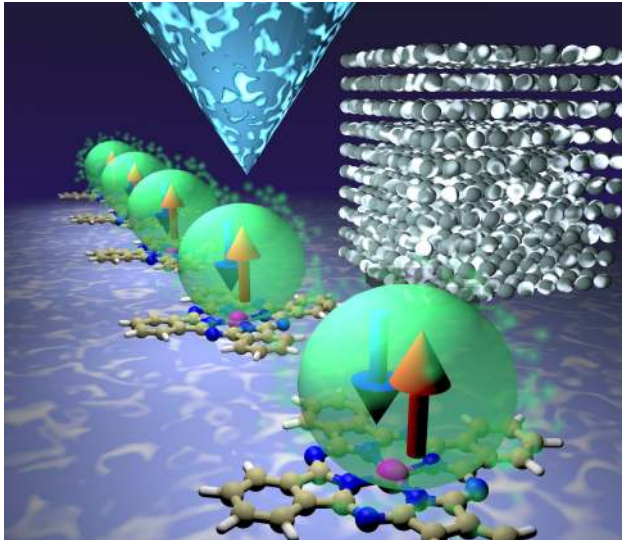
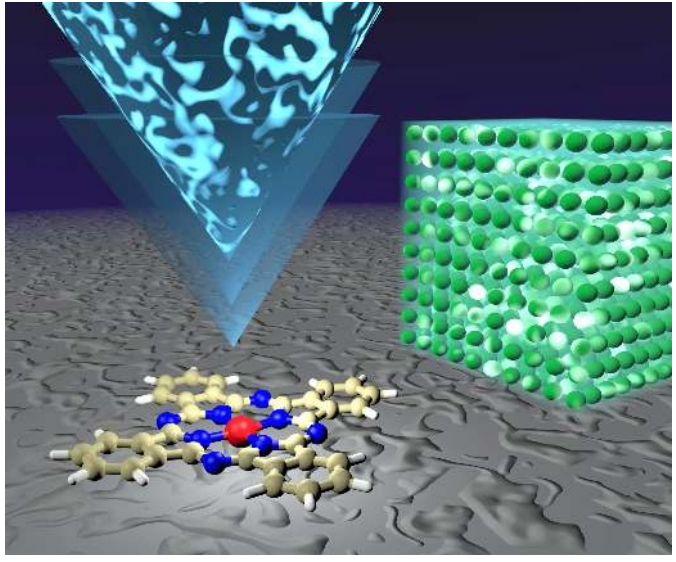
Hamiltonian learning nanoscale quantum magnets and multiorbital excitations with setpoint-dependent scanning tunneling spectroscopy

Jose Lado

Department of Applied Physics, Aalto University, Finland





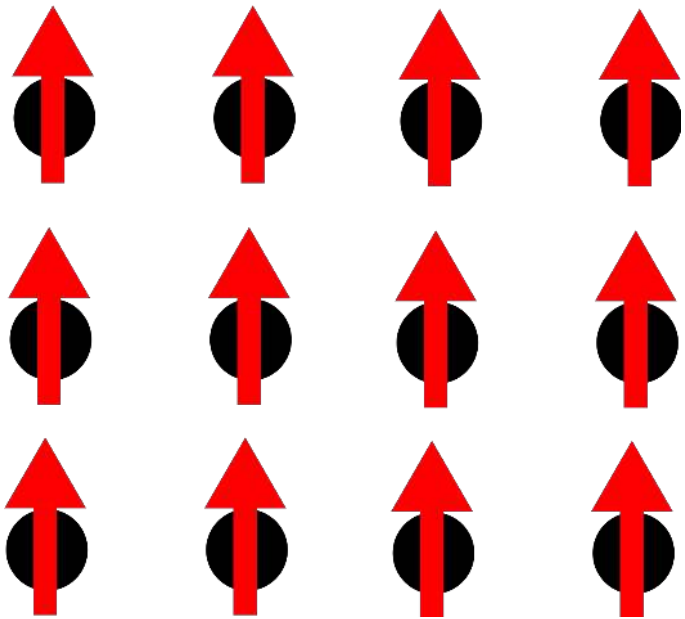
Nano Letters 25, 36, 13435-13440 (2025)

arXiv:2601.19371 (2026)

Atomic structure of nanosystems from first-principles simulations and microscopy experiments (AS-SIMEX 2026), June 2nd 2026

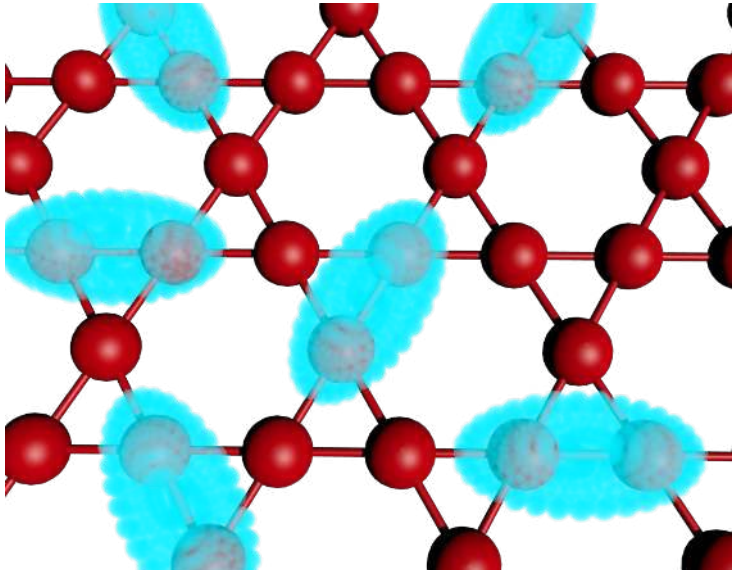
Classical and quantum magnetism

Ferromagnet & antiferromagnets



Classical ground state $\langle \vec{S}_n \rangle \neq 0$

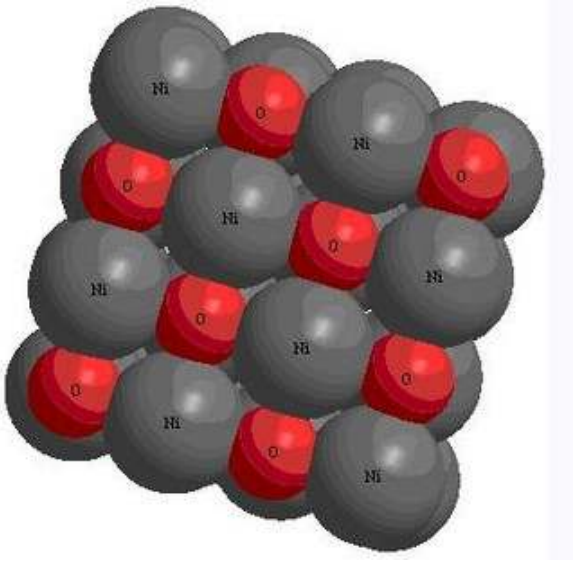
Quantum spin-liquids



Quantum ground state $\langle \vec{S}_n \rangle = 0$

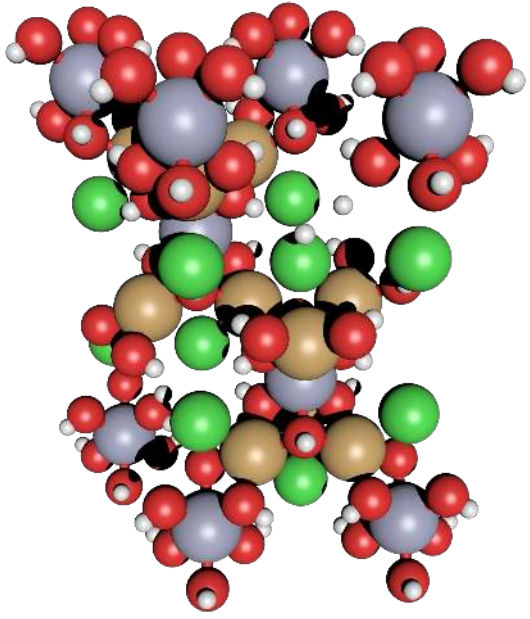
Classical and quantum magnetic materials

Ferromagnets & antiferromagnets



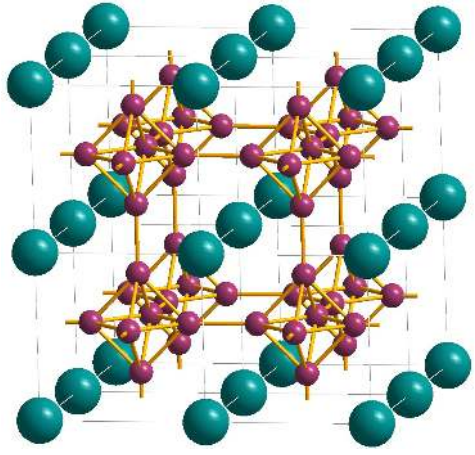
NiO2

Quantum spin-liquids



Herbertsmithite

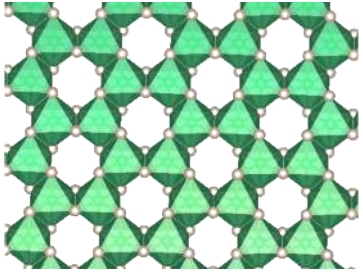
Heavy-fermion Kondo insulators



SmB_6

Classical and quantum magnetic materials, for STM

Ferromagnet, antiferromagnets

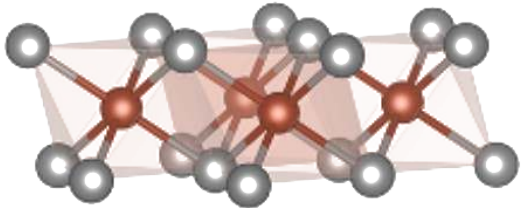


CrI_3, $CrCl_3$, $CrBr_3$

Nature 546, 270–273 (2017)

Break time-reversal

Multiferroics

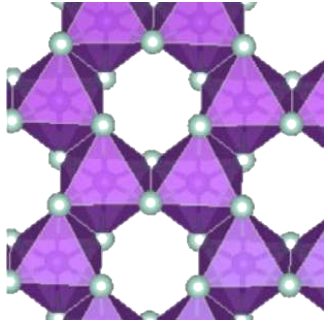


NiI_2

Nature 602, 601–605 (2022)

Break time-reversal and inversion symmetry

(proximal) Quantum spin-liquids

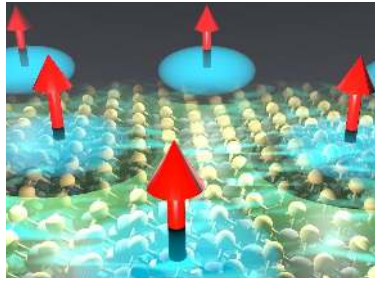


$RuCl_3$, 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Do not break time-reversal

Heavy-fermion Kondo insulators

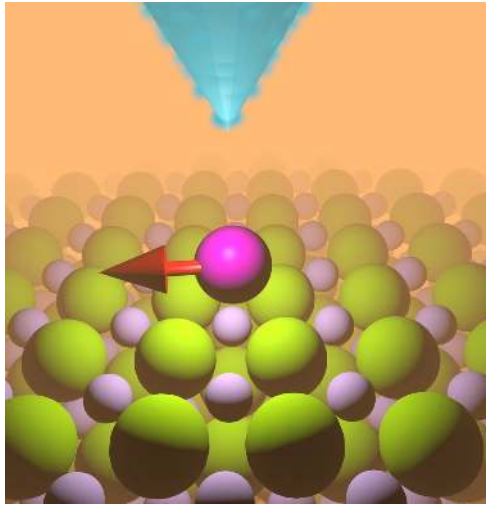


1T-TaS_2/1H-TaS_2

Nature 599, 582–586 (2021)

STM in artificial quantum materials

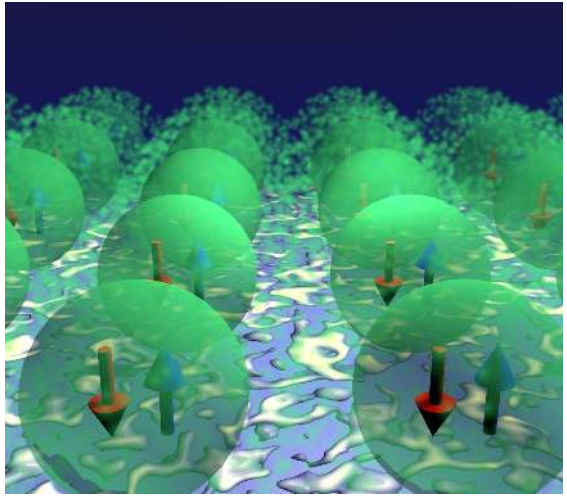
Atomic lattices



a = 0.5 nm

Nature Physics 7 (9), 713-718 (2011)
Nature Physics 13 (7) 668 (2017)
Nature Nano 17 (4), 384-389 (2022)
Science 335 (6065), 196-199 (2012)

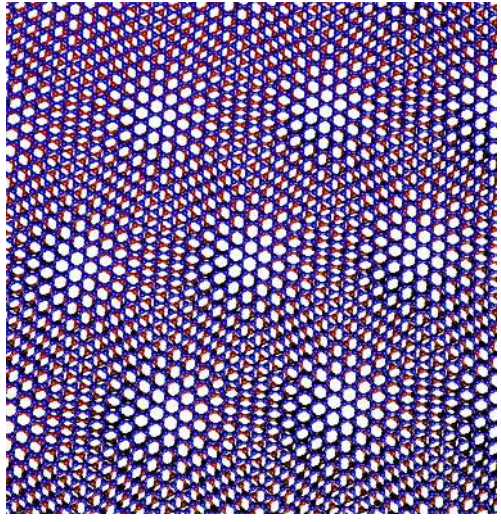
Molecular lattices



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)

2D moire materials

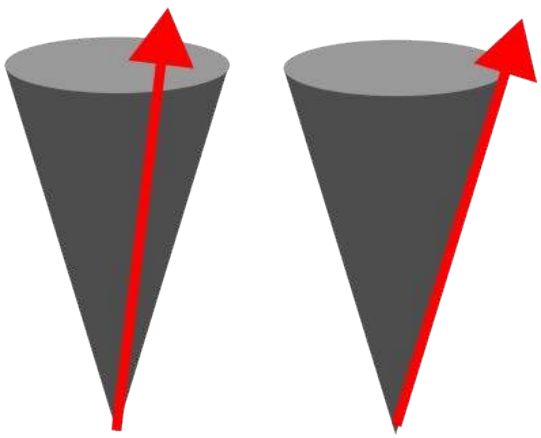


a = 20 nm

Phys. Rev. Lett. 99, 256802 (2007)
Nature Physics 6, 109–113 (2010)
PNAS 108 (30) 12233-12237 (2011)
Nature 556, 80–84 (2018)

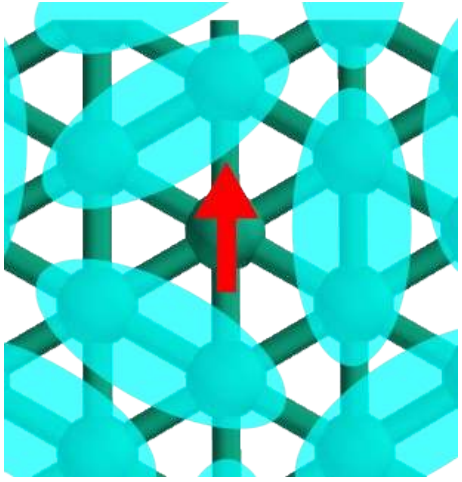
Excitations in correlated magnets

Magnons



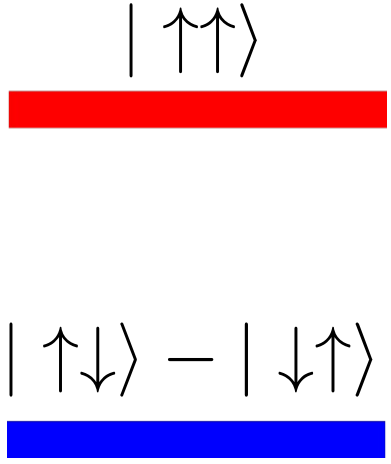
S=1

Spinons



S=1/2

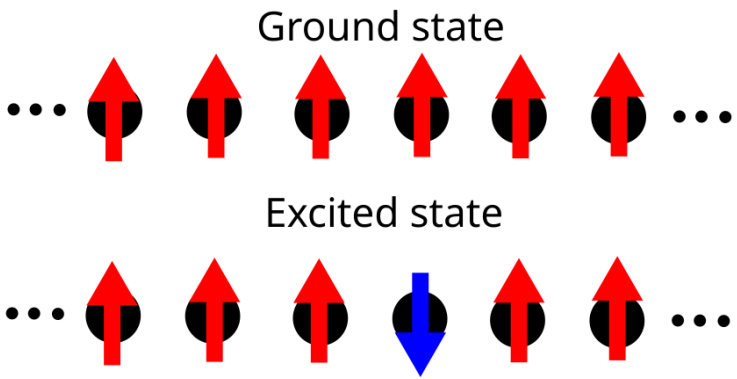
Triplons



S=1

Two excitations in magnetic materials

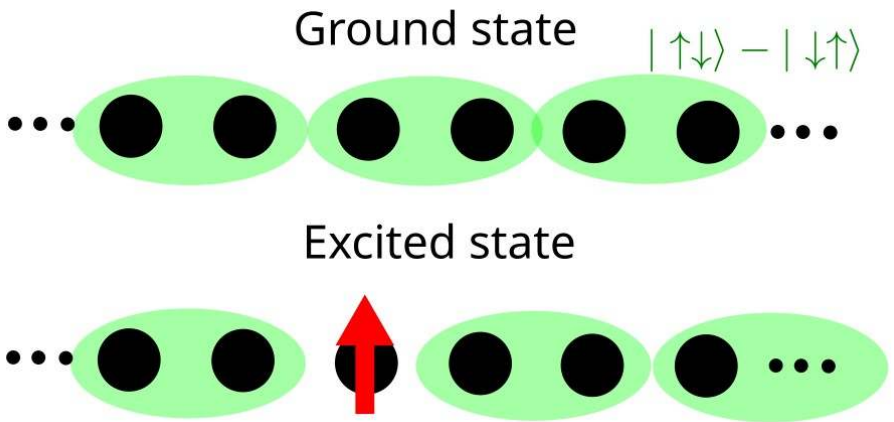
Excitations in a symmetry broken magnet



$$\Delta S = 1$$

Magnons

Excitations in a quantum spin liquid



$$\Delta S = 1/2$$

Spinons

A minimal quantum magnet: 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

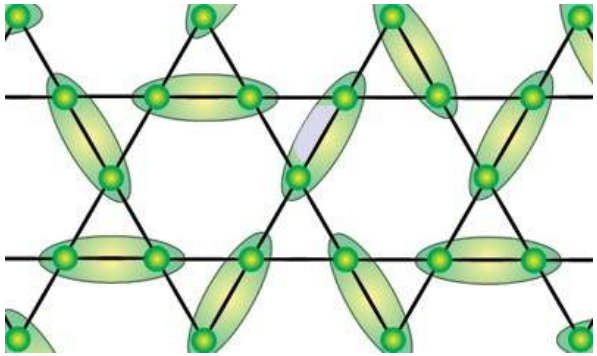
$$|GS\rangle = \frac{1}{\sqrt{2}}[|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle] \qquad \langle \vec{S}_i \rangle = 0$$

The state is maximally entangled

Quantum magnets are macroscopic versions of these entangled states

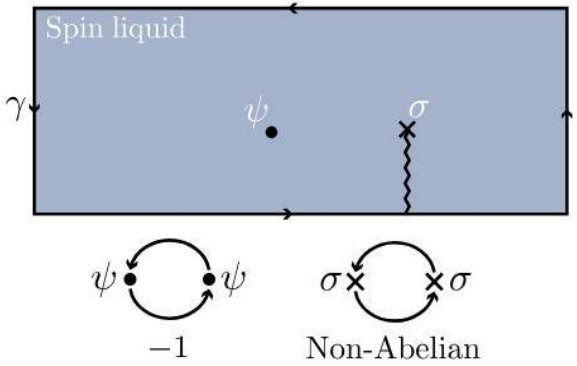
Quantum spin liquid materials: opportunity and challenge

Macroscopically entangled magnetic state



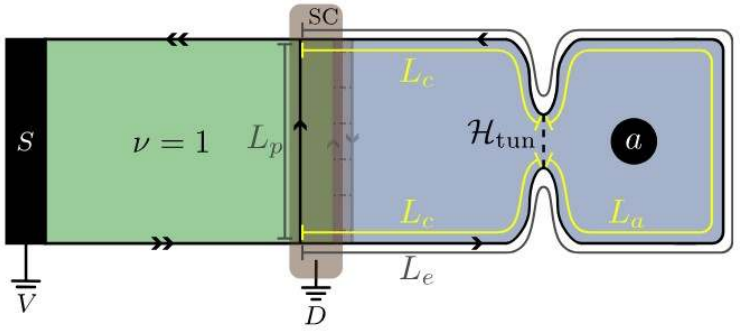
Rep. Prog. Phys. 80, 016502 (2017)

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, $RuCl_3$)

However, quantum spin liquids are exceptionally challenging to identify experimentally

How can we learn the microscopic description of quantum magnets?

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 H —*Many-body solver*→ **Compute an observable** $\langle\Psi|O|\Psi\rangle$

The problem we often find experimentally

Measure an observable $\langle\Psi|O|\Psi\rangle$ → **Determine its Hamiltonian** H

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

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)

Two strategies for Hamiltonian learning

Uniquely defined problems

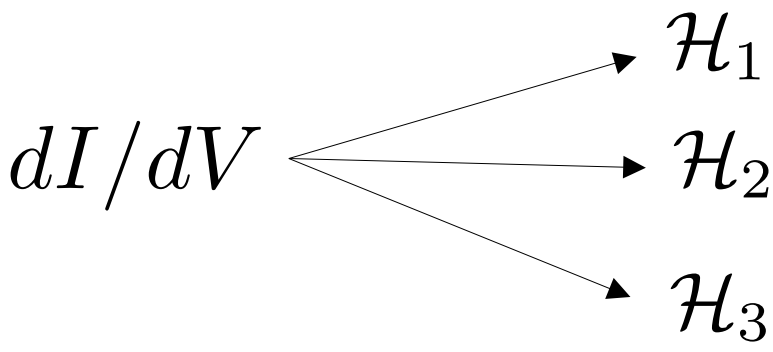
$$dI/dV \longrightarrow \mathcal{H}$$

A set of experiments is consistent with a single Hamiltonian

Supervised learning

Nano Letters 25, 36, 13435-13440 (2025)
arXiv:2601.19371 (2026)

Multiply-defined problems



A set of experiments constrain the Hamiltonian, but it is non-unique

Generative learning

Phys. Rev. Applied 20, 044081 (2023)
to appear (2026)

Supervised 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

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

Hamiltonian

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

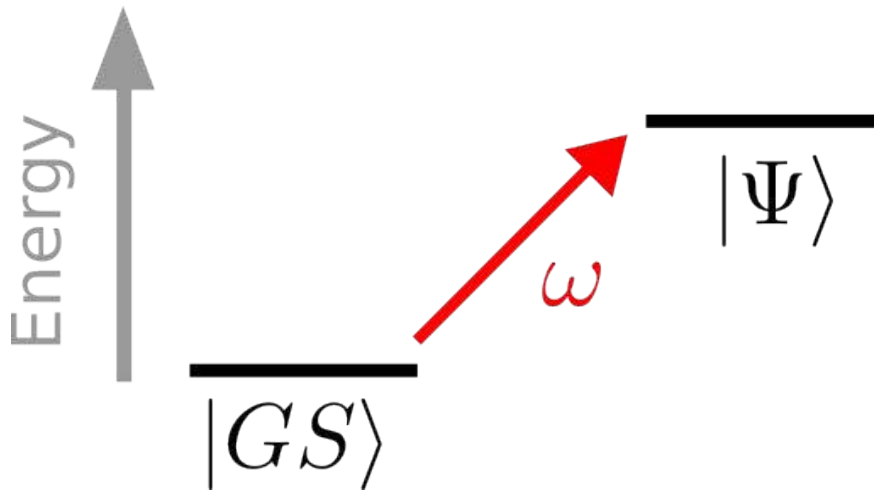
$\langle O \rangle_{\text{exp}} \longrightarrow$ Neural network $\longrightarrow \lambda_i^{\text{exp}}$

Hamiltonian

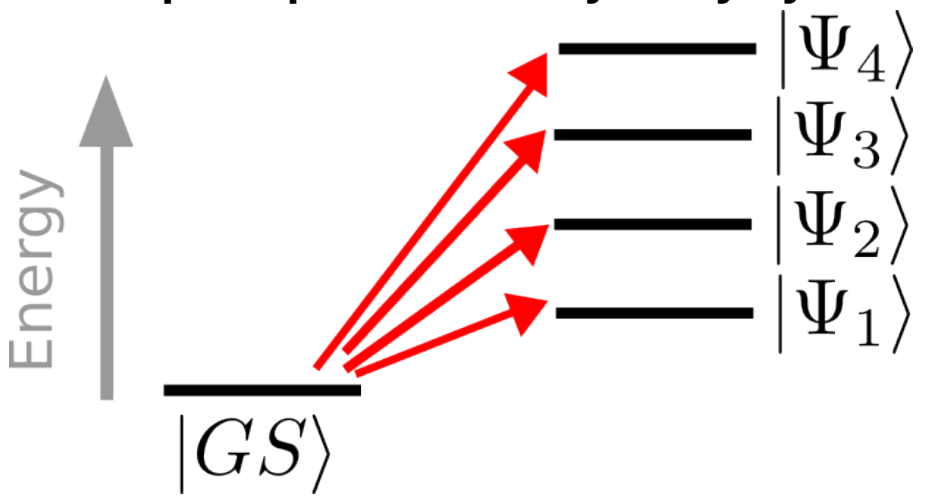
This strategy combines quantum many-body solvers, machine learning, and experimental measurements

Spin excitations in collective quantum magnets

Minimal quantum spin system



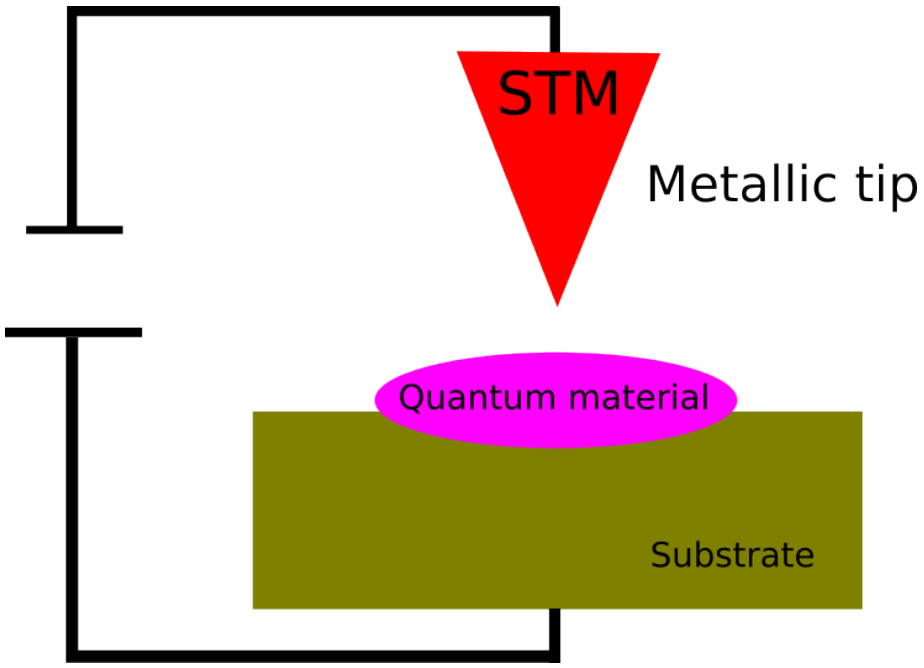
Complex quantum many-body system



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

That can be measured with scanning tunneling microscopy

Spectroscopy atomic-scale quantum matter with STM

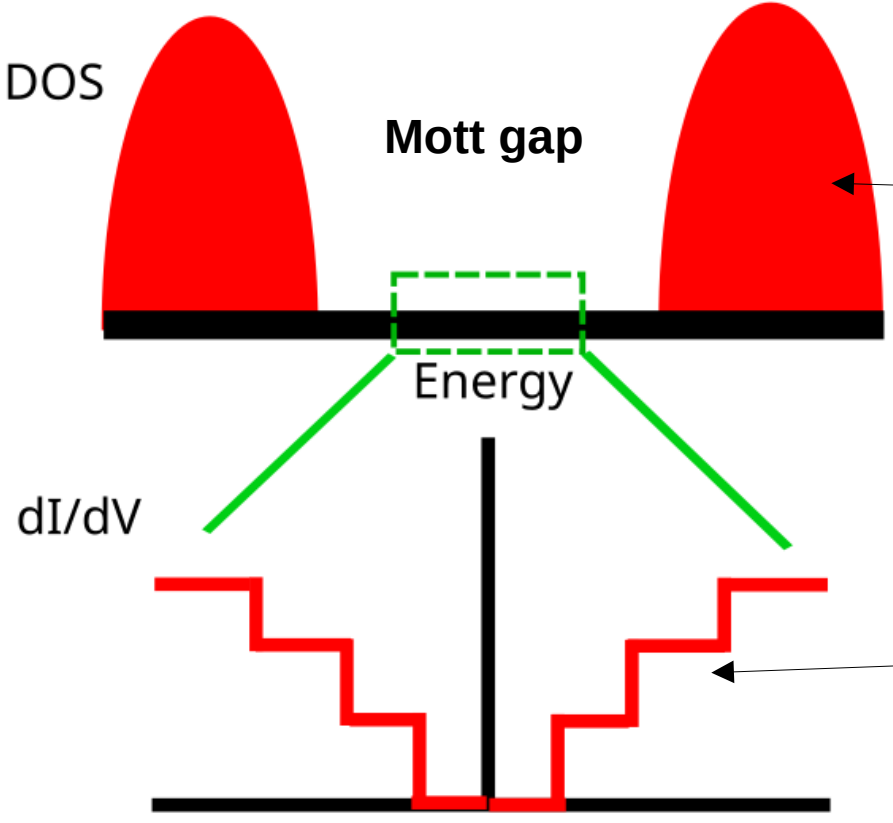


Atomic real-space resolution
Energy/frequency and time resolution

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

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

Single particle VS many-body excitations in STM



Single particle tunneling

$$dI/dV \sim DOS(\omega, \mathbf{r})$$

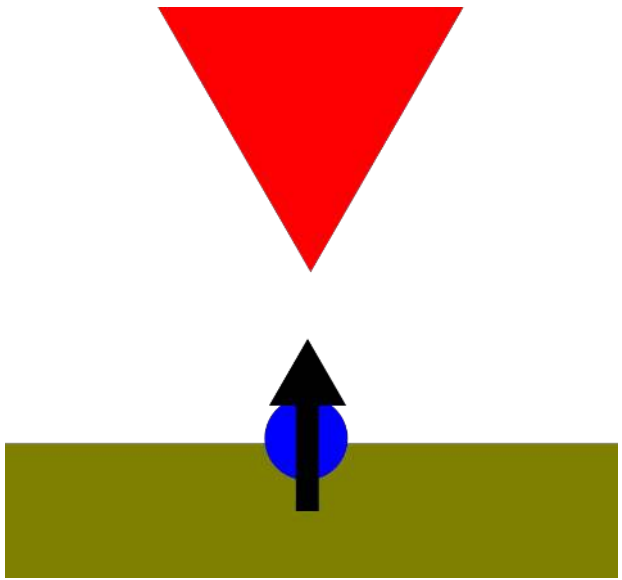
Many-body inelastic tunneling

$$d^2I/dV^2 \sim A(\omega, \mathbf{r})$$

Many-body spectral function

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

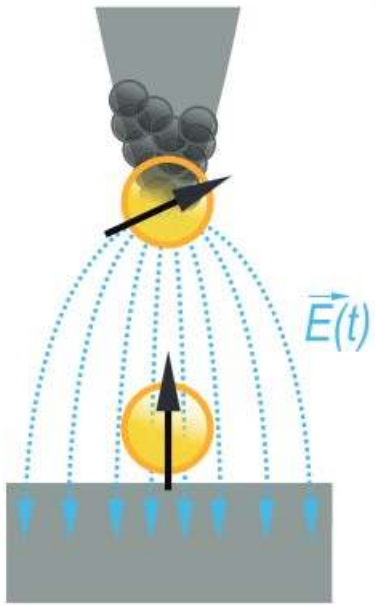
Inelastic electron tunneling spectroscopy (IETS)



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

Generating the training data: Solving the quantum many-body problem

Let us take 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?

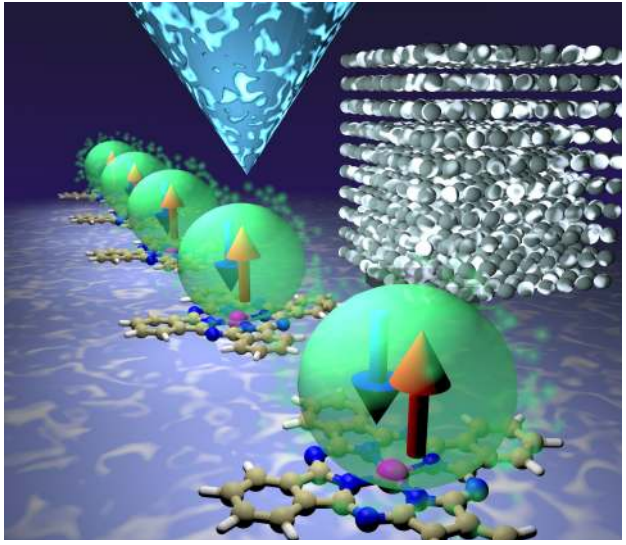
And ultimately compute the dynamical spin correlator

Dealing with very large spaces with tensor networks

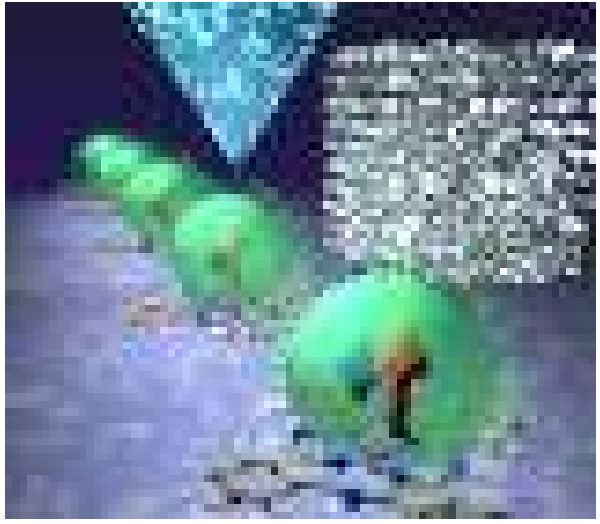
A many-body wavefunction a is a very high dimensional object (2^L coefficients)

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

Tensor-networks allow “compressing” exponentially large information with linear resources



“True wavefunction”



“Tensor-network wavefunction”

Dealing with very large spaces with tensor networks

Typical many-body wavefunction $$|\Psi\rangle = \sum c_{s_1,s_2,\ldots,s_L} |s_1, s_2, \ldots s_L\rangle$$

Tensor networks allow parametrizing many-body wavefunctions as

$$|\Psi\rangle = \sum_{\{s\}} M_1^{(s_1)} M_2^{(s_2)} \cdots M_N^{(s_N)} |s_1 s_2 \ldots s_N\rangle$$

$L\chi^2$ parameters

Full wavefunction  *Matrix product state*

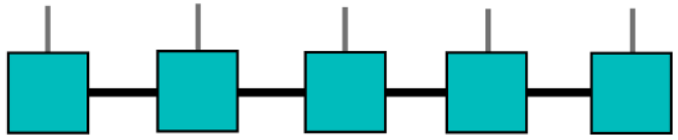
Tensor networks allow to drastically reduce the memory required to store a many-body state

Annals of Physics 326, 96 (2011)

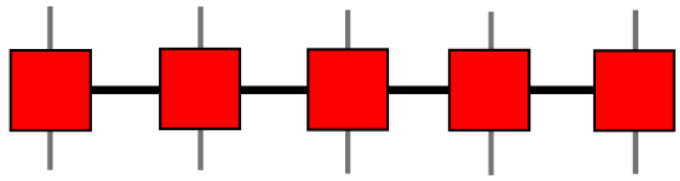
Exponentially large algebra with tensor networks

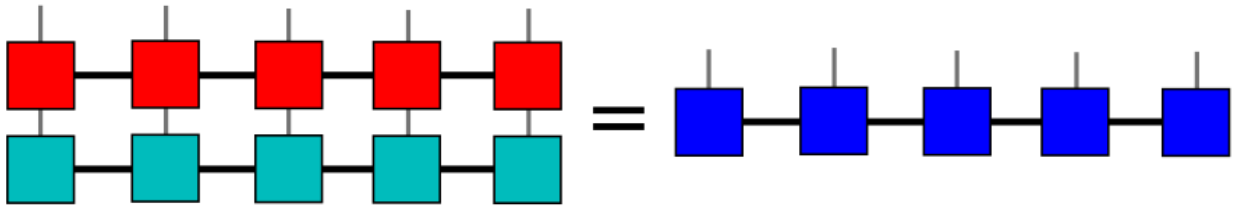
Tensor network allow to (approximately) operate in exponentially large vector spaces

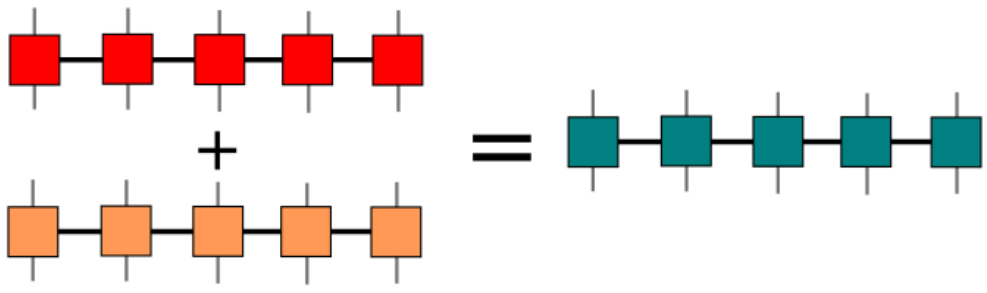
vector

$|\Psi\rangle \equiv$ 

matrix

$\hat{A} \equiv$ 

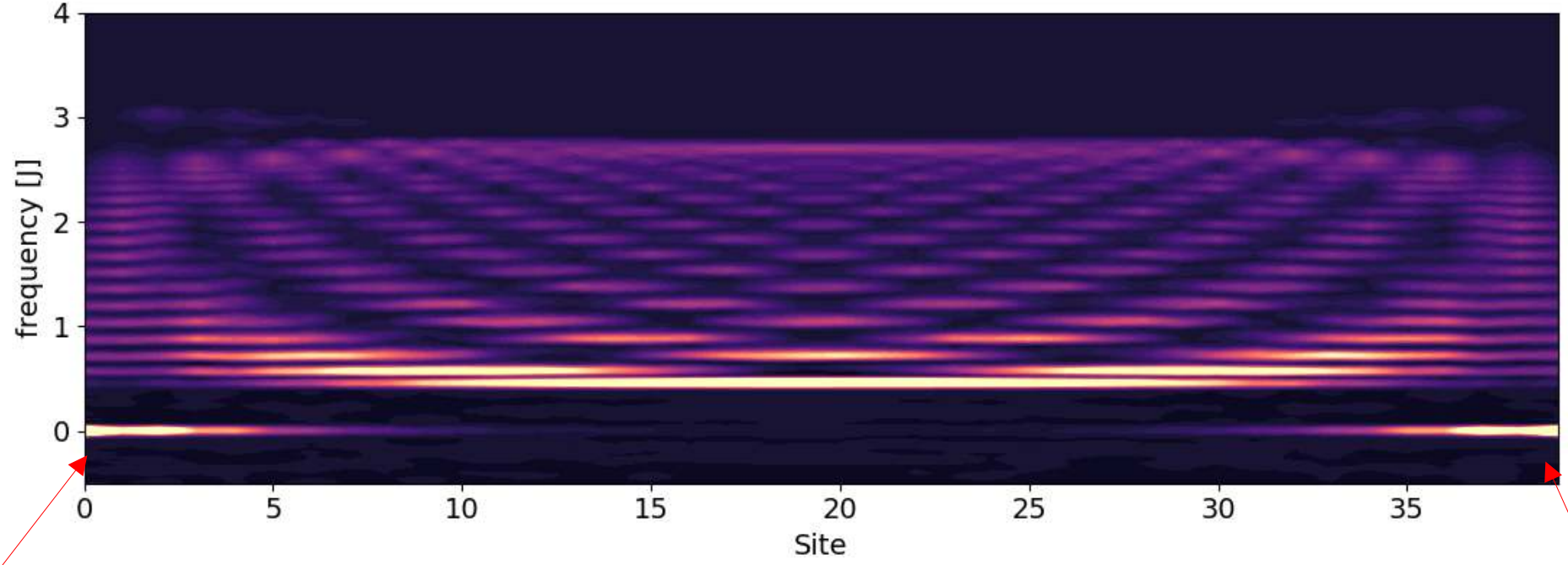
$|\Psi_2\rangle = \hat{A}|\Psi_1\rangle \equiv$ 

$\hat{A}_3 = \hat{A}_1 + \hat{A}_2 \equiv$ 

This allows us to compute observables in generic quantum many-body problems

Dynamical spectral functions in a 1D topological quantum magnet

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



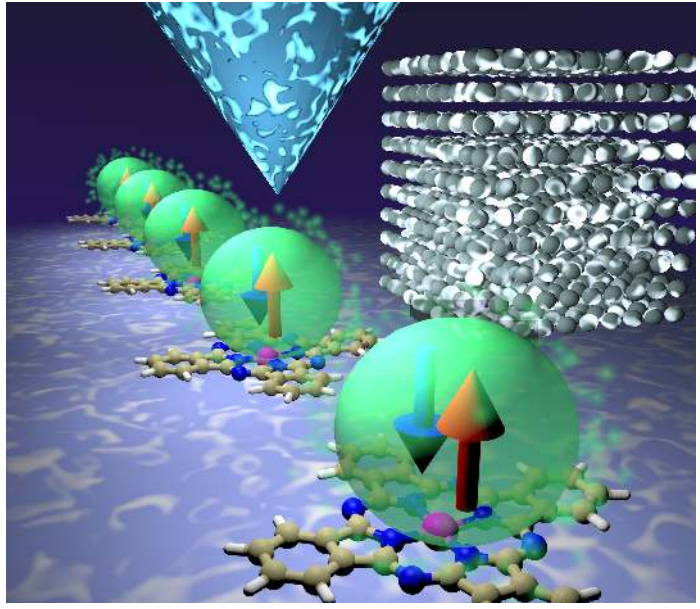
Edge mode

Open source software for spin models with tensor networks
https://github.com/joselado/dmrgpy

Edge mode

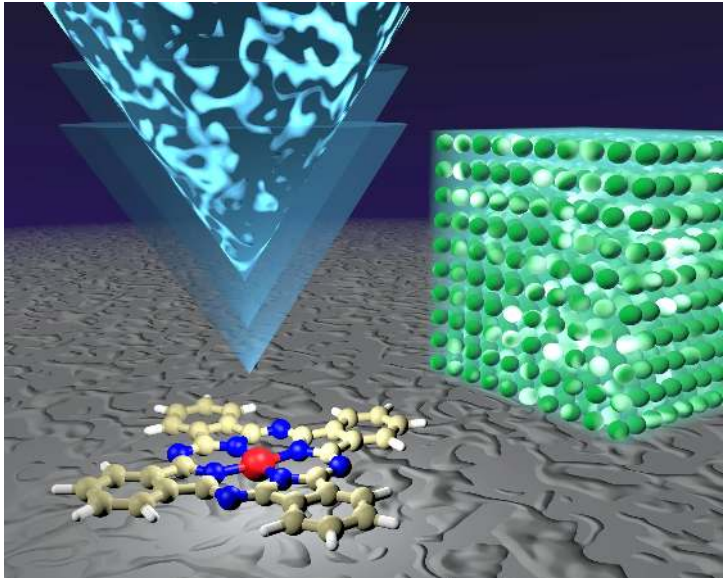
Hamiltonian learning correlated system form inelastic STM spectroscopy

Hamiltonian learning from spin excitation



Phys. Rev. Lett. 131, 086701 (2023)
Nano Letters 25, 36, 13435-13440 (2025)

Hamiltonian learning from setpoint dependent spectroscopy



arXiv:2601.19371 (2026)

Hamiltonian learning correlated system form inelastic STM spectroscopy

Rouven Koch

Greta Lupi

Mohammad Amini

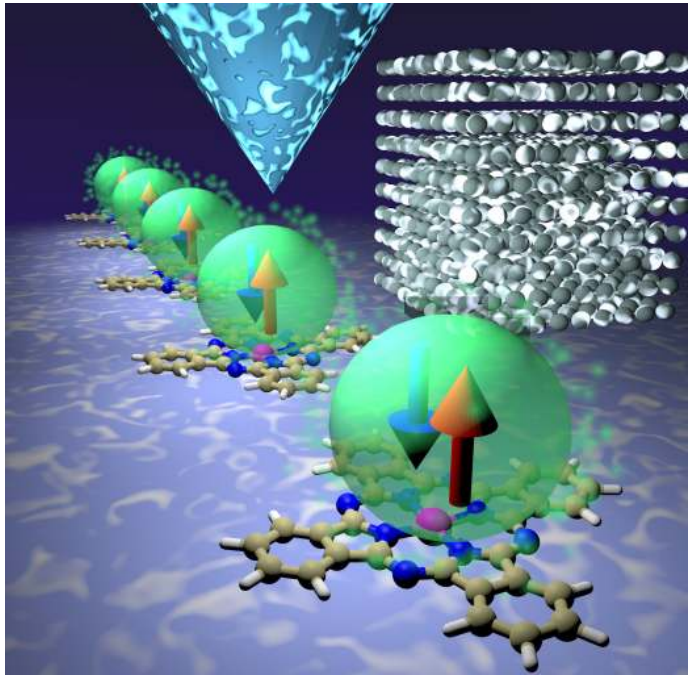
Robert Drost

Shawulienu Kezilebieke

Adolfo Fumega

Peter Liljeroth

Hamiltonian learning triplons in a molecular quantum magnet



Rouven Koch

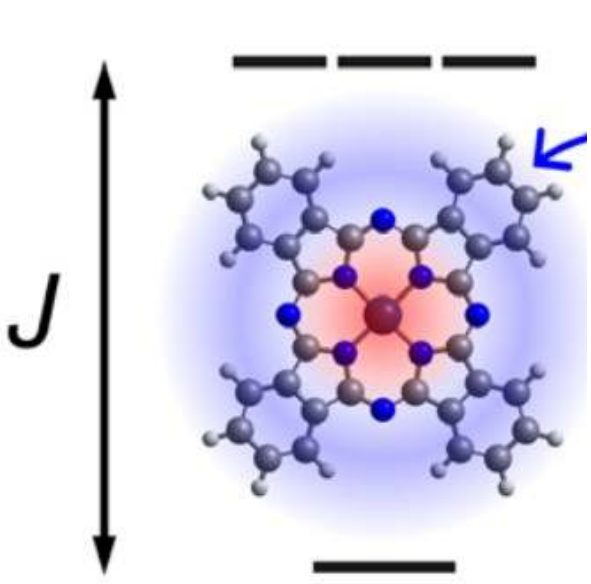
Robert Drost

Peter Liljeroth

Shawulienu Kezilebieke

Phys. Rev. Lett. 131, 086701 (2023)
Nano Letters 25, 36, 13435-13440 (2025)

Triplet excitations in a molecule



$S_T = 1$

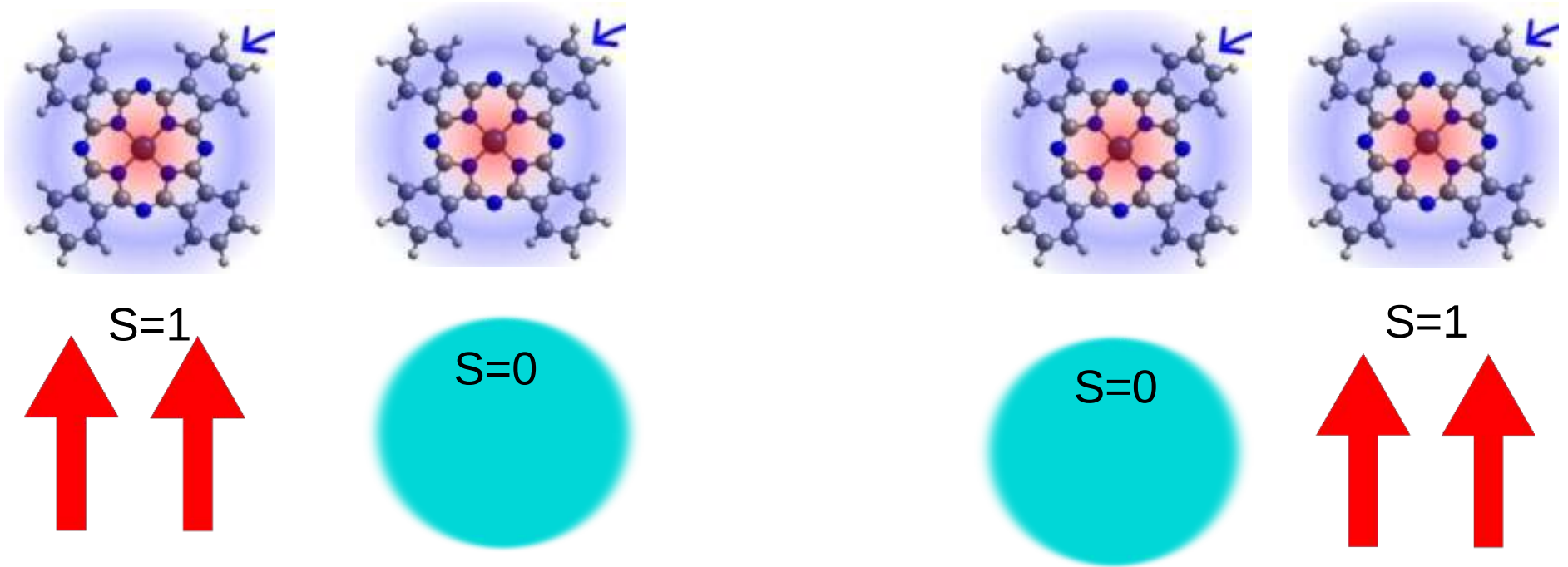
$S_T = 0$

Internal antiferromagnetic coupling

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

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

Triplons in a dimer

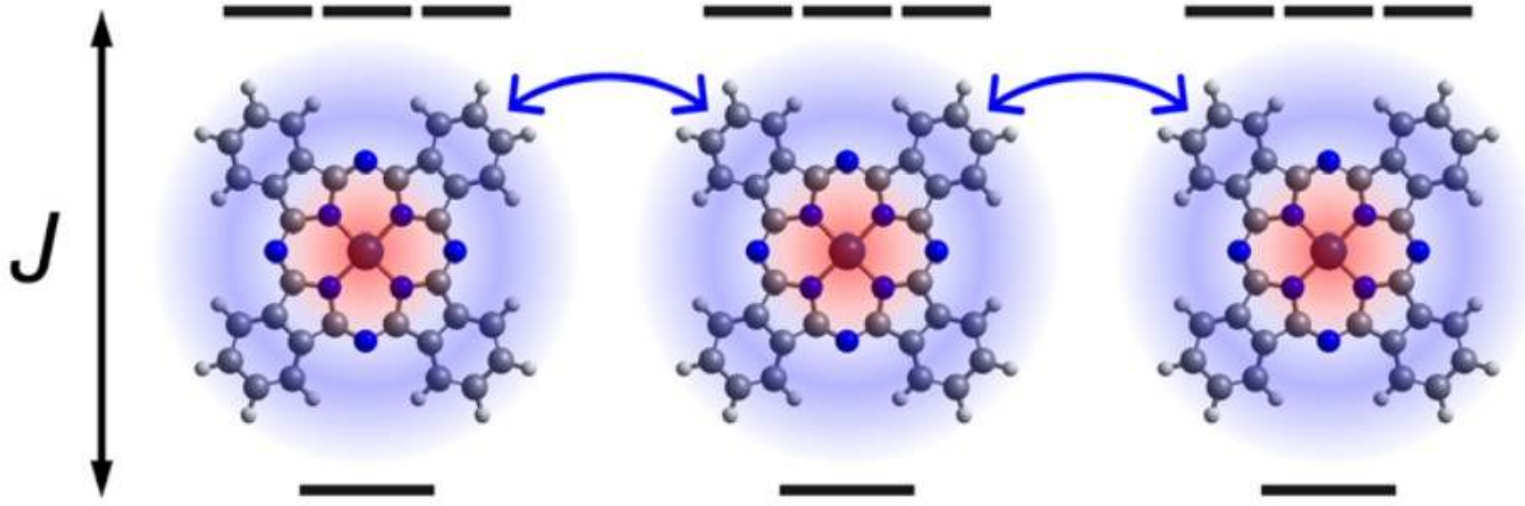


A triplet in the first molecule

Can jump to the second molecule

Exchange coupling between molecules leads to propagating triplon excitations

A chain of triplons



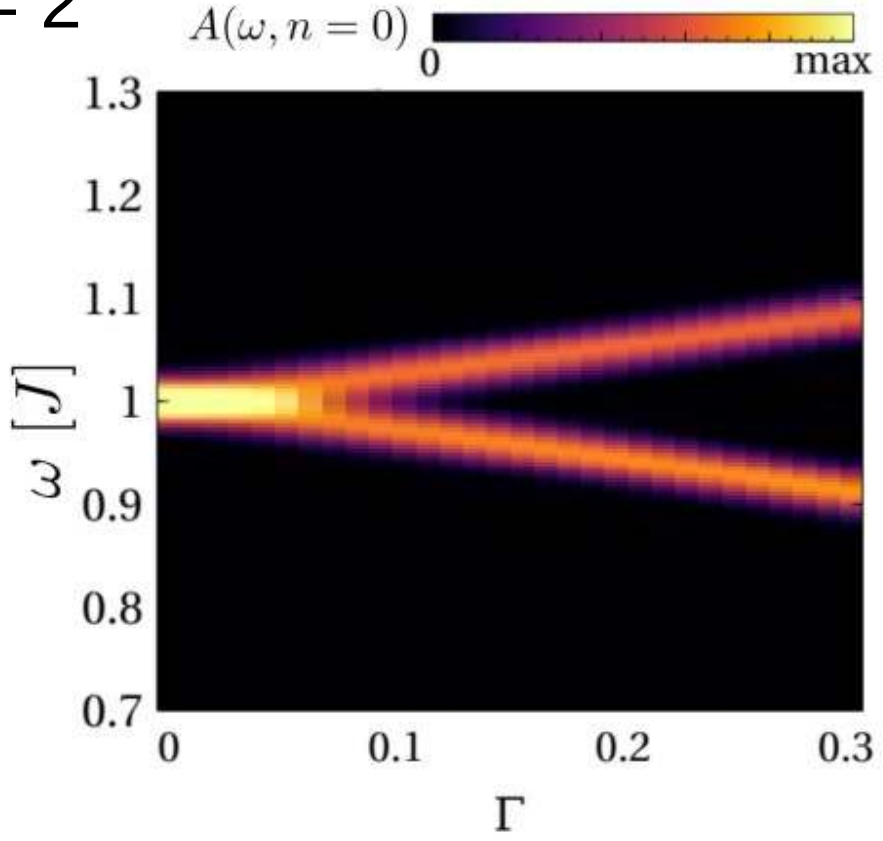
$S_T = 1$

$S_T = 0$

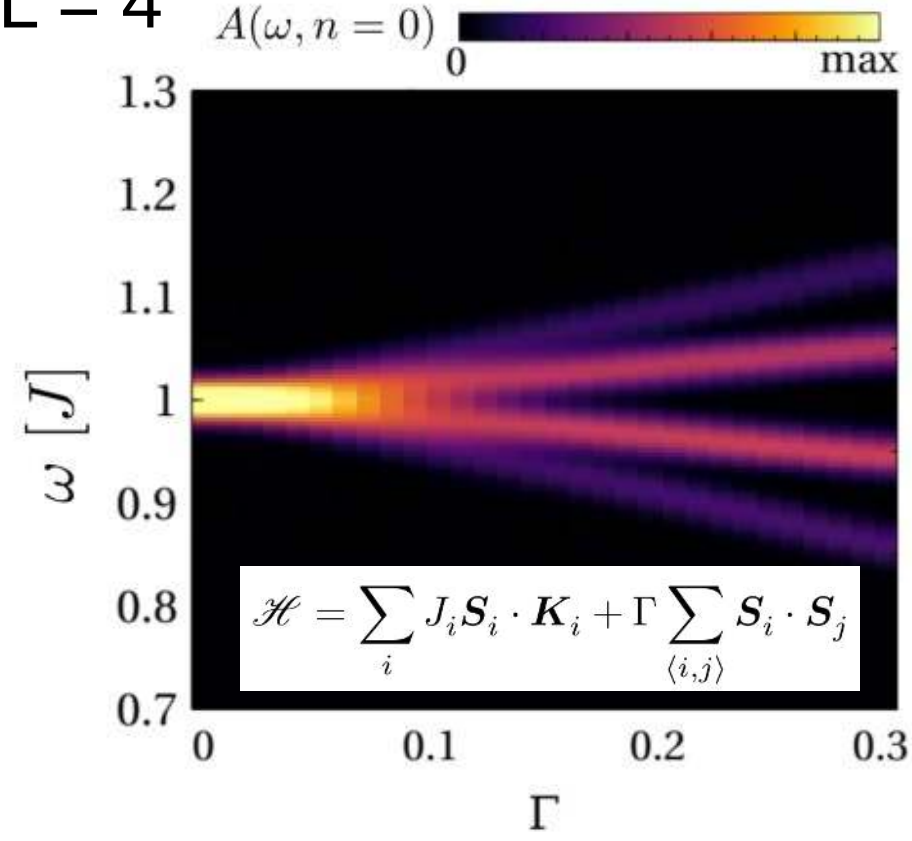
$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Chains of triplons

L = 2

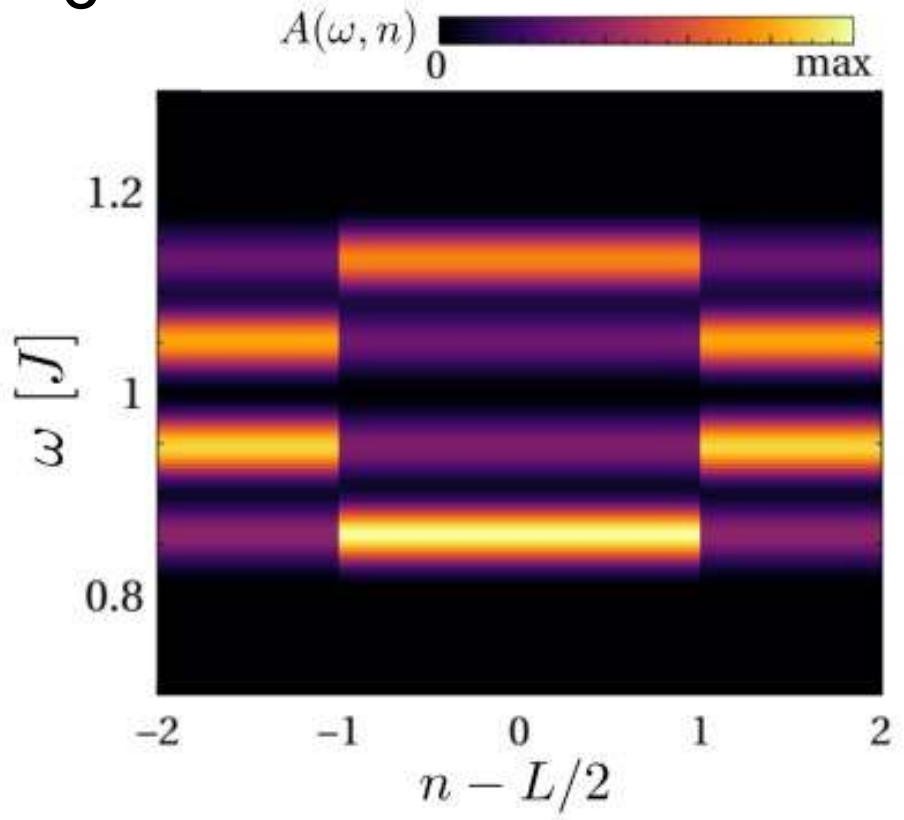


L = 4

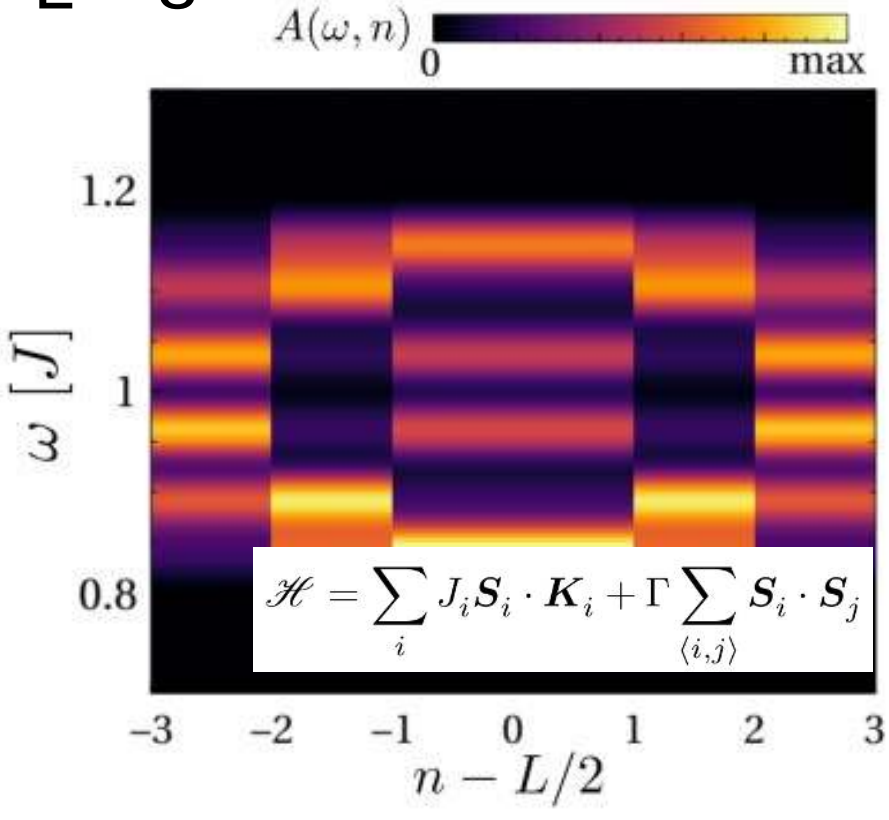


Chains of triplons

L = 6

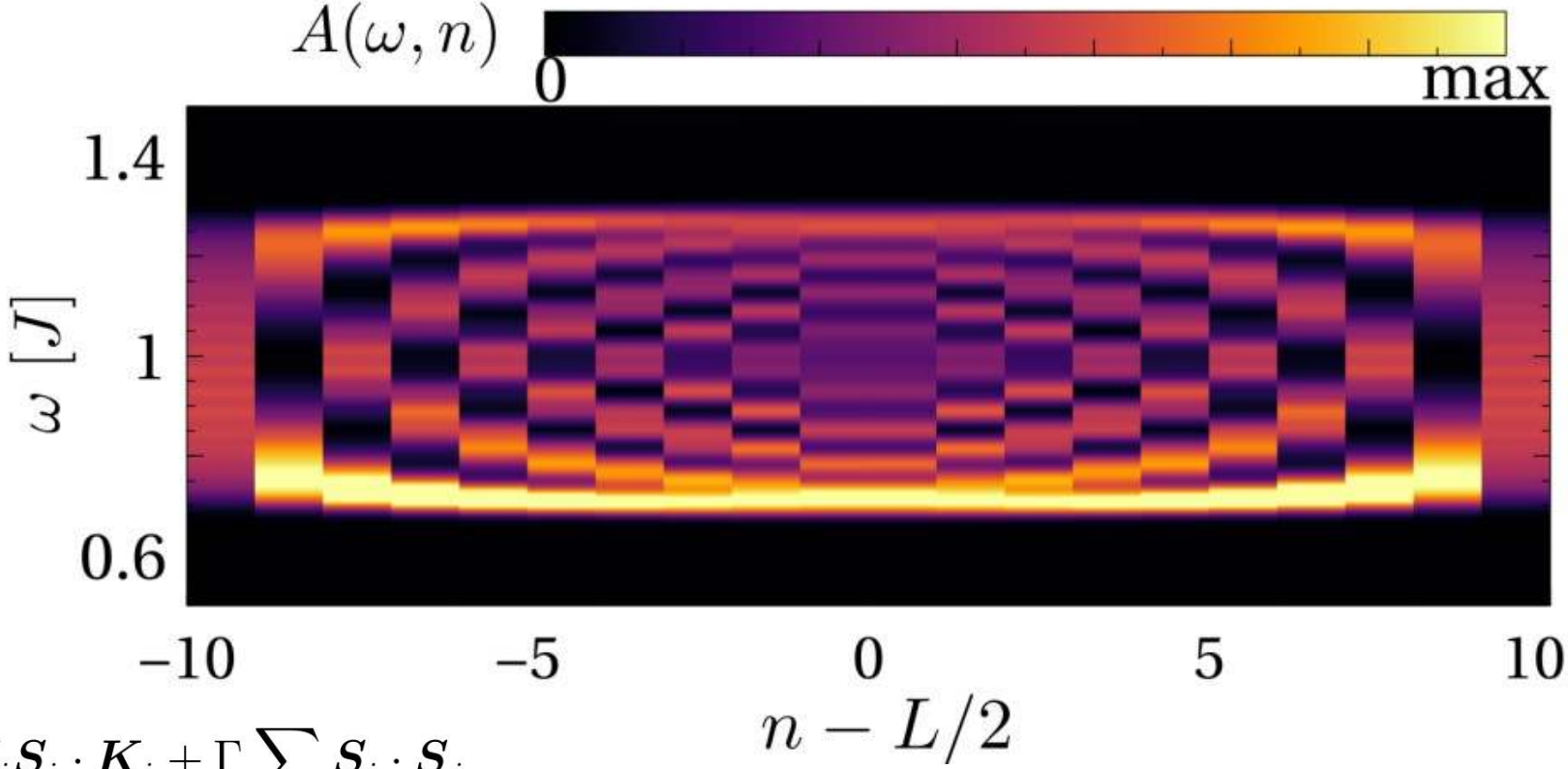


L = 8



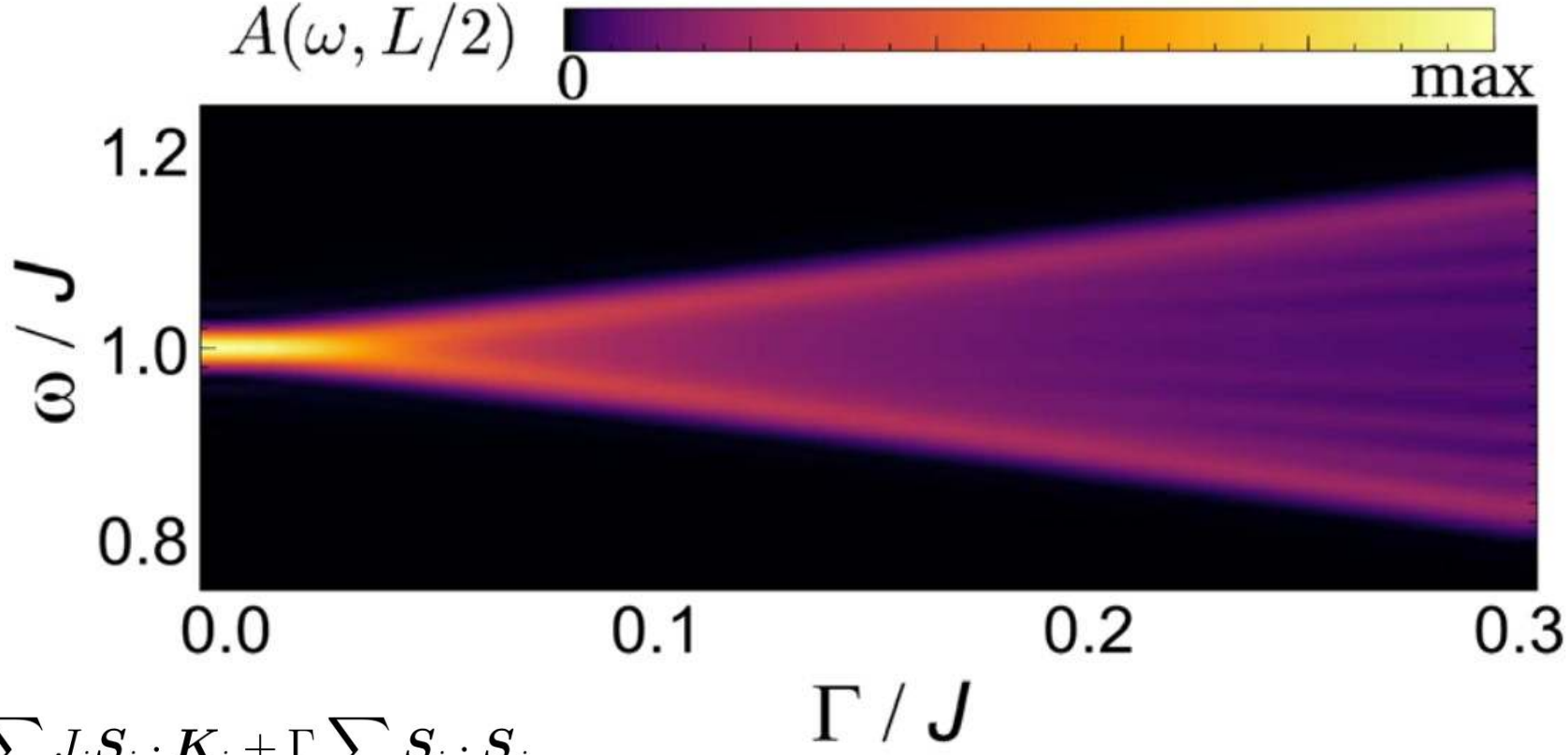
Chains of triplons

L = 20



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

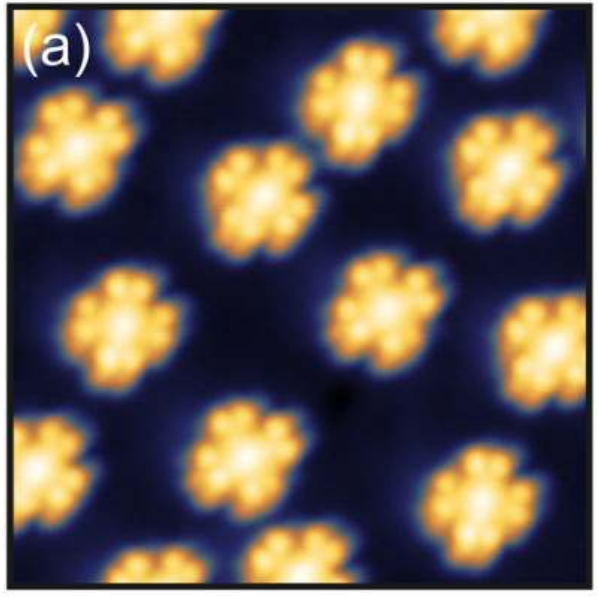
Chains of triplons

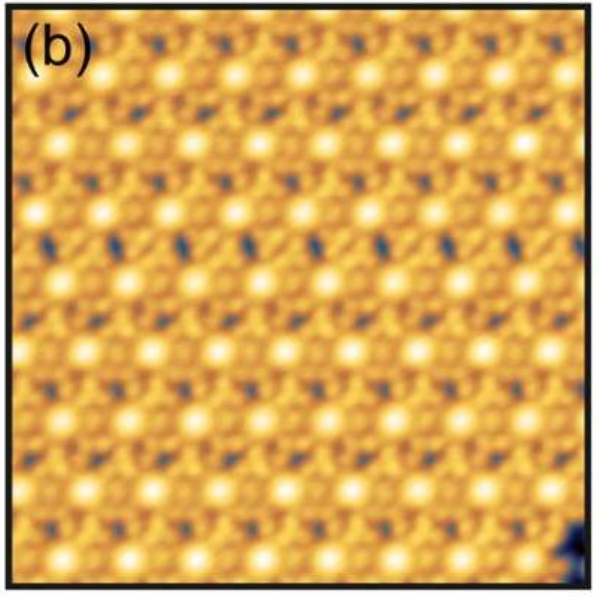


$$\mathscr{H} = \sum_i J_i \boldsymbol{S}_i \cdot \boldsymbol{K}_i + \Gamma \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

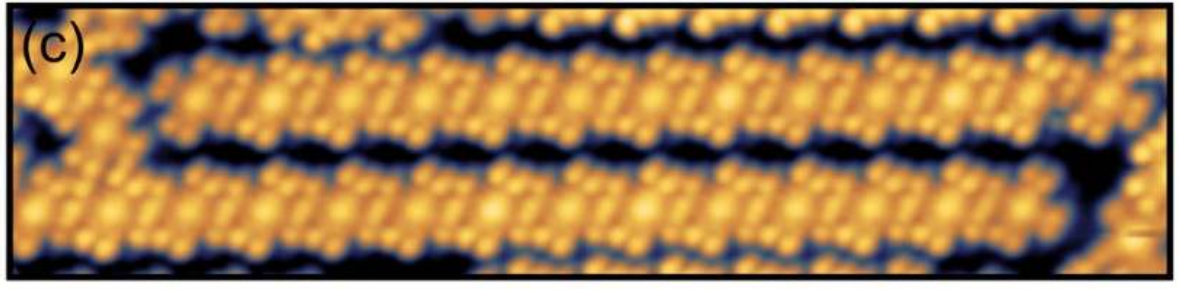
0D, 1D and 2D arrays

0D



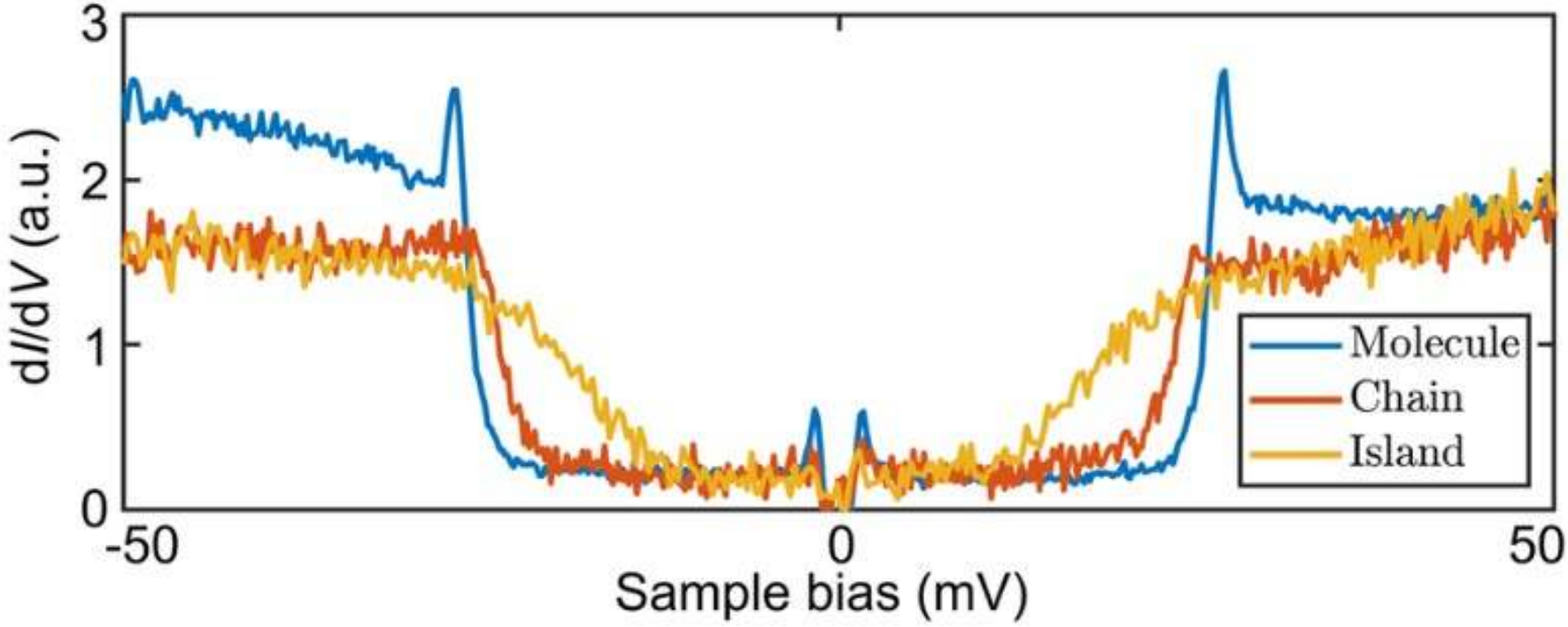


2D



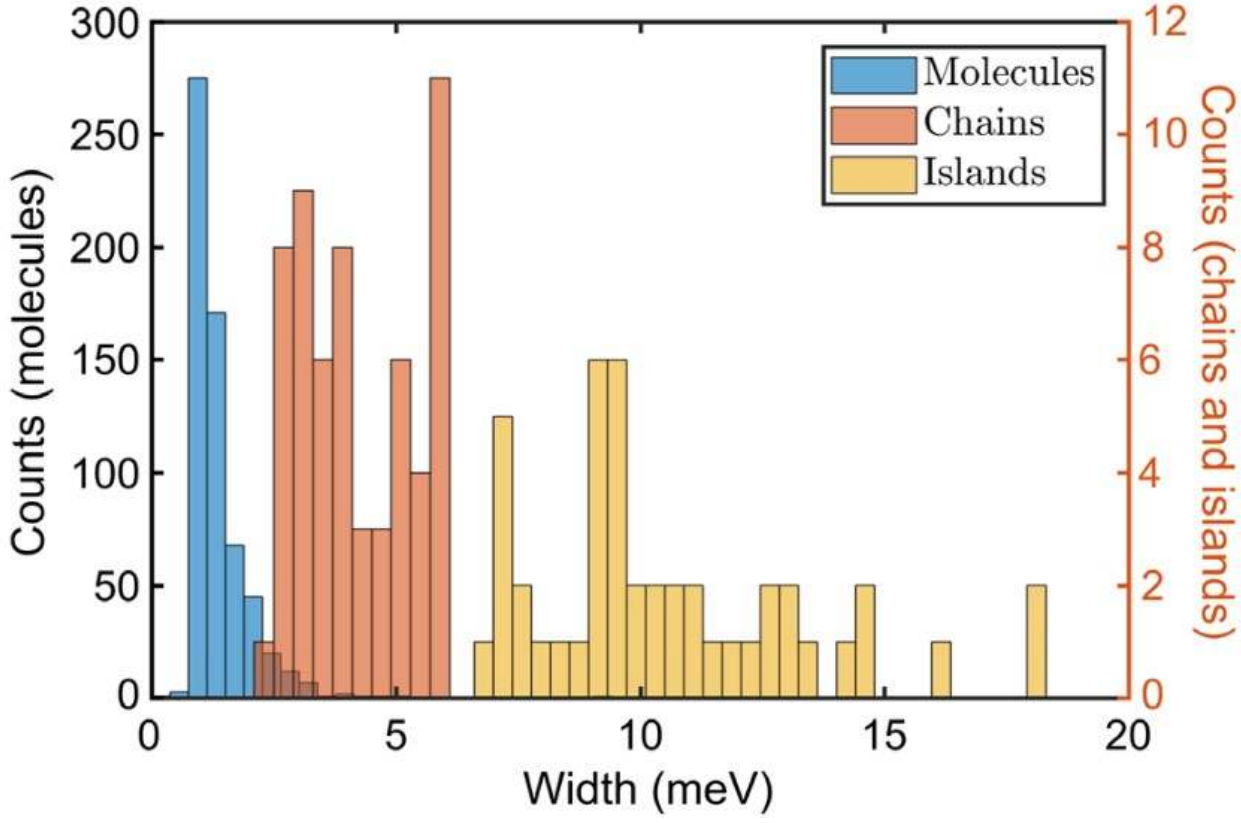
1D

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



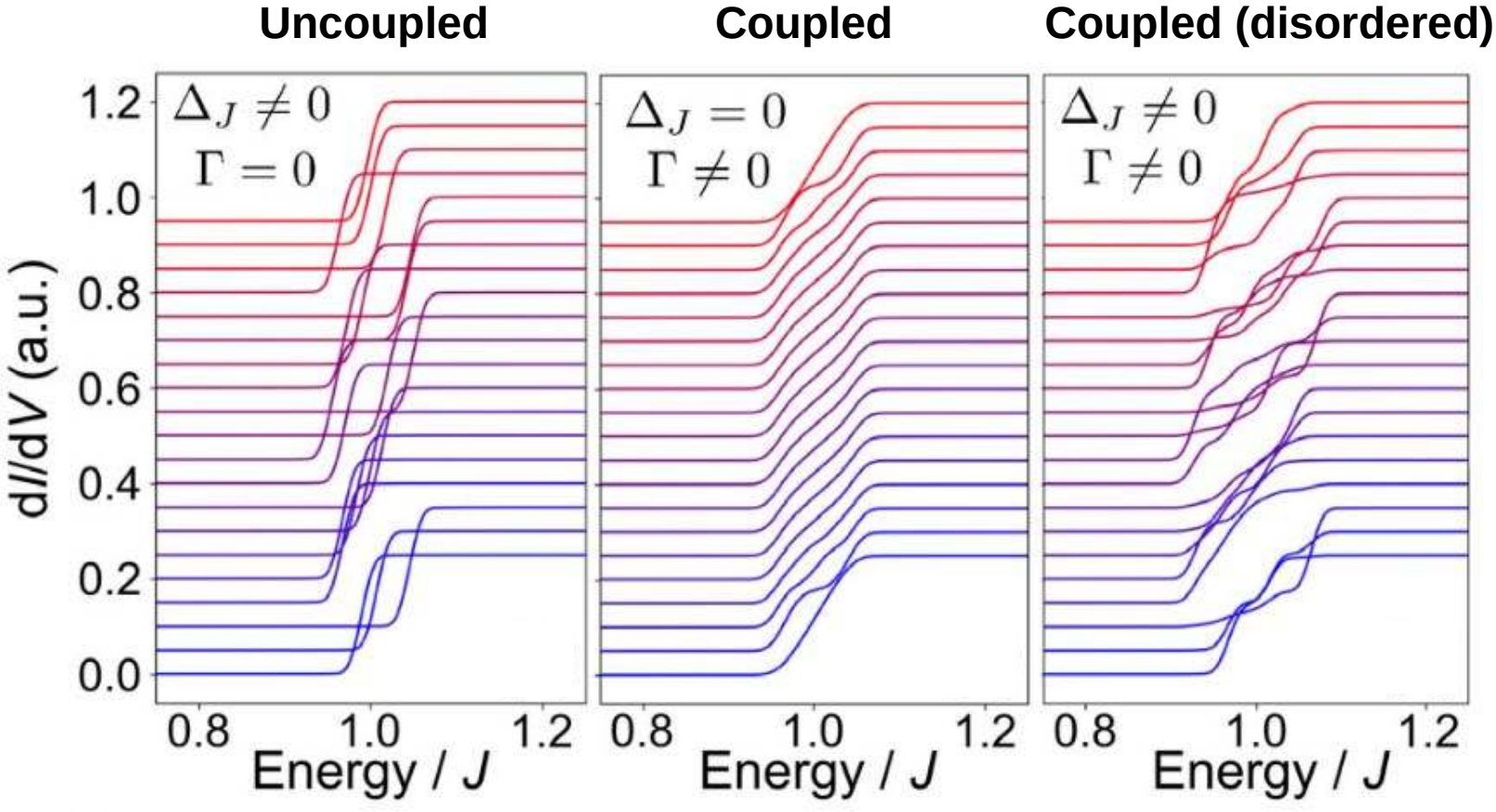
The width of the step is bigger for islands than for molecules

Statistic of the inelastic steps

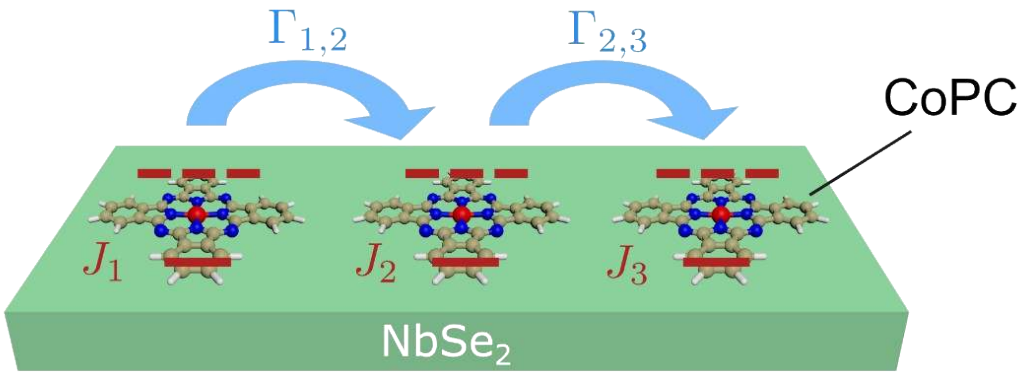


2D arrays have the largest width of the spin excitation

Coupled and uncoupled inelastic excitation



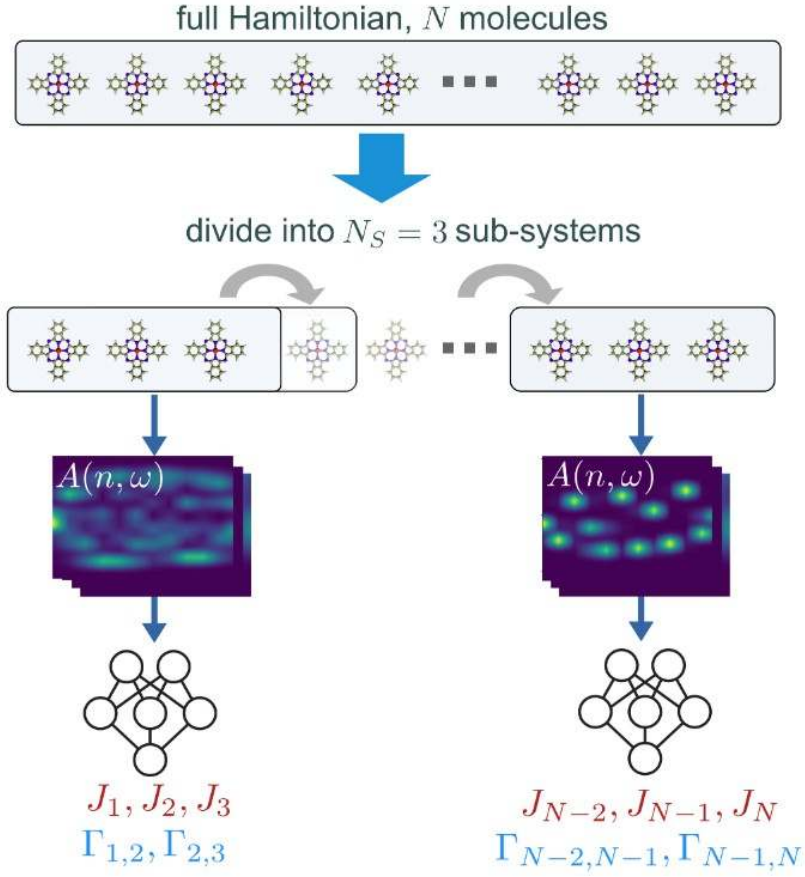
A strategy for machine learning quantum magnets



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

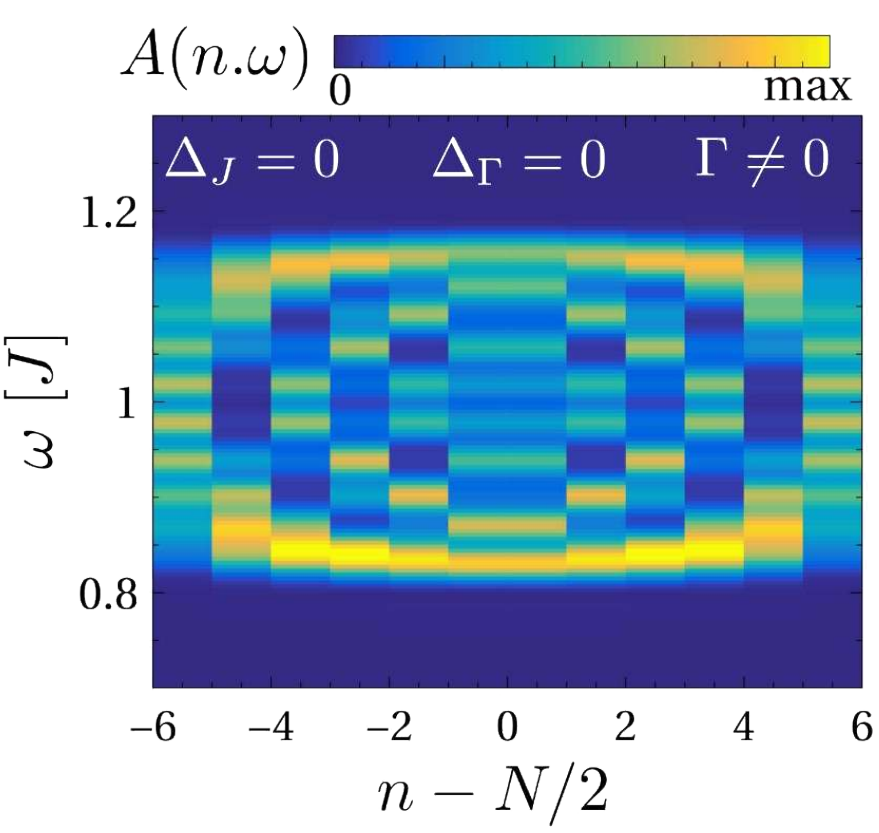
J_n : intramolecular exchange

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

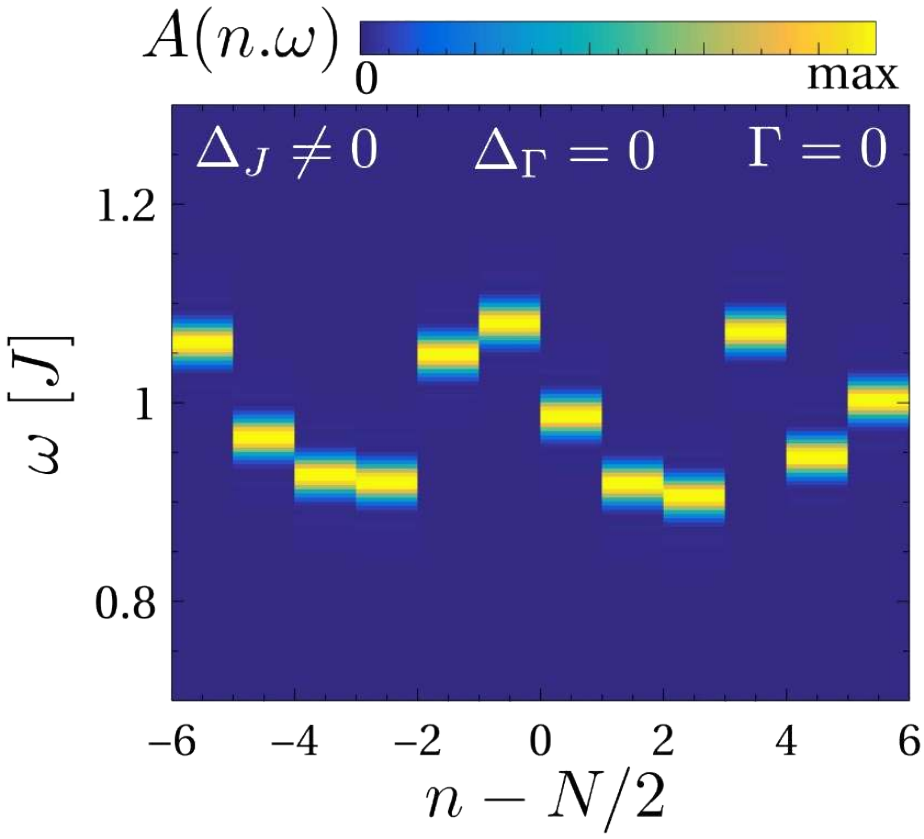


Pristine triplon excitations

Pristine and coupled molecules

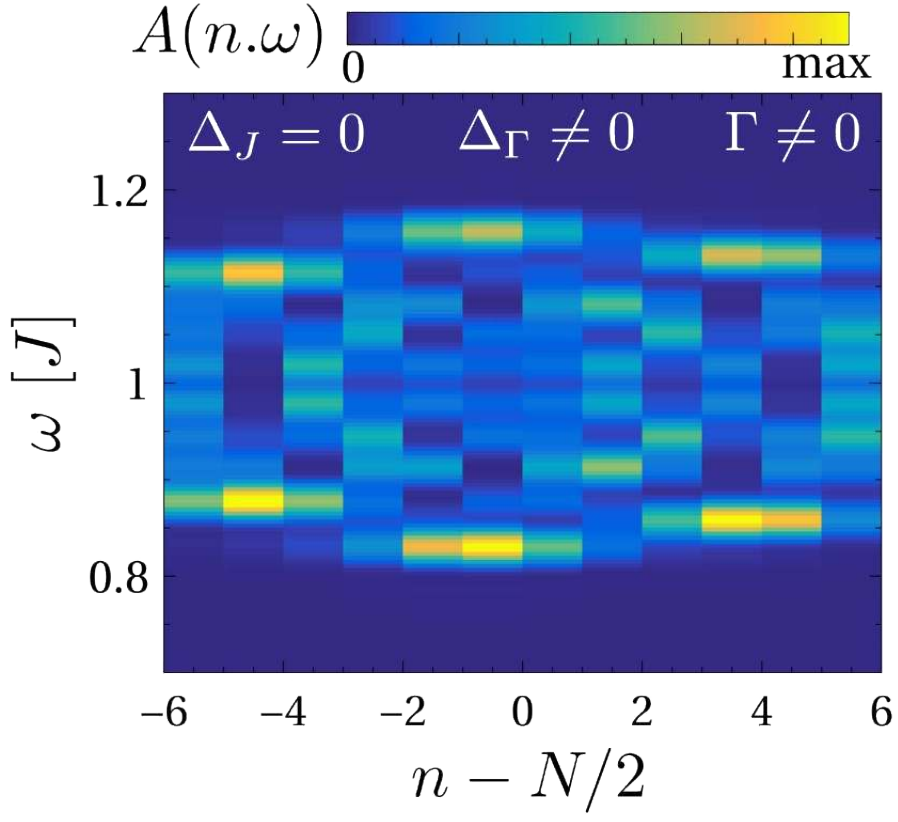


Decoupled and disordered molecules

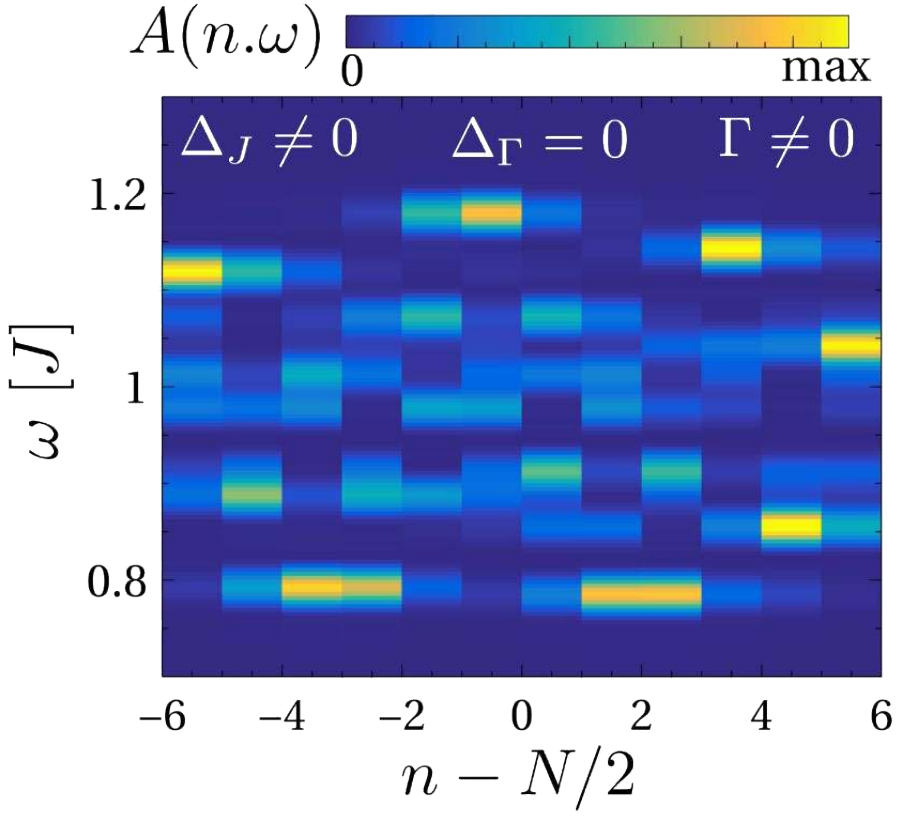


The different impact of molecular disorder

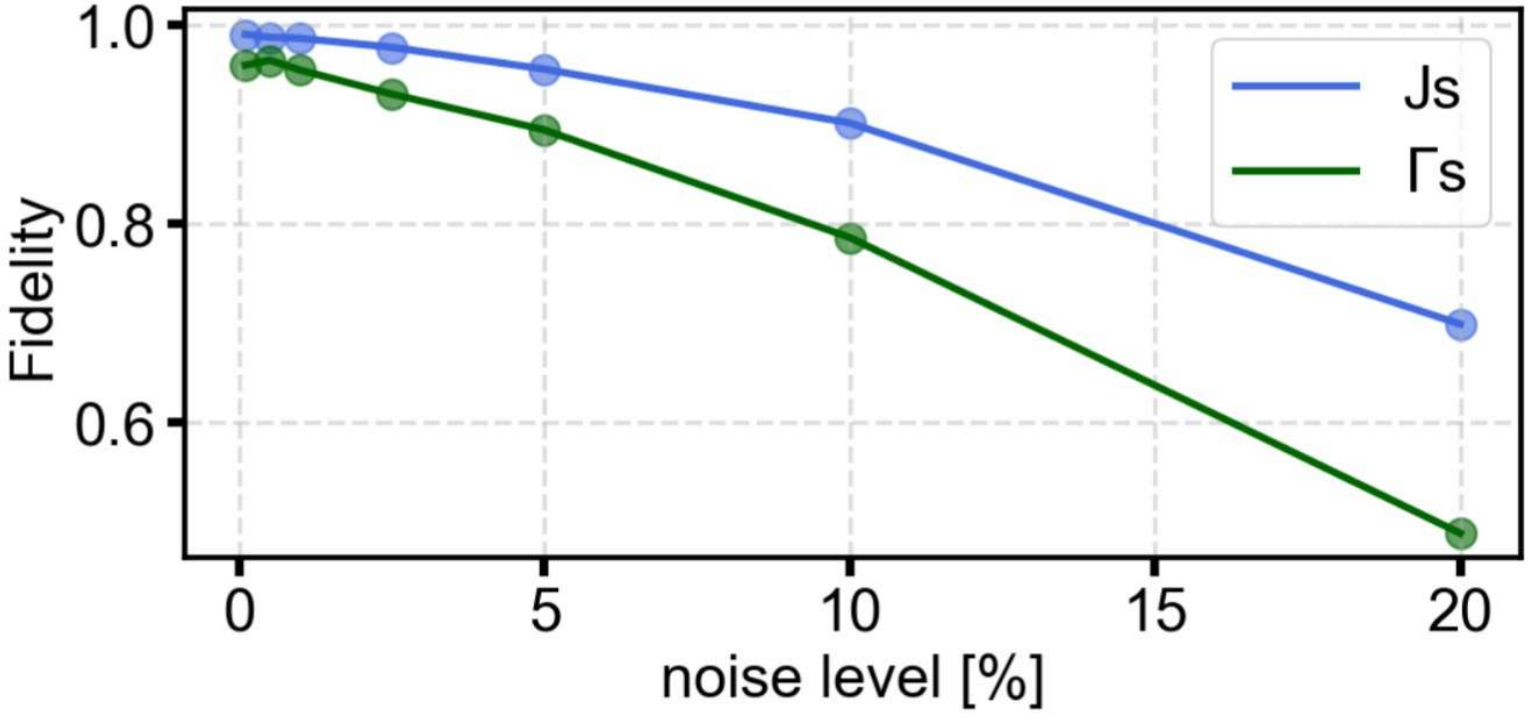
Disorder in inter-molecule coupling



Disorder in intra-molecule coupling

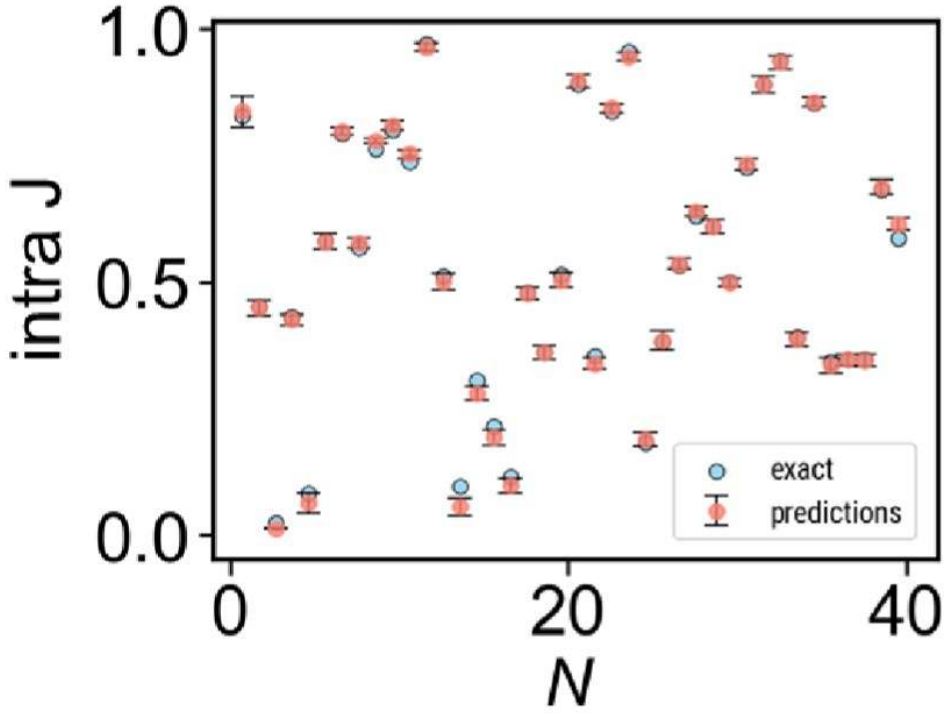


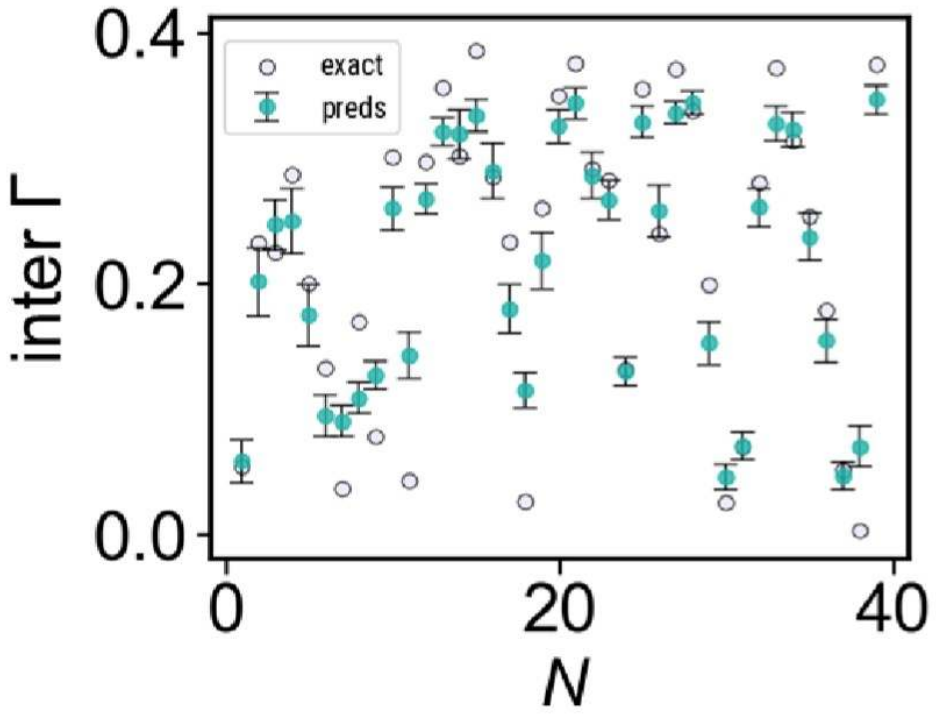
Resilience to noise of Hamiltonian learning



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

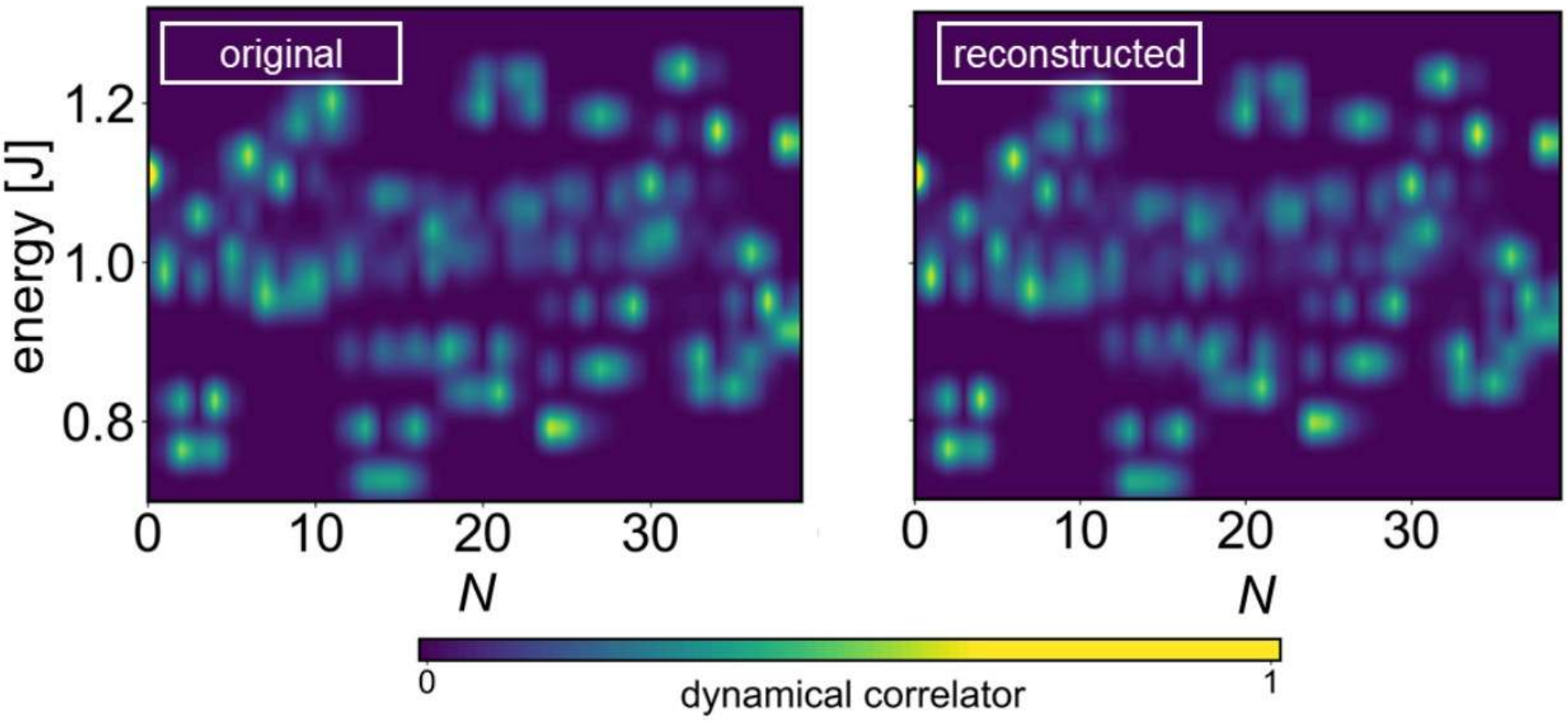
Inferring spatially dependent exchange couplings



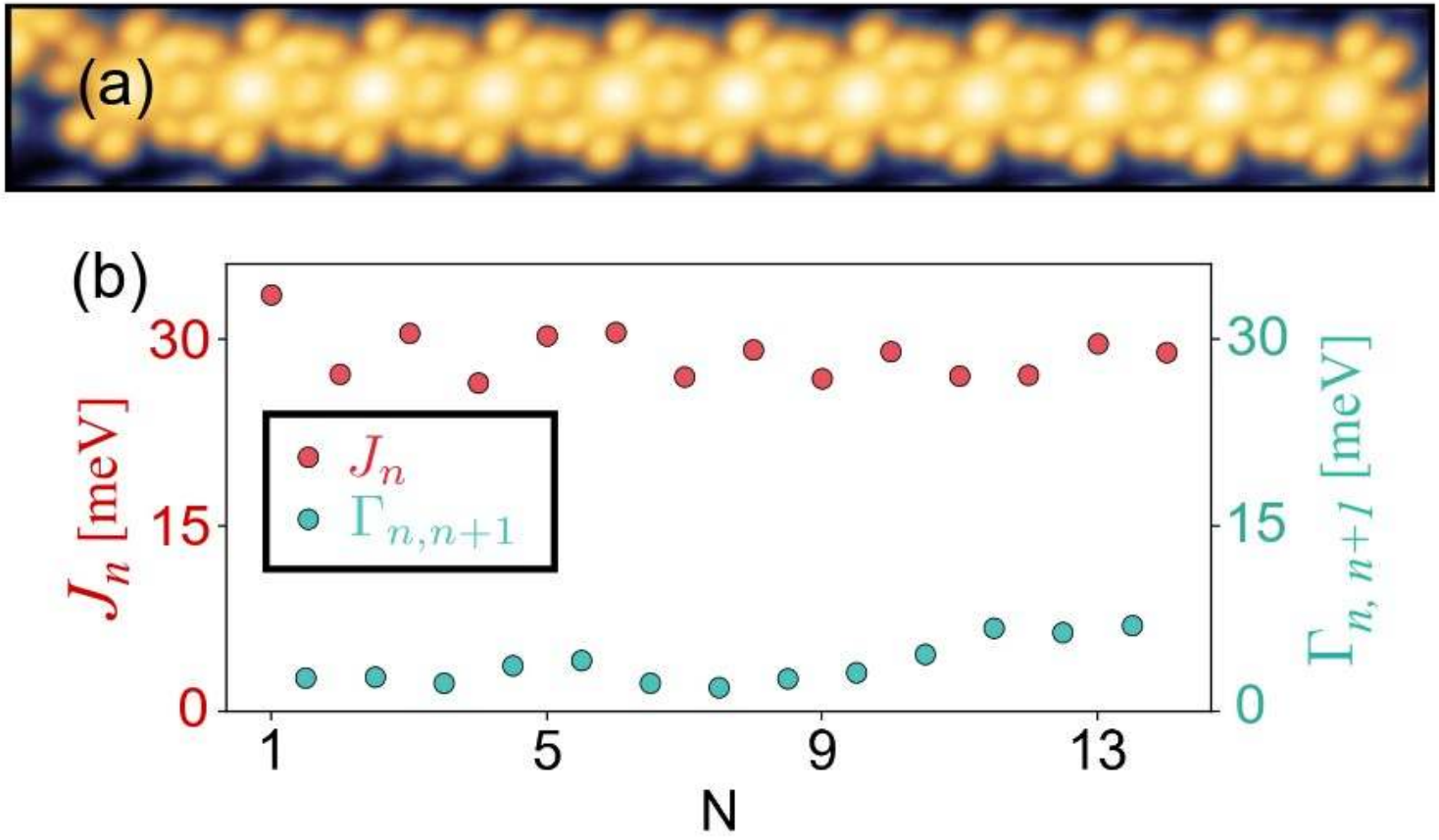


Intramolecular J_n exchange variations can be predicted robustly
Intermolecular Γ_n exchange variations are more subtle to predict

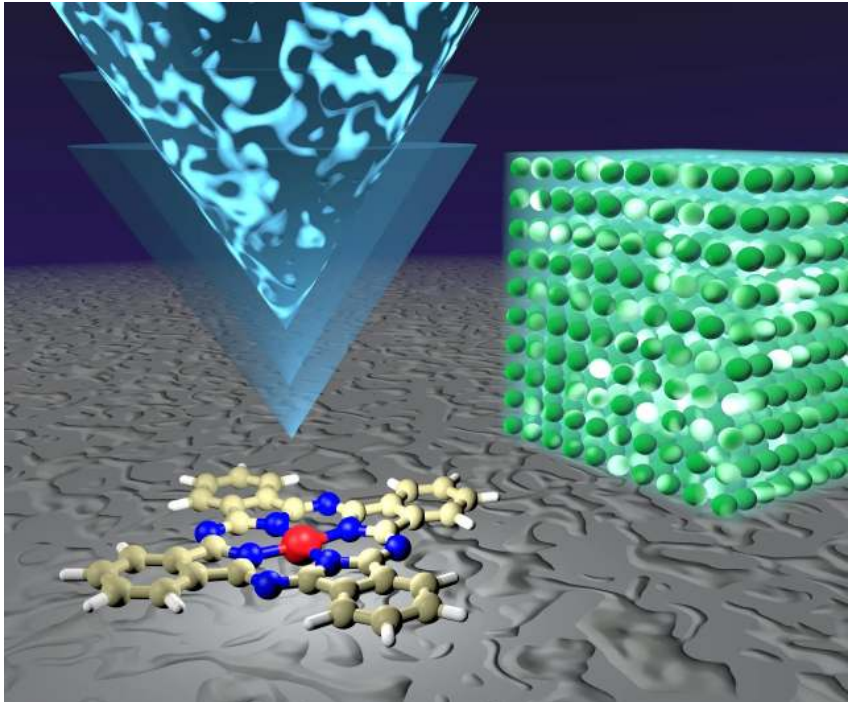
Reconstructing a quantum magnet Hamiltonian



Machine learning Hamiltonian of the experimental triplon chain



Hamiltonian learning from setpoint dependent spectroscopy



arXiv:2601.19371 (2026)

Greta Lupi

Mohammad Amini

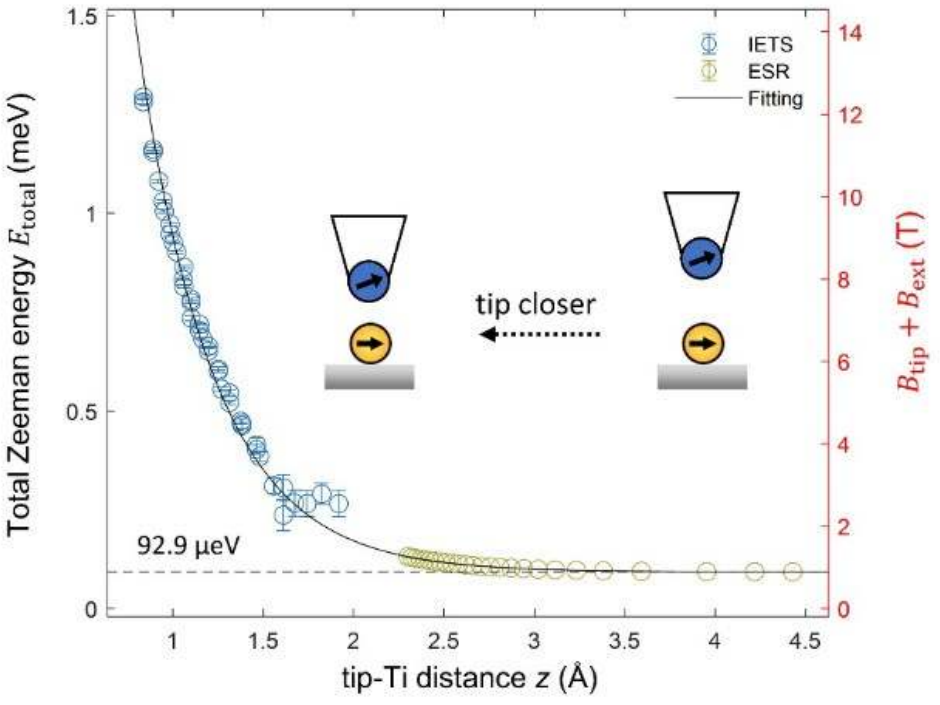
Robert Drost

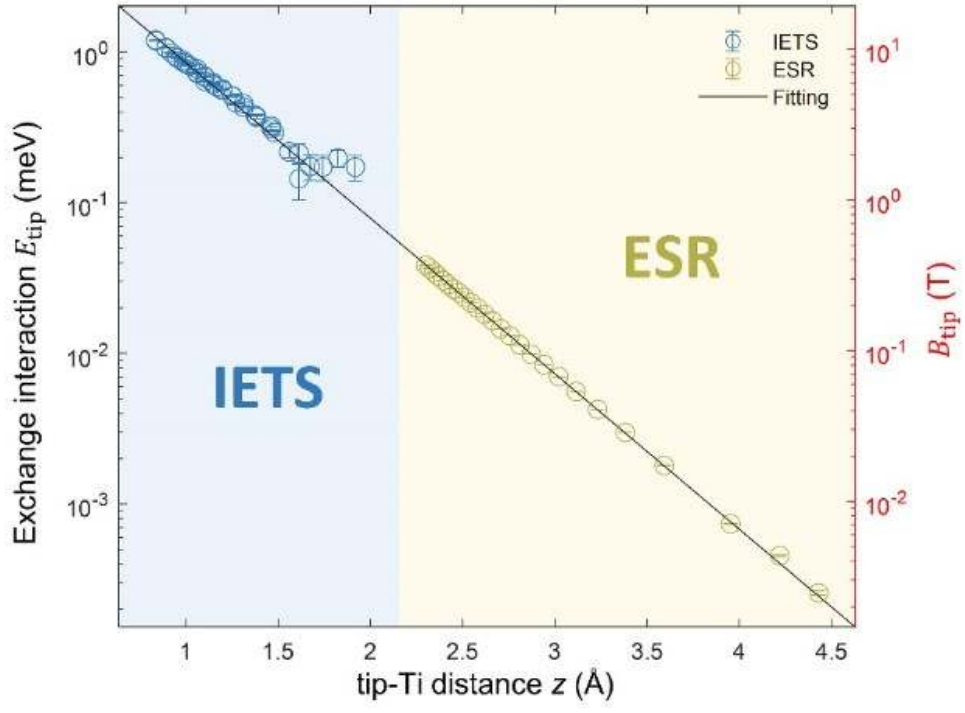
Adolfo Fumega

Peter Liljeroth

Many-body excitation energies can be distance dependent

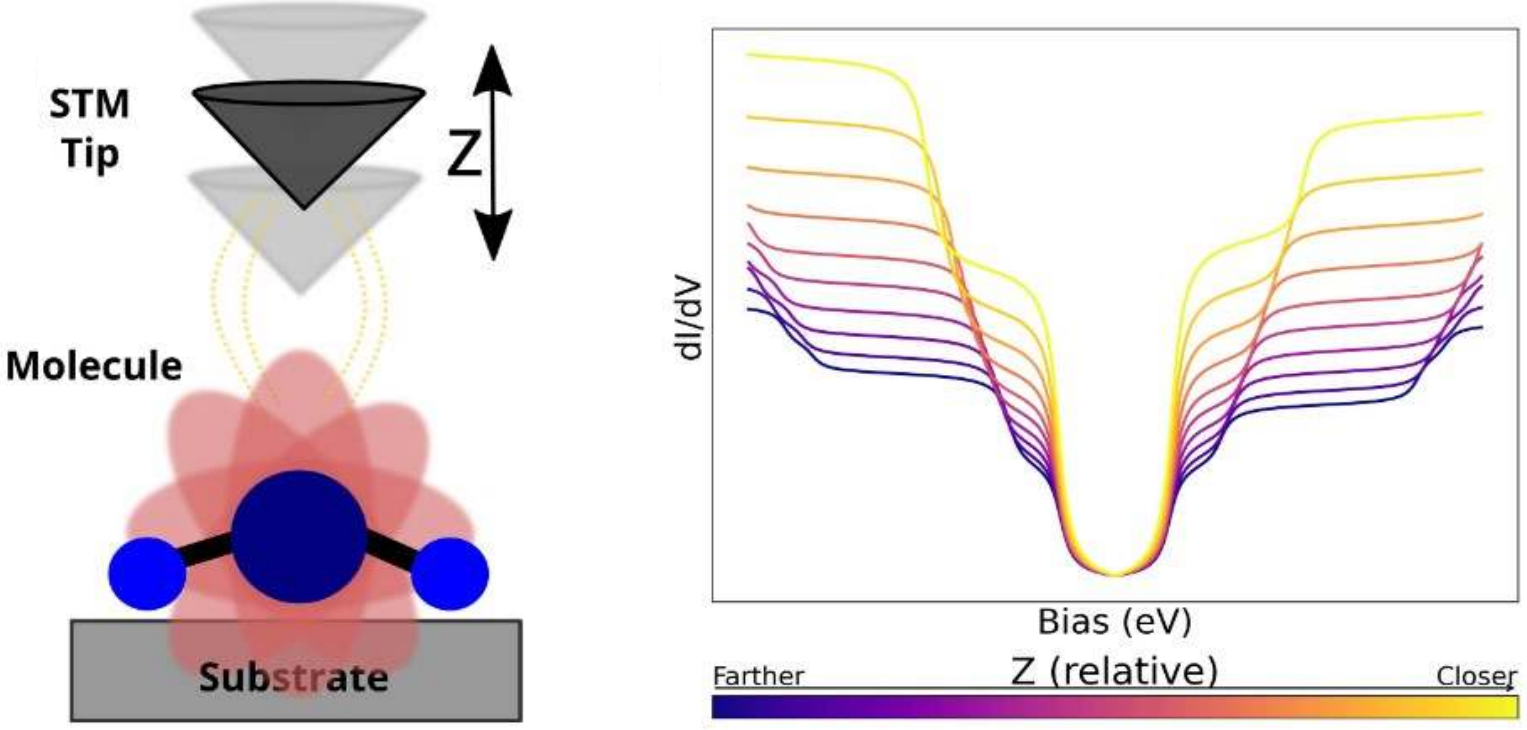
The magnetic field of a spin-polarized STM tip affects the energy of the spin flip of a S=1/2





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

Distance-dependent spectroscopy in STM

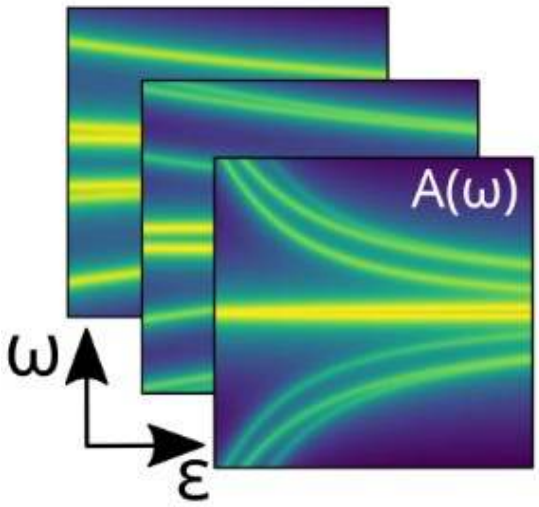


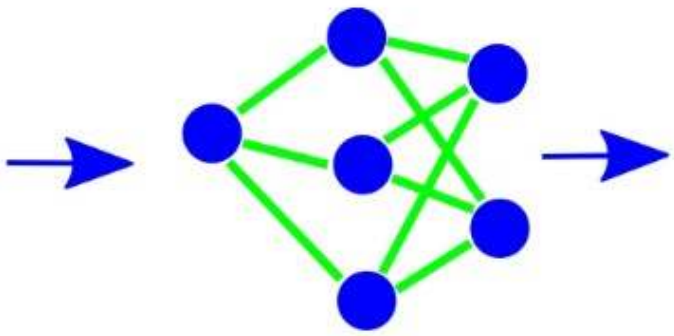
Spectroscopy can be distance dependent for specific systems

Can we learn the many-body Hamiltonian from this dependence?

Learning Hamiltonians from distance-dependent spectroscopy

Setpoint dependent spectroscopy



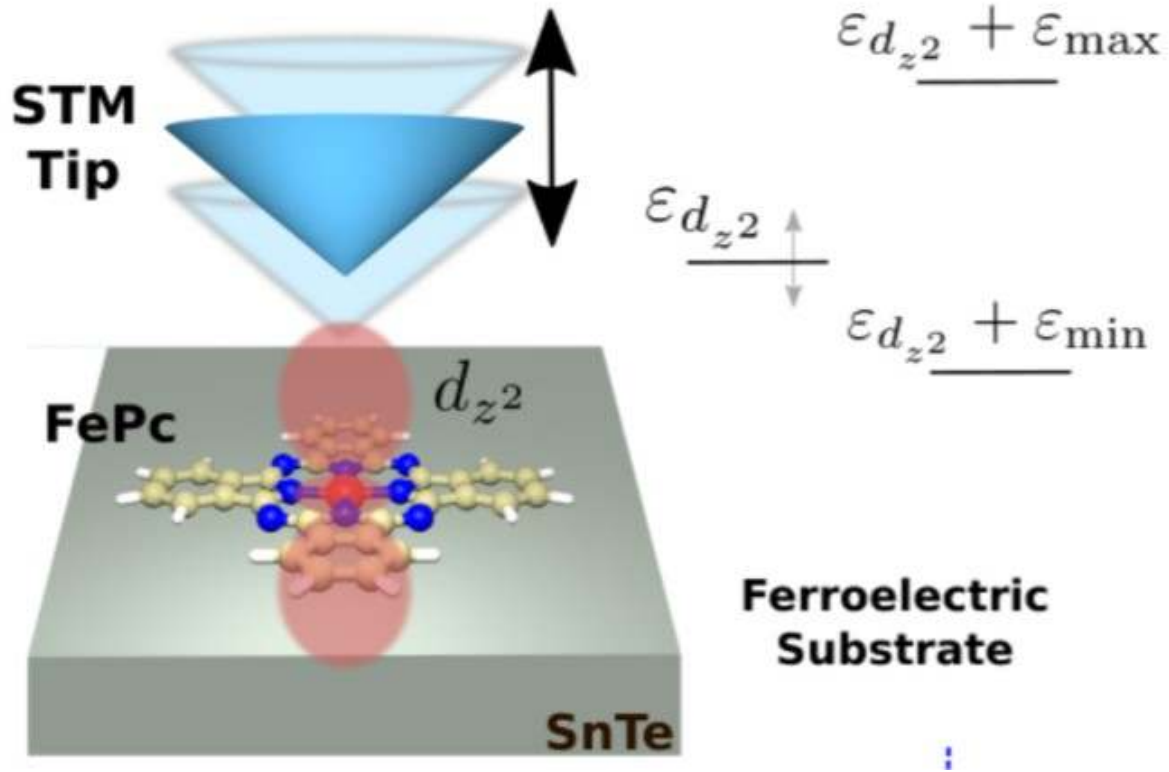


Trained Algorithm

Hamiltonian Inferrence:

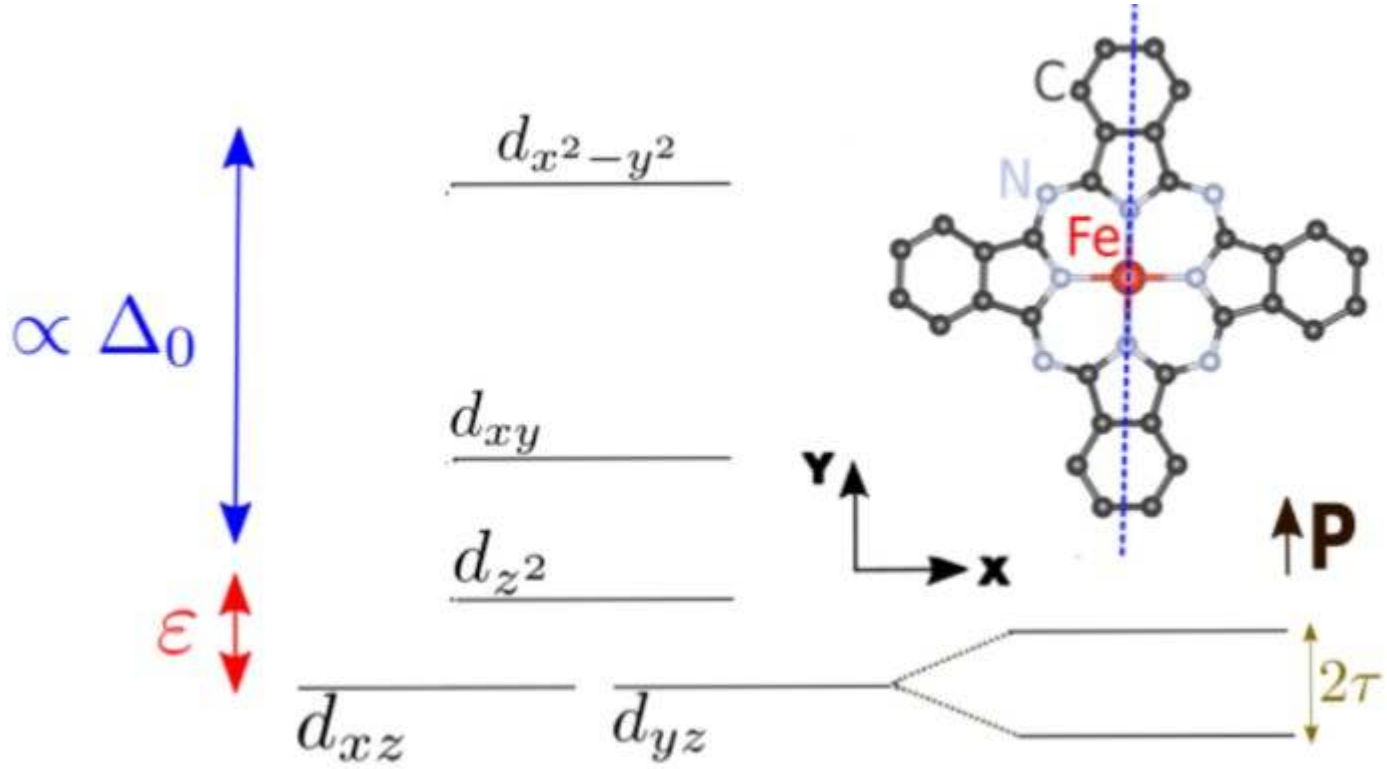
Crystal-field
Spin-Orbit Coupling
Orbital mixing
External parameters

The impact of STM in a molecular many-body Hamiltonian



In a multiorbital system, the STM tip effectively impact different orbitals differently

Setpoint-dependent Hamiltonian



In a multiorbital system, the STM tip effectively impact different orbitals differently

Setpoint-dependent Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\mathrm{CF}}(z) + \mathcal{H}_{\mathrm{Coulomb}} + \mathcal{H}_{\mathrm{SOC}}$$

$$\mathcal{H}_{\mathrm{CF}}(z) = \mathcal{H}^{0}_{\mathrm{CF}} + \mathcal{H}^{\mathrm{tip}}_{\mathrm{CF}}(z)$$

$$\mathcal{H}^{0}_{\mathrm{CF}} = \sum_{i,j,s} t_{ij}\, c^{\dagger}_{i,s} c_{j,s},$$

$$\mathcal{H}_{\mathrm{Coulomb}} = \sum_{ijkl ss'} V_{ijkl}\, c^{\dagger}_{i,s} c^{\dagger}_{k,s'} c_{l,s'} c_{j,s}$$

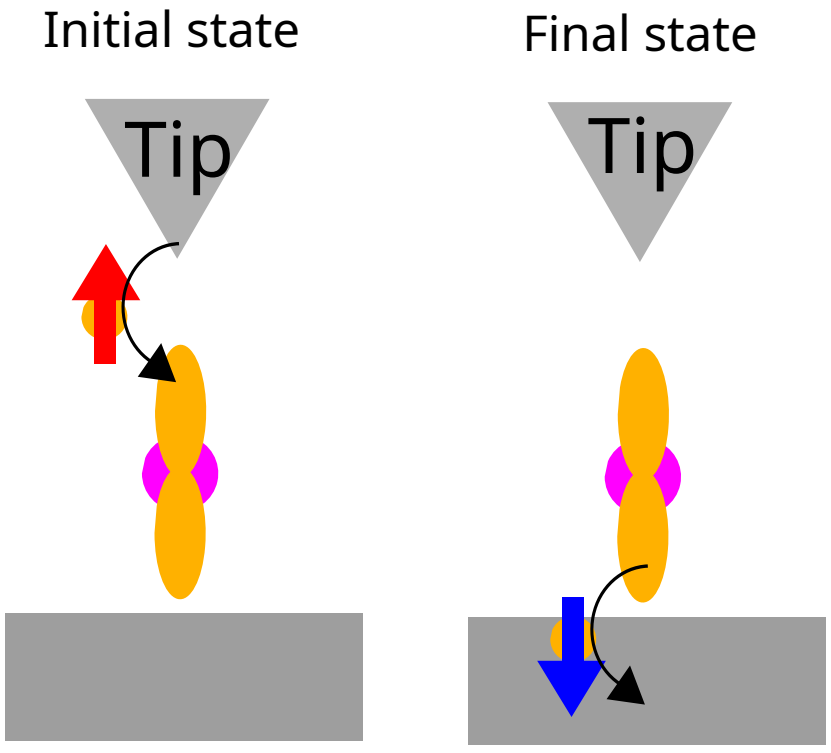
$$\mathcal{H}^{\mathrm{tip}}_{\mathrm{CF}}(z) = \sum_{i,j,s} \gamma_{ij}(z)\, c^{\dagger}_{i,s} c_{j,s}$$

$$\mathcal{H}_{\mathrm{SOC}} = \lambda_{\mathrm{SOC}} \sum_{ij\alpha ss'} \ell^{\alpha}_{ij}\, \sigma^{\alpha}_{ss'}\, c^{\dagger}_{i,s} c_{j,s'},$$

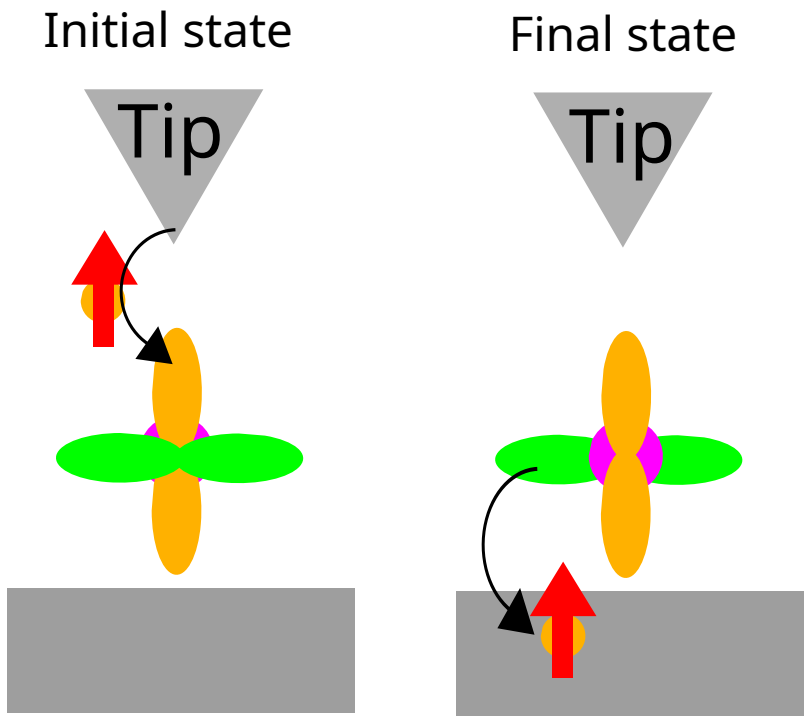
Distance dependence

Setpoint-dependent inelastic excitations

Spin flip excitations



Orbital many-body excitations



Setpoint-dependent inelastic excitations

$$\mathcal{H} = \mathcal{H}_{\mathrm{CF}}(z) + \mathcal{H}_{\mathrm{Coulomb}} + \mathcal{H}_{\mathrm{SOC}}$$

Spin-flip excitations

$$A_{\mathrm{flip}}(\omega) = \sum_{\nu} \langle\Omega|\, \hat{S}^{\nu}_{d_{z^2}}\, \delta(\omega - \mathcal{H} + E_{\Omega})\, \hat{S}^{\nu}_{d_{z^2}}\, |\Omega\rangle$$

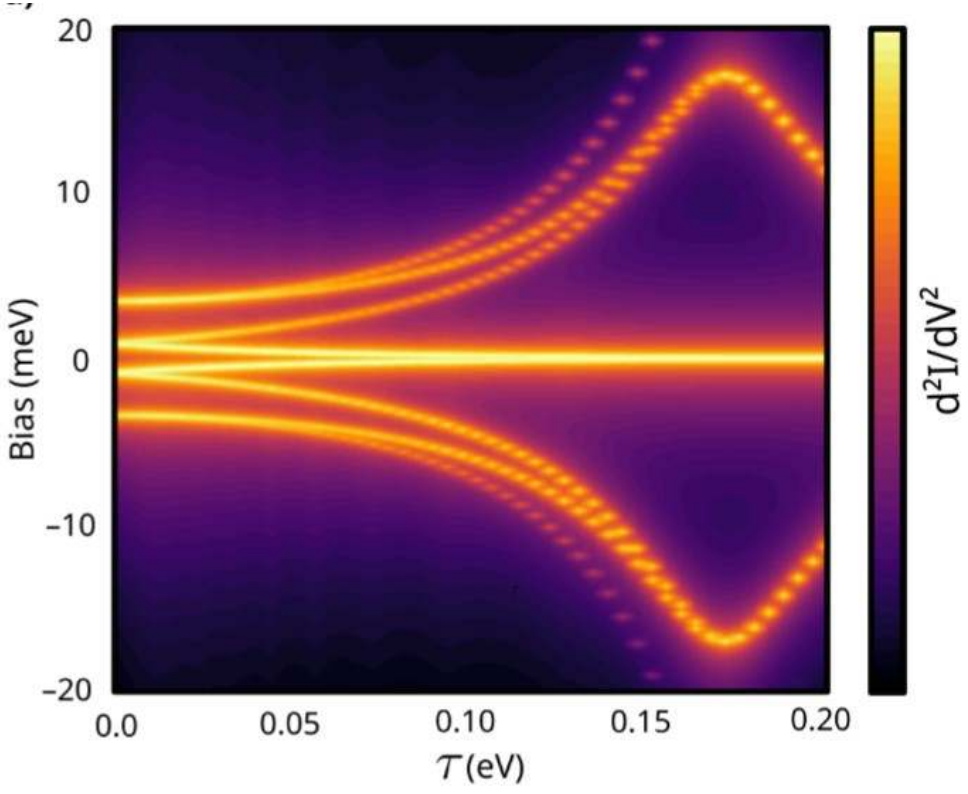
Orbital many-body excitations

$$A_{\mathrm{orb}}(\omega) = \sum_{\alpha,s,s'} \Theta(\omega)\, C^{s,s'}_{d_{z^2},\alpha}(\omega) + [1 - \Theta(\omega)]\, C^{s,s'}_{\alpha,d_{z^2}}(\omega)$$

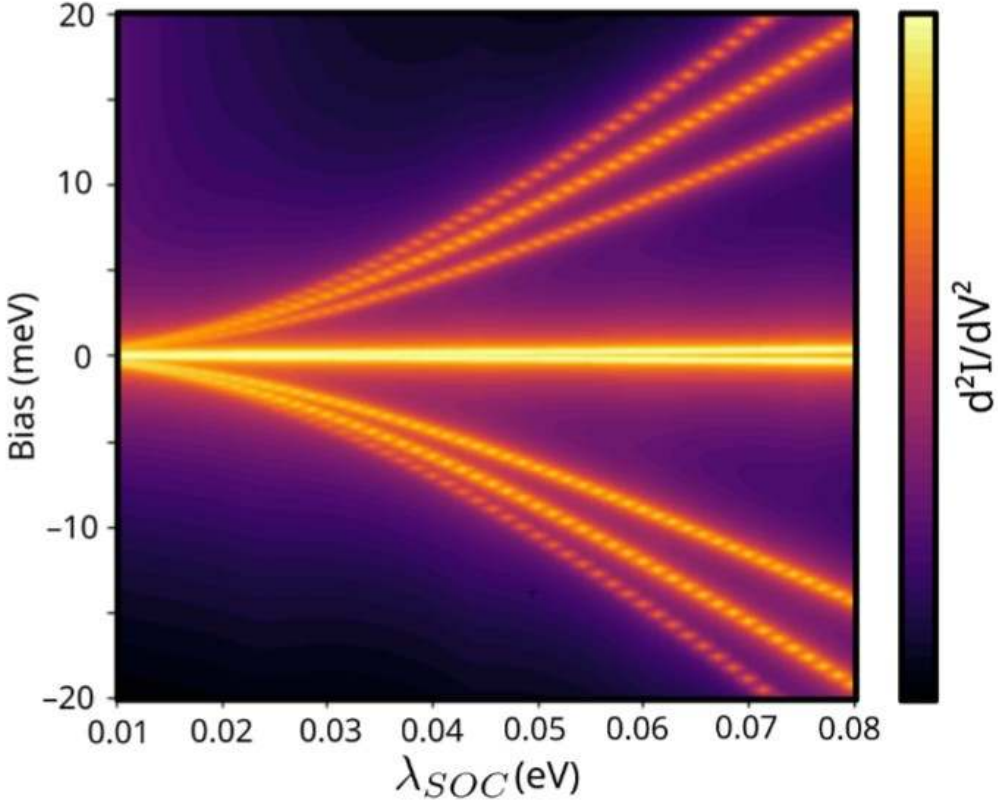
$$C^{s,s'}_{\alpha,\beta}(\omega) = \langle\Omega|\, c^{\dagger}_{\beta,s} c_{\alpha,s'}\, \delta(\omega - \mathcal{H} + E_{\Omega})\, c^{\dagger}_{\alpha,s'} c_{\beta,s}\, |\Omega\rangle$$

Setpoint-dependent inelastic excitations

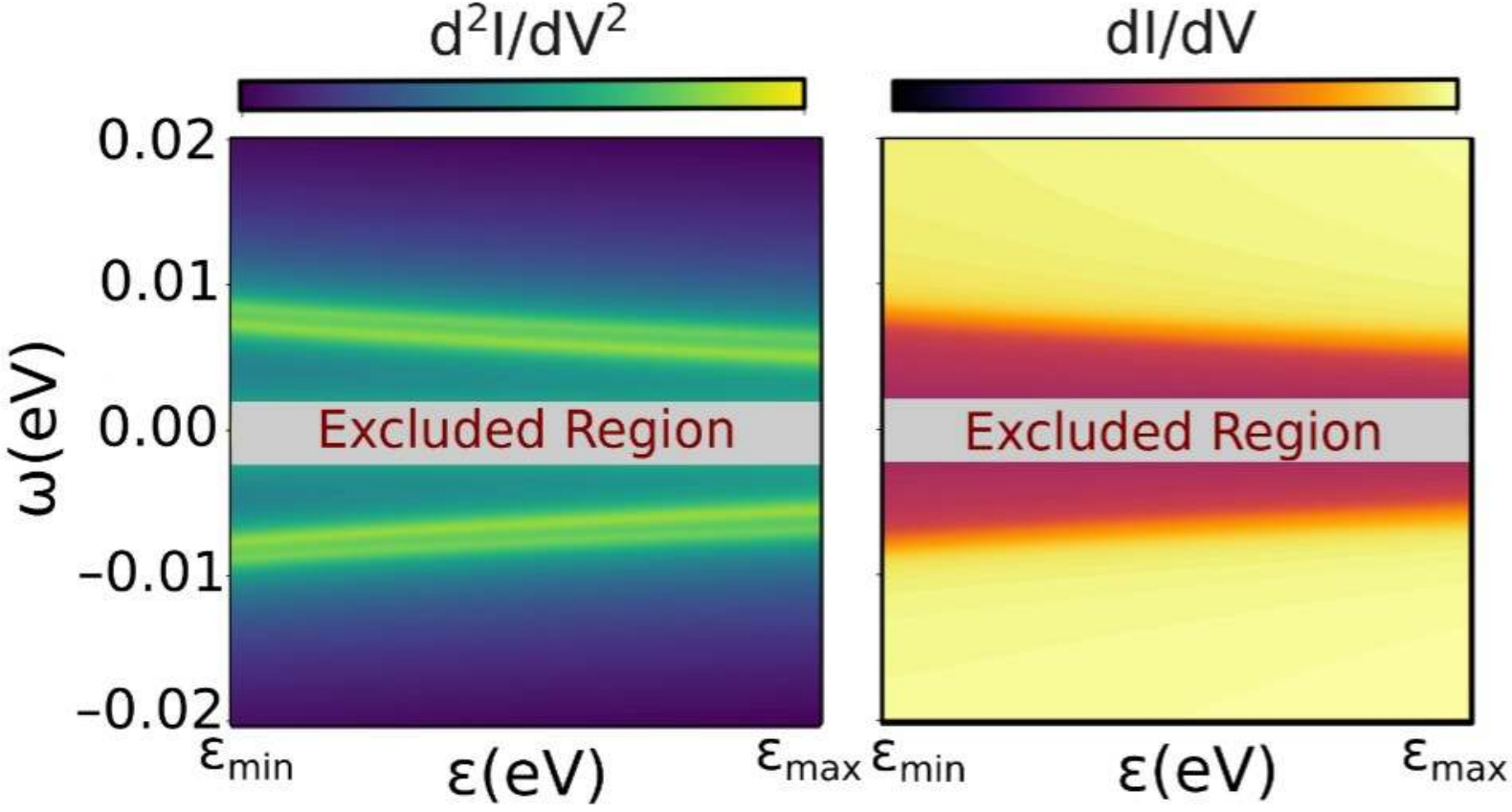
Dependence with crystal field



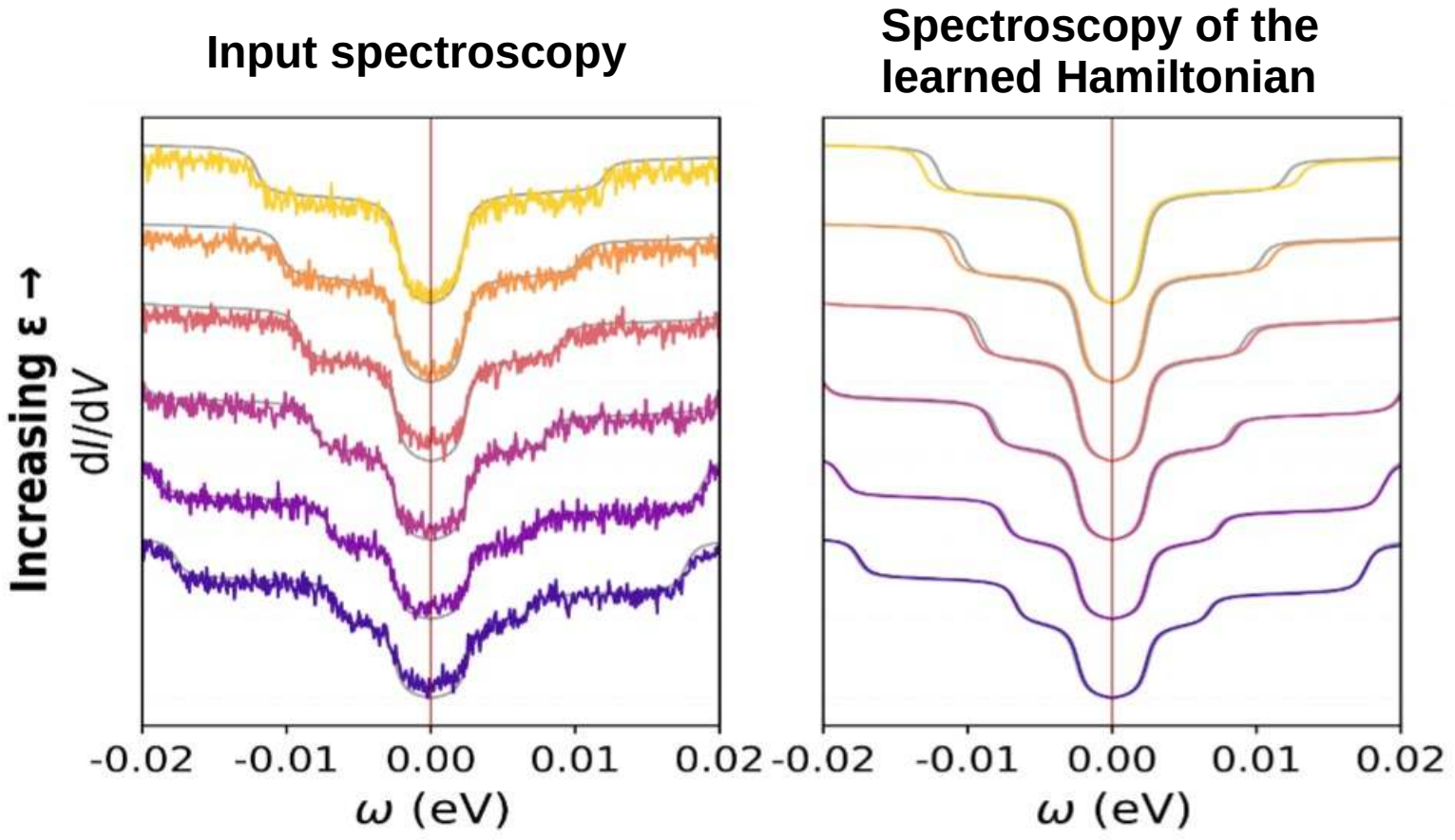
Dependence with spin-orbit coupling



Inelastic spectroscopy of the molecular magnet



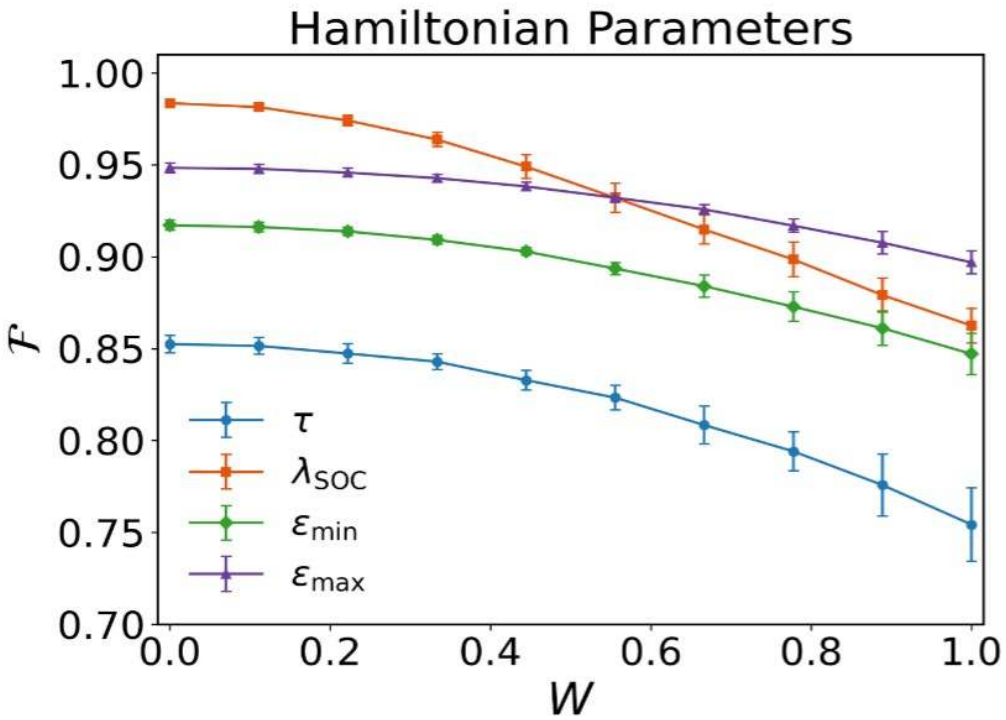
Original spectroscopy and learned Hamiltonian



Robustness of the Hamiltonian learning

Including noise $$dI/dV_{\mathrm{noisy}}(\varepsilon,\omega) = dI/dV_{\mathrm{test}}(\varepsilon,\omega) + \chi_{\mathrm{noise}}(\varepsilon,\omega)$$

$$\langle \chi^2_{\mathrm{noise}}(\varepsilon,\omega)\rangle = W^2$$

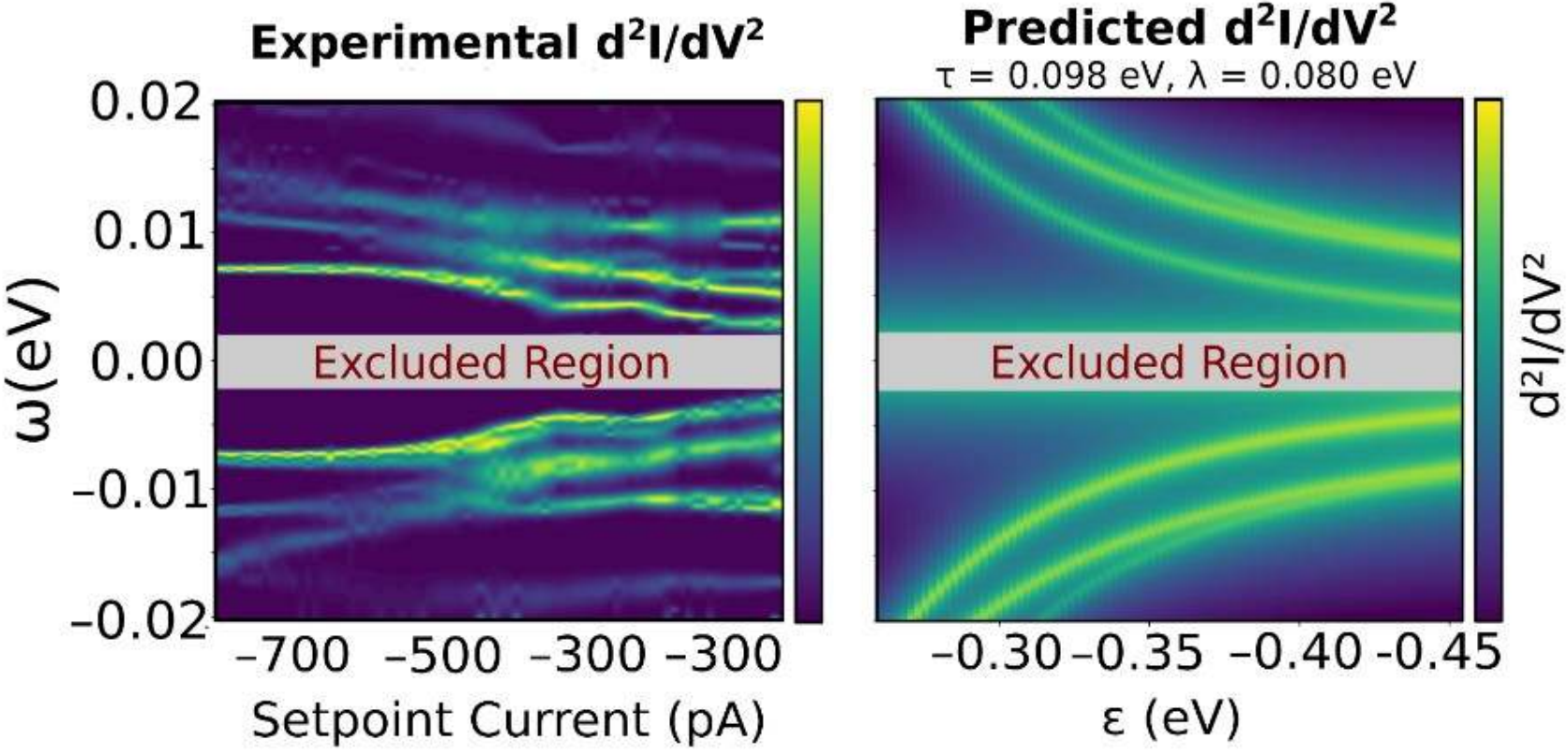


Fidelity

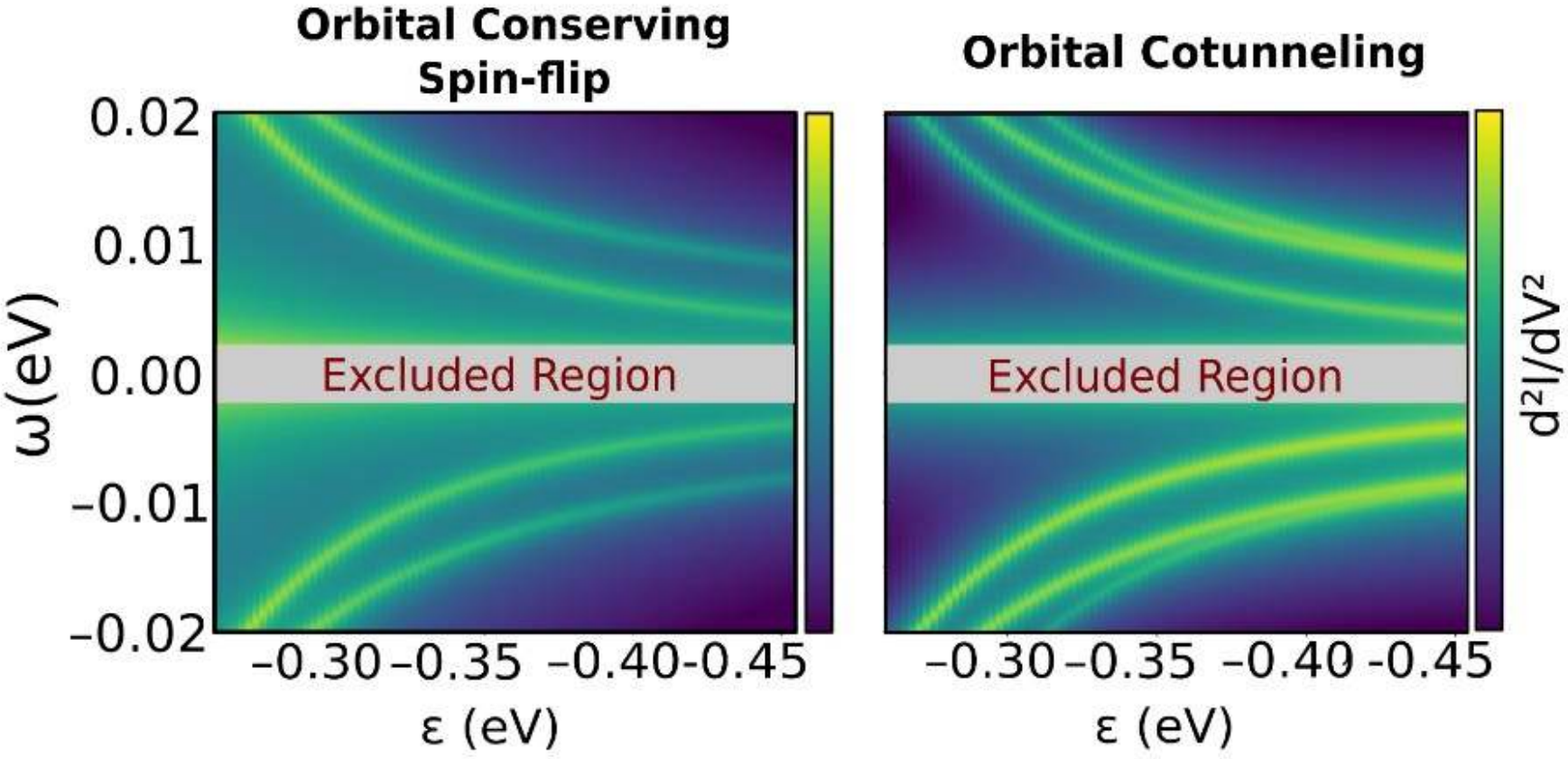
$\mathcal{F} = 1$ Perfect prediction

$\mathcal{F} = 0$ No prediction ability

Hamiltonian molecular learning from experimental spectroscopy

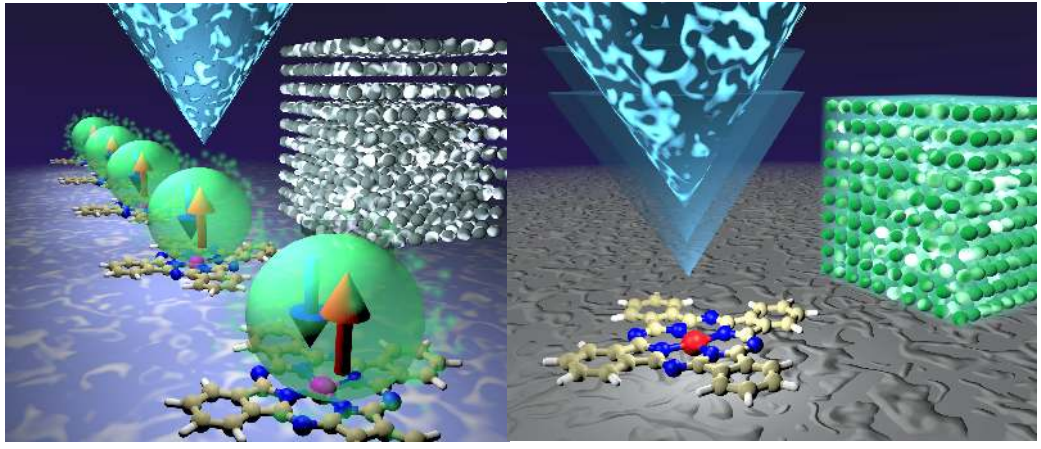


Disentangling inelastic channels



Take home

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



Nano Letters 25, 36, 13435-13440 (2025)
arXiv:2601.19371 (2026)

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