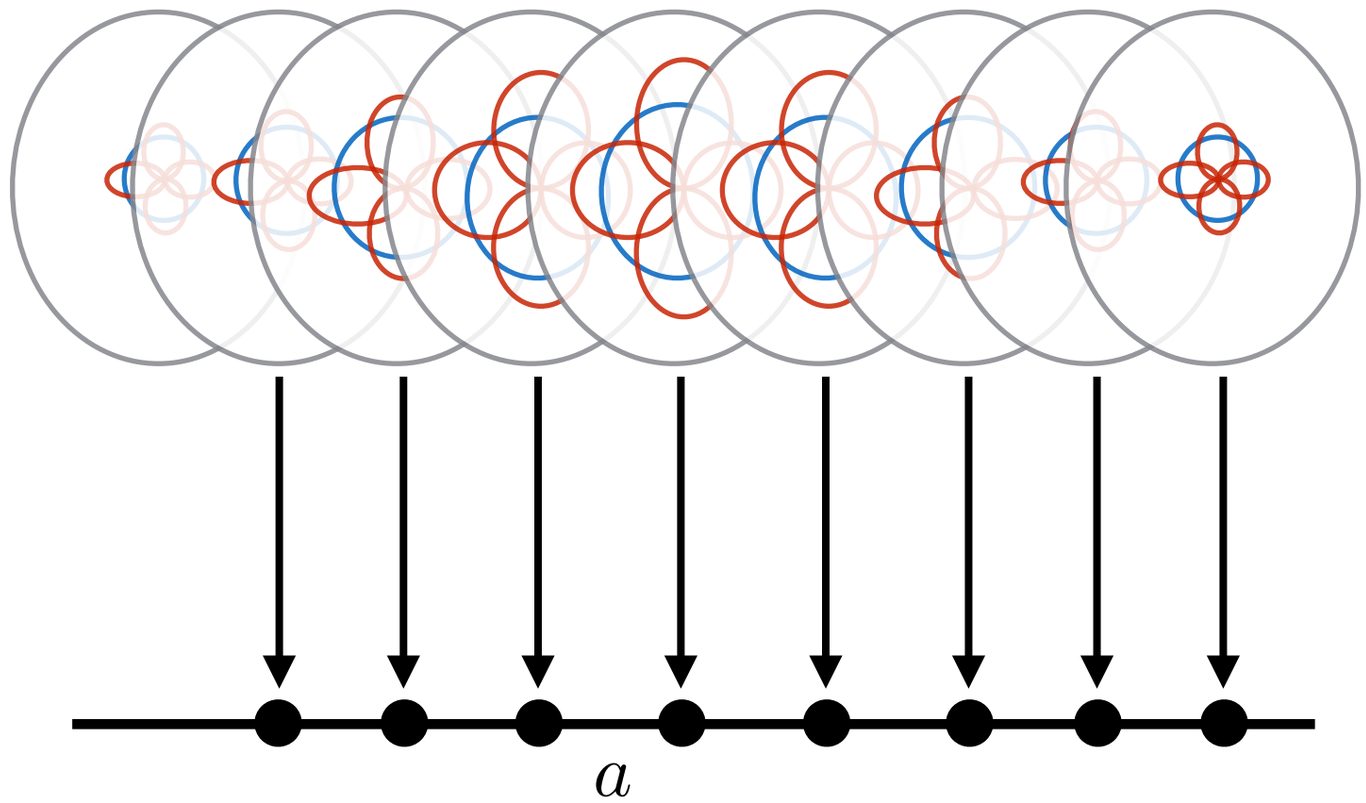
Tensor Networks for Ab Initio Quantum Systems



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Sep 2019 - Jülich

FLATIRON INSTITUTE
SIMONS FOUNDATION

This talk:

The many-electron problem

Tensor networks

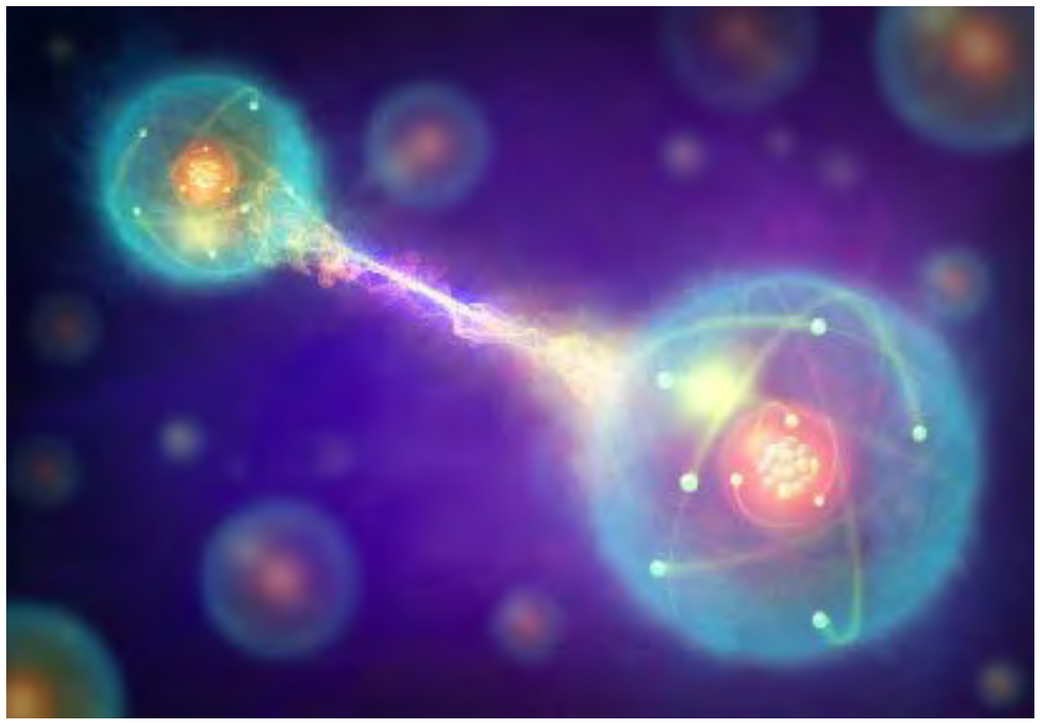
Intro to quantum chemistry

Chemistry approaches for tensor networks

The Many-Electron Problem

Behavior of electrons in matter:

- continuum problem
- three dimensional
- strong interaction (repulsion) between electrons



Credit: MARK GARLICK/SCIENCE PHOTO LIBRARY/Getty Images

Can simplify various ways:

- Born-Oppenheimer approximation (classical nuclei)
- ignore most relativistic effects

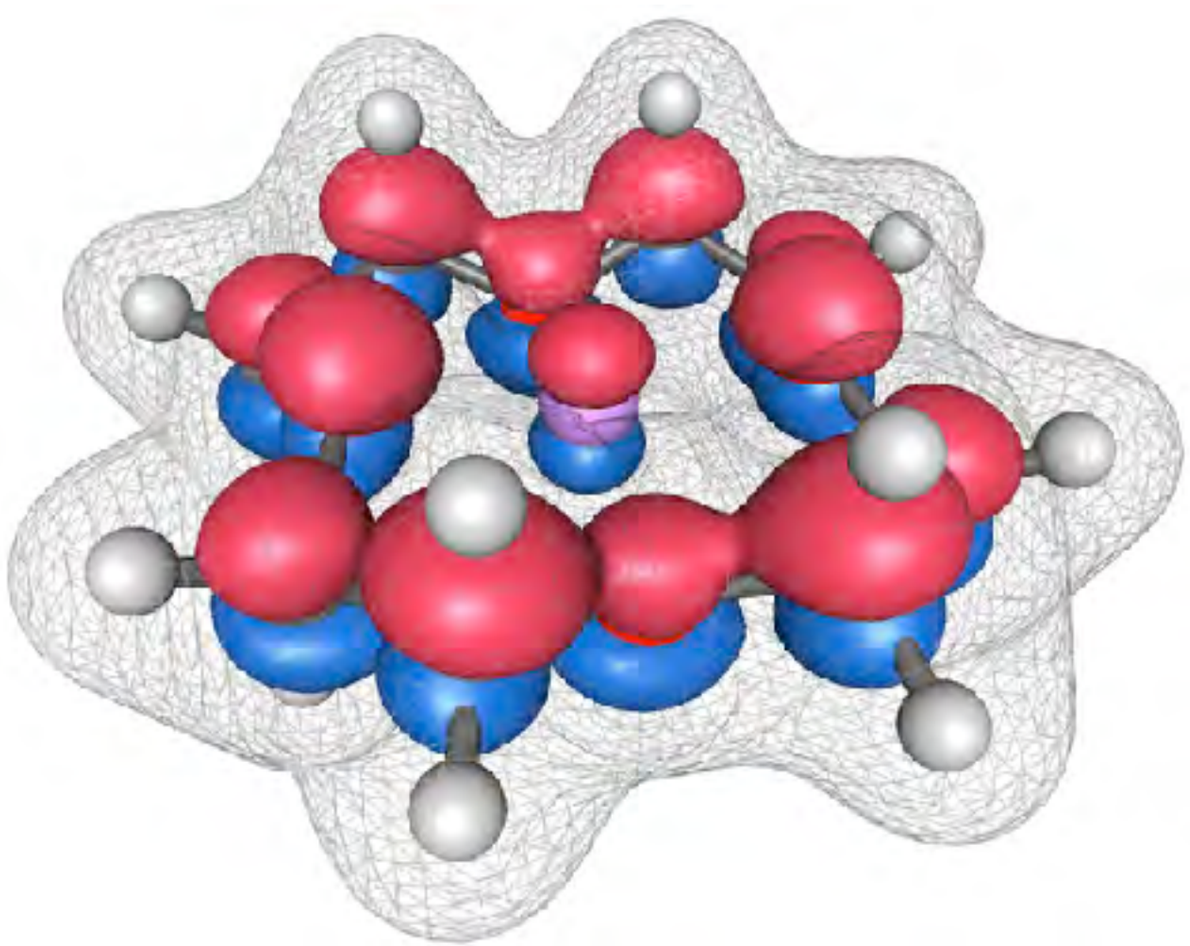


Figure credit: iqmol.org

Then problem simplifies to

$$i\frac{\partial}{\partial t}|\Psi\rangle = \hat{H}|\Psi\rangle$$

with Hamiltonian

$$\hat{H} = \frac{1}{2}\int_{\mathbf{r}} \hat{\psi}^{\dagger}_{\mathbf{r}} \left[-\nabla^2 + v(\mathbf{r})\right] \hat{\psi}_{\mathbf{r}} + \frac{1}{2}\int_{\mathbf{r}\mathbf{r}'} u(\mathbf{r},\mathbf{r}')\hat{\psi}^{\dagger}_{\mathbf{r}}\hat{\psi}^{\dagger}_{\mathbf{r}'}\hat{\psi}_{\mathbf{r}'}\hat{\psi}_{\mathbf{r}}$$

$v(\mathbf{r})$ = one-body potential

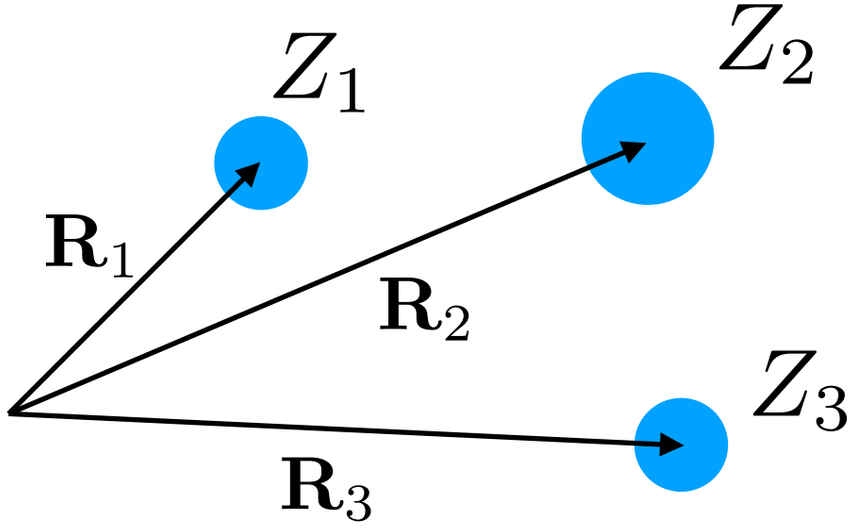
$u(\mathbf{r},\mathbf{r}')$ = two-body interaction

The *"electronic structure problem"*

Form of the one-body potential

$$v(\mathbf{r}) = -\sum_a \frac{Z_a}{|\mathbf{r} - \mathbf{R}_a|}$$

● = nucleus



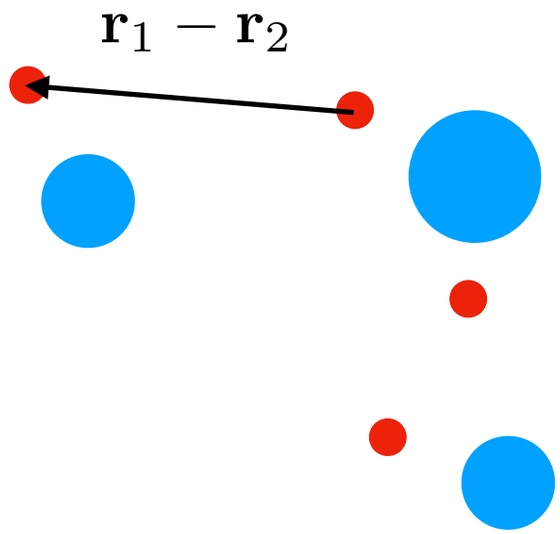
Attraction to classical point nuclei, atomic number Z

Form of the two-body potential

$$u(\mathbf{r}, \mathbf{r}') = \frac{1}{|\mathbf{r} - \mathbf{r}'|}$$

= nucleus

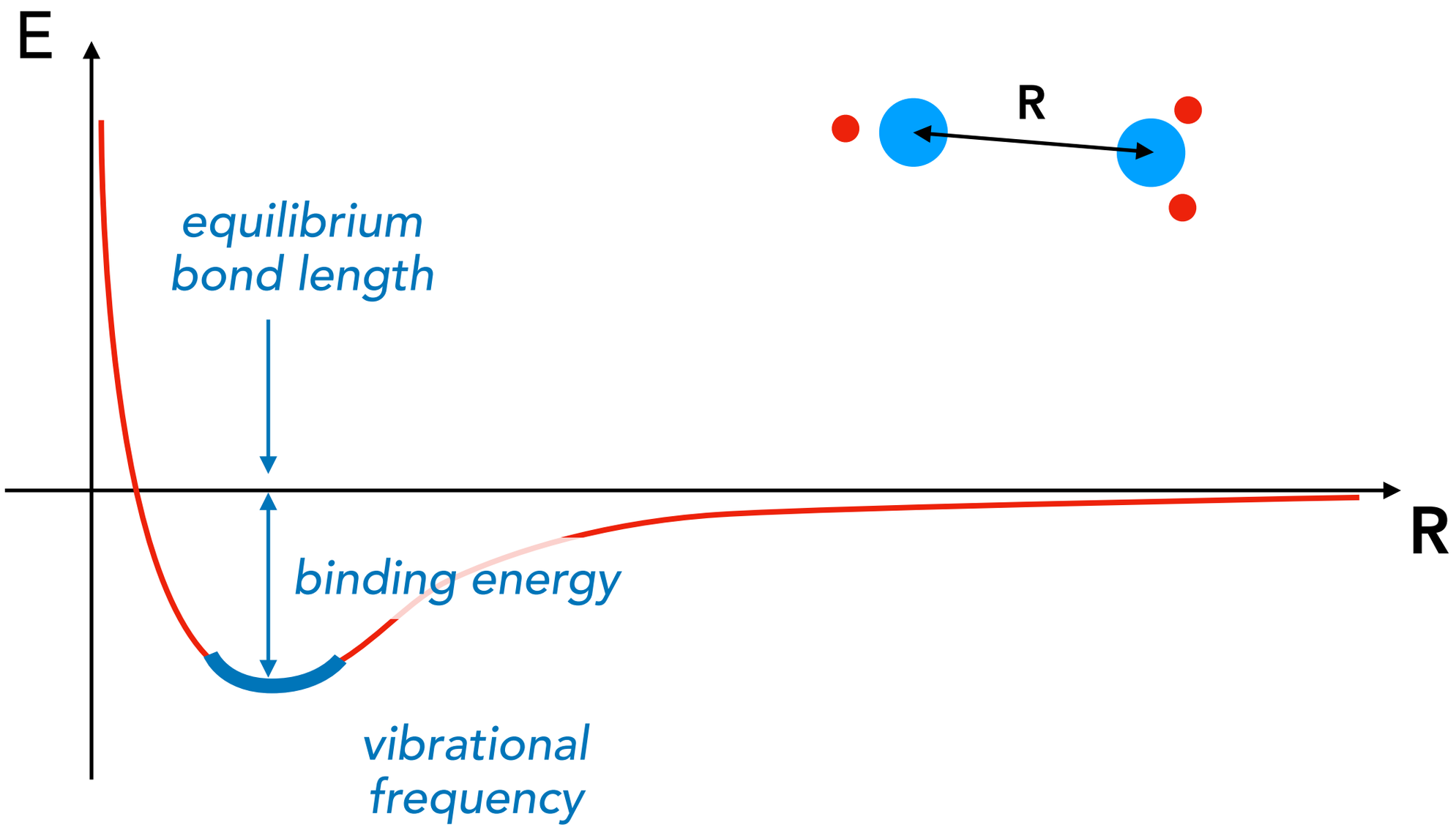
= electron



Coulomb repulsion of electrons

Accurate ground state energy
of electronic structure problem extremely useful

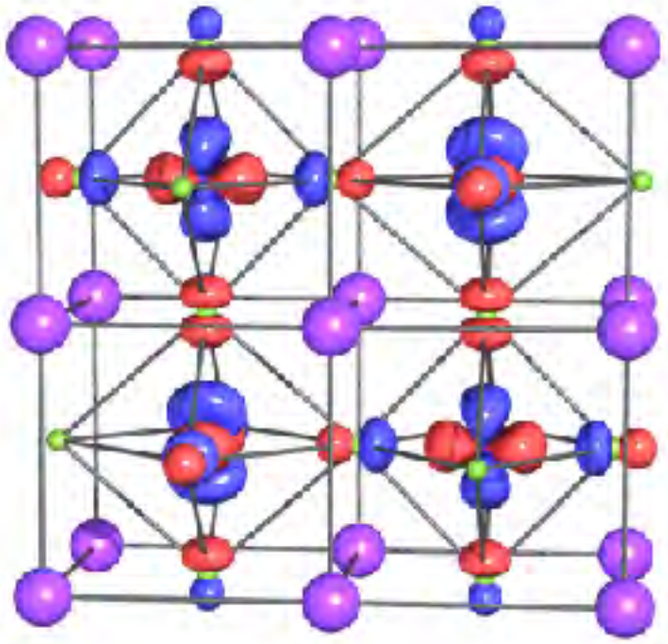
Example: energy of two atoms, distance **R**



Most techniques require discrete and finite system

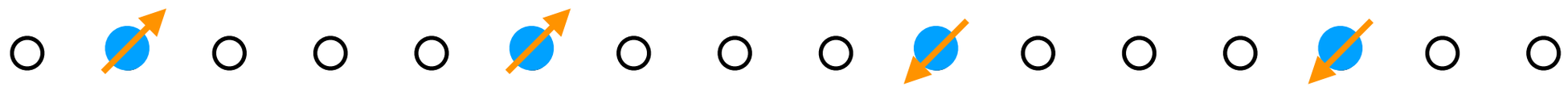
To achieve this can:

- integrate out "core" electrons (pseudopotentials)
- treat high energy states with approximations such as perturbation theory
- project electron motion to certain orbitals



Credit: EVA PAVARINI

Regardless of discretization, system becomes
a "lattice" with four states per "site"



Four states of a site are $\{0,\ \uparrow,\ \downarrow,\ \uparrow\downarrow\}$

Regardless of discretization, electronic structure Hamiltonian takes following discrete form

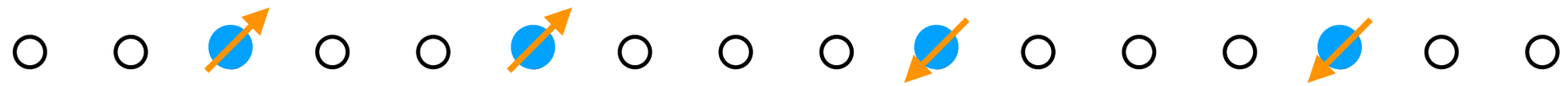
$$H = \sum_{ij} t_{ij} \hat{c}^\dagger_{i\sigma} \hat{c}_{j\sigma} + \sum_{ijkl} V_{ijkl} \hat{c}^\dagger_{i\sigma} \hat{c}^\dagger_{j\sigma'} \hat{c}_{k\sigma'} \hat{c}_{l\sigma}$$

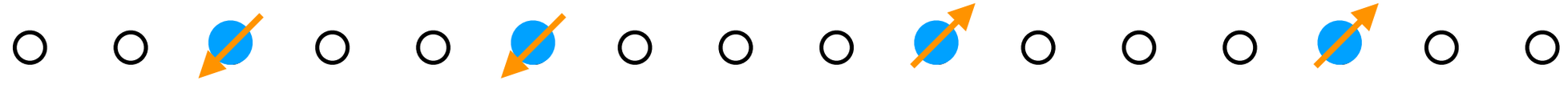
E.g. if projecting into orbitals $\{\phi_i(\mathbf{r})\}$, $\hat{c}_{i\sigma} = \int_{\mathbf{r}} \phi_i(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r})$

$$t_{ij} = \int_{\mathbf{r}} \phi_i(\mathbf{r}) \left[-\frac{1}{2}\nabla^2 + v(\mathbf{r}) \right] \phi_j(\mathbf{r})$$

$$V_{ijkl} = \int_{\mathbf{r}_1, \mathbf{r}_2} \frac{\phi_i(\mathbf{r_1}) \phi_j(\mathbf{r_2}) \phi_k(\mathbf{r_2}) \phi_l(\mathbf{r}_1)}{|\mathbf{r}_1 - \mathbf{r}_2|}$$

Quantum wavefunction assigns an amplitude to each configuration:

1: 

2: 

3: 

Four states per site $\{0,\ \uparrow,\ \downarrow,\ \uparrow\downarrow\}$

So 4^N configurations given N sites

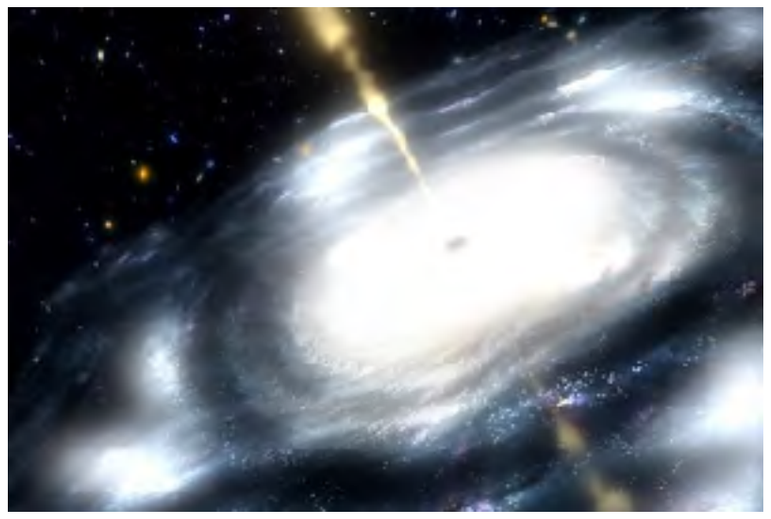
Could try to store all the amplitudes, but

N = 10, $4^{10} \sim 10^6$

N = 20, $4^{20} \sim 10^{12}$

N = 30, $4^{30} \sim 10^{18}$

For N > 130, number of amplitudes greater than **number of atoms in the known universe**



But can "nature's computer" really work this way?

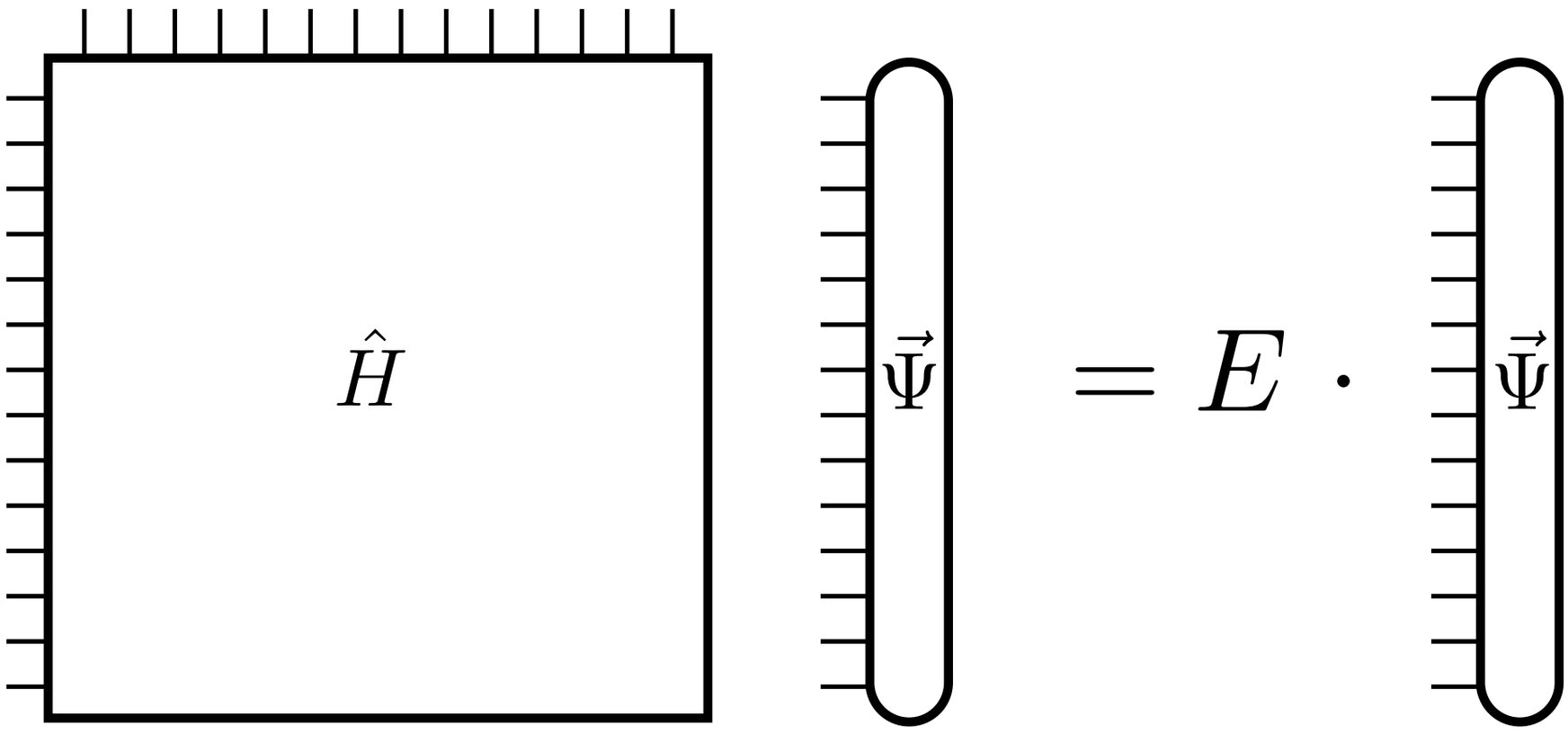
Are the amplitudes of a realistic wavefunction all different?

Or is there some relationship between them?

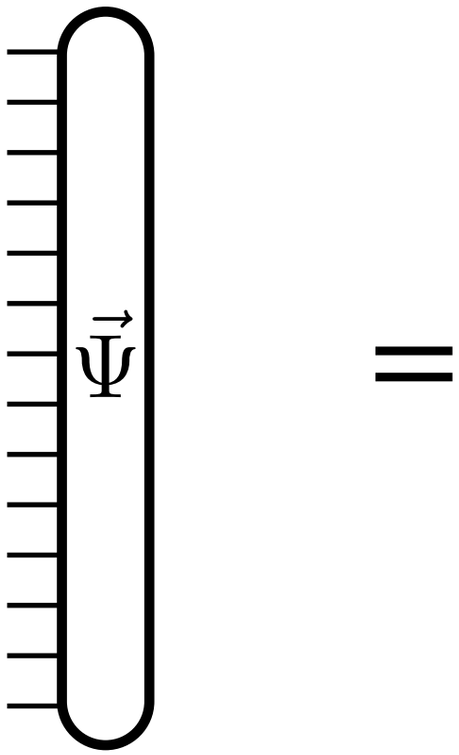
Tensor Network Wavefunctions

$\hat{H}$ is a 4^N x 4^N matrix

$\Longrightarrow$ wavefunction $\vec{\Psi}$ has 4^N components

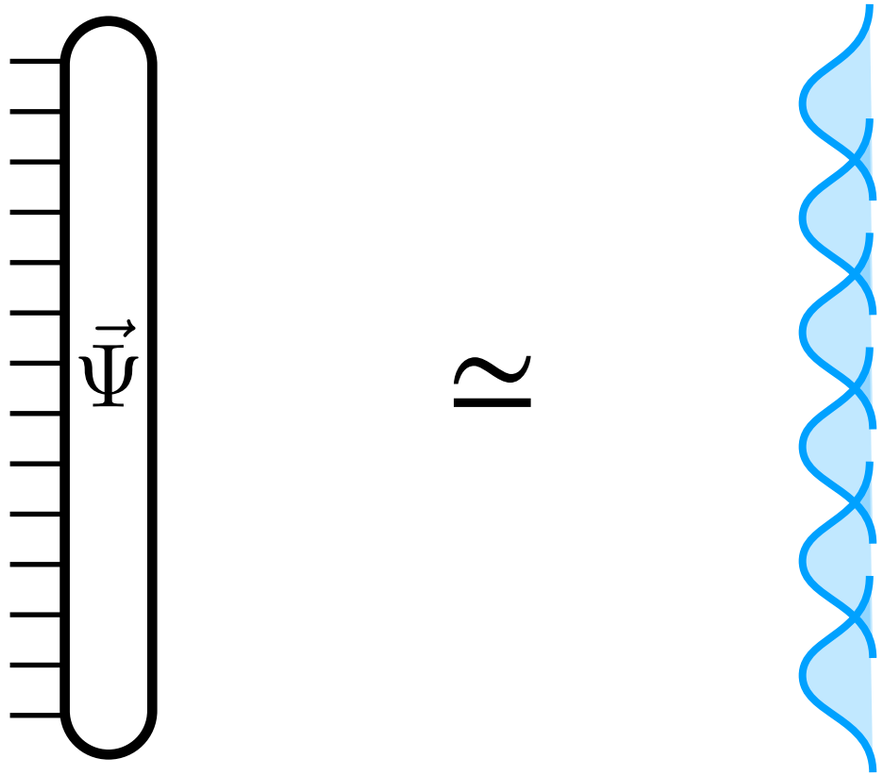


Most of these eigenvectors / wavefunctions
are indeed exponentially complex



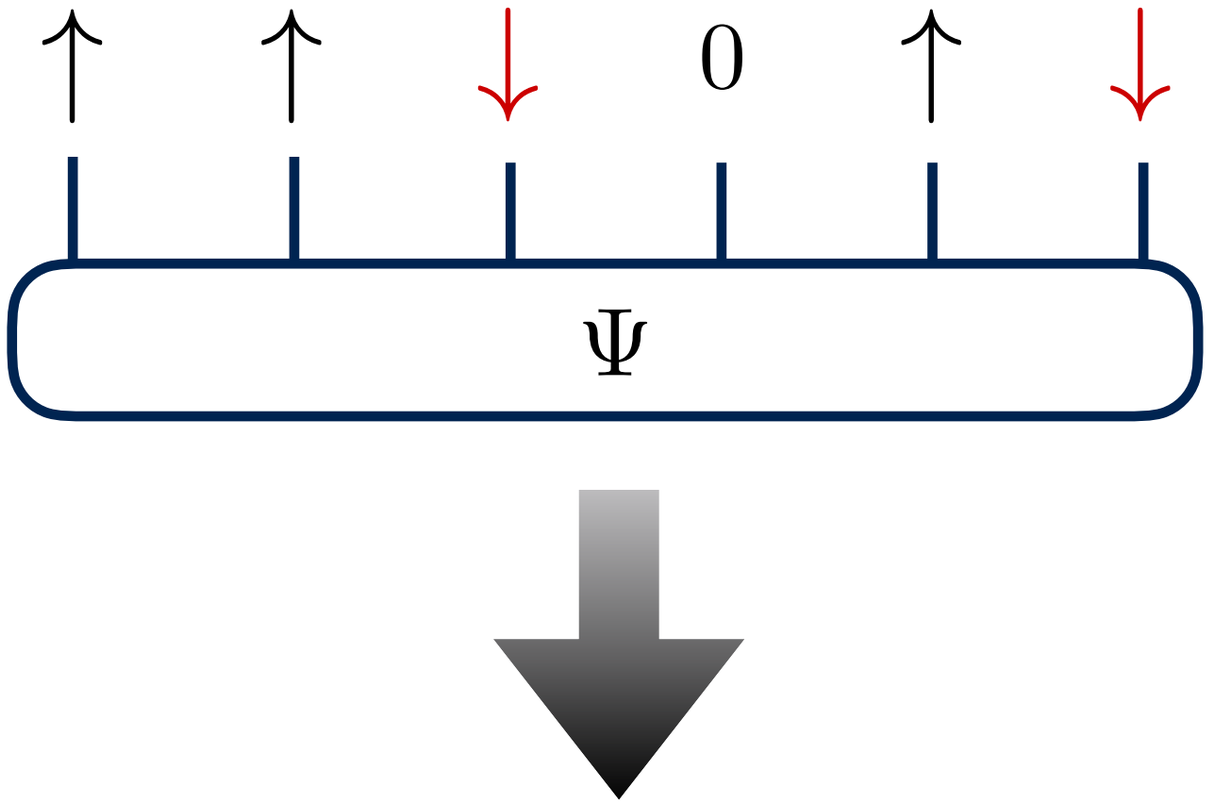
=

But some have hidden structure that makes them *tractable*



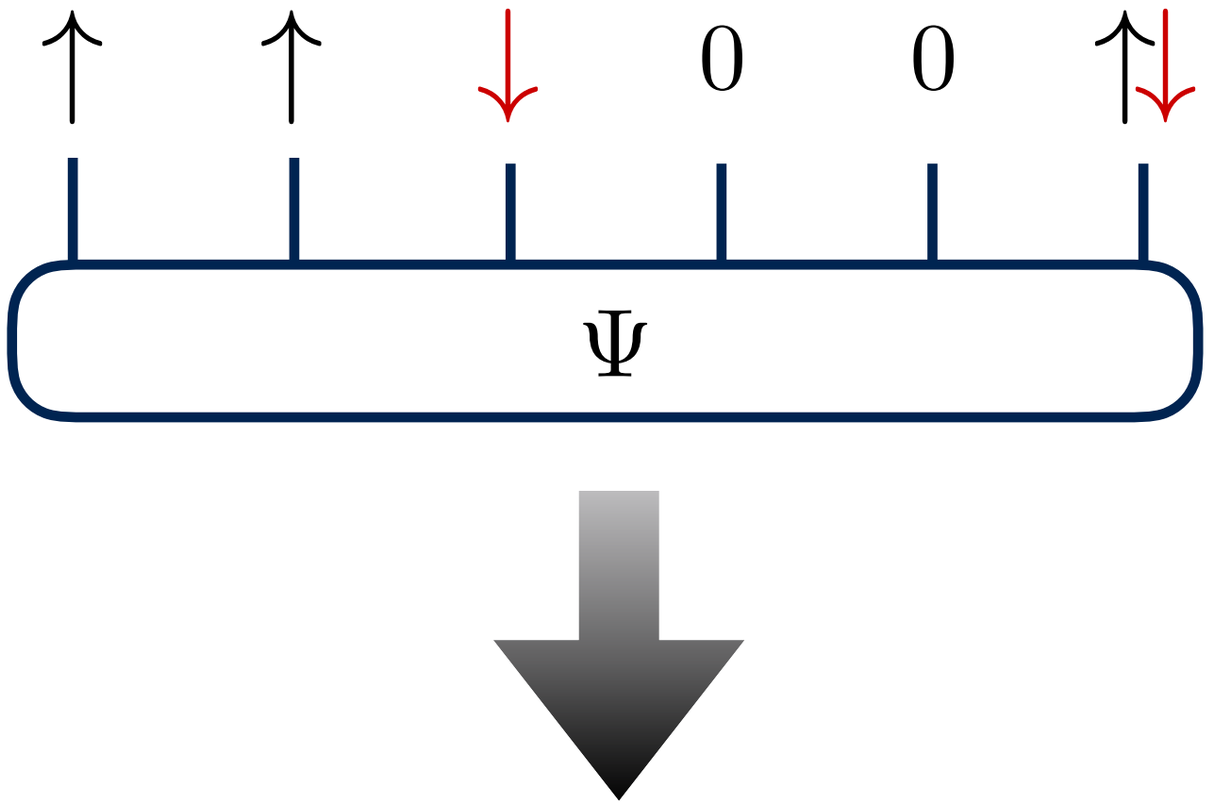
- low-energy states
- equilibrium states
- short-time dynamics

Wavefunction a rule, mapping configurations to numbers



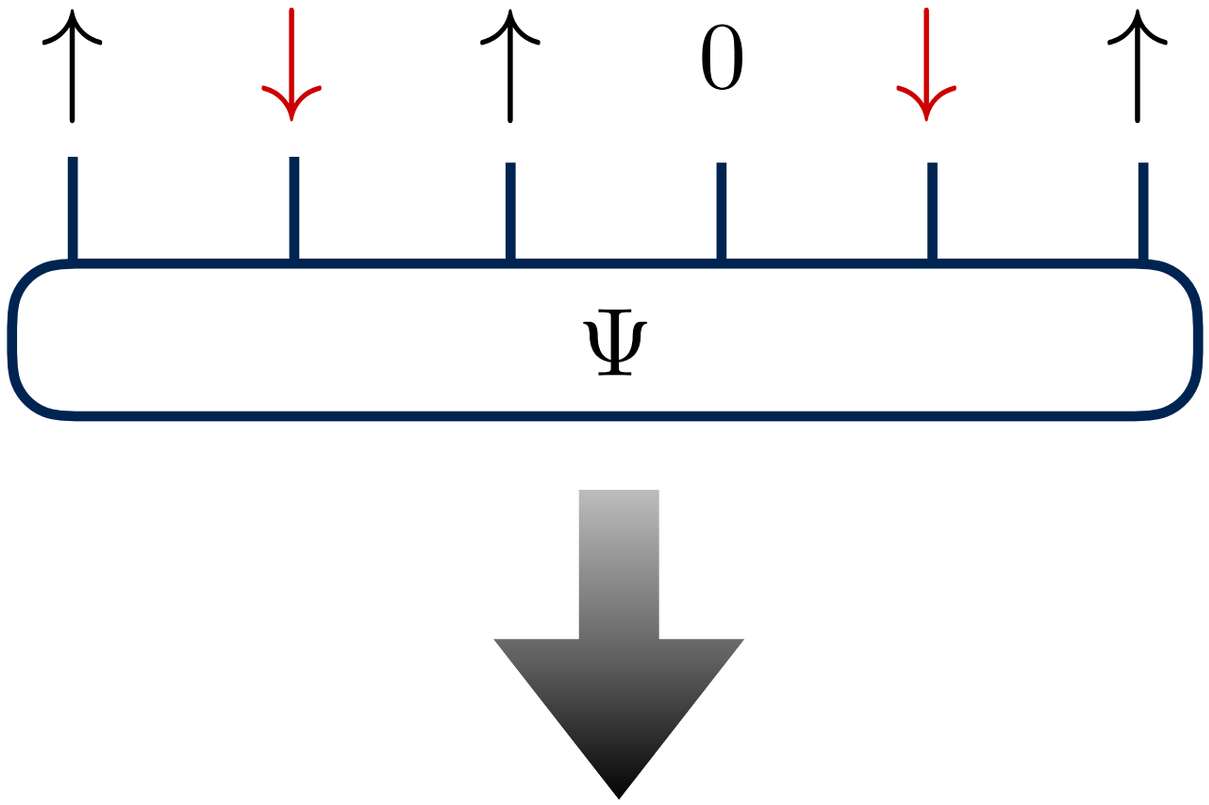
0.1

Wavefunction a rule, mapping configurations to numbers



0.05

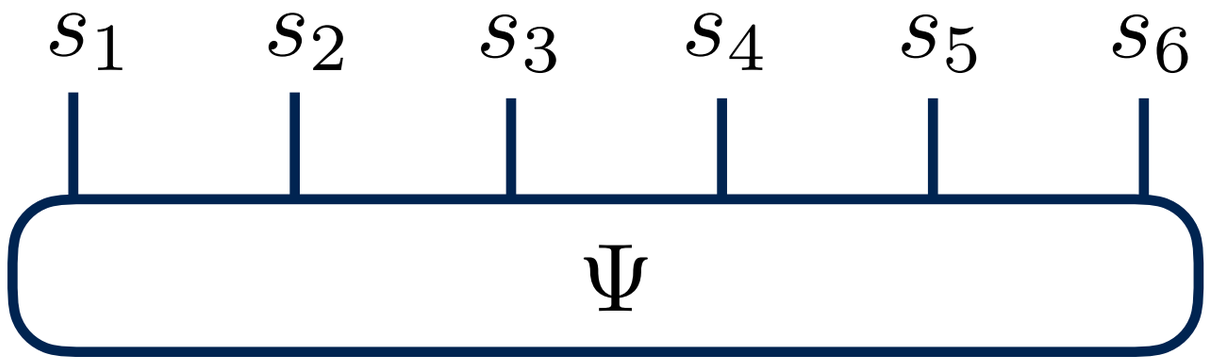
Wavefunction a rule, mapping configurations to numbers



-0.2

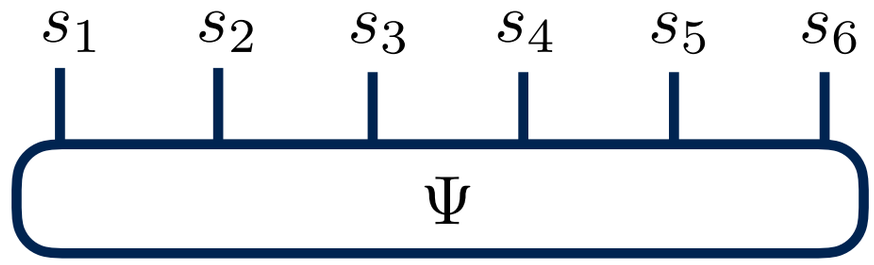
Formally a *tensor* with N indices

$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} =$$



4^N numbers inside

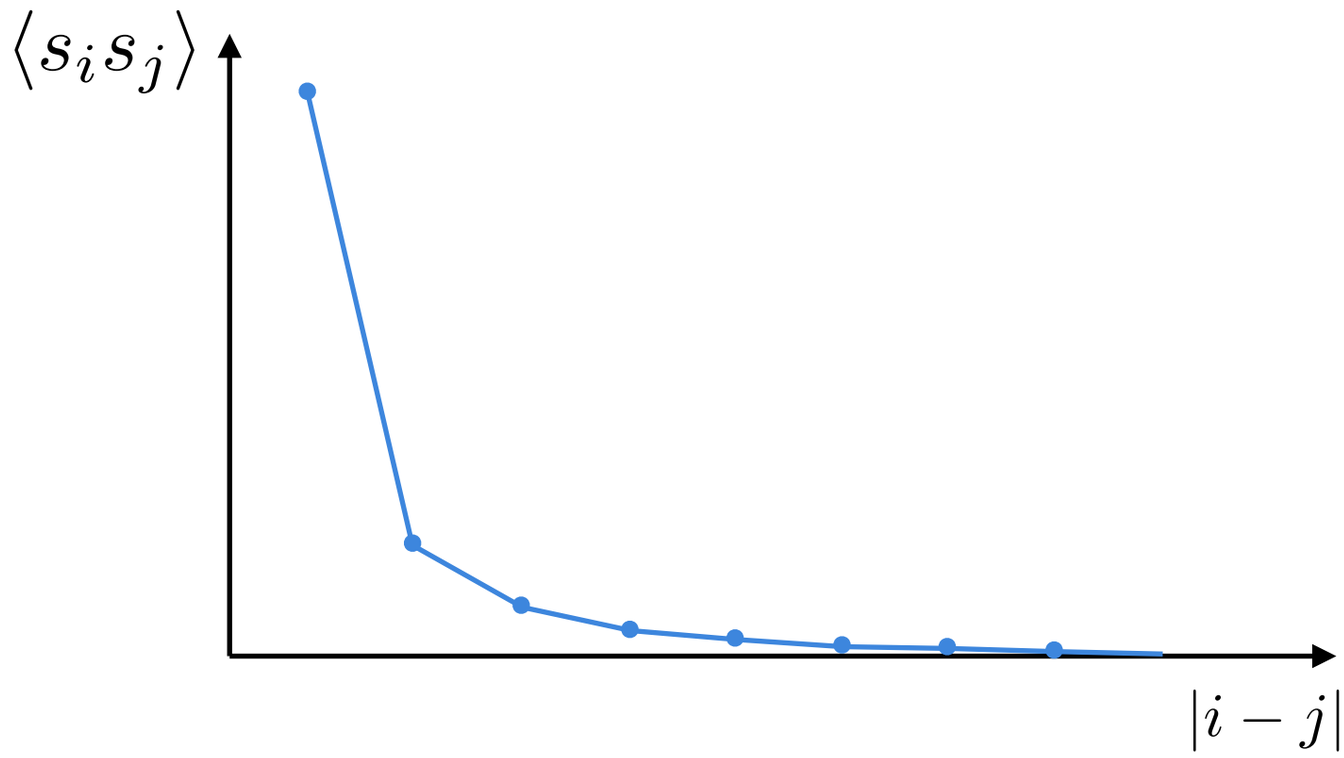
$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} =$$



Problem seems hopeless (maximum N ~ 20)

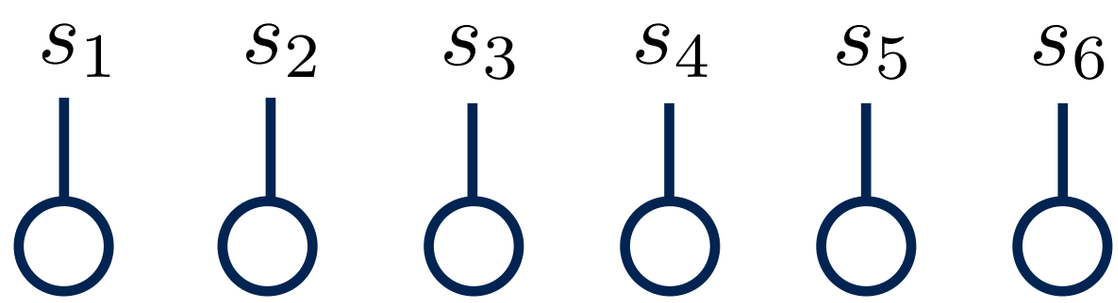
Physical intuition:

weak correlations between distant particles



Neglect correlations

$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} \simeq \psi^{s_1}\, \psi^{s_2}\, \psi^{s_3}\, \psi^{s_4}\, \psi^{s_5}\, \psi^{s_6}$$

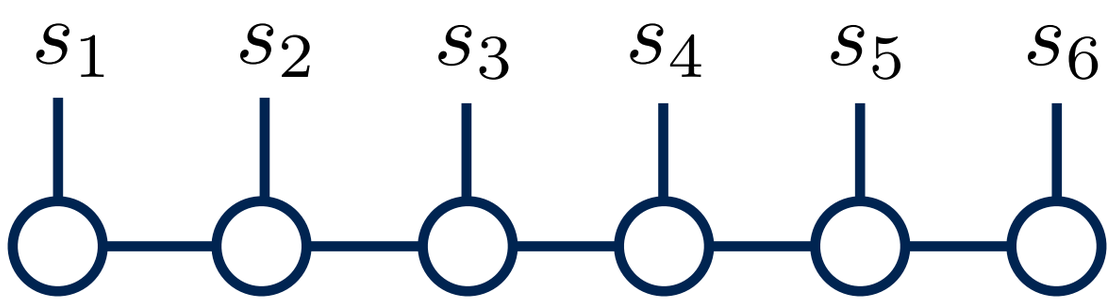


✓ Expected values of individual sites ok

✗ Missing correlations

Restore correlations locally

$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} \simeq \psi^{s_1}_{i_1} \psi^{s_2}_{i_1 i_2} \psi^{s_3}_{i_2 i_3} \psi^{s_4}_{i_3 i_4} \psi^{s_5}_{i_4 i_5} \psi^{s_6}_{i_5}$$



matrix product state (MPS)

✓ Local expected values accurate

✓ Exponentially decaying correlations

Name *matrix product state* derives from

$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} \simeq \psi^{s_1}_{i_1} \underbrace{\psi^{s_2}_{i_1 i_2}}\underbrace{\psi^{s_3}_{i_2 i_3}}\underbrace{\psi^{s_4}_{i_3 i_4}}\underbrace{\psi^{s_5}_{i_4 i_5}}\underbrace{\psi^{s_6}_{i_5}}$$

$$\psi^{s_3=0}_{i_2 i_3} = M^{0}_{i_2 i_3}$$

$$\psi^{s_3=\uparrow}_{i_2 i_3} = M^{\uparrow}_{i_2 i_3}$$

$$\psi^{s_3=\downarrow}_{i_2 i_3} = M^{\downarrow}_{i_2 i_3}$$

$$\psi^{s_3=\uparrow\downarrow}_{i_2 i_3} = M^{\uparrow\downarrow}_{i_2 i_3}$$

Compute wavefunction by multiplying matrices together

$$\Psi^{\uparrow\downarrow\uparrow\uparrow\downarrow} \approx M_1^{\uparrow} M_2^{\downarrow} M_3^{\uparrow} M_4^{\uparrow} M_5^{\downarrow}$$

$$\Psi^{\uparrow\uparrow 0\downarrow\downarrow} \approx M_1^{\uparrow} M_2^{\uparrow} M_3^{0} M_4^{\downarrow} M_5^{\downarrow}$$

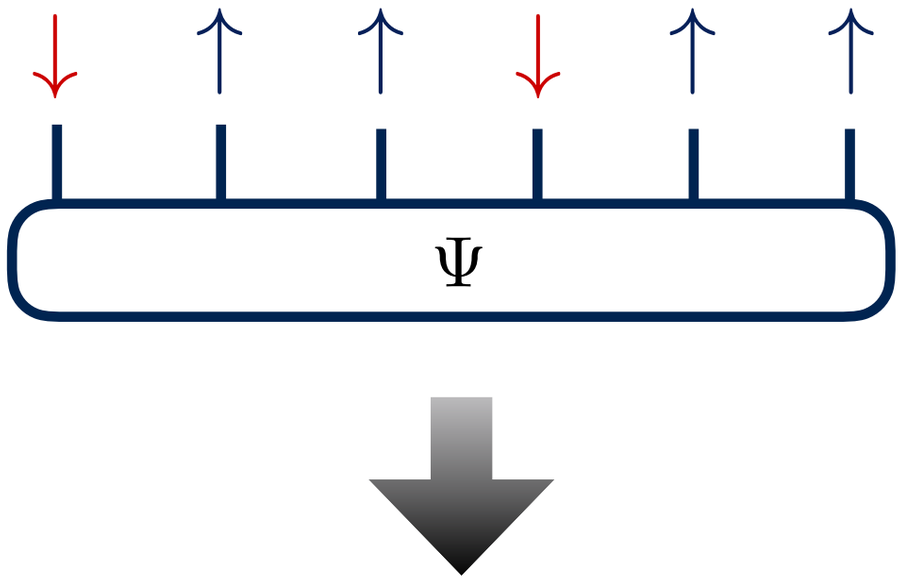
$$\Psi^{\uparrow\downarrow\downarrow\uparrow\Updownarrow} \approx M_1^{\uparrow} M_2^{\downarrow} M_3^{\downarrow} M_4^{\uparrow} M_5^{\Updownarrow}$$

matrix product state compresses a tensor

$$\Psi^{s_1 s_2 s_3 s_4 s_5} = M_1^{s_1} M_2^{s_2} M_3^{s_3} M_4^{s_4} M_5^{s_5}$$

For typical matrix size $m \times m$

4^N parameters $\longrightarrow$ $4Nm^2$ parameters



$$M_1^{\downarrow} \; M_2^{\uparrow} \; M_3^{\uparrow} \; M_4^{\downarrow} \; M_5^{\uparrow} \; M_6^{\uparrow}$$

Why this rule?

1. **Principled**

Large enough matrices, represent *any* wavefunction

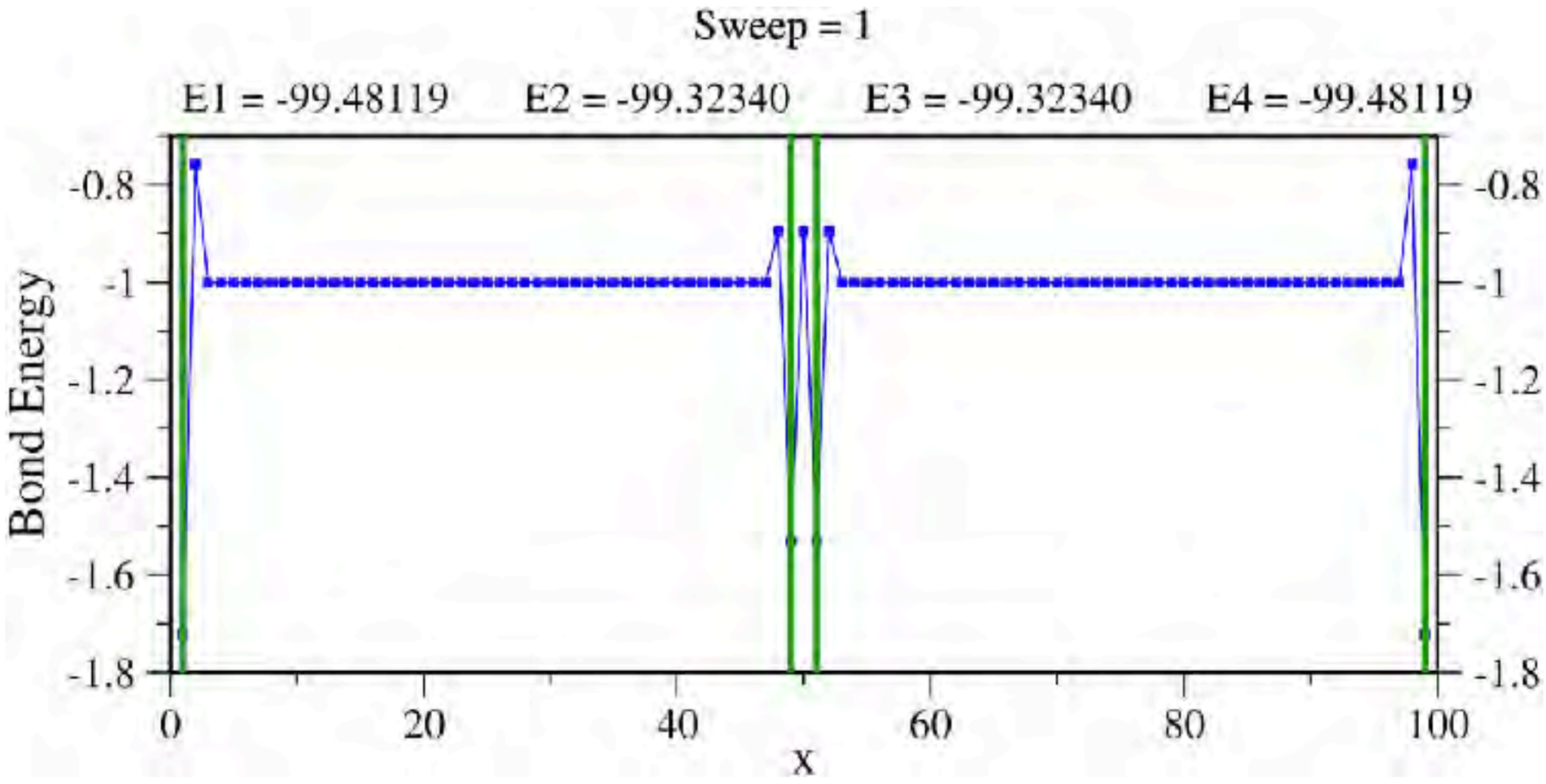
Proof (Hastings, 2007) that one-dimensional systems
with exponentially decaying correlations*
are 'close' to matrix product states

Error decreases rapidly (exponentially in practice)
and *error per site* is constant

*(technically: gap between ground state and excited states)

2. Powerful Algorithms

Matrix product ansatz comes with sophisticated optimization techniques



DMRG algorithm

White, PRL **69**, 2863 (1992)

Stoudenmire, White, PRB **87**, 155137 (2013)

"In one dimension... it is at the moment the closest to an ultimate weapon as one can dream of."

– Thierry Giamarchi

"Quantum Physics in One Dimension"

Very often get exact answer

Only takes as many parameters as needed

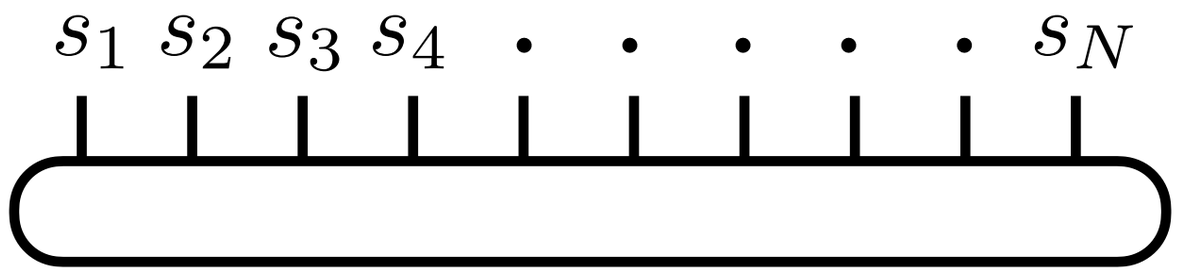
3. Extendable

Generalize to ***tensor networks***

Apply to two-dimensional systems, infinite systems, ...

Draw N-index tensor as blob with N lines

$$\Psi^{s_1 s_2 s_3 \cdots s_N} =$$

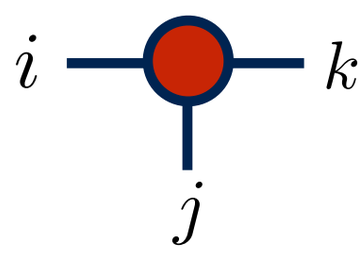


Diagrams for simple tensors

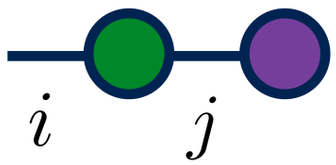
vector v_j

matrix M_{ij}

3-index tensor T_{ijk}

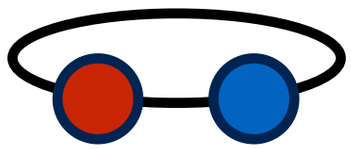


Joining lines means contraction, omit names



$$\longleftrightarrow \quad \sum_j M_{ij} v_j$$

$$\longleftrightarrow \quad A_{ij} B_{jk} = AB$$

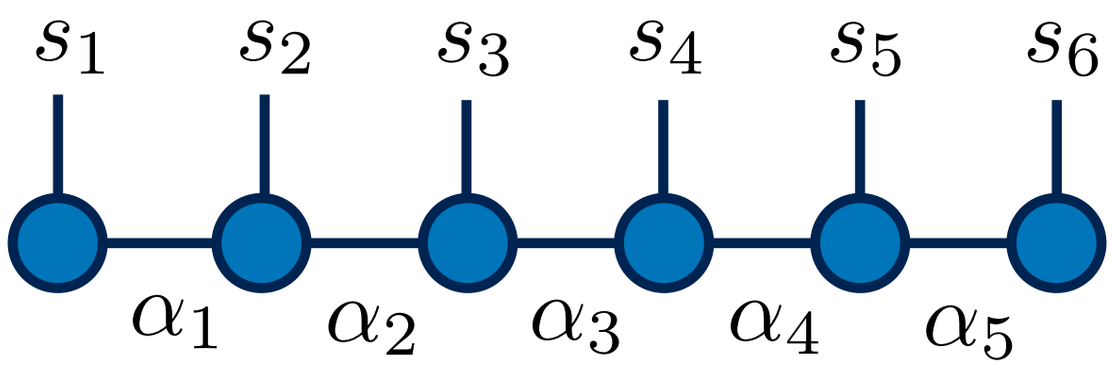


$$\longleftrightarrow \quad A_{ij} B_{ji} = \mathrm{Tr}[AB]$$

Matrix product state in diagram notation

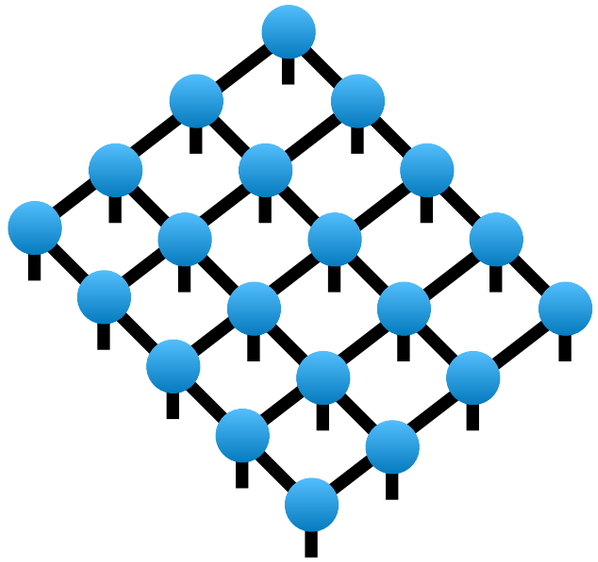
$$\Psi^{s_1 s_2 s_3 s_4 s_5 s_6} = \sum_{\alpha} M^{s_1}_{\alpha_1} M^{s_2}_{\alpha_1 \alpha_2} M^{s_3}_{\alpha_2 \alpha_3} M^{s_4}_{\alpha_3 \alpha_4} M^{s_5}_{\alpha_4 \alpha_5} M^{s_6}_{\alpha_5}$$

$=$



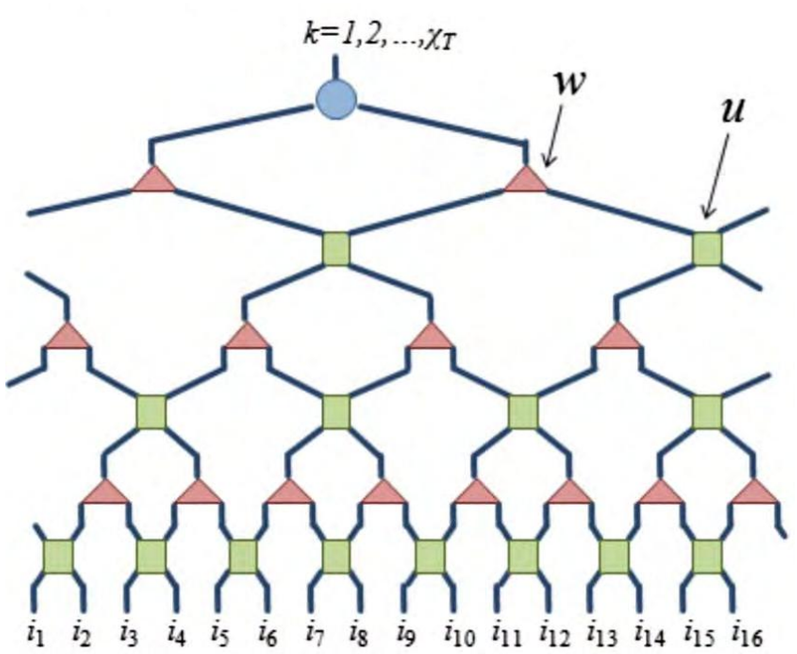
Suppress index names, very convenient

Besides matrix product state network, other very interesting networks are PEPS and MERA



PEPS

(2D systems)



MERA

(critical systems)

Evenbly, Vidal, PRB **79**, 144108 (2009)

Verstraete, Cirac, cond-mat/0407066 (2004)

Orus, Ann. Phys. **349**, 117 (2014)

In addition to physics, tensor networks useful as an applied math technique

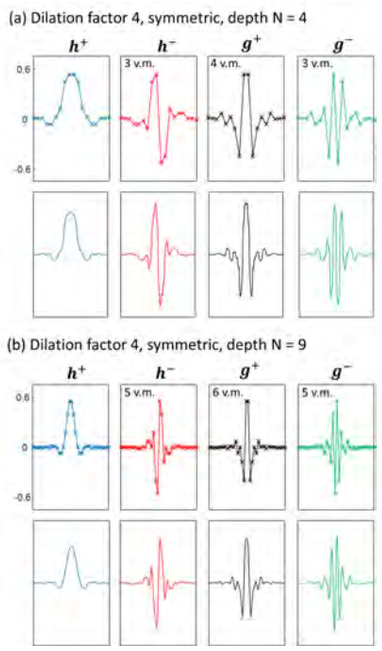


FIG. 12. Plots of dilation $m = 4$ orthogonal wavelets from (a) depth $N = 4$ and (b) depth $N = 9$ quarternary circuits with angles θ_k as given in Tab. VIII. The top three panels of each group denote the (exactly symmetric) scaling sequence $\boldsymbol{h}^+$ and wavelet sequences $\boldsymbol{h}^-, \boldsymbol{g}^+, \boldsymbol{h}^-$, which possess the number of vanishing moments (v.m.) as indicated. The bottom three panels of each group depict the scaling functions and wavelets in the continuum limit (windowed to include only the non-vanishingly small part of the functions).

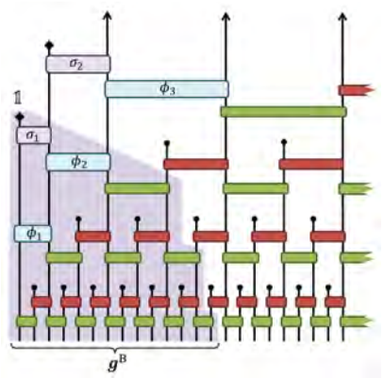
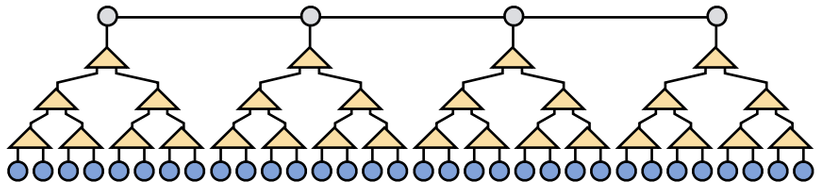


FIG. 13. Depiction of a multi-scale circuit (formed from composition of depth $N = 2$ binary unitary circuits) with an open boundary on the left, where a double layer of (scale dependent) unitary gates $u(\phi_z)$ and $u(\sigma_z)$ is introduced. Boundary wavelets $\boldsymbol{g}^B$ are given by transforming the unit vector $\mathbb{1}$ located on a boundary index as indicated. The angles ϕ_z and σ_z are chosen to ensure the boundary wavelets each have two vanishing moments.

	ϕ_z	σ_z
$z = 1$	0.615479708	0.261157410
$z = 2$	0.713724378	0.316335000
$z = 3$	0.752040089	0.339836909
$z = 4$	0.769266332	0.350823961
$z = 5$	0.777461322	0.356148400
$z = 6$	0.781461114	0.358770670
$z = 7$	0.783437374	0.360072087
$z = 8$	0.784419689	0.360720398
$z = 9$	0.784909404	0.361043958
$z = 10$	0.785153903	0.361205590
$z = 11$	0.785276063	0.361286369
$z = 12$	0.785337120	0.361326749

TABLE IX. Angles ϕ_z and σ_z parameterizing the boundary

Evenbly, White,
"Representation and design of wavelets using unitary circuits"



89% accuracy on Fashion MNIST data set

Stoudenmire,
"Learning Relevant Features of Data with Multi-scale Tensor Networks"

Brief Note on Fermions

Typical tensor network approach uses
second quantization

This means:

$$|\Psi\rangle = \psi^{s_1 s_2 s_3 s_4} |s_1 s_2 s_3 s_4\rangle \qquad s_j = 0, 1$$

$$= \psi^{s_1 s_2 s_3 s_4} \; \underbrace{(\hat{c}_1^\dagger)^{s_1} (\hat{c}_2^\dagger)^{s_2} (\hat{c}_3^\dagger)^{s_3} (\hat{c}_4^\dagger)^{s_4}}_{\text{can be any tensor}} \underbrace{|0\rangle}_{\text{all antisymmetry handled in this part}}$$

No need to antisymmetrize (or symmetrize)
amplitude tensor represented by tensor network

When do the signs enter in?

When using operators:

- applying Hamiltonian
- computing observables

$$\hat{c}_2 \left[\psi^{s_1 s_2 s_3 s_4} \; (\hat{c}_1^\dagger)^{s_1} (\hat{c}_2^\dagger)^{s_2} (\hat{c}_3^\dagger)^{s_3} (\hat{c}_4^\dagger)^{s_4} \right] |0\rangle$$

Sign of result will depend on value of s_1 index

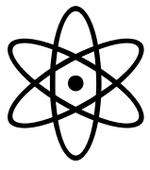
Fermion minus signs & tensor networks

Programming approaches – 3 alternatives:

- map fermionic operators to non-local bosonic operators (Jordan-Wigner transformation); work only with these

- choose canonical, reference ordering of sites and always permute basis states to this order

- anti-commuting tensor indices (newest approach)



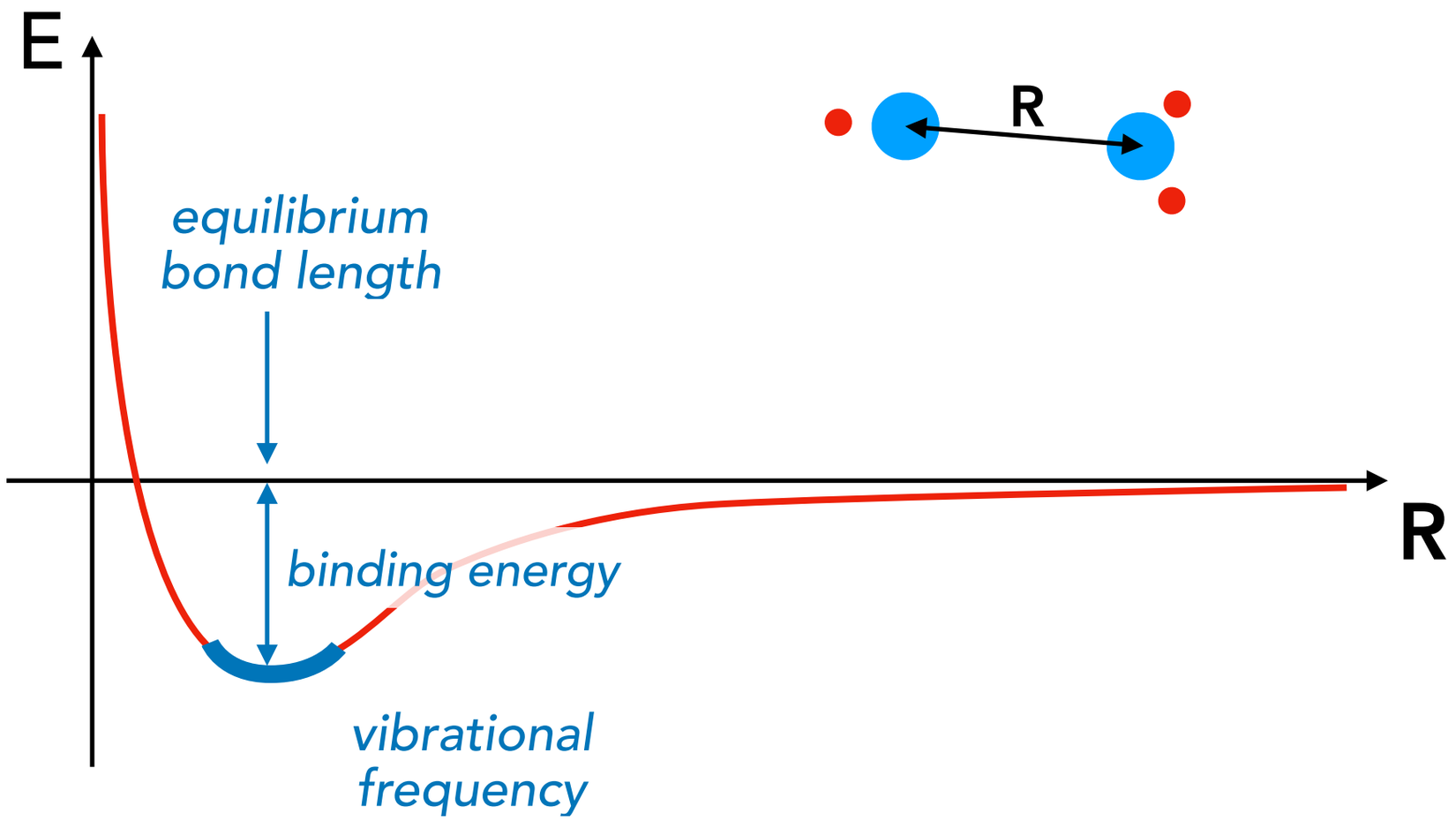
Quantum Chemistry

Chemistry an instance of the many-electron problem:

$$\hat{H} = \frac{1}{2}\int_{\mathbf{r}} \hat{\psi}^{\dagger}_{\mathbf{r}} \left[-\nabla^2 + v(\mathbf{r})\right] \hat{\psi}_{\mathbf{r}} + \frac{1}{2}\int_{\mathbf{r}\mathbf{r}'} u(\mathbf{r},\mathbf{r}') \hat{\psi}^{\dagger}_{\mathbf{r}} \hat{\psi}^{\dagger}_{\mathbf{r}'} \hat{\psi}_{\mathbf{r}'} \hat{\psi}_{\mathbf{r}}$$

electronic structure Hamiltonian

Unlike some areas of condensed matter,
mostly after **energies** & **quantitative** properties



But qualitative properties also important:

Will two molecules or atoms bind?

State of atoms during a reaction?

Biggest challenges in quantum chemistry:

- continuum nature of problem

- strong correlation

Tensor network methods don't suffer from **strong correlation**

Strong correlation is fact that some wavefunctions
(e.g. stretched diatomic molecules) are sum of
exponentially many Slater determinants 😬

Tensor networks do not use sums of Slater determinants ✅

Continuum is the bigger issue for tensor networks...

Standard approach pioneered by John Pople is to use *Gaussian basis functions* to approximate the continuum

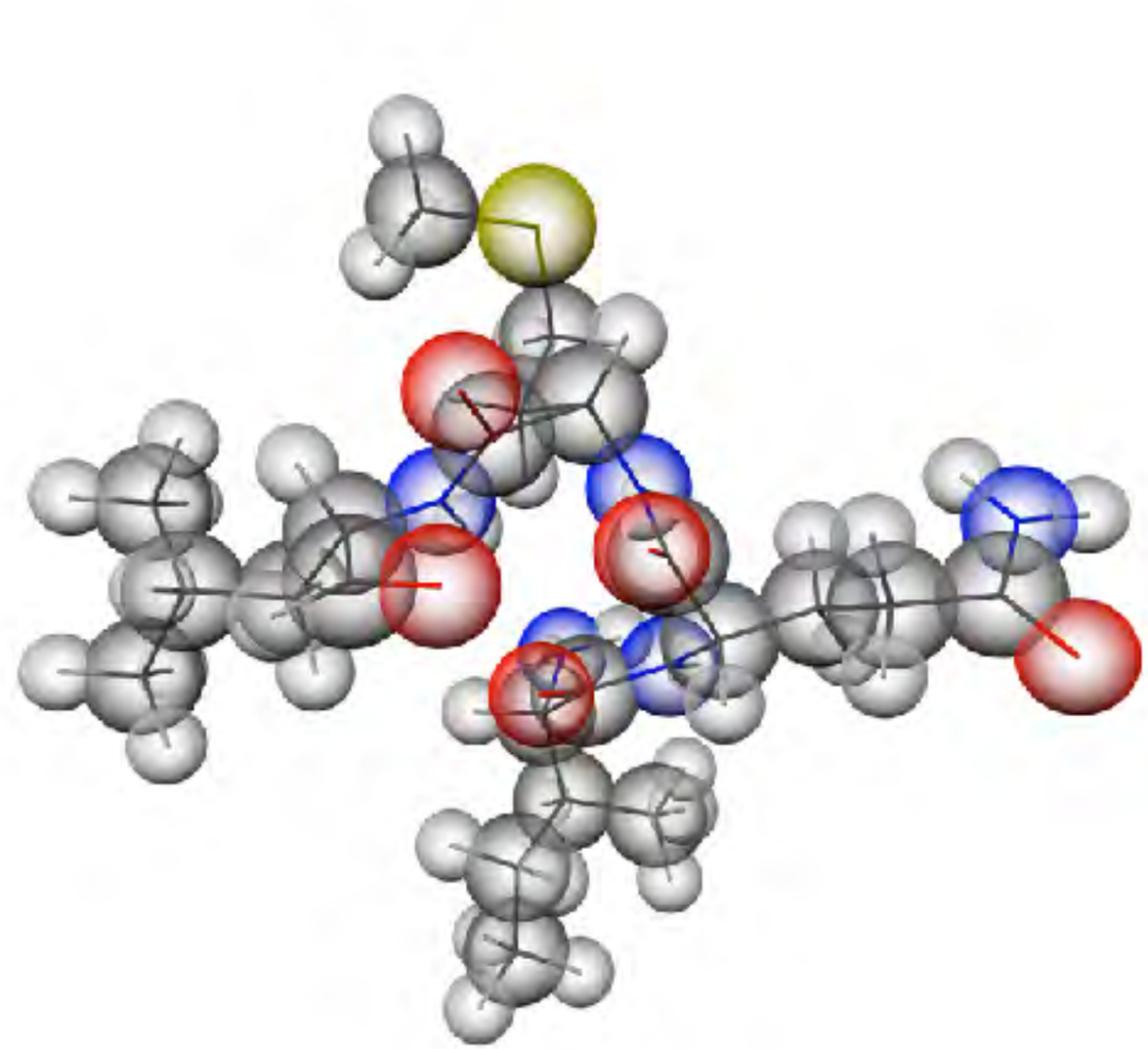
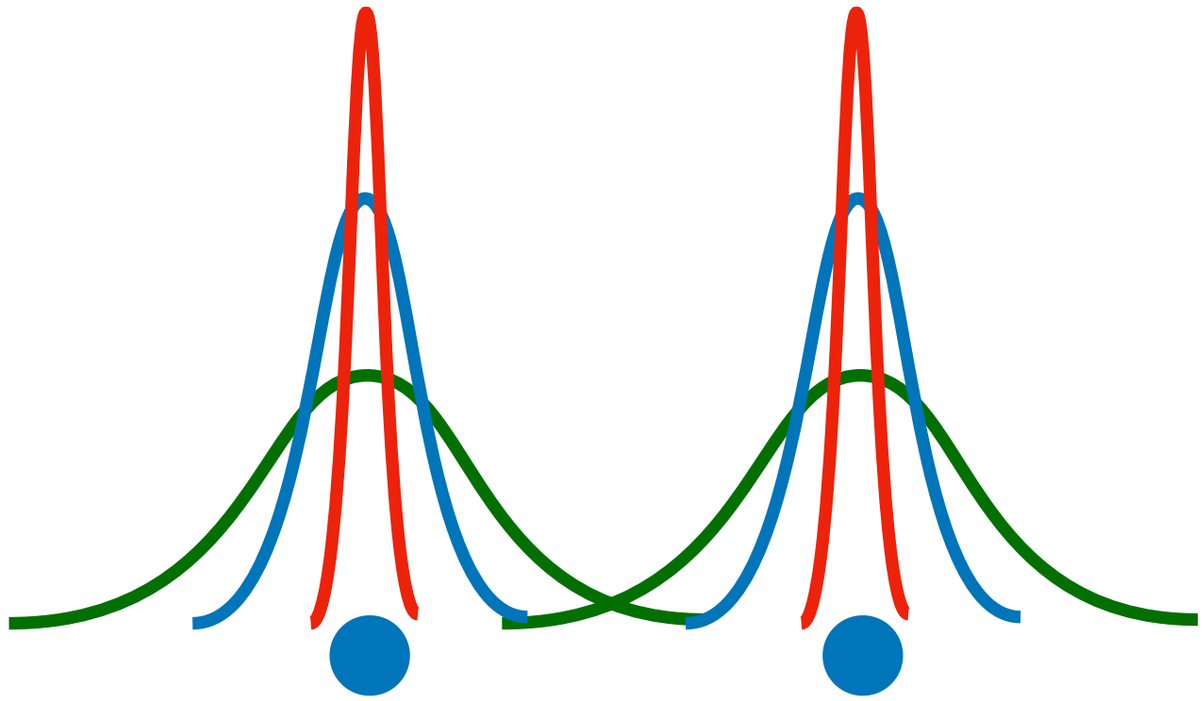


Figure credit: iqmol.org

Consider H_2 molecule

Cartoon of Gaussian **basis set**:



Basis sets also include linear combinations of Gaussians:

$$b_n(\mathbf{r}) = \sum_{i=1}^{N_n} c_{n,i} e^{-\zeta_{n,i}(\mathbf{r}-\mathbf{r}_A)^2}$$

And multiplicative factors: $x^p y^q z^s$

Orbital basis Hamiltonian (*i,j,k,l* label orbital 'sites'):

$$H = \sum_{ij} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \sum_{ijkl} V_{ijkl} c^\dagger_{i\sigma} c^\dagger_{j\sigma'} c_{k\sigma'} c_{l\sigma}$$

$$t_{ij} = \int_{\mathbf{r}} \phi_i(\mathbf{r}) \left[-\frac{1}{2}\nabla^2 + v(\mathbf{r}) \right] \phi_j(\mathbf{r})$$

Basis functions, or "orbitals"

$$V_{ijkl} = \int_{\mathbf{r}_1, \mathbf{r}_2} \frac{\phi_i(\mathbf{r_1}) \phi_j(\mathbf{r_2}) \phi_k(\mathbf{r_2}) \phi_l(\mathbf{r_1})}{|\mathbf{r}_1 - \mathbf{r}_2|}$$

Coulomb "integrals"

The coefficients V_{ijkl} are called Coulomb *integrals*

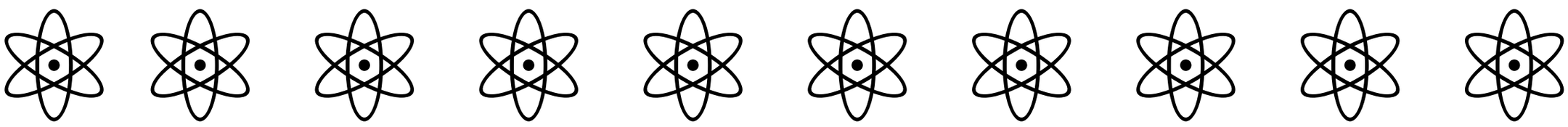
For N orbitals, there are N^4 of these....

Say N=100, then N^4 = *one hundred million*!

Just constructing the Hamiltonian is a serious business...

Point of Gaussians is computing integrals quickly!
(Especially on 1990's computers...)

DMRG & MPS for Quantum Chemistry



DMRG and MPS require system to be discrete*

Finite basis is needed

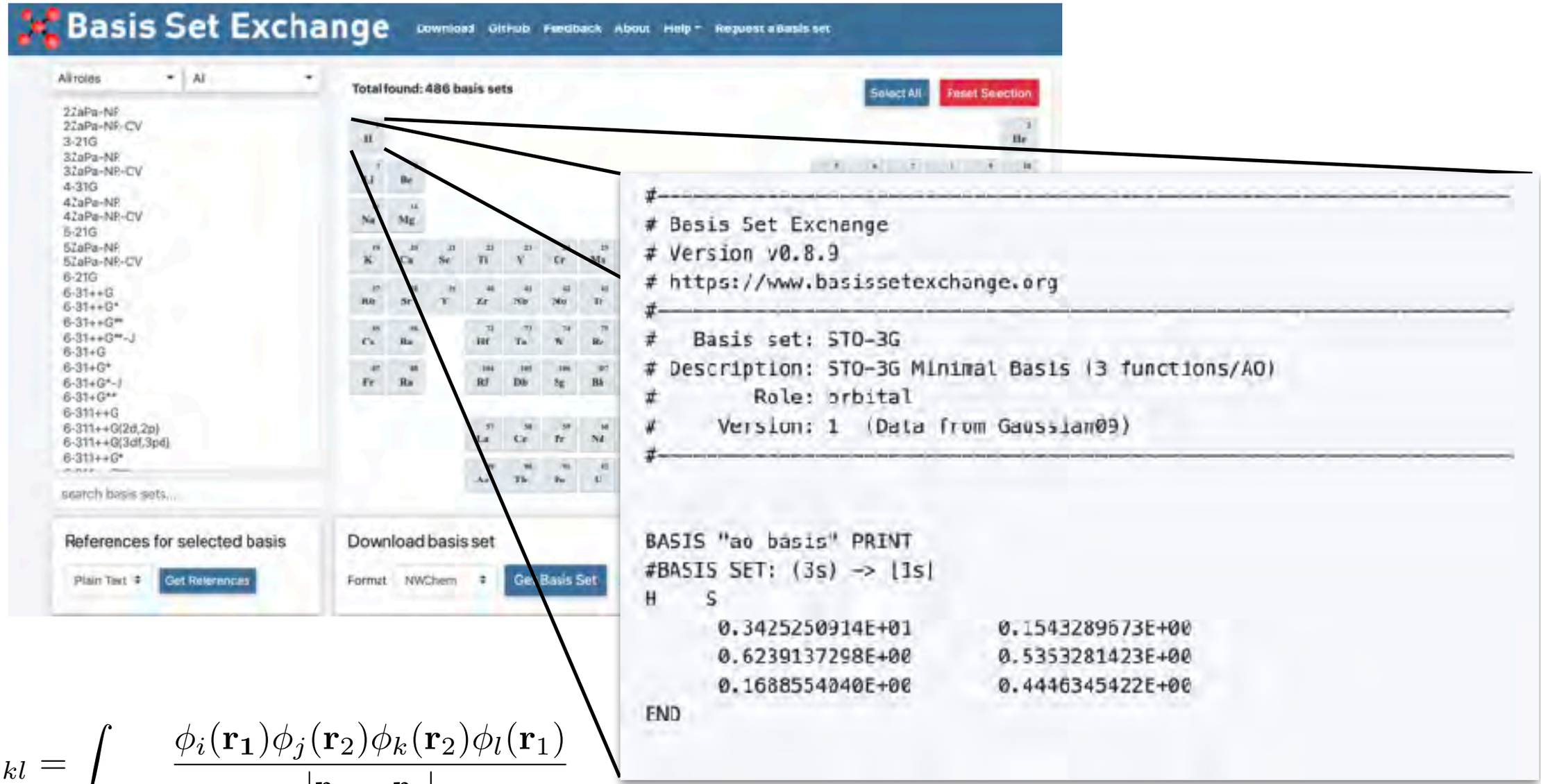
Let's briefly discuss 3 types:

1. Gaussian basis sets [standard]
2. sliced basis sets [new]
3. gausslet basis [new]

* exception are continuous MPS, but still new topic

1. Gaussian basis DMRG (MPS tensor network)

a. choose a Gaussian basis set, orthogonalize basis, & compute integrals t_{ij}, V_{ijkl}

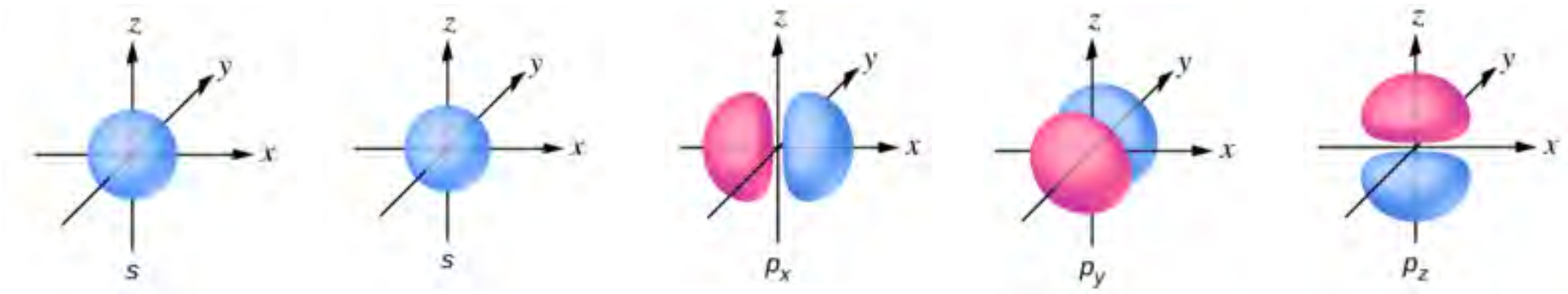


$$V_{ijkl} = \int_{\mathbf{r}_1,\mathbf{r}_2} \frac{\phi_i(\mathbf{r_1})\phi_j(\mathbf{r}_2)\phi_k(\mathbf{r}_2)\phi_l(\mathbf{r}_1)}{|\mathbf{r}_1 - \mathbf{r}_2|}$$

1. Gaussian basis DMRG (MPS tensor network)

b. treat orbitals as "sites" of a pseudo-1D system

$$H = \sum_{ij} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \sum_{ijkl} V_{ijkl} c^\dagger_{i\sigma} c^\dagger_{j\sigma'} c_{k\sigma'} c_{l\sigma}$$



1. Gaussian basis DMRG (MPS tensor network)

c. if converged, obtain state of the art results!

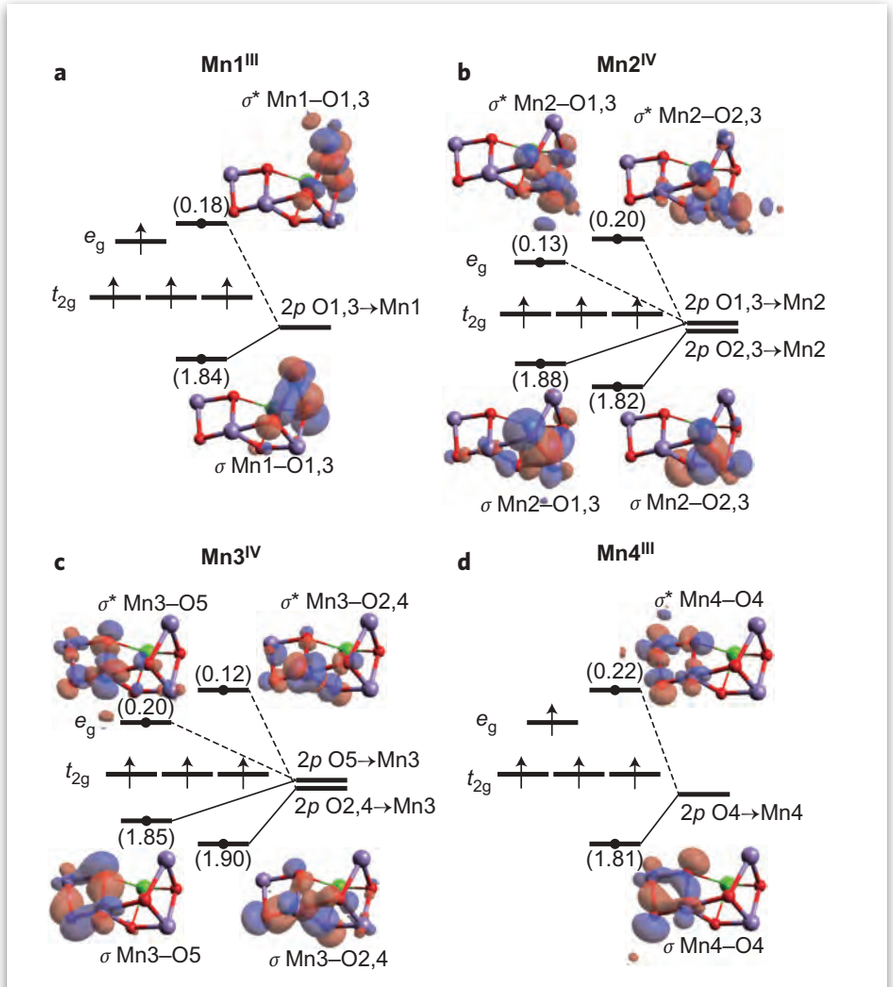
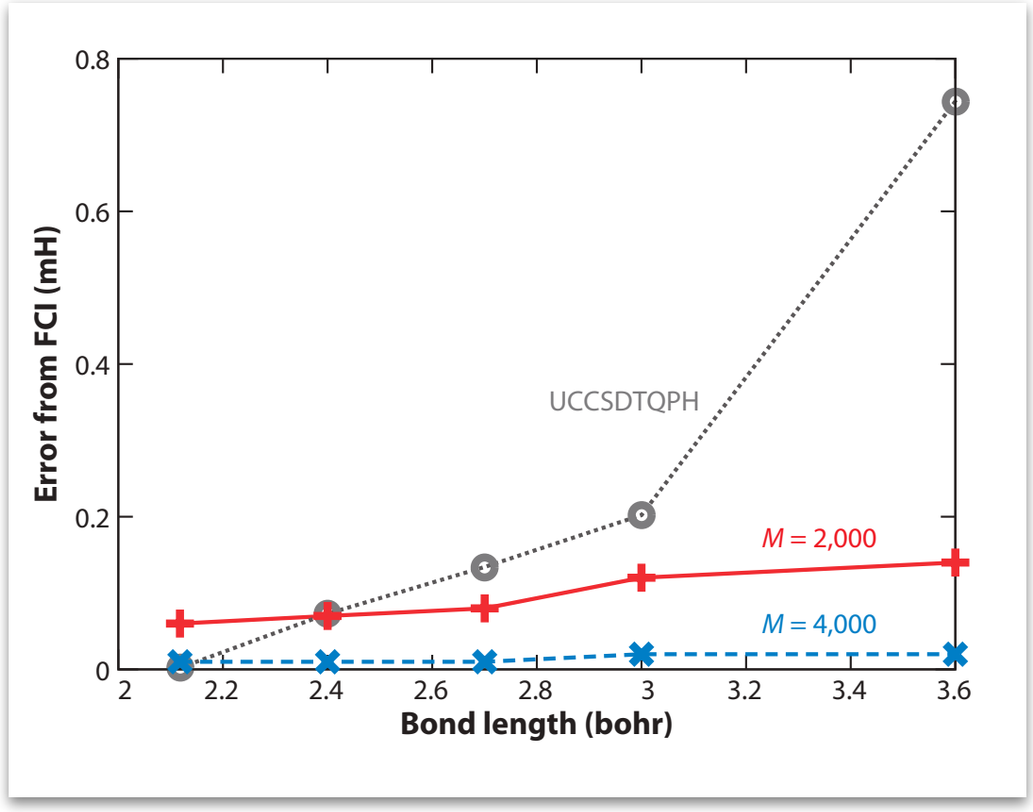


Figure 3 | Orbital energy levels and electron occupancies of four Mn sites for the refined structure. a–d, Orbital energy levels and electron occupancies for Mn1 (**a**), Mn2 (**b**), Mn3 (**c**) and Mn4 (**d**). The layout and labelling follows Fig. 1 (XRD model).

Kurashige, Chan, Yanai, *Nat. Chem.* (2013)
Study of photosystem-II

Error from exact soln of binding energy of N_2

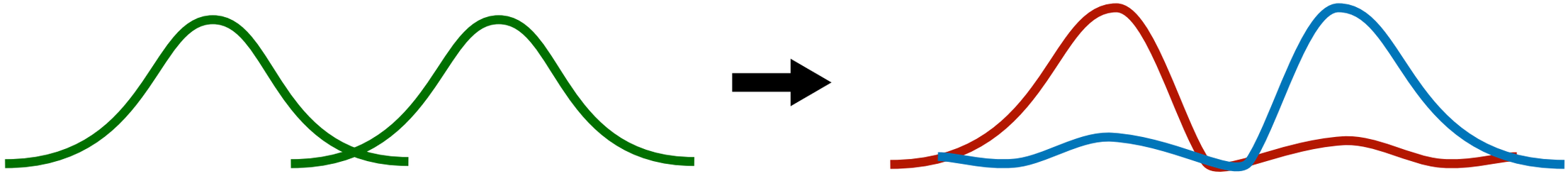


Chan, Sharma, *Ann. Rev. Phys. Chem.* (2011)
Review: DMRG in Quantum Chemistry

1. Gaussian basis DMRG (MPS tensor network)

Drawbacks of this approach?

Gaussian basis functions overlap significantly
(especially after orthogonalization)



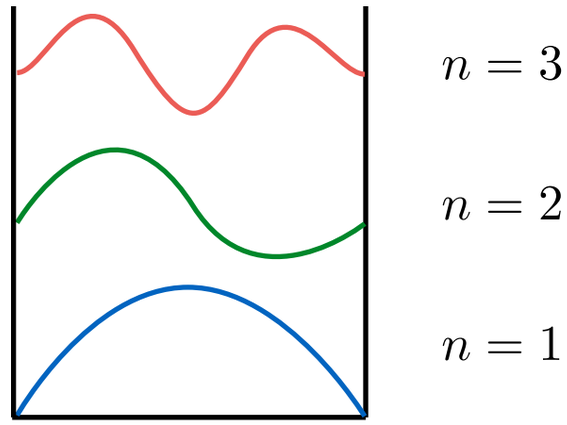
Must keep all N^4 Coulomb integrals (can't truncate)

Hamiltonian non-local; DMRG scales poorly
(large MPS bond dimension required)

Alternatives to Gaussian basis set approach?

Consider 1D particles in a box:

Approach 1: basis set $c_n = \int_x \phi_n(x)\hat{\psi}(x)$

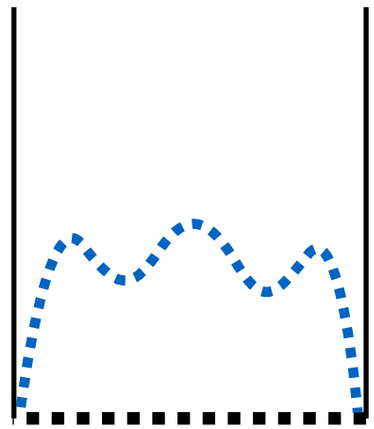


$$H = -\frac{1}{2}\int_x \hat{\psi}^\dagger(x)\frac{\partial^2}{\partial x^2}\hat{\psi}(x) \quad \rightarrow \quad H = \sum_{nm} t_{nm} c_n^\dagger c_m$$

- Loss of locality
- Must compute integrals
+ Variational

Consider 1D particles in a box:

Approach 2: grid approximation $c_j = \sqrt{a}\ \ \hat{\psi}(x_j)$



$$H = -\frac{1}{2}\int_x \hat{\psi}^\dagger(x)\frac{\partial^2}{\partial x^2}\hat{\psi}(x)$$

↘

$$H \approx -\frac{1}{2a^2}\sum_j (c_j^\dagger c_{j+1} - 2n_j + c_{j+1}^\dagger c_j) + \mathcal{O}(a^2)$$

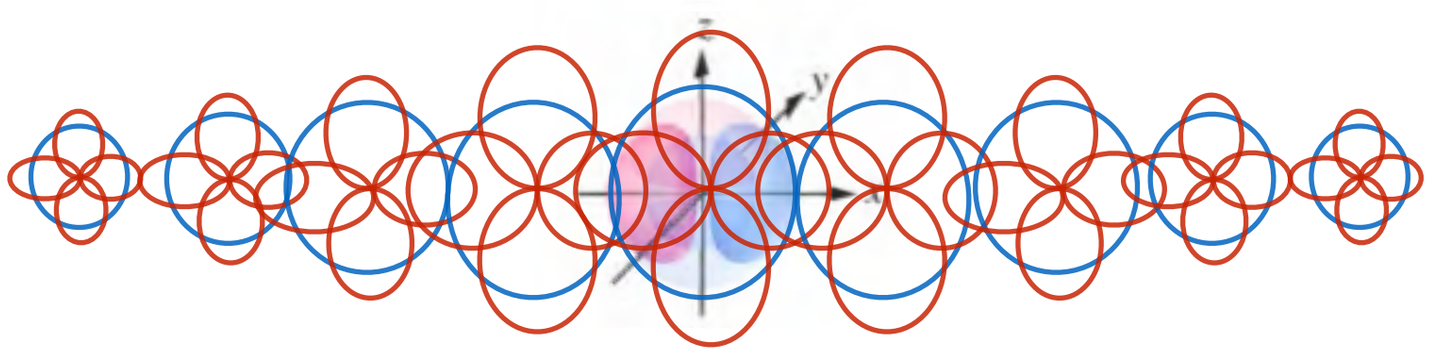
+ Local / short range
+ No integrals to compute!
- Not variational

Possible to mix basis set and grid approaches?

Yes...

2. sliced-basis DMRG [new]

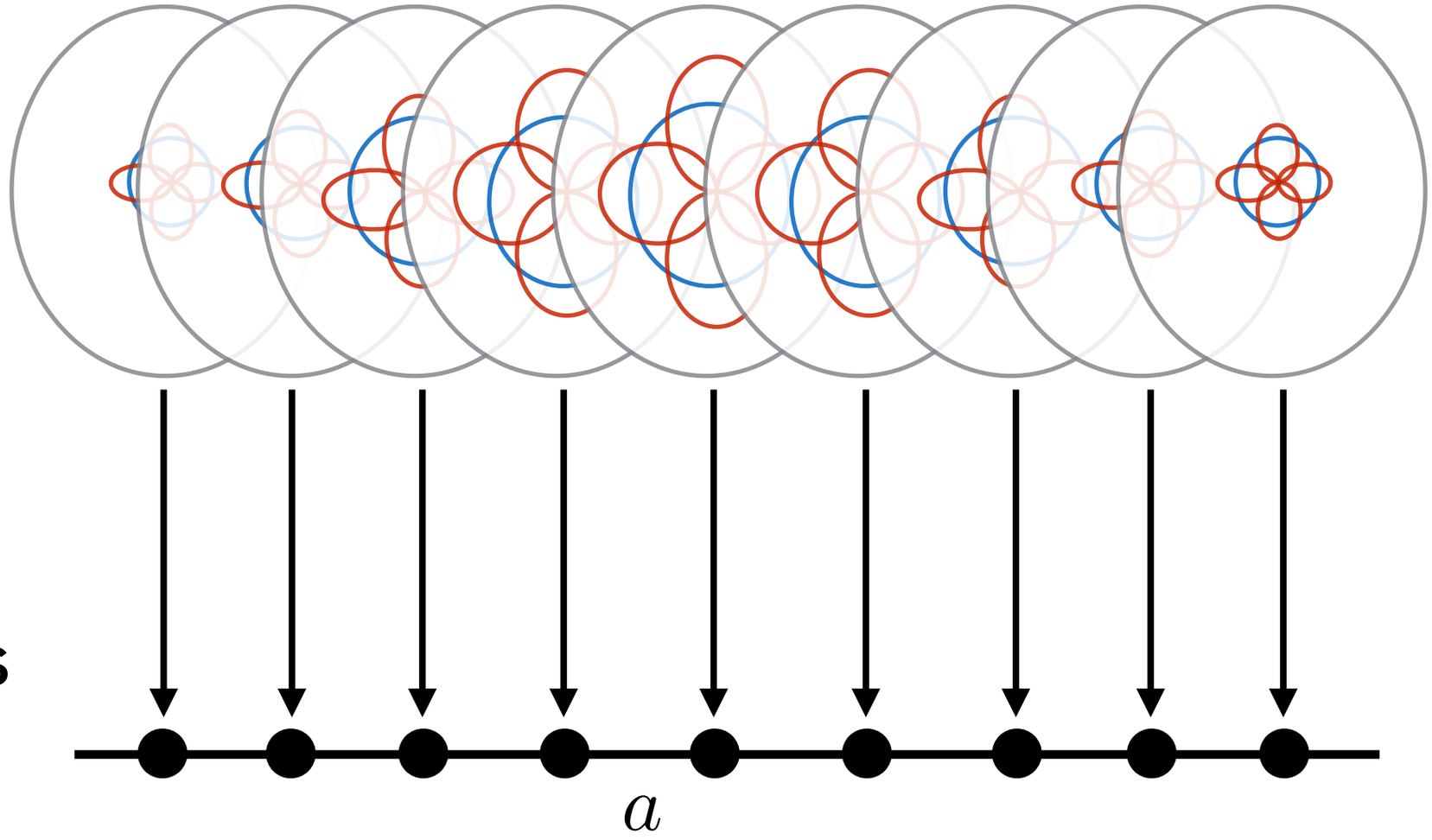
Slice 3D basis sets along z-direction:



Stoudenmire, White, arxiv:1702.03650

2. sliced-basis DMRG [new]

Slice 3D basis sets along z-direction:



Map to 1D 'chain' with 1000's of sites (small $\Delta z = a$):

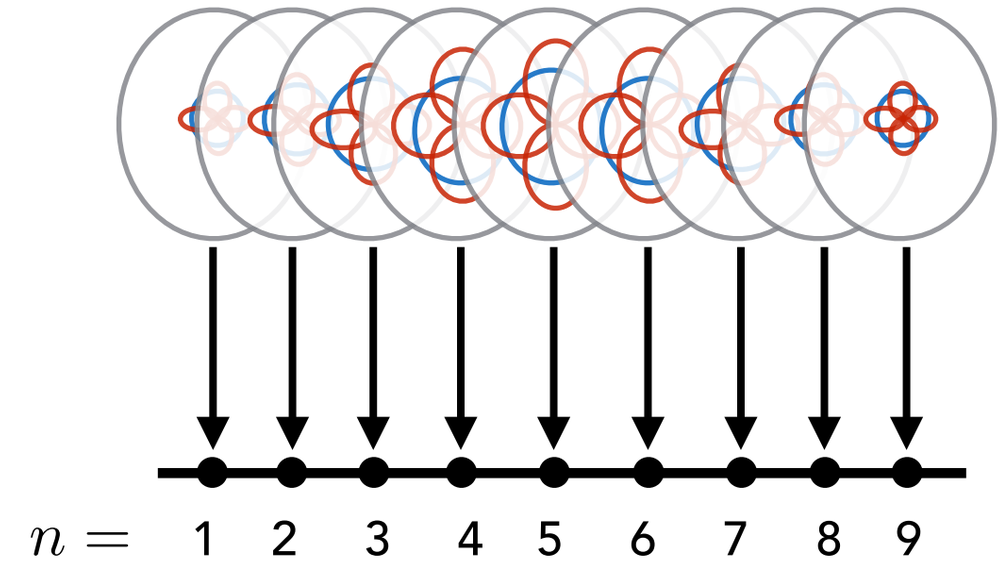
- Leverage ability of DMRG to scale to long systems
- Can reach high (chemical) accuracy for $a \lesssim 0.1$
- Scalable to 1000's of atoms

Stoudenmire, White, arxiv:1702.03650

2. sliced-basis DMRG [new]

Slices roughly equivalent to using basis set of "functions":

$$\phi_{nj}(\mathbf{r}) = \delta^{\frac{1}{2}}(z - n\cdot a)\,\varphi_{nj}(x,y)$$



Stoudenmire, White, arxiv:1702.03650

2. sliced-basis DMRG [new]

$$\phi_{nj}(\mathbf{r}) = \delta^{\frac{1}{2}}(z - n\cdot a)\,\varphi_{nj}(x,y)$$

Interaction Energy:

Normally must deal with N^4 interaction terms

$$V_{ijkl} = \int_{\mathbf{r}_1,\mathbf{r}_2} \frac{\phi_i(\mathbf{r_1})\phi_j(\mathbf{r}_2)\phi_k(\mathbf{r}_2)\phi_l(\mathbf{r}_1)}{|\mathbf{r}_1 - \mathbf{r}_2|}$$

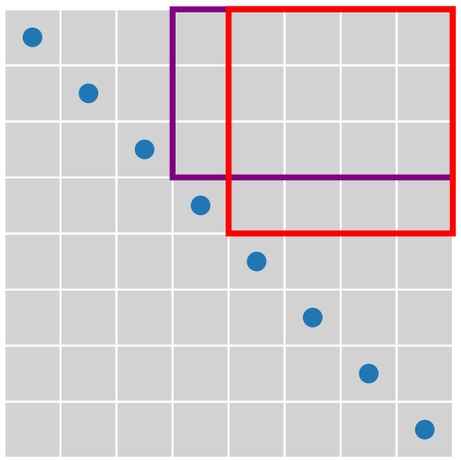
But treat slices as orthogonal. Then V_{ijkl} non-zero only if i,l on same slice and j,k on same slice

Number of terms: $N^4 \to N_z^2 N_{\mathrm{orb}}^4$ *Better Scaling*

Stoudenmire, White, arxiv:1702.03650

2. sliced-basis DMRG [new]

[Technical Slides] – Important for Practitioners



$$V_{ij} = \int_{\mathbf{r},\mathbf{r}'} \frac{|\phi_i(\mathbf{r})|^2 |\phi_j(\mathbf{r}')|^2}{|\mathbf{r}-\mathbf{r}'|}$$

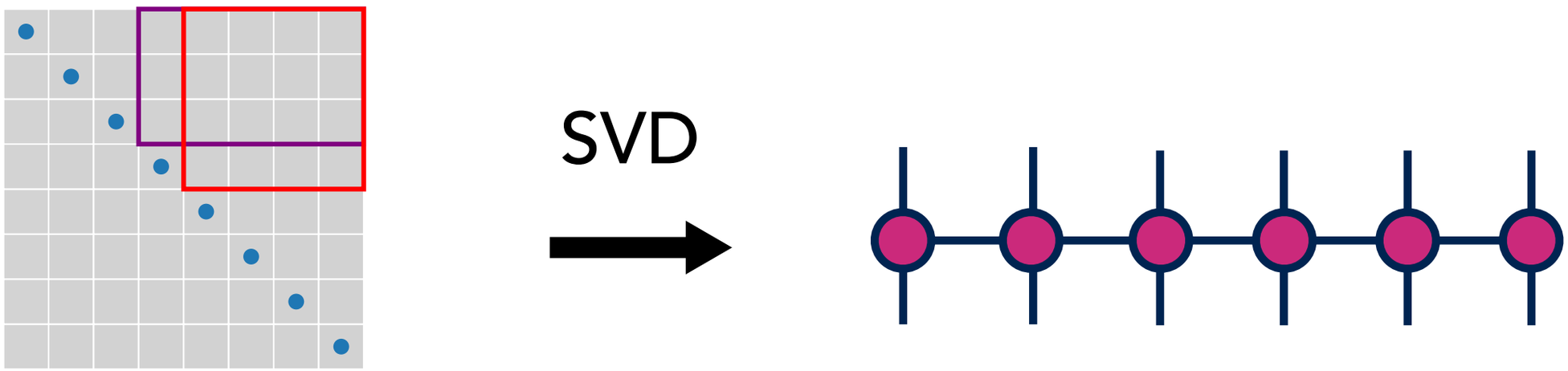
Sliced Coulomb integrals have upper-triangular-low-rank property (SVD of upper triangles compresses well)

Lin, Tong, arxiv:1909.02206

Stoudenmire, White, arxiv:1702.03650

2. sliced-basis DMRG [new]

[Technical Slides] – Important for Practitioners



From SVD's of all upper triangle blocks,
construct efficient matrix product operator (MPO)
of Coulomb Hamiltonian terms

Lowers scaling all the way to *linear* in number of z slices (number of atoms)

Lin, Tong, arxiv:1909.02206

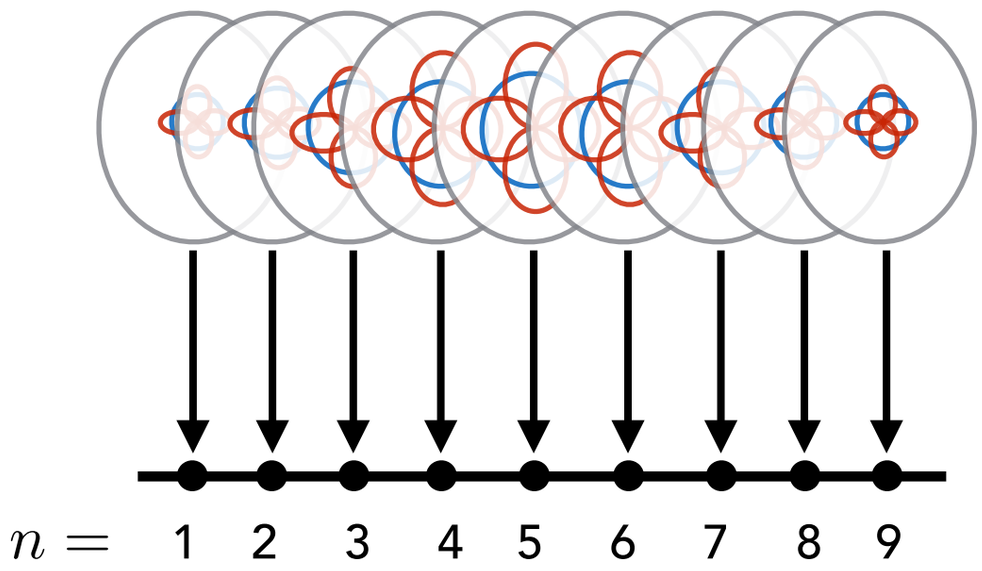
Stoudenmire, White, arxiv:1702.03650

Possible to do better?

3. multi-sliced gausslet basis [new]

Drawback of sliced bases include:

- require very many slices to resolve z-direction
- problems with Gaussians in x,y directions



3. multi-sliced gausslet basis [new]

Seek functions which are local (compact), orthogonal, and sums of them can represent any smooth function

Can find such functions using *theory of wavelets*

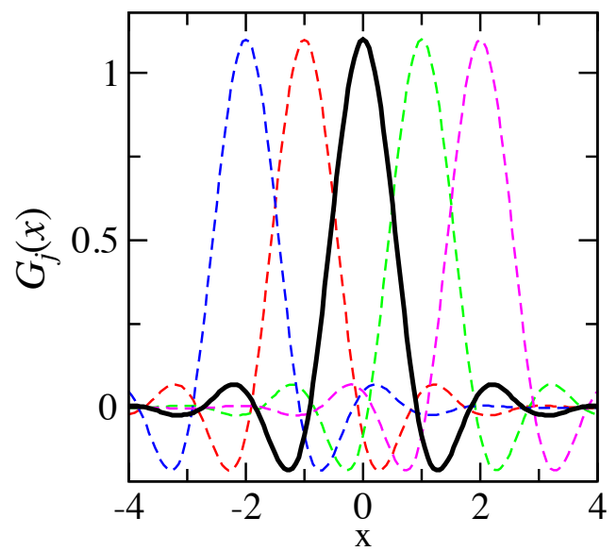
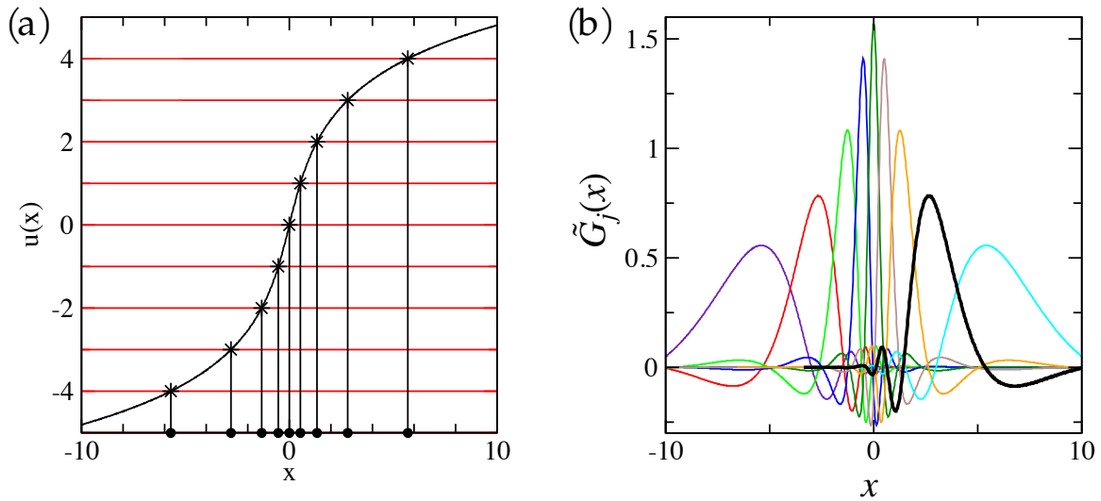


Fig. 4: *One-dimensional array of gausslet functions with a length scale of 1.0. The gausslet centered at the origin is highlighted in solid black to emphasize details.*

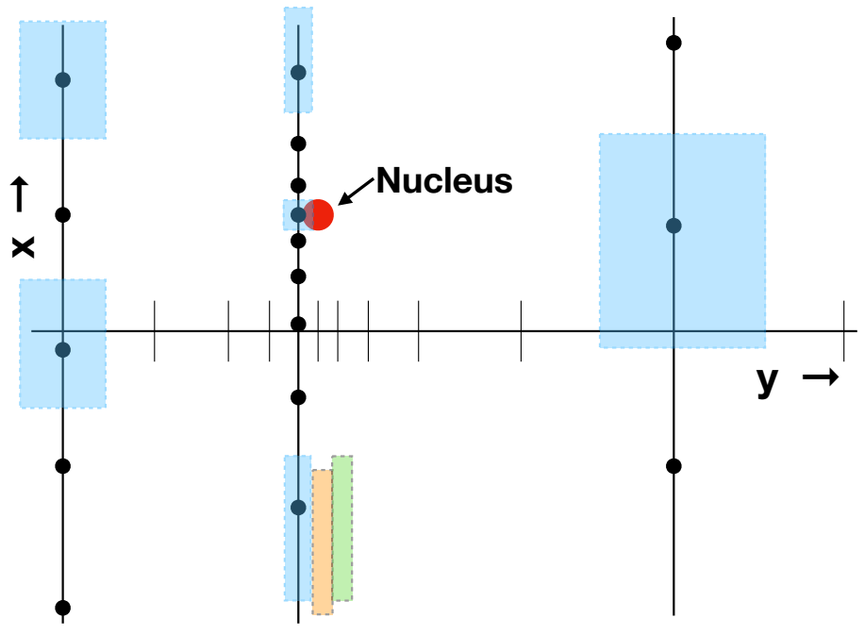
Gausslets = wavelet (scaling functions) built from Gaussians

3. multi-sliced gausslet basis [new]

By scale-adapting and "multi-slicing"
can represent smooth 3D functions



scale-adaptation

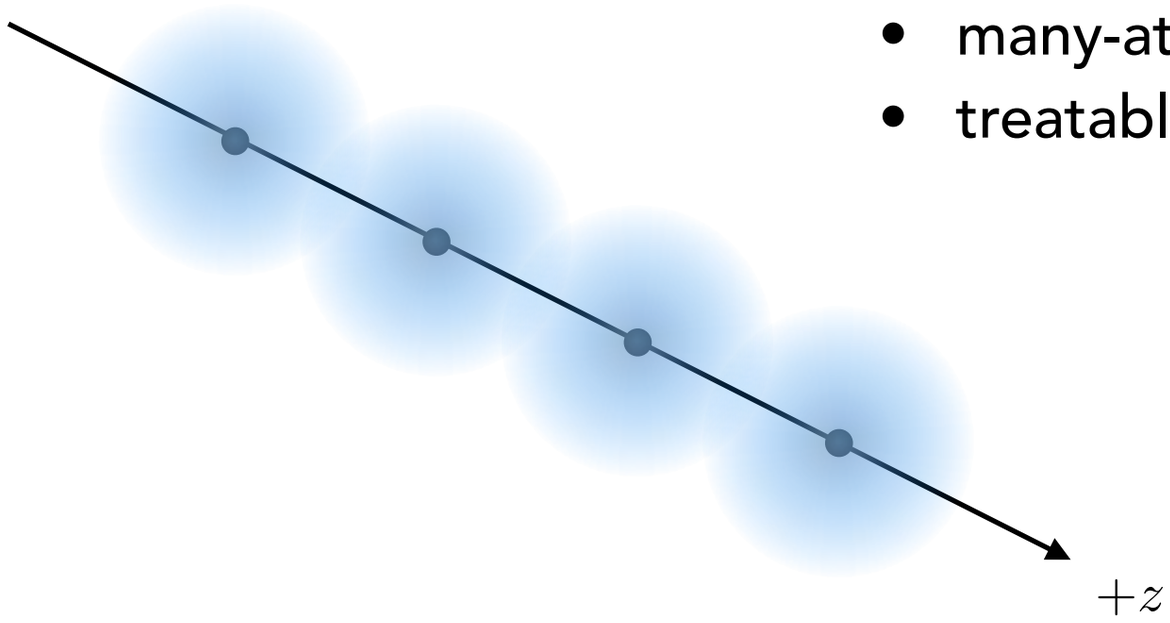


multi-slicing

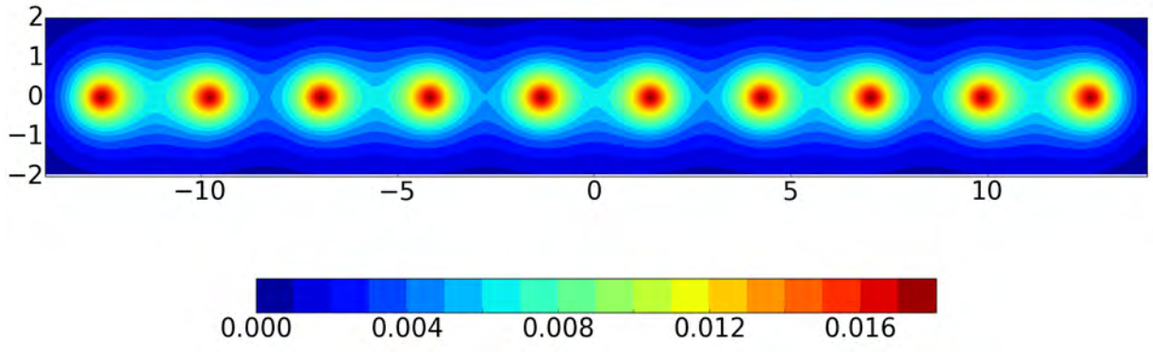
Results of sliced-basis and multi-sliced gausslets

Hydrogen chains make good benchmark systems

- continuum limit
- strong correlation
- many-atom (thermodynamic) limit
- treatable by most methods



Density cross-section of H_{10} hydrogen chain:



Sliced-basis results for H_{10}:

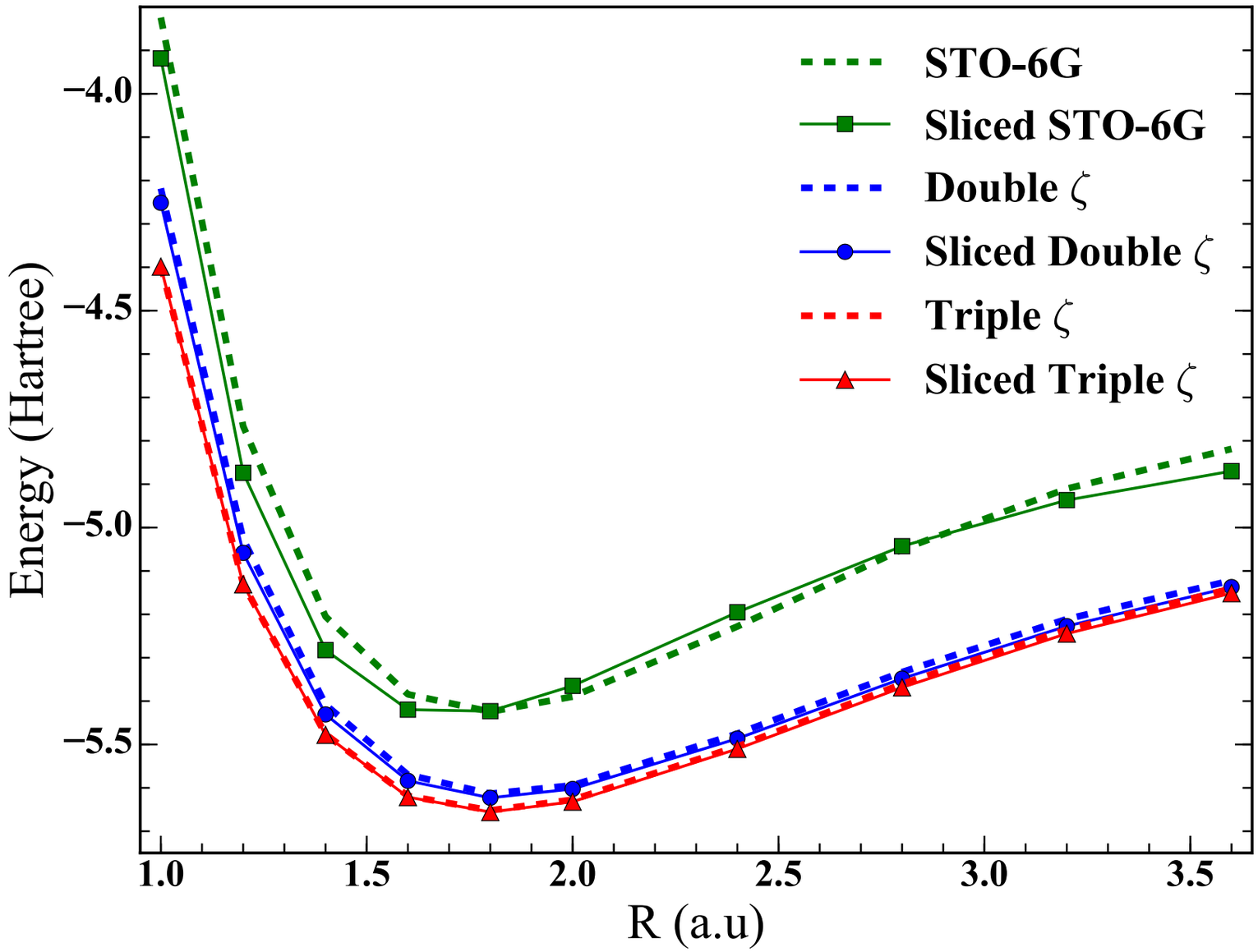


Fig. 2: *Ground-state energies of H_{10} chains as a function of inter-atomic spacing R calculated using DMRG within standard Gaussian basis sets (dashed curves) and sliced basis sets (solid curves and points) using a uniform grid spacing of $a = 0.1$ atomic units [26].*

Sliced-basis results for H_{1000}:

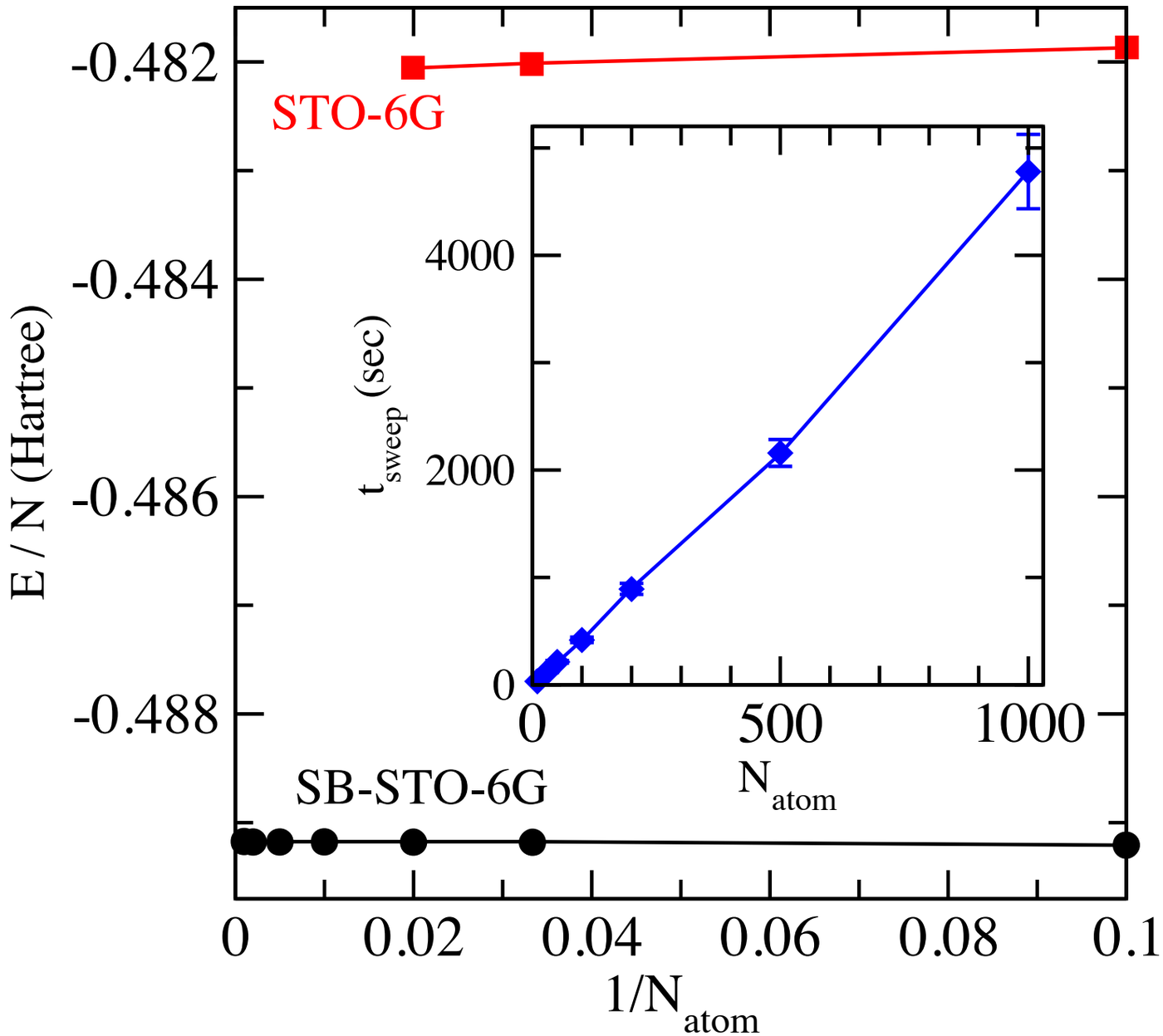
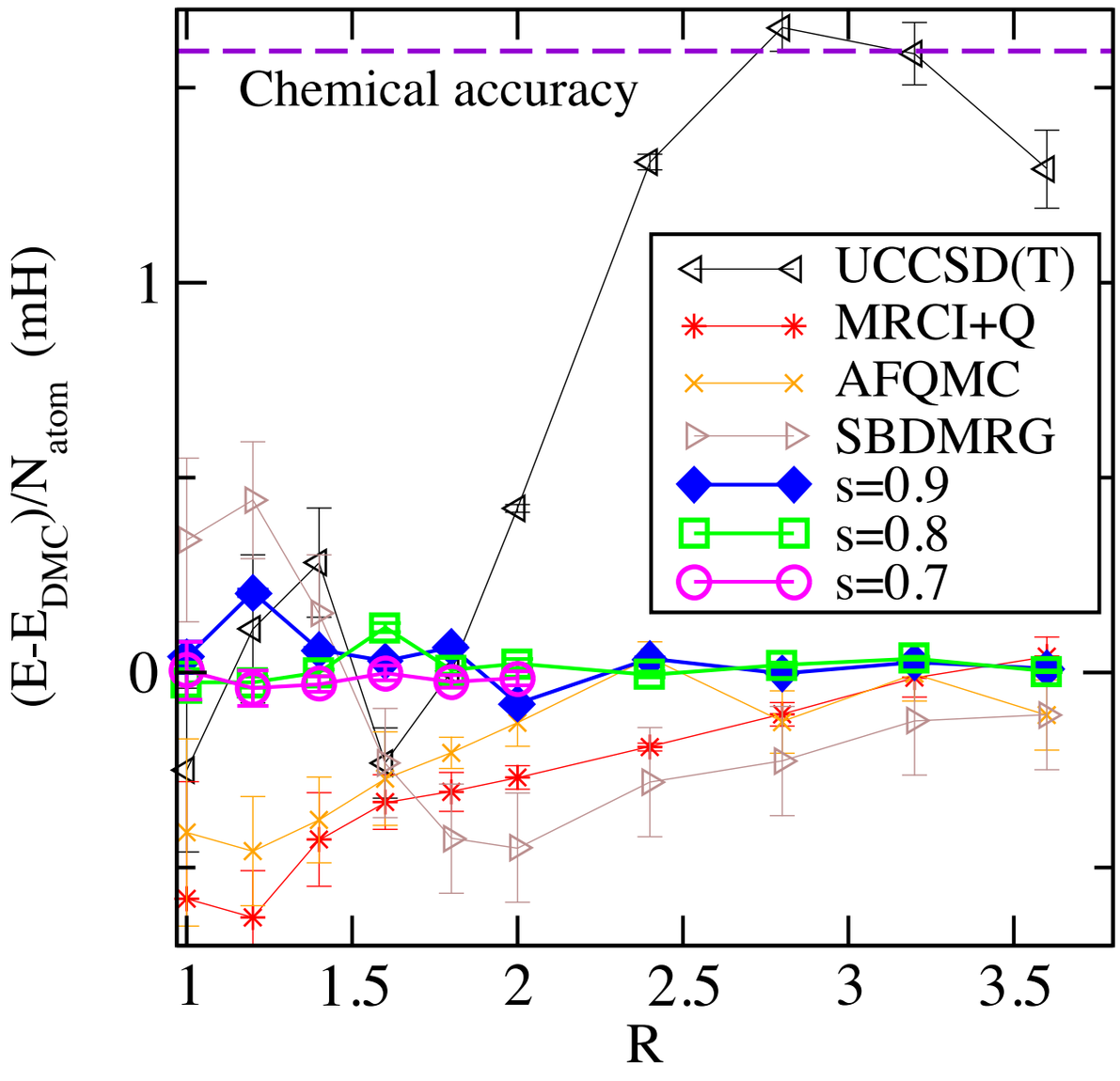


Fig. 3: *Scaling with number of atoms of sliced-basis calculations up to 1000 hydrogen atoms. The inter-atomic spacing is fixed to* $R = 3.6$ *and a sliced basis derived from the STO-6G Gaussian basis was used. The outer plot shows the ground state energy from DMRG using the standard STO-6G basis and the sliced version (SB-STO-6G). The inset shows the average time per DMRG sweep, taking a bond dimension of* $m = 100$.

Multi-sliced-gausslet (MSG) results for H_{10}:

Comparison to diffusion Monte Carlo (DMC)

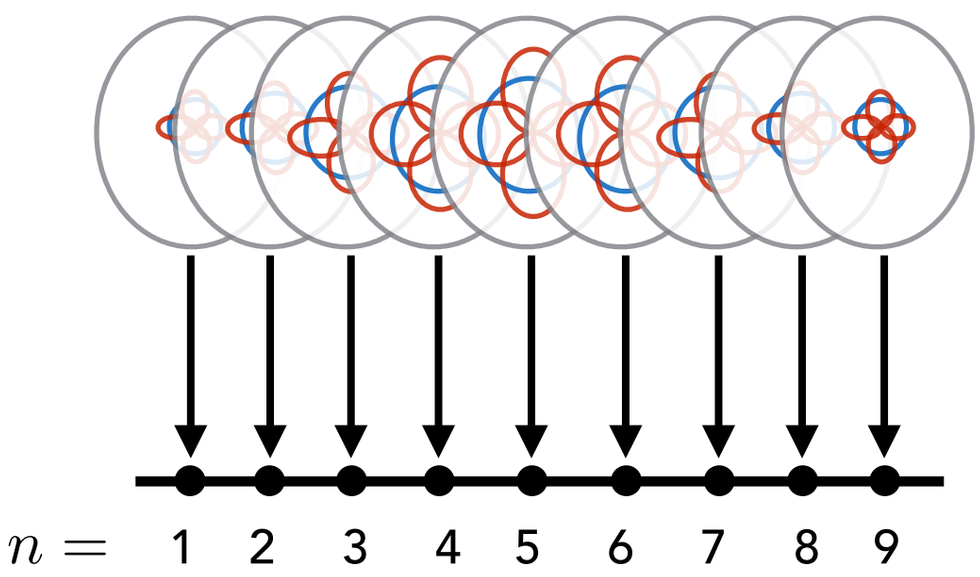


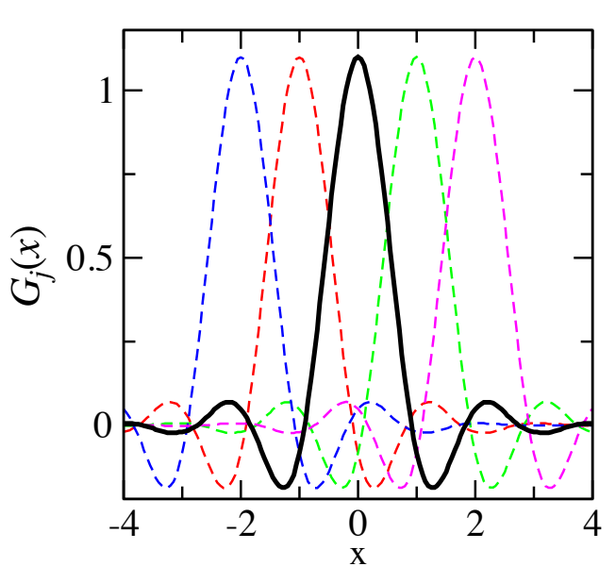
} MSG-DMRG

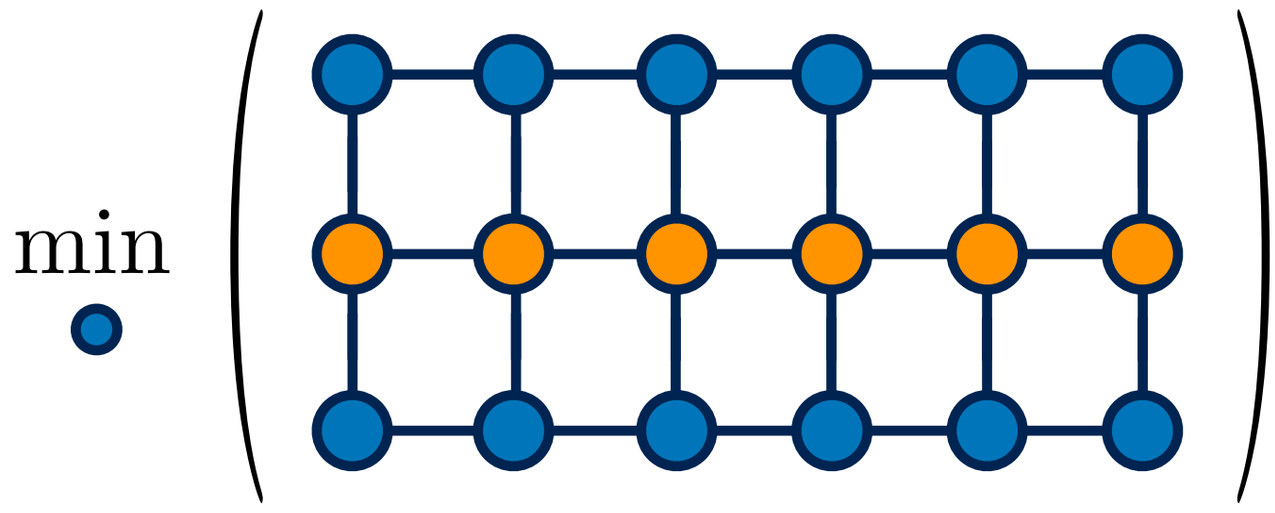
Future Directions & Conclusions

A lot of creativity is possible in going beyond standard Gaussian approach to quantum chemistry

Sliced-basis and gausslet basis are just two of many ideas to be tried





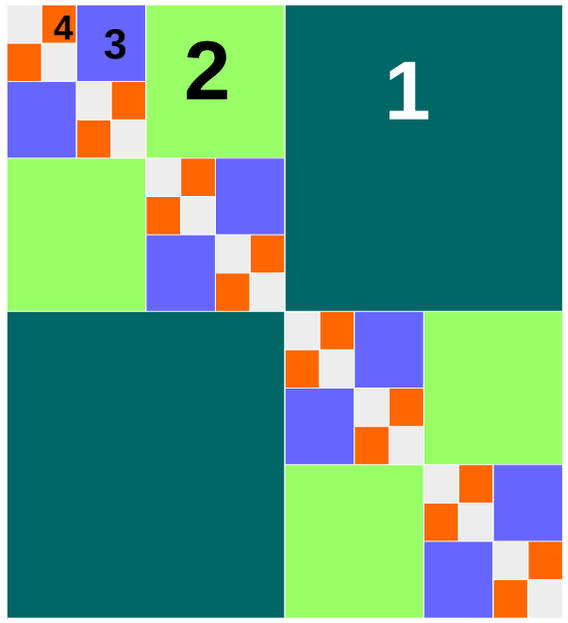


Tensor networks are real-space oriented, don't use Slater determinants

Require re-thinking conventional choices to get best performance

Recent work by Lin and Tong for compressing Coulomb interactions into "PEPO" tensor networks

V_{ij}



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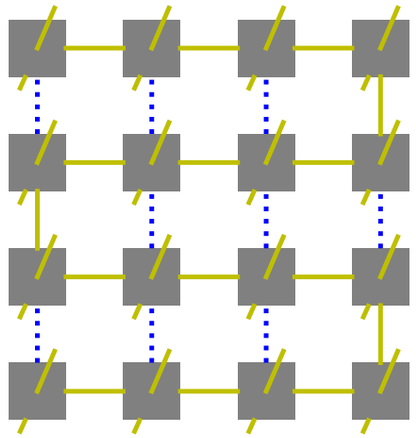

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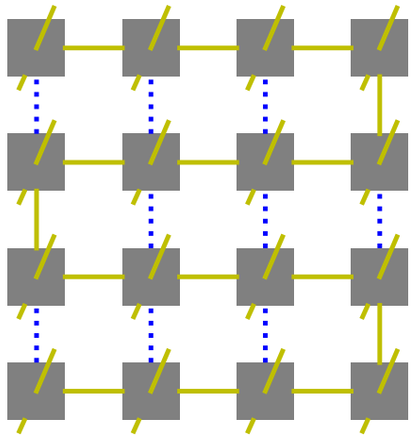

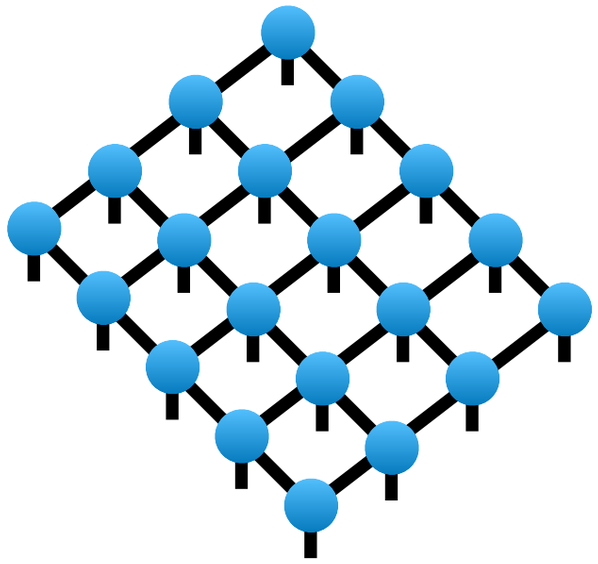

Combined with recent progress in optimizing PEPS*,
could soon see PEPS quantum chemistry!

In principle scalable to huge 2D planes of atoms,
controlled & accurate, handling strong correlation

* Liao, Liu, Wang, Zhang, arxiv:1903.09650
Zaletel, Pollmann, 1902.05100
Haghshenas, O'Rourke, Chan, 1903.03843
Liu, Huang, Gong, Gu, 1908.09359
Hyatt, Stoudenmire, 1908.08833