NECI Tutorial

April 2023

1 Introduction

Here we will discuss the practical aspects of running FCIQMC calculations using the NECI software. For a more complete introduction and overview of the software, please see the documentation:
`www2.fkf.mpg.de/alavi/neci/stable/`[1]

The goal of this tutorial is to provide a practical introduction to the software. We will calculate the energy of the nitrogen dimer in the cc-pVDZ basis using *i*-FCIQMC, and do the same but with transcorrelated integrals calculated by the TCHInt software with the XTC approximation, thereby demonstrating NECI's robustness to non-Hermiticity.

2 Required files

To run NECI, all you need is an FCIDUMP (a standard means by which to store necessary 1- and 2-body integrals) and an input file. Of course, you also need the binary for NECI, which you can find here:
`/home/ws-p.haupt/workshop/neci/build/bin/neci`

For this tutorial, you can find the input files in
`/home/ws-p.haupt/workshop/run/n2_notc_demo`

This contains three files: the NECI input, the FCIDUMP, and a submit script for the SLURM scheduler.

3 Anatomy of a NECI Input File

Let's take a look at the input file, `neci.inp`:

```
title
    # comments may be added like this
    # and line breaks with the \ character

    system read
        electrons 10
```

[1] At the time of writing, the public site is somewhat outdated and may be difficult to use with this tutorial.

```
        spin-restrict 0
        nobrillouintheorem
        nonuniformrandexcits pchb
        molpromimic
    endsys

    calc
        methods
            method vertex fcimc
        endmethods
        seed -7
        totalwalkers 1.e5
        nmcyc 1.e4
        definedet 1-10

        startsinglepart 1000

        tau-values \
            start user-defined 0.001

        tau-search \
            algorithm conventional \
            maxWalkerBloom 1

        truncinitiator
        addtoinitiator 3

        semi-stochastic 100
        allrealcoeff
        pops-core 1000

        realspawncutoff 1.0

        time 2800
    endcalc
end
```

All these keywords (and plenty more) are described in detail on the website. They are all case-insensitive. An overview:

- Input must begin with the `title` keyword, and end with `end` (i.e. wrap your input in this block). This is mandatory.
- Next, we have the `system` block, which is also mandatory.
  - The `system` keyword has a mandatory argument which comes directly after it on the same line, which in this case is `read`, indicating we are reading the system integrals from an `FCIDUMP` file.

  - The number of electrons (14, minus 4 core electrons) is specified with `electrons`, and the spin-projection is specified with `spin-restrict`.
  - The excitation generator used in this case is `pchb` or the precomputed heat bath. This just makes the calculation faster, but should not affect the result.
  - In this case, the FCIDUMP was produced with Molpro; to accommodate its formatting of the integrals, we use the `molpromimic` keyword.

- Next, we have the `calc` block, which is also mandatory, necessarily terminated with `endcalc`. This block in particular has many options and potential keywords; here we use only a small subset.
  - The `methods` subblock specifies which algorithm we are using. Here we are using FCIQMC.
  - We set the number of total walkers, the number of cycles for which to run the calculation and the determinant on which to start, as well as the number of walkers on this determinant.
  - We define the time step, `tau`, and the search algorithm for finding the optimal time step.
  - We specify to use *i*-FCIQMC with the `truncinitiator` keyword, with a population of 3 walkers required to be promoted to initiator.
  - We use a "semistochastic" space, which means part of the space is run deterministically (this reduces the stochastic noise in the calculation).

- There are also other optional blocks here, such as `integral` which controls properties of the integrals (e.g. freezing) and `logging` which controls output.

4 Running NECI and Analysing Results

To submit your job to the cluster, just run `sbatch submit_neci`. You can also run it locally, directly with the binary, but make sure to load OpenMPI and specify the number of MPI processes with the `-np` flag.

Data is output to a file in your working directory called `FCIMCStats`. This contains several columns useful for checking the results, and is output in a format convenient to most plotting software, such as gnuplot.

Particularly important columns are:

- Column 1, the iteration count. This is just the number of cycles for which the calculation has run; often used as the x-axis in plots.
- Column 23, the total projected energy.
- Columns 2 and 10, the current shift energy and the averaged shift energy. Useful for checking agreement with the projected energy.
- Column 9, the projected energy.

- Column 5, the total number of walkers. Useful for checking the dynamics of the calculation.
- Column 12, the number of walkers on the reference determinant. Useful for checking stability (e.g. reference changes) and multi-reference character.

An estimate for the final projected and shift energies are also printed at the end of the standard output of the calculation.

5 Additional Remarks

In order to properly analyse the results, it is important to perform a blocking analysis. This is included in the NECI source code, here:
`/home/ws-p.haupt/workshop/neci/utils/blocking.py`

However, for this calculation it is not necessary, as the final printed energy from NECI is a good estimate.

In order to make sure we have sampled the Hilbert space sufficiently, we should also run several calculations at different walker numbers and ensure the energy converges with the number of walkers. This was not done here out of the interest of time.

6 Transcorrelated Calculation

Input for the same system, except all electron and with integrals calculated with the TCHInt library under the XTC approximation is available here:
`/home/ws-p.haupt/workshop/run/n2_xtc_demo`

The most important additional information here is in the `system` block: `nonhermitian 2-body`, which tells NECI that the 2-body integrals are not Hermitian, and so treats H_{ij} and H_{ji} as distinct. Note in the transcorrelated method the 1-body integrals are still Hermitian.

If you run this calculation, you will see the energy estimate to be much better at about twice the cost of the non-TC calculation, as the determinant graph is now directed (i.e. spawning from D_i to D_j is different from spawning from D_j to D_i).

7 Bonus: Formaldehyde

Time permitting, you may be interested in running calculations for the ground and first excited states for the formaldehyde (COH_2) molecule. The input files are available here (aug-cc-pVDZ basis):
`/home/ws-p.haupt/workshop/run/coh2/excited_state`
`/home/ws-p.haupt/workshop/run/coh2/excited_state`