Backflow corrections in QMC

Going beyond the Slater-Jastrow wave function in fermionic systems

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Backflow corrections in QMC

What are trial wave functions used for in QMC?

1. VMC: trial wave functions are used to evaluate the energy:

$$E_{VMC} = \frac{1}{N} \sum_{i=1}^{N} \frac{H \Psi_T(\boldsymbol{R}_i)}{\Psi_T(\boldsymbol{R}_i)}$$

Quality of E_{VMC} **determined by quality of** Ψ_T

2. VMC allows for optimization of Ψ_T**, as**

$$E_0 \leq E_{VMC} = \frac{\langle \Psi_T | H | \Psi_T \rangle}{\langle \Psi_T | \Psi_T \rangle}$$

Backflow corrections in QMC

What are trial wave functions used for in QMC?

3. DMC: high energy components of Ψ_T are projected out using iTDSE

If DMC unconstrained, Ψ_T 'unwraps' into the bosonic ground state

The fixed-node approximation (FNA) cures this by preventing the fermions from crossing the nodes of Ψ_T

Quality of $E_{FN\text{-}DMC}$ determined by quality of the nodal surface of Ψ_T

Backflow corrections in QMC

Which trial wave functions are used in QMC?

1. Hartree-Fock

$$\Psi_{HF} = D_{\uparrow}(\boldsymbol{R}_{\uparrow}) D_{\downarrow}(\boldsymbol{R}_{\downarrow}) \quad ; \quad D_{\sigma}(\boldsymbol{r}_1^{\sigma}, \ldots, \boldsymbol{r}_{N_\sigma}^{\sigma}) = \begin{vmatrix} \phi_1^{\sigma}(\boldsymbol{r}_1^{\sigma}) & \phi_2^{\sigma}(\boldsymbol{r}_1^{\sigma}) & \vdots & \phi_{N_\sigma}^{\sigma}(\boldsymbol{r}_1^{\sigma}) \\ \phi_1^{\sigma}(\boldsymbol{r}_2^{\sigma}) & \phi_2^{\sigma}(\boldsymbol{r}_2^{\sigma}) & \vdots & \phi_{N_\sigma}^{\sigma}(\boldsymbol{r}_2^{\sigma}) \\ \cdots & \cdots & \ddots & \cdots \\ \phi_1^{\sigma}(\boldsymbol{r}_{N_\sigma}^{\sigma}) & \phi_2^{\sigma}(\boldsymbol{r}_{N_\sigma}^{\sigma}) & \vdots & \phi_{N_\sigma}^{\sigma}(\boldsymbol{r}_{N_\sigma}^{\sigma}) \end{vmatrix}$$

*** Correct anti-symmetry (exchange)**
*** Constructed from one-particle orbitals**

*** No correlations taken into account (except multi-determinants)**
*** Local energy diverges when two particles coalesce**

Backflow corrections in QMC

Which trial wave functions are used in QMC?

2. Slater-Jastrow

$$\Psi_{SJ} = e^{J(\boldsymbol{R})} \Psi_S(\boldsymbol{R}) \quad ; \qquad J(\boldsymbol{R}) = J_{e-e}(\boldsymbol{R}) + J_{e-N}(\boldsymbol{R}) + J_{e-e-N}(\boldsymbol{R}) + \ldots$$

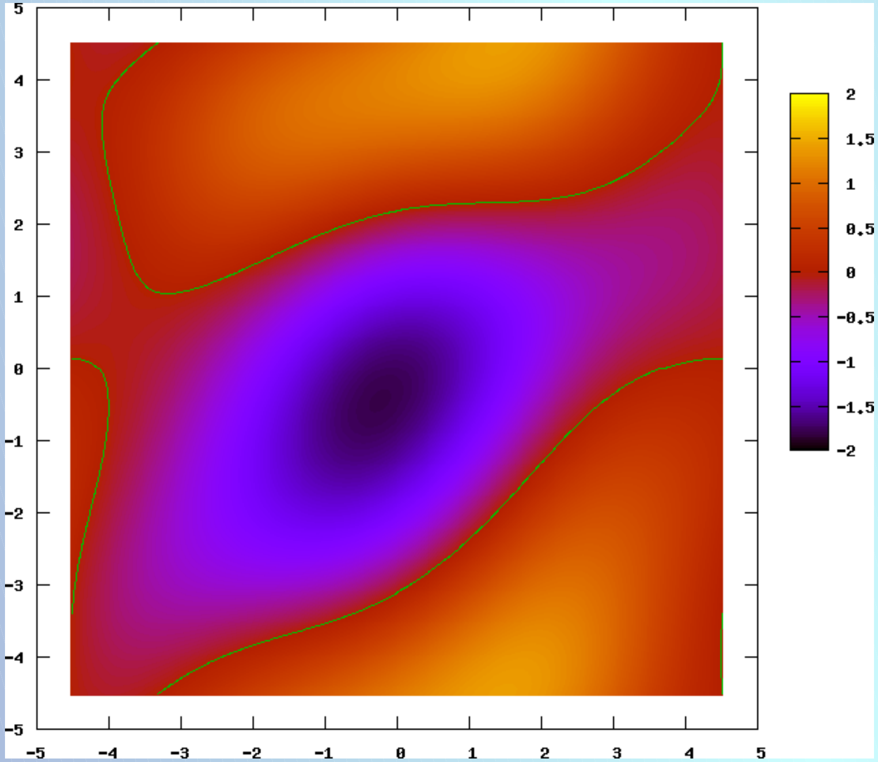
* Ability to introduce arbitrary correlations (2-body, 3-body, ...)
* Ability to remove divergencies: stable calculations
* Well-known functional forms

* Inability to modify the nodal surface

Backflow corrections in QMC

The Slater-Jastrow wave function

1. What is the effect of exp(J) on the HF wave function?



26-ELECTRON 2D HEG AT RS=1.0
MOVING ONE ELECTRON WITH THE REST FIXED.

Backflow corrections in QMC

The Slater-Jastrow wave function

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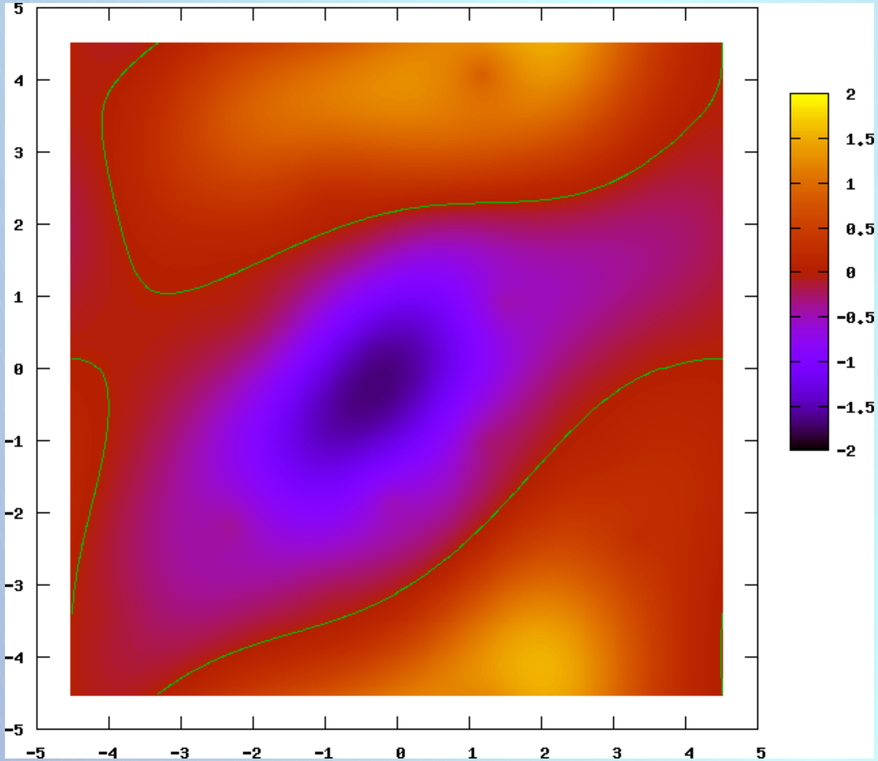

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Backflow corrections in QMC

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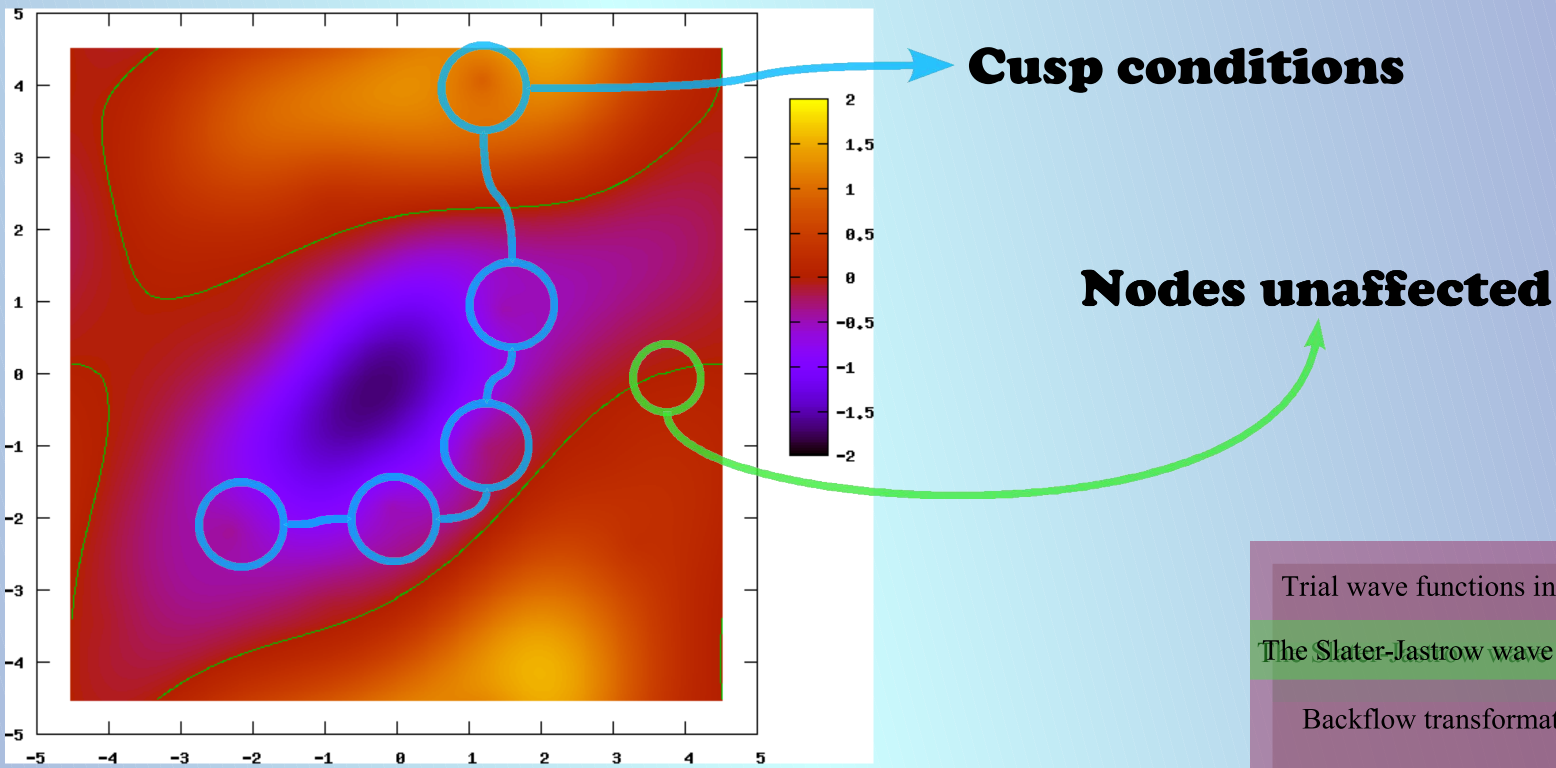


26-electron 2D HEG at rs=1.0
Moving one electron with the rest fixed.

Backflow corrections in QMC

The Slater-Jastrow wave function

2. What is the effect of exp(J) on the energy?

	Energy (au)	σ^2 (au)	CE (%)
HF	0.5694(5)	19.3(6)	0.0
J=U(minimal)	0.5346(1)	1.61(3)	88.0(5)
J=U	0.5332(1)	1.22(1)	91.5(5)
J=U+P	0.53201(9)	0.72(1)	94.5(4)
Best (DMC)	0.52985(5)	N/A	100.0

3D 54-electron HEG at rs=1.0
VMC results using 30,000 configurations

Backflow corrections in QMC

Backflow transformations

1. History of backflow

Backflow formulated:
Feynman, Cohen, PR 102, 1189 (1956)

Backflow first used in QMC:
Panoff, Carlson, PRL 62, 1130 (1989)

Backflow first applied to electronic systems:
Kwon, Ceperley, Martin, PRB 48, 12037 (1993)

Backflow applied to inhomogeneous systems:
Present work

Backflow transformations

2. What is backflow?

First introduced to conserve particle current in trial WFs

the current at a point $\mathbf{a}$ is $\mathbf{j}(\mathbf{a})=\hbar m^{-1}|h(\mathbf{a})|^2$. The wave function (11) therefore leads to the picture of a total current $\hbar\mathbf{k}m^{-1}$ (assume $\int|h(\mathbf{a})|^2 d\mathbf{a}=1$) distributed over a small region and having everywhere the same direction, with no appreciable change in the number density anywhere. Such a picture clearly cannot represent anything like a stationary state, since in a stationary state the current is divergence-free and there would necessarily be a return flow directed oppositely to $\mathbf{k}$.

One way to incorporate such a backflow into (11) is to multiply the wave function by $\exp[i\sum g(\mathbf{r}_i)]$, obtaining

$$\psi=\varphi\exp[i\sum g(\mathbf{r}_i)]\sum h(\mathbf{r}_i)\exp(i\mathbf{k}\cdot\mathbf{r}_i). \quad (12)$$

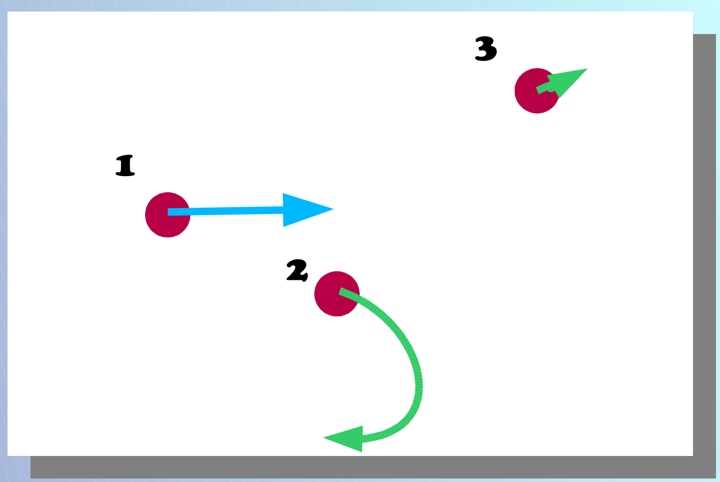
Application of the velocity operator $-i\hbar m^{-1}\nabla_i$ shows

"Energy spectrum of excitations in liquid Helium"
R. P. Feynman, M. Cohen, Phys. Rev. 102, 1189 (1956)

Backflow transformations

2. What is backflow?

A set of collective coordinates $\{x_i(R)\}$ is defined so that the resulting quasi-particles "avoid" each other:



Dynamical view of how quasi-particles ought to behave.
As 1 moves, 2 clears the way, while 3 barely notices

Backflow corrections in QMC

Backflow transformations

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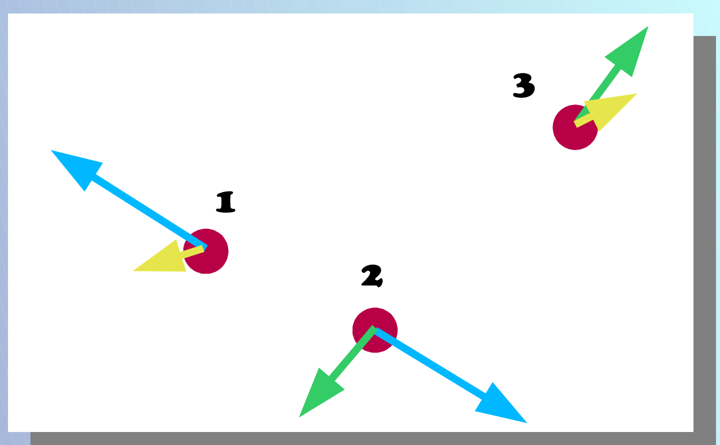


Static view

$$x_i = r_i + \sum_{j \neq i}^{N} \eta(r_{ij})\, r_{ij}$$

Backflow expression

$$\xi_i = \sum_{j \neq i}^{N} \eta(r_{ij})\, r_{ij}$$

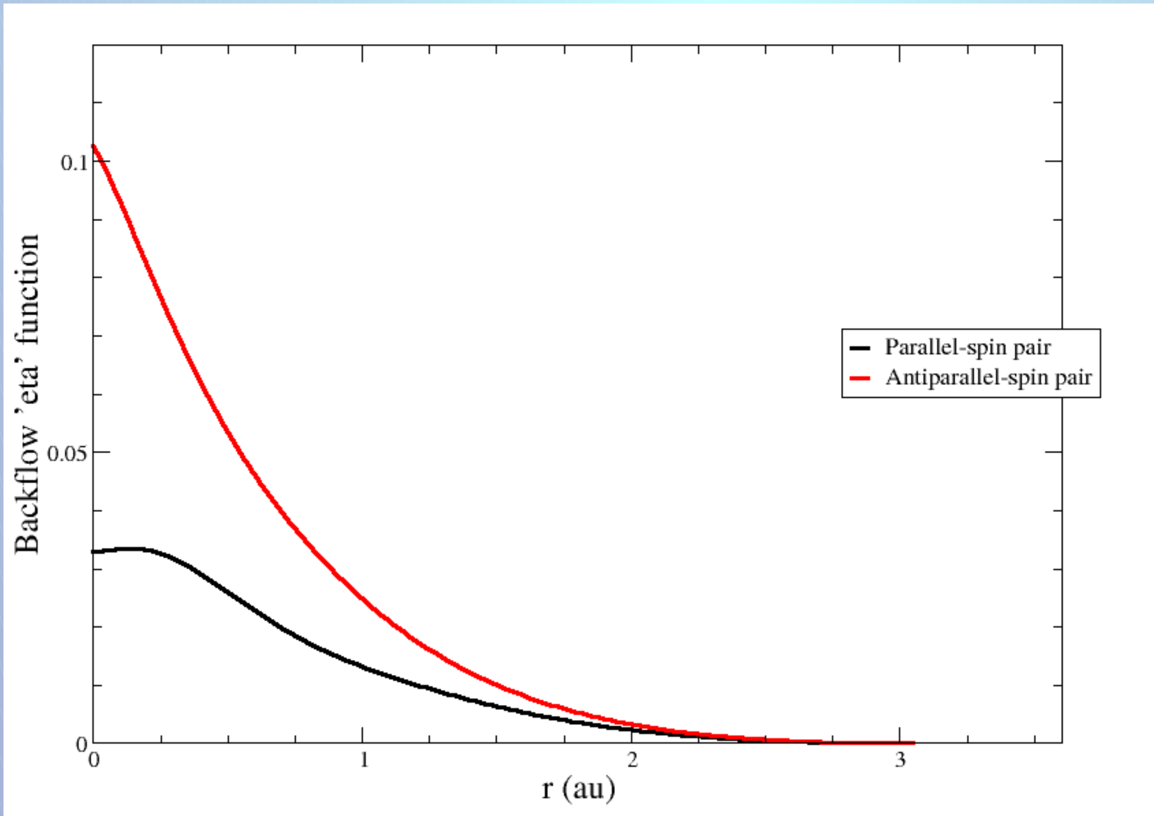
Backflow displacement: a vector field

Particle i sees a set of preferred directions, given by $\{\mathbf{r}_{ij}\}_{j \neq i}$

Pablo López Ríos

Backflow transformations

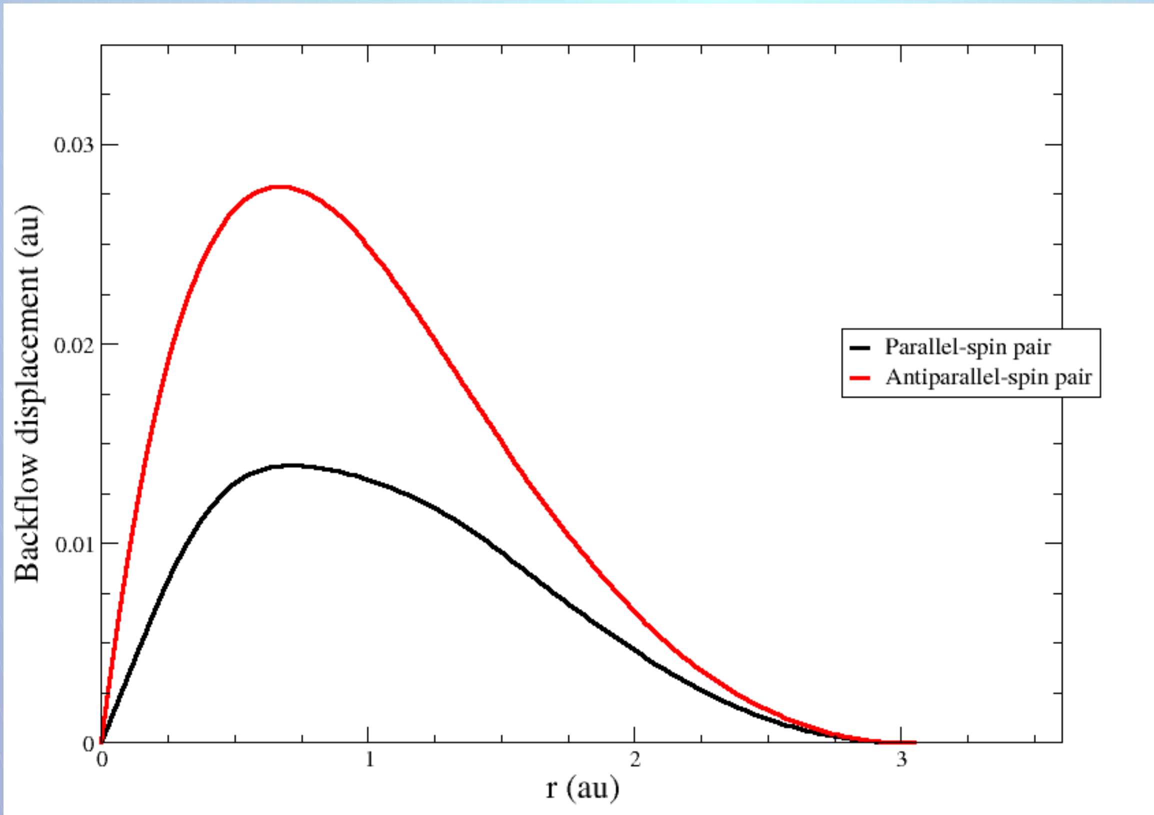
3. The backflow η(r) function



Backflow corrections in QMC

Backflow transformations

3. The backflow $\eta(r)$ function



Backflow corrections in QMC

Backflow transformations

4. Origin of backflow: derivations and justification

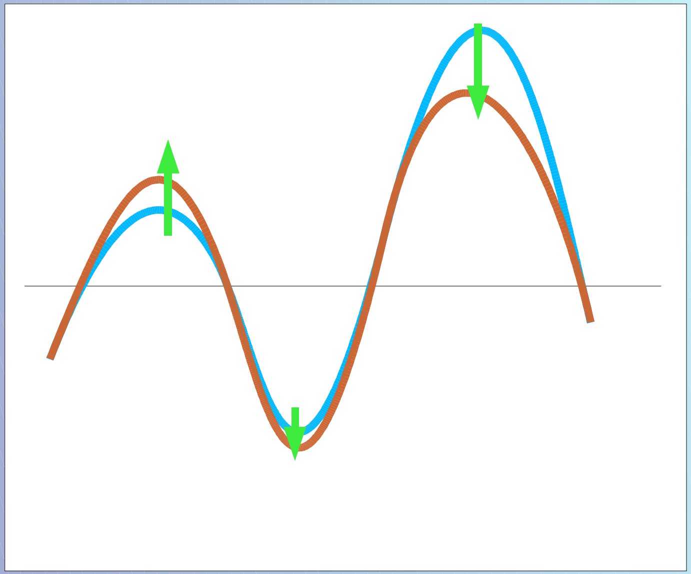
* **Feynman & Cohen: conservation of particle current**

* **Ceperley et al.: imaginary time propagation argument**

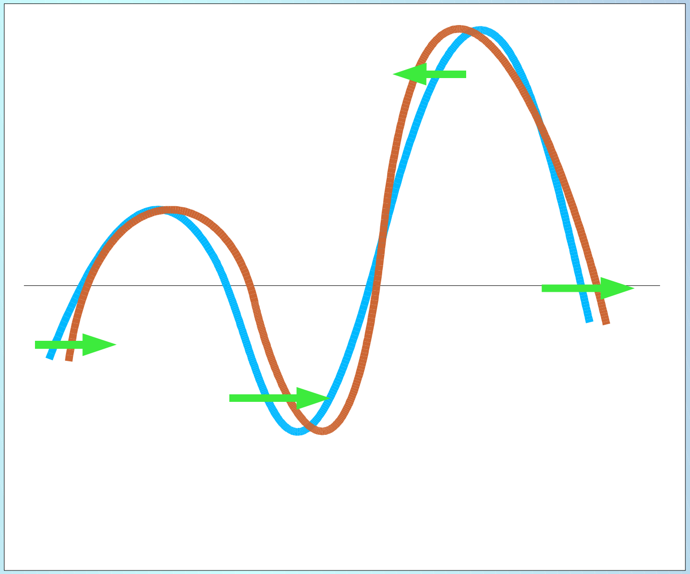
Backflow corrections in QMC

Backflow transformations

4. Origin of backflow: derivations and justification



Effect of a Jastrow factor



Complementary effect.

The most general way of doing this is given by backflow

Backflow corrections in QMC

Backflow transformations

5. Using backflow in QMC

To avoid interference between Jastrow & backflow, use:

$$\Psi_{BF} = e^{J(\boldsymbol{R})} \Psi_S(\boldsymbol{X})$$

Parametrize the backflow $\eta(r_{ij})$ **function appropriately:**

$$\eta(r) = \lambda \frac{1 + s r}{\rho + \omega r + r^{\alpha}}$$

Rational form. Kwon et al. (1993)

$$\eta(r) = \lambda \exp\left[-\left(\frac{r-\rho}{\omega} \right)^2 \right]$$

Gaussian form. Holzmann et al. (2003)

$$\eta(r) = \sum_{m=0}^{N_{\exp}} c_m r^m$$

Polynomial form. Present work

Backflow corrections in QMC

Backflow transformations

5. Using backflow in QMC

Must:

*** Smoothly truncate the backflow function**

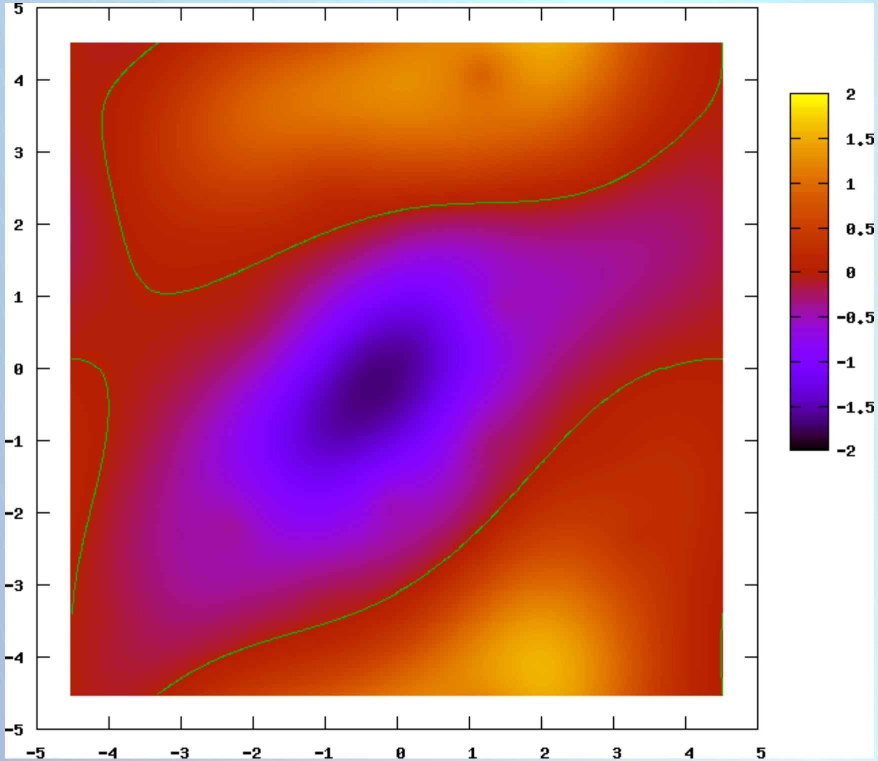
*** Not use single-electron update algorithms (hence scales as N^4)**

*** Constrain parameters so that cusp conditions are still verified**

*** Optimize Jastrow+backflow altogether**

Backflow transformations

6. What is the effect of backflow on the SJ wave function?

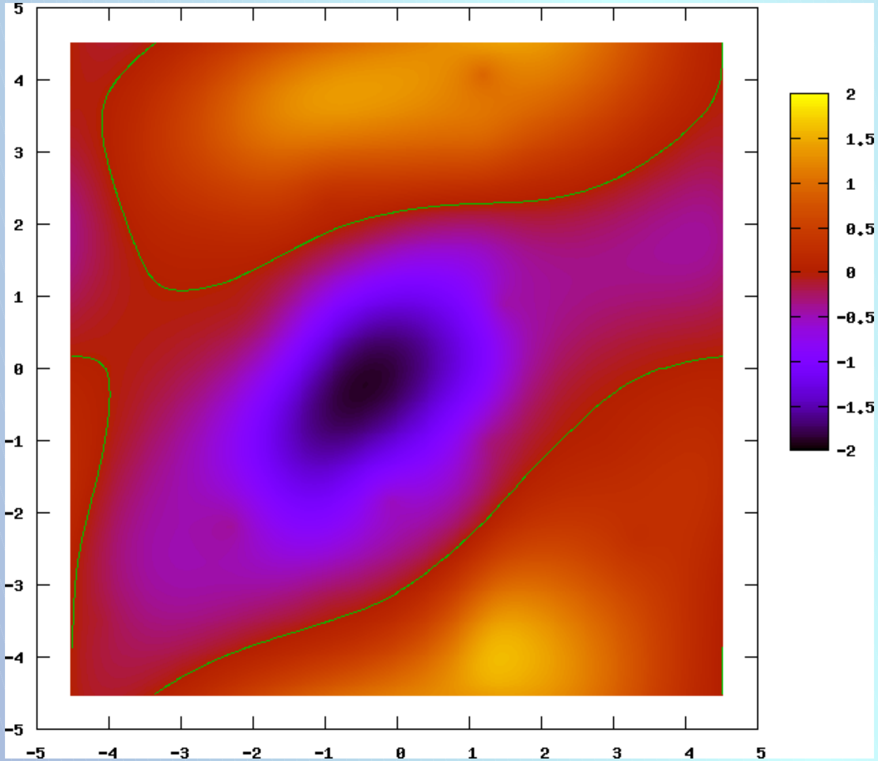


26-electron 2D HEG at rs=1.0
Moving one electron with the rest fixed.

Backflow corrections in QMC

Backflow transformations

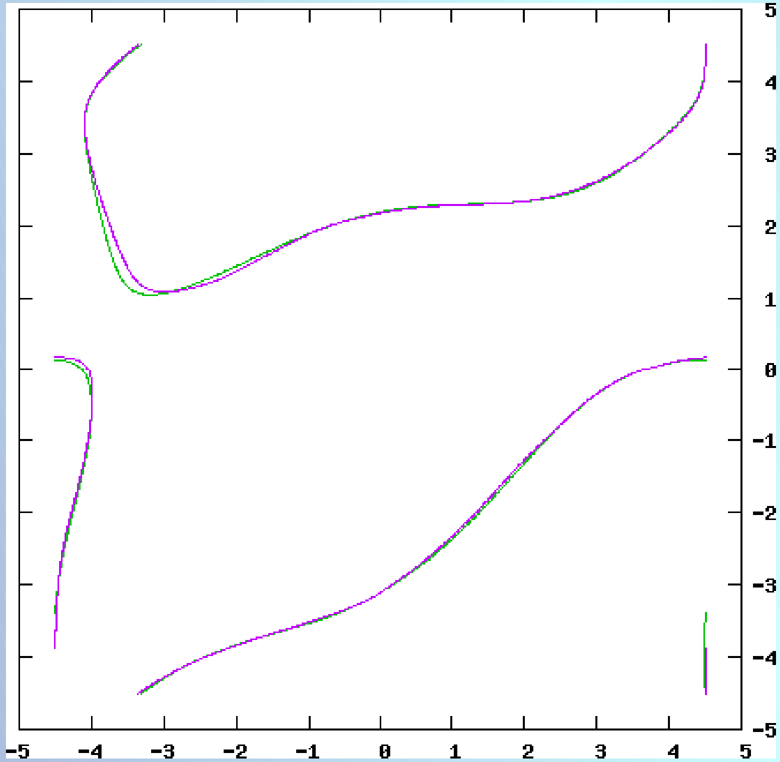
6. What is the effect of backflow on the SJ wave function?



26-electron 2D HEG at rs=1.0
Moving one electron with the rest fixed.

Backflow transformations

6. What is the effect of backflow on the SJ wave function?



26-electron 2D HEG at rs=1.0
SJ (green) and BF (purple) nodal surfaces

Backflow corrections in QMC

Backflow transformations

7. What is the effect of backflow on the energy?

	Minimal Jastrow			J=U			J=U+P		
	Energy (au)	σ^2 (au)	CE (%)	Energy (au)	σ^2 (au)	CE (%)	Energy (au)	σ^2 (au)	CE (%)
No backflow	0.5346(1)	1.61(3)	88.0(5)	0.5332(1)	1.22(1)	91.5(5)	0.53201(9)	0.72(1)	94.5(4)
Polynomial (8)	0.53186(9)	0.723(8)	94.9(4)	0.53171(9)	0.663(8)	95.3(4)	0.53014(4)	0.168(4)	99.3(2)
Gaussian	0.53194(9)	0.736(7)	94.7(4)	0.53151(8)	0.628(6)	95.8(4)	0.53034(5)	0.208(3)	98.8(3)*
Rational	0.53203(9)	0.769(8)	94.5(4)	0.53151(8)	0.625(7)	95.8(4)	0.53019(6)	0.171(3)	99.1(2)

Best (DMC)	0.52985(5)	N/A	100.0

3D 54-electron HEG at rs=1.0
VMC results using 30,000 configurations

Backflow corrections in QMC

Backflow transformations

8. Key concepts

* Backflow as a transformation complementary to the Jastrow

* There exists a set of preferred directions seen by each electron

* Backflow displacement expanded using the former

* Use of expansions (e.g. polynomials) advisable

Backflow corrections in QMC

Backflow in real systems

1. What do we mean by “real systems”?

Systems with inhomogeneous external potentials:

* **Systems with atoms (atoms, molecules, solids)**

* **Systems with arbitrary external potentials (infinite wells, harmonic potentials...)**

Backflow corrections in QMC

Backflow in real systems

2. How to apply the “key concepts”

a) Find set of “preferred directions” seen by each electron

b) Write backflow displacement as a vector field expanded in such set

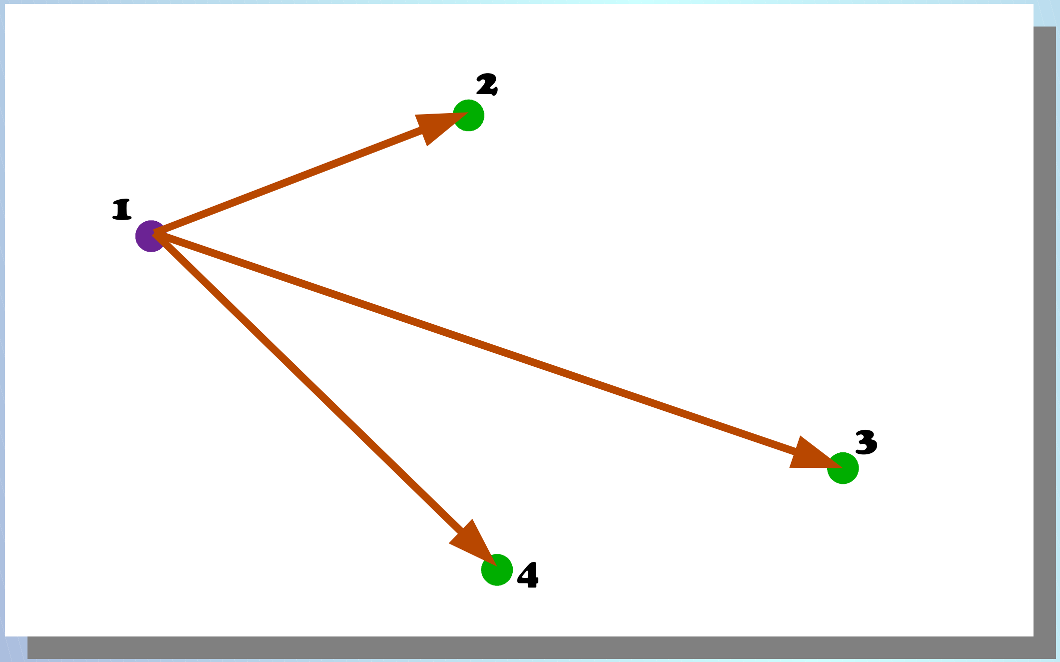
c) Use power expansions to parametrize

Backflow corrections in QMC

Backflow in real systems

2. How to apply the "key concepts"

a) Find set of "preferred directions" seen by each electron (systems with atoms)



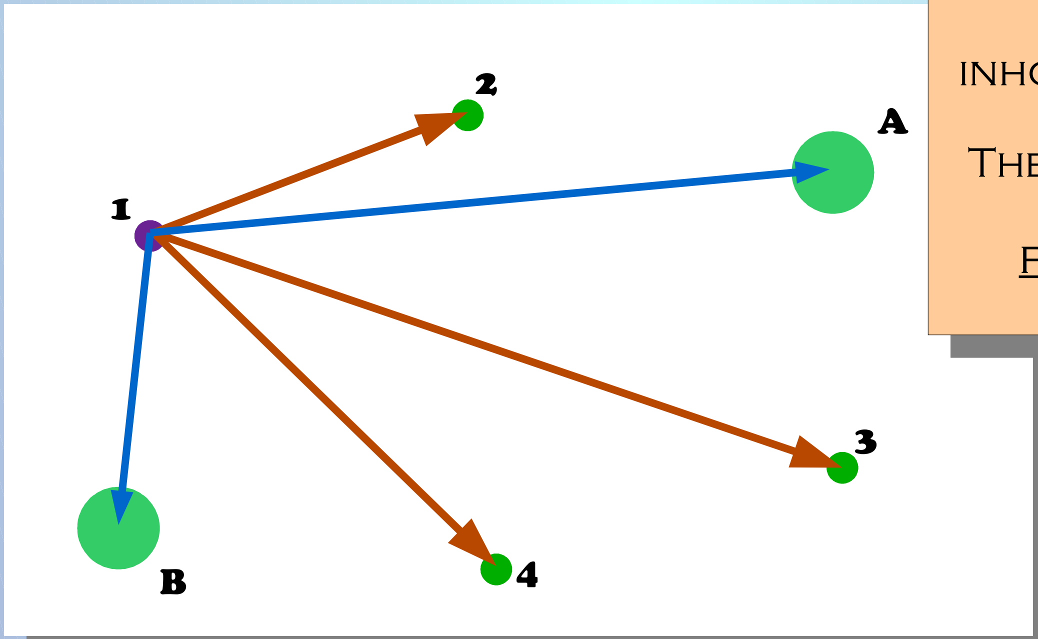
SET OF DIRECTIONS SEEN BY PARTICLE 1 IN THE PRESENCE OF PARTICLES 2, 3 AND 4

Backflow corrections in QMC

Backflow in real systems

2. How to apply the "key concepts"

a) Find set of "preferred directions" seen by each electron (systems with atoms)



Set of directions seen by particle 1 in the presence of particles 2, 3 and 4, and nuclei A and B

Key to other inhomogeneous systems:

These are the distances involved in the <u>potential energy</u>

Backflow corrections in QMC

Backflow in real systems

2. How to apply the "key concepts"

b) Write backflow displacement as a vector field expanded in such set

$$\xi_i = \xi_i^{e-e} + \xi_i^{e-N} + \xi_i^{e-e-N}$$

$$\xi_i^{e-e} = \sum_{j \neq i}^{N} \eta(r_{ij}) \mathbf{r}_{ij}$$

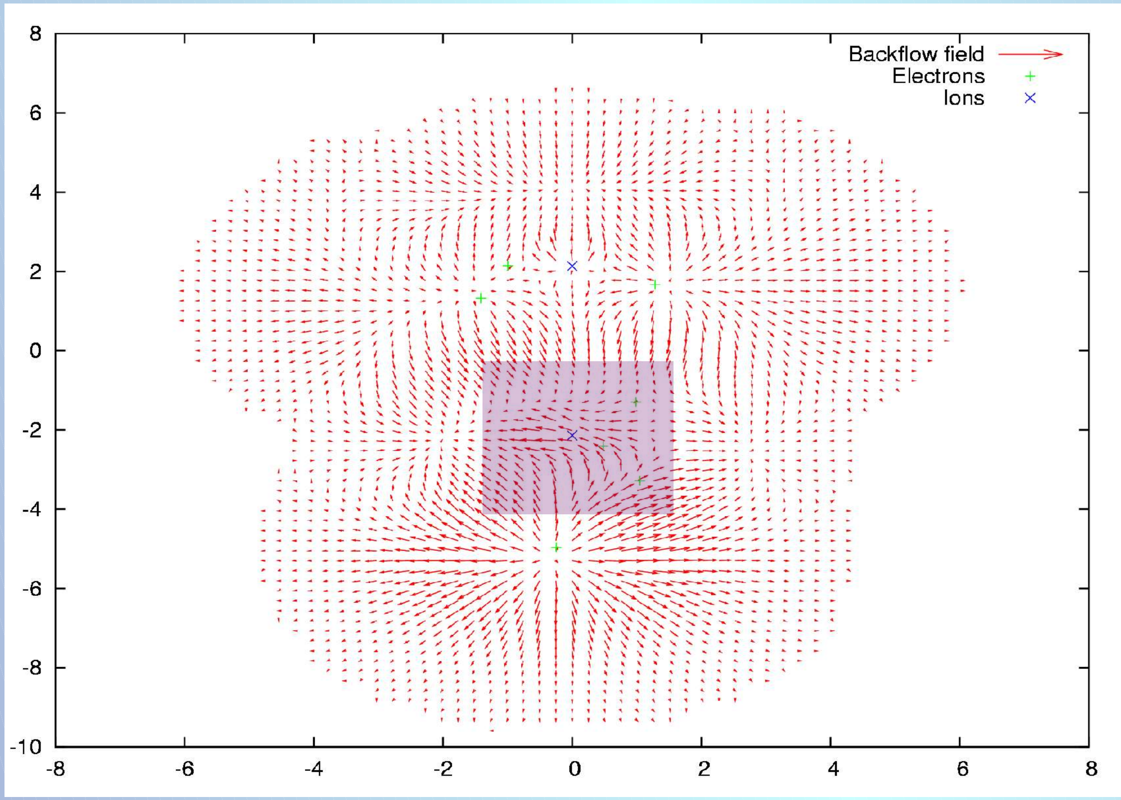
$$\xi_i^{e-N} = \sum_{I}^{N_{ion}} \mu(r_{iI}) \mathbf{r}_{iI}$$

$$\xi_i^{e-e-N} = \sum_{j \neq i}^{N} \sum_{I}^{N_{ion}} \left[\Phi(r_{ij}, r_{iI}, r_{jI}) \mathbf{r}_{ij} + \Theta(r_{ij}, r_{iI}, r_{jI}) \mathbf{r}_{iI} \right]$$

Backflow corrections in QMC

Backflow in real systems

3. What does inhomogeneous backflow look like?

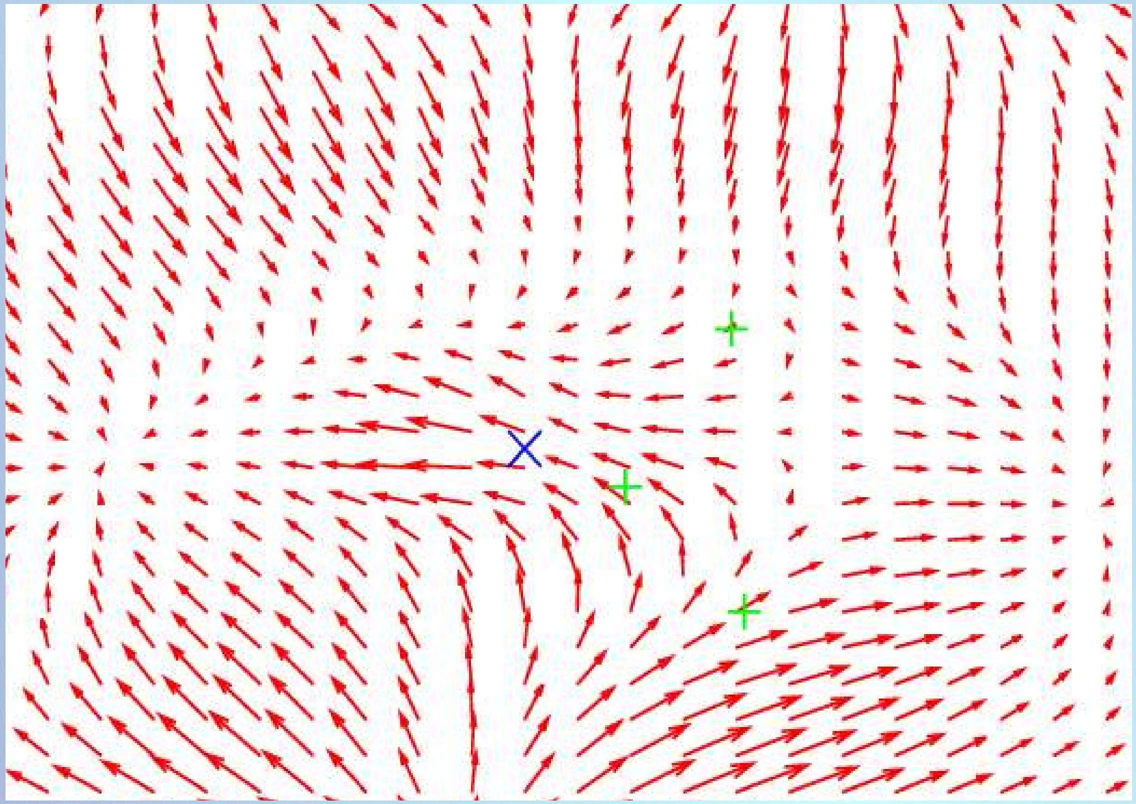


Backflow displacement field for Si_2 molecule (pseudo).
Moving one electron on the plane y=0 with the rest fixed

Backflow corrections in QMC

Backflow in real systems

3. What does inhomogeneous backflow look like?

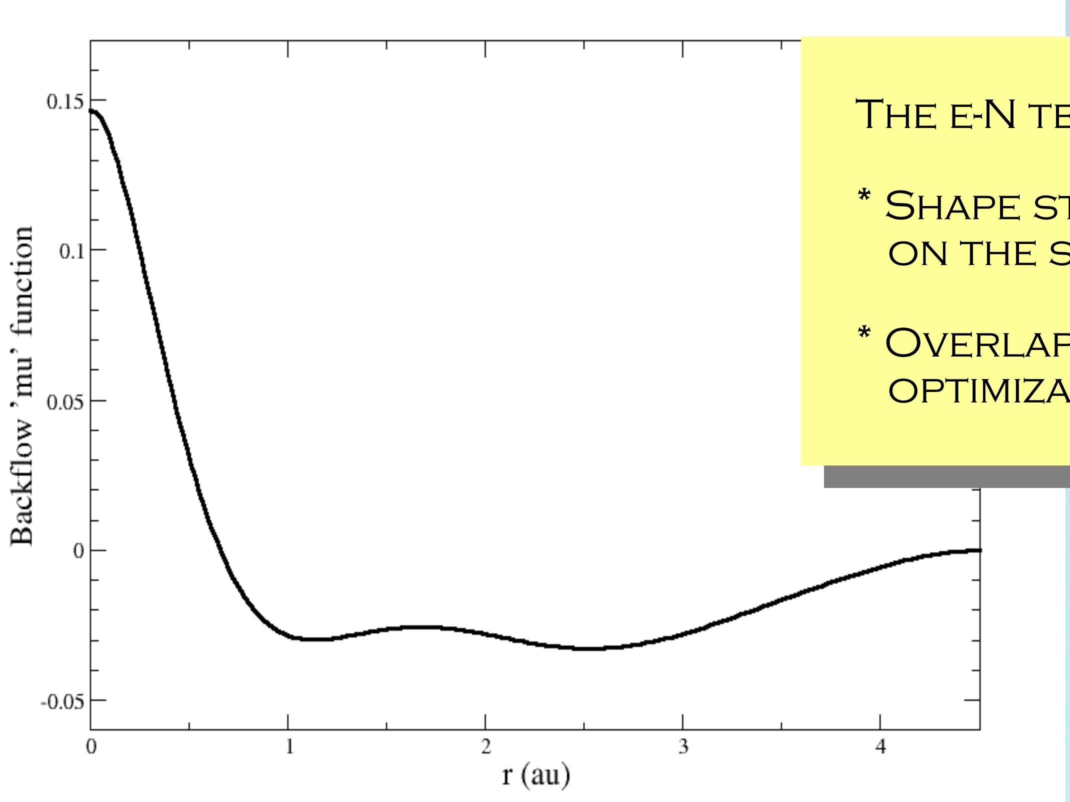


Close-up of previous figure

Backflow corrections in QMC

Backflow in real systems

3. What does inhomogeneous backflow look like?



The e-N term:

* Shape strongly depends on the system

* Overlaps with orbital optimization

Electron-Nucleus $\mu(r_{iI})$ backflow function for the Si_2 molecule

Backflow corrections in QMC

Backflow in real systems

4. Inhomogeneous backflow: limitations

* e-N cusp conditions are satisfied by the orbitals.
  3-body term has to be greatly constrained in systems with bare nuclei.

* Use of pseudo-potentials in QMC has some known issues.
  The ability to reduce the variance is limited.

Backflow in real systems

5. Inhomogeneous backflow: results

	Slater-Jastrow			Backflow		
	Energy (au)	σ^2 (au)	CE (%)	Energy (au)	σ^2 (au)	CE (%)
HF	-128.5471	N/A	0.0			
VMC	-128.898(3)	1.134(9)	89.9(8)	-128.919(2)	0.413(3)	95.2(5)
DMC	-128.9238(7)	N/A	96.5(2)	-128.928(1)	N/A	97.5(3)
Exact	-128.9376	N/A	100.0			

Results for all-electron Ne atom

Backflow in real systems

5. Inhomogeneous backflow: results

		Slater-Jastrow			Backflow		
		Energy (au)	σ^2 (au)	CE (%)	Energy (au)	σ^2 (au)	CE (%)
Silicon atom	HF	-3.677(1)	0.206(8)	0	-	-	-
	VMC	-3.7537(5)	0.0232(4)	90(1)	-3.7579(3)	0.0100(3)	95(1)
	DMC	-3.7599(4)	N/A	97(1)	-3.7624(5)	N/A	100
Silicon dimer	HF	-7.377(2)	0.56(2)	0	-	-	-
	VMC	-7.5813(9)	0.0750(8)	83.0(8)	-7.5906(7)	0.0480(5)	86.8(7)
	DMC	-7.6232(9)	N/A	100	-7.618(3)	N/A	98(2)

Results for Si atom and Si dimer using pseudopotentials

Backflow corrections in QMC

Backflow in real systems

6. What else needs to be done?

* Ceperley's derivation yields $\xi_i = \nabla_i f(R)$. Would this improve the calculations? Is the rotational part important?

* Try on other inhomogeneous systems

* Produce a large-ish set of benchmark results

* Study overlap with other methods for wave function optimization

* Etcetera

Backflow corrections in QMC

Conclusions

*Importance of having good trial wave functions in QMC

* Homogeneous backflow transformation

* Systematic extension to inhomogeneous systems

* Backflow combined with Jastrow optimization – excellent results

* Further study – how to combine with other methods

Backflow corrections in QMC

The End

"Going beyond the Slater-Jastrow wave function in fermionic systems"

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