

Exploring phase diagrams of quantum matter with neural quantum states

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Equilibrium (Zero temperature $T=0$): Variational Monte Carlo Simulations

- Many-body quantum system:
Hamiltonian

$$H = \sum_{i=1}^N \left(-\frac{\hbar^2 \nabla_i^2}{2m} + u(\mathbf{r}_i) \right) + \sum_{i < j} v(|\mathbf{r}_i - \mathbf{r}_j|)$$

Extended Systems:

- liquid/solid Helium
- electron gas
- liquid/solid hydrogen

(Finite systems:)

- Atoms
- Molecules

- Variational principle for ground state energy

$$E_0 \leq \frac{\int d\mathbf{R} \Psi_T(\mathbf{R}) H \Psi_T(\mathbf{R})}{\int d\mathbf{R} \Psi_T^2(\mathbf{R})}$$

$$\mathbf{R} = (\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)$$

Trial wave function $\Psi_T(\mathbf{R})$
contains **variational parameters**
to be optimized

$$E_0/N = \epsilon_0 + \Delta/N^\delta + \dots$$

Trial wave functions: from mean-field to pair product wave functions

- Mean field (**single particle**) wave functions:

$$\Psi_{mf} = \prod_i \phi_1(\mathbf{r}_i) \equiv \exp \left[- \sum_i u_1(\mathbf{r}_i) \right]$$

Fermions: Slater-determinant $D(\mathbf{R}) = \det_{\mathbf{k}i} \phi_{\mathbf{k}}(\mathbf{r}_i) \rightarrow$ **Hartree-Fock**

- Trial wave function based on **pair correlations**:

$$\Psi_J = \prod_{i < j} \phi_2(\mathbf{r}_i - \mathbf{r}_j) \equiv \exp \left[- \sum_{i < j} u_2(\mathbf{r}_i - \mathbf{r}_j) \right]$$

“*Jastrow*” ansatz Phys. Rev. 98, 1479 (1955), [A. Bijl, Physica 7, 869 \(1940\)](#)...

Standard form for variational Monte-Carlo [McMillan, Phys. Rev. 175, 266 \(1968\)](#)

Fermions: “**Slater-Jastrow**” $\Psi_{SJ} = D(\mathbf{R}) \exp[-U(\mathbf{R})]$ [Ceperley, Chester, Kalos, Phys. Rev. B 16, 3081 \(1977\)](#)

Guidelines for Improving trial wave functions (I):

Density
operator:

$$\rho(\mathbf{r}) = \sum_i \delta(\mathbf{r}_i - \mathbf{r})$$

Re-write the trial wave function
as an explicit functional of the density operator

$$U[\rho(\mathbf{r})] = -\log \Psi_T[\rho(\mathbf{r})]$$

$$U[\rho(\mathbf{r})] = \int d\mathbf{r} u_1(\mathbf{r}; n) \rho(\mathbf{r}) + \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' \rho(\mathbf{r}) u_2(\mathbf{r} - \mathbf{r}'; n) \rho(\mathbf{r}') + \dots$$

Single-particle (mean-field)

Two-particle correlations (Jastrow)

Potentials u_1 and u_2 contain variational parameters,
optimal values are functional of the mean density:

$$n(\mathbf{r}) = \langle \rho(\mathbf{r}) \rangle$$

Three-body correlations? $U_3 = \int d\mathbf{r} d\mathbf{r}' d\mathbf{r}'' u_3(\mathbf{r} - \mathbf{r}', \mathbf{r} - \mathbf{r}''; n) \rho(\mathbf{r}) \rho(\mathbf{r}') \rho(\mathbf{r}'')$

Gradient terms? $U_g = \int d\mathbf{r} d\mathbf{r}' [\nabla \rho(\mathbf{r})] u_g(\mathbf{r} - \mathbf{r}'; n) [\nabla \rho(\mathbf{r}')]$



more insight needed!

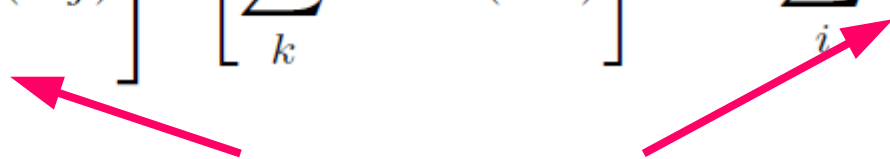
How to reduce systematic bias due to the variational ansatz?

Systematic improvement using higher order ($n > 3$) many-body correlations
 $\Rightarrow$ how to calculate efficiently ????

Look at 3body term

$$U_3[\mathbf{R}] = \sum_i \sum_{j,k} \mathbf{r}_{ij} \cdot \mathbf{r}_{ik} u_3(r_{ij}) u_3(r_{ik})$$

rewrite

$$= \sum_i \left[\sum_j \mathbf{r}_{ij} u_3(r_{ij}) \right] \cdot \left[\sum_k \mathbf{r}_{ik} u_3(r_{ik}) \right] = \sum_i \mathbf{q}_i^{(1)} \cdot \mathbf{q}_i^{(1)}$$


using quasi-particle (“backflow”) coordinates

$$\mathbf{q}_i^{(1)}[\mathbf{R}] = \sum_j \mathbf{r}_{ij} u_3(r_{ij})$$

- Explicitly n-body correlation terms:
Quasi-particle (“backflow”) tensors:

$$\underline{\mathbf{T}}_i^{\alpha\beta} = \sum_j \mathbf{r}_{ij}^\alpha \mathbf{r}_{ij}^\beta t(r_{ij}) \quad U_4 = \sum_i \mathbf{q}_i \cdot \underline{\mathbf{T}}_i \cdot \mathbf{q}_i$$

M.H., B.Bernu, D.Ceperley, Phys. Rev. B 74, 104510 (2006)

- Non-linear correlations:
Iteratively backflow-network states:

$$U_2^{(1)}[\mathbf{Q}^{(1)}] = \sum_{i < j} u_2(q_{ij}^{(1)})$$

M. Taddei, M. Ruggeri, S. Moroni, M.H.,
Phys. Rev. B 91, 115106 (2015)

Trial wave functions: Many-body correlations (U_n with $n > 2$)

- Analytical forms/ efficient calculations:

Local energy method



$$E_L(\mathbf{R}|\Psi = \text{const}) \rightarrow U_2(\mathbf{R}) \sim \sum_{i < j} u(r_{ij})$$

- Most general 3body form

$$U_3(\mathbf{R}) = \sum_{i < j < k} u_3(\mathbf{r}_{ij}, \mathbf{r}_{ik})$$

- Local energy method suggests

$$E_L(\mathbf{R}|U_2) \rightarrow U_3[\mathbf{R}] \sim \sum_i \nabla_i [U_2(\mathbf{R})]^2$$

$$U_3[\mathbf{R}] = \sum_i \sum_{j,k} \mathbf{r}_{ij} \cdot \mathbf{r}_{ik} u_3(r_{ij}) u_3(r_{ik}) = \sum_i \left[\sum_j \mathbf{r}_{ij} u_3(r_{ij}) \right] \cdot \left[\sum_k \mathbf{r}_{ik} u_3(r_{ik}) \right]$$

- Introduce generalized vector/tensor forms

$$E_L(\mathbf{R}|U_3) \rightarrow U_4[\mathbf{R}]$$

“backflow vector”

$$\mathbf{d}_i = \sum_{j \neq i} \mathbf{r}_{ij} d(r_{ij})$$

1D functions to be optimized

“backflow tensor”

$$\underline{T}_i|_{\alpha\beta} = \sum_{j \neq i} \mathbf{r}_{ij}|_{\alpha} \mathbf{r}_{ij}|_{\beta} t(r_{ij})$$

$\alpha = x, y, z$

- n-body correlations** must be scalars:

$$U_3 = \lambda \sum_i \mathbf{d}_i \cdot \mathbf{d}_i \quad U_4 = \sum_i \mathbf{d}_i \cdot \underline{T}_i \cdot \mathbf{d}_i$$

n-body backflow $\mathbf{x}_i = \underline{T}_i \mathbf{d}_i$

- easily automatized
- Chain rules for derivatives
- 1D function per vector/tensor
- Computational cost remains $\sim N^2$

Backflow: history

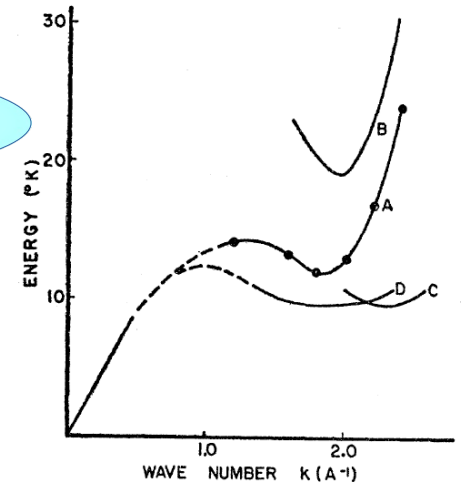
- Hydrodynamic considerations: Feynman, Cohen, Phys. Rev. 102, (1956)
“backflow” ansatz to describe excitations of liquid helium

$$\Phi_{\mathbf{k}}(\mathbf{R}) = \exp \left[i\mathbf{k} \cdot \mathbf{r}_A + \sum_{i \neq A} g(\mathbf{r}_i - \mathbf{r}_A) \right] \Psi_0(\mathbf{R})$$

$$= \exp [i\mathbf{k} \cdot \mathbf{q}_A] \Psi_0(\mathbf{R})$$

$$g(r) \sim \mathbf{k} \cdot \mathbf{r} / r^3$$

“backflow” coordinates $\mathbf{q}_A(\mathbf{R}) = \mathbf{r}_A + \sum_i (\mathbf{r}_A - \mathbf{r}_i) \eta(\mathbf{r}_A - \mathbf{r}_i)$



- Antisymmetrized form: “backflow” wave function for normal Fermi liquids (He³)
Schmidt, Pandharipande, Phys. Rev. B 19, 2504 (1979)

$$\Psi_{SJ}(\mathbf{R}) = \det_{\mathbf{k}i} \phi_{\mathbf{k}}(\mathbf{r}_i) \exp [-U(\mathbf{R})] \longrightarrow \Psi_{bf}(\mathbf{R}) = \det_{\mathbf{k}i} \phi_{\mathbf{k}}(\mathbf{q}_i) \exp [-U(\mathbf{R})]$$

- Liquid ³He Schmidt, Lee, Kalos, Chester, Phys. Rev. Lett. 47, 807 (1981)
- Homogeneous electron gas Kwon, Ceperley, Martin, Phys. Rev. B 48, 12037 (1993)
- Liquid/solid hydrogen: M.H., Ceperley, Pierleoni, Esler, Phys. Rev. E 68, 046707 (2003)
- Atoms, molecules, solids: López Ríos, Ma, Drummond, Towler, Needs, Phys. Rev. E 74, 066701 (2006)

Iterative backflow (renormalization)

M. Taddei, M. Ruggeri, S. Moroni, M.H., Phys. Rev. B 91, 115106 (2015)

M. Ruggeri, S. Moroni, M.H., Phys. Rev. Lett. 120, 205302 (2018)

M. H. and S. Moroni, Phys. Rev. B 99, 085121 (2019)

Fermions: He³

Bosons He⁴

Atoms

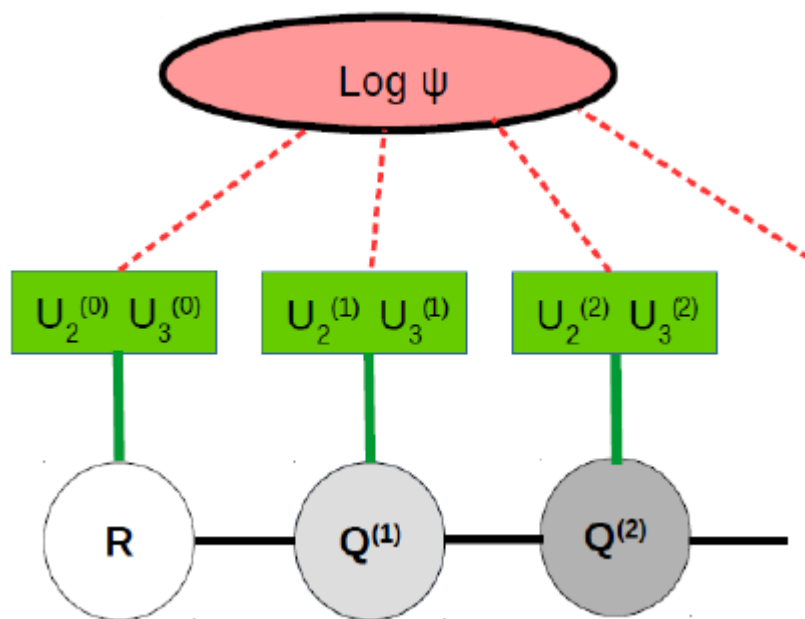
Use generalized backflow coordinates calculated iteratively:

$$\{Q^{(n)}\} = (q_1^{(n)}, q_2^{(n)}, \dots, q_N^{(n)}) \Rightarrow q_i^{(n+1)}[Q^{(n)}] = \sum_{j \neq i} q_{ij}^{(n)} \eta(q_{ij}^{(n)})$$

Improving the trial wave function iteratively:

$$\Psi_T = \exp \left[- \sum_n U_2^{(n)} \right]$$

$$U_2^{(n)}[Q^{(n)}] = \sum_{i < j} u_2(q_{ij}^{(n)})$$

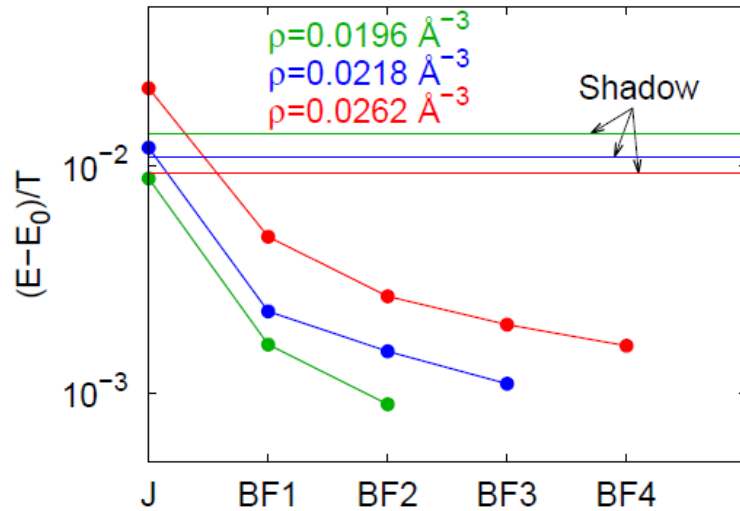


Original motivation of
Iterative backflow (deep backflow):
"renormalize" states at Fermi surface

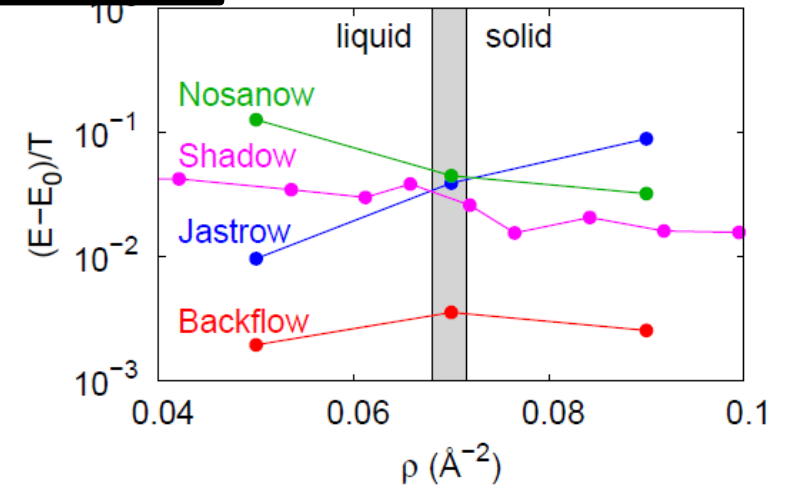
Benchmark for ground state energies

3D

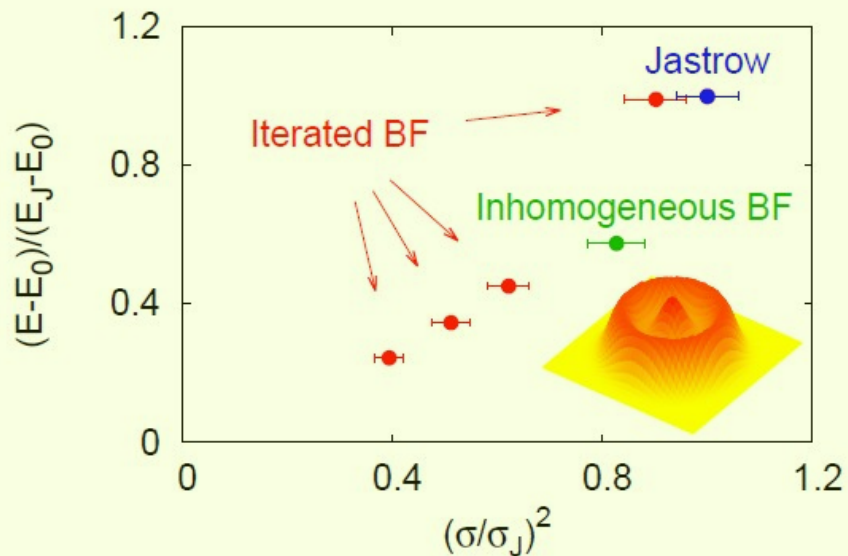
Bosonic liquid/ solid
 ^4He



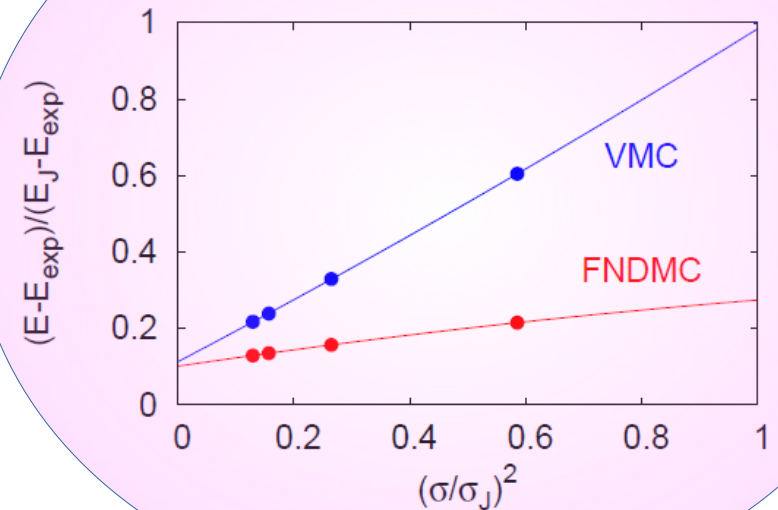
2D



N=7 Charged Bosons in external harmonic trap



Fermionic liquid ^3He



Iterative backflow - Neural quantum states for fermions in the continuum

General backflow determinant:

$$\Psi_T(\mathbf{R}) = \det \begin{pmatrix} \phi_1(\mathbf{q}_1) & \phi_1(\mathbf{q}_2) & \cdots & \phi_1(\mathbf{q}_N) \\ \phi_2(\mathbf{q}_1) & \phi_2(\mathbf{q}_2) & \cdots & \phi_2(\mathbf{q}_N) \\ \cdots & \cdots & \cdots & \cdots \\ \phi_N(\mathbf{q}_1) & \phi_N(\mathbf{q}_2) & \cdots & \phi_N(\mathbf{q}_N) \end{pmatrix}$$

general Many-body backflow:
MH, Bernu, Ceperley PRB 74, 104510 (2005)

'iterative backflow" (deep network)
Taddei, Ruggeri, Moroni, MH,
Phys. Rev. B 91, 115106 (2015).

Backflow coordinates depend on all particles: $\mathbf{q}_j = f(\mathbf{r}_j | \mathbf{R})$

- Exchange of particle j and n exchanges corresponding backflow coordinates:

$$P_{jn} \mathbf{q}_j = \mathbf{q}_n$$

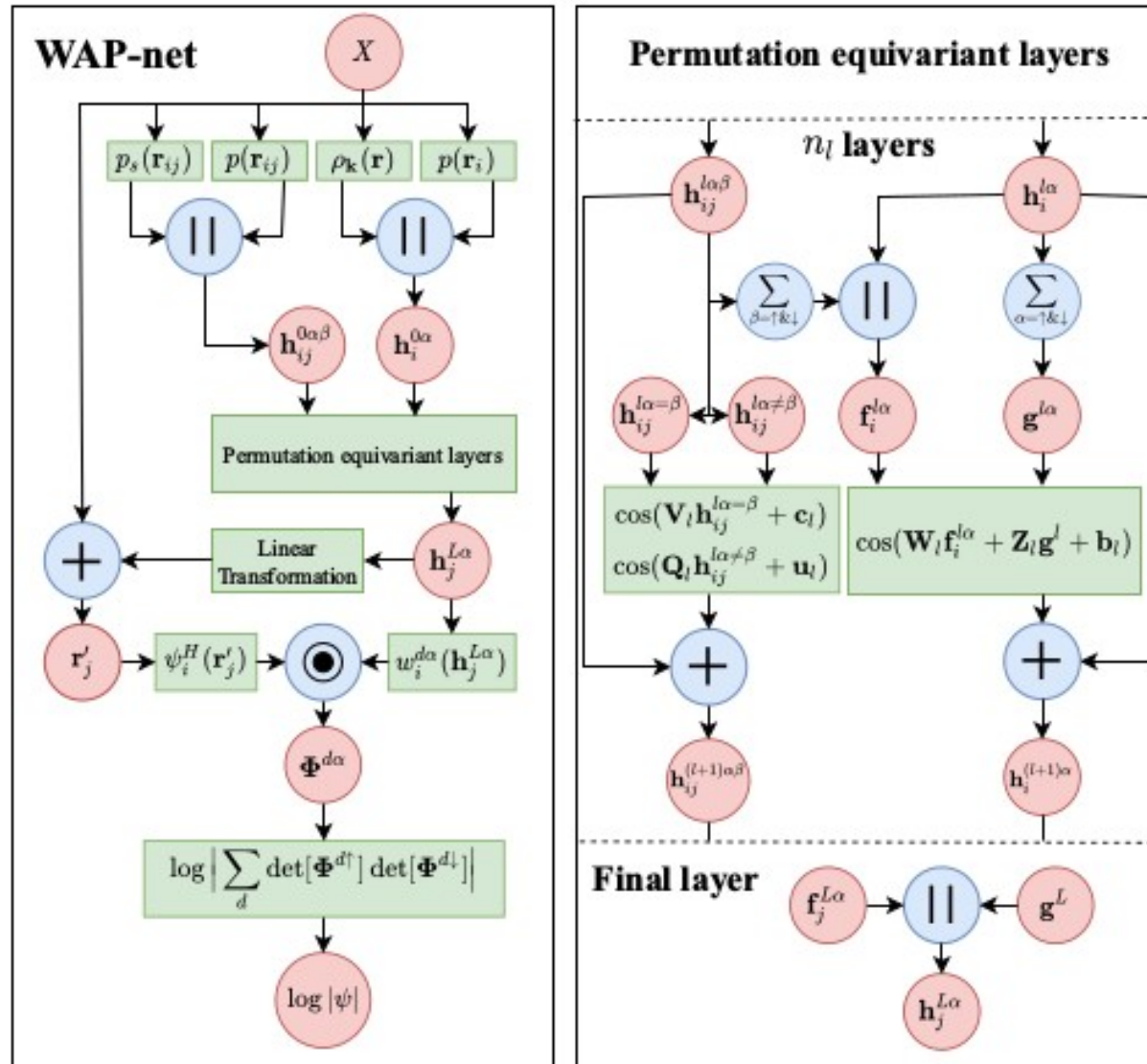
- Symmetric with respect to exchanges of $N-1$ particles

$$P_{nm} \mathbf{q}_j = f(\mathbf{r}_j | P_{nm} \mathbf{R}) = f(\mathbf{r}_j | \mathbf{R}) \quad n, m \neq j$$

- Although can represent arbitrary fermionic wave functions in principle, representation (universal approximation) theorems do not apply for extensive systems as fermionic wave functions are not sufficiently smooth!

Neural network based wave functions: WAP-net

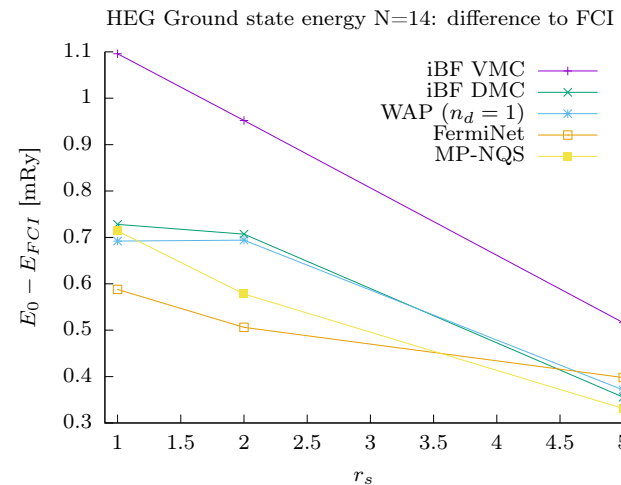
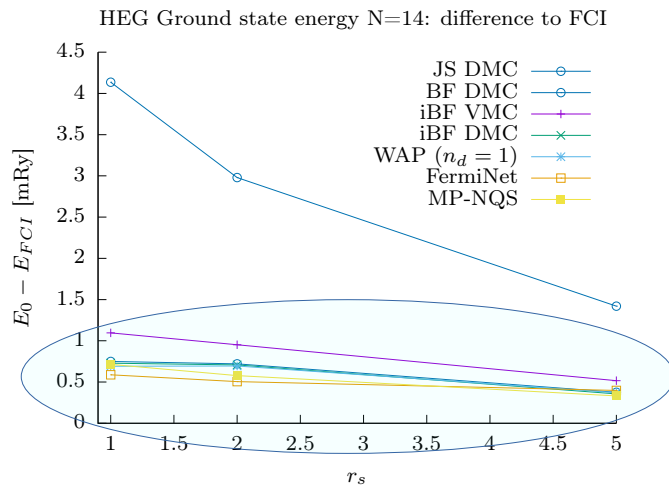
M. Wilson, S. Moroni, M. H., N. Gao, F. Wudarski, T. Vegge, A. Bhowmik
Phys. Rev. B 107, 235139 (2023).



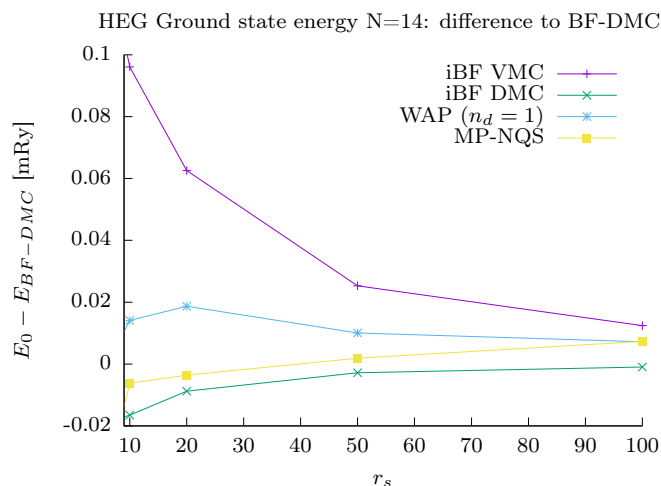
Benchmark: Homogeneous Electron Gas 3D N=14

IBF, WAP-net, FermiNet, MP-NQS, FCI...

High density $r_s < 5$



low density $r_s > 5$



FCI:

K. Liao, T. Schraivogel, H. Luo, D. Kats, A. Alavi, Phys. Rev. Research 3, 033072 (2021).

WAP-net, iterated backflox IBF:

M. Wilson, S. Moroni, M. H., N. Gao, F. Wudarski, T. Vegge, A. Bhowmik Phys. Rev. B 107, 235139 (2023).

FermiNet:

G. Cassella, H. Sutterud, S. Azadi, N. D. Drummond, D. Pfau, J. S. Spencer, W. M. C. Foulkes, Phys. Rev. Lett. 130, 036401 (2023).

MP-NQS:

G. Pescia, J. Nys, J. Kim, A. Lovato, G. Carleo, Phys. Rev. B 110, 035108 (2024).

Benchmarks beyond energies: Momentum distribution and $g_1(r)$

M. Wilson, S. Moroni, M. H., N. Gao, F. Wudarski,
T. Vegge, A. Bhowmik Phys. Rev. B 107, 235139 (2023).

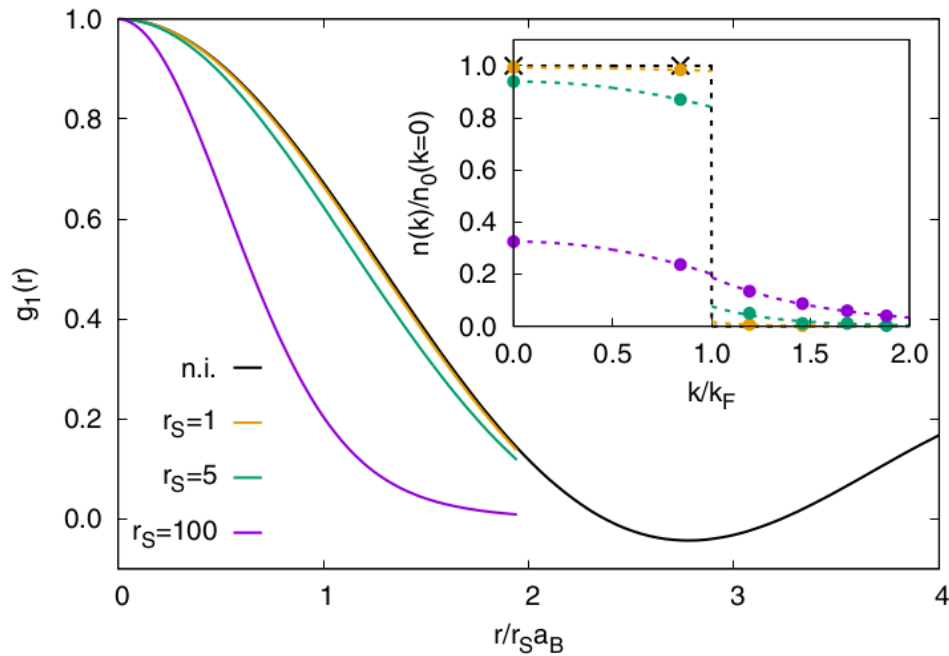


FIG. 5. The reduced single-particle density matrix $g_1(r)$ of the paramagnetic HEG. The colored lines are extrapolated estimates calculated with IB wave functions for $r_s = 1, 5$, and 100 at $N_e = 14$. The black line is the analytic result for the noninteracting Fermi gas at the same system size. The inset shows the corresponding momentum distributions (dotted lines are a guide to the eye).

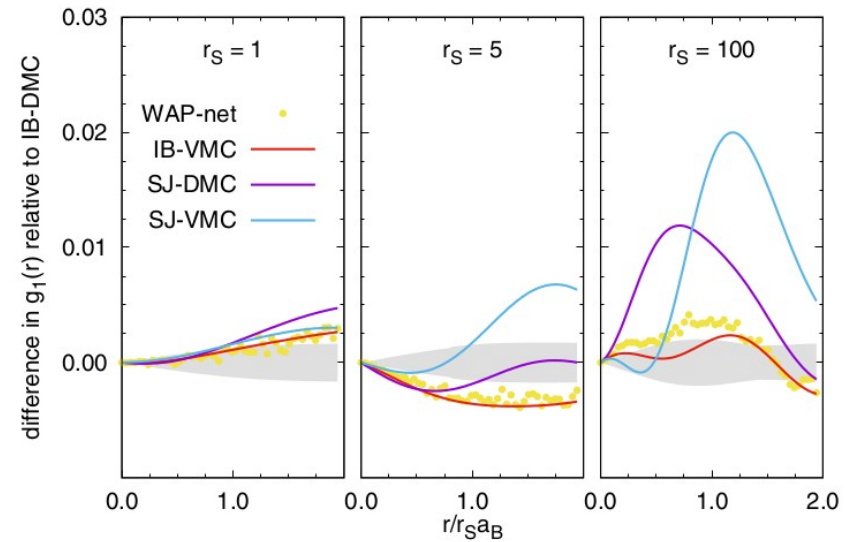


FIG. 6. The difference between various calculations of $g_1(r)$ and the IB extrapolated estimate IB-DMC of Fig. 5. The shaded area indicates the statistical uncertainty of IB-DMC. The WAP-net result is very close to the IB variational estimate IB-VMC.

Phase diagram of the homogeneous electron gas:

Electron gas:
(jellium, HEG, OCP)

$$H = -\frac{\hbar^2}{2m_e} \sum_i \nabla_i^2 + \sum_{i < j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}$$

$$r_s = a/a_B$$

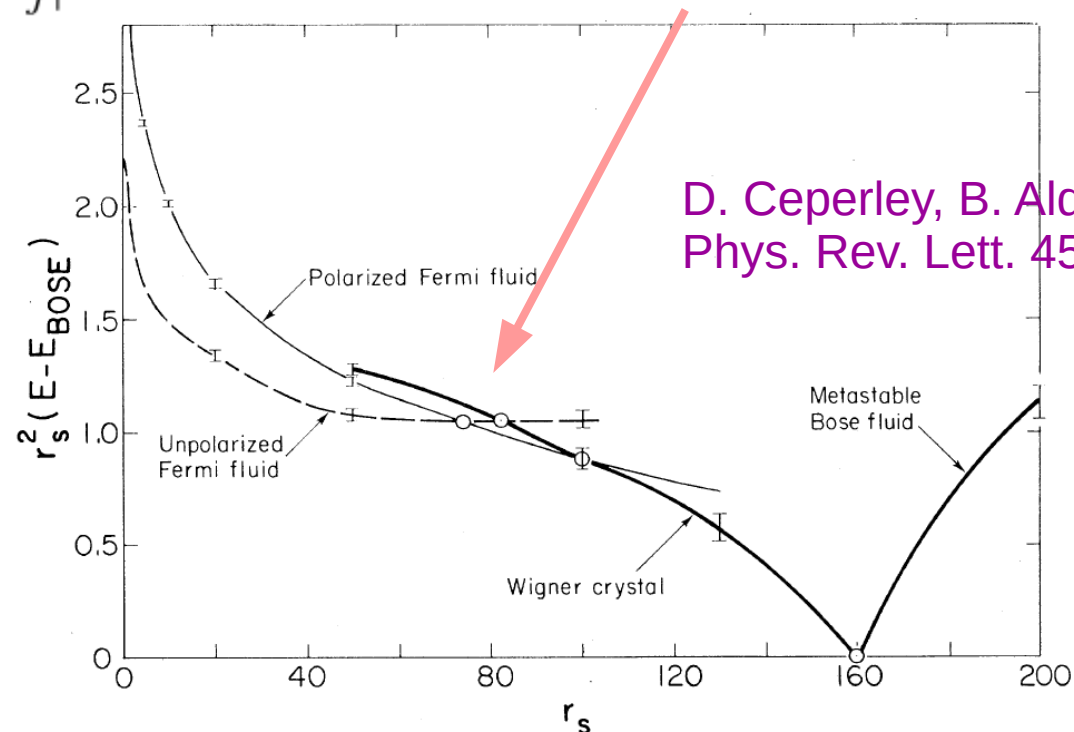
$a \sim n^{-1/3}$: mean distance

Itinerant magnetism in 3D electron gas?

F. Bloch, Zeitschrift f. Physik 57, 545 (1929)

E. P. Wigner, Phys. Rev. 46, 1002 (1934)

E.C. Stoner, Proc. R. Soc. Lond. A 165, 372 (1938)



D. Ceperley, B. Alder,
Phys. Rev. Lett. 45 (1980)

FIG. 2. The energy of the four phases studied relative to that of the lowest boson state times r_s^2 in rydbergs vs r_s in Bohr radii. Below $r_s = 160$ the Bose fluid is the most stable phase, while above, the Wigner crystal is most stable. The energies of the polarized and unpolarized Fermi fluid are seen to intersect at $r_s = 75$. The polarized (ferromagnetic) Fermi fluid is stable between $r_s = 75$ and $r_s = 100$, the Fermi Wigner crystal above $r_s = 100$, and the normal paramagnetic Fermi fluid below $r_s = 75$.

Application of iterated backflow renormalization:

Itinerant magnetism in 3D electron gas? Many-body correlations vs Stoner (polarization) transition

M.H. and S. Moroni, Phys. Rev. Lett. 124, 206404 (2020)

Many-body correlations in the low Density region can be accurately Extrapolated by iterative backflow

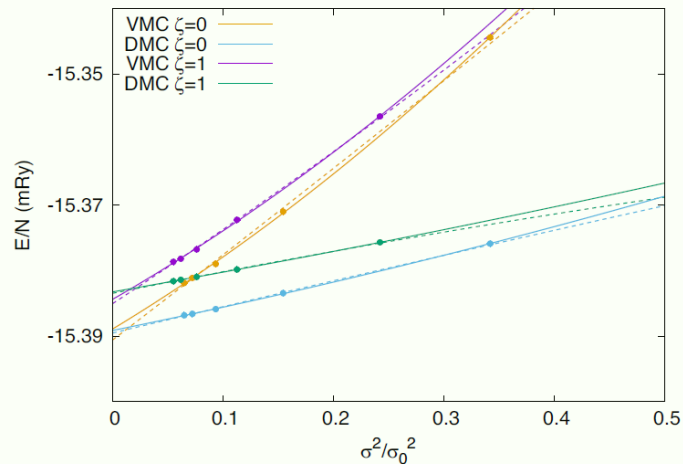


FIG. 1. Extrapolation of the energy to zero variance for $r_s = 100$ at polarizations $\zeta = 0$ and 1 . The data are calculated with VMC and DMC using SJ, BF0, ..., BF4 wave functions in order of decreasing energy. The reference value σ_0^2 is the variance of the local energy at $\zeta = 0$ with the SJ wave function. The curves are quadratic fits; for each set of data points (VMC and DMC for $\zeta = 0$ and 1) there are two curves, one of which (solid line) excludes the SJ energy from the fit.

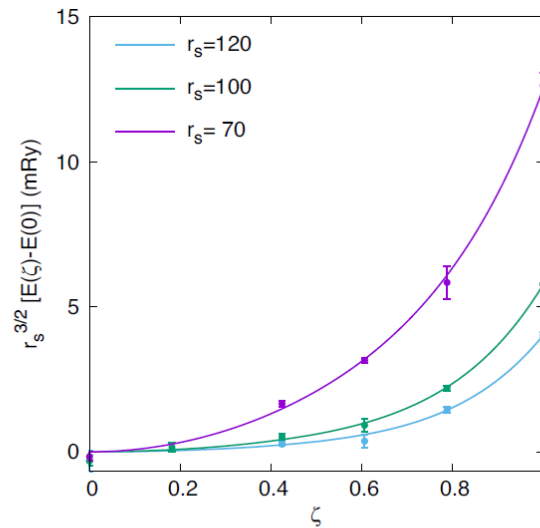
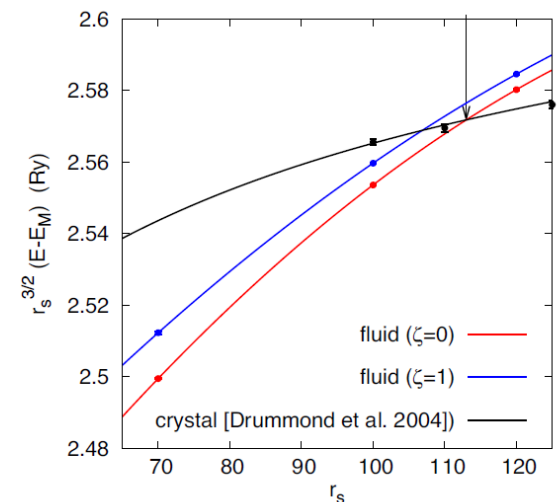


FIG. 3. The polarization energy $E(\zeta) - E(0)$ obtained from zero-variance extrapolations of the DMC energies without the SJ result. The lines are polynomial fits with terms of order 0, 2, and 6. Alternative functional forms and ensuing confidence levels for

Wigner crystallization
sets in before
Polarization transition



Does a Stoner transition occur in nature?

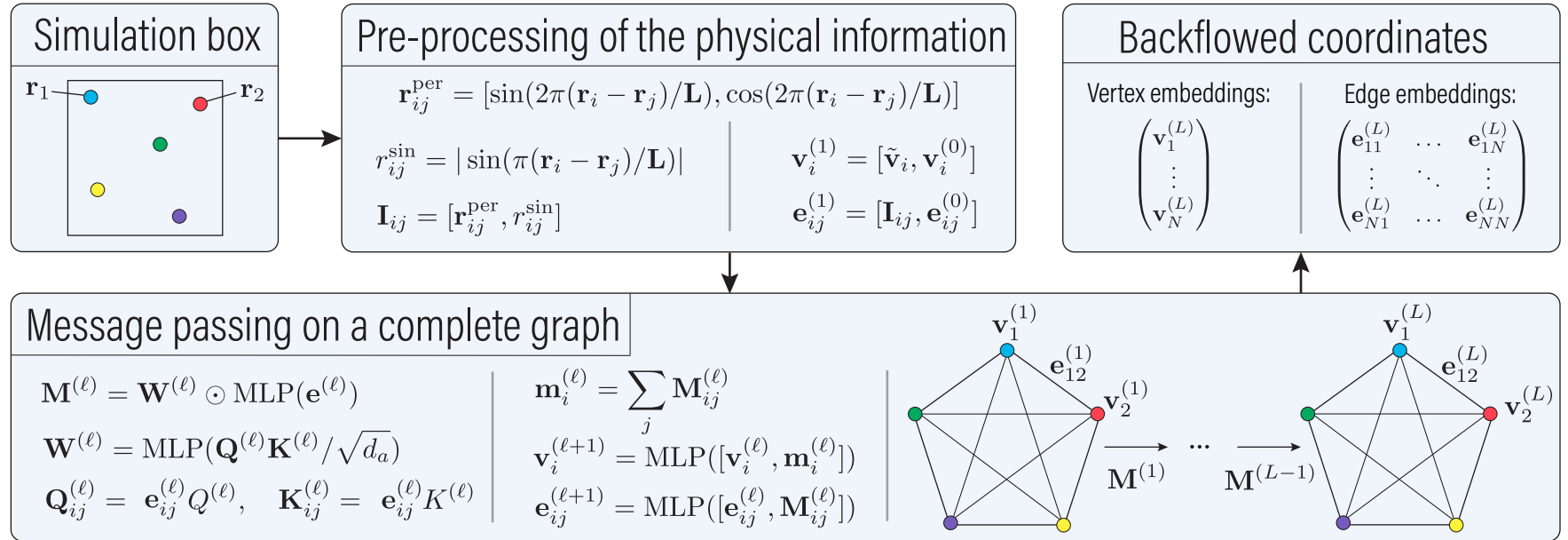
- **Electronic systems:**
itinerant ferromagnetism: **correlations** + **band structure**
- different **isotropic** systems with **spin-independent** ?
(ultracold atoms/ molecules)
 - Change scattering length $a_s \rightarrow \infty$?
=> difficulties/controversals due to 2-body bound state leading to instability!
 - Dipolar interactions (heteronuclear molecules, Rydberg atoms)?
=> no polarization transition in 2D before freezing
T. Comparin, R. Bombin, M. H., F. Mazzanti, J. Boronat, and S. Giorgini,
Phys. Rev. A 99, 043609 (2019)
- **Liquid helium** ? No
experiment / higher order backflow calculations...

Conjecture:

For an isotropic normal Fermi liquid with
spin-independent isotropic interactions
the ground state remains unpolarized up to freezing.

Liquid/solid He⁴ at T=0: Graph neural network results

D. Linteau, G. Pescia, J. Nys, G. Carleo, M. H. Phys. Rev. Lett 134, 246001 (2025)



Size consistency!

N	n [Å ⁻²]	VMC U2		VMC U3		DMC	VMC NQS	
		E/N	σ ² /N	E/N	σ ² /N	E/N	E/N	σ ² /N
30	0.065	0.632(5)	45.0(5)	0.132(3)	17.8(1)	-0.047(1)	-0.046(1)	0.11
	0.068	1.073(8)	55.3(9)	0.477(4)	9.7(1)	0.245(2)	0.246(2)	0.25
	0.070	0.684(4)	14.9(5)	0.581(2)	4.28(7)	0.461(2)	0.450(2)	0.31
	0.072	0.867(4)	16.1(2)	0.765(2)	4.52(7)	0.661(2)	0.659(2)	0.29
	0.075	1.226(3)	19.9(3)	1.101(2)	4.49(8)	1.020(1)	1.020(1)	0.24
80	0.065	0.667(6)	43.1(1)	0.268(4)	15.1(2)	-0.021(1)	-0.020(1)	0.15
	0.068	1.133(8)	53(1)	0.640(4)	18.9(2)	0.287(1)	0.290(1)	0.20
	0.070	0.738(3)	14.8(2)	0.650(2)	4.52(7)	0.504(2)	0.518(3)	1.17
	0.075	1.305(3)	18.1(2)	1.195(2)	4.08(4)	1.073(1)	1.085(3)	0.97

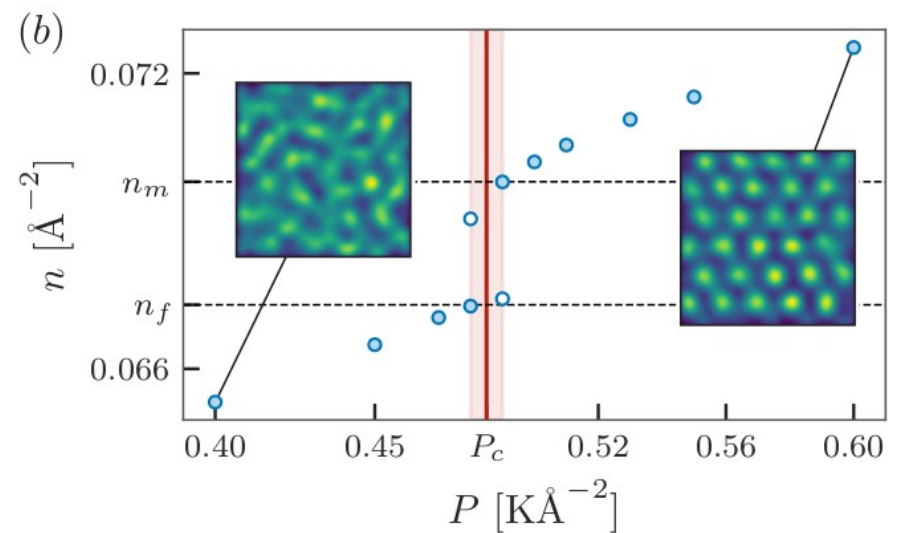
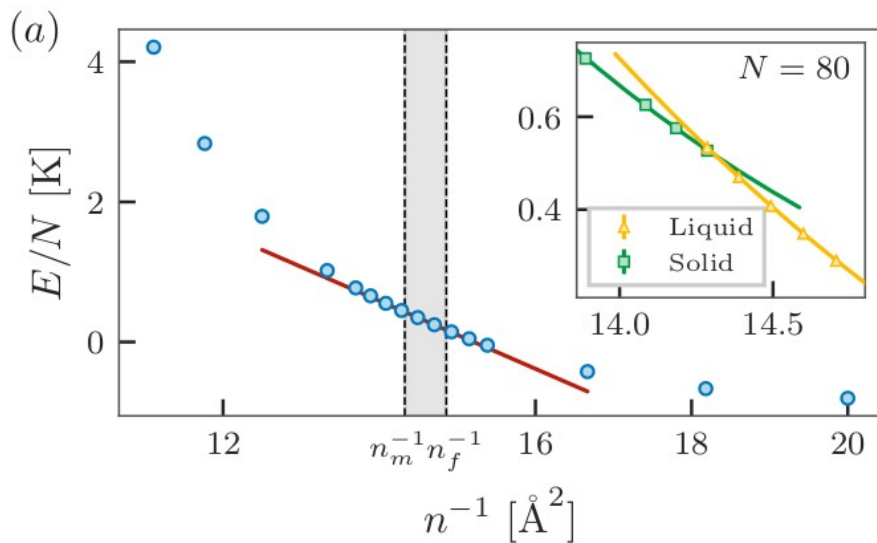
Table I. Ground state energies using a truncated and shifted version of the Aziz potential (described in Appendix A 2). Energy (per particle) E/N are in units of kelvin while energy variances (per particle) σ^2/N are in units of kelvin squared. The new quantum state results (VMC NQS) obtained in this work are compared with two simpler wavefunctions, including two-body pair correlations (VMC U2), as well as additional three body correlations (VMC U3) (see Eq. (C1)). In the solid phase, the

Liquid/solid He⁴ at T=0: constant pressure calculations

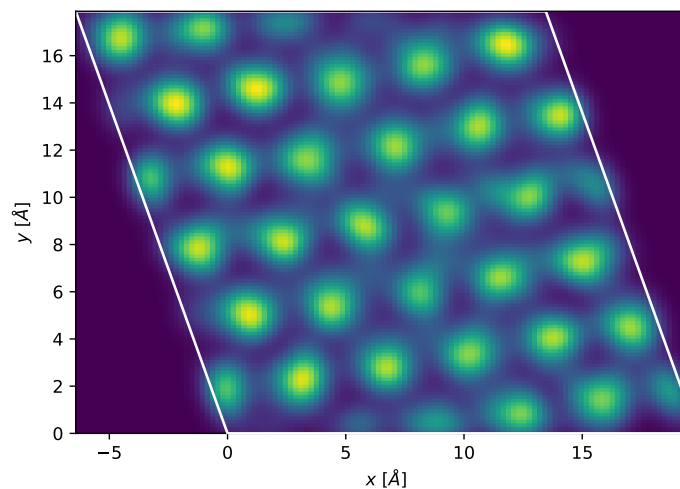
D. Linteau, G. Pescia, J. Nys, G. Carleo, M. H. Phys. Rev. Lett 134, 246001 (2025)

Canonical ensemble (fixed density)

Isobaric ensemble (fixed pressure)



Isobaric ensemble:
varying angle of box
N=31 atoms



Liquid/solid He⁴ at T=0: Crystalline vs hexatic order

An old question: Is there a phase with hexatic order?

- Crystalline order: structure factor $S(G)$, Bragg peaks

- Hexatic order: $\Psi_6(\mathbf{r}_i) = \frac{1}{6} \sum_{j \in \mathcal{N}_6(i)} e^{6i\theta_{ij}}$

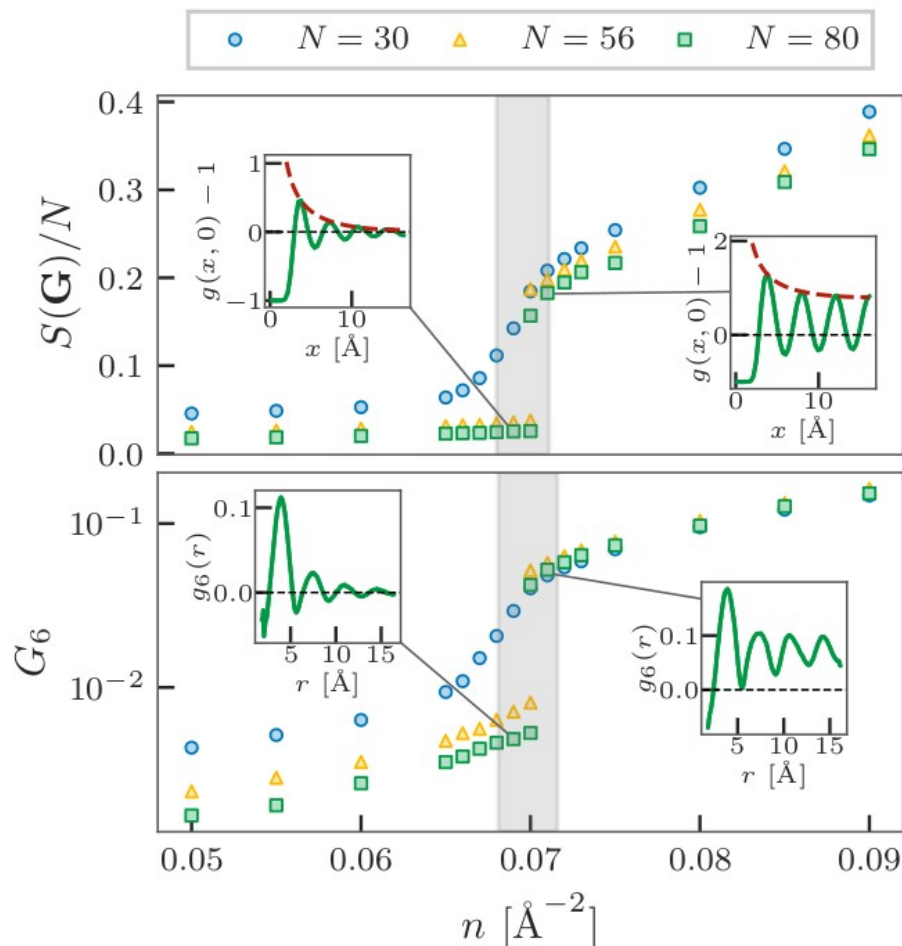
$$g_6(r) = \langle \Psi_6^*(\mathbf{r}_i) \Psi_6(\mathbf{r}_j) \rangle,$$

$$g_6(r \rightarrow \infty) \sim \text{cst.} \rightarrow \text{solid,}$$

$$g_6(r) \sim r^{-\eta} \rightarrow \text{hexatic,}$$

$$g_6(r) \sim e^{-r/\xi} \rightarrow \text{liquid,}$$

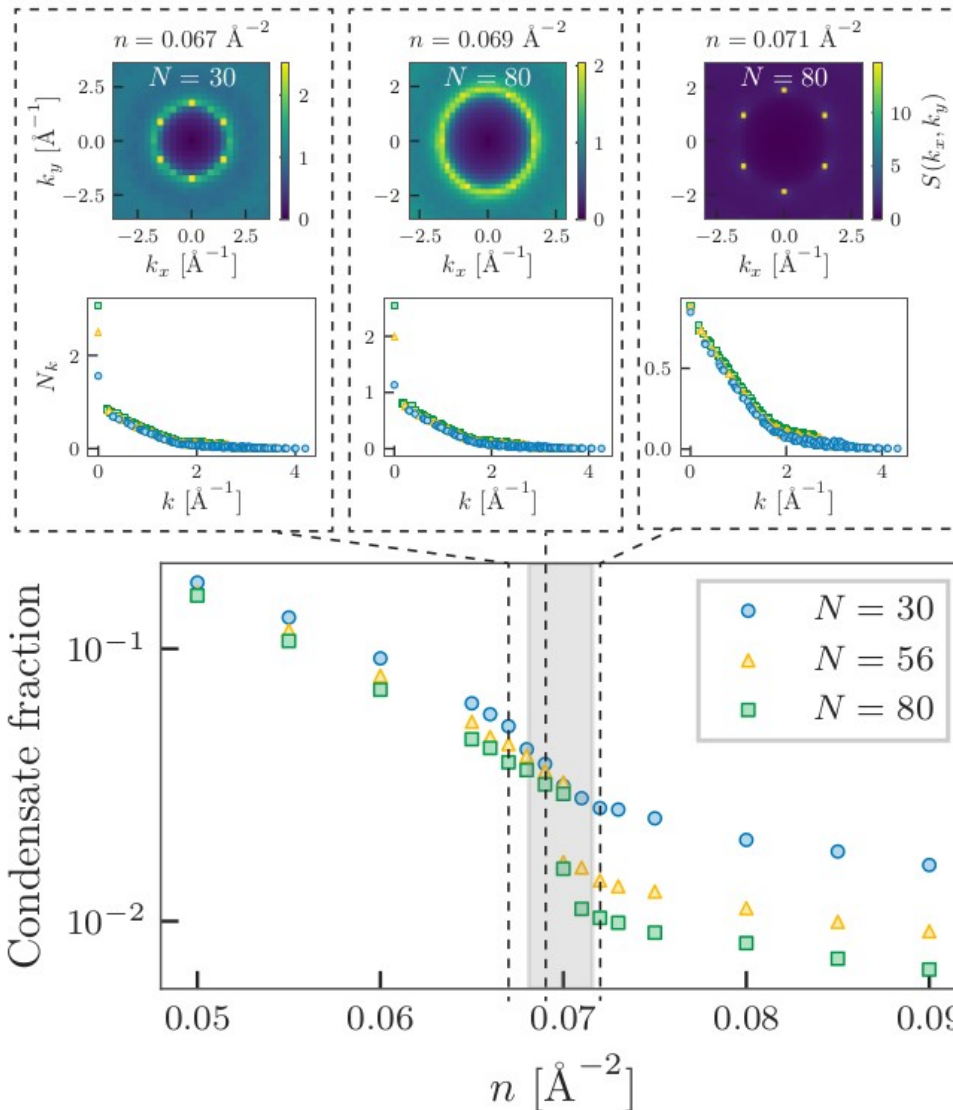
N likely too small to be conclusive....



$$G_6 = (2\pi/V) \int_0^{\min(\mathbf{L})/2} r dr g_6(r)$$

Liquid/solid He⁴ at T=0: Off-diagonal long-range order, supersolid?

• ODLRO:
$$\frac{n(\mathbf{r})}{n} = \mathbb{E} \left[\frac{\Psi_0(\mathbf{r}_1 + \mathbf{r}, \mathbf{r}_2, \dots, \mathbf{r}_N)}{\Psi_0(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N)} \right]$$



Structure factor $S(\mathbf{k})$

Momentum distribution:

$$N_{\mathbf{k}} = \int d\mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} n(\mathbf{r})$$

Condensate fraction n_0

Large finite size effects:

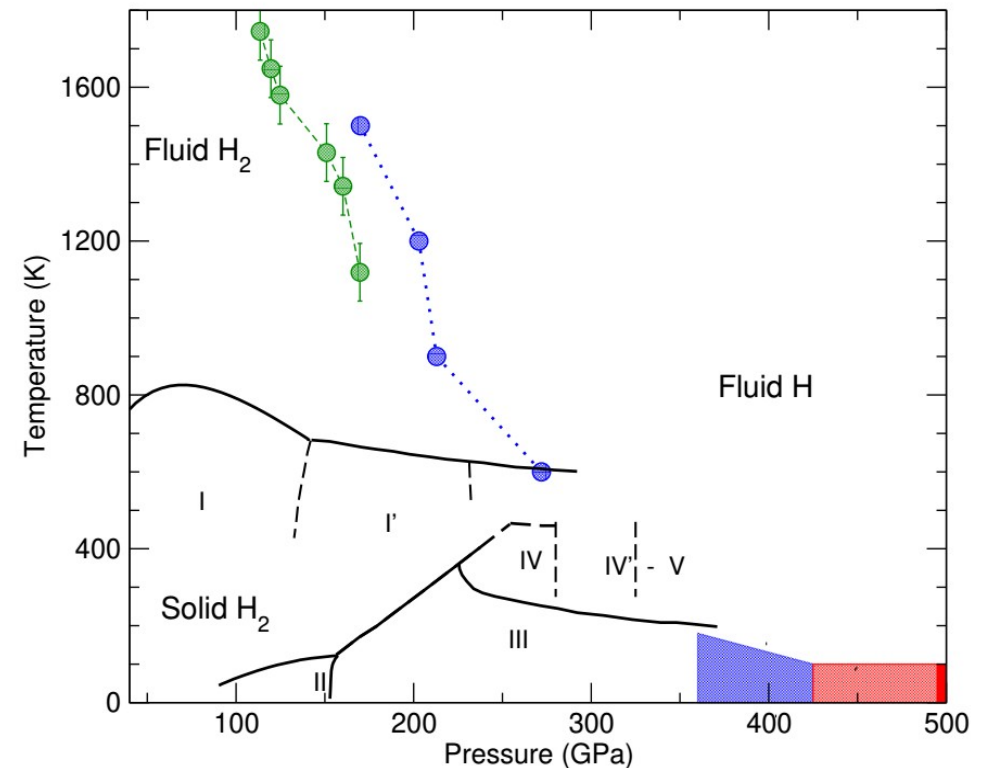
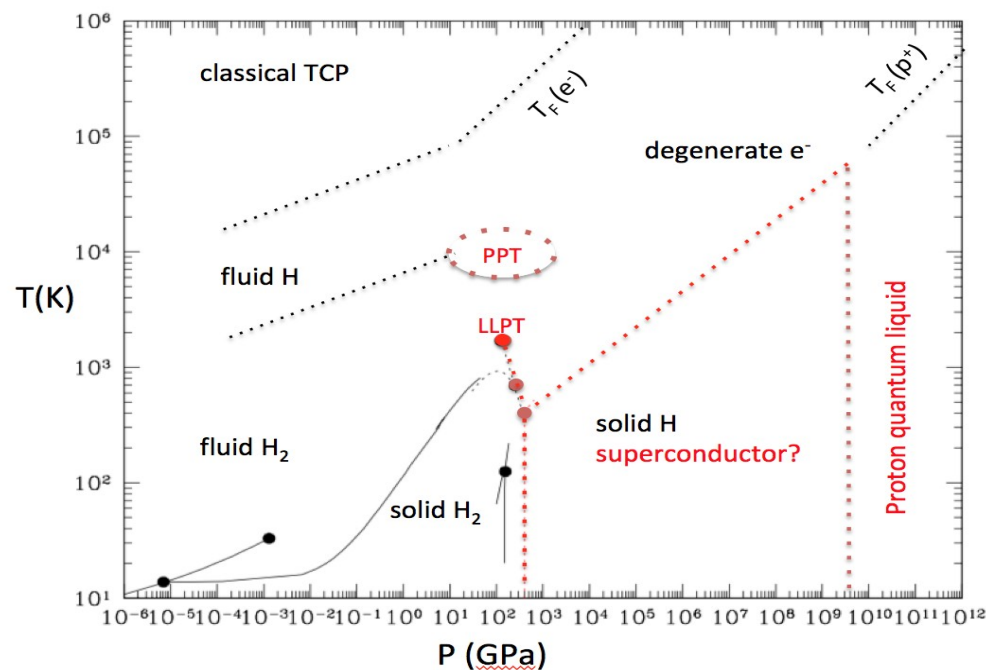
$$n_0 = N_0/V = n - V^{-1} \sum_{\mathbf{k} \neq 0} N_{\mathbf{k}}$$

Liquid/solid hydrogen under pressure:

- Non-relativistic Hamiltonian for ions and electrons

$$H = - \sum_n \frac{\hbar^2 \nabla_n^2}{2M} - \sum_i \frac{\hbar^2 \nabla_i^2}{2m} + \sum_{n < m} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_{n,i} \frac{Ze^2}{|\mathbf{r}_i - \mathbf{R}_n|} + \sum_{n < m} \frac{Z^2 e^2}{|\mathbf{R}_n - \mathbf{R}_m|}$$

$$= - \sum_n \frac{\hbar^2 \nabla_n^2}{2M} + H_e(\mathbf{R})$$



Liquid/solid hydrogen under pressure: $T < \sim 10^3 \text{K}$

- Born-Oppenheimer approximation ($T \ll T_F$):
electrons in ground state for fixed ion positions

$$H_e(\mathbf{R})\Psi(\mathbf{r}) = E_0(\mathbf{R})\Psi(\mathbf{r})$$

- $T > 0$ distribution of ions (within Born-Oppenheimer)

Coupled Electron-Ion Monte Carlo (CEIMC)

- Classical Boltzmann distribution of nuclei (deuterium?):

C. Pierleoni, M. Morales, MH, D. Ceperley...

Molecular dynamics (VMC-MD)

C. Attaccalite, M. Casula, S. Sorella...

$$p(\mathbf{R}) \sim e^{-\beta E_0(\mathbf{R})}$$

- Quantum nuclei: thermal density matrix

→ Lei Wang..

$$\rho(\mathbf{R}) \propto \langle \mathbf{R} | e^{-H/T} | \mathbf{R} \rangle \approx \left\langle \mathbf{R} \left| \exp \left[\sum_n \frac{\hbar^2 \nabla_n^2}{2MT} - \frac{E_0(\mathbf{R})}{T} \right] \right| \mathbf{R} \right\rangle$$

- $T=0$: quantum nuclei without Born-Oppenheimer (full electron-proton ground state)

$$H\Psi(\mathbf{r}, \mathbf{R}) = \left[-\sum_n \frac{\hbar^2 \nabla_n^2}{2M} + H_e(\mathbf{R}) \right] \Psi(\mathbf{r}, \mathbf{R}) = E_0 \Psi(\mathbf{r}, \mathbf{R})$$

Diffusion Monte Carlo (DMC): Ceperley Alder, PRB 36, 2092 (1987)

Liquid/solid hydrogen under pressure: Quantum Nuclear + Electrons

- Addressing full Schrödinger equation:

$$H\Psi(\mathbf{r}, \mathbf{R}) = \left[-\sum_n \frac{\hbar^2 \nabla_n^2}{2M} + H_e(\mathbf{R}) \right] \Psi(\mathbf{r}, \mathbf{R}) = E_0 \Psi(\mathbf{r}, \mathbf{R})$$

→ $M \gg m_e$: different scale in (imaginary time) dynamics
bottleneck of stochastic projection Monte Carlo methods (DMC, RMC..)
 (solution $T > 0$: Born-Oppenheimer), but electron-phonon etc...?

- Ground state wave function (for atomic phase):
 including electrons, nuclear structure, phonons, electron-phonon effects...

“nuclear wfn”
Symmetric correlations (Phonons)

$\Psi(\mathbf{r}, \mathbf{R}) = \Phi(\mathbf{R}) \det \phi_{\mathbf{k}}(\mathbf{r}_i | \mathbf{r}, \mathbf{R}) e^{-U(\mathbf{r}, \mathbf{R})}$

Electronic (Bloch) orbitals (electron-phonon)

$$\phi_{\mathbf{k}}^{\sigma}(\mathbf{r}_i) = \sum_{\mathbf{n} \in \mathbb{Z}^3} \sum_{I=1}^N e^{i\mathbf{k} \cdot (\mathbf{R}_I + \mathbf{n}L)} \chi(\mathbf{r}_i^{\sigma} - \mathbf{R}_I - \mathbf{n}L)$$

Atomic hydrogen: Neural ground state

D. Linteau, S. Moroni, G. Carleo, M. H., Phys. Rev. Res.8, L022007 (2026)

$$\phi_{\mathbf{k}}^{\sigma}(\mathbf{r}_i) = \sum_{\mathbf{n} \in \mathbb{Z}^3} \sum_{I=1}^N e^{i\mathbf{k} \cdot (\mathbf{R}_I + \mathbf{n}L)} \chi(\mathbf{r}_i^{\sigma} - \mathbf{R}_I - \mathbf{n}L)$$

$$\Psi(\mathbf{r}, \mathbf{R}) = \Phi(\mathbf{R}) \det \phi_{\mathbf{k}}(\mathbf{r}_i | \mathbf{r}, \mathbf{R}) e^{-U(\mathbf{r}, \mathbf{R})}$$

Data– Given a Monte Carlo configuration $((\mathbf{r}_1, s_1), \dots, (\mathbf{r}_N, s_N), \mathbf{R}_1, \dots, \mathbf{R}_N)$, where s_i is an electron spin label, the following electron-electron and electron-proton input tensors can be constructed:

$$\mathbf{I}_{ij} = [\sin(2\pi \mathbf{r}_{ij}/L), \cos(2\pi \mathbf{r}_{ij}/L), |\sin(\pi \mathbf{r}_{ij}/L)|, s_i s_j] \in \mathbb{R}^{2d+2}, \quad (\text{A1})$$

$$\mathbf{I}_{il} = [\sin(2\pi \mathbf{r}_{il}/L), \cos(2\pi \mathbf{r}_{il}/L), |\sin(\pi \mathbf{r}_{il}/L)|] \in \mathbb{R}^{2d+1}, \quad (\text{A2})$$

where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, $\mathbf{r}_{il} = \mathbf{r}_i - \mathbf{R}_l$ and the square brackets correspond to the concatenation operation. The following hyperparameters have to be fixed by the user: the hidden dimension h , the attention dimension a , the MLP output dimension e , and the number of message-passing iteration(s) b .

(The vector or matrix dimension is shown below as a comment on the right.)

Initialization– instantiate “hidden” embedding parameters $\mathbf{h}_i^{(0)}, \mathbf{H}_{ij}^{(0)}, \mathbf{H}_{il}^{(0)}$ $\triangleright h$

Define vertex states $\mathbf{y}_i^{(0)} = \mathbf{h}_i^{(0)}$ $\triangleright h$

Define edge states $\mathbf{Y}_{ij}^{(0)} = [\mathbf{I}_{ij}, \mathbf{H}_{ij}^{(0)}], \mathbf{Y}_{il}^{(0)} = [\mathbf{I}_{il}, \mathbf{H}_{il}^{(0)}]$ $\triangleright 2d + 2 + h, 2d + 1 + h$

for $0 \leq \ell \leq b$ **do**

Electron-proton edge update:

 Initialize query and key matrices $\mathbf{Q}_{ep}^{(\ell)}, \mathbf{K}_{ep}^{(\ell)}$ $\triangleright \dim(\mathbf{Y}_{il}^{(\ell)}) \times a$

 Compute queries and keys $\mathbf{Q}_{il}^{(\ell)} = \mathbf{Q}_{ep}^{(\ell)} \mathbf{Y}_{il}^{(\ell)}, \mathbf{K}_{il}^{(\ell)} = \mathbf{K}_{ep}^{(\ell)} \mathbf{Y}_{il}^{(\ell)}$ $\triangleright a$

 Compute attention weights $\mathbf{W}_{il}^{(\ell)} = \text{MLP}_{ep,1}^{(\ell)}(\mathbf{Q}_{il}^{(\ell)} \mathbf{K}_{il}^{(\ell)} / \sqrt{a})_{il}$ $\triangleright e$

 Compute messages $\mathbf{M}_{il}^{(\ell)} = \mathbf{W}_{il}^{(\ell)} \odot \text{MLP}_{ep,2}^{(\ell)}(\mathbf{Y}_{il}^{(\ell)})$ $\triangleright e$

 Update edge hidden states $\mathbf{H}_{il}^{(\ell+1)} = \text{MLP}_{ep,3}^{(\ell)}([\mathbf{Y}_{il}^{(\ell)}, \mathbf{M}_{il}^{(\ell)}])$ $\triangleright e$

 Update edge states $\mathbf{Y}_{il}^{(\ell+1)} = [\mathbf{I}_{il}, \mathbf{H}_{il}^{(\ell+1)}]$ $\triangleright 2d + 1 + e$

Electron-electron edge and vertex update:

 Initialize query and key matrices $\mathbf{Q}_{ee}^{(\ell)}, \mathbf{K}_{ee}^{(\ell)}$ $\triangleright \dim(\mathbf{Y}_{ij}^{(\ell)}) \times a$

 Compute queries and keys $\mathbf{Q}_{ij}^{(\ell)} = \mathbf{Q}_{ee}^{(\ell)} \mathbf{Y}_{ij}^{(\ell)}, \mathbf{K}_{ij}^{(\ell)} = \mathbf{K}_{ee}^{(\ell)} \mathbf{Y}_{ij}^{(\ell)}$ $\triangleright a$

 Compute attention weights $\mathbf{W}_{ij}^{(\ell)} = \text{MLP}_{ee,1}^{(\ell)}(\mathbf{Q}_{ij}^{(\ell)} \mathbf{K}_{ij}^{(\ell)} / \sqrt{a})_{ij}$ $\triangleright e$

 Compute messages $\mathbf{M}_{ij}^{(\ell)} = \mathbf{W}_{ij}^{(\ell)} \odot \text{MLP}_{ee,2}^{(\ell)}(\mathbf{Y}_{ij}^{(\ell)})$ $\triangleright e$

 Compute vertex cross-species contributions: $\mathbf{C}_i^{(\ell)} = \sum_l \text{MLP}_{ee,3}^{(\ell)}(\mathbf{Y}_{il}^{(\ell+1)})$ $\triangleright e$

 Compute edge cross-species contributions: $\mathbf{C}_{ij}^{(\ell)} = \sum_l \text{MLP}_{ee,4}^{(\ell)}(\mathbf{Y}_{il}^{(\ell+1)}) \odot \text{MLP}_{ee,5}^{(\ell)}(\mathbf{Y}_{jl}^{(\ell+1)})$ $\triangleright e$

 Update vertex hidden states $\mathbf{h}_i^{(\ell+1)} = \text{MLP}_{ee,6}^{(\ell)}([\mathbf{h}_i^{(\ell)}, \sum_j \mathbf{M}_{ij}^{(\ell)}, \mathbf{C}_i^{(\ell)}])$ $\triangleright e$

 Update edge hidden states $\mathbf{H}_{ij}^{(\ell+1)} = \text{MLP}_{ee,7}^{(\ell)}([\mathbf{Y}_{ij}^{(\ell)}, \mathbf{M}_{ij}^{(\ell)}, \mathbf{C}_{ij}^{(\ell)}])$ $\triangleright e$

 Update vertex states $\mathbf{y}_i^{(\ell+1)} = \mathbf{h}_i^{(\ell+1)}$ $\triangleright e$

 Update edge states $\mathbf{Y}_{ij}^{(\ell+1)} = [\mathbf{I}_{ij}, \mathbf{H}_{ij}^{(\ell+1)}]$ $\triangleright 2d + 2 + e$

end for

return final vertex and edge states $\mathbf{y}_i^{(b)}, \mathbf{Y}_{ij}^{(b)}, \mathbf{Y}_{il}^{(b)}$

Add propagator like backflow:

$$\mathbf{r}_i \rightarrow \mathbf{r}_i + W \mathbf{y}_i^{(b)}$$

With electron vertex $\mathbf{y}$ from MPP and matrix W and electron edge $\mathbf{Y}$ in Jastrow

$$U(\mathbf{x}) \rightarrow U(\mathbf{x}) + \sum_{i < j} [w \text{MLP}(\mathbf{Y}_{ij}^{(b)})]$$

2889 parameters!

Liquid/solid hydrogen under pressure: Neural quantum states

D. Linteau, S. Moroni, G. Carleo, M. H., Phys. Rev. Res.8, L022007 (2026)

Static protons (Born-Oppenheimer)

N	wave function	E/N	σ^2/N
16	SJ-PW (DMC)	-0.4857(1)	0.0773(25)
	SJ-LDA (DMC)	-0.4890(5)	-
	BF-PW (DMC)	-0.4905(1)	0.0232(1)
	NQS (VMC)	-0.49154(1)	0.0062(1)
54	SJ-PW (DMC)	-0.5329(1)	0.0642(9)
	BF-PW (DMC)	-0.5382(1)	0.0222(2)
	SJ-LDA (DMC)	-0.5390(5)	-
	NQS (VMC)	-0.54007(2)	0.00479(7)
128	SJ-PW	-0.4900(2)	0.0656(23)
	BF-PW (DMC)	-0.4978(4)	0.030(1)
	SJ-LDA (DMC)	-0.4978(2)	-
	NQS (VMC)	-0.49991(7)	0.00771(4)

Table I. Static hydrogen with protons pinned to a BCC lattice at $r_s = 1.31$ using PBC. The energy per atom, E/N , is given in units of Hartree, and σ^2 denotes the energy variance. The NQS energy, obtained from VMC calculations using the ansatz given in Eq. (2), is compared to previous DMC reference energies taken from [20], based on Slater-Jastrow (SJ-PW) and backflow (BF-PW) plane wave orbitals as well as Slater-Jastrow calculations using DFT-LDA (SJ-LDA) orbitals.

dynamic protons (No Born-Oppenheimer)

N	wave function	E/N	σ^2/N
16	SJ-LDA (VMC)	-0.46785(2)	-
	BF-PW (VMC)	-0.4724(1)	0.030(2)
	BF-PW (DMC)	-0.4792(1)	-
	NQS (VMC)	-0.48091(5)	0.0114(1)
54	SJ-LDA (VMC)	-0.5195(2)	-
	SJ-LDA (DMC)	-0.52415(5)	-
	BF-PW (VMC)	-0.52194(5)	0.025(1)
	BF-PW (DMC)	-0.52610(7)	-
	NQS (VMC)	-0.52854(9)	0.0088(1)

Table II. Ground state energies for dynamic hydrogen at $r_s = 1.31$, for different wave functions obeying PBC. The reference VMC and DMC energies for Slater-Jastrow with LDA-DFT orbitals (SJ-LDA) and backflow plane wave orbitals (BF-PW) are taken from [15]. For all calculations, the nuclear part of the trial wave function is given by gaussians localized around BCC lattice sites.

Neural-backflow wave functions: Exploring crystalline structures

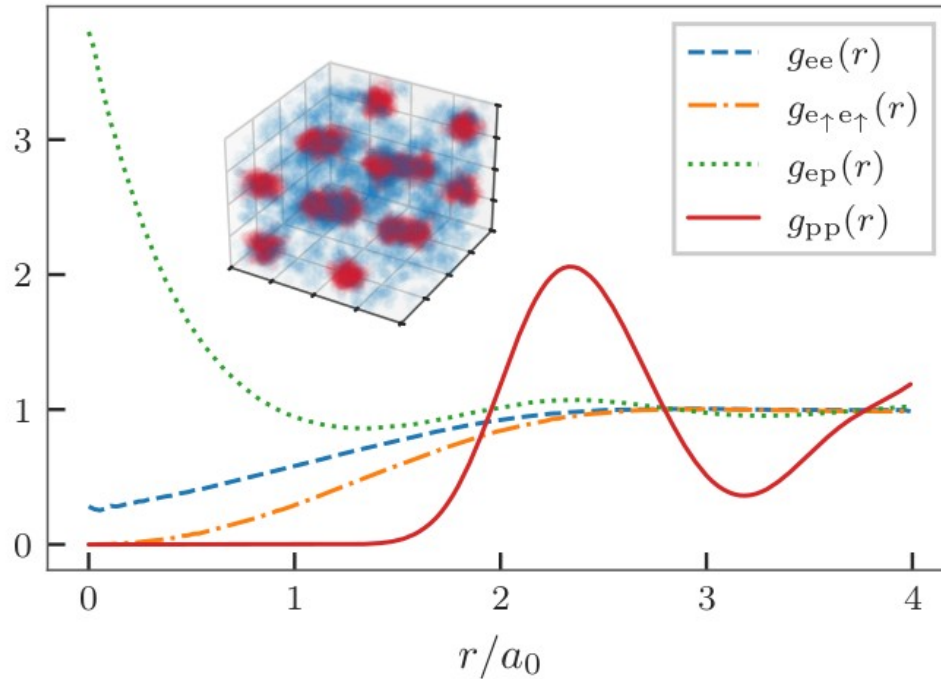


Figure 2. Four pair correlation functions are shown for $N = 54$ between three different types of particles: “e”, “p” and “e $\uparrow$ ”, corresponding to electrons, protons and spin-up electrons respectively. An inset with 300 Monte Carlo configurations in the BCC crystal is depicted (for $N = 16$, for clarity), where protons form the localized red balls while electrons form the delocalized blue cloud.

Pressure induced melting

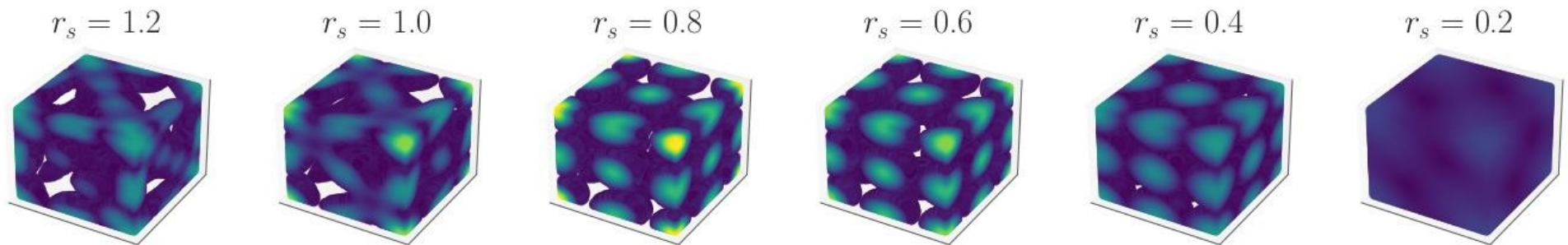


Figure 3. Pair correlation function for $N = 8$ protons at different densities under PBC. The same color scale is used in all subplots, globally adjusted with a power law normalization to enhance visibility of the structural features. A fixed threshold is manually set to prevent plotting areas where the pair correlation function has low values. At $r_s = 0.2$, the structure is drastically reduced suggesting that the system turned liquid.

Exploring atomic structures: Simple force and pressure estimators

D. Linteau, S. Moroni, G. Carleo, M. H., cond-mat/2603.14521

- Use expected value for observable $O(\mathbf{r})$
$$\tilde{O}(\mathbf{r}_i, \mathbf{r}_f) = aO(\mathbf{r}_f) + (1 - a)O(\mathbf{r}_i)$$

With Metropolis acceptance of Markov chain

$$a \equiv a(\mathbf{r}_i \rightarrow \mathbf{r}_f) = \min \left(1, \frac{\pi(\mathbf{r}_f)p(\mathbf{r}_i|\mathbf{r}_f)}{\pi(\mathbf{r}_i)p(\mathbf{r}_f|\mathbf{r}_i)} \right)$$

→ Unbiased, but reduced variance in general

$$\mathbb{E} [\tilde{O}(\mathbf{r}_i, \mathbf{r}_f)] = \langle O \rangle \quad \text{Var} [\tilde{O}(\mathbf{r}_i, \mathbf{r}_f)] \leq \langle O^2 \rangle - \langle O \rangle^2$$

- For general parameter optimization of wfn: energy gradient (or Pulay force term):

$$\frac{\partial E(p)}{\partial p} = 2\text{Re} \left\langle [E_L(\mathbf{r}) - E(p)] \partial_p \log \Psi^*(\mathbf{r}|p) \right\rangle$$

- Divergence of variance due to wfn nodes reduced to logarithmic

See also Z.-Q. Wan, R. Wiersema, S. Shang, arXiv:2603.18148
“blurred sampling”

Various reduction for “Pulay-forces”

- Toy model for Pulay-force:

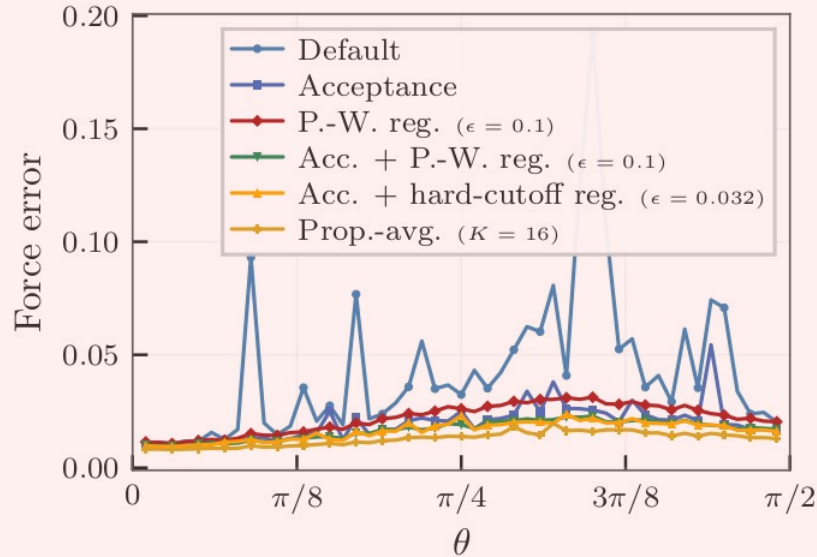
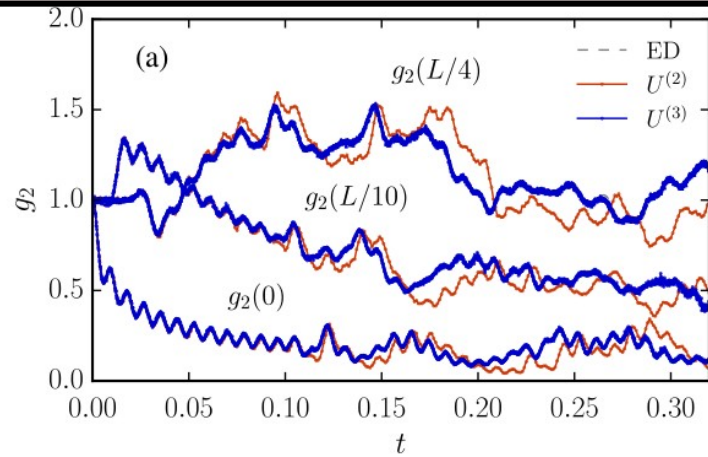


Figure 7. Statistical error of the toy-model force F_θ as a function of θ for the Gaussian trial state $\psi_\theta(x, y) = e^{-2x^2 - y^2}(x \sin \theta + y \cos \theta)$. The curves compare the default, acceptance-weighted (or “Acc.” for short) [Eq. (28)], finite- K proposal-averaged (or Prop.-avg.) [Eq. (32)], hard-cutoff regularized (or “reg.”) [Eq. (34)] estimators, as well as the extrapolation scheme from Pathak and Wagner³⁴ (denoted “P.-W.”).



- Time-dependent VMC benchmark:

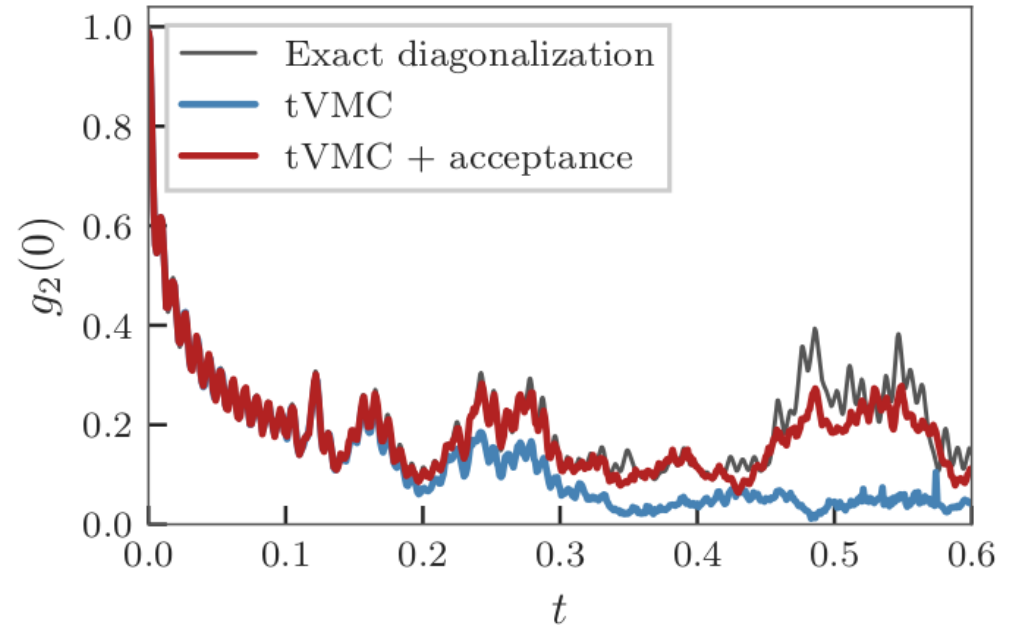


Figure 8. Time evolution of the double occupancy $g_2(r=0)$ for a quench of $N=3$ particles on a one-dimensional lattice with 40 sites, from the non-interacting state at $t=0$ to an on-site interaction strength $g=8$. The curves compare bare tVMC, tVMC using the acceptance trick [Eq. (28)] in the equations of motion, and the exact diagonalization benchmark.

T-VMC benchmark 1D hard-core bosons, $N=3$ quench with generalized 3body as in appendix of

G. Carleo, L. Cevolani, L. Sanchez-Palencia, and M. H., Phys. Rev. X 7, 031026 (2017)

Beyond ground state energies:

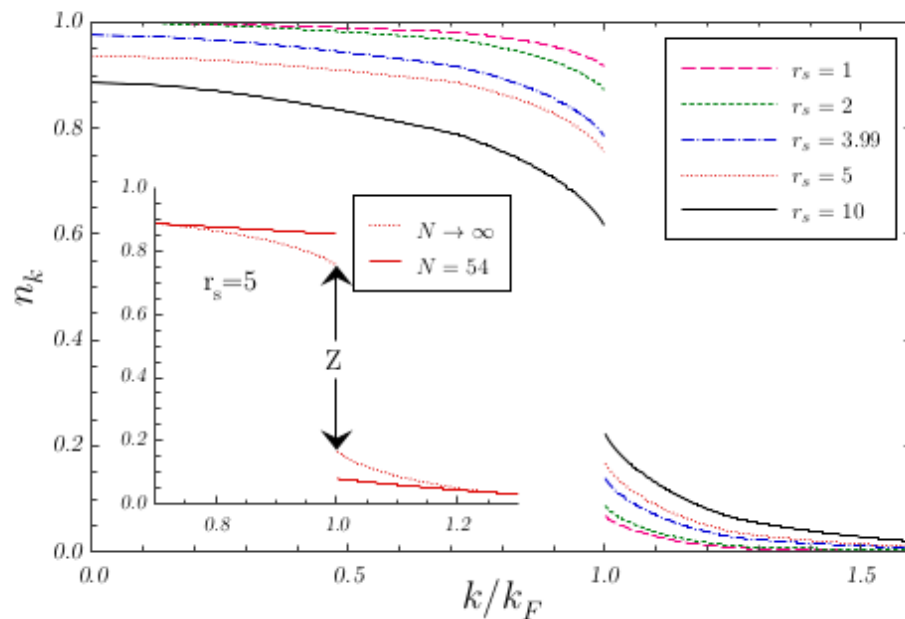
- Quantum Monte Carlo calculations are based on variational principle
 - Energy is true upper bound, but other (ground) state properties are accessible: pair correlation functions, static structure factor give structural information

Momentum distribution gives complementary information:
Extended or localized electronic states, Fermi liquid behavior?

- Extensions to excited state properties:
effective mass, gaps?
- (New) difficulties for computing spectral properties:
 - QMC extensions for excited states to guarantee orthogonality
 - Extended systems/thermodynamic limit: continuous spectrum: important finite size effects may be expected, transition from discrete to continuous spectra might not be straightforward

$n(k)$, Z of jellium (3DEG) at various densities

M.H., B. Bernu, C. Pierleoni, J. McMinis, D.M. Ceperley, V. Olevano, L. Delle Site, PRL **107**, 110402 (2011) (2011).



from analytical expressions of Ψ :
exact behavior of n_k around k_F

Quantitative agreement of QMC with G_0W_0 over broad density region $1 \leq r_s \leq 5 \dots 10$

r_s	1	2	3.99	5	10
BF-RMC	0.84(2)	0.77(1)	0.64(1)	0.58(1)	0.40(1)
SJ-VMC	0.894(9)	0.82(1)	0.69(1)	0.61(2)	0.45(1)
BF-VMC	0.86(1)	0.78(1)	0.65(1)	0.59(1)	0.41(1)
G_0W_0 [25]	0.859	0.768	0.646*	0.602	0.45
GW_0 [26]		0.804	0.702*		
GW [27]		0.846	0.793*		
Lam [28]	0.896	0.814	0.615*	0.472	
RPA [28]	0.843	0.700	0.442*	0.323	
SJ-DMC [6]	0.952	0.889		0.725	0.593

[25] L. Hedin, Phys.Rev. **139**, A796 (1965).

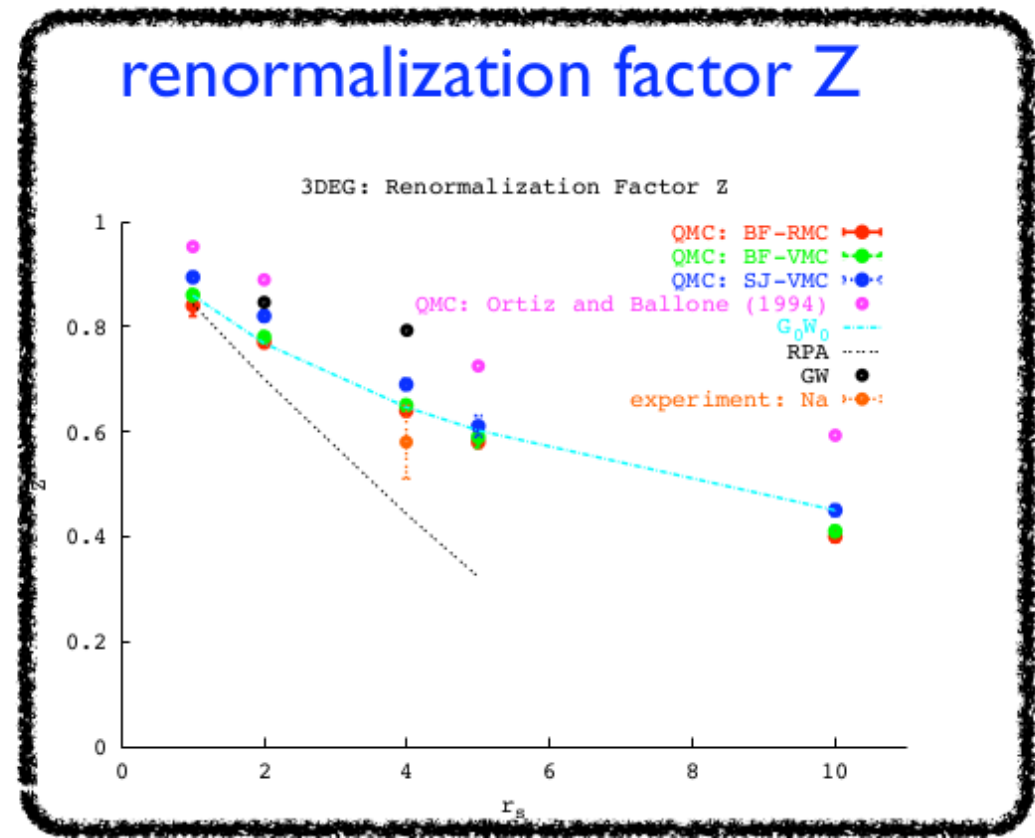
[26] U. von Barth, B. Holm, PRB **54**, 8411 (1996).

[27] B. Holm, U. von Barth, PRB **57**, 2108 (1998).

[28] J. Lam, PRB **3**, 3243 (1971).

[6] G. Ortiz, P. Ballone, PRB **50**, 1391 (1994).

renormalization factor Z



Momentum distribution and Z: Experiment vs QMC

solid Na ($r_s=3.99$)

S. Huotari, J. A. Soininen, T. Pylkkänen, A. Titov, A. Issolah, K. Hämäläinen, J. McMinis, J. Kim, K. Esler, D.M. Ceperley, M. H., V. Olevano, Phys. Rev. Lett. 105, 086403 (2010).

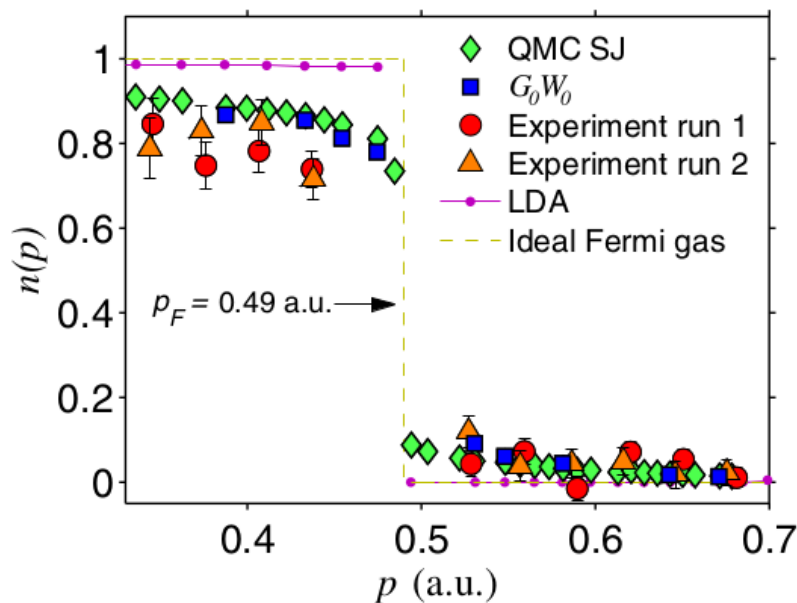


FIG. 1 (color online). The momentum distribution of Na determined by experiment, QMC SJ, G_0W_0 , and LDA calculations. The ideal-Fermi gas step function is also shown.

Li: Use of **Pseudo-potentials** to eliminate core electrons gives systematic (small) deviations: **all electron calculations needed!**

solid and liquid Li ($r_s=3.25$)

Yang, Hiraoka, Matsuda, M. H., Ceperley, Phys. Rev. B 101, 165125 (2020);
Hiraoka, Yang, Hagiya, Niozu, Matsuda, Huotari, M. H., Ceperley, Phys. Rev. B 101, 165124 (2020).

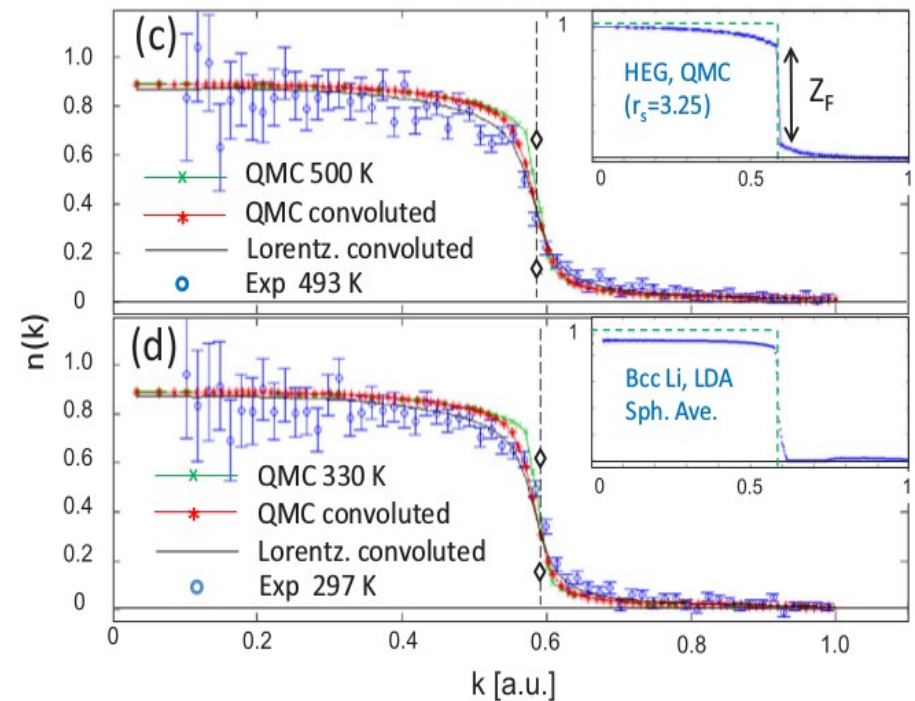


FIG. 2. Valence CPs of liquid (a) and solid Li (b), compared with theory convoluted by the broadening function. The insets show the difference between the experimental and theoretical CPs. EMDs in liquid (c) and solid Li (d), compared to theory with or without the convolutions. The insets show EMD for HEG and the spherically averaged EMD from band theory (LDA). The $\diamond$'s in (c) and (d) show the $n(k_F)$ obtained by RPA fits near k_F .

Microscopic Definition of m^*

Def. from single particle
Green's function

$$\frac{m}{m^*} = \frac{1 + \frac{m}{k_F} \frac{\partial \Sigma(k_F, \mu)}{\partial k}}{1 - \frac{\partial \Sigma(k_F, \mu)}{\partial \omega}}$$

Fermi liquids have jump Z in
momentum distribution:

$$Z = \frac{1}{1 - \frac{\partial \Sigma(k_F, \mu)}{\partial \omega}}$$

✗ we calculate Z within QMC

□ how to calculate k -derivative of self-energy? $\frac{\partial \Sigma(k_F, \mu)}{\partial k}$

↳ note that $\Sigma(k, \mu)$ is a **static** quantity

and $G(k, \mu)$ is a **static response** function!

Static self-energy

M.H., F. Calcavecchia, D.M. Ceperley, V. Olevano, PRL 131, 186501 (2023) (2023)

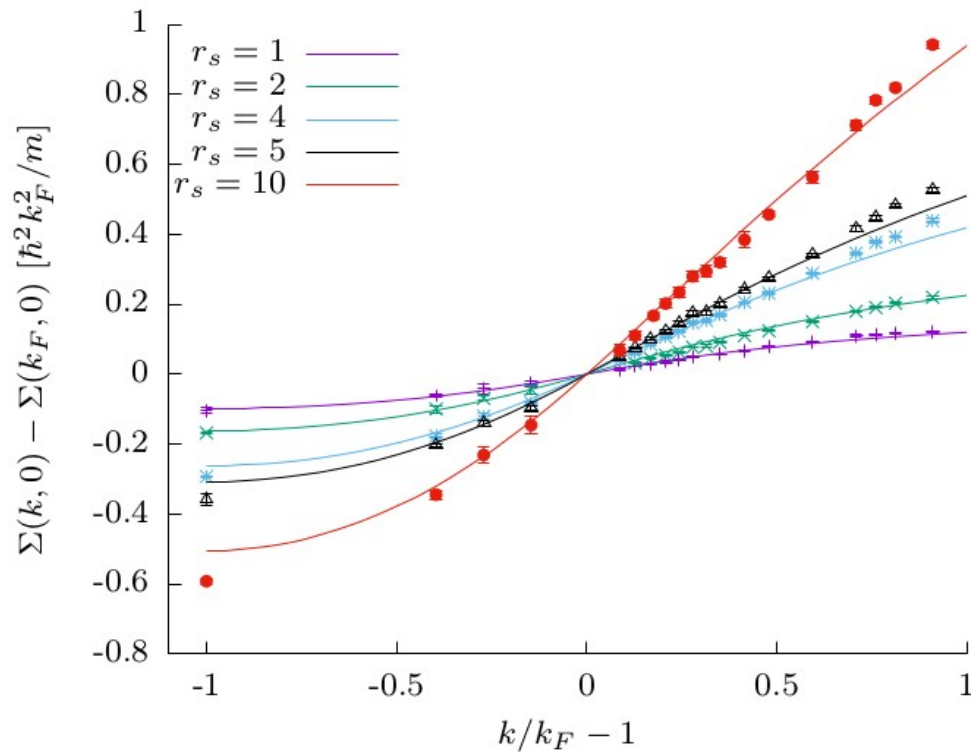


FIG. 1: Static self-energy for various densities (r_s) using back-flow (BF) trial wave functions and GC-TABC simulations for $N = 38$ electrons. They include size corrections. The color lines are from G_0W_0 calculations.

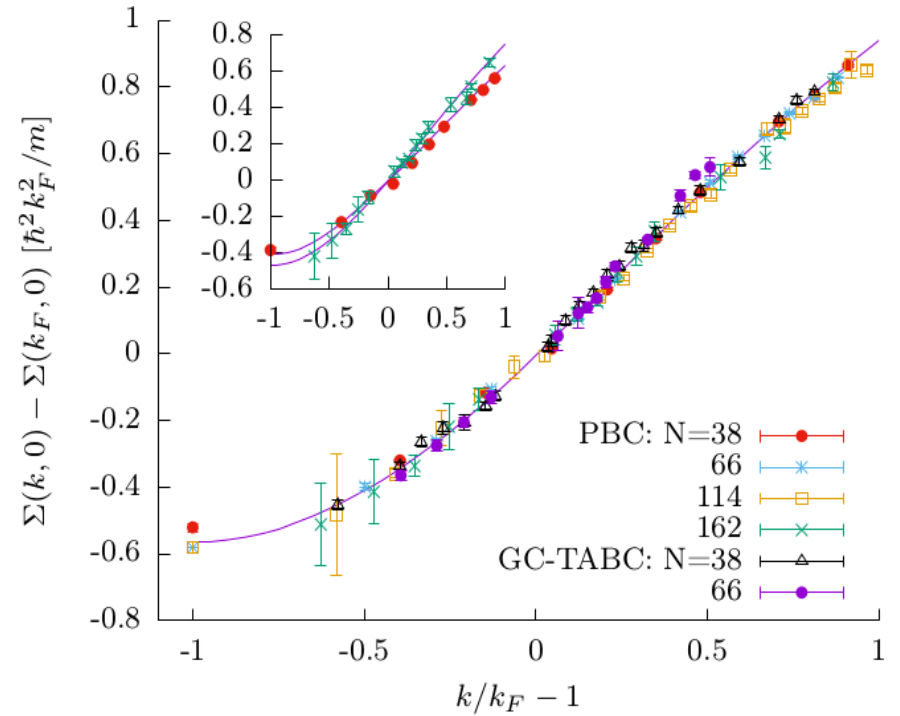


FIG. 2: Static self-energy for $r_s = 10$ using SJ-VMC trial wave functions for simulations with periodic boundary conditions (PBC) and GC-TABC for various sizes ranging from $N = 38$ to $N = 162$, size corrected according to Eq. (17), the line is a fit to the data. The inset shows the uncorrected values for $N = 38$ and $N = 162$ (PBC), the lines indicate the size corrections of the fit based on Eq. (17).

Z + static self-energy = m*

M.H., F. Calcavecchia, D.M. Ceperley, V. Olevano, PRL 131, 186501 (2023)

r_s	method	Z	$mk_F^{-1}\partial_k\Sigma$	m^*/m
1	BF-VMC	0.86(1) [13]	$0.17_{0.15}^{0.18}(1)$	$1.00_{0.99}^{1.01}(1)$
	SJ-VMC	0.894(9) [13]	$0.17_{0.15}^{0.18}(1)$	$0.96_{0.95}^{0.97}(1)$
	G_0W_0 (RPA)	0.859 [31]	0.200	0.970 [10]
	VDiagMC [12]	0.8725(2)	0.200(1)	0.955(1)
2	BF-VMC	0.78(1)	$0.309_{0.280}^{0.361}(6)$	$0.98_{0.94}^{1.00}(1)$
	SJ-VMC	0.82(1)	$0.30_{0.28}^{0.31}(2)$	$0.94_{0.93}^{0.95}(2)$
	G_0W_0 (RPA)	0.768 [31]	0.313	0.992 [10]
	VDiagMC [12]	0.7984(2)	0.328(4)	0.943(3)
4	BF-VMC	0.65(1)	$0.538_{0.530}^{0.549}(7)$	$1.00_{0.99}^{1.01}(2)$
	SJ-VMC	0.69(1)	$0.55_{0.45}^{0.65}(2)$	$0.94^{1.00}(2)$
	G_0W_0 (RPA)	0.646 [31]	0.490	1.039 [10]
	VDiagMC [12]	0.6571(2)	0.528(5)	0.996(3)
5	BF-VMC	0.59(1)	$0.56^{0.65}(1)$	$1.09_{1.03}(3)$
	SJ-VMC	0.61(1)	$0.610_{0.596}^{0.624}(9)$	$1.02_{1.01}^{1.03}(2)$
	G_0W_0 (RPA)	0.602 [31]	0.569	1.059 [10]
10	BF-VMC	0.41(1)	$0.90_{0.88}^{0.98}(2)$	$1.28_{1.23}^{1.30}(3)$
	SJ-VMC	0.45(1)	$0.97_{0.91}^{1.03}(3)$	$1.13_{1.09}^{1.16}(3)$
	G_0W_0 (RPA)	0.45 [31]	0.98	1.13

TABLE I: Our QMC results with backflow (BF) and Slater-Jastrow (SJ) trial functions as compared to those of G_0W_0 (RPA) and variational diagrammatic Monte Carlo (VDiagMC) [12] [32] [33]. Upper and low indices indicate systematic errors assuming different fitting functions/ranges to determine $\partial_k\Sigma(k_F, 0)$.

Summary - Conclusions:

- ⇒ Improvement of ground state wave functions:
iterated backflow, machine learned network ansätze...
- ⇒ Helium atoms: 2D liquid-solid: first order transition....
Hexatic, supersolid phase only for small systems ($N=30$)
- ⇒ Hydrogren under pressure: ground state, gaps,...
- ⇒ Fermi Liquid parameters: Z , m^*

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