SAND2018-10072C

Optimization of low-valence Ga PAWs for faster DFT calculations of point defects in GaN

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Introduction

- Want to study defects in gallium nitride (GaN) using plane-wave density functional theory (DFT) and projector augmented wave data sets (PAWs)
- However, for the large system sizes required for accurate defect studies, there is a strong need to reduce computational expense of the DFT calculations
- If quantum molecular dynamics (QMD) studies are required, existing Ga PAWs are too slow because of supercell sizes and time steps required
- **Objective:** use optimization to tune parameters of a new 3-electron Ga PAW, evaluating scattering properties and zb-GaN lattice parameter for each PAW. Then, evaluate accuracy and speedup of new PAW in defect calculations in GaN to see if it speeds up the calculations without adversely affecting the results

Methods: PAW Optimizations

generate all-electron reference solution (lattice constant)

generate PAWs with parameter sweep

compare to reference

for each PAW

test lattice constant accuracy

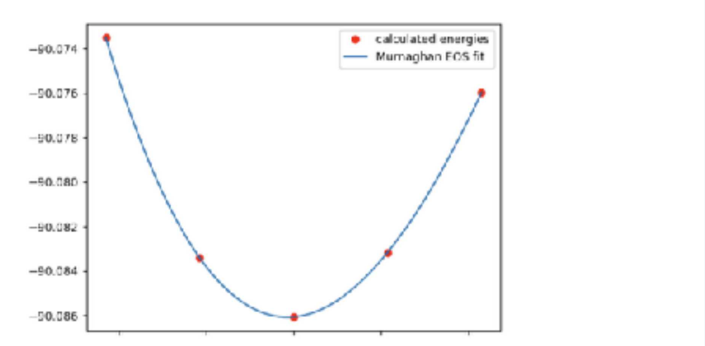
test scattering accuracy

choose best PAW

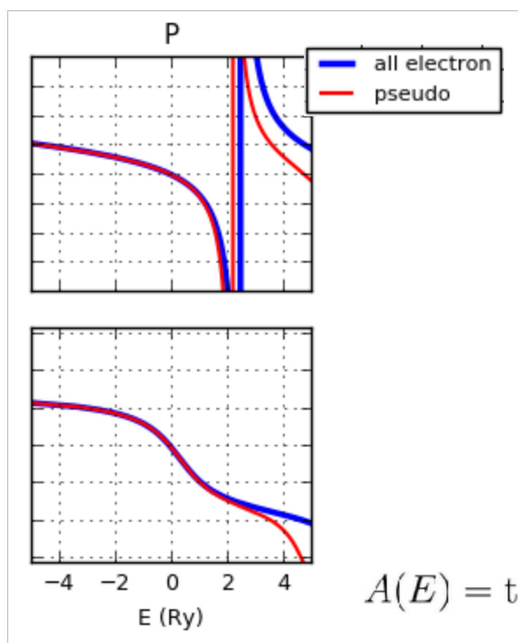
test in further calculations in materials of interest

```
parameter sweep:
     min max steps
r_c    2   3    11
E_P    0  10    11
E_D    0  10    11
E_D_2  0  10    11
total points: 14641
```

Parameter sweep space where r_c is the PAW radius in Bohr, and E_P, E_D1, and E_D2 are the projector energies for the additional P and two additional D projectors



$$S_s = |a_{\text{PAW}} - a_{\text{elk}}|$$



$$A(E) = \tan^{-1} \left.\frac{\psi'(E,r)}{\psi(E,r)}\right|_{r=r_c}$$

$$S_{\text{atom}} = \sum_l \sqrt{\frac{1}{N} \sum_E (A_{\text{PS}}(E) - A_{\text{AE}}(E))^2}$$

Methods: Defect Calculations

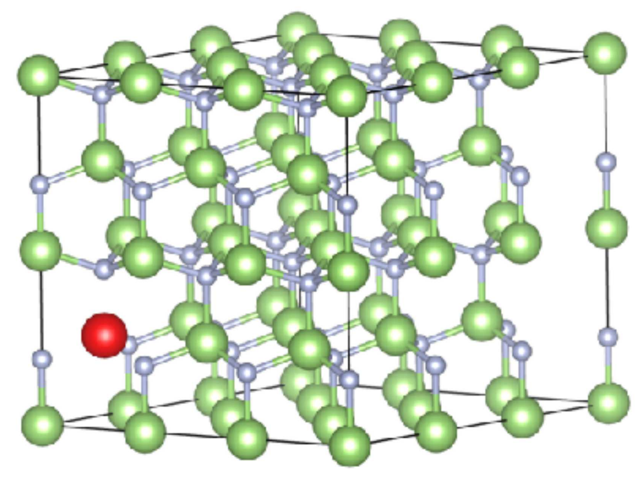
To get defect transition levels:

- remove Ga atom from unit cell
- relax structure
- calculate total energy
- repeat relaxation and total energy calculation for different charge states
- Calculate transition levels:

$$E_L[q-1,q] = E[q-1] - E[q] - E_{\text{shift}}$$

where q is charge state and E_{shift} is *usually* the valence band edge

- the charge is no longer localized on the defect when the levels are at the bounds (red lines in results)

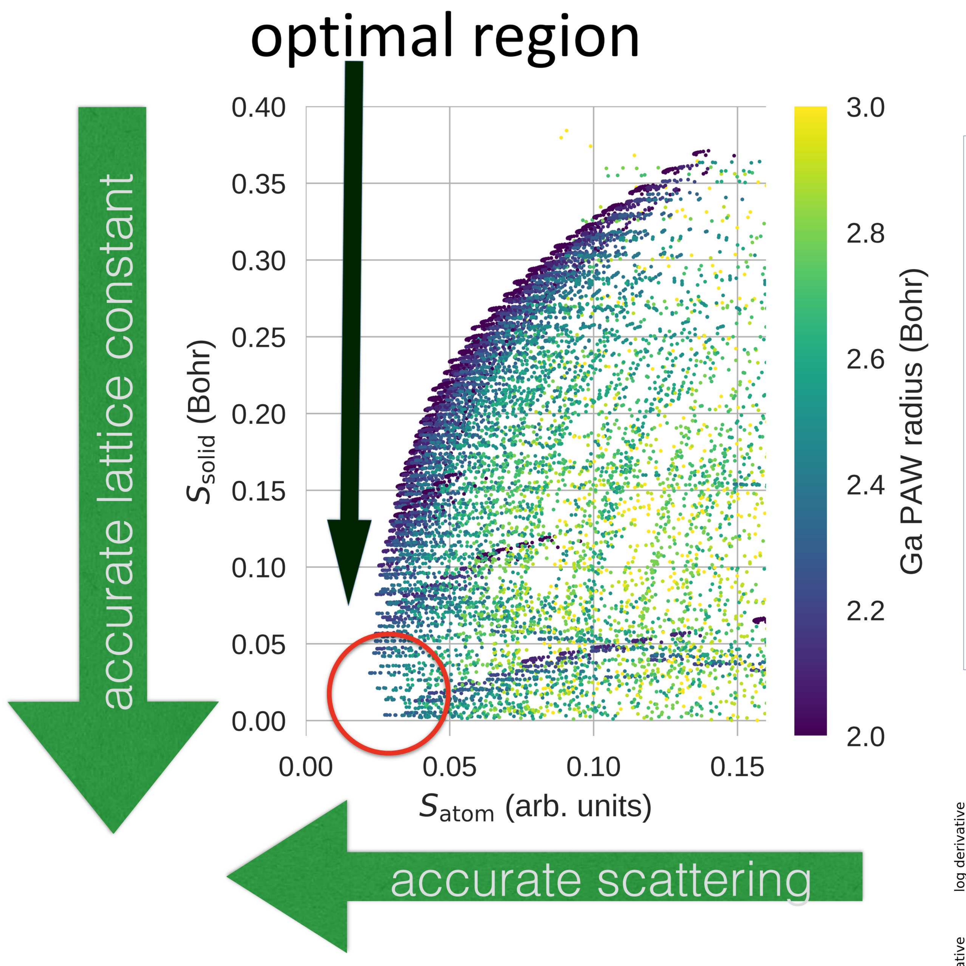


Ga vacancy in wurtzite GaN

To get band gap and bounds:

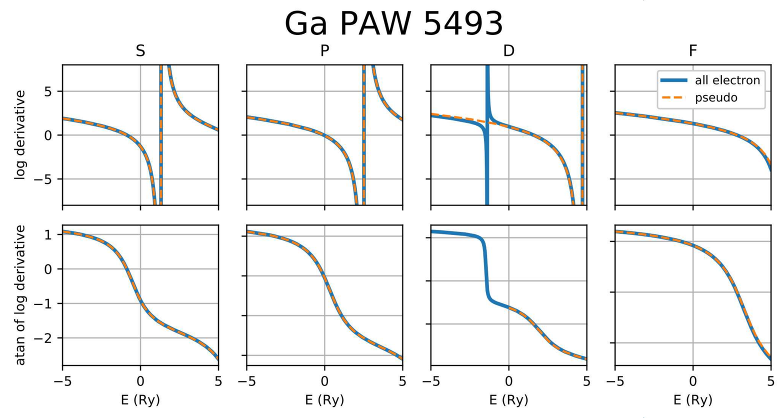
- Band gap comes from neutral bulk structure with no defect
- The bounds are the energies required to add or remove an electron from the unit cell, *which are only equivalent to the band edges in an infinite size cell*

Results



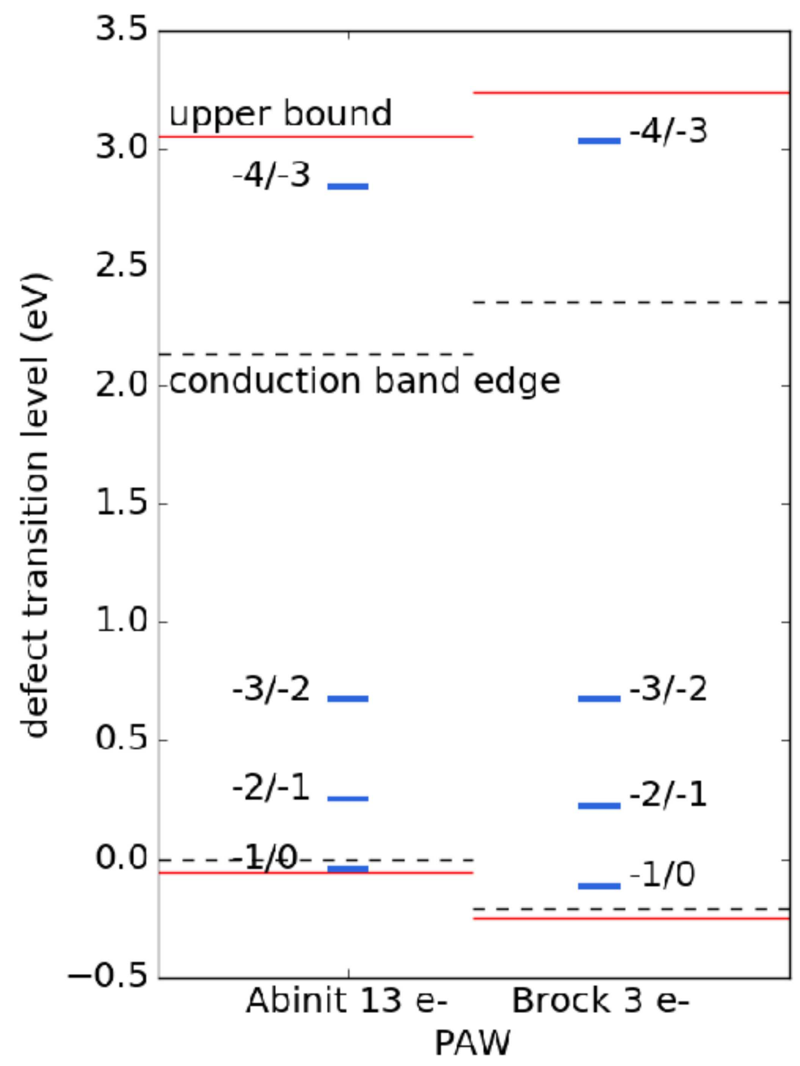
Found optimal PAW

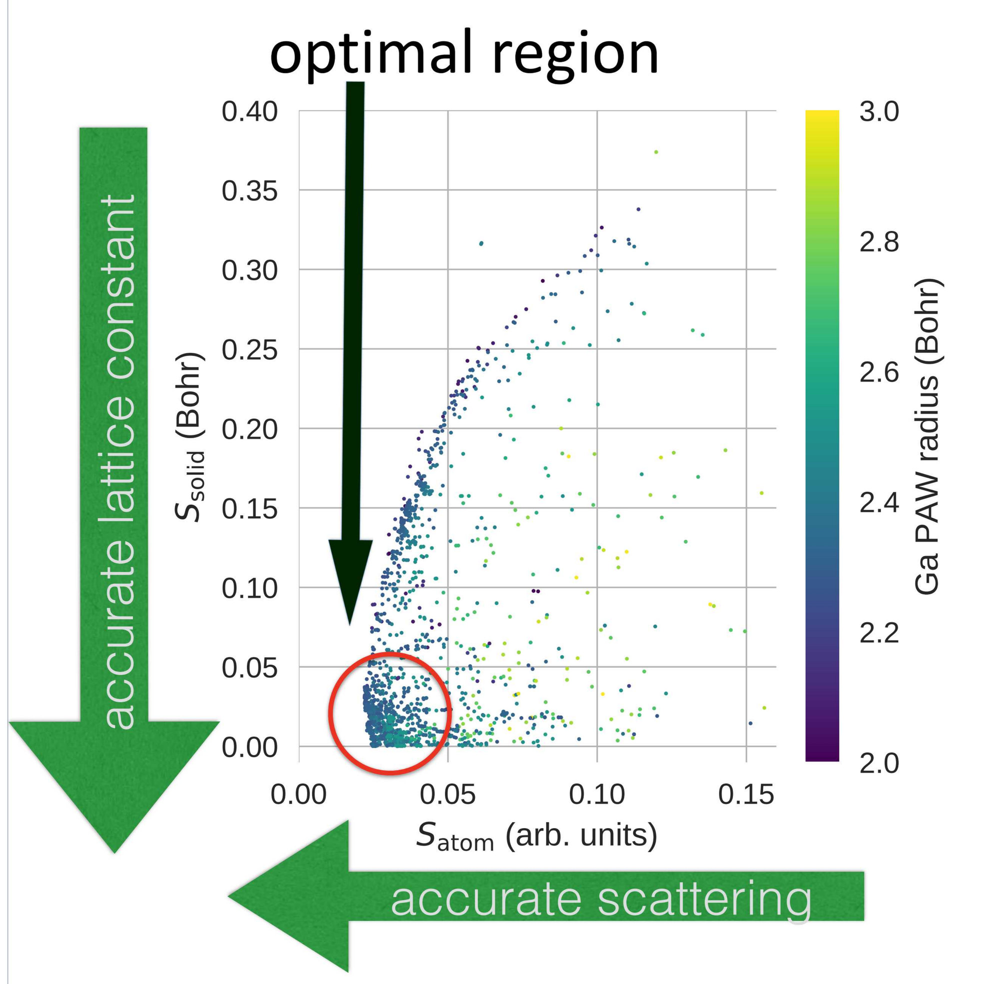
- The parameter sweep found optimal PAWs, which have low scattering errors and low lattice constant errors
- Next, need to make sure the optimal PAW is actually accurate in defect calculations



Test new PAW accuracy in defect calculations

- Compare defect transition levels between new 3-electron PAW and existing 13-electron
- set -3/-2 levels equal for reference
- -2/-1 levels agree within 30 meV, meaning there is good agreement between the PAWs
- Run times in test calculations with new 3-electron PAW reduced about 40% compared to same calculations with 13-electron PAW
- difference in band gap can be partially explained by artificial interaction between Ga 3d and N 2s electrons in the 13-electron case
- **Takeaway:** new 3-electron PAW reduces run times by 40% without sacrificing accuracy of defect calculations





Genetic Algorithm can find optimal PAWs faster

- Searched same parameter space as the parameter sweep, but using a genetic algorithm (GA) implemented in the DAKOTA software instead of a parameter sweep
- the GA found more optimal points, while reducing the number of iterations (computational expense) required by about an order of magnitude

Conclusions

- Generated a 3-electron Ga PAW that reduces computational expense of defect calculations in GaN by about 40%
- A genetic algorithm can improve optimization results over a parameter sweep with an order of magnitude fewer iterations
- This optimization process could be applied to other systems where custom PAWs would improve accuracy or speed of plane-wave DFT calculations

Acknowledgements

Thanks to Normand Modine, Steve Lee, Adam Stephens, and Alan Tackett for helpful discussions, and Natalie Holzwarth for merging our new scattering metric into the Atompaw code.

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists, Office of Science Graduate Student Research (SCGSR) program. The SCGSR program is administered by the Oak Ridge Institute for Science and Education for the DOE under contract number DE-SC0014664.

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