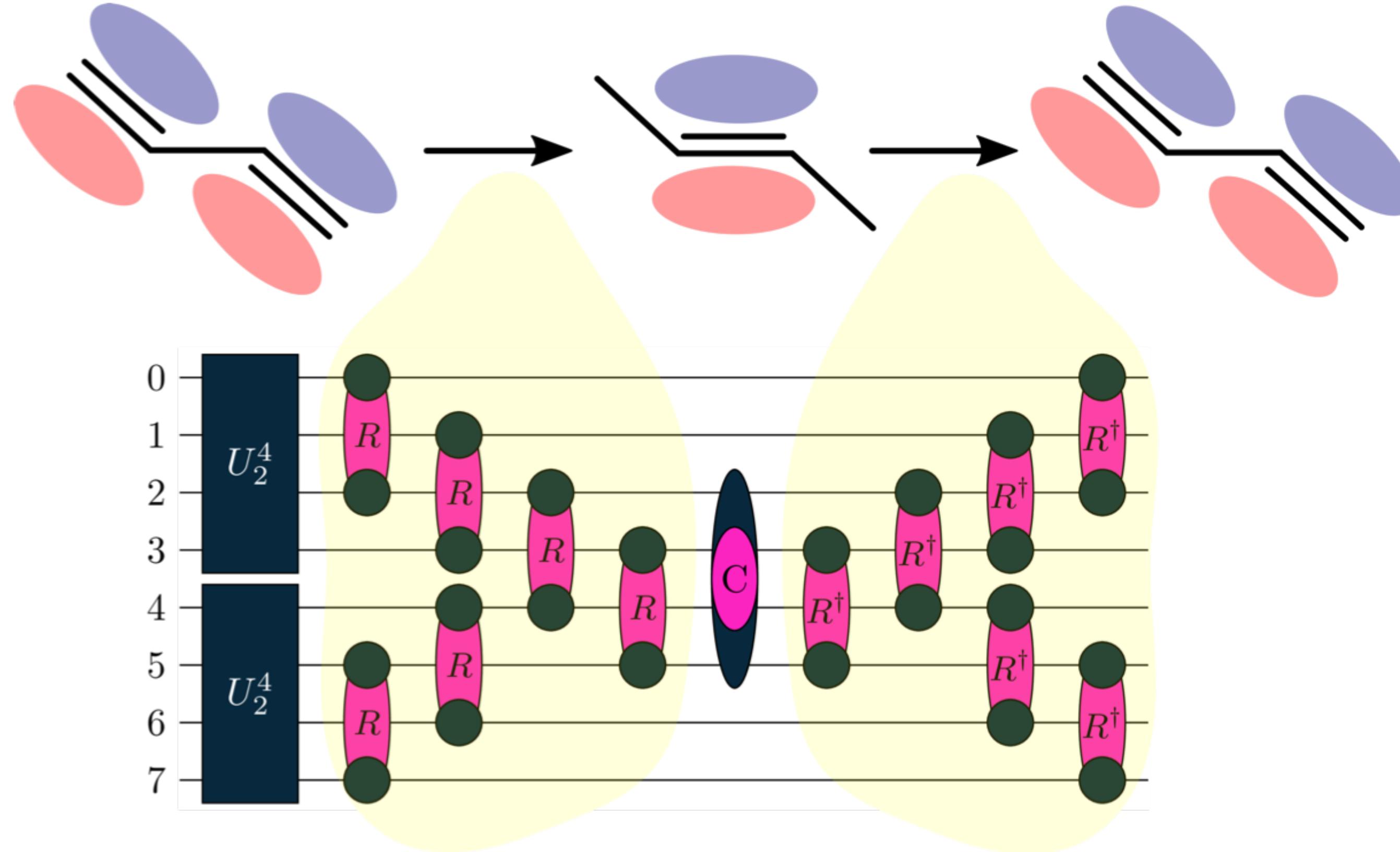


# Molecular Quantum Circuit Design

 @JakobKottmann

 [github/tequilahub](https://github.com/tequilahub)

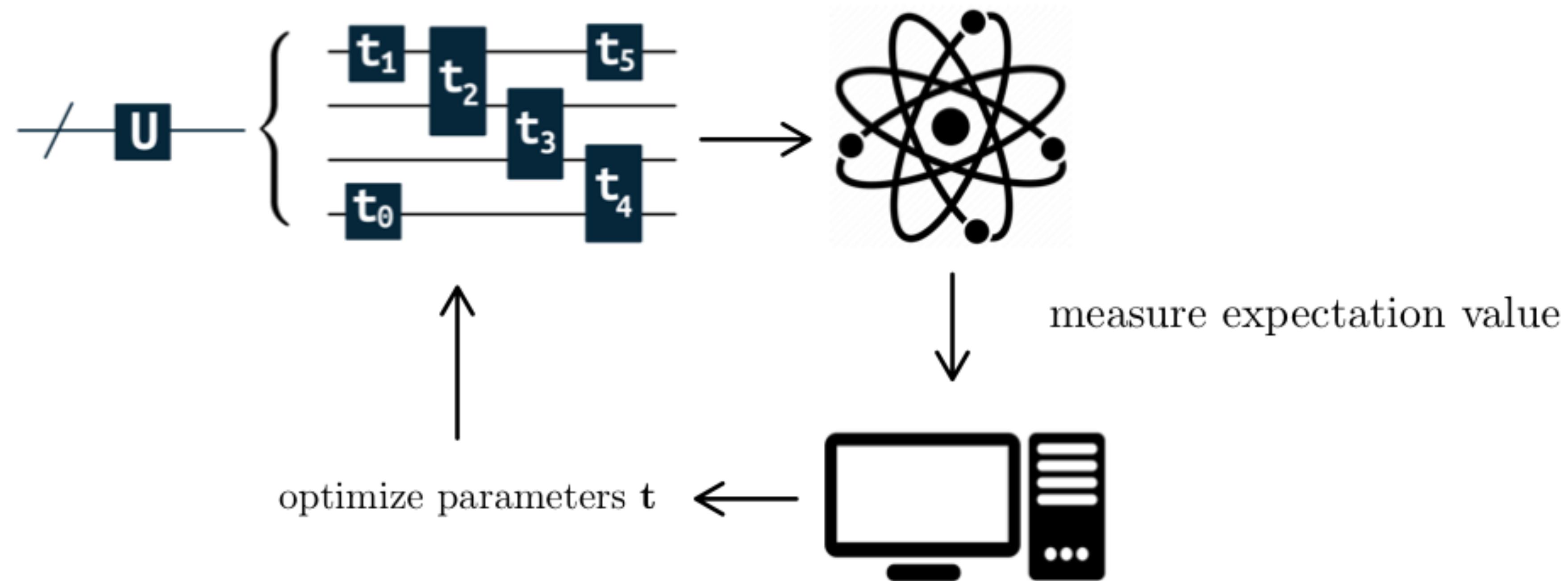


# Graph Based Molecular Quantum Circuit Design

Jakob S. Kottmann<sup>\*</sup>  
(Dated: July 20, 2022)

Optimized Low-Depth Quantum Circuits for Molecular Electronic Structure using  
a Separable Pair Approximation

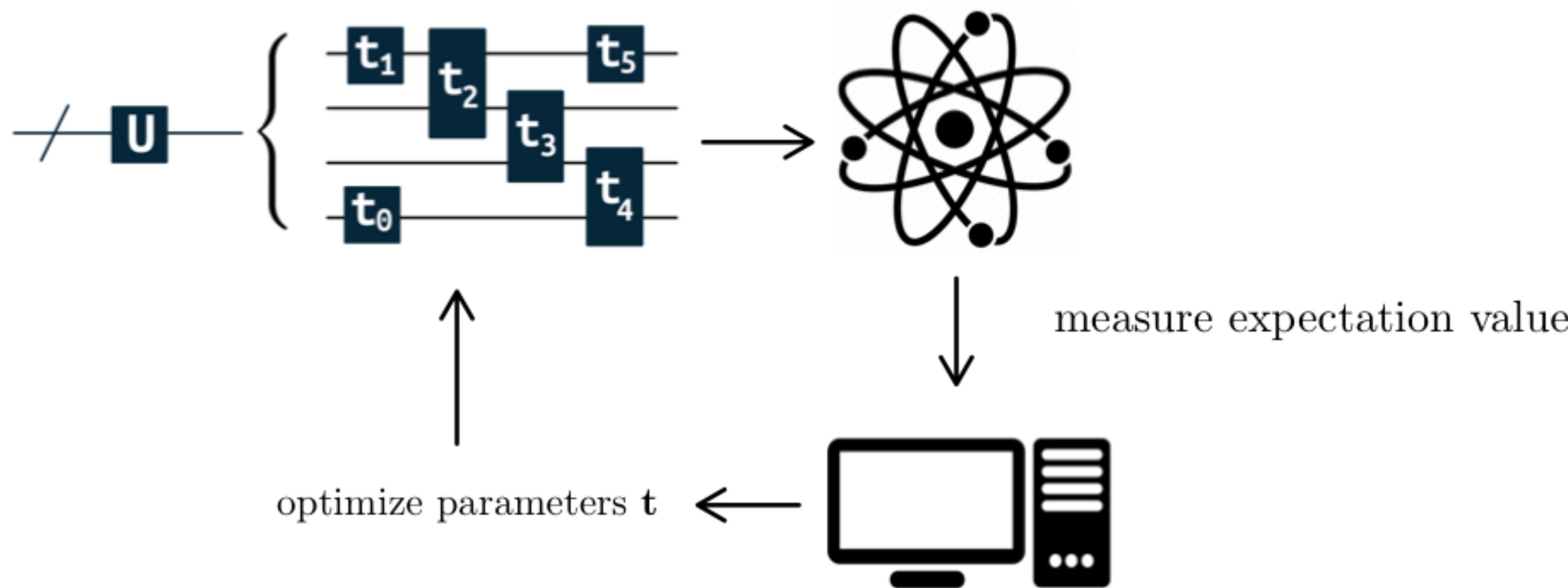
Jakob S. Kottmann<sup>1, 2, \*</sup> and Alán Aspuru-Guzik<sup>1, 2, 3, 4, †</sup>

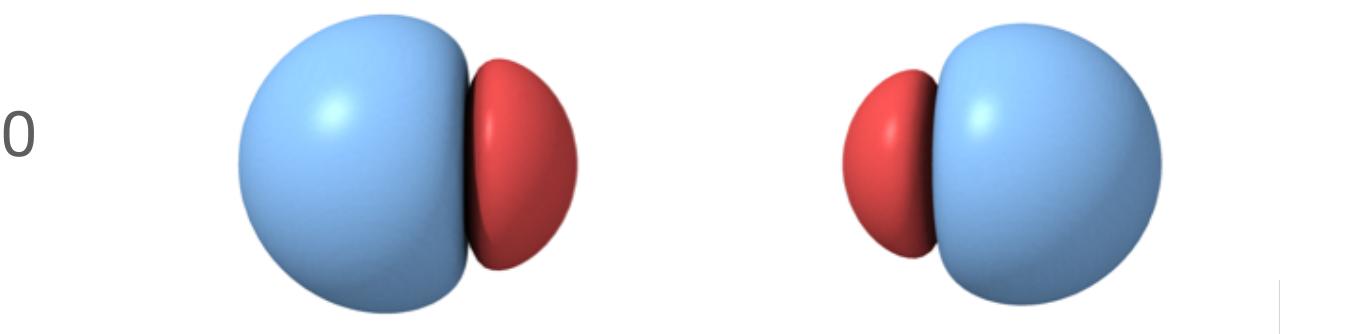
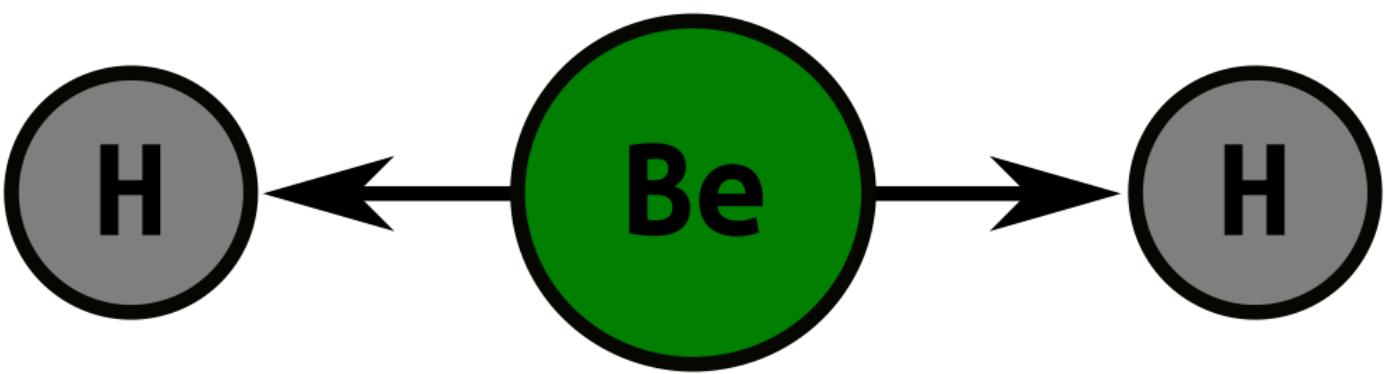


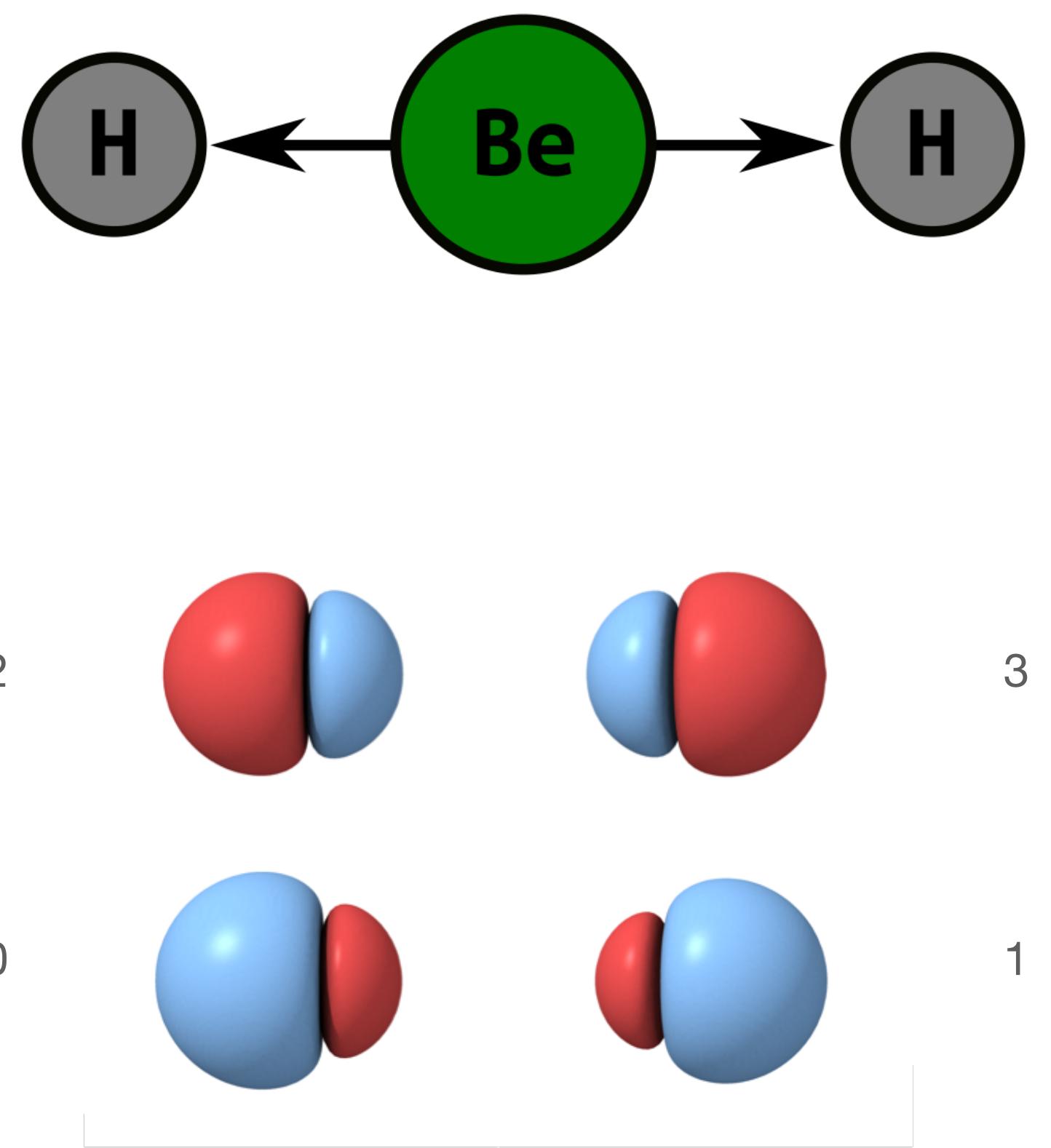
Design principles



- Locality (circuit connections)
- Shallow depth
- Hardware efficient
- Good convergence
- Initialisation heuristics



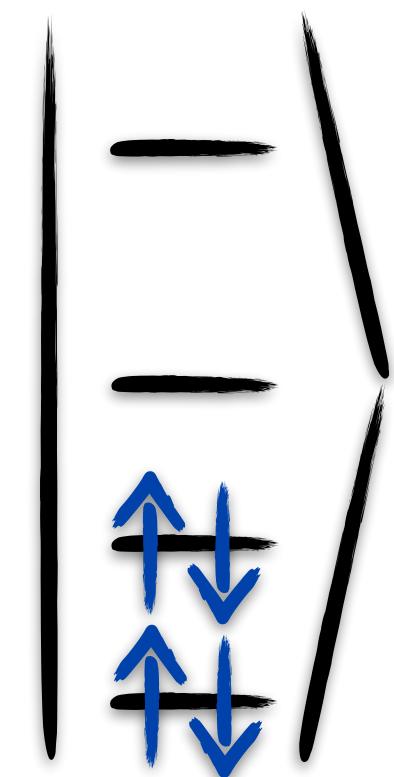




# Empty orbitals 2,3

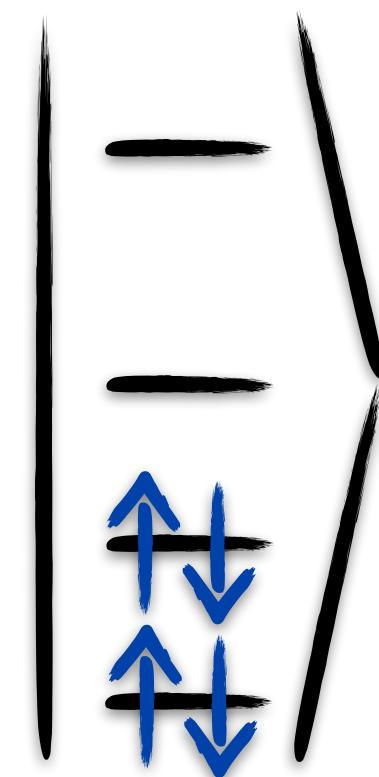
# Two electrons in orbital 1

# Two electrons in orbital 0



$|11\ 11\ 00\ 00\rangle$ 

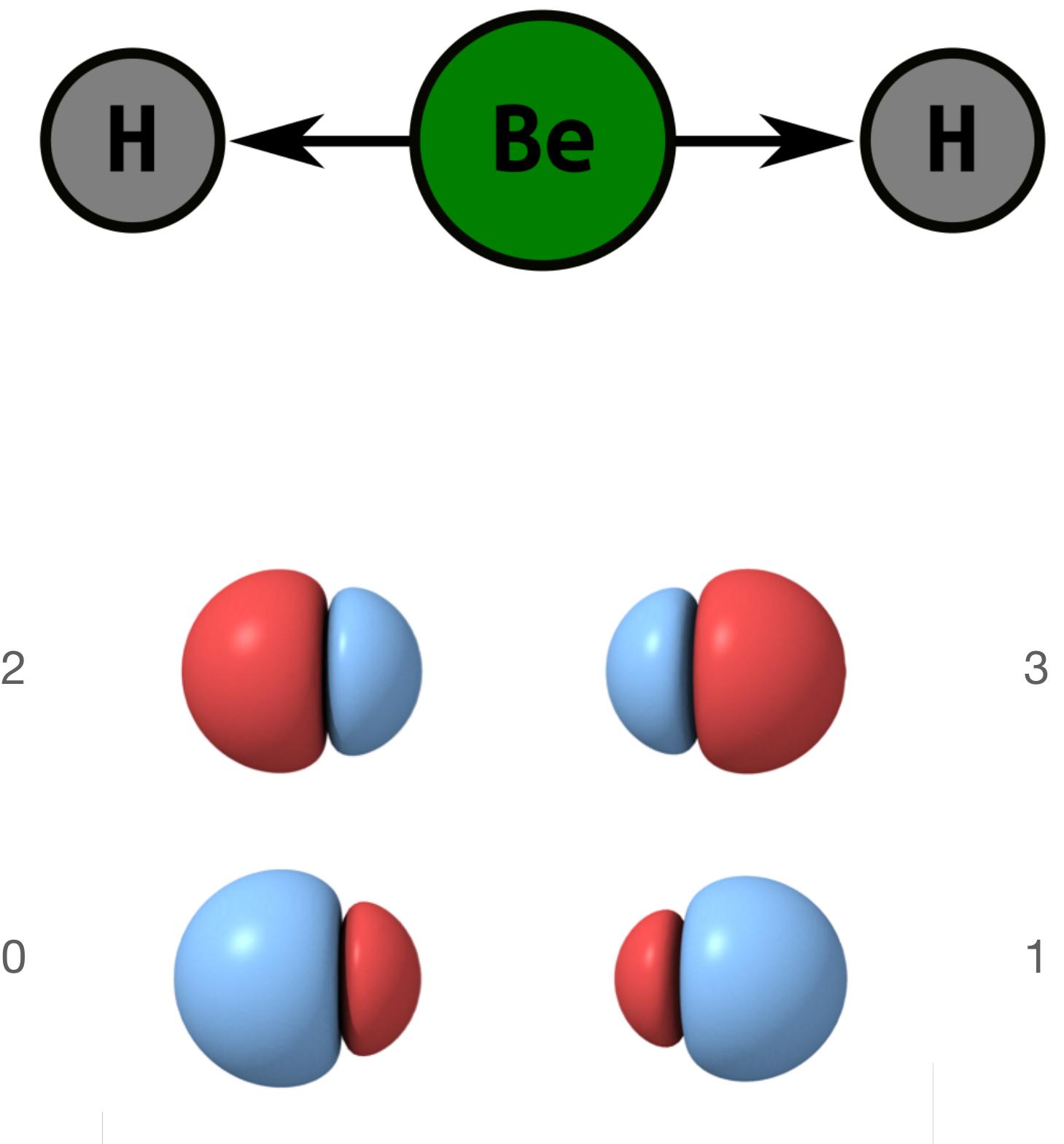
Qubit encoding



Empty orbitals 2,3

Two electrons in orbital 1

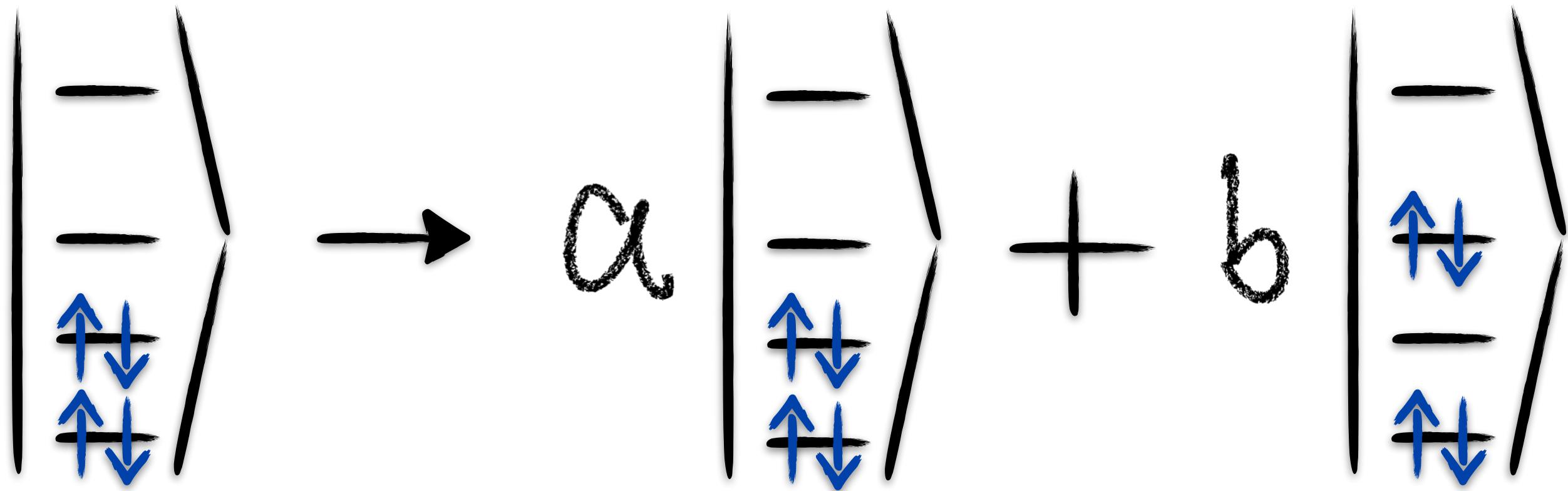
Two electrons in orbital 0



$$\left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \downarrow \end{array} \right\rangle \rightarrow a \left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \downarrow \end{array} \right\rangle + b \left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \end{array} \right\rangle + c \left| \begin{array}{c} + \\ - \\ \uparrow \downarrow \\ - \\ \downarrow \end{array} \right\rangle$$

# **Building Blocks**

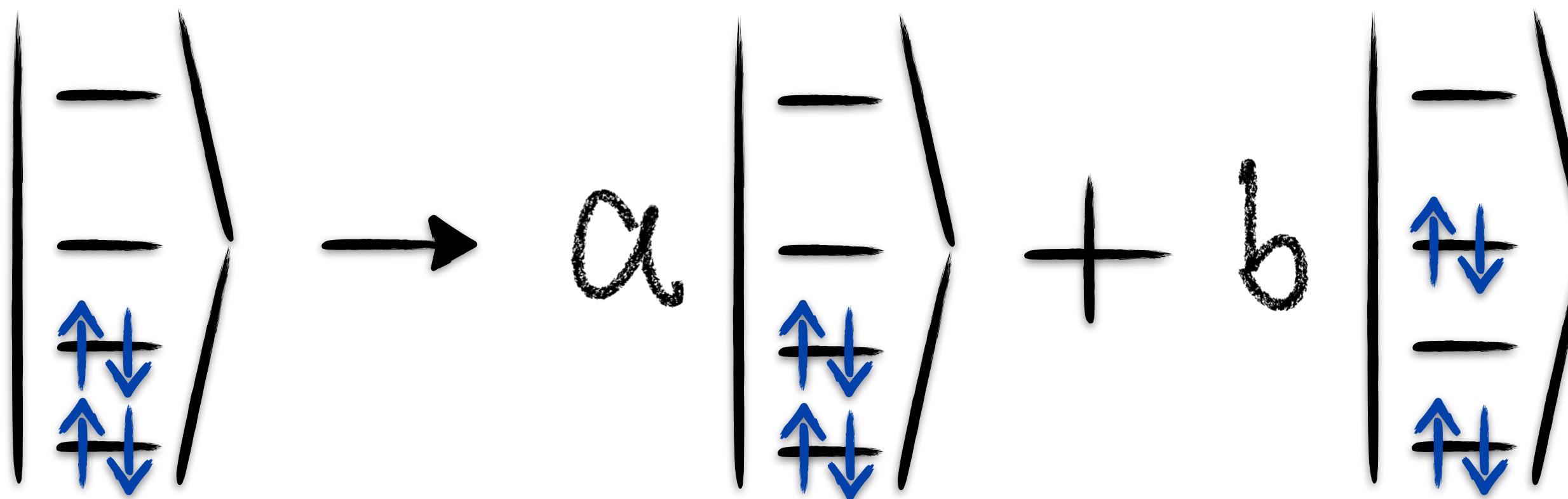
$$\left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \downarrow \end{array} \right\rangle \rightarrow a \left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \downarrow \end{array} \right\rangle + b \left| \begin{array}{c} - \\ - \\ \uparrow \downarrow \\ \uparrow \downarrow \end{array} \right\rangle$$



$$U_{\mathbf{pq}}(\theta) = e^{-i \frac{\theta}{2} G_{\mathbf{pq}}}$$

$$G_{\mathbf{pq}} = i \left( \prod_k a_{p_k}^\dagger a_{q_k} - h.c. \right)$$

$$a_p^\dagger \xrightarrow[Wigner]{Jordan} \sigma_p^+ \prod_{k < p} \sigma_k^z.$$



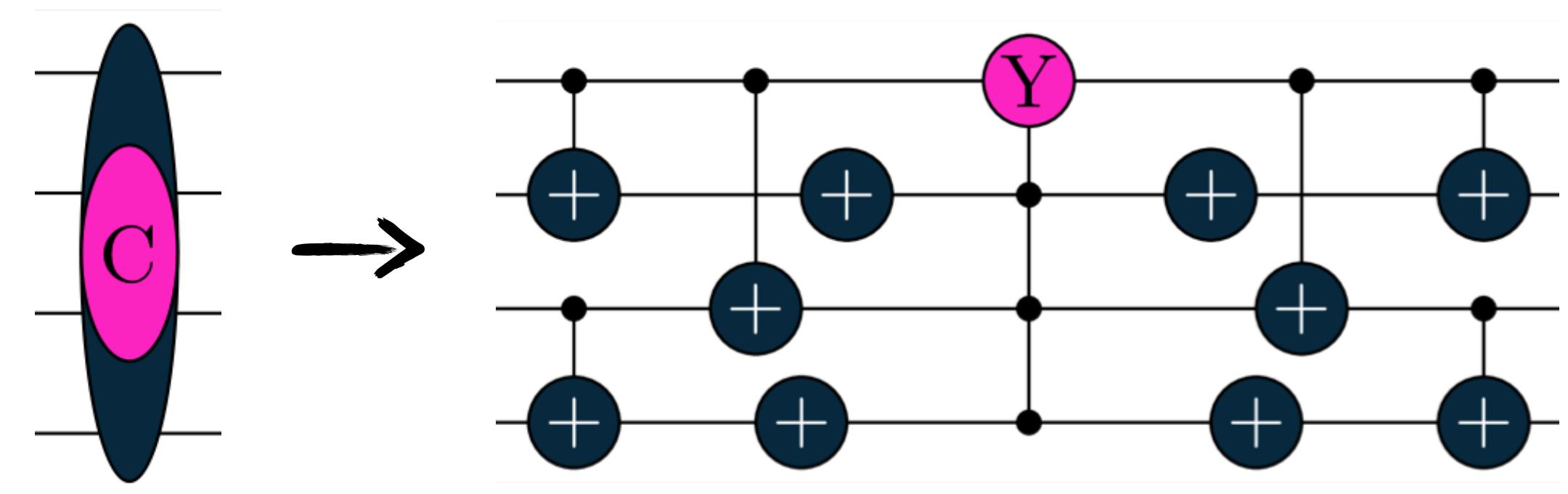
$$U_{\mathbf{pq}}(\theta) = e^{-i\frac{\theta}{2}G_{\mathbf{pq}}}$$

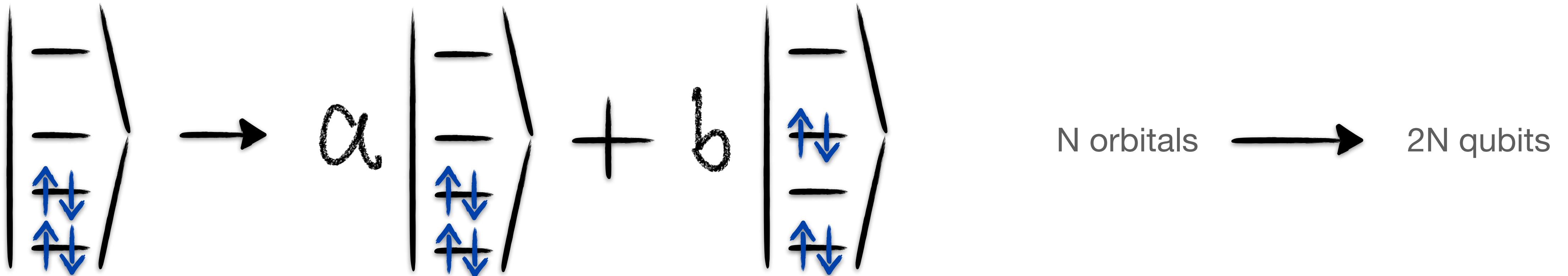
Example

$$G_{p\uparrow p\downarrow q\uparrow q\downarrow} \equiv \tilde{G}_{pq} = i \left( a_{p\uparrow}^\dagger a_{q\uparrow} a_{p\downarrow}^\dagger a_{q\downarrow} - h.c \right)$$

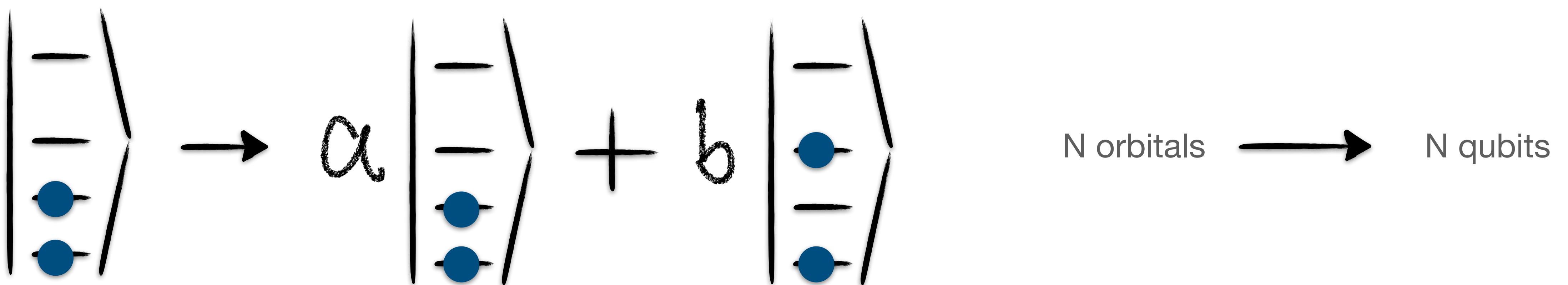
$$G_{\mathbf{pq}} = i \left( \prod_k a_{p_k}^\dagger a_{q_k} - h.c. \right)$$

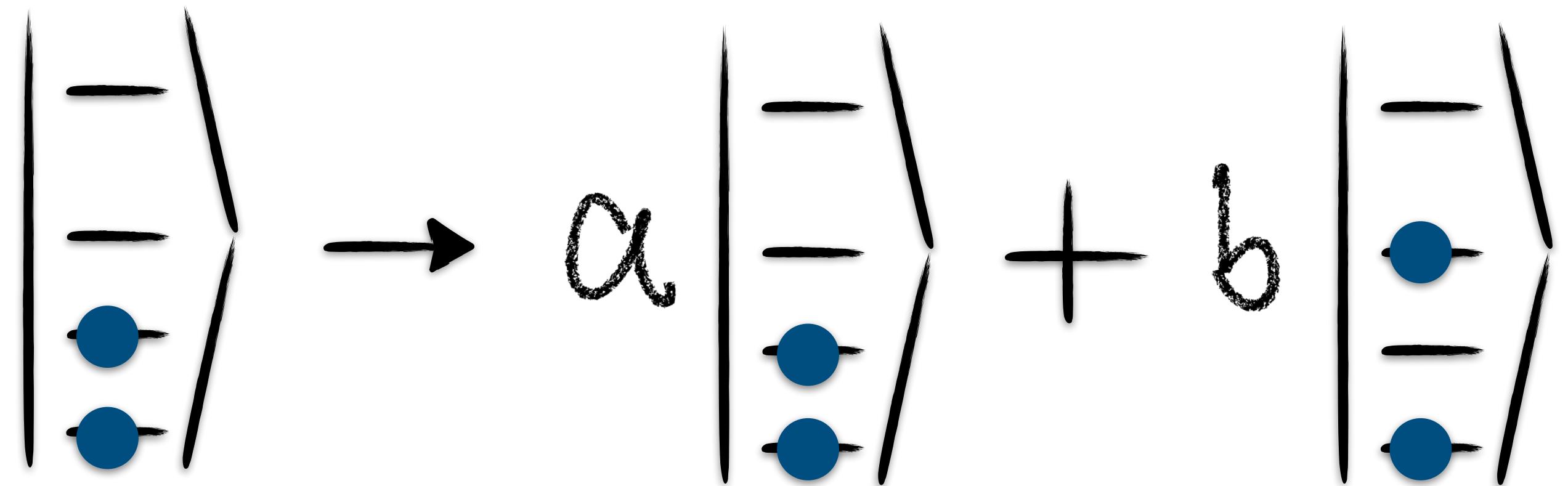
$$a_p^\dagger \xrightarrow[\text{Wigner}]{\text{Jordan}} \sigma_p^+ \prod_{k < p} \sigma_k^z.$$





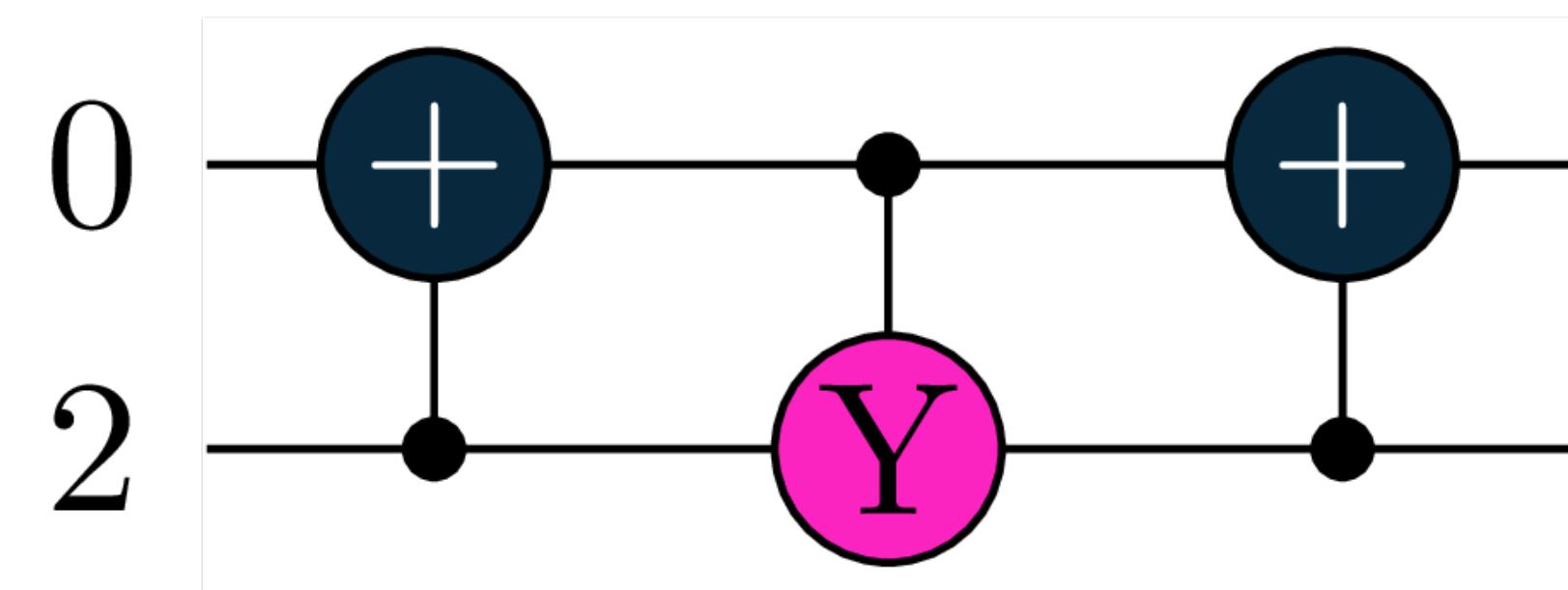
“Hard-Core Boson”, “seniority zero”, “Doubly Occupied ...”



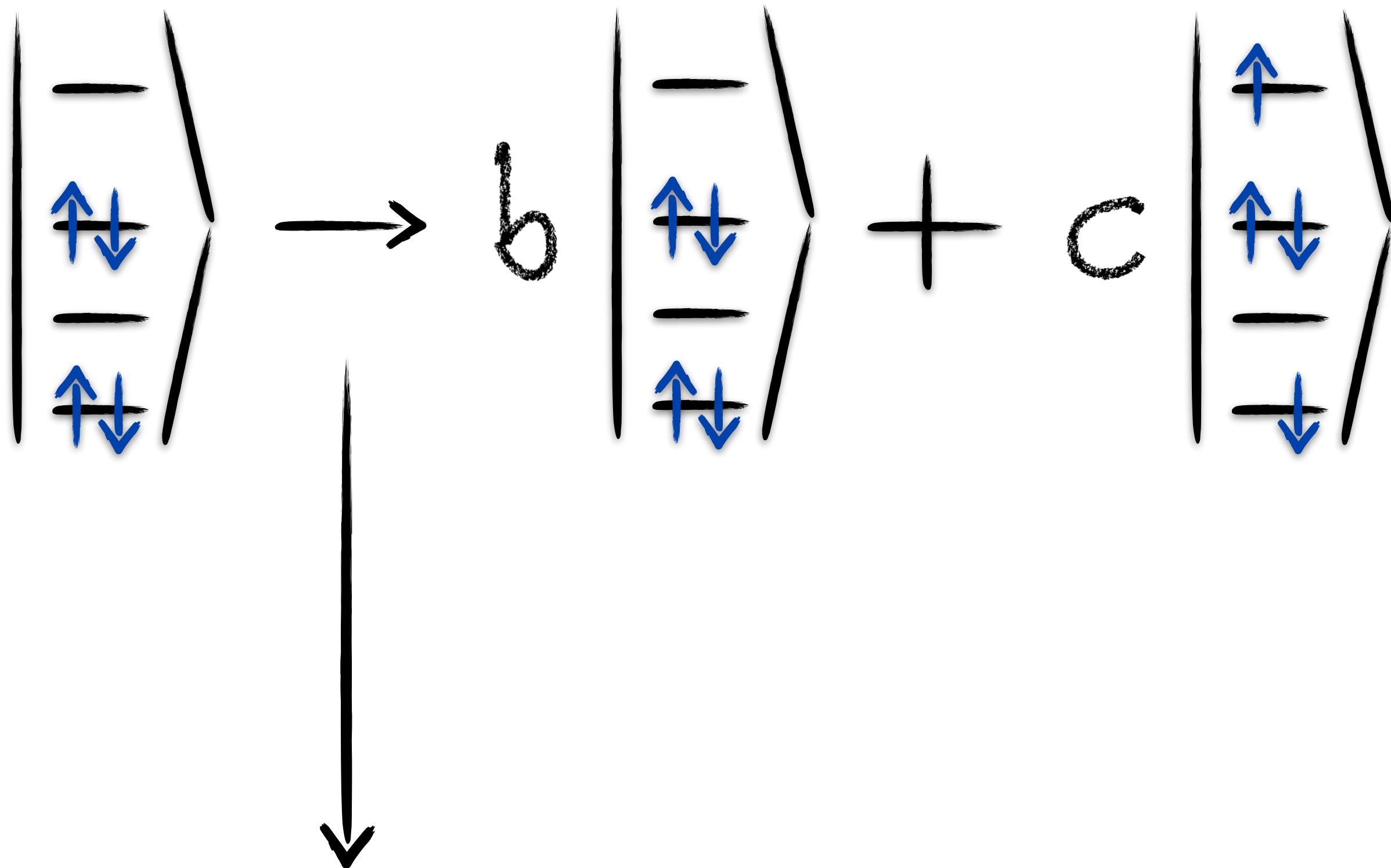


$$\tilde{G}_{pq} \xrightarrow[\text{Wigner}]{\text{Jordan-}} \tilde{G}_{pq}^{\text{JW}} = i \left( \sigma_{p\uparrow}^+ \sigma_{q\uparrow}^- \sigma_{p\downarrow}^+ \sigma_{q\downarrow}^- - h.c. \right)$$

$$\tilde{G}_{pq} \xrightarrow[\text{Boson}]{\text{hard-core}} \tilde{G}_{pq}^{\text{HCB}} = i \left( \sigma_p^+ \sigma_q^- - h.c. \right)$$

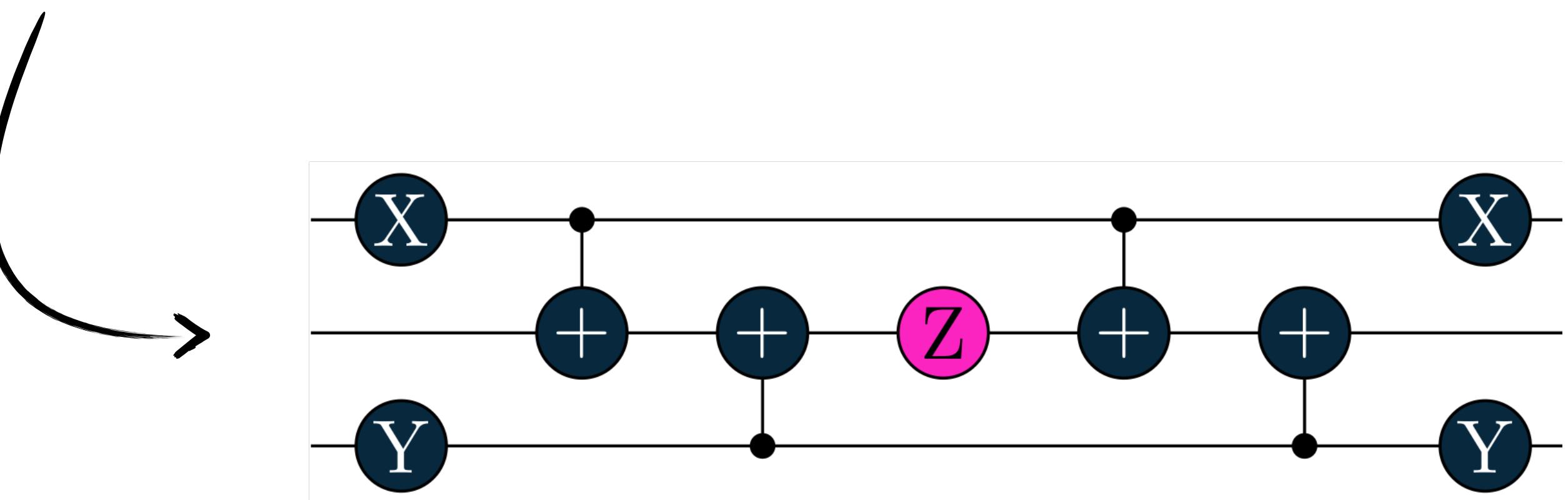
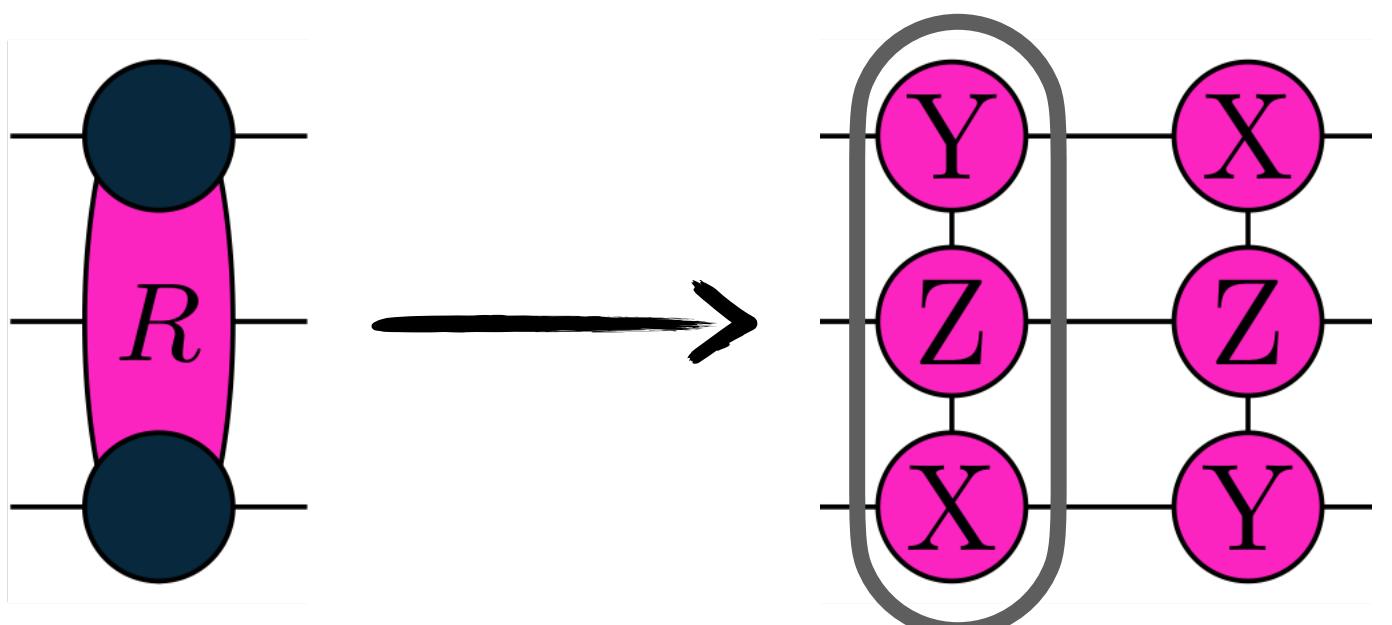
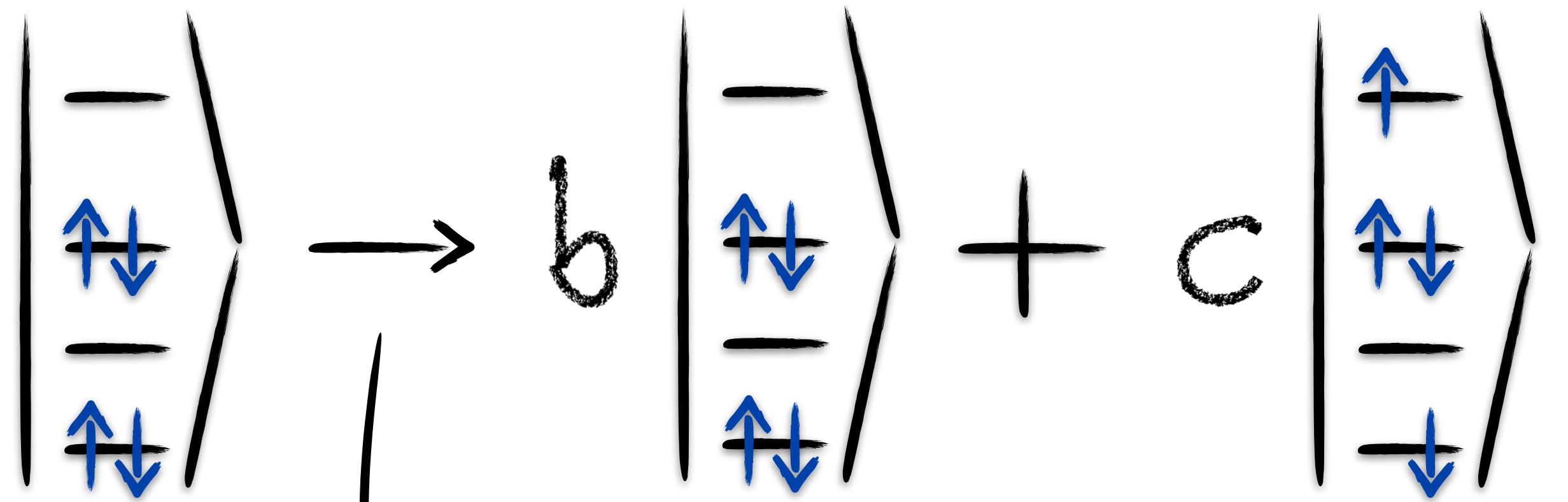


$$\left| \begin{array}{c} - \\ \uparrow \downarrow \\ - \\ \uparrow \downarrow \end{array} \right\rangle \rightarrow b \left| \begin{array}{c} - \\ \uparrow \downarrow \\ - \\ \uparrow \downarrow \end{array} \right\rangle + c \left| \begin{array}{c} + \\ \uparrow \downarrow \\ - \\ \downarrow \end{array} \right\rangle$$



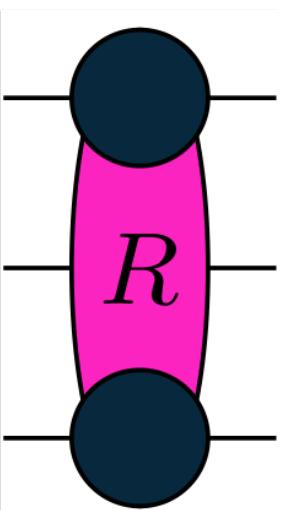
$$U_{i\uparrow}^{j\uparrow} = e^{-i\frac{\theta}{2}(a_{i\uparrow}^\dagger a_{j\uparrow} - a_{j\uparrow}^\dagger a_{i\uparrow})}$$

$$e^{-\frac{\theta}{2}\sigma_0^x\sigma_1^z\sigma_2^y} e^{-\frac{\theta}{2}\sigma_0^y\sigma_1^z\sigma_2^x}$$

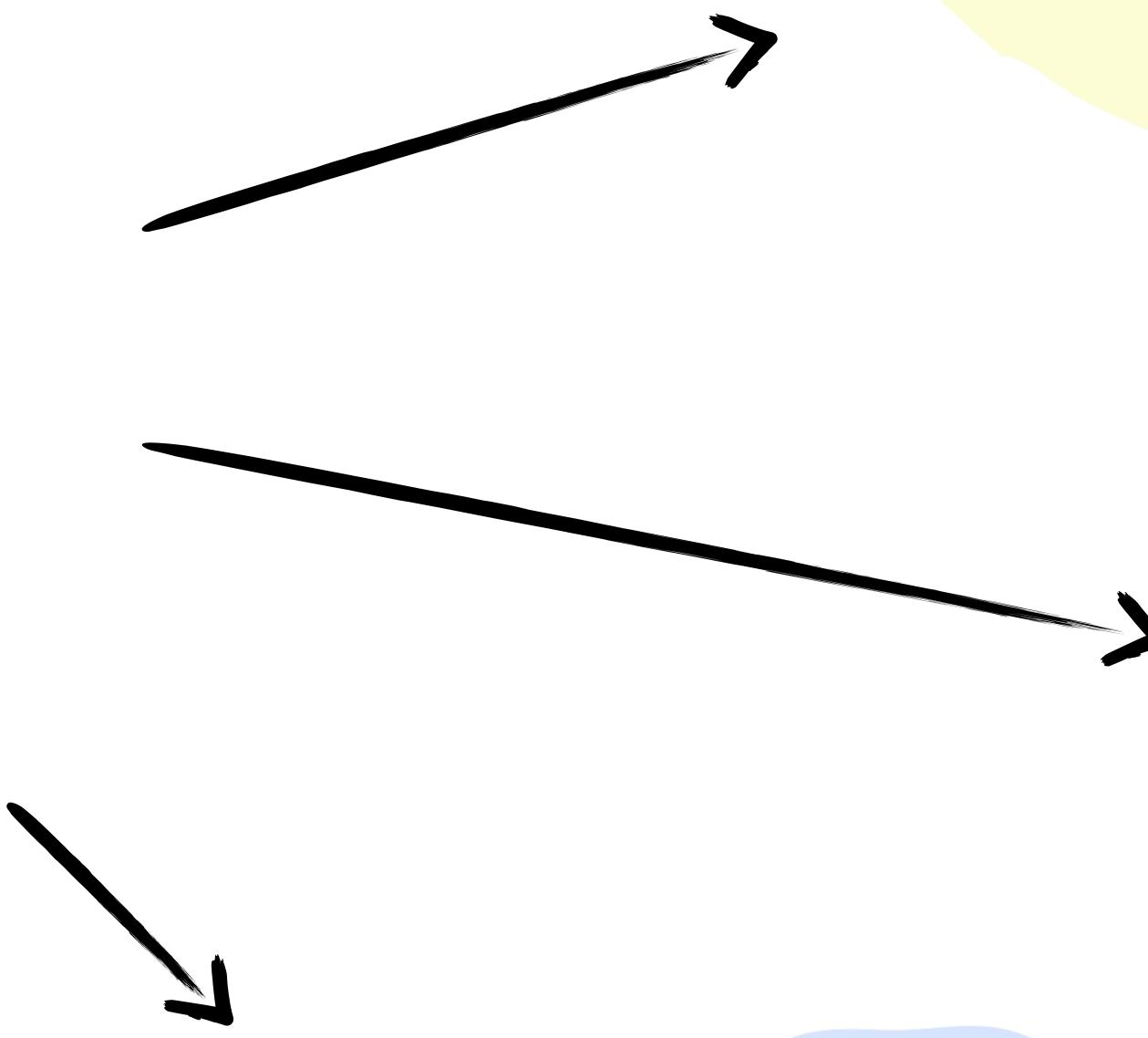
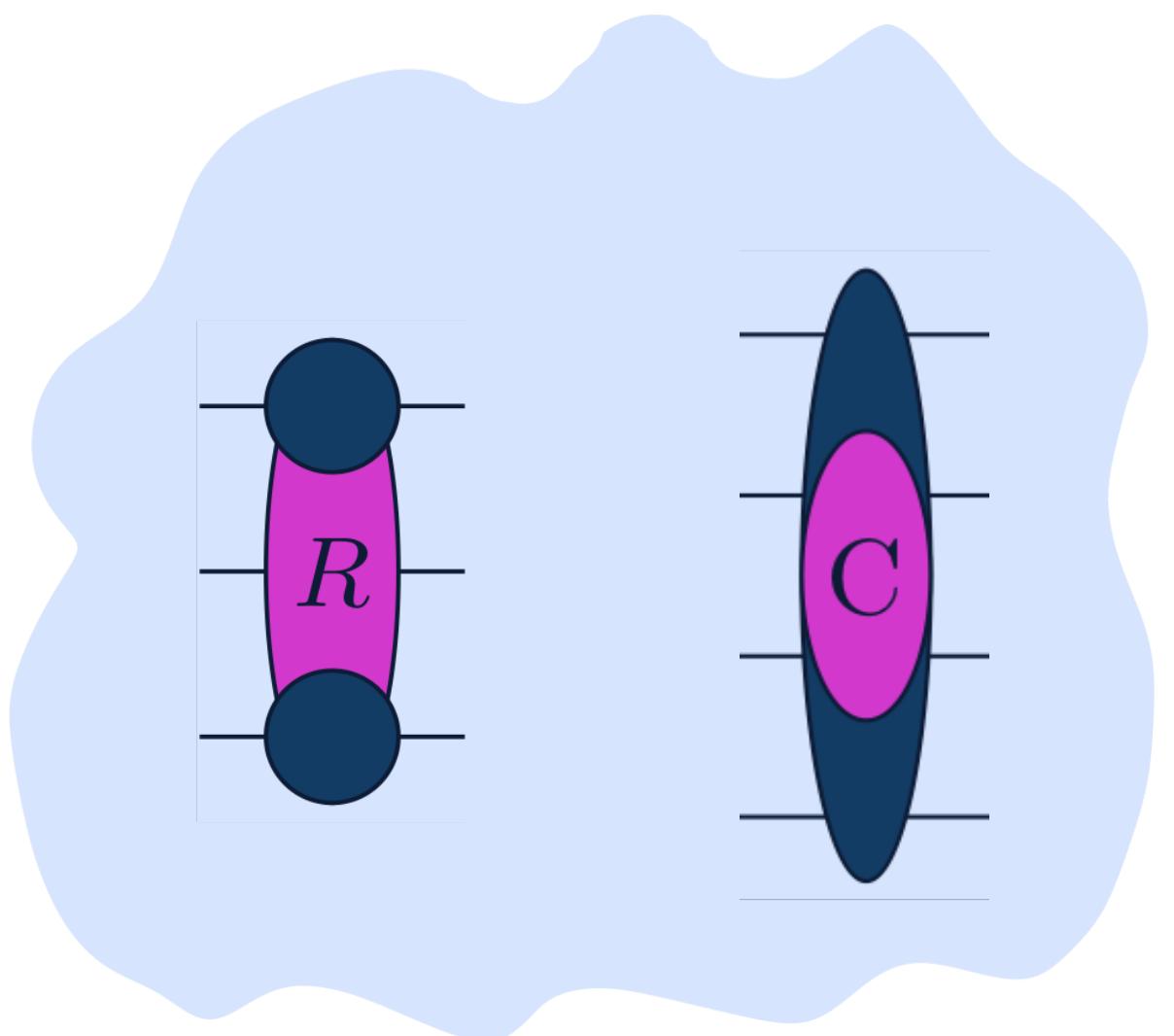


orbital rotations

$$\left| \begin{array}{c} \text{-} \\ \text{-} \\ \text{+} \end{array} \right\rangle_{\psi}^{\phi} \xrightarrow{\quad} a \left| \begin{array}{c} \text{-} \\ \text{-} \\ \text{+} \end{array} \right\rangle + b \left| \begin{array}{c} \text{-} \\ \text{-} \\ \text{-} \end{array} \right\rangle \xrightarrow{\quad} \left| \begin{array}{c} \text{-} \\ \text{-} \\ \text{+} \end{array} \right\rangle^{\tilde{\phi}} = a\psi - b\phi$$
$$\tilde{\psi} = a\psi + b\phi$$



Rotators  
and  
Correlators



Standard methods  
UCCSD  
k-UpCCGSD  
...

Adaptive Methods  
ADAPT-VQE  
Qubit-Coupled-Cluster  
...

High-Level-Design  
Now....

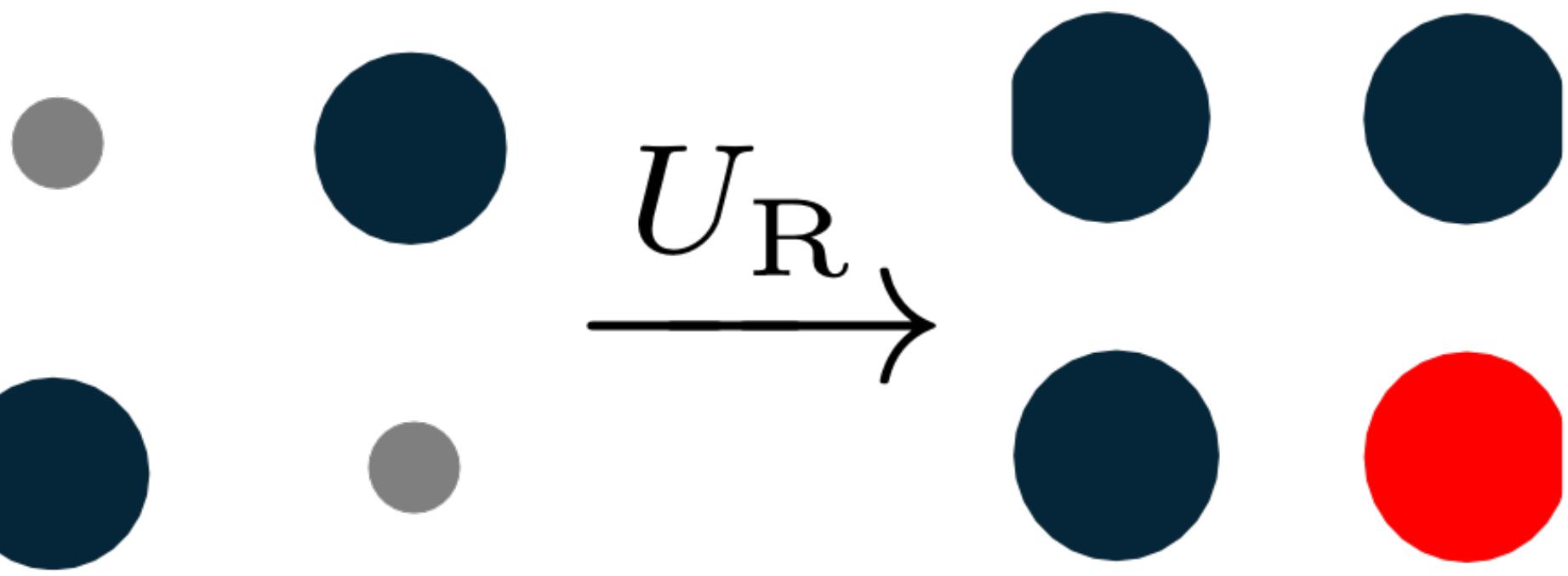
# High-Level Design

# Example: Hydrogen Molecule



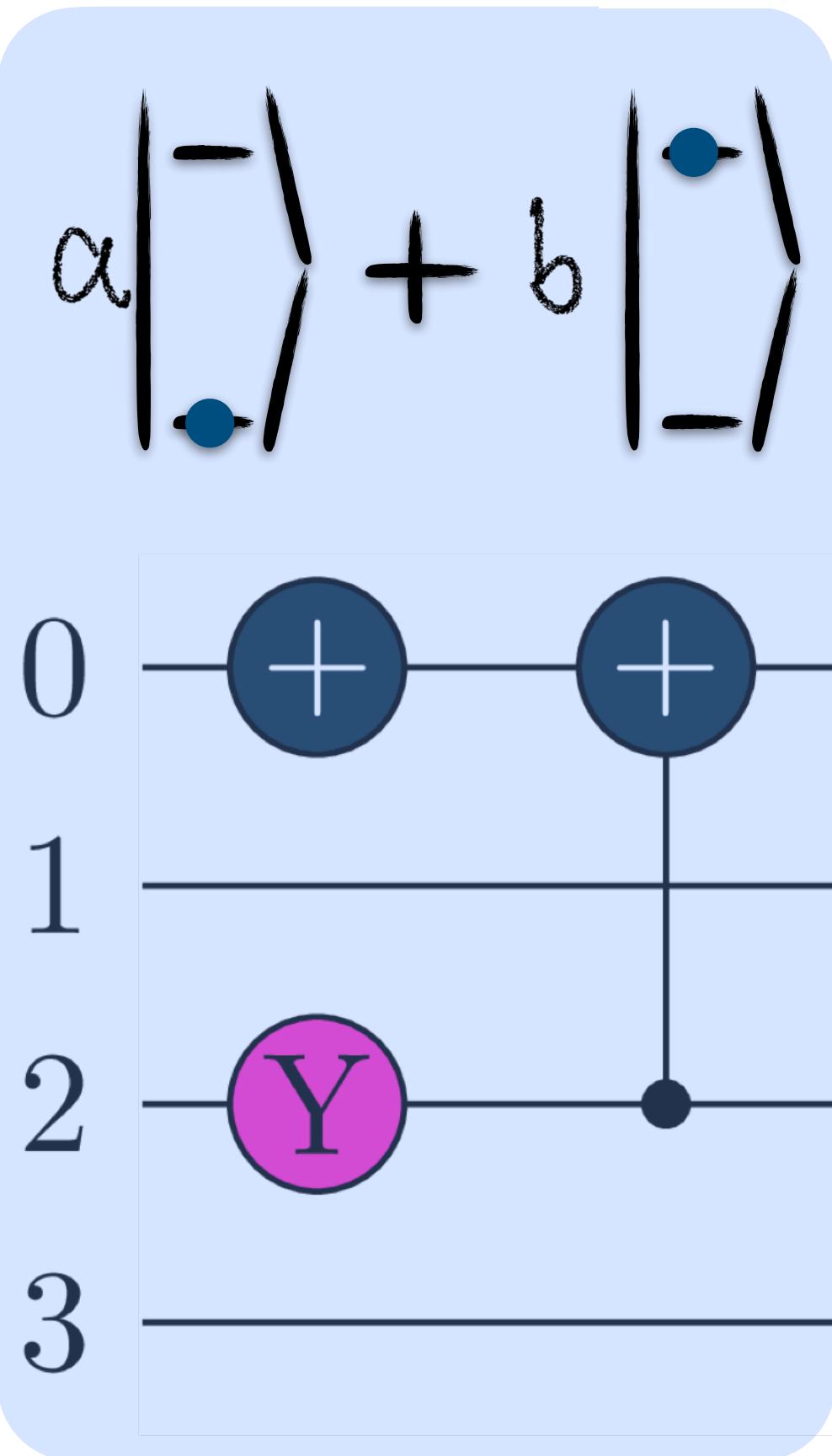
**Simple system: 2-Electrons**

Atomic orbital basis  
e.g. STO-3G basis set

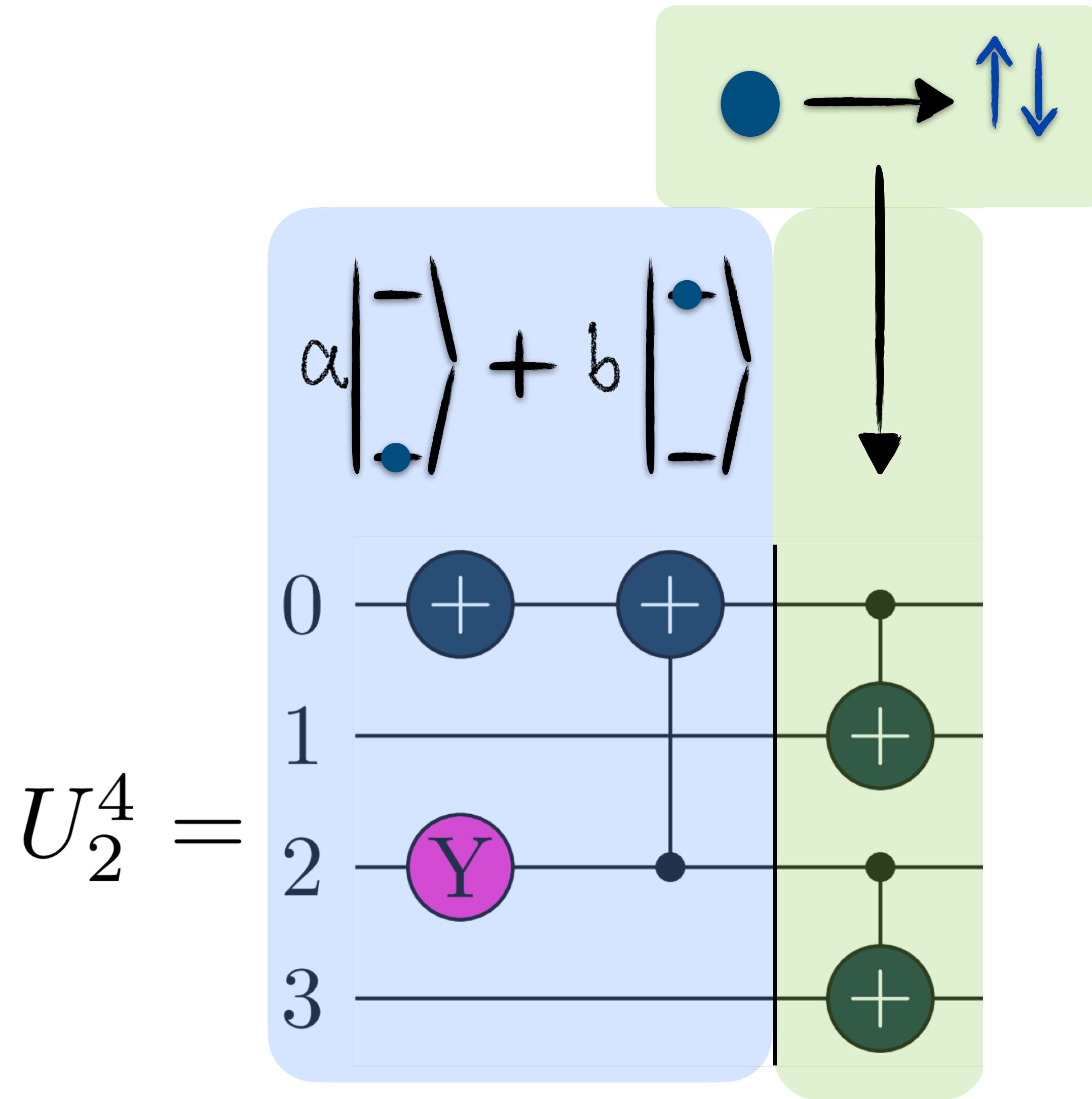


## Simple system: 2-electrons in 2 orbitals (4 qubits)

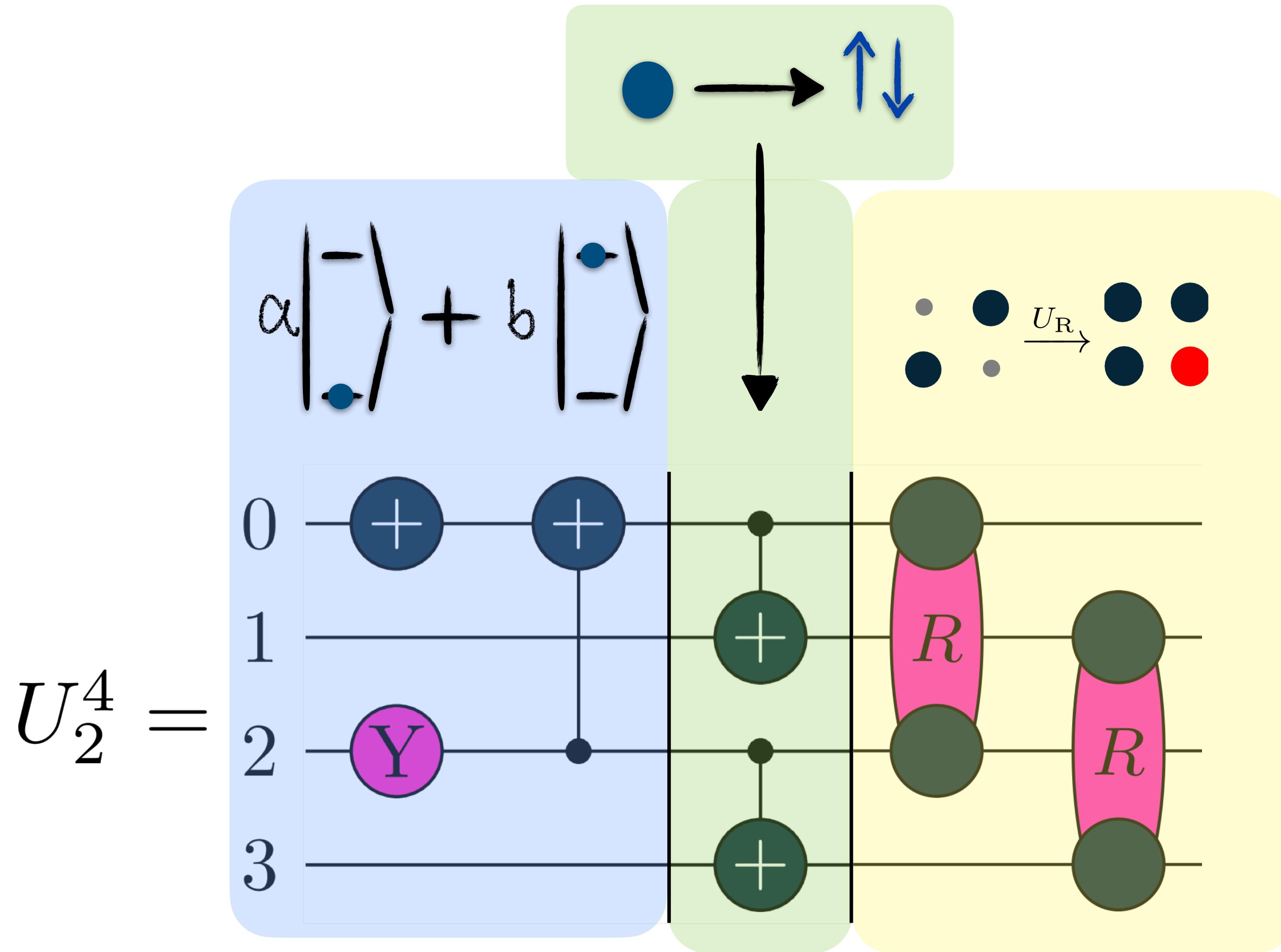
$$U_2^4 =$$



## Simple system: 2-electrons in 2 orbitals (4 qubits)

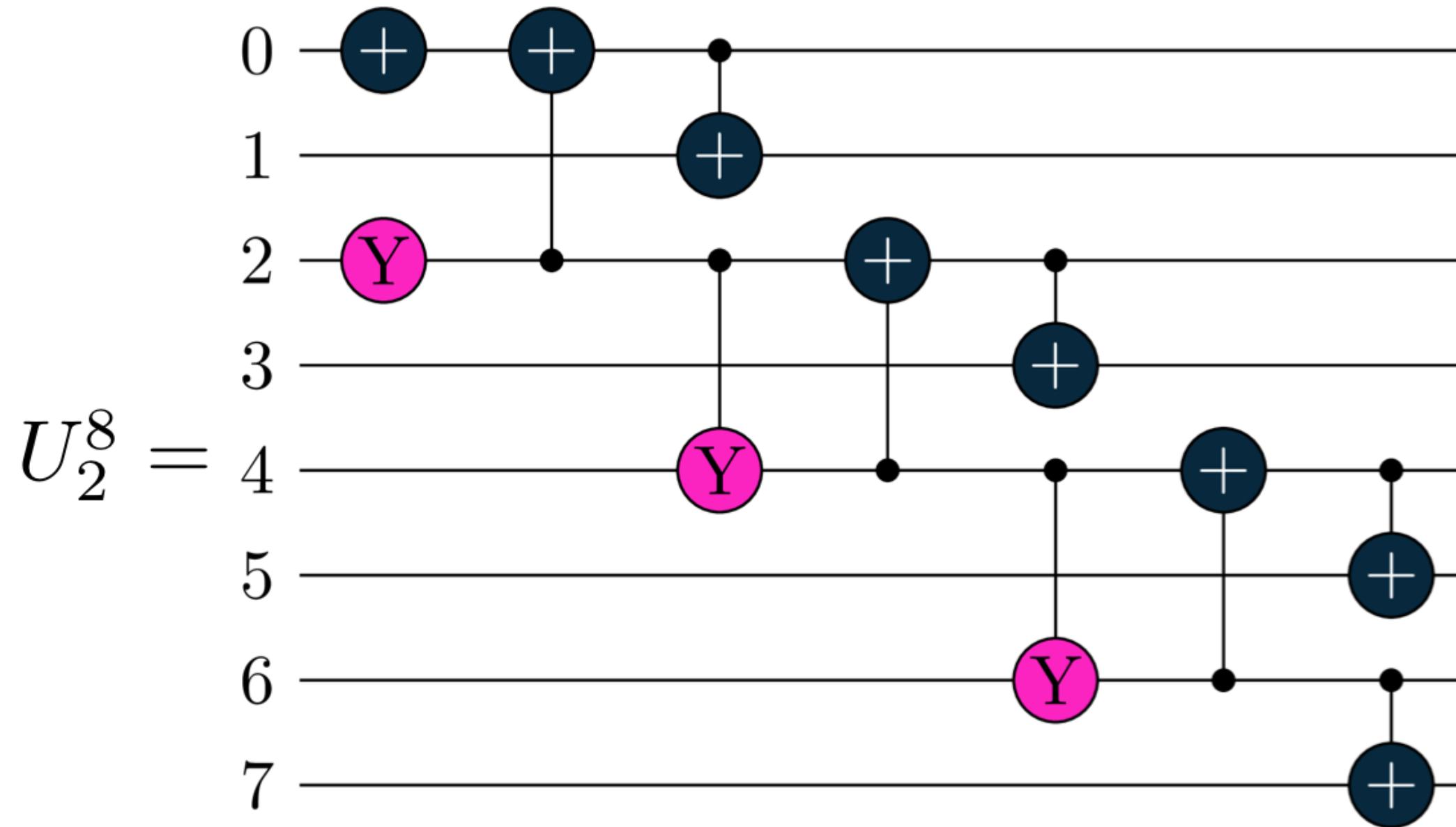


## Simple system: 2-electrons in 2 orbitals (4 qubits)



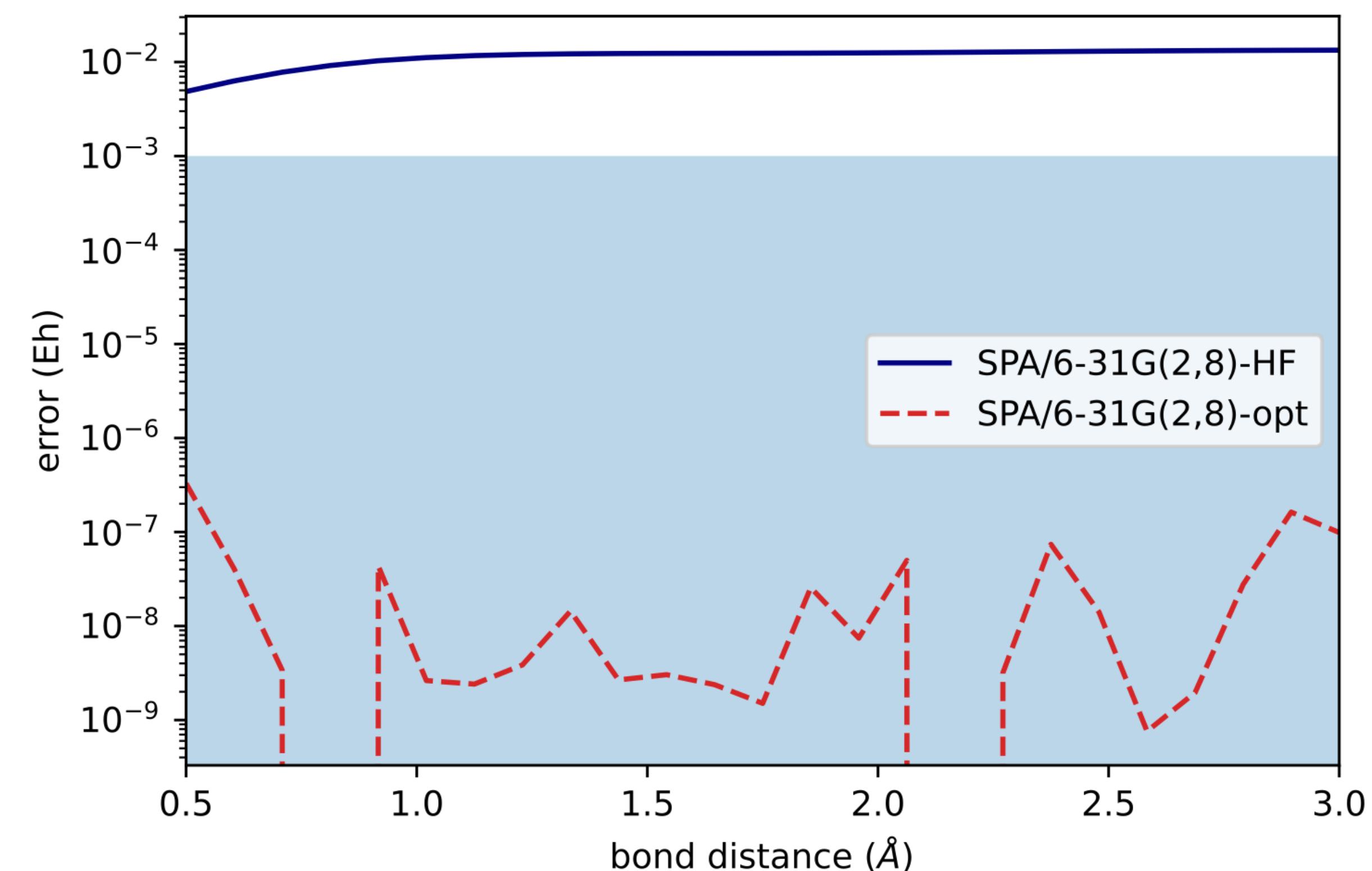
Orbital rotation  
can be absorbed  
into the  
Hamiltonian

# Simple system: 2-electrons in 4 orbitals (8 qubits)



Larger basis (8 qubits)

Same behaviour  
for other (effective) two electron systems  
He, LiH, H<sub>3</sub>+ ...



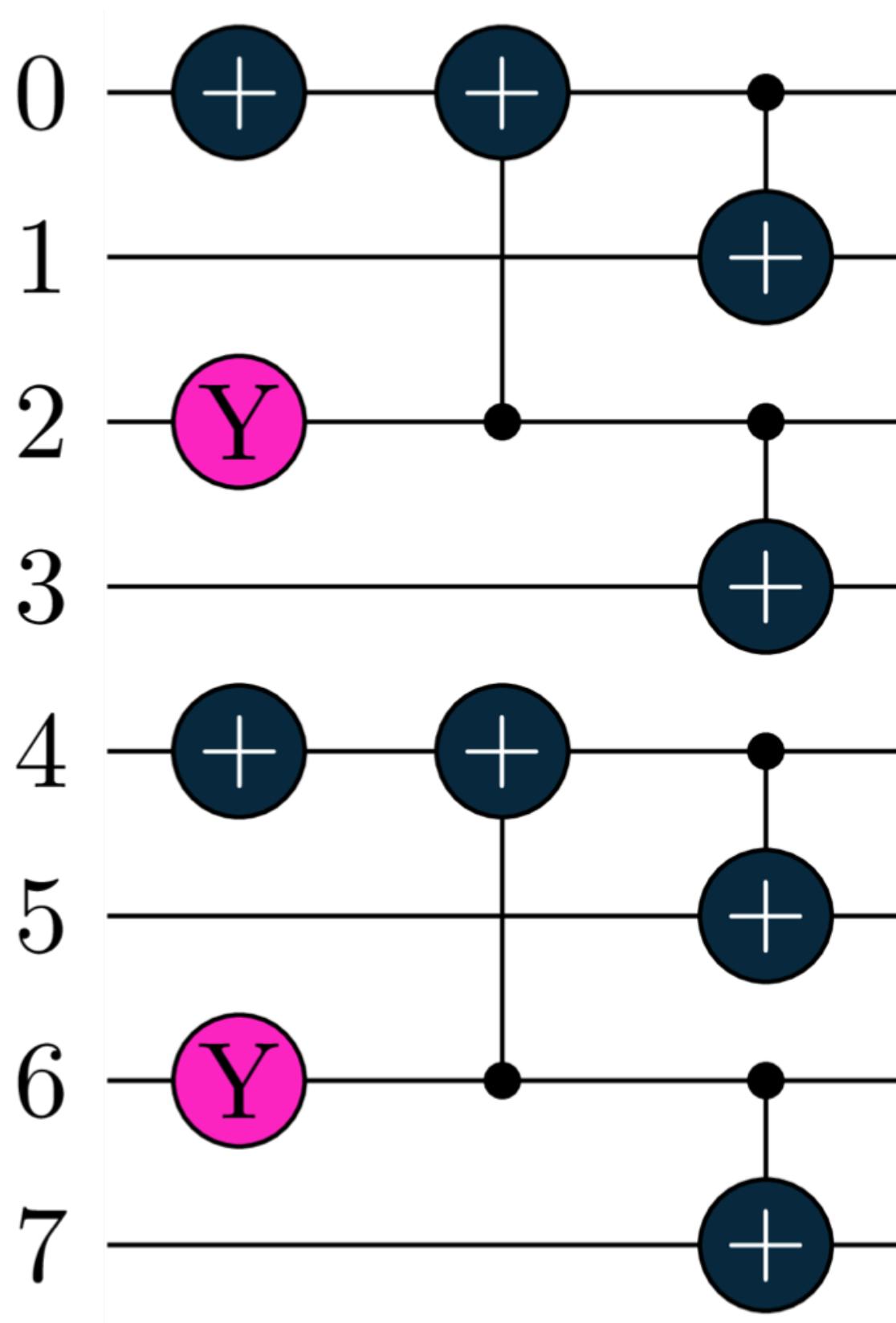
# H<sub>4</sub> Molecule



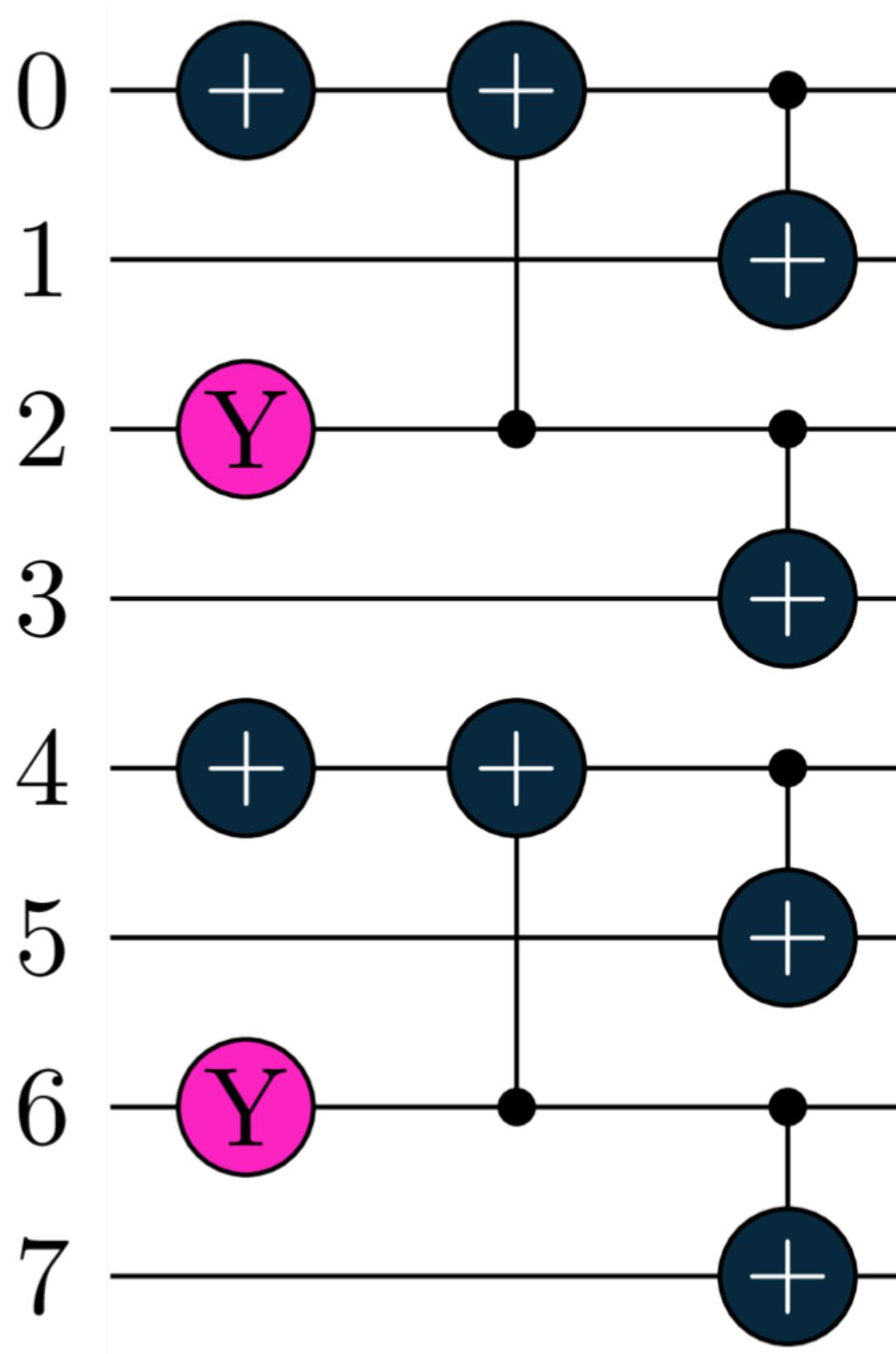
As before: Minimal basis STO-3G. One orbital per Hydrogen

SPA: Separable Pair Ansatz

$$U_{\text{SPA}}^{(4,8)} = U_2^4 \otimes U_2^4 =$$



$$U_{\text{SPA}}^{(4,8)} = U_2^4 \otimes U_2^4 =$$



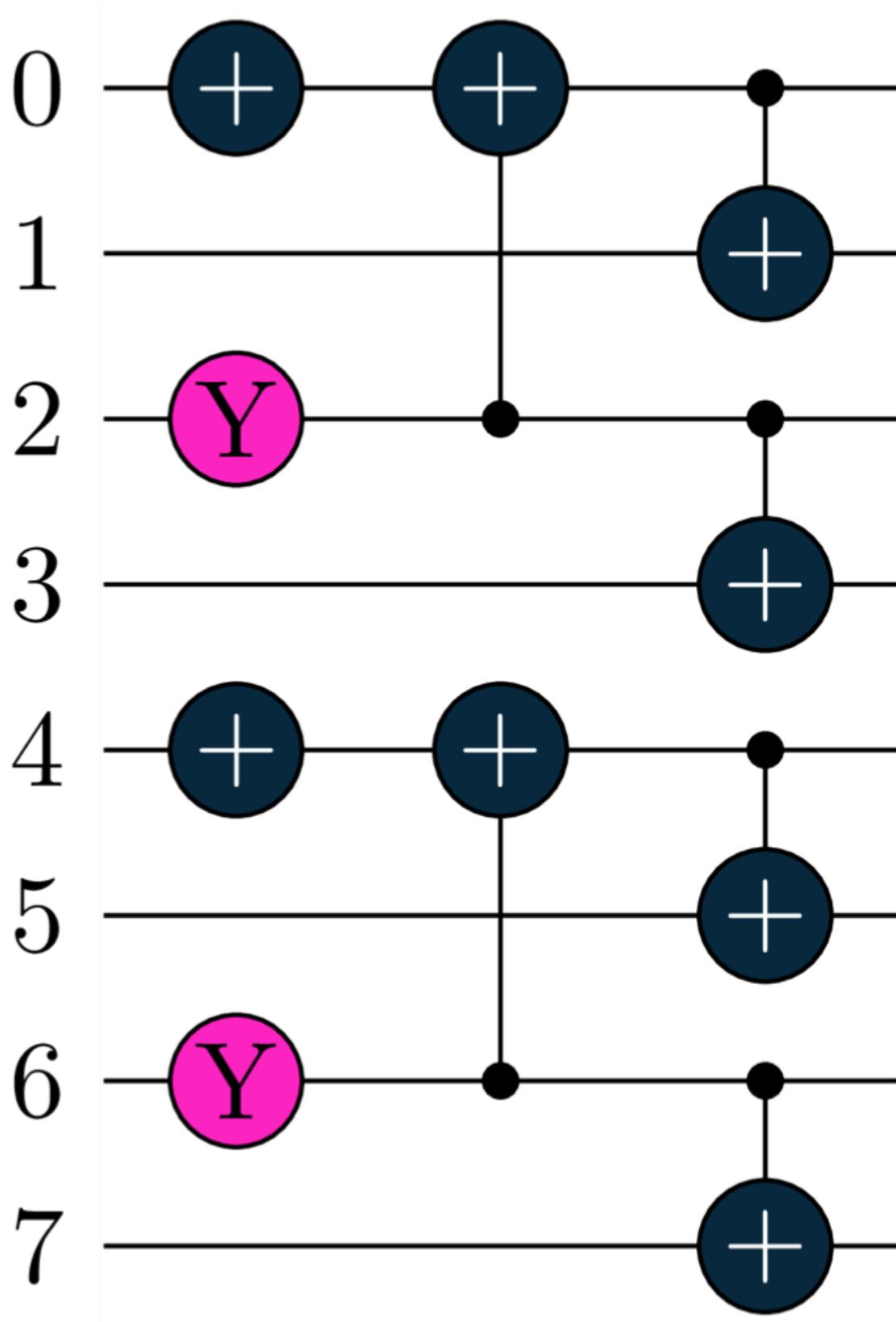
- Locality (circuit connections)
- Shallow depth
- Parallelizable
- Hardware efficient
- Classically tractable wavefunction



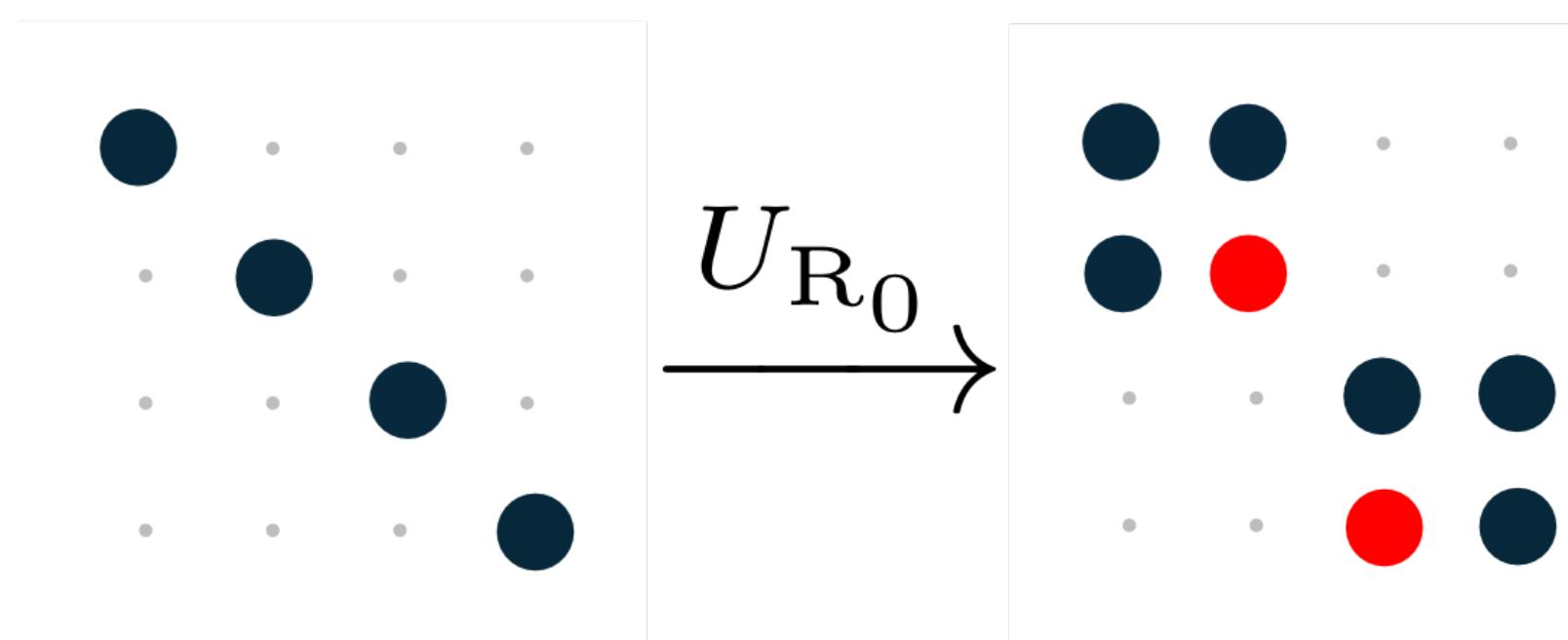
becomes a classical pre-compilation step

Hartree–Fock error: 167 mEH

$$U_{\text{SPA}}^{(4,8)} = U_2^4 \otimes U_2^4 =$$



Guess orbitals:  
error of 40 mEh

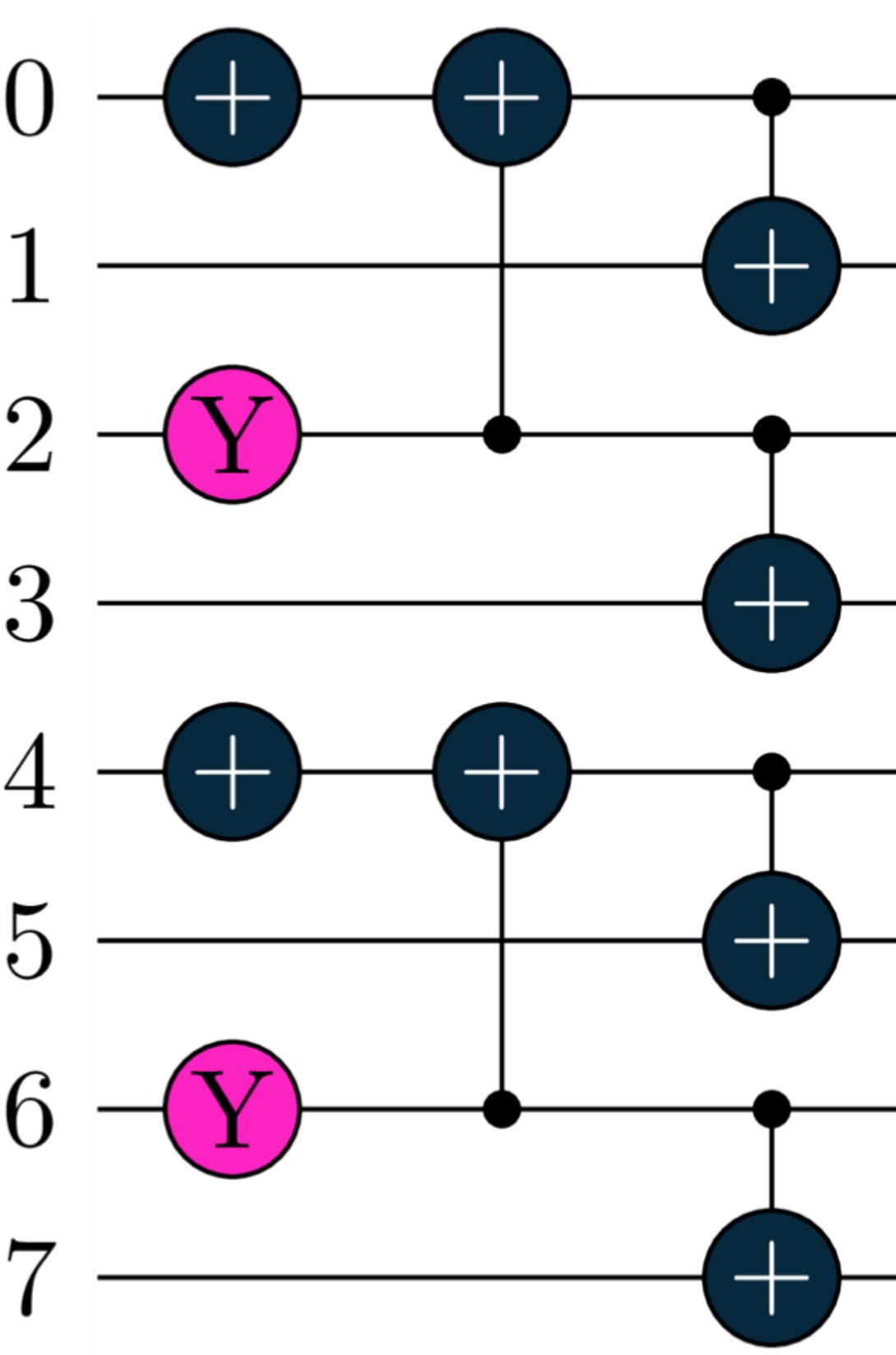


SPA: Separable Pair Ansatz

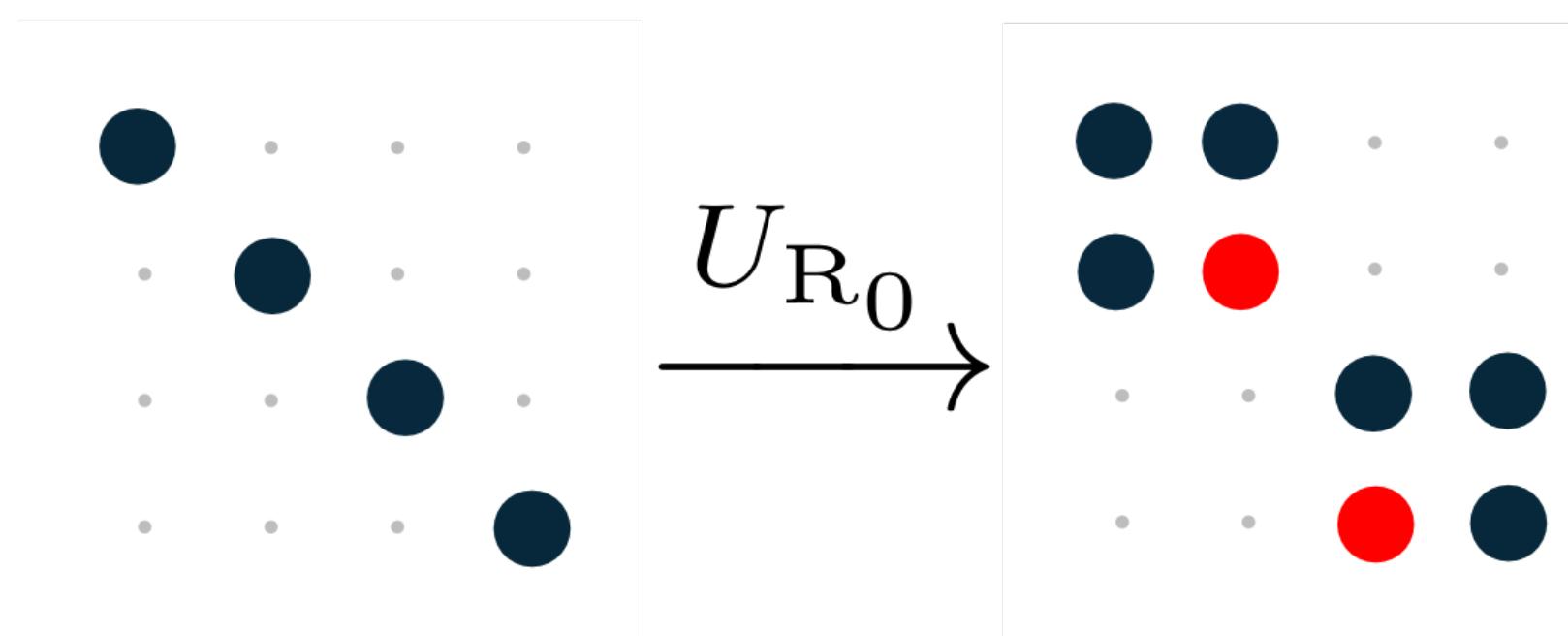


Hartree–Fock error: 167 mEH

$$U_{\text{SPA}}^{(4,8)} = U_2^4 \otimes U_2^4 =$$



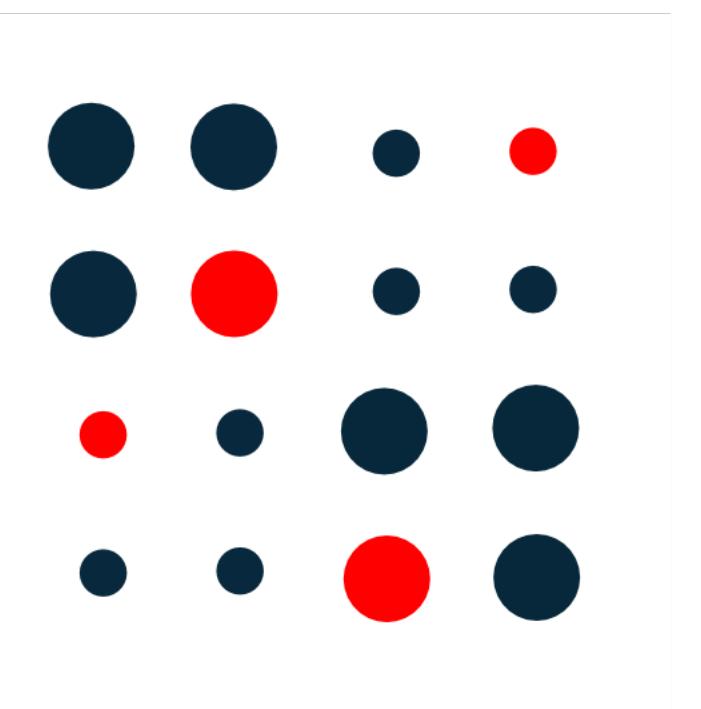
Guess orbitals:  
error of 40 mEh



SPA: Separable Pair Ansatz



Optimal orbitals:  
error of 16 mEh

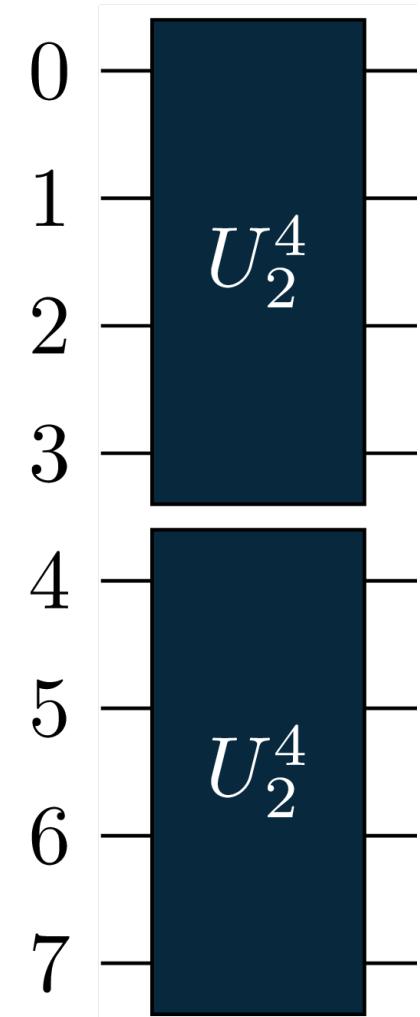


## Alternative Graph

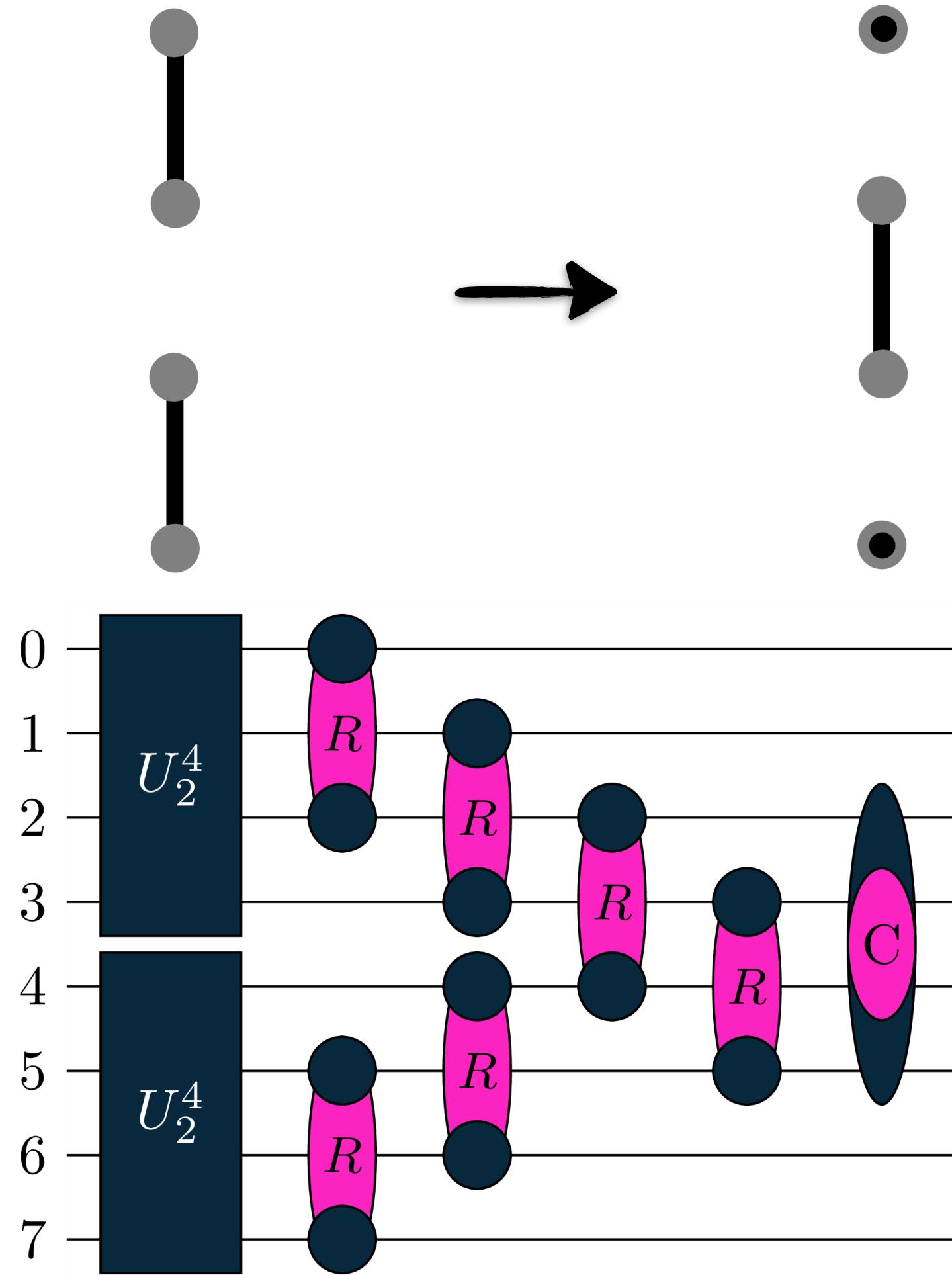


Hartree–Fock error: 167 mEH

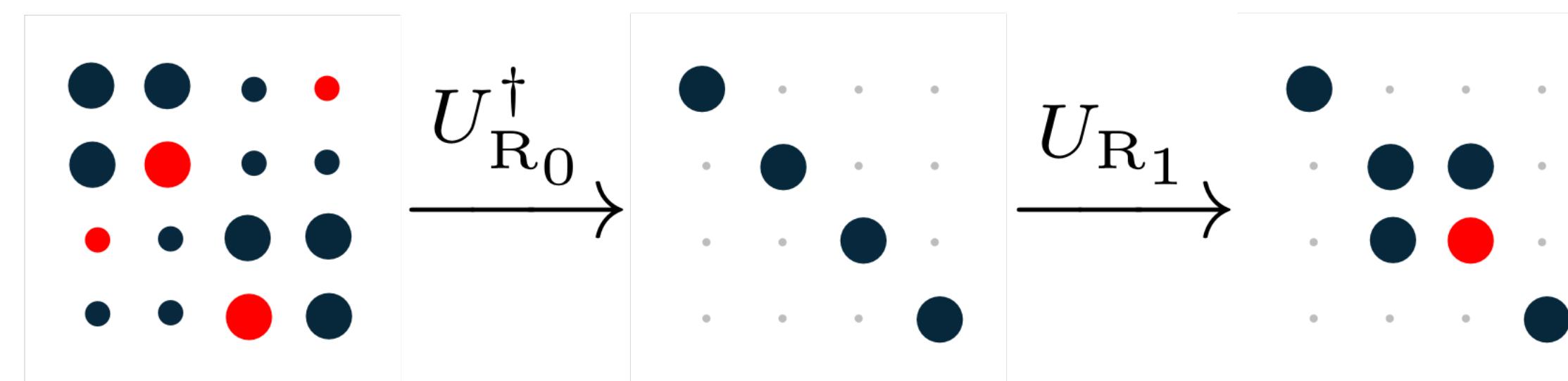
method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*



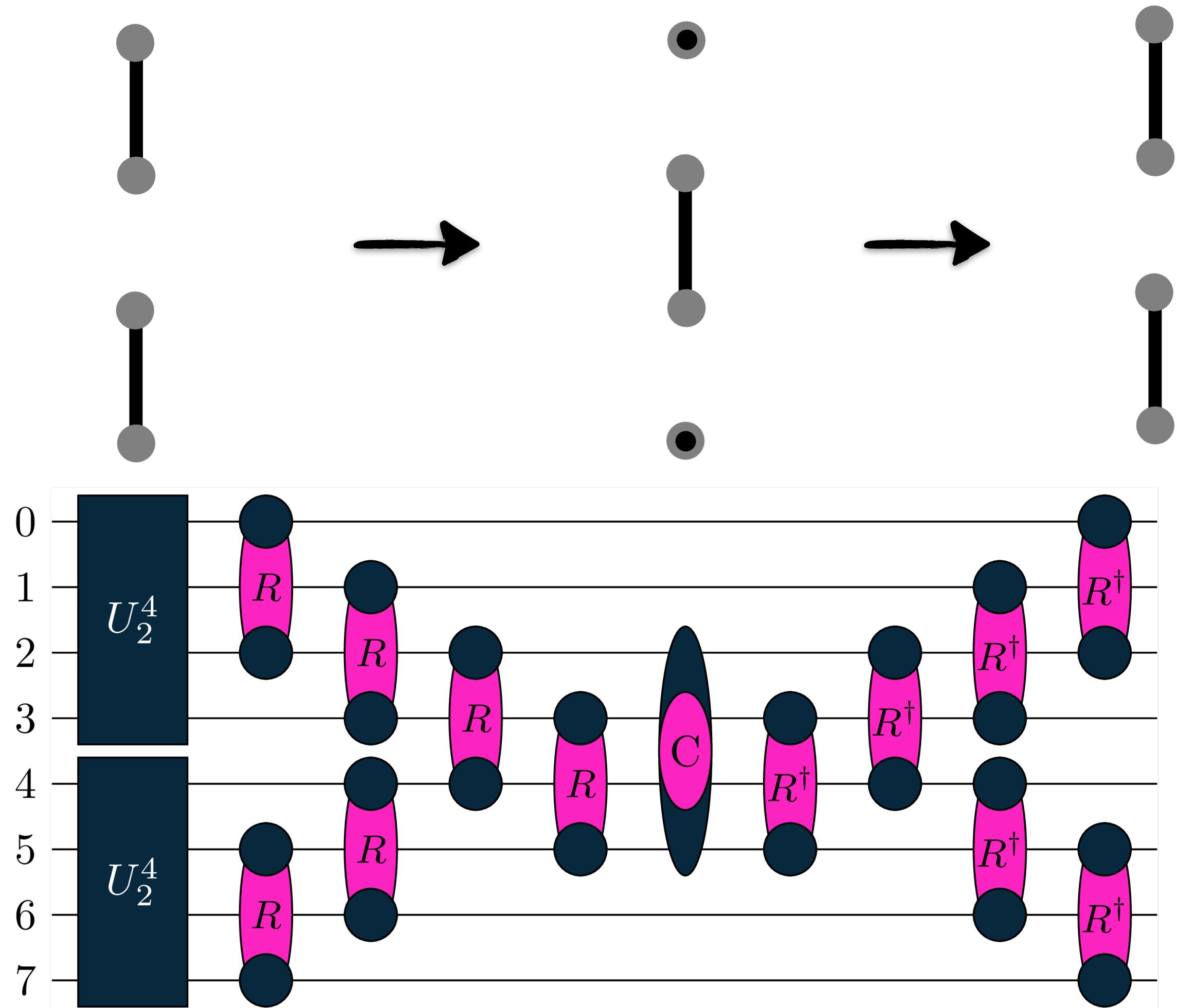
Hartree–Fock error: 167 mEH



method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*

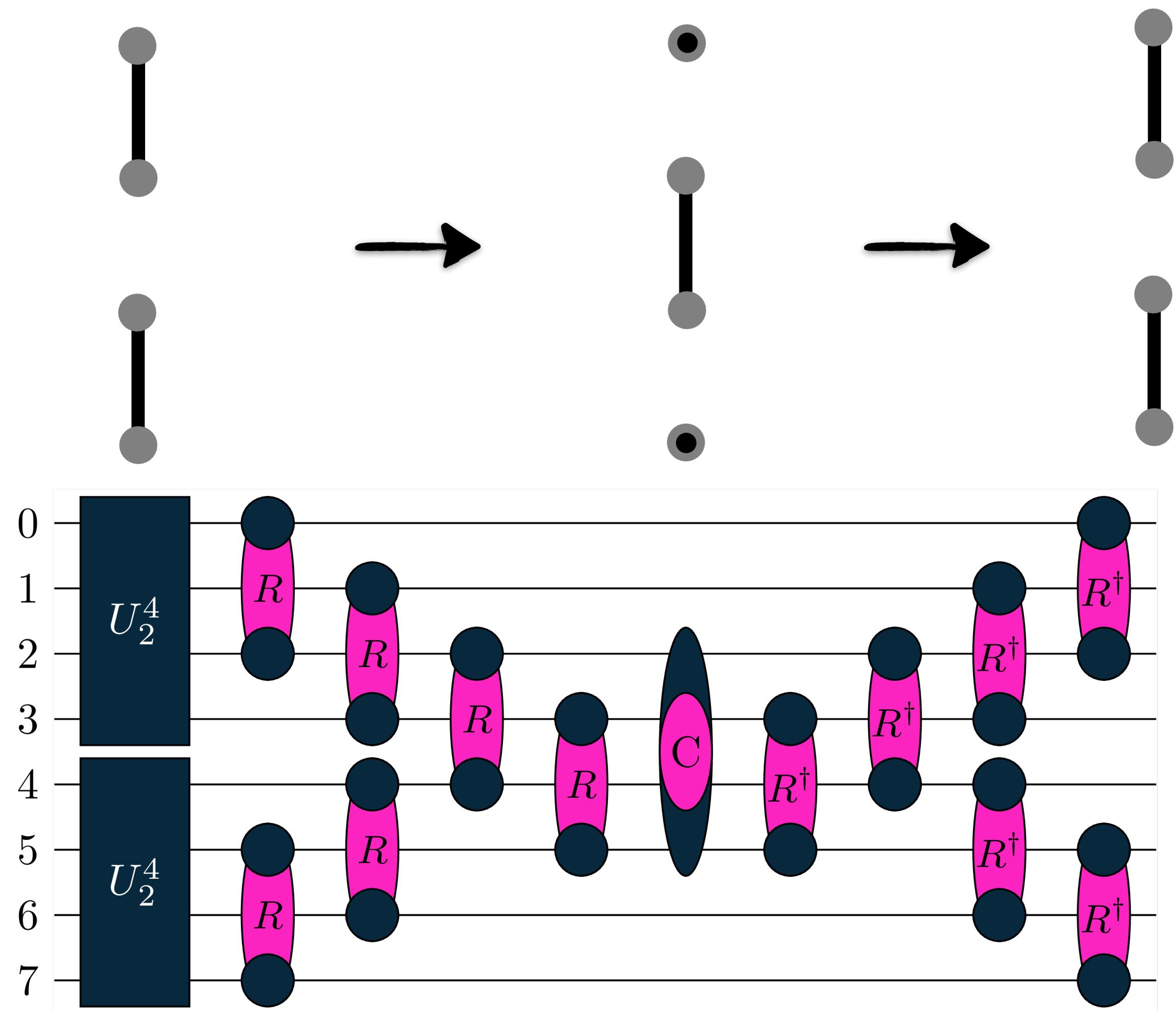


Hartree–Fock error: 167 mEH

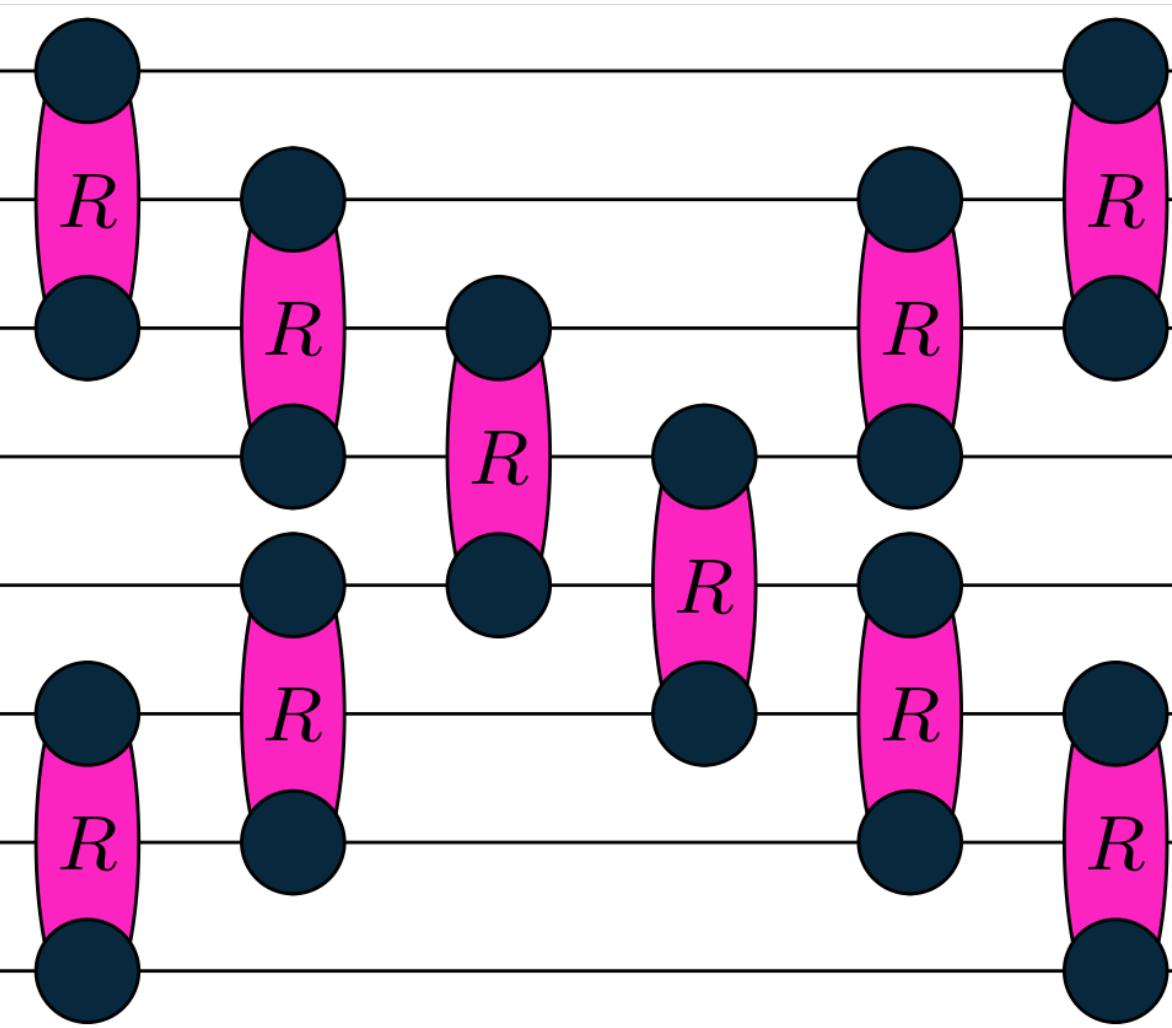
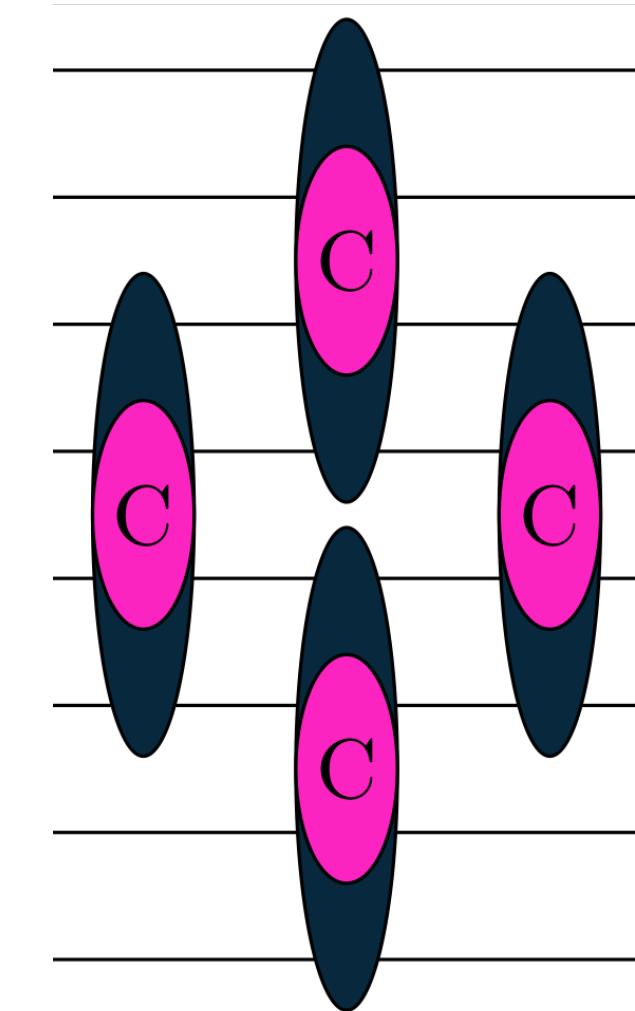


method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*
SPA+	8	96	6	116	131	10

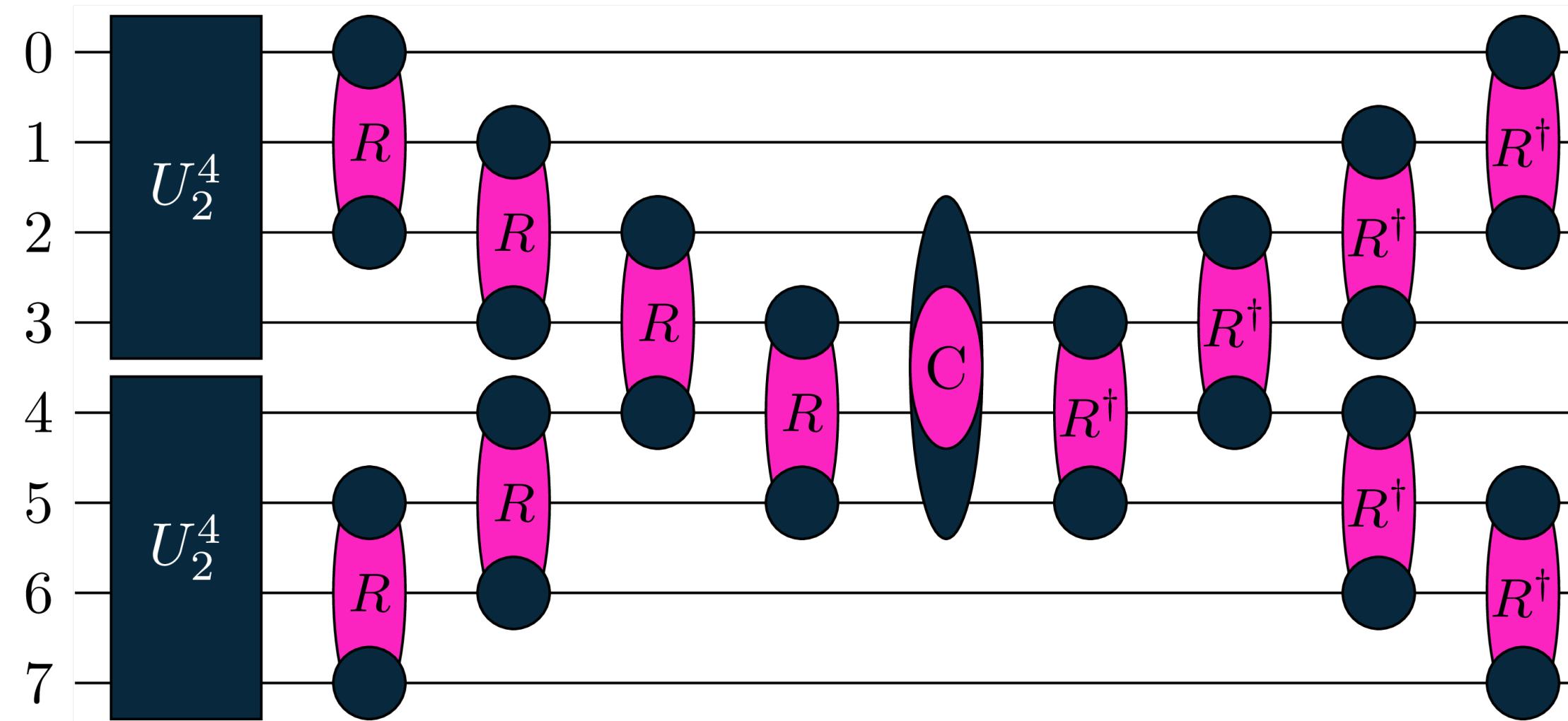
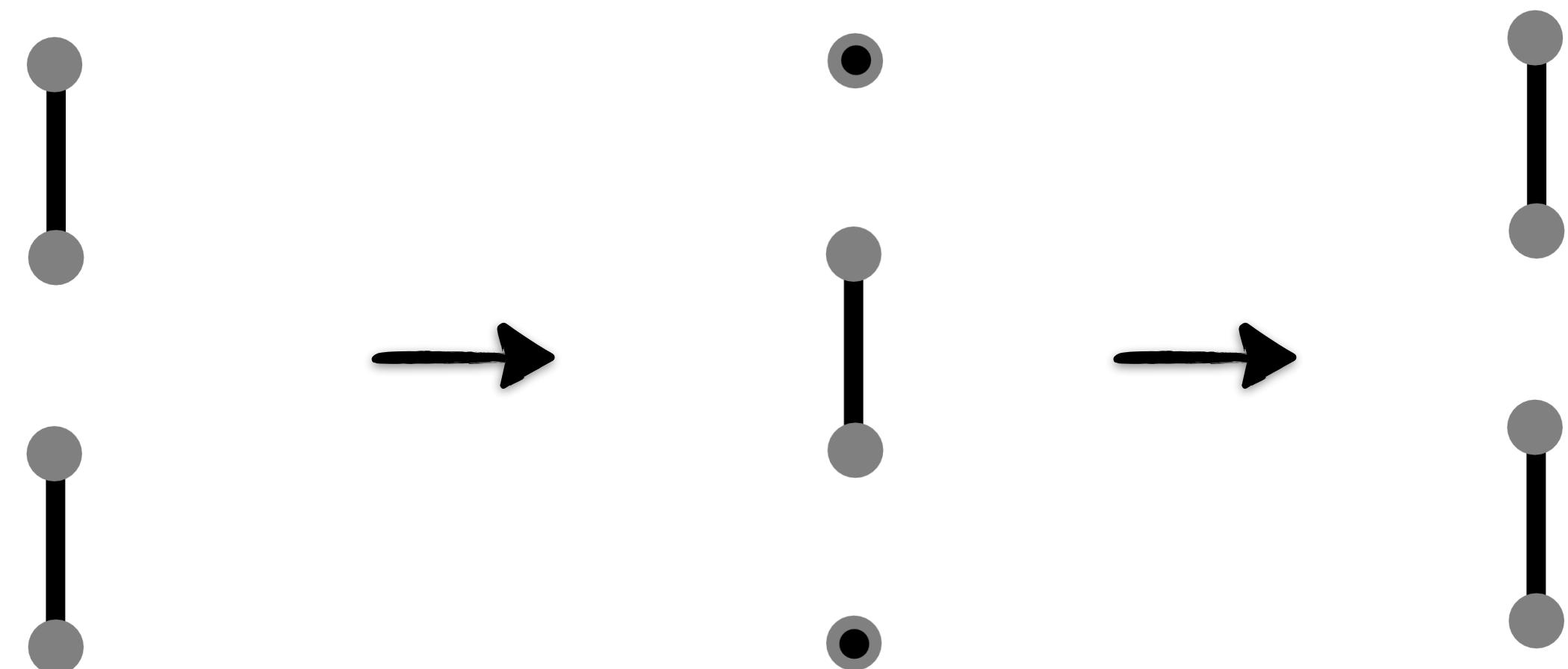
Hartree–Fock error: 167 mEH



method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*
SPA+	8	96	6	116	131	10
SPA+CRRCRRC	0	100	19	334	367	160

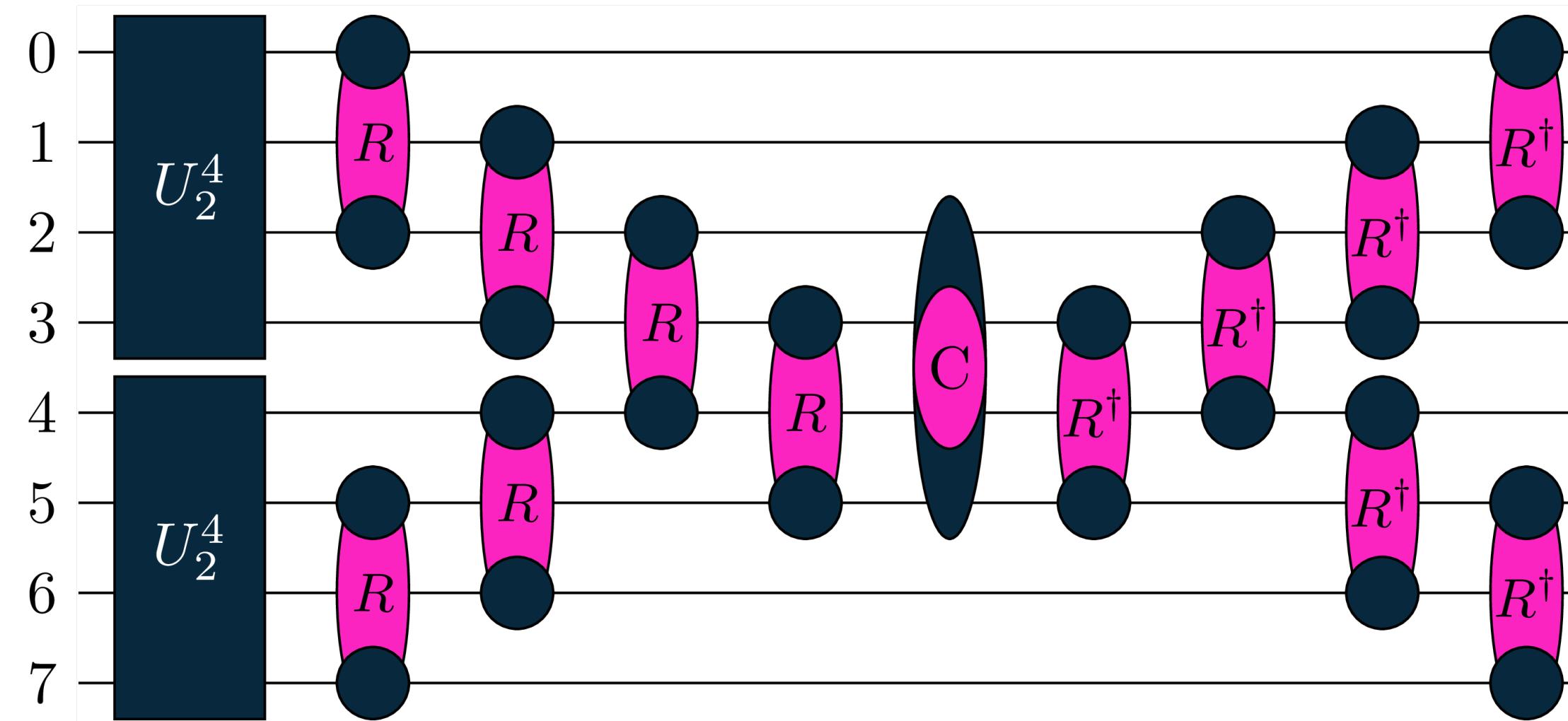
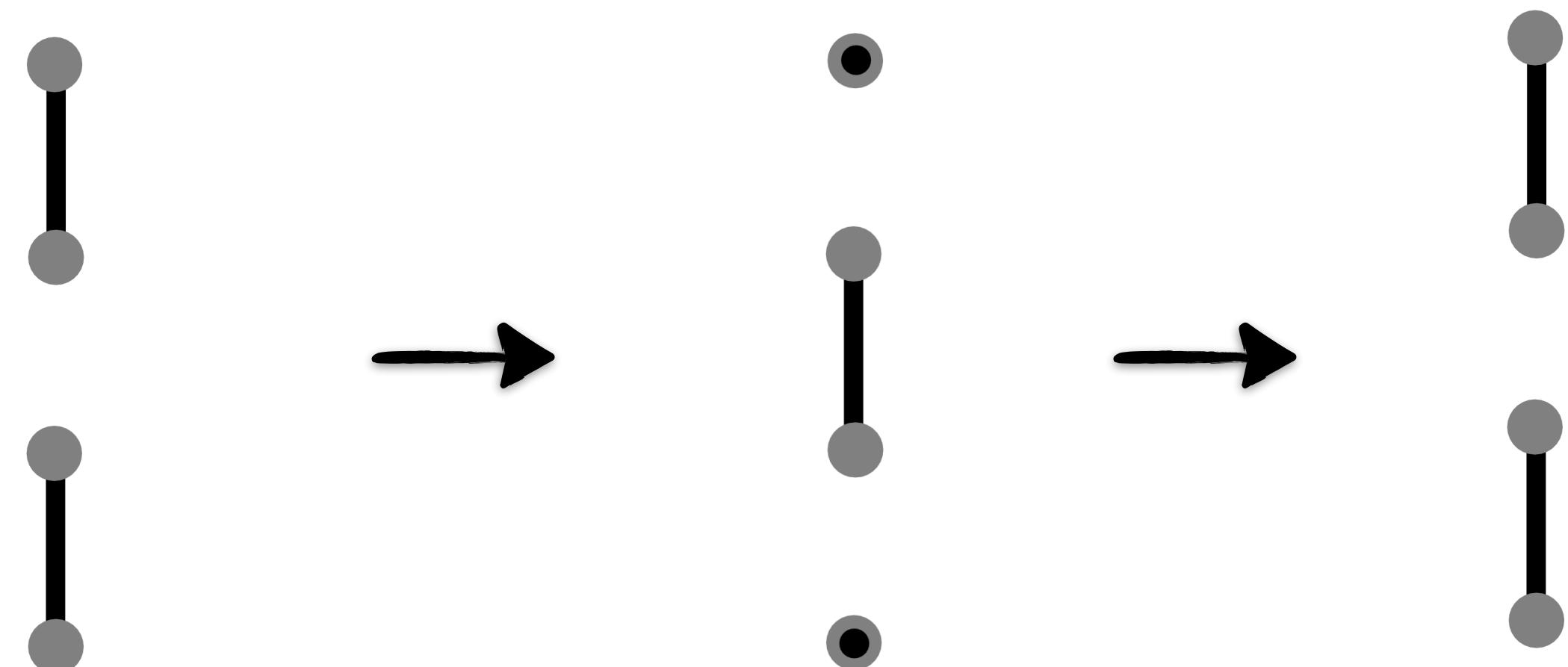


Hartree–Fock error: 167 mEH



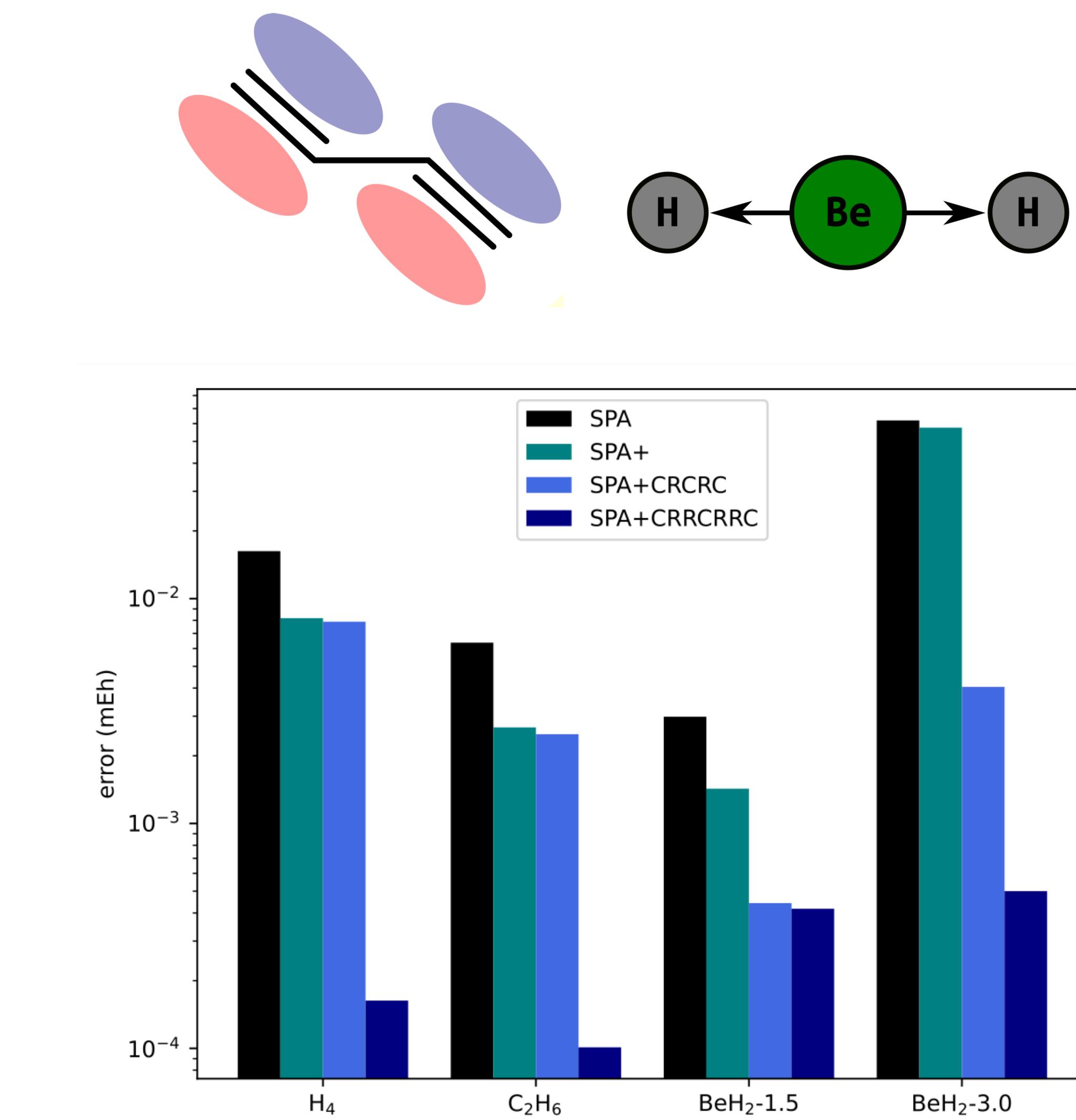
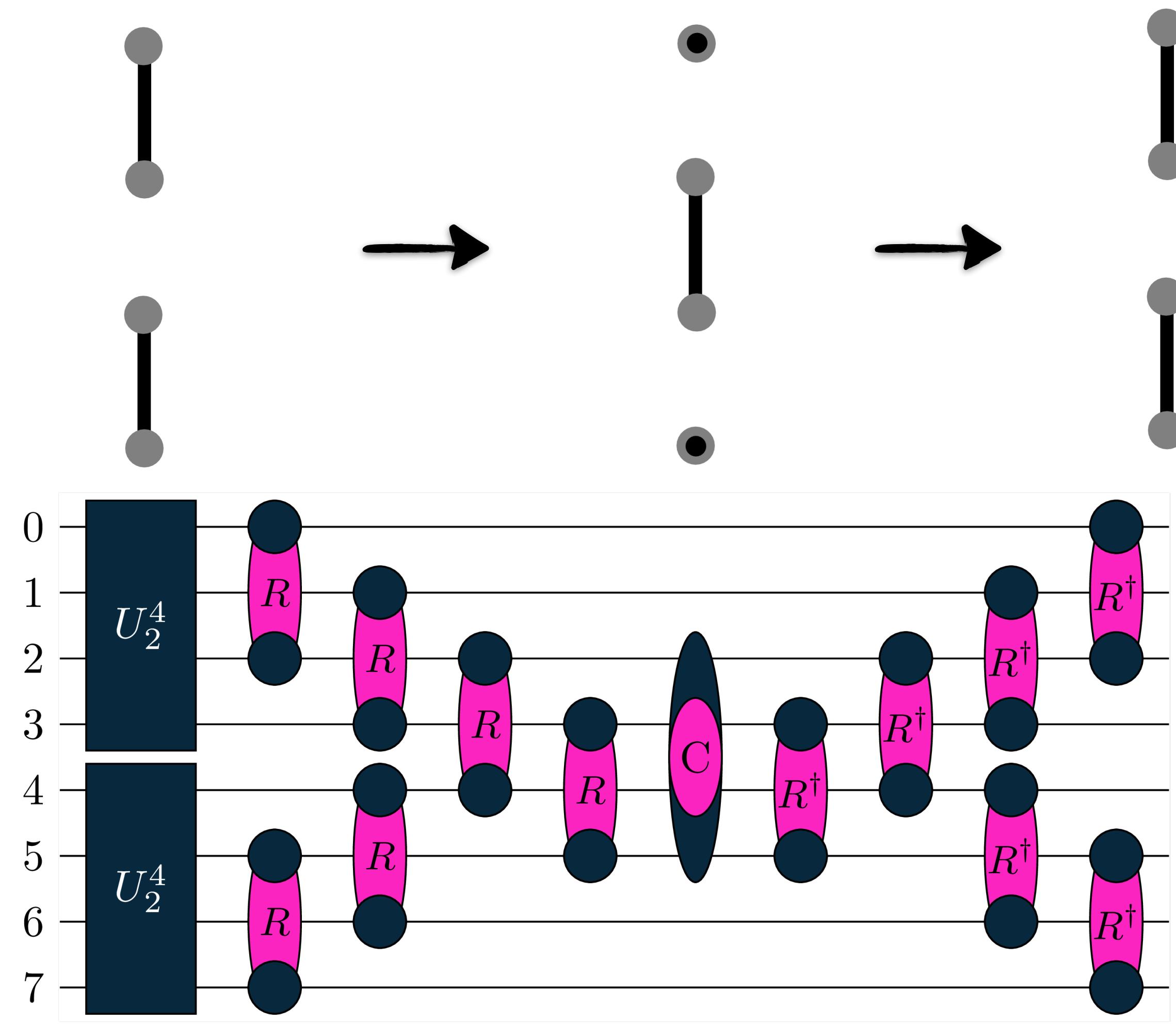
method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*
SPA+	8	96	6	116	131	10
SPA+CRRCRRC	0	100	19	334	367	160
UpCCD	103	88	4	20	26	8
UpCCSD	86	74	12	148	193	13
UpCCGSD	86	74	18	188	254	13
2-UpCCGSD	32	90	36	432	540	48

Hartree–Fock error: 167 mEH

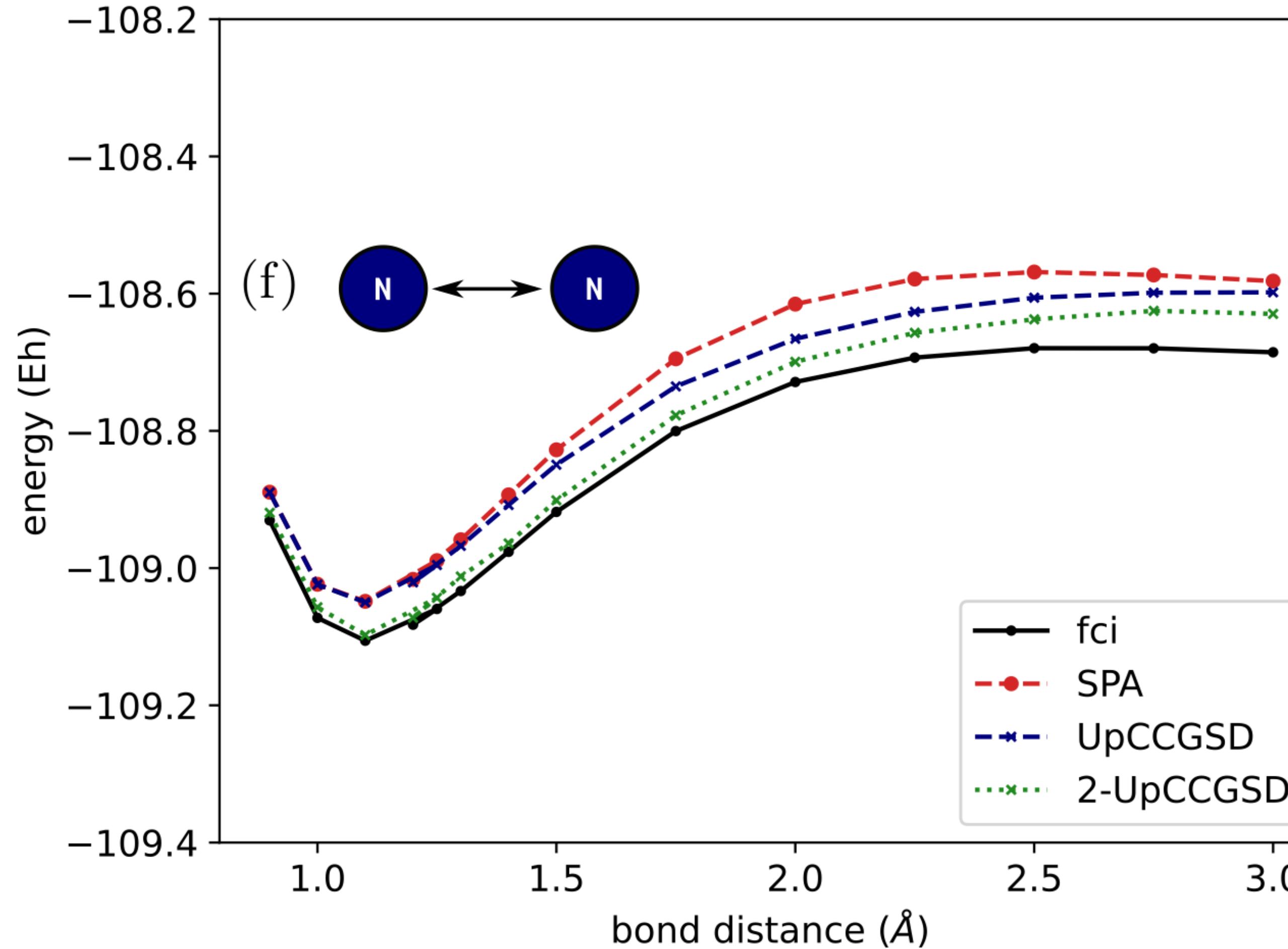


method	error	$F$	$N_v$	cnots	depth	iter
SPA	16	94	2	6	3	17*
SPA+	8	96	6	116	131	10
SPA+CRRCRRC	0	100	19	334	367	160
UpCCD	103	88	4	20	26	8
UpCCSD	86	74	12	148	193	13
UpCCGSD	86	74	18	188	254	13
2-UpCCGSD	32	90	36	432	540	48
ADAPT(UpCCGSD)	32	90	12	448	442	113
ADAPT(UCCGSD)	0	100	21	1360	1705	58

Hartree–Fock error: 167 mEH



# Limitations



- Well behaved SPA
- Improvement through other graphs not obvious

# Implementation: Tequila



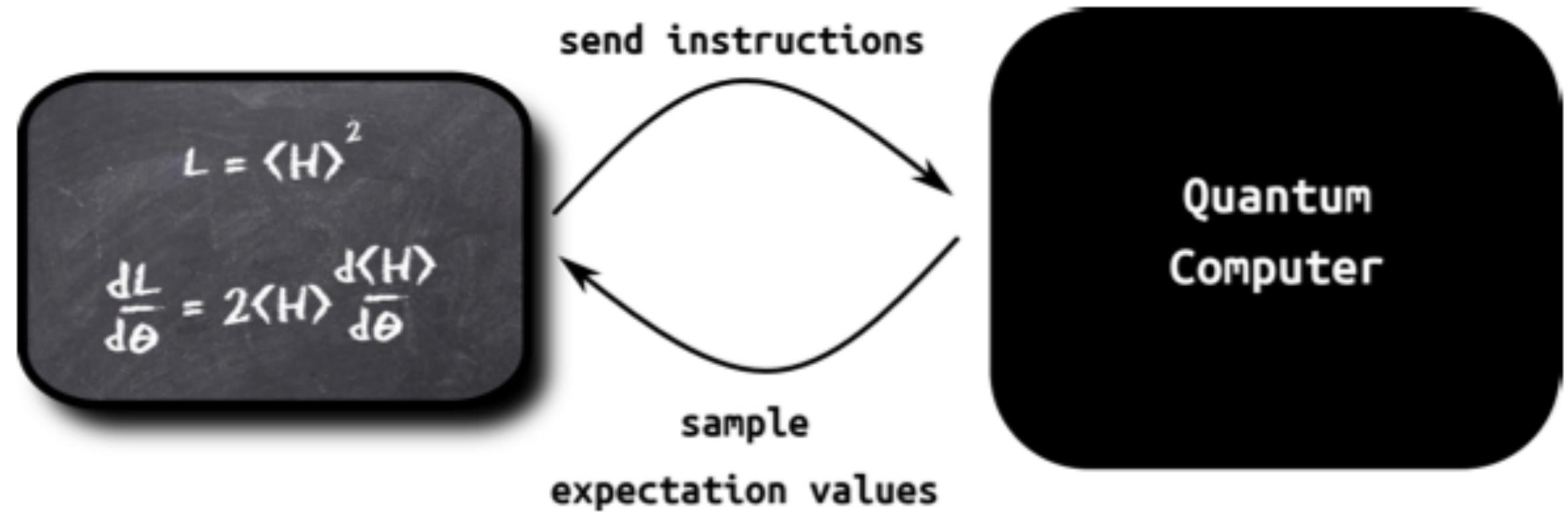
Sumner  
Alperin-Lea  
UofT/Chem



Alba  
Cervera-Lierta  
Barcelona Supercomputing Center



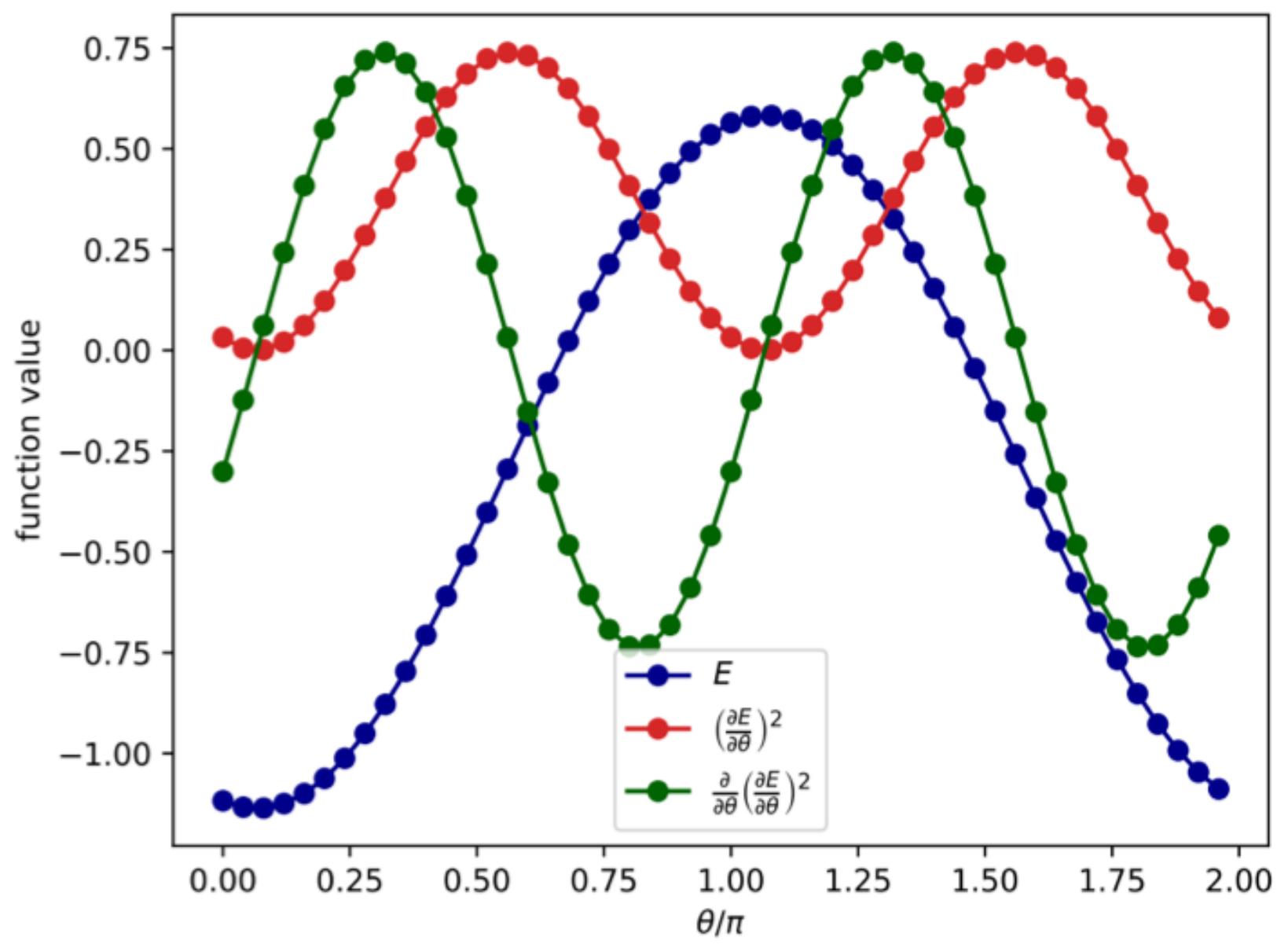
Teresa  
Tamayo-Mendoza  
Harvard



[github.com/tequilahub](https://github.com/tequilahub)

API inspired by `madness` library

```
E    = ExpectationValue(H,U)
dE   = grad(E, "a")
dE2  = dE**2
ddE2 = grad(dE2, "a")
```



# Automatisation

```

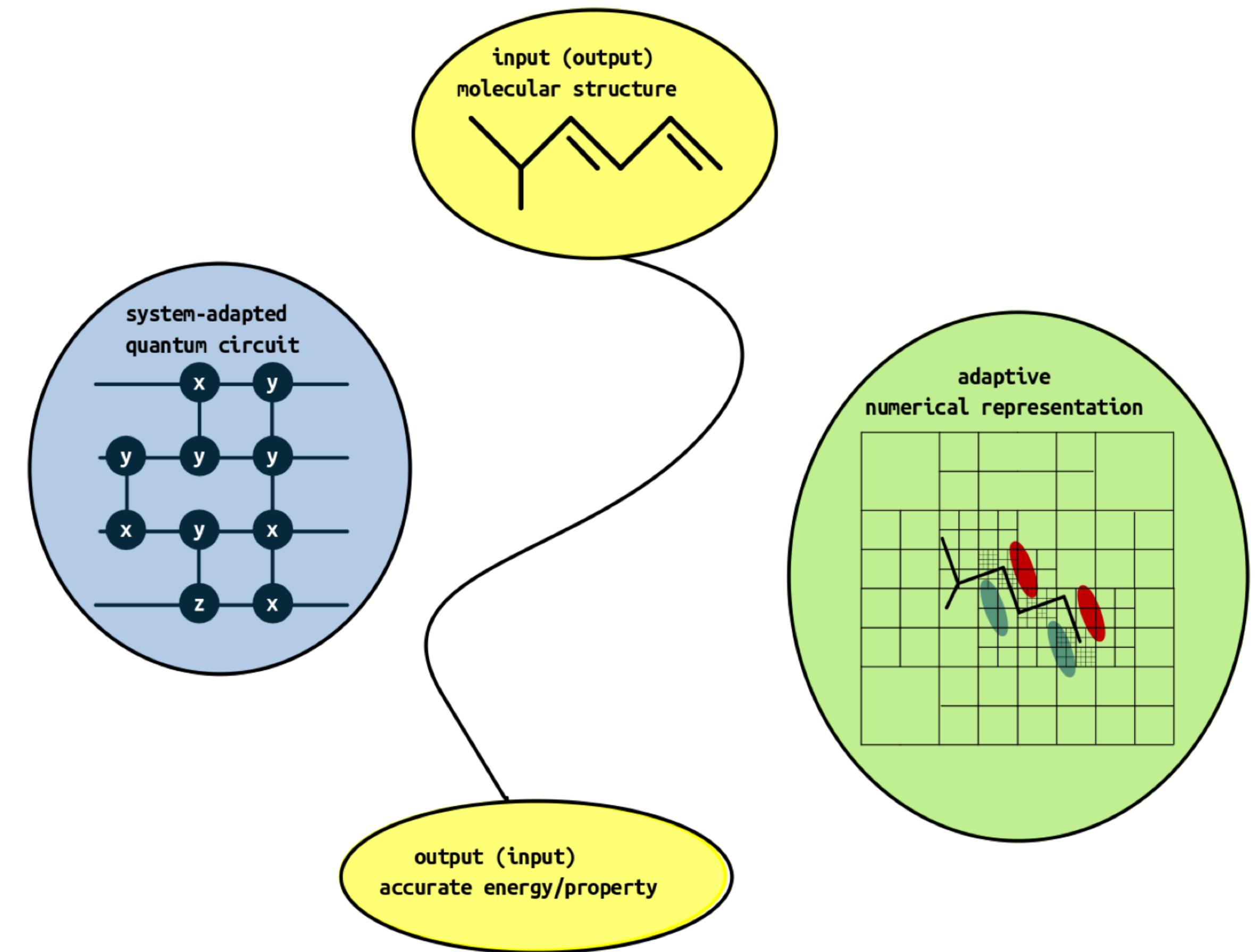
import tequila as tq

mol = tq.Molecule(geometry="beh2.xyz")

H = mol.make_hamiltonian()
U = mol.make_ansatz(name="SPA")
E = tq.ExpectationValue(H=H,U=U)

result = tq.minimize(E)

```



## Optimized Low-Depth Quantum Circuits for Molecular Electronic Structure using a Separable Pair Approximation

```

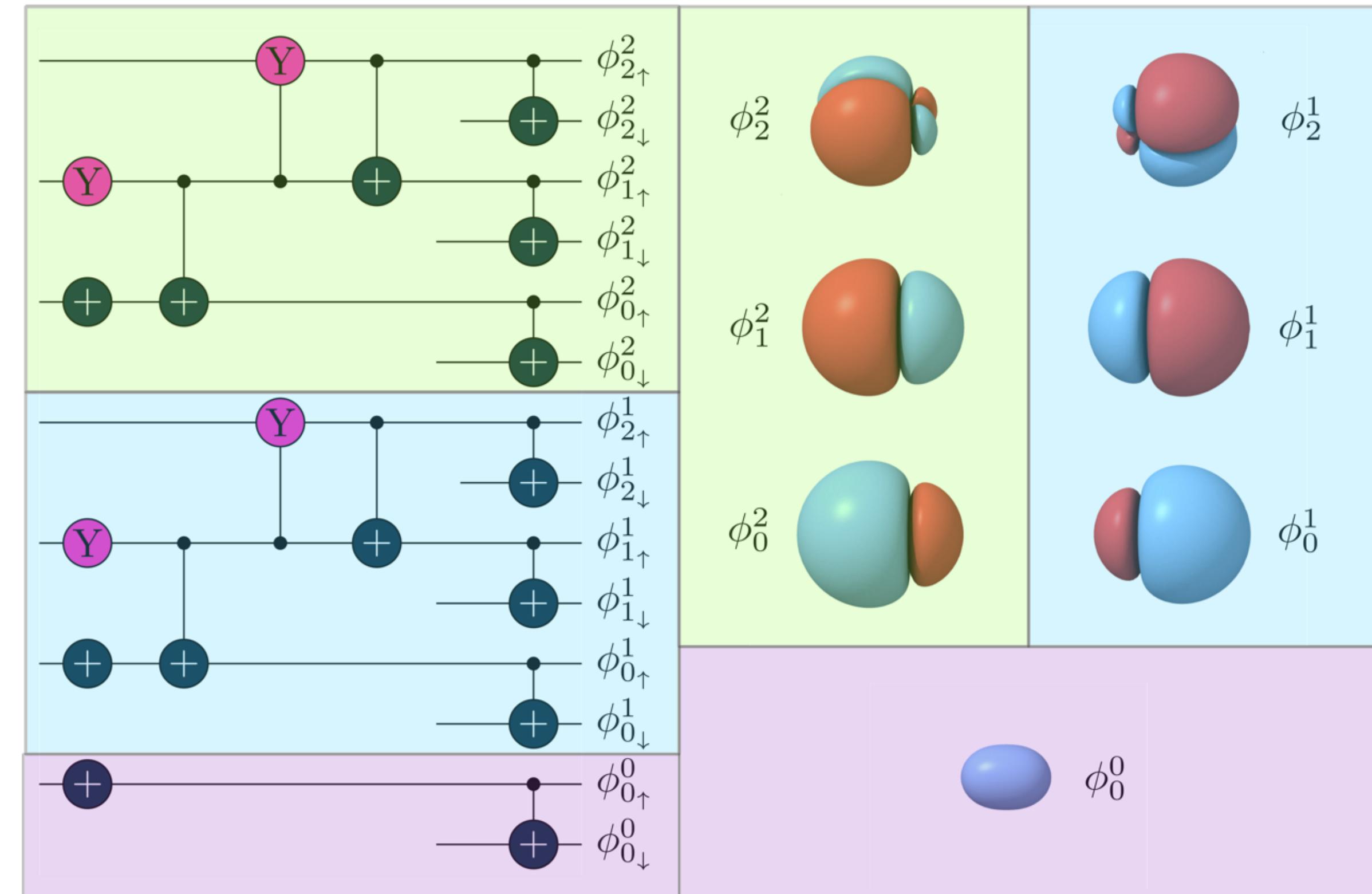
import tequila as tq

mol = tq.Molecule(geometry="beh2.xyz")

H = mol.make_hamiltonian()
U = mol.make_ansatz(name="SPA")
E = tq.ExpectationValue(H=H,U=U)

result = tq.minimize(E)

```

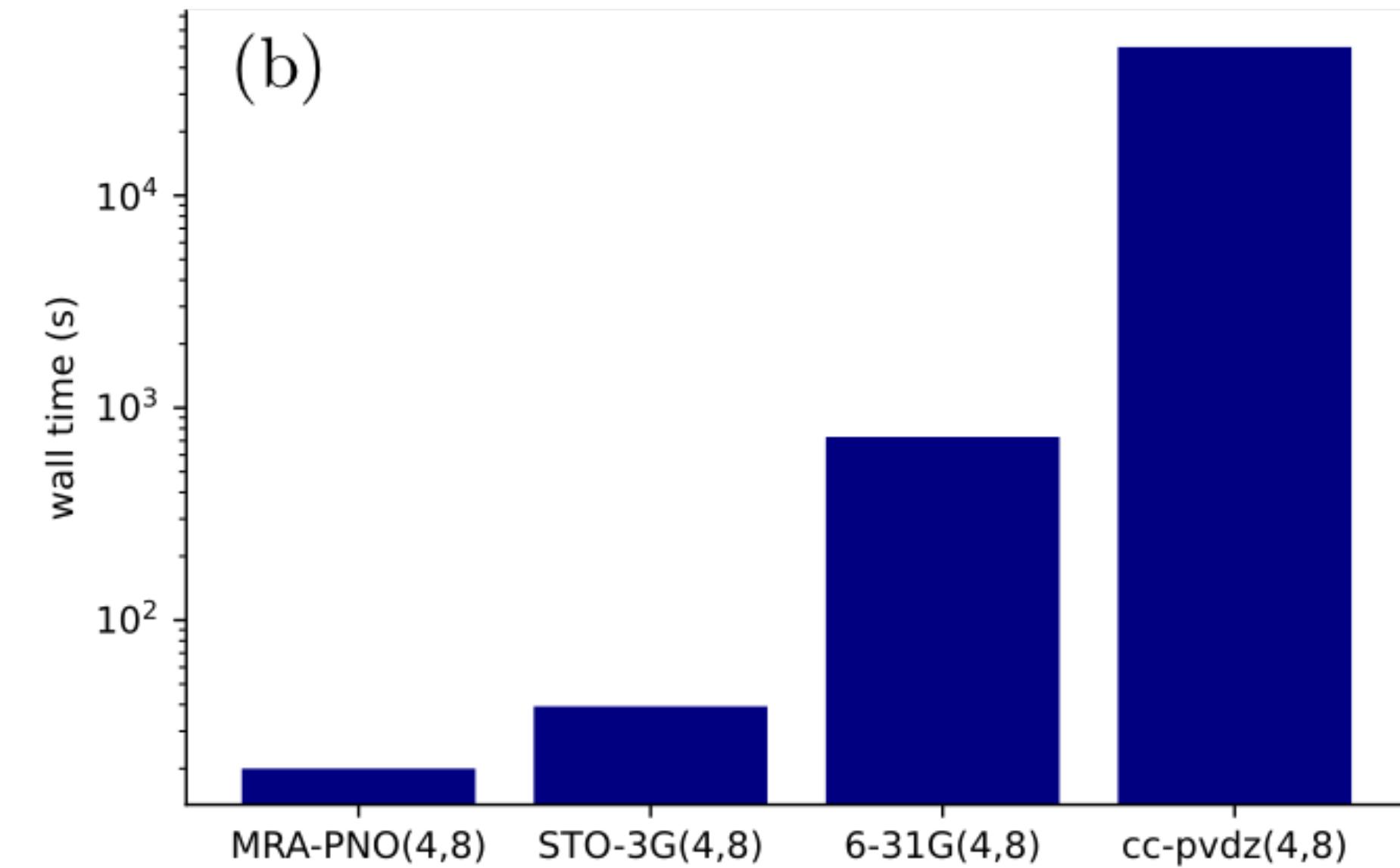
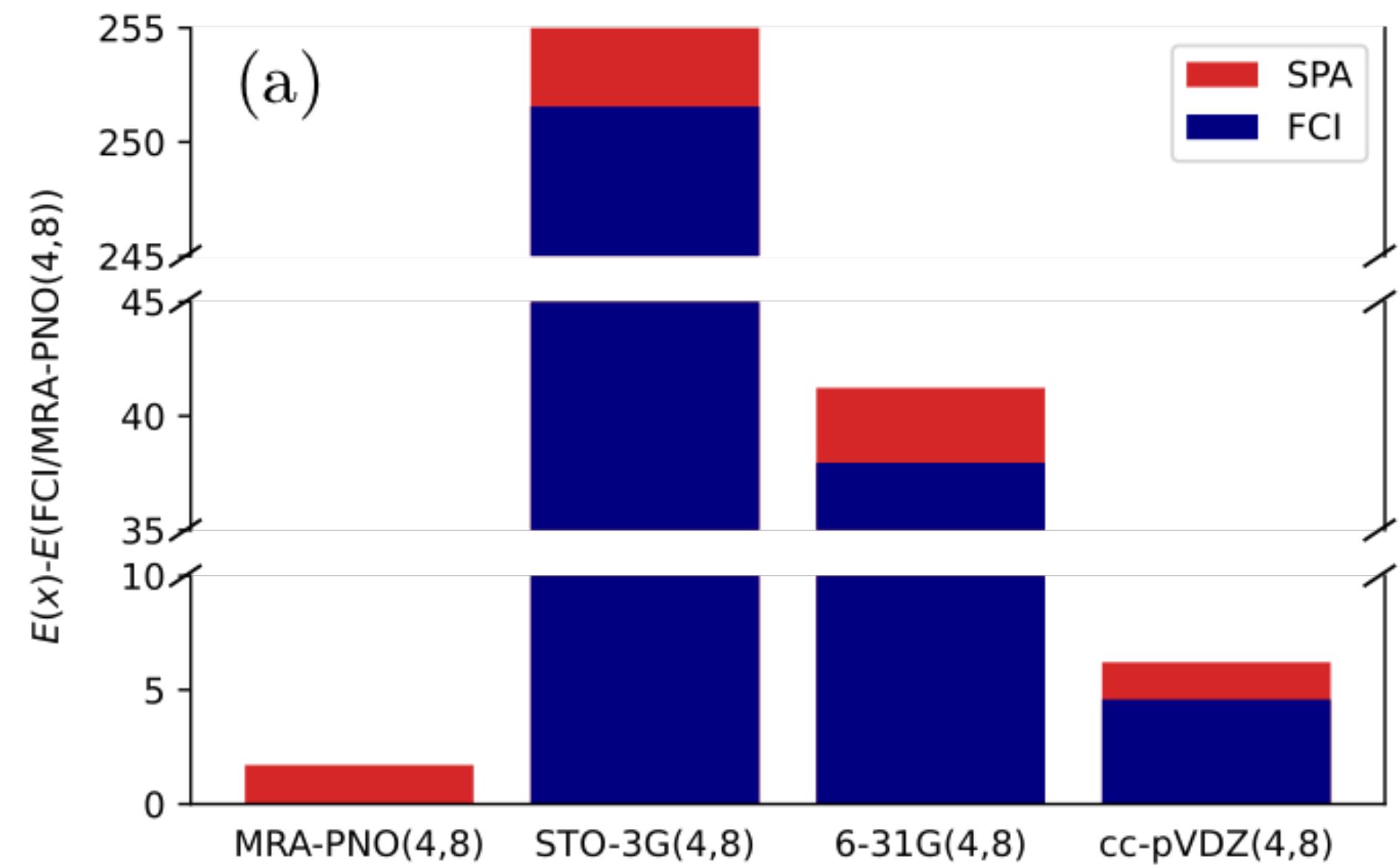


Optimized Low-Depth Quantum Circuits for Molecular Electronic Structure using  
a Separable Pair Approximation

Jakob S. Kottmann<sup>1, 2, \*</sup> and Alán Aspuru-Guzik<sup>1, 2, 3, 4, †</sup>

# Why Basis-Set-Free?

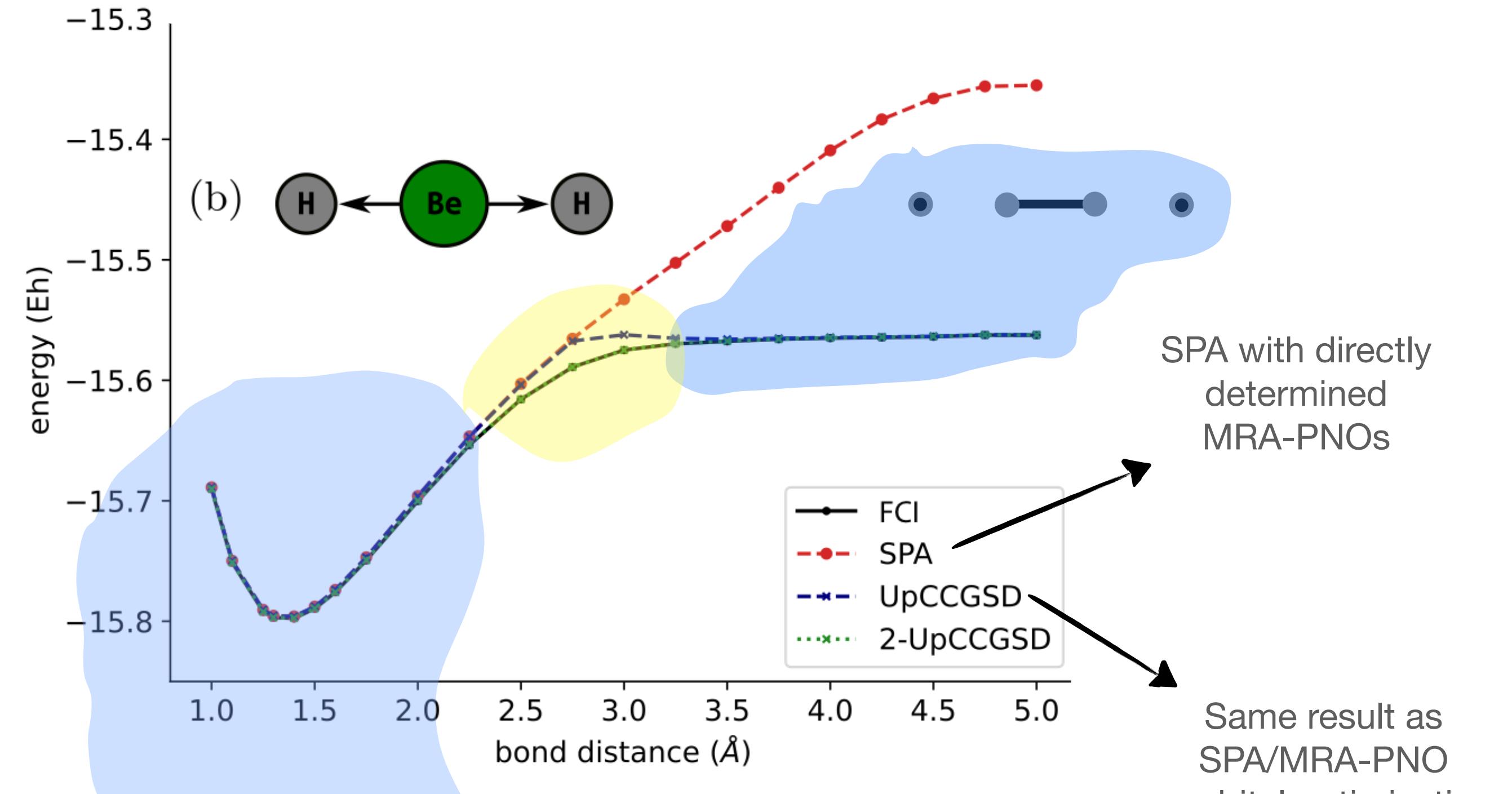
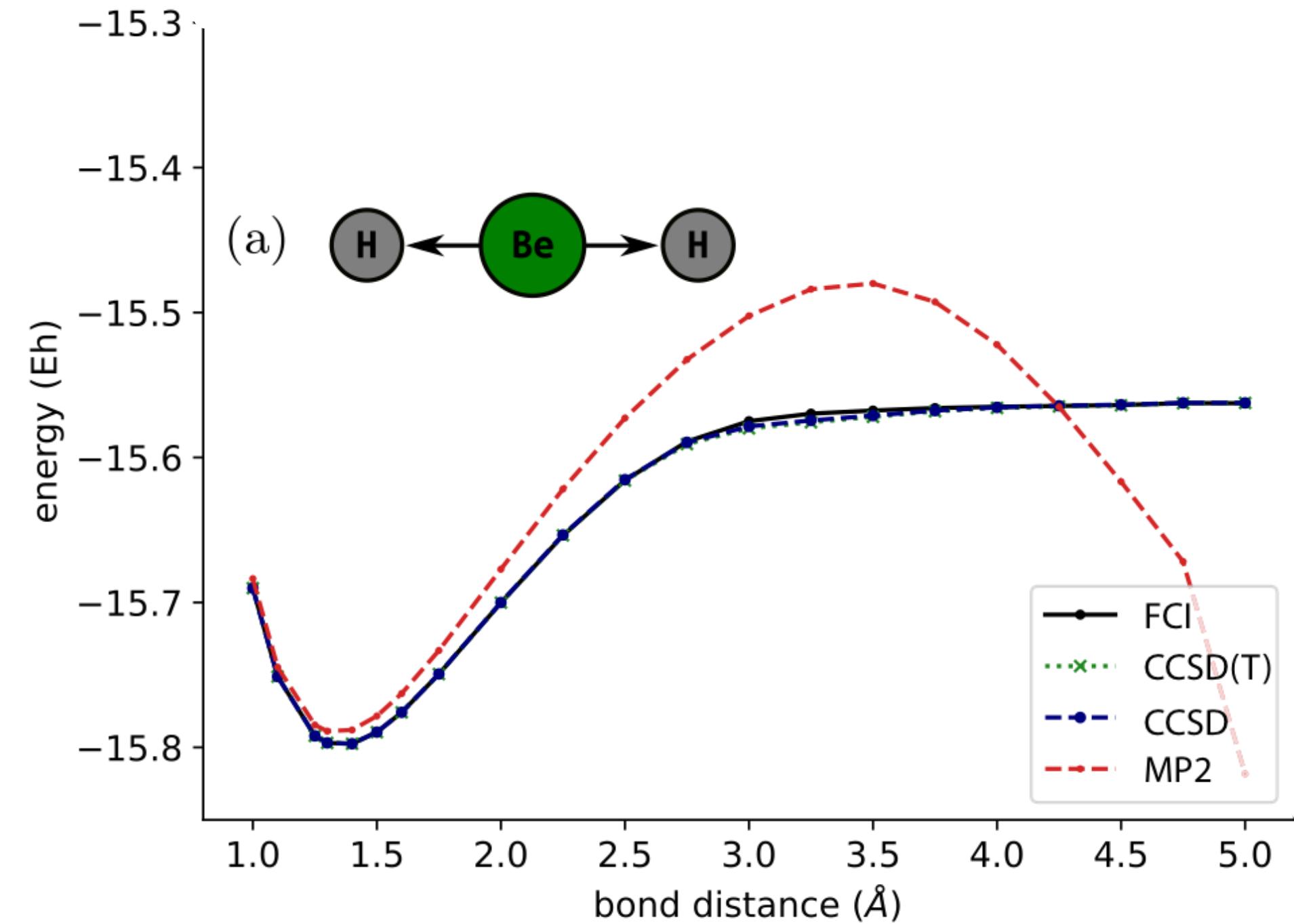
faster and more accurate



BeH<sub>2</sub> with 8 qubits

Standard basis sets: tequila + pyscf

MRA-PNOs: tequila + madness



## Optimized Low-Depth Quantum Circuits for Molecular Electronic Structure using a Separable Pair Approximation

Jakob S. Kottmann<sup>1, 2, \*</sup> and Alán Aspuru-Guzik<sup>1, 2, 3, 4, †</sup>

SPA with directly determined MRA-PNOs

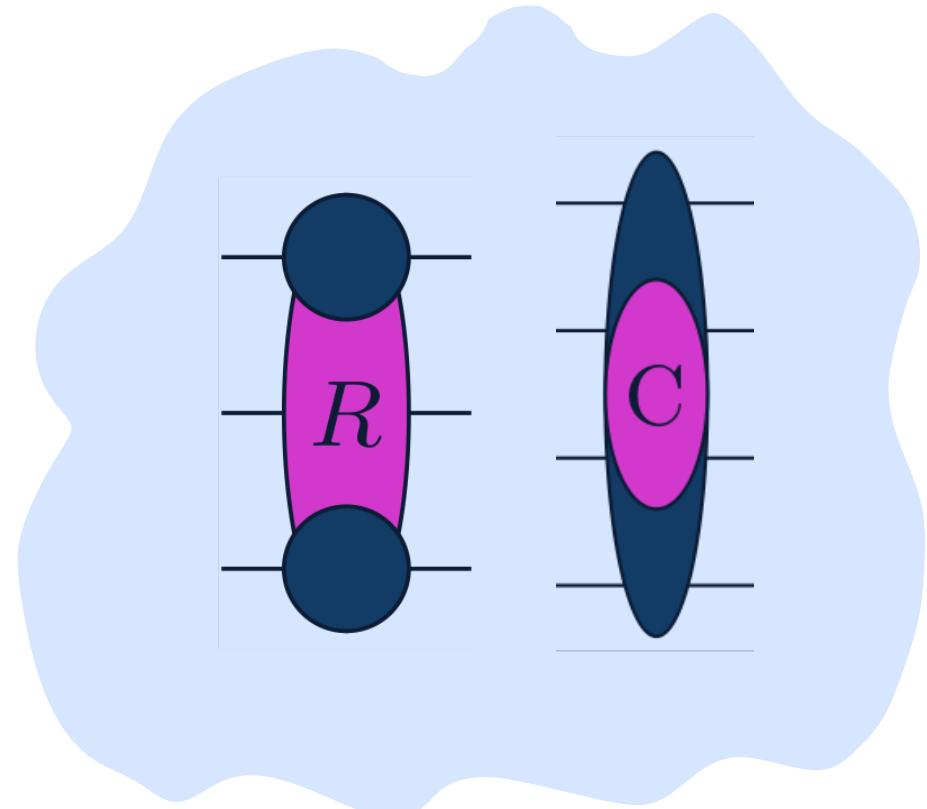
Same result as SPA/MRA-PNO + orbital optimisation

Classically simulable

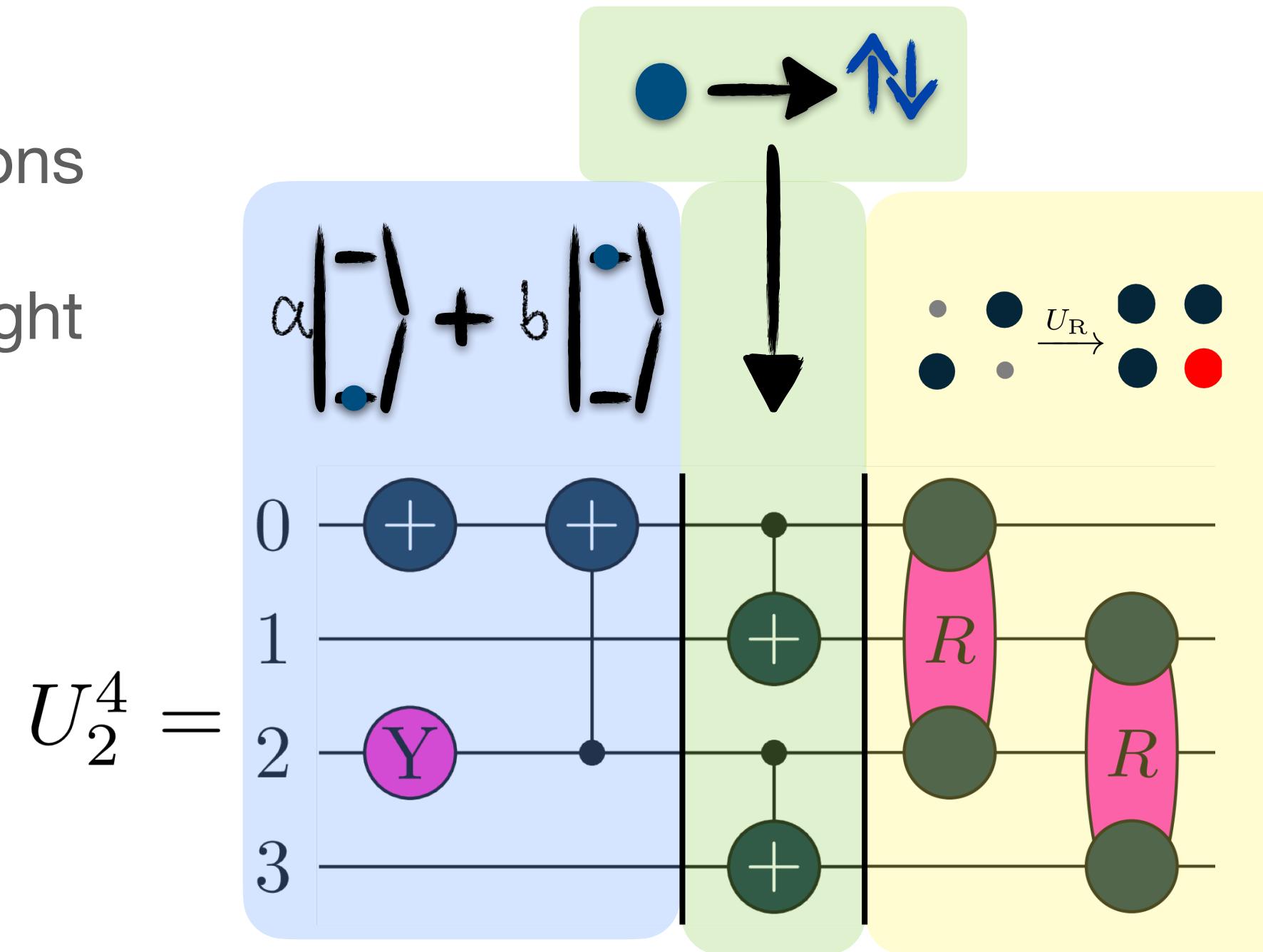
Interesting region

# Summary

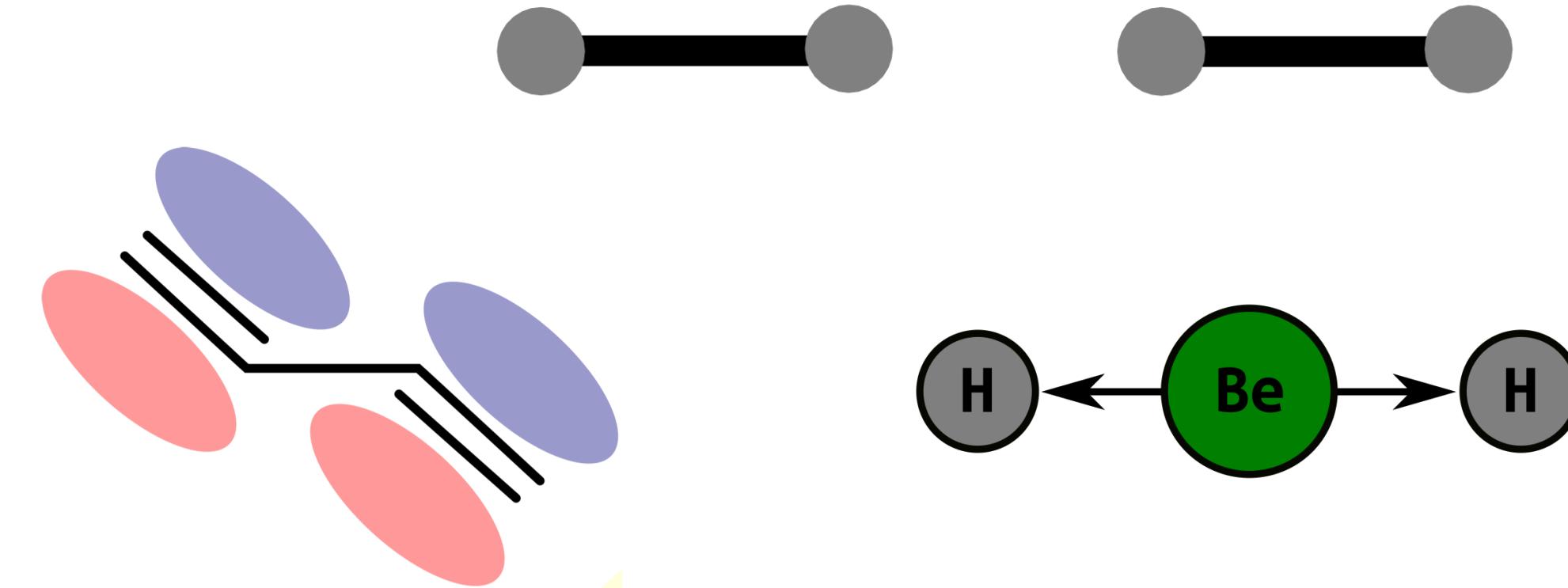
Basic Building Blocks



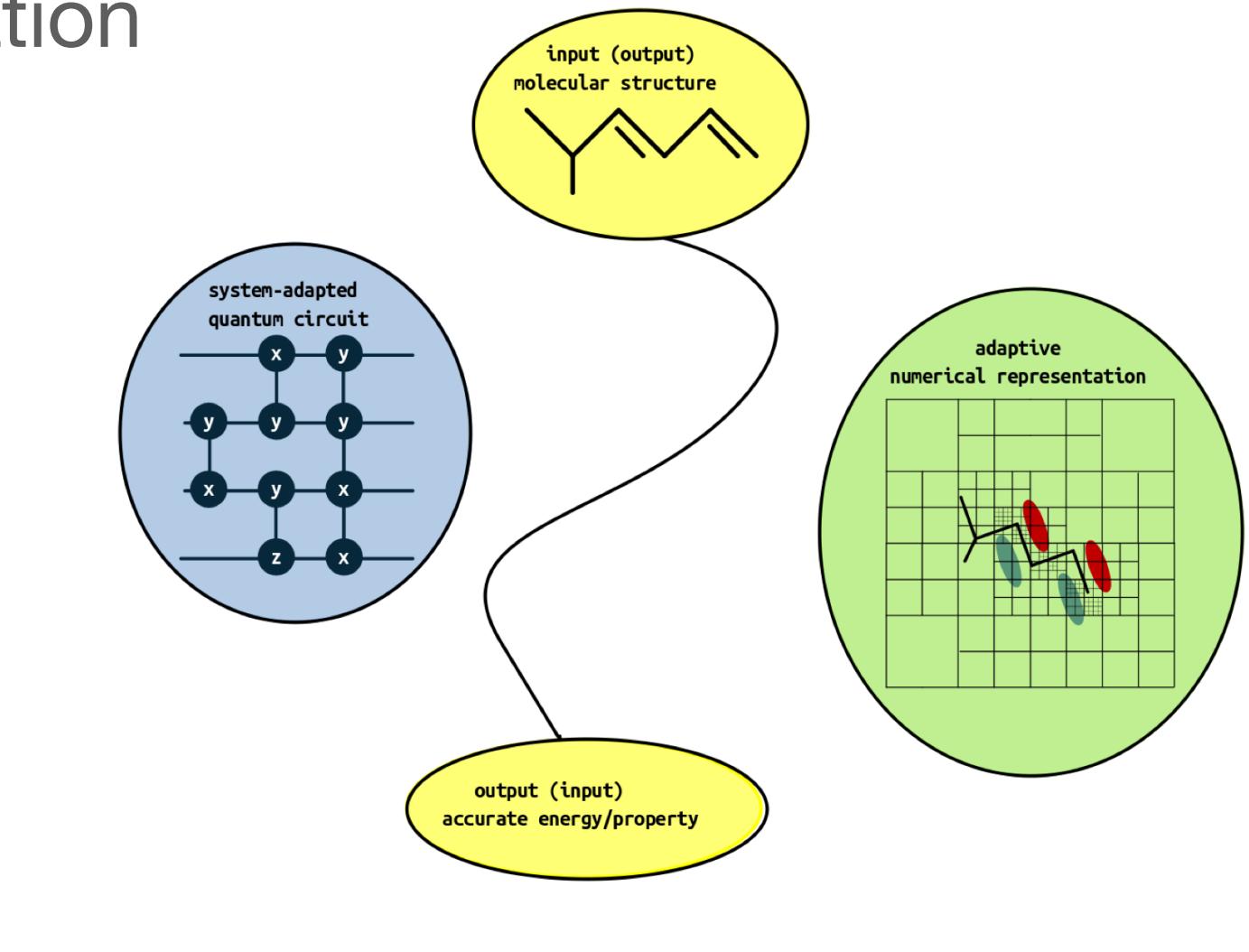
Approximations and Physical Insight



Abstraction & Transferability



Automatisation





Universität  
Augsburg  
University

**Positions available  
(PhD and Postdoc)  
Starting end of 2022**

