Hamiltonian learning quantum magnets with scanning tunnel microscopy

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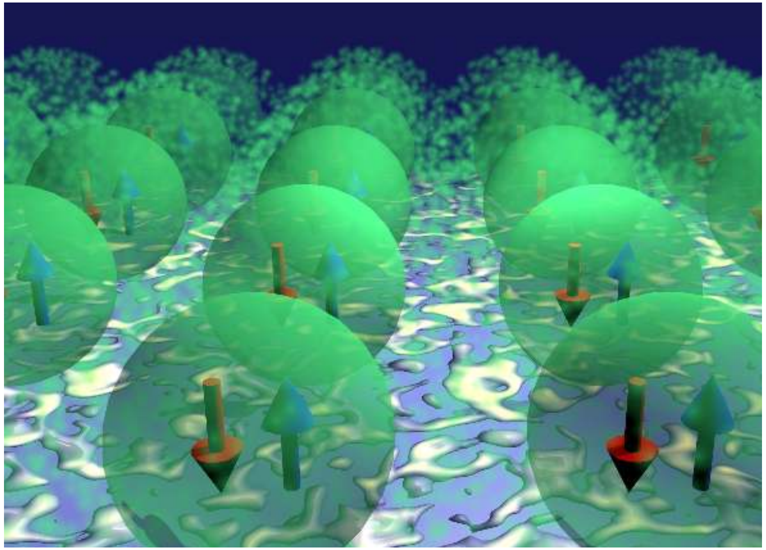
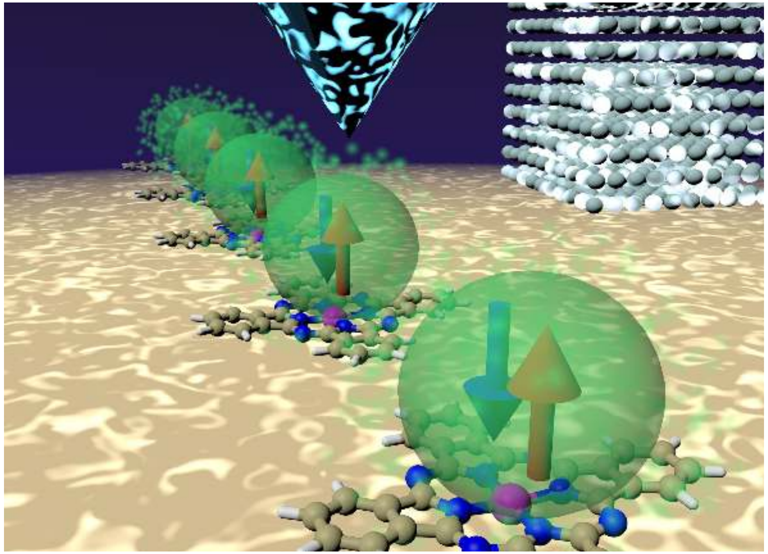


Phys. Rev. Lett. 131, 086701 (2023)



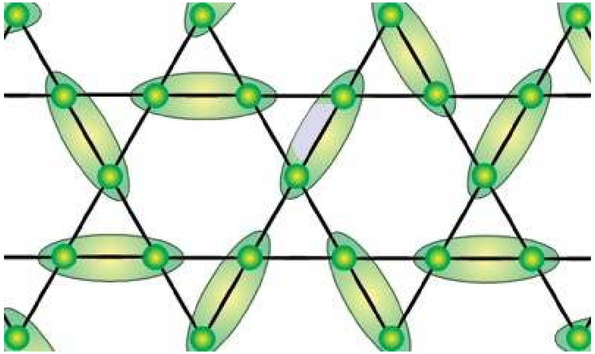
arXiv:2504.20711 (2025)

Low-Temperature Quantum Detectors, Helsinki

August 6th 2025

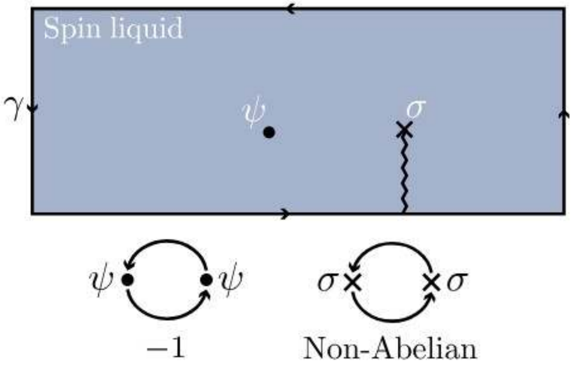
Quantum spin liquid quantum materials: opportunity and challenge

Macroscopically entangled magnetic state



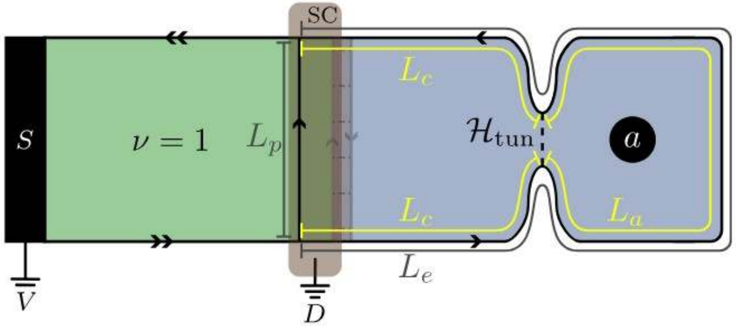
Rep. Prog. Phys. 80, 016502 (2017)

Featuring fractional excitations



Phys. Rev. X 10, 031014 (2020)

Potential candidates for topological quantum computing



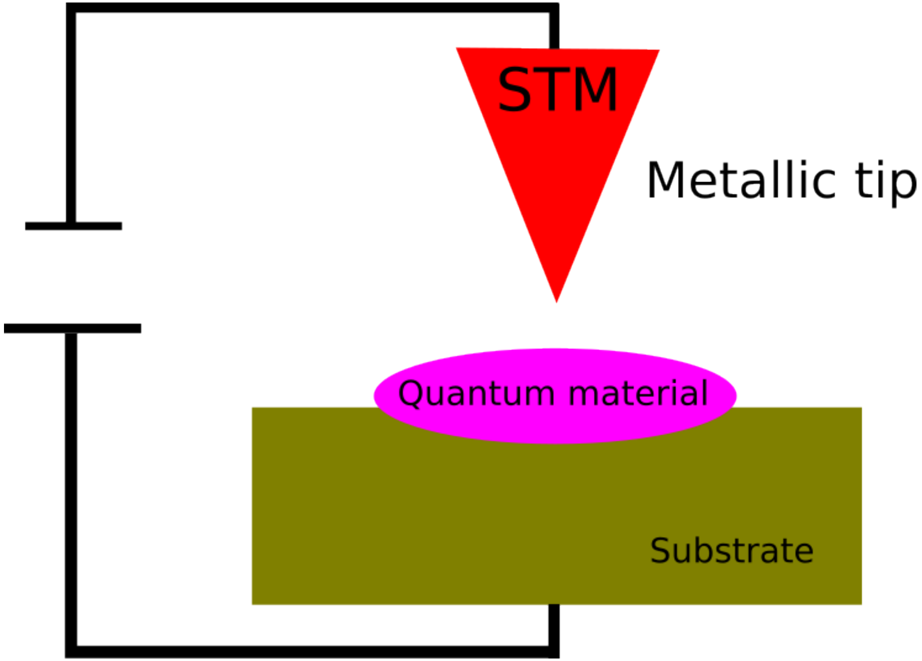
Phys. Rev. X 12, 011034 (2022)

Many potential QSL candidates (Herbertsmithite, 1T-TaS_2, RuCl_3)

<u>**However, quantum spin liquids are exceptionally challenging to identify experimentally**</u>

How can we learn the microscopic description of quantum magnets?

Sensing atomic-scale quantum matter with scanning tunneling microscopy



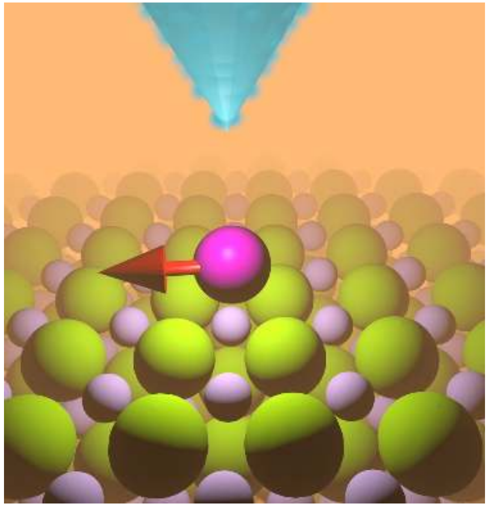
Atomic real-space resolution
Energy/frequency and time resolution

- Electronic states
- Phonons
- Superconducting pairing
- Orbital order
- Spin excitations

Rev. Mod. Phys. 91, 041001 (2019)
Nat. Rev. Materials 9, 460–480 (2024)

Probing quantum matter with scanning tunneling microscopy

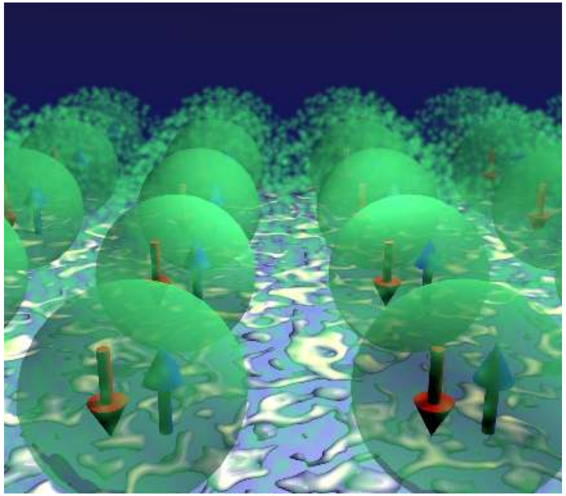
Atomic lattices



a = 0.5 nm

Nature Materials 13, 782–785 (2014)
Rev. Mod. Phys. 91, 041001 (2019)

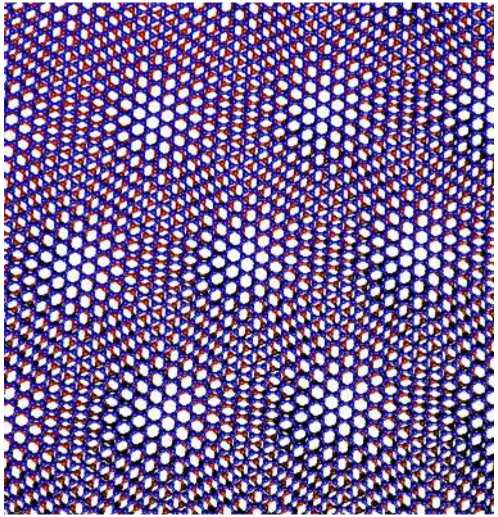
Molecular lattices



a = 3 nm

Nature Rev. Mat. 5 (2), 87-104 (2020)
Nature 598, 287–292 (2021)

Twisted 2D moire materials

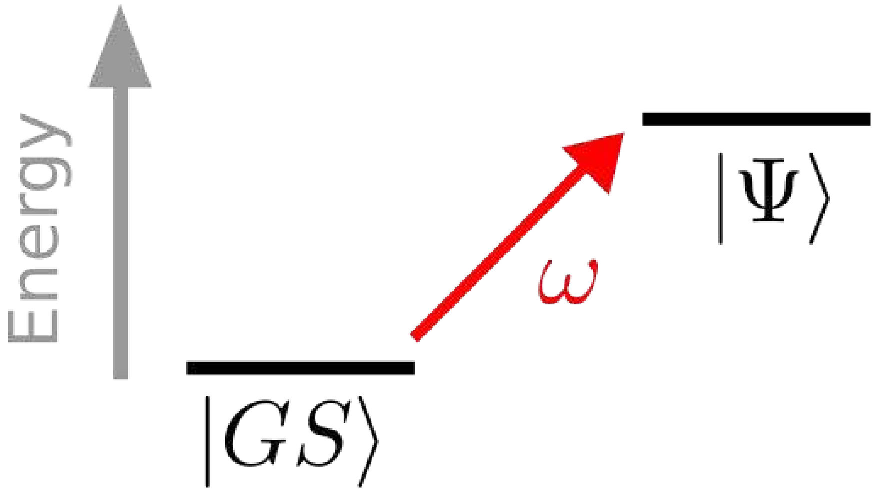


a = 30 nm

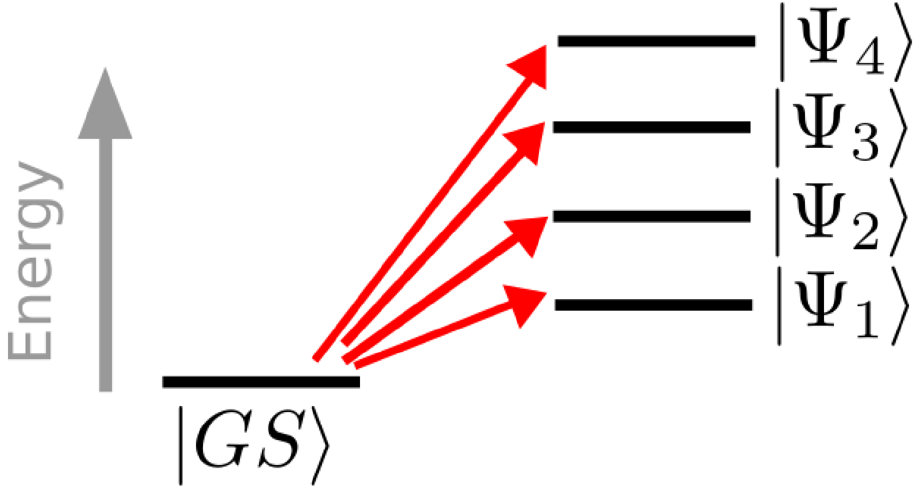
Nature 600, 240-245 (2021)
Nat. Rev. Materials 9, 460–480 (2024)

Spin excitations in collective quantum matter

Minimal quantum spin system



Complex quantum many-body system



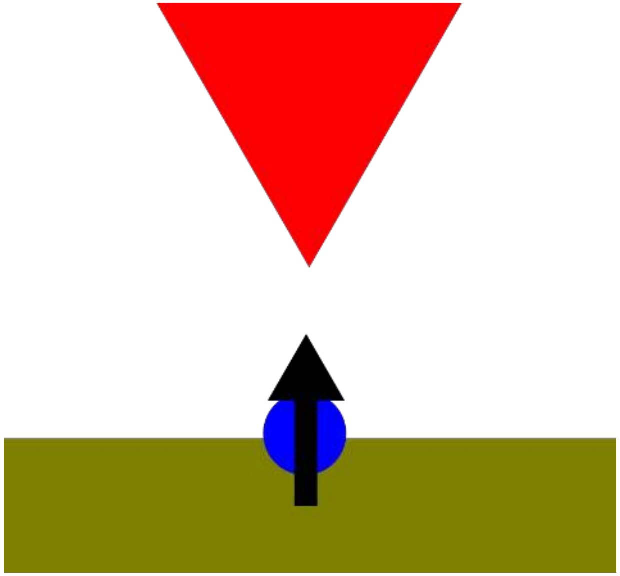
Spin excitations in quantum materials are described by the dynamical spin correlator

$$A(\mathbf{r},\omega) = \sum_\alpha \langle GS|S^\alpha_{\mathbf{r}}\delta(\omega - H + E_{GS})S^\alpha_{\mathbf{r}}|GS\rangle$$

That can be measured with scanning tunneling microscopy

Two ways of measuring spin-excitations in atomic-scale magnets

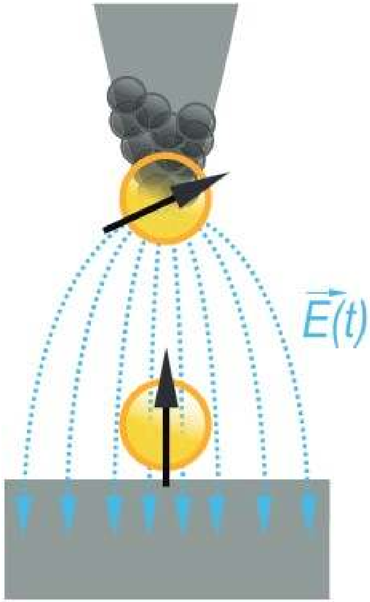
Inelastic electron tunnelling Spectroscopy (IETS)



Science 306 (5695), 466-469 (2004)

Without spin-polarized tip

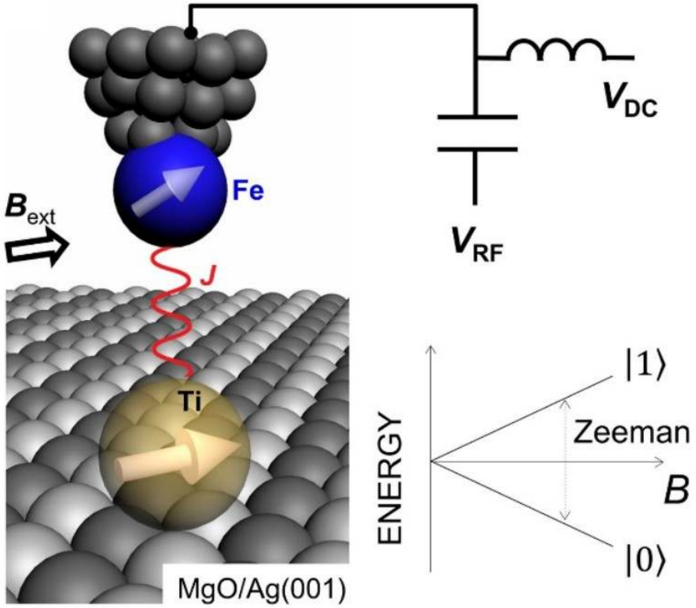
Electrically driven electron spin resonance (ESR)



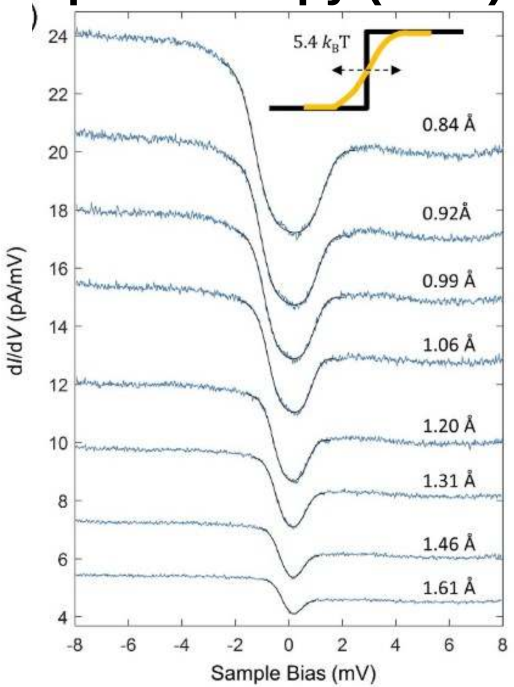
Science 350 (6259), 417-420 (2015)

With spin-polarized tip

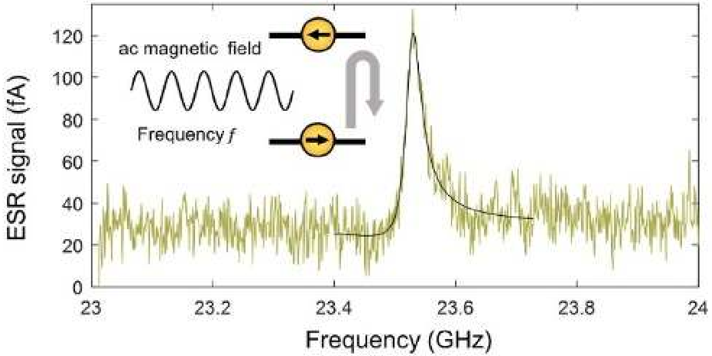
Measuring spin excitations with IETS and ESR



Inelastic electron tunneling spectroscopy (IETS)



Electron spin resonance (ESR) with STM

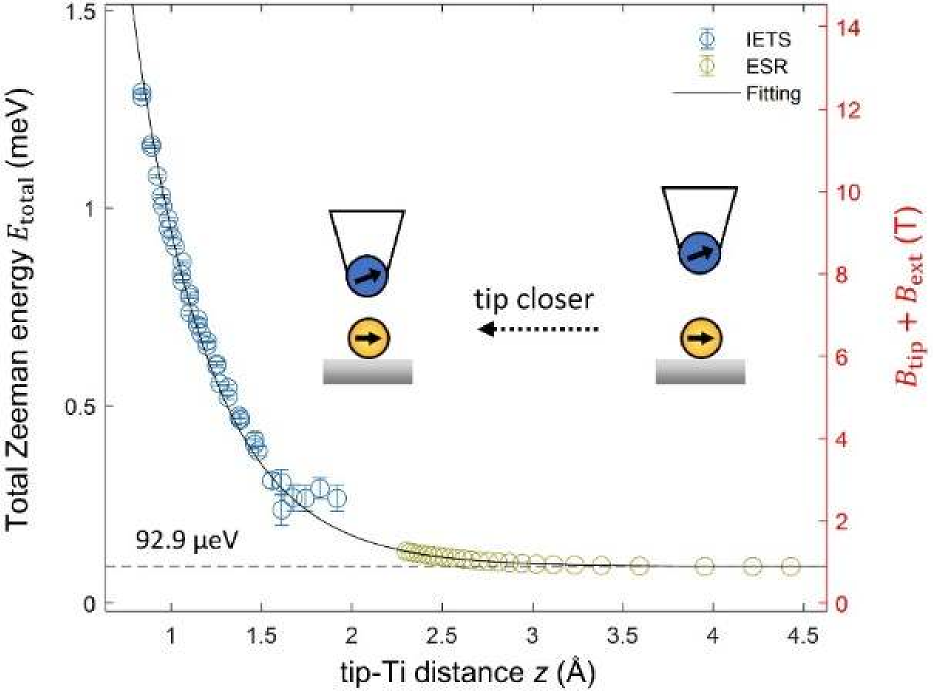


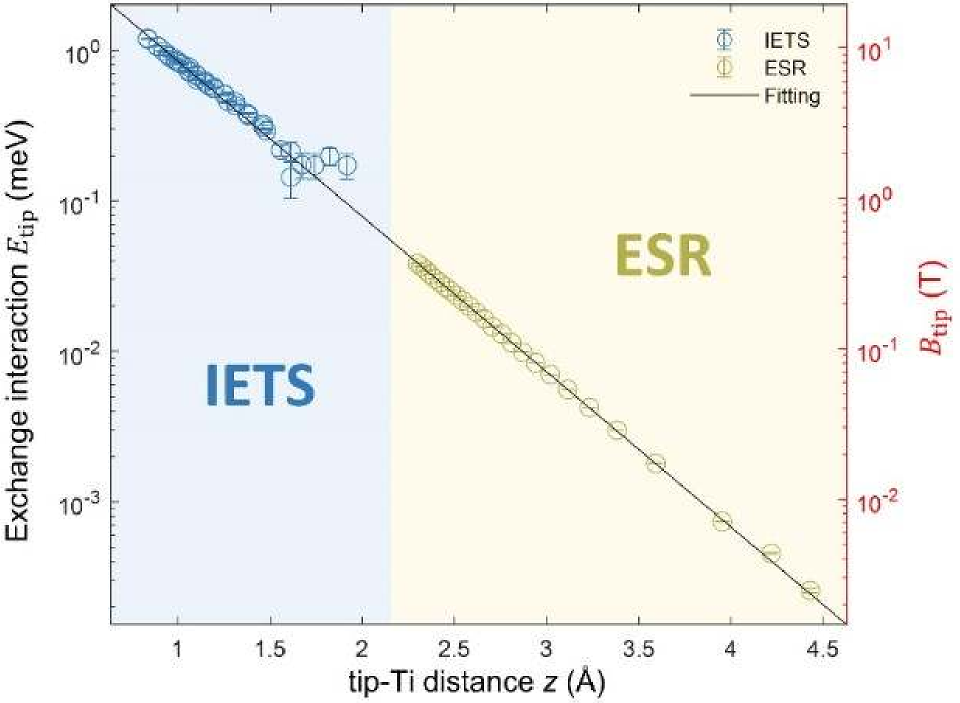
Spin excitations can probed with IETS or ESR

Phys. Rev. Lett. 122, 227203 (2019)

Measuring spin excitations with IETS and ESR

Excitation energies ranging 3 orders of magnitude can be measured with STM

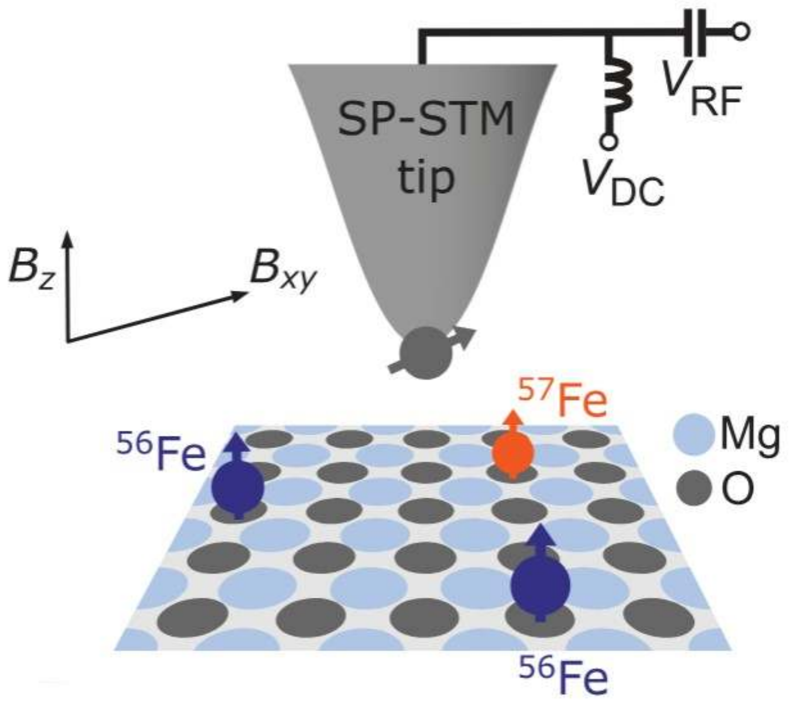




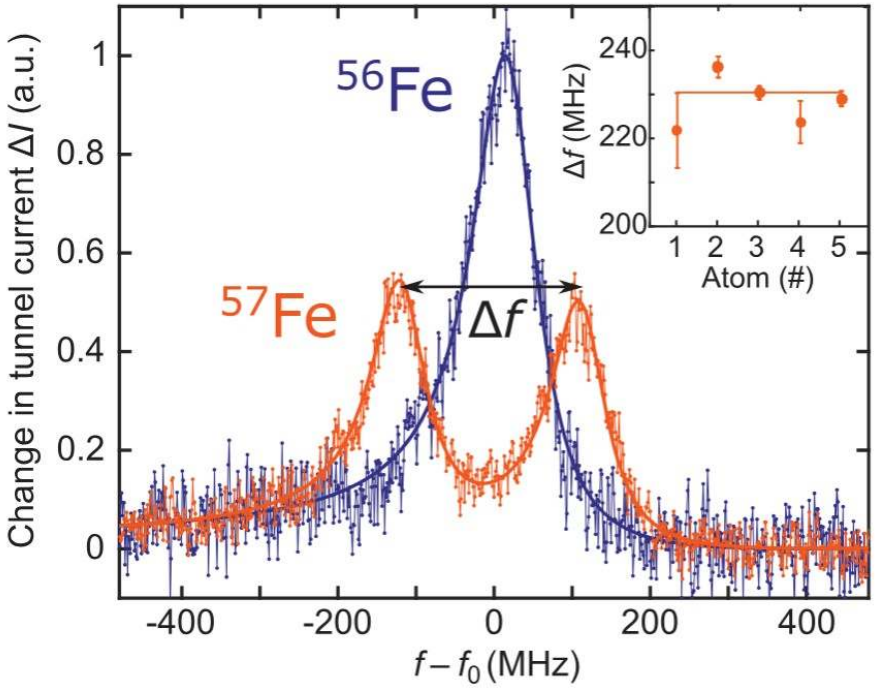
Phys. Rev. Lett. 122, 227203 (2019)

Hyperfine coupling in individual atoms with STM

Individually addressing each isotope



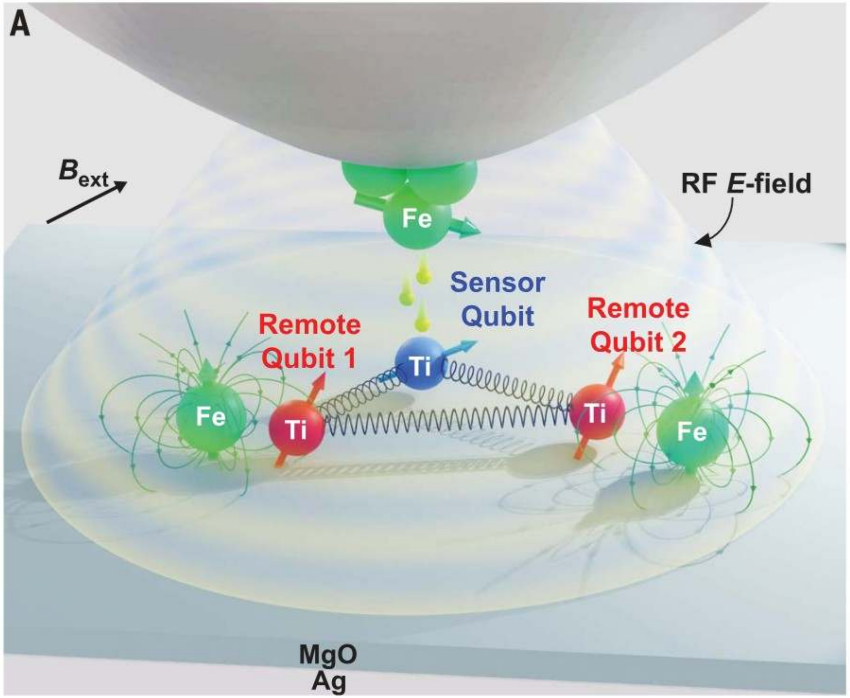
Hyperfine splitting in ESR



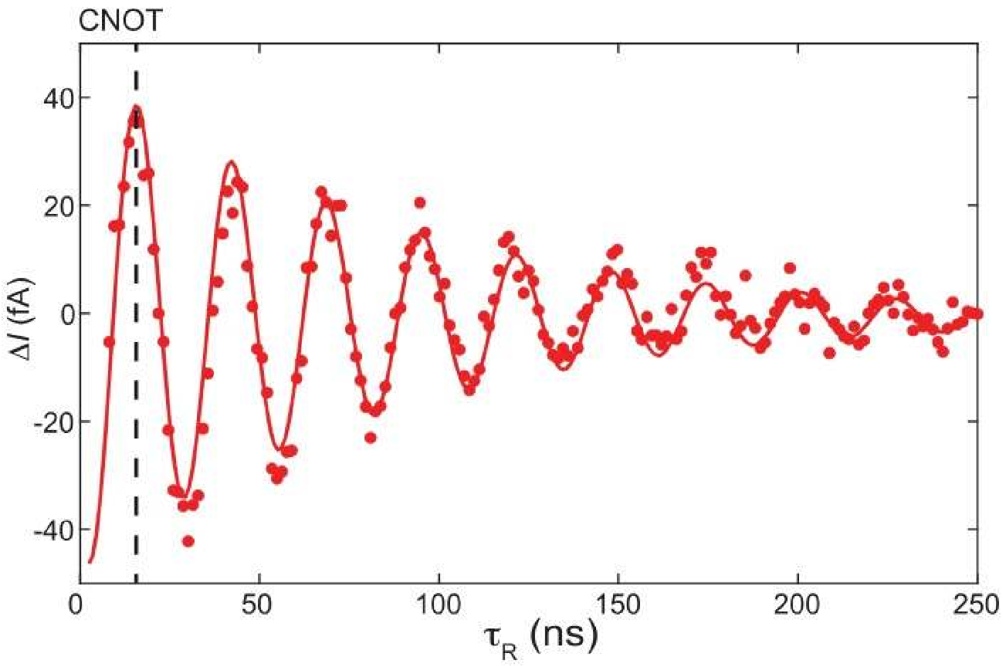
Science 362 (6412), 336-339 (2018)

Qubit operations between individual atoms with STM

Atomic multiqubit platform



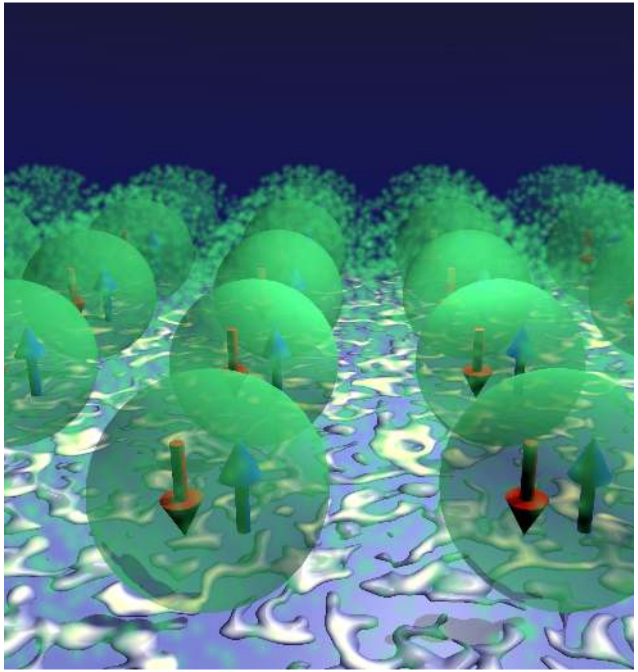
Conditional controlled qubit rotation



Science 382 (6666), 87-92 (2023)

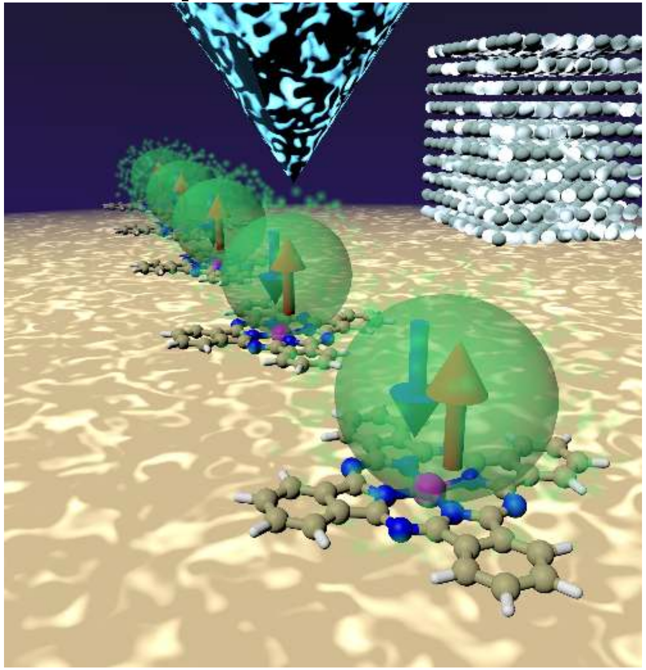
Engineering and Hamiltonian learning of an artificial quantum magnet

A triplon molecular quantum magnet



Phys. Rev. Lett. 131, 086701 (2023)

Hamiltonian learning from spin excitation



arXiv:2504.20711 (2025)

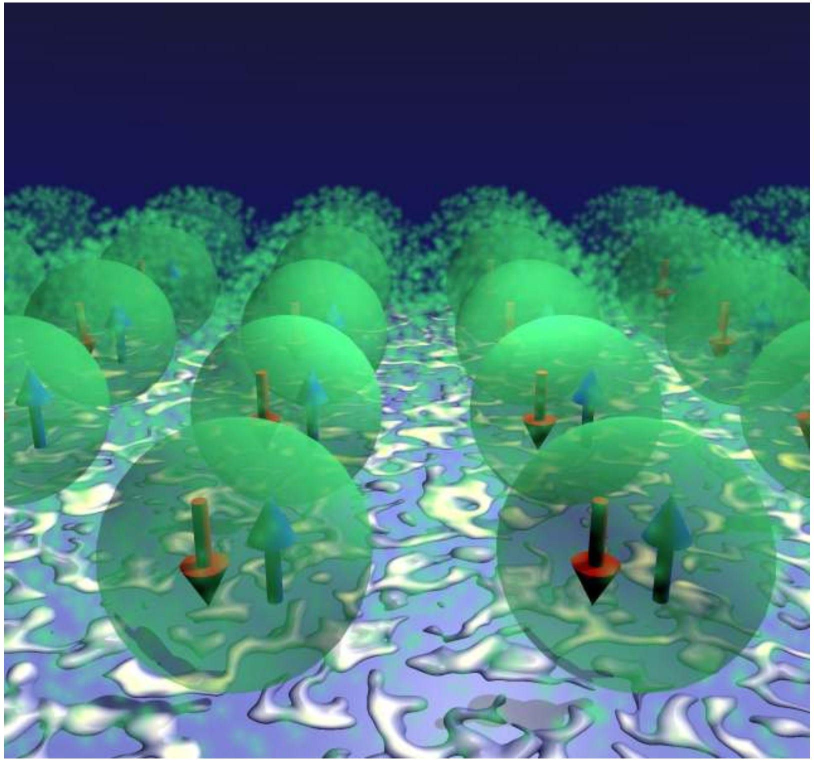
Robert Drost

Shawulienu Kezilebieke

Rouven Koch

Peter Liljeroth

Triplons in an artificial molecular quantum magnet



Phys. Rev. Lett. 131, 086701 (2023)

Triplons in an atomic-scale quantum magnet

Robert Drost

Shawulienu Kezilebieke

Peter Liljeroth

The quantum Heisenberg dimer

Let us take a minimal quantum magnet

$$\mathcal{H} = \vec{S}_0 \cdot \vec{S}_1$$

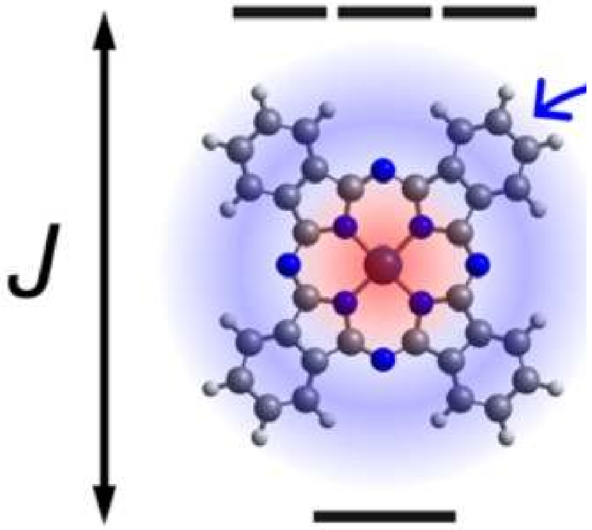
The ground state is unique, and does not break time-reversal

$$|GS\rangle = \frac{1}{\sqrt{2}}[|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle] \qquad \langle \vec{S}_i \rangle = 0$$

The state is maximally entangled

How does this generalize to a macroscopic system, and how can we see its quasiparticles?

Triplet excitations in a molecule



S = 1

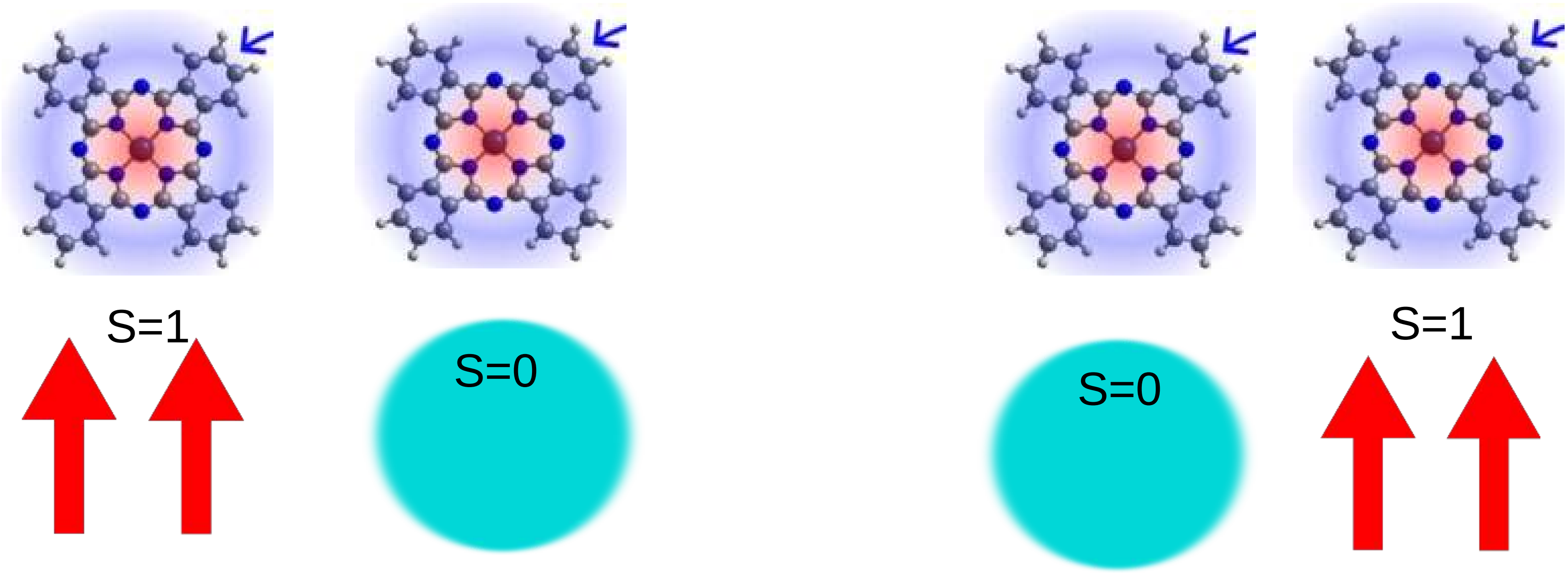
S = 0

Internal antiferromagnetic coupling

$$H = J\mathbf{S} \cdot \mathbf{K}$$

A single molecule (CoPC) has a singlet-triplet transition

Triplons in a dimer

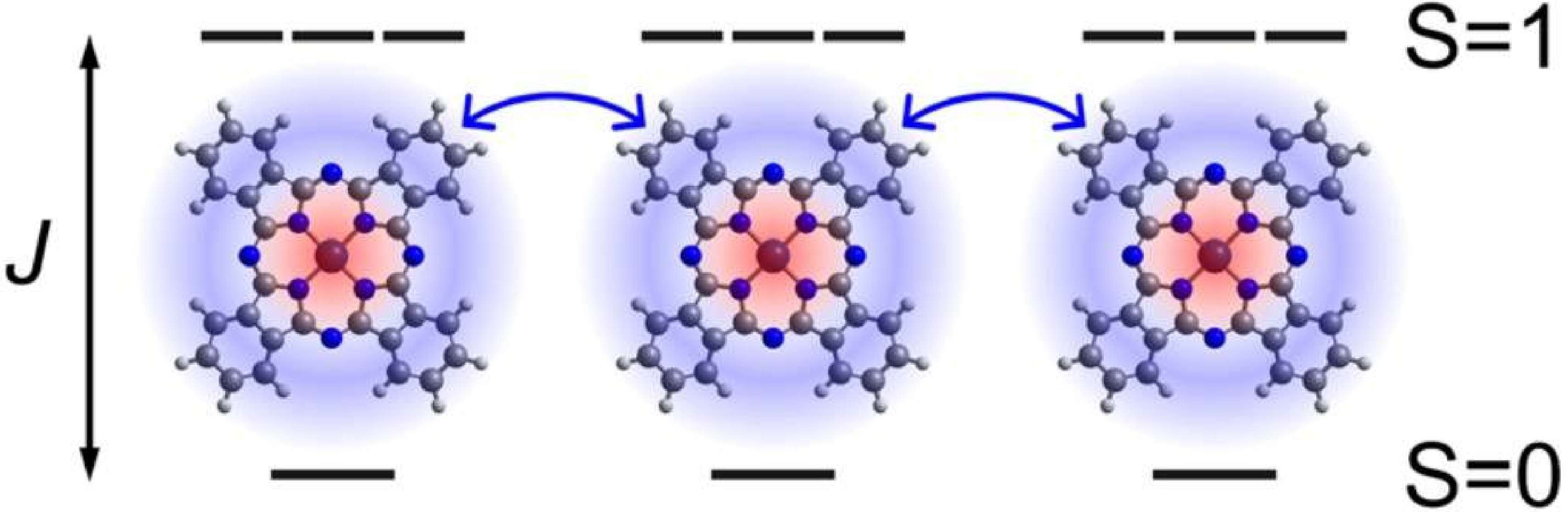


A triplet in the first molecule

Can jump to the second molecule

Exchange coupling between molecules leads to propagating triplon excitations

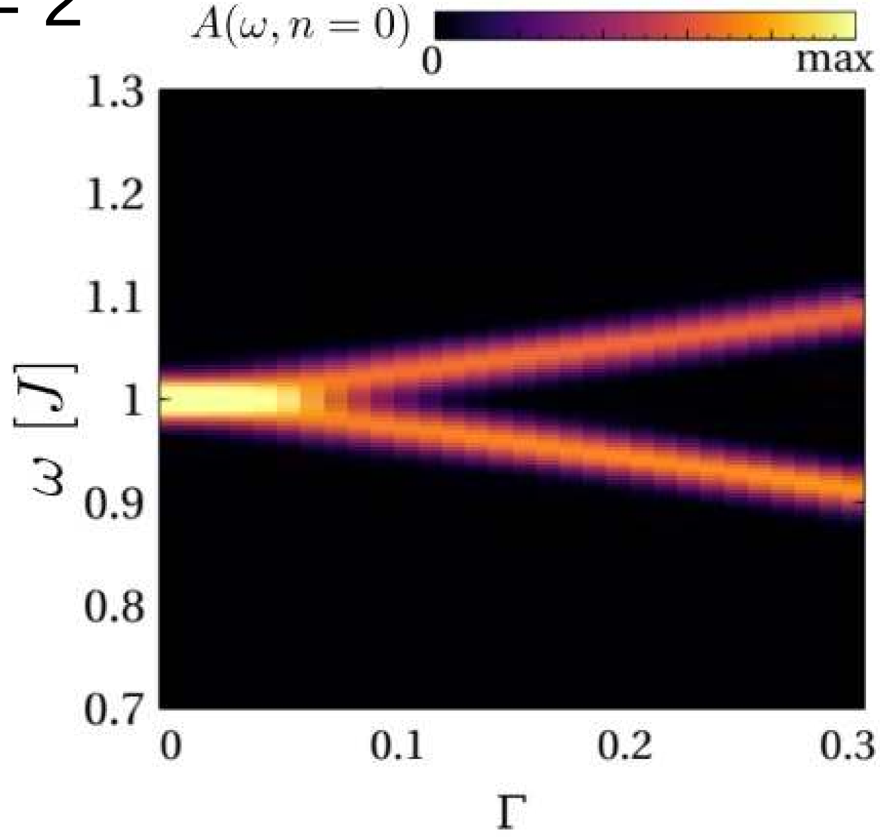
A chain of triplons



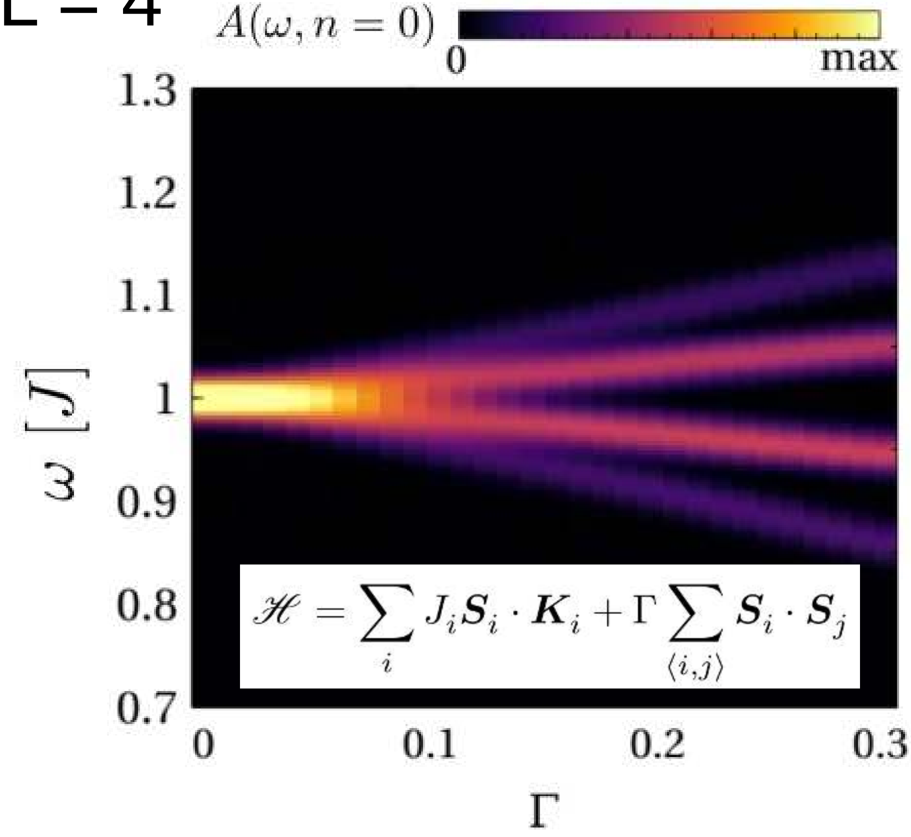
$$\mathscr{H} = \sum_{i} J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Chains of triplons

L = 2

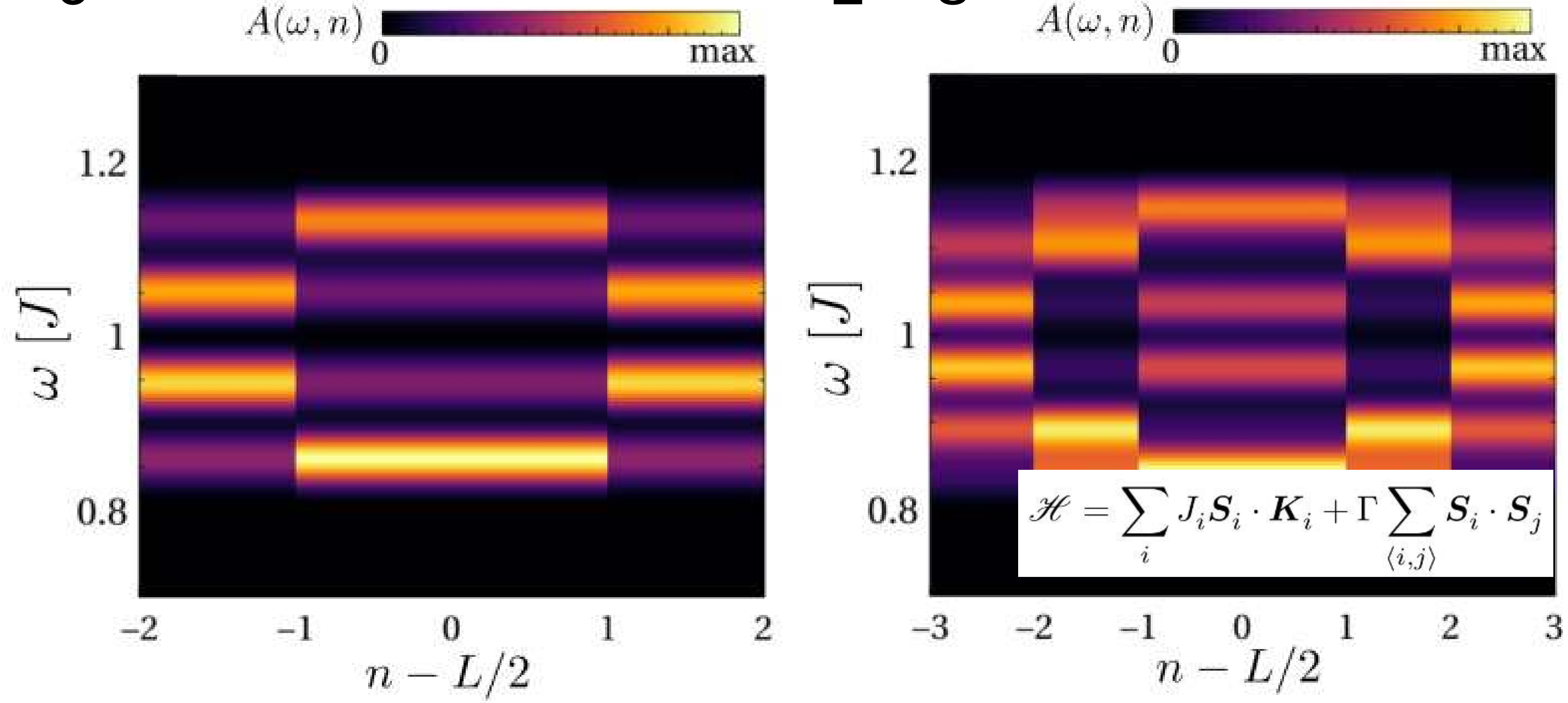


L = 4



Chains of triplons

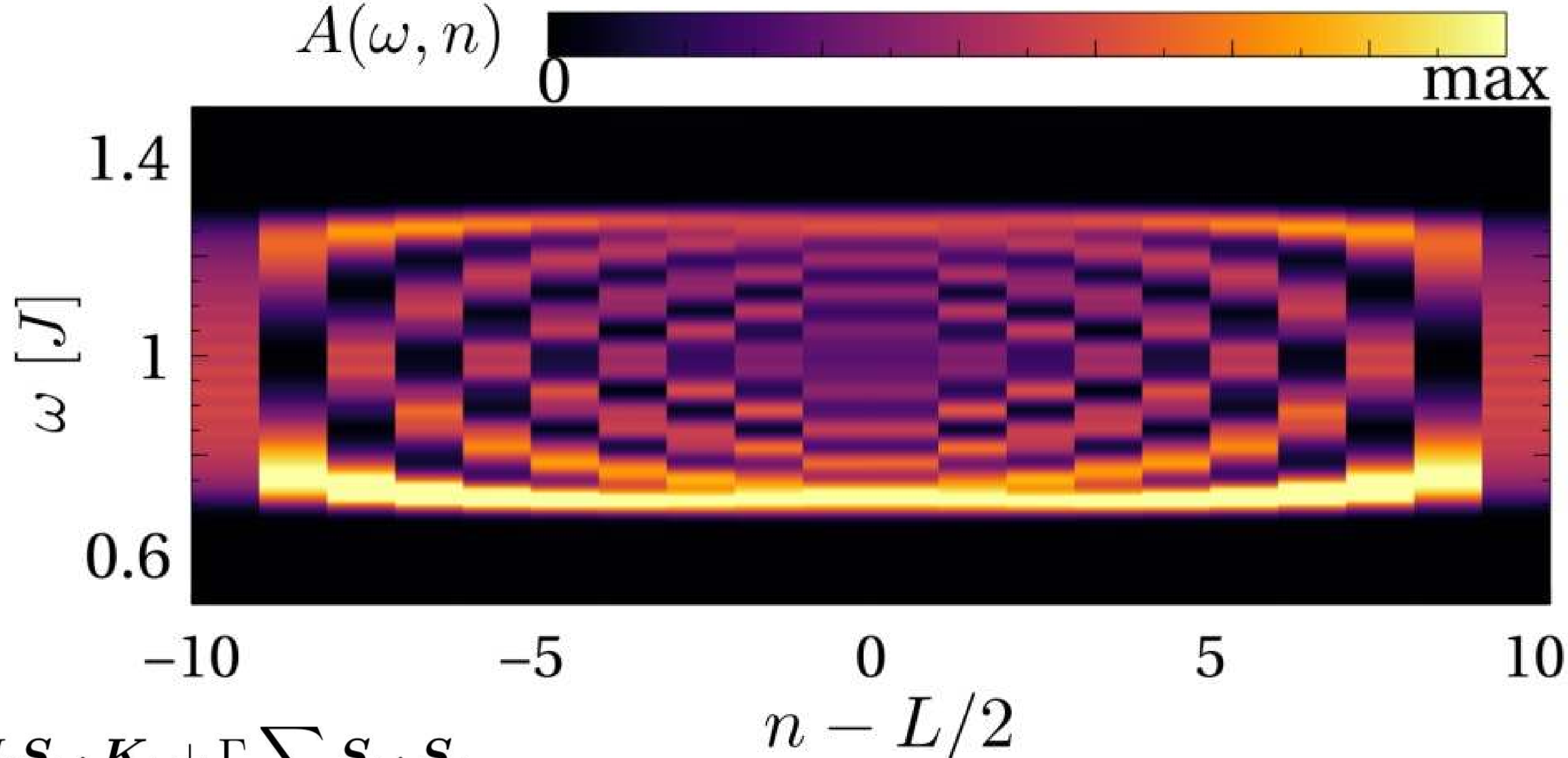
L = 6 L = 8



$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

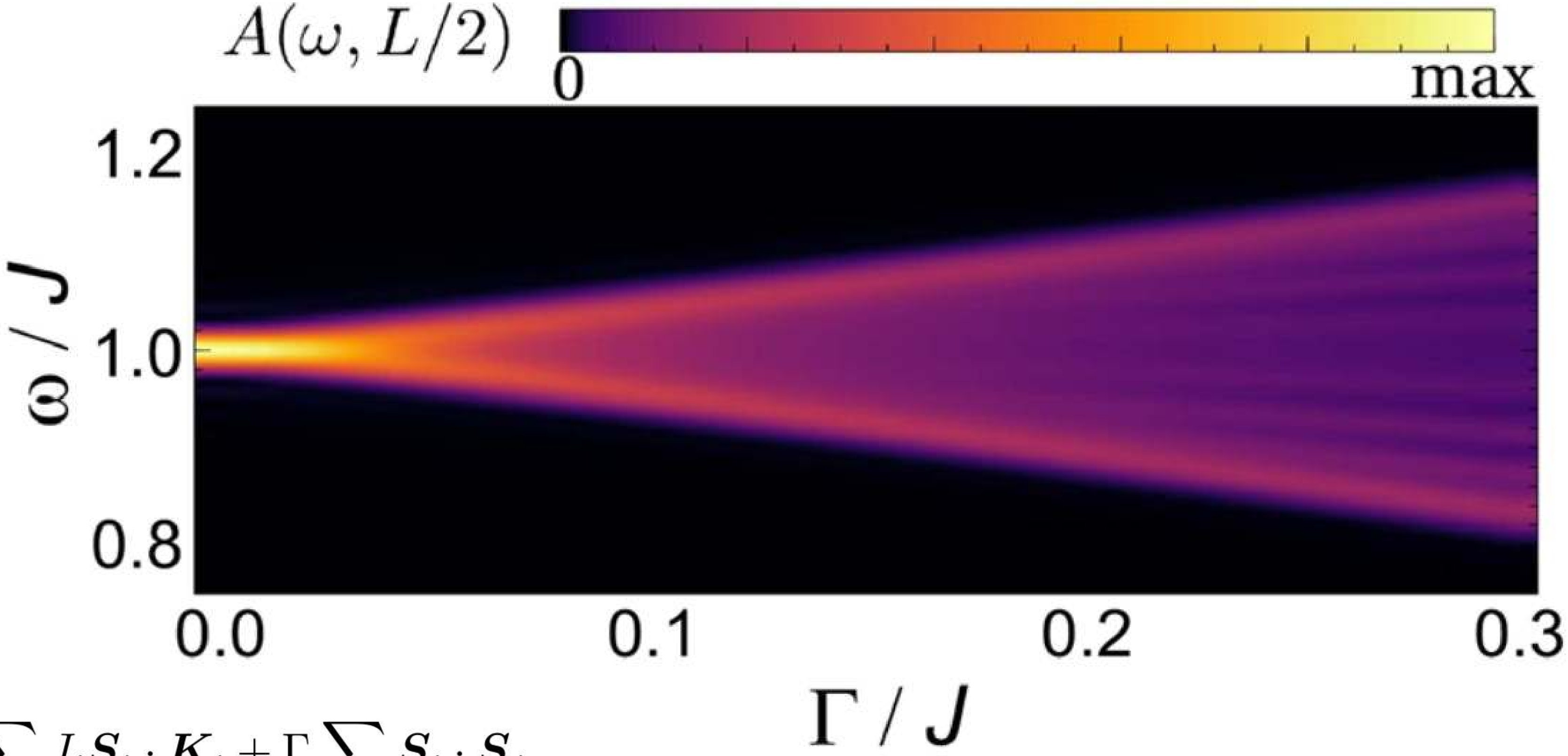
Chains of triplons

L = 20



$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

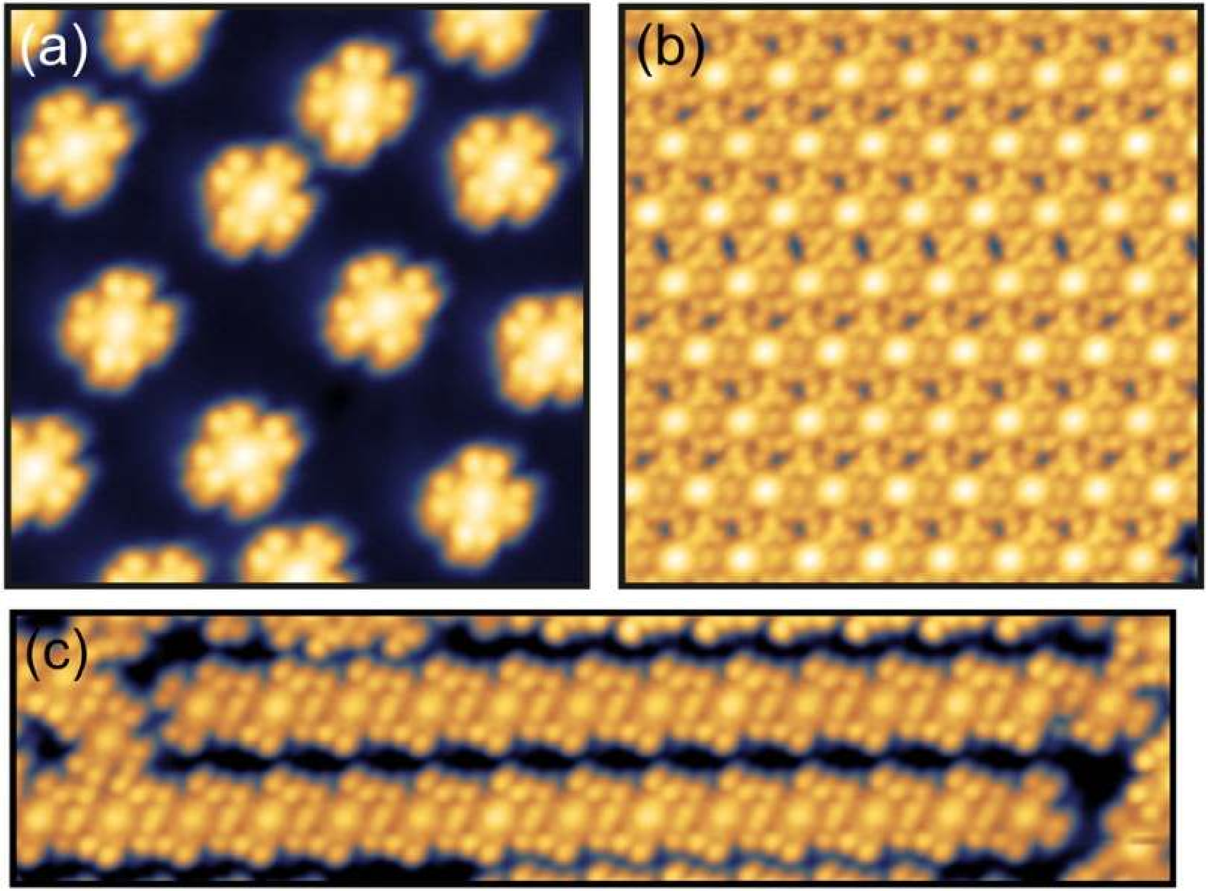
Chains of triplons



$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

0D, 1D and 2D arrays

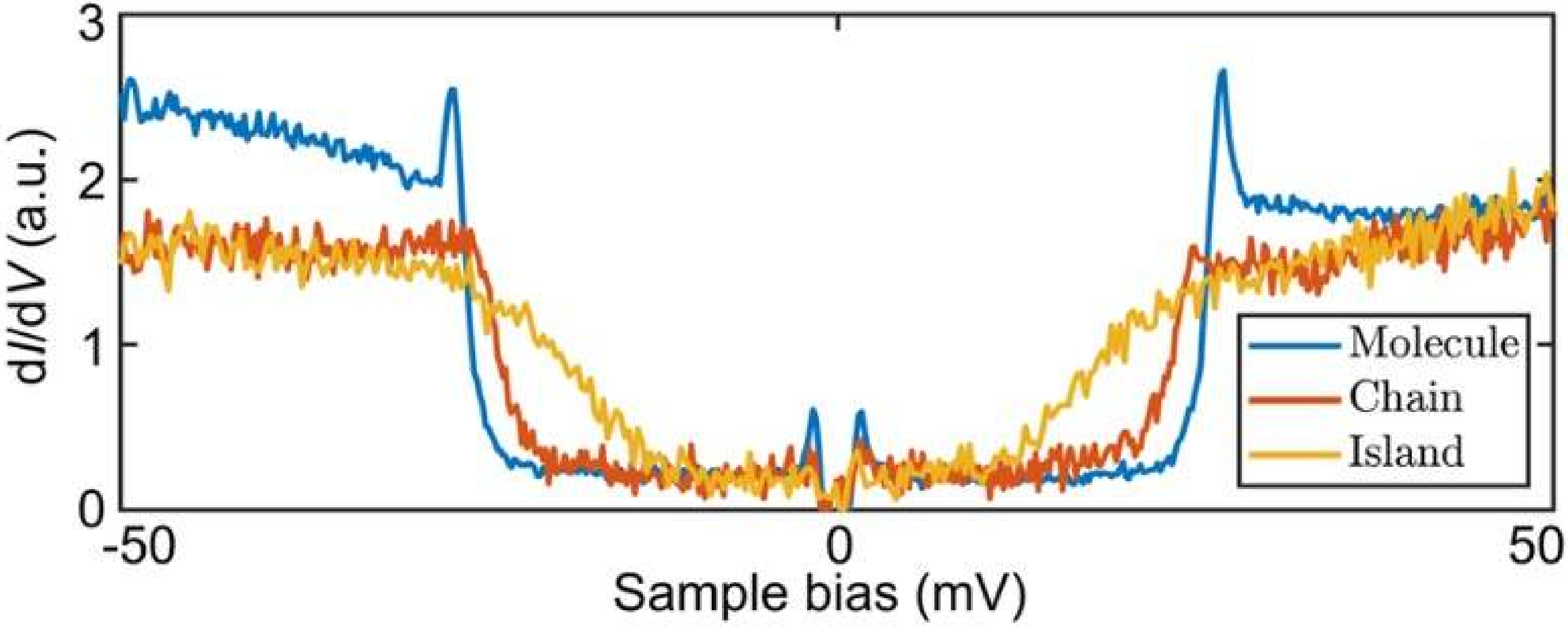
0D



2D

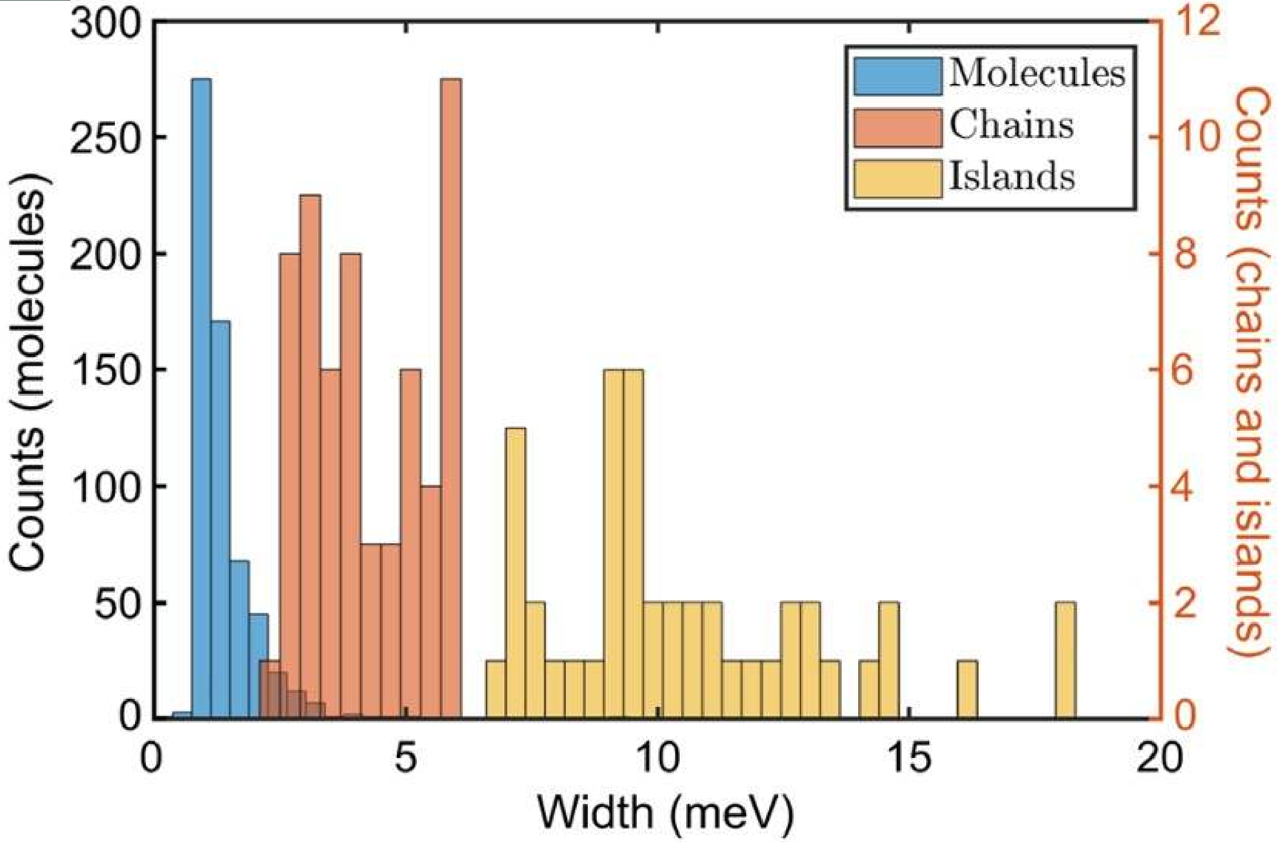
1D

Inelastic excitations for 0D, 1D and 2D arrays



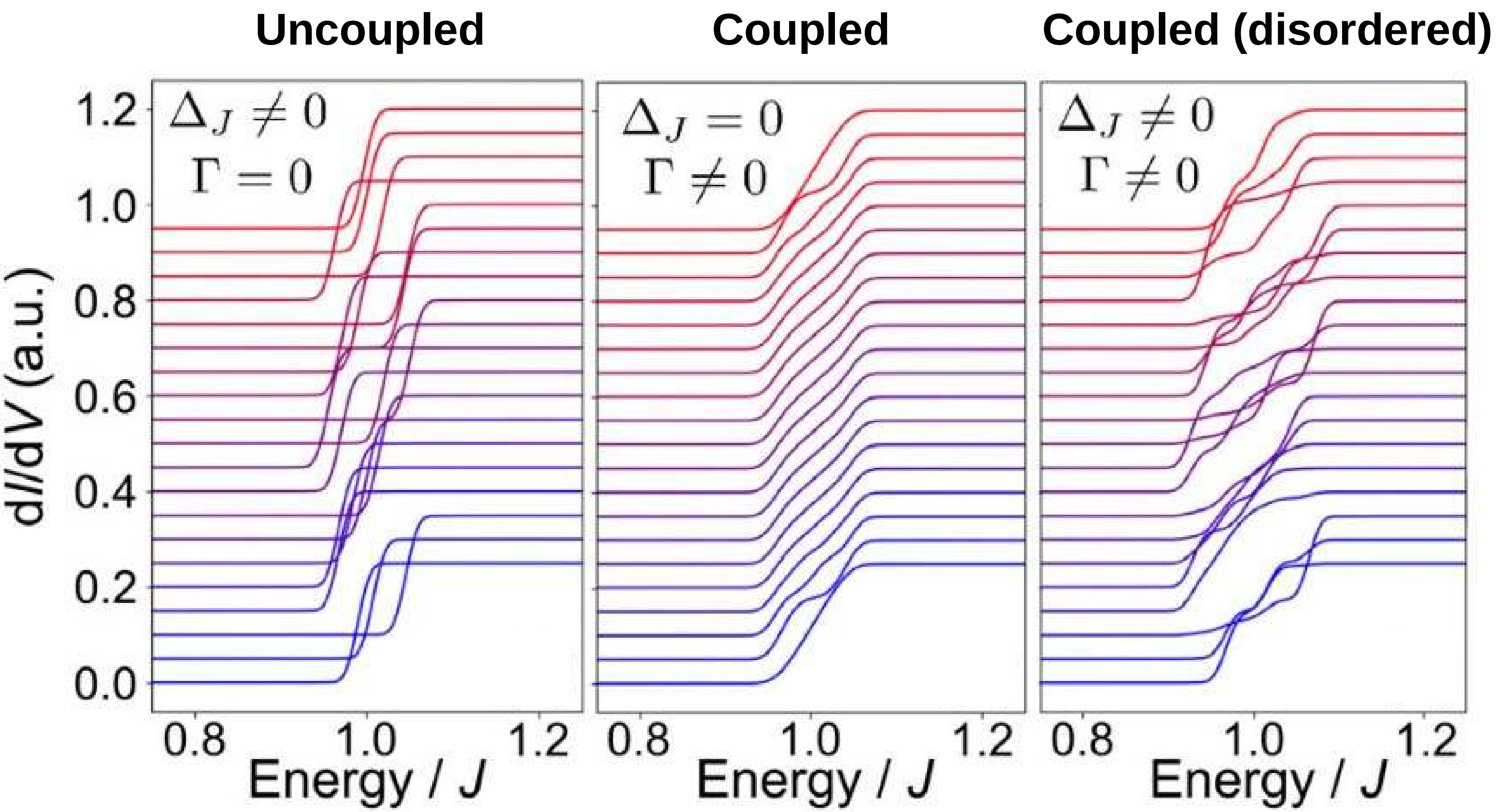
The width of the step is bigger for islands than for molecules

Statistic of the inelastic steps

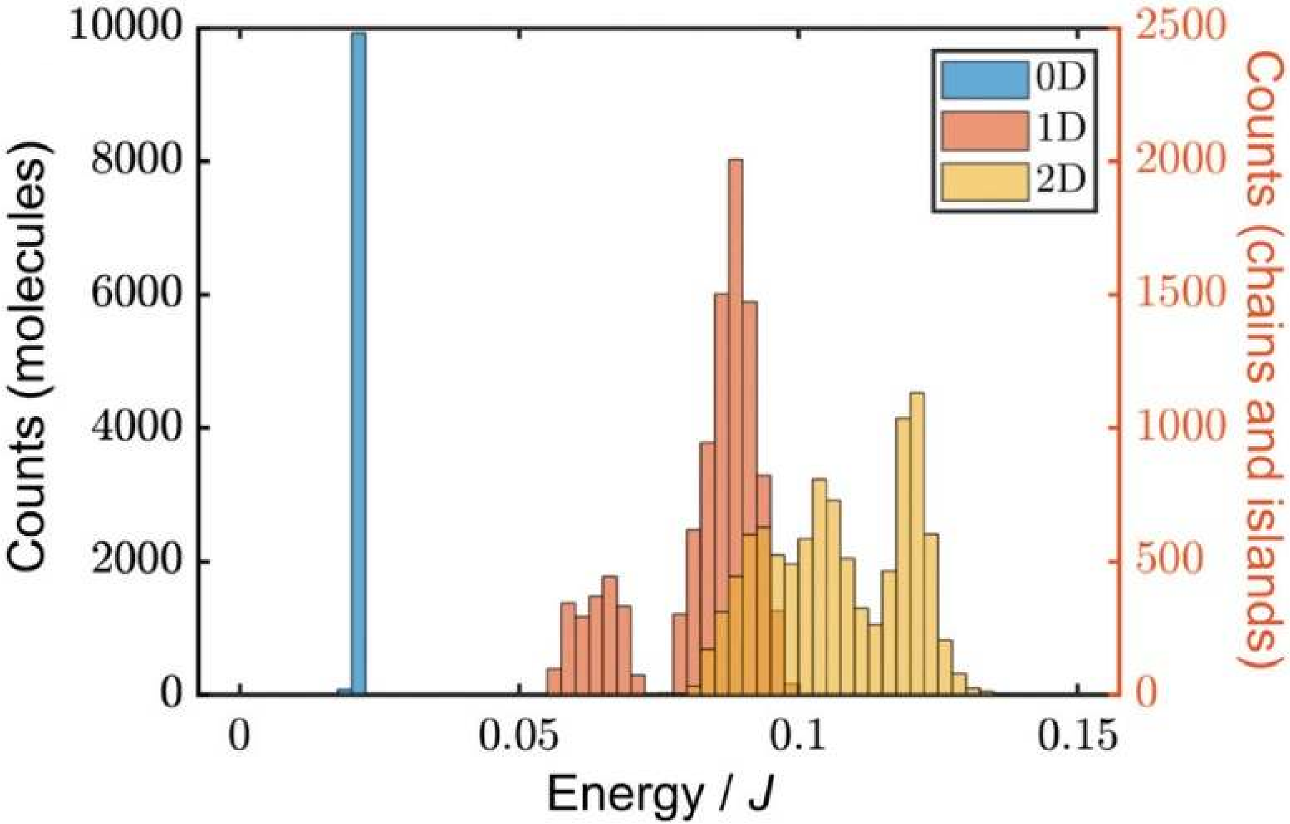


2D arrays have the largest width of the spin excitation

Coupled and uncoupled inelastic excitation

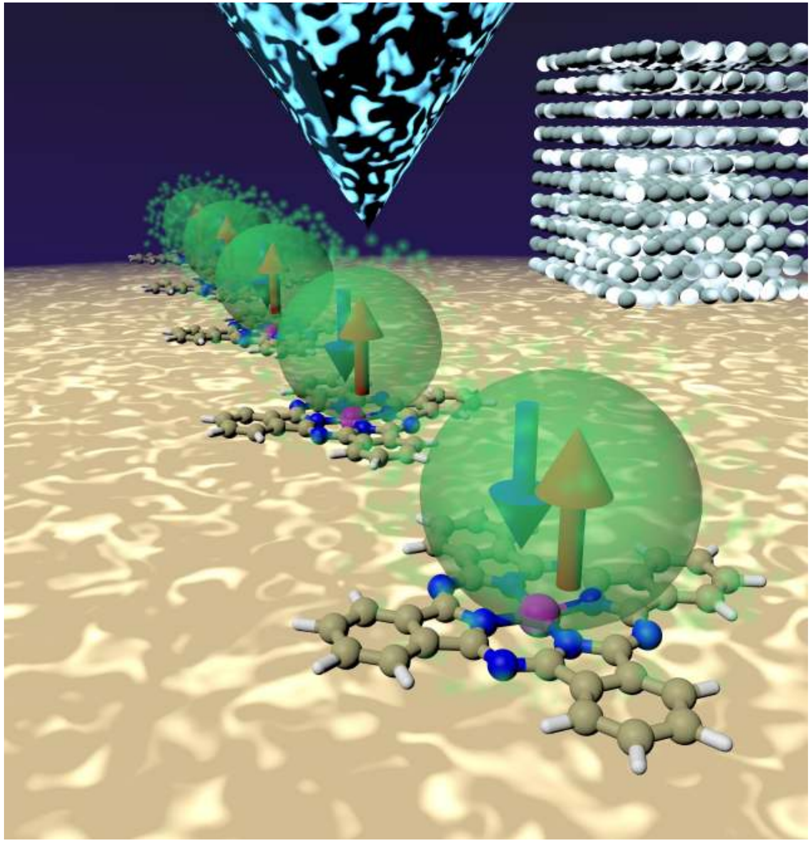


Statistics of the theoretical model



2D arrays have the largest width of the spin excitation

Hamiltonian learning triplons in a molecular quantum magnet



Rouven Koch

Robert Drost

Peter Liljeroth

arXiv:2504.20711 (2025)

The problem of Hamiltonian learning

If we measure a quantum magnet, how can we obtain its microscopic description?

Conventional workflow in theoretical physics

Solve the Hamiltonian

Compute an observable

$$H \longrightarrow \langle \Psi | O | \Psi \rangle$$

The problem we often find experimentally

Measure an observable

Determine its Hamiltonian

$$\langle \Psi | O | \Psi \rangle \longrightarrow H$$

How can we extract a complex Hamiltonian from its physical observables?

Hamiltonian learning: combining theory and experiment

Train a machine learning algorithm with **simulated quantum many-body theory data**

so that the parameters of each Hamiltonian are explicitly know

$$H(\lambda_i^{\text{theo}}) \longrightarrow \langle O \rangle_{\text{theo}} \longrightarrow \boxed{\text{Neural network}} \longrightarrow \lambda_i^{\text{theo}}$$

Hamiltonian

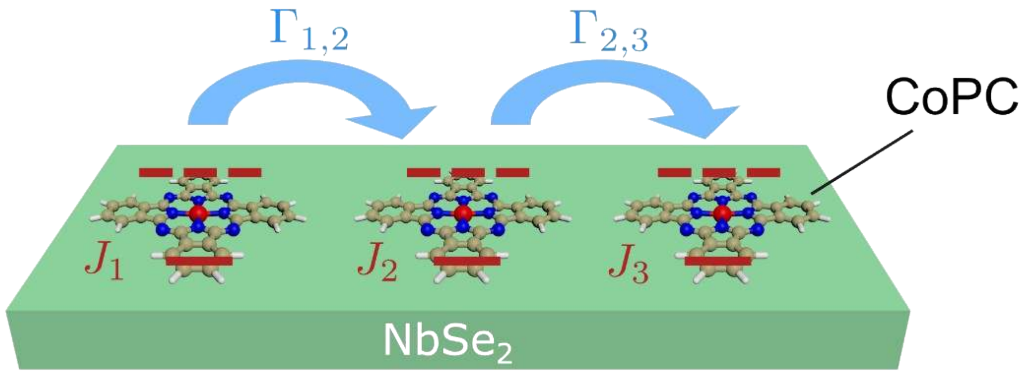
With the trained algorithm, **evaluated in experimental data** to extract the parameters

$$\langle O \rangle_{\text{exp}} \longrightarrow \boxed{\text{Neural network}} \longrightarrow \lambda_i^{\text{exp}}$$

Hamiltonian

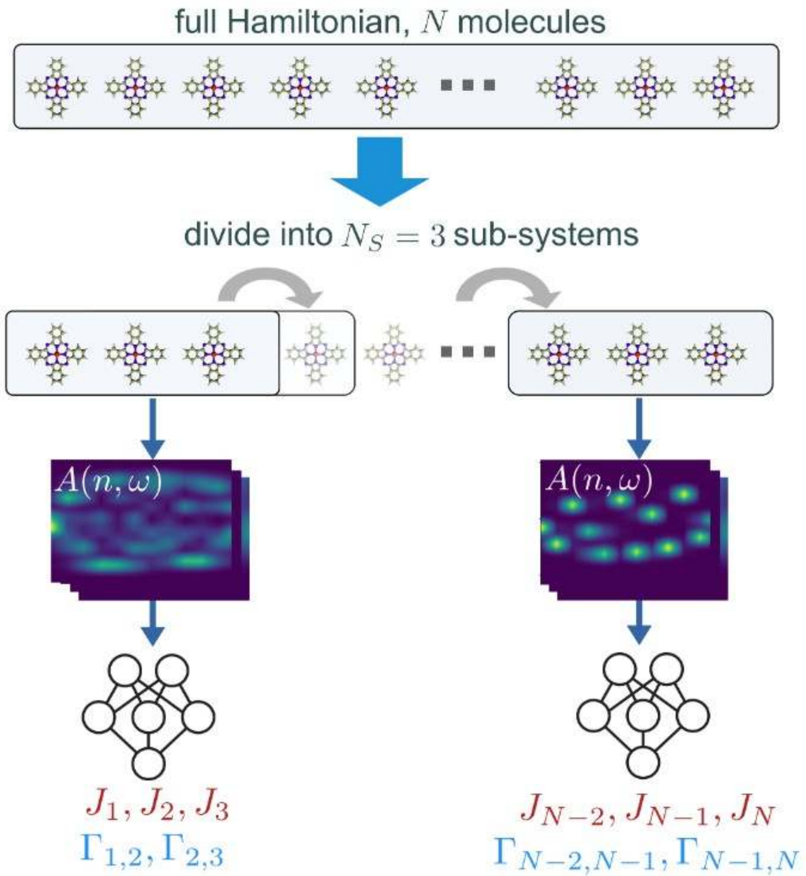
This strategy combines quantum many-body solvers, machine learning, and experimental measurements

A strategy for machine learning quantum magnets



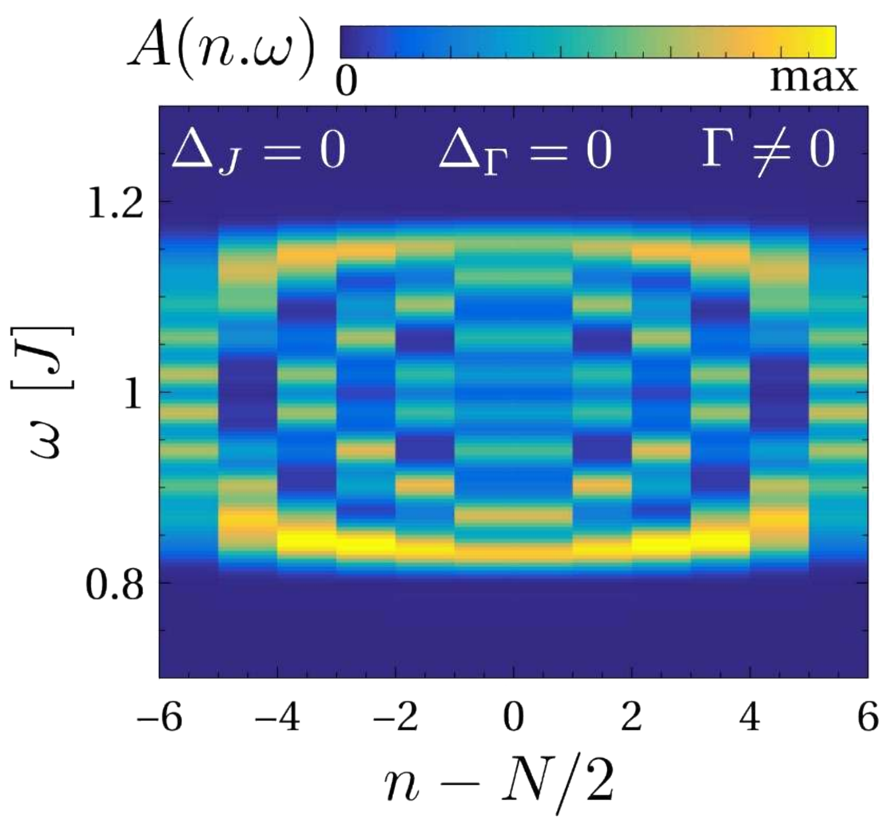
$\Gamma_{n,n+1}$: intermolecular exchange
J_n: intramolecular exchange

$$H = \sum_n J_n \mathbf{S}_n \cdot \mathbf{K}_n + \sum_n \Gamma_{n,n+1} \mathbf{S}_n \cdot \mathbf{S}_{n+1}$$

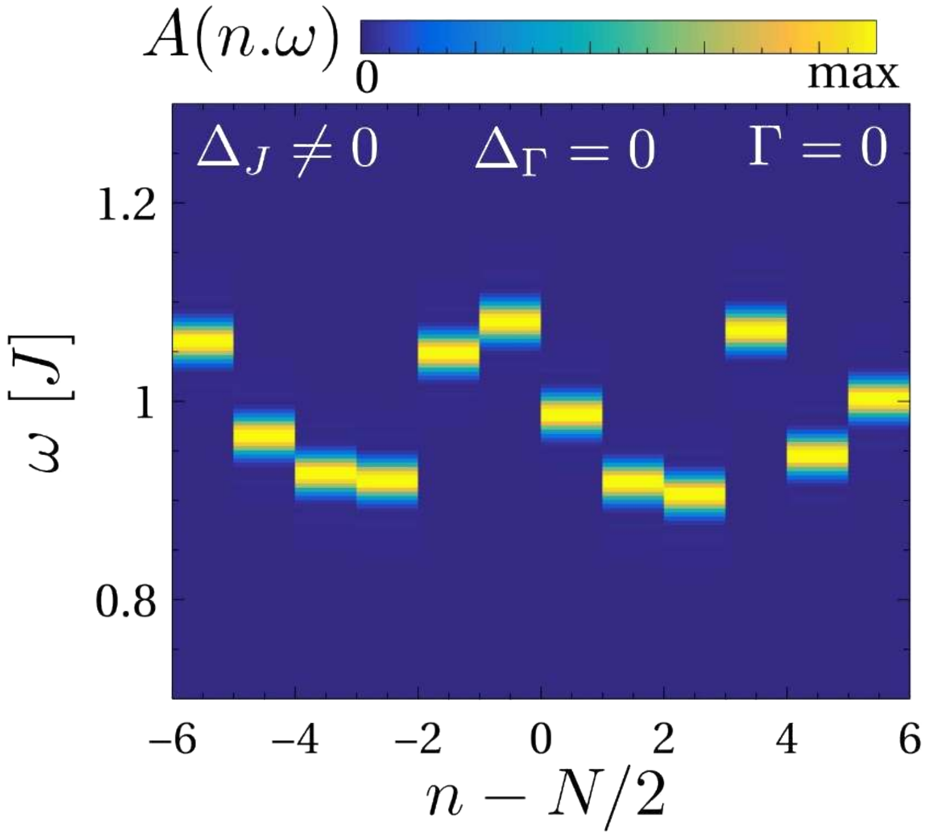


Pristine triplon excitations

Pristine and coupled molecules

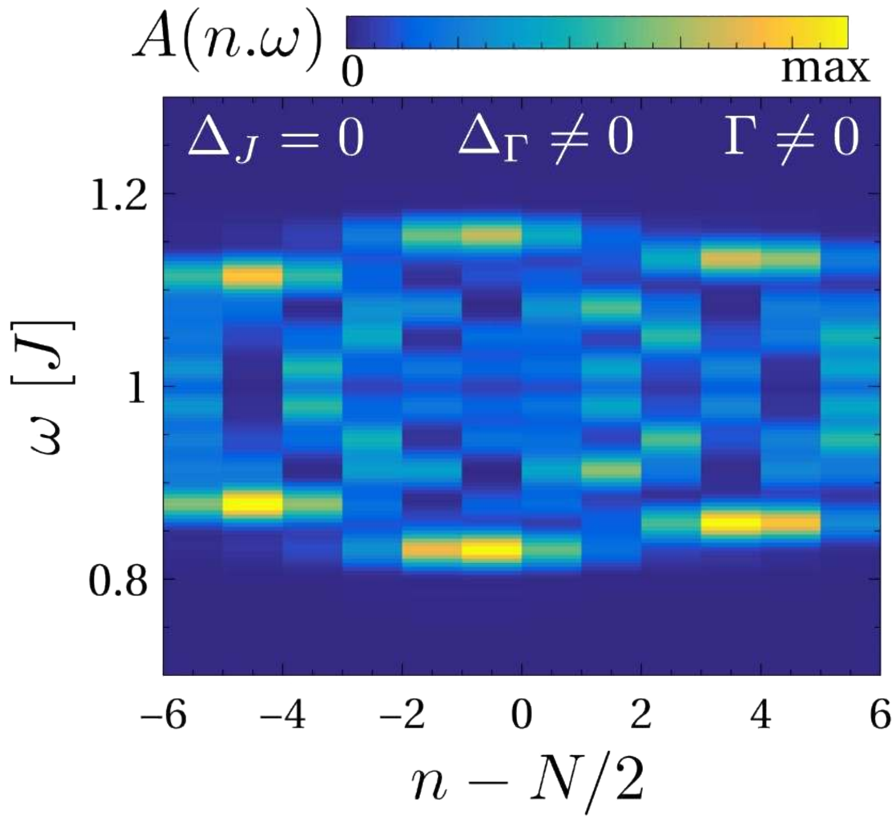


Decoupled and disordered molecules

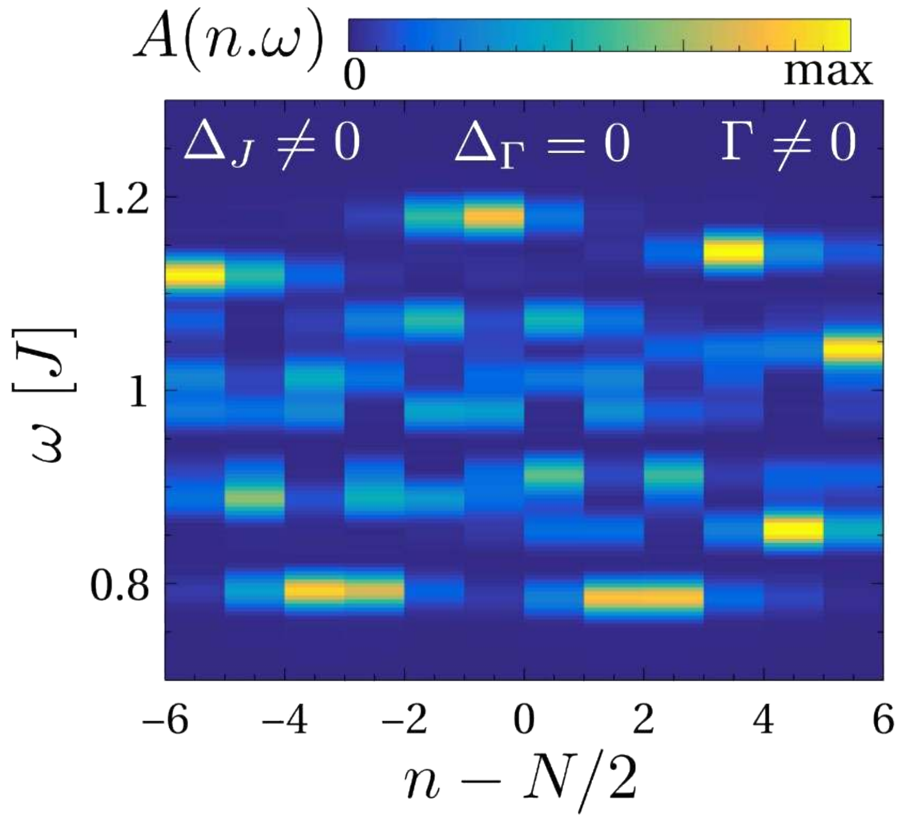


The different impact of molecular disorder

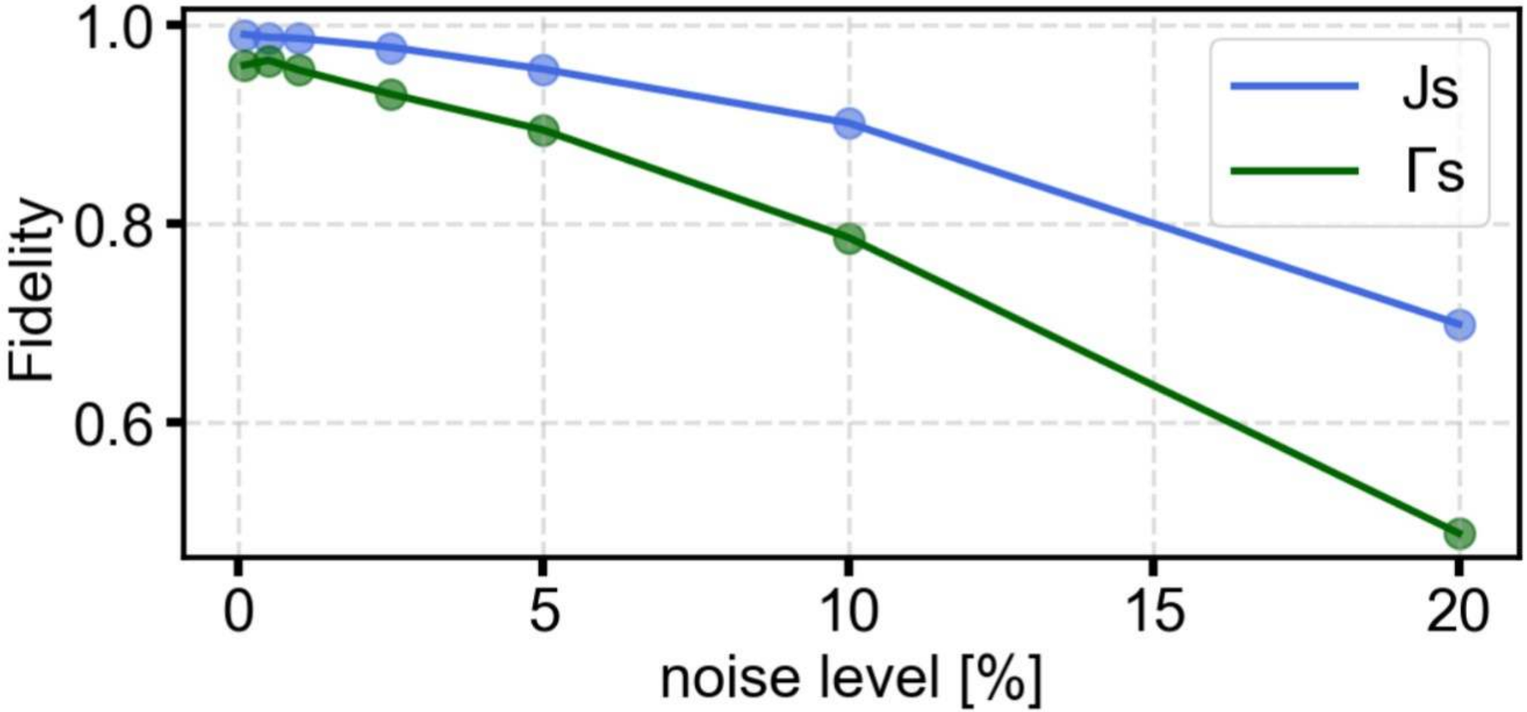
Disorder in inter-molecule coupling



Disorder in intra-molecule coupling

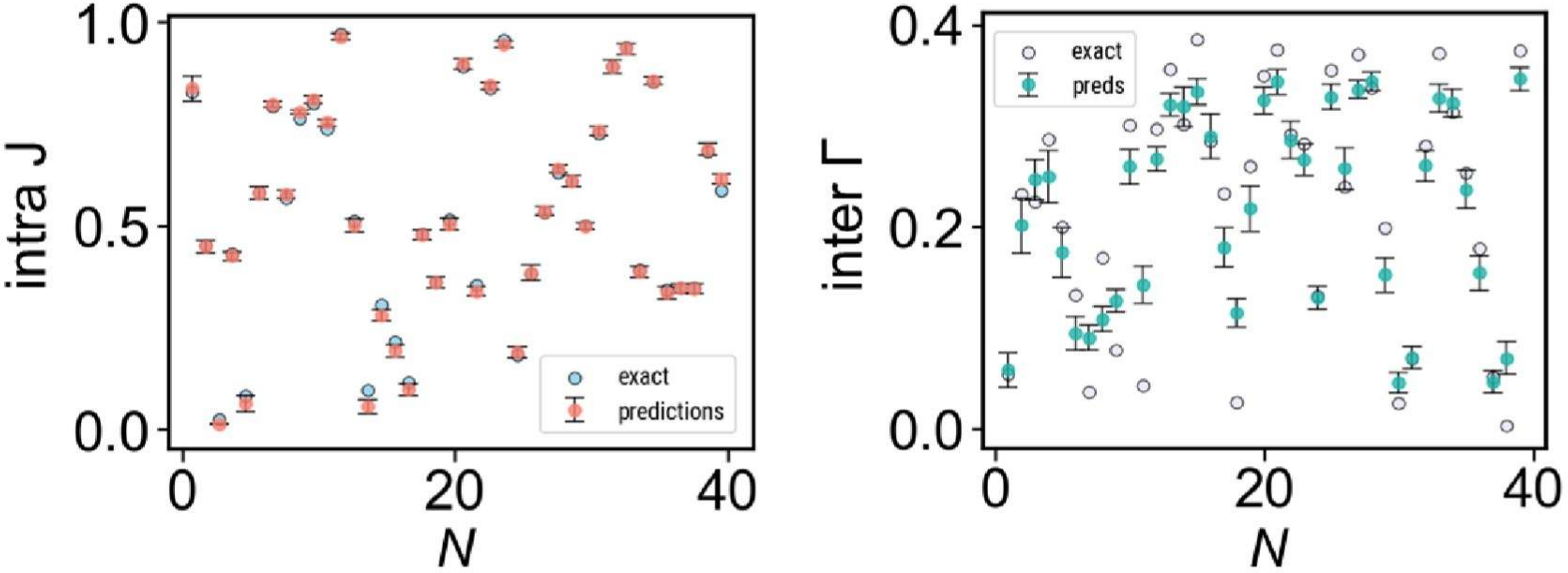


Resilience to noise of Hamiltonian learning



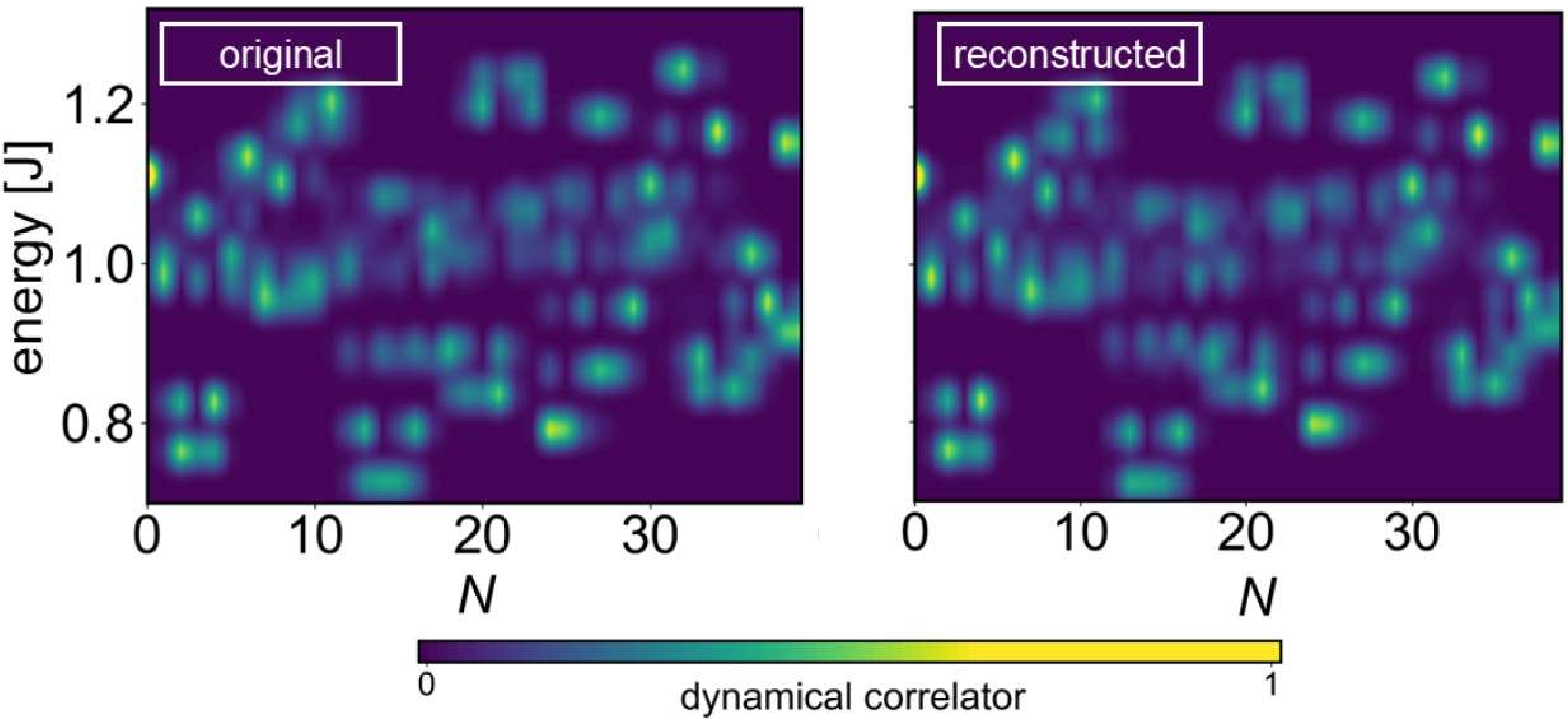
Noise model $$\left(\frac{dI}{dV}\right)_{\text{noise}} = \left(\frac{dI}{dV}\right)_{\text{data}} + \eta \cdot \mathbf{R}$$

Inferring spatially dependent exchange couplings

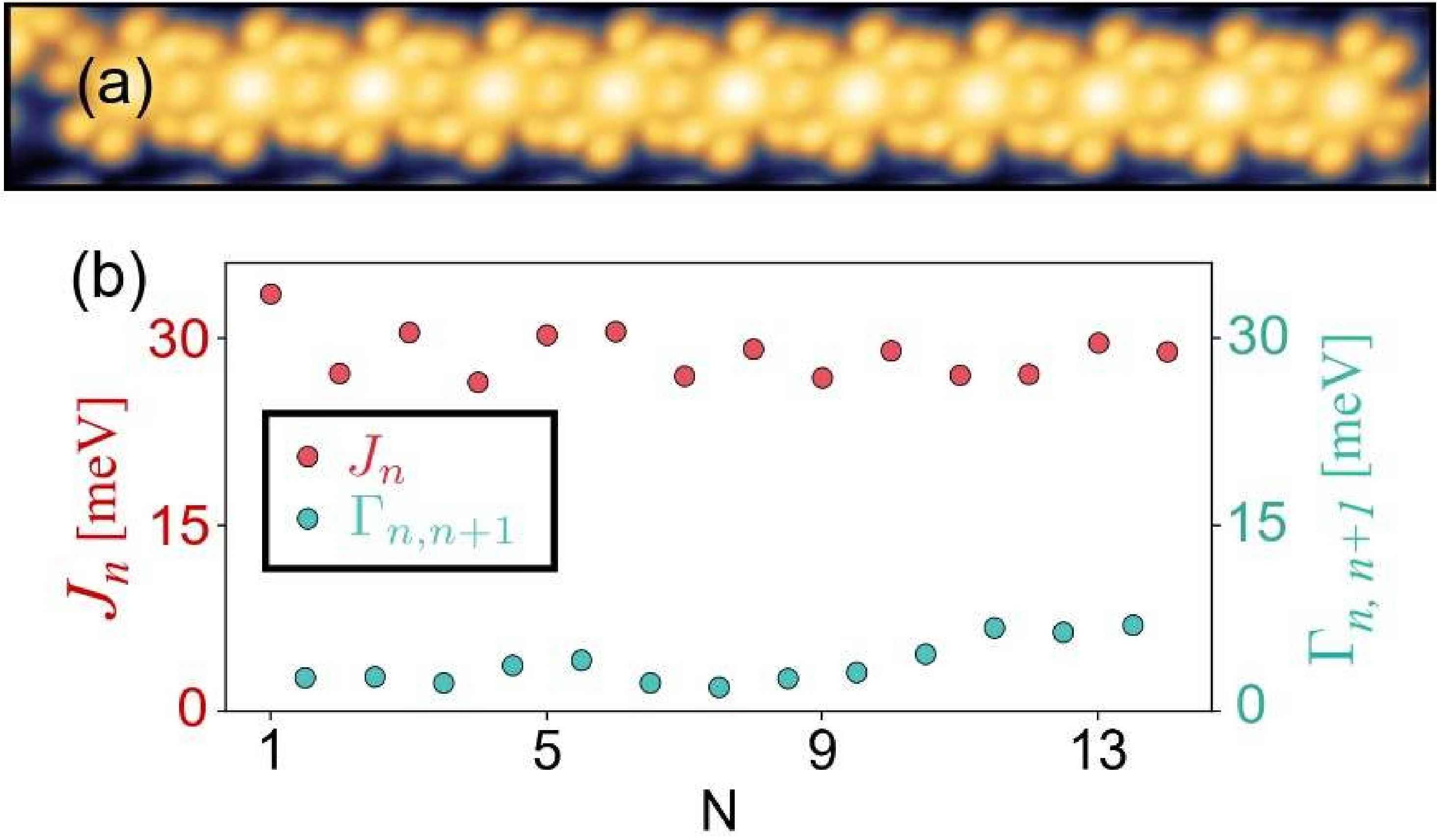


Intramolecular exchange variations can be predicted robustly
Intermolecular exchange variations are more subtle to predict

Reconstructing a quantum magnet Hamiltonian

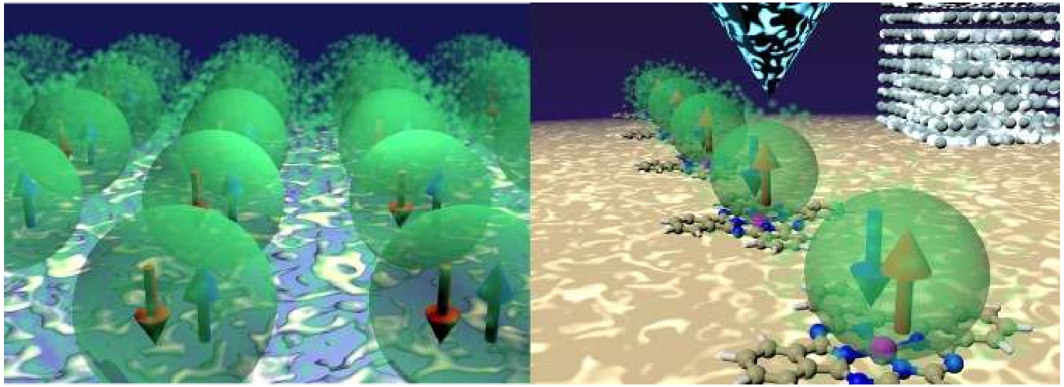


Machine learning triplons experimentally



Take home

STM sensing allows learning the Hamiltonian of complex collective quantum magnets, by leveraging machine learning trained with quantum many-body methods



Phys. Rev. Lett. 131, 086701 (2023)
arXiv:2504.20711 (2025)

Funding from

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Finnish Quantum Flagship
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J&AE JANE AND AATOS ERKKO FOUNDATION

Institute Q

Research Council of Finland