ABACUS LCAO2QO

Hands-on manual

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Introduction

- Quasiatomic Orbital (QO) analysis can project all selected bands as much as possible to basis sets that are atom-centered and manually defined by user, unlike the MLWF (Maximally Localized Wannier Function) that may yield bond-like Wannier functions.
- It takes the idea that QO, which is the projection of manually constructed atom-centered orbitals onto a super-plane (or subspace of the one spanned by original basis) that is spanned by manually selected states and several constructed unoccupied states, should have the maximal norm. Orthonormality is hold during representation transform from original one to QO.

Usage

- In ABACUS INPUT file, for a `calculation scf` run add QO controlling keywords like:

```
qo_switch 1
# To switch on QO analysis
qo_basis hydrogen
# construct hydrogen-like radial function as atom-center orb.
qo_thr 1e-10
# controls real space spreading of orbital constructed
qo_strategy energy-valence
# only generate valence occupied orbitals according to Aufbau principle
#qo_screening_coeff 0.1
# turn on Slater screening to mimic many-electron effect
# comment out to switch off
```

Usage

- In ABACUS KPT file, select to generate a Gamma-centered k-point meshes by Monkhorst-Pack (MP) method, usually dense k-point mesh will reproduce bands better, but certainly will be more expensive:

```
K_POINTS
0
MP
9 9 9 0 0 0
```

Usage

- Run this calculation, on screen or redirected standard output (stdout), after scf converged, you will find output like:

```
---- Quasiatomic Orbital (QO) Analysis Initialization ----
type 0 Si strategy: minimal-valence
Build numerical atomic orbital basis done.
Searching for the cutoff radius for n = 3, l = 0, conv_thr = 1.0000e-10
  Step Nr. Rmax (a.u.)          Norm       Delta Norm
...
        11        5.00  1.0000018602  1.8668178114e-11
...
Build arbitrary atomic orbital basis done.
---- Quasiatomic Orbital (QO) Analysis Initialization Done ----
Calculating overlap integrals for kpoints.
Calculating overlap integrals for kpoints done.
```

Usage

- In OUT.${suffix} (default to be OUT.ABACUS) folder, you will find there are plenty of files:

```
QO_ovlp_0.dat ...
data-H-0 ...
data-S-0 ...
LOWF_K_1.txt ...
QO_supercells.dat
kpoints
istate.info
running_scf.log
```

- Modify the Python script postprocess.py by specifying path to this folder and other parameters you like, then simply run the Python script.

Result: Si fcc crystal

- Several sets of orbitals are tested, relevant parameters are: number of k-points, `qo_basis`, `qo_strategy`, `qo_screening_coeff`

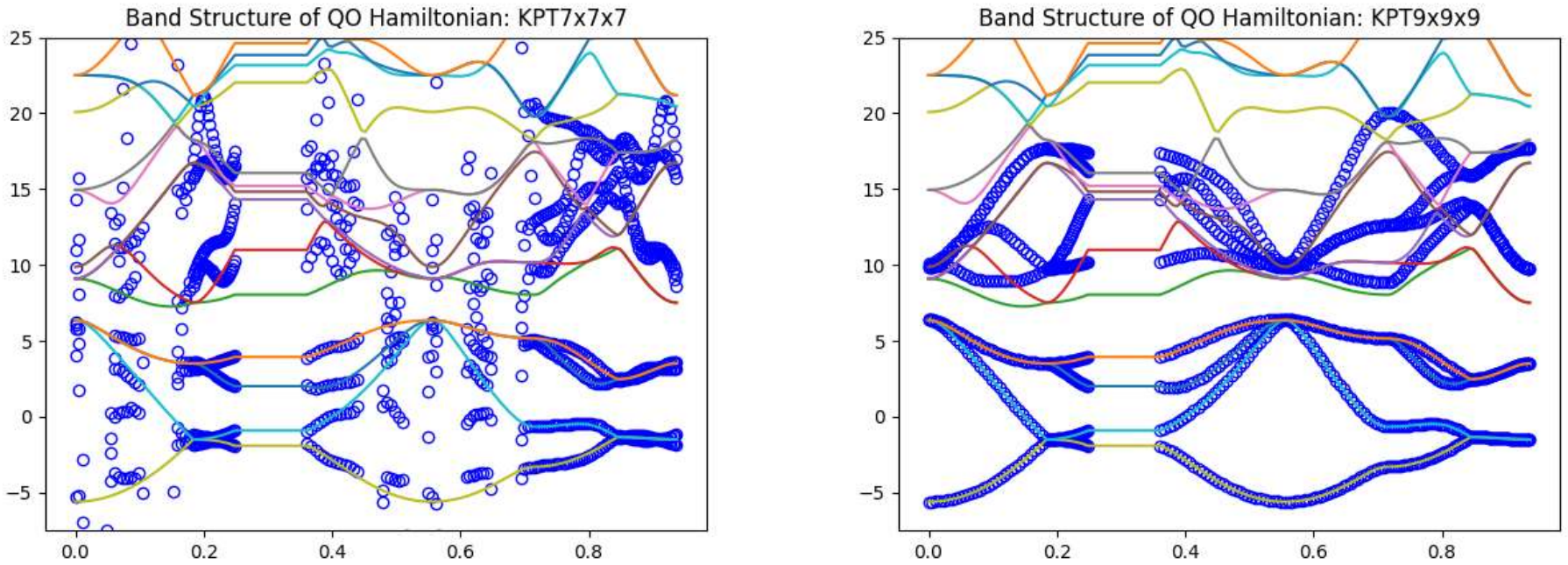

Figure 1. Band structures extrapolated from QO constructed from ABACUS numerical atomic orbital single zeta. Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- The numerical stability for k-point extrapolated H(k) and S(k) in representation of QO is subtle, present acts worse than SZ (single zeta)

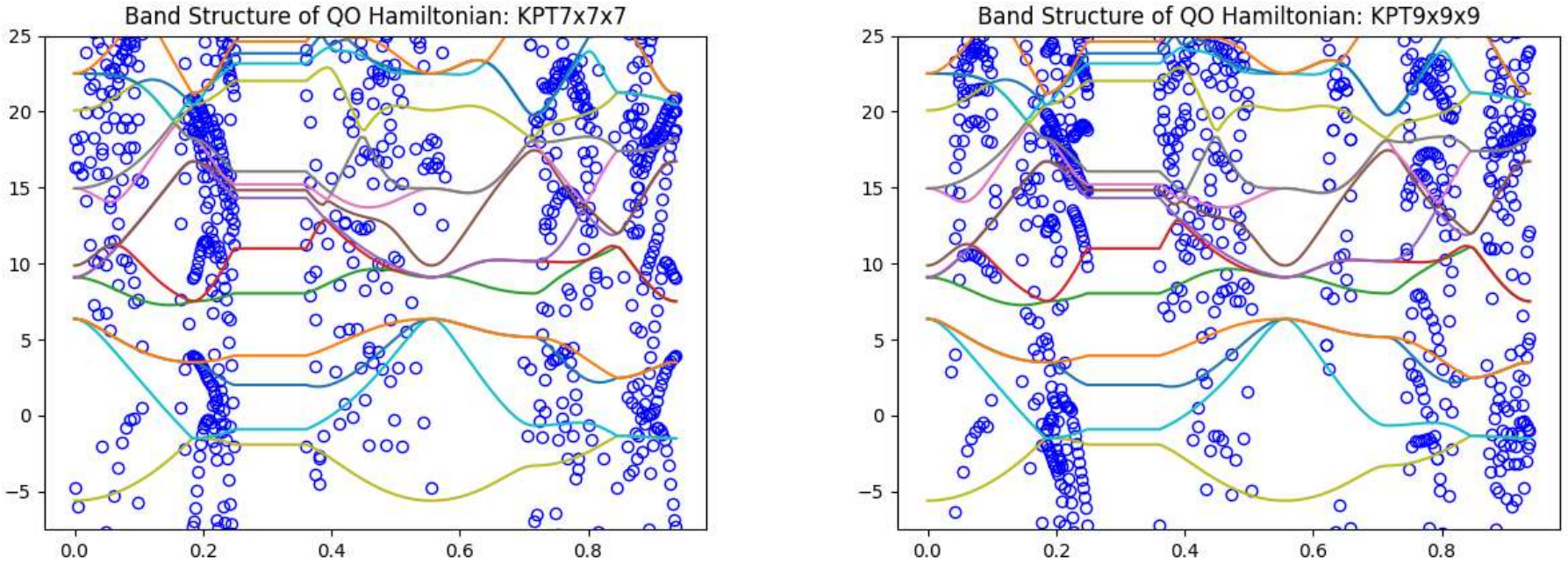

Figure 2. Band structures extrapolated from QO constructed from `qo_basis hydrogen`, `qo_strategy energy-full` (for Si, 1s, 2s, 2p, 3s, 3p). Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- Slater screening appears to be less helpful for `energy-full` way construct hydrogen-like atom-center orbitals

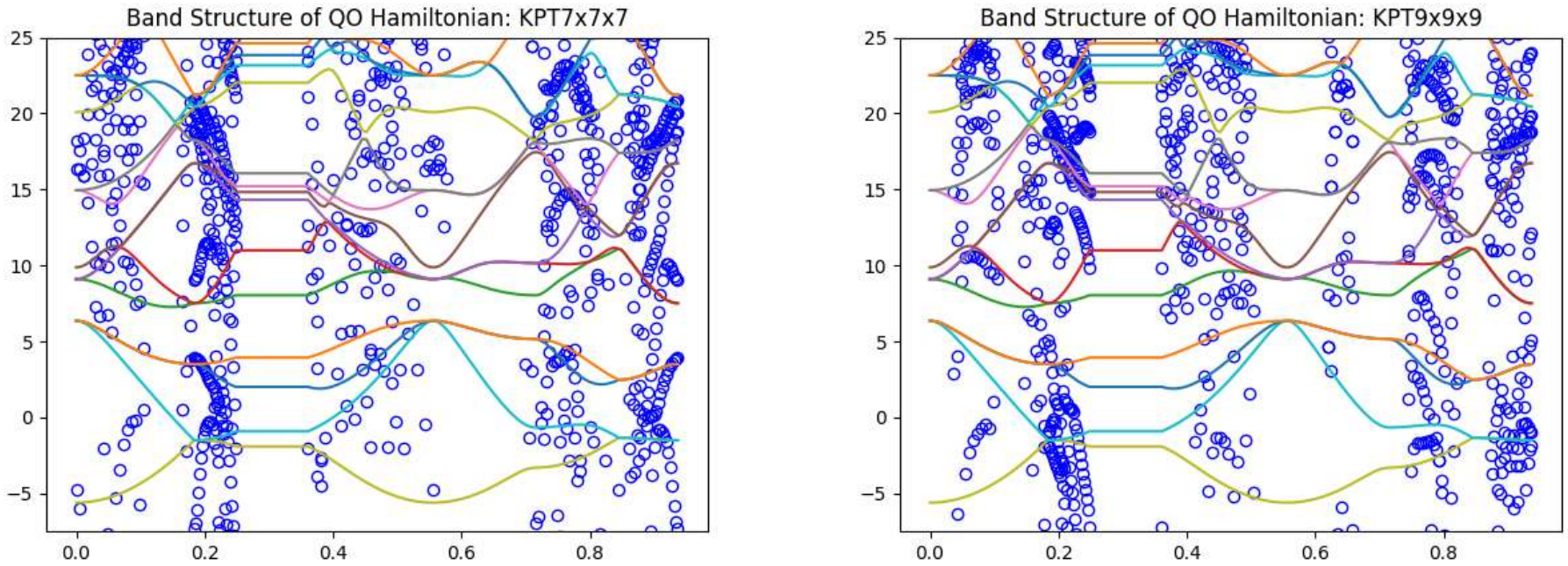

Figure 3. Band structures extrapolated from QO constructed from `qo_basis hydrogen`, `qo_strategy energy-full` with Slater screening. Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- However, if avoid to include non-valence orbitals, the band-reproduction efficiency is much better, which indicates those orbital make diagonalization ill-conditioned.

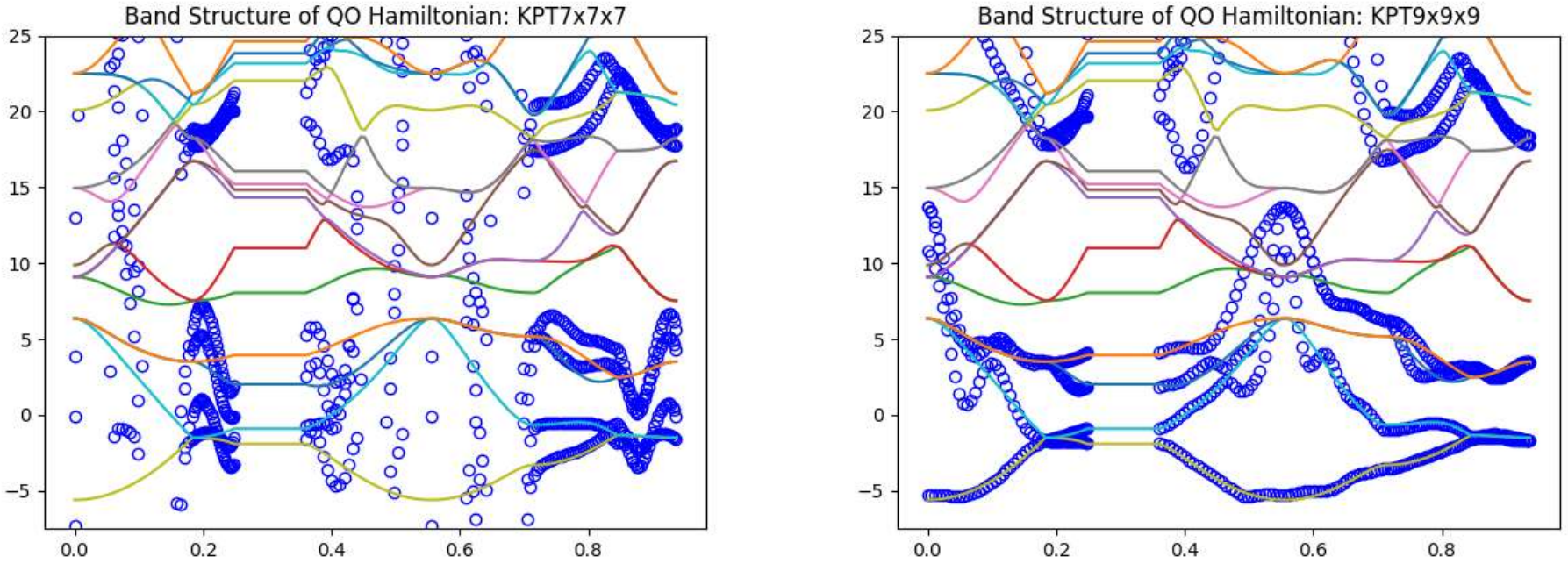

Figure 4. Band structures extrapolated from QO constructed from `qo_basis hydrogen`, `qo_strategy energy-valence` (for Si, 3s, 3p). Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- Together with Slater screening, k-point MP mesh 7*7*7 can get result roughly as “good” as 9*9*9.

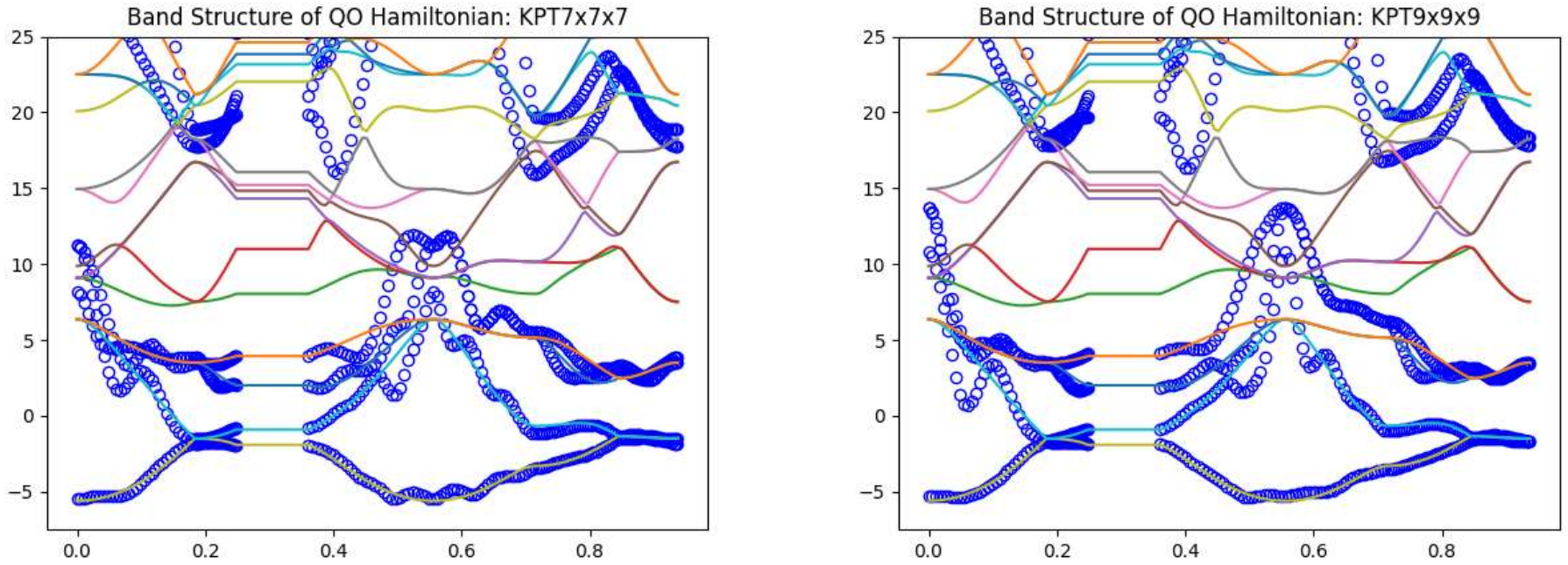

Figure 5. Band structures extrapolated from QO constructed from `qo_basis hydrogen`, `qo_strategy energy-valence` with Slater screening. Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- `qo_strategy minimal-nodeless` generate hydrogen-like orbitals according to maximal principle quantum number. Extra inclusion of d orbital did not make result better.

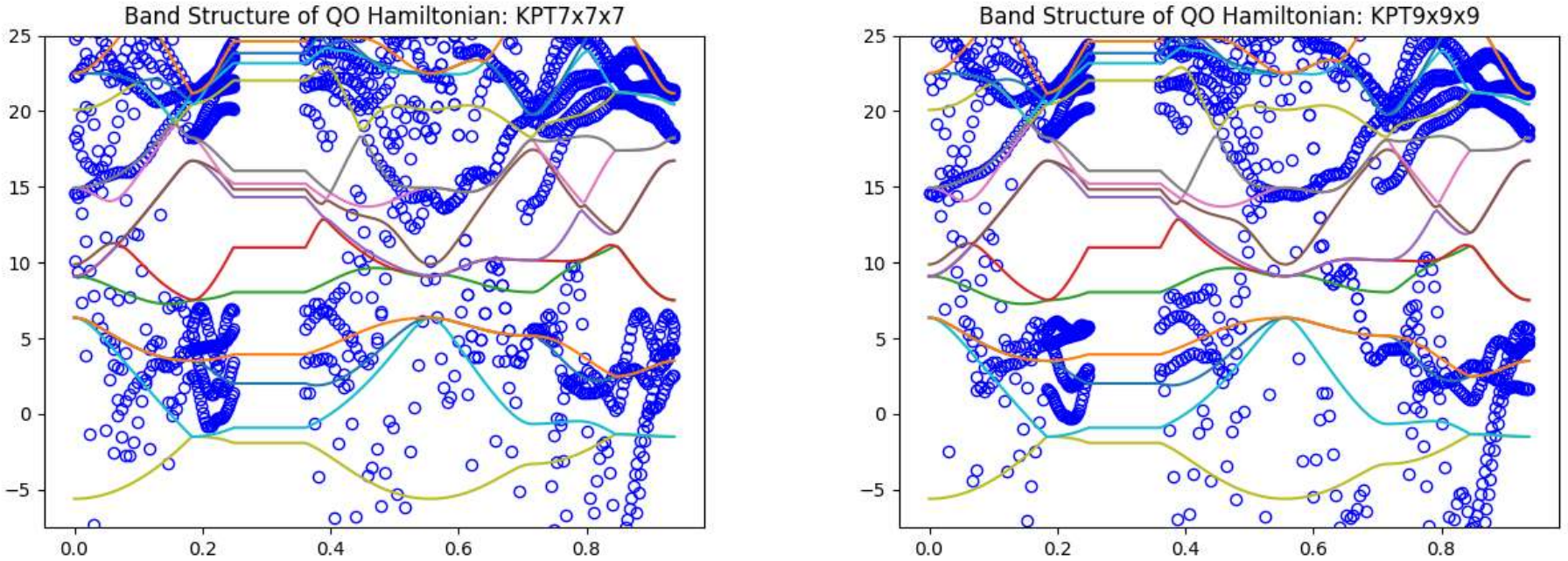

Figure 6. Band structures extrapolated from QO constructed from `qo_basis hydrogen`, `qo_strategy minimal-nodeless` (for Si, 1s, 2p, 3d). Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

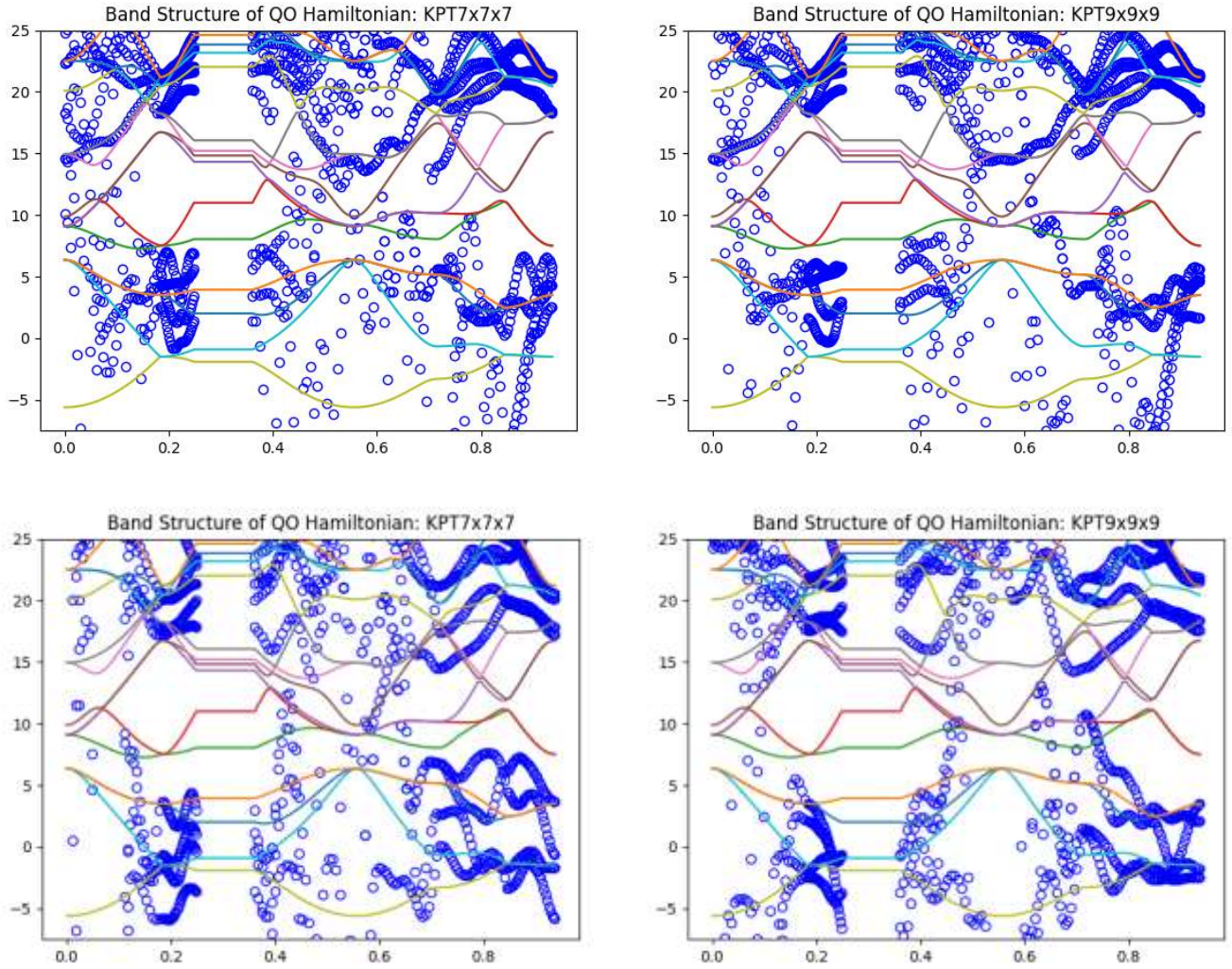

Figure 7. Comparison on the efficiency of band structure extrapolation ability between orbitals constructed by `minimal-nodeless` (upper two) and `minimal-valence` (lower two). For Si, `minimal-nodeless` strategy will generate Si 1s, 2p and 3d hydrogen-like orbitals, `minimal-valence` will generate 3s, 3p and 3d.
Although not significant, but still worthwhile to point out that the “valence”-like hydrogen-like orbital has better ability of expression of band structure.
Blue circle: QO, colored line: full LCAO calculation.

Result: Si fcc crystal

- At last change `qo_basis` to `pswfc` to use pseudowavefunction in pseudopotential as atom-centered orbital. An adjustable parameter `qo_screening_coeff`, which controls the magnitude of exponential decay is tested (`qo_strategy` set to `all` by default).

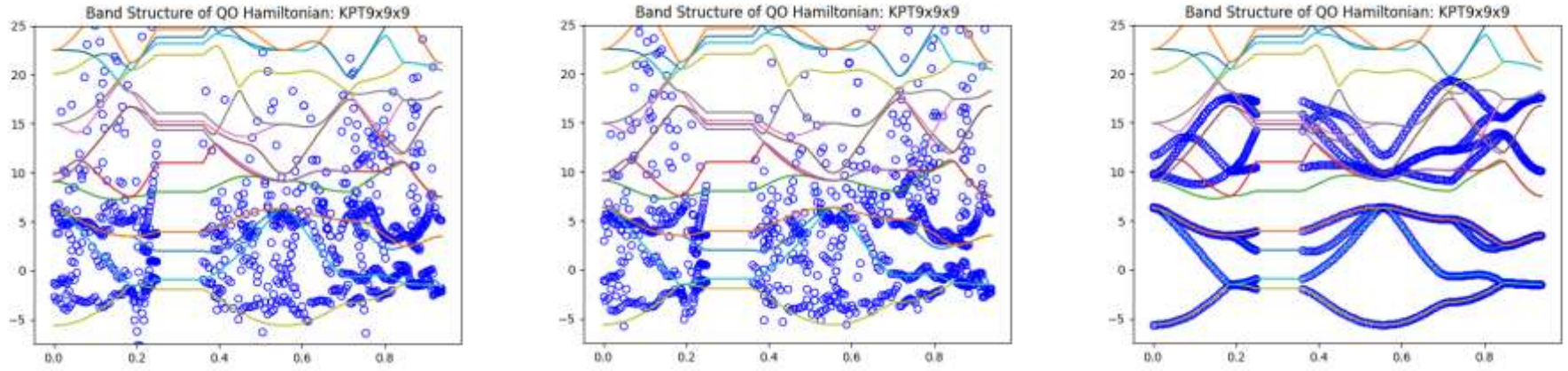

Figure 8. Band structures extrapolated from QO constructed from `qo_basis pswfc`, `qo_screening_coeff` varies from 0.05, 0.10 to 0.50 from left to right. Blue circle: QO, colored line: full LCAO calculation.

Figure 9. Comparison on band structure in energy range (5 – 15 eV) between the one predicted by SZ (left) and pseudowavefunction with `qo_screening_coeff` 0.50.

Result: Si fcc crystal

- Comparison between SZ and pseudowavefunction is also interesting in K-point convergence aspect. For SZ, 9*9*9 can reproduce band structure but 7*7*7 cannot. For pseudowavefunction, 7*7*7 also works well. For 5*5*5, `qo_screening_coeff 0.5` does not work at all but work roughly for `1.0`, which corresponds to the case orbitals are more localized.

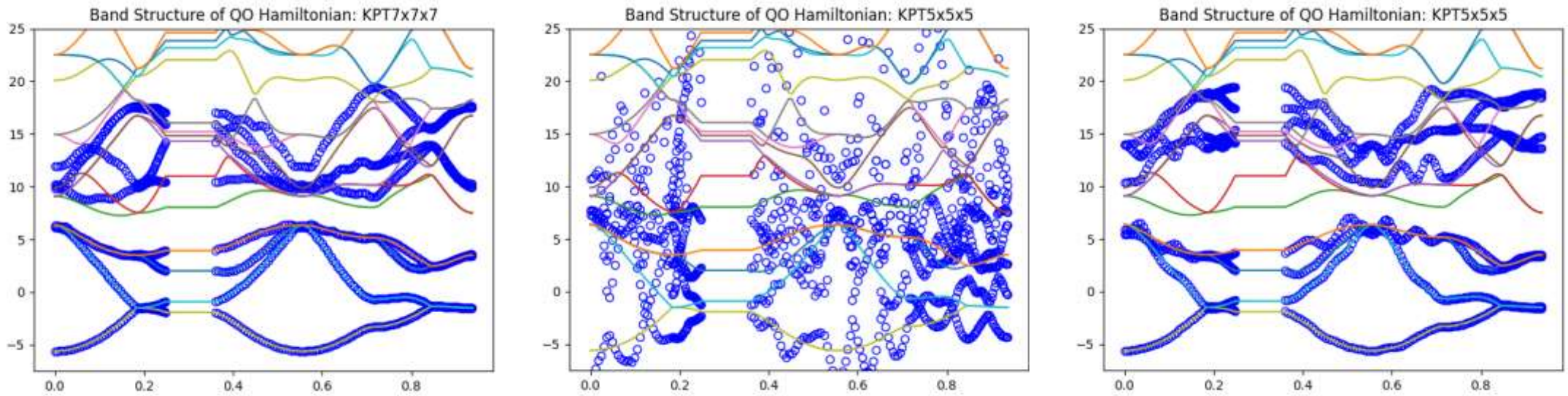

Figure 10. Localization-k correlated convergence test on band structure QO constructed from `qo_basis pswfc`, `qo_screening_coeff` varies from 0.5 (left and middle) to 1 (right).Blue circle: QO, colored line: full LCAO calculation.

Conclusion

- Several methods to construct atom-centered orbitals are supported, for Si fcc crystal system the `pswfc` method works approximately as good as SZ (Single Zeta of numerical atomic orbital).
- `qo_screening_coeff` controls the behavior of `pswfc` method, a higher value may support a k-point mesh not as dense as SZ, but drawback is system-dependent and need user to test as reported in original QO paper.
- Selection of orbital is critical, Slater screening correction can improve the performance of valence hydrogen-like orbital.

For more features & Bug report

ABACUS develop team encourages user to submit issue on Github:

https://github.com/deepmodeling/abacus-develop/issues

Acknowledgement

ABACUS LCAO2QO module uses two-center-integrator module refactored by @jinzx10, who also helps in code debugging. @mohanchen, @dyzheng and @WHUzhouweiqing provide suggestions on code review and technical aspects. @QG-phys provides Si fcc crystal example for test.

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