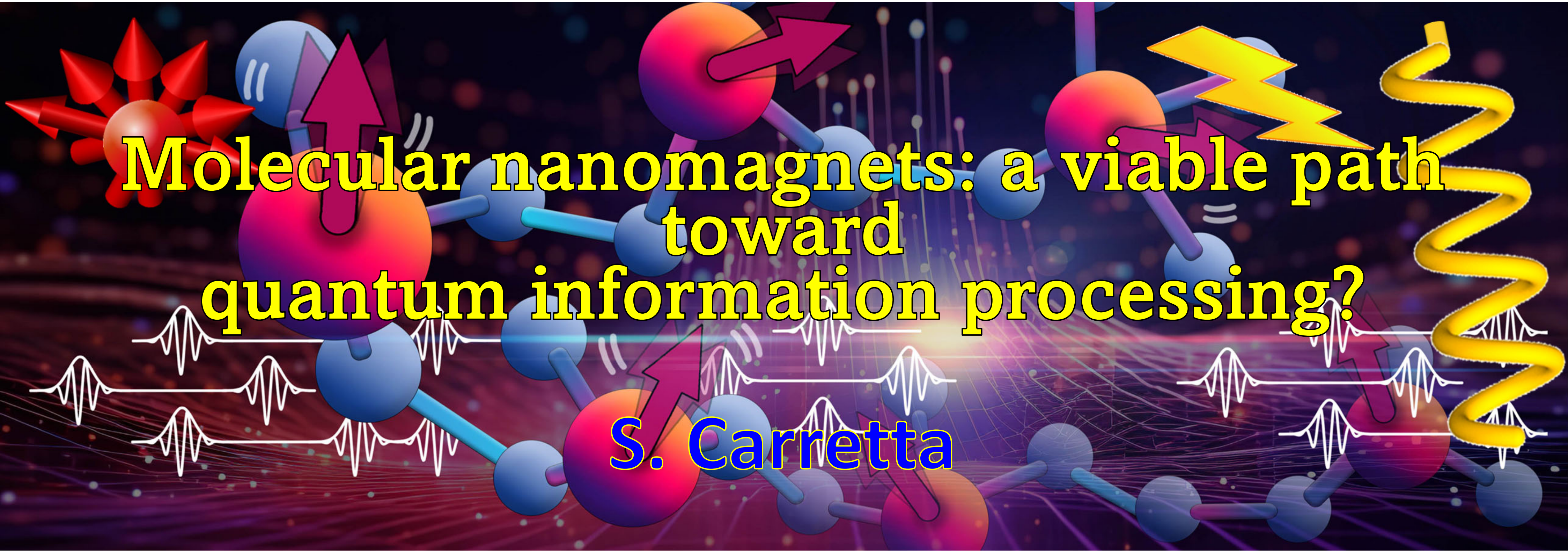
# Molecular nanomagnets: a viable path toward quantum information processing?



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spin QS spin-based QUANTUM SCIENCE

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- Matteo Mezzadri
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UNIVERSITÀ DI PARMA

spin QS
spin-based QUANTUM SCIENCE

https://spinquantumscience.unipr.it/

European Research Council
Established by the European Commission

project n. 101071533

CASTLe

**PNRR MUR project PE0000023-NQSTI**
National Quantum Science and Technology Institute

novo nordisk **foundation**

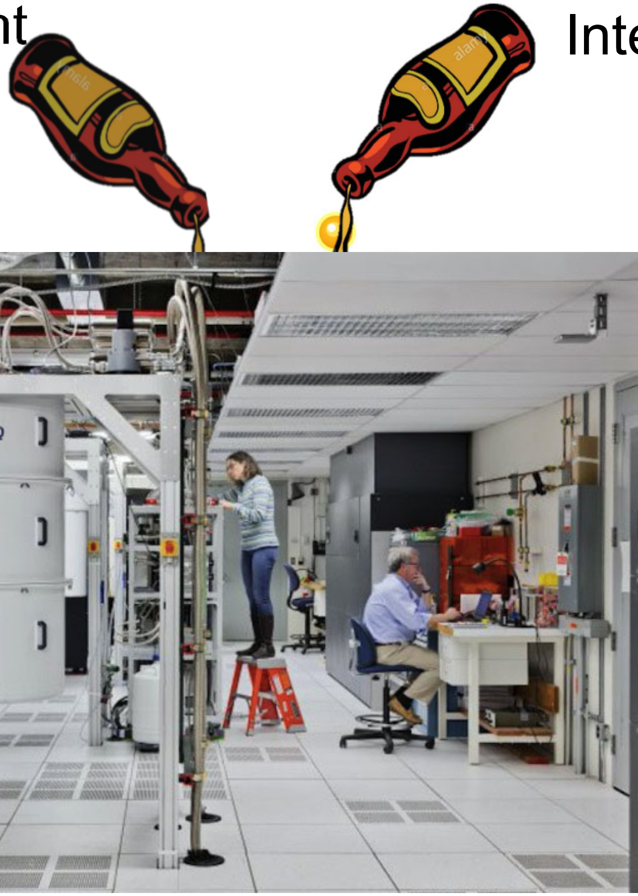
Coherent addressing of isotopically pure lanthanide complexes by photons and efficient quantum error correction for Quantum Information Technologies

# Basic Ingredients of Quantum Computation

Entanglement

Interference

Superp




Applications:

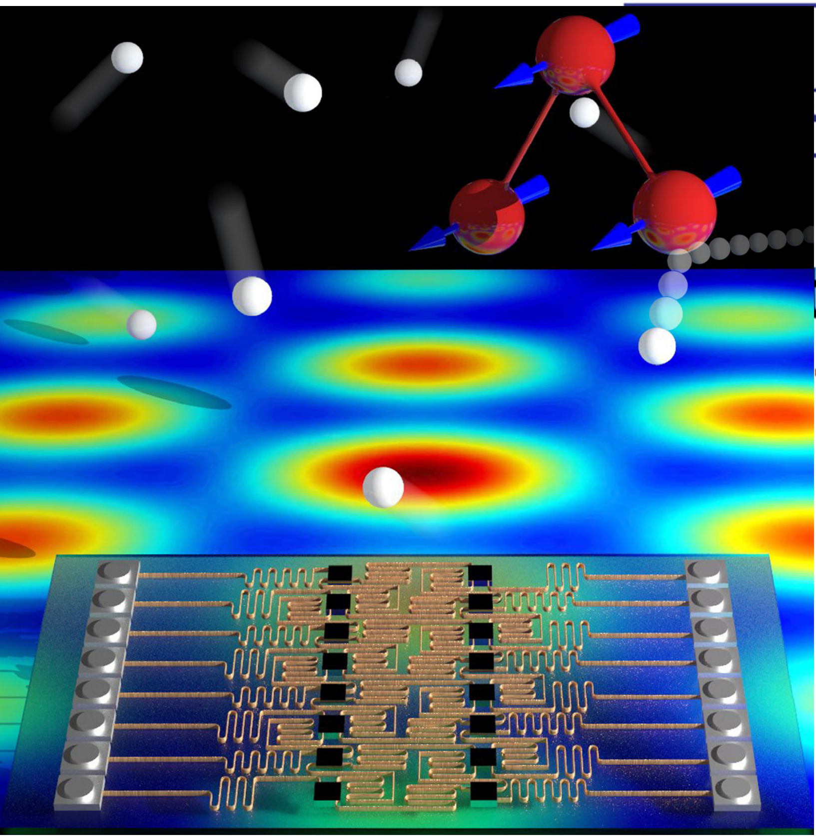
- Quantum simulation
- Drug and material design
- Traffic flow
- Protein folding

- Factorization
- Cryptography and security
- Search
- Optimization

# Qubits

Qubits are two-level quantum systems and can be realized using a variety of physical objects.

Superconducting qubits and Ion traps probably represent the most advanced platform.



cs

LETTERS

https://doi.org/10.1038/s41567-019-0437-4

## tum hardware simulating four-dimensional tic neutron scattering

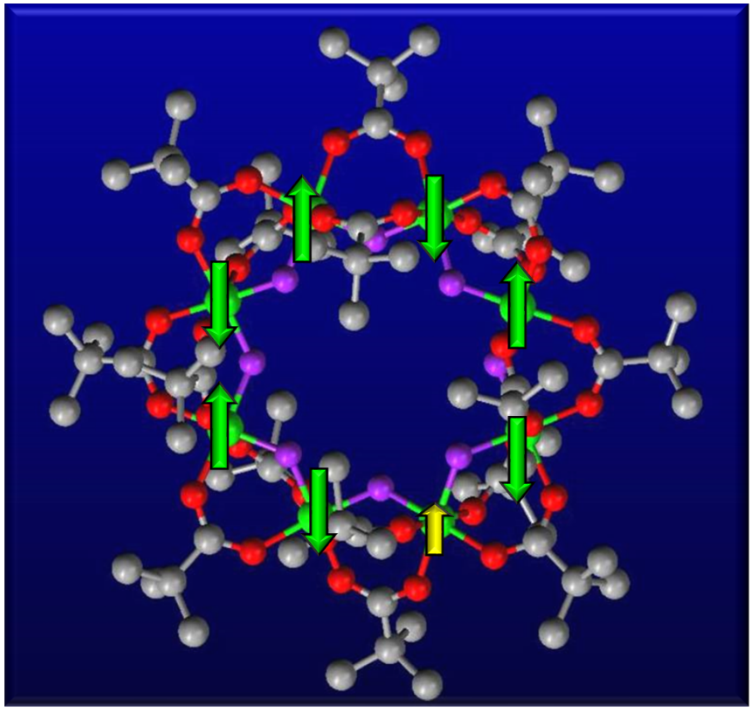
F. Tacchino[2,5], M. Grossi[2,3], P. Santini[1], I. Tavernelli[4], D. Gerace[2] and S. Carretta[1*]

**Quantum simulations** of the **spin dynamics** of prototypical spin systems and calculation of the **4D inelastic neutron cross-section**

# Molecular Nanomagnets

Molecules in which the magnetic core is constituted by a small number of exchange-coupled magnetic ions.

A. Chiesa, P. Santini, E. Garlatti, F. Luis, S. Carretta,
**Reports on Progress in Physics,** 87, 034501 (2024).



**Cr7Ni**

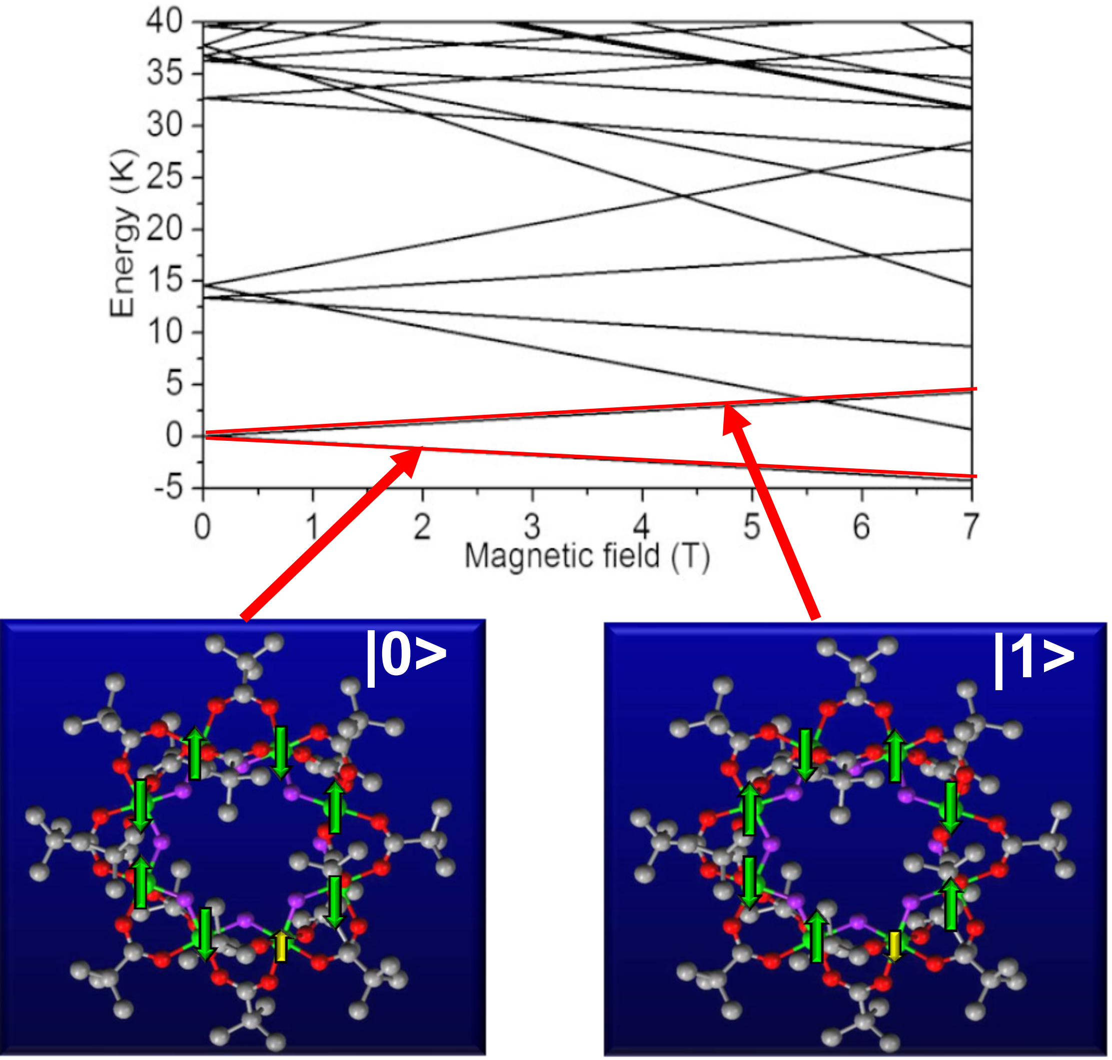
***Phys. Rev. Lett. 94, 207201 (2005)***

***Phys. Rev. Lett. 98, 167401 (2007).***

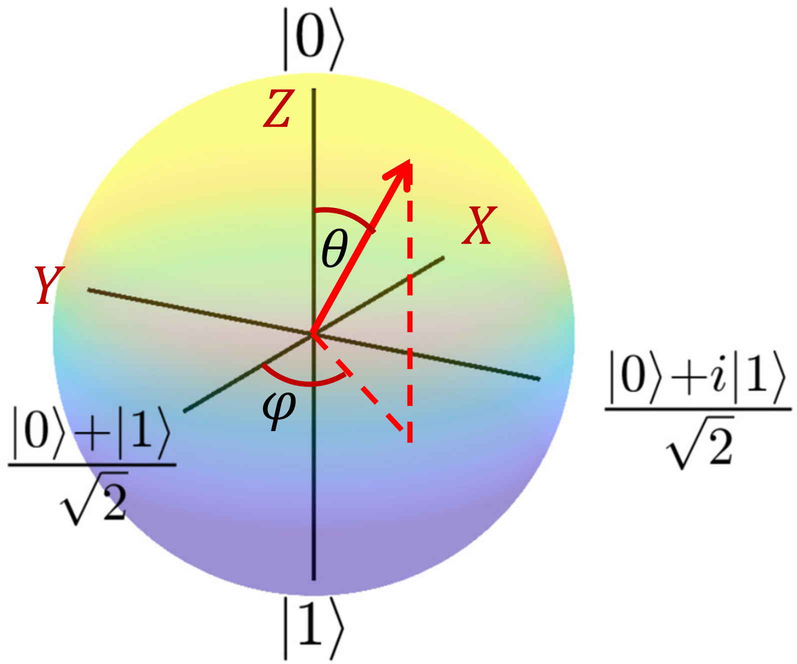
$$H = \sum_i J_i \mathbf{s}(i) \cdot \mathbf{s}(i+1) + \sum_i d_i [s_z^2(i) - s_i(s_i+1)/3] + \sum_{i>j} \mathbf{s}(i) \cdot \mathbf{D}_{ij} \cdot \mathbf{s}(j) + \mu_B \sum_i g_i \mathbf{B} \cdot \mathbf{s}(i),$$

# Molecular Nanomagnets as Qubits: 1) computational basis

A. Chiesa, P. Santini, E. Garlatti, F. Luis, S. Carretta,
**Reports on Progress in Physics,** 87, 034501 (2024).

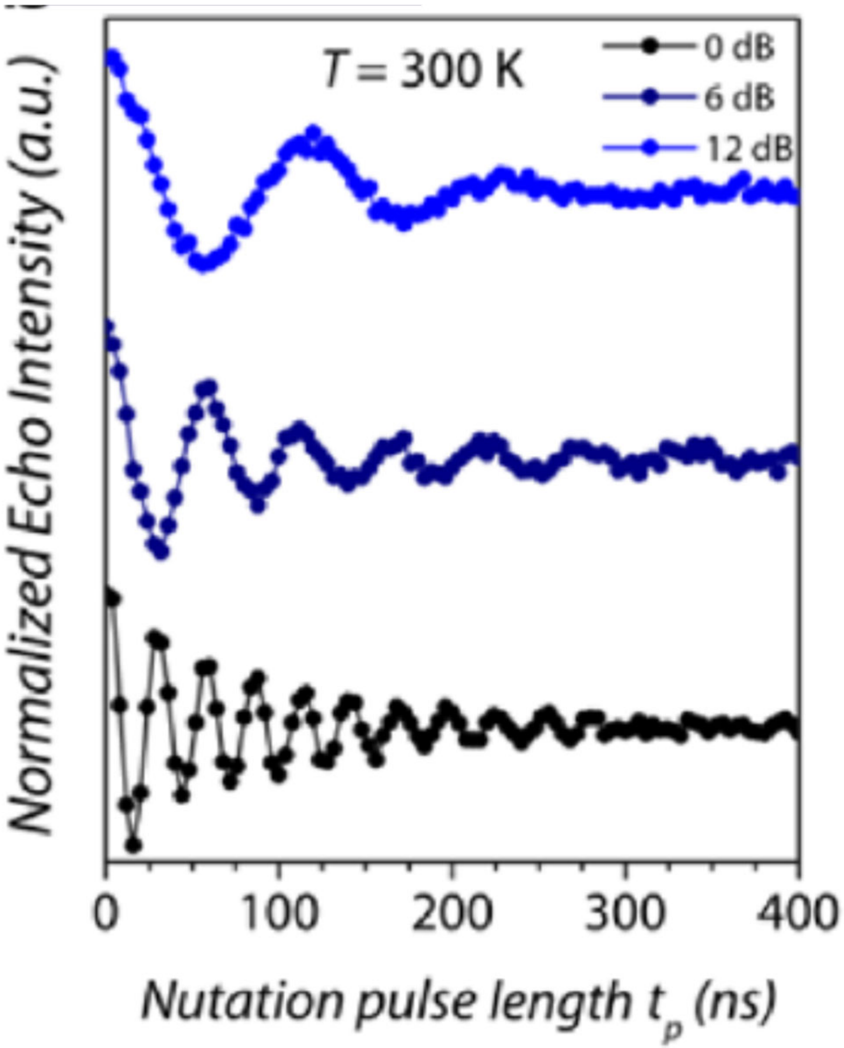


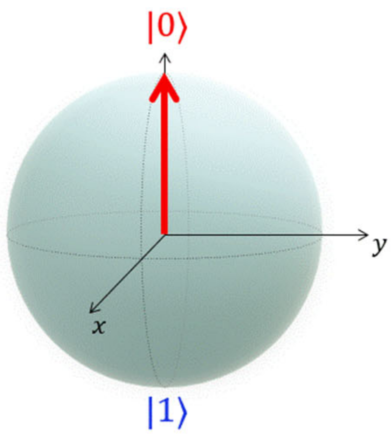

$$|\psi(\theta,\varphi)\rangle = \cos\frac{\theta}{2}|0\rangle + e^{i\varphi}\sin\frac{\theta}{2}|1\rangle$$

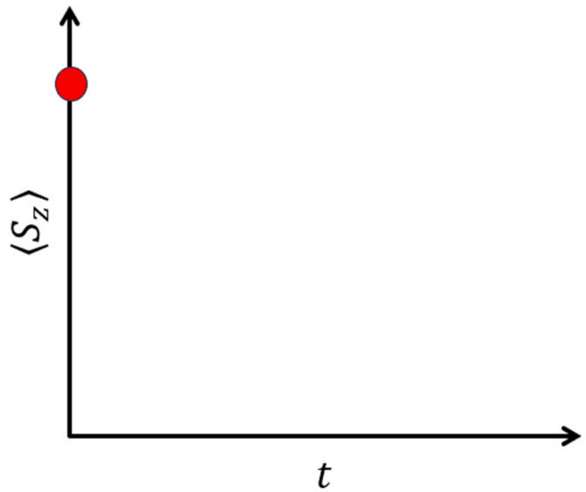


# Molecular Nanomagnets as Qubits: 2) single-qubit gates

Single-qubit logical operations in molecular qubits can be produced by **resonant magnetic pulses**



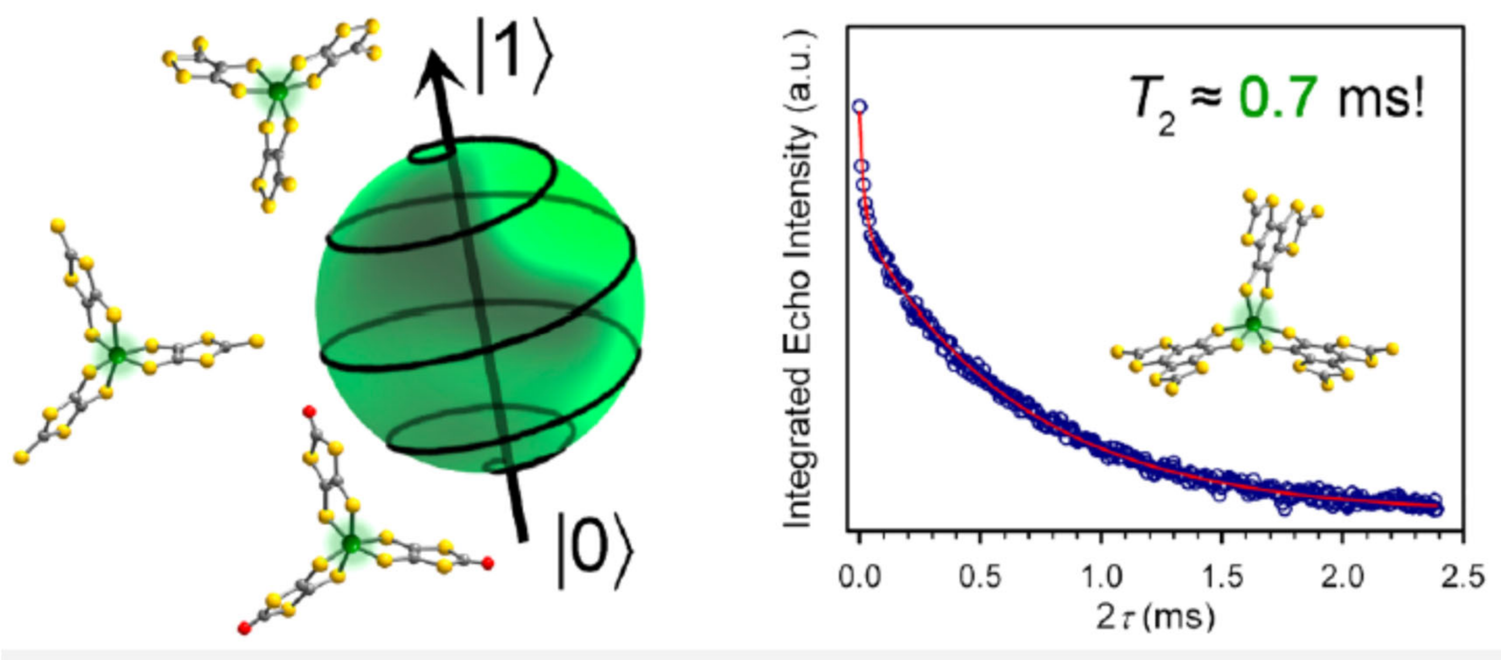




Room temperature coherence in a Vanadyl Phthalocyanine

***Atzori et al., J. Am. Chem. Soc. 138, 2154 (2016)***

# Molecular Nanomagnets as Qubits: 2) single-qubit gates

Coherence times can be very long!




*Zadrozny et al, ACS Cent. Sci.* ***1****, 488 (2015)*

# Molecular Nanomagnets as Qubits: 3) two-qubit gates

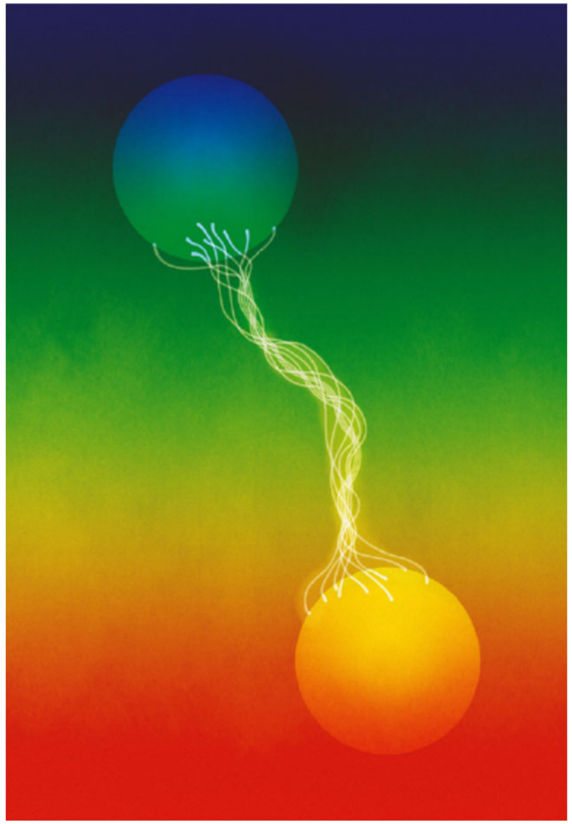
- The state of a qubit is changed in a way which depends on the state of another

CNOT

| CNOT | $\|00\rangle$ | $\|01\rangle$ | $\|10\rangle$ | $\|11\rangle$ |
|---|---|---|---|---|
| $\langle 00\|$ | 1 | 0 | 0 | 0 |
| $\langle 01\|$ | 0 | 1 | 0 | 0 |
| $\langle 10\|$ | 0 | 0 | 0 | 1 |
| $\langle 11\|$ | 0 | 0 | 1 | 0 |

$$\frac{|00\rangle + |10\rangle}{\sqrt{2}} \longrightarrow \frac{|00\rangle + |11\rangle}{\sqrt{2}}$$

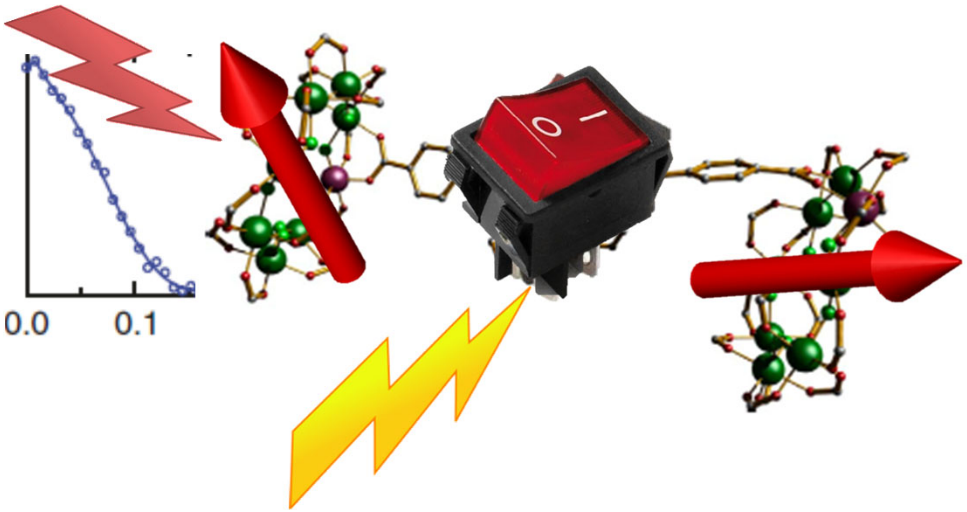
product → entangled



*It is still a 2-qubit state but cannot be written as product.*

# Molecular Nanomagnets as Qubits: 3) two-qubit gates

- The state of a qubit is changed in a way which depends on the state of another




Phys. Rev. Lett. **107**, 230502 (2011).
Sci. Rep. **4**, 7423 (2014)
Chem **1**, 727 (2016).
Nat. Commun. **7**, 11377 (2016).

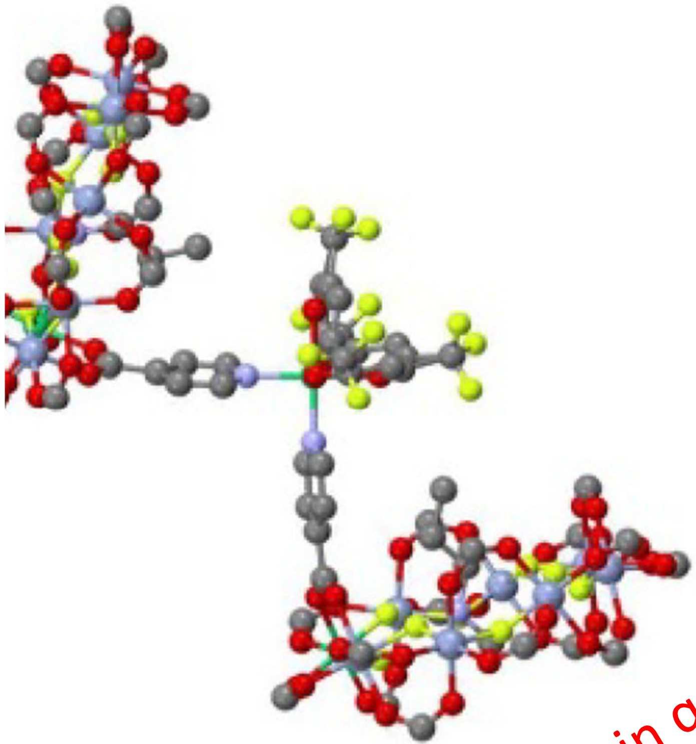
Ardavan et al., npj Quantum Information 1, 15012 (2015);
Nat. Comm. 14, 7029 (2023)

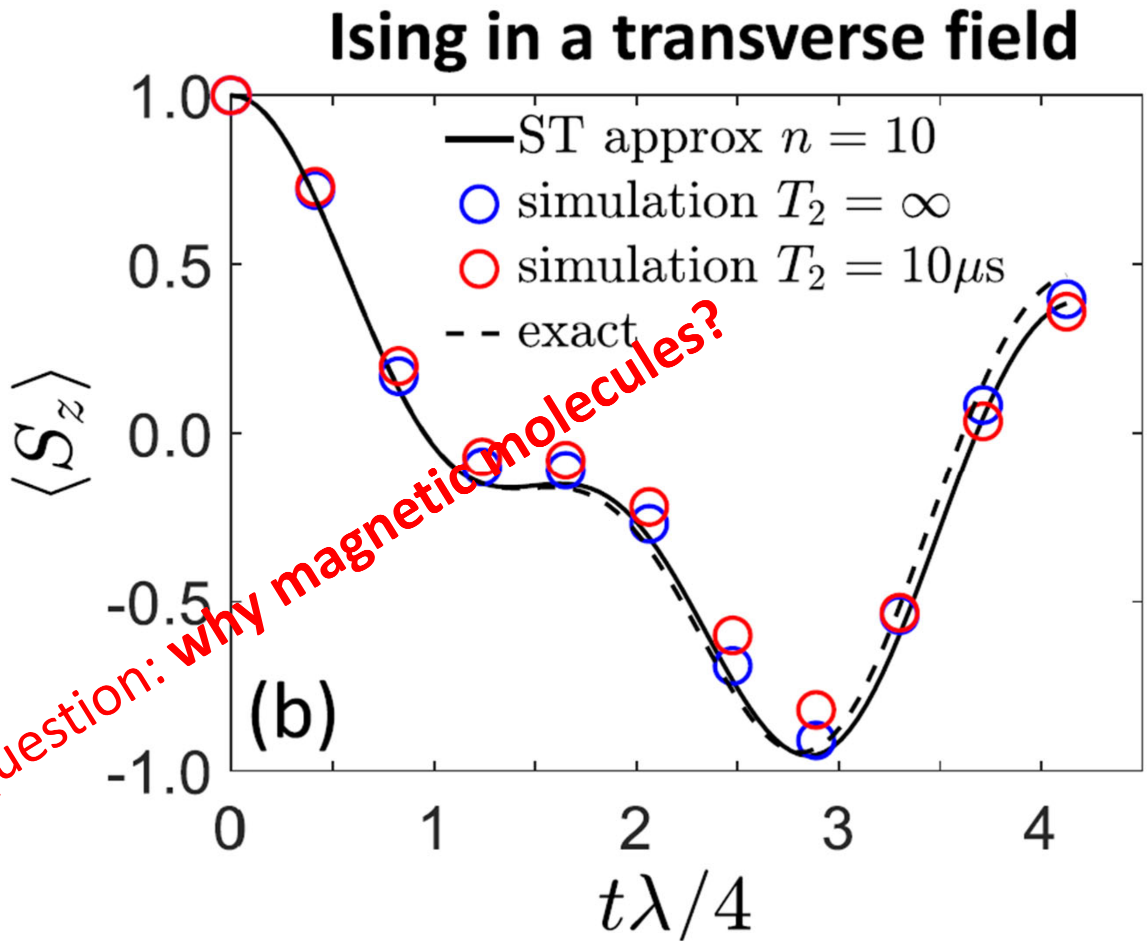
When the switch is in the ground state, the switch-Q interaction act as a magnetic field -> Single-qubit gates

The excitation energy of the switch depends on the state of both Qubits -> Controlled Z gate

- A **switchable interaction** is mandatory to efficiently implement quantum computing algorithms

# Molecular Nanomagnets as Qubits: 3) two-qubit gates






Main question: **why magnetic molecules?**

The idea can work well even with limited coherence times

# Molecular Nanomagnets as Qudits

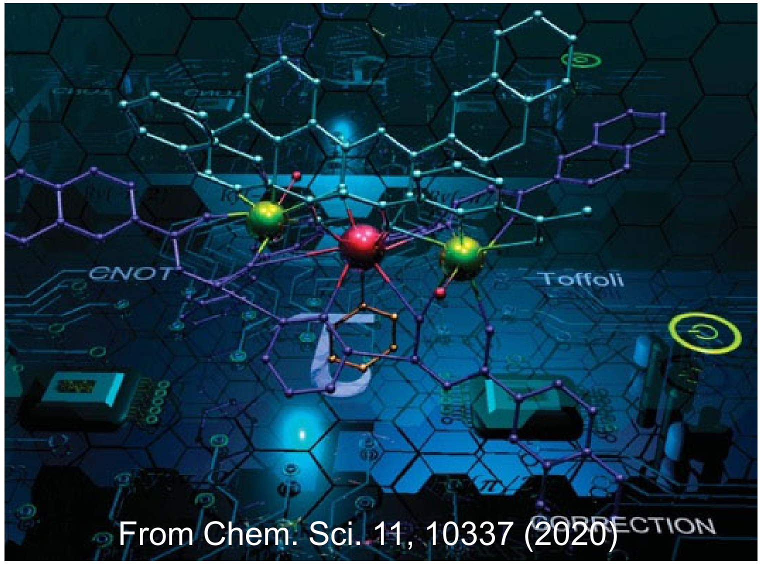
MNMs are typically characterized by a sizeable number of accessible low-energy levels

A. Chiesa, P. Santini, E. Garlatti, F. Luis, S. Carretta,
**Reports on Progress in Physics,** 87, 034501 (2024).

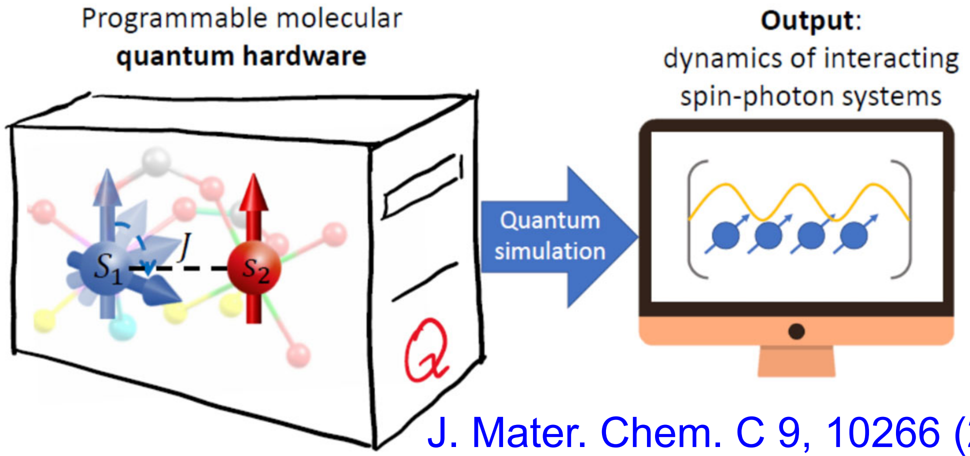
Quantum systems with d > 2 levels, called qudits, can enhance the power of quantum logic:

a) Integrate multiple quantum resources **Quantum Error Correction**

b) reduce the computational costs of some applications **Quantum Simulations**.



From Chem. Sci. 11, 10337 (2020)




J. Mater. Chem. C 9, 10266 (2021).

J. Am. Chem. Soc. 144, 10266 (2022).

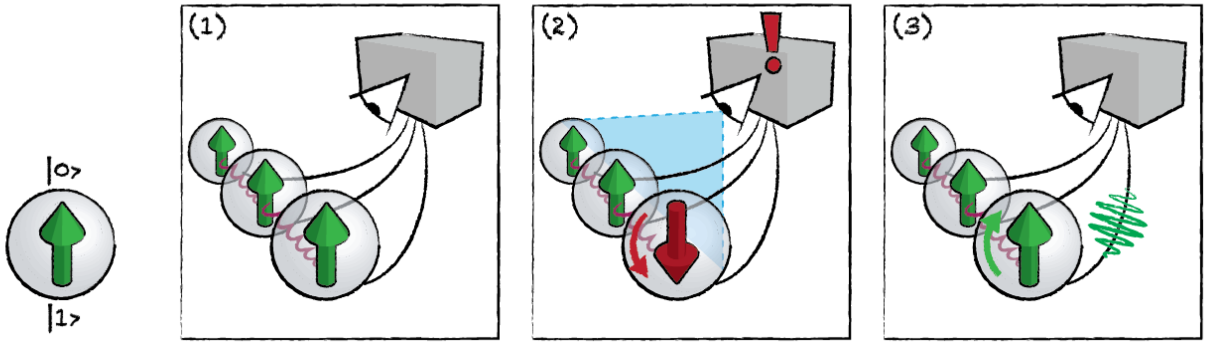
# Quantum Error Correction

**Many physical qubits** to encode a single logical qubit.

**Qudit-encoding**: a **single multi-level object** to encode an error protected qubit.

**less demanding!**

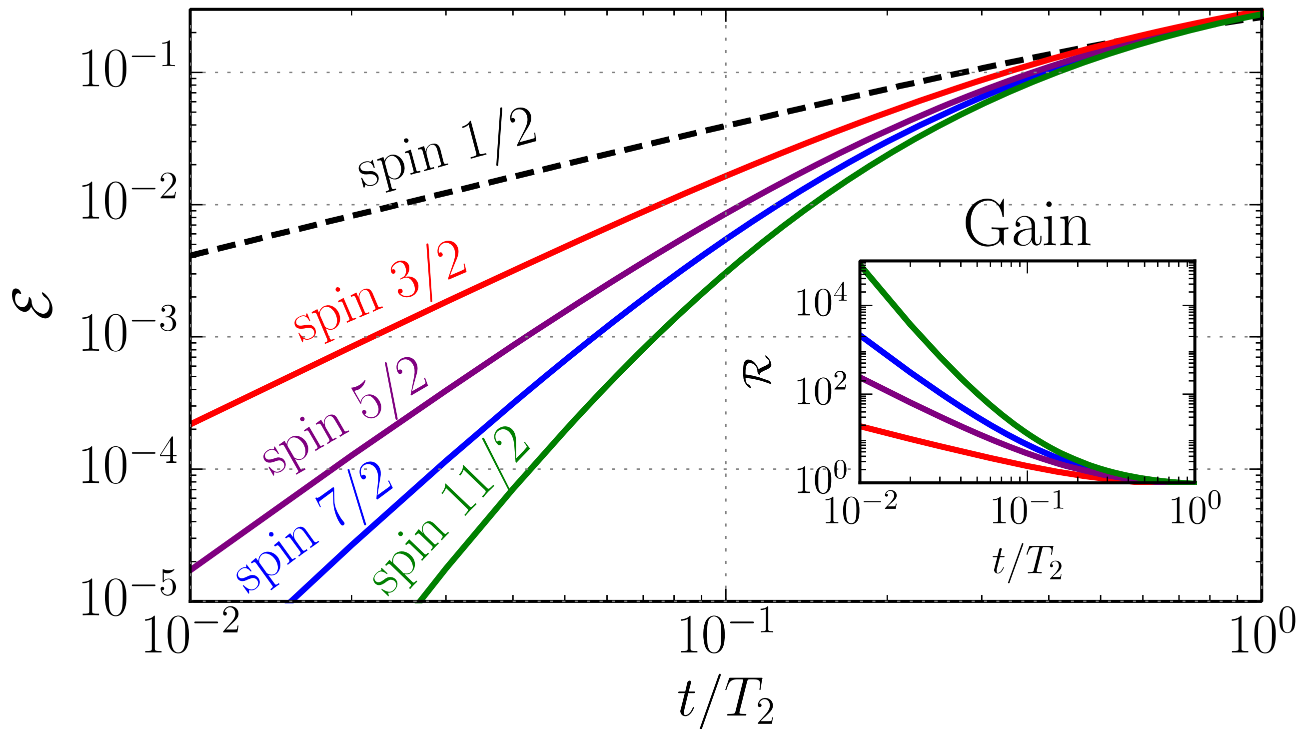
Nature Nanotech. 9, 171–176 (2014)

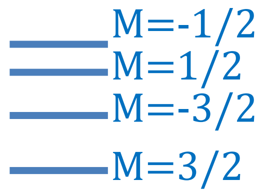



Large overhead of physical qubits and operations

# Quantum error correction with spin S qudits

We design a code correcting **main error** occurring in molecular qubits: **pure dephasing**.




$$= \frac{\sqrt{3}|3/2\rangle - |-1/2\rangle}{2} \equiv |e_0\rangle$$

$$= \frac{|1/2\rangle - \sqrt{3}|-3/2\rangle}{2} \equiv |e_1\rangle$$

- Error brings $|0_L\rangle$ and $|1_L\rangle$ to distinguishable states
- Preserves the superposition

Errors are **detectable** and **correctable**

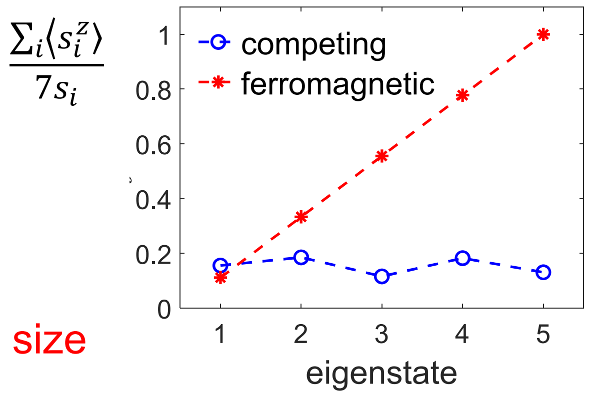
$$|\psi\rangle = \alpha|0_L\rangle + \beta|1_L\rangle \xrightarrow{S_z \text{ error}} \alpha|e_0\rangle + \beta|e_1\rangle$$

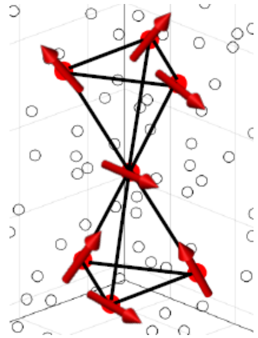

Chiesa, Macaluso, Petiziol, Wimberger, Santini, Carretta, ***J. Phys. Chem. Lett.* 11**, 8610 (2020)

Petiziol, Chiesa, Wimberger, Santini, Carretta, ***NPJ Quantum Information*** *7, 133 (2021)*

# Which are the best molecular qudits?

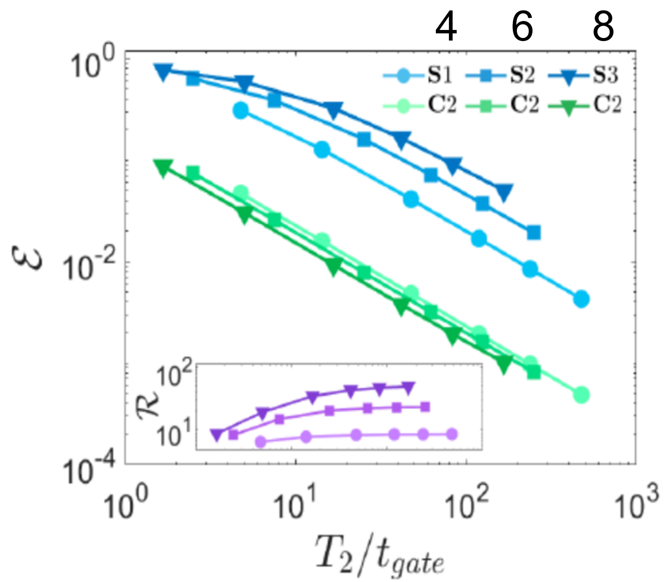
$$\gamma_{\mu\nu} = \sum_{jj'=1}^{7} C_{jj'}^{zzz}[-2\langle\mu|s_j^z|\mu\rangle\langle\nu|s_{j'}^z|\nu\rangle + \langle\mu|s_j^z|\mu\rangle\langle\mu|s_{j'}^z|\mu\rangle + \langle\nu|s_j^z|\nu\rangle\langle\nu|s_{j'}^z|\nu\rangle]$$



Small $\langle s_i^z \rangle$

Not increasing with the size



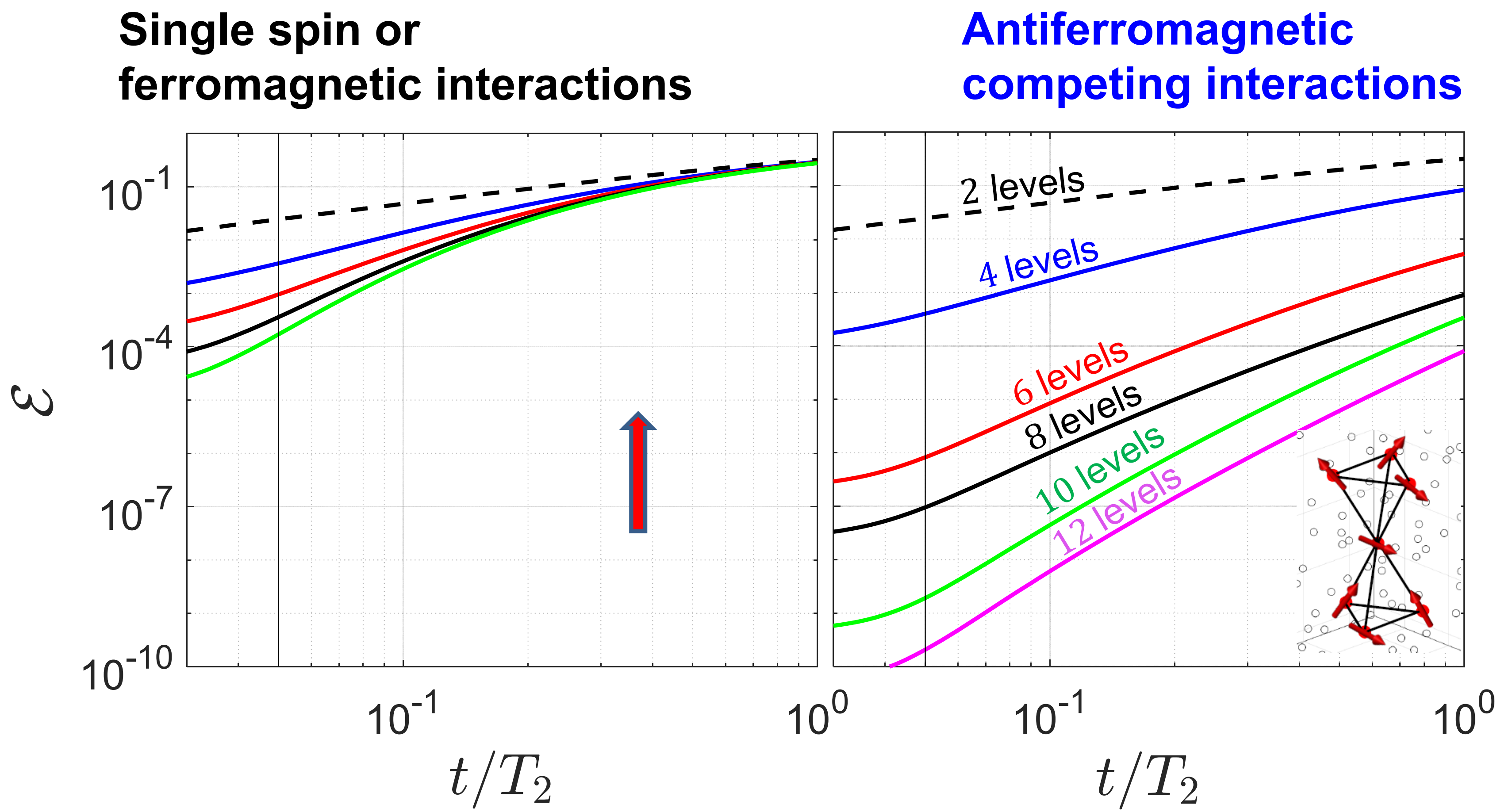

Quantum Fourier Transform

$$|\tilde{b}\rangle = \hat{U}_{\mathrm{QFT}}|b\rangle = 2^{-n/2}\sum_{a=0}^{2^n-1}\exp\left(i\frac{2\pi ab}{2^n}\right)|a\rangle$$

*Chiesa, Petiziol, Chizzini, Santini, Carretta,*
***J. Phys. Chem. Lett. 13****, 6468 (2022)*

*Chizzini, Crippa, Chiesa, Tacchino, Petiziol, Tavernelli,*
*Santini, Carretta,* ***Phys. Rev. Res. 4,*** *043135 (2022)*

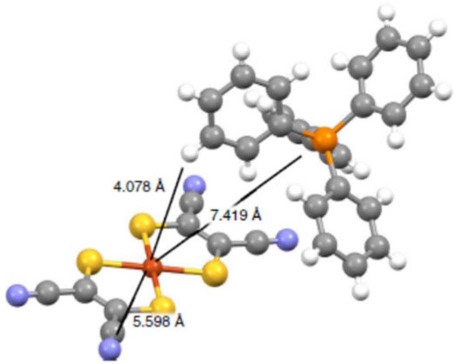
# Optimal molecular qudits for QEC



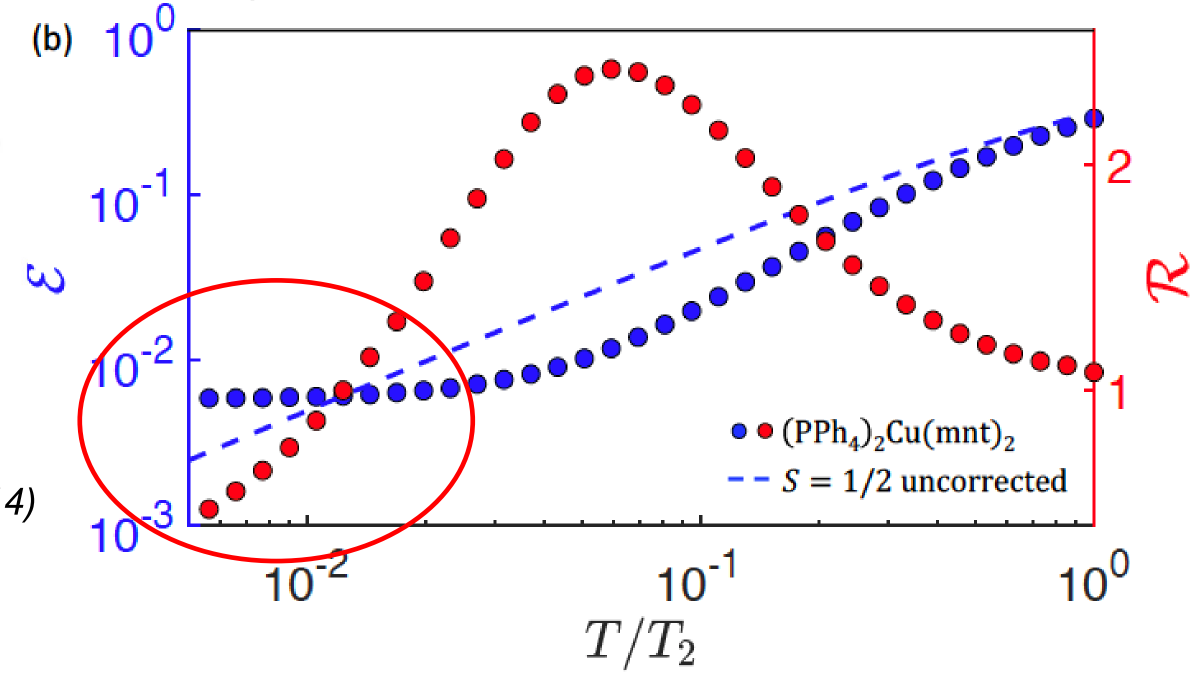

A. Chiesa, F. Petiziol, M. Chizzini, P. Santini, S. Carretta, **J. Phys. Chem. Lett.** 13, 6468 (2022).

# Error accumulation during correction and gates

$Cu^{2+}$ molecule ($I = 3/2$ qudit + $S = 1/2$ electron ancilla).




Parameters from
*Nat. Commun.* **5**, *5304 (2014)*



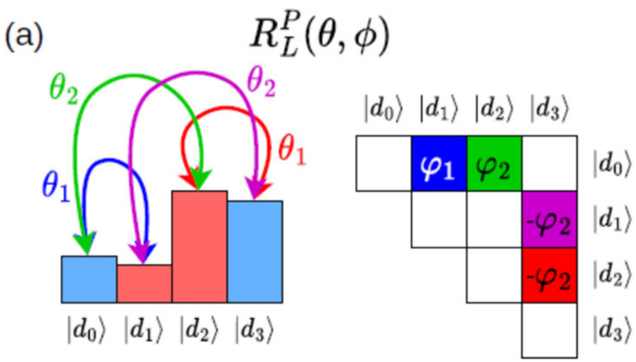

- Dephasing acts also during detection/correction
- Error detection/correction bring the qudit state out of the protected subspace

The actual **correcting power is largely reduced**

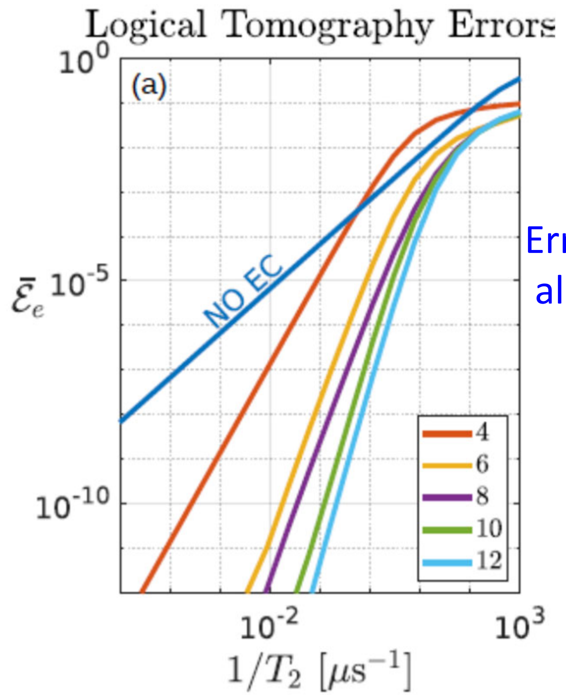
# Qudit Fault-Tolerant Quantum Computing

A methodology that allows us to **tolerate faults**, allowing QEC to remain effective while CORRECTION and quantum GATES are being performed.

We can exploit **all-all connectivity between spin levels and parallel pulses**



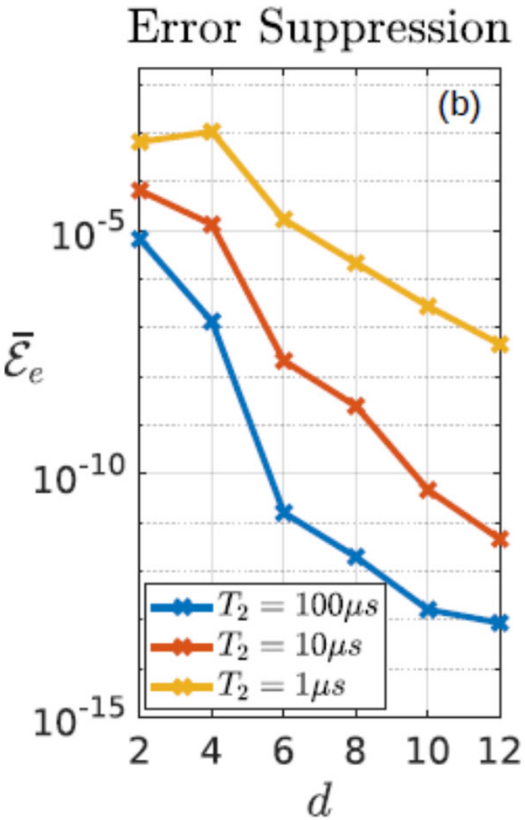

E.g., protected logical qubit gate with 4 levels




Errors acting all the time

M. Mezzadri, A. Chiesa, L. Lepori & S. Carretta, submitted, du[ly=563:143:94

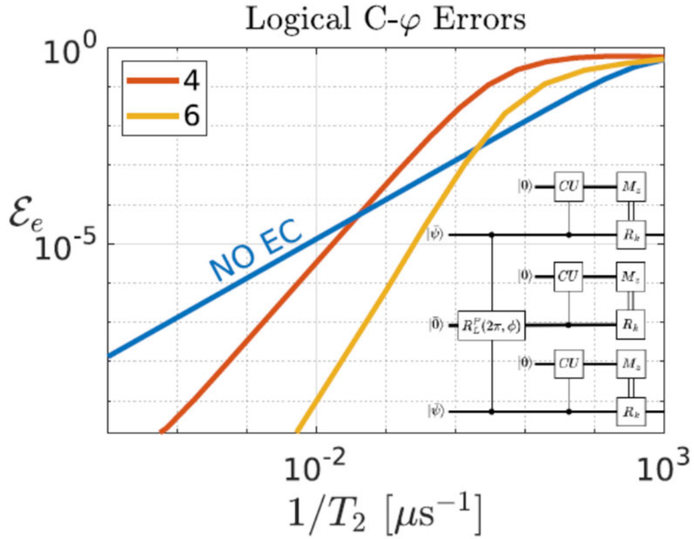
# Qudit Fault-Tolerant Quantum Computing




-Almost exponential suppression of the error with the number d of qudit levels

-Duration does not increase with d

-Strikingly error suppression, more than 5000 qubits are needed by surface codes with the same elementary error

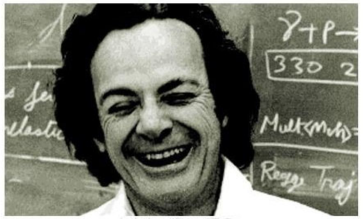



Two-qubit logical gate

M. Mezzadri, A. Chiesa, L. Lepori & S. Carretta, submitted, du[ly=563:143:94

# Quantum Simulations

The simulation of quantum systems by a classical computer is intrinsically inefficient, because the required number of bits and operations grow exponentially with the system size.



***"I think I can safely say that nobody understands quantum mechanics."***

**QUANTUM SIMULATORS**: encode the information in a hardware which operates according to quantum mechanics and whose dynamics can be controlled to mimic the evolution of the target system.

$$|\phi(t=0)> \overset{\hat{U}(t)}{\rightarrow} |\phi(t)> \; = \; ?$$

**Target quantum system (e.g., interacting fermions on lattice)**

**Suitable mapping**

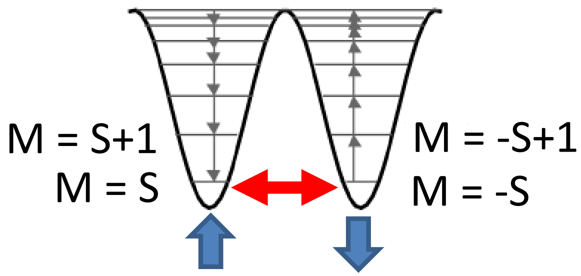
$$|\psi(t=0)> \overset{\hat{U}'(t)}{\rightarrow} |\psi(t)>$$

**Controllable quantum system (e.g., MNMs under pulsed fields)**

# Quantum Simulation of Tunneling in S = 1

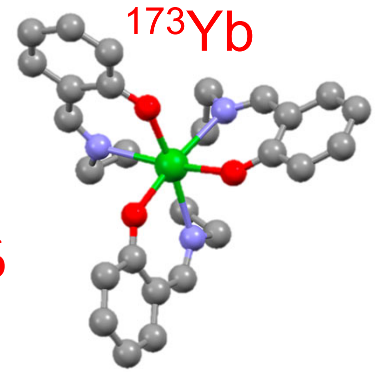
We map the target system (with S = 1) into 3 levels of a **nuclear spin qudit** **Yb(trensal)**

$$H = DS_z^2 + E(S_x^2 - S_y^2)$$



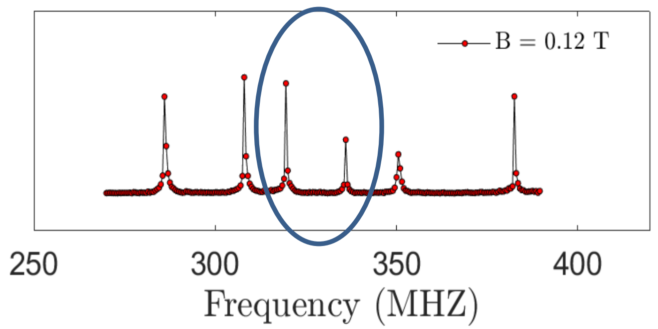

$$S = \frac{1}{2}$$

$$I = \frac{5}{2} \rightarrow d = 6$$




J. Am. Chem. Soc. 2018, 140, 9814

We implement a quantum simulation sequence using
2 different driving radio-frequencies

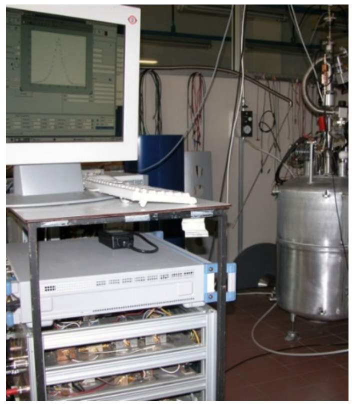



$\rightarrow |1\rangle$

$\rightarrow |0\rangle$

$\rightarrow |-1\rangle$

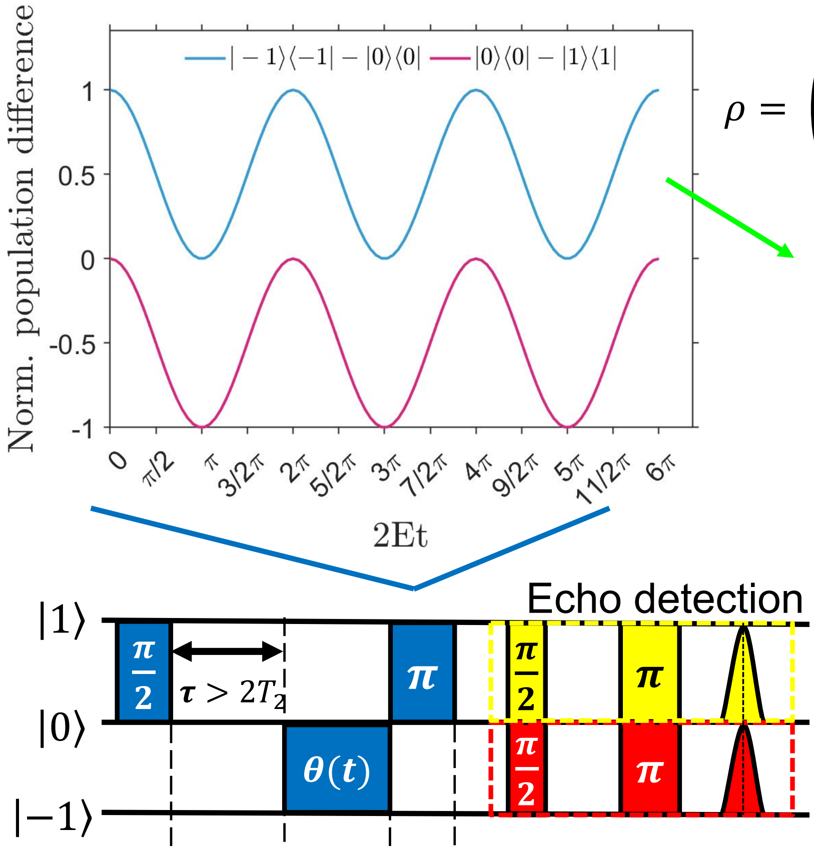
$B_x = 0.12\ T \quad T = 1.4\ K$



"HyReSpect" NMR spectrometer
@UNIPR
*Rev. Sci. Instrum.* **2005***,* 76, 083911

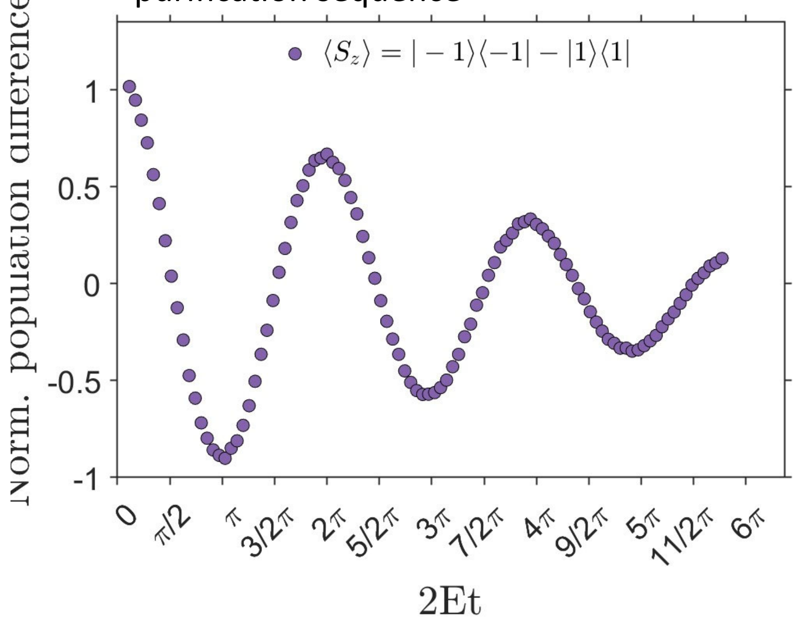
# Quantum Simulation of Tunneling in S=1

Initial state of the molecular qudit: pseudo-pure states




$$\rho = \begin{pmatrix} 1/3 + 2\varepsilon & 0 & 0 \\ 0 & 1/3 - \varepsilon & 0 \\ 0 & 0 & 1/3 - \varepsilon \end{pmatrix} = c\mathbb{I} + \begin{pmatrix} 3\epsilon & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Rabi oscillations cannot be driven between the two equipopulated states after the purification sequence



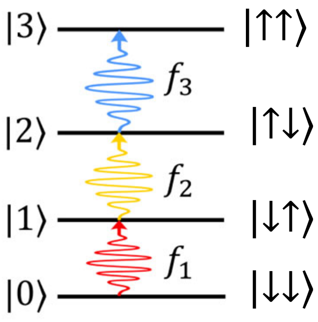

S. Chicco, G. Allodi, A. Chiesa, E. Garlatti, C. Buch, P. Santini, R. De Renzi, S. Piligkos, S. Carretta, **J. Am. Chem. Soc.** 2023.

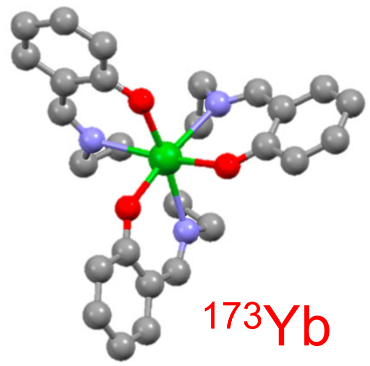
# Quantum Simulation of Transverse-field Ising model

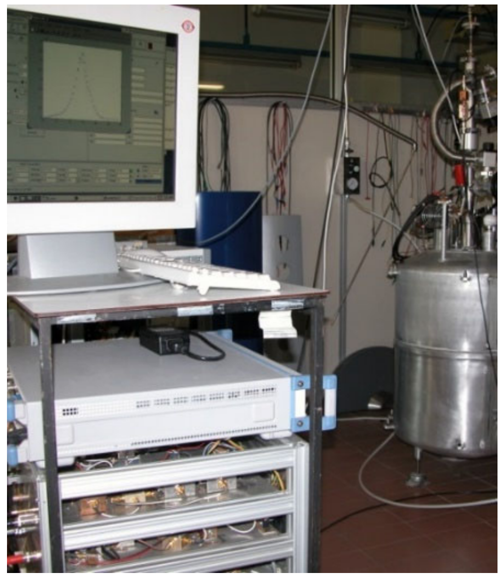
A multi-spin system can be encoded into a single-qudit to **reduce the number of error-prone two-body gates**

$$\mathcal{H} = b\left(s_{y1} + s_{y2}\right) + J s_{z1} s_{z2}$$

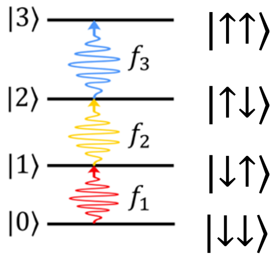
We map the target system (two spin 1/2) into **4 levels** of the **Yb(trensal) qudit**



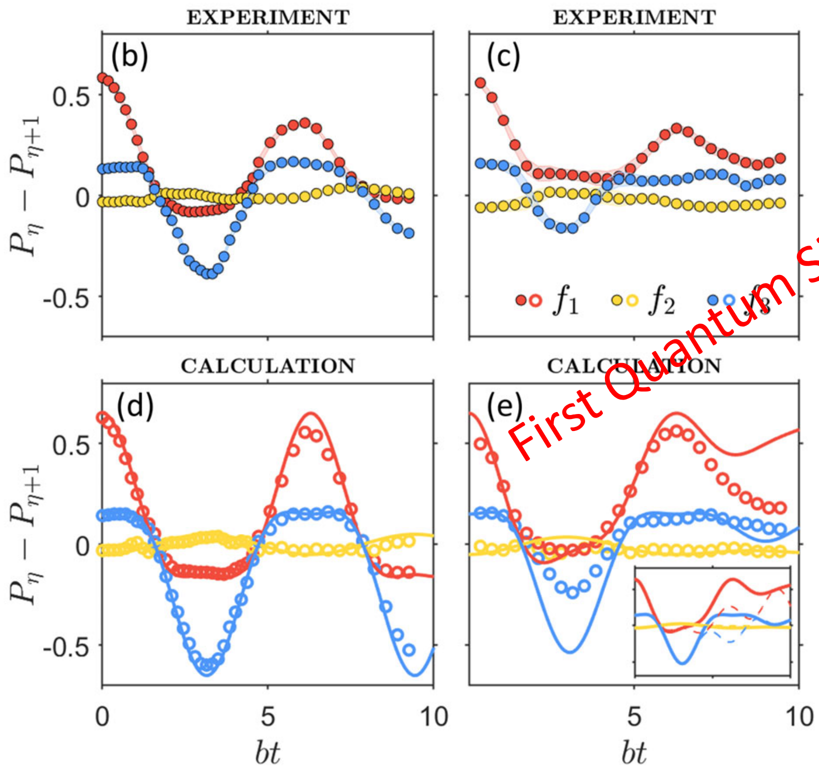



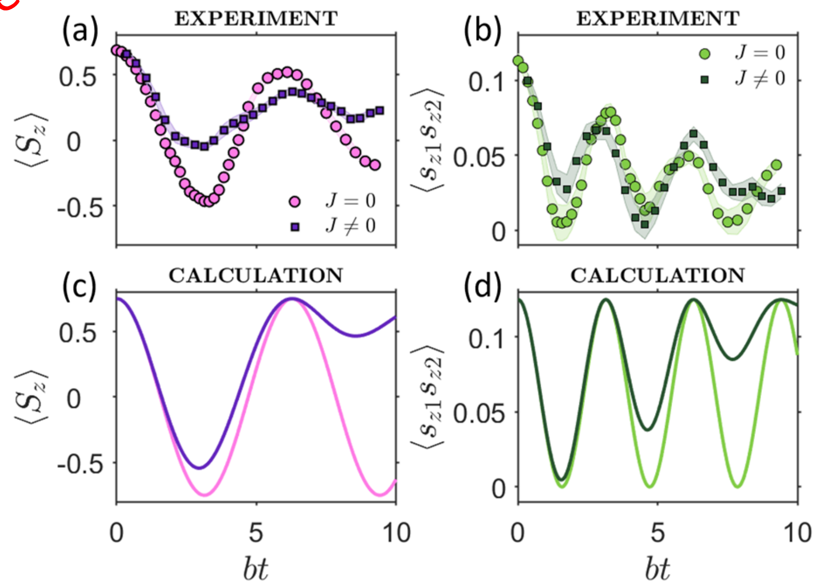
# Quantum Simulation of Transverse-field Ising model




$$U = e^{-i\mathcal{H}t} \approx \left(e^{-\frac{ib(s_{y1}+s_{y2})t}{n}} e^{-\frac{iJs_{z1}s_{z2}t}{n}}\right)^n$$

First Quantum Simulations with Molecular Nanomagnets




S. Chicco, G. Allodi, A. Chiesa, E. Garlatti, C. Buch, P. Santini, R. De Renzi, S. Piligkos, S. Carretta, **J. Am. Chem. Soc.** 2023.

# Scalability: the Molecular spin Quantum Processor

Scalable setup to: -perform gates on individual molecular qudits
-read-out the state of individual qudits

PHYSICAL REVIEW APPLIED **19**, 064060 (2023)

Editors' Suggestion    Featured in Physics

**Blueprint for a Molecular-Spin Quantum Processor**

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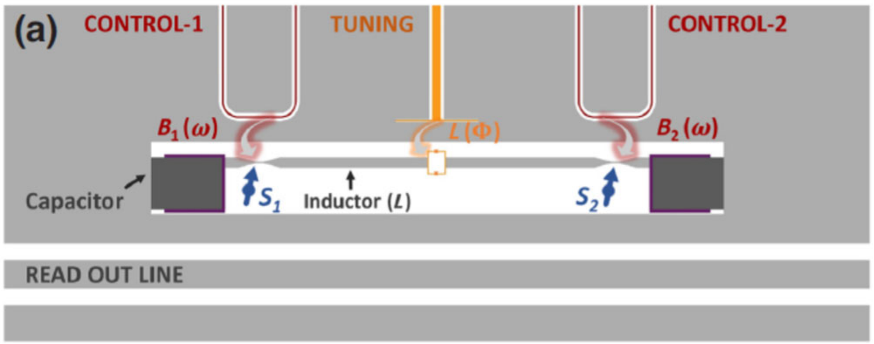
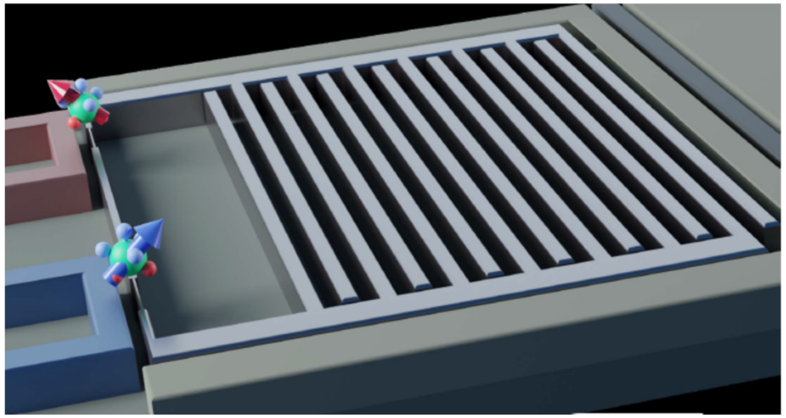
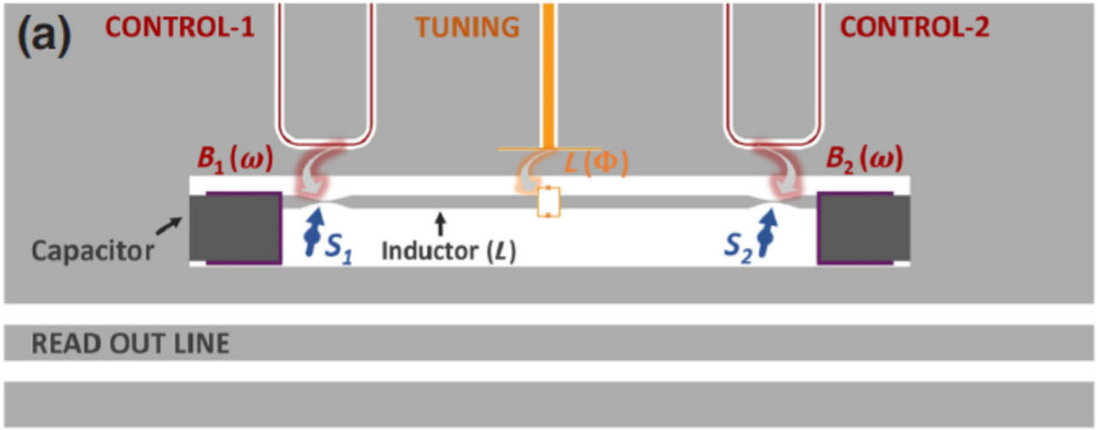
D. Zueco,[4,5] F. Luis,[4,5,*] and S. Carretta[1,2,3,†]



Superconducting resonators with individual MNMs on constrictions (**largely enhanced coupling**) + auxiliary wave guides. The resonator reads out the spin states and mediates an effective interaction between the qudits.

# Scalability: the Magnetic Quantum Processor




$$H_S = \mu_B B \sum_{i=1,2} g_i S_{zi} + \sum_{i=1,2} D_i S_{zi}^2$$

Single-qudit gates

$$H_1(t) = B_1 \theta(|t - t_0| - \tau) \mu_B \cos(\omega t + \phi) \, (g_1 S_{y1} + g_2 S_{y2})$$

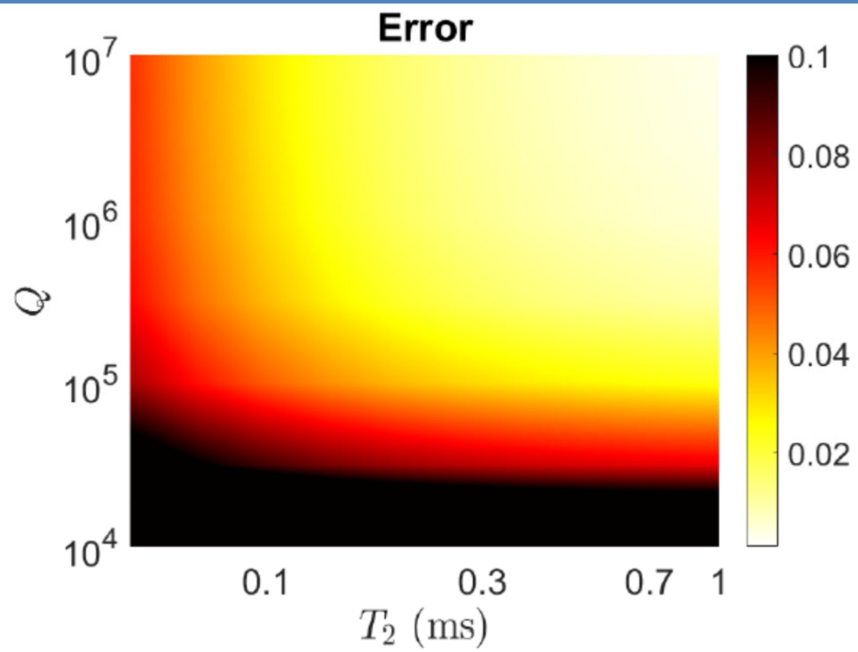
$$H_p = \hbar \omega_r(t) \left(a^\dagger a + 1/2\right)$$

$$H_{Sp} = \sum_{i=1,2} 2G_i (a + a^\dagger) S_{xi}$$

Switchable coupling between distant molecules through **exchange of photons -> two-qudit gates**

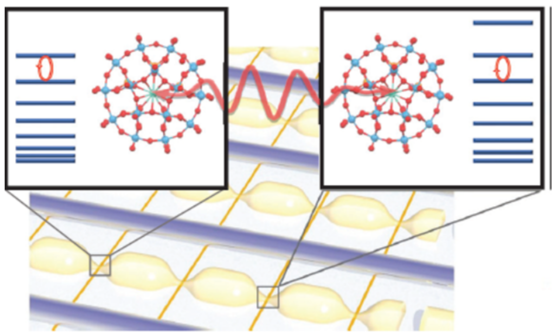
Chiesa, Roca, Chicco, de Ory, Gomez-Leon, Gomez, Zueco, Luis, Carretta, **Phys. Rev. Applied** 19, 064060 (2023).

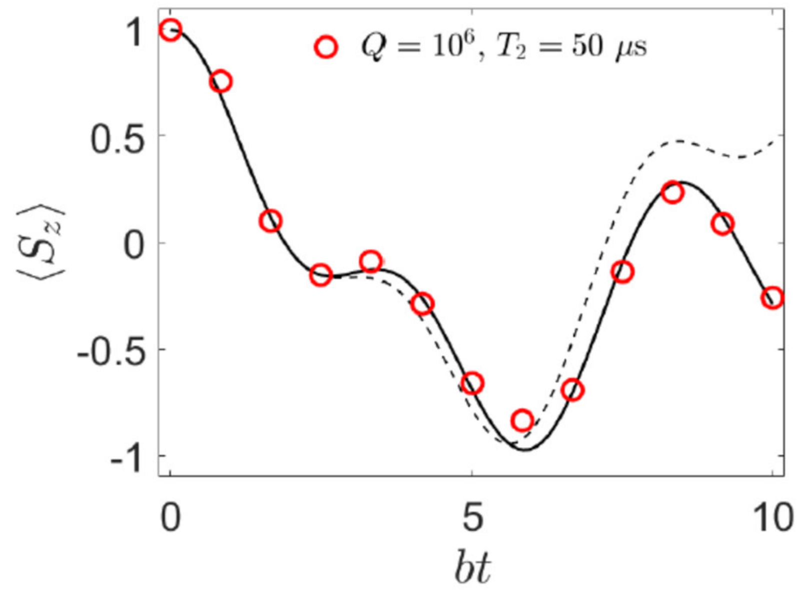
# Scalability: the Molecular spin Quantum Processor




Realistic design of resonators

Two-qudit controlled phase gate






Quantum Simulation of spin models

Chiesa, Roca, Chicco, de Ory, Gomez-Leon, Gomez, Zueco, Luis, Carretta, **Phys. Rev. Applied** 19, 064060 (2023).

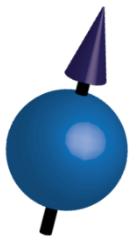
S. Carretta, D. Zueco, A. Chiesa, A. Gomez-Leon, F. Luis, **Appl. Phys. Lett.** 118, 240501 (2021).

# Chiral-Induced Spin Selectivity: a new tool

Interaction of molecular spins with magnetic (and electric) fields is weak

- Very low temperatures
- Difficult to perform a fast read out of single molecules

## Possible solution: Chiral-Induced Spin Selectivity

Spin  +  Chirality

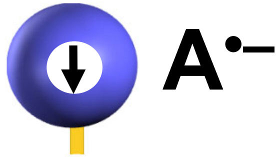
✓ Works at room T

X Not really understood

A. Chiesa, A. Privitera, E. Macaluso, M. Mannini, R. Bittl, R. Naaman, M. R. Wasielewski, R. Sessoli, S. Carretta, **Adv. Mater.** 2300472 (2023).

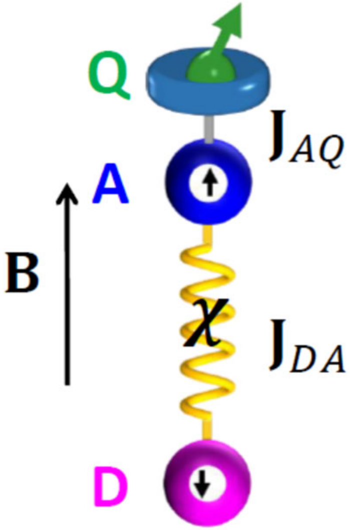
# Chiral-Induced Spin Selectivity in electron transfer

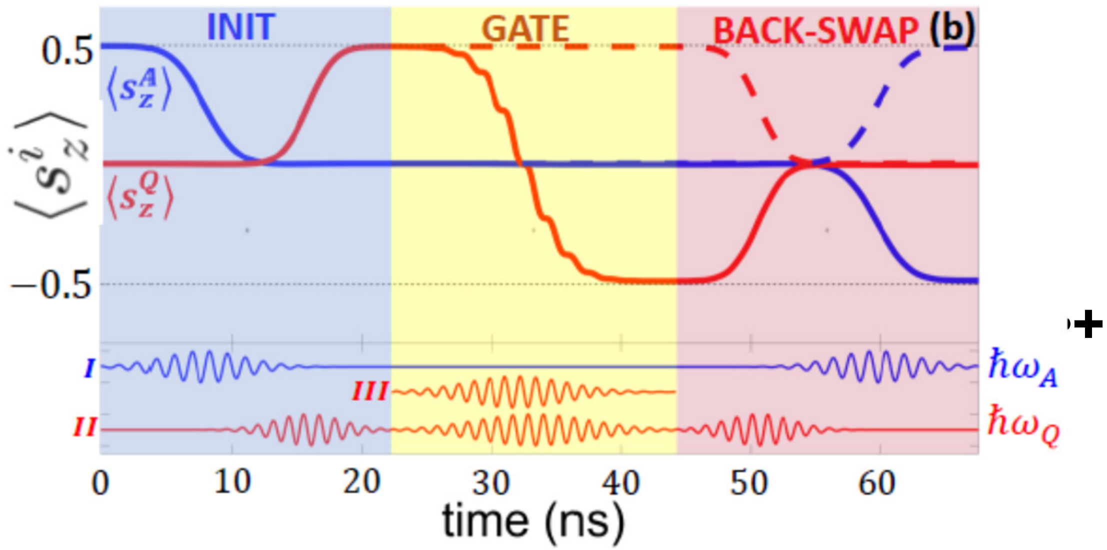
Symplify the setup to distentangle the various ingredients:




A molecular Donor-Chiral Bridge-Acceptor system

By adding a qubit

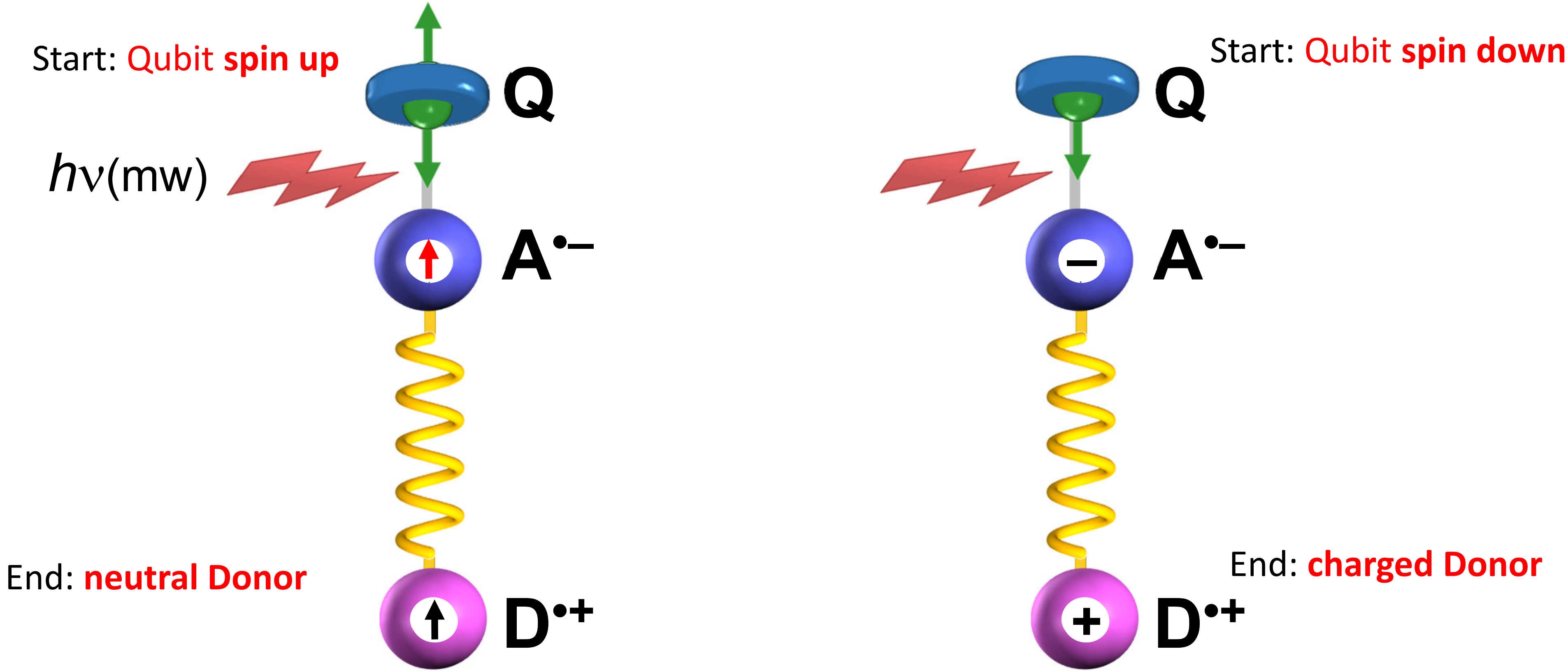



- High-temperature inizialization of the qubit
- Enables also two-qubit gates


A. Chiesa, A. Privitera, E. Macaluso, M. Mannini, R. Bittl, R. Naaman, M. R. Wasielewski, R. Sessoli, S. Carretta, **Adv. Mater.** 2300472 (2023).

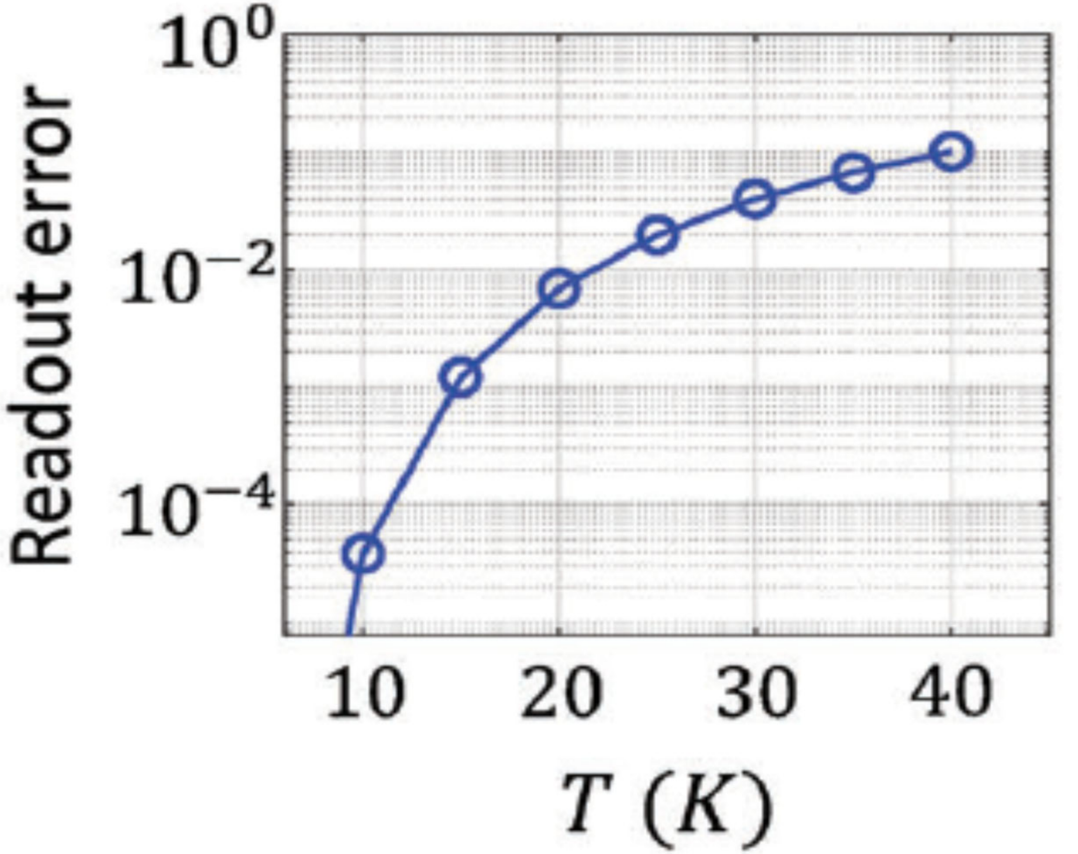
# Spin to charge conversion: readout




The spin information has been transduced into a charge information, it can be readout by a single-electron transistor

A. Chiesa, A. Privitera, E. Macaluso, M. Mannini, R. Bittl, R. Naaman, M. R. Wasielewski, R. Sessoli, S. Carretta, **Adv. Mater.** 2300472 (2023).

# Spin to charge conversion: readout



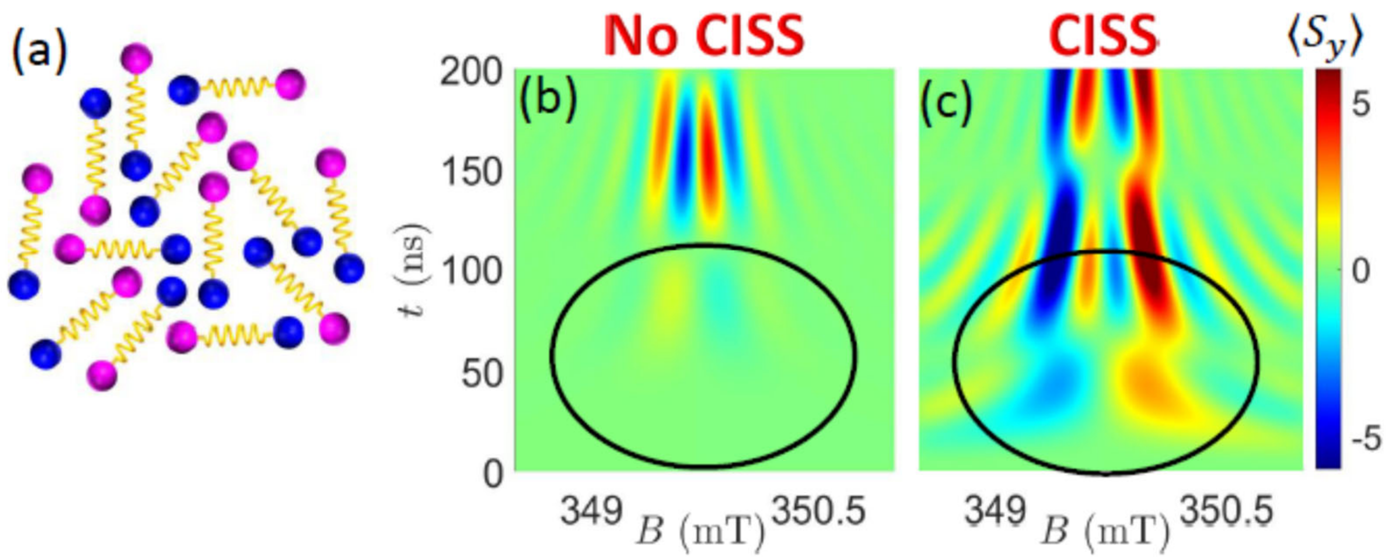


A. Chiesa, A. Privitera, E. Macaluso, M. Mannini, R. Bittl, R. Naaman, M. R. Wasielewski, R. Sessoli, S. Carretta, **Adv. Mater.** 2300472 (2023).

# Chiral-Induced Spin Selectivity in electron transfer?

The nature and the dynamics of spin states after ET can be investigated by Transient EPR

In the presence of an anisotropic dipolar DA interaction, characteristic features of CISS are already present in the spectrum of an isotropic solution

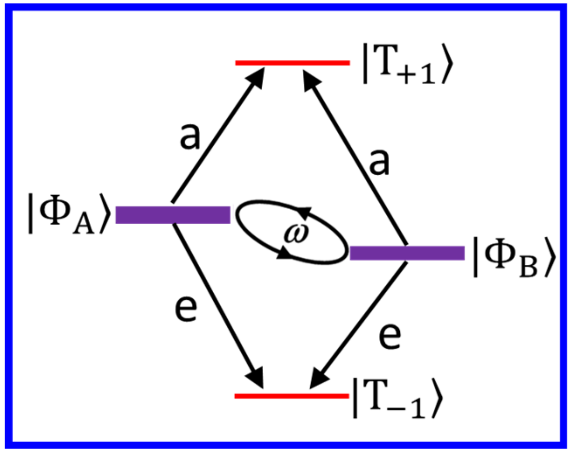




A. Chiesa, M. Chizzini, E. Garlatti, E. Salvadori, F. Tacchino, P. Santini, I. Tavernelli, R. Bittl, M. Chiesa, R. Sessoli, S. Carretta, **J. Phys. Chem. Lett.** 12, 6341 (2021)

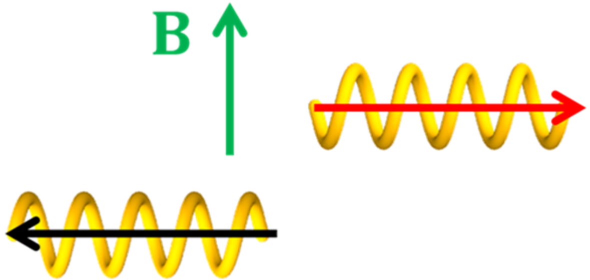
A. Privitera, E. Macaluso, A. Chiesa, A. Gabbani, D. Faccio, D. Giuri, M. Briganti, N. Giaconi, F. Santanni, N. Jarmouni, L. Poggini, M. Mannini, M. Chiesa, C. Tomasini, F. Pineider, E. Salvadori, S. Carretta, R. Sessoli, **Chem. Sci.** 13, 12208 (2022).

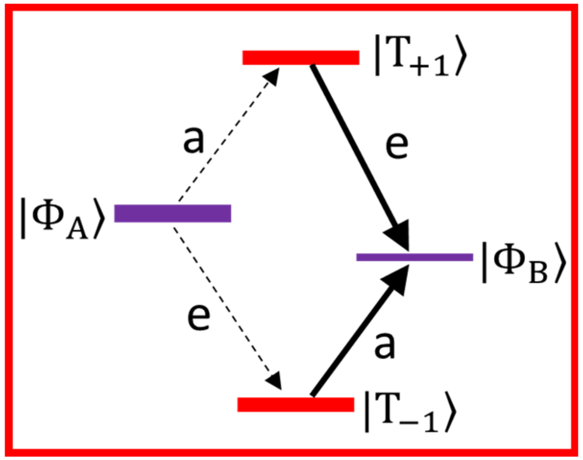
# Intramolecular CISS in EPR




SINGLET

$$\theta = 90°$$




100 % CISS

# CISS in electron transfer!

RESEARCH

Science

PHYSICAL CHEMISTRY

## Direct observation of chirality-induced spin selectivity in electron donor–acceptor molecules

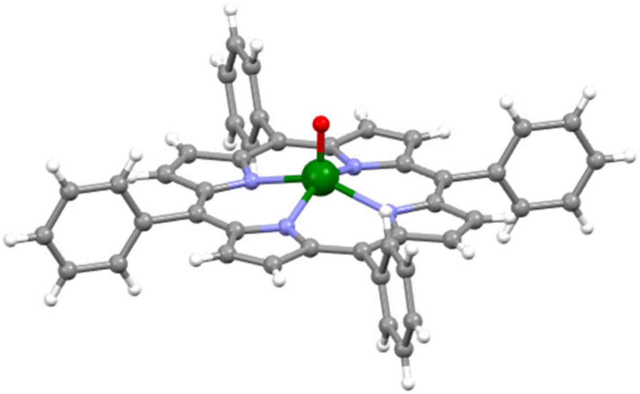
Hannah J. Eckvahl[1]†, Nikolai A. Tcyrulnikov[1]†, Alessandro Chiesa[2]†, Jillian M. Bradley[1], Ryan M. Young[1], Stefano Carretta[2]*, Matthew D. Krzyaniak[1]*, Michael R. Wasielewski[1]*

# Unveiling phonons in molecular qubits with IXS

Main source of relaxation in MNMs: → **PHONONS**

**Inelastic X-ray Scattering**

**High-resolution beamline → ID28 @ ESRF**



**[VO(TPP)]**

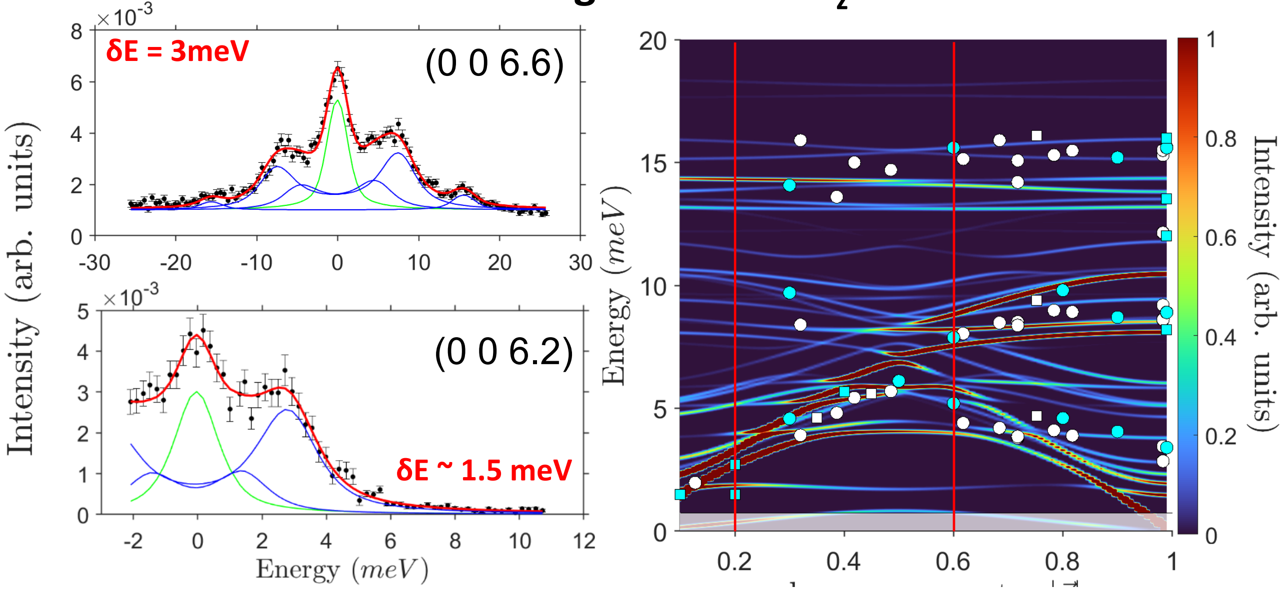
**First IXS measurements of phonons in a molecular qubit**

E. Garlatti, A. Albino, S. Chicco, V.H.A. Nguyen, F. Santanni, L. Paolasini, C. Mazzoli, R. Caciuffo, F. Totti, P. Santini, R. Sessoli, A. Lunghi, S. Carretta, **Nat. Comm.** 14, 1653 (2023).

# Unveiling phonons in molecular qubits with IXS

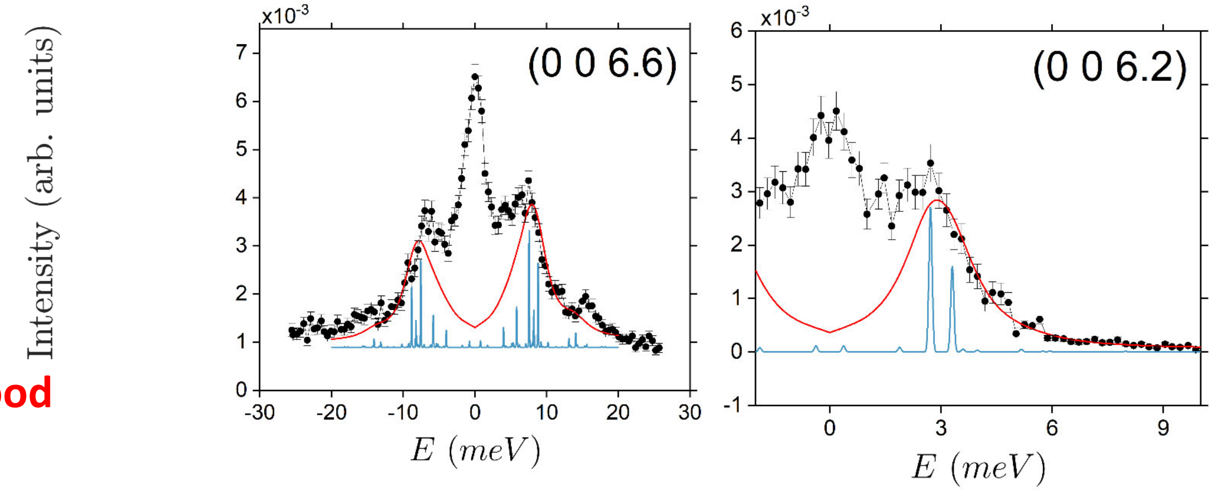
**Longitudinal Γ - $K_z$**

**ID28 DATA**



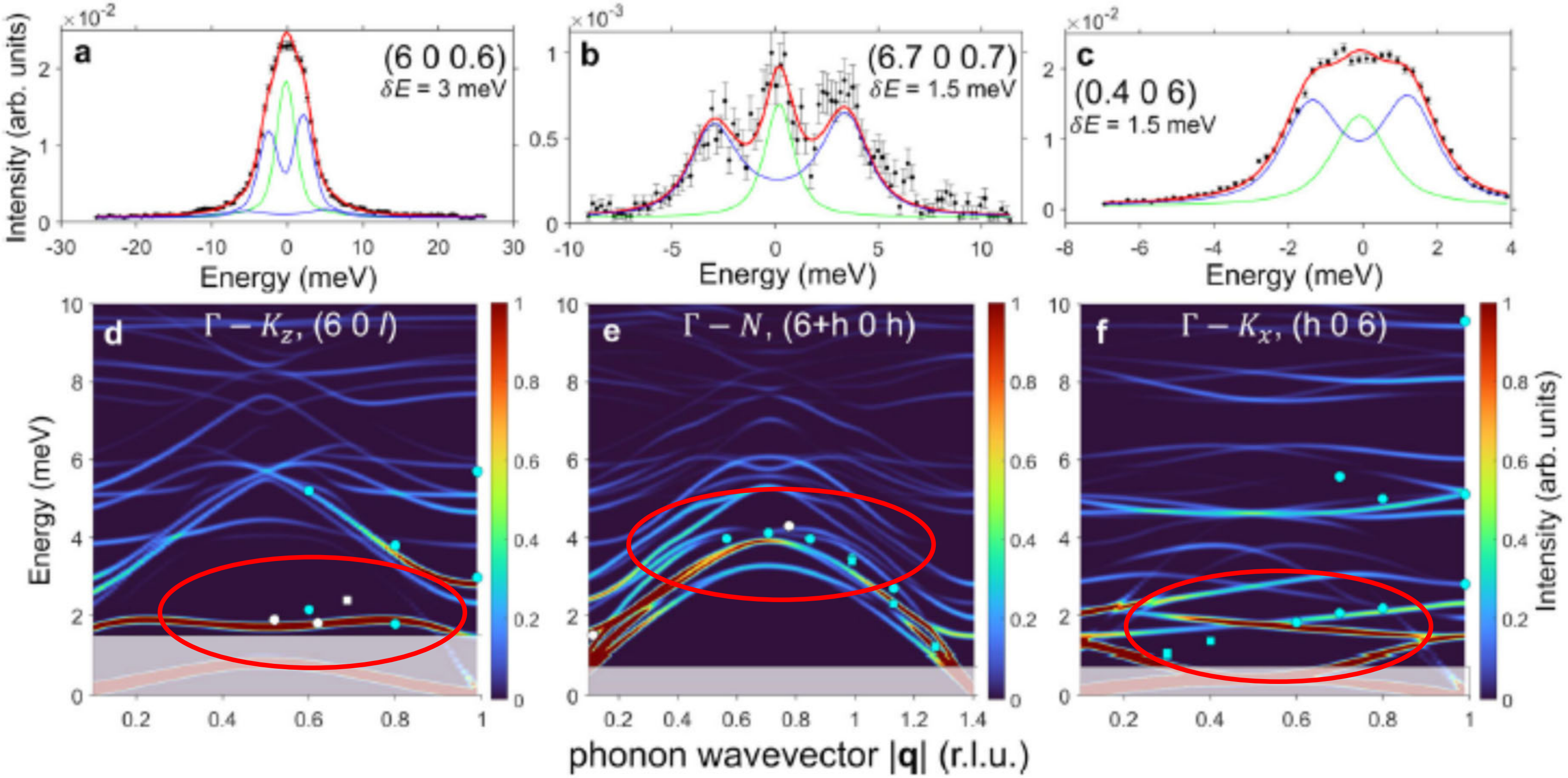

**DFT**
**PBE +**
**Grimme's D3 vdW**

Simulation of the IXS cross-section starting from DFT-calculated phonon energies and eigenvectors




**DFT calculations are in very good agreement**

# The critical role of ultra-low-energy vibrations



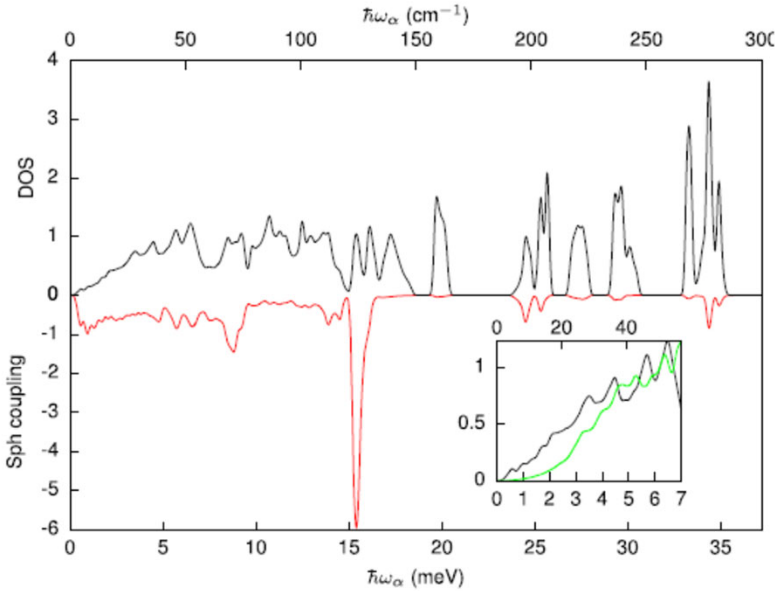

**We have found** **very-low energy optical phonon modes**

→ **they can deeply** **affect relaxation.**


E. Garlatti, A. Albino, S. Chicco, V.H.A. Nguyen, F. Santanni, L. Paolasini, C. Mazzoli, R. Caciuffo, F. Totti, P. Santini, R. Sessoli, A. Lunghi, S. Carretta, **Nat. Comm**. 14, 1653 (2023).

# The critical role of ultra-low-energy vibrations

- IXS data validated DFT → **calculations of spin-phonon couplings**
- Resource- and time-consuming → **neural networks approach**

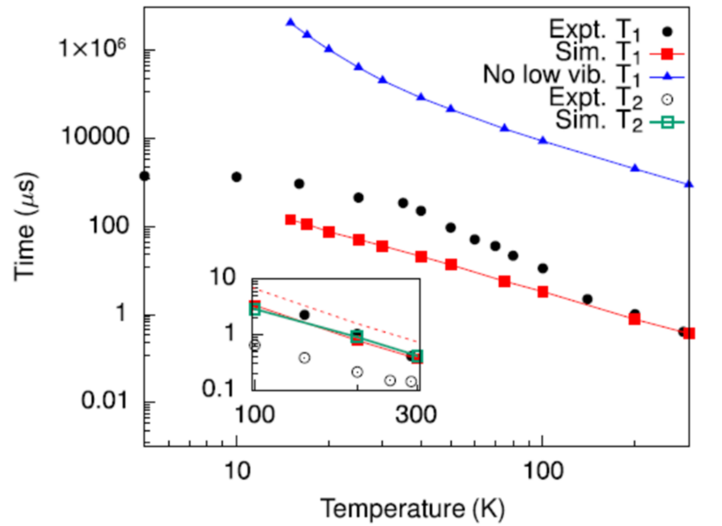



phonon-induced modulation of the g-tensor and of the hyperfine coupling constant A

**A. Lunghi & co-workers**
Trinity College Dublin
The University of Dublin

low-energy optical phonons have very strong spin-phonon couplings and they are the main trigger of magnetic relaxation

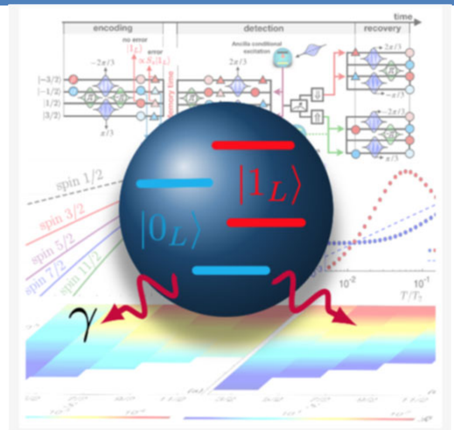
fully ab initio calculation of the spin relaxation time of [VO(TPP)]

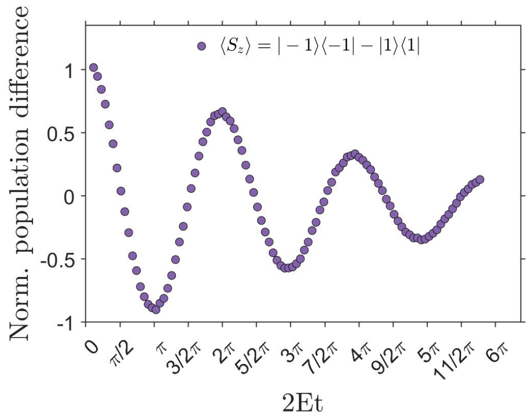



E. Garlatti, A. Albino, S. Chicco, V.H.A. Nguyen, F. Santanni, L. Paolasini, C. Mazzoli, R. Caciuffo, F. Totti, P. Santini, R. Sessoli, A. Lunghi, S. Carretta, **Nat. Comm.** 14, 1653 (2023).

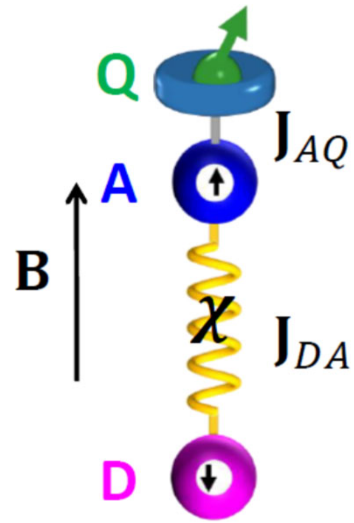
# Conclusion: MNMs are a viable path toward QIP

Molecular qudits can be used as qubits with embedded Quantum Error Correction (in a fault-tolerant manner!)

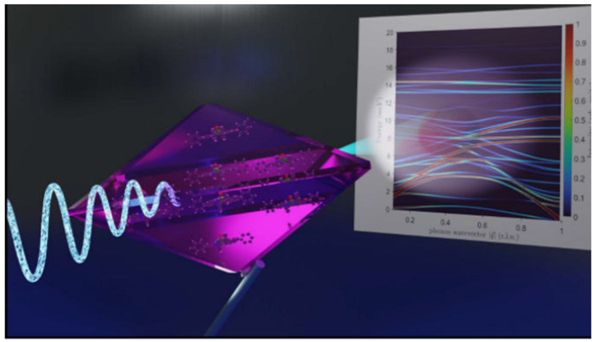



First quantum simulation experiments with MNMs




Chiral-induced spin selectivity: an enabling technology for quantum applications



Critical role of ultra-low-energy vibrations unveiled by IXS and DFT

A. Chiesa, P. Santini, E. Garlatti, F. Luis, S. Carretta, **Reports on Progress in Physics,** 87, 034501 (2024).

# Projects

CASTLe

project n. 101071533

European Research Council
Established by the European Commission

**PNRR MUR project PE0000023-NQSTI**
National Quantum Science and Technology Institute

Finanziato dall'Unione europea NextGenerationEU | Ministero dell'Università e della Ricerca | Italiadomani PIANO NAZIONALE DI RIPRESA E RESILIENZA | NQSTI National Quantum Science and Technology Institute

FATMOLS

HORIZON 2020

grant n. 862893
**FET - Open**

NUOVO BANDO PRIN
Progetti di ricerca di Rilevante Interesse Nazionale

Project2017CR5WCH Q-chiSS

SUMO QUANTERA

novo nordisk foundation

Coherent addressing of isotopically pure lanthanide complexes by photons and efficient quantum error correction for Quantum Information Technologies

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