Simulations of state-of-the-art fermionic neural network wave functions with diffusion Monte Carlo

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Recently developed neural network-based *ab-initio* solutions (Pfau et. al arxiv:1909.02487v2) for finding ground states of fermionic systems can generate state-of-the-art results on a broad class of systems. In this work, we improve the results for this Ansatz with Diffusion Monte Carlo. Additionally, we introduce several modifications to the network (Fermi Net) and optimization method (Kronecker Factored Approximate Curvature) that reduce the number of required resources while maintaining or improving the modelling performance. In terms of the model, we remove redundant computations and alter the way data is handled in the permutation equivariant function. The Diffusion Monte Carlo results exceed or match state-of-the-art performance for all systems investigated: atomic systems Be-Ne, and the carbon cation C^+.

Outline

The problem

- What are we trying to do?
- Why would we want to do that?

Background

- What has been done before?
- What is related now?

The algorithms

- What algorithms were used to solve the problem?

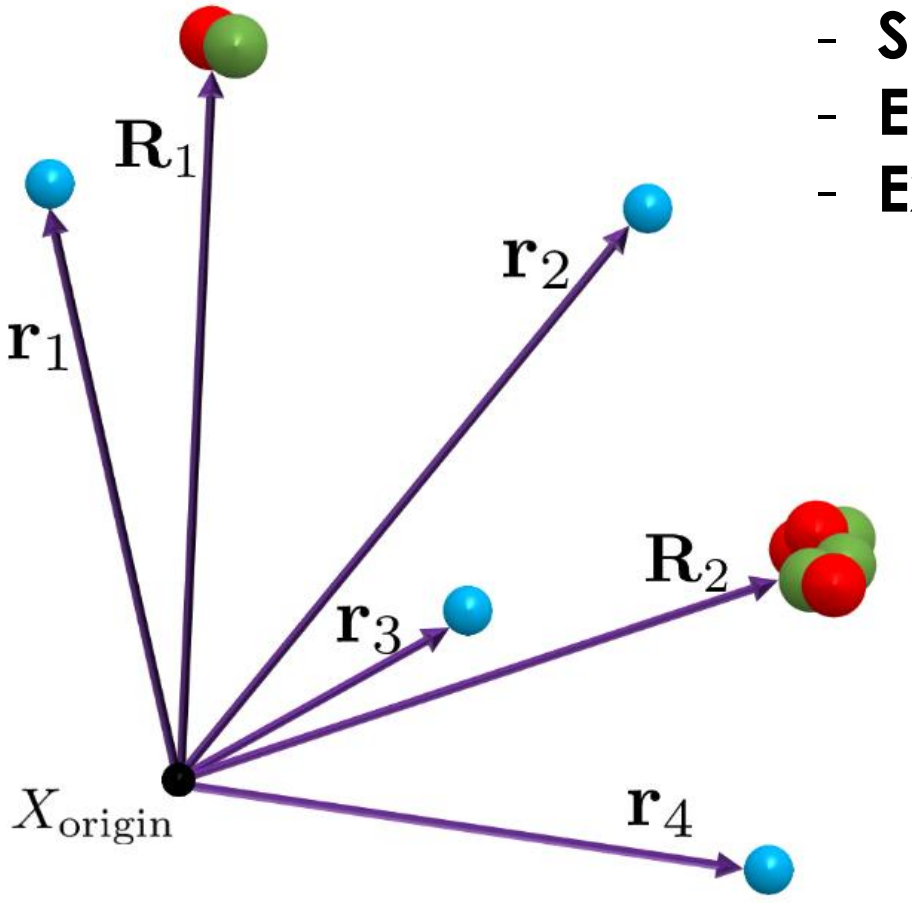
Some results

- What results did we get?

Next steps

- Where is this going?

The problem



Quantum particles/systems

- **Superposition** (waves)
- **Entanglement** (non-classical correlations)
- **Exponential** amount of information to model

$$\hat{H}\psi(X) = E\psi(X)$$

Related work

Solving the QMB with AANs
Science 10 2017

- Spin systems
- Boltzmann machines
- First demonstration

SchNet
J. Chem. Phys. 148 2019

- Atomic/Molecular properties predictions
- Graph NNs

Fermi Net
Nature Chemistry12 2020

- Atomic/molecular wave functions
- Multi-electron orbitals
- Convolutional layers
- Expensive, but very accurate

PauliNet
Nature Chemistry12 2020

- Atomic/molecular wave functions
- Slater-Jastrow-Backflow wave function
- Graph NNs
- Fast, but less accurate

Related work

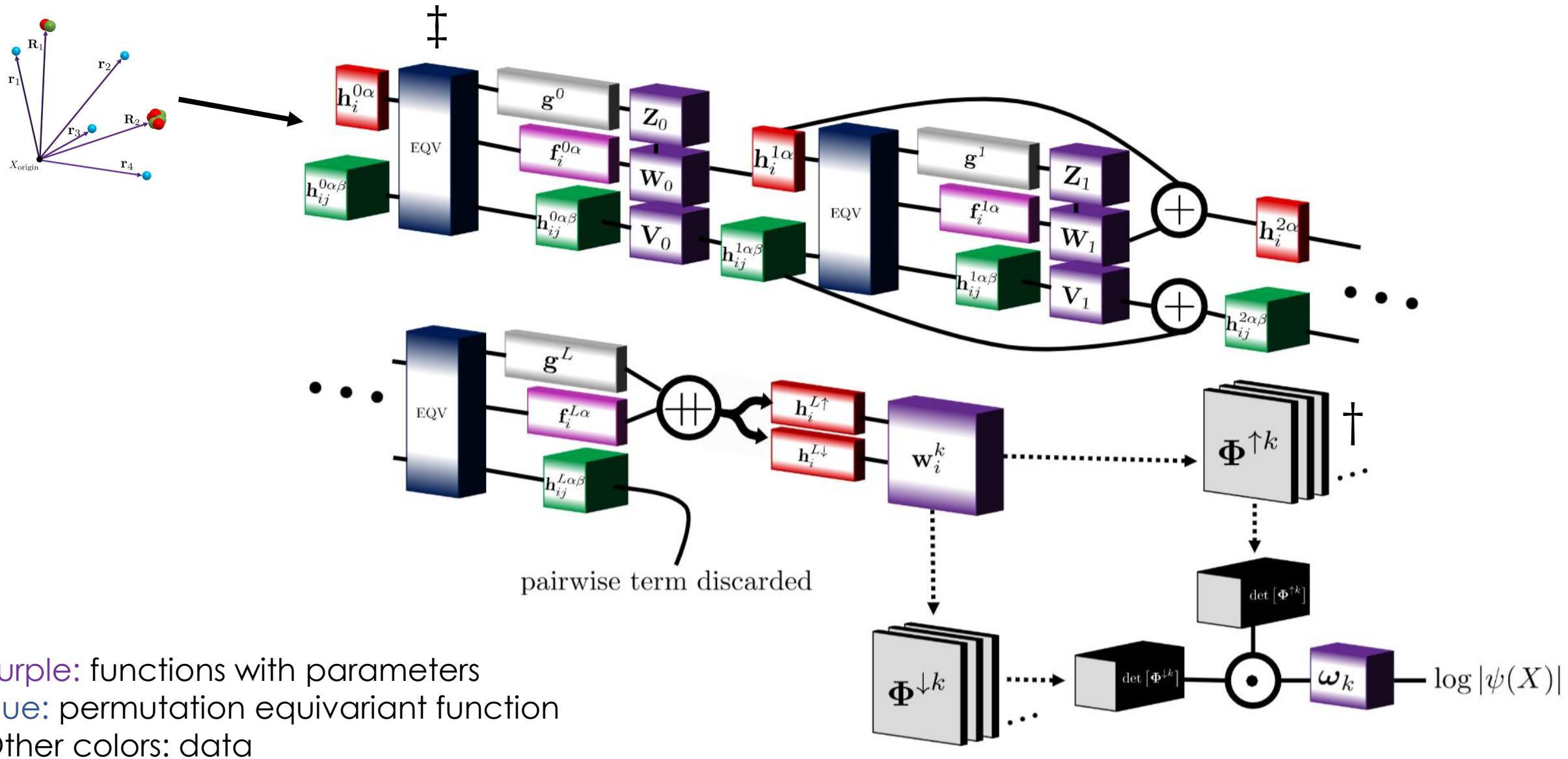
Contributions of this work

- **Improvements in efficiency** to Fermi Net
- **First (known to us) application of DMC** to neural network Ansatz
- **Matching or exceeding state-of-the-art results** on atomic systems Be-Ne and ion C^+

Other differences

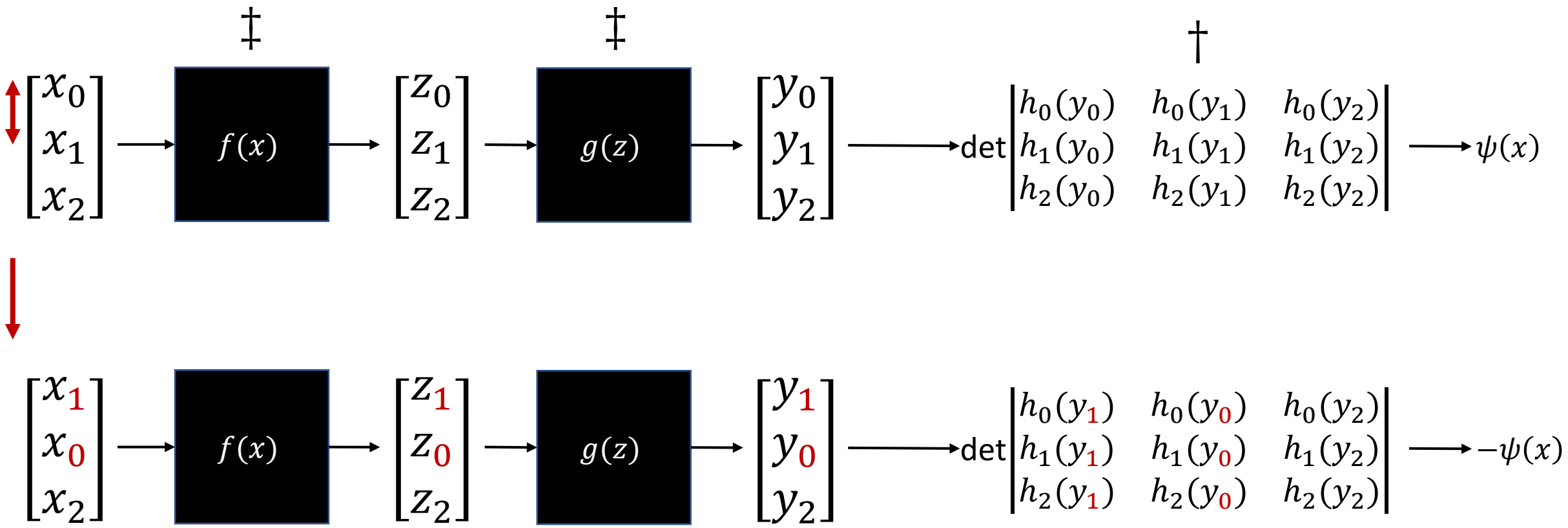
- Changes to the KFAC optimisation

The model



Purple: functions with parameters
Blue: permutation equivariant function
Other colors: data

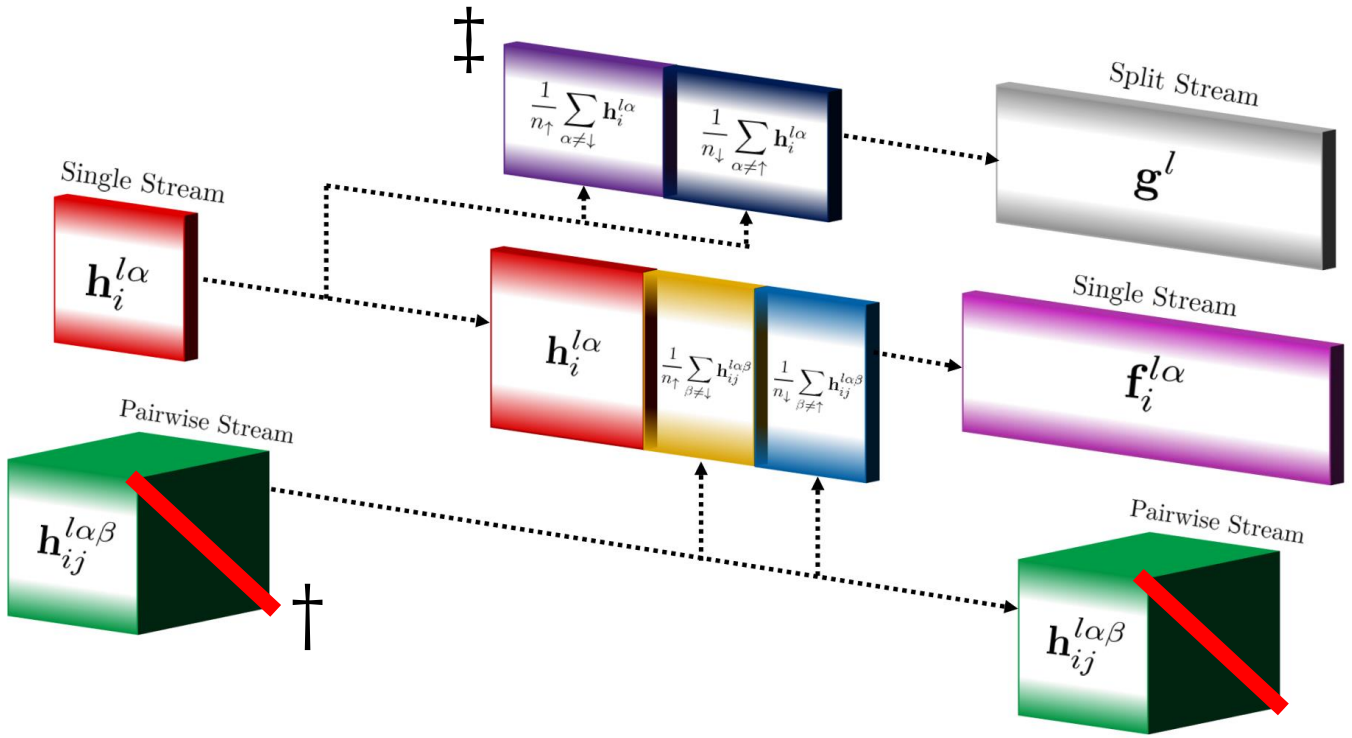
The model



Swapping inputs results in change of sign of the wave function = antisymmetric = fermionic system

- Pfaffian wave function https://pubmed.ncbi.nlm.nih.gov/16711968/
- Vandermond wave function https://ieeexplore.ieee.org/document/9297114/
- …

The model:
Anti-symmetry & the permutation equivariant function



† Remove redundant diagonal elements $i = j$

‡ Split data which is the same in all indices

The algorithms

Variational Monte Carlo
VMC

Iterative improvement of the wave function Ansatz by changing the parameters performed under the Born-Oppenheimer approximation (fixed nuclei)

Kronecker Factored Approximate Curvature
KFAC

Approximate second order optimization related to stochastic configuration

Diffusion Monte Carlo
DMC

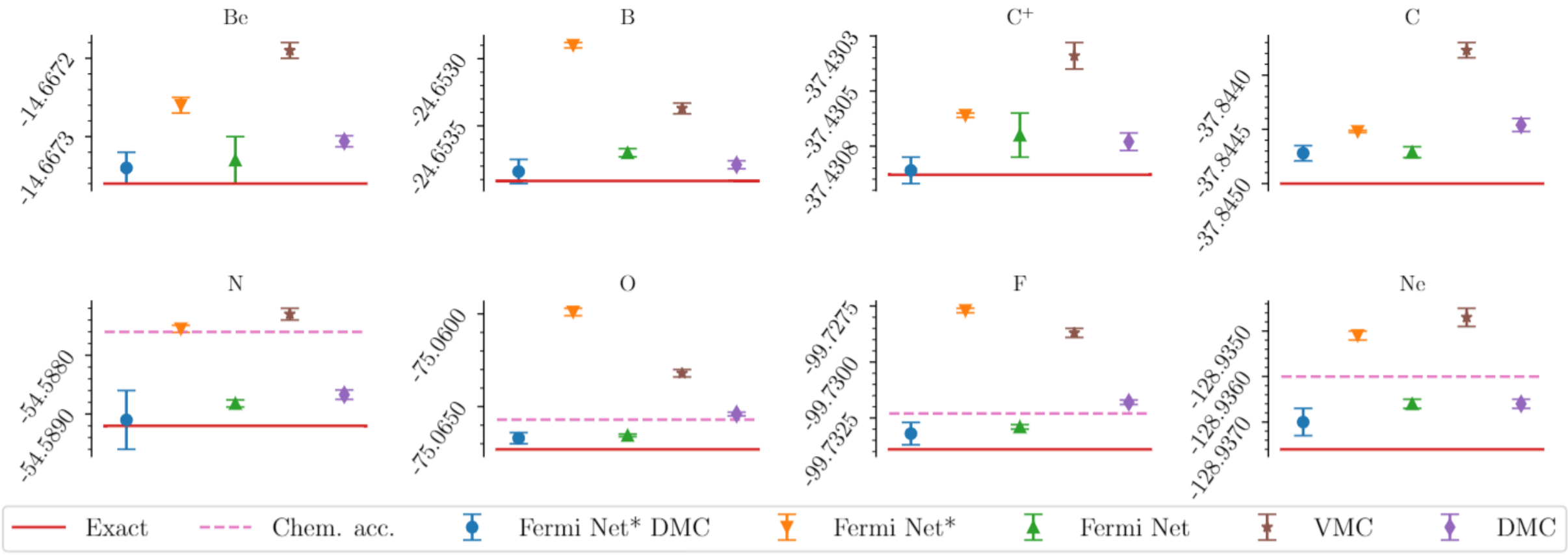
Projector method iteratively improving the wave function by reweighting the walkers performed under the fixed-node approximation

Approximate stochastic configuration

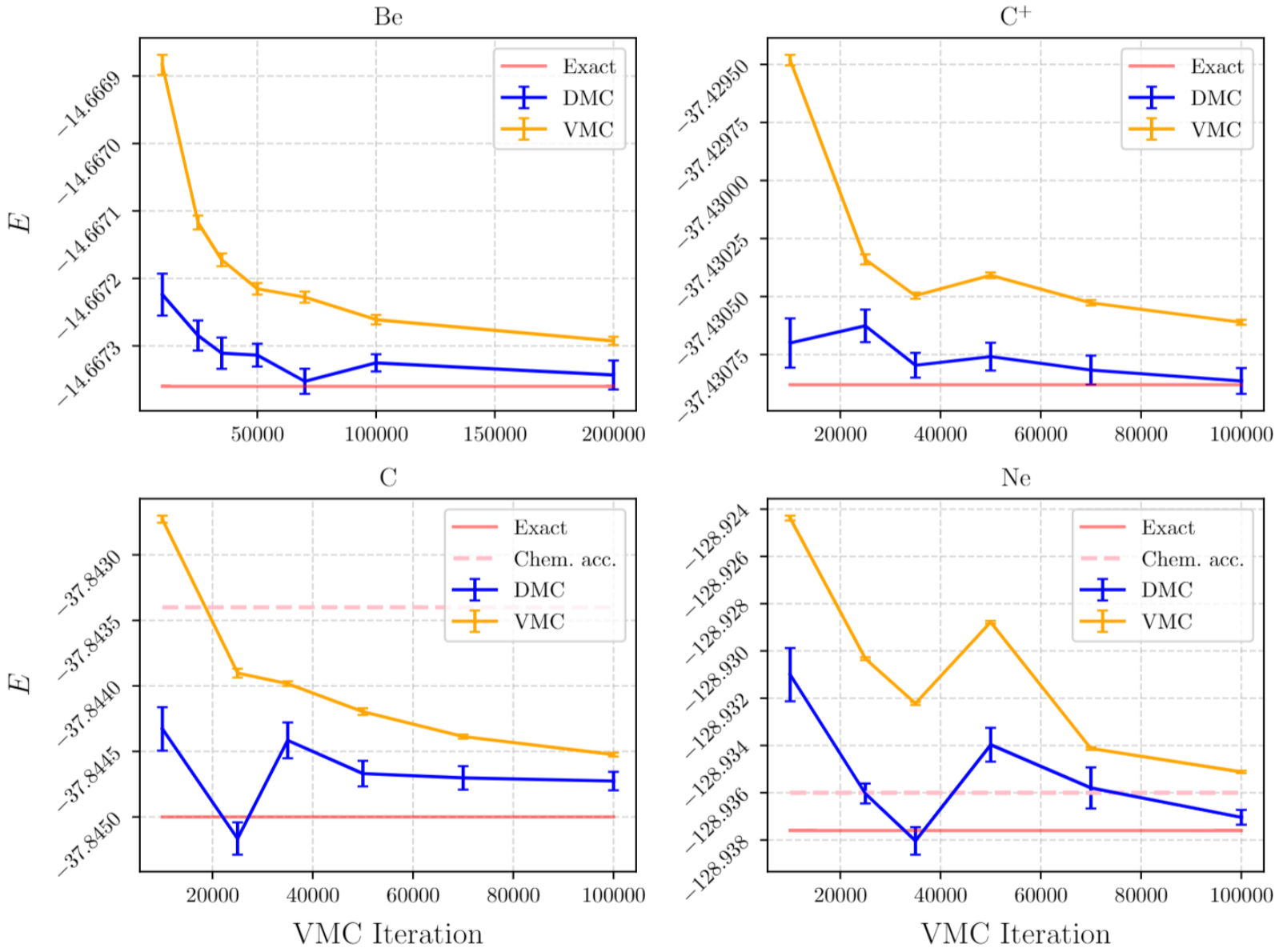
Some results

Atom	Fermi Net* + DMC (This work)	Fermi Net* (This work)	Fermi Net [7]	VMC [31]	DMC [31]	HF	Exact [32]
Be	-14.66734(2)	-14.66726(1)	-14.66733(3)	-14.66719(1)	-14.667306(7)	-14.35188	-14.66736
B	-24.65384(9)	-24.65290(2)	-24.65370(3)	-24.65337(4)	-24.65379(3)	-24.14899	-24.65391
C^+	-37.43086(6)	-37.43061(1)	-37.4307(1)†	-37.43034(6)	-37.43073(4)	-36.87037	-37.43088
C	-37.84472(7)	-37.84452(1)	-37.84471(5)	-37.84377(7)	-37.84446(6)	-37.08959	-37.8450
N	-54.5891(5)	-54.58755(6)	-54.58882(6)	-54.5873(1)	-54.58867(8)	-53.5545	-54.5892
O	-75.0667(3)	-75.0599(2)	-75.06655(7)	-75.0632(2)	-75.0654(1)	-73.6618	-75.0673
F	-99.7332(5)	-99.7277(1)	-99.7329(1)	-99.7287(2)	-99.7318(1)	-97.9865	-99.7339
Ne	-128.9370(3)	-128.9351(1)	-128.9366(1)	-128.9347(2)	-128.9366(1)	-126.6045	-128.9376

Some results



Some results



Next steps

Improved Ansatz design

- Can other changes be made to Fermi Net?
- Other wave functions?

Improved VMC

- CG SR?
- KFAC features?
- Mixed precision training?

Other systems

- Solids?
- Where else is VMC used?

Other VMC methods

- Pseudopotentials?

Scaling

- Most efficient way to model large systems?
- What are the technical challenges of scaling?

Final Comments

Long term vision

- Infrastructure (GPUs and deep learning frameworks) will continue to improve
- Quantum chemistry neural network methods will adopt other standard quantum chemistry methods
- Neural network methods will supplement or replace standard quantum chemistry methods