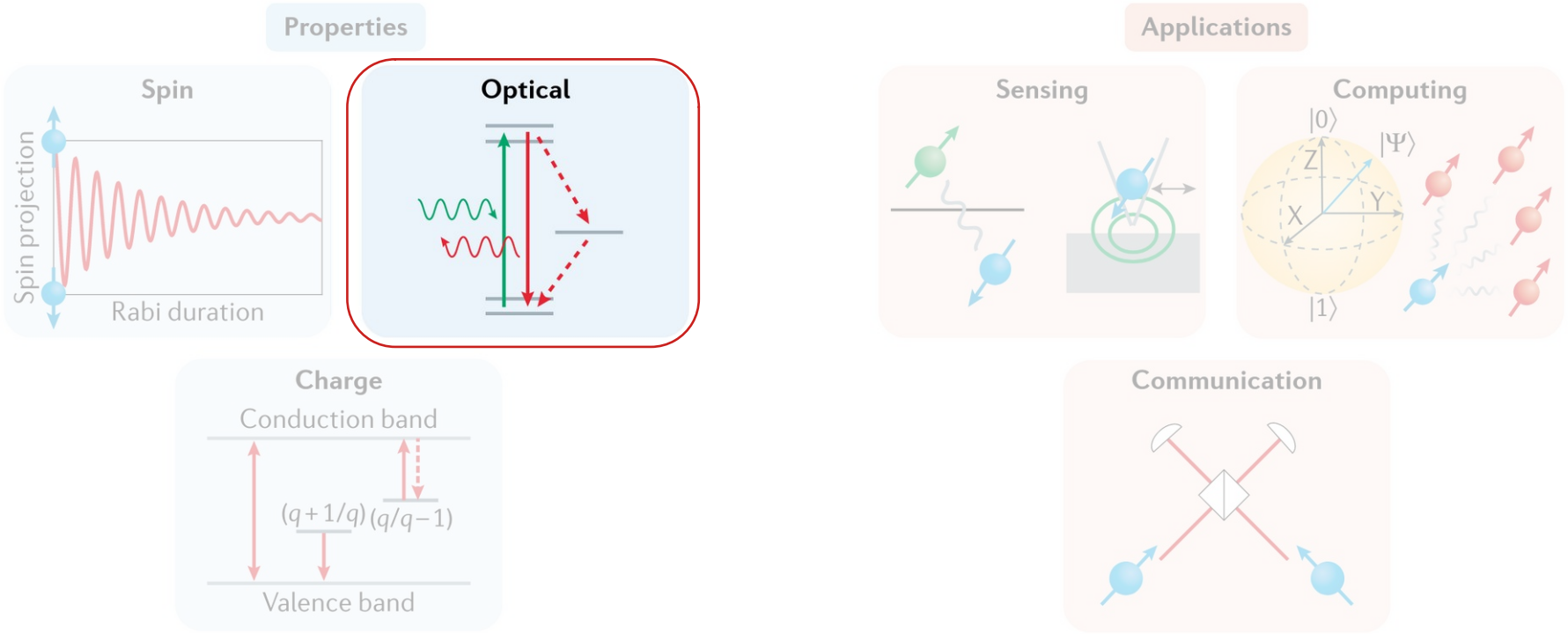
H. Ma, M. Govoni, and G. Galli, npj Comput. Mat. **6**, 85 (2020).

Relevance of Spin Defects in Semiconductors



G. Wolfowicz *et al.*, Nature Rev. Mat. **6**, 906 (2021).
J.R. Weber *et al.*, PNAS **19**, 8513 (2010).

Relevance of Spin Defects in Semiconductors



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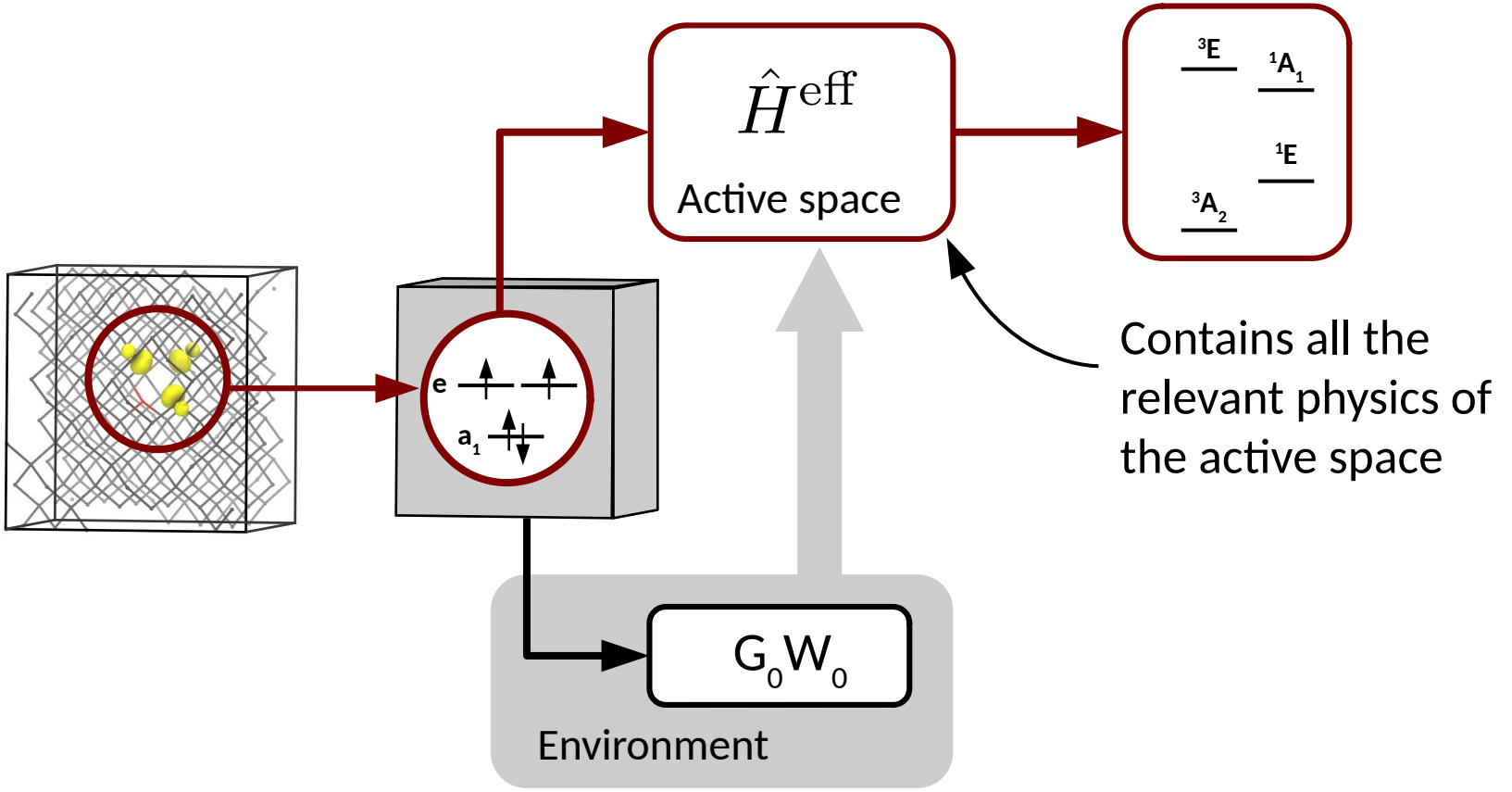
Quantum Defect Embedding Theory for Correlated Defect States



[1] N. Sheng, C. Vorwerk *et al.*, J. Chem. Theory Comput. **18**, 6, 3512 (2022).

Effective Hamiltonian in Quantum Defect Embedding

$$\hat{H}^{\mathrm{eff}} = \sum_{ij}^{A} t_{ij}^{\mathrm{eff}} \hat{a}_i^\dagger \hat{a}_j + \frac{1}{2} \sum_{ijkl}^{A} v_{ijkl}^{\mathrm{eff}} \hat{a}_i^\dagger \hat{a}_j^\dagger \hat{a}_l \hat{a}_k$$

Effective one-body terms

$$t^{\mathrm{eff}} = H^{\mathrm{KS}} - t^{\mathrm{dc}}$$

Double counting accounts for exchange and correlation within G_0W_0

Effective two-body terms

$$v^{\mathrm{eff}} = W_0^R(\omega = 0)$$

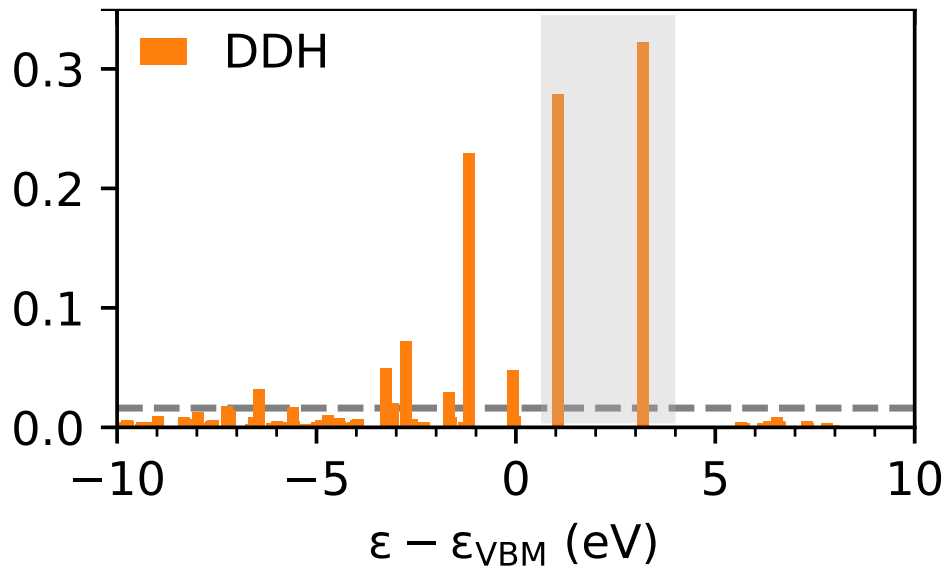
Partial screening due to the environment within random-phase approximation (RPA)

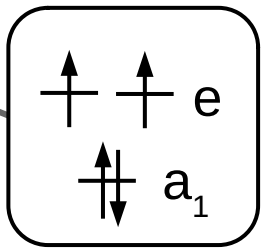
[1] N. Sheng, C. Vorwerk *et al.*, J. Chem. Theory Comput. **18**, 6, 3512 (2022).

Localized Defect Orbitals of the NV^- Center in Diamond

$$L_V = \int d^3r |\phi_i^{\mathrm{KS}}(\mathbf{r})|^2$$

Integral over charge density around defect





Strongly localized in-gap states

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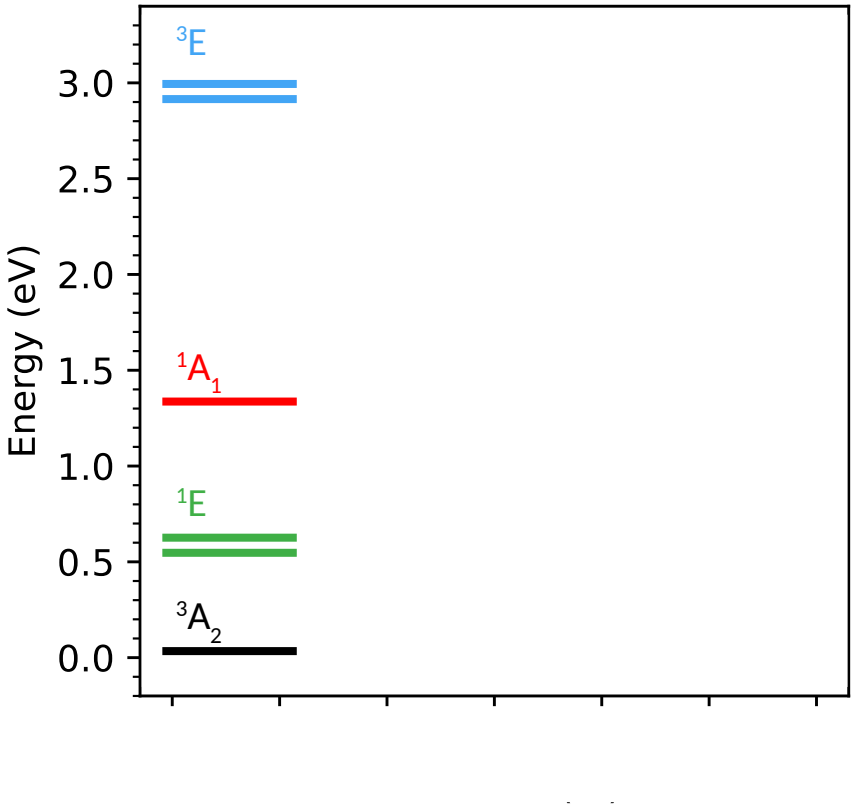
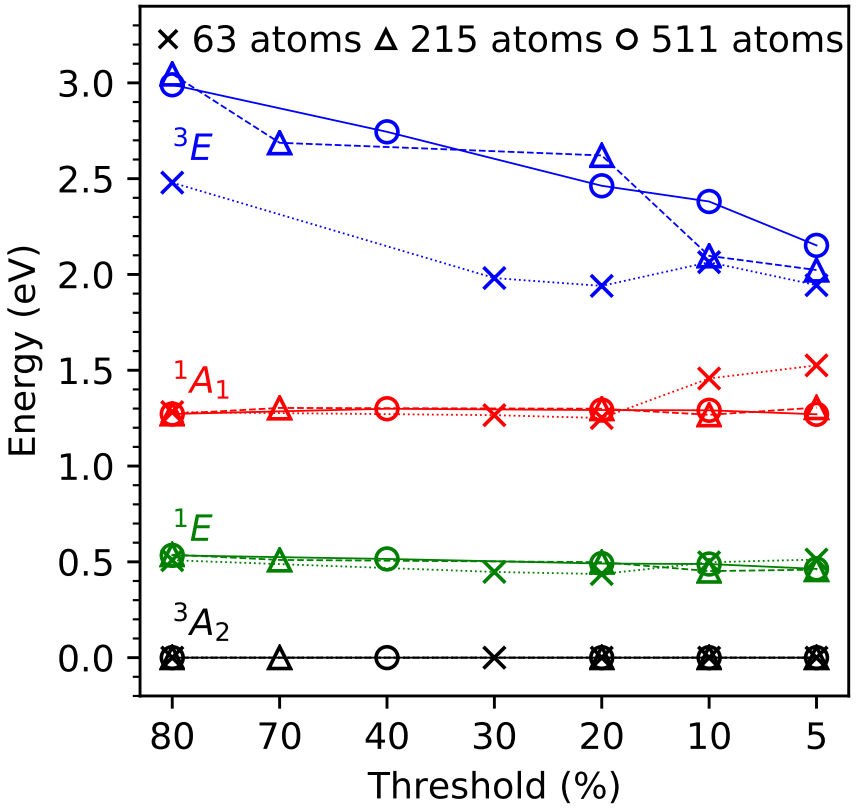
Correlated Excitations of the NV^- Center in Diamond



Correlated Excitations of the NV^- Center in Diamond



Slow convergence

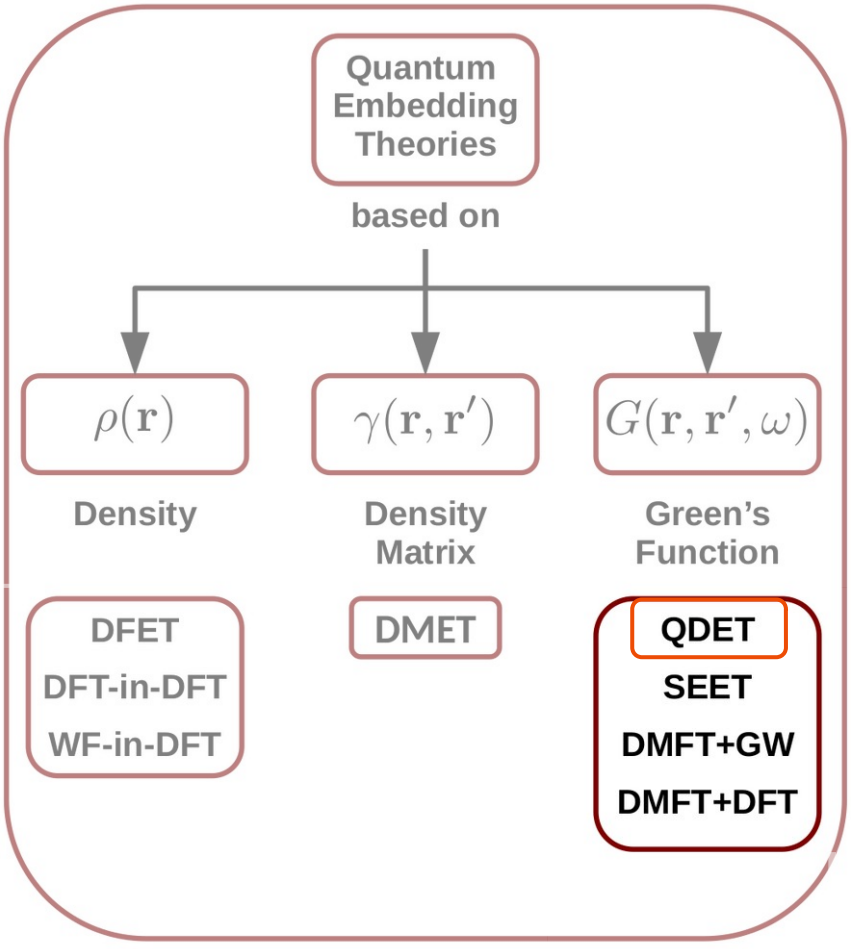
Low-energy excitations converge quickly

Correlated Excitations of the NV^- Center in Diamond

	^1E	$^1A_1$	^3E
Exp.[1]	0.34 - 0.43*	1.51-1.60*	2.18
QDET[1]	0.46	1.27	2.15
GW + BSE[2]	0.40	0.99	2.32
$C_{85}H_{76}N^-$ CASSCF(6,6)[3]	0.25	1.60	2.14

[1] N. Sheng, C. Vorwerk *et al.*, J. Chem. Theory Comput. **18**, 6, 3512 (2022).
[2] Y. Ma *et al.*, Phys. Rev. B **81**, 041204 (2010).
[3] C. Bhandari *et al.*, Phys. Rev. B 103, 014115 (2021).

Overview of Quantum Embedding Theories



C. Vorwerk, N. Sheng *et al.*, Nat. Comp. Sci. **2**, 424 (2022).

Summary

- QDET is an embedding theory to describe the electronic structure of strongly localized orbitals in solids.
- QDET is rigorously derived in the framework of Green's function theory, and is efficiently implemented in WEST.
- QDET yields accurate results in good agreement with experimental values for a wide range of spin defects in semiconductors.

NOW YOU CAN TRY QDET YOURSELF!

How to Get Started

Terminal 1

```
$ ssh <username>@bebop.lcrc.anl.gov

$ srun --pty -A MICCOM-TRAIN --reservation miccom_day2_pm
  -p knlall -N 1 -t 01:30:00 /bin/bash

$ source /lcrc/project/MICCoM-train/load_bebop_env.sh

$ miccom_start_jupyter

  Your compute node is  :  knld-0019
  Your port number is   :  27055
  Starting jupyter notebook ...


  To access the notebook, open this file in a browser:
    file:///gpfs/fs1/home/yuw/.local/...
  Or copy and paste one of these URLs:
    http://localhost:27055/?token=f86350...
   or http://127.0.0.1:27055/?token=f86350...
```

Terminal 2

```
$ ssh -L 27055:localhost:27055 <username>@bebop.lcrc.anl.gov

$ ssh -L 27055:localhost:27055 knld-0019


$ cp -r /lcrc/project/MICCoM-train/qdet_tutorial $HOME
```