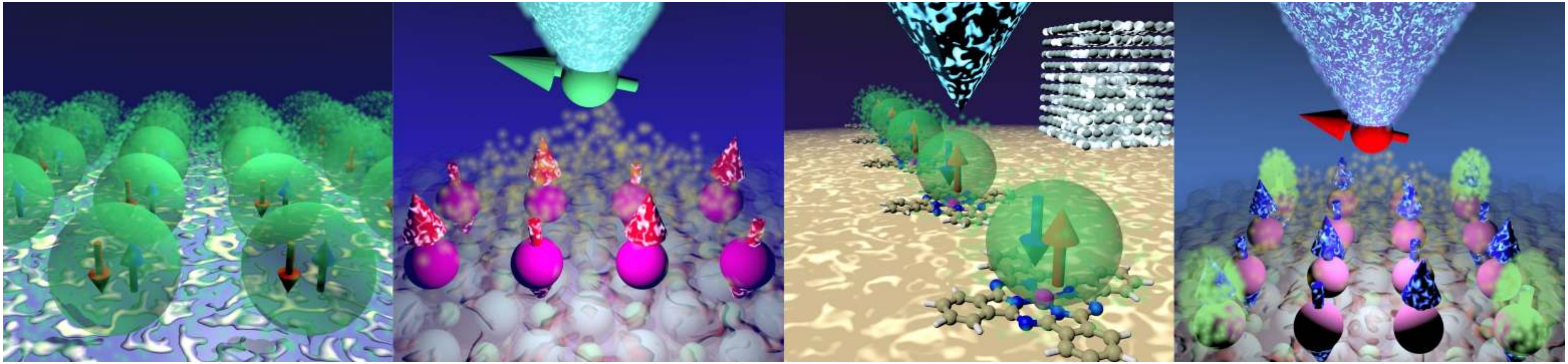
Hamiltonian learning triplons and high-order topological order in nanoscale quantum magnets

Jose Lado

Department of Applied Physics, Aalto University, Finland



Phys. Rev. Appl 20, 024054 (2023)

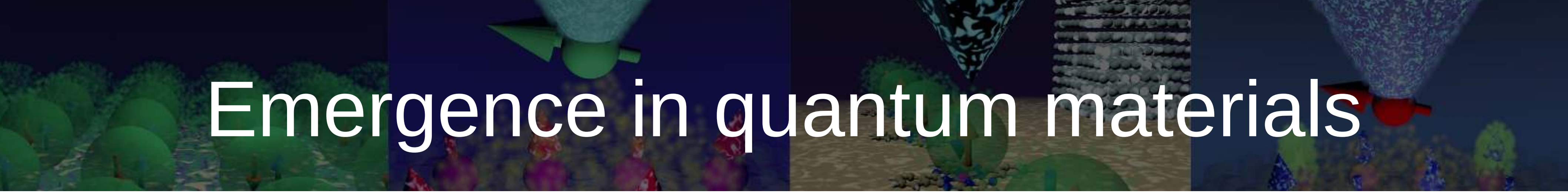
Nature Nano 19, 1782–1788 (2024)

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

arXiv:2504.20711 (2025)

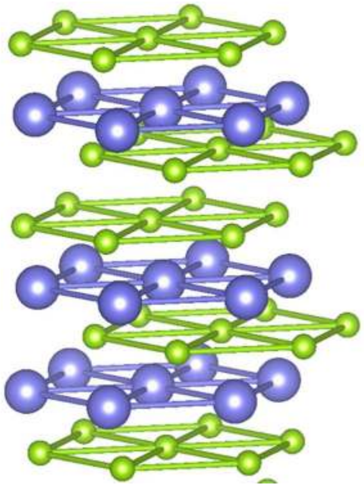
4th European Conference on Molecular Spintronics, Trinity College Dublin, Ireland

June 4th, 2025



Emergence in quantum materials

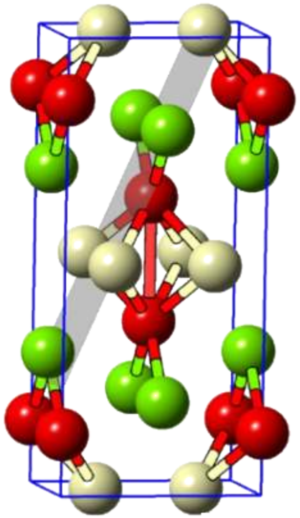
Topological materials



Bi_2Se_3

Science, 325(5937), 178-181 (2009)

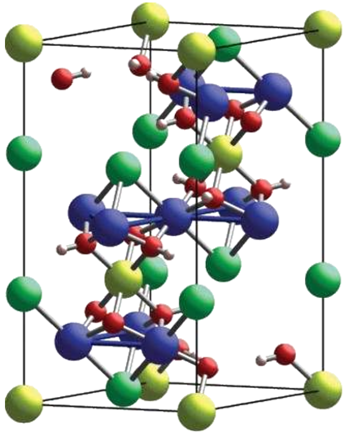
Unconventional superconductors



UTe_2

Nature 579, 523–527 (2020)

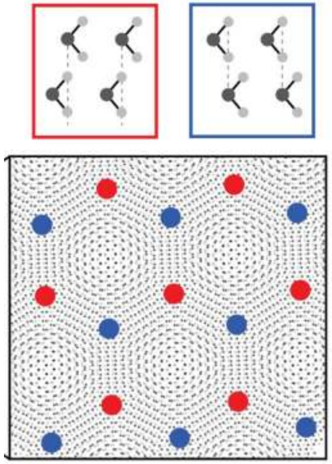
Quantum spin liquids



$ZnCu_3(OH)_6Cl_2$

Science 350(6261), 655-658 (2015)

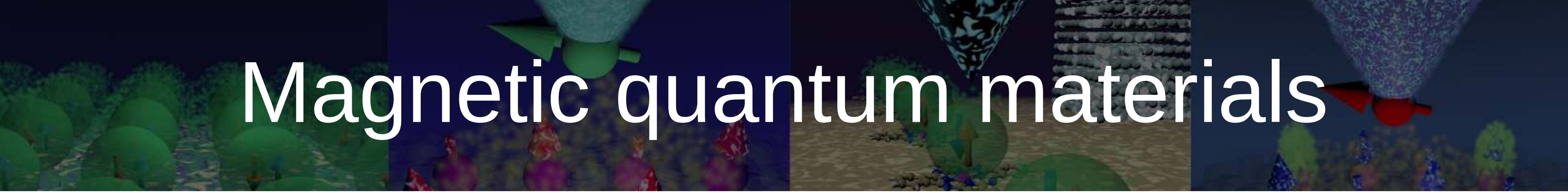
Fractional topological matter



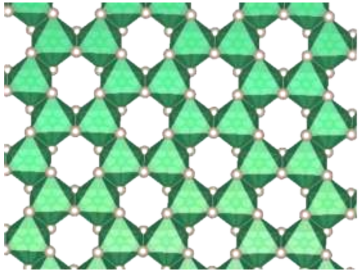
Twisted $MoTe_2$

Nature 622, 63–68 (2023)

Magnetic quantum materials



Ferromagnet, antiferromagnets

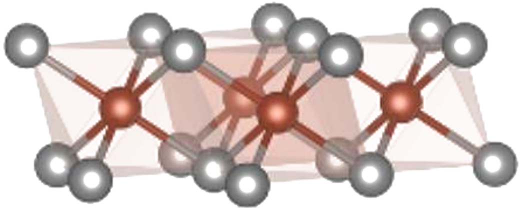


CrI_3, $CrCl_3$, $CrBr_3$

Nature 546, 270–273 (2017)

Break time-reversal

Multiferroics

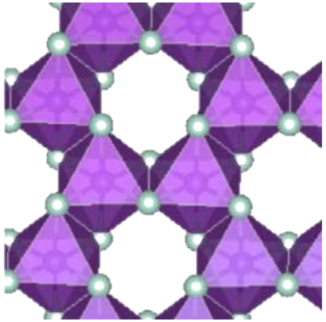


NiI_2

Nature 602, 601–605 (2022)

Break time-reversal and inversion symmetry

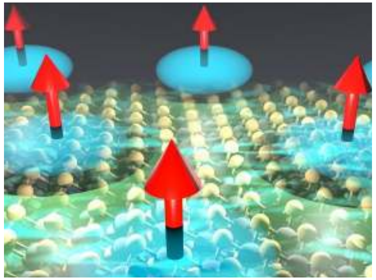
(proximal) Quantum spin-liquids



$RuCl_3$, 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Heavy-fermion Kondo insulators



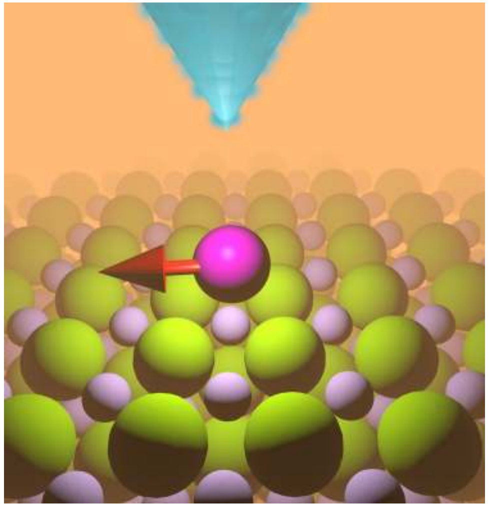
1T-TaS_2/1H-TaS_2

Nature 599, 582–586 (2021)

Do not break time-reversal

Building quantum matter with artificial lattices

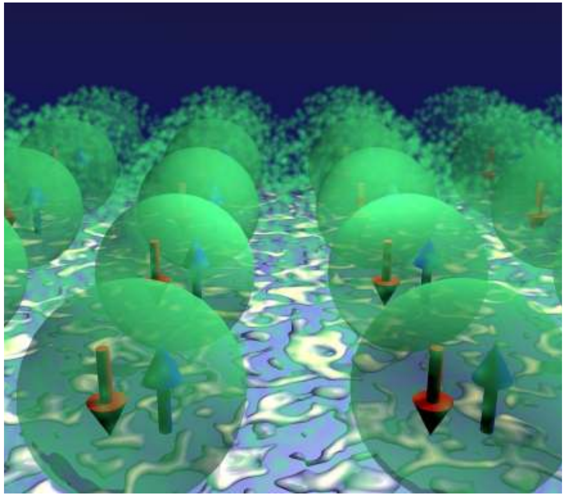
Atomic lattices



a = 0.5 nm

Nature Physics 12 (7), 656 (2016)
Nature Physics 13 (7) 668 (2017)
Science 335 (6065), 196-199 (2012)

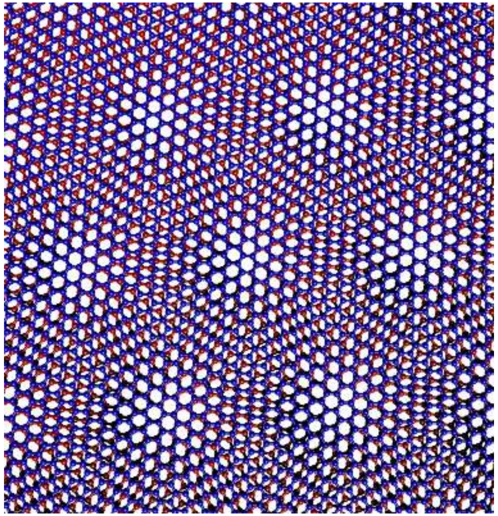
Molecular lattices



a = 3 nm

Nature 408, 447–449 (2000)
Nature Rev. Mat. 5 (2), 87-104 (2020)
Nature 598, 287–292 (2021)
Phys. Rev. Lett. 130, 100401 (2023)

Twisted 2D moire materials

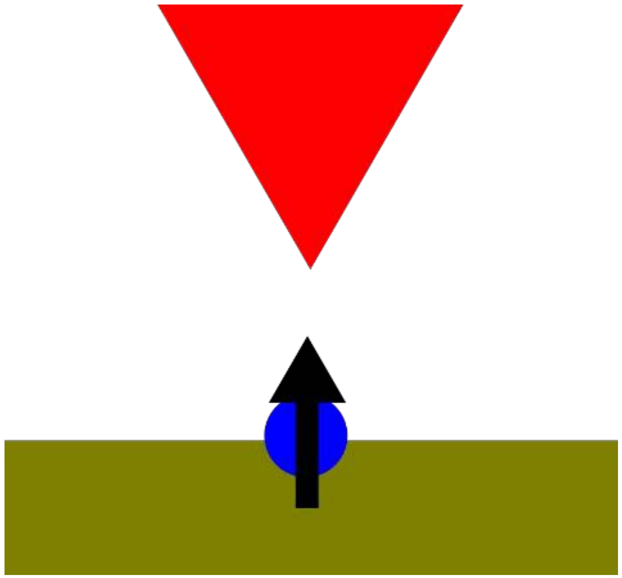


a = 30 nm

Phys. Rev. Lett. 99, 256802 (2007)
Nature Physics 6, 109–113 (2010)
Nature 556, 80–84 (2018)
Nature 599, 582–586 (2021)

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

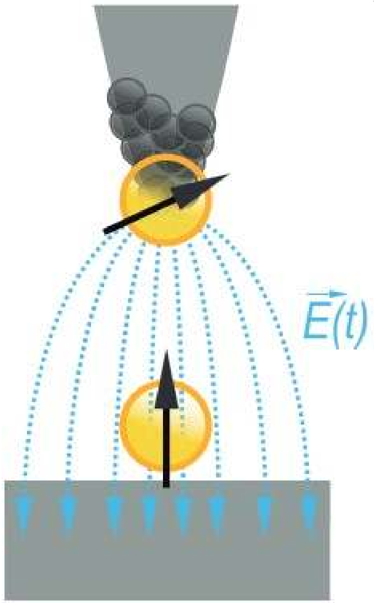
Inelastic spectroscopy



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

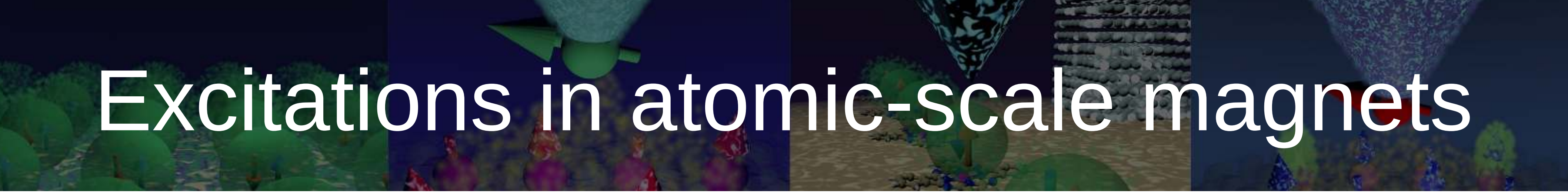
Without spin-polarized tip

Electrically driven paramagnetic resonance



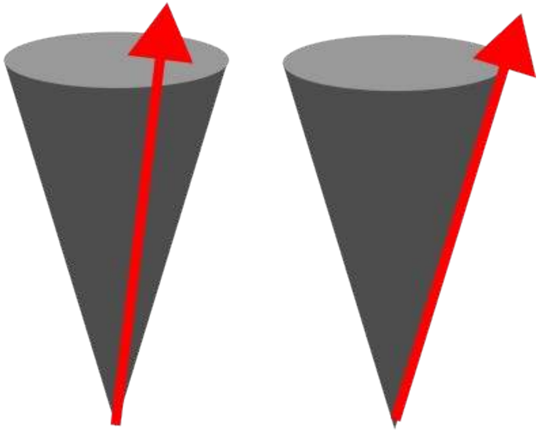
Science 350 (6259), 417-420 (2015)

With spin-polarized tip



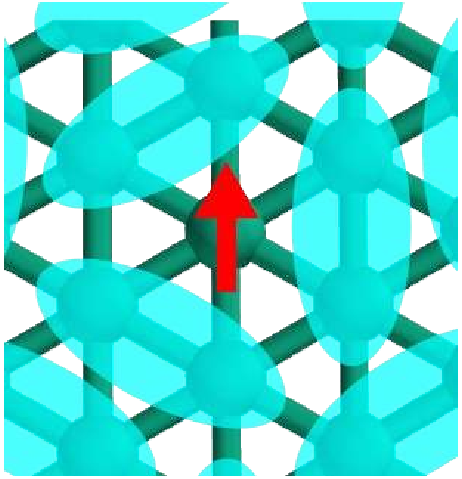
Excitations in atomic-scale magnets

Magnons



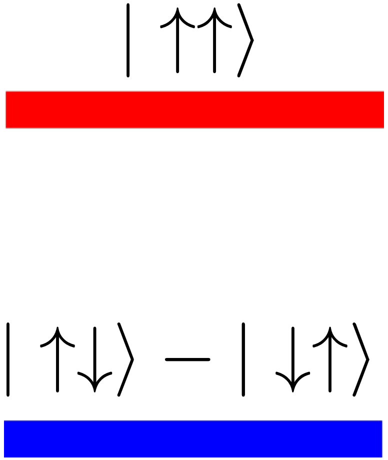
S=1

Spinons



S=1/2

Triplons



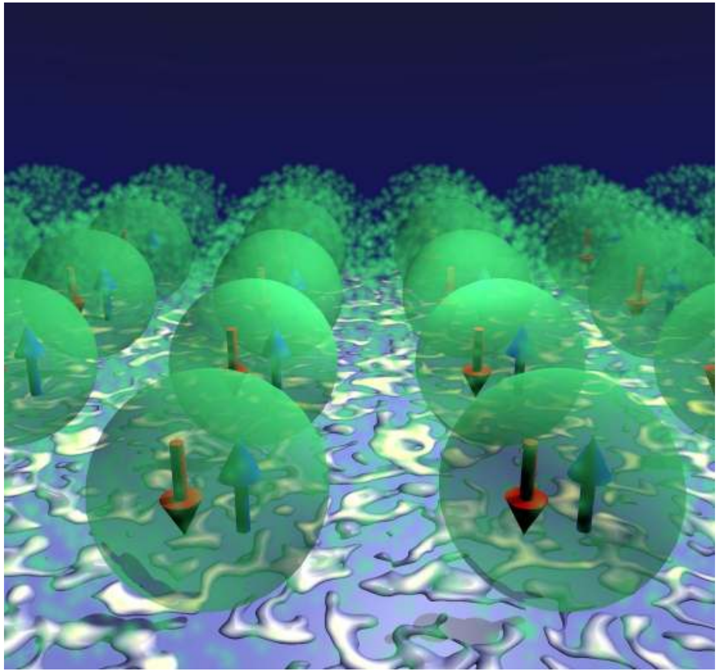
$$|\uparrow\uparrow\rangle$$

$$|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$$

S=1

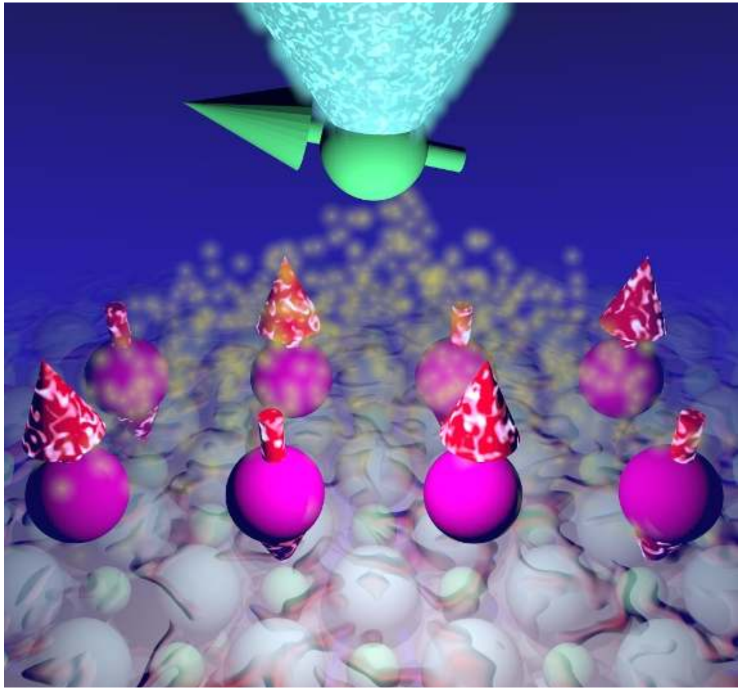
Plan for today

Triplons in a molecular quantum magnet



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

Hamiltonian learning nanoscale magnets



Phys. Rev. Applied 20, 024054 (2023)

Behind the scenes

Triplons in a molecular quantum magnet

Robert Drost

Shawulienu Kezilebieke

Peter Liljeroth

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

Hamiltonian learning nanoscale magnets

Netta Karjalainen

Zina Lippo

Rouven Koch

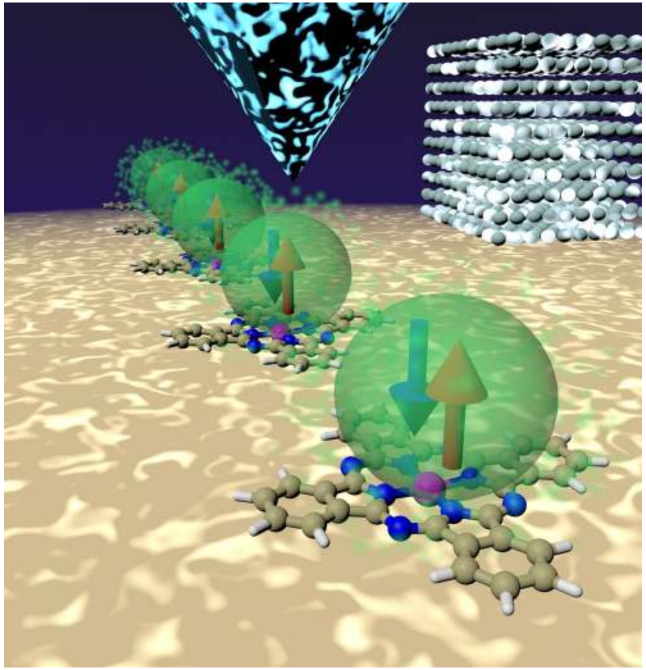
Guangze Chen

Adolfo Fumega

Phys. Rev. Applied 20, 024054 (2023)

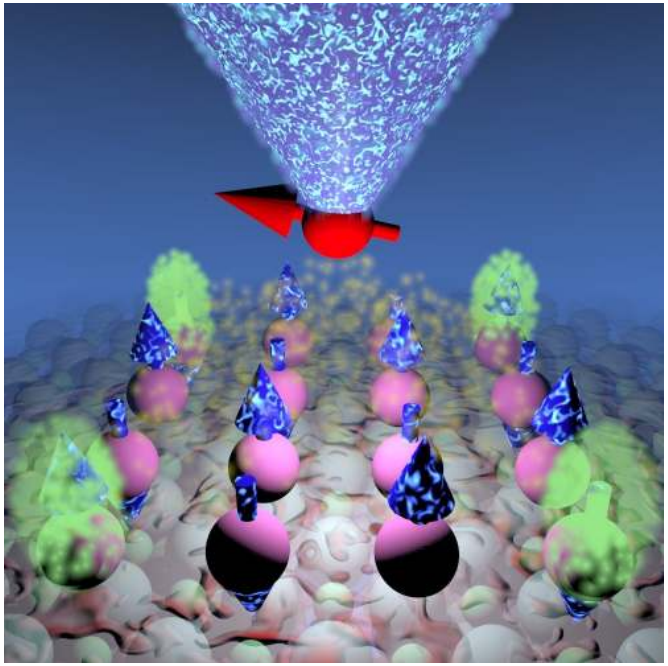
Plan for today

Hamiltonian learning triplons



arXiv:2504.20711 (2025)

Realization of a higher order topological quantum magnet



Nature Nanotechnology 19, 1782–1788 (2024)

Behind the scenes

Hamiltonian learning triplons

Rouven Koch

Robert Drost

Peter Liljeroth

arXiv:2504.20711 (2025)

Realization of a higher order topological quantum magnet

Hao Wang

Jing Chen

Lili Jiang

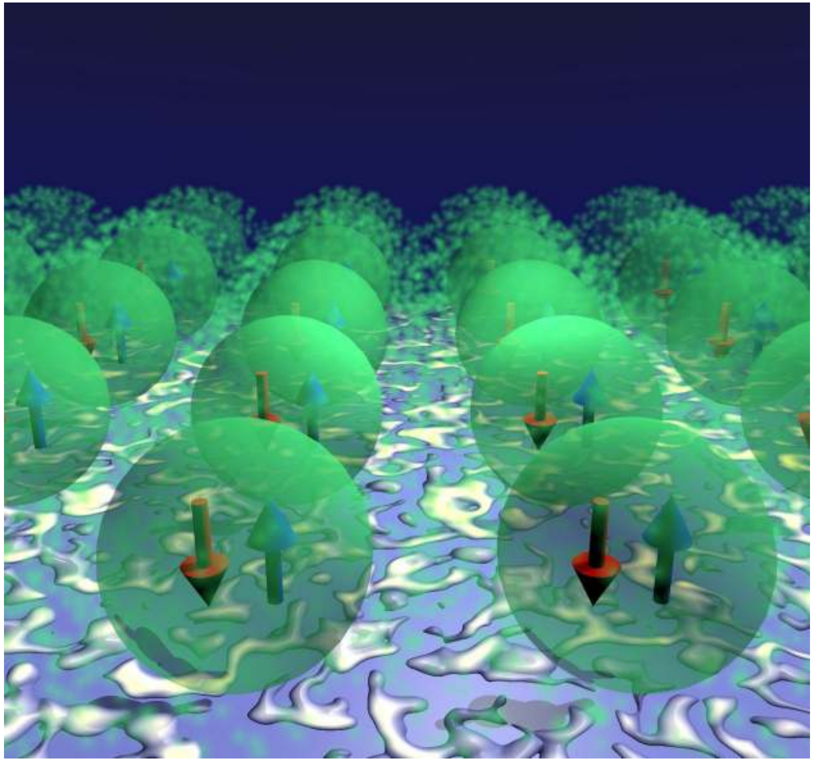
Peng Fang

Hong-Jun Gao

Kai Yang

Nature Nanotechnology 19, 1782–1788 (2024)

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

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

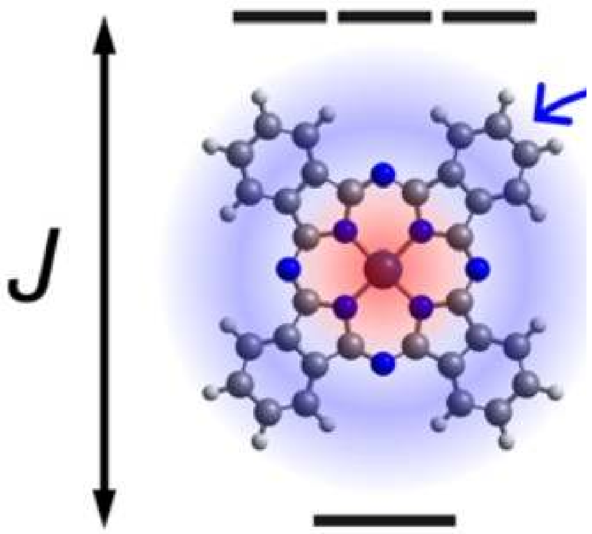
The ground state is unique, and does not break time-reversal

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

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

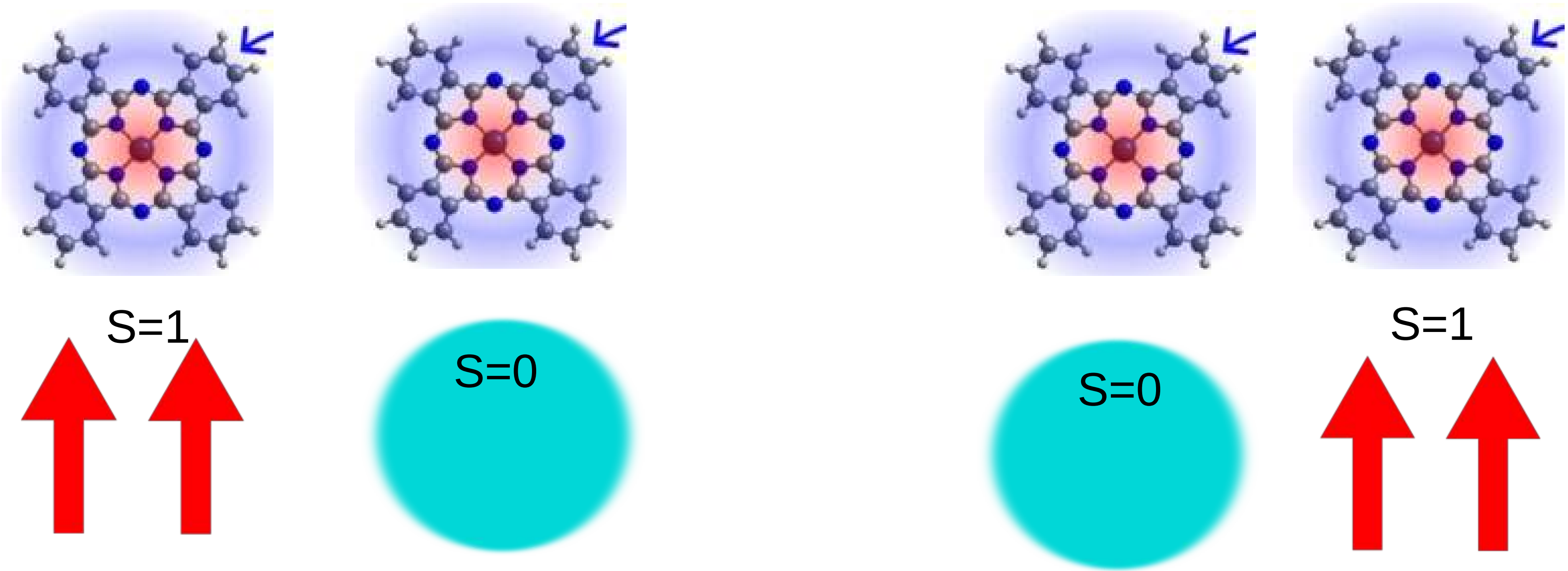
S = 0

Internal antiferromagnetic coupling

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

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

Triplons in a dimer

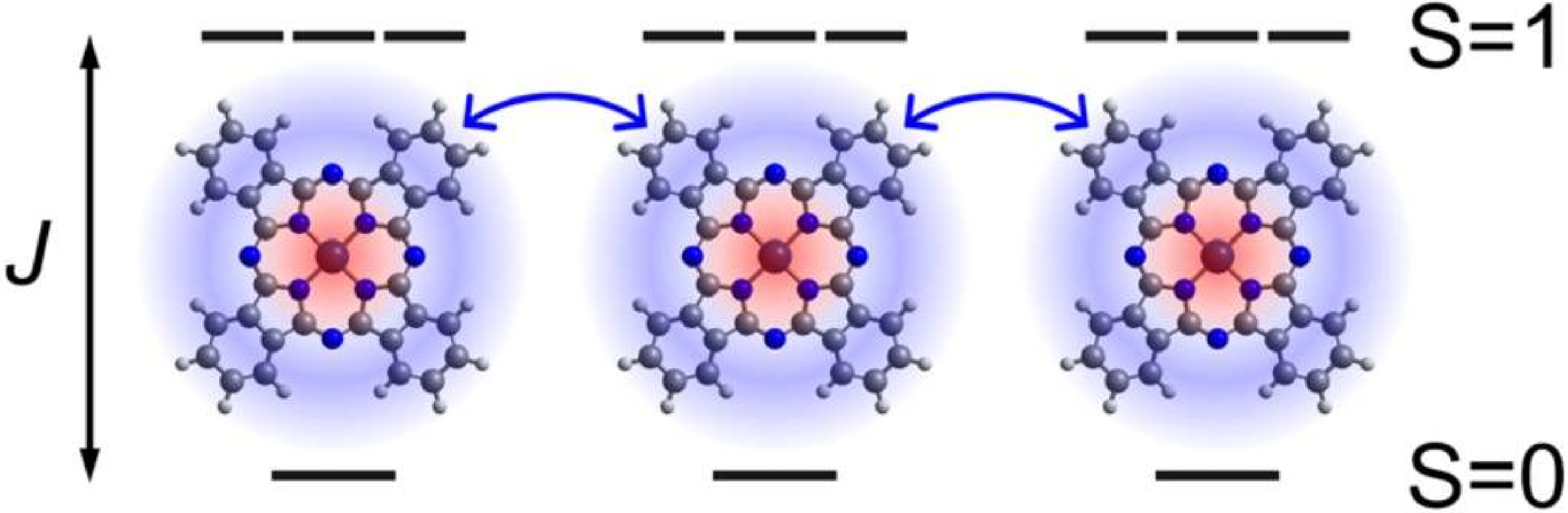


A triplet in the first molecule

Can jump to the second molecule

Exchange coupling between molecules leads to propagating triplon excitations

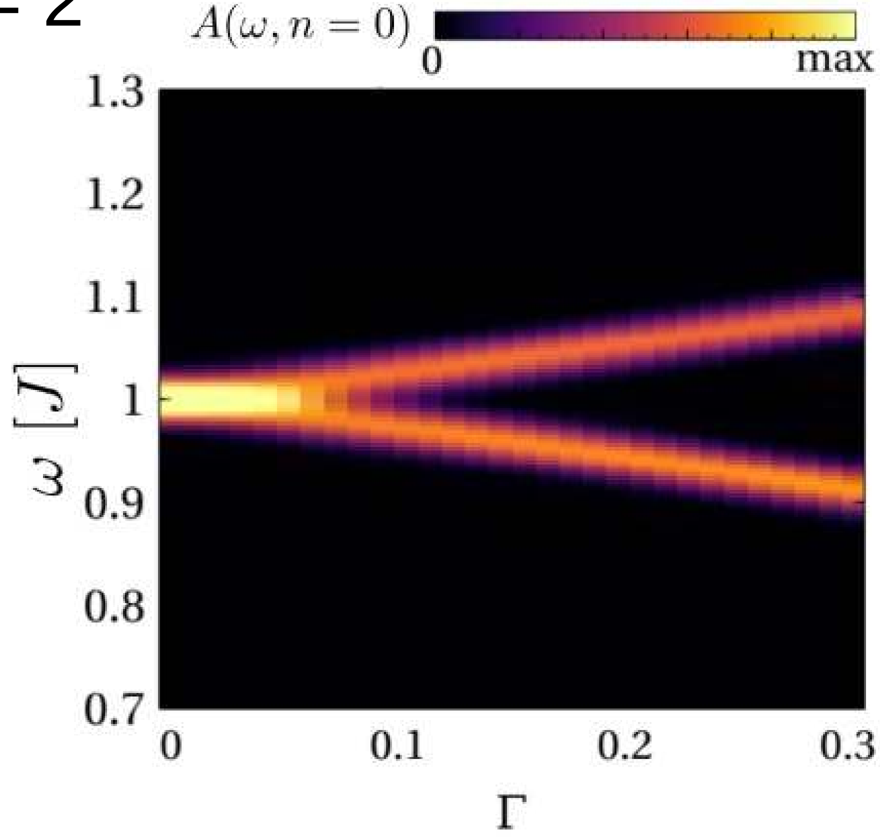
A chain of triplons



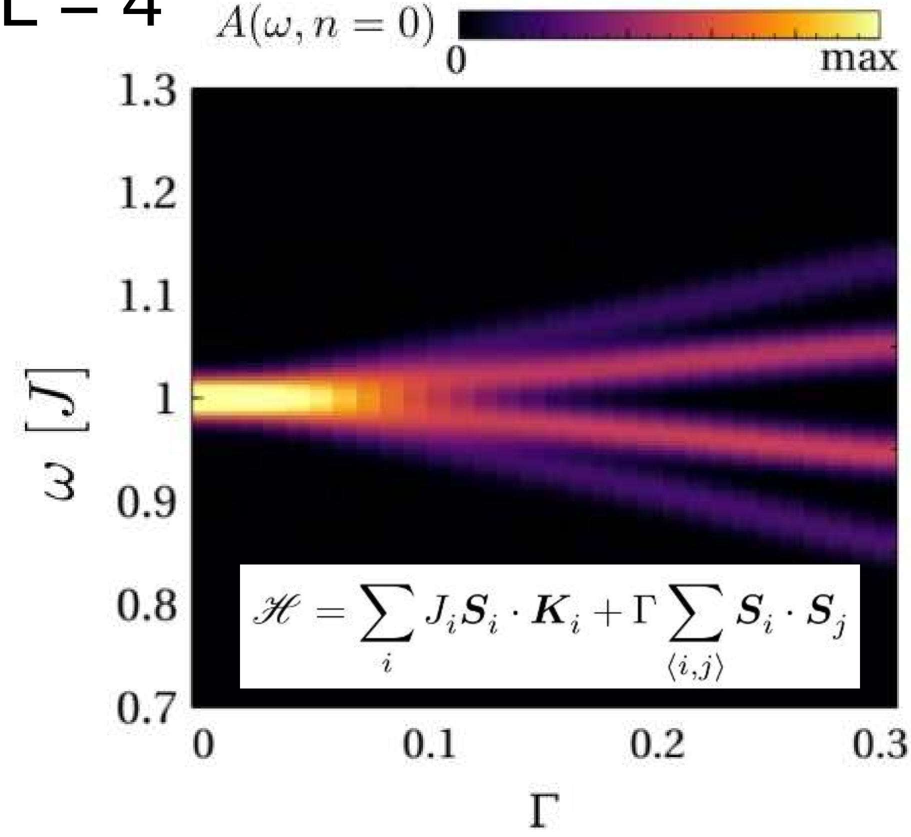
$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Chains of triplons

L = 2

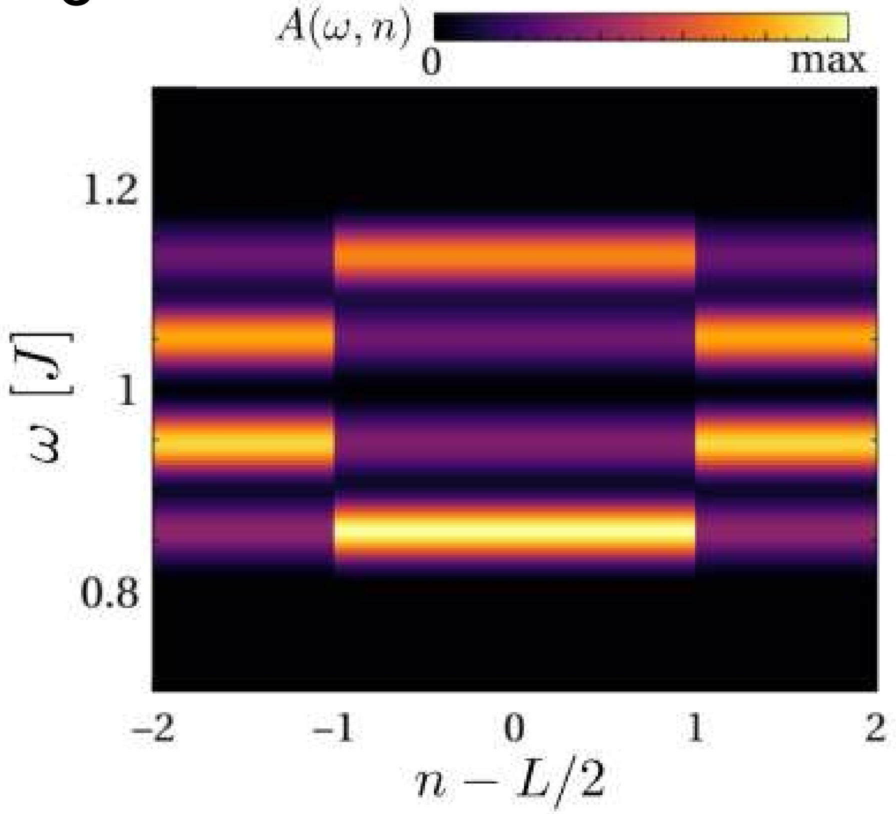


L = 4

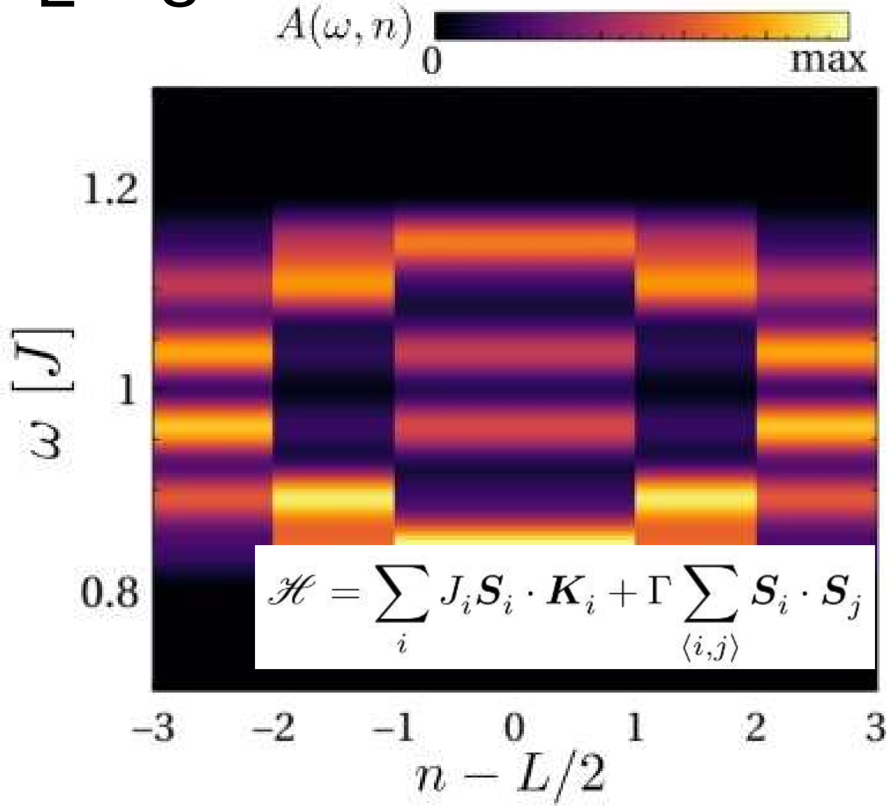


Chains of triplons

L = 6



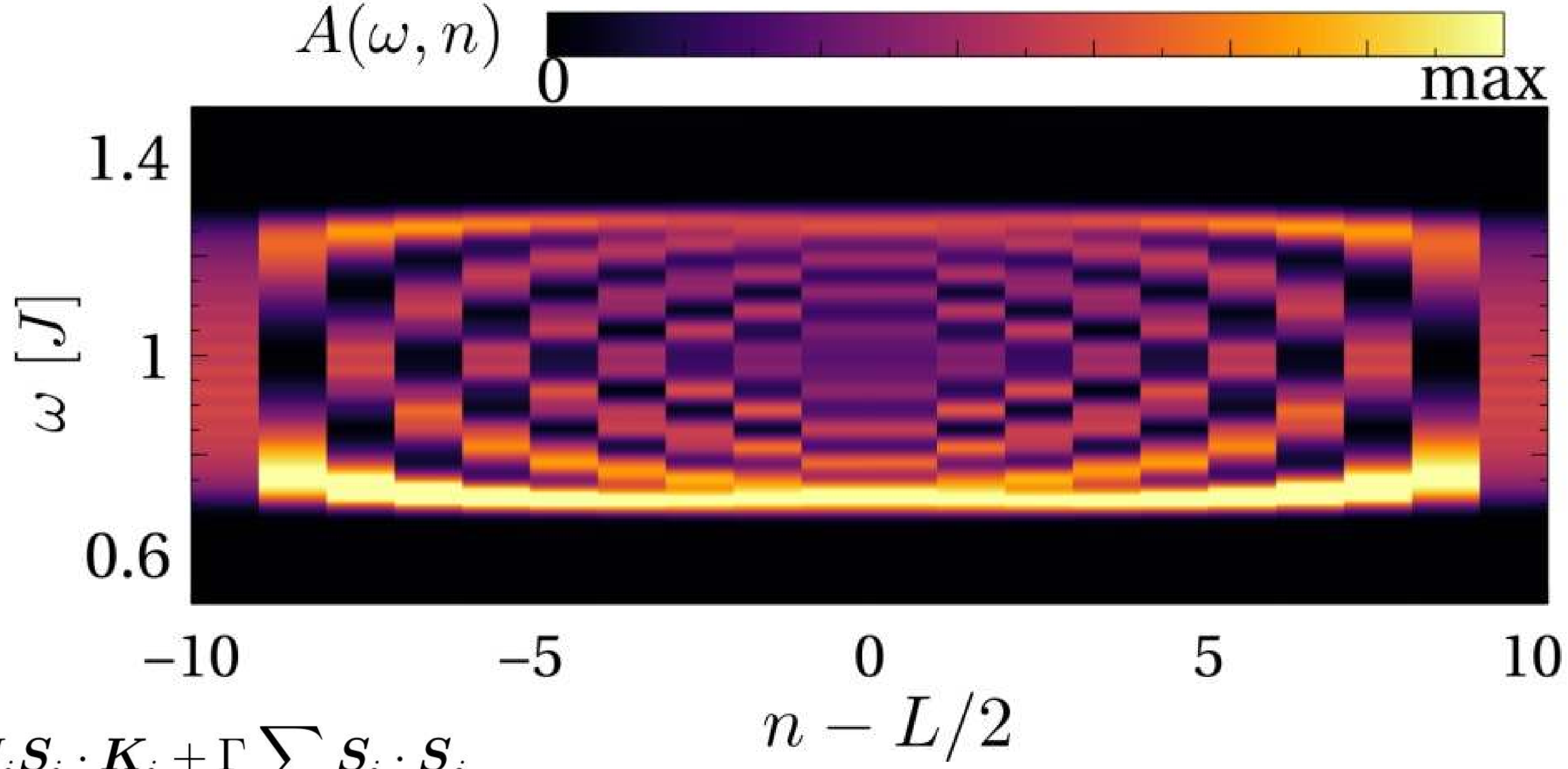
L = 8



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

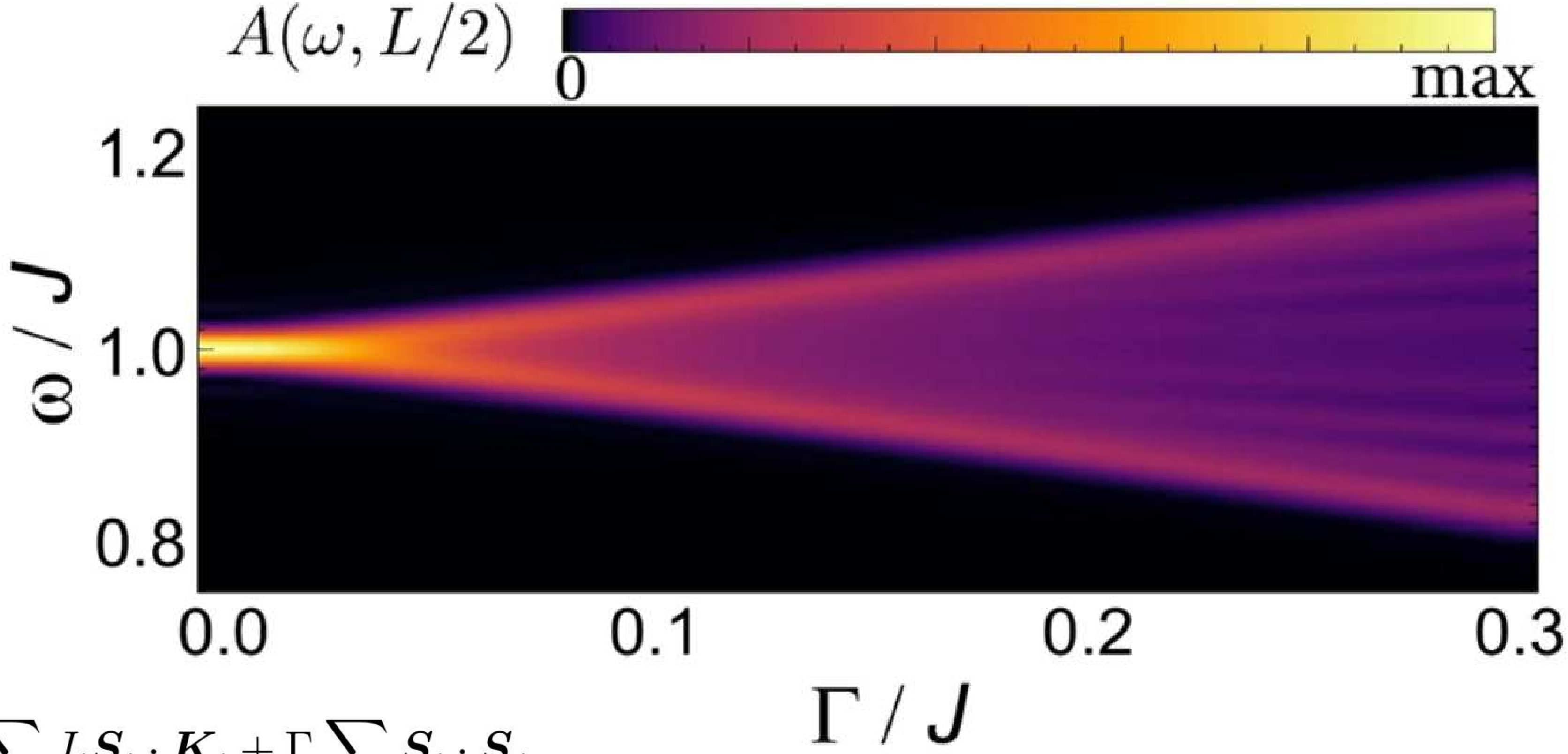
Chains of triplons

L = 20

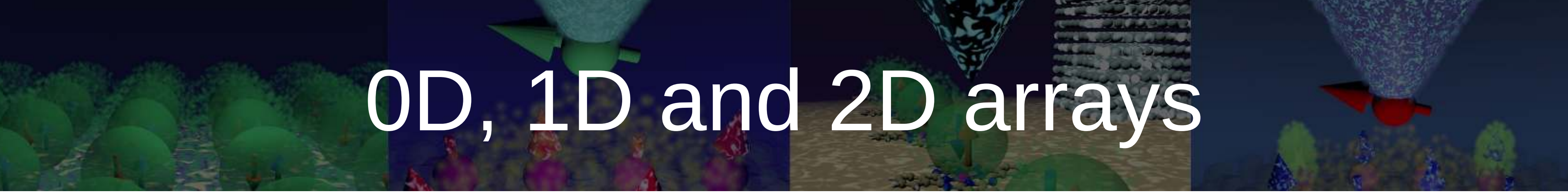


$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Chains of triplons

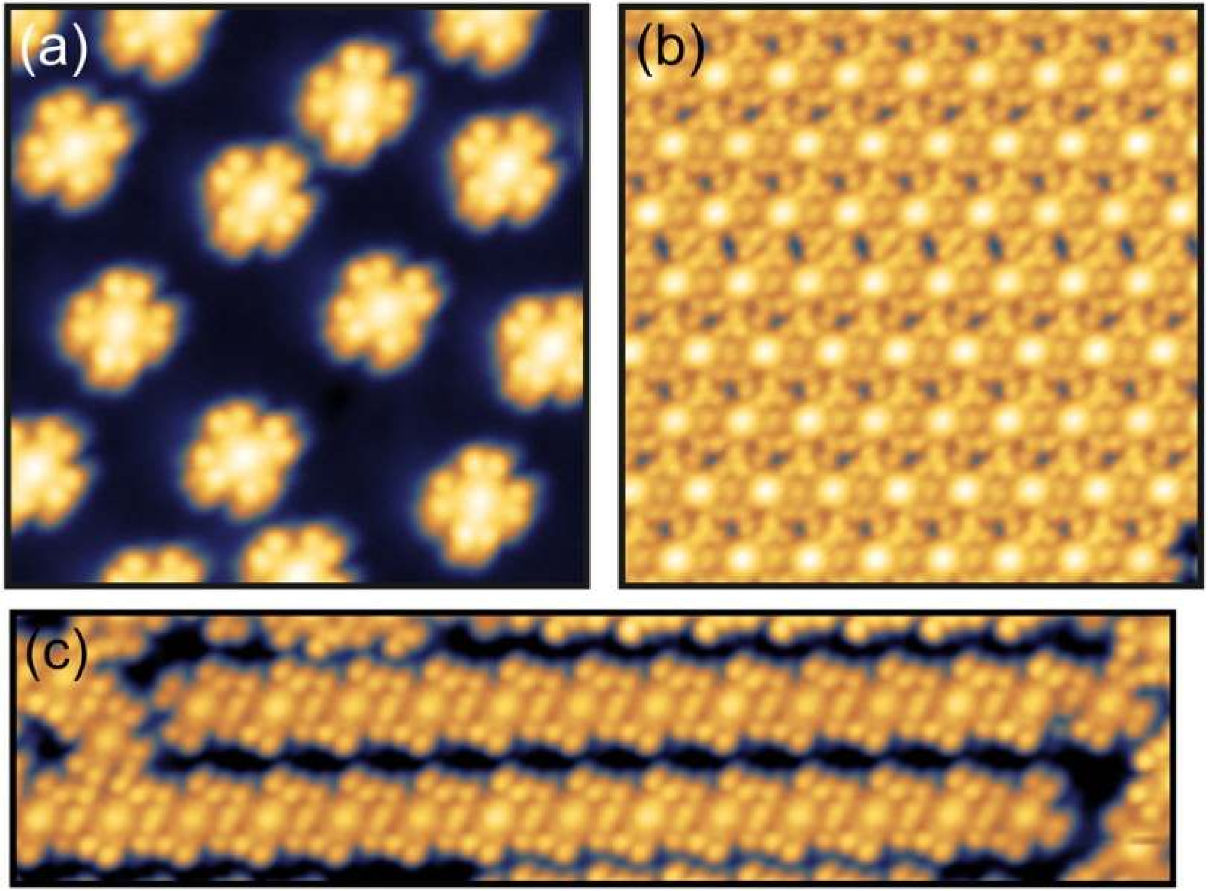


$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$



0D, 1D and 2D arrays

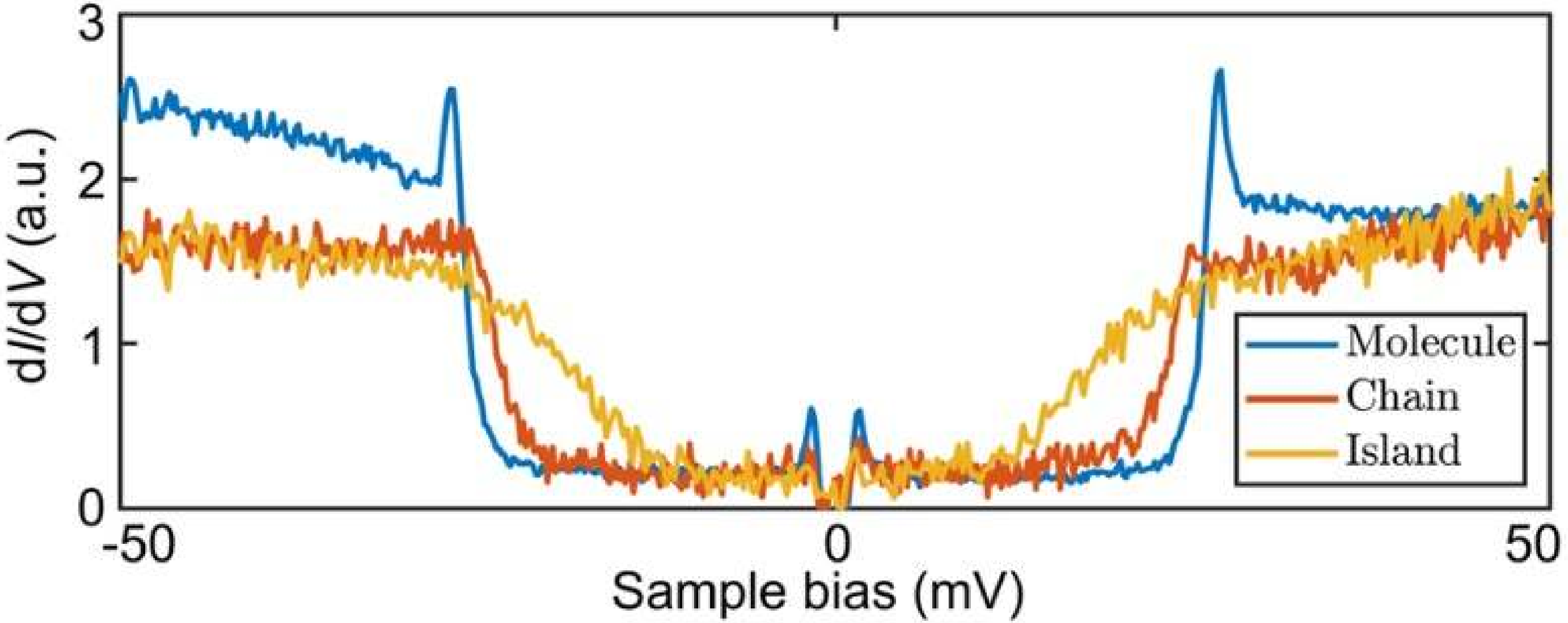
0D



2D

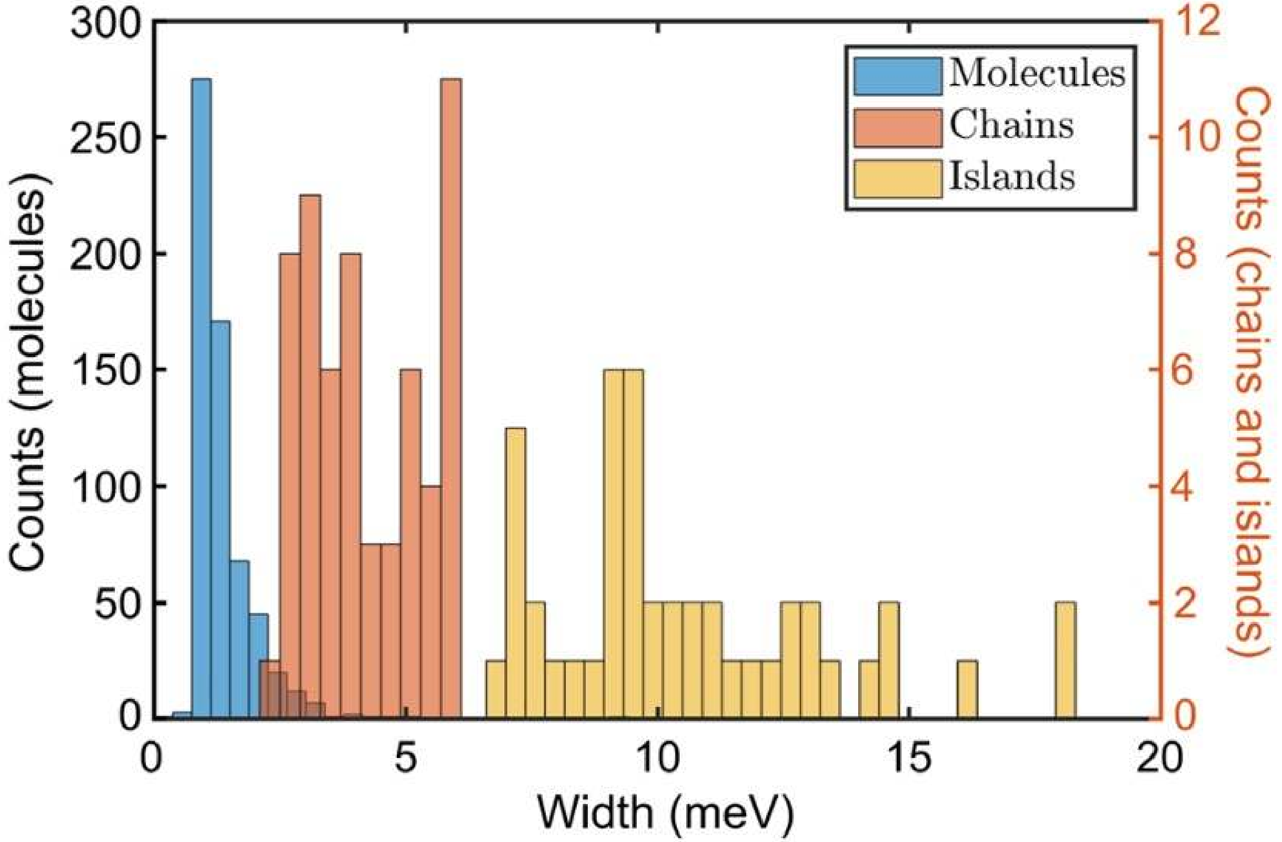
1D

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



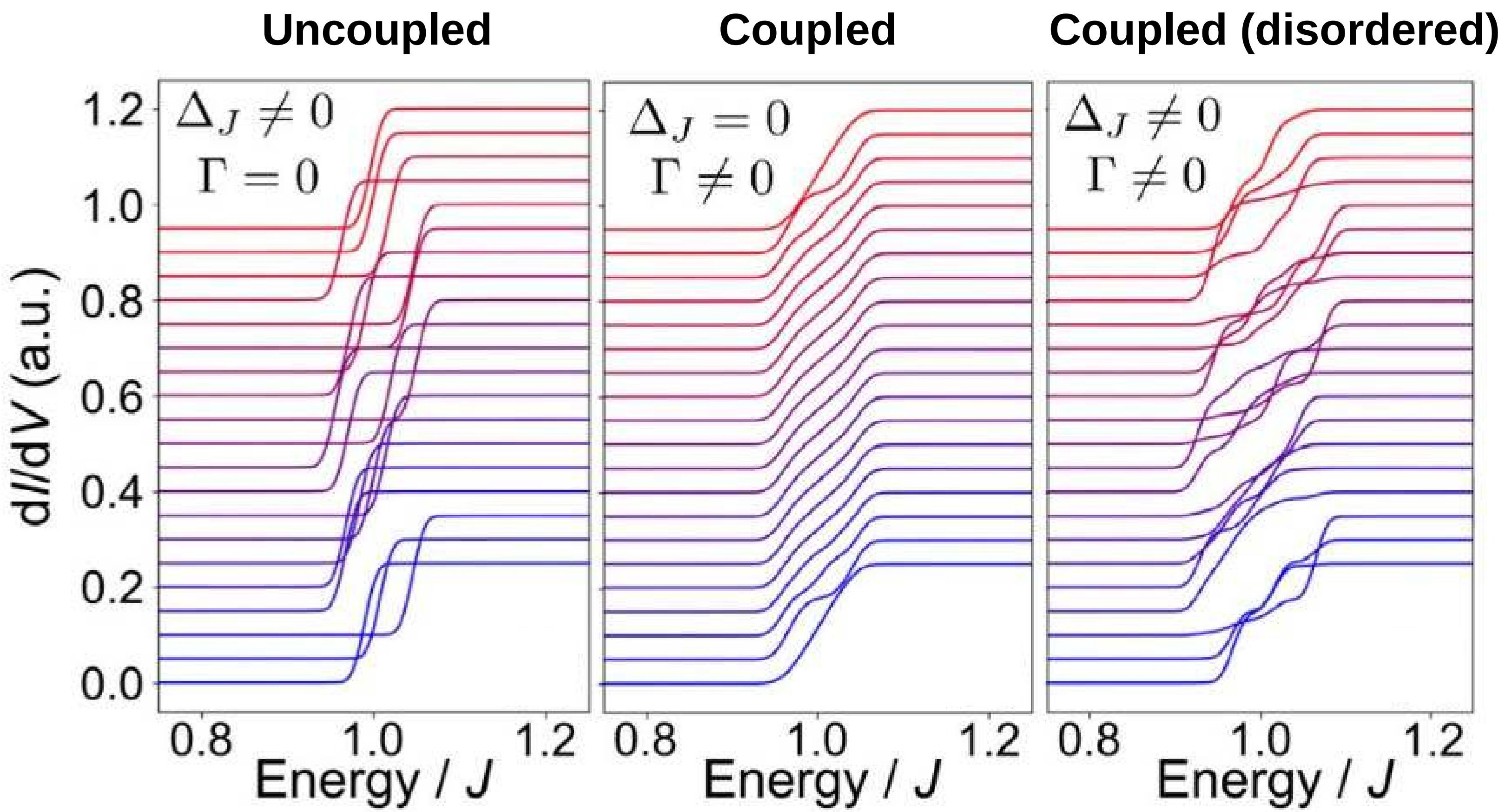
The width of the step is bigger for islands than for molecules

Statistic of the inelastic steps

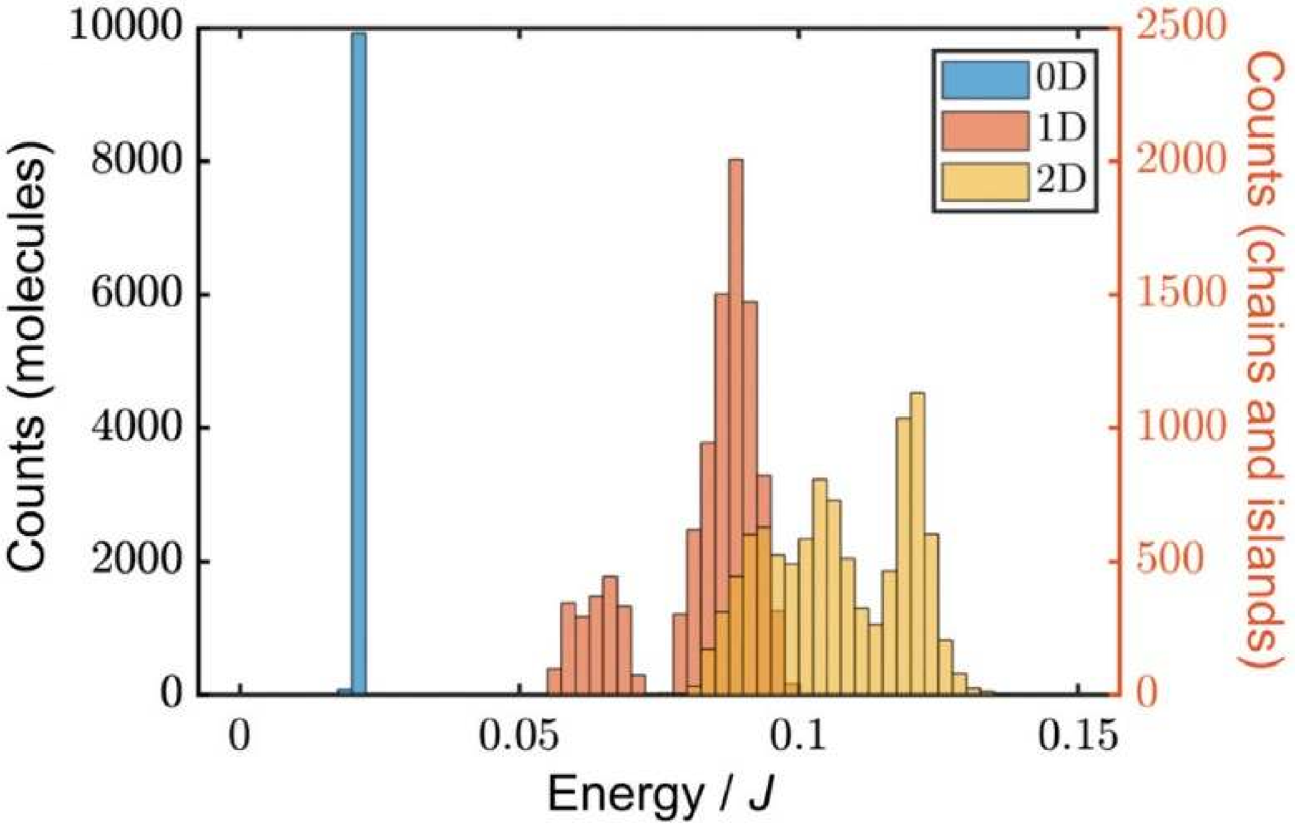


2D arrays have the largest width of the spin excitation

Coupled and uncoupled inelastic excitation

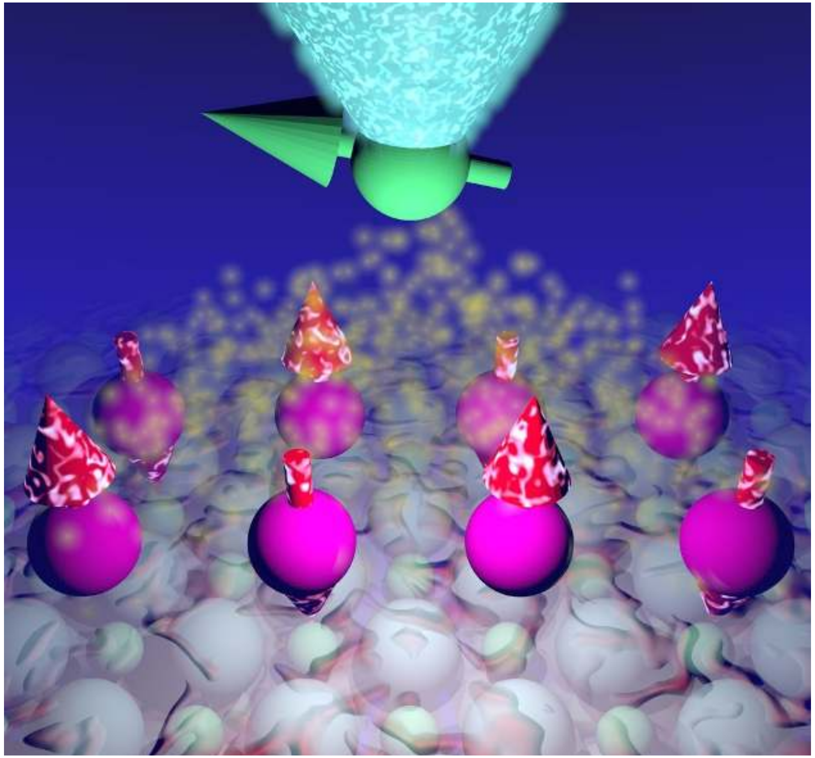


Statistics of the theoretical model



2D arrays have the largest width of the spin excitation

Hamiltonian learning nanoscale magnets



Hamiltonian learning nanoscale magnets

Netta Karjalainen

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Phys. Rev. Applied 20, 024054 (2023)

The problem of Hamiltonian learning

How can we go from observables to the Hamiltonian?

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 perform this task?

Can we use machine learning to solve problems in quantum matter?

Supervised learning

"Dog"

Labeled prediction

Generative learning

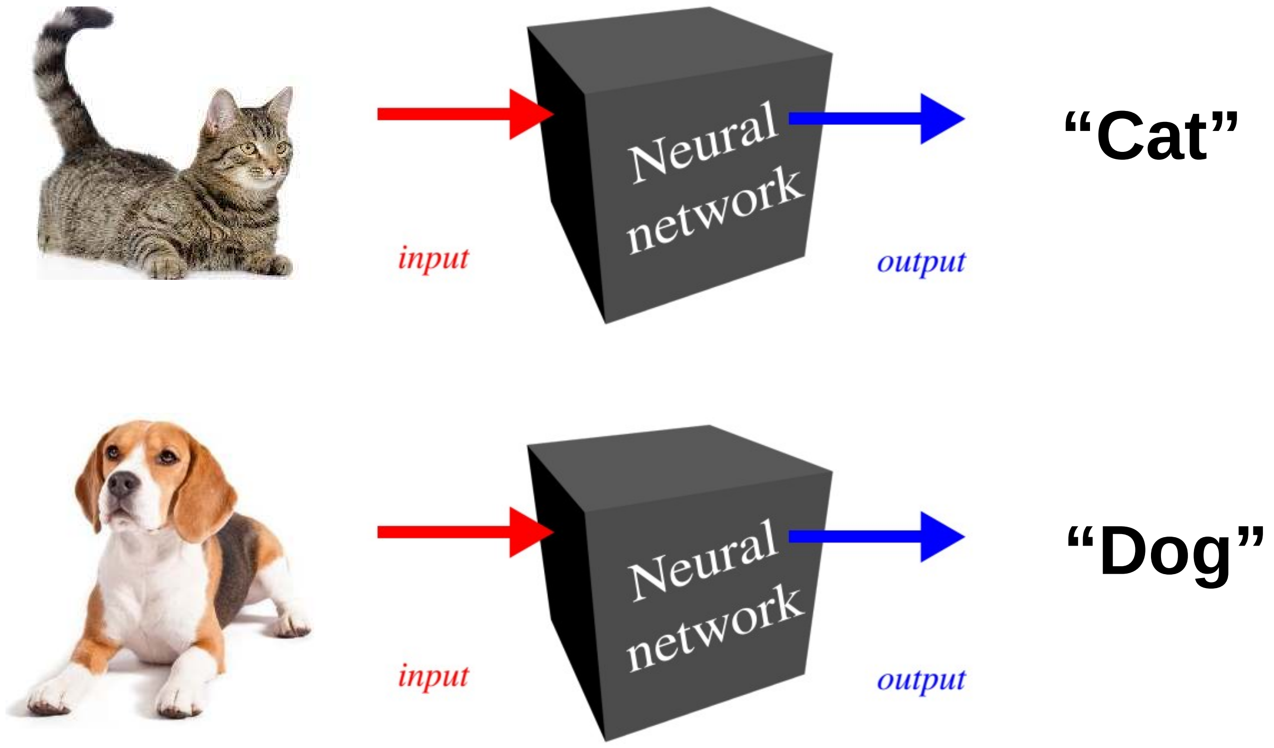
Probability Modeling

Reinforcement learning

Reward-based decision

(and others)

Machine learning in classification problems



Hamiltonian learning: combining theory and experiment

Train a machine learning algorithm with simulated data

so that the parameters of each Hamiltonian are explicitly know

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

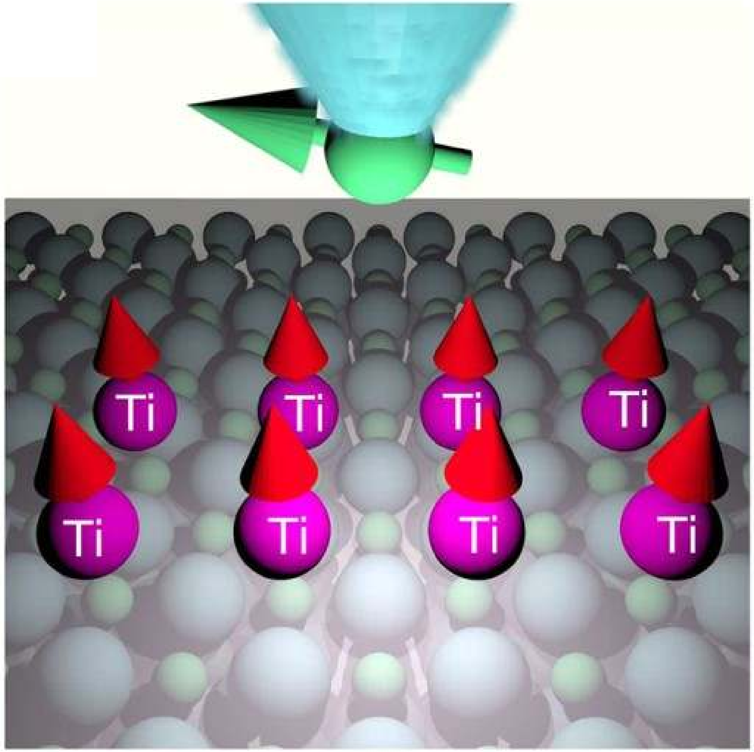
Hamiltonian

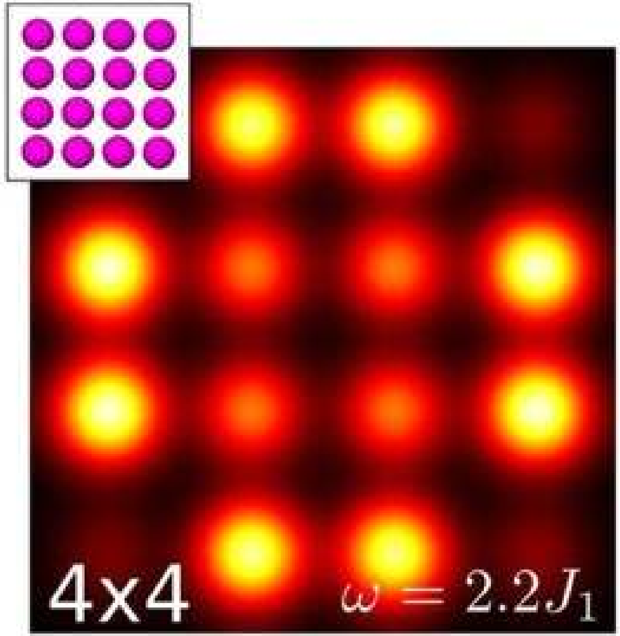
With the trained algorithm, evaluated in experimental data to extract the parameters

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

Hamiltonian

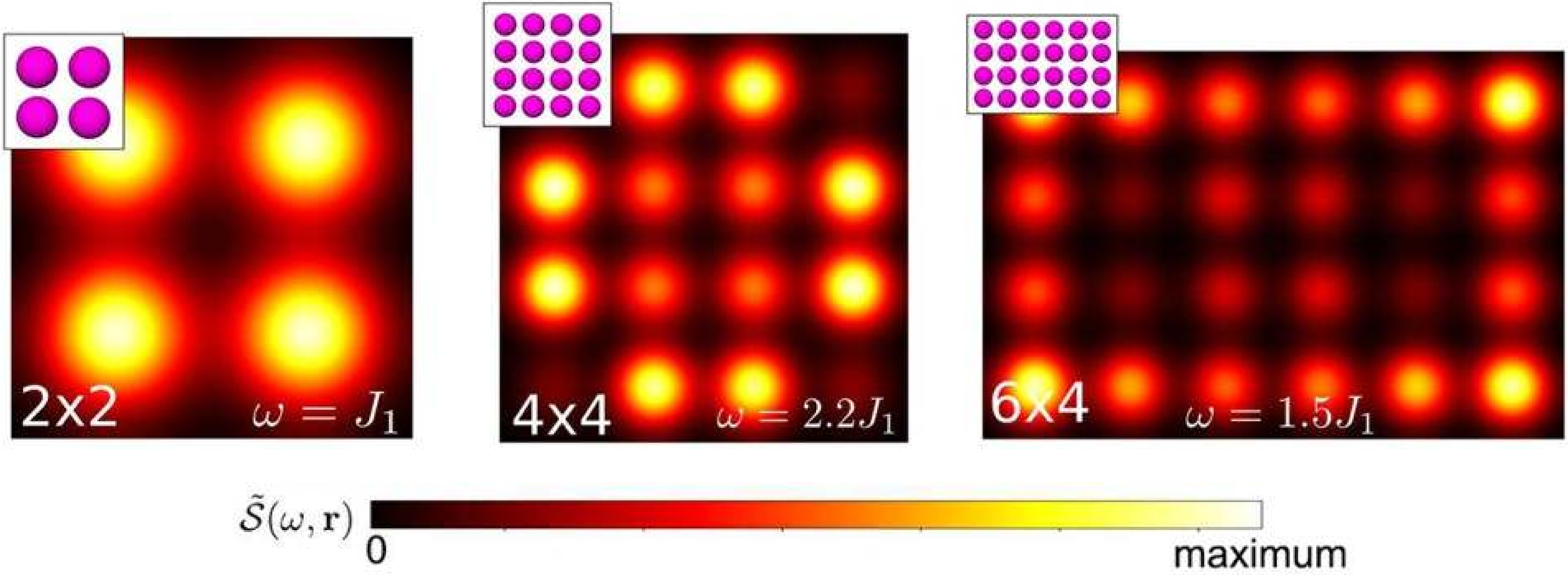
Spatially resolved excitations





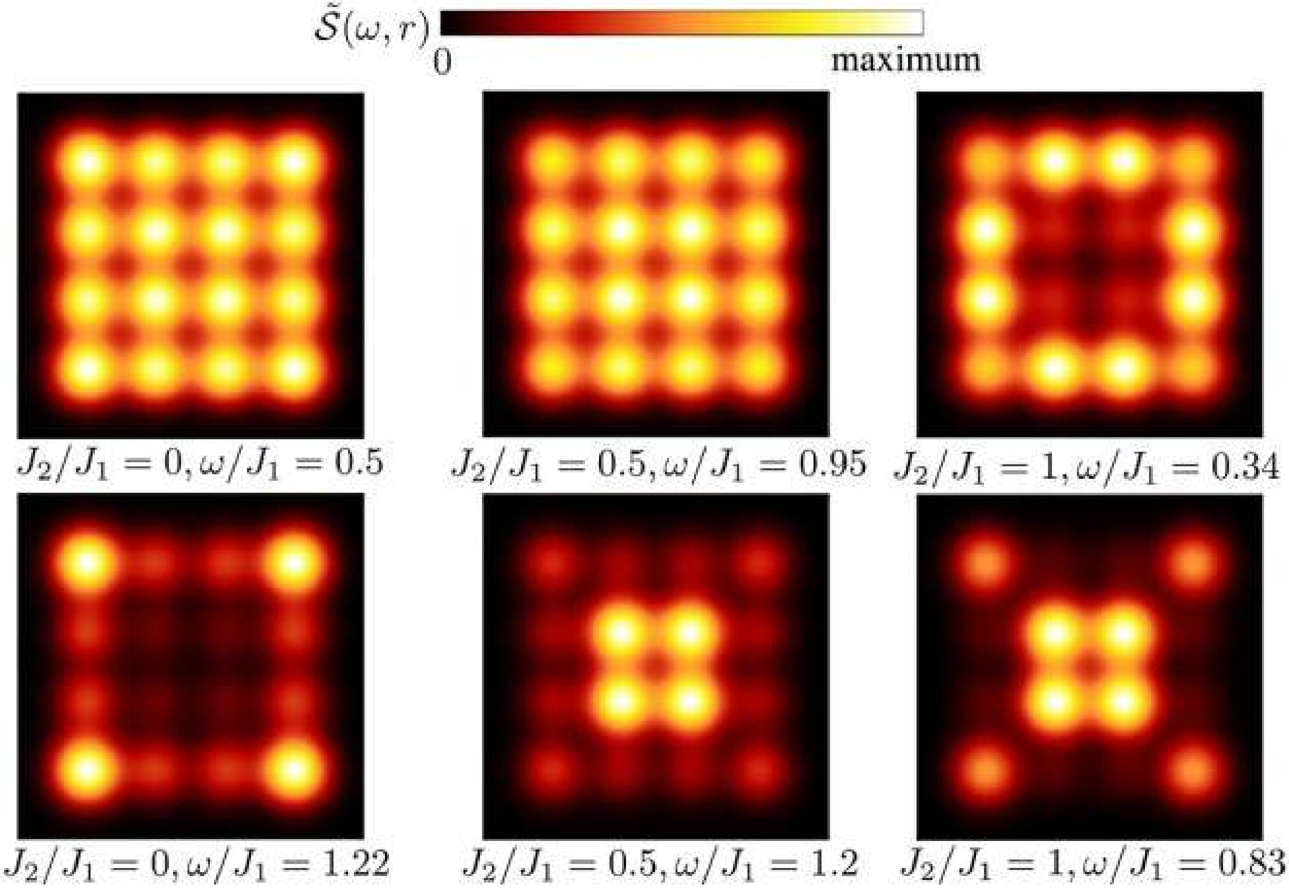
What can we learn from the Hamiltonian using real-space information?

Spatially resolved excitations



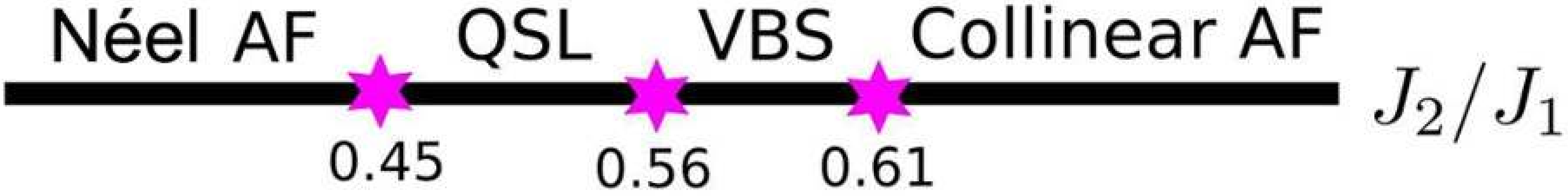
Spin excitations behave like particle in a box states

Spin excitation modes in spin islands



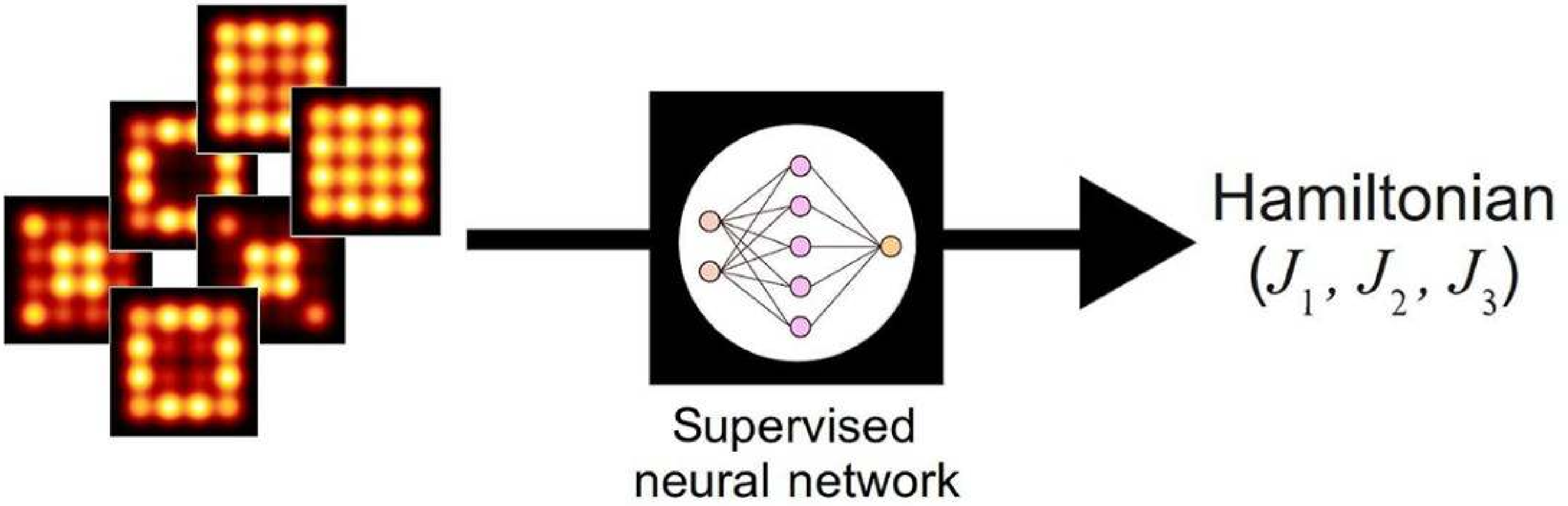
The extended Heisenberg model

$$\mathcal{H} = J_1 \sum_{\langle \mathbf{r}_i , \mathbf{r}_j \rangle} \mathbf{S}_{\mathbf{r}_i} \cdot \mathbf{S}_{\mathbf{r}_j} + J_2 \sum_{\langle\langle \mathbf{r}_i , \mathbf{r}_j \rangle\rangle} \mathbf{S}_{\mathbf{r}_i} \cdot \mathbf{S}_{\mathbf{r}_j}$$



Can we learn the exchange couplings from atomic-scale spin islands?

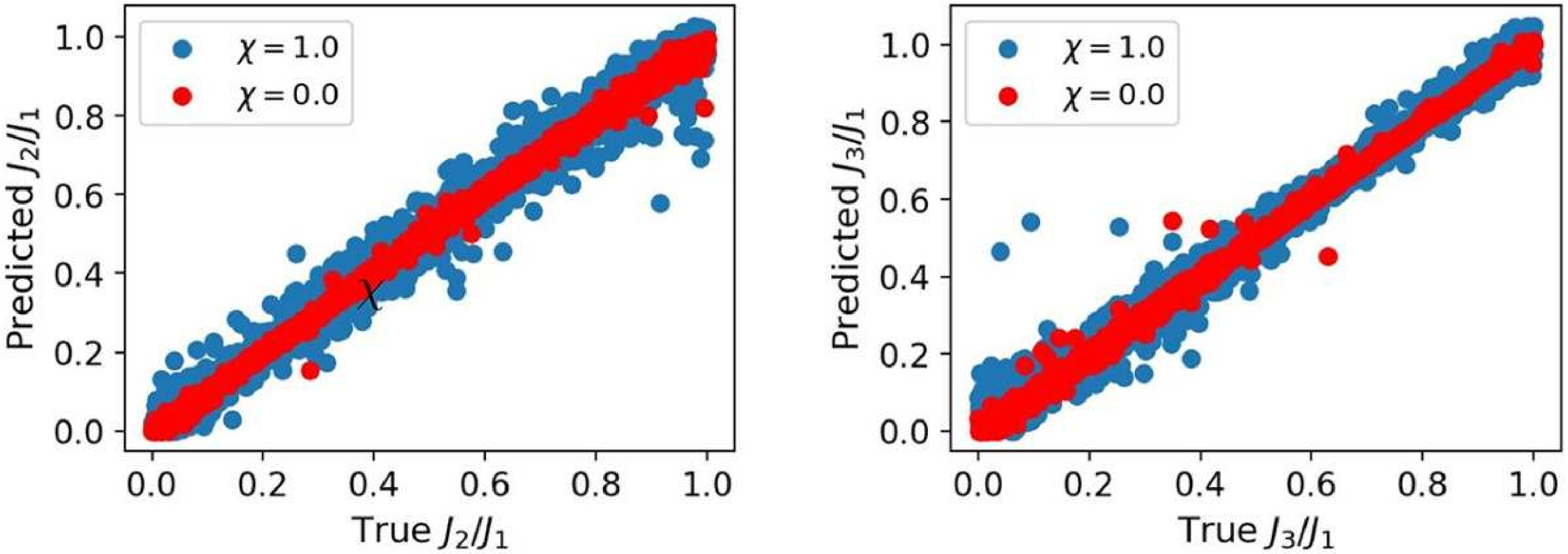
The idea of Hamiltonian learning



Provide the spectroscopy as input, obtain the Hamiltonian as output

Real VS predicted exchange parameters

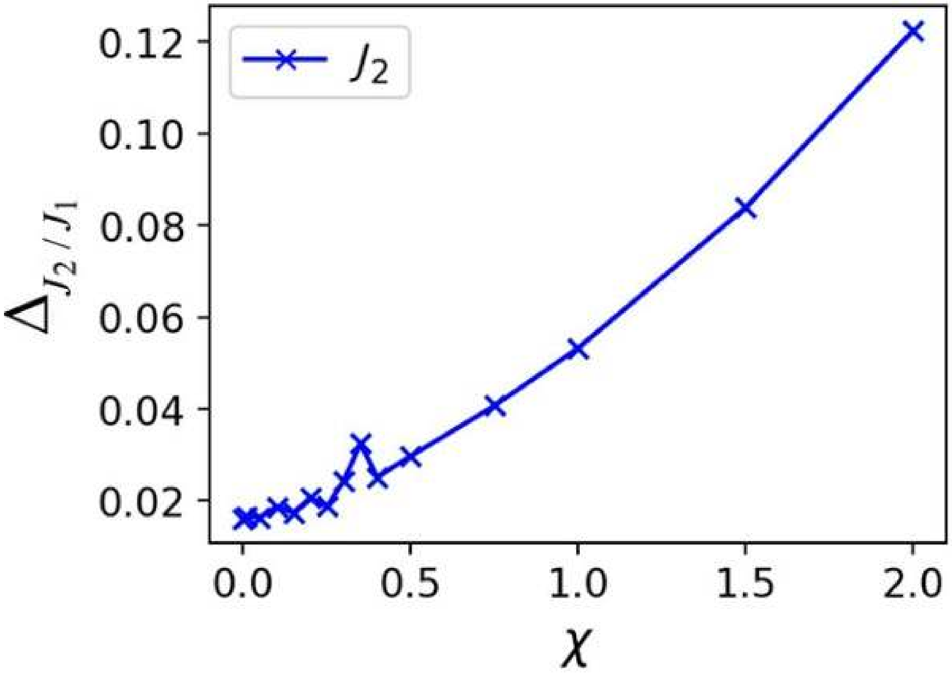
With noise $$\mathcal{S}(\omega, \mathbf{r}_i)_{\text{noisy}} = \mathcal{S}(\omega, \mathbf{r}_i) \cdot |1 + \zeta(\omega)| \quad \zeta(\omega) \in [-\chi, \chi]$$

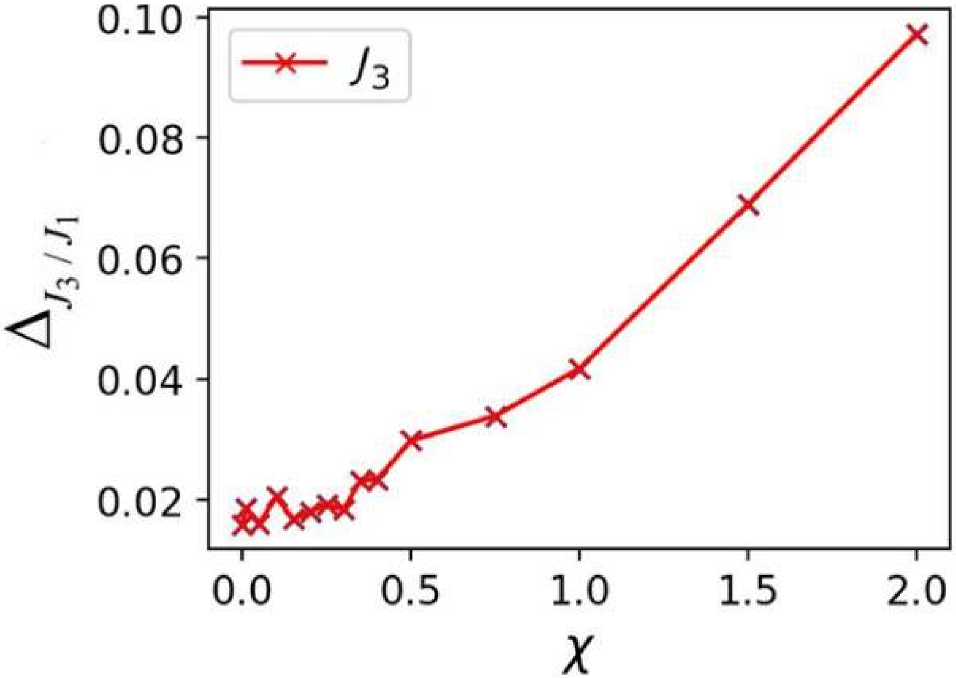


A supervised learning algorithm extracts faithfully the exchange parameters

Predictions with noise

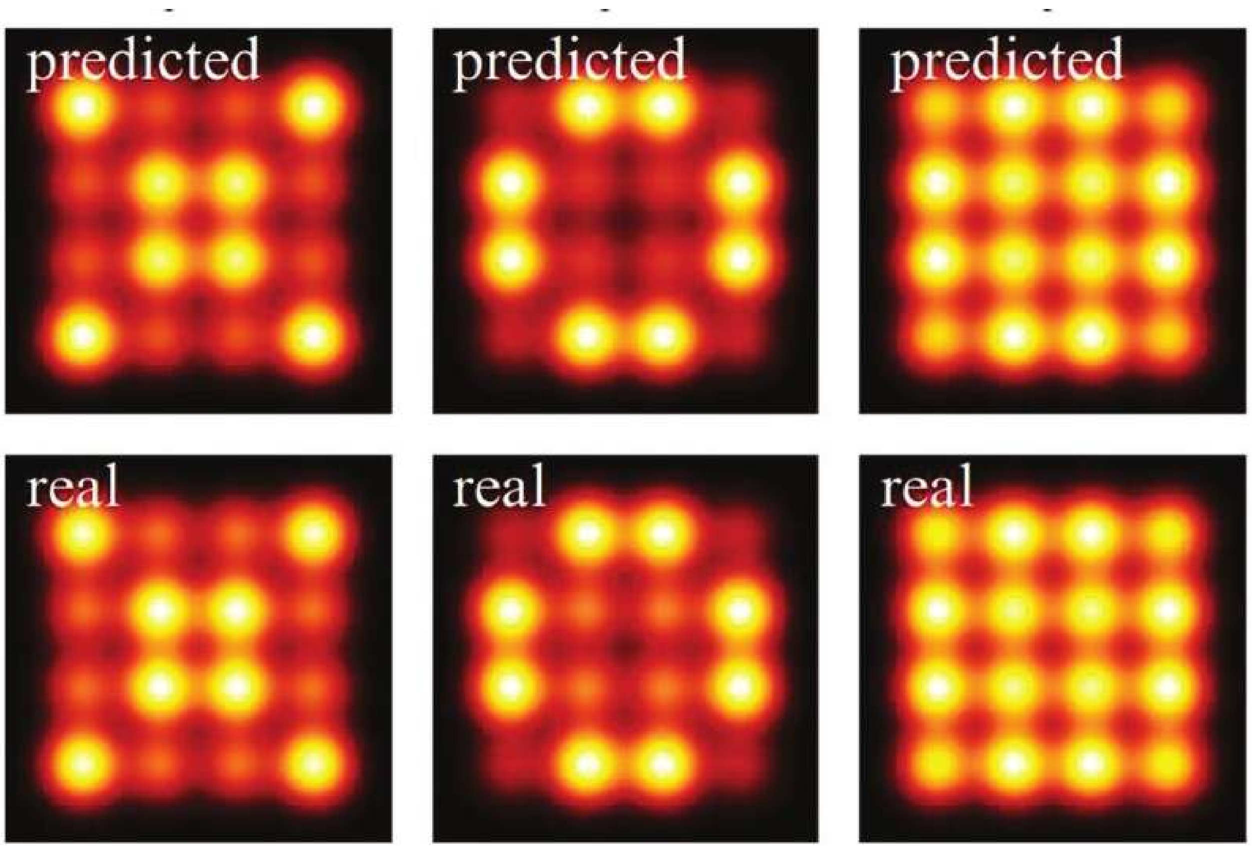
$$\mathcal{S}(\omega, \mathbf{r}_i)_{\text{noisy}} = \mathcal{S}(\omega, \mathbf{r}_i) \cdot |1 + \zeta(\omega)| \qquad \zeta(\omega) \in [-\chi, \chi]$$



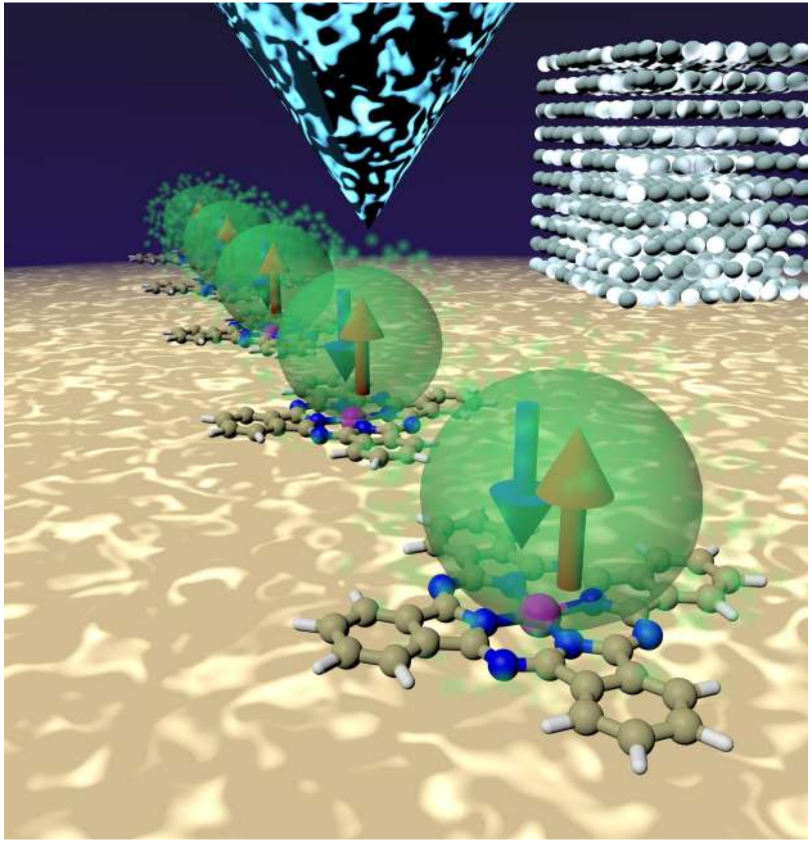


Quality of prediction remain good with sizable noise levels

Predicted VS real spin excitations



Hamiltonian learning triplons in a molecular quantum magnet



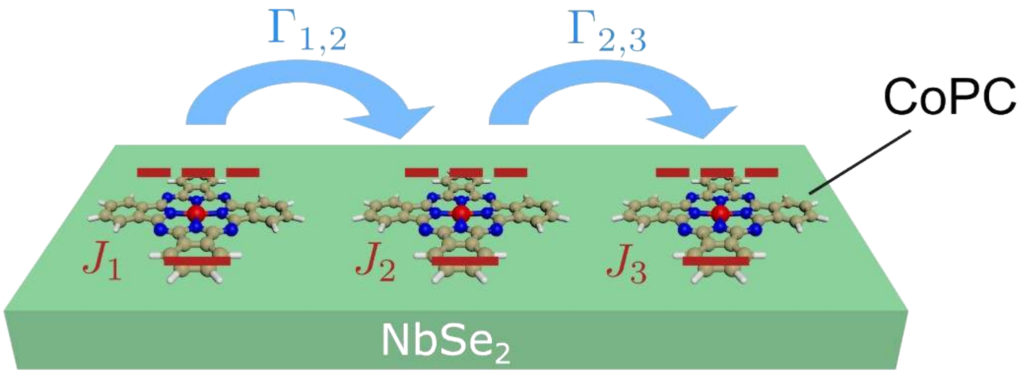
Rouven Koch

Robert Drost

Peter Liljeroth

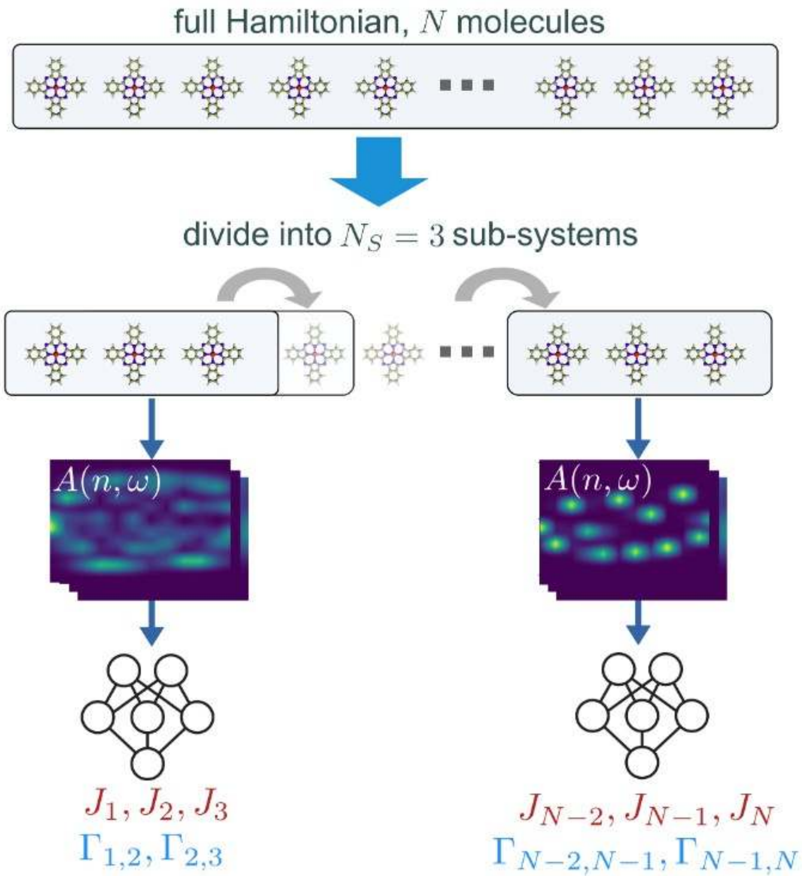
arXiv:2504.20711 (2025)

A strategy to machine learning quantum magnets



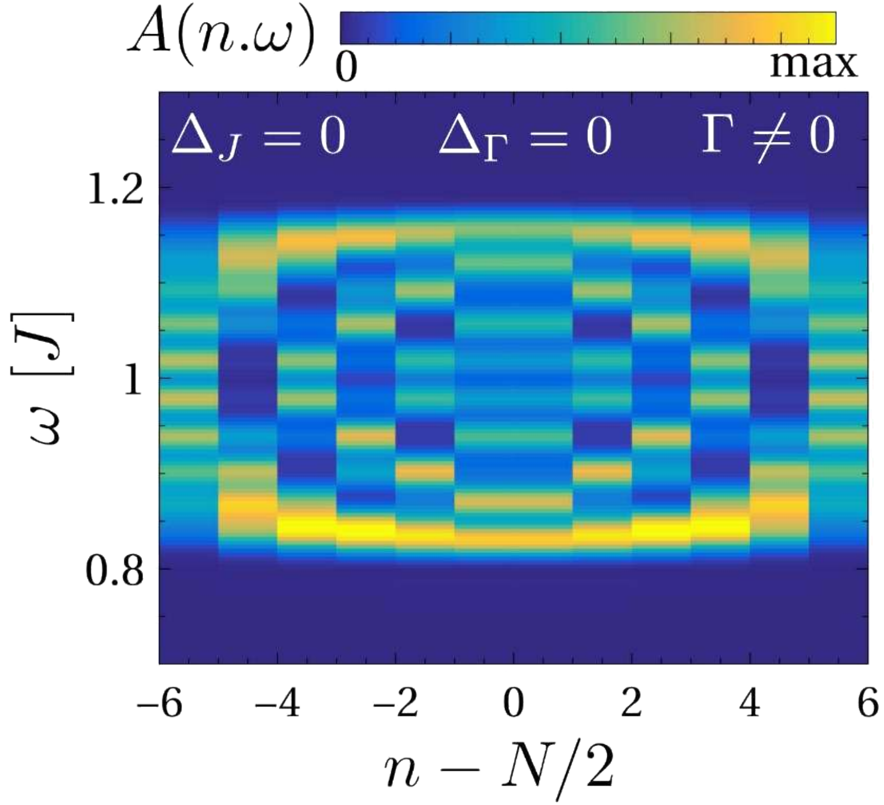
$\Gamma_{n,n+1}$: intermolecular exchange
J_n : intramolecular exchange

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

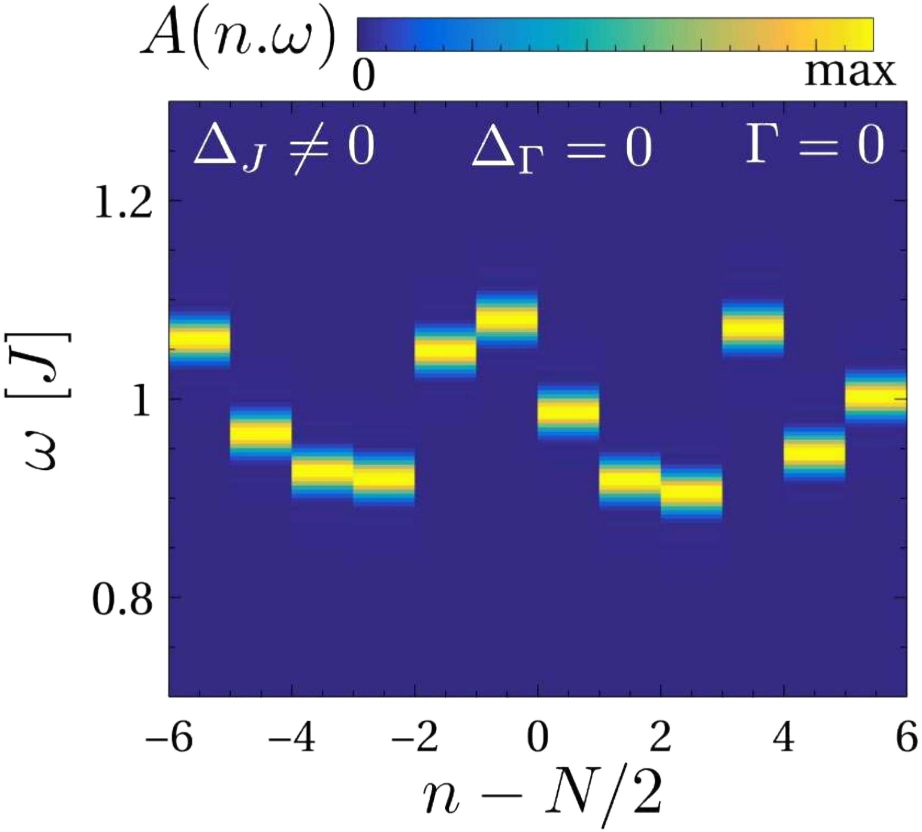


Pristine triplon excitations

Pristine and coupled molecules

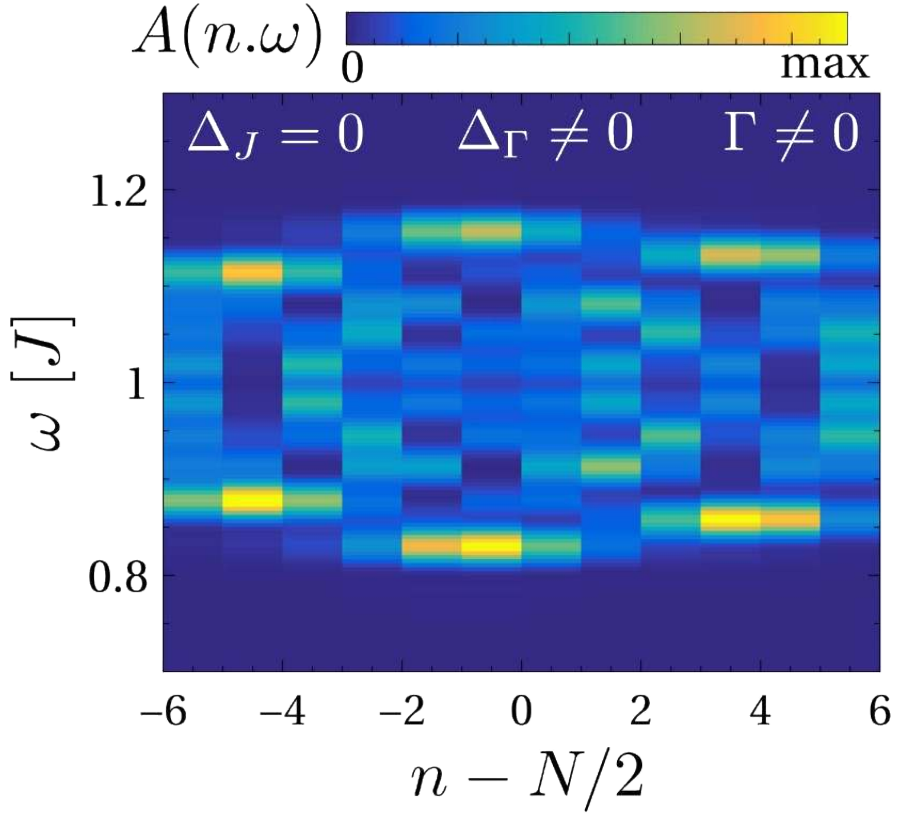


Decoupled and disordered molecules

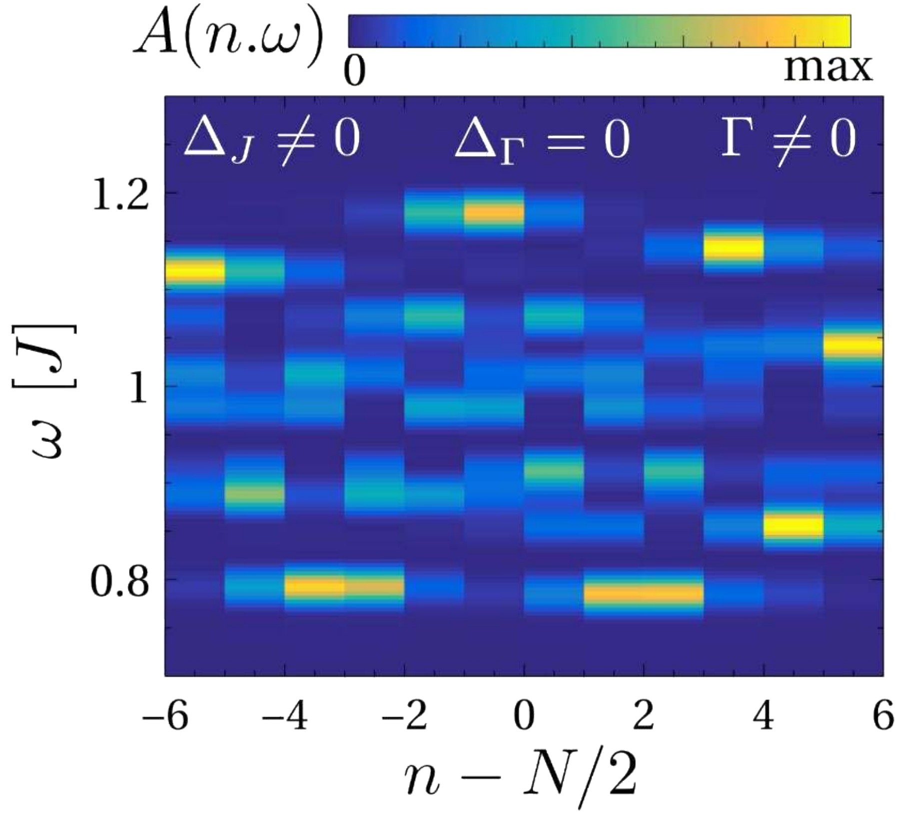


The different impact of molecular disorder

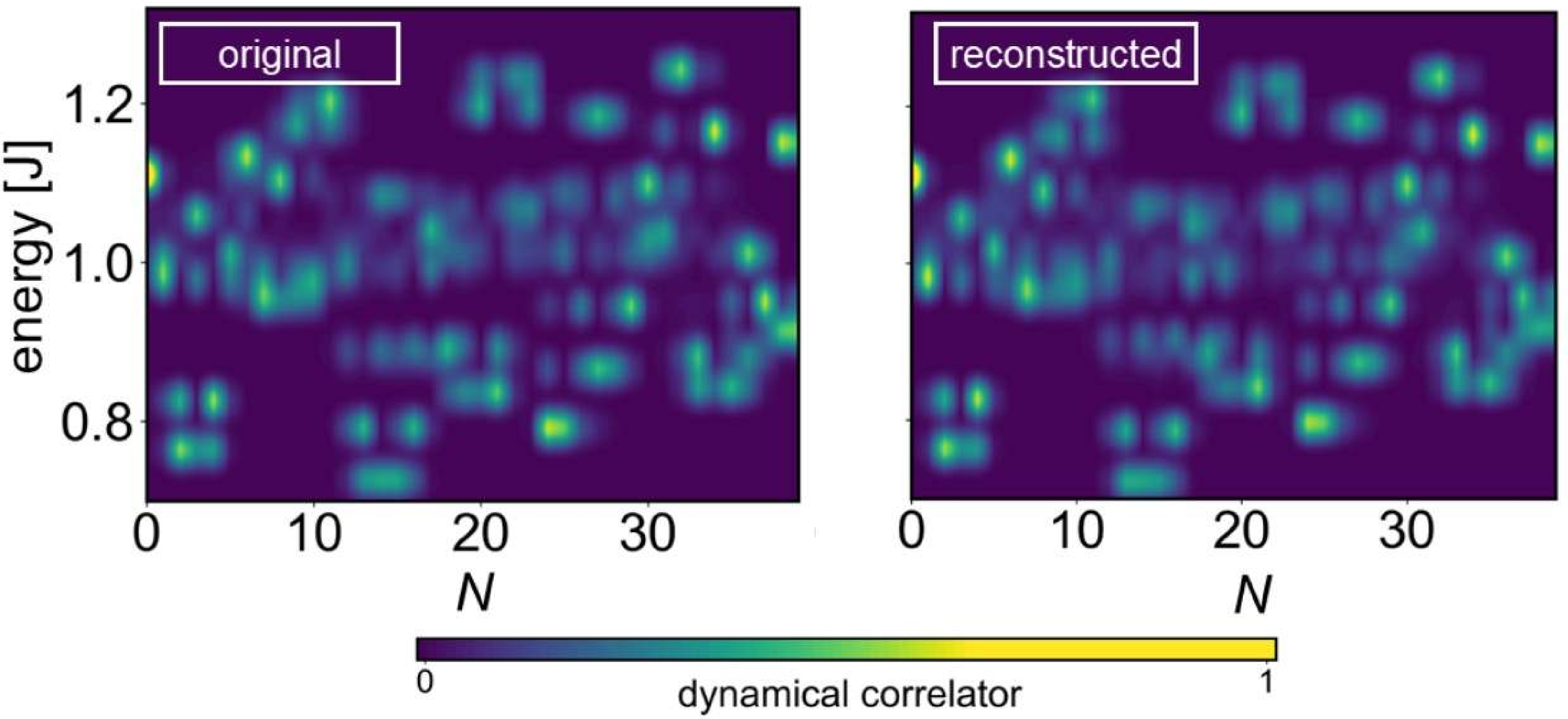
Disorder in inter-molecule coupling



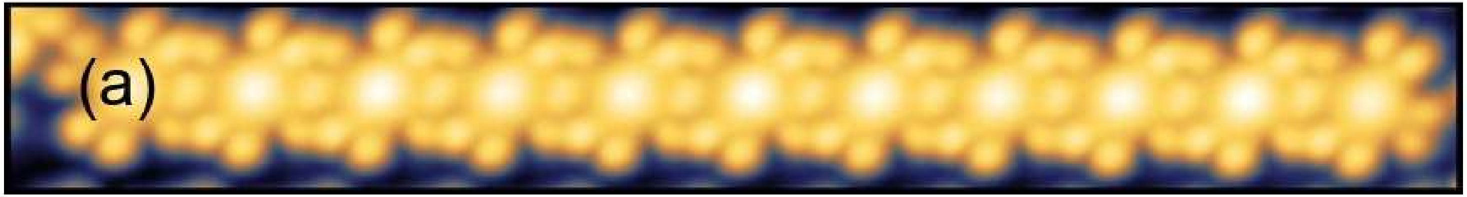
Disorder in intra-molecule coupling

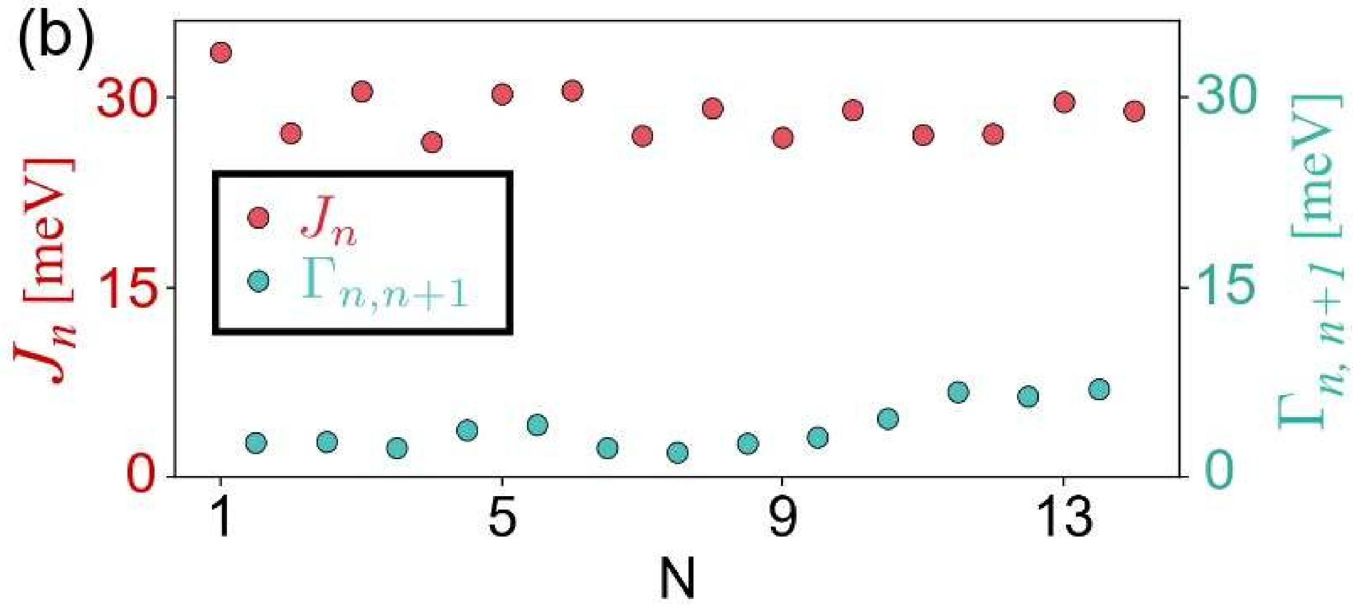


Reconstructing a quantum magnet Hamiltonian

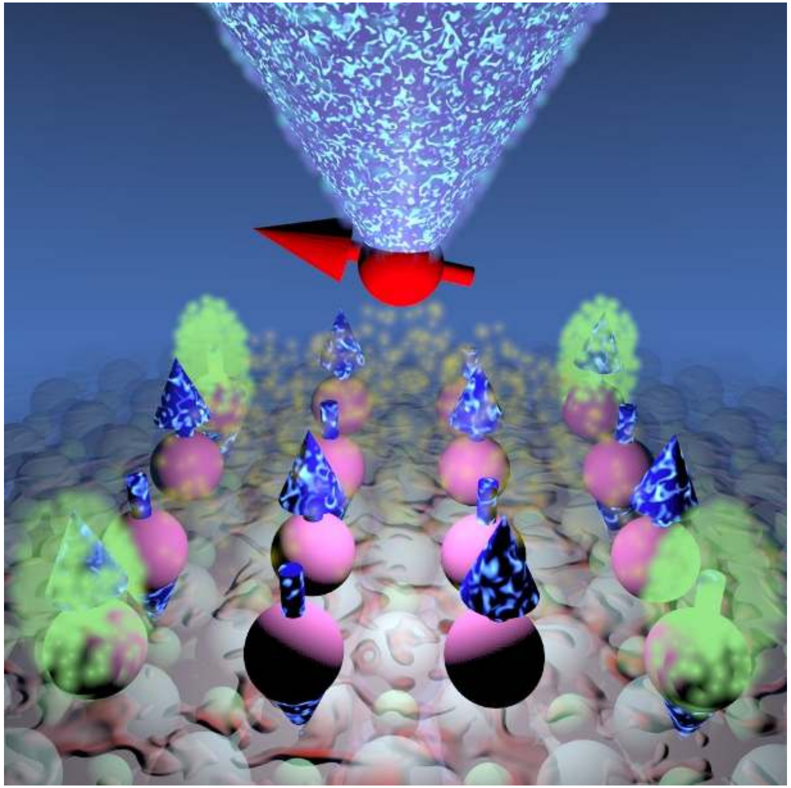


Machine learning triplons experimentally





Engineering a higher order topological quantum magnet



Hao Wang

Jing Chen

Lili Jiang

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Kai Yang

Nature Nanotechnology 19, 1782–1788 (2024)

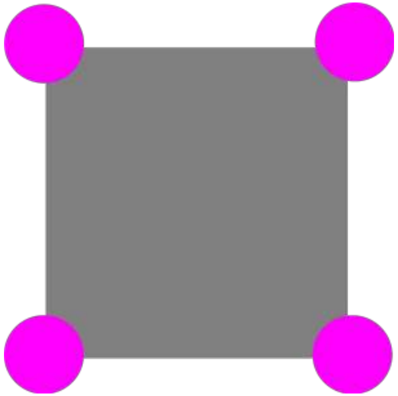
Topological quantum magnets

First order topological quantum magnet

1D gaped bulk
0D fractional edge modes

Nature 598, 287–292 (2021)

Second order topological quantum magnet

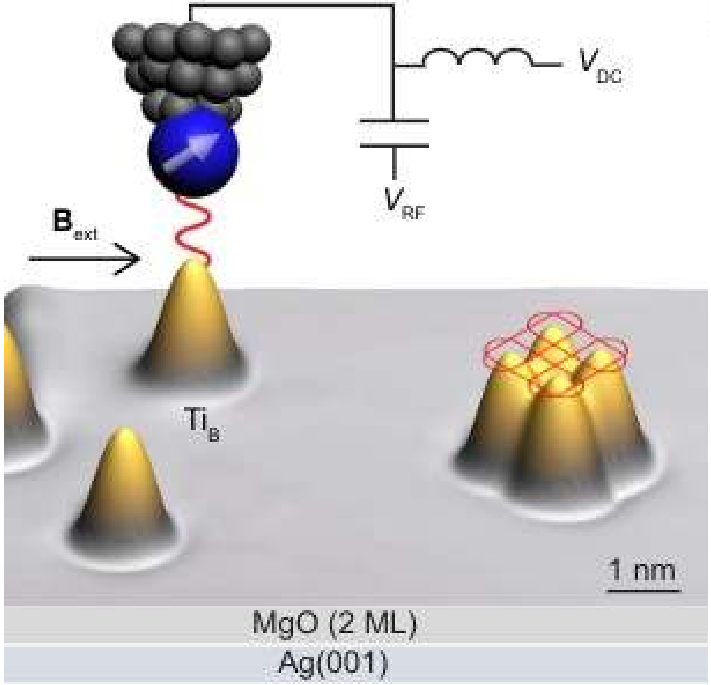


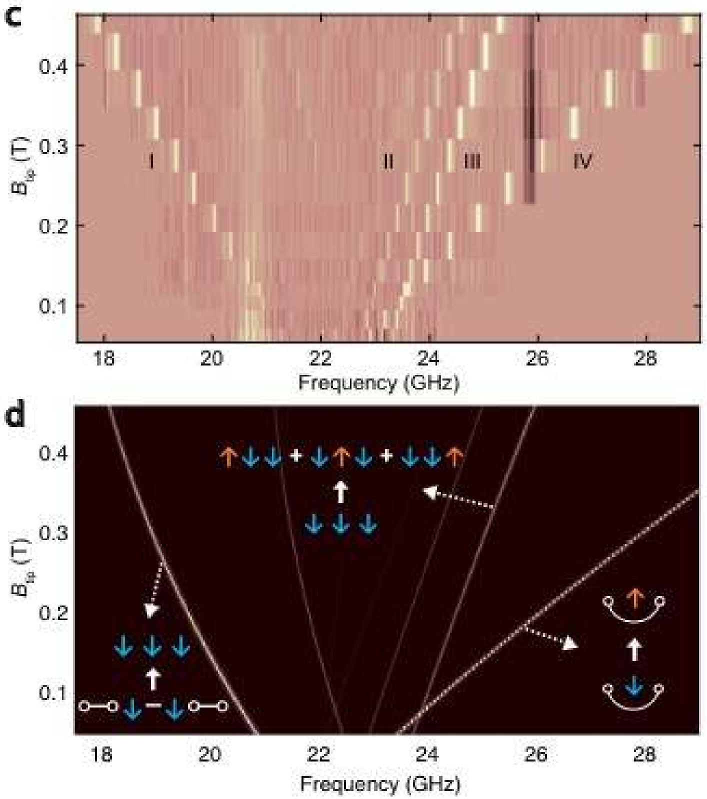
2D gaped bulk
0D fractional edge modes

Nature Nanotechnology (2024)
10.1038/s41565-024-01775-2

Artificial spin models with Ti@MgO

Ti@MgO (2x2 lattice)

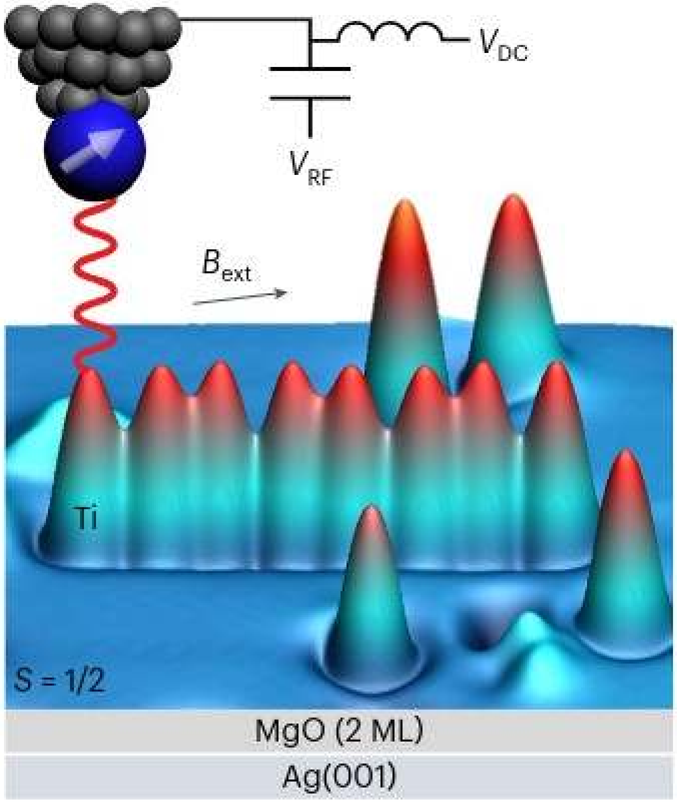




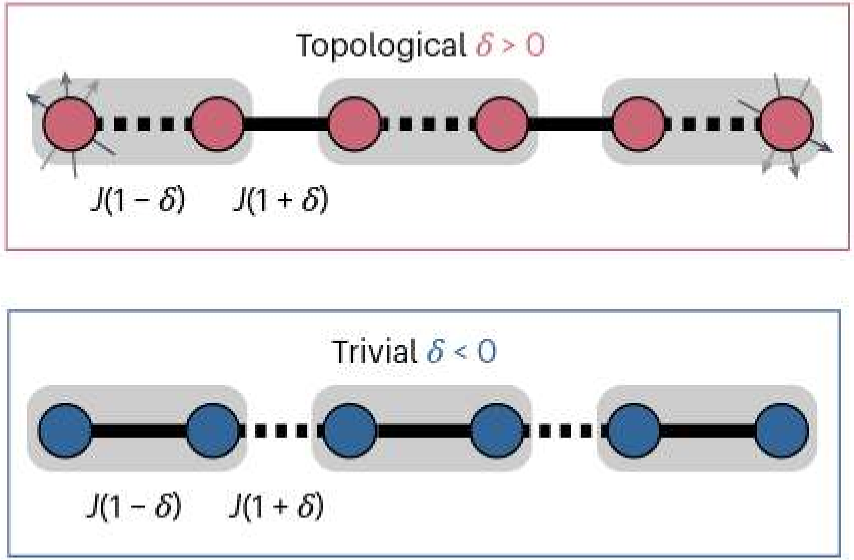
Nature Communications 12, 993 (2021)

Can we create new states of matter?

First order topological quantum magnets



Two possible configurations

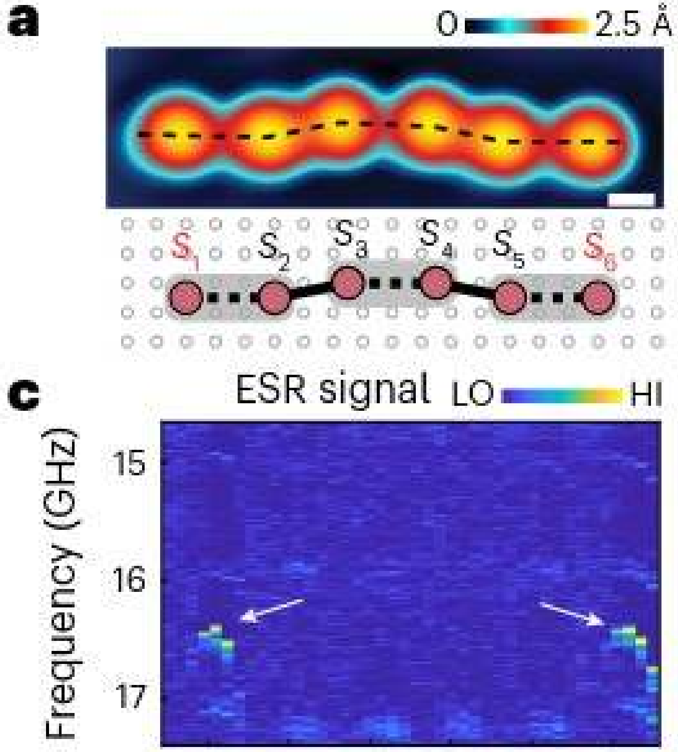


Nature Nanotechnology (2024)

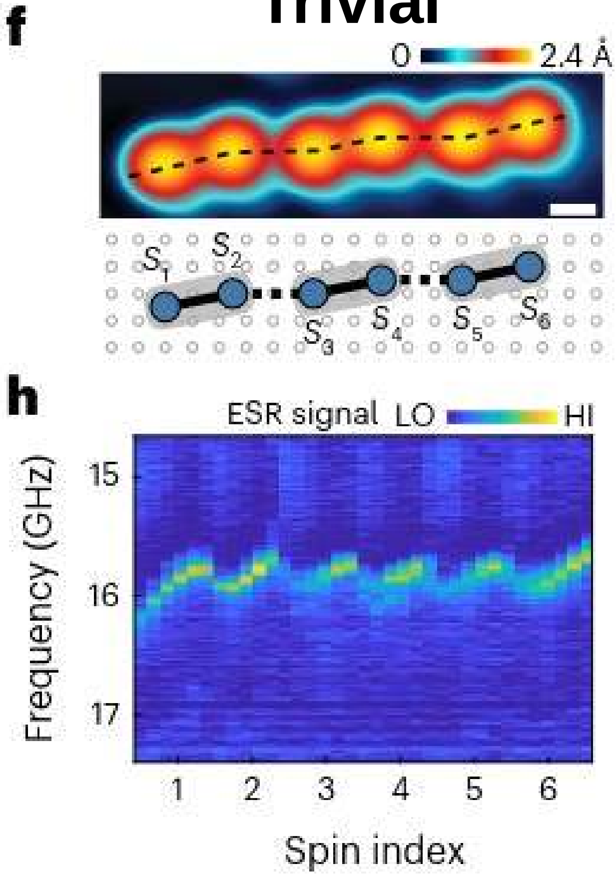
10.1038/s41565-024-01775-2

First order topological quantum magnets

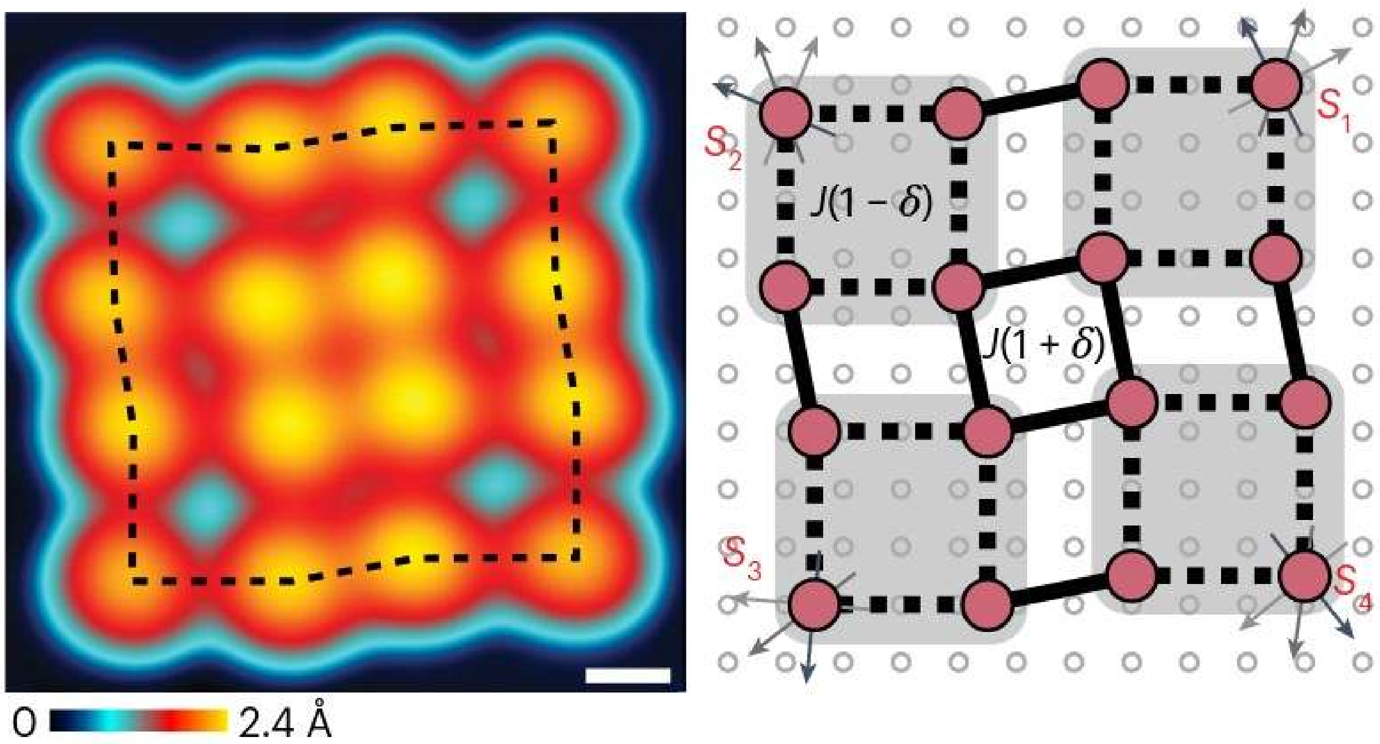
Topological



Trivial

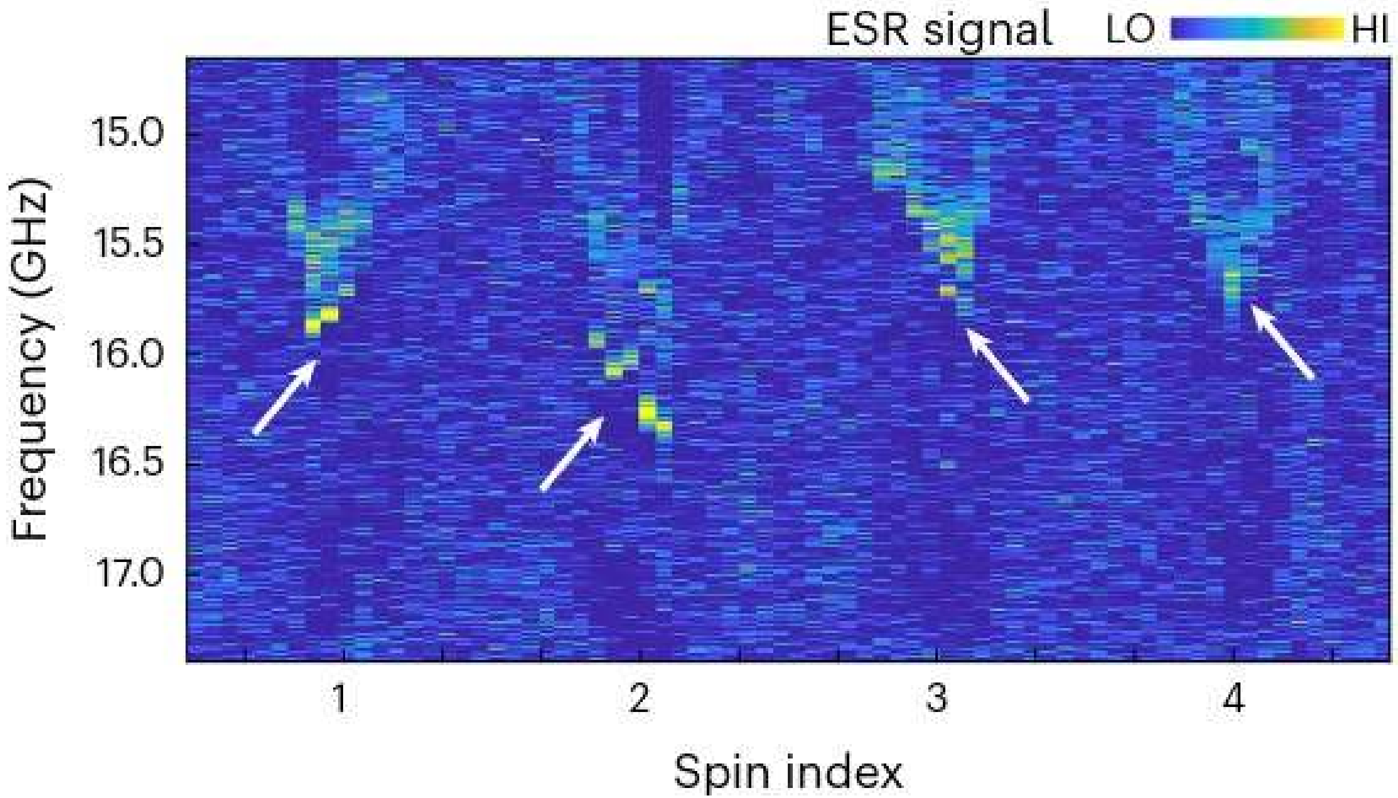


A higher order topological quantum magnet



The higher order topological quantum magnet shows topological corner modes

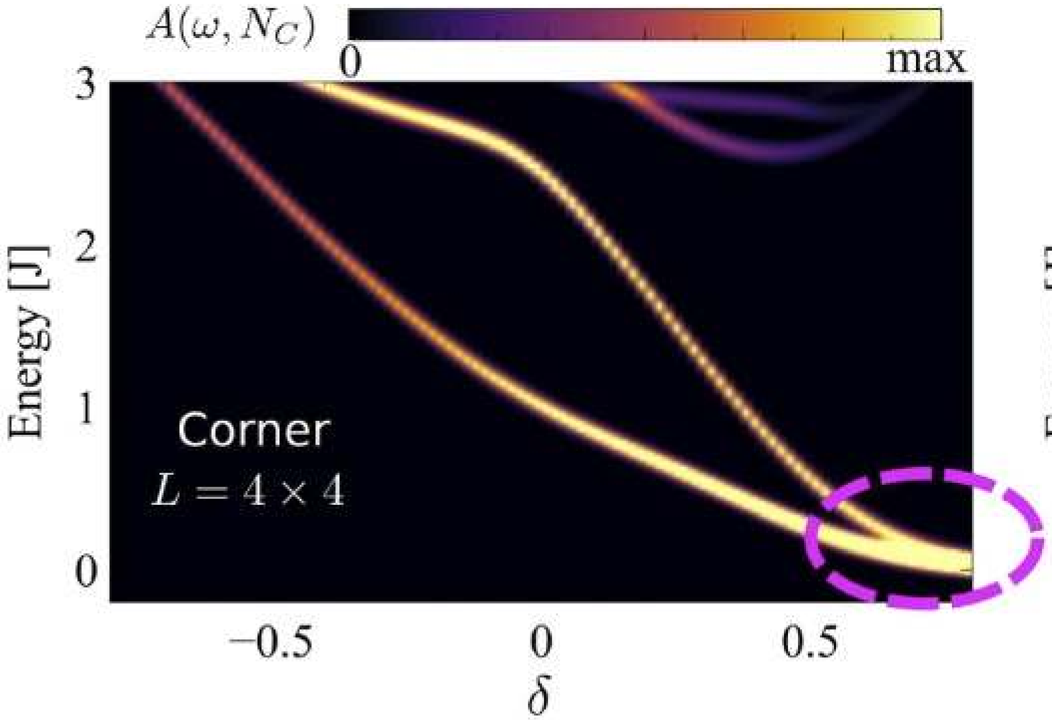
A higher order topological quantum magnet



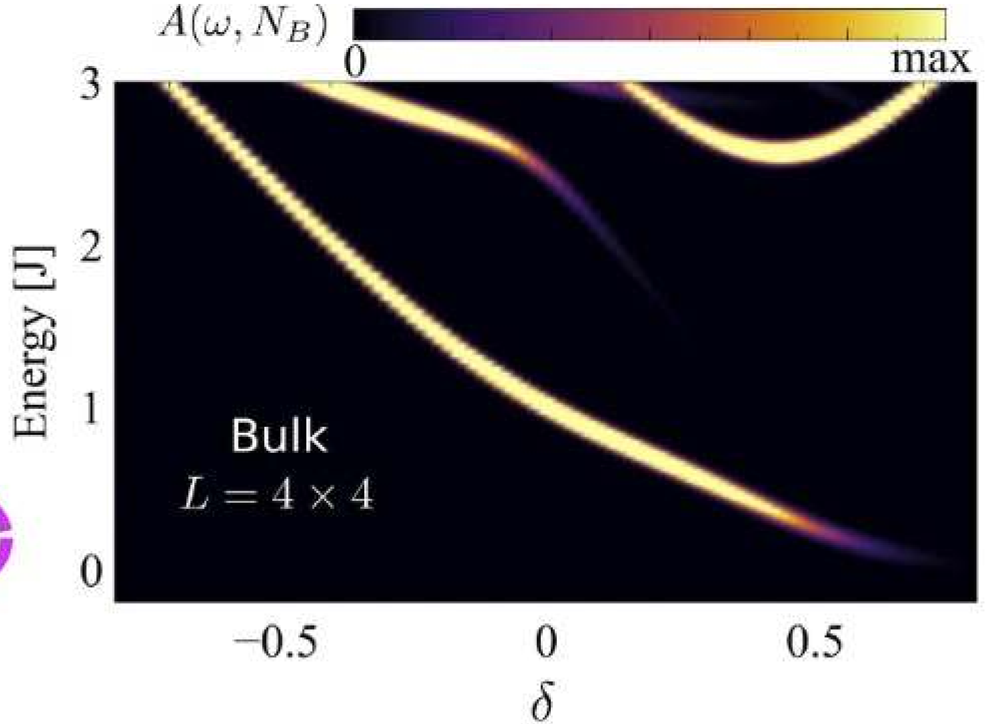
Topological corner modes are measured coexisting with a bulk spectral gap

A higher order topological quantum magnet

Corner spin excitations



Bulk spin excitations



The tetramerization of the lattice creates topological corner spin excitations

Open-source software for artificial quantum materials

Open-source software for many-body quantum magnets

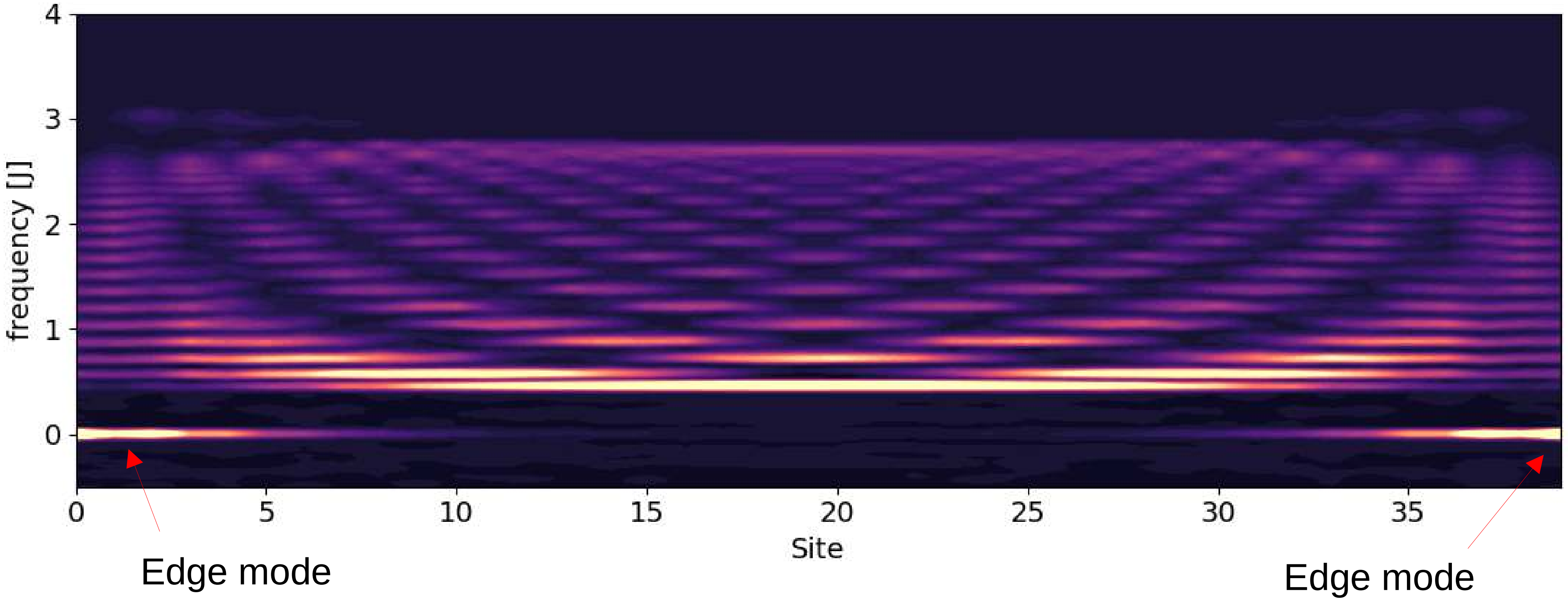
Python library for tensor-network kernel polynomial algorithms for spins, fermions, parafermions, with static solvers for Hermitian and non-Hermitian modes

```
from dmrgpy import spinchain
spins = ["S=1" for i in range(40)] # S=1 chain
sc = spinchain.Spin_Chain(spins) # create spin chain object
h = 0 # initialize Hamiltonian
for i in range(len(spins)-1):
  h = h + sc.Sx[i]*sc.Sx[i+1]
  h = h + sc.Sy[i]*sc.Sy[i+1]
  h = h + sc.Sz[i]*sc.Sz[i+1]
sc.set_hamiltonian(h)
sc.get_dynamical_correlator(name=(sc.Sz[0],sc.Sz[0]))
```

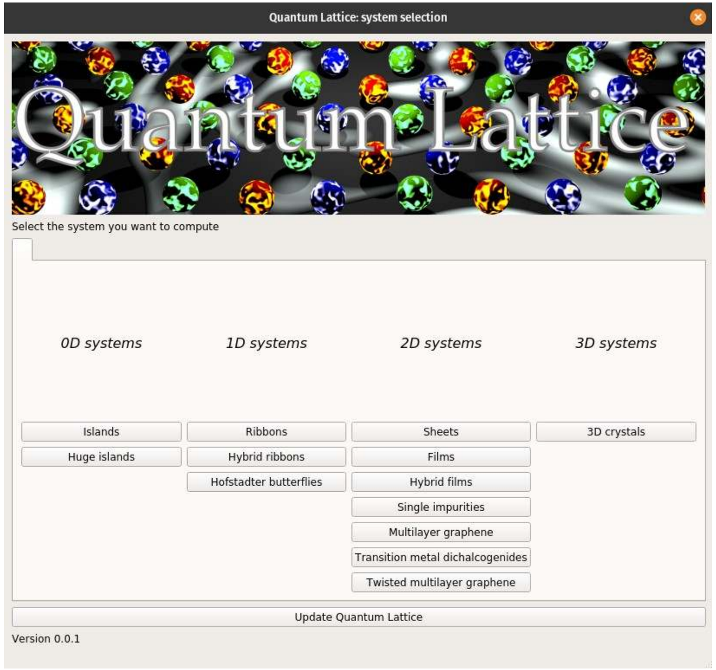
dmrgpy https://github.com/joselado/dmrgpy

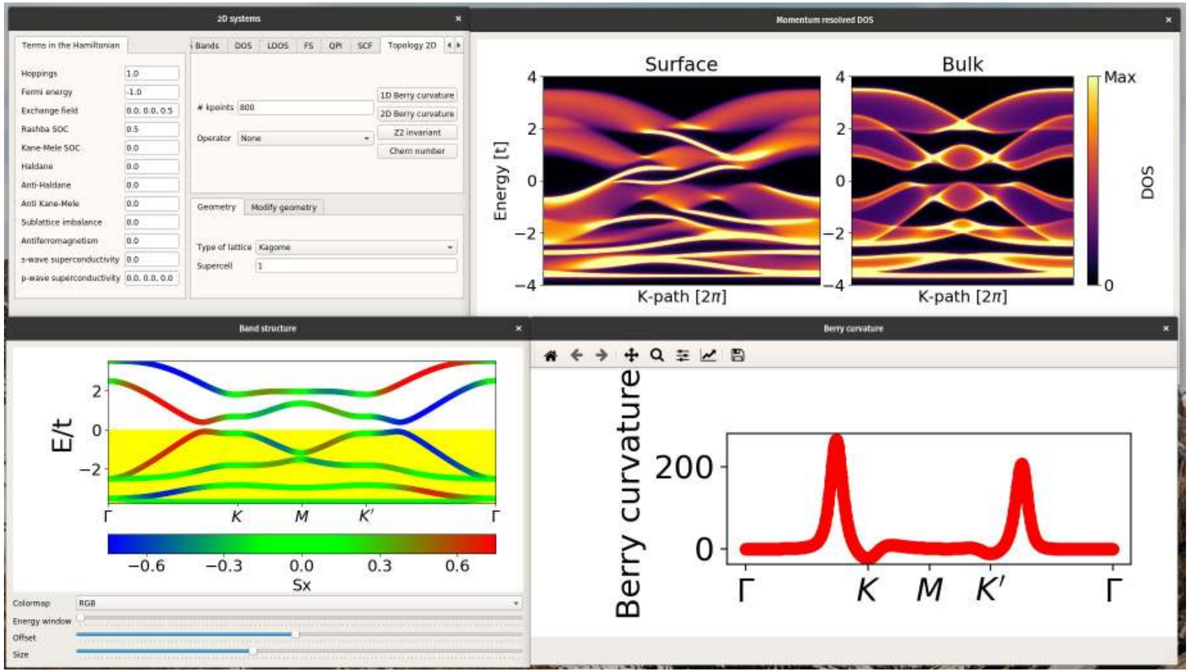
Open-source software for many-body quantum magnets

The spin spectral function of the S=1 Heisenberg model (L=40 sites)



Quantum Lattice: A user interface to compute electronic properties



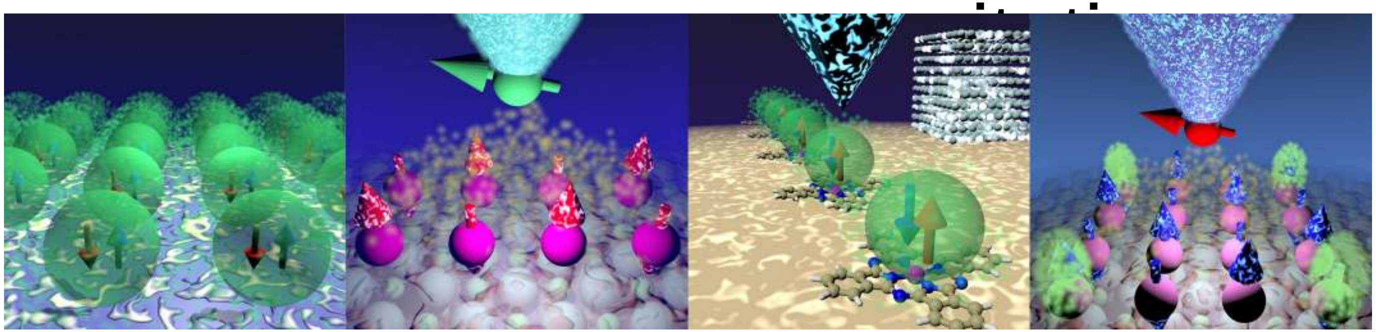


Quantum Lattice: open source interactive interface for tight binding modeling

https://github.com/joselado/quantum-lattice

Take home

Atomically engineered spin lattices allows us to create and learn quantum matter with emergent quantum



Phys. Rev. Lett. 131, 086701 (2023)
Phys. Rev. Applied 20, 024054 (2023)
arXiv:2504.20711 (2025)
Nature Nano 19, 1782–1788 (2024)

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Finnish Quantum Flagship

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