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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James Shee
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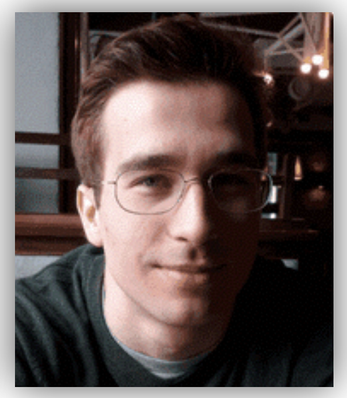

Marcin Kalinowski
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→ IBM Quantum)

Anna Krylov
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(Harvard)

Martin Head-Gordon
(UC Berkeley)

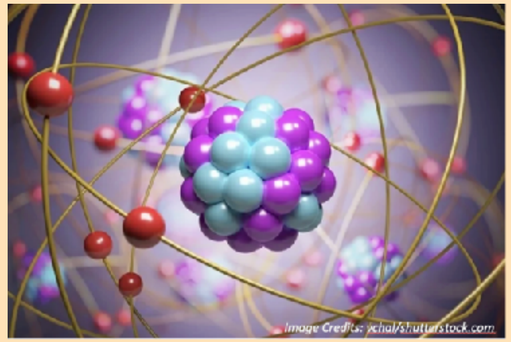
Mikhail Lukin
(Harvard)

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:

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

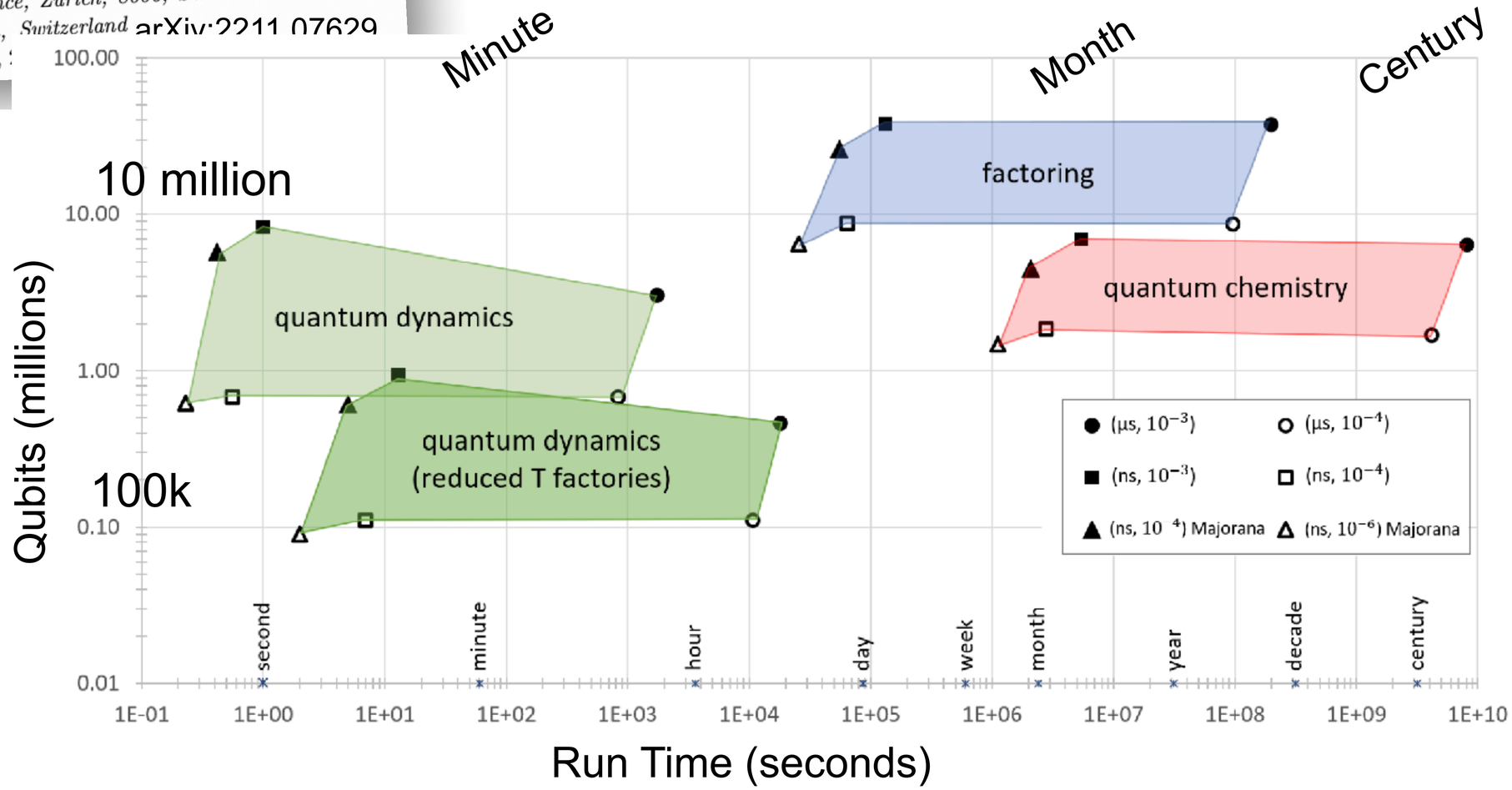
Challenge: Simulating systems with **strong correlations**

➤ 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,[1] P. Murali,[1] M. Troyer,[1] K. M. Svore,[1] T. Hoefler,[2]
V. Kliuchnikov,[1] G. H. Low,[1] M. Soeken,[3] A. Sundaram,[1] and A. Vaschillo[1]
[1] *Microsoft Quantum, Redmond, WA 98052, USA*
[2] *ETH Zurich, Department of Computer Science, Zürich, 8006, Switzerland*
[3] *Microsoft Quantum, Zurich, Switzerland*
arXiv:2211.07629
(Dated: November 19,



Quantum Chemistry on Quantum Computers

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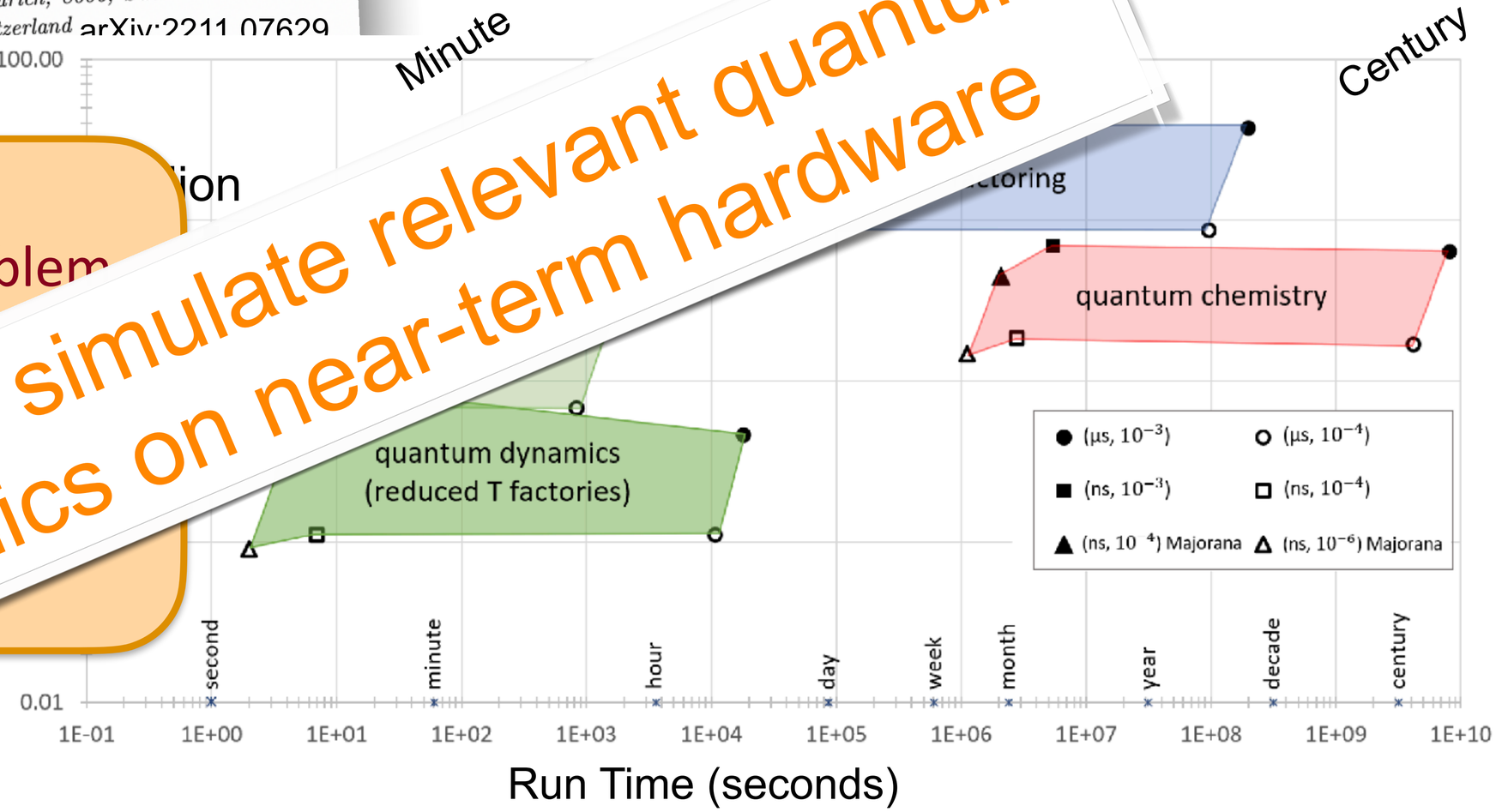
Sources of overhead:

- Mapping of target problem to quantum hardwa
- Expensive alg (quant
- Large

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



Advancing Computational Quantum Chemistry

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?

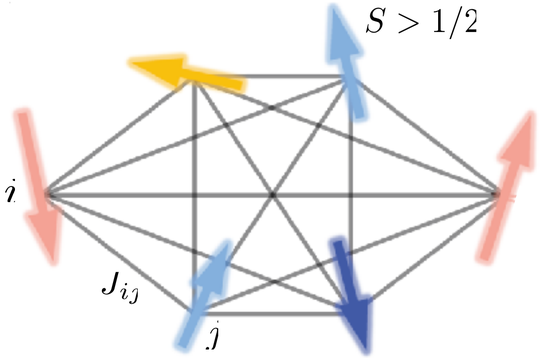
problems with strong correlations

Hybrid Quantum-Classical Workflow

High-performance computing

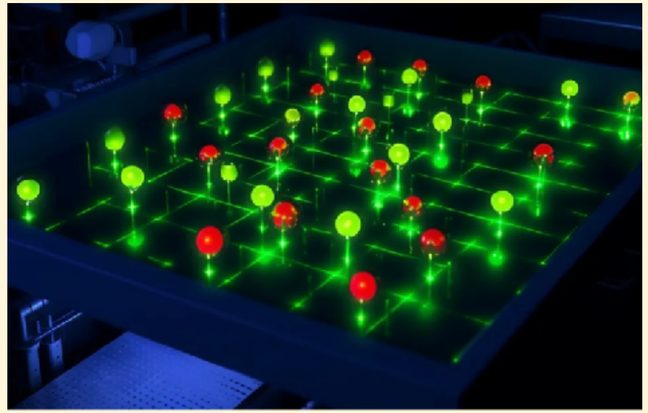
- Employ state-of-the art algorithms (Coupled Cluster, DMRG, etc.)

Model Hamiltonians for quantum chemistry and material science



capture the strong correlations (at low energies)

Large scale analog-digital quantum simulator

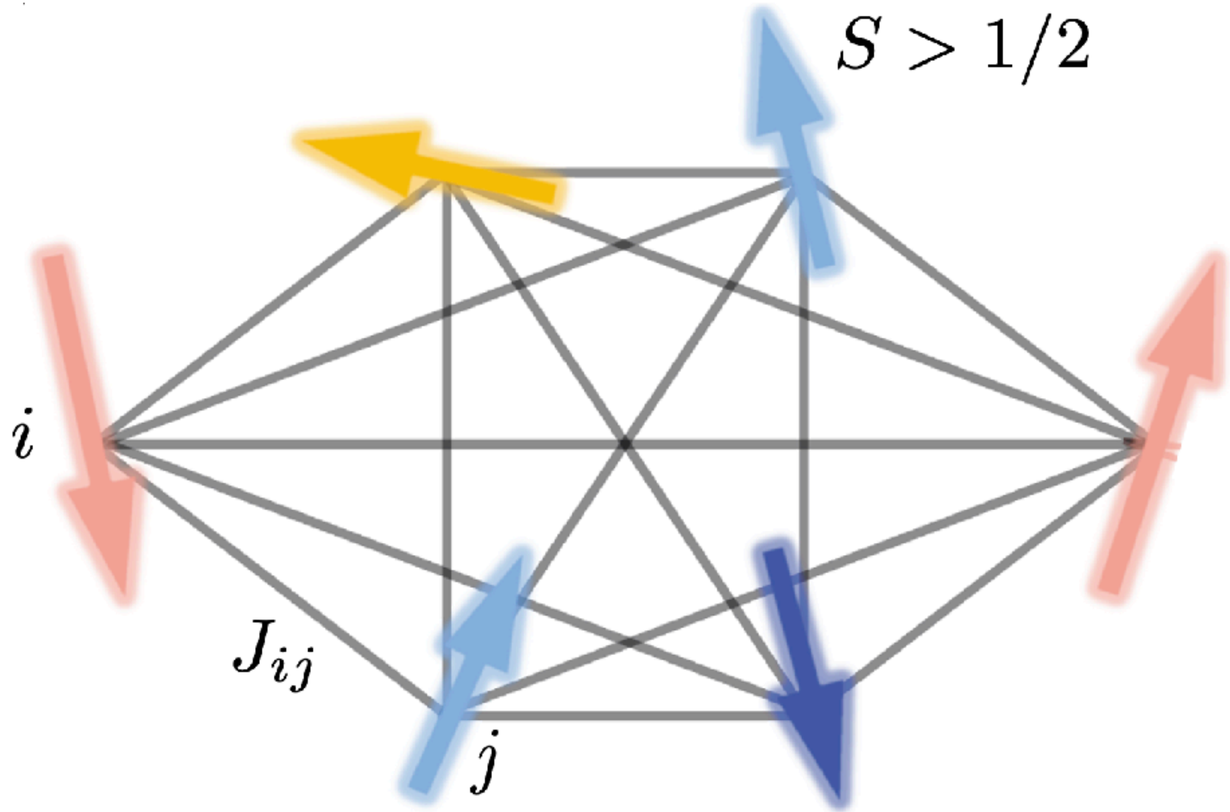


- Efficient multi-qubit gates
- High-degree of programmability

→ Simulate time dynamics of many-particle systems

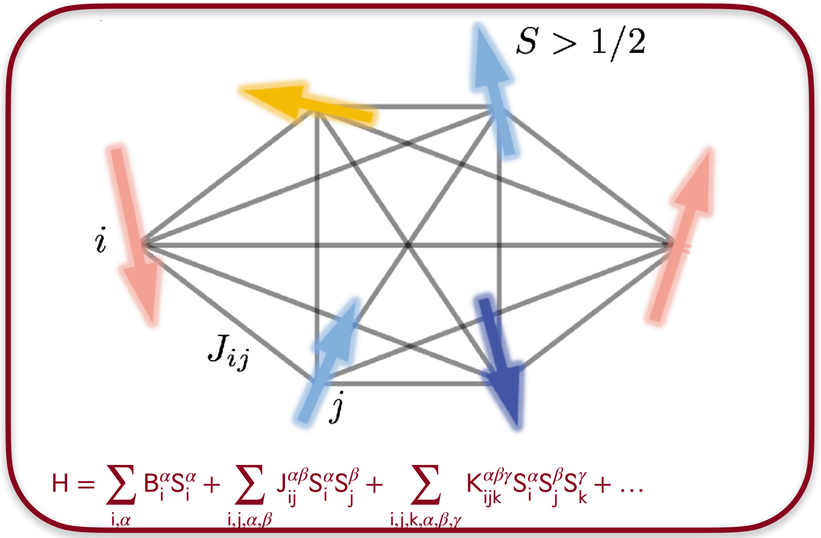
Target Problem

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



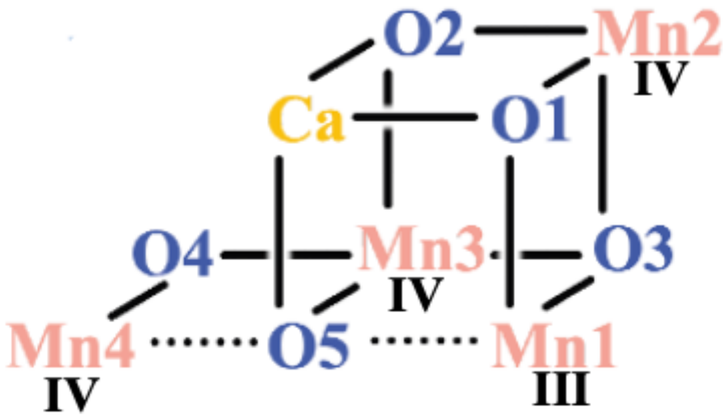
$$H = \sum_{i,\alpha} B_i^{\alpha} S_i^{\alpha} + \sum_{i,j,\alpha,\beta} J_{ij}^{\alpha\beta} S_i^{\alpha} S_j^{\beta} + \sum_{i,j,k,\alpha,\beta,\gamma} K_{ijk}^{\alpha\beta\gamma} S_i^{\alpha} S_j^{\beta} S_k^{\gamma} + \ldots$$

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.

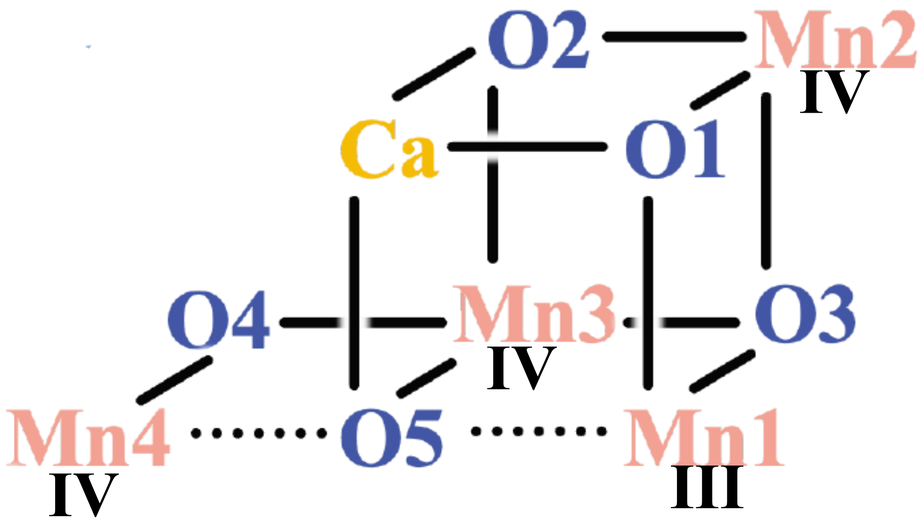
- Good model for materials or molecules with localized valence electrons



- 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)



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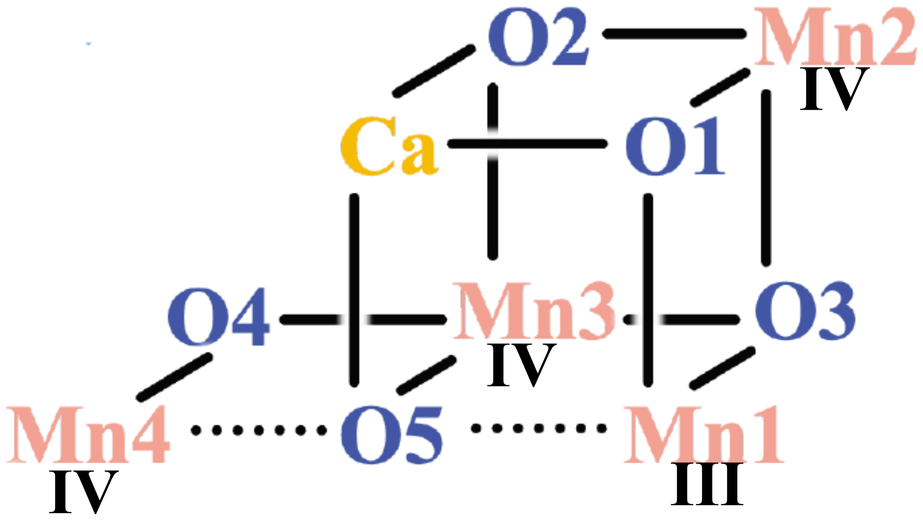

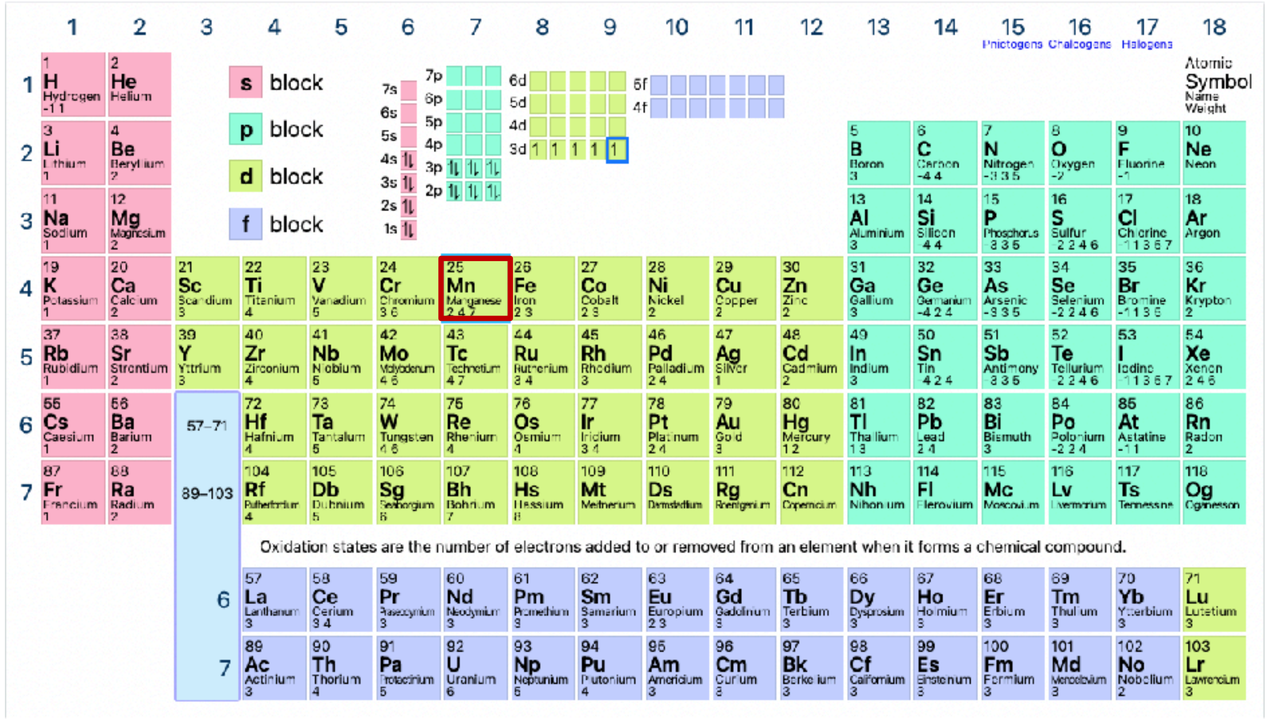

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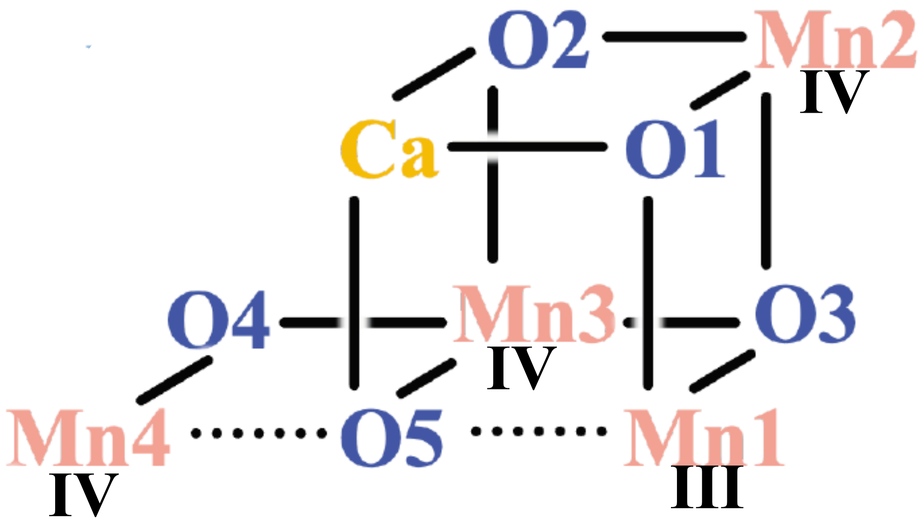

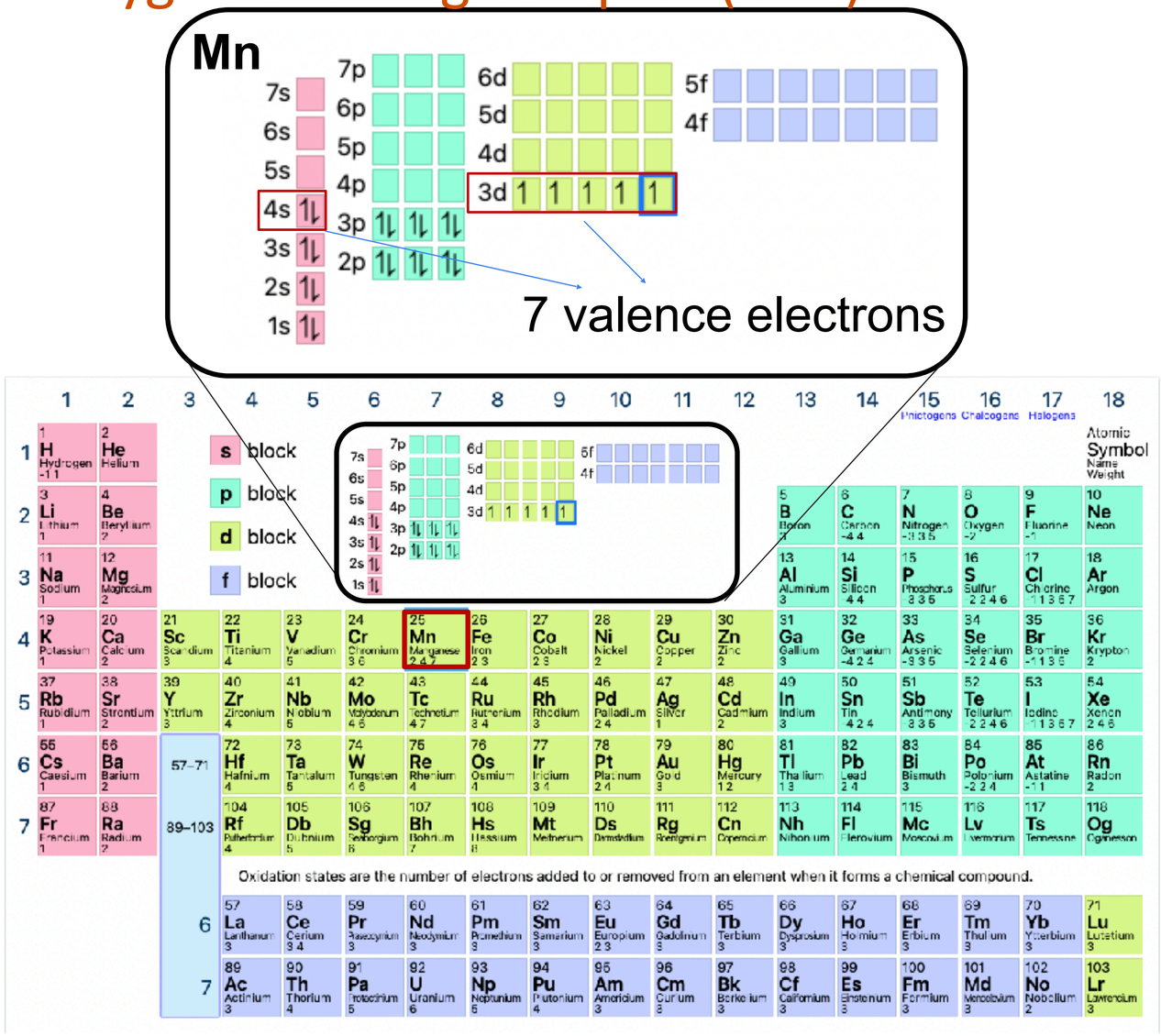

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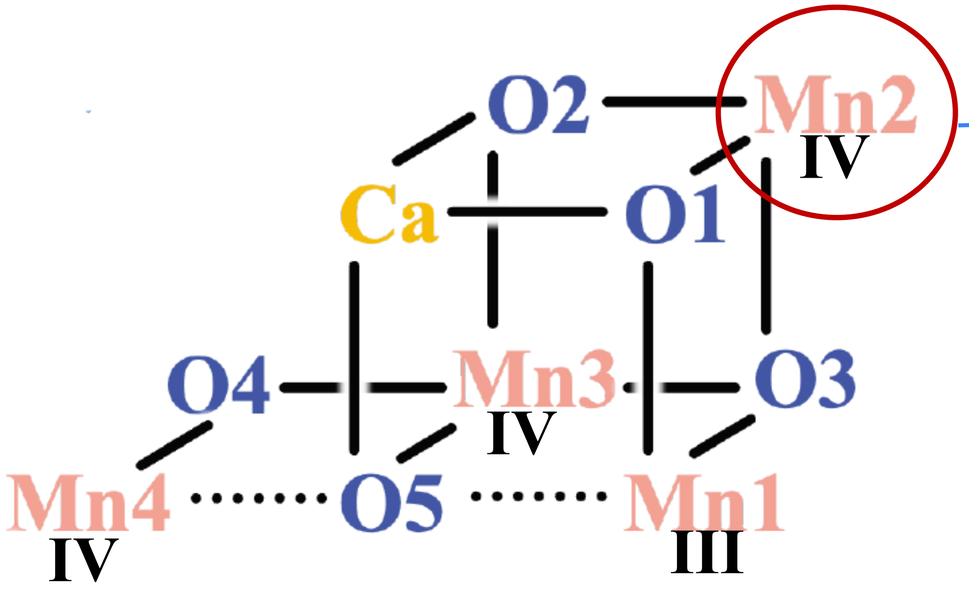

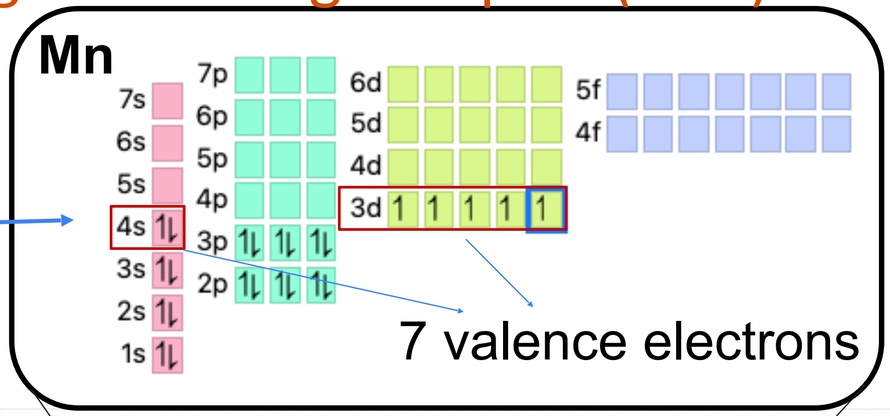

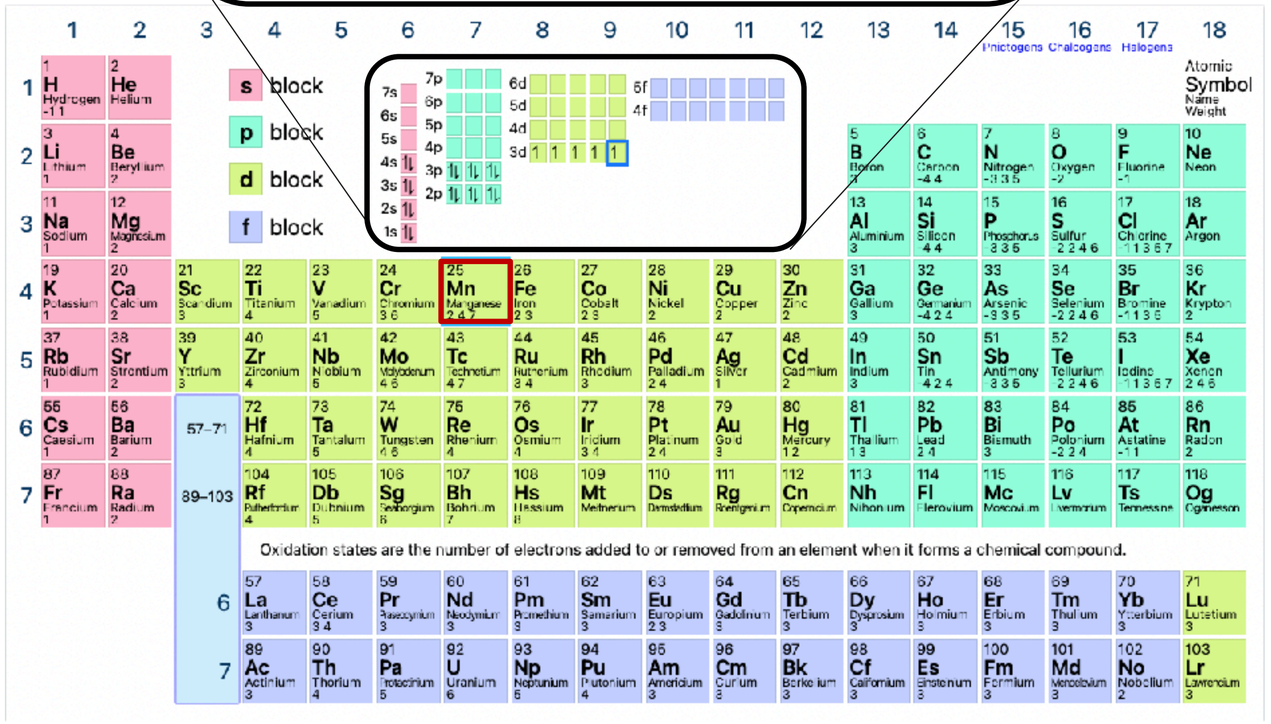

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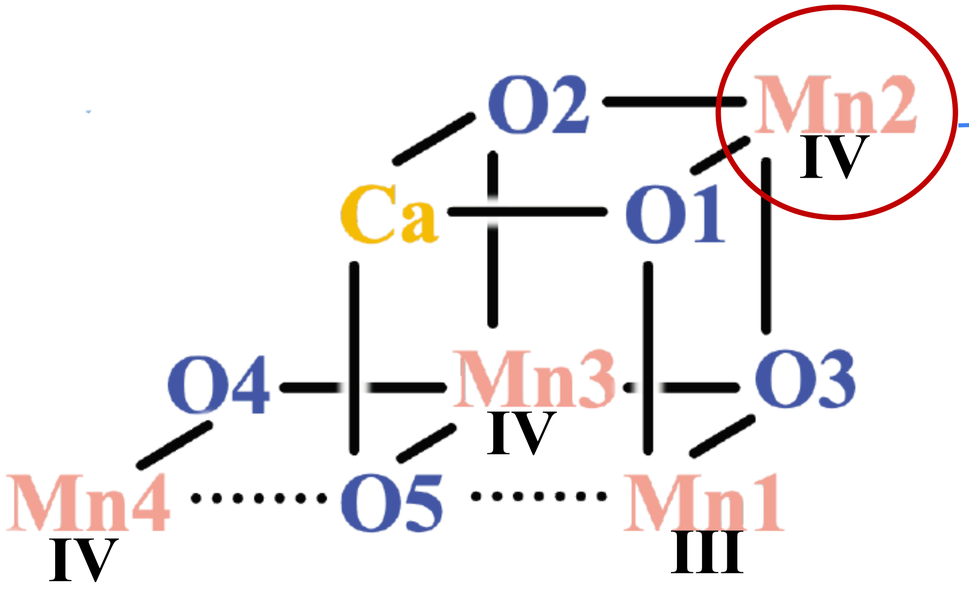

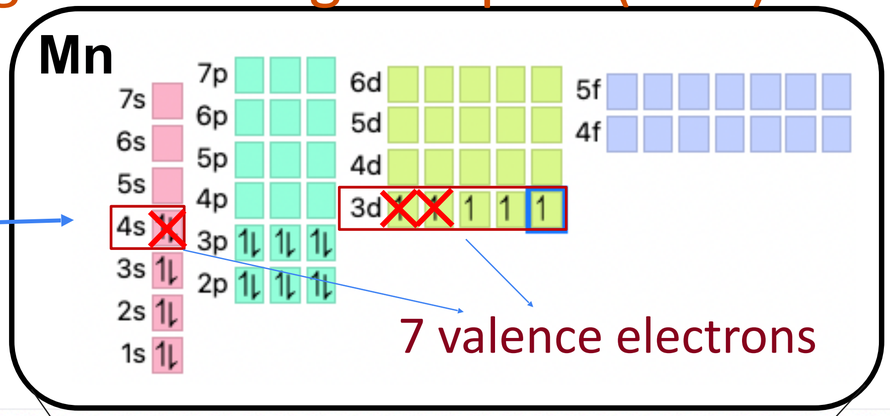

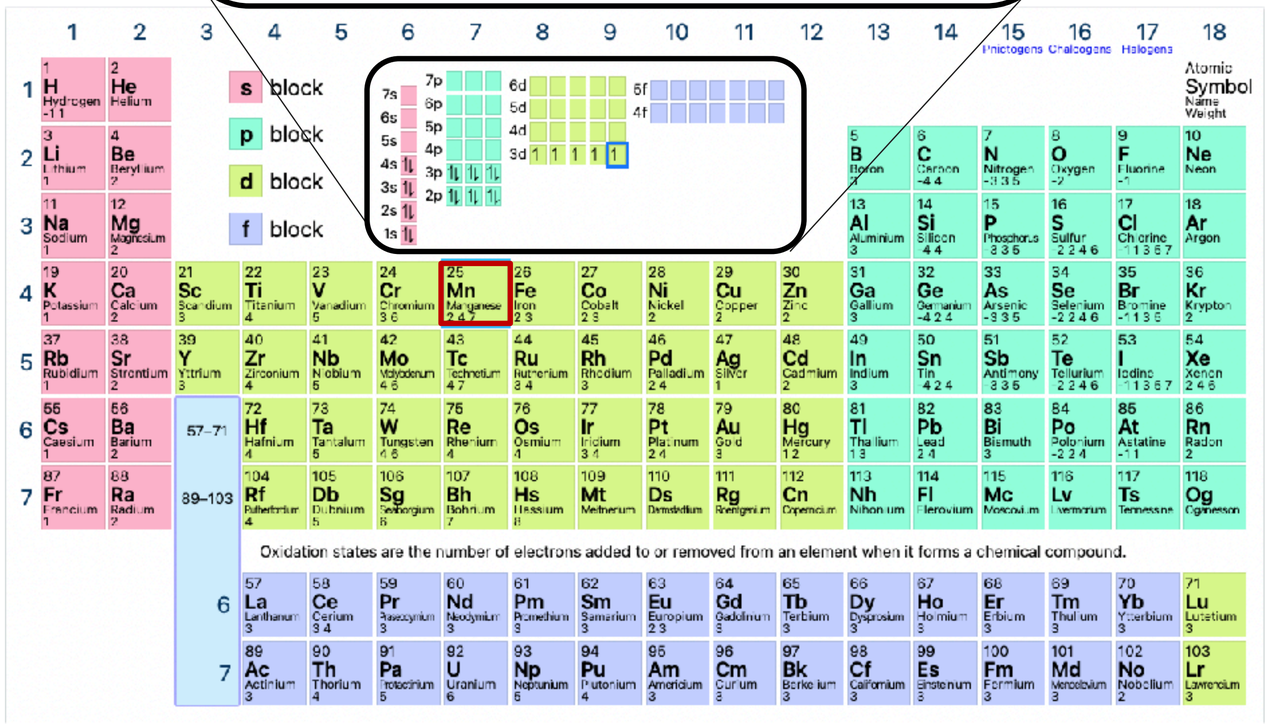

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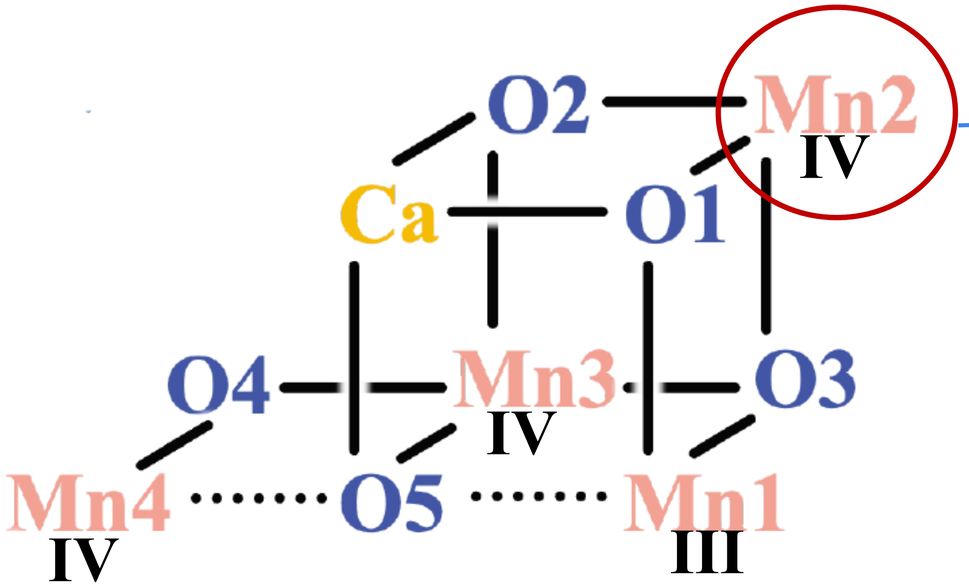

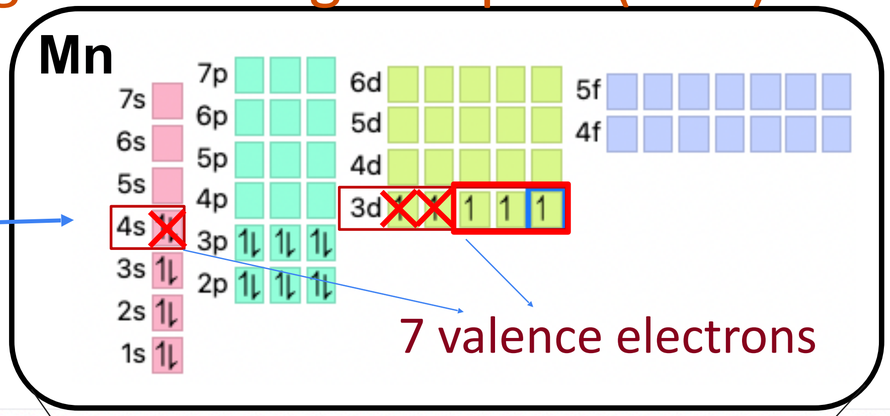

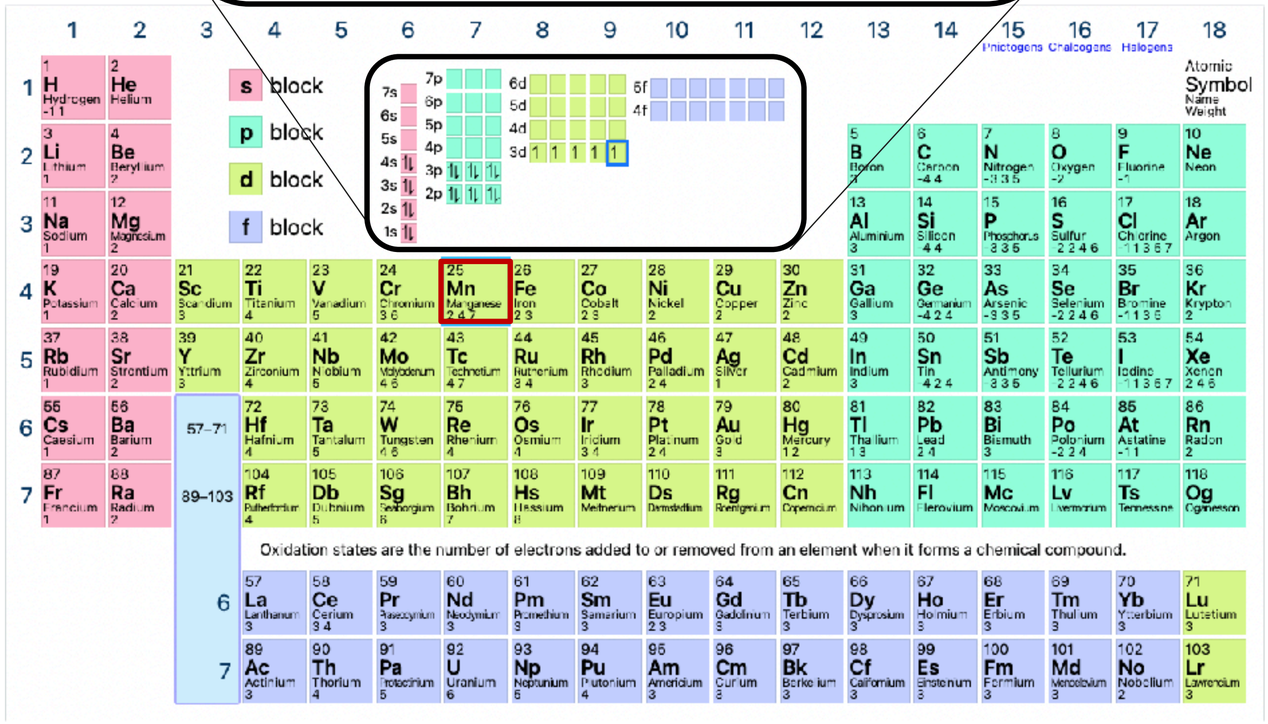

→ Mn2 has 3 **active** electrons

Model Hamiltonians

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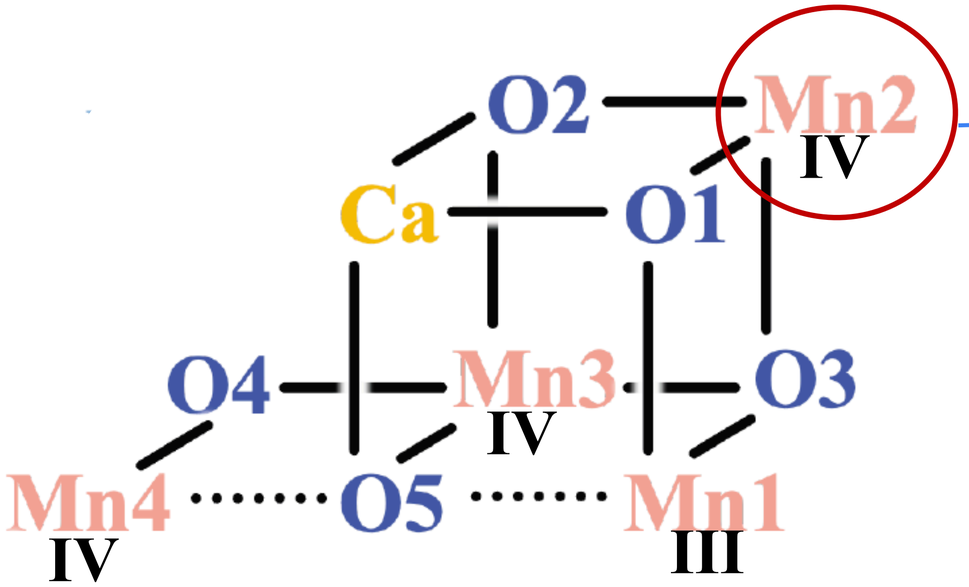
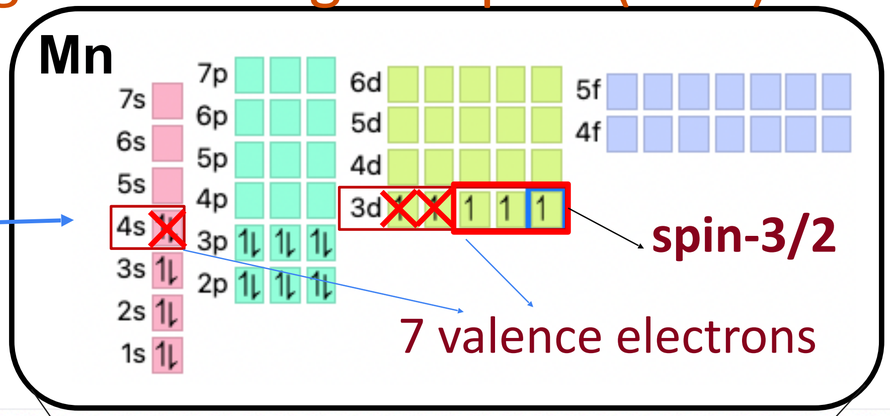
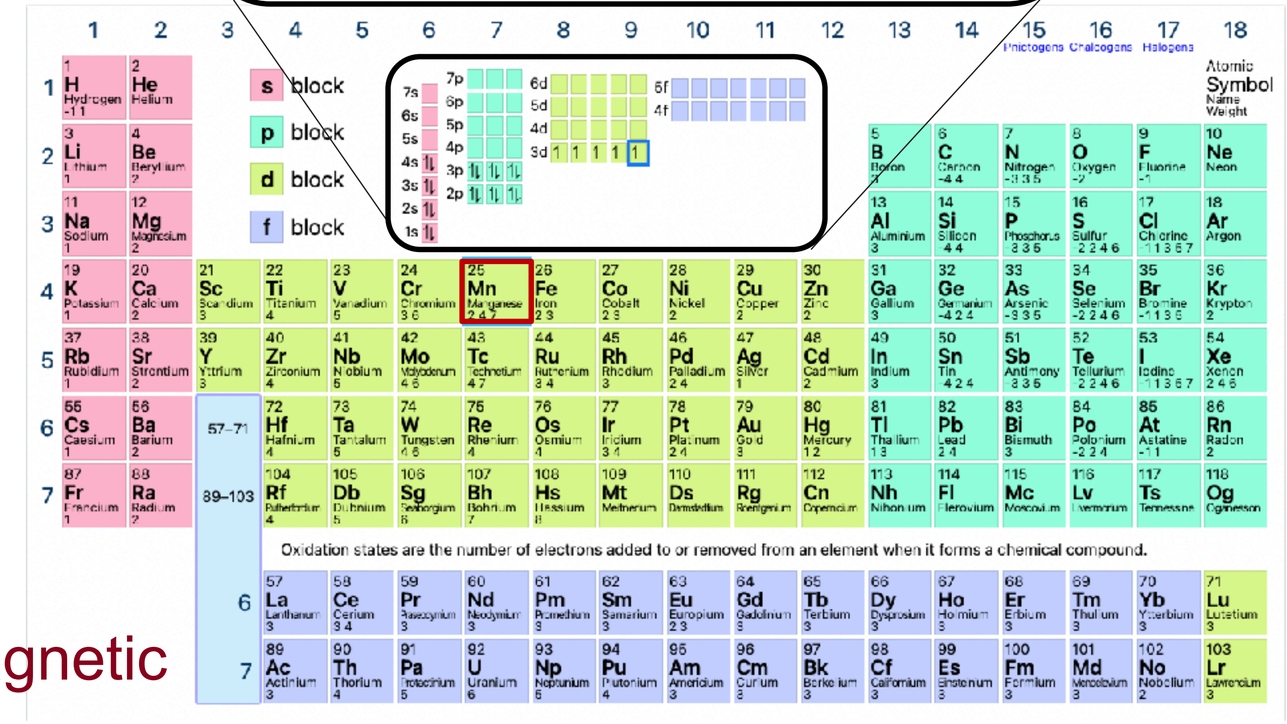




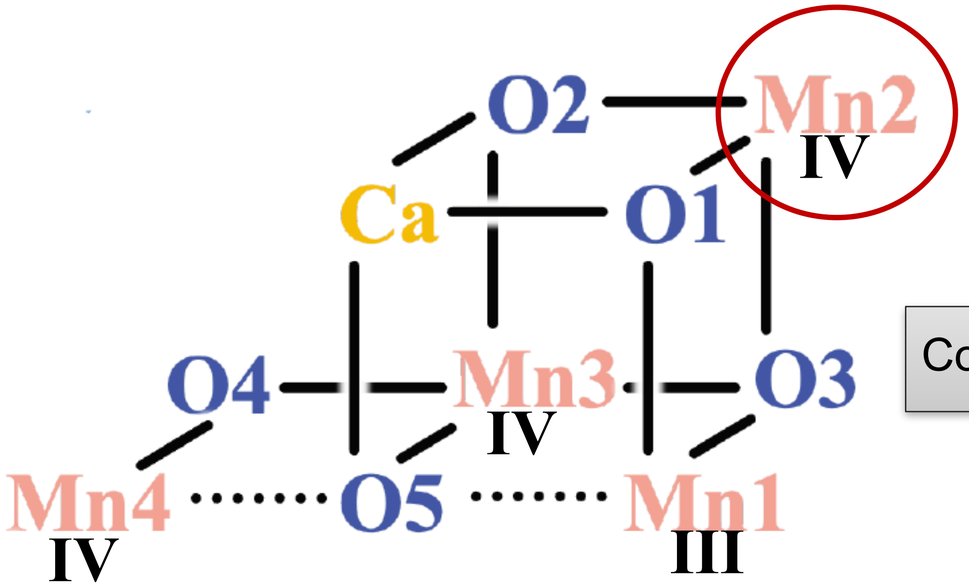


→ Mn2 has 3 **active** electrons

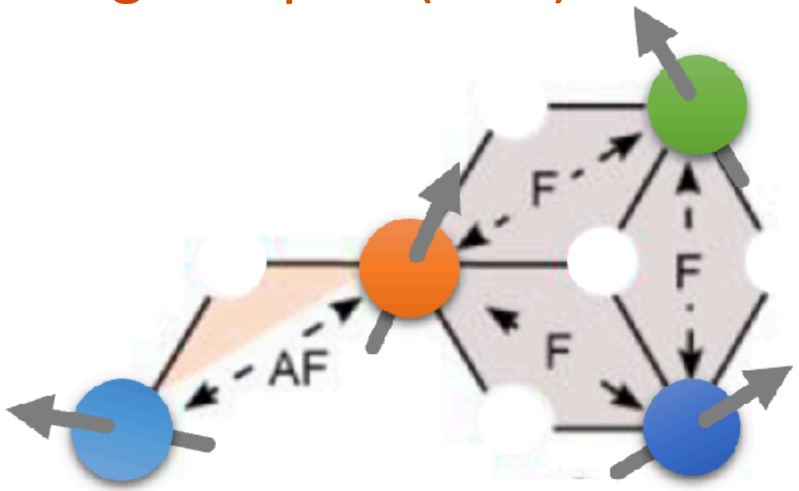
- Coulomb interaction - localizes electrons
- Exchange interaction - ferromagnetic
- Super-exchange interaction - anti-ferromagnetic

Model Hamiltonians

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



Computational Chemistry



→ Mn2 has 3 **active** electrons

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

- Represent high spins …

  ➡ How to implement high spins?

- … and let them interact

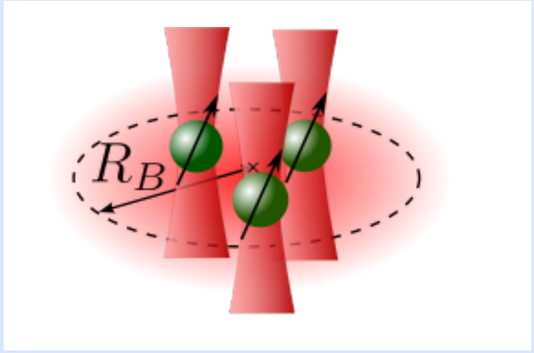
  ➡ How to implement non-local connectivity?

- Read out chemically relevant quantities

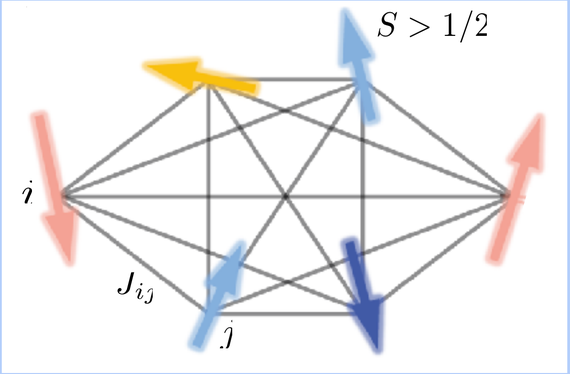
  ➡ Quantum-classical co-processing

Necessary Ingredients

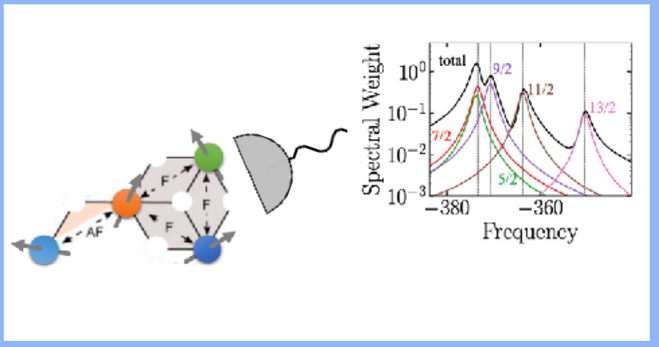
Control high spin (S>1/2)



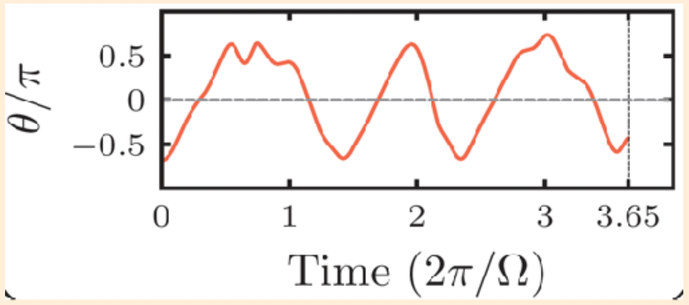
Non-local connectivity



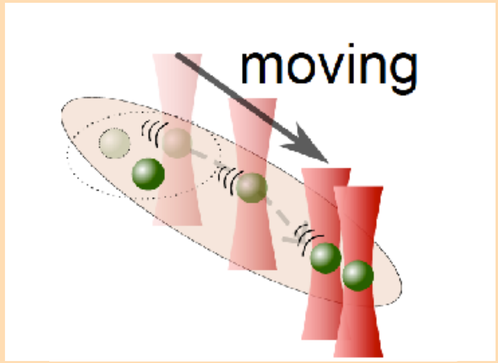
Solution read-out



Native multi-qubit gates

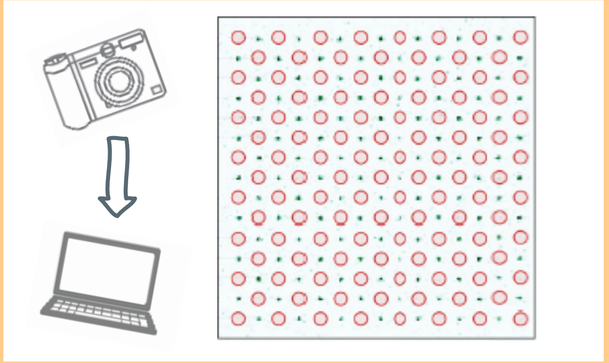


Atom moving



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

Co-processing

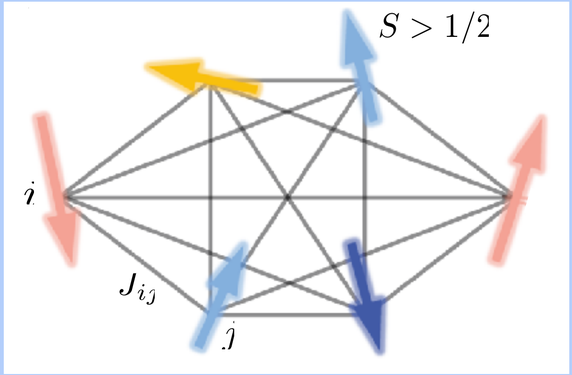


H.-Y. Huang et al., Nat. Phys. 16, 1050–1057 (2020)

Necessary Ingredients

Control high spin (S>1/2)

Non-local connectivity



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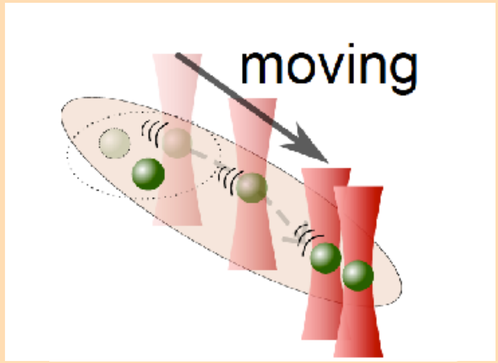

D. Bluvstein et al., Nature **604**, 451–456 (2022)

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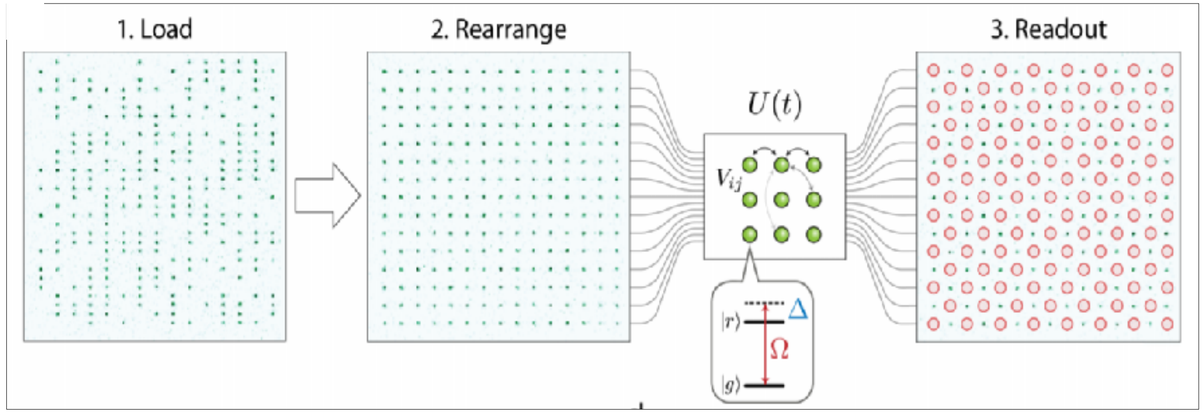
H.-Y. Huang et al., Nat. Phys. 16, 1050–1057 (2020)

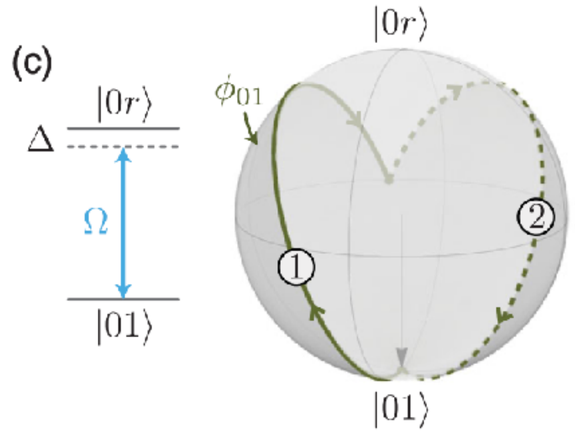
Example implementation: Reconfigurable atom arrays

Analog quantum simulation



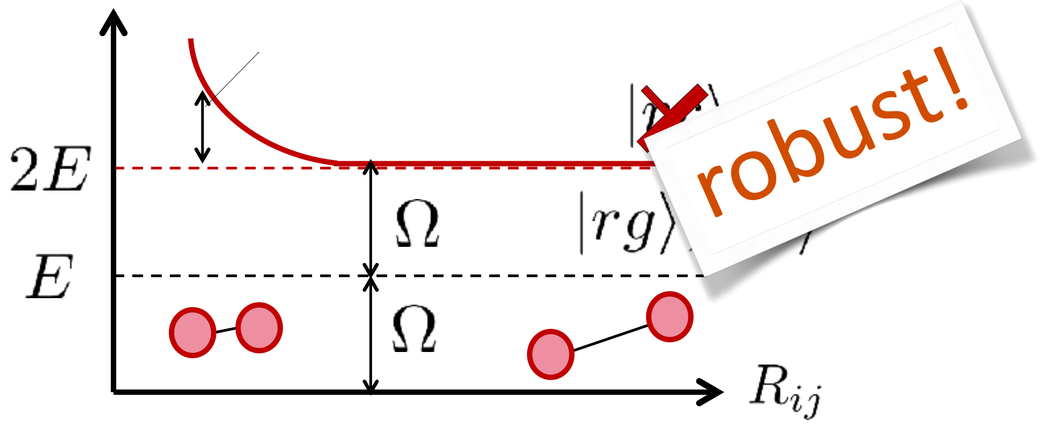
Ebadi, S., et al., Nature (2020)

Digital Quantum Computing (gates)



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

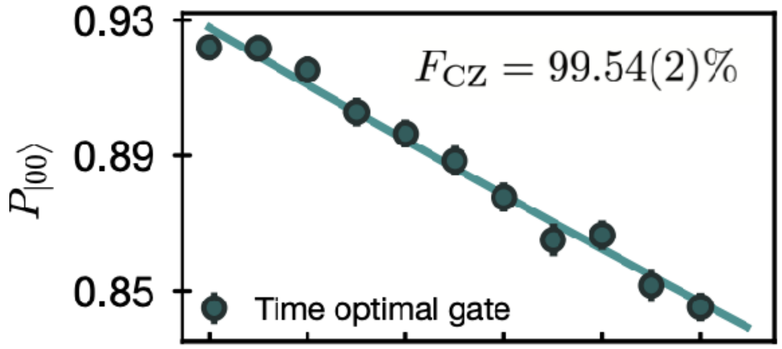
Rydberg blockade



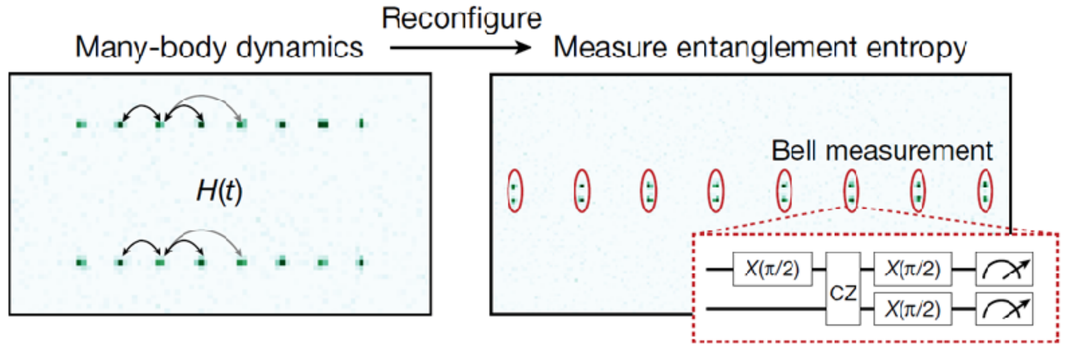
Atom Array Platform in Analog-Digital Mode

Reconfigurable architecture

High-fidelity gates



Hybrid analog-digital control



High degree of programmability

Interactions manipulated via geometric configuration

+

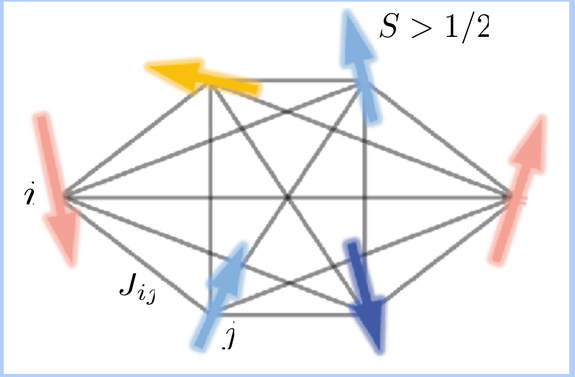
Global control pulses

Bluvstein *et al. Nature* **604**, 451 (2022).
Evered, *et al. Nature* **622**, 268n (2023)

Necessary Ingredients

Control high spin (S>1/2)

Non-local connectivity



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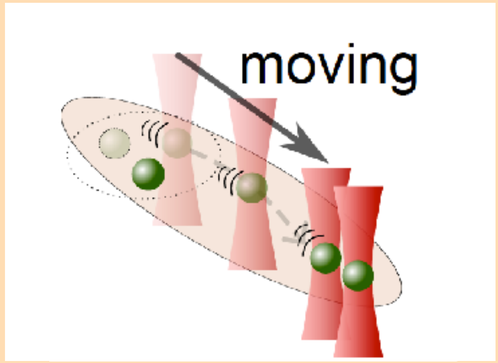

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Co-processing

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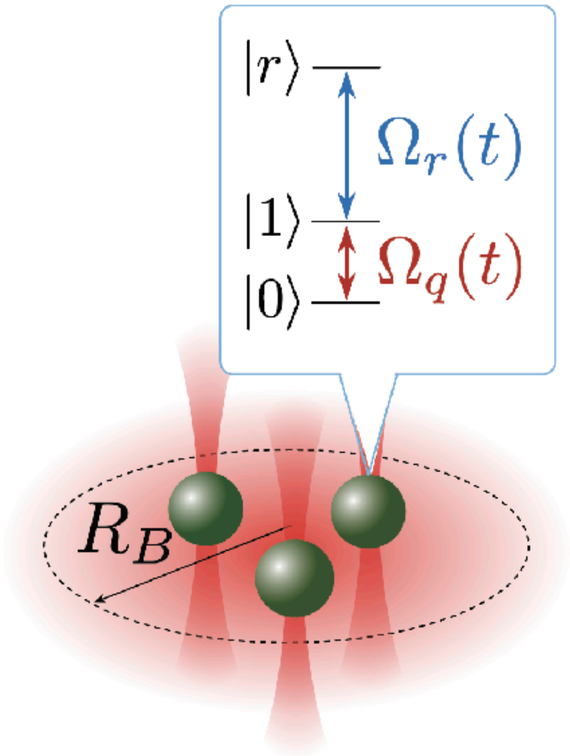
Engineering Large Spins on Rydberg Platform

Hardware Efficient Multi-Qubit Operations with a global drive



Encode spin-S variables into 2S (spin-1/2) qubits ("clusters"):

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

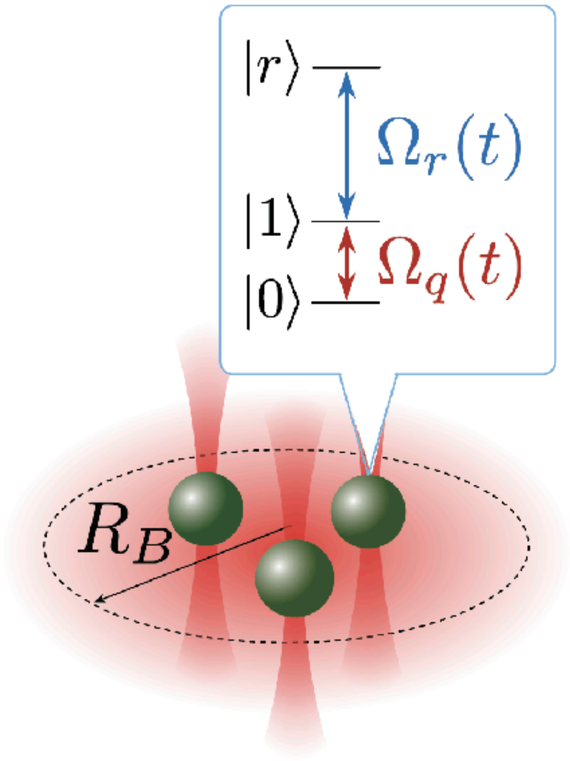
Engineer two-field pulses (using Rydberg blockade) to implement any 2S-qubit gate!

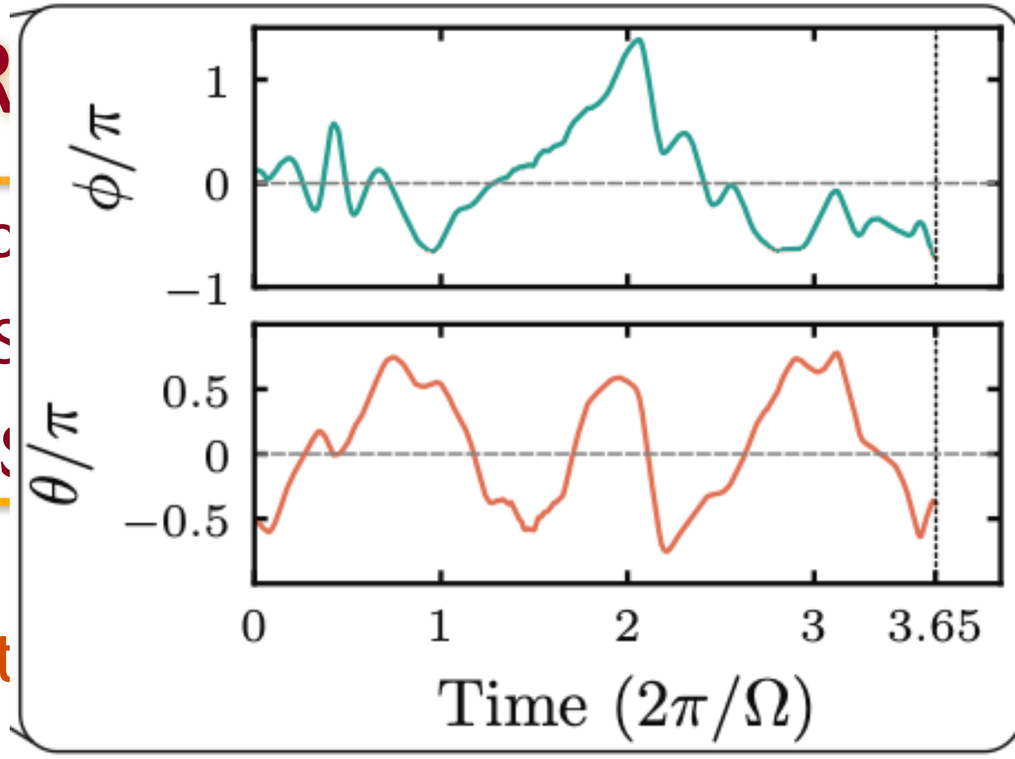
Engineering Large Spins on R

Hardware Efficient Multi-Qubit Operations with a glc

Encode spin-S variables into 2S

$\Rightarrow$ valid spin-S states: $\langle \hat{\mathbf{S}}_i^2 \rangle = S$





Two important examples:

1. Engineer all-to-all interact

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

2. Projection into symmetric subspace:

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

Realize multi-qubit gates via time-dependent

use GrAPE (Gradient Ascent Pulse Eng

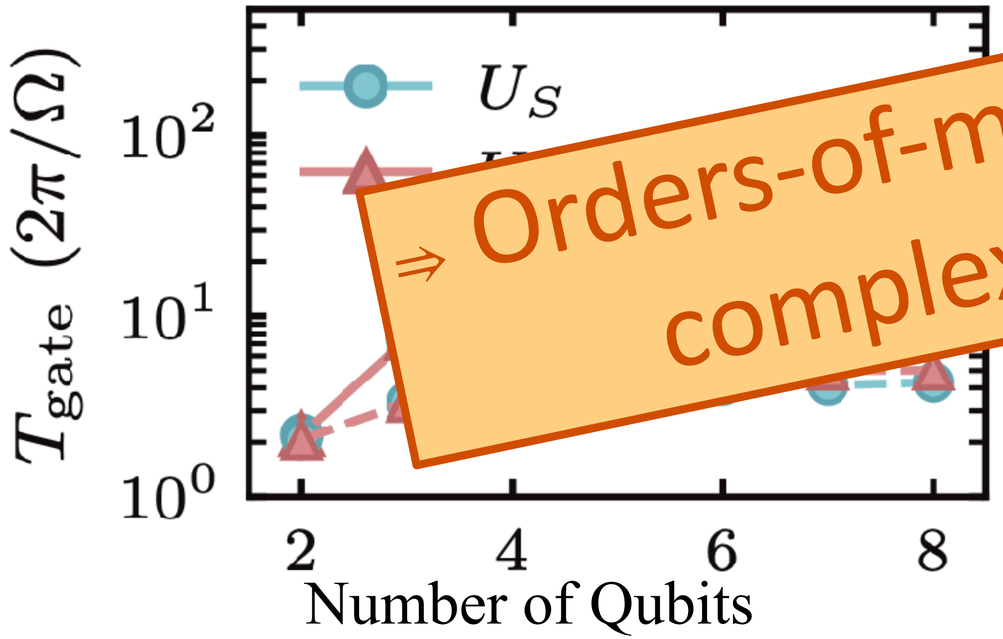
Khaneja, et al., J. Magn. Reson. **172**, 296 (2005)
Jandura et al., Quantum **6**, 712 (2022)
Evered et al., Nature **622**, 268 (2023)
Katz, et al., Nat. Phys. **19**, 1452 (2023)

Important Metric: Gate Times

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

Multi-Qubit Gates via Global Drive

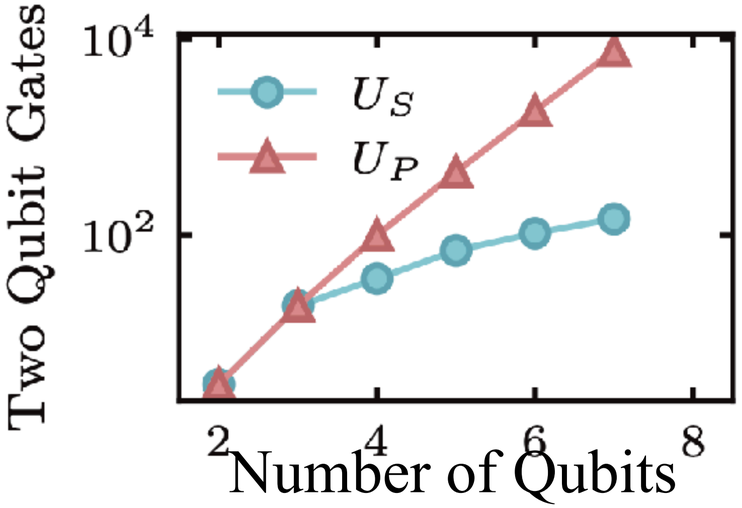
(for error $\epsilon = 10^{-3}$)



⇒ Almost no scaling with cluster size

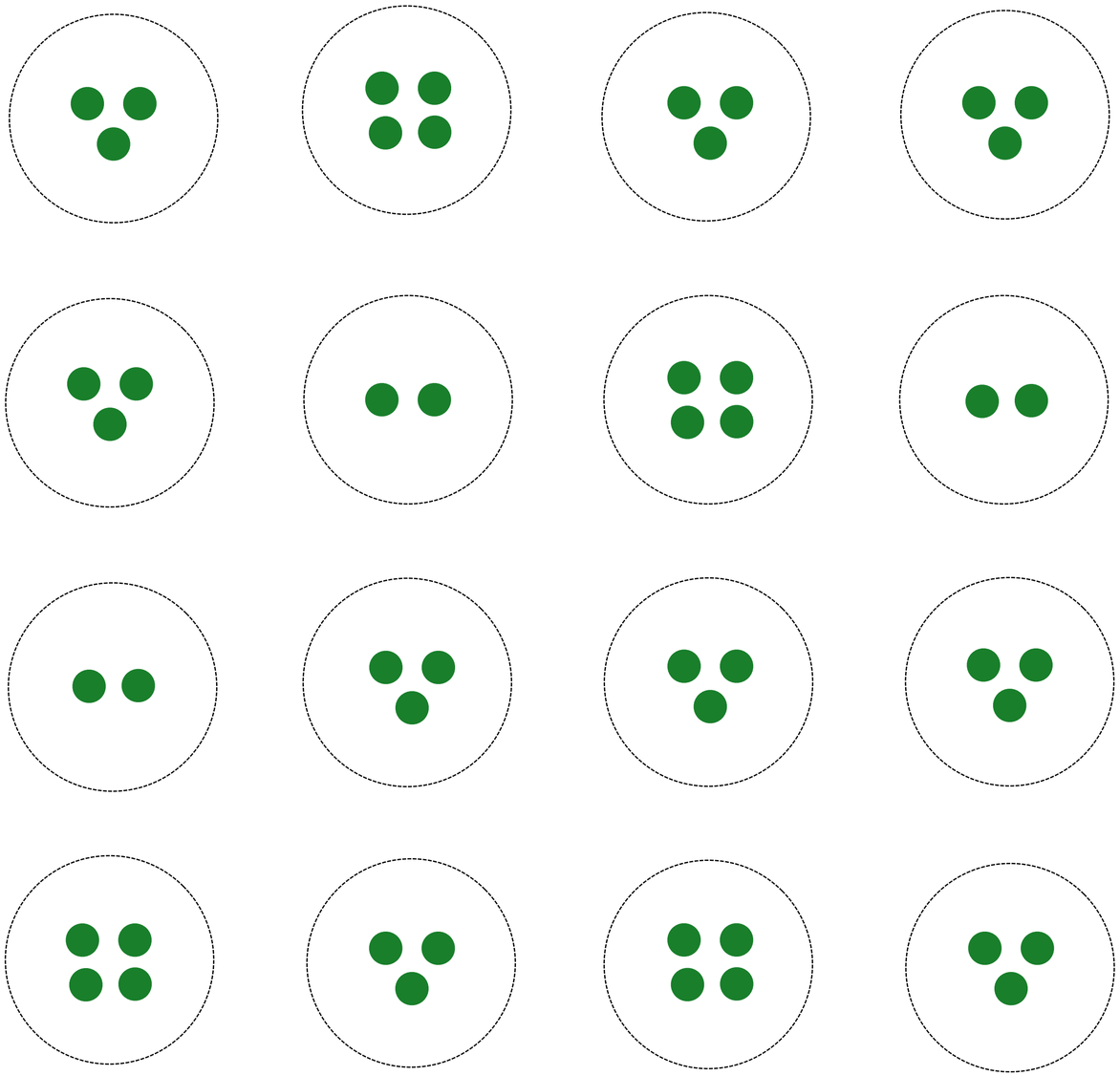
Comparison: two-qubit operations

(for error ϵ [illegible])



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

Engineering Interactions



Target Hamiltonian:

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

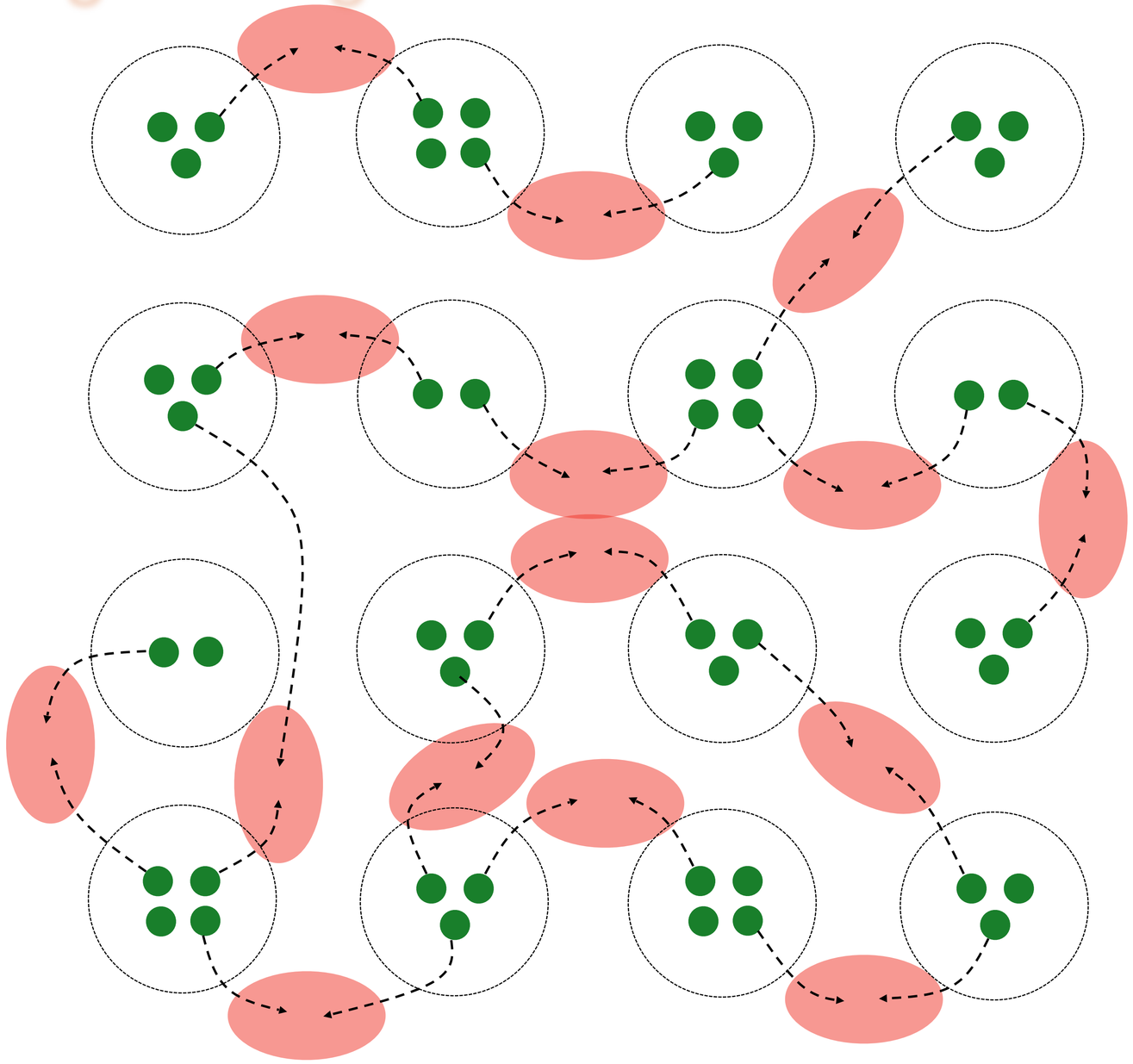
Highly programmable!

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:

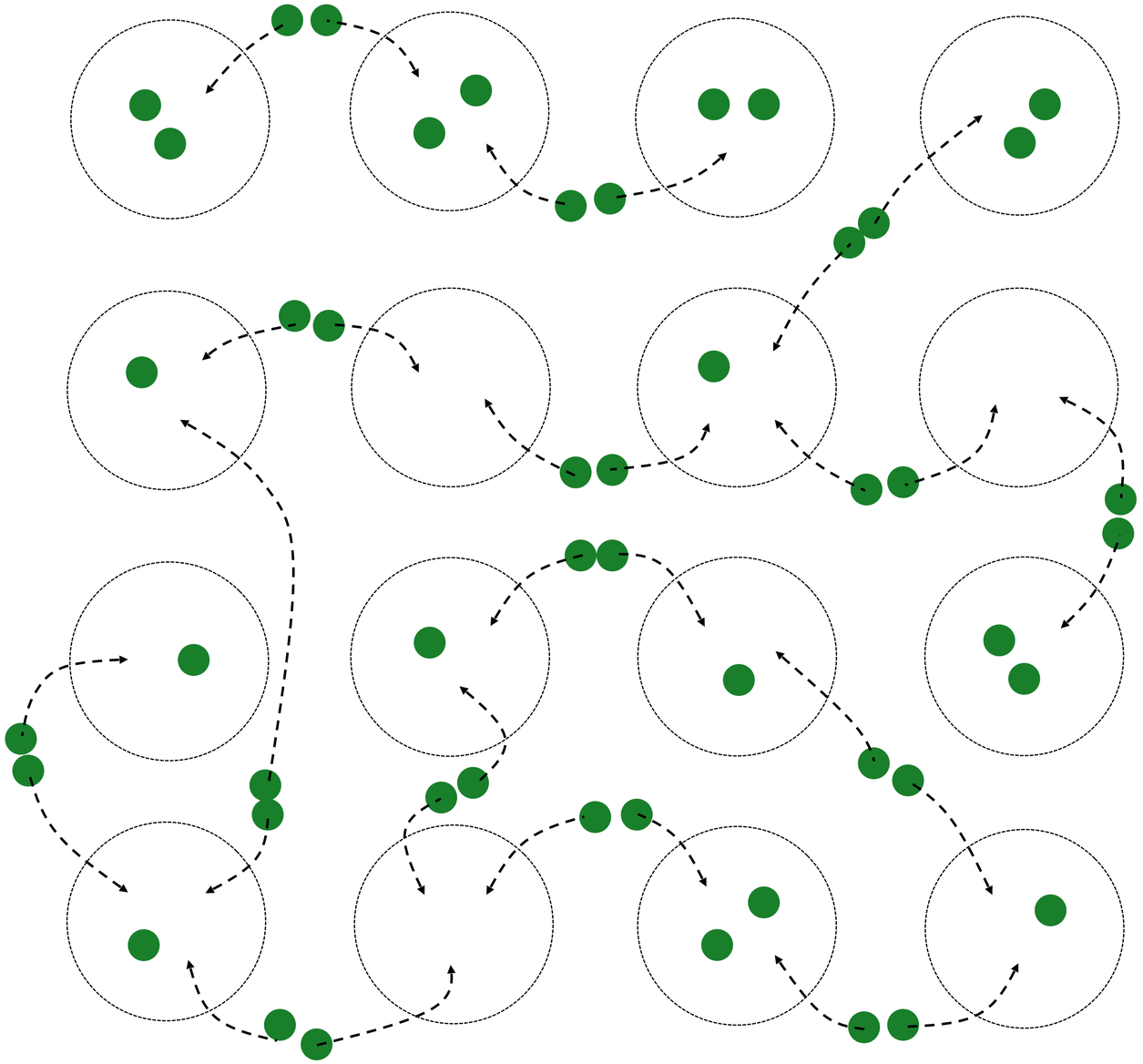
$$H_{target} = \sum_{i,j} J_{ij}^{\alpha\beta} \hat{S}_i^{\alpha} \hat{S}_j^{\beta}$$

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

Engineering Interactions



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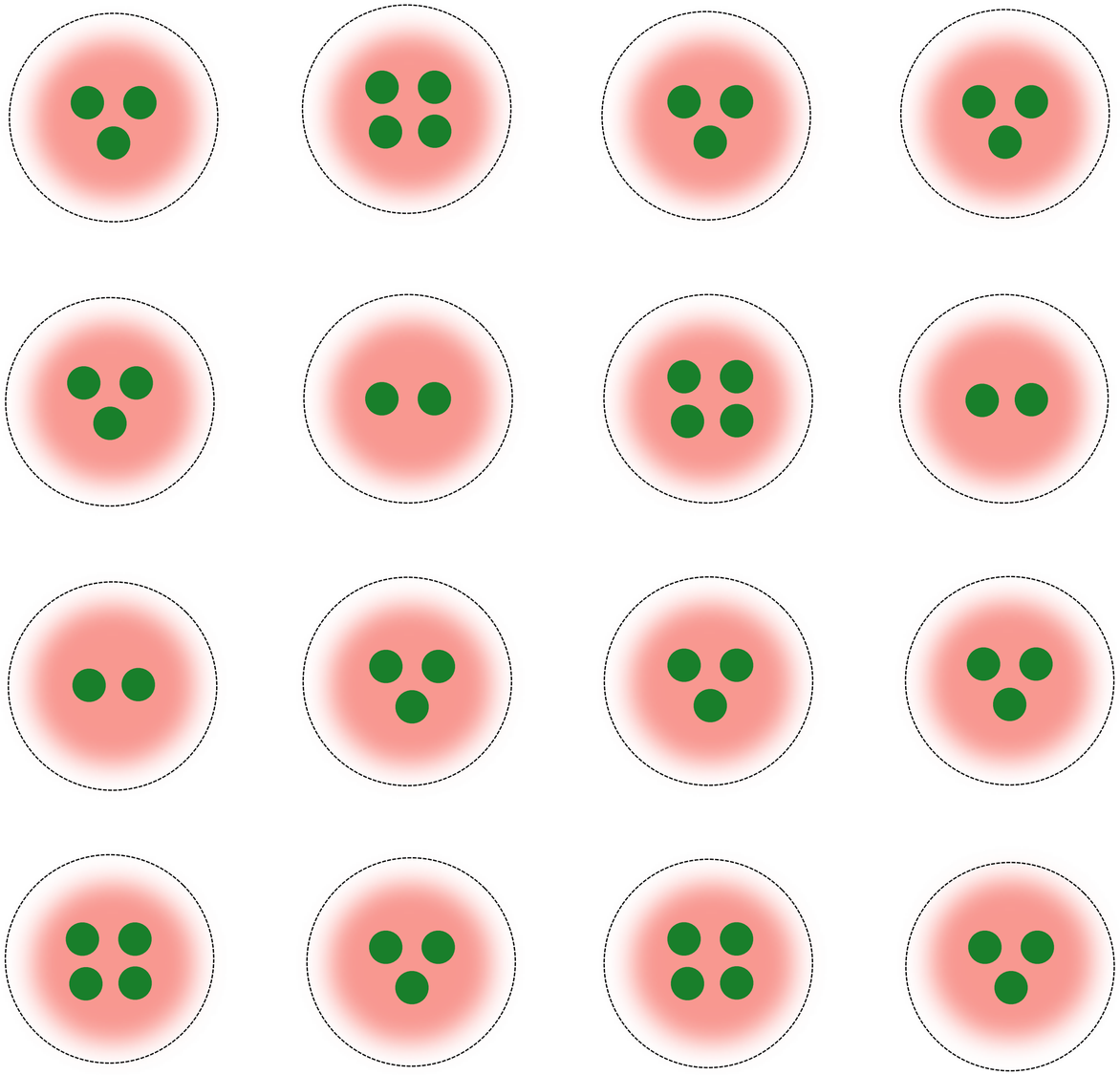
- Spin-1/2 gates and local rotations
- Mediates generic, long-range connectivity
- Violates large-spin encoding

3. Reconfigure

Engineering Interactions



Target Hamiltonian:

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

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: $$H_C = -\sum_i P_{\mathrm{sym}}[(\vec{S}_i)^2]$$

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

Floquet Sequence to Implement Model Hamiltonian

Effective evolution operator:

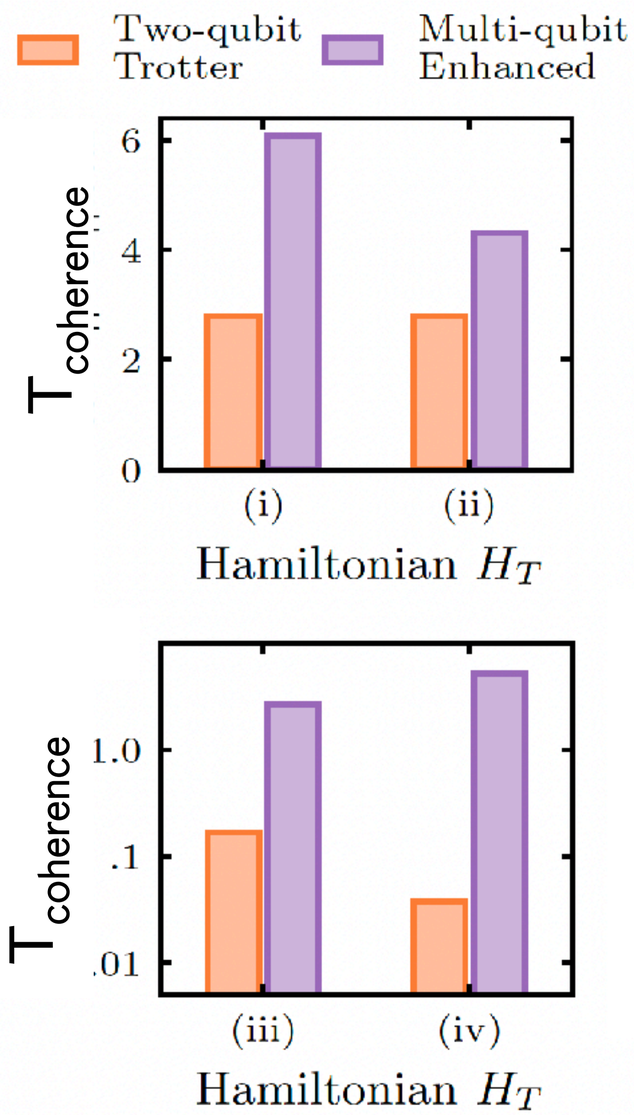
$$U_F = \prod_k e^{-i\theta_k H_C} e^{-i\tau H_I}$$

can be large-angle rotations

$\Rightarrow$ Realizes target Hamiltonian on average

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

Performance of the Approach

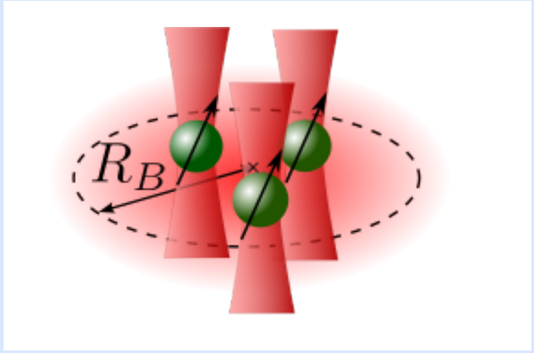


Takeaway:

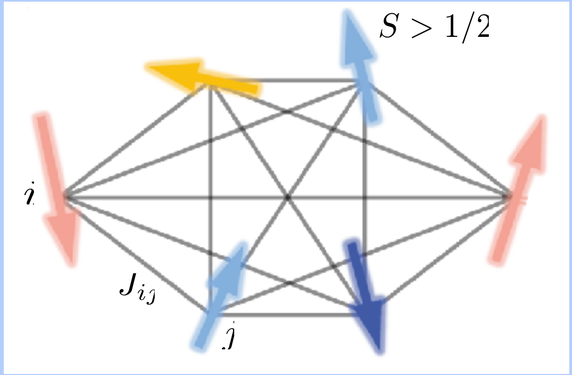
- Hardware-optimized multi-qubit operations to efficiently generate interactions
- Floquet/Hamiltonian engineering to efficiently combine operations.

Necessary Ingredients

Control high spin (S>1/2)

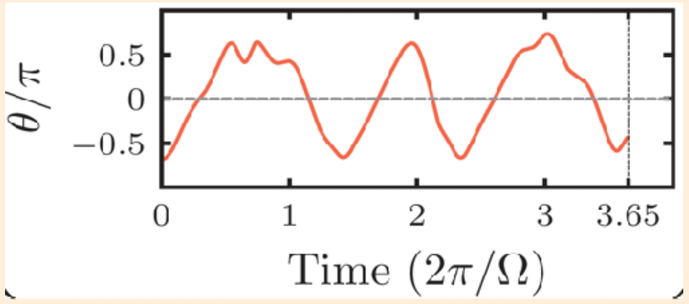


Non-local connectivity

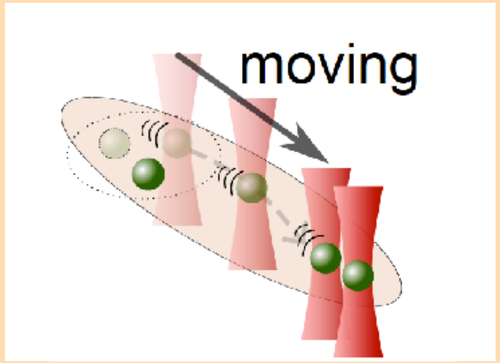


Solution read-out

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?

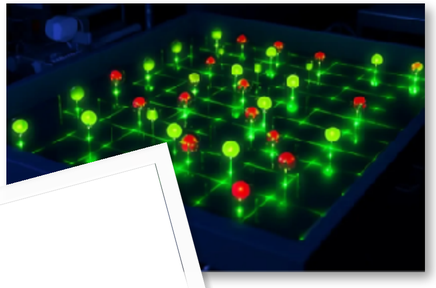
peaks at state energies

Such measurements are ve… den…

Efficient readout: Quantum-Classical Co-Processing

Hardware-efficient toolbox to compute spectral functions

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



$$\int \quad \mathrm{Corr}_{\mathrm{classical}} \; \mathrm{Corr}_{\mathrm{quantum}}$$

evolution times

stat

o

time evol

kno

superposition state

D^A

$O_{s,R}(0)e^{-iE_S t}$

$C_{O_s,R}(t) + O(1/\sqrt{M})$

Makes procedure sample efficient:

D_A can be found for different A with same sample set

Uniformly sample perturbe

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

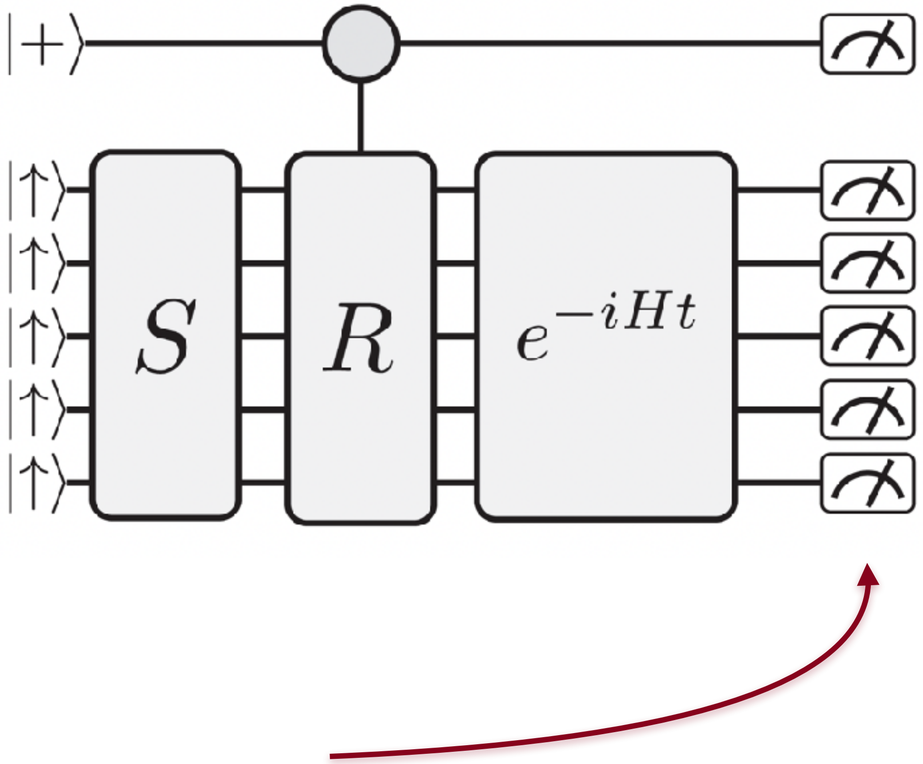
Sum over observables

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

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

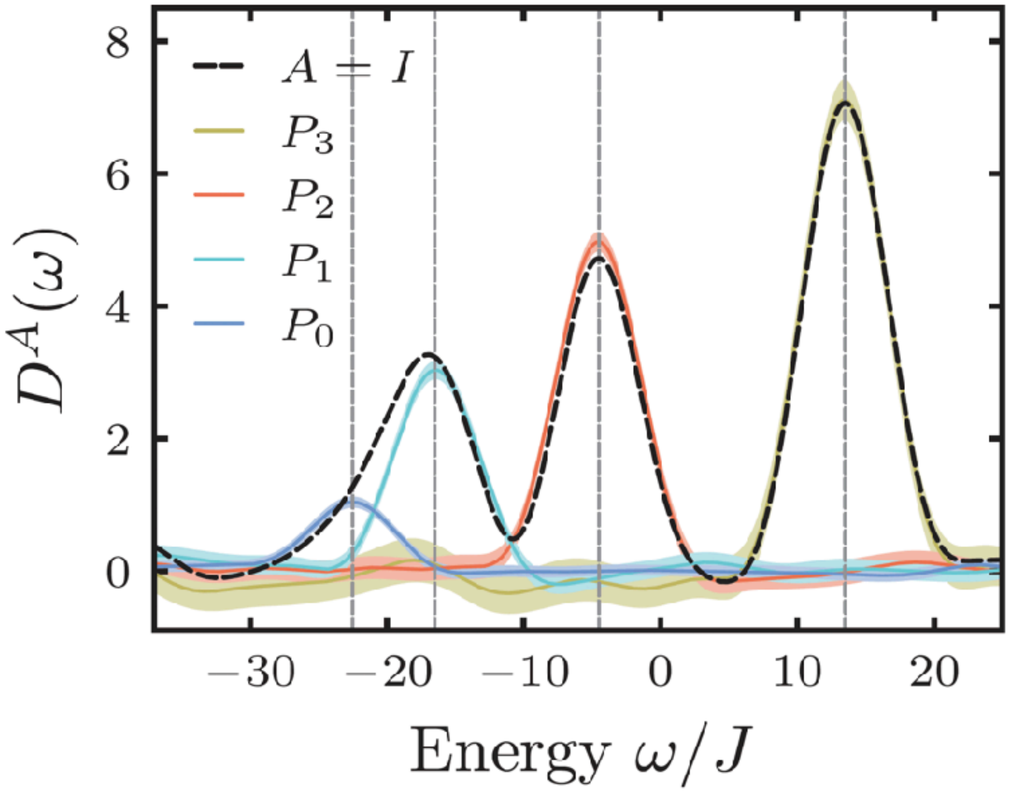
Efficient readout: Quantum Circuit + Spectrum



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

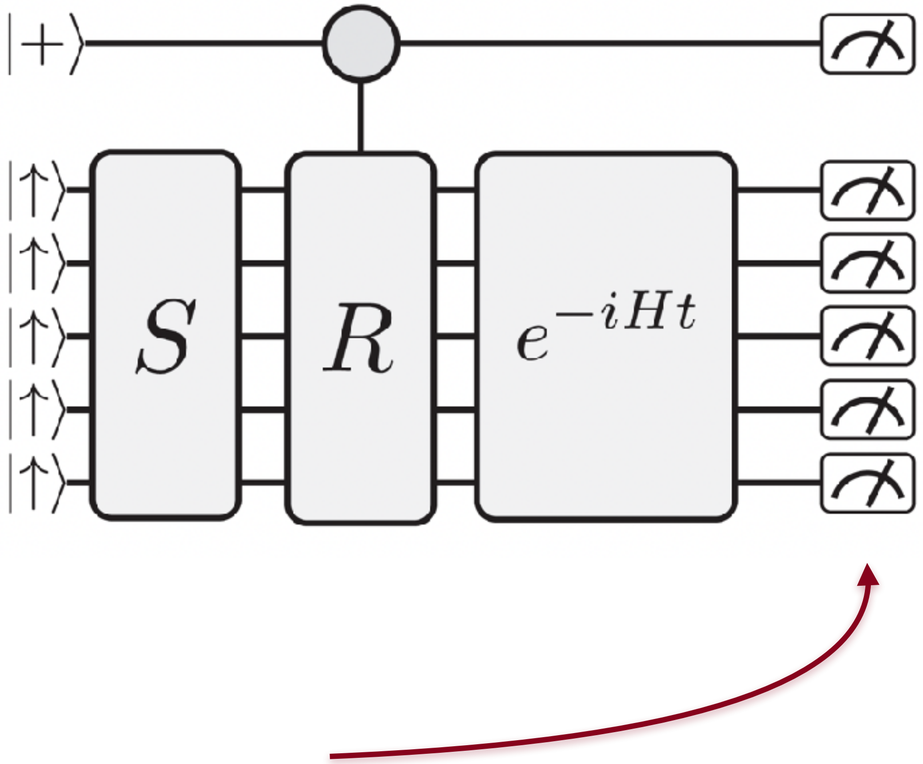
Example: two interacting spin-3/2s

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



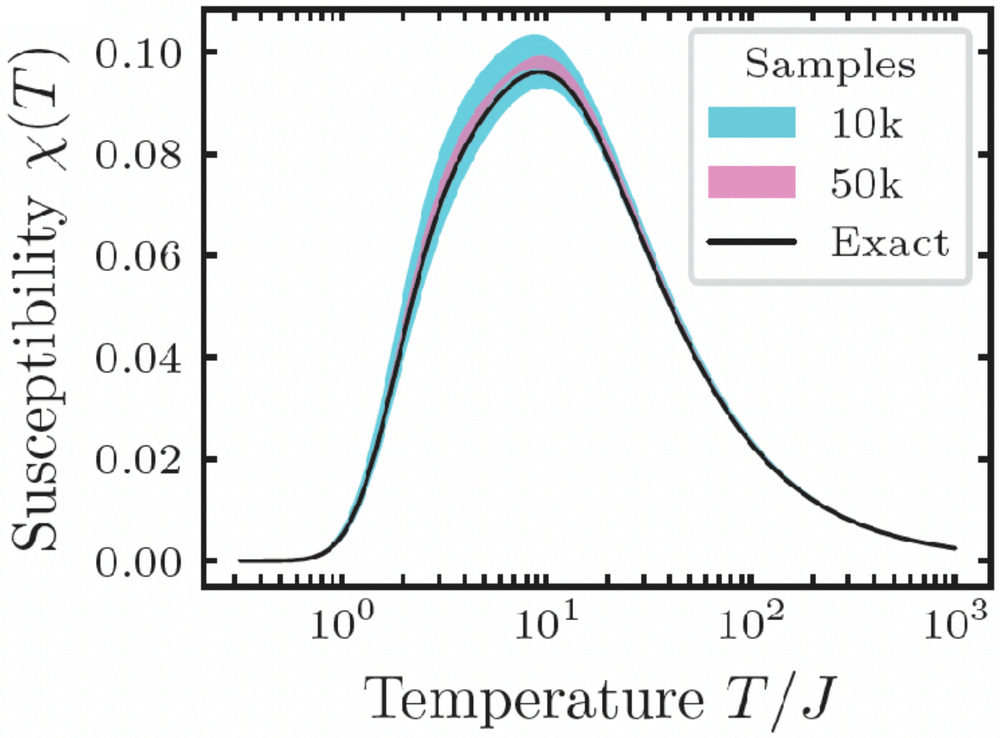
20,000 samples

Efficient readout: Quantum Circuit + Spectrum

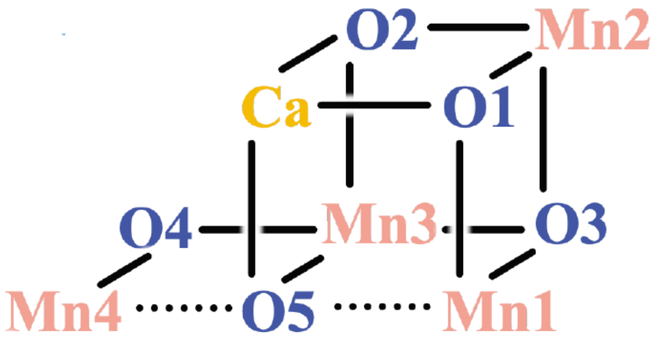


Parallel measurement of 2^n observables
(any operator diagonal in measurement basis)

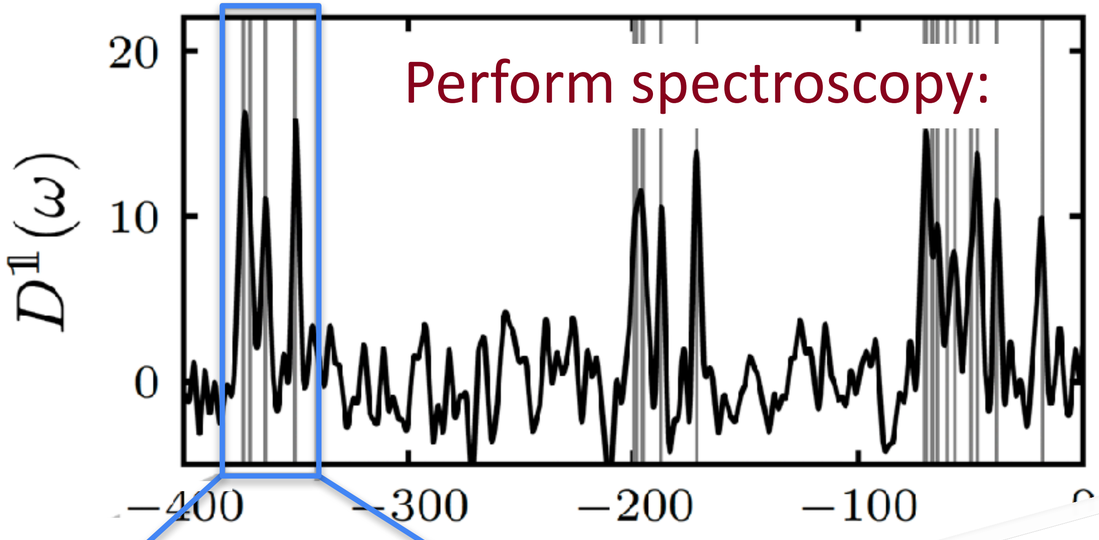
Access to full spectral information
⇒ finite temperature properties!



Benchmark on Larger Molecule (OEC Complex)

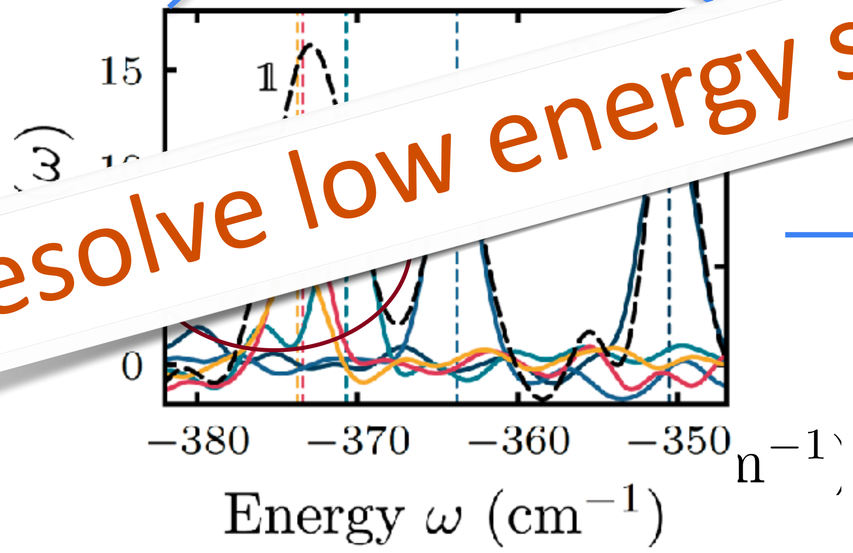


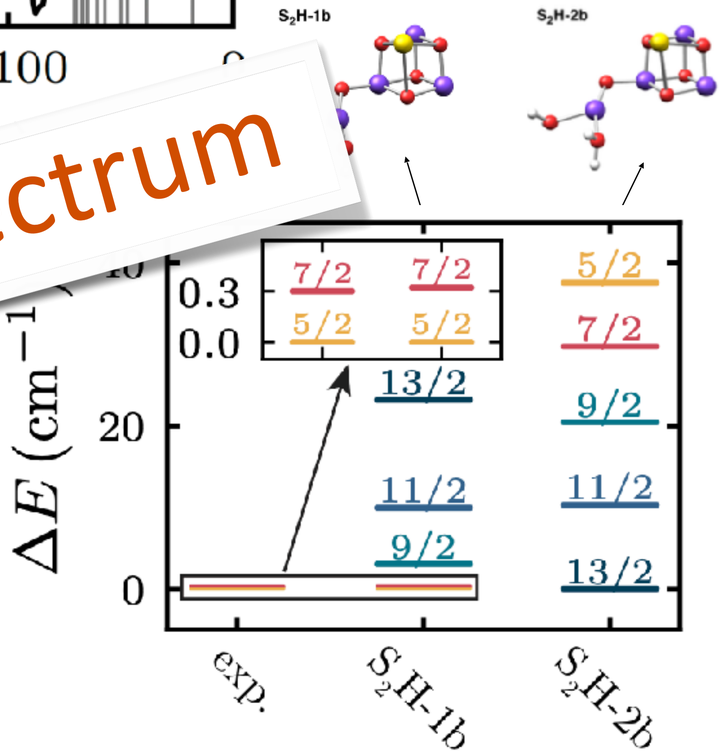
Perform spectroscopy:



resolve low energy spectrum

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



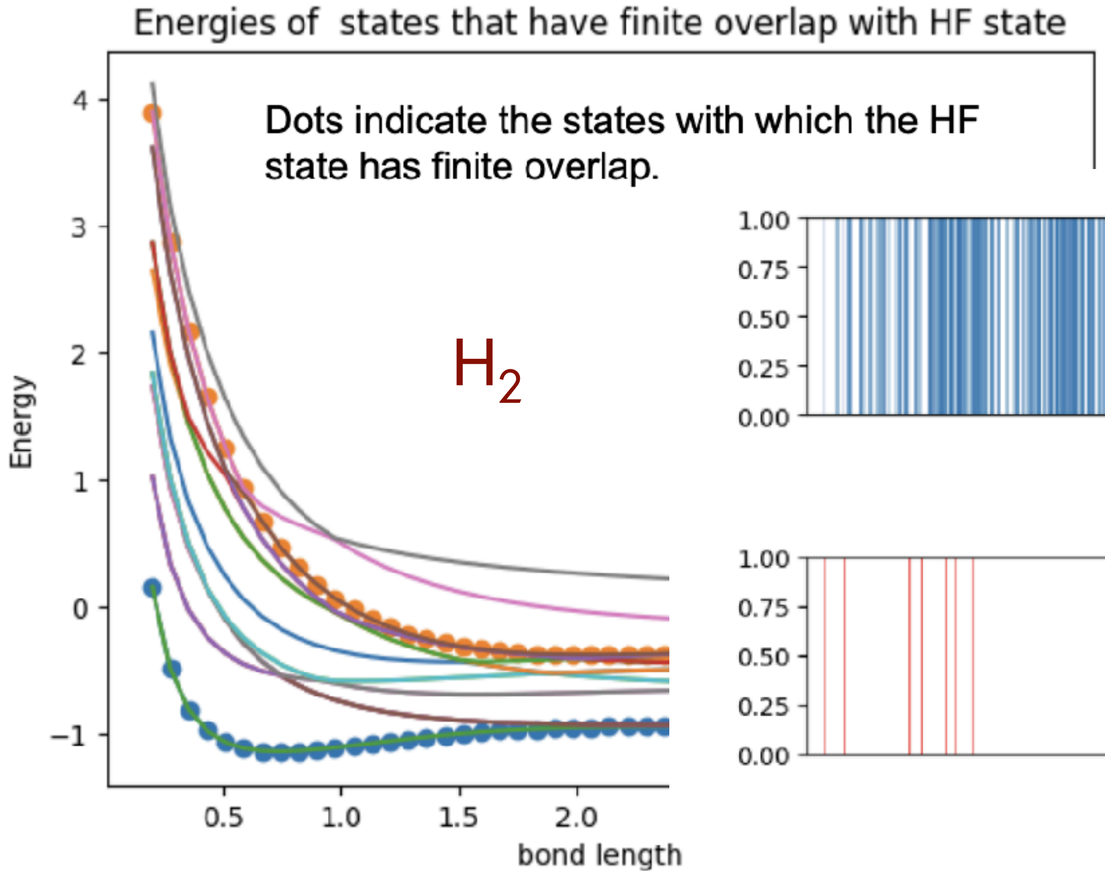


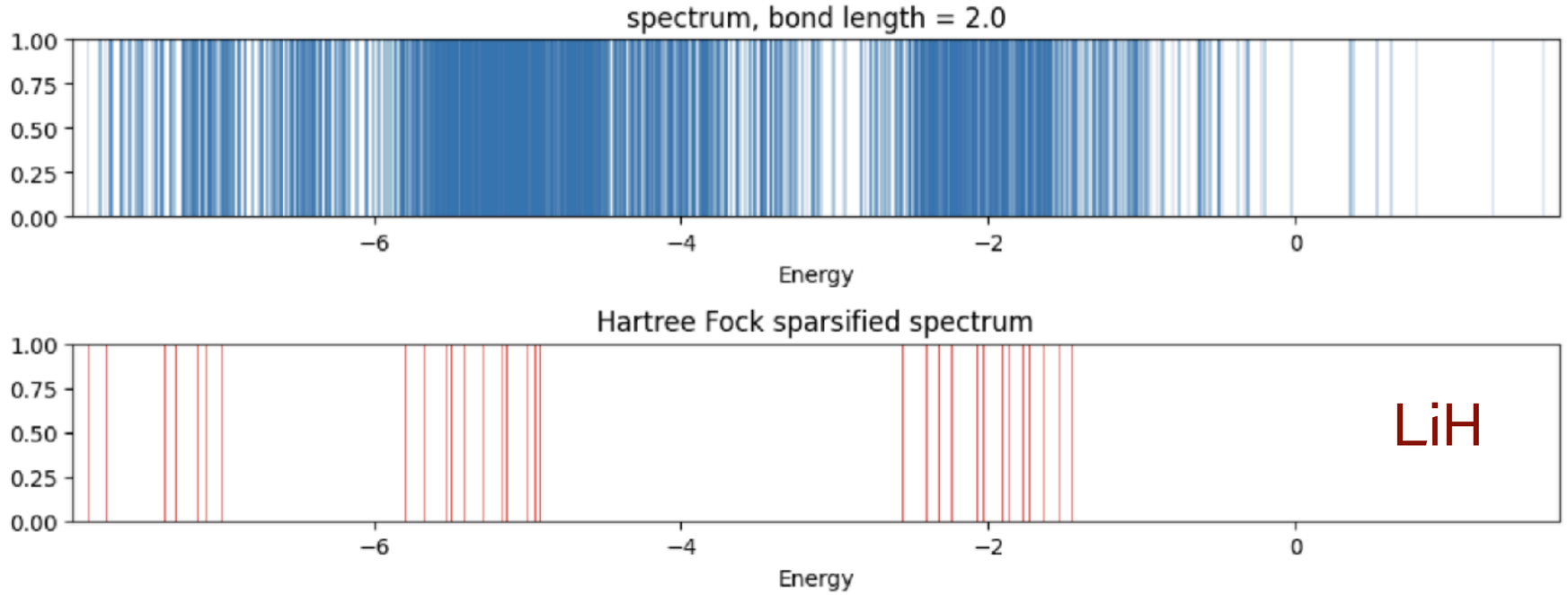
Newer results

Access to full spectrum + matrix elements of operators = perturbation theory

⇒ enhance accuracy of energy estimates for approximated Hamiltonians

⇒ Time evolving fermionic Hamiltonians for small molecules

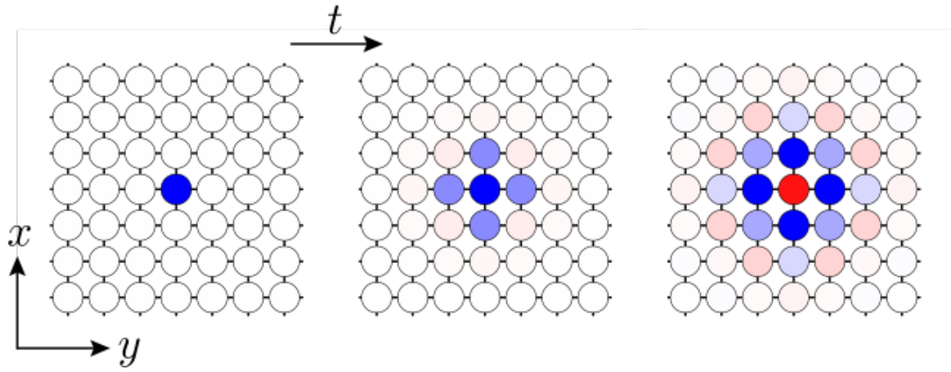




Additional Applications and Outlook

Application to 2D magnetic materials

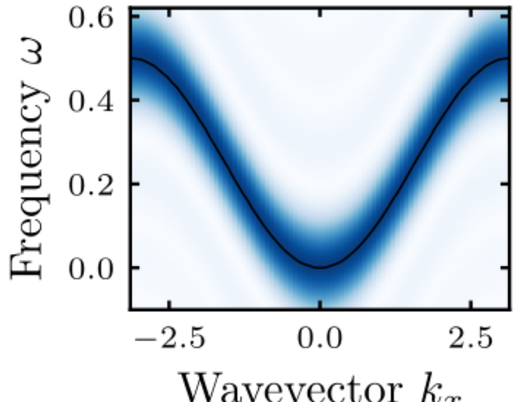
Single-particle Green's function of FM Heisenberg:

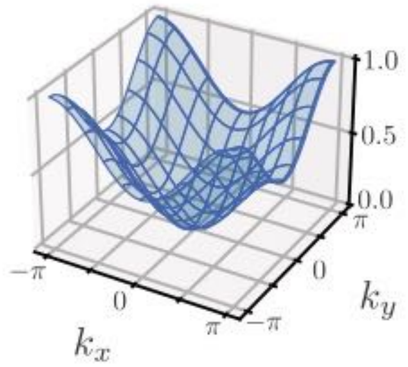


Quasi-particle properties encoded in spectral function

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

Dispersion $\epsilon(k)$

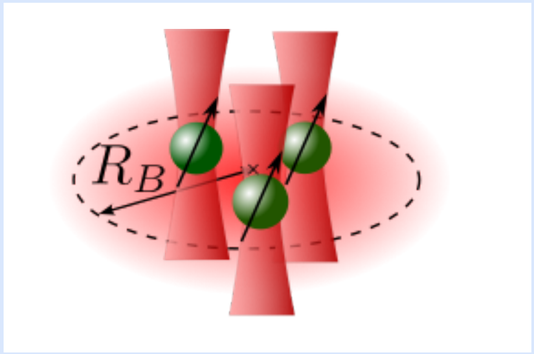




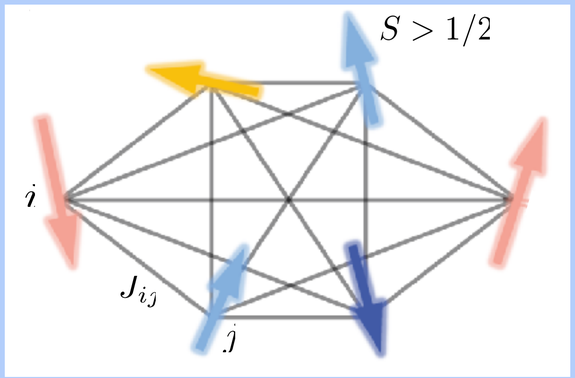
Next steps:

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

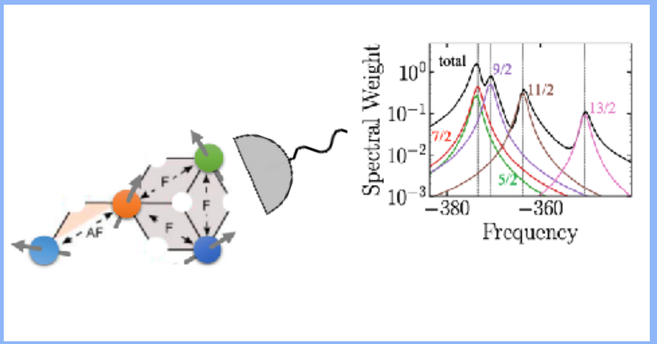
Control high spin (S>1/2)



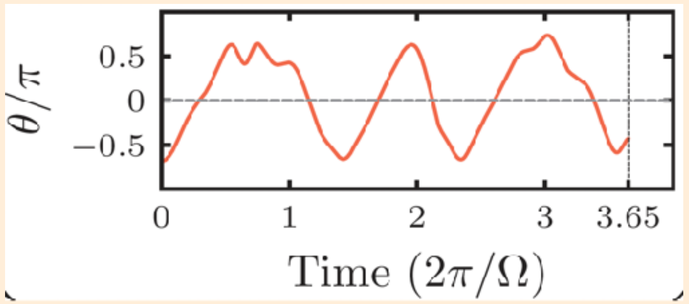
Non-local connectivity



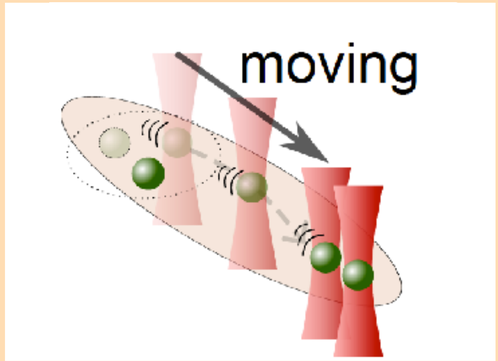
Solution read-out



Native multi-qubit gates

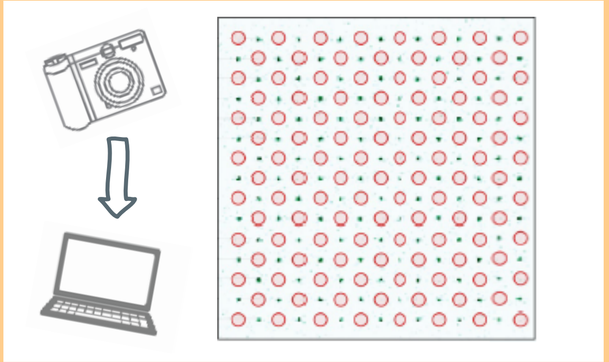


Atom moving



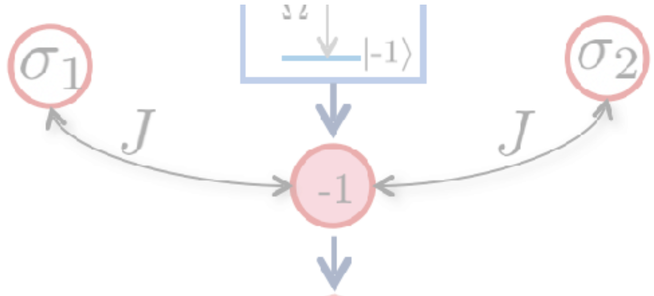
D. Bluvstein et al., Nature **604**, 451–456 (2022)

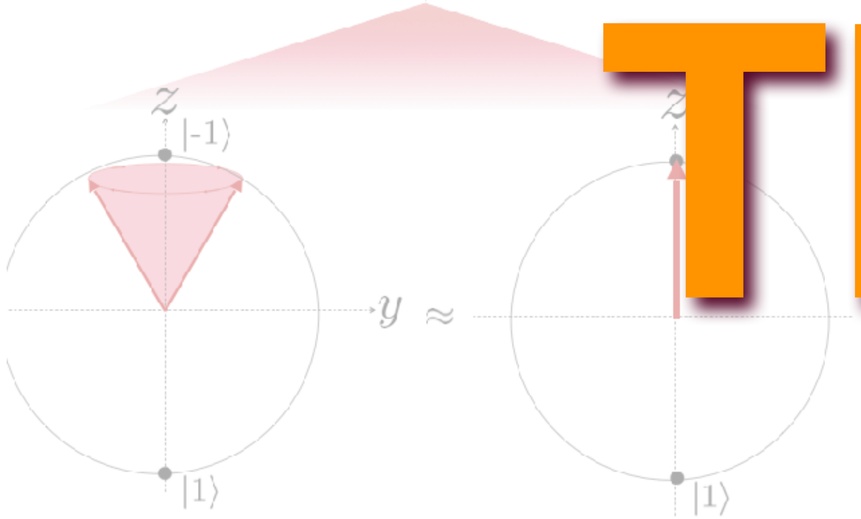
Co-processing



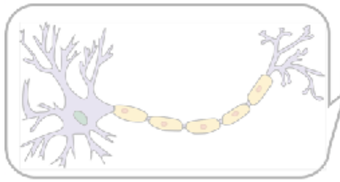
H.-Y. Huang et al., Nat. Phys. 16, 1050–1057 (2020)

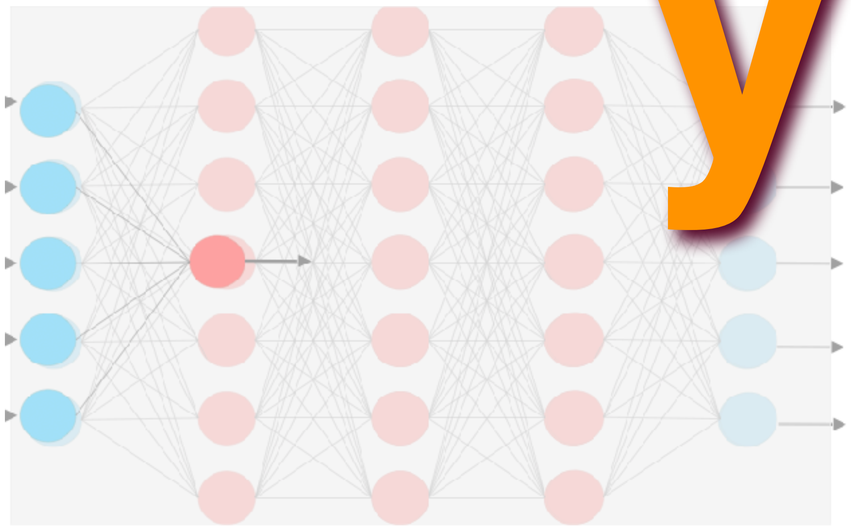
Maskara, Ostermann, Shee, Kalinowski, McClain Gomez, Araiza Bravo, Wang, Krylov, Yao, Head-Gordon, Lukin, Yelin, arXiv:2312.02265





Thank you!





Energy measurement and entanglement detection