# Hamiltonian learning and triplon quantum matter in atomically assembled spin lattices

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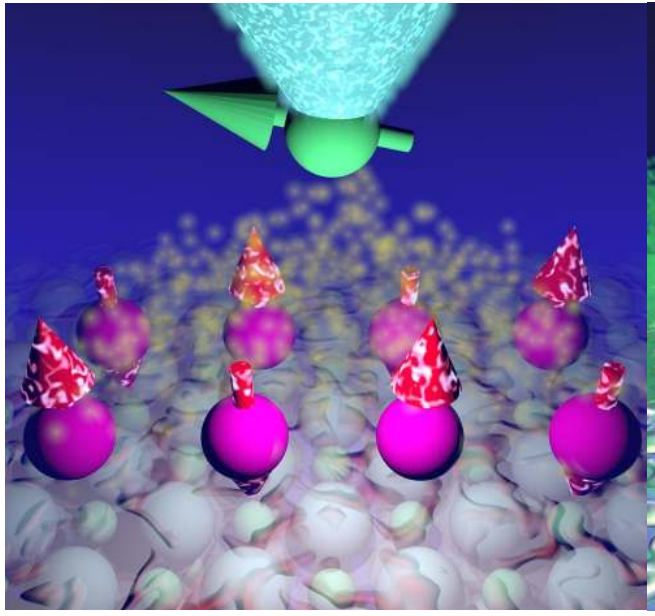
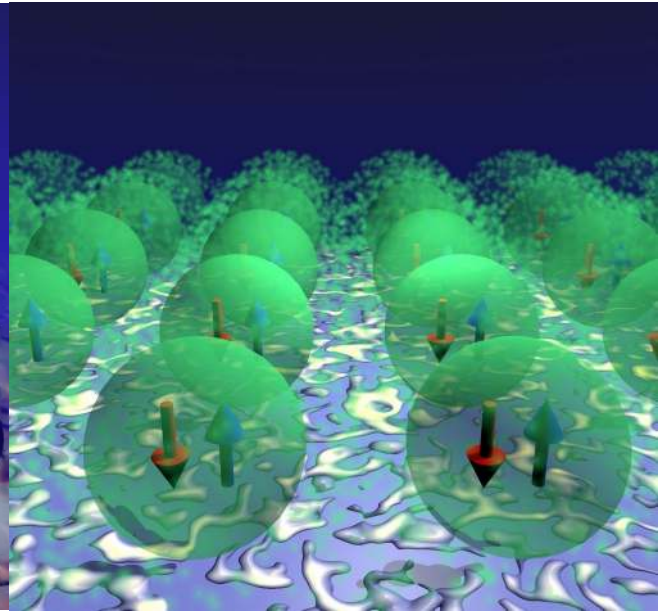


*Phys. Rev. Applied 20, 024054 (2023)*

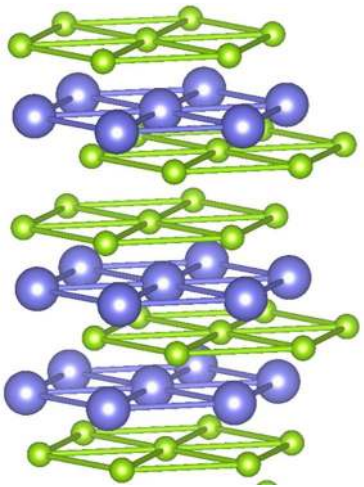


*Phys. Rev. Lett. 131, 086701 (2023)*

Plenty of Room at the Bottom – New Developments in Scanning Probe Tools
November 14th 2023, Germany

# Emergence in quantum materials

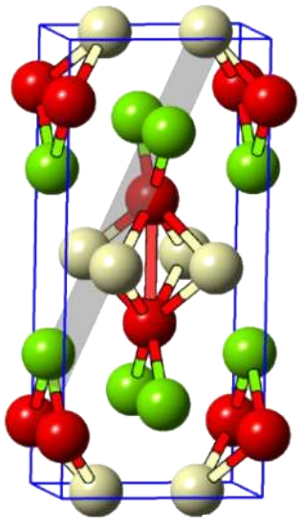
**Topological materials**



$Bi_2Se_3$

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

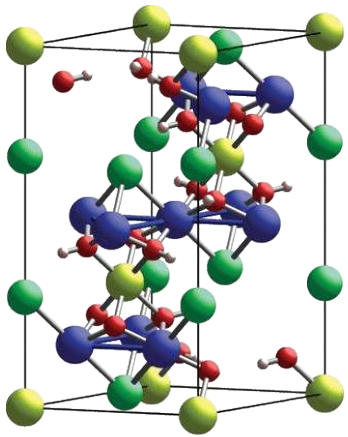
**Unconventional superconductors**



$UTe_2$

*Nature 579, 523–527 (2020)*

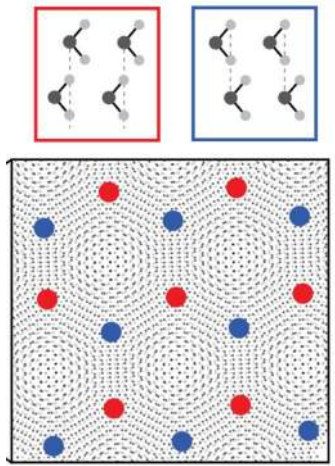
**Quantum spin liquids**



$ZnCu_3(OH)_6Cl_2$

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

**Fractional topological matter**

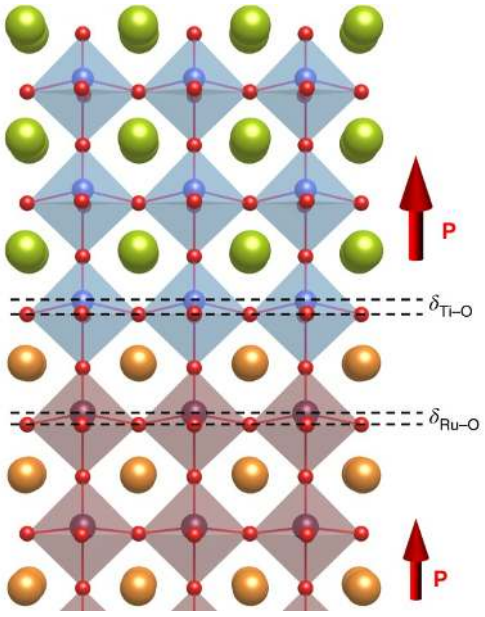


Twisted $MoTe_2$

*Nature (2023)*

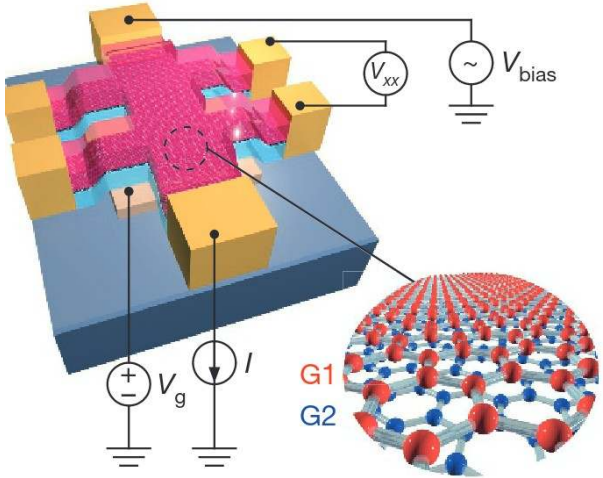
# Artificial quantum materials

**Oxide interfaces**



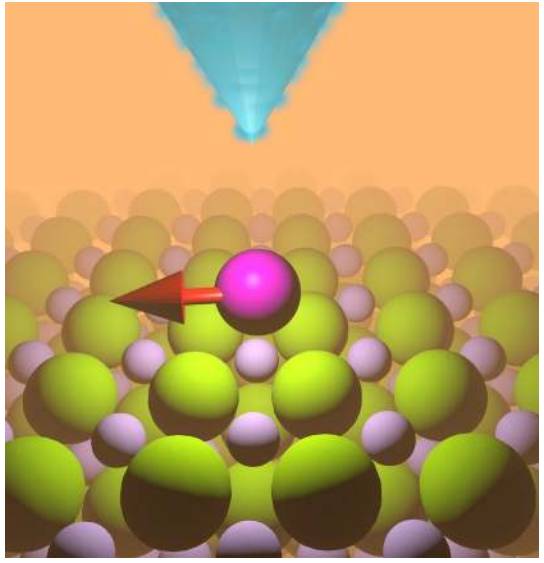

*Nature Materials 17, 1087–1094 (2018)*

**Twisted van der Waals materials**




*Nature 556, 43–50 (2018)*

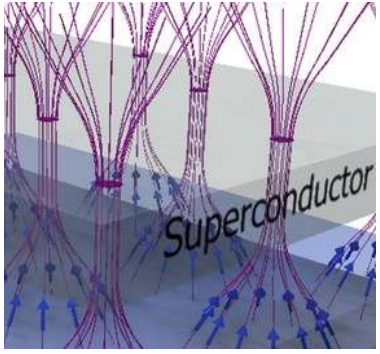
**Atomic lattices**



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

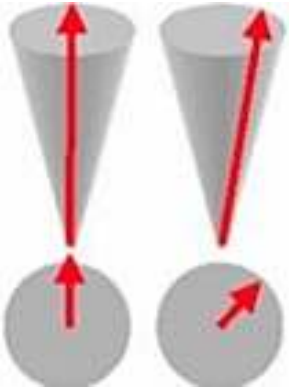
# Emergent excitations in quantum materials

Vortexes




***Superconductors***

Magnons

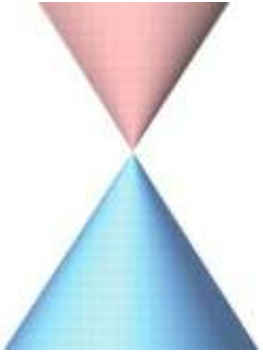


***Ordered magnets***

Skyrmions

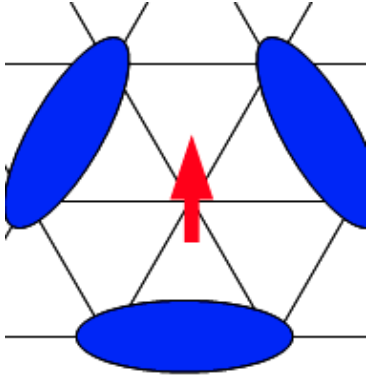
**Non-collinear magnets**

Dirac fermions



***Topological materials***

Spinons

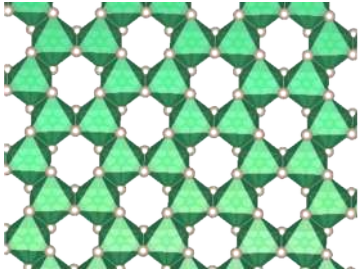


***Quantum spin liquids***

***The discovery of new quasiparticles opens new possibilities in quantum technologies***

# Magnetic quantum materials

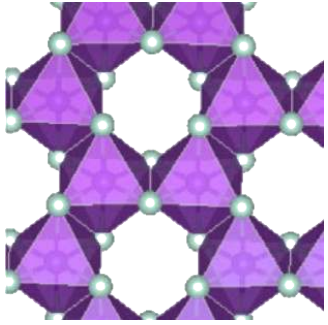
**Ferromagnet, antiferromagnets**



$CrI_3$, $CrCl_3$, $CrBr_3$

*Nature 546, 270–273 (2017)*

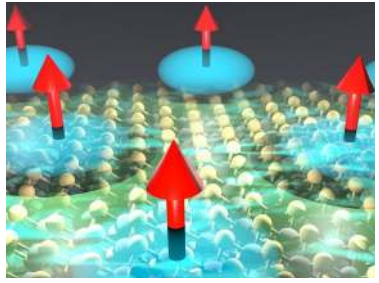
**(proximal) Quantum spin-liquids**



$RuCl_3$, 1T-$TaS_2$

*Nature Physics 17, 1154–1161 (2021)*

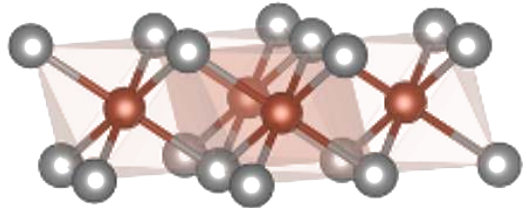
**Heavy-fermion Kondo insulators**



1T-$TaS_2$/1H-$TaS_2$

*Nature 599, 582–586 (2021)*

**Multiferroics**



$NiI_2$

*Nature 602, 601–605 (2022)*

Break time-reversal

Do not break time-reversal

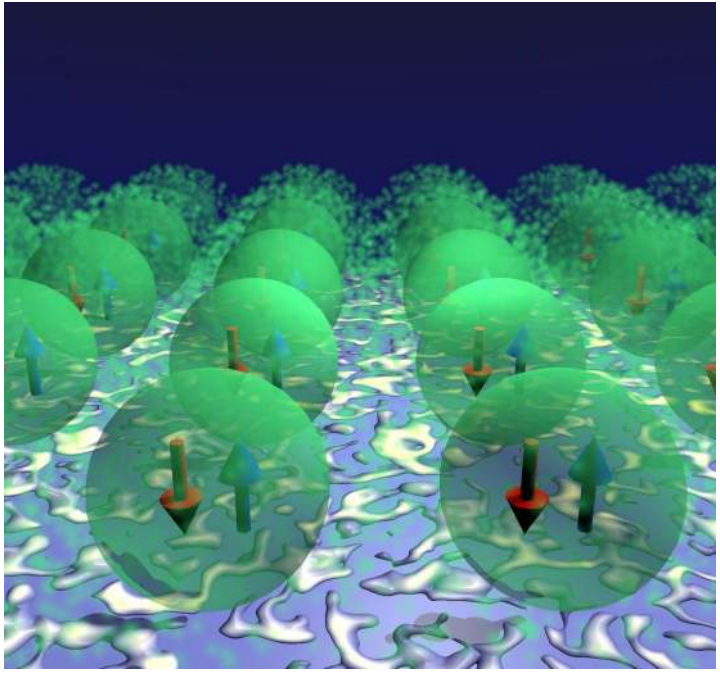
Break time-reversal and inversion symmetry

# Plan for today

- Excitations in atomic-scale quantum magnets
- The theoretical challenge of quantum magnetism
- Detecting triplons in an atomic-scale magnet
- Hamiltonian learning atomic-scale magnets

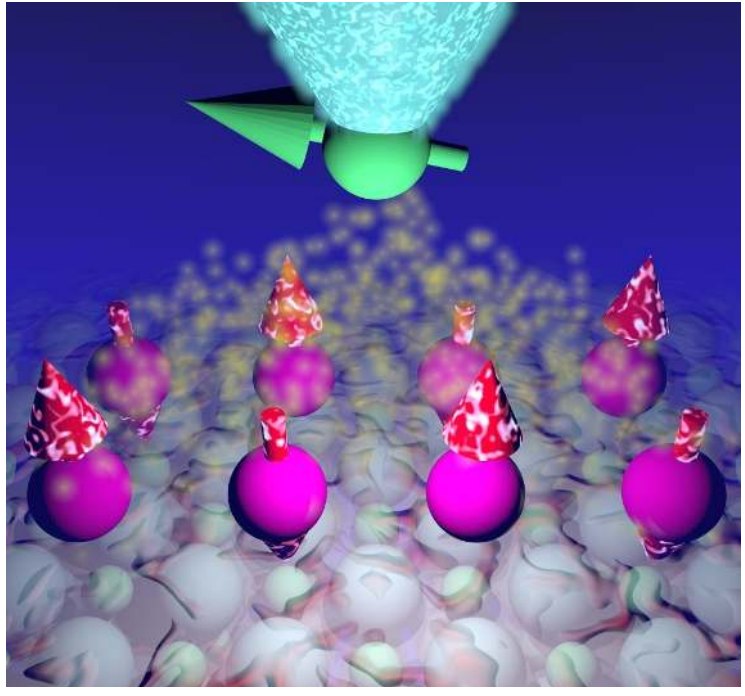
# Plan for today

**Triplons in an atomic-scale quantum magnet**



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

**Hamiltonian learning atomic-scale magnets**



Phys. Rev. Applied 20, 024054 (2023)

# Behind the scenes

**Triplons in an atomic-scale quantum magnet**

Robert Drost

Shawulienu Kezilebieke

Peter Liljeroth

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

**Hamiltonian learning atomic-scale magnets**

Netta Karjalainen

Zina Lippo

Rouven Koch

Guangze Chen

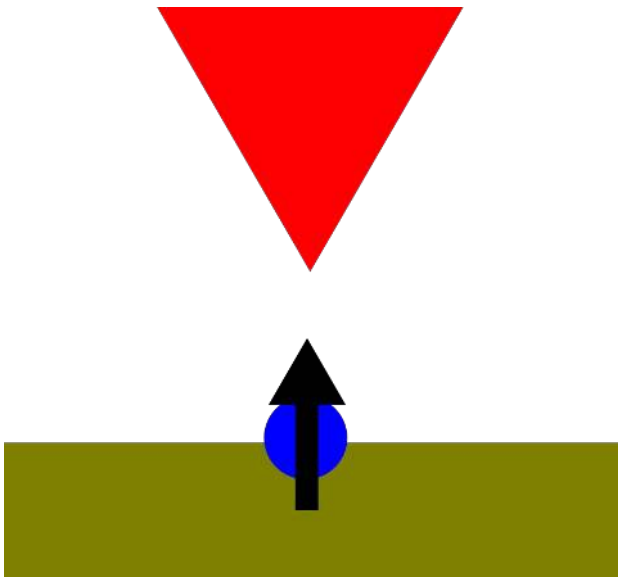
Adolfo Fumega

Phys. Rev. Applied 20, 024054 (2023)

# Quantum excitations in atomic-scale magnets

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

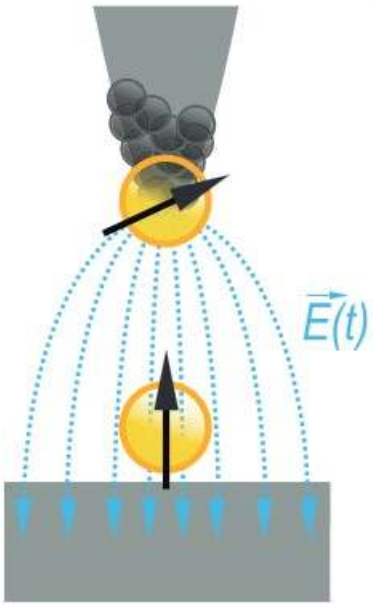
**Inelastic spectroscopy**



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

*Without spin-polarized tip*

**Electrically driven paramagnetic resonance**

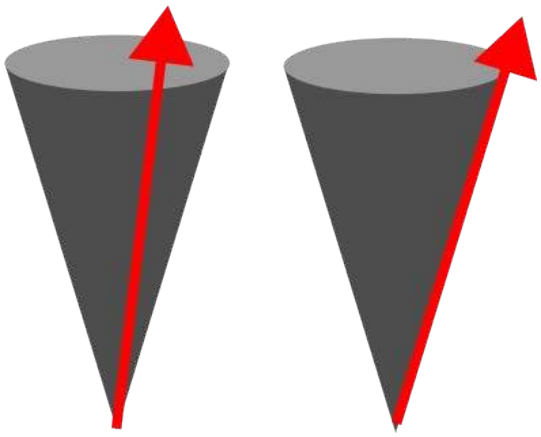



*Science 350 (6259), 417-420 (2015)*

*With spin-polarized tip*

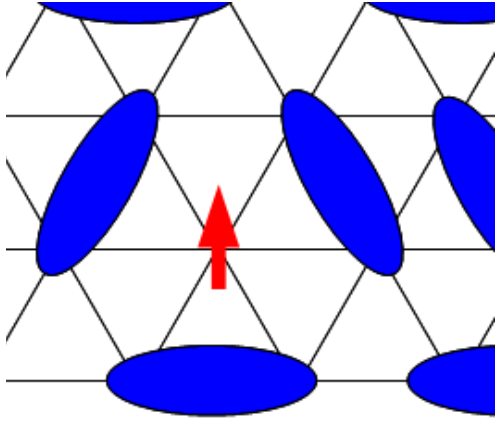
# Excitations in atomic-scale magnets

**Magnons**



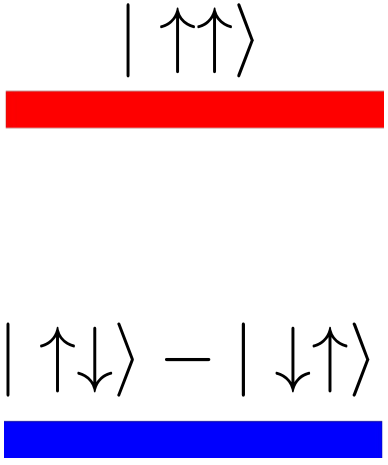
S=1

**Spinons**



S=1/2

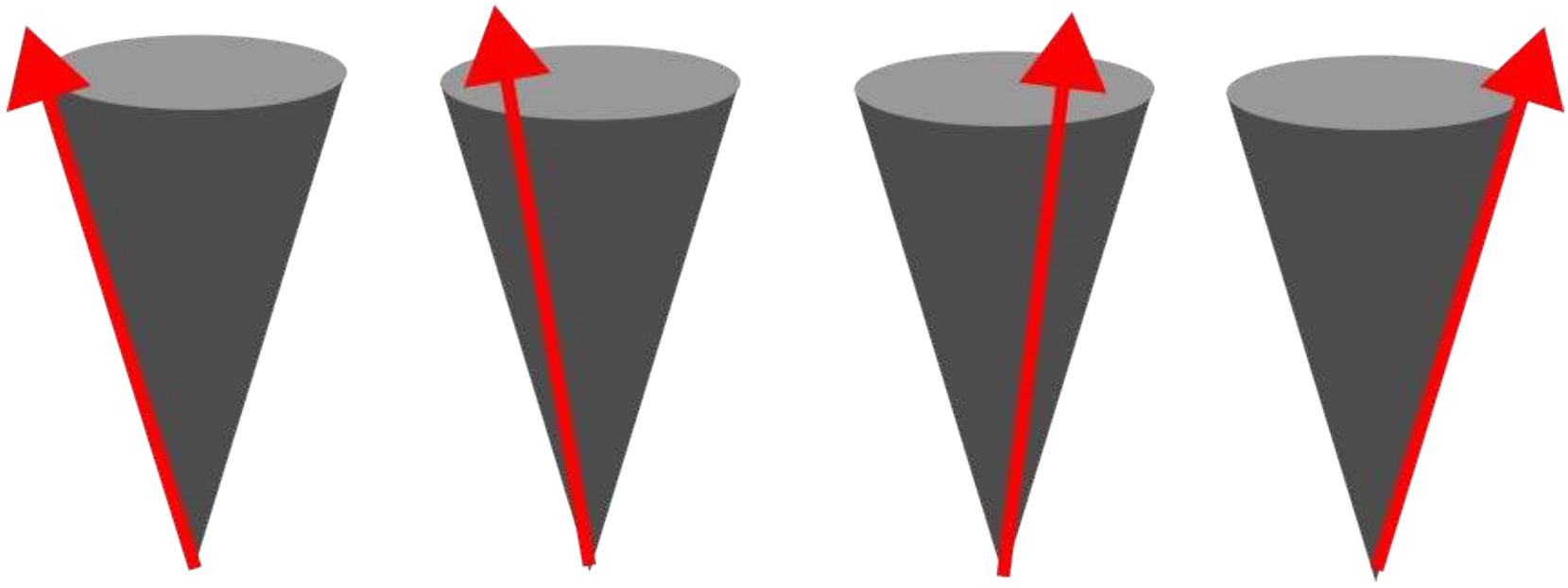
**Triplons**




S=1

# Magnons: quasiparticles in magnets

Qualitatively, magnons are the fluctuations of the order parameter



# Excitations in the Heisenberg model

The Heisenberg model is a full-fledged many-body problem

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

Algebraic commutation relations $[S_j^\alpha, S_j^\beta] = i\epsilon_{\alpha\beta\gamma} S_j^\gamma$

$$S = 1/2, 1, 3/2, 2, ...$$

**How do we compute its many-body excitations?**

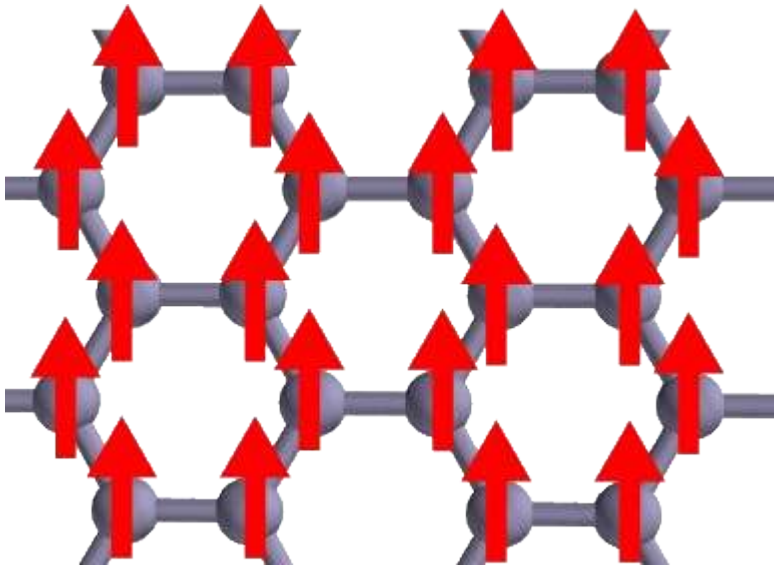
# The ferromagnetic Heisenberg model

In the case of a ferromagnetic Heisenberg model, we know the ground state

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$J_{ij} < 0$$

$$|GS\rangle = |\uparrow\uparrow\uparrow\uparrow\uparrow \ldots.\rangle$$



**We take perturbations around this classical ground state**

# Magnons as quasiparticles

Replace the spin Hamiltonian by a bosonic Hamiltonian

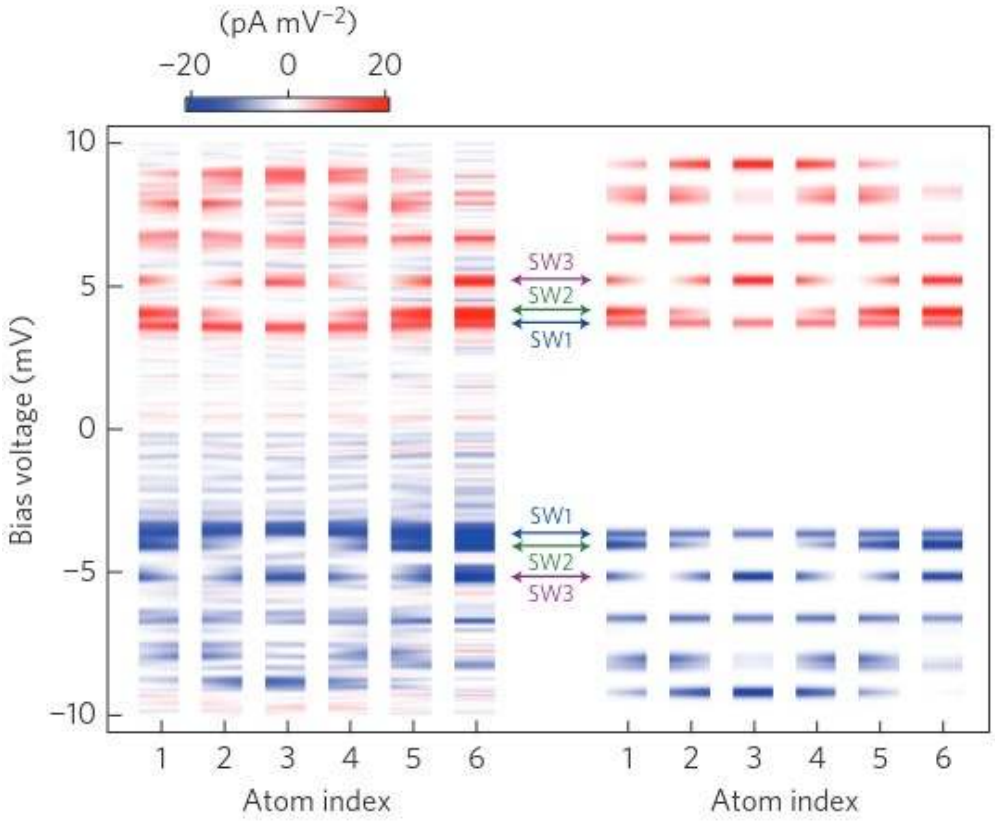
$$S_+ = \sqrt{2s}\sqrt{1 - \frac{a^\dagger a}{2s}}a, \quad S_- = \sqrt{2s}a^\dagger\sqrt{1 - \frac{a^\dagger a}{2s}}, \quad S_z = \left(s - a^\dagger a\right)$$

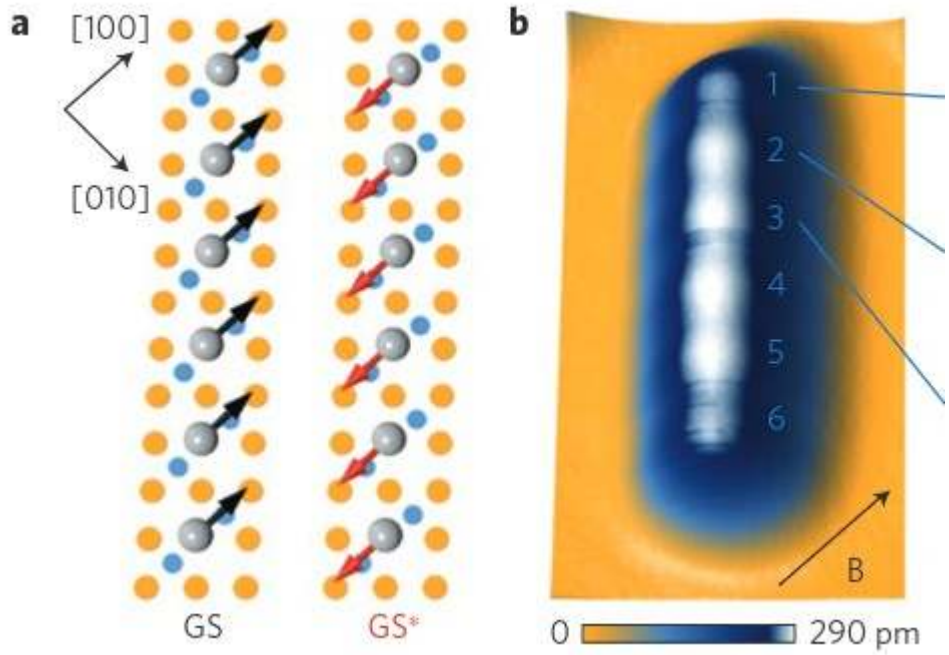
Make the replacement and decouple with mean-field assuming $\langle a_i^\dagger a_i \rangle \ll s$

$$\mathcal{H} = \sum_{ij} J_{ij}\vec{S}_i \cdot \vec{S}_j \longrightarrow \mathcal{H} = \sum_{ij} \gamma_{ij} a_i^\dagger a_j$$

**Spins** **Magnon**

# Spin waves in atomic-scale magnets




Nature Materials 13, 782–785 (2014)

# Quasiparticles in a quantum magnet

Take a Hamiltonian for a quantum magnet $\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$

Quantum magnets require $\langle \vec{S}_i \rangle = 0$

The approximation used for magnons breaks down

$$\langle S_i^z \rangle = S - \langle a_i^\dagger a_i \rangle$$

$$\langle a_i^\dagger a_i \rangle \ll S$$

**We need a new approximation for the quantum excitations**

# The parton transformation for spinons

Transform spin operators to auxiliary fermions

$$S_i^\alpha = \frac{1}{2}\sigma_{s,s'}^\alpha f_{i,s}^\dagger f_{i,s'}$$

The fermions f (spinons) have S=1/2 but no charge

This transformation artificially enlarges the Hilbert space, thus we have to put the constraint

$$\sum_s f_{i,s}^\dagger f_{i,s} = 1$$

**This transformation allow to turn a spin Hamiltonian into a fermionic Hamiltonian**

# Spinons as quasiparticles

We can insert the auxiliary fermions $S_i^\alpha \sim \sigma^\alpha_{s,s'} f^\dagger_{i,s} f_{i,s'}$

And perform a mean-field in the auxiliary fermions (spinons)

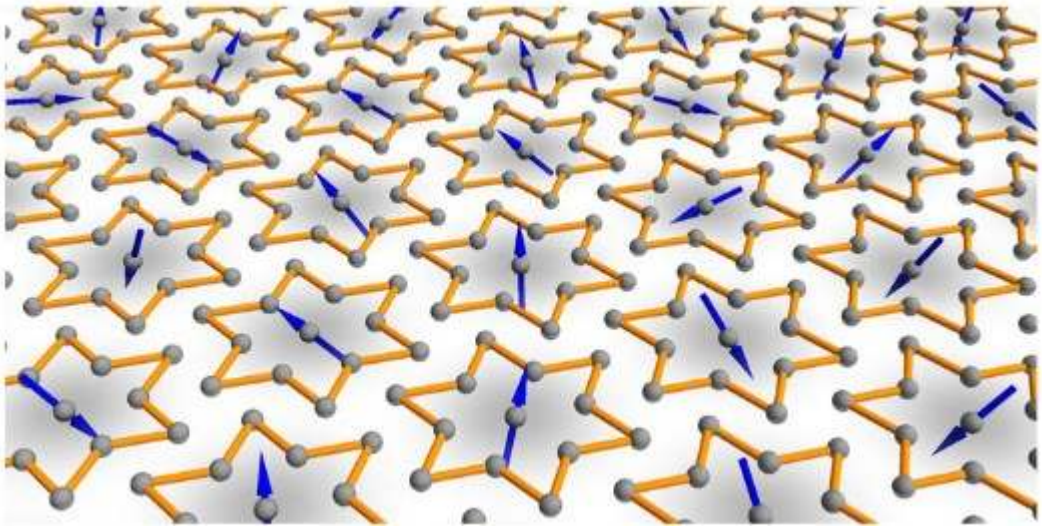
$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \longrightarrow \mathcal{H} = \sum_{ij,s} \chi_{ij} f^\dagger_{i,s} f_{j,s}$$

Enforcing time-reversal symmetry $\langle \vec{S}_i \rangle = 0$

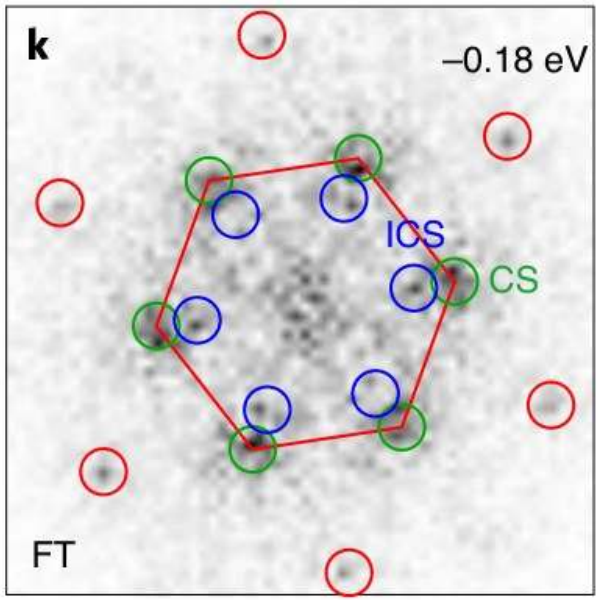
**The exitations of the QSL are described by a single particle spinon Hamiltonian**

# Signatures of spinons with STM

Frustrated magnetism in $TaSe_2$



Quasiparticle interference




Nature Physics 17, 1154–1161 (2021)

# The theoretical challenge of quantum magnetism

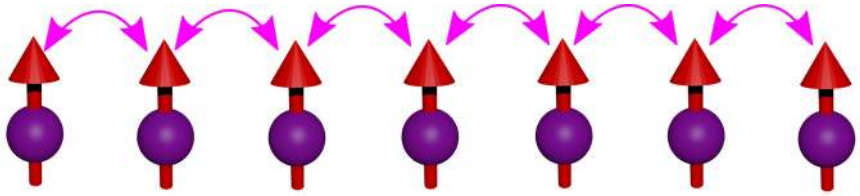
# The problem of dimensionality

In a many-body problem, the size of our vectors grows as

$$2^L$$

where L is the number of sites

$$\mathcal{H} = \sum_{\langle ij \rangle} J \vec{S}_i \cdot \vec{S}_j$$



For a single-particle tight binding problem, we can reach up to $10^8$ sites in a laptop

For a many-problem, we cannot even store states for systems bigger than $L = 30$ sites

$$2^{30} \sim 10^9$$

# The quantum many-body problem

Let us go back to a simple many-body problem $\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$

A typical wavefunction is written as

$$|\Psi\rangle = \sum c_{s_1, s_2, \ldots, s_L} |s_1, s_2, \ldots s_L\rangle$$

We need to determine in total $2^L$ coefficients

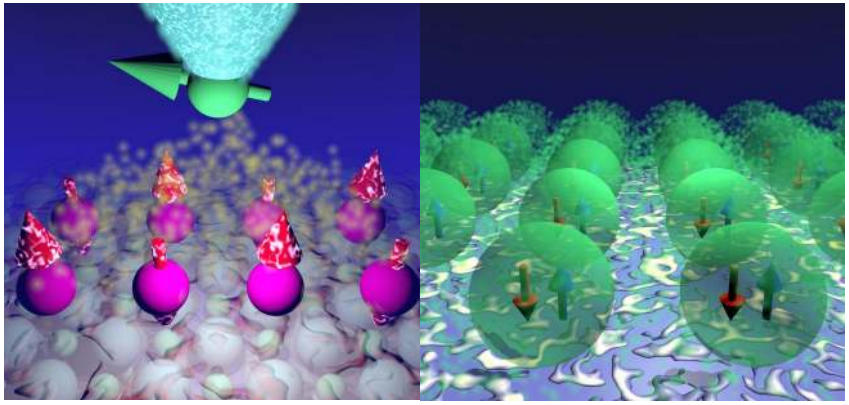
**Is there an efficient way of storing so many coefficients?**

# The fundamental idea of tensor-networks

A many-body wavefunction a is a very high dimensional object

$$|\Psi\rangle = \sum c_{s_1,s_2,\ldots,s_L} |s_1, s_2, \ldots s_L\rangle$$

Tensor-networks allow “compressing” all that information in a very efficient way



“True wavefunction”

“Tensor-network wavefunction”

# The matrix-product state ansatz

For this wavefunction $\left|\Psi\right\rangle = \sum c_{s_1,s_2,\ldots,s_L} \left|s_1, s_2, \ldots s_L\right\rangle$

Let us imagine to propose a parametrization in this form

$$c_{s_1,s_2,\ldots,s_L} = M_1^{s_1} M_2^{s_2} \ldots . M_L^{s_3}$$

dimension $2^L$

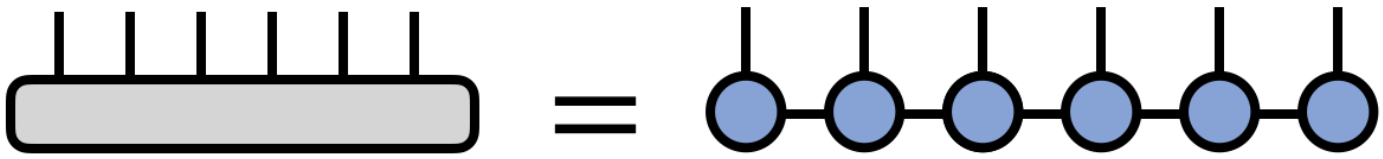
dimension $\sim Lm^2$

(m dimension of the matrix)

# State compression with tensor-networks

Given a many-body wavefunction, we can parametrice the components as

$$|\Psi\rangle = \sum_{\{s\}} \mathrm{Tr}\left[M_1^{(s_1)} M_2^{(s_2)} \cdots M_N^{(s_N)}\right] |s_1 s_2 \ldots s_N\rangle$$



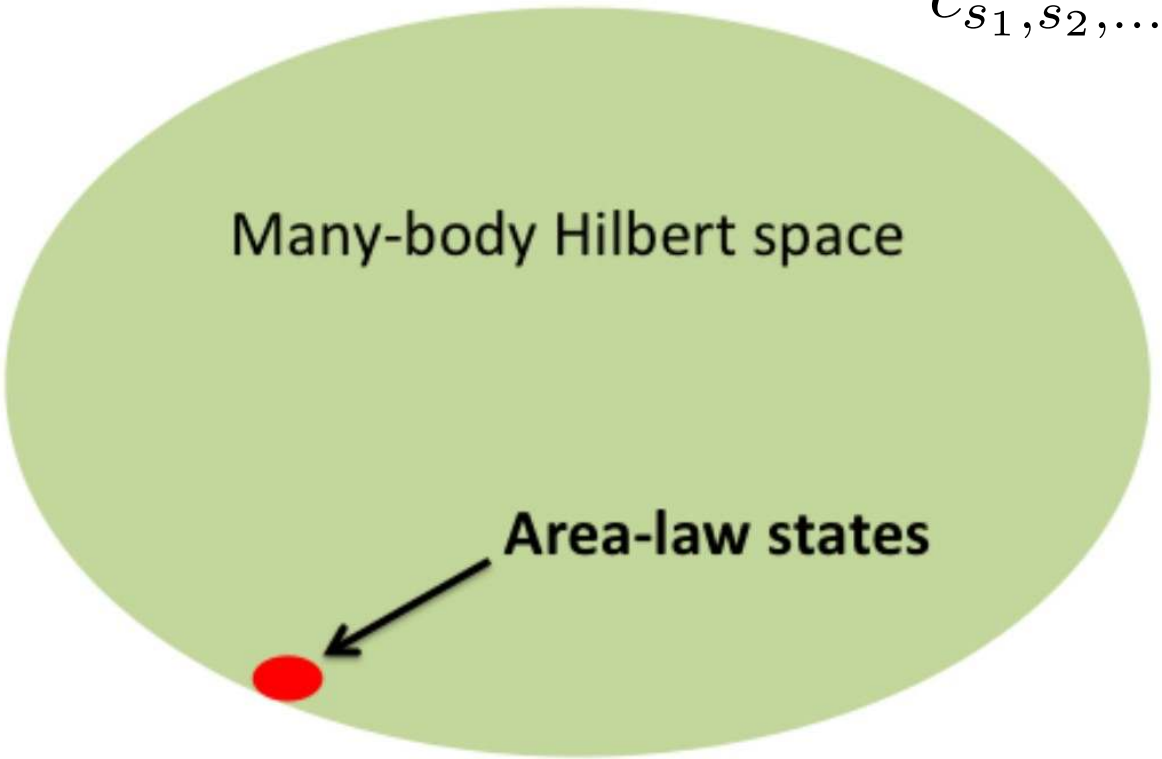
*Matrix product state*

The previous representation allows drastically reducing the memory required to store a state

*Annals of Physics 326, 96 (2011)*

# Matrix product states as a parametrization of area law states

$$c_{s_1,s_2,\ldots,s_L} = M_1^{s_1} M_2^{s_2} \ldots M_L^{s_3}$$



# The matrix-product state ansatz

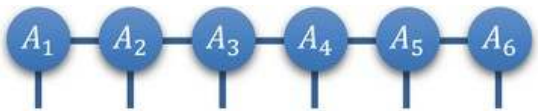
- This ansatz enforces a maximum amount of entanglement entropy in the state $S \sim \log m$
- One-dimensional problems have ground states that can be captured with this ansatz

$$c_{s_1,s_2,\ldots,s_L} = M_1^{s_1} M_2^{s_2} \ldots M_L^{s_3}$$

This ansatz can be generalized for time-evolution, excited states, or typical thermal states

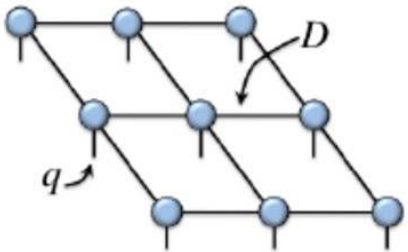
# Many-body state compression

**Matrix-product states**



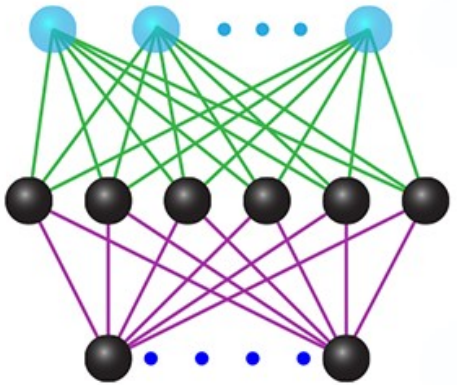

*Phys. Rev. Lett. 69, 2863 (1992)*

**Projected entangled pair-states**




*Annals of Physics 326, 96 (2011)*

**Neural-network quantum states**



*Science 355.6325 (2017): 602-606.*

***Other compressed many-body states could be potentially used for quantum spin models in atomic-scale magnets***

# Dynamical excitations in quantum many-body systems

# Dynamical correlators

**For a single particle model**

$L$ eigenstates/eigenenergies

*Hilbert space*

*One particle excitations*

**For a many-body model**

$2^L$ eigenstates/eigenenergies

*Fock space*

*One, two, three... particle excitations*

**How do we identify single particle excitations in a many-body Hamiltonian?**

A single particle Hamiltonian can be solved in any of the two spaces $H = \sum_{ij} t_{ij} c_i^\dagger c_j$

# Why dynamical correlators

Take a many-body Hamiltonian $\mathcal{H}$

With ground state $\mathcal{H}|\Omega\rangle = E_0|\Omega_0\rangle$

And excited states $\mathcal{H}|\Psi_n\rangle = E_n|\Psi_n\rangle$

**A way to “filter” single particle excitations is by defining**

$$A(\omega, l) = \sum_n \delta(\omega - (E_n - E_0))|\langle\Psi_n|c_l^\dagger|\Omega\rangle|^2$$

Dirac delta function

Picks energy differences $\omega$

Only non-zero if they differ by one electron

This is a dynamical correlator

# Why dynamical correlators

**Dynamical correlator (for single particle electron excitations)**

$$A(\omega, n) = \langle \Omega | c_n \delta(\omega - (\mathcal{H} - E_0)) c_n^\dagger | \Omega \rangle$$

*for fermionic models*

**Dynamical correlator (for spin excitations)**

$$A(\omega, n) = \langle \Omega | S_n^+ \delta(\omega - (\mathcal{H} - E_0)) S_n^- | \Omega \rangle$$

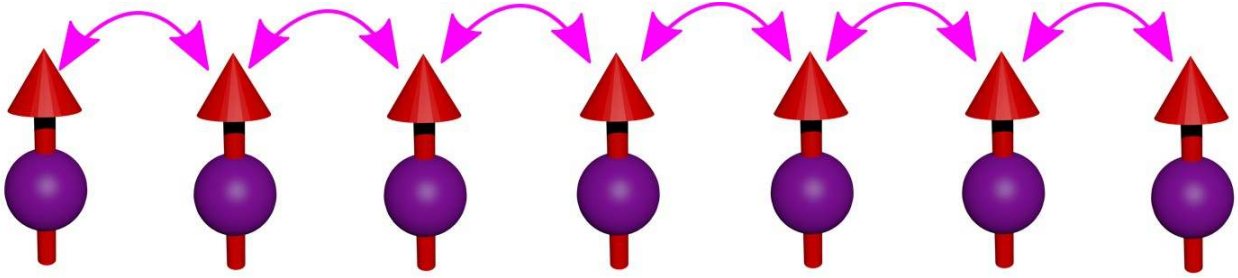
*for spin models*

Both can be related by a fermion-spin mapping (Jordan-Wigner transformation)

# Many-body dynamical correlators

***One dimensional Heisenberg Hamiltonian***

$$\mathcal{H} = \sum_{\langle ij \rangle} J \vec{S}_i \cdot \vec{S}_j$$



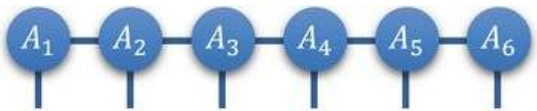
**We want to compute the dynamical correlator**

$$\mathcal{S}(N,\omega) = \langle GS | S^z_N \delta(\omega - \mathcal{H} + E_0) S^z_N | GS \rangle$$

# The kernel-polynomial tensor-network algorithm

Take a many-body Hamiltonian $\mathcal{H} = \sum_{\langle ij \rangle} J \vec{S}_i \cdot \vec{S}_j$

Obtain a tensor-network representation of the many-body ground state

$|GS\rangle =$ 


And apply the kernel polynomial algorithm in the compressed representation

$$\bar{\chi}(\omega) = \langle \mathrm{GS} | S_N^z \delta(\omega - \bar{H}) S_N^z | \mathrm{GS} \rangle$$

$$\bar{\chi}(\omega) = \frac{1}{\pi \sqrt{1-\omega^2}} \left[ \mu_0 + 2 \sum_{l=1}^{N_P} \mu_l T_l(\omega) \right]$$

*Rev. Mod. Phys. 78, 275 (2006)*

$$\mu_l = \langle \mathrm{GS} | S_N^z T_l(\bar{H}) S_N^z | \mathrm{GS} \rangle$$

$$\mu_l = \langle \mathrm{GS} | S_N^z | w_l \rangle$$

$$|w_0\rangle = S_N^z |\mathrm{GS}\rangle,$$

$$|w_1\rangle = \bar{H} |w_0\rangle,$$

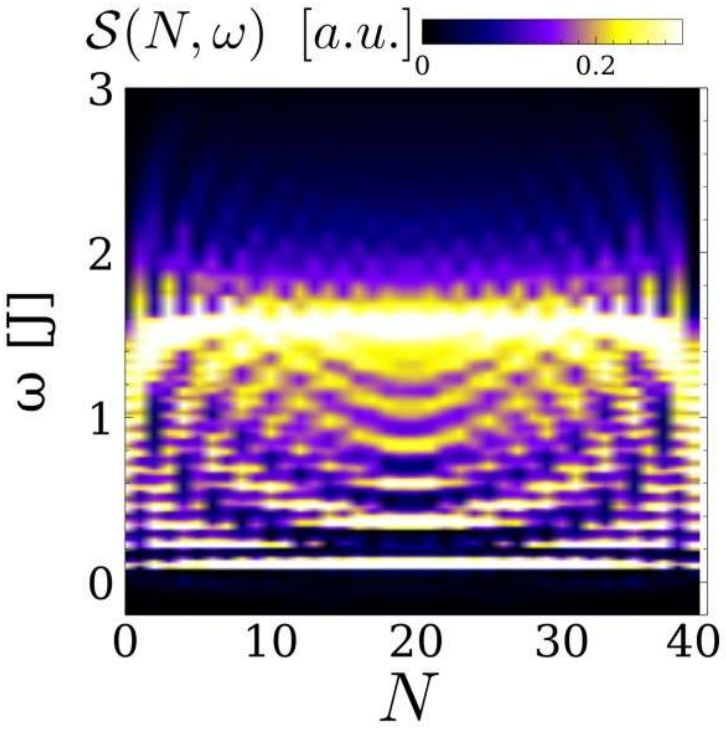
$$|w_{l+1}\rangle = 2\bar{H}|w_l\rangle - |w_{l-1}\rangle,$$

*Phys. Rev. Research 1, 033009 (2019)*

***Open-source implementation in*** *https://github.com/joselado/dmrgpy*

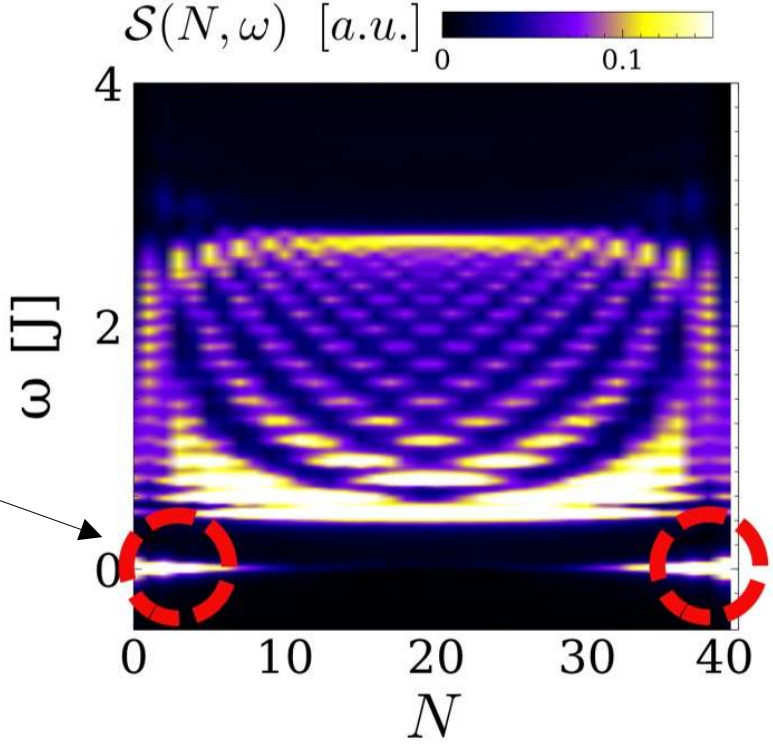
# Dynamical structure factor of a Heisenberg model

**S=1/2 chain**



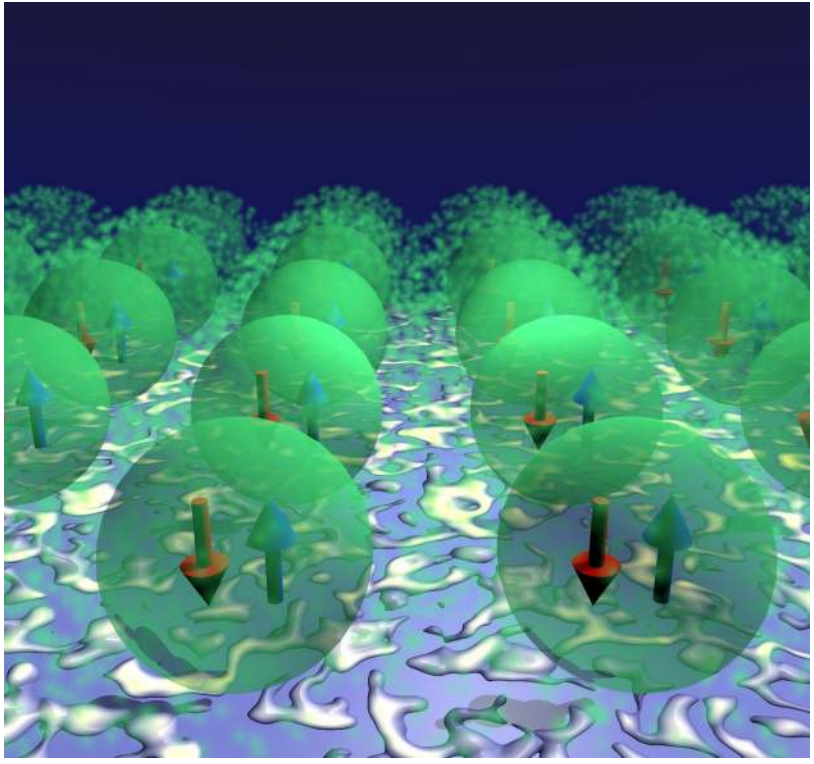

Edge modes

**S=1 chain**



# Triplons in an artificial quantum magnet

# Triplons in an artificial quantum magnet



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

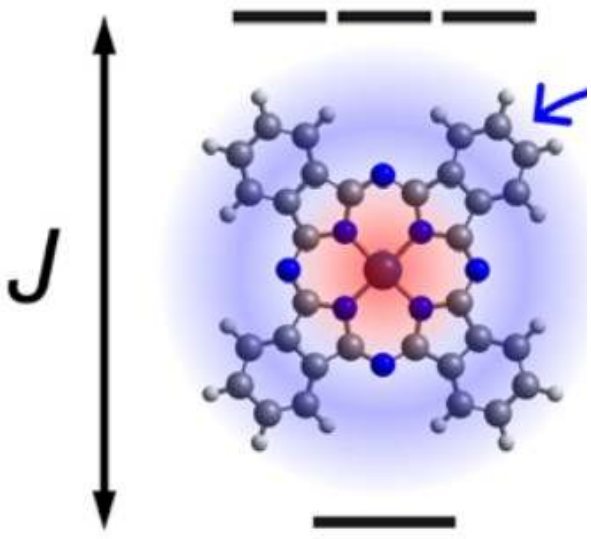
**Triplons in an atomic-scale quantum magnet**

Robert Drost

Shawulienu Kezilebieke

Peter Liljeroth

# Triplet excitations in a molecule




S = 1

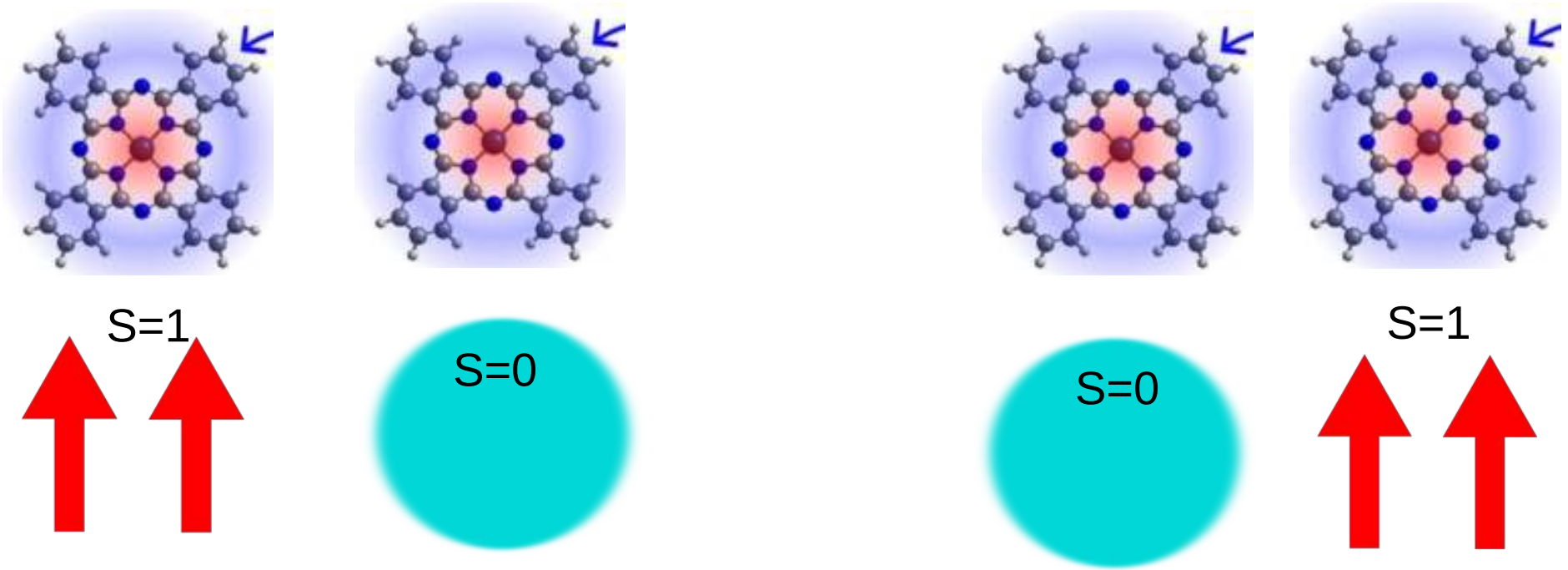
S = 0

Internal antiferromagnetic coupling

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

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

# Triplons in a dimer

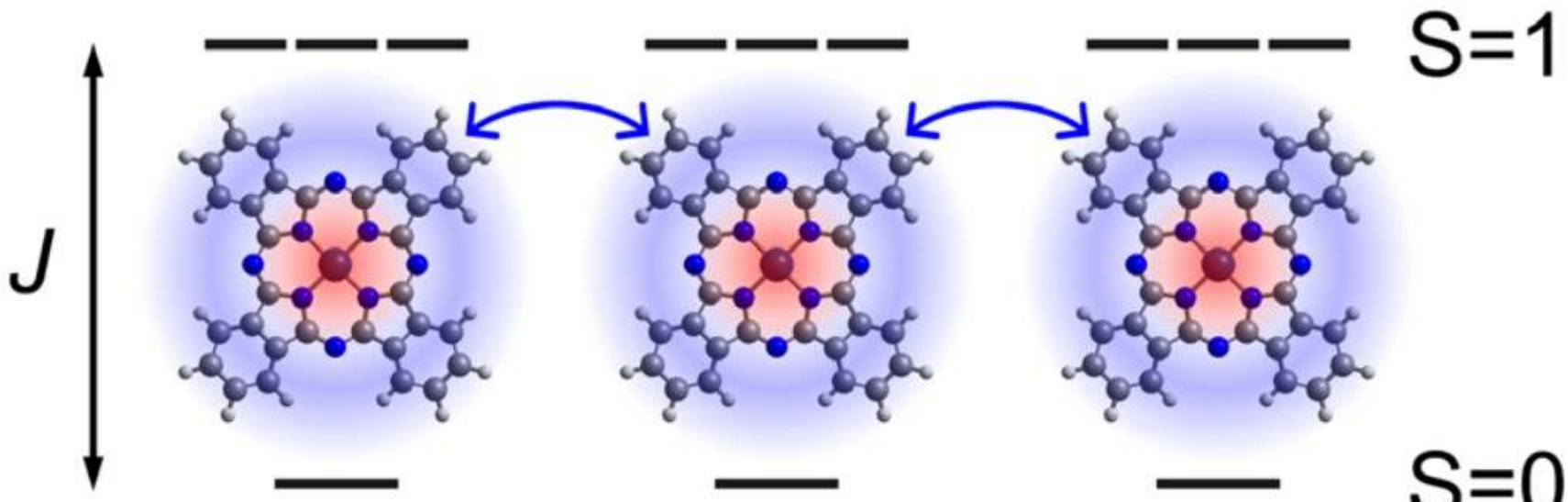



A triplet in the first molecule

Can jump to the second molecule

**Exchange coupling between molecules leads to propagating triplon excitations**

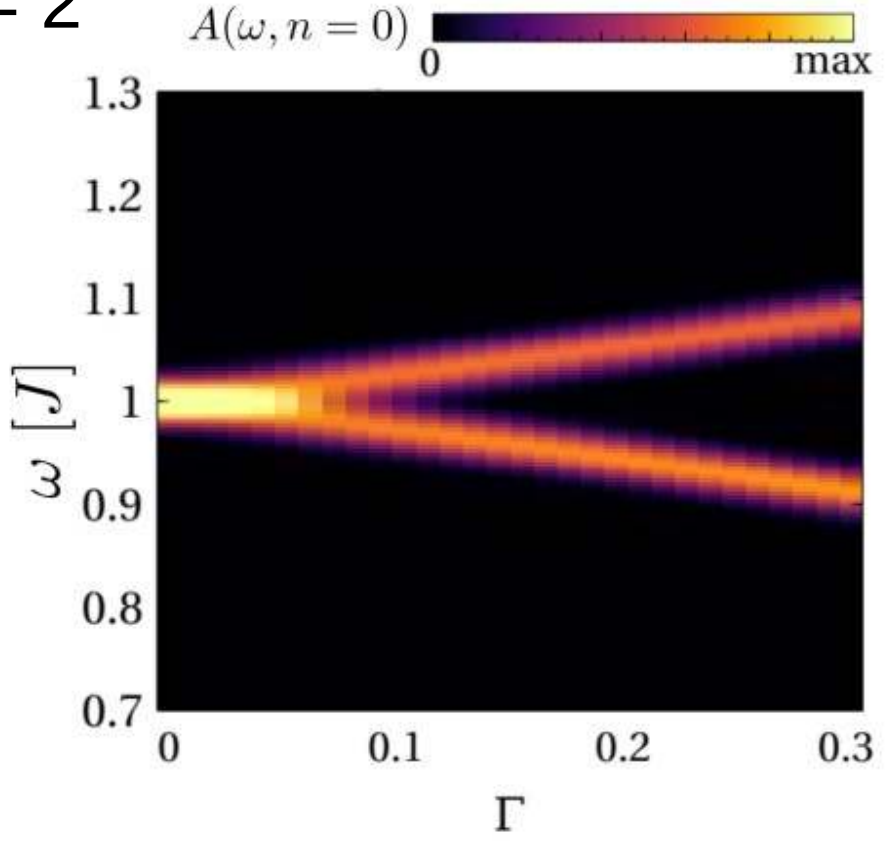
# A chain of triplons



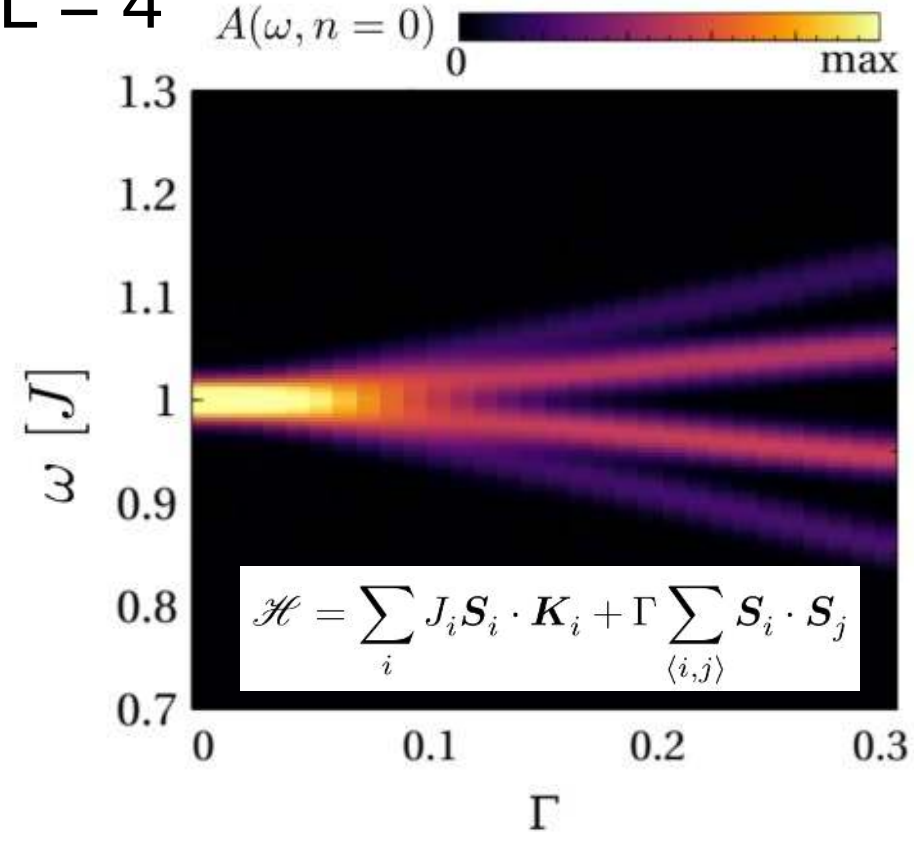

$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

# Chains of triplons

L = 2

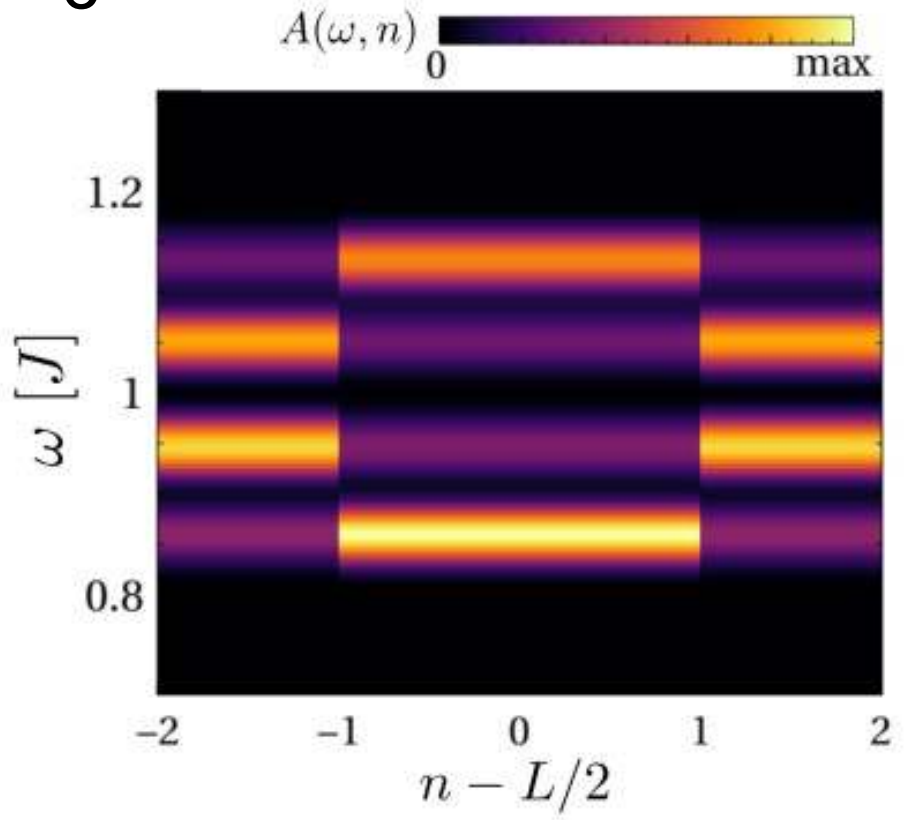



L = 4

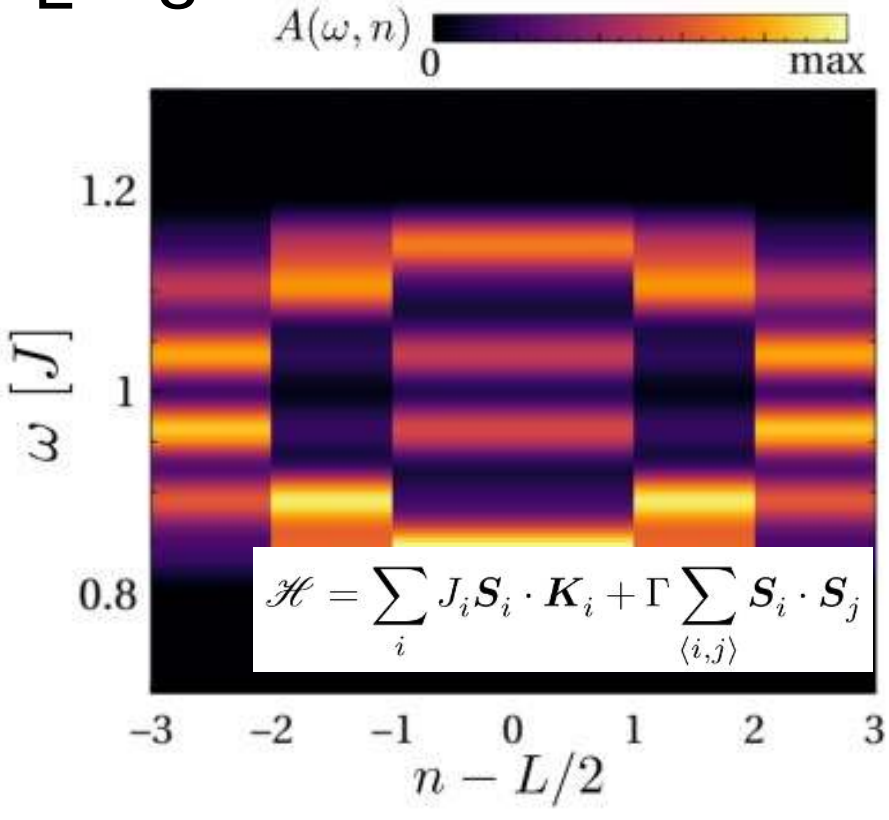


# Chains of triplons

L = 6

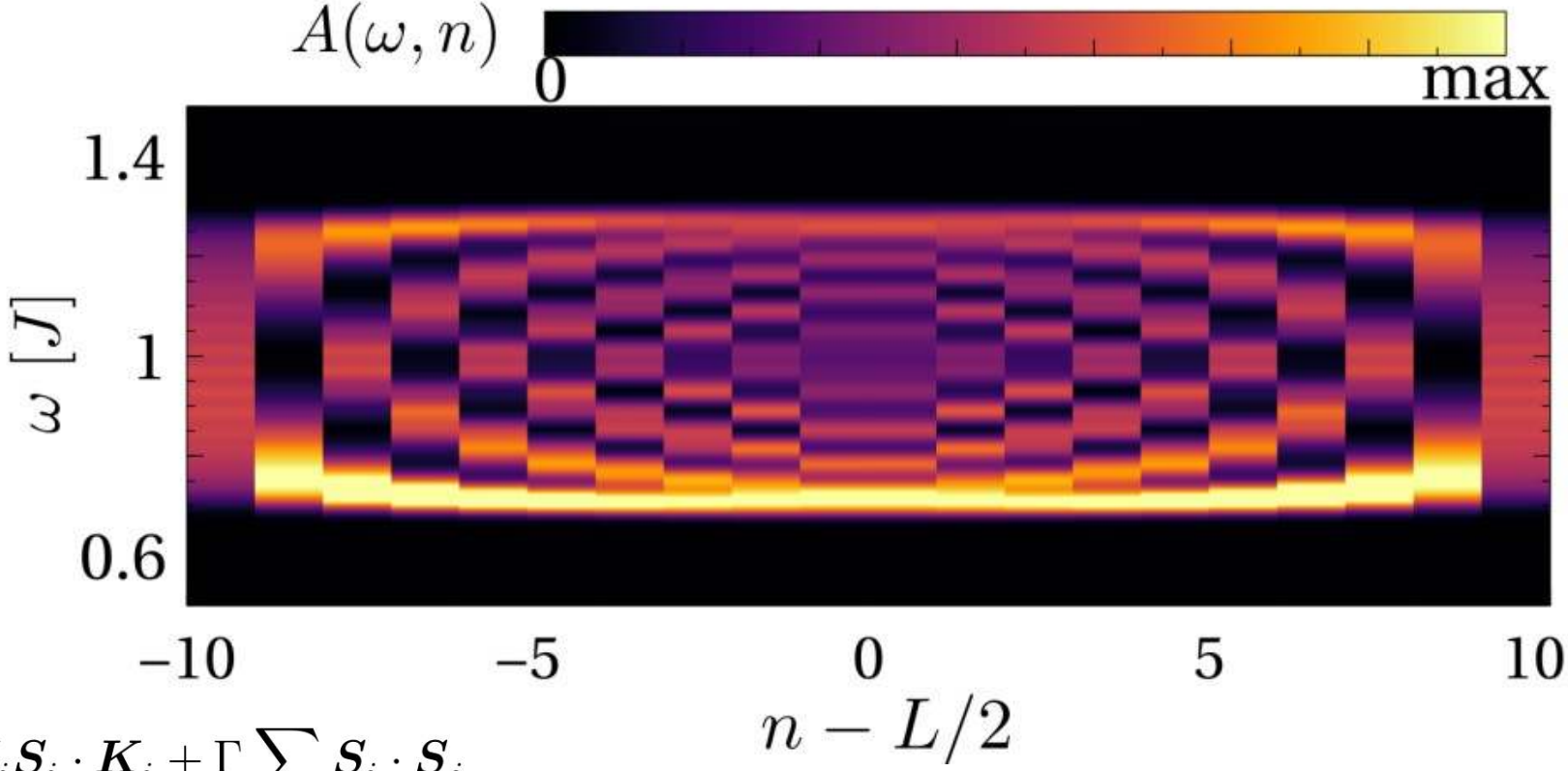



L = 8



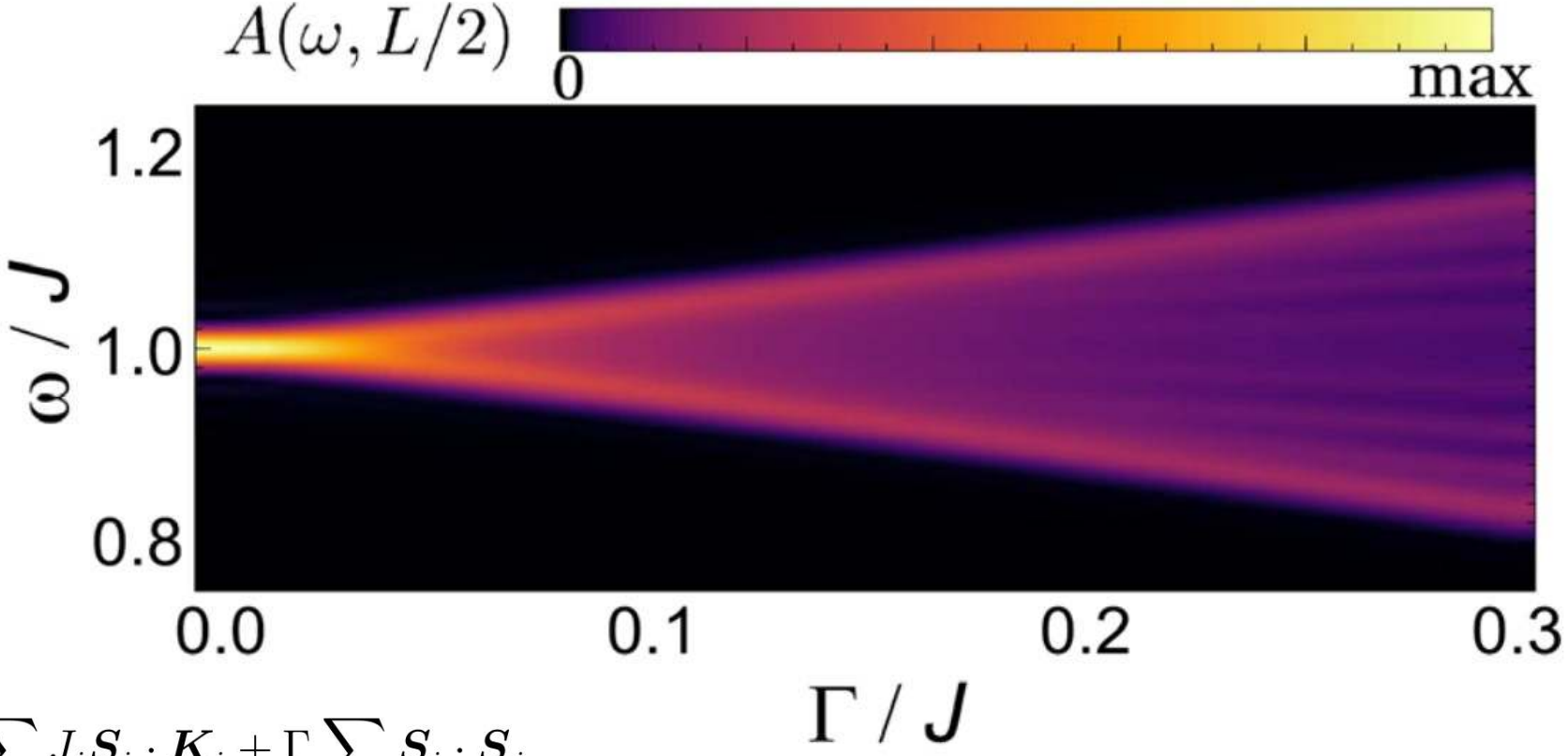
# Chains of triplons

L = 20



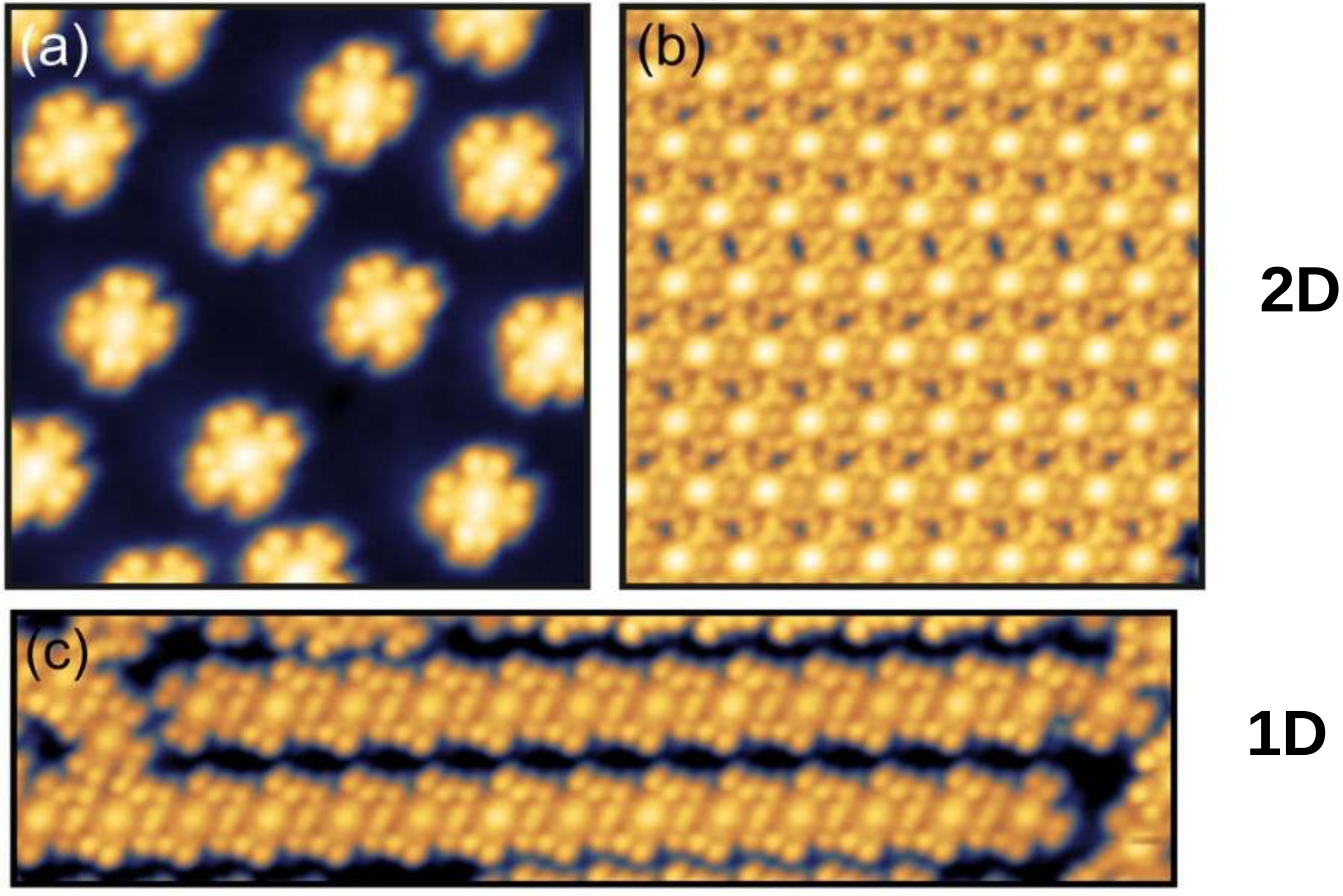

$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

# 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

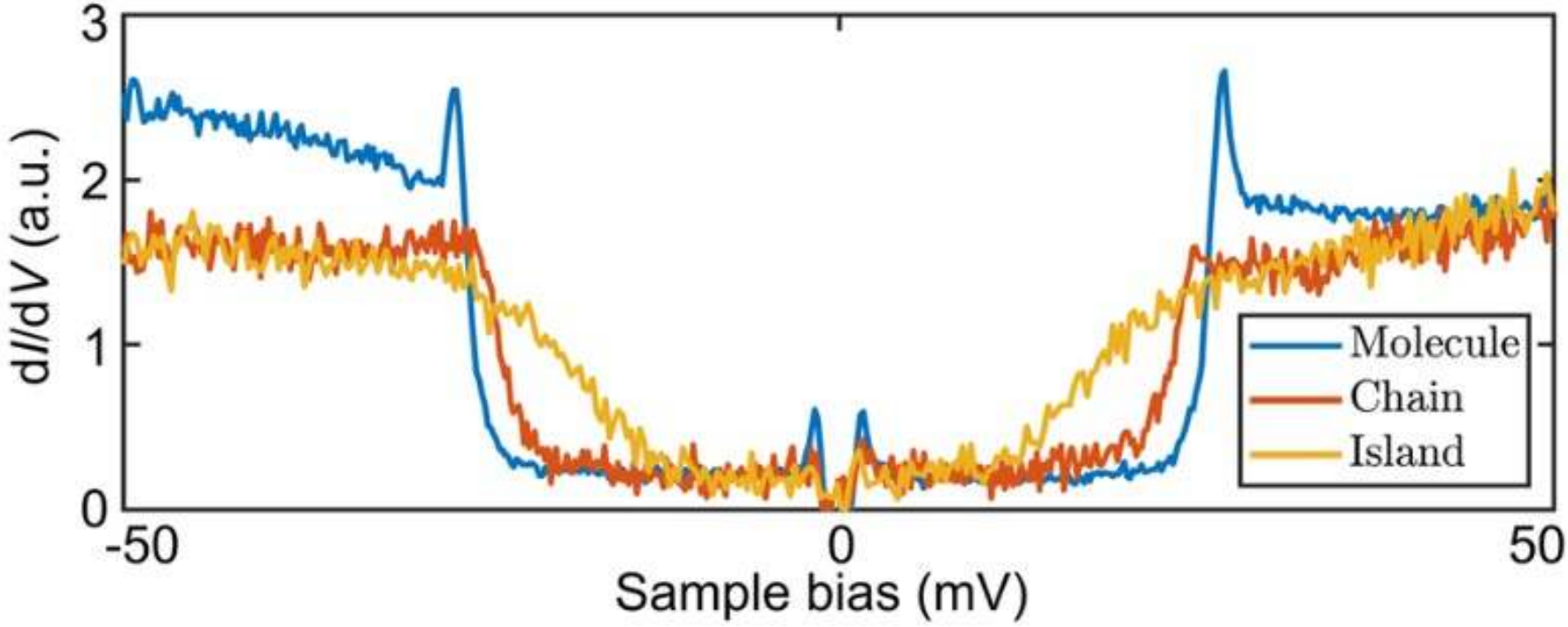



**0D**

**2D**

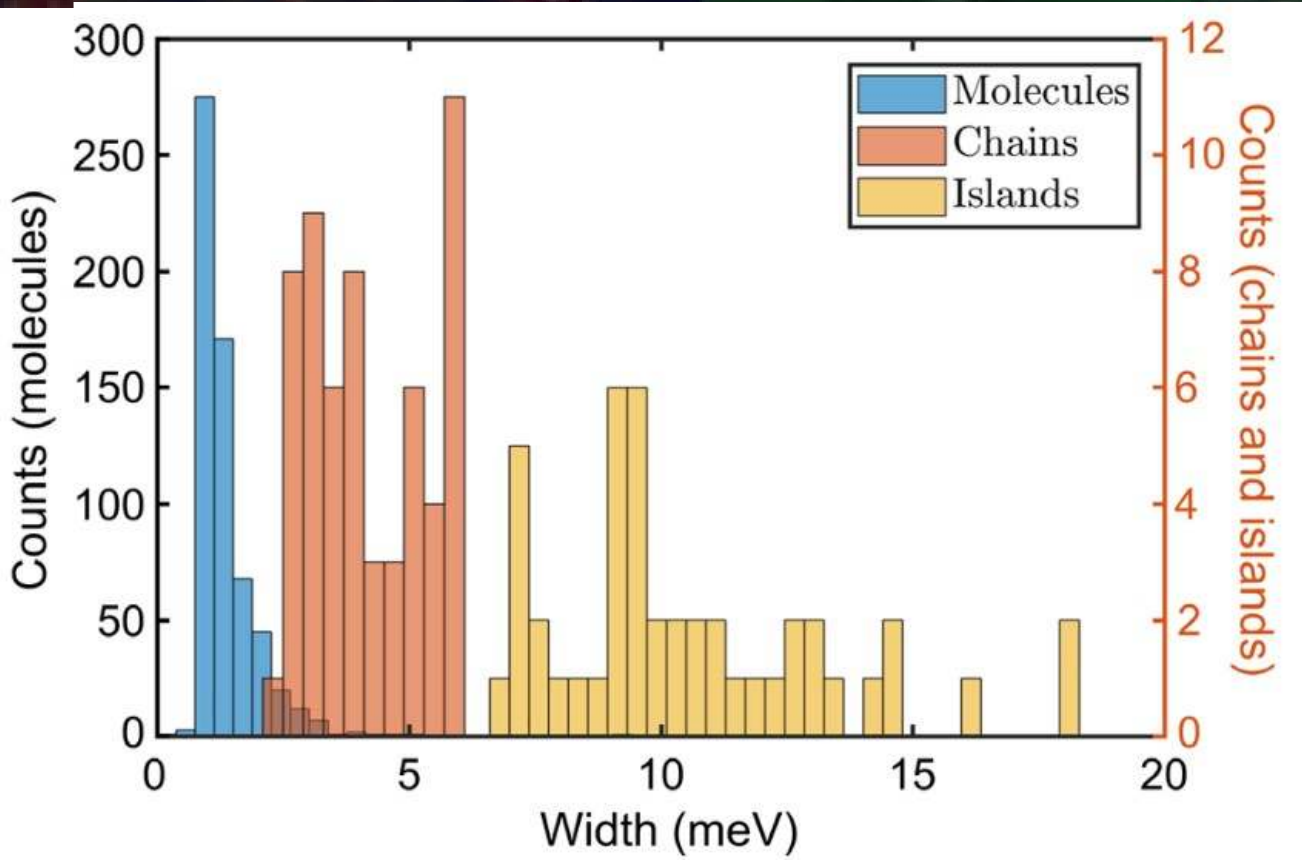
**1D**

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



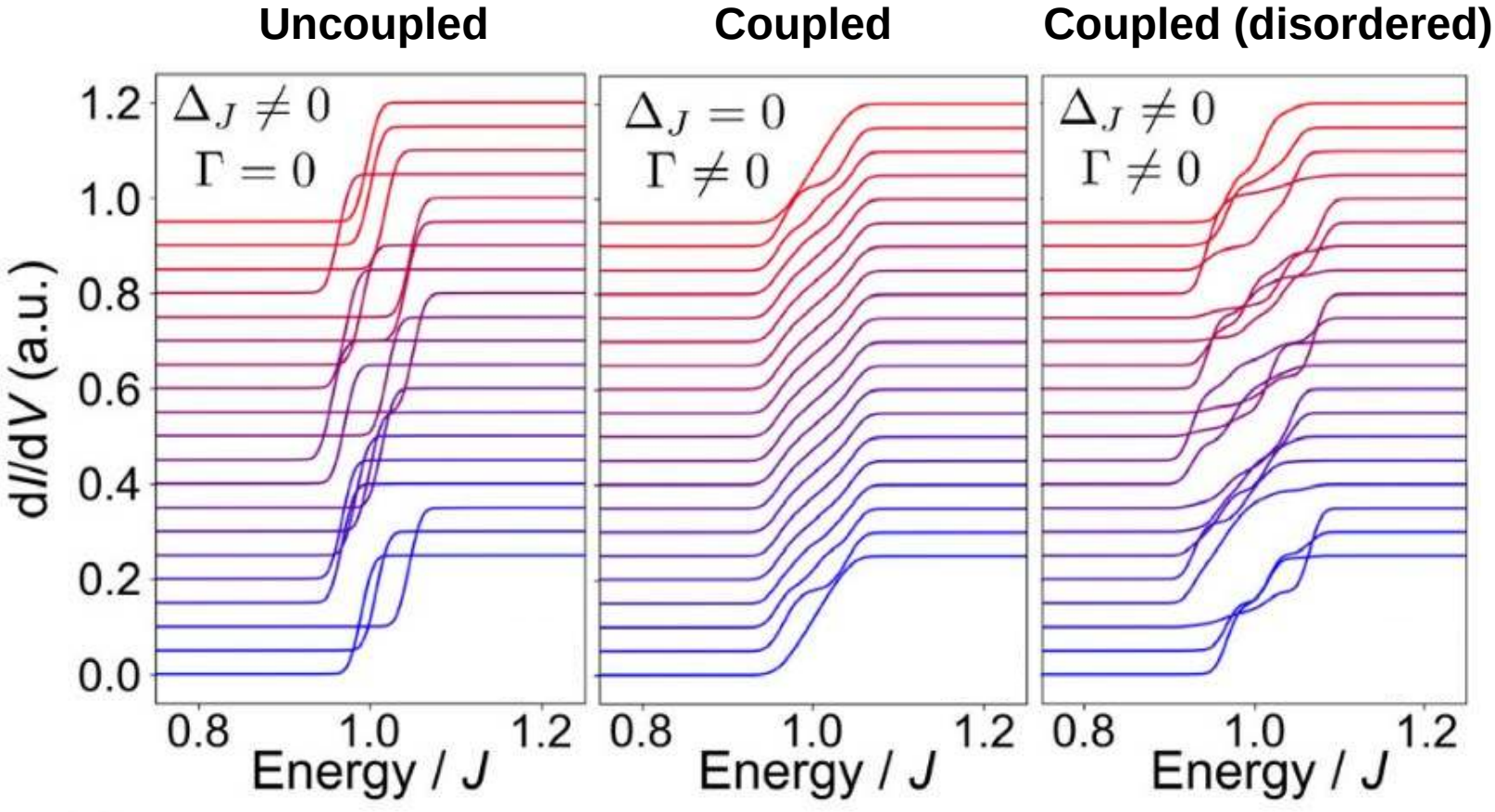

**The width of the step is bigger for islands than for molecules**

# Statistic of the inelastic steps

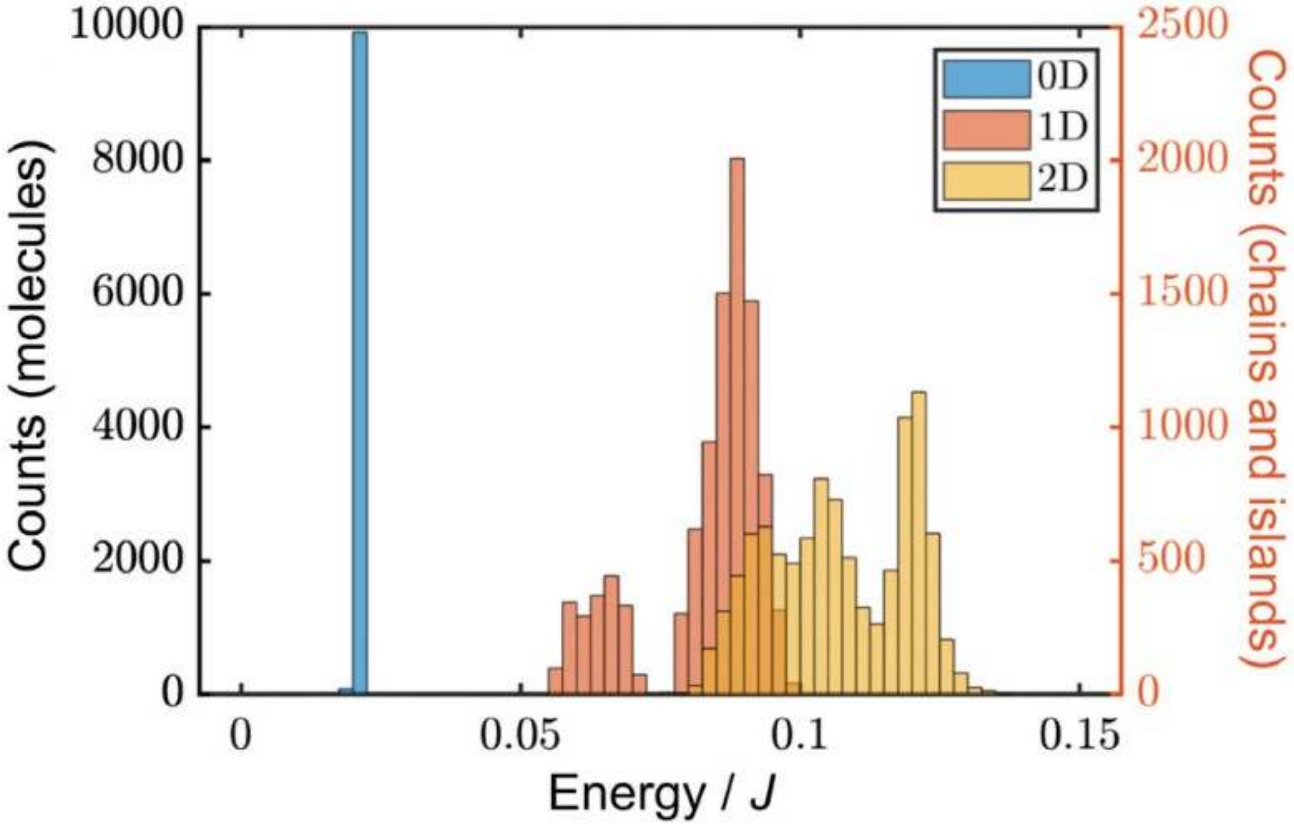



**2D arrays have the largest width of the spin excitation**

# Coupled and uncoupled inelastic excitation



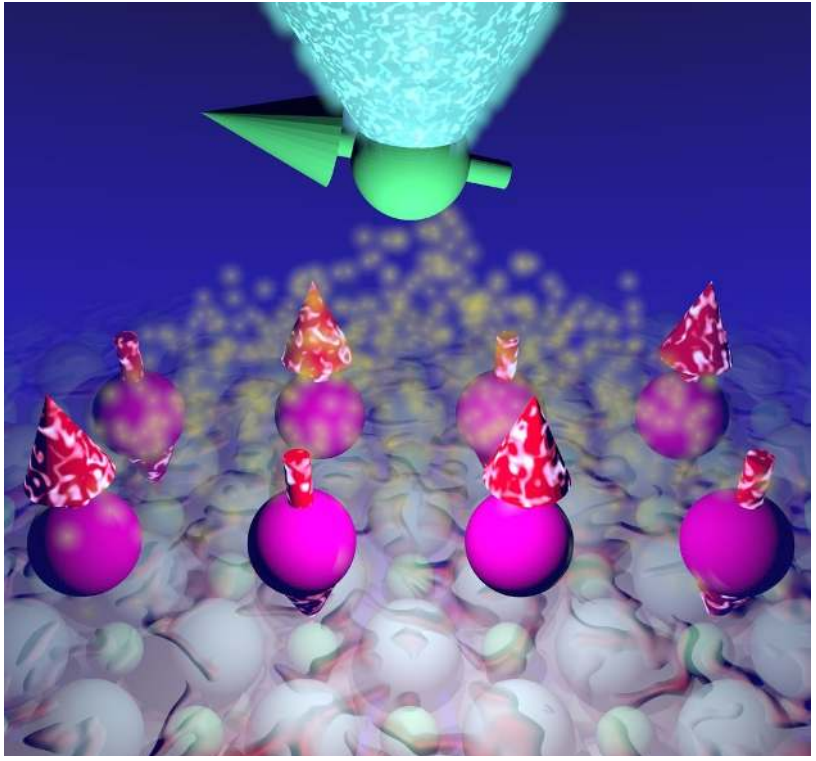
# Statistics of the theoretical model




**2D arrays have the largest width of the spin excitation**

# Hamiltonian learning an atomic-scale magnet

# Hamiltonian learning an atomic-scale magnet



**Hamiltonian learning atomic-scale magnets**

Netta Karjalainen

Zina Lippo

Rouven Koch

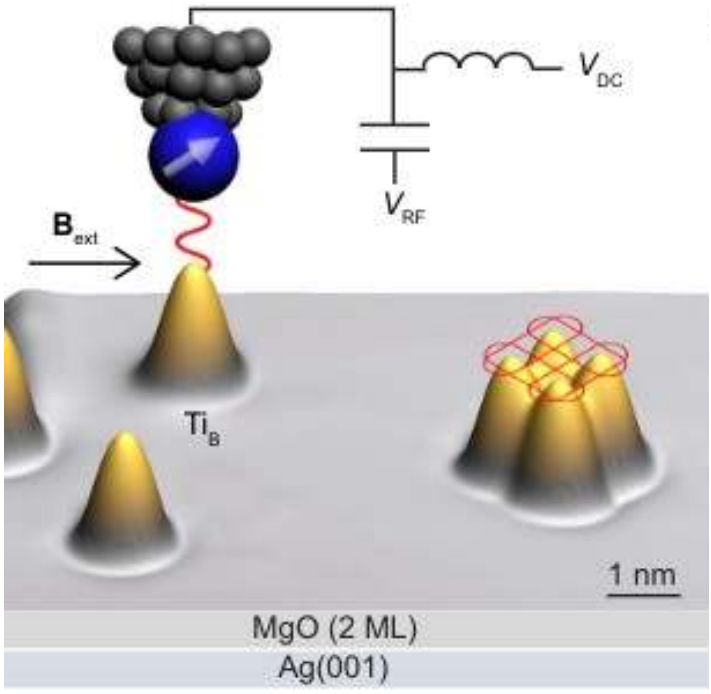
Guangze Chen

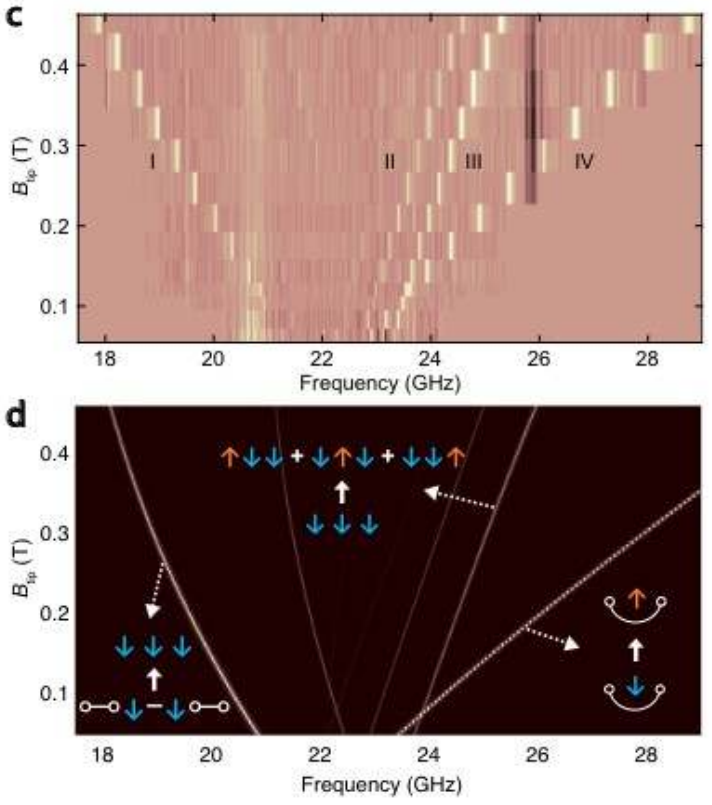
Adolfo Fumega

Phys. Rev. Applied 20, 024054 (2023)

# Spin excitations in S=1/2 islands

Ti@MgO (2x2 lattice)

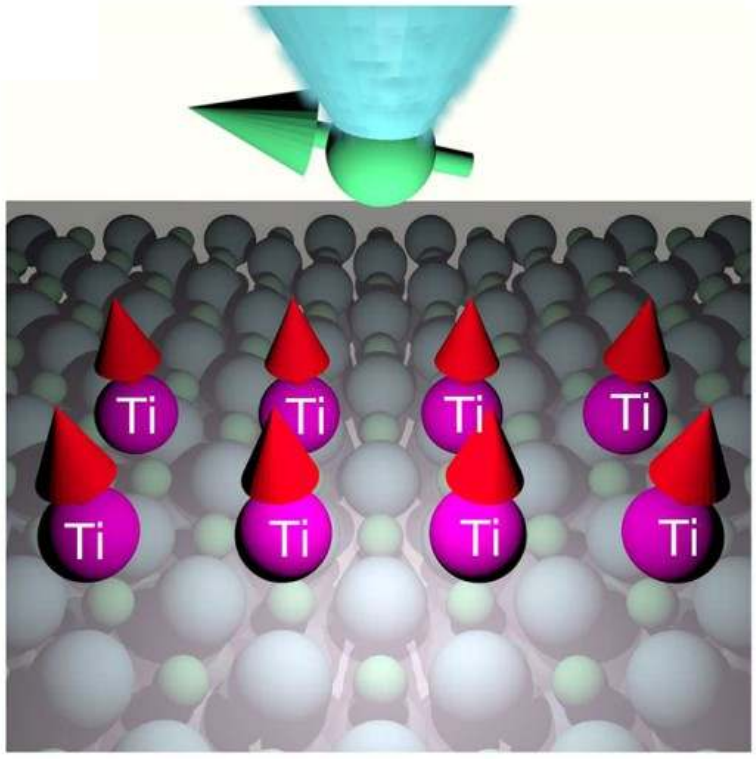


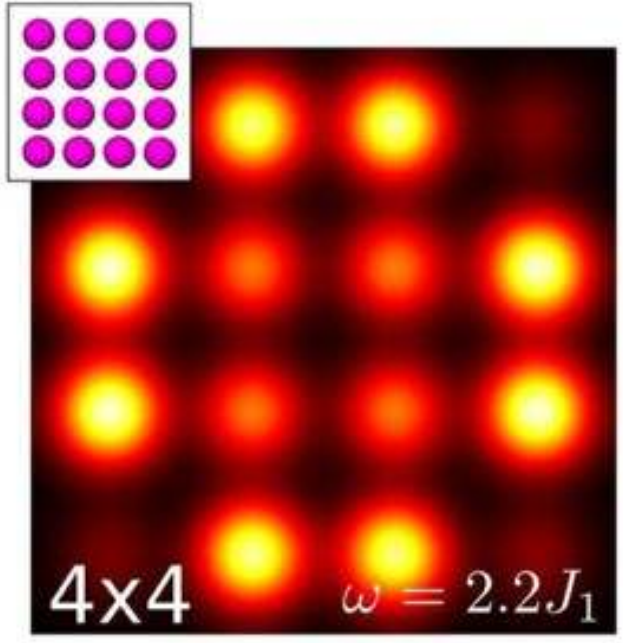




Nature Communications 12, 993 (2021)

**Lets assume we can go to bigger islands!**

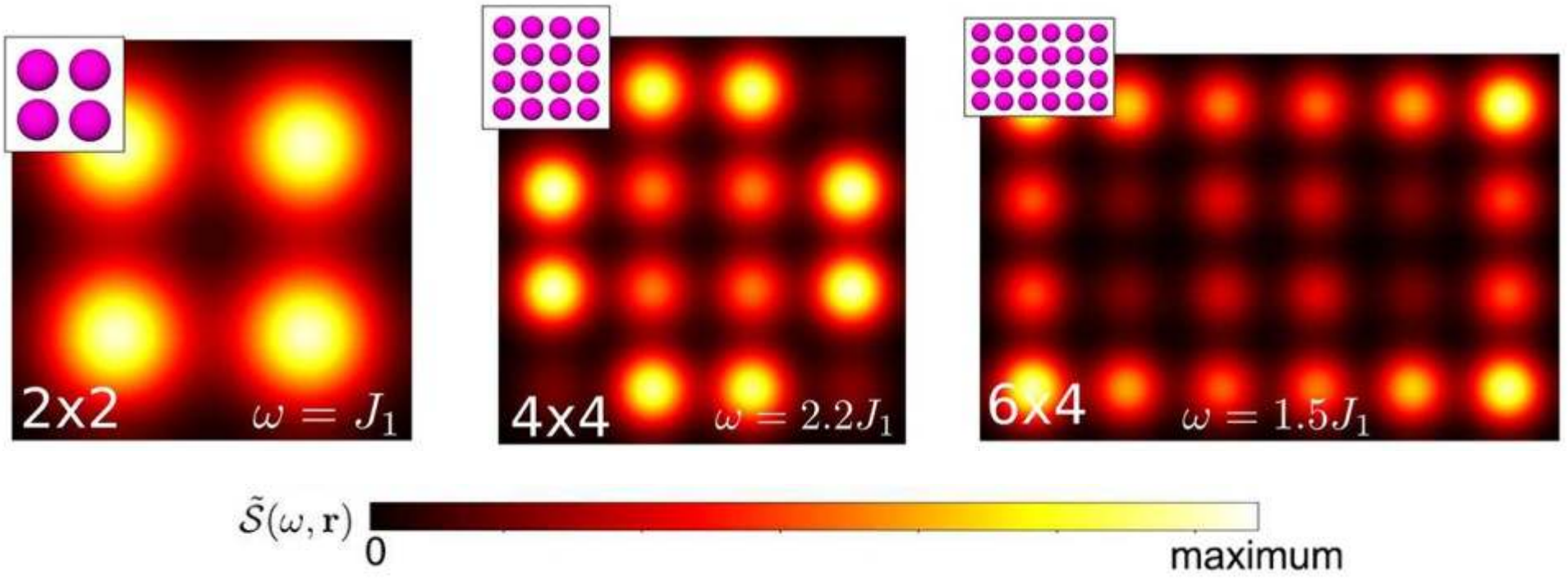
# Spatially resolved excitations



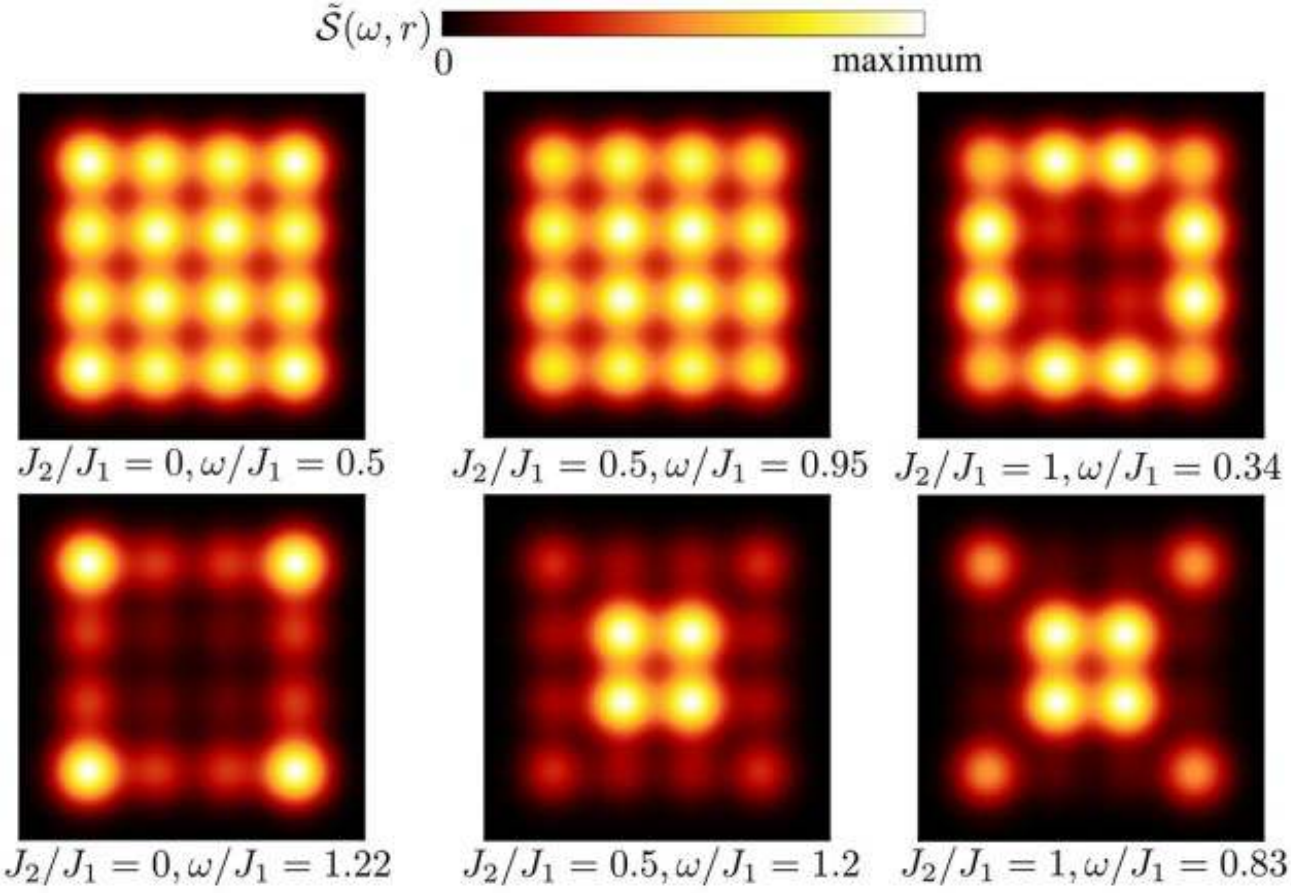

***What can we learn from the Hamiltonian using real-space information?***

# Spatially resolved excitations



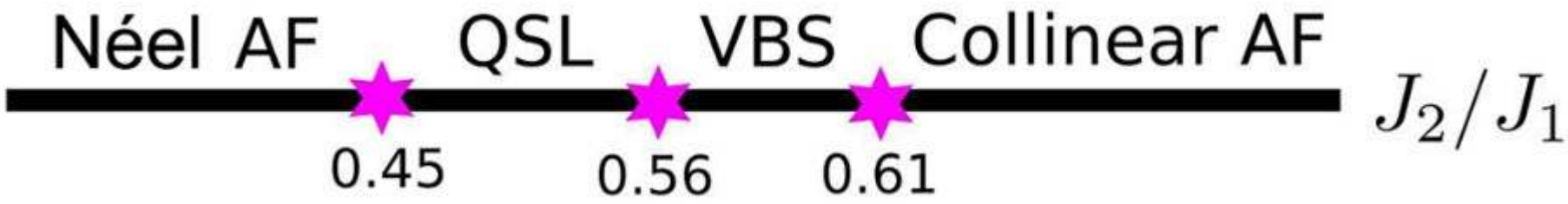

***Spin excitations behave like particle in a box states***

# Spin excitation modes in spin islands



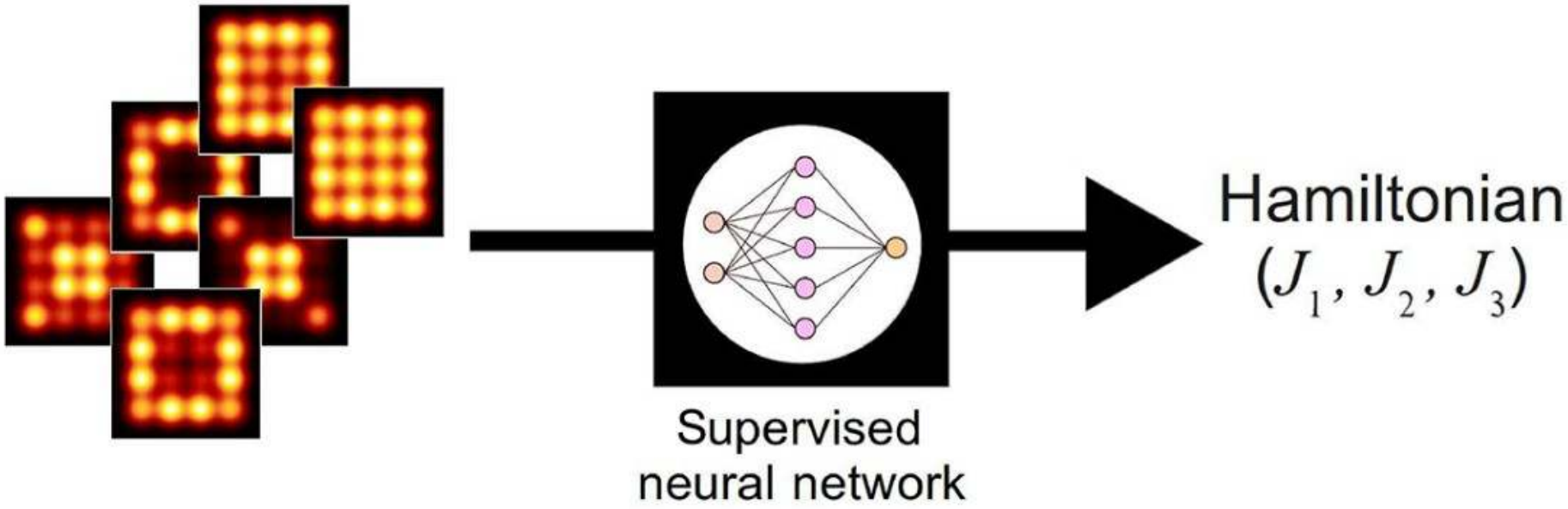
# The extended Heisenberg model

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




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

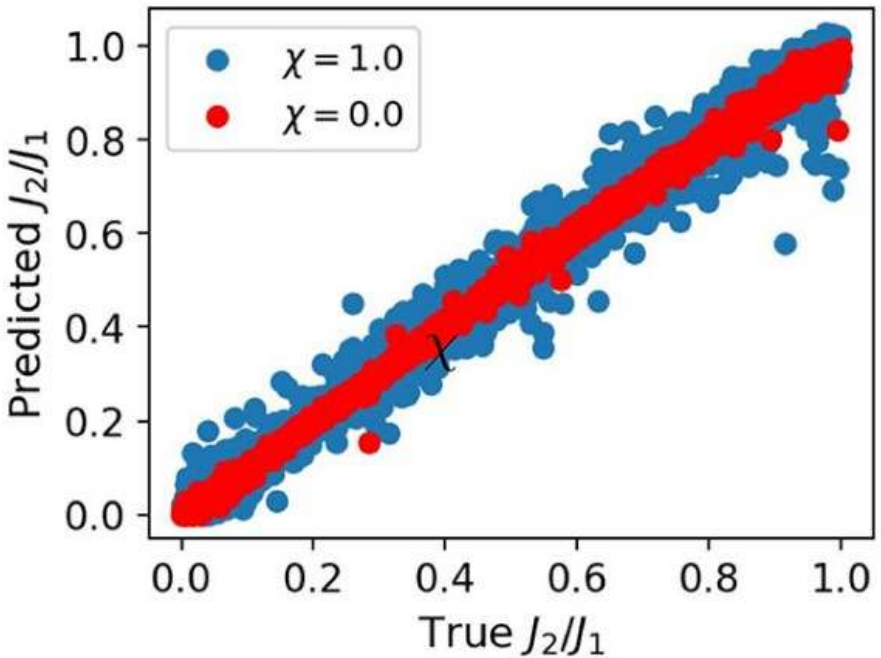
# The idea of Hamiltonian learning

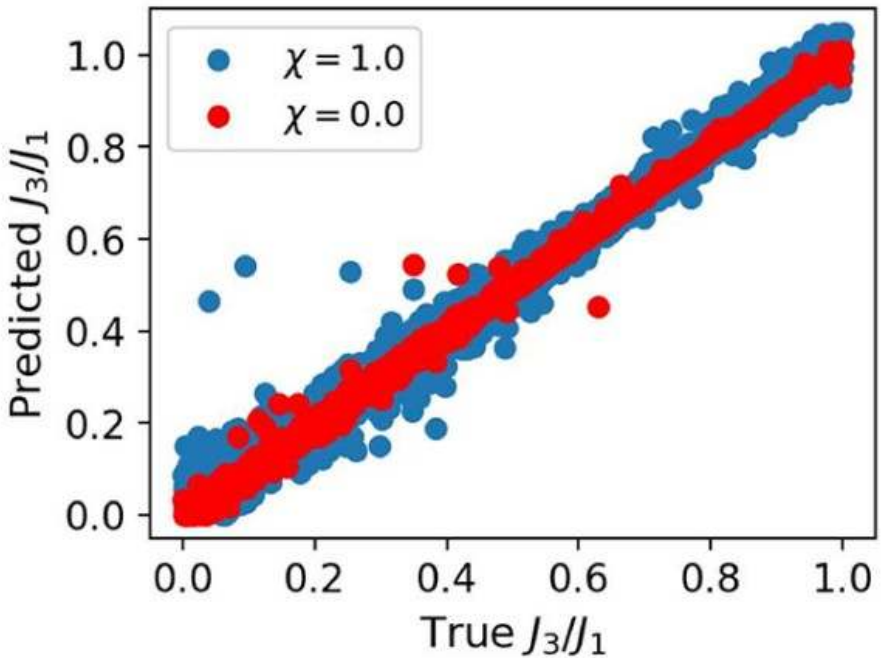



**Provide the spectroscopy as input, obtain the Hamiltonian as output**

# Real VS predicted exchange parameters

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

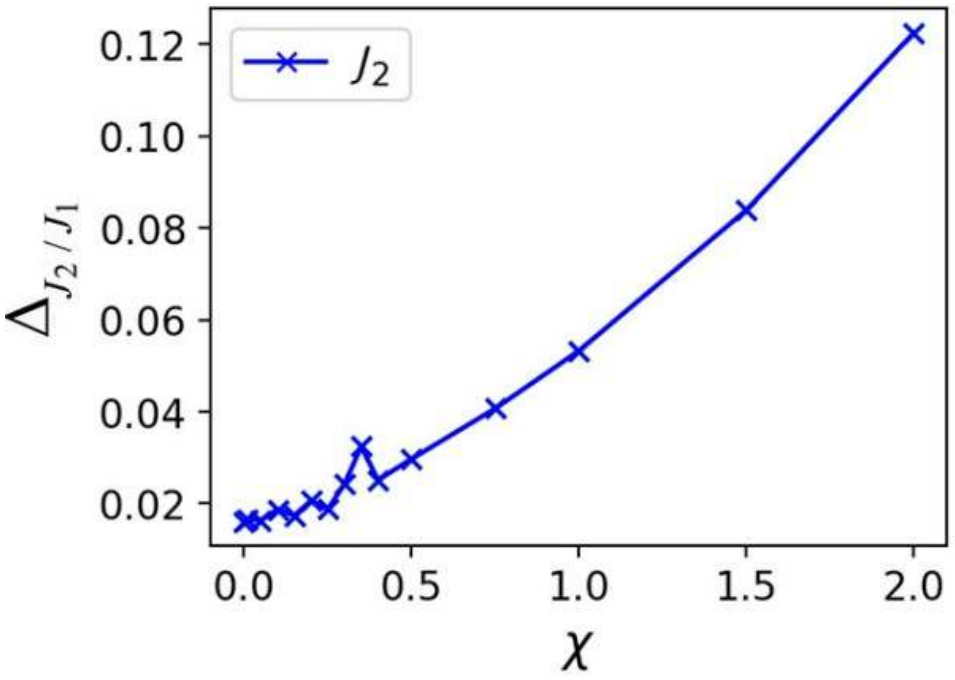


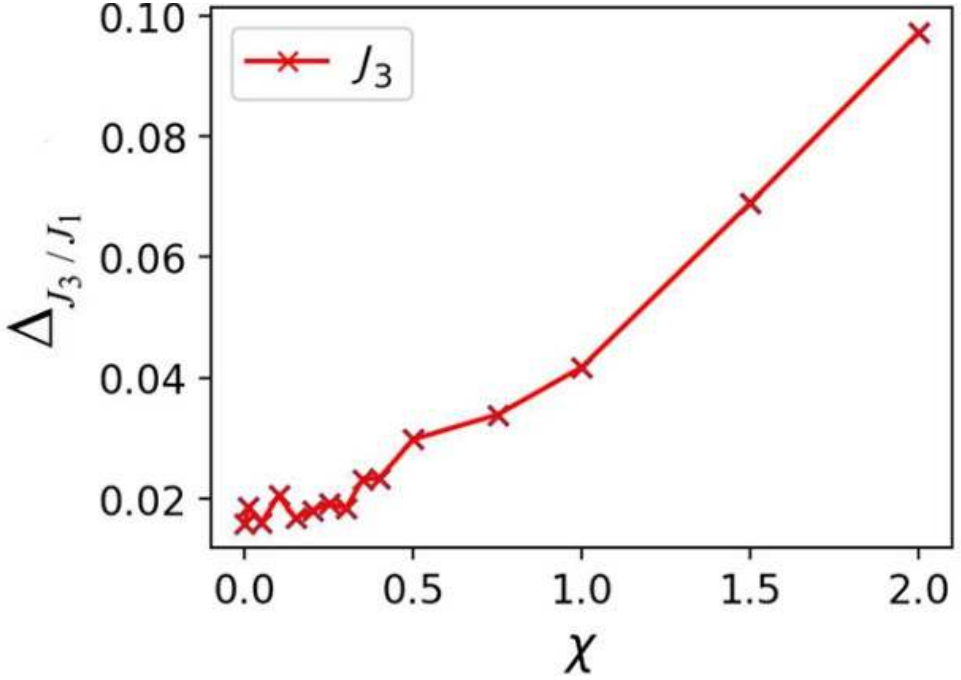




***A supervised learning algorithm extracts faithfully the exchange parameters***

# Predictions with noise

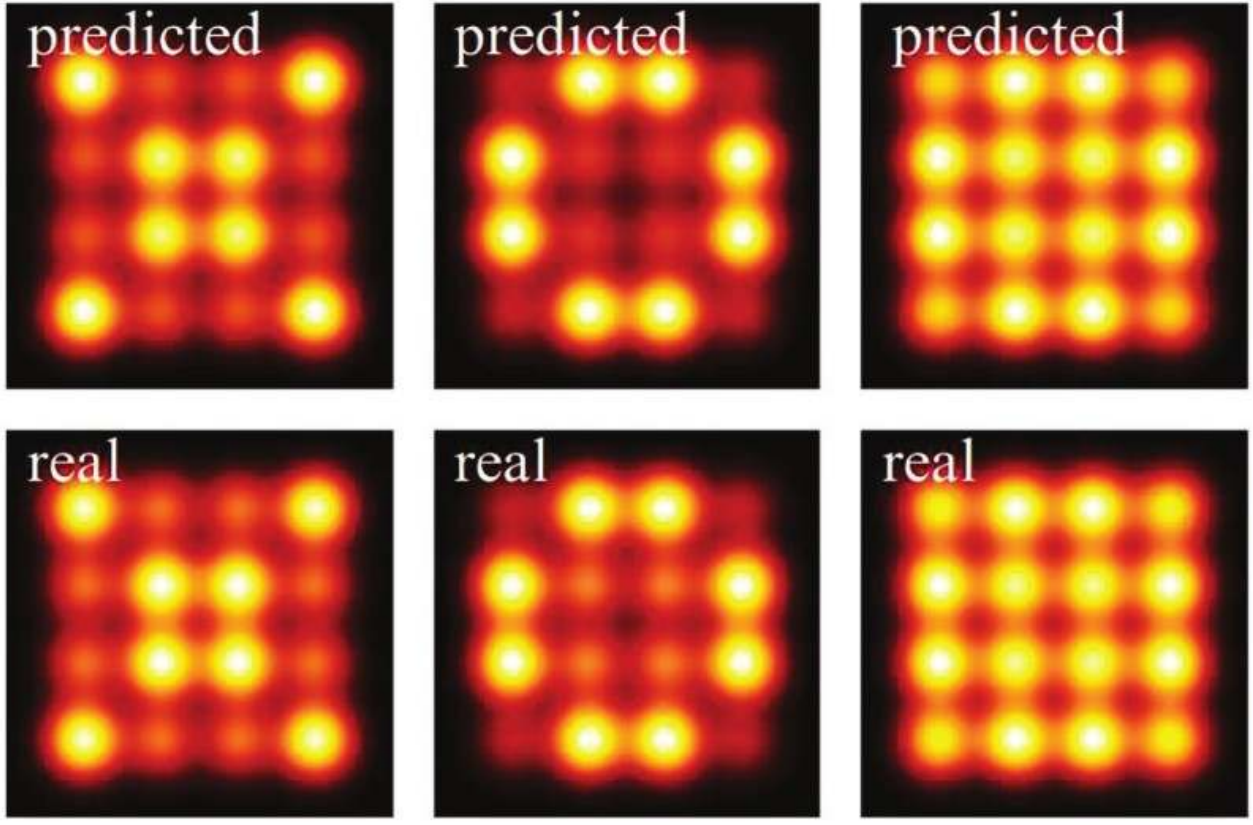
$$\mathcal{S}(\omega, \mathbf{r}_i)_{\text{noisy}} = \mathcal{S}(\omega, \mathbf{r}_i) \cdot |1 + \zeta(\omega)| \qquad \zeta(\omega) \in [-\chi, \chi]$$




***Quality of prediction remain good with sizable noise levels***

# Predicted VS real spin excitations



# Open-source software for artificial quantum materials

# Open-source software for many-body quantum magnets

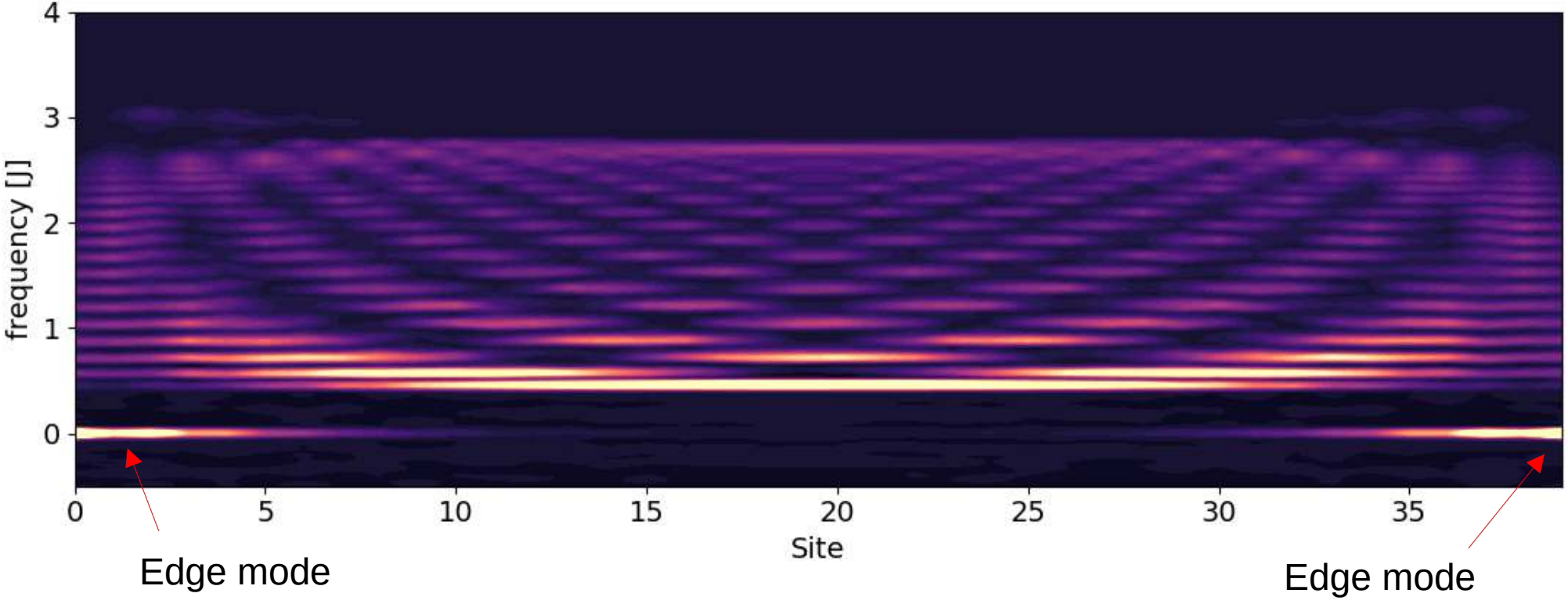
*Python library for tensor-network kernel polynomial algorithms for spins, fermions, parafermions, with static solvers for Hermitian and non-Hermitian modes*

```
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]))
```

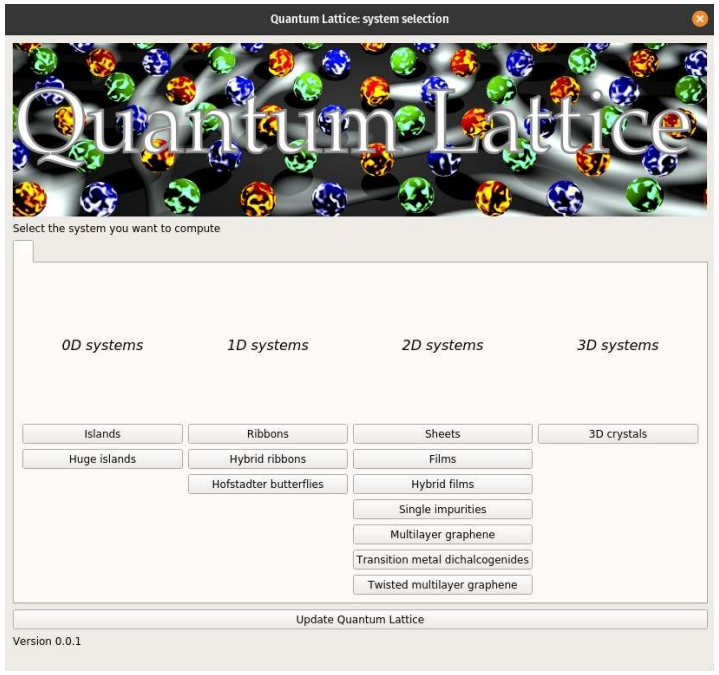
**dmrgpy** https://github.com/joselado/dmrgpy

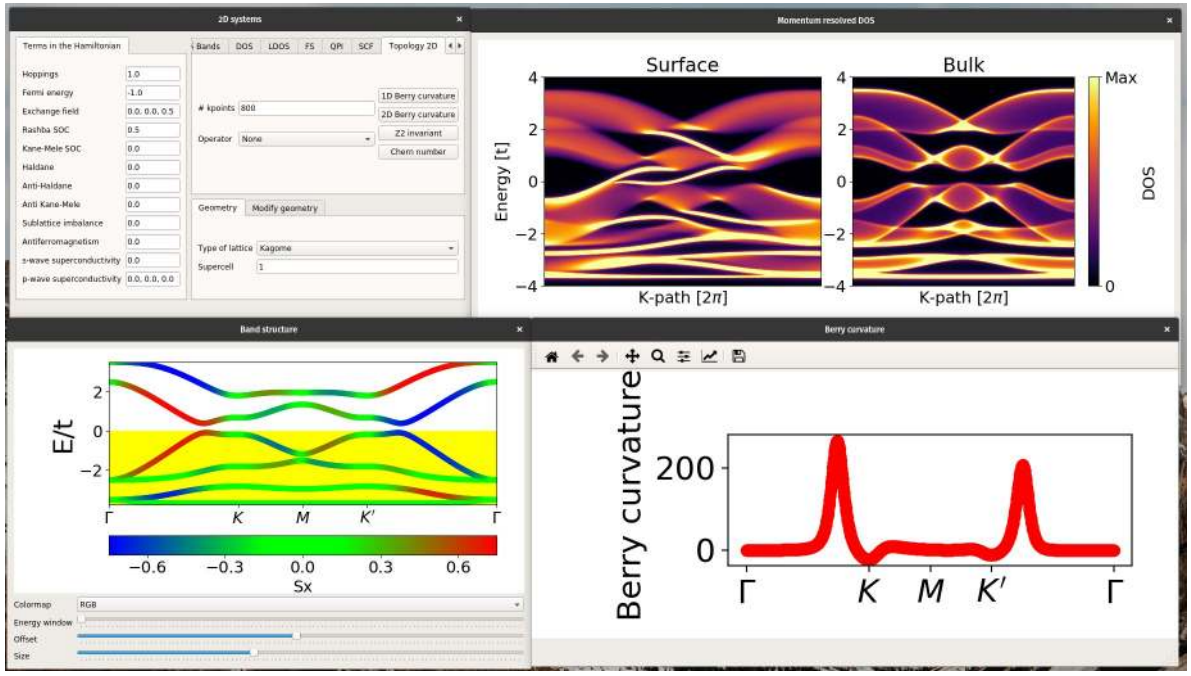
# Open-source software for many-body quantum magnets

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



# Quantum Lattice: A user interface to compute electronic properties

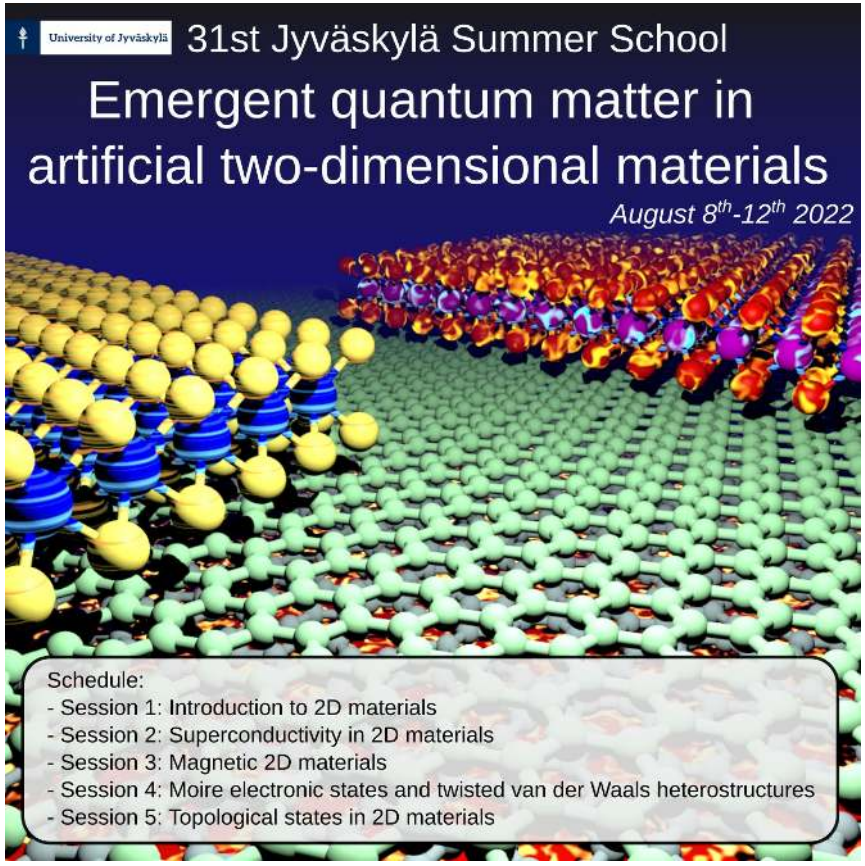



**Quantum Lattice**: open source interactive interface for tight binding modeling

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

# Quantum matter in van der Waals materials school



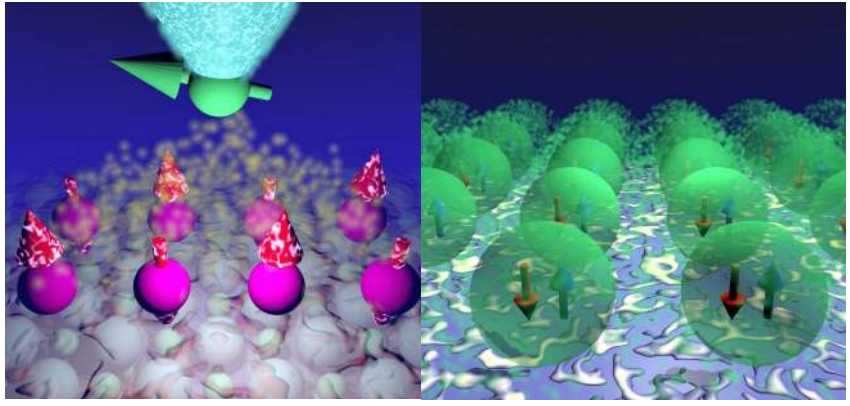

**Recordings available at**

https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ

- Session 1: Introduction to 2D materials
- Session 2: Superconductivity in 2D materials
- Session 3: Magnetic 2D materials
- Session 4: Moire electronic states and twisted van der Waals heterostructures
- Session 5: Topological states in 2D materials

# Take home

Atomically-assembled spin lattices allows us to create quantum matter with emergent quantum excitations



Phys. Rev. Lett. 131, 086701 (2023)
Phys. Rev. Applied 20, 024054 (2023)

Funding from

ACADEMY OF FINLAND

JANE AND AATOS ERKKO FOUNDATION

**Thank you**