Tutorial: Quantum Defect Embedding Theory

Christian Vorwerk\textsuperscript{1} and Marco Govoni\textsuperscript{2}

\textsuperscript{1}Pritzker School of Molecular Engineering, University of Chicago
\textsuperscript{2}Materials Science Division and Center for Molecular Engineering, Argonne National Laboratory
Spin Defects in Semiconductors

(a) NV in diamond (b) SiV in diamond (c) Cr in 4H-SiC

Relevance of Spin Defects in Semiconductors

J.R. Weber et al., PNAS 19, 8513 (2010).
Relevance of Spin Defects in Semiconductors

J.R. Weber et al., PNAS 19, 8513 (2010).
Quantum Defect Embedding Theory for Correlated Defect States

$\hat{H}^{\text{eff}}$

Active space

Contains all the relevant physics of the active space

$G_0W_0$

Environment

$^1E$ $^1A_1$

$^3E$ $^1E$

$^3A_2$

$^3A_2$

$e$ $a_1$

$^1A_1$

$^1E$

$^3E$

Effective Hamiltonian in Quantum Defect Embedding

\[ \hat{H}^{\text{eff}} = \sum_{ij}^{A} t_{ij}^{\text{eff}} \hat{a}_i \hat{a}_j + \frac{1}{2} \sum_{ijkl}^{A} v_{ijkl}^{\text{eff}} \hat{a}_i \hat{a}_j \hat{a}_l \hat{a}_k \]

Effective one-body terms

\[ t^{\text{eff}} = H^{\text{KS}} - t^{\text{dc}} \]

Double counting accounts for exchange and correlation within G_0W_0

Effective two-body terms

\[ v^{\text{eff}} = W_0^R(\omega = 0) \]

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

---

Localized Defect Orbitals of the NV Center in Diamond

\[ L_V = \int d^3 r |\phi_i^{KS}(r)|^2 \]

Integral over charge density around defect

Strongly localized in-gap states

Correlated Excitations of the NV\textsuperscript{−} Center in Diamond
Correlated Excitations of the NV Center in Diamond

Low-energy excitations converge quickly

Slow convergence

Increasing number of orbitals in active space
## Correlated Excitations of the NV\(^{-}\) Center in Diamond

<table>
<thead>
<tr>
<th>Method</th>
<th>(^1E)</th>
<th>(^1A_1)</th>
<th>(^3E)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exp.(^1)</td>
<td>0.34 - 0.43(^*)</td>
<td>1.51-1.60(^*)</td>
<td>2.18</td>
</tr>
<tr>
<td>QDET(^1)</td>
<td>0.46</td>
<td>1.27</td>
<td>2.15</td>
</tr>
<tr>
<td>GW + BSE(^2)</td>
<td>0.40</td>
<td>0.99</td>
<td>2.32</td>
</tr>
<tr>
<td>C(<em>{85})H(</em>{76})N(^-) CASSCF(6,6)(^3)</td>
<td>0.25</td>
<td>1.60</td>
<td>2.14</td>
</tr>
</tbody>
</table>

Overview of Quantum Embedding Theories

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