

# Quantum Chemistry on a Quantum Computer

Susanne Yelin

Harvard University

Simons Institute, Berkeley, April 25, 2024

Programmable Simulations of Molecules and Materials with  
Reconfigurable Quantum Processors, arXiv:2312.02265

# Collaborators



Nishad Maskara  
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→ Rice)



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(Harvard)



Abigail McClain  
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(Harvard)



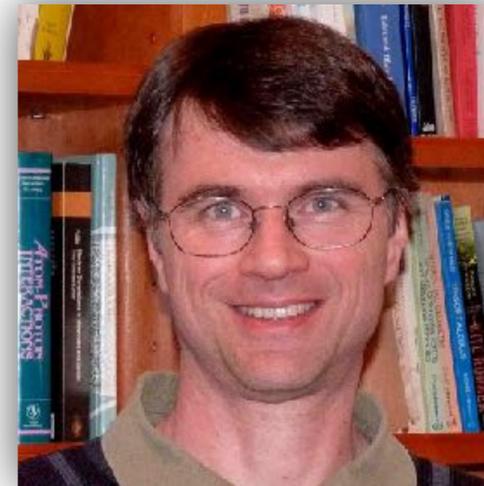
Derek Wang  
(Harvard  
→ IBM Quantum)



Anna Krylov  
(USC, Los Angeles)



Norman Yao  
(Harvard)



Martin Head-Gordon  
(UC Berkeley)



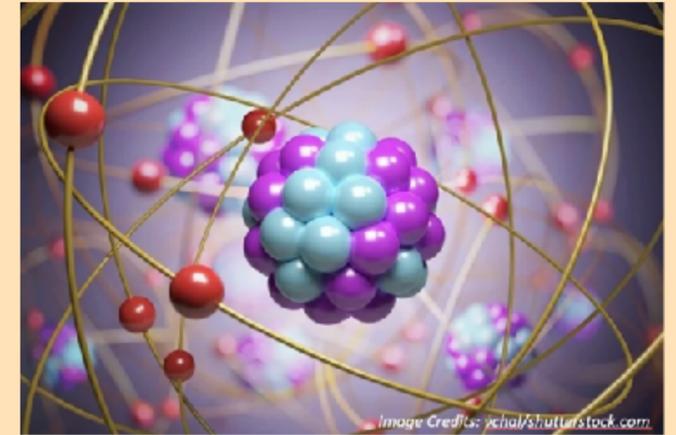
Mikhail Lukin  
(Harvard)

**And many thanks to:** Dolev Bluvstein, Madelyn Cain, Joonho Lee, Nathan Leitao, Kushal Seethram, and many more.

# Quantum Chemistry and Material Science

## Quantum Chemistry

= solving an electronic structure problem for a configuration of electrons and nuclei



Major thrust of quantum chemistry: quantitative prediction of material or molecular properties

Full Hamiltonian:

$$H = - \sum_i \frac{\hbar^2 \nabla^2}{2m_i} - \sum_{k \neq i} \frac{Z_k e^2}{|\mathbf{R}_k - \mathbf{r}_i|} + \sum_{i \neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Challenge: Simulating systems with **strong correlations**

➤ Unfavorable Hilbert space scaling motivates use of **quantum computers**

# Quantum Chemistry on Quantum Computers

Assessing requirements to scale to practical quantum advantage

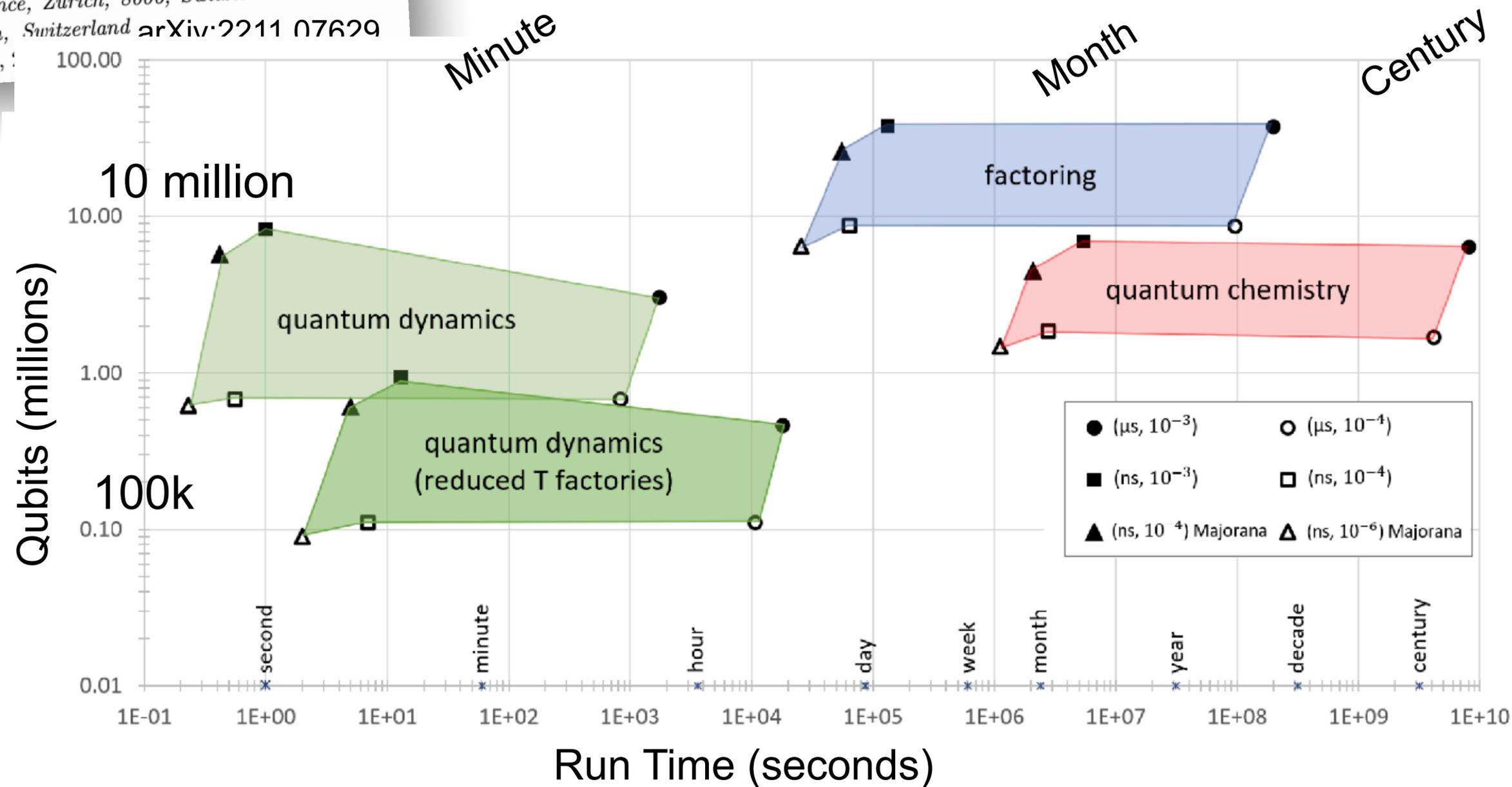
M. E. Beverland,<sup>1</sup> P. Murali,<sup>1</sup> M. Troyer,<sup>1</sup> K. M. Svore,<sup>1</sup> T. Hoefler,<sup>2</sup>  
 V. Kliuchnikov,<sup>1</sup> G. H. Low,<sup>1</sup> M. Soeken,<sup>3</sup> A. Sundaram,<sup>1</sup> and A. Vaschillo<sup>1</sup>

<sup>1</sup>Microsoft Quantum, Redmond, WA 98052, USA

<sup>2</sup>ETH Zurich, Department of Computer Science, Zürich, 8006, Switzerland

<sup>3</sup>Microsoft Quantum, Zurich, Switzerland arXiv:2211.07629

(Dated: November 19, 2022)



# Quantum Chemistry on Quantum Computers

Assessing requirements to scale to practical quantum advantage

M. E. Beverland,<sup>1</sup> P. Murali,<sup>1</sup> M. Troyer,<sup>1</sup> K. M. Svore,<sup>1</sup> T. Hoefler,<sup>2</sup>  
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<sup>2</sup>ETH Zurich, Department of Computer Science, Zürich, 8006, Switzerland

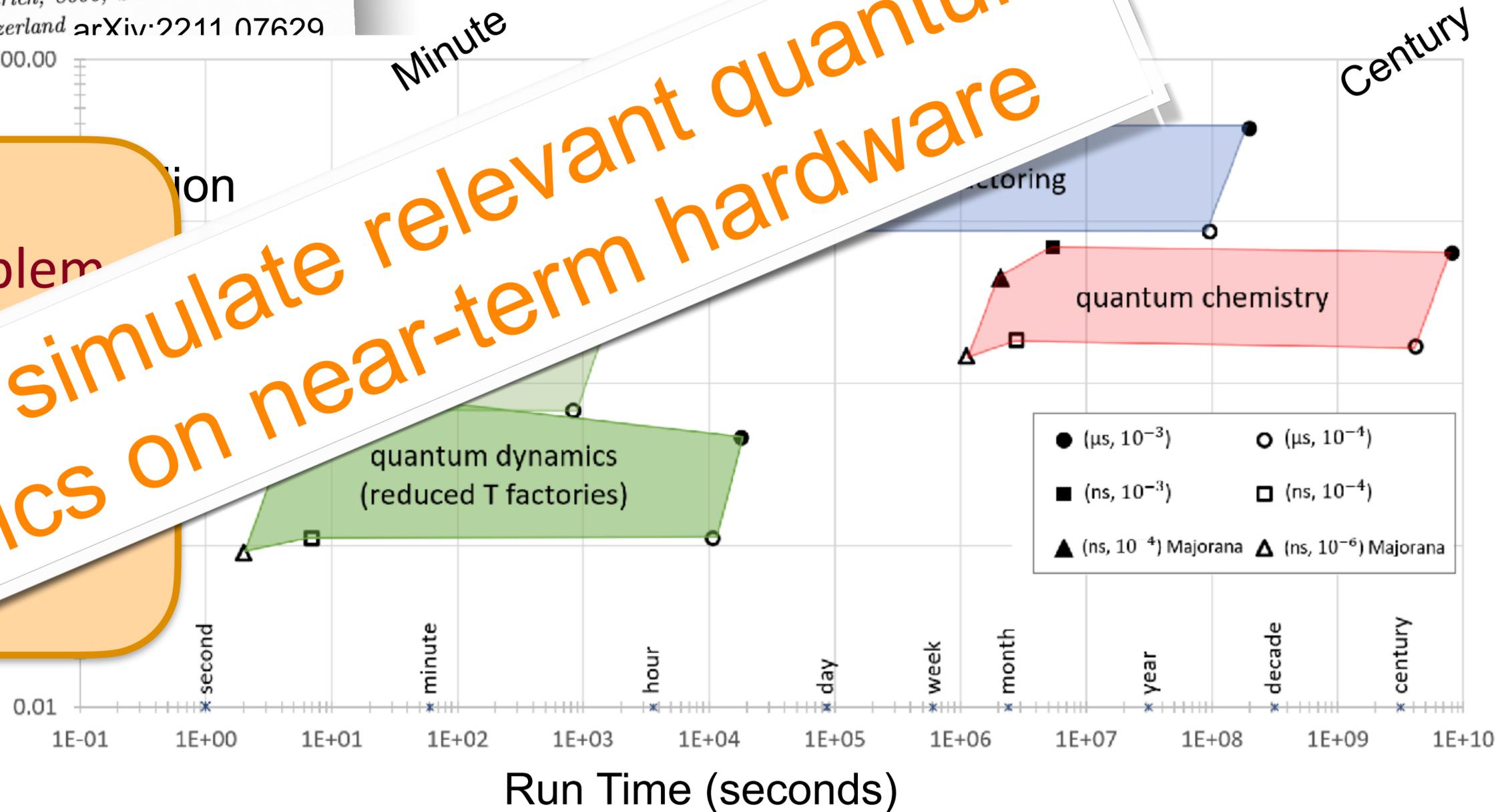
<sup>3</sup>Microsoft Quantum, Zurich, Switzerland arXiv:2211.07629

(Dated: November 19, 2022)

## Sources of overhead:

- Mapping of target problem to quantum hardware
- Expensive algorithms (quantum chemistry)
- Large overheads

Our goal: simulate relevant quantum dynamics on near-term hardware



# Advancing Computational Quantum Chemistry

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## Our approach:

- Leverage insights obtained from state-of-the art **classical computational algorithms**.
- Use state-of-the art **programmable quantum simulators** (e.g., Rydberg atom arrays)
- Focus on **hardware-efficient** implementations on near term devices.

## What problems do need a quantum computer?

problems with strong correlations

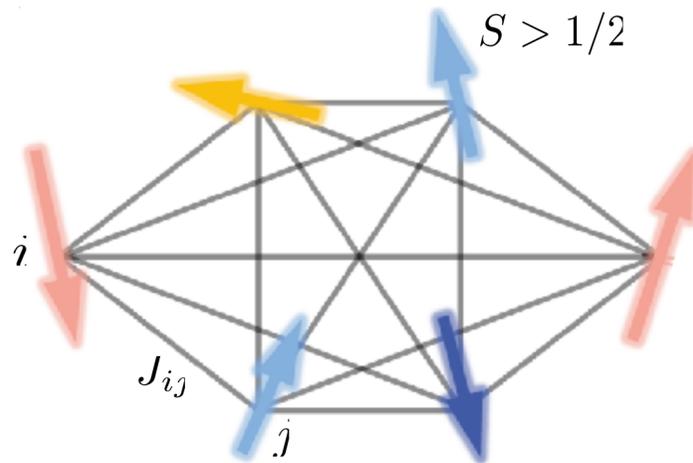
# Hybrid Quantum-Classical Workflow

## High-performance computing



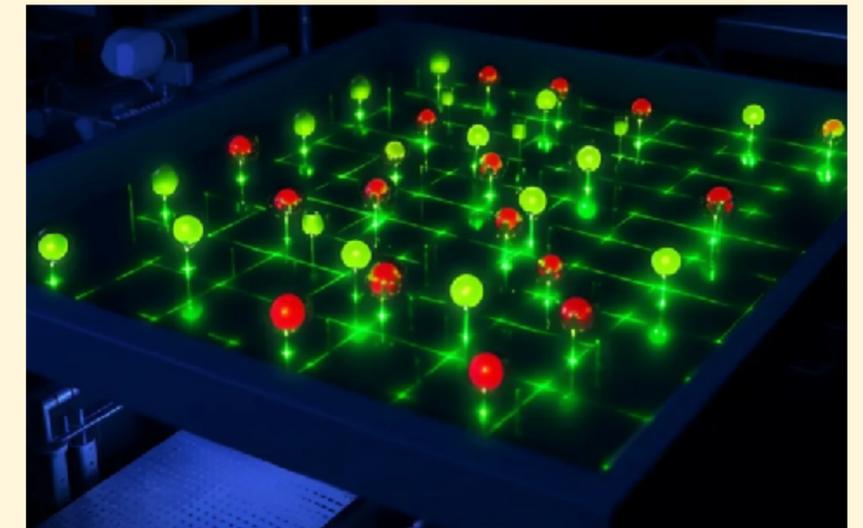
- Employ state-of-the-art algorithms (Coupled Cluster, DMRG, etc.)

## Model Hamiltonians for quantum chemistry and material science



capture the strong correlations (at low energies)

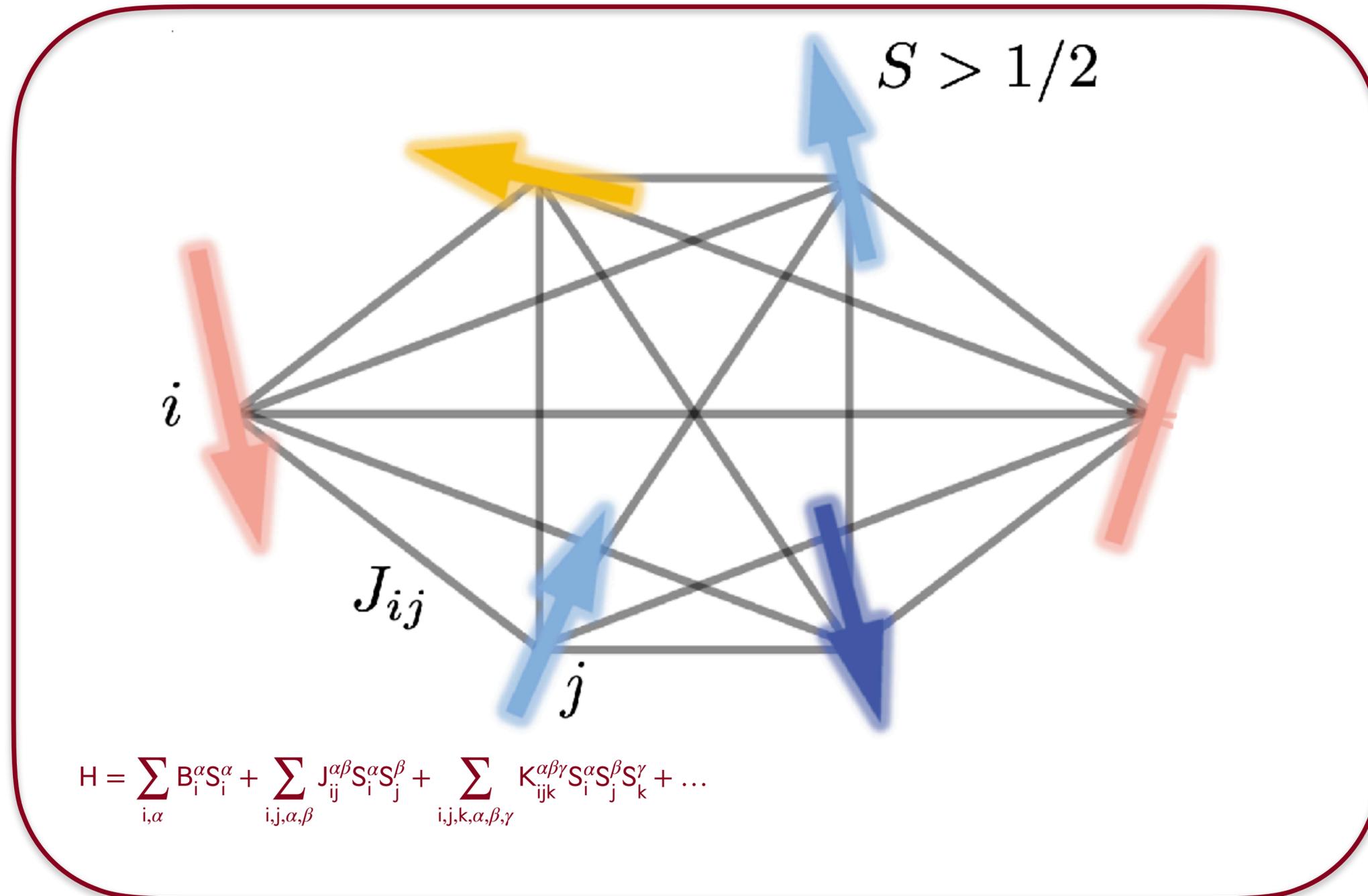
## Large scale analog-digital quantum simulator



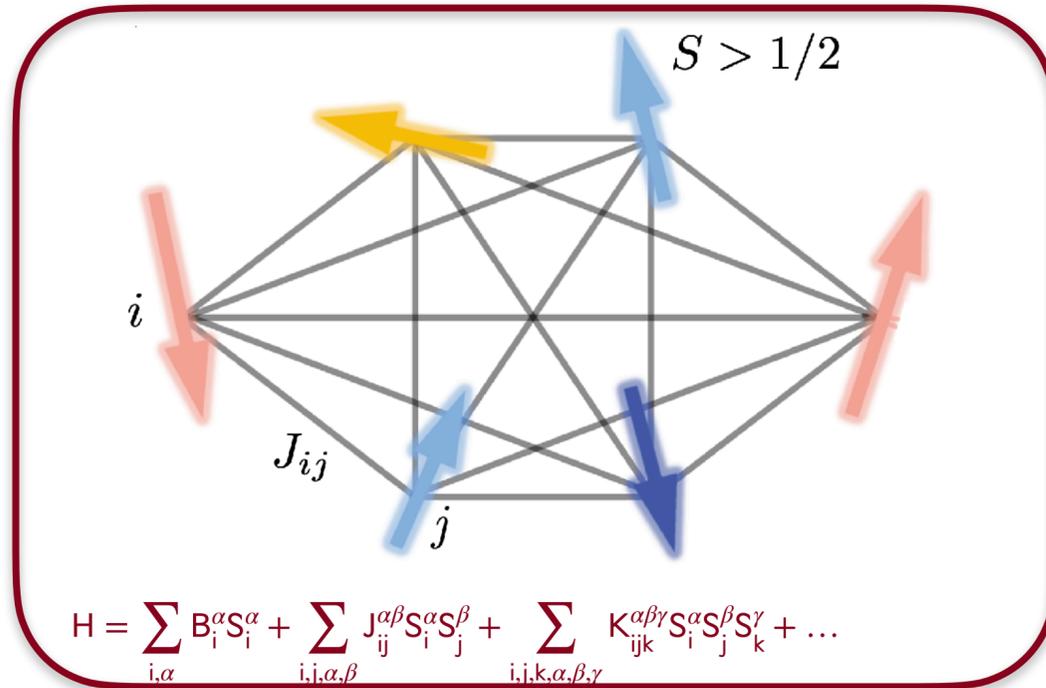
- Efficient multi-qubit gates
- High-degree of programmability  
→ Simulate time dynamics of many-particle systems

# Target Problem

Model Hamiltonian with spin  $S > 1/2$  and arbitrary connectivity



# Target Problem



see for example:

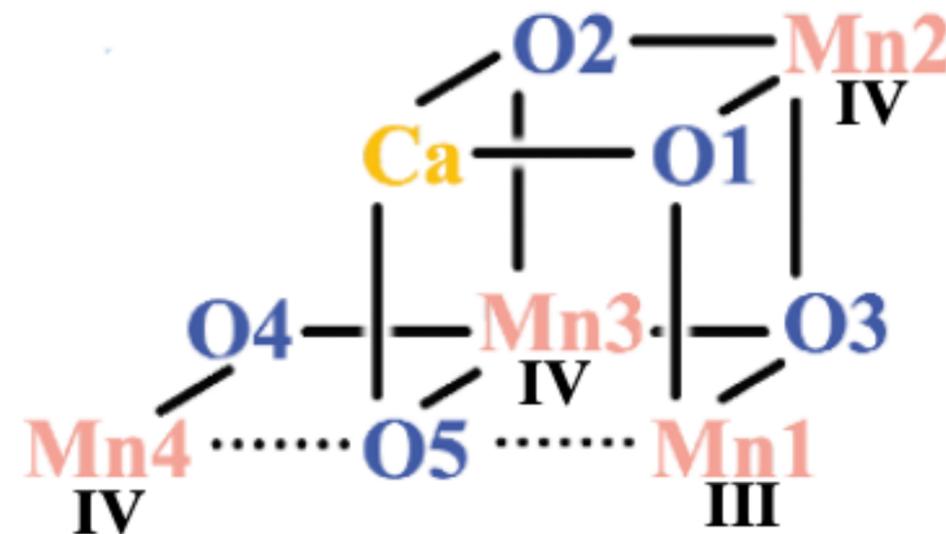
Mayhall, Head-Gordon, JPCL **6**, 10, 1982

(2015), JPC **141**, 134111 (2014)

V. Krewald, ..., D.A. Pantazis, Chem. Sci., **6**, 1676 (2015)

S. Kotaru, S. Kähler, M. Alessio, A. I. Krylov, J Comp Chem **44**, 367 (2022), etc.

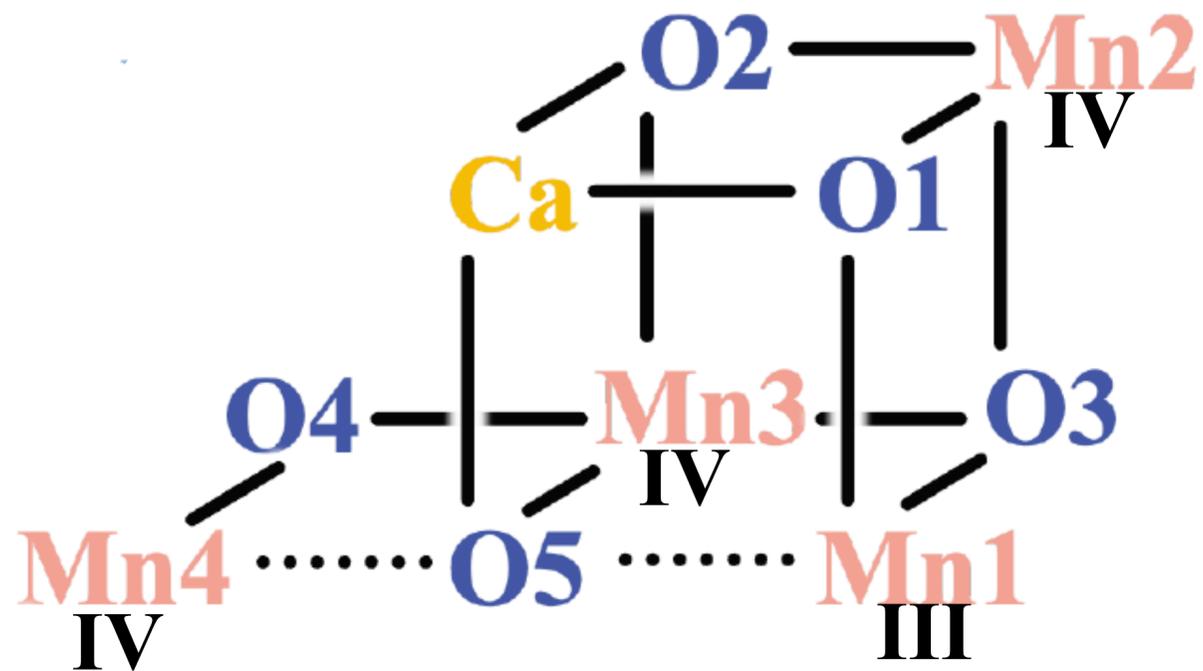
- Good model for materials or molecules with localized valence electrons



- Difficult to solve but easy to obtain from ab-initio quantum chemistry methods
- More natural mapping to quantum hardware

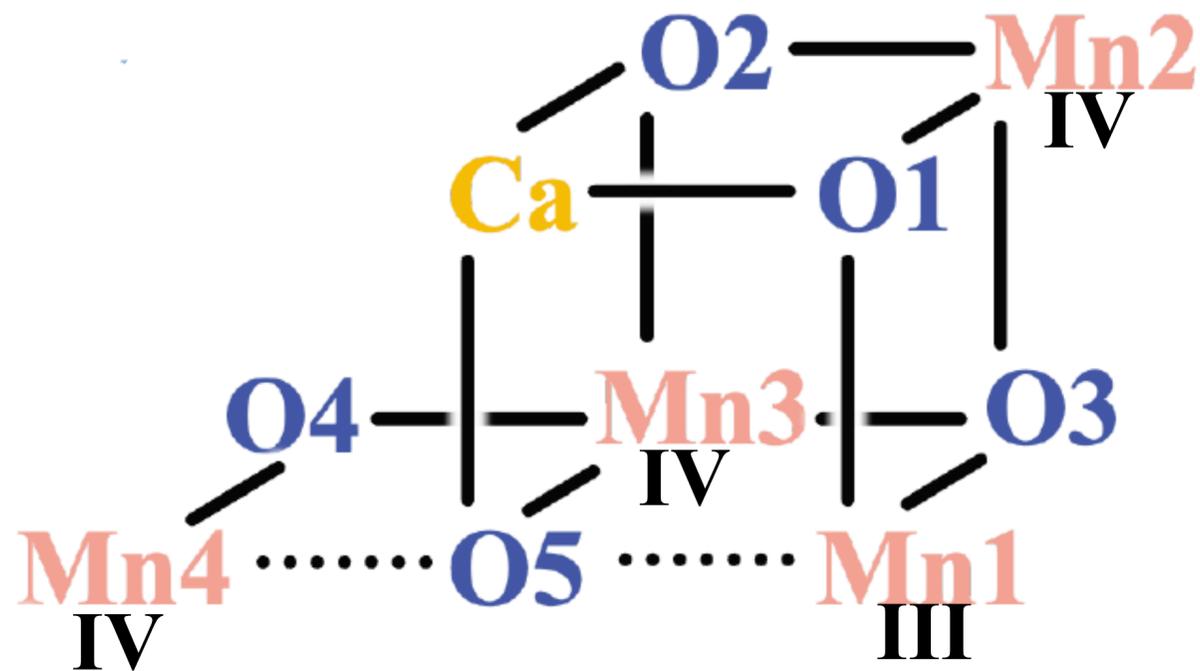
# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



# Model Hamiltonians

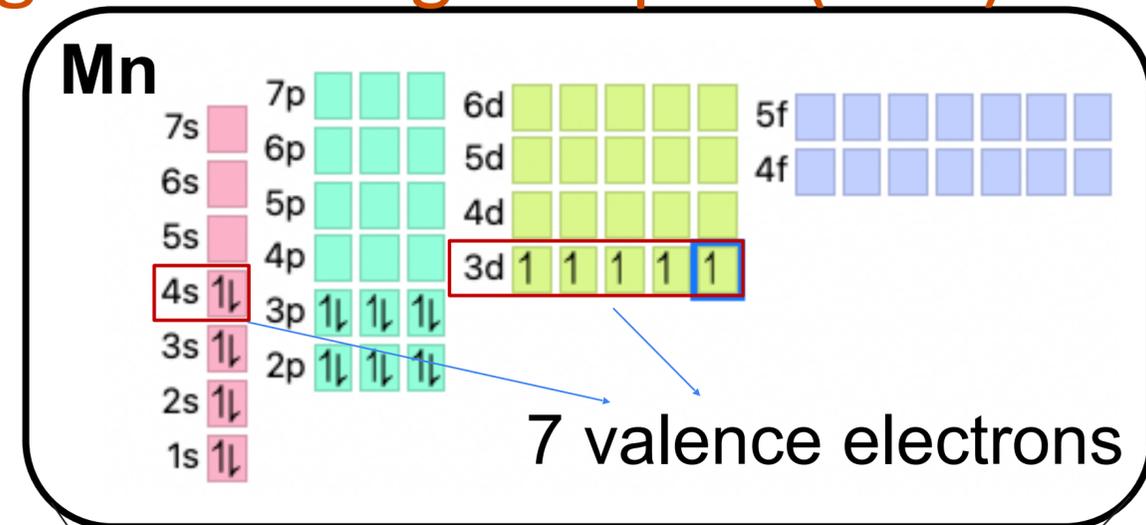
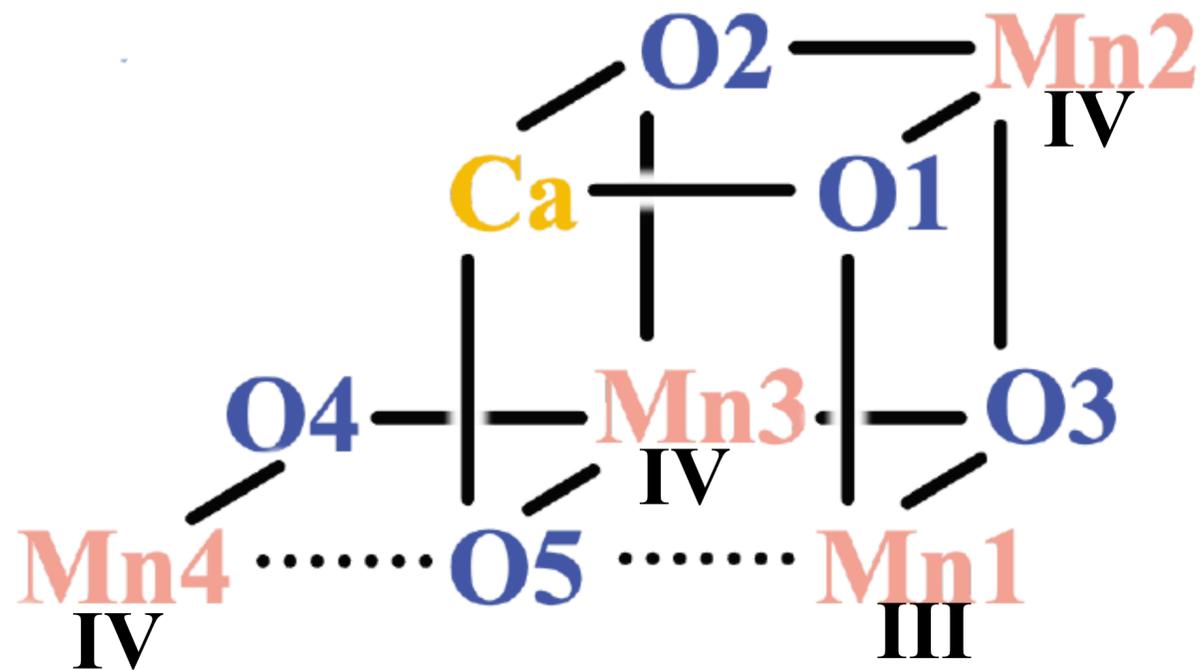
Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18						
												Pnictogens		Chalcogens		Halogens									
1	H Hydrogen -1.1	2	He Helium																	Atomic Symbol Name Weight					
2	3 Li Lithium 1	4	Be Beryllium 2																	5 B Boron 3	6 C Carbon -4.4	7 N Nitrogen -3.35	8 O Oxygen -2	9 F Fluorine -1	10 Ne Neon
3	11 Na Sodium 1	12 Mg Magnesium 2																	13 Al Aluminum 3	14 Si Silicon 4.4	15 P Phosphorus 3.35	16 S Sulfur 2.246	17 Cl Chlorine 1.367	18 Ar Argon	
4	19 K Potassium 1	20 Ca Calcium 2	21 Sc Scandium 3	22 Ti Titanium 4	23 V Vanadium 5	24 Cr Chromium 3.6	25 Mn Manganese 2.47	26 Fe Iron 2.3	27 Co Cobalt 2.3	28 Ni Nickel 2	29 Cu Copper 2	30 Zn Zinc 2	31 Ga Gallium 3	32 Ge Germanium -4.24	33 As Arsenic -3.35	34 Se Selenium -2.246	35 Br Bromine -1.367	36 Kr Krypton							
5	37 Rb Rubidium 1	38 Sr Strontium 2	39 Y Yttrium 3	40 Zr Zirconium 4	41 Nb Niobium 5	42 Mo Molybdenum 4.6	43 Tc Technetium 4.7	44 Ru Ruthenium 3.4	45 Rh Rhodium 3	46 Pd Palladium 2.4	47 Ag Silver 1	48 Cd Cadmium 2	49 In Indium 3	50 Sn Tin -4.24	51 Sb Antimony 3.35	52 Te Tellurium 2.246	53 I Iodine 1.367	54 Xe Xenon 2.46							
6	55 Cs Caesium 1	56 Ba Barium 2	57-71	72 Hf Hafnium 4	73 Ta Tantalum 5	74 W Tungsten 4.6	75 Re Rhenium 4	76 Os Osmium 4	77 Ir Iridium 3.4	78 Pt Platinum 2.4	79 Au Gold 3	80 Hg Mercury 1.2	81 Tl Thallium 1.3	82 Pb Lead 2.4	83 Bi Bismuth 3	84 Po Polonium -2.24	85 At Astatine -1.1	86 Rn Radon							
7	87 Fr Francium 1	88 Ra Radium 2	89-103	104 Rf Rutherfordium 4	105 Db Dubnium 5	106 Sg Seaborgium 6	107 Bh Bohrium 7	108 Hs Hassium 8	109 Mt Meitnerium	110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson							
Oxidation states are the number of electrons added to or removed from an element when it forms a chemical compound.																									
		6	57 La Lanthanum 3	58 Ce Cerium 3.4	59 Pr Praseodymium 3	60 Nd Neodymium 3	61 Pm Promethium 3	62 Sm Samarium 3	63 Eu Europium 2.3	64 Gd Gadolinium 3	65 Tb Terbium 3	66 Dy Dysprosium 3	67 Ho Holmium 3	68 Er Erbium 3	69 Tm Thulium 3	70 Yb Ytterbium 3	71 Lu Lutetium 3								
		7	89 Ac Actinium 3	90 Th Thorium 4	91 Pa Protactinium 5	92 U Uranium 6	93 Np Neptunium 6	94 Pu Plutonium 4	95 Am Americium 3	96 Cm Curium 3	97 Bk Berkelium 3	98 Cf Californium 3	99 Es Einsteinium 3	100 Fm Fermium 3	101 Md Mendelevium 3	102 No Nobelium 2	103 Lr Lawrencium 3								

# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)

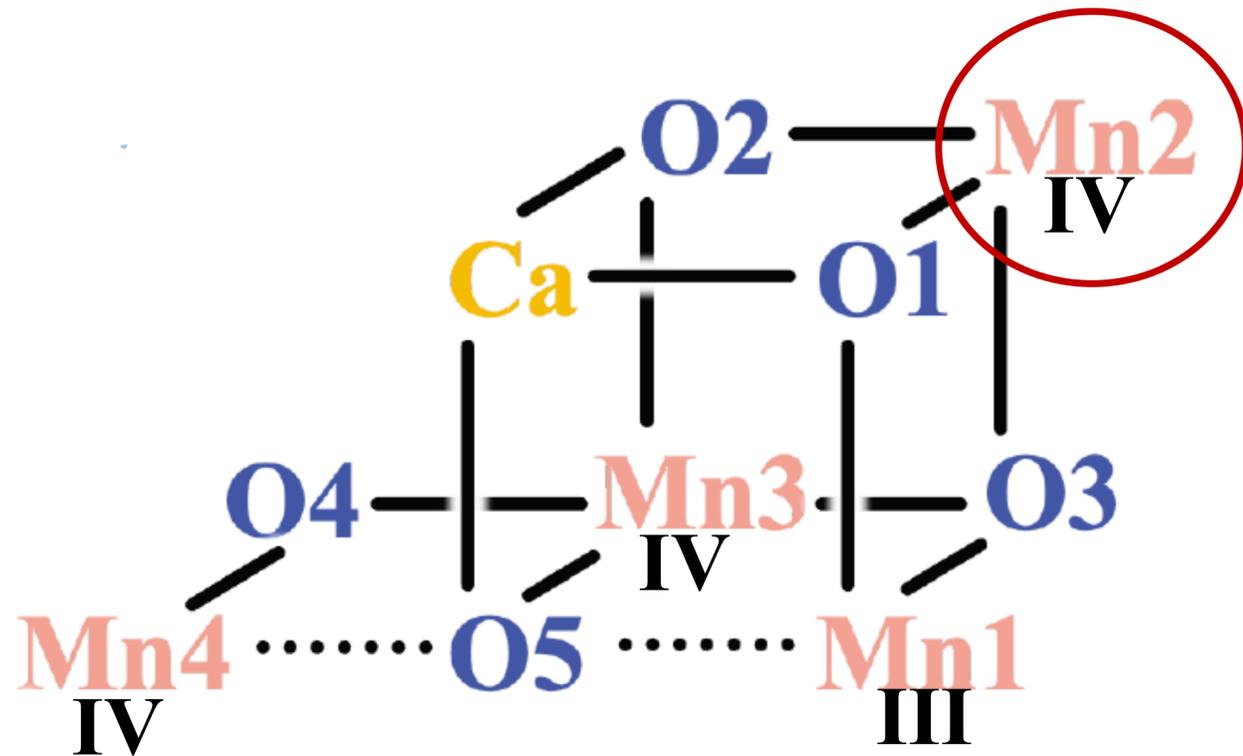


	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18		
1	1 H Hydrogen -1.1	2 He Helium																		Atomic Name Symbol Weight
2	3 Li Lithium 1	4 Be Beryllium 2																		
3	11 Na Sodium 1	12 Mg Magnesium 2												5 B Boron 3	6 C Carbon -4.4	7 N Nitrogen -3.35	8 O Oxygen -2	9 F Fluorine -1	10 Ne Neon	
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# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



**Mn**

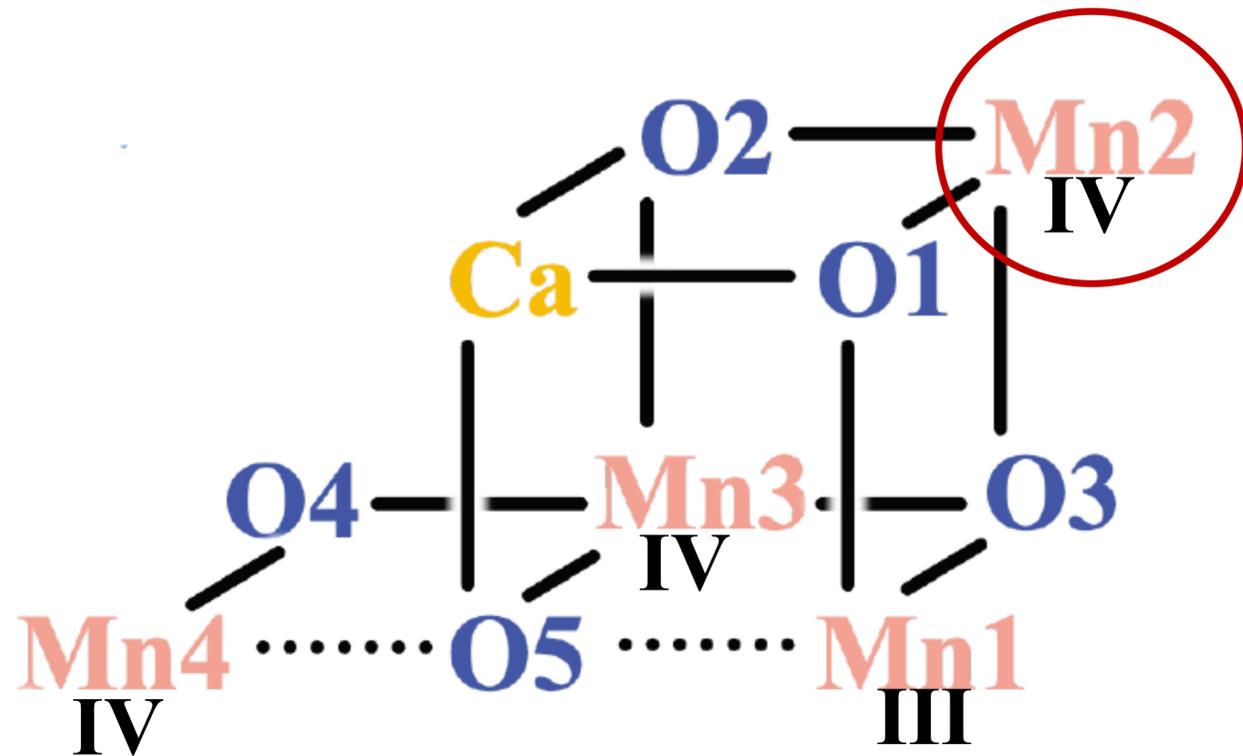
Orbital diagram for Manganese (Mn) showing the following configuration: 1s<sup>2</sup> 2s<sup>2</sup> 2p<sup>6</sup> 3s<sup>2</sup> 3p<sup>6</sup> 3d<sup>5</sup> 4s<sup>1</sup>. The 3d orbitals are highlighted with a red box and labeled "7 valence electrons".

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	Atomic Name	Symbol	Weight	
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# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



**Mn**

Orbital diagram for Manganese (Mn) showing the following configuration: 1s<sup>2</sup> 2s<sup>2</sup> 2p<sup>6</sup> 3s<sup>2</sup> 3p<sup>6</sup> 3d<sup>5</sup> 4s<sup>1</sup>. The 4s orbital is crossed out with a red 'X', and the 3d orbitals are highlighted with a red box. A label indicates "7 valence electrons".

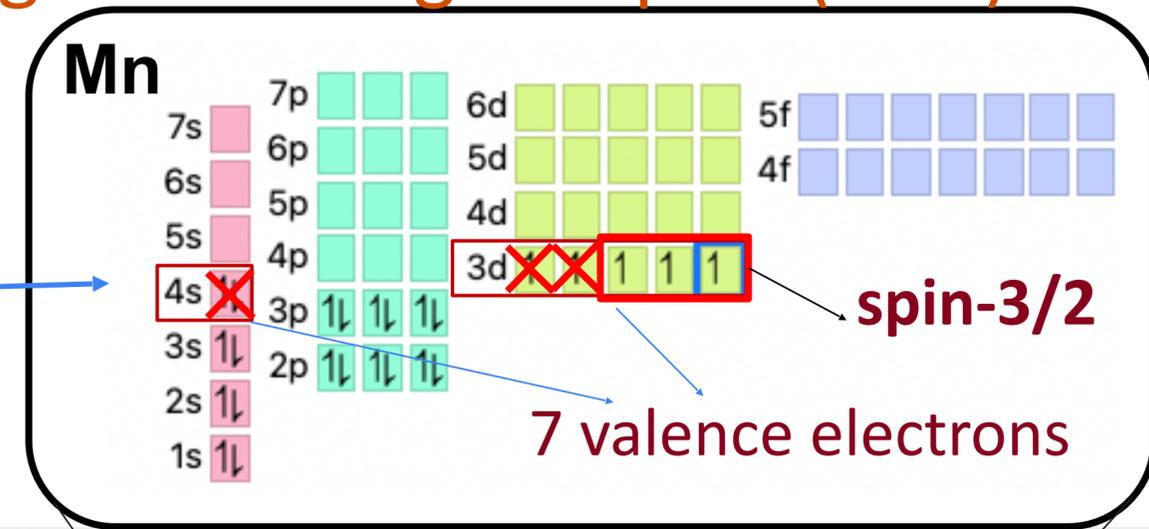
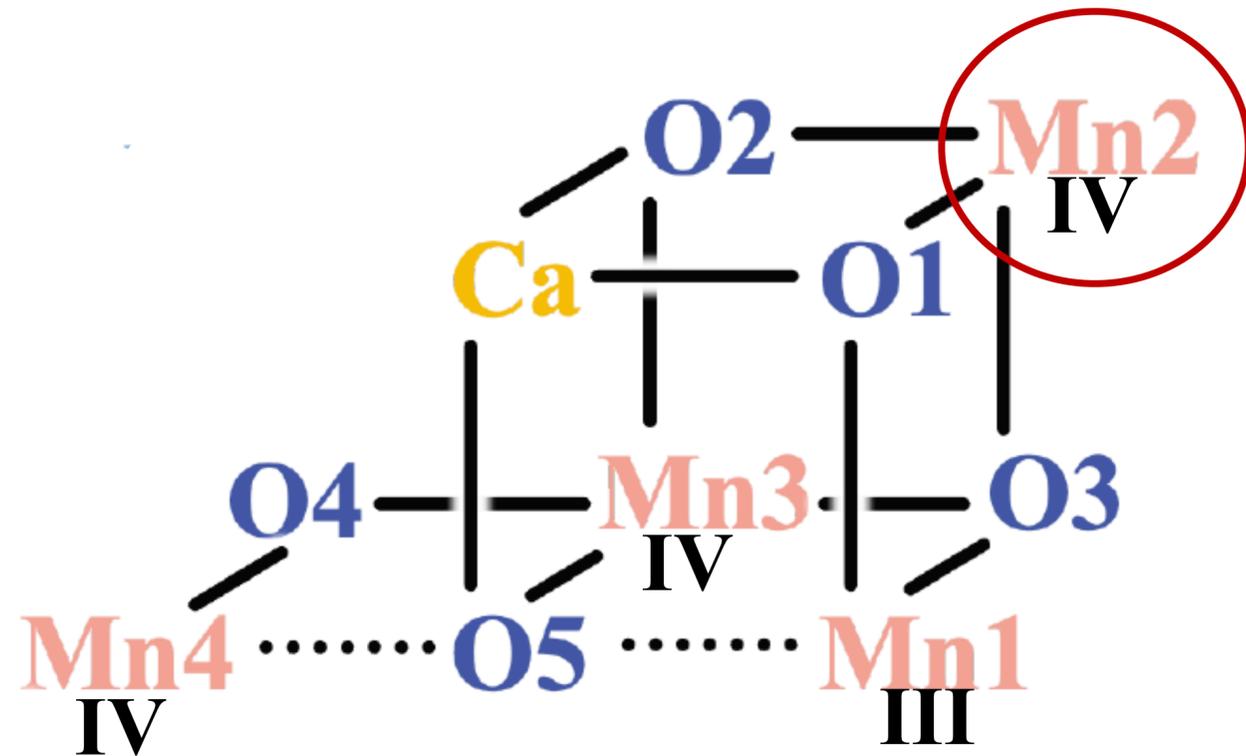
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												Pnictogens		Chalcogens		Halogens			
1	2	1 H Hydrogen -1.1	2 He Helium															Atomic Name	
2	3	3 Li Lithium 1	4 Be Beryllium 2															Symbol	
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Oxidation states are the number of electrons added to or removed from an element when it forms a chemical compound.



# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



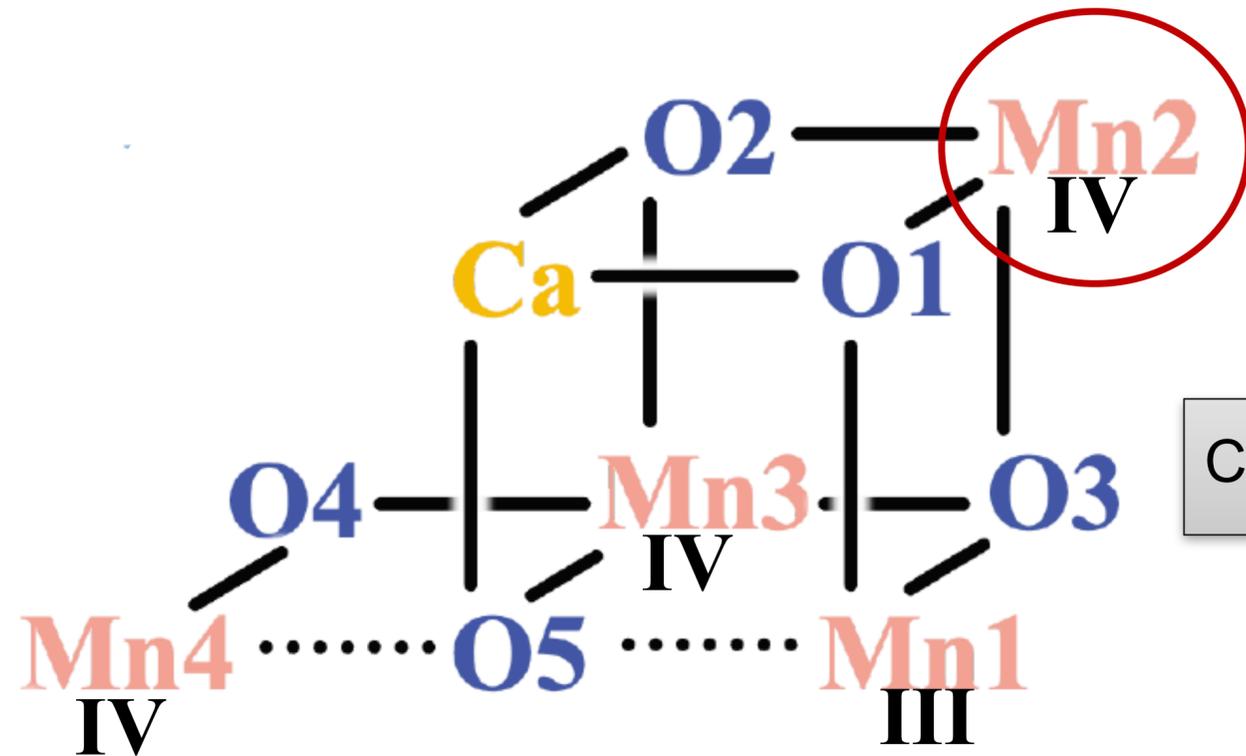
Periodic table showing the position of Manganese (Mn, atomic number 25) in the d-block. An inset orbital diagram shows the 3d orbitals with 5 unpaired electrons. The table includes element symbols, names, atomic numbers, and oxidation states.

→ Mn<sup>2+</sup> has 3 active electrons

- Coulomb interaction - localizes electrons
- Exchange interaction - ferromagnetic
- Super-exchange interaction - anti-ferromagnetic

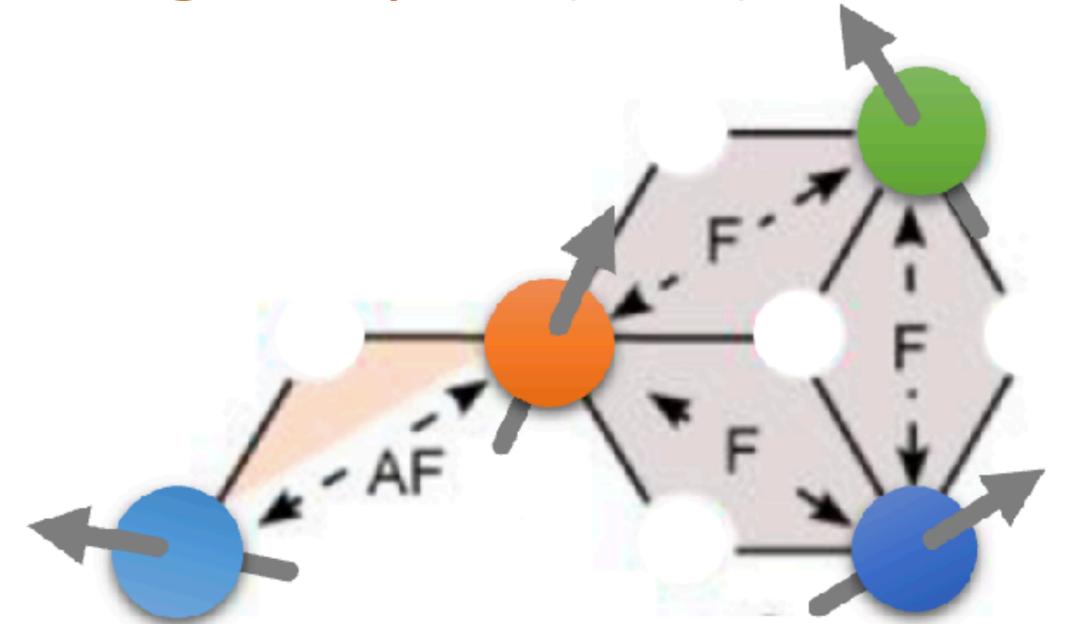
# Model Hamiltonians

Example: Biochemical catalyst involved in the oxygen evolving complex (OEC)



→ Mn2 has 3 **active** electrons

Computational Chemistry



Hilbert space scaling:

$$\propto (2S + 1)^N$$

V. Krewald, M. Retegan, F. Neese, W. Lubitz,  
D. A. Pantazis, N. Cox, Inorg. Chem. 55, 488–501 (2016)

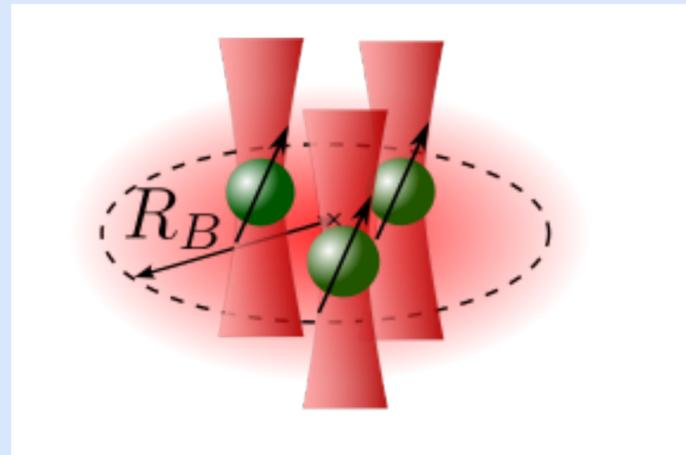
# Approach

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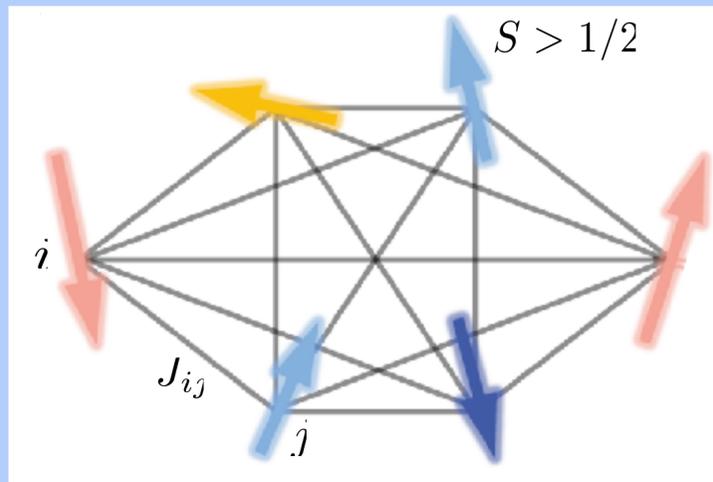
- Represent high spins ...
  - ➔ How to implement high spins?
- ... and let them interact
  - ➔ How to implement non-local connectivity?
- Read out chemically relevant quantities
  - ➔ Quantum-classical co-processing

# Necessary Ingredients

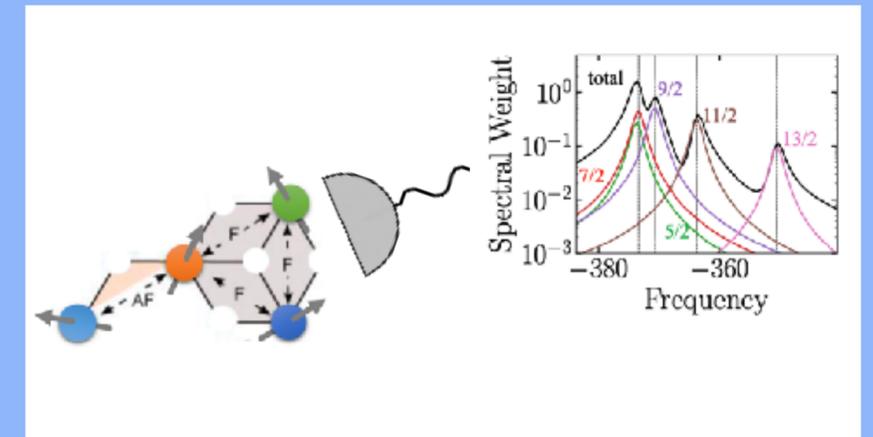
Control high spin ( $S > 1/2$ )



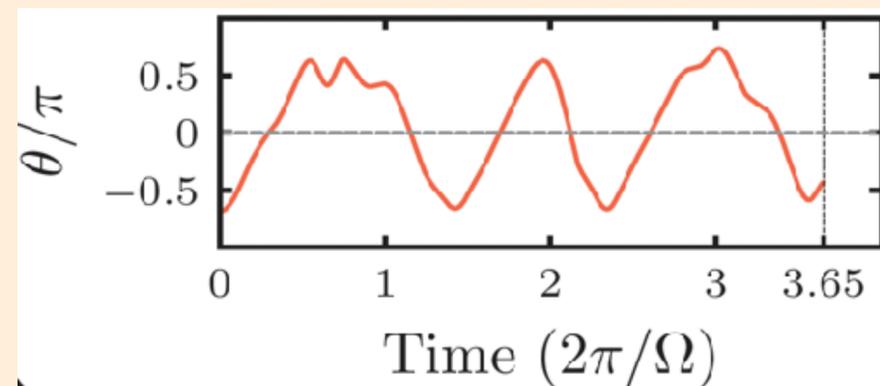
Non-local connectivity



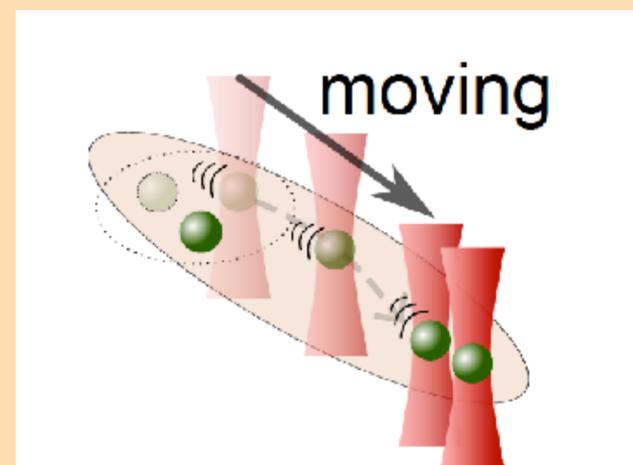
Solution read-out



Native multi-qubit gates

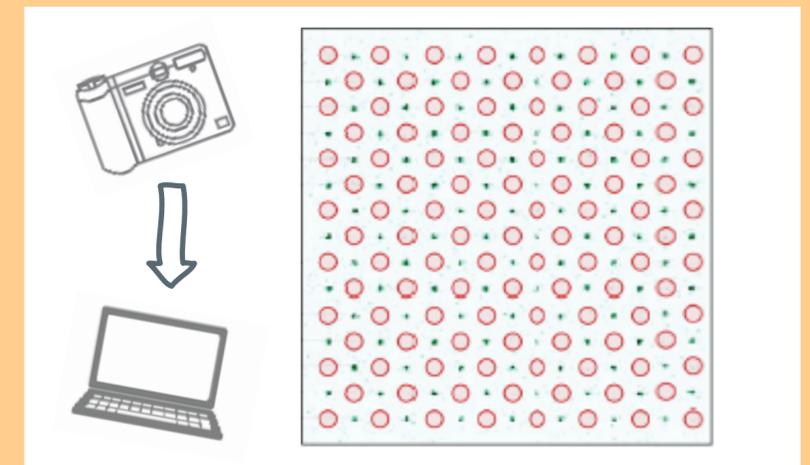


Atom moving



D. Bluvstein et al., Nature **604**, 451–456 (2022)

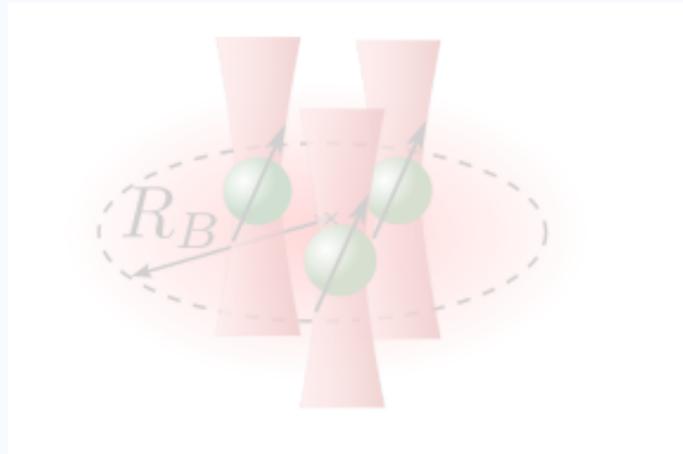
Co-processing



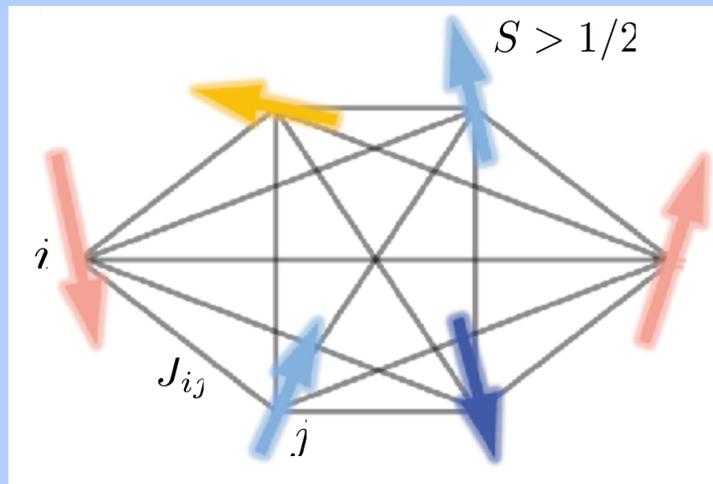
H.-Y. Huang et al., Nat. Phys. **16**, 1050–1057 (2020)

# Necessary Ingredients

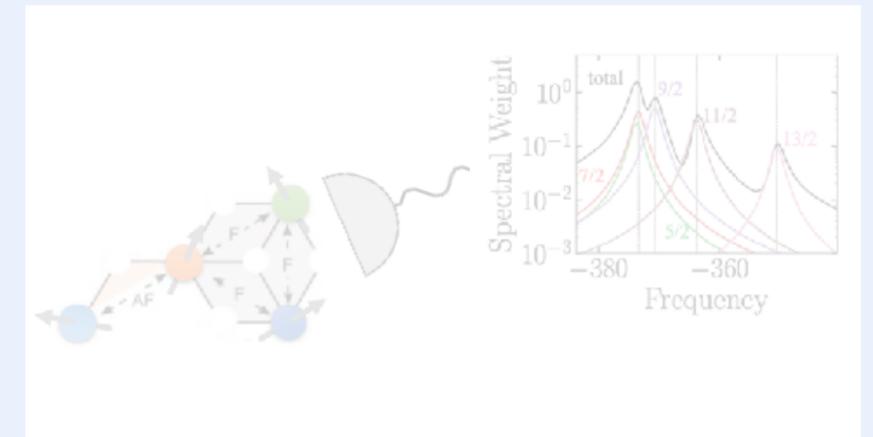
Control high spin ( $S > 1/2$ )



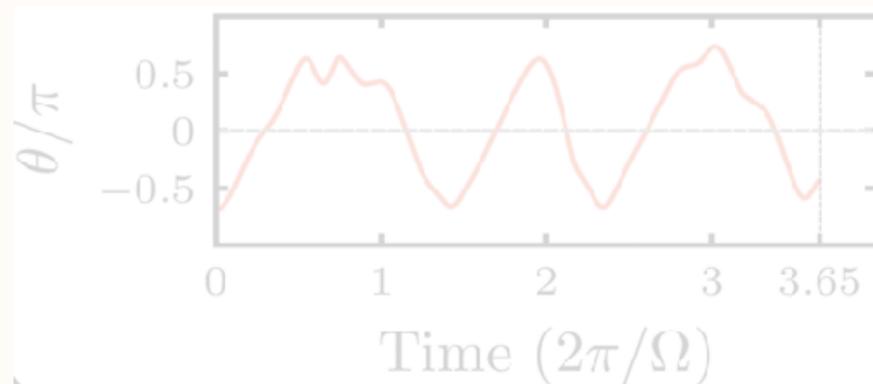
Non-local connectivity



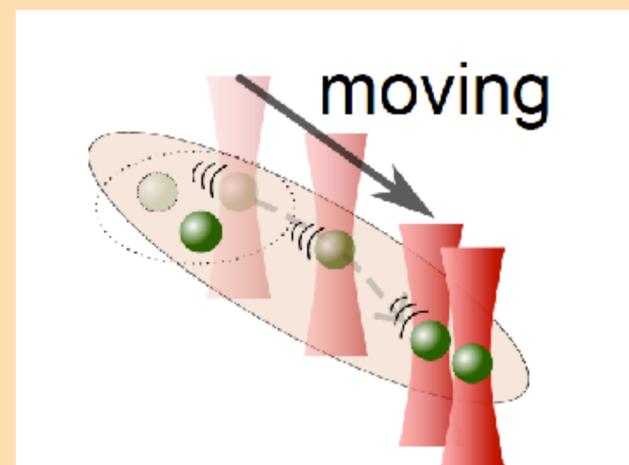
Solution read-out



Native multi-qubit gates

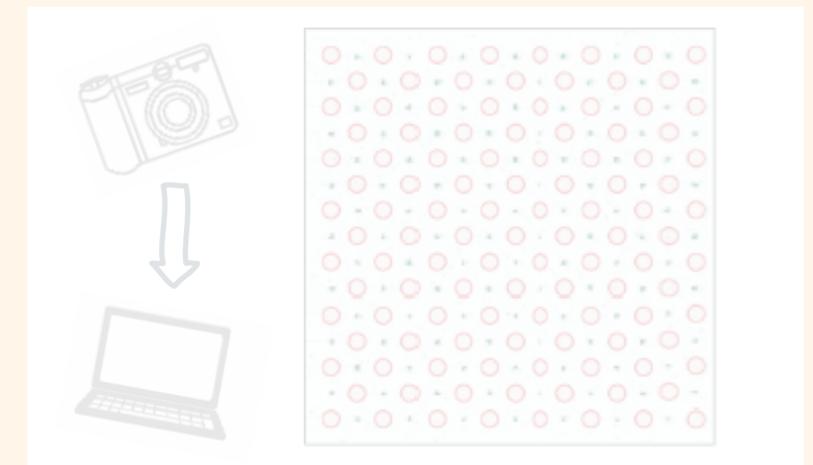


Atom moving



D. Bluvstein et al., Nature **604**, 451–456 (2022)

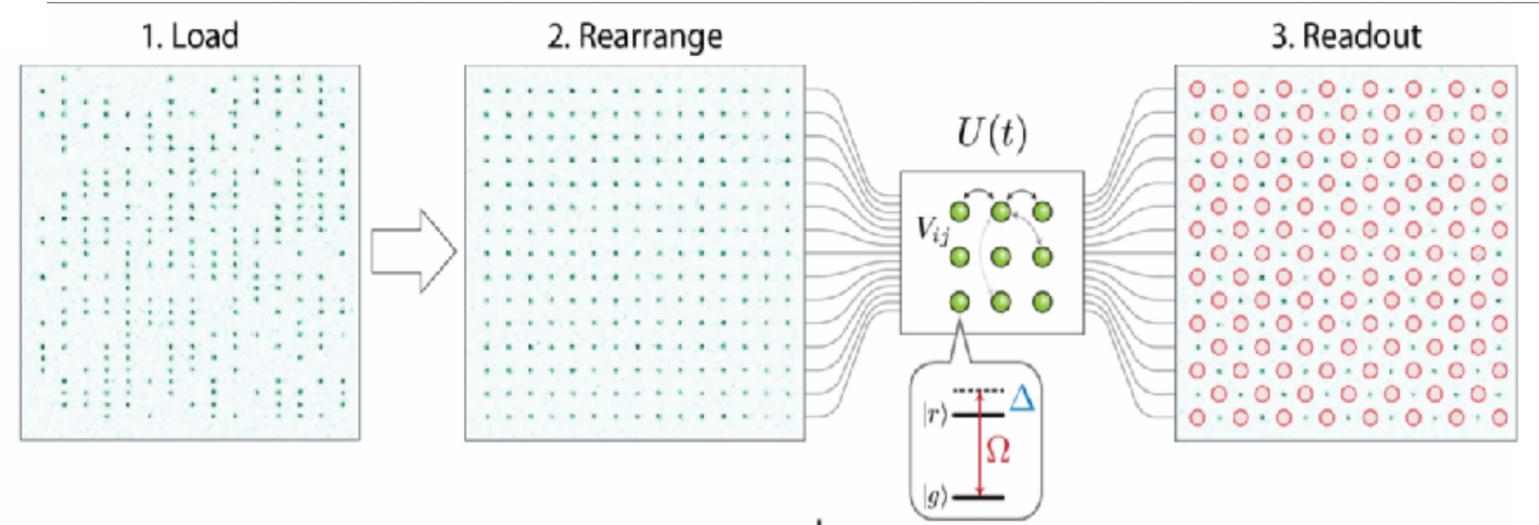
Co-processing



H.-Y. Huang et al., Nat. Phys. **16**, 1050–1057 (2020)

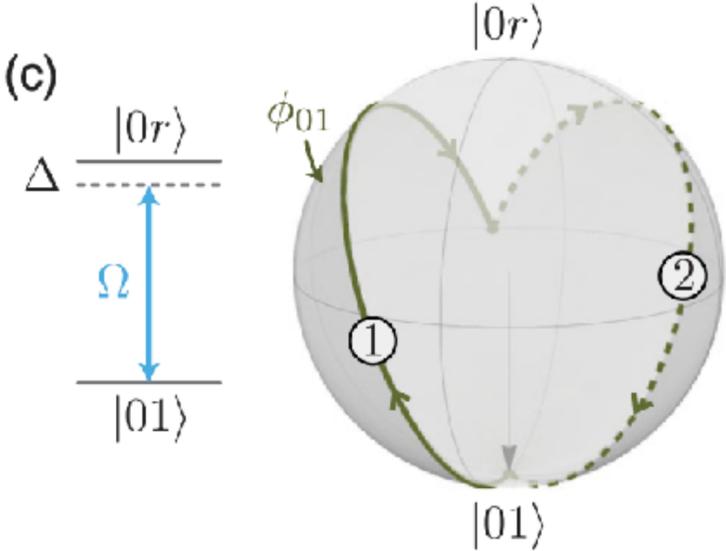
# Example implementation: Reconfigurable atom arrays

## Analog quantum simulation



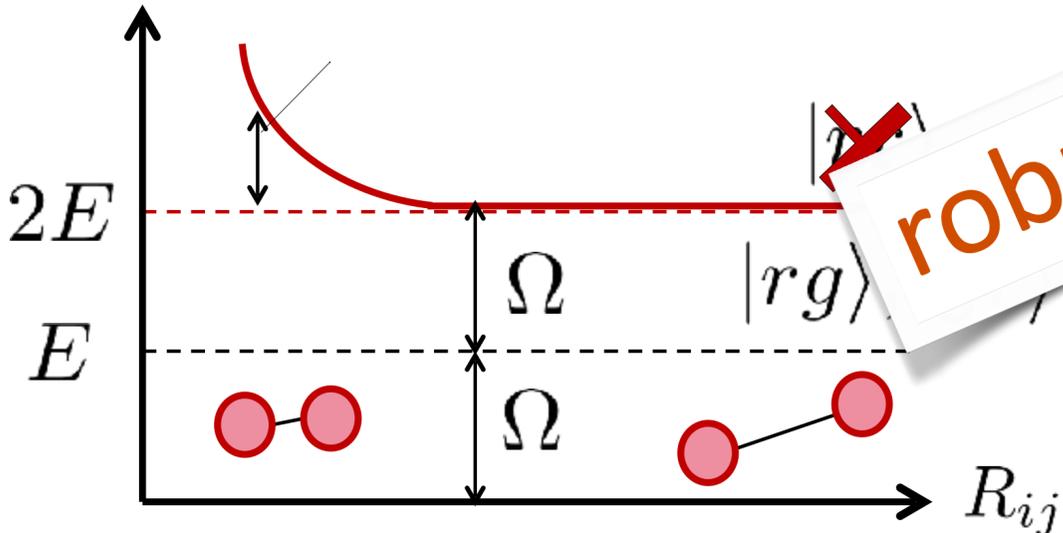
Ebadi, S., et al., Nature (2020)

## Digital Quantum Computing (gates)



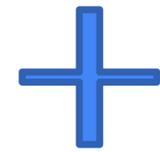
H. Levine et al., Phys, Rev. Lett. **123**, 170503 (2019)

## Rydberg blockade

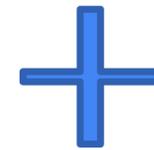
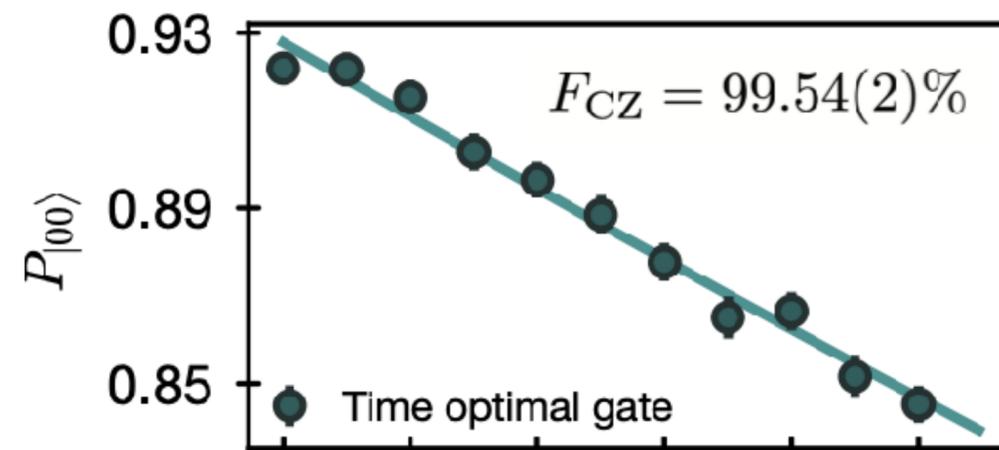


# Atom Array Platform in Analog-Digital Mode

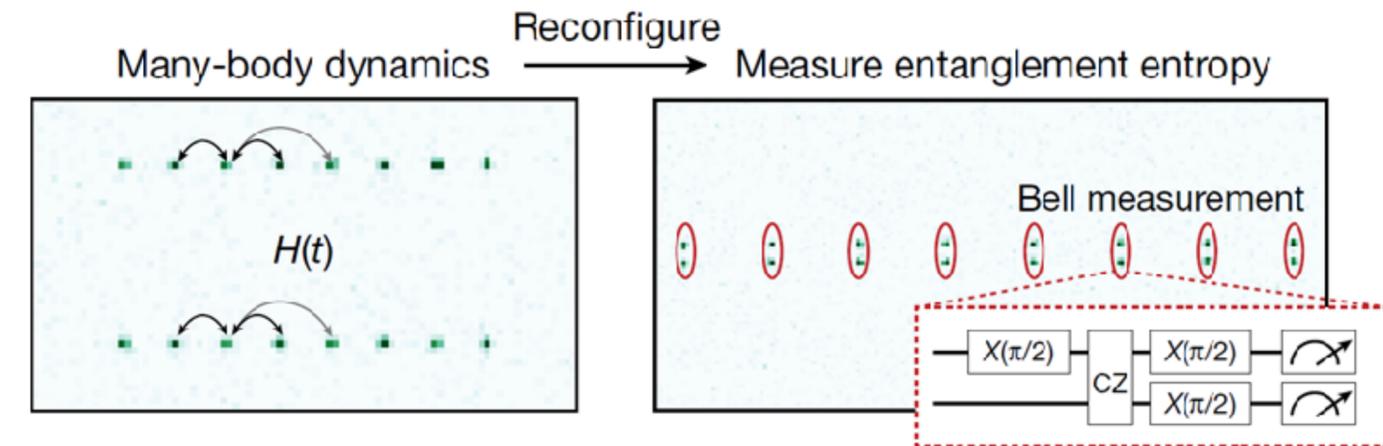
Reconfigurable architecture



High-fidelity gates



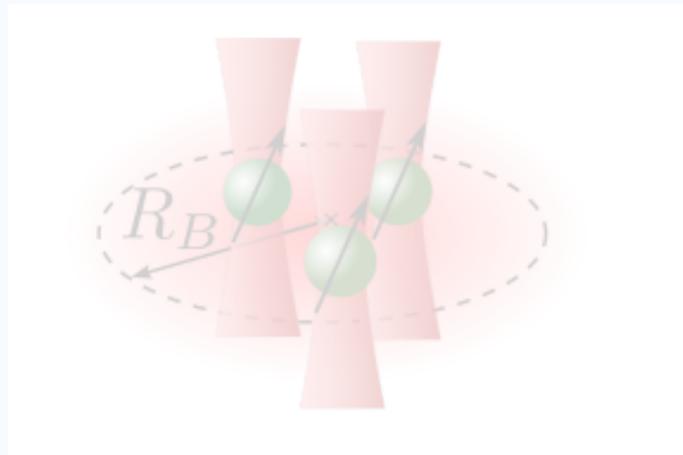
Hybrid analog-digital control



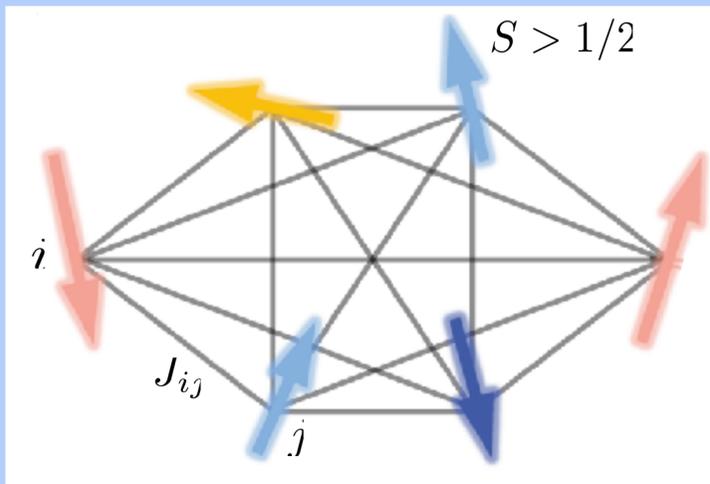
High degree of programmability  
Interactions manipulated via  
geometric configuration  
+  
Global control pulses

# Necessary Ingredients

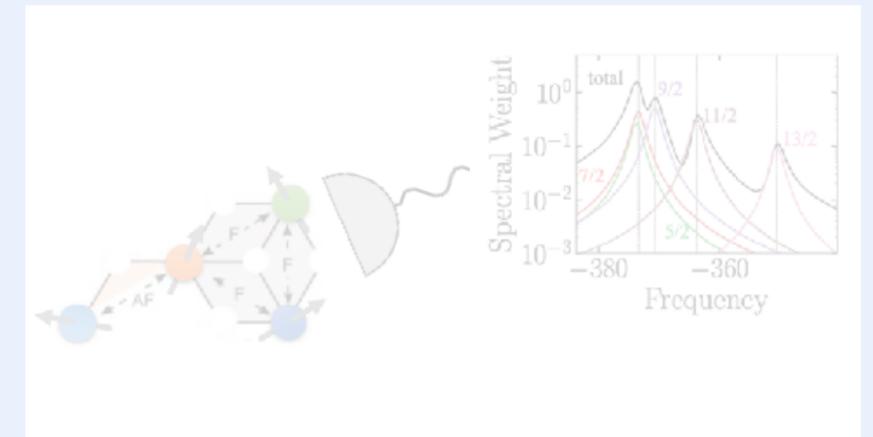
Control high spin ( $S > 1/2$ )



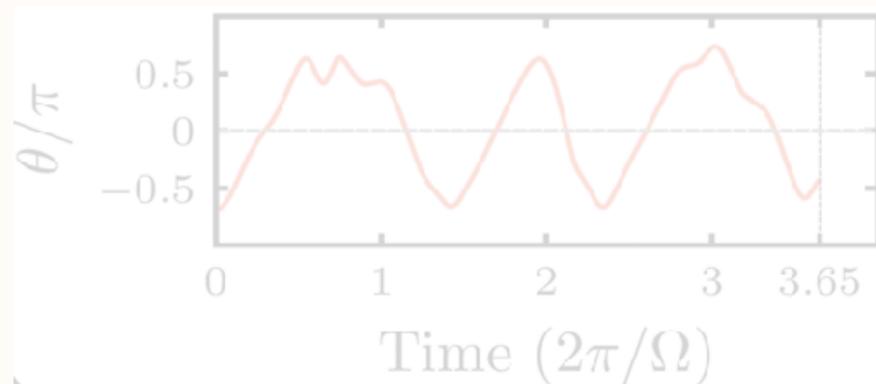
Non-local connectivity



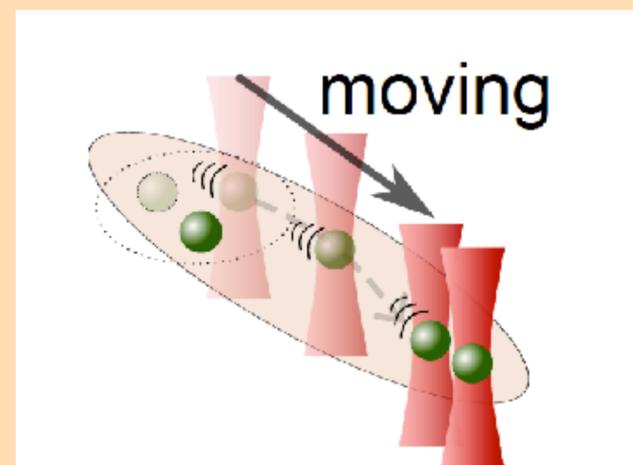
Solution read-out



Native multi-qubit gates

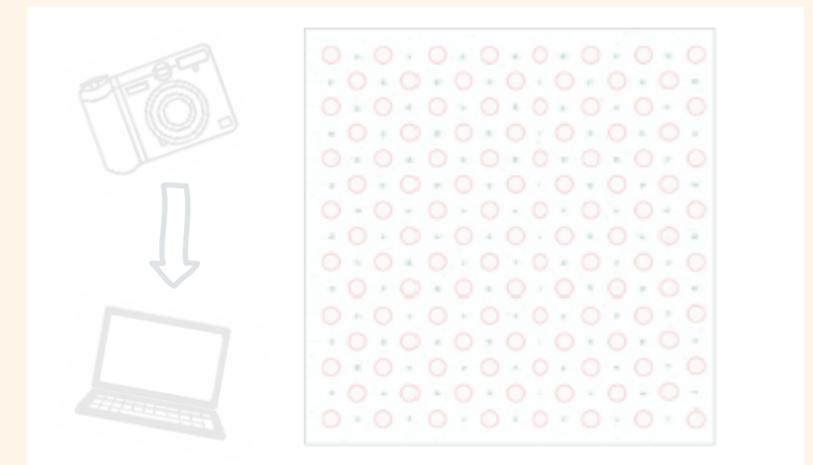


Atom moving



D. Bluvstein et al., Nature **604**, 451–456 (2022)

Co-processing



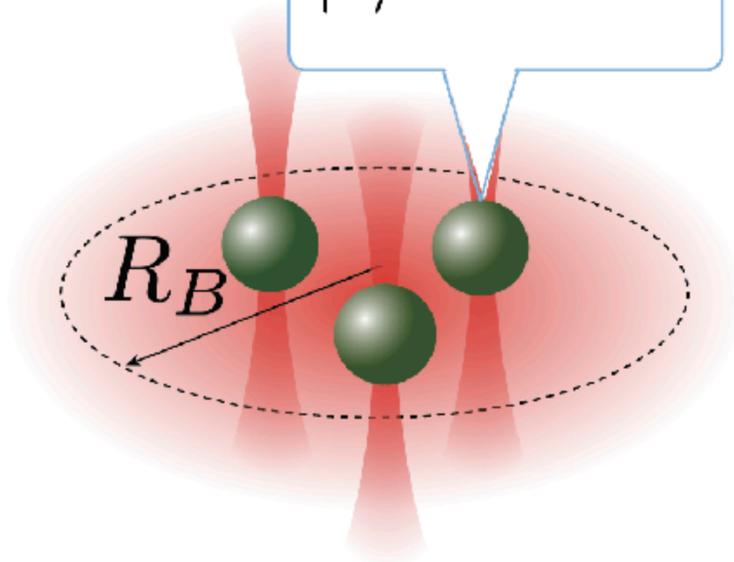
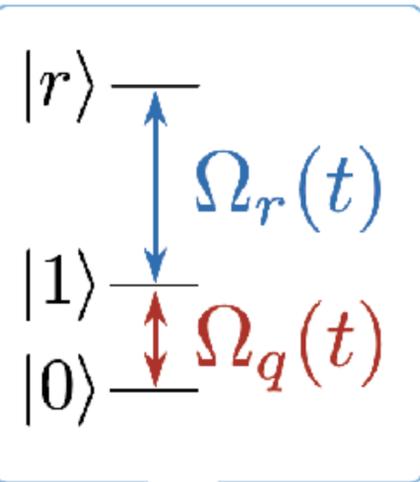
H.-Y. Huang et al., Nat. Phys. **16**, 1050–1057 (2020)

# Engineering Large Spins on Rydberg Platform

Hardware Efficient Multi-Qubit Operations with a global drive

Encode spin- $S$  variables into  $2S$  (spin- $1/2$ ) qubits (“clusters”):

⇒ valid spin- $S$  states:  $\langle \hat{S}_i^2 \rangle = S_i(S_i + 1)$



Engineer two-field pulses (using Rydberg blockade) to implement any  $2S$ -qubit gate!

# Engineering Large Spins on R

Hardware Efficient Multi-Qubit Operations with a glc

Encode spin-S variables into 2S

⇒ valid spin-S states:  $\langle \hat{S}_i^2 \rangle = S(S+1)$

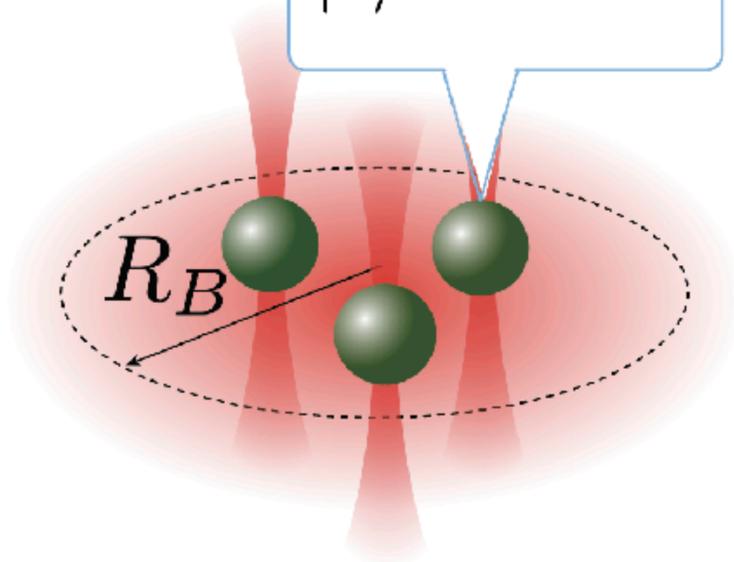
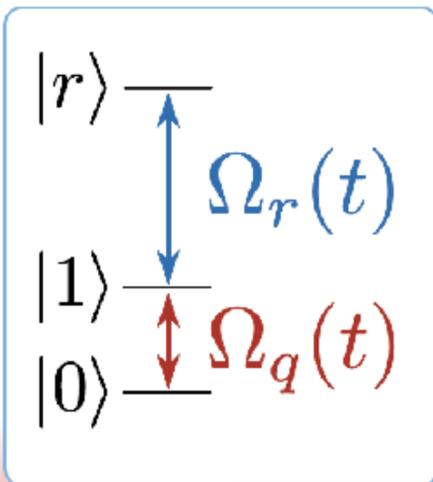
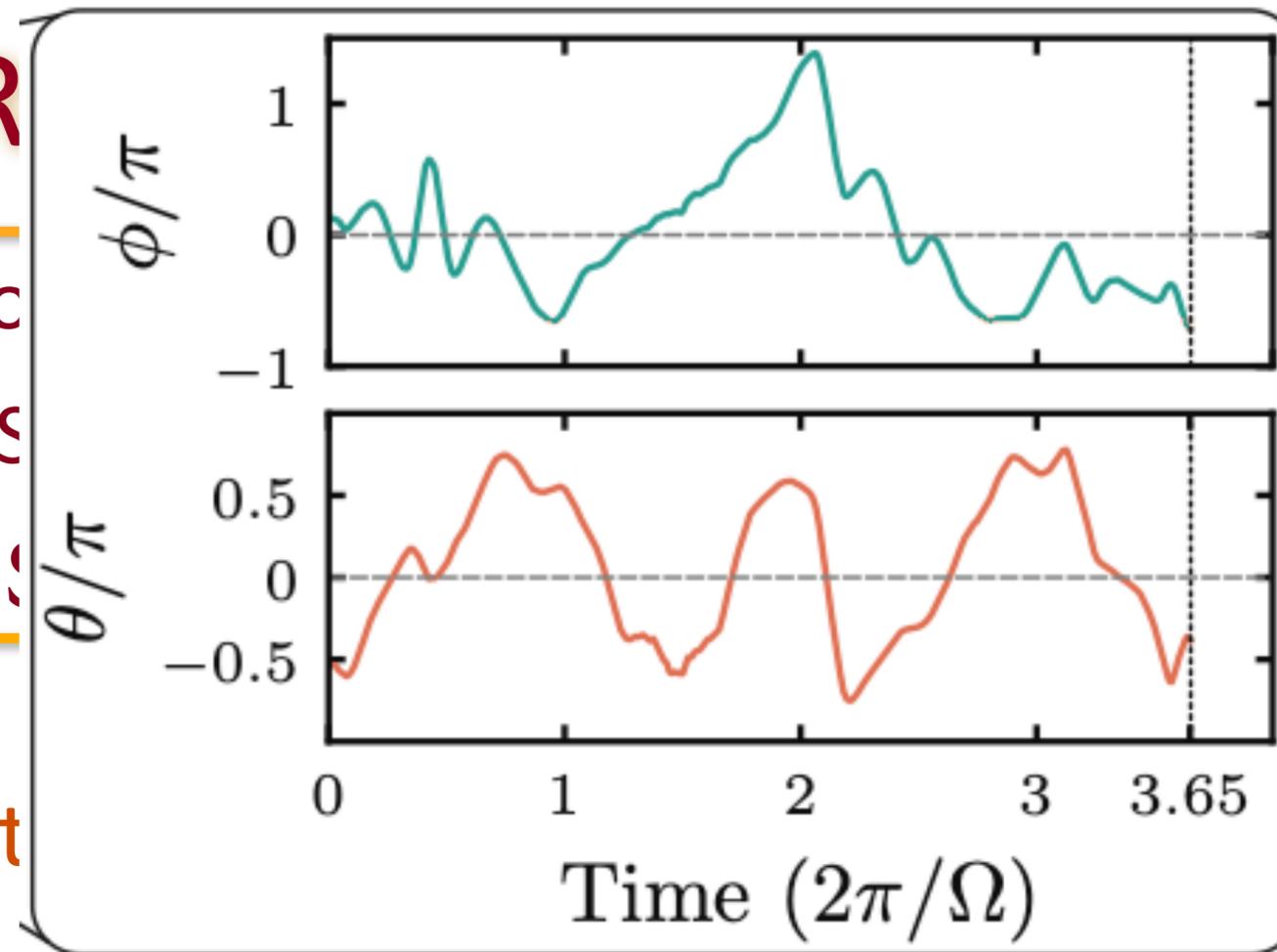
Two important examples:

1. Engineer all-to-all interact

$$\text{e.g., } U_S(\theta) = e^{-i\theta \frac{\hat{S}^2}{2S}}$$

2. Projection into symmetric subspace:

$$U_P(\theta) = e^{-i\theta P[\hat{S}^2]}$$



Realize multi-qubit gates via time-dependent  
use GRAPE (Gradient Ascent Pulse Eng

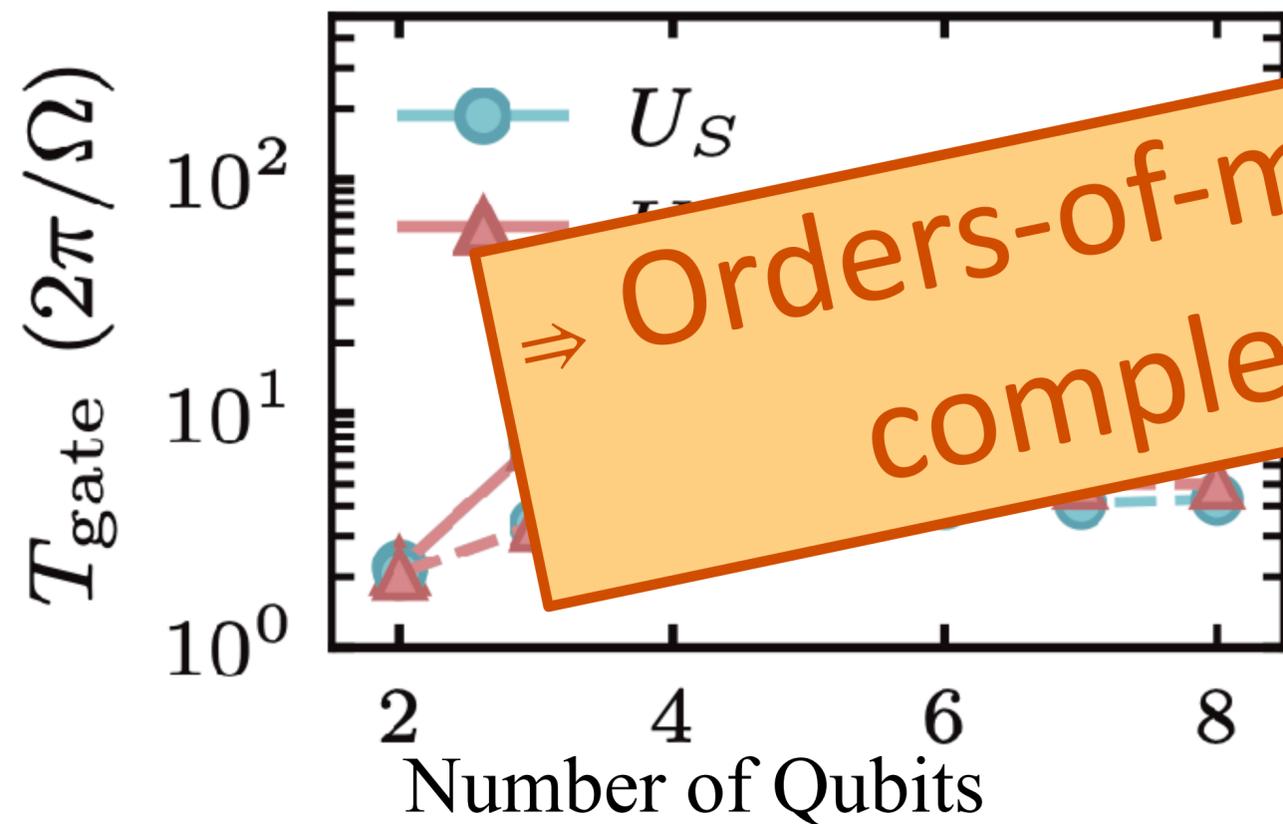
Khaneja, et al., J. Magn. Reson. **172**, 296 (2005)  
 Jandura et al., Quantum **6**, 712 (2022)  
 Evered et al., Nature **622**, 268 (2023)  
 Katz, et al., Nat. Phys. **19**, 1452 (2023)

# Important Metric: Gate Times

The shorter the gates, the more sequences one can run (until system decoheres)

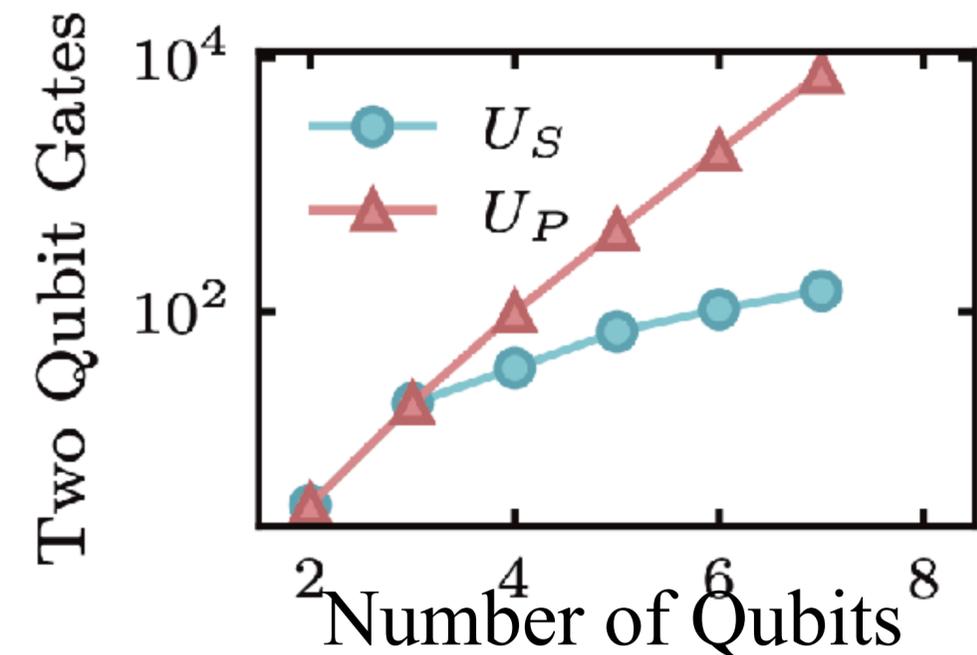
Multi-Qubit Gates via Global Drive  
(for error  $\epsilon = 10^{-3}$ )

Comparison: two-qubit operations  
(for error  $\epsilon = 10^{-3}$ )



⇒ Orders-of-magnitude speedup for complex spin-operations

⇒ Almost no scaling with cluster size



# Engineering Interactions



Target Hamiltonian:

$$H_{\text{target}} = \sum_{i,j} J_{ij}^{\alpha\beta} \hat{S}_i^\alpha \hat{S}_j^\beta$$

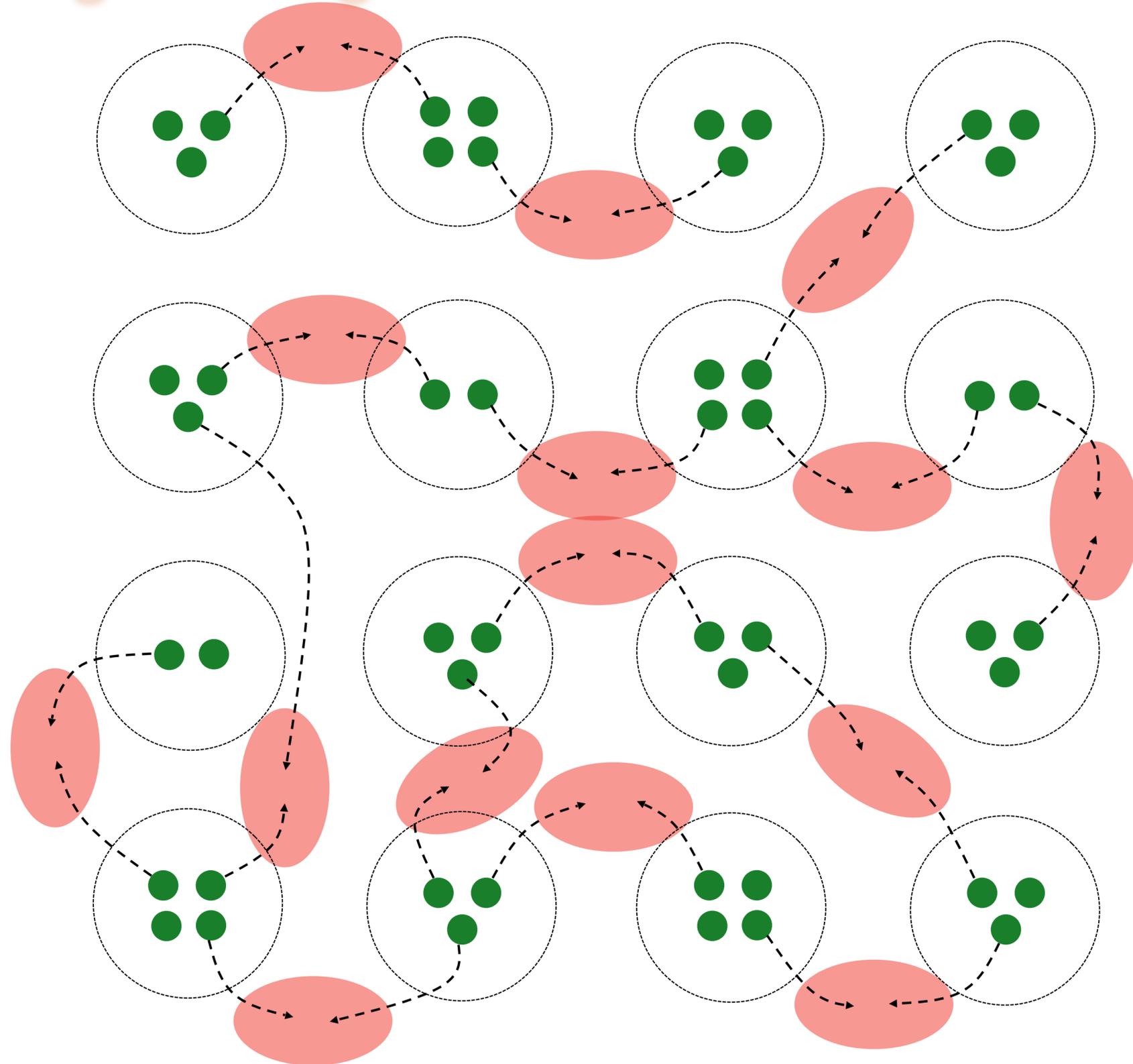
Highly programmable!

Example:

$$J_{ij}^{\alpha\beta} = \underbrace{J_{ij} \delta_{\alpha\beta}}_{\text{Heisenberg}} + \underbrace{D_{ij}^\gamma \epsilon_{\alpha\beta}^\gamma}_{\text{DM}}$$

(e.g., Malrieu et al, Chem. Rev **114**, 429 (2014))

# Engineering Interactions



Target Hamiltonian:

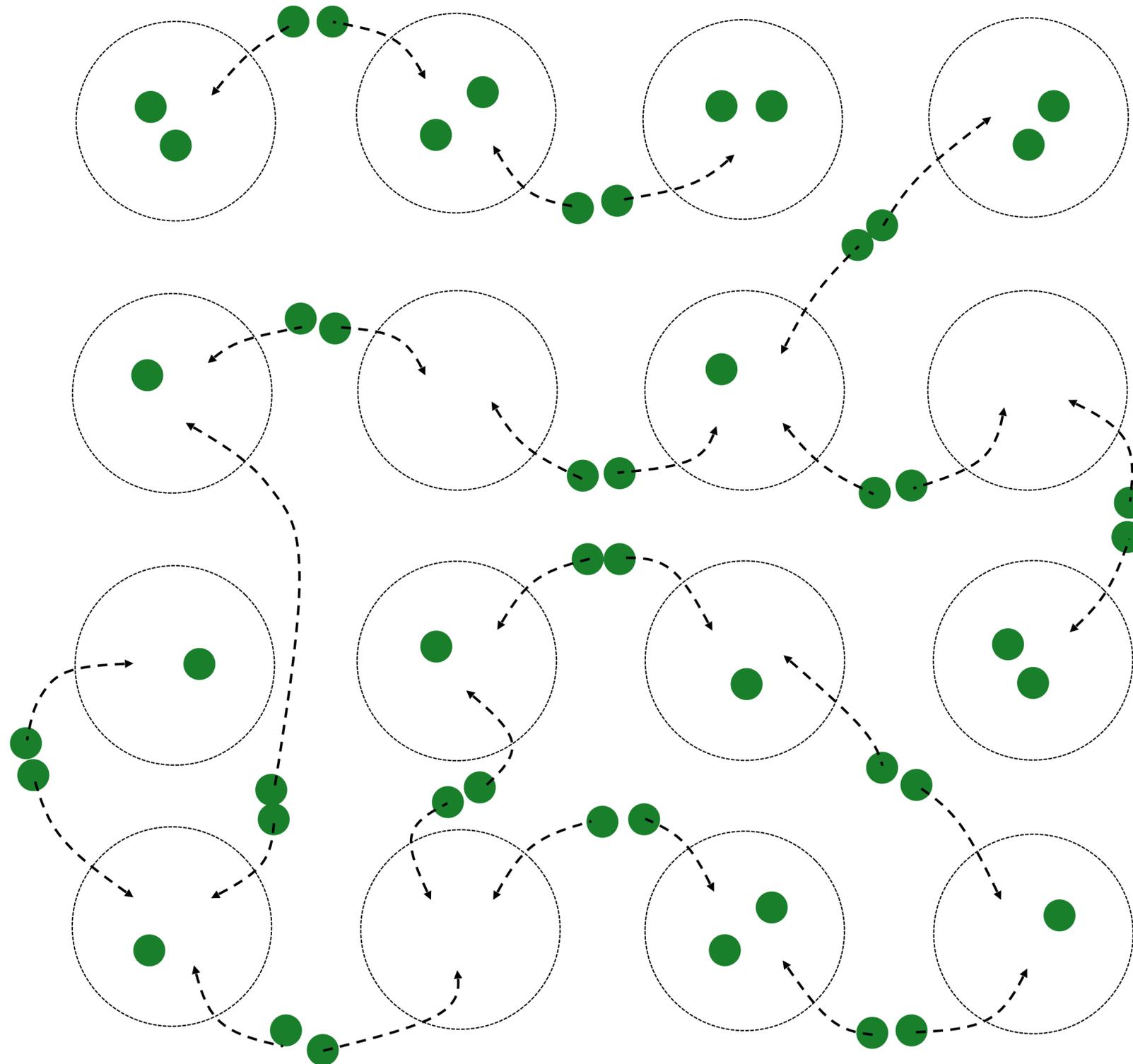
$$H_{\text{target}} = \sum_{ij} J_{ij}^{\alpha\beta} \hat{S}_i^\alpha \hat{S}_j^\beta$$

1. Reconfigure

2. Inter-cluster gate:  $H_I = \sum_{ij} J_{ij}^{\alpha\beta} \hat{S}_{i,a_{ij}}^\alpha \hat{S}_{j,b_{ij}}^\beta$

- Spin-1/2 gates and local rotations
- Mediates generic, long-range connectivity
- Violates large-spin encoding

# Engineering Interactions



Target Hamiltonian:

$$H_{\text{target}} = \sum_{i,j} J_{ij}^{\alpha\beta} \hat{S}_i^\alpha \hat{S}_j^\beta$$

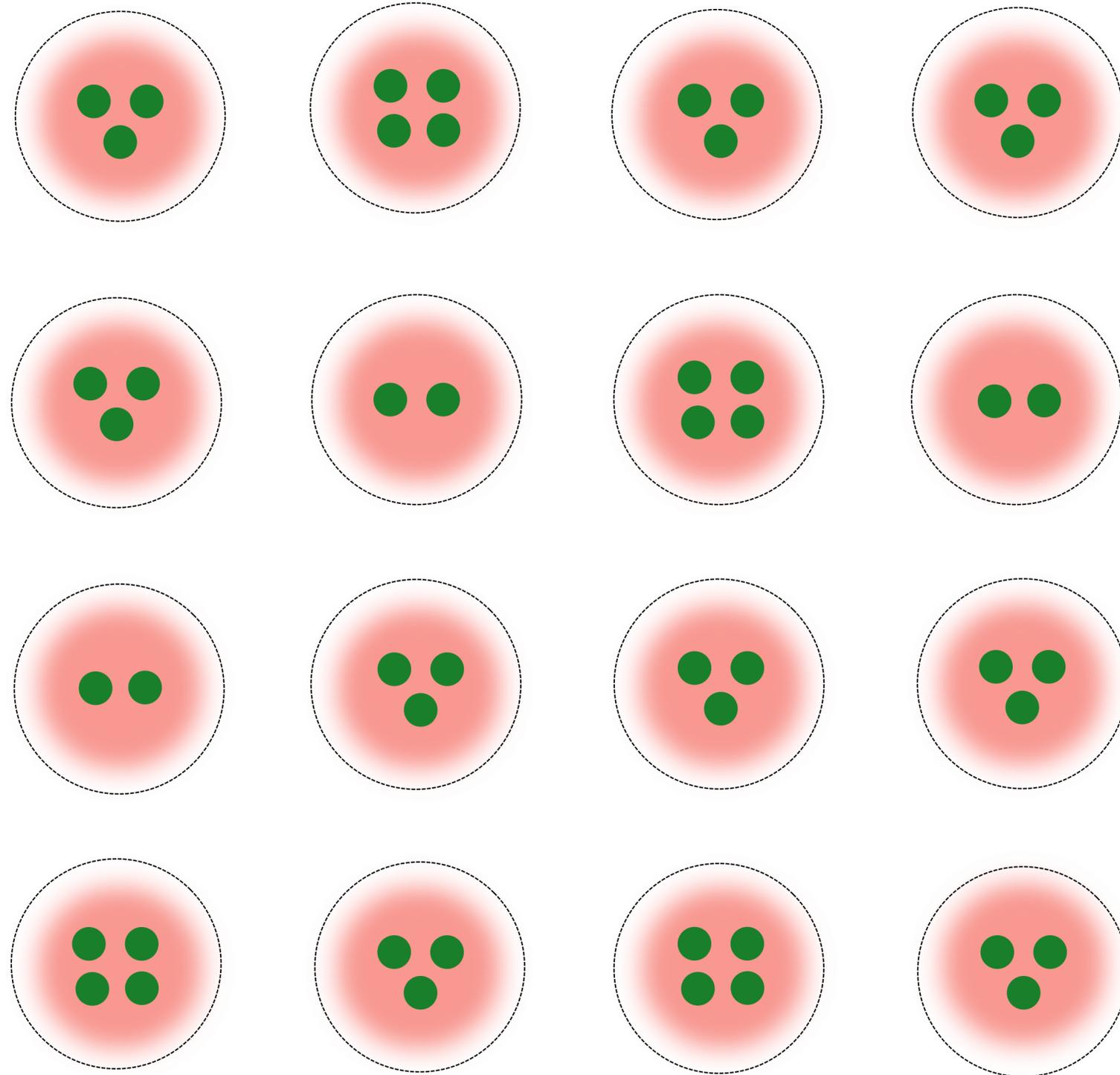
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- Spin-1/2 gates and local rotations
- Mediates generic, long-range connectivity
- Violates large-spin encoding

3. Reconfigure

# Engineering Interactions



Target Hamiltonian:

$$H_{\text{target}} = \sum_{i,j} J_{ij}^{\alpha\beta} \hat{S}_i^\alpha \hat{S}_j^\beta$$

1. Reconfigure

2. Inter-cluster gate:  $H_I = \sum_{ij} J_{ij}^{\alpha\beta} \hat{S}_{i,a_{ij}}^\alpha \hat{S}_{j,b_{ij}}^\beta$

- Spin-1/2 gates and local rotations
- Mediates generic, long-range connectivity
- Violates large-spin encoding

3. Reconfigure

4. Intra-cluster gate:  $H_C = - \sum_i P_{\text{sym}}[(\vec{S}_i)^2]$

- Encoding space is gapped ground state
- Applies phase to encoding violating terms

# Floquet Sequence to Implement Model Hamiltonian

---

Effective evolution operator:

$$U_F = \prod_k e^{-i\theta_k H_C} e^{-i\tau H_I}$$

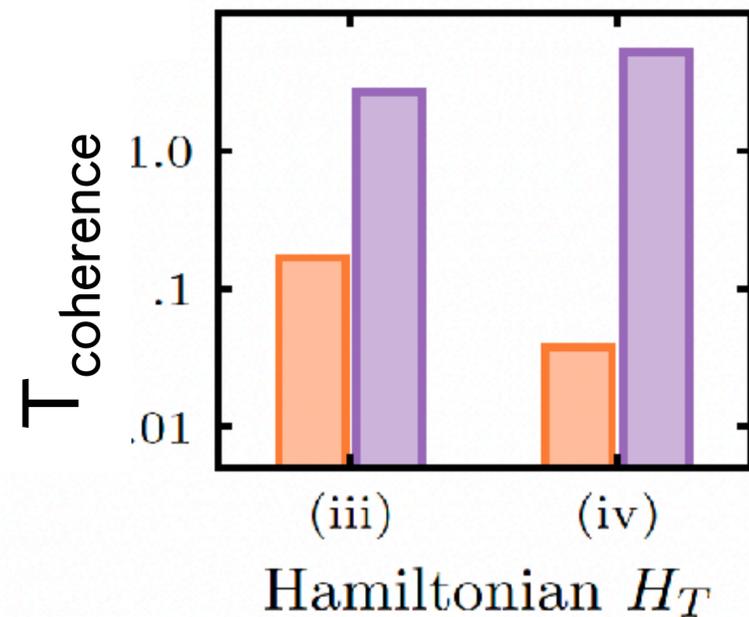
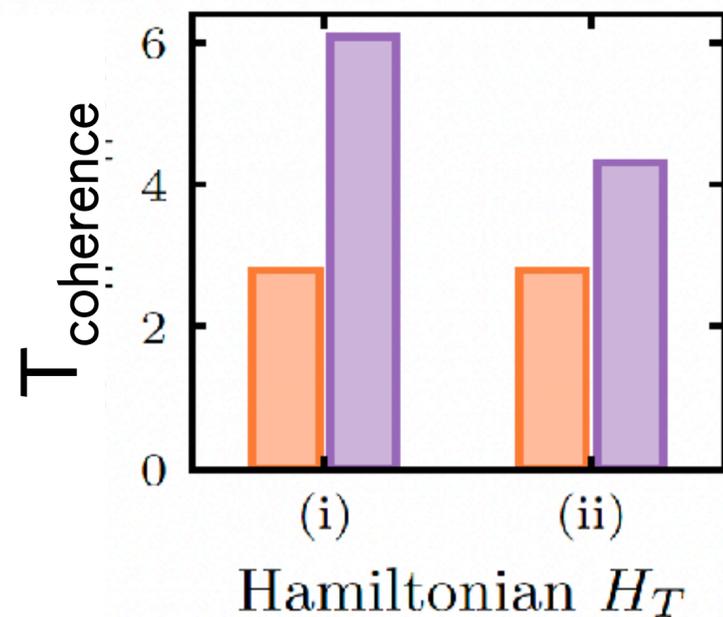
can be large-angle rotations

⇒ Realizes target Hamiltonian on average

Higher-order errors can be cancelled out, or controlled via Floquet engineering.

# Performance of the Approach

Two-qubit Trotter      Multi-qubit Enhanced

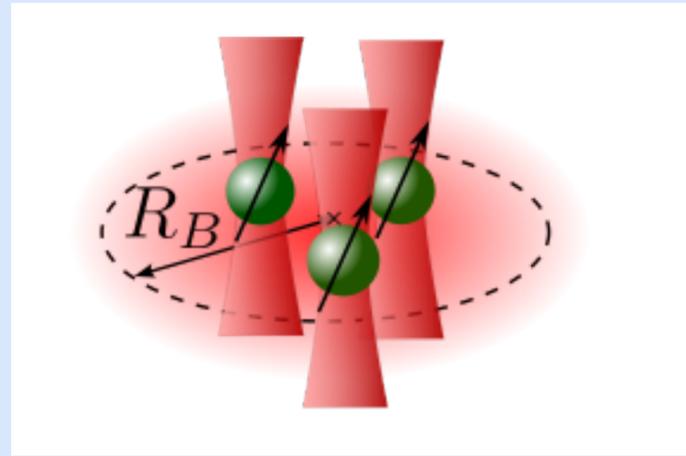


Takeaway:

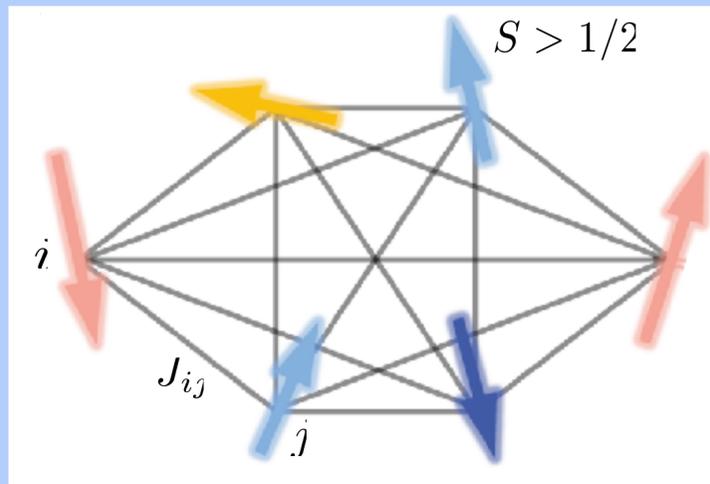
- Hardware-optimized multi-qubit operations to efficiently generate interactions
- Floquet/Hamiltonian engineering to efficiently combine operations.

# Necessary Ingredients

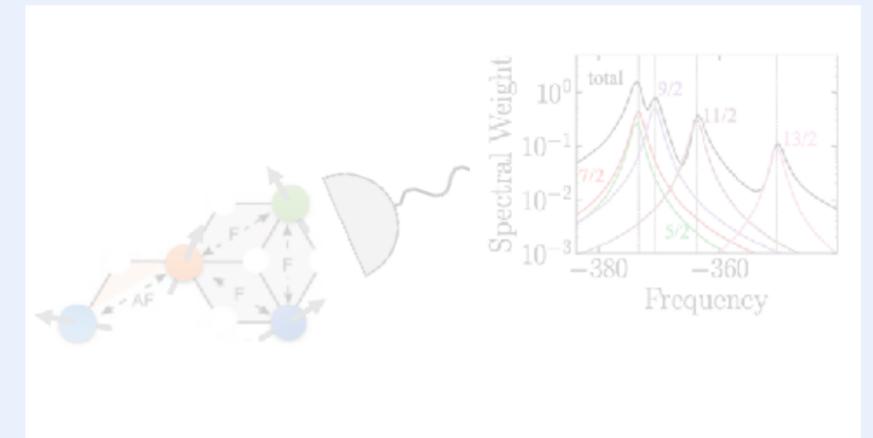
Control high spin ( $S > 1/2$ )



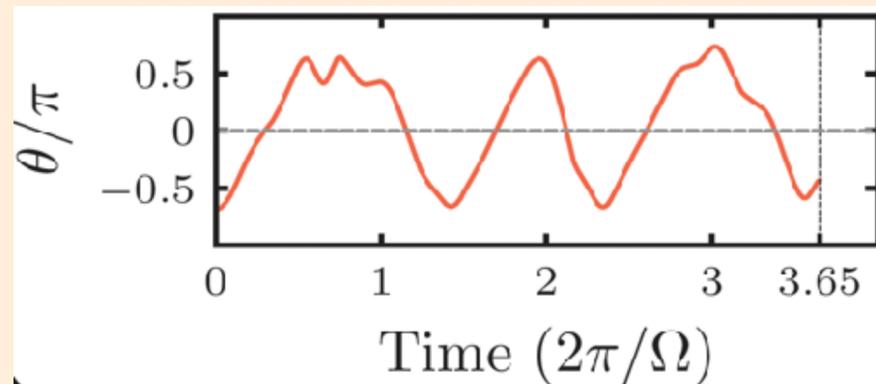
Non-local connectivity



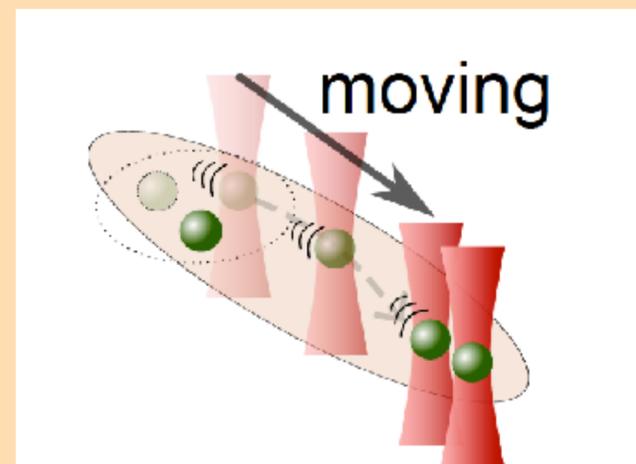
Solution read-out



Native multi-qubit gates

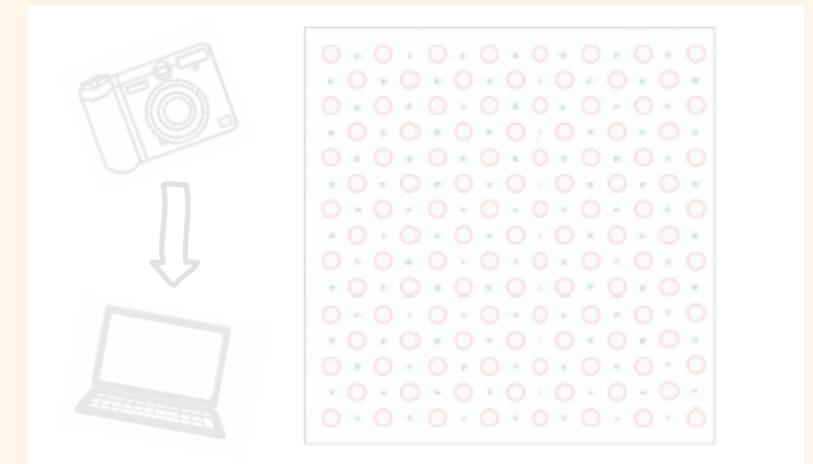


Atom moving



D. Bluvstein et al., Nature **604**, 451–456 (2022)

Co-processing



H.-Y. Huang et al., Nat. Phys. **16**, 1050–1057 (2020)

# Efficient Read-Out Based on Snapshots

## Leverage

- ability for efficient time evolution of Rydberg simulator
- ability to perform snapshot measurements

Key quantity: Operator-resolved density of states

$$D_A(\omega) = \sum_n \langle n | A | n \rangle \delta(\omega - \epsilon_n)$$

How to obtain?



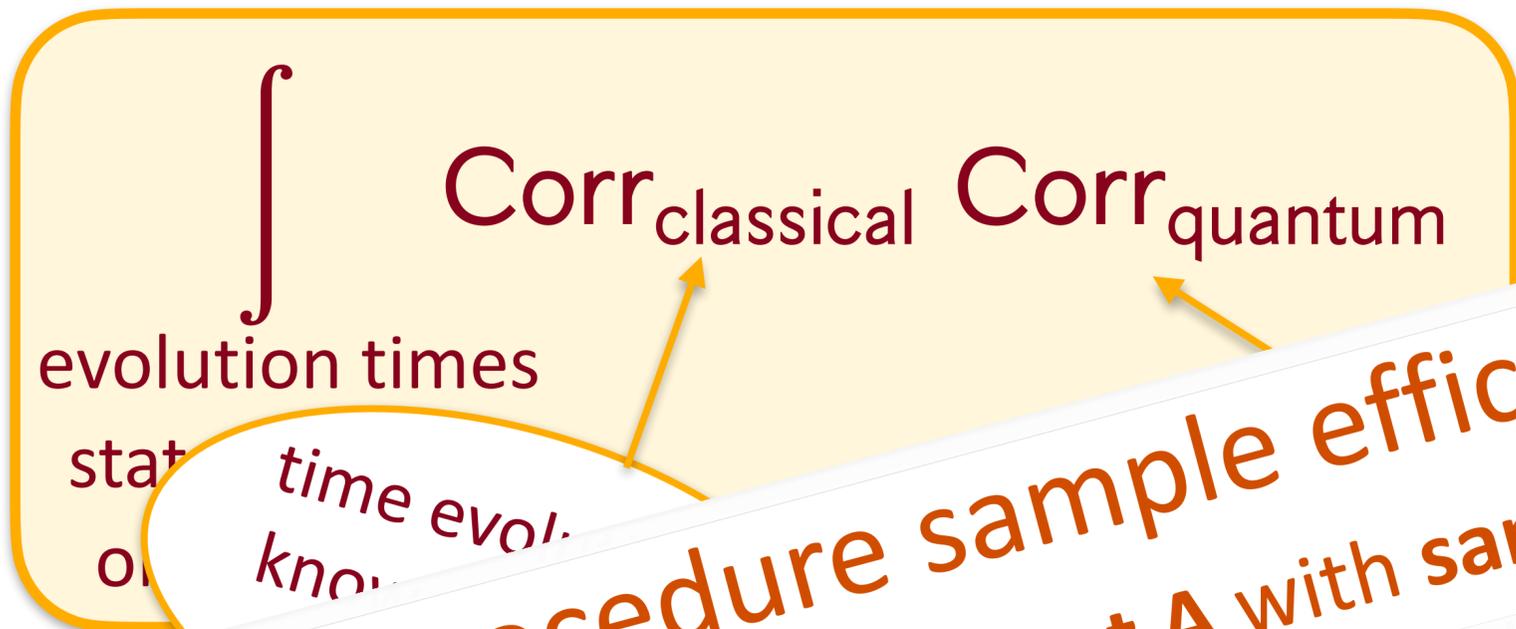
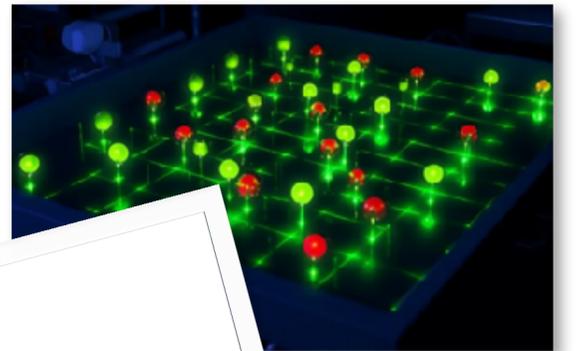
peaks at state energies

Such measurements are very sensitive to disorder

# Efficient readout: Quantum-Classical Co-Processing

Hardware-efficient toolbox to compute spectral functions

$$D_A(\omega) = \sum \langle n | A | n \rangle \delta(\omega - \epsilon_n) =$$



**Makes procedure sample efficient:  
 $D_A$  can be found for different  $A$  with same sample set**

$$D_A(\omega) = \sum_{s,r} \langle S | O_{s,R}(0) e^{-iE_S t} O_{s,R}(t) | S \rangle + O(1/\sqrt{M})$$

Uniformly sample perturbed states

$$\mathbb{E}_{R \sim \mathcal{R}} [R | S \rangle \langle S | R] = I$$

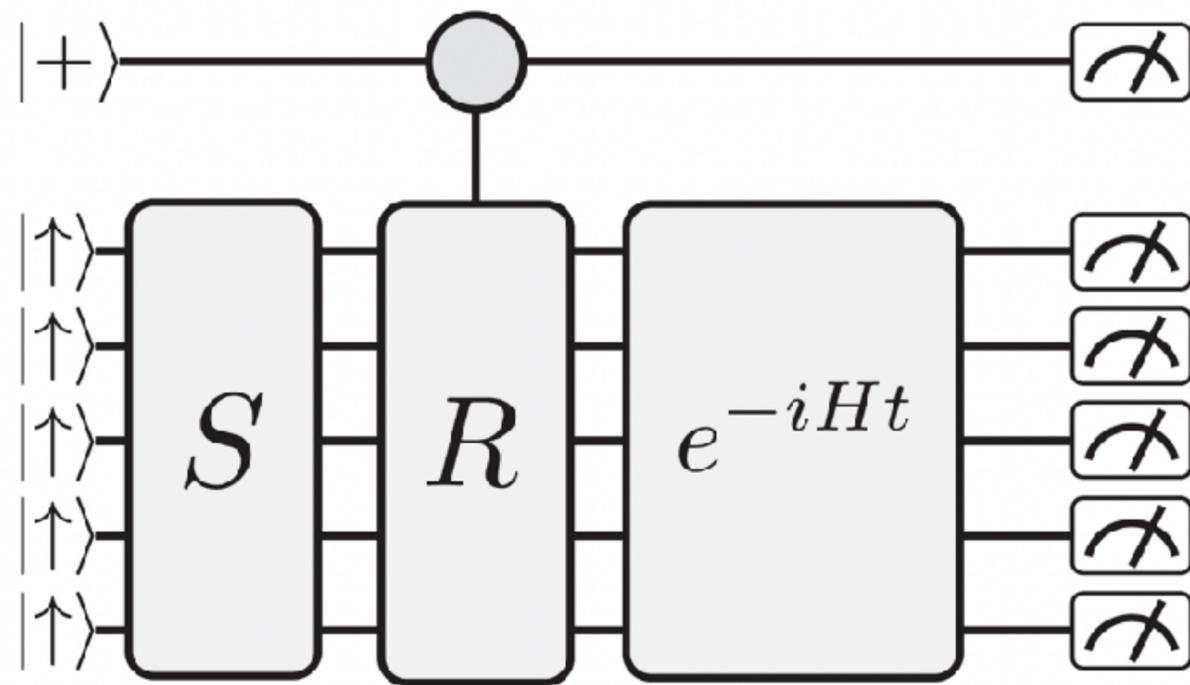
Sum over observables

$$\sum [O_s | S \rangle \langle S | O_s] = I$$

Estimate via **quantum** time evolution on simulator and  $M$  snapshots.

Evaluate **classically** using an efficient representation e.g. MPS  $|S\rangle, R|S\rangle$  and MPO  $A$

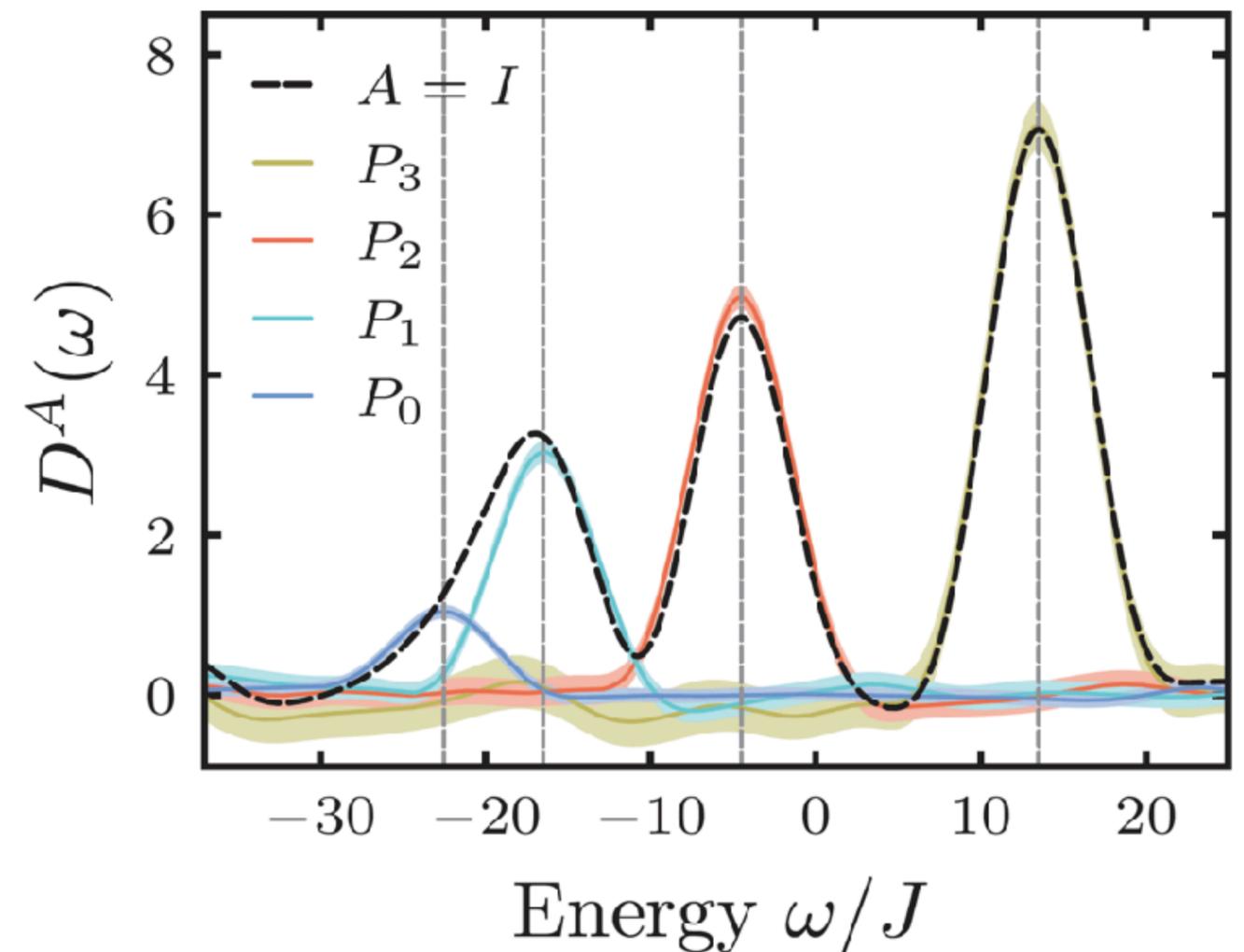
# Efficient readout: Quantum Circuit + Spectrum



Parallel measurement of  $2^n$  observables  
(any operator diagonal in measurement basis)

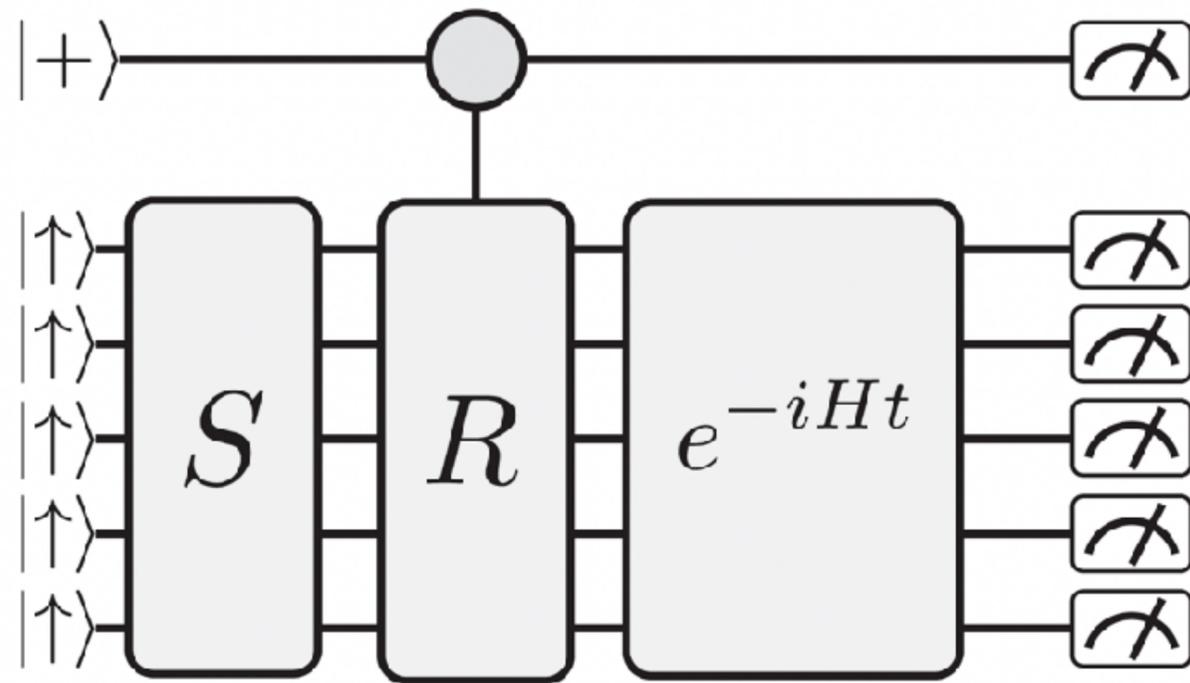
Example: two interacting spin-3/2s

$$H = J \mathbf{S}_1 \cdot \mathbf{S}_2$$



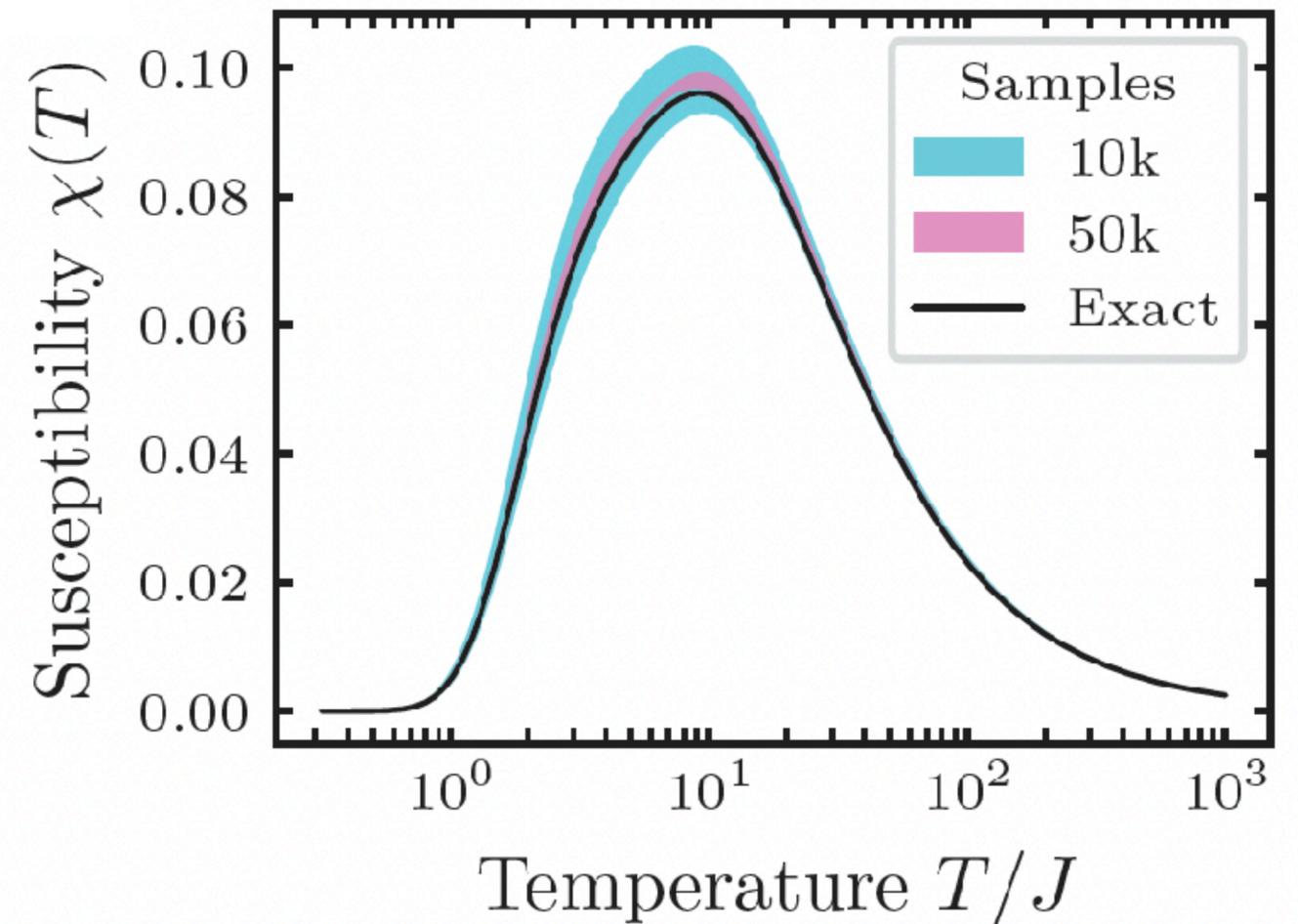
20,000 samples

# Efficient readout: Quantum Circuit + Spectrum

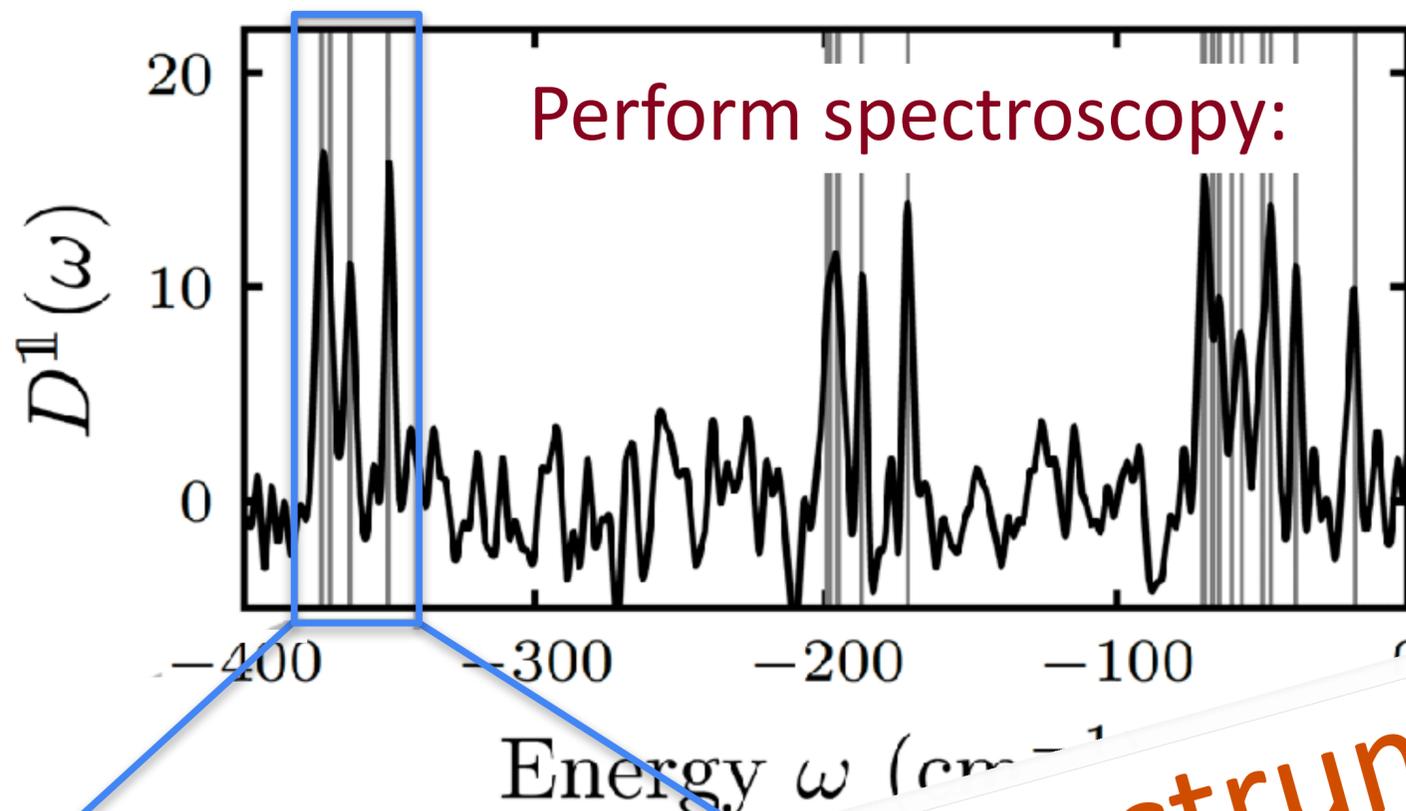
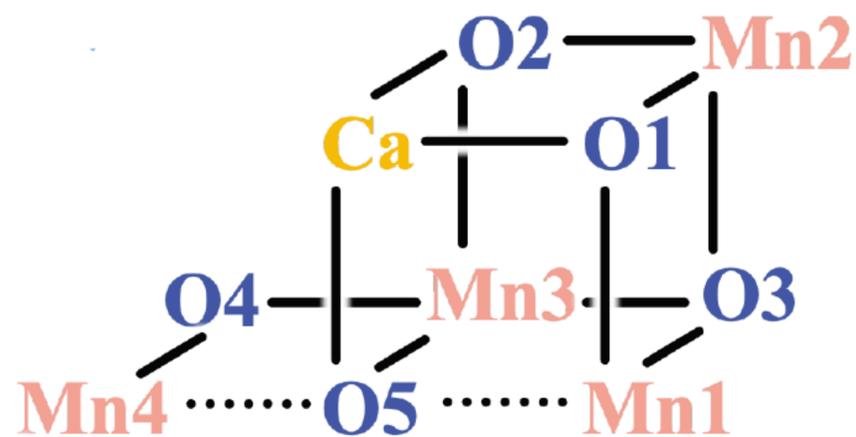


Parallel measurement of  $2^n$  observables  
(any operator diagonal in measurement basis)

Access to full spectral information  
⇒ finite temperature properties!

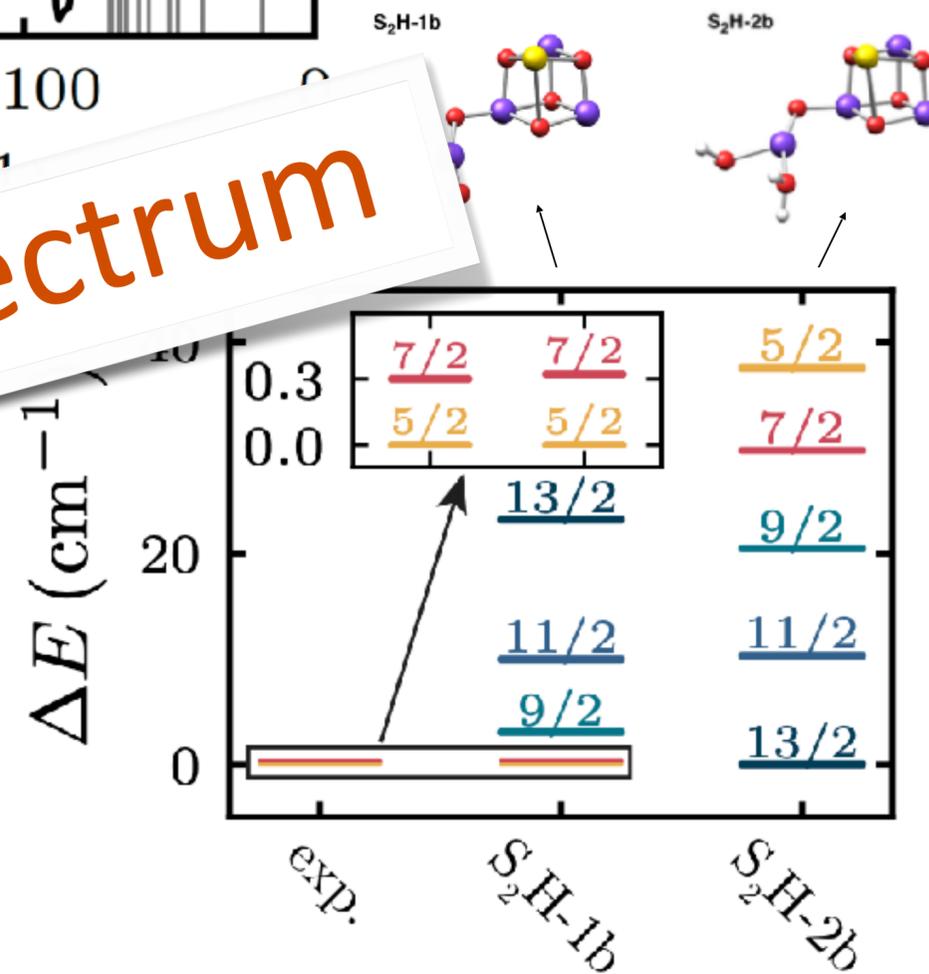
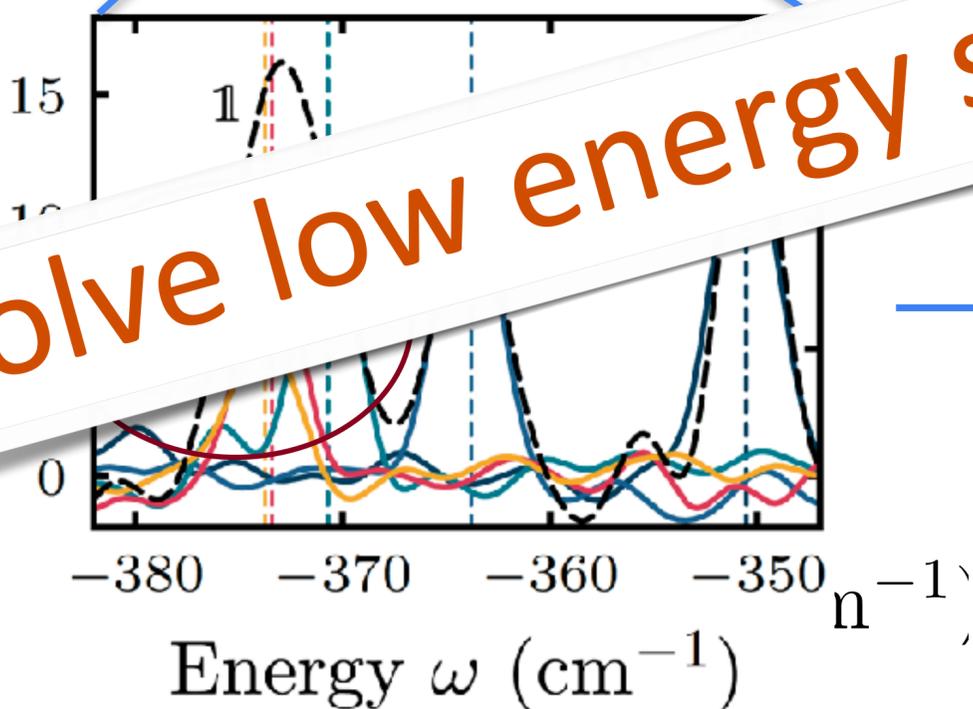


# Benchmark on Larger Molecule (OEC Complex)



resolve low energy spectrum

spin-projection allows detection of tiny energy differences ( $0.3 \text{ cm}^{-1}$ )

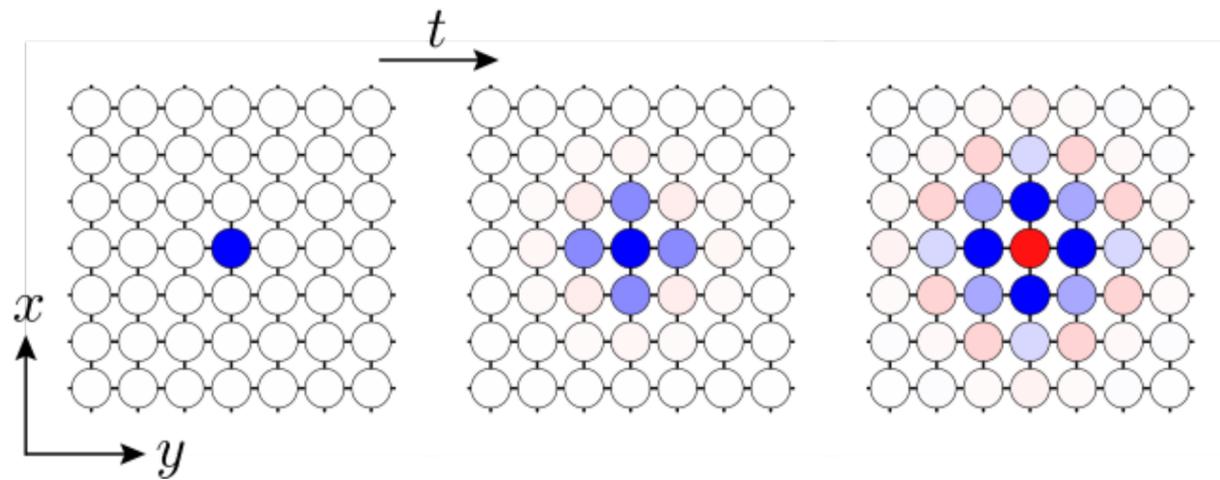




# Additional Applications and Outlook

## Application to 2D magnetic materials

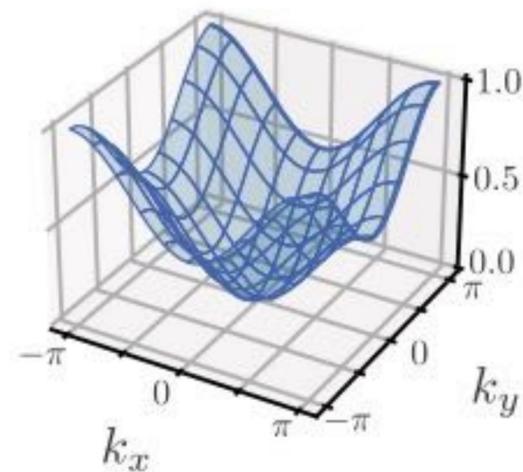
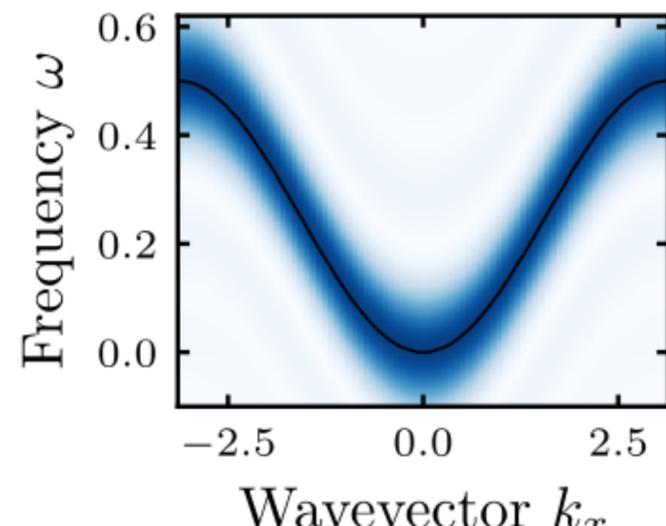
Single-particle Green's function of FM Heisenberg:



Quasi-particle properties encoded in spectral function

$$S(k, \omega) = |G(k, \omega)|^2$$

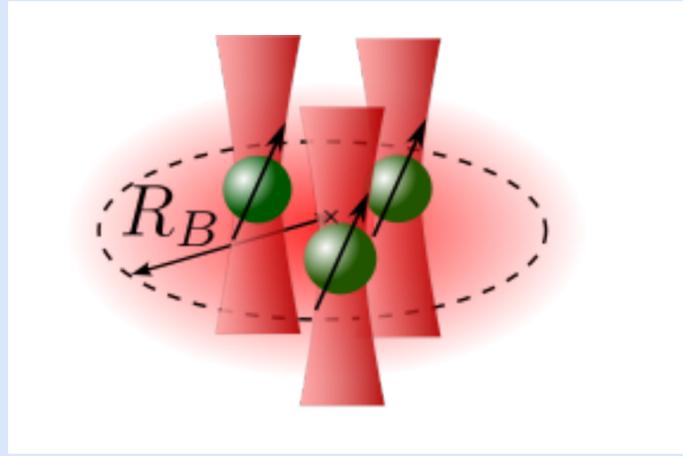
Dispersion  $\epsilon(k)$



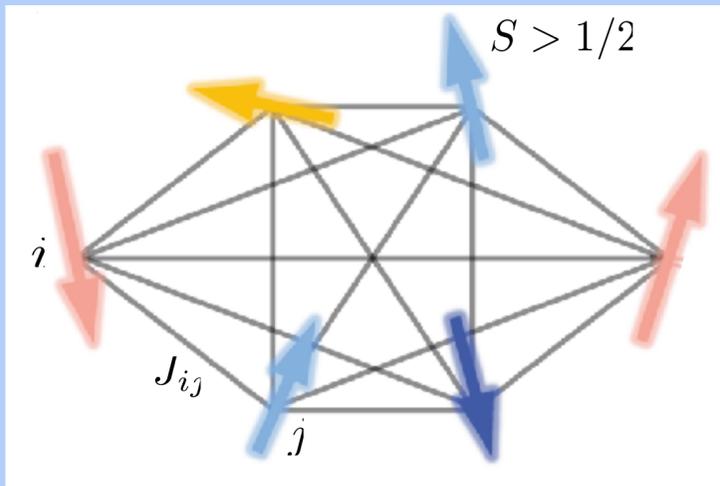
Next steps:

- Expand operator-resolved density of states
- include error correction
- dynamics of chemical reactions
- simulate fermions (e.g., Coulomb Hamiltonian)

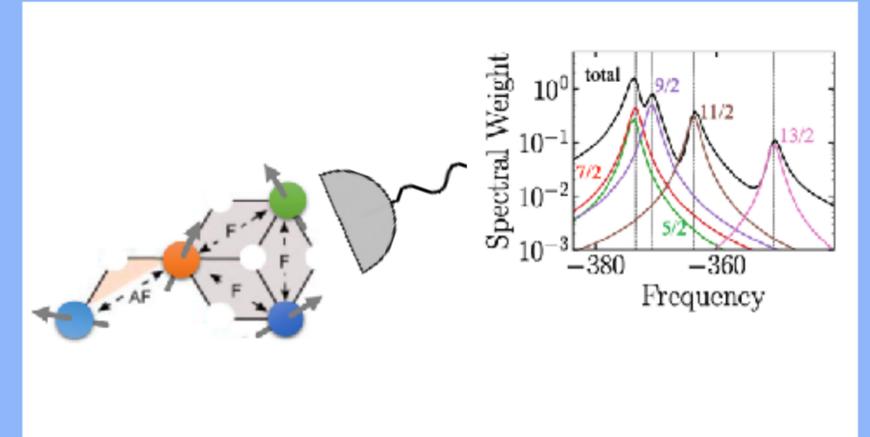
## Control high spin ( $S > 1/2$ )



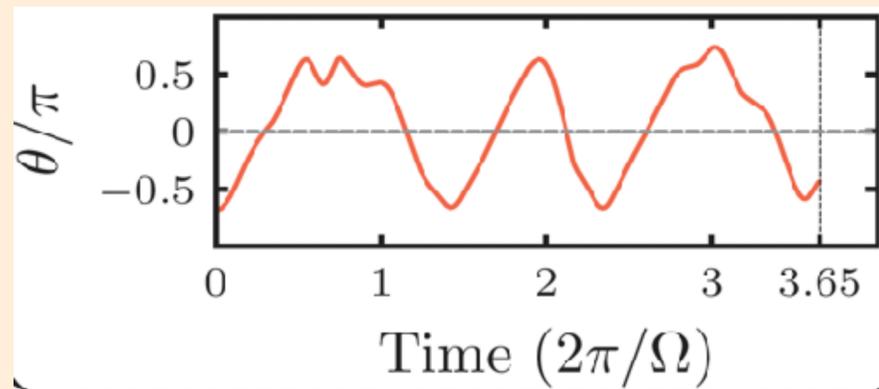
## Non-local connectivity



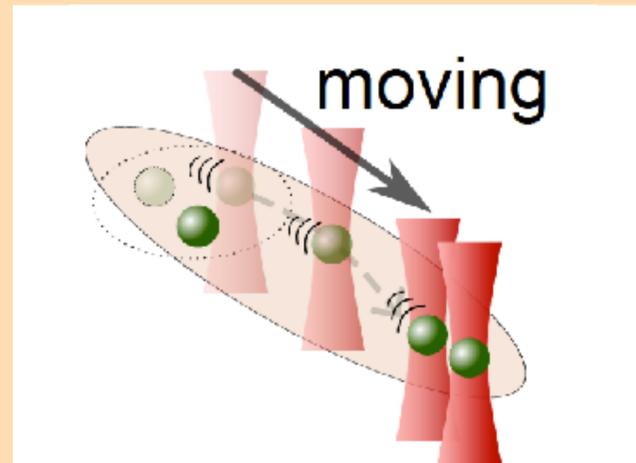
## Solution read-out



## Native multi-qubit gates

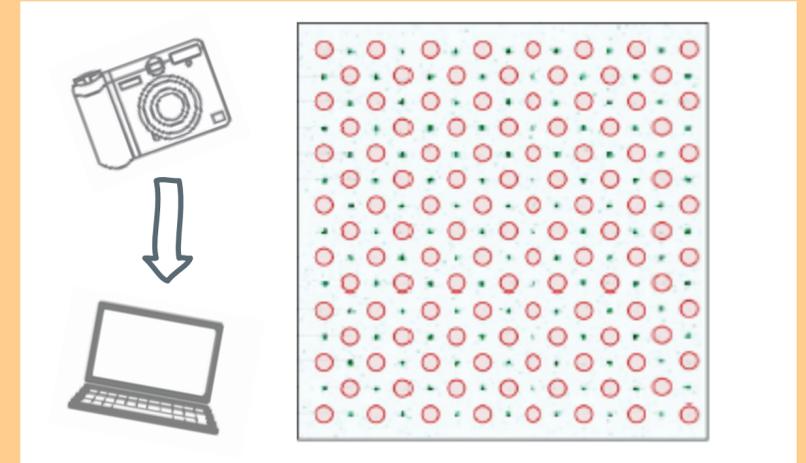


## Atom moving

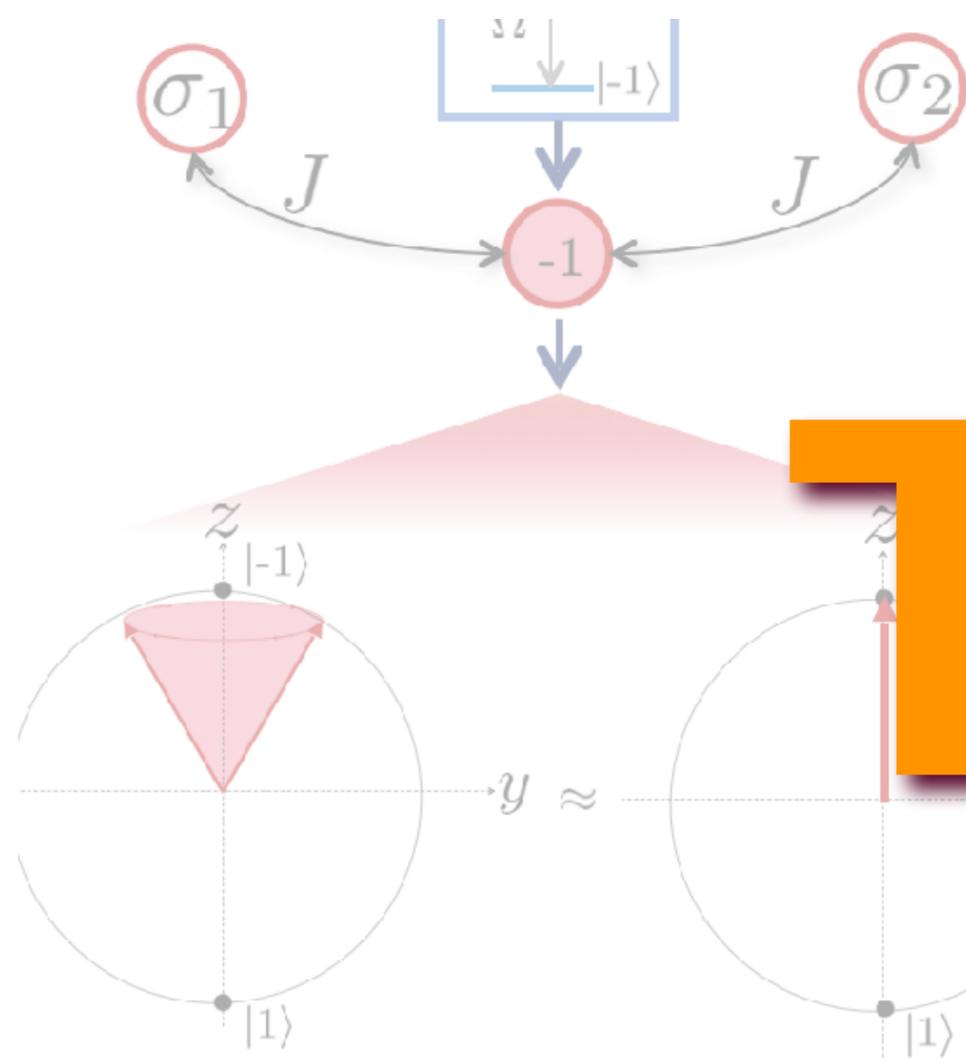


D. Bluvstein et al., Nature **604**, 451–456 (2022)

## Co-processing



H.-Y. Huang et al., Nat. Phys. **16**, 1050–1057 (2020)



# Thank you!

