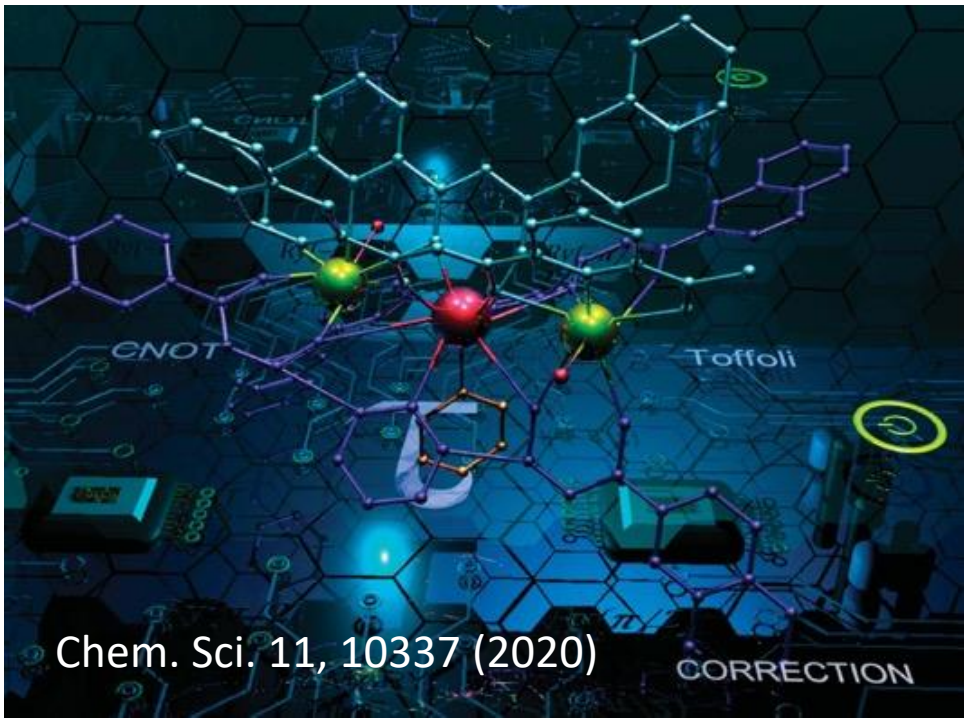
Molecular spin qudits: a promising ingredient for quantum computers

Stefano Carretta

Dipartimento di Scienze Matematiche, Fisiche e Informatiche,
Università di Parma

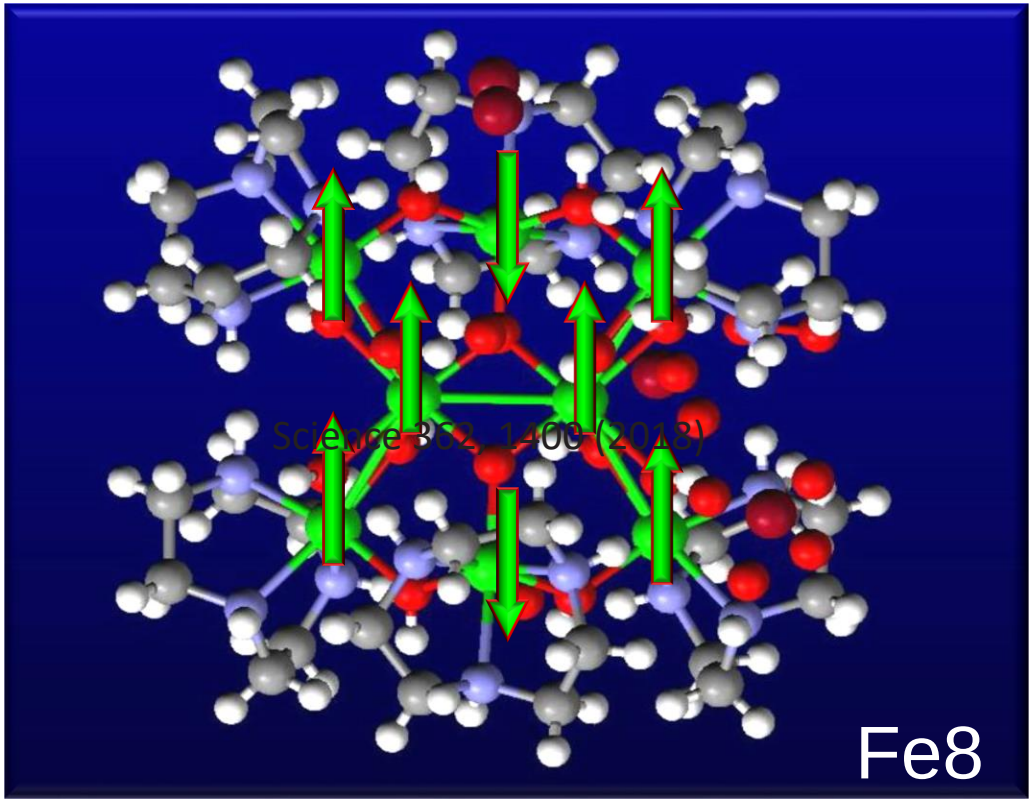


Chem. Sci. 11, 10337 (2020)

Molecular Nanomagnets

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

Organic ligands provide a **magnetic separation** between neighboring molecules and the crystal behaves as a collection of **non-interacting magnetic particles.** Single-molecule properties can be probed by bulk measurements.



Molecular Nanomagnets

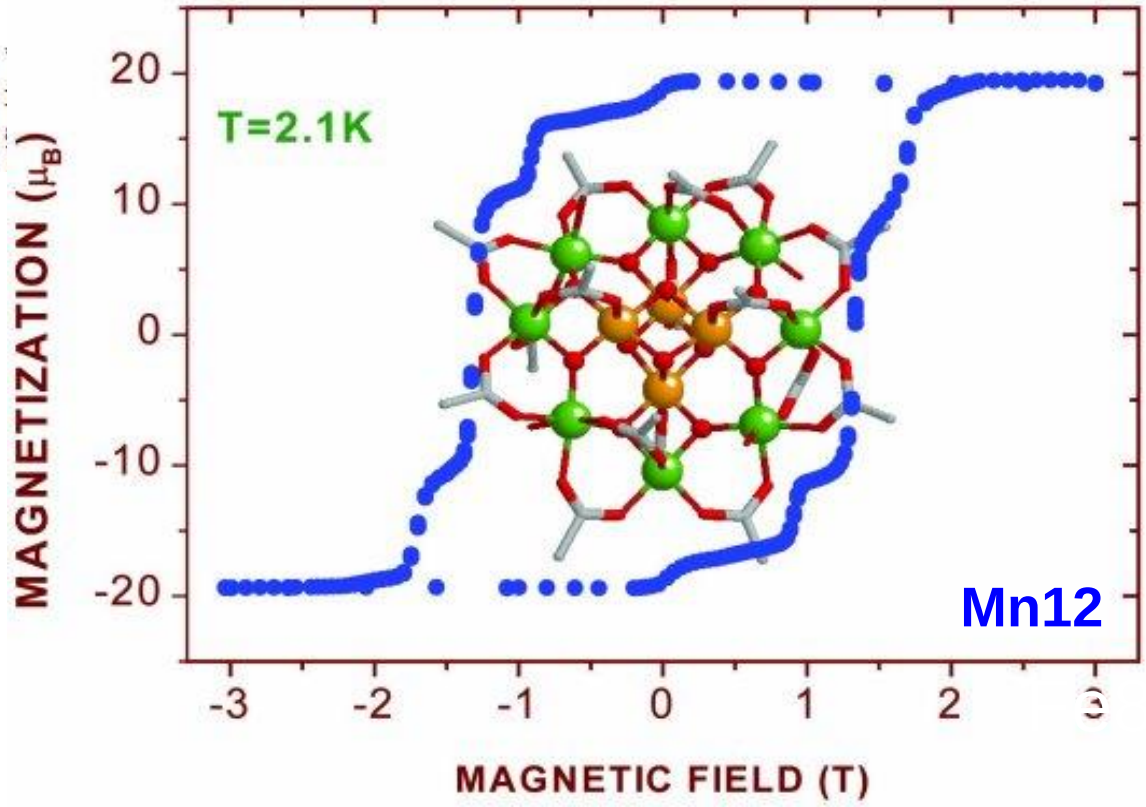
Molecules in which the magnetic core is constituted by a small number of exchange-coupled magnetic ions.

letters to nature

Nature **365**, 141 - 143 (09 September 1993); doi:10.1038/365141a0

Magnetic bistability in a metal-ion cluster

R. SESSOLI*, D. GATTESCHI*‡, A. CANESCHI* & M. A. NOVAK†§



Mn12 displays hysteresis and behaves as a tiny magnet

Possibility of storing information in single molecules

Molecular Nanomagnets

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

letters to nature

LETTER

doi:10.1038/nature23447

Molecular magnetic hysteresis at 60 kelvin in dysprosocenium

Conrad A. P. Goodwin[1], Fabrizio Ortu[1], Daniel Reta[1], Nicholas F. Chilton[1] & David P. Mills[1]

RESEARCH

MOLECULAR MAGNETS

Science 362, 1400 (2018)

Magnetic hysteresis up to 80 kelvin in a dysprosium metallocene single-molecule magnet

Fu-Sheng Guo[1], Benjamin M. Day[1,2], Yan-Cong Chen[3], Ming-Liang Tong[3]*, Akseli Mansikkamäki[4]*, Richard A. Layfield[1]*

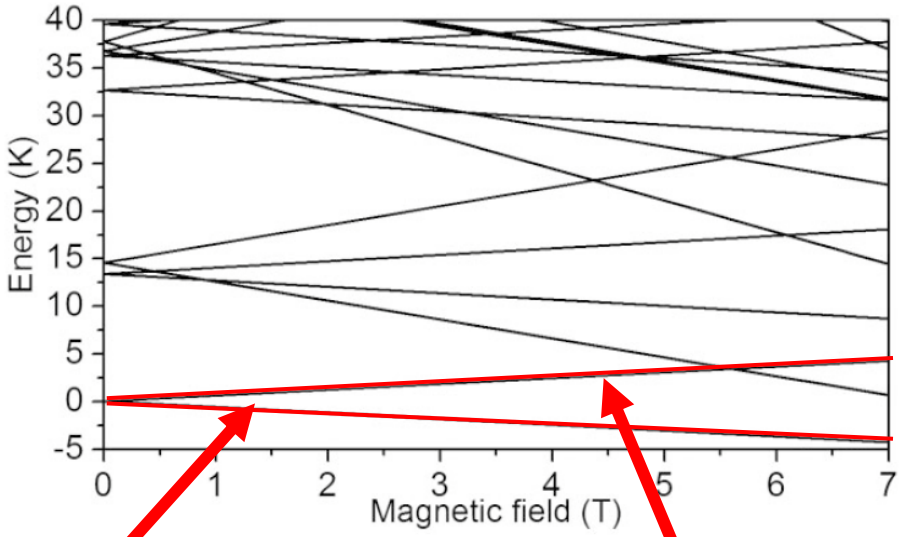
Possibility of storing information in single molecules

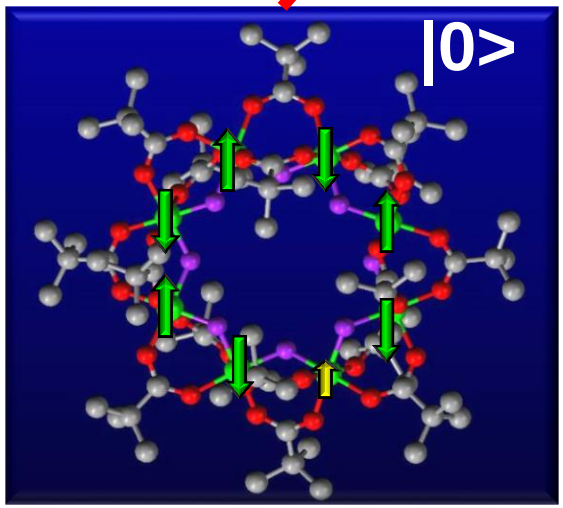
Molecular Nanomagnets

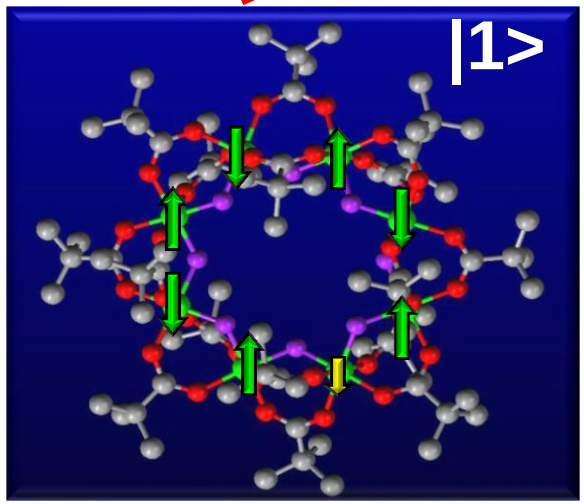
Molecular magnetic materials on the one hand show magnetic hysteresis as ordinary magnets do and, on the other hand, they are small enough to show quantum effects.

Magnetic molecules can be exploited to encodes qubits, the basic units of a quantum computer

Molecular Nanomagnets







Cr7Ni

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

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

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: chips with dozens of qubits enable to implement non trivial quantum algorithms.

nature physics

LETTERS

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

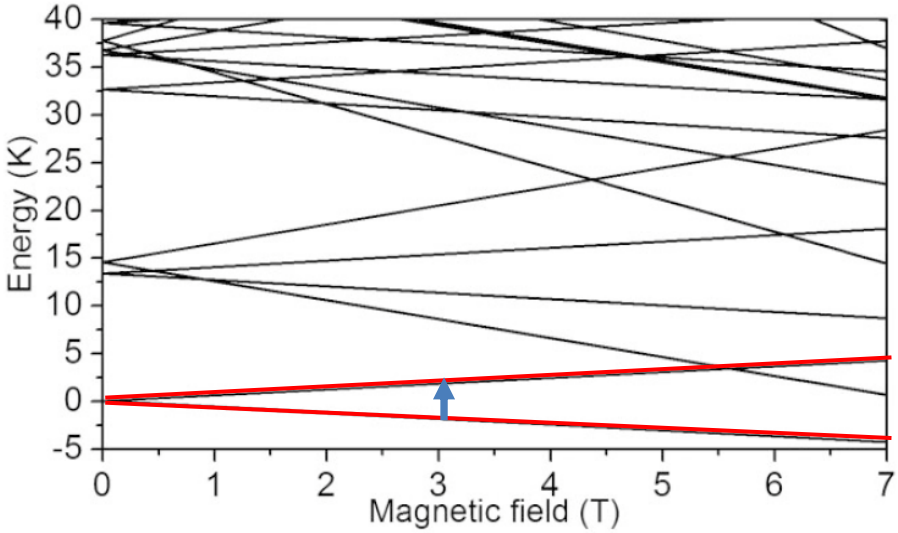
Quantum hardware simulating four-dimensional inelastic neutron scattering

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

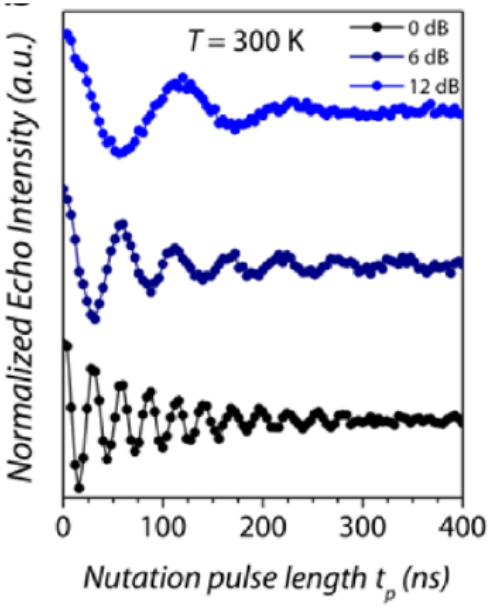
Quantum simulations of the **spin dynamics** of prototypical spin systems and calculation of the **4D inelastic neutron cross-section**

Molecular Qubits: single-qubit gates

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



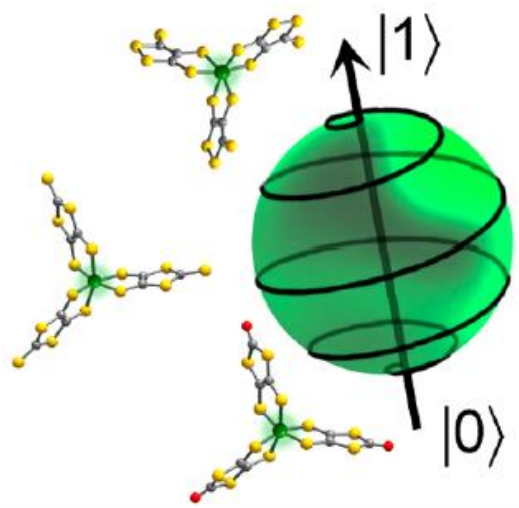
Molecular Qubits: single-qubit gates

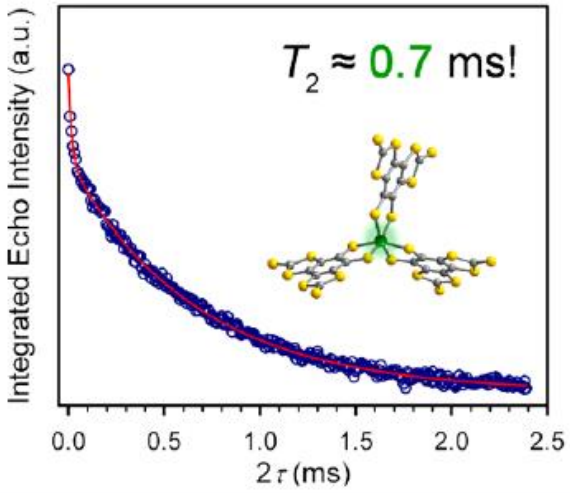


Single-qubit logical operations in molecular qubits can be produced by **resonant magnetic pulses**

Room temperature coherence in a Vanadyl Phthalocyanine

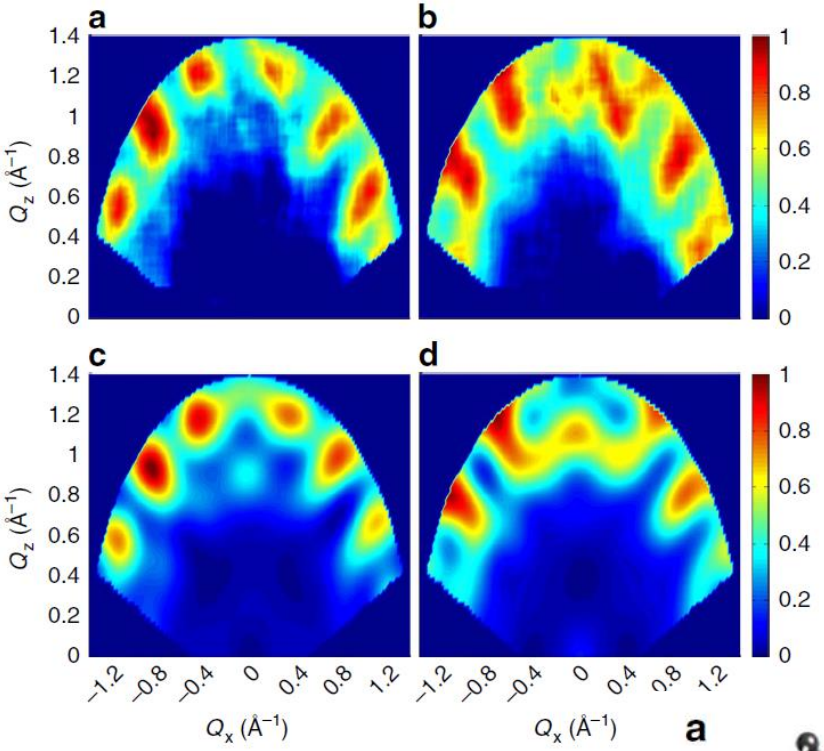
Atzori et al., J. Am. Chem. Soc. 138, 2154 (2016)





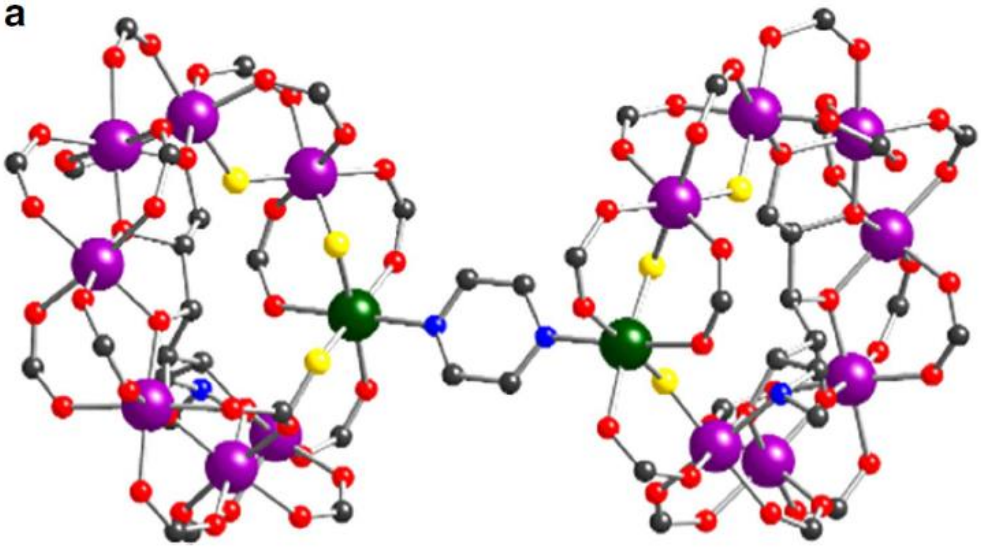
Zadrozny et al, ACS Cent. Sci. ***1****, 488 (2015)*

Molecular Qubits: two-qubits gates

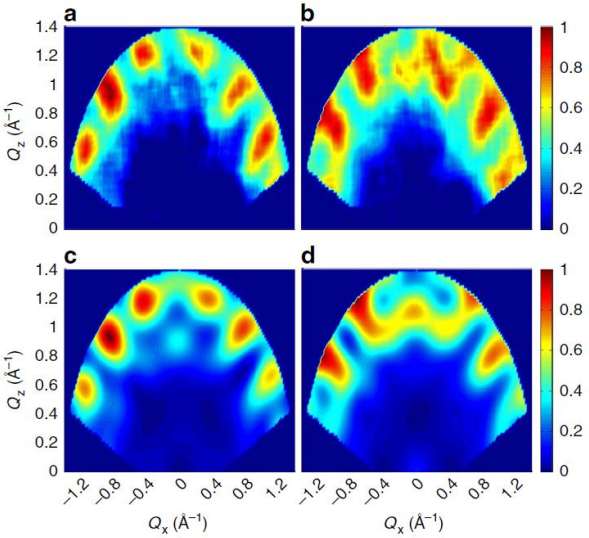


Portraying entanglement between molecular qubits

Nature Communications 8, 14543 (2017)



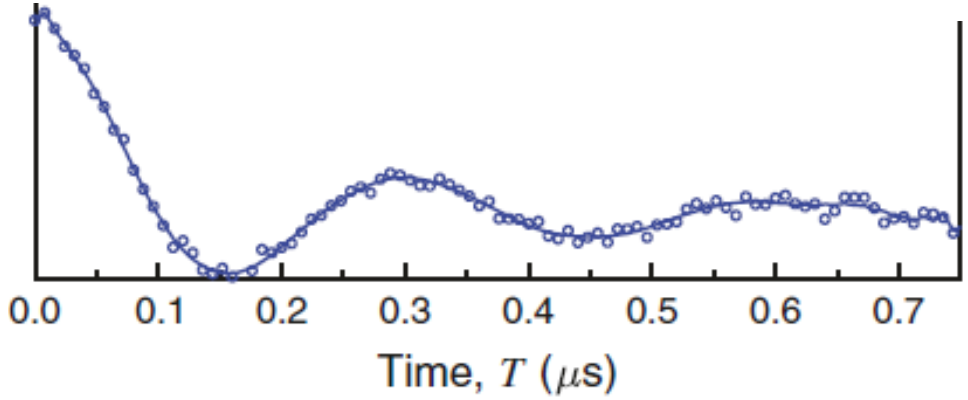
Molecular Qubits: two-qubits gates



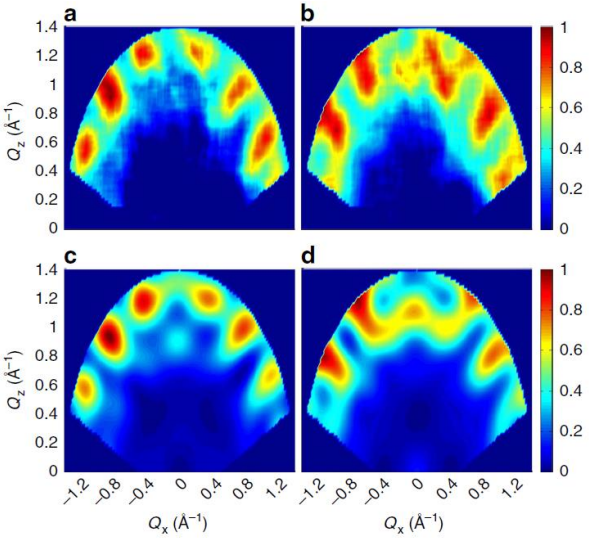
Nature Communications 8, 14543 (2017)

Two-qubit gates with permanent qubit-qubit coupling

Luis et al., PRL 107, 117203 (2011), A. Ardavan et al., npj Quantum Information 1, 15012 (2015)



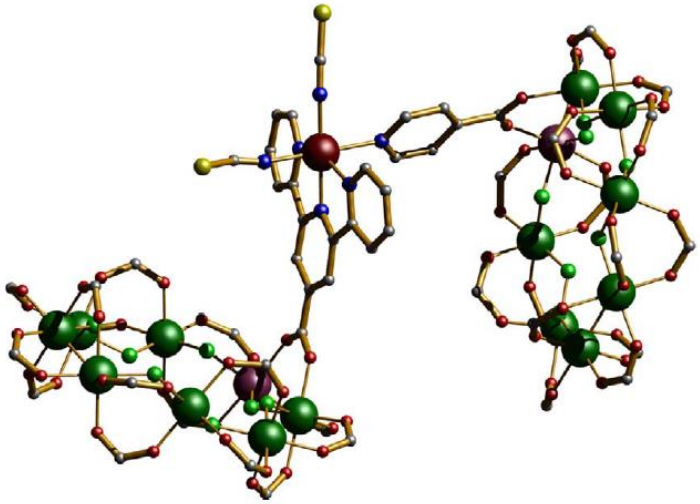
Molecular Qubits: two-qubits gates



Nature Communications 8, 14543 (2017)

Two-qubit gates with permanent qubit-qubit coupling

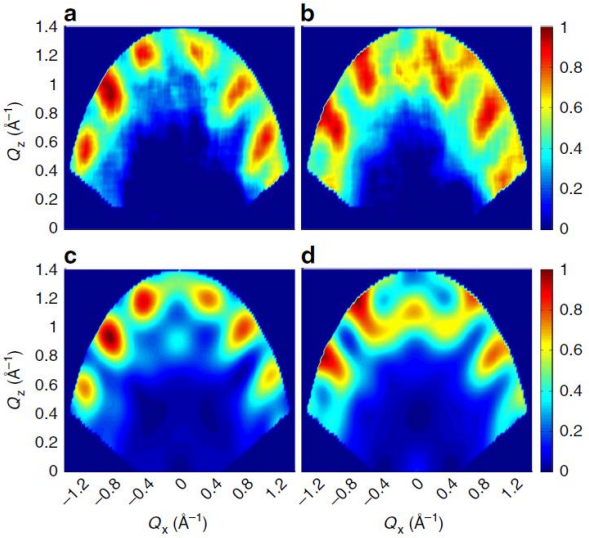
Luis et al., PRL 107, 117203 (2011), A. Ardavan et al., npj Quantum Information 1, 15012 (2015)



Switchable couplings can be obtained in supramolecular compounds

Nature Nanotechnology 4, 173 (2009).
Phys. Rev. Lett. 107, 230502 (2011).
Nature Communications 7, 11377 (2016).

Molecular Qubits: two-qubits gates

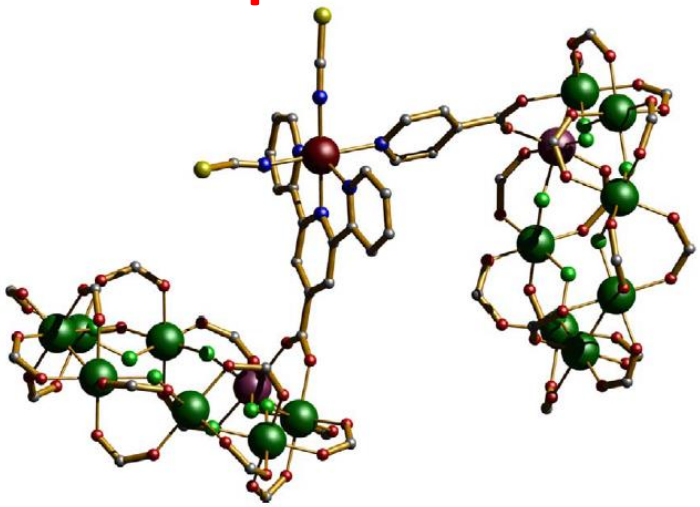


Nature Communications 8, 14543 (2017)

Two-qubit gates with permanent qubit-qubit coupling

Luis et al., PRL 107, 117203 (2011), A. Ardavan et al., npj Quantum Information 1, 15012 (2015)

Main question: **why magnetic molecules?**



Switchable couplings can be obtained in supramolecular compounds

Nature Nanotechnology 4, 173 (2009).
Phys. Rev. Lett. 107, 230502 (2011).
Nature Communications 7, 11377 (2016).

Molecular Nanomagnets as Qudits

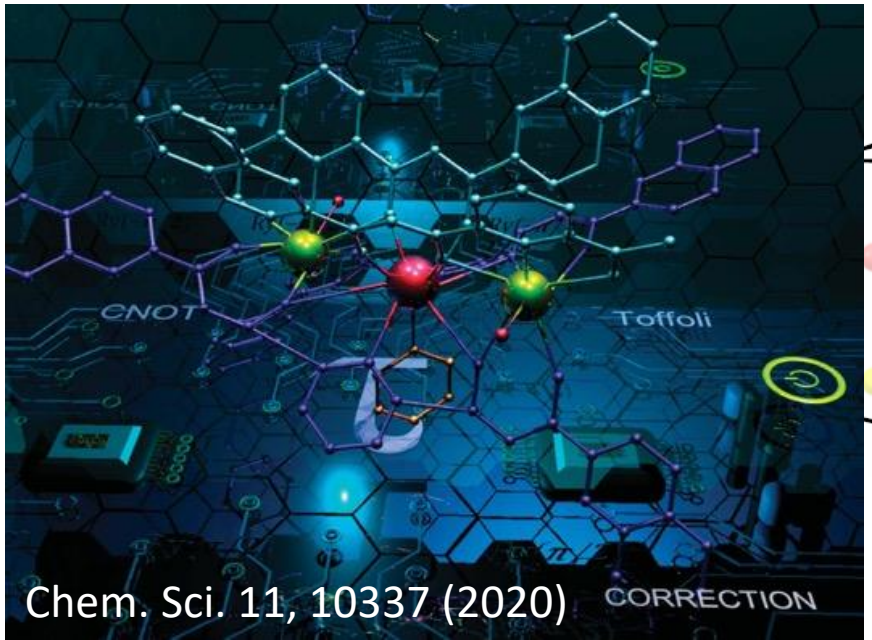
MNMs are typically characterized by a sizeable number of accessible low-energy levels

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

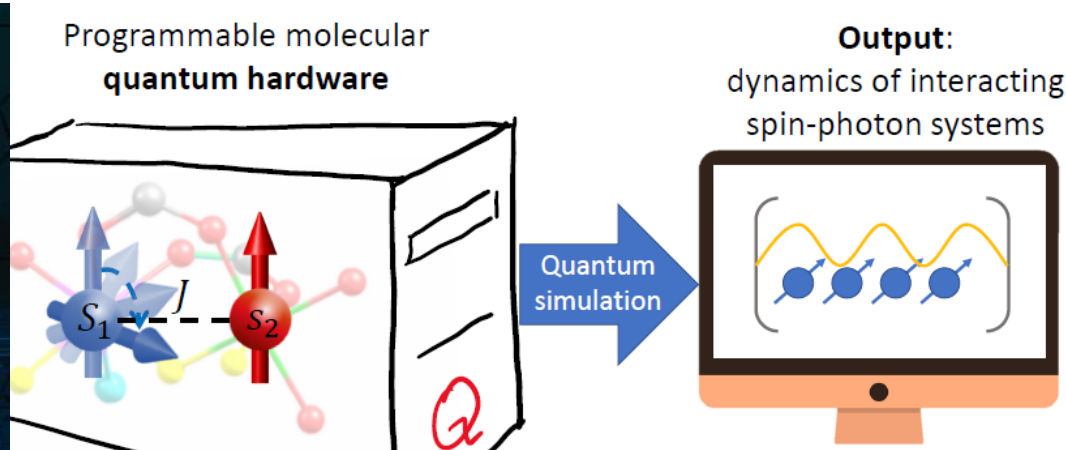
Quantum systems with d > 2 levels, called qudits, can enhance the power of quantum logic:

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

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



Chem. Sci. 11, 10337 (2020)



Quantum Error Correction

Quantum superposition states are fragile. In order to **scale up** quantum computing we **need ERROR-PROTECTED (LOGICAL) QUBITS**

"While highly demanding, **this achievement is nothing short of the equivalent to being the first nation to land on the moon**."

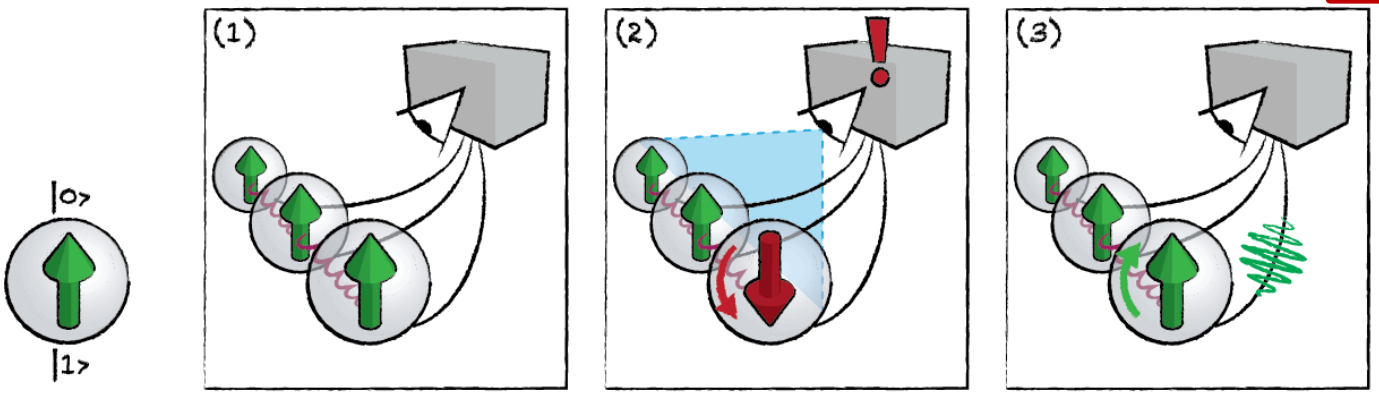
Quantum Error Correction

Block-encoding: **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

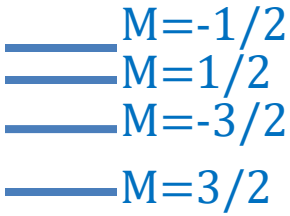
- We use only a fraction of the available space to encode the logical state. If errors bring the qubit wave-function **outside the computational basis** they can be detected and corrected.

Quantum error correction with spin S qudits

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

To first order $\rho(t) = \rho(0) + \frac{2t}{T_2} S_z \rho(0) S_z$

No error $\rho(0)$ — S_z error $S_z \rho(0) S_z$



$$|0_L\rangle = \frac{|3/2\rangle + \sqrt{3}|-1/2\rangle}{2} \longrightarrow S_z|0_L\rangle/\|S_z|0_L\rangle\| = \frac{\sqrt{3}|3/2\rangle - |-1/2\rangle}{2} \equiv |e_0\rangle$$

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

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

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

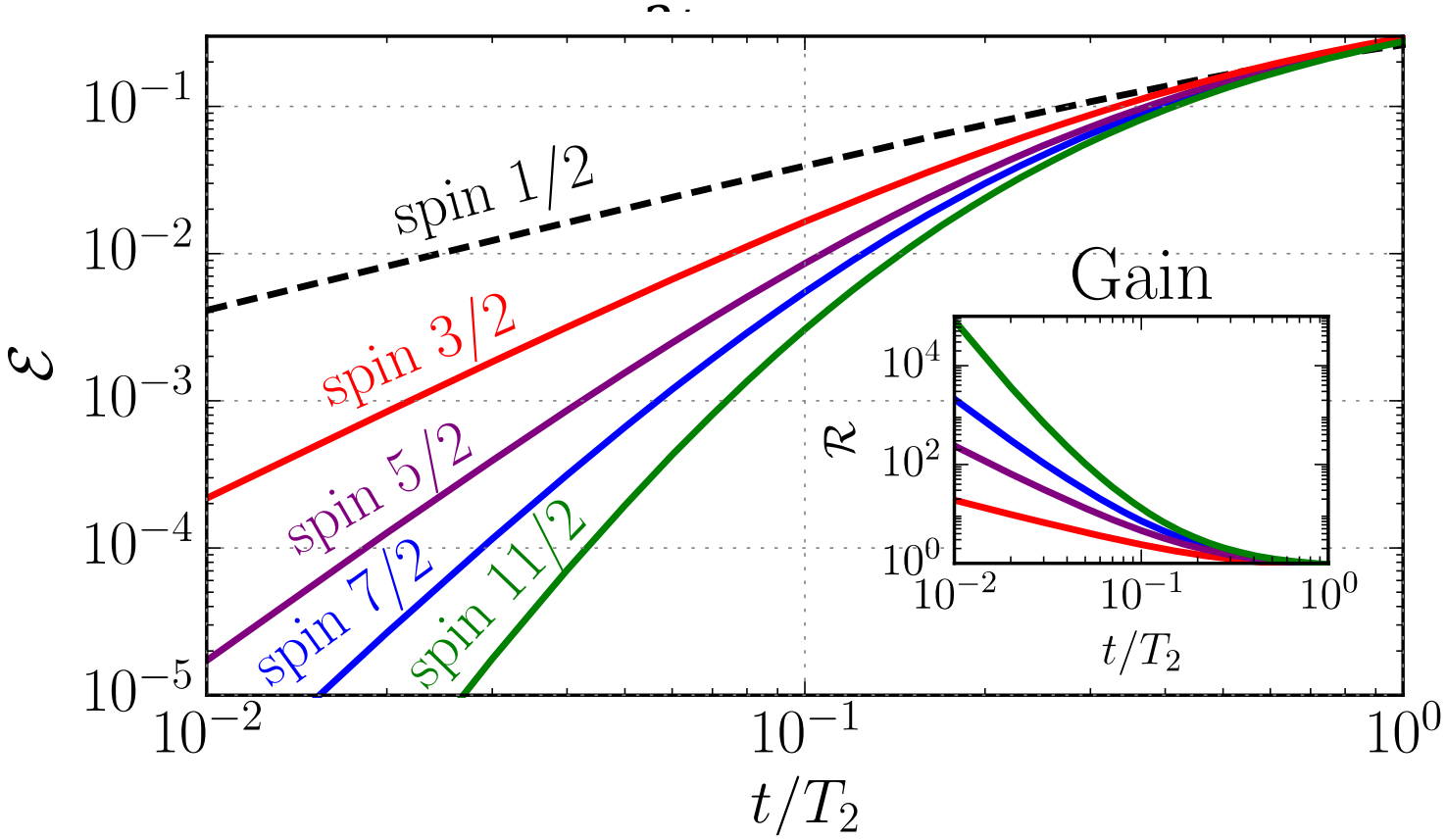
Errors are **detectable** and **correctable**

A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

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

Quantum error correction with spin S qudits

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



M=-1/2
M=1/2
M=-3/2
M=3/2

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

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

s $|0_L\rangle$ and $|1_L\rangle$ to
e states

he superposition

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

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

A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

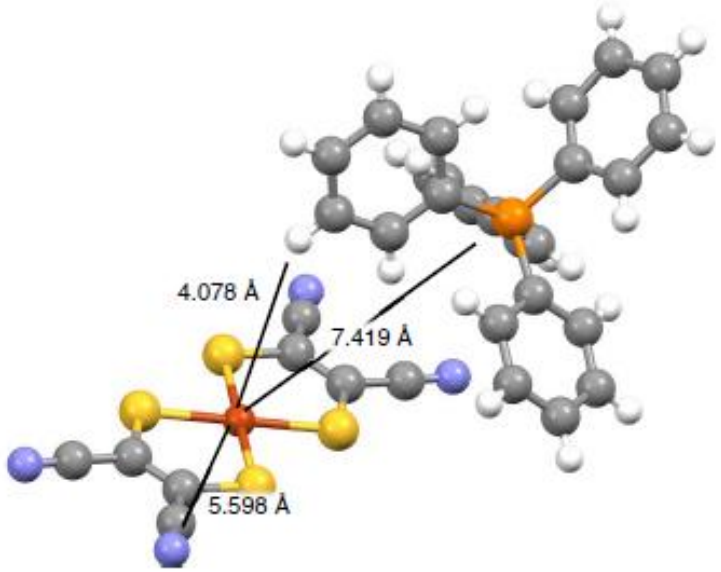
Petiziol, Chiesa, Wimberger, Santini, Carretta, ***NPJ Quantum Information*** *7, 133 (2021)*

Simple physical implementation

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

We developed the **sequence of** mw/rf **pulses**:

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



A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

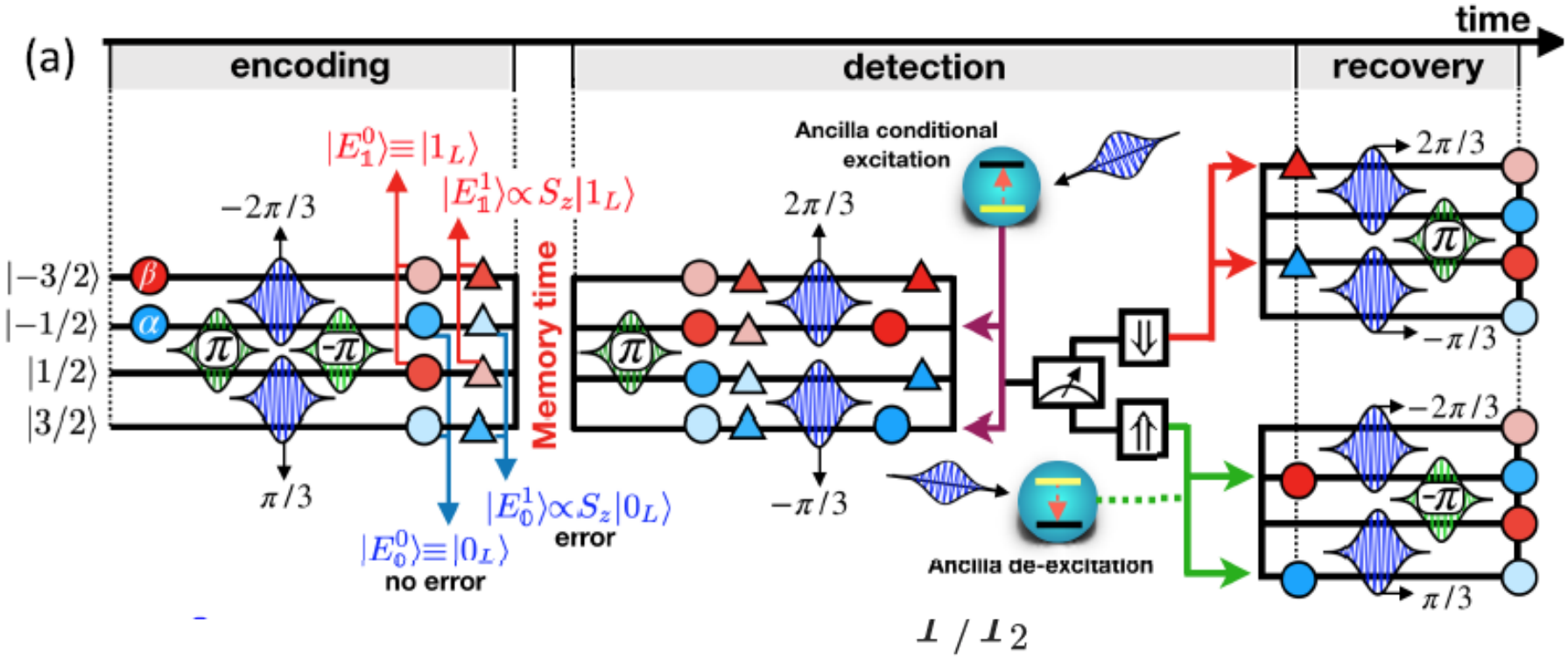
Petiziol, Chiesa, Wimberger, Santini, Carretta, ***NPJ Quantum Information*** *7, 133 (2021)*

Simple physical implementation

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

We developed the **sequence of** mw/rf **pulses**:

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



A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

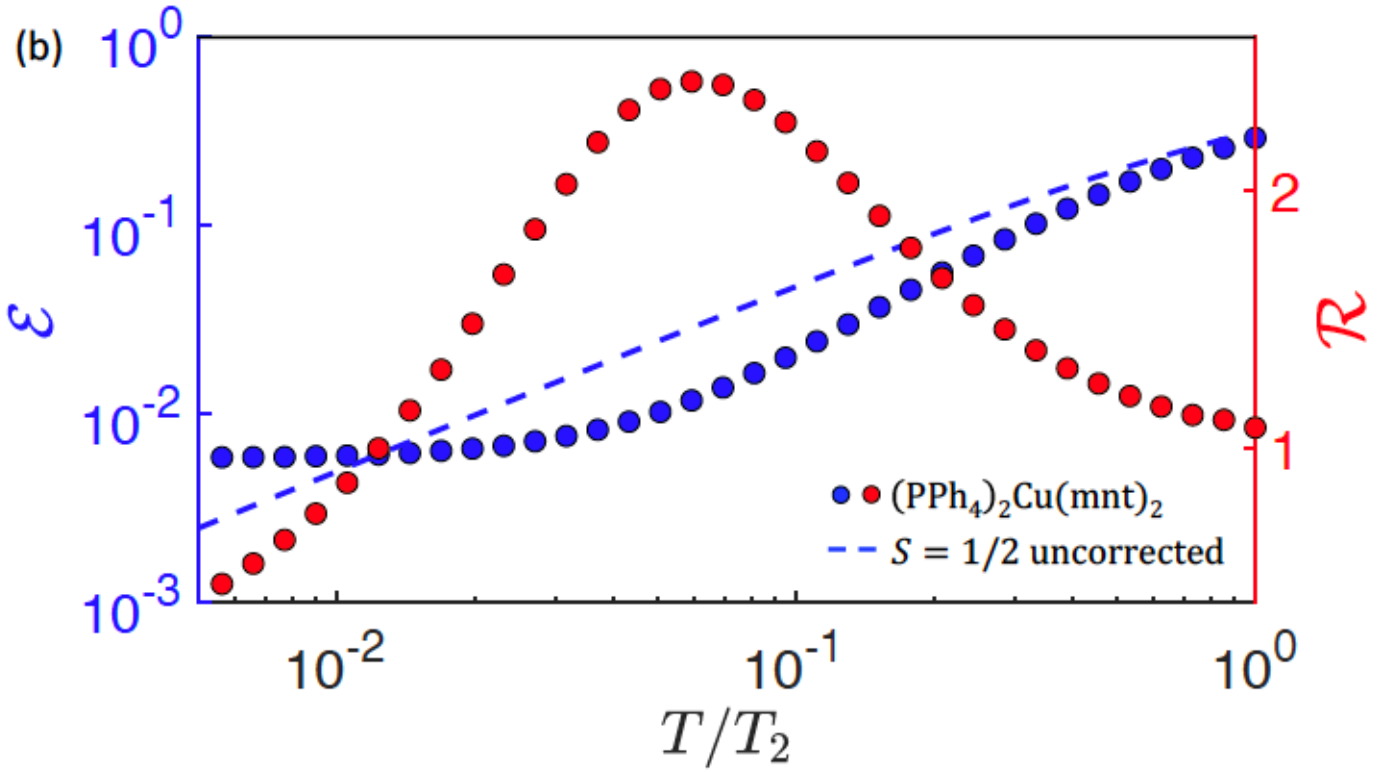
Petiziol, Chiesa, Wimberger, Santini, Carretta, ***NPJ Quantum Information*** *7, 133 (2021)*

Simple physical implementation

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

We developed the **sequence of** mw/rf **pulses**:

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

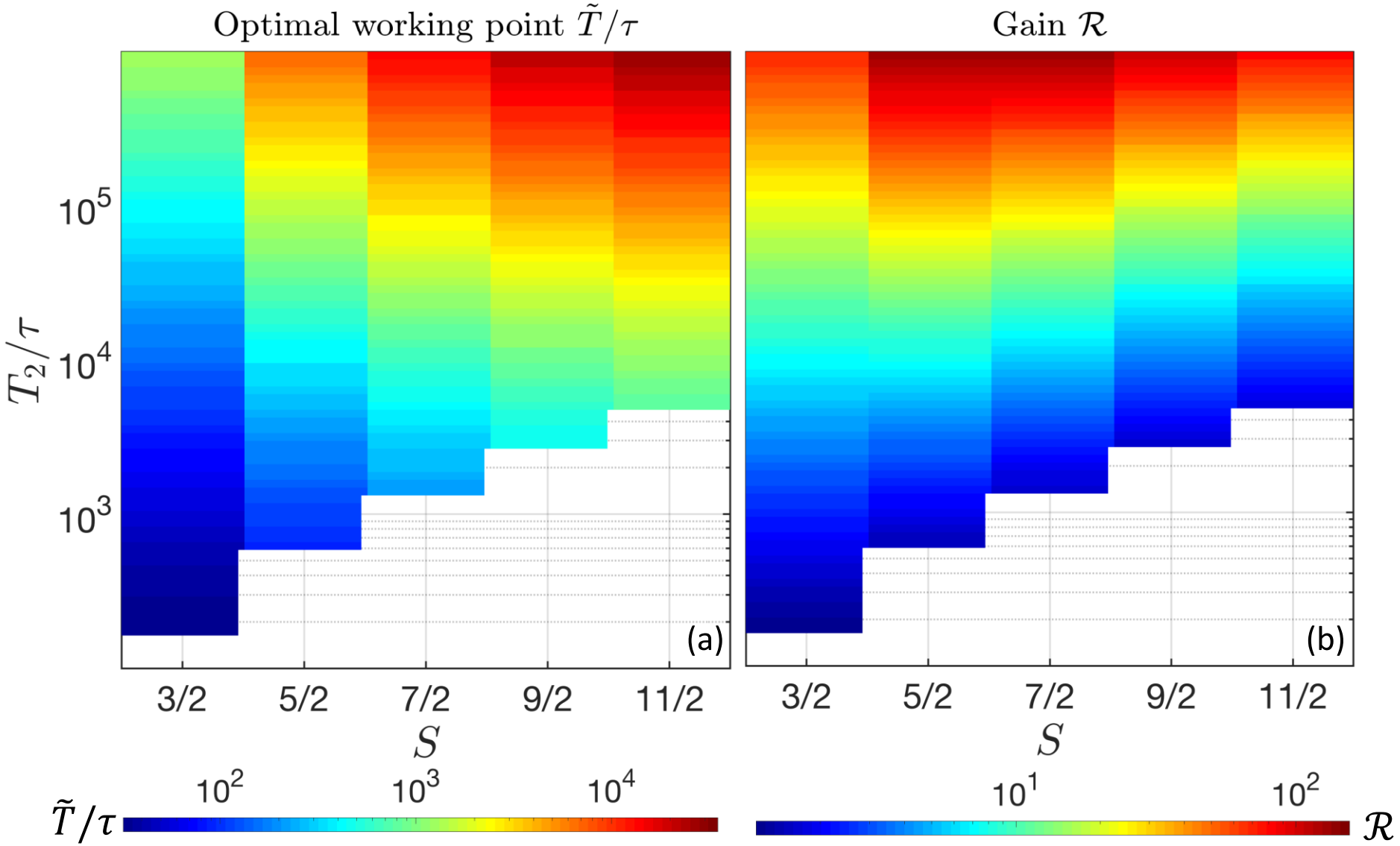


A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

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

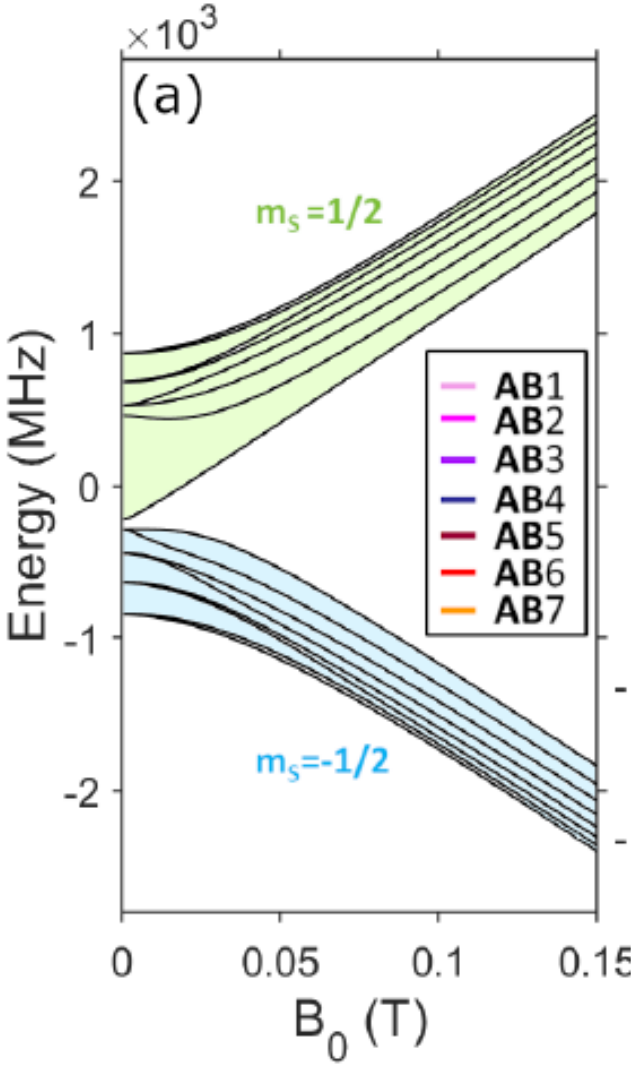
Scaling to larger spins

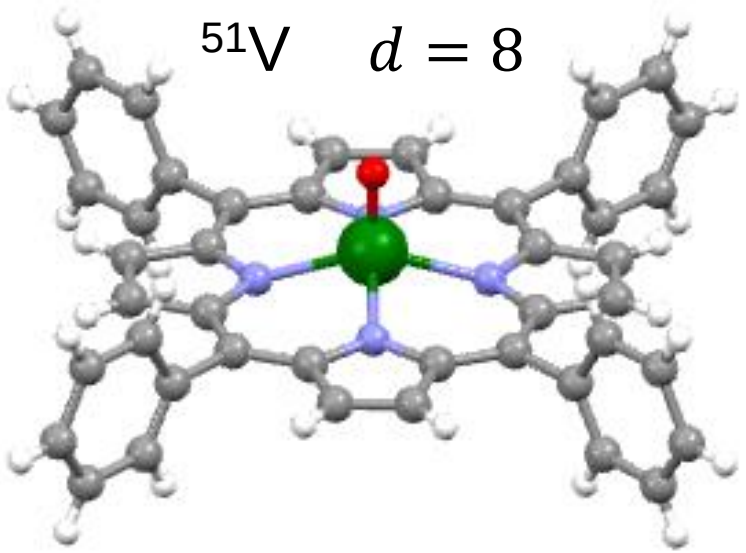
Larger spins allow one to correct more errors (i.e., larger powers of S_z)



A. Chiesa, E. Macaluso, F. Petiziol, S. Wimberger, P. Santini, S. Carretta, ***J. Phys. Chem. Lett.*** **11**, 8610 (2020)

Promising molecular qudits

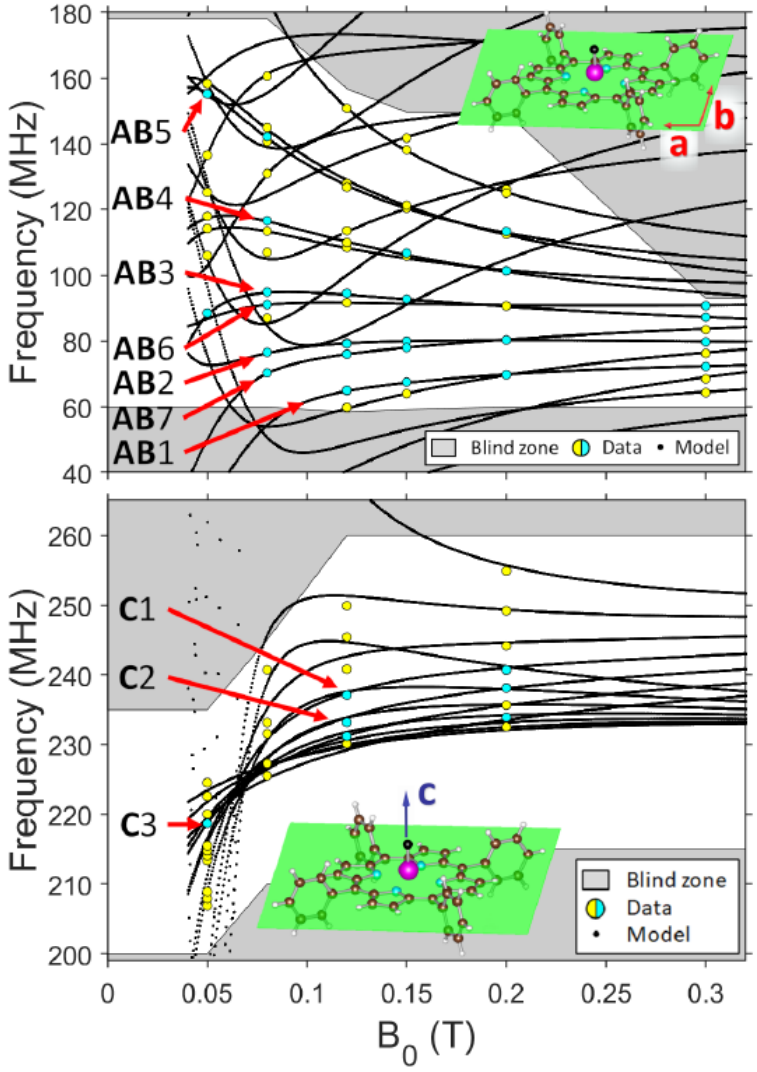


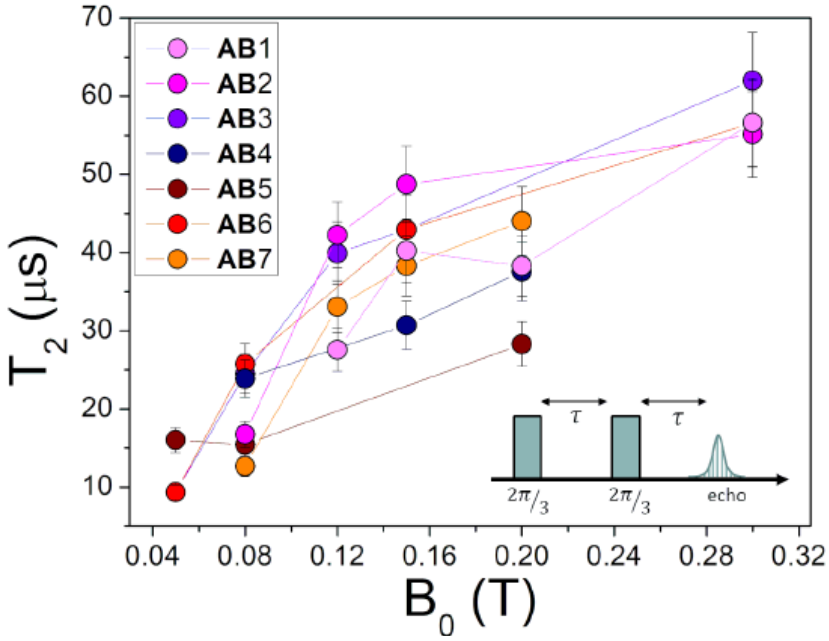


Electronic S=½ - nuclear S= 7/2

Chicco, Chiesa, Allodi, Garlatti, Atzori, Sorace, De Renzi, Sessoli, Carretta**, Chem. Sci., 2021**, DOI: 10.1039/d1sc01358k

Promising molecular qudits

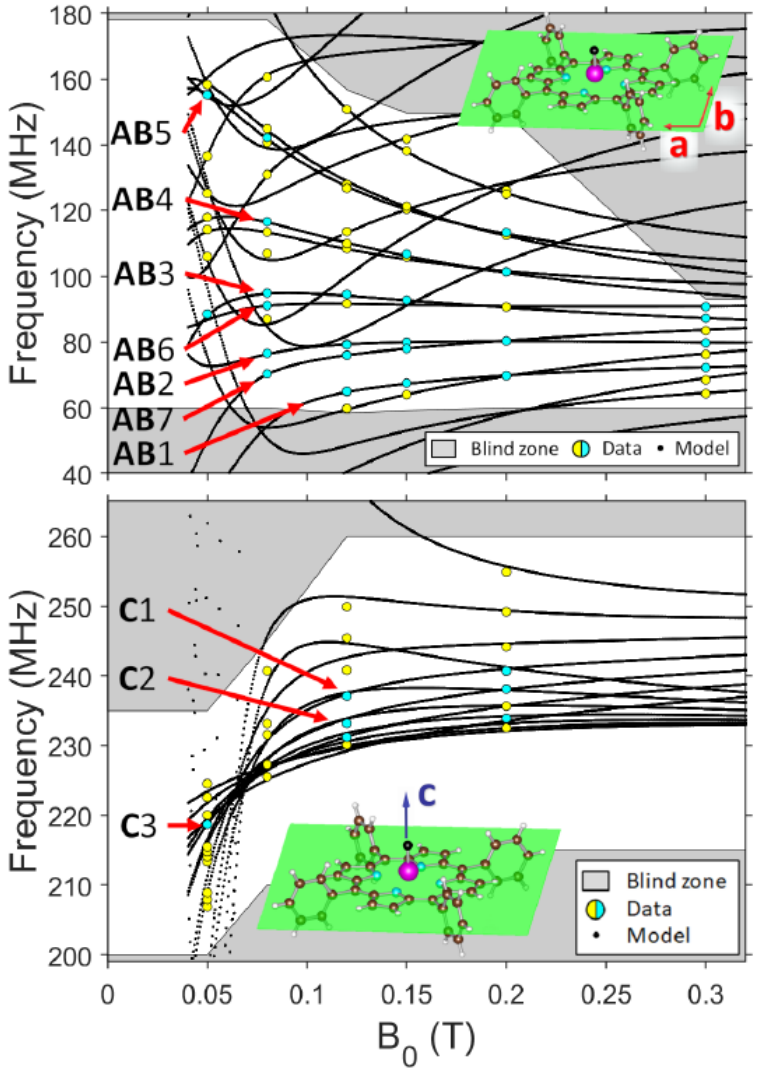


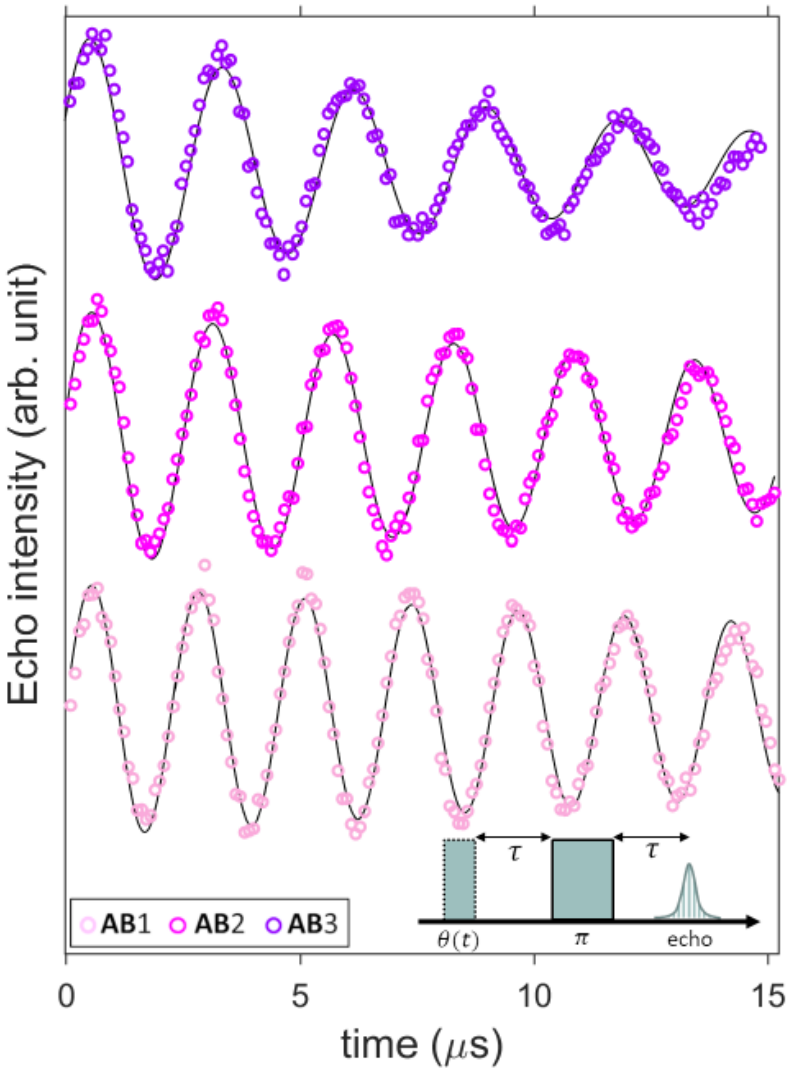


Electronic S=½ - nuclear S= 7/2

Chicco, Chiesa, Allodi, Garlatti, Atzori, Sorace, De Renzi, Sessoli, Carretta**, Chem. Sci., 2021**, DOI: 10.1039/d1sc01358k

Promising molecular qudits



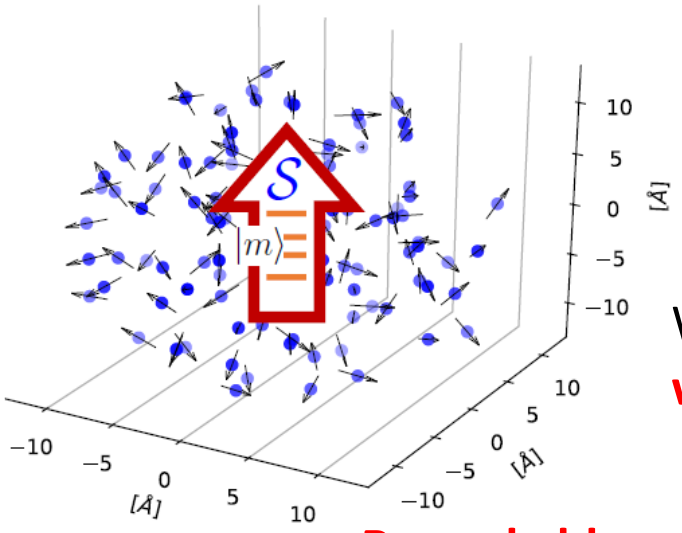


Electronic S=½ - nuclear S= 7/2

Chicco, Chiesa, Allodi, Garlatti, Atzori, Sorace, De Renzi, Sessoli, Carretta**, Chem. Sci., 2021**, DOI: 10.1039/d1sc01358k

Nuclear spin dynamics and optimized qudit encoding

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



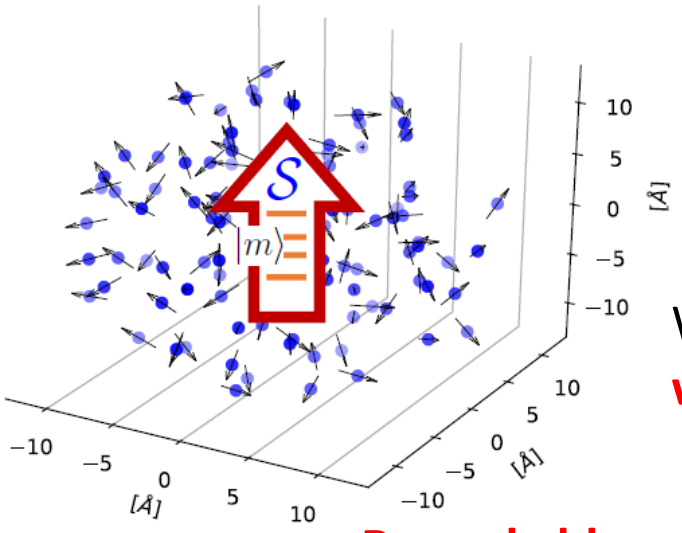
We performed quasi-exact simulations of the coupled dynamics of a molecular qudit and a nuclear spin bath using cluster-correlation expansion

We have designed **numerically optimized QEC code-words** for the errors occuring in real molecules.

Remarkable performance (increasing with S) even at rather long times

Nuclear spin dynamics and optimized qudit encoding

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

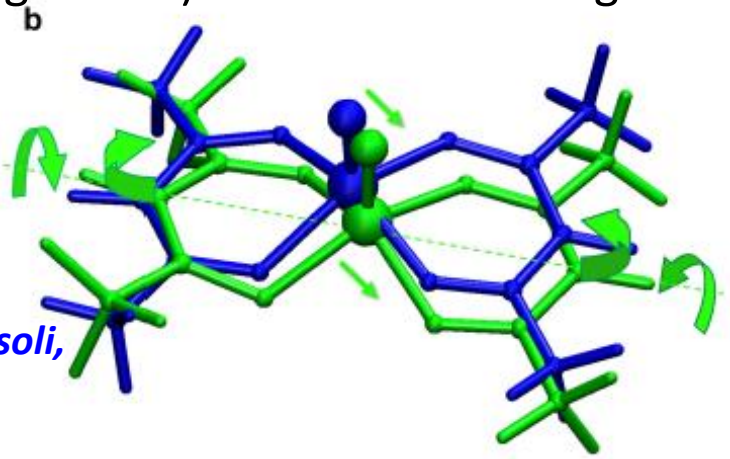


We performed quasi-exact simulations of the coupled dynamics of a molecular qudit and a nuclear spin bath using cluster-correlation expansion

We have designed **numerically optimized QEC code-words** for the errors occuring in real molecules.

Remarkable performance (increasing with S) even at rather long times

Role of vibrations



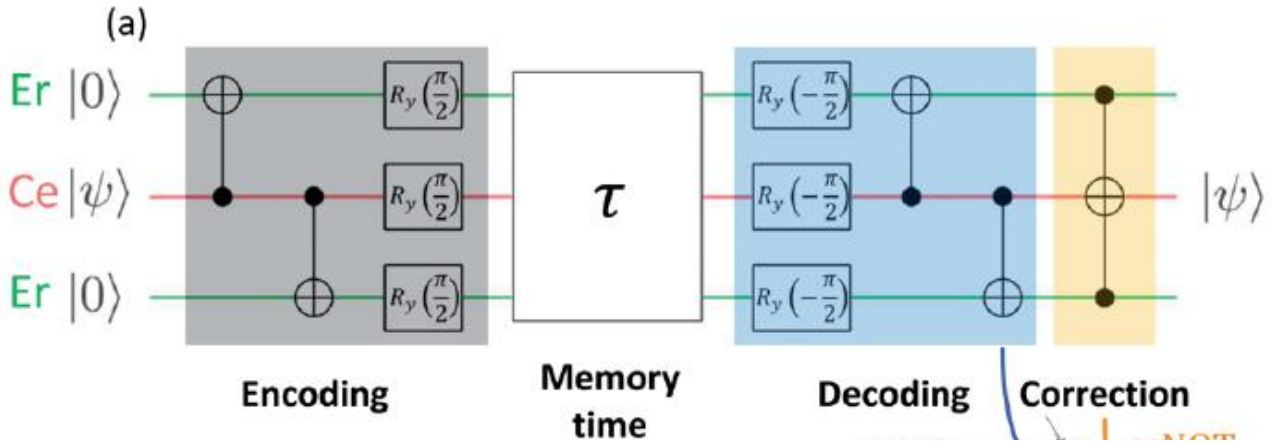
Garlatti, Tesi, Lunghi, Atzori, Voneshen, Santini, Sanvito, Guidi, Sessoli, Carretta, NATURE COMMUNICATIONS 11, 1751 (2020)

Garlatti, et al The Journal of Physical Chemistry Letters 12, 8826 (2021)

M. Briganti et al., J. Am. Chem. Soc. 2021, 143, 34, 13633

Alternative: repetition code with Er-Ce-Er trimer

Weakly interacting (effective) spin ½ + standard QEC codes for phase flip errors



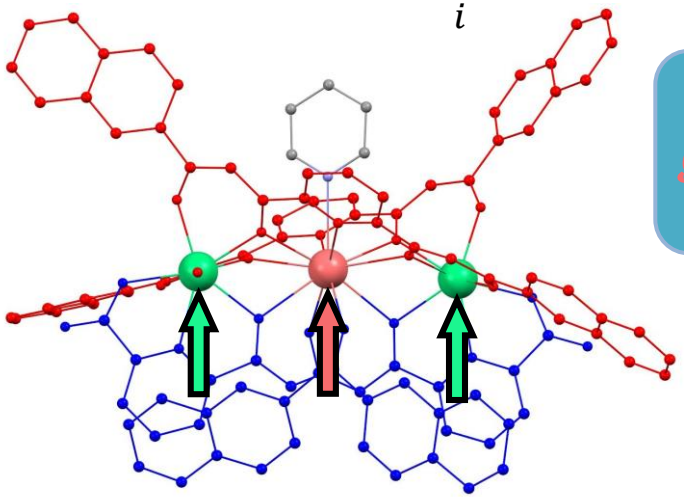
This requires:

-Interacting spin 1/2.
-Factorized eigenstates.
-Fast coherent transitions between eigenstates.

Macaluso, Rubin, Aguilà, Chiesa, Barrios, Martinez, Alonso, Roubeau, Luis, Aromi, Carretta, ***Chem. Sci.*** **11**, 10337 (2020)

Er-Ce-Er: experimental characterization

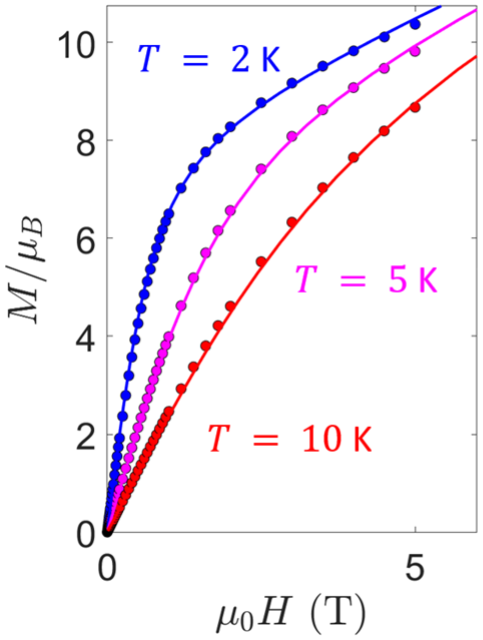
$$H = \mu_B \sum_i \boldsymbol{S}_i \cdot \boldsymbol{g}_i \cdot \boldsymbol{B} + \boldsymbol{S}_1 \cdot \boldsymbol{J}_{12} \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{J}_{23} \cdot \boldsymbol{S}_3 + \boldsymbol{S}_1 \cdot \boldsymbol{J}_{13} \cdot \boldsymbol{S}_3$$

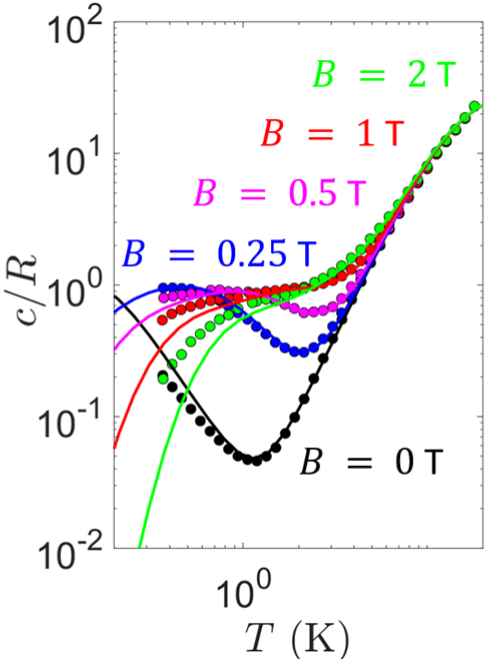


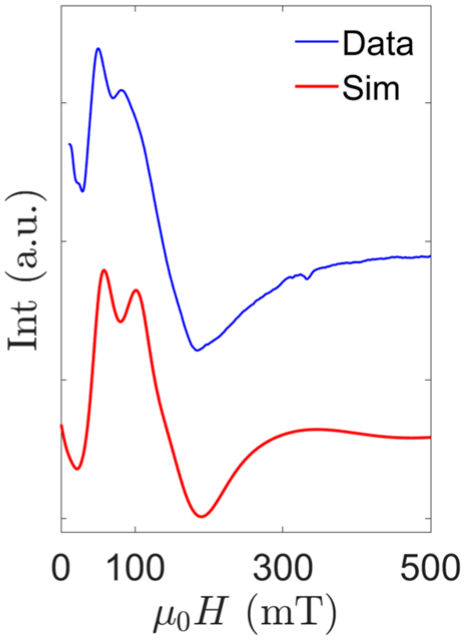
$$g_{Ce} = \begin{pmatrix} 1.7 & 0 & 0 \\ 0 & 1.7 & 0 \\ 0 & 0 & 2.2 \end{pmatrix}$$

$$g_{Er,1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 5 & 0 \\ 0 & 0 & 11.5 \end{pmatrix}$$

g tensors of Ce and Er ions have been determined on the isostructural molecules that contain either one or the other (by using Lu or La).







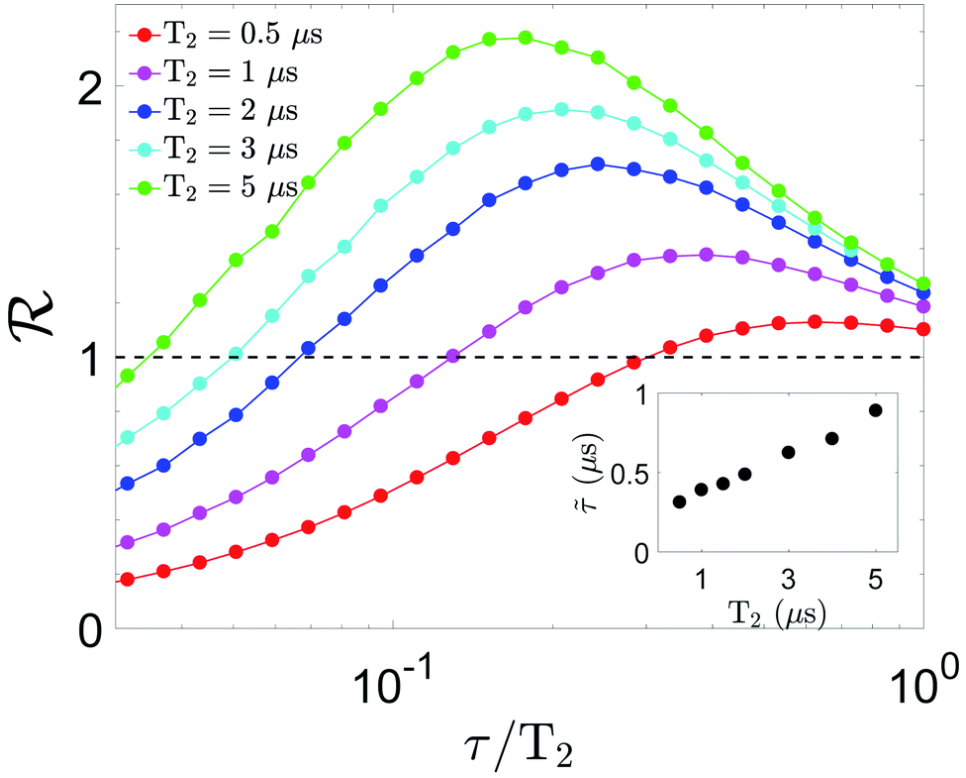
Chem. Sci. 11, 10337 (2020)

Er-Ce-Er: QEC

Gain compared to an uncorrected qubit:

$$\mathcal{R} = \mathcal{E}_U / \mathcal{E}_C$$

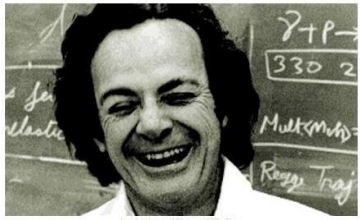
Maximum gain $\tilde{\mathcal{R}}$ at **optimal working point** $\tilde{\tau}$



Macaluso, Rubin, Aguilà, Chiesa, Barrios, Martinez, Alonso, Roubeau, Luis, Aromi, Carretta, ***Chem. Sci.*** **11**, 10337 (2020)

Quantum Simulators

The simulation of quantum systems by a classical computer is intrinsically inefficient, 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 quantum system whose dynamics can be controlled to mimic the evolution of the target system.

PRL **107,** 230502 (2011) PHYSICAL REVIEW LETTERS week ending 2 DECEMBER 2011

Molecular Nanomagnets as Quantum Simulators

P. Santini,[1] S. Carretta,[1] F. Troiani,[2] and G. Amoretti[1]

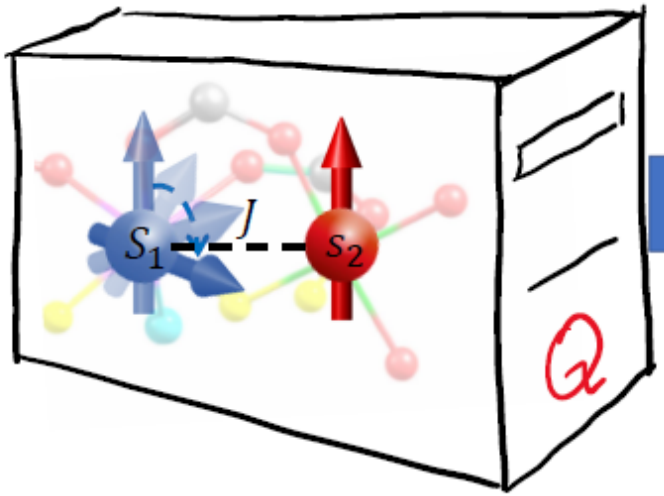
[1]*Dipartimento di Fisica, Università di Parma, Viale G. P. Usberti 7/A, I-43124 Parma, Italy*

[2]*Istituto Nanoscienze-CNR, S3, via G. Campi 213/a, I-41125 Modena, Italy*

(Received 24 June 2011; published 30 November 2011)

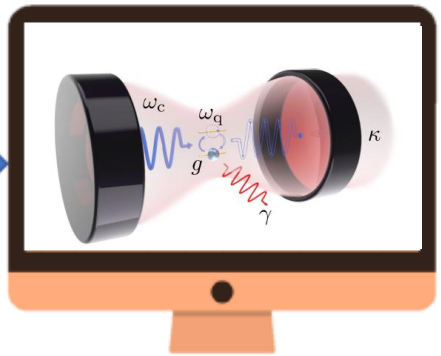
Qudits for simulations of light-matter interaction

Programmable molecular **quantum hardware**



Quantum simulation

Output: dynamics of interacting spin-photon systems

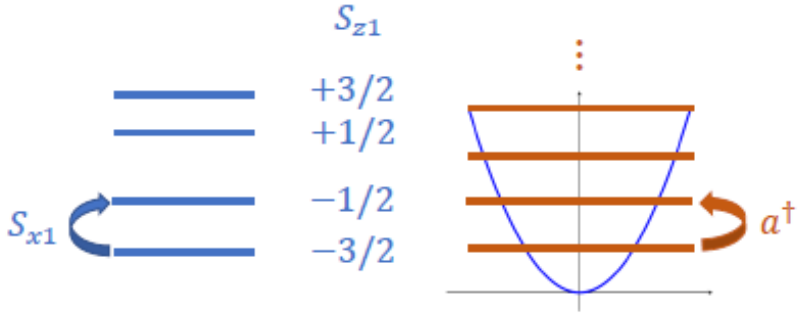


Molecular qudits can simplify the relization of quantum simulation algorithms.

Internal figure from Nat. Rev Phys 1, 19 (2019)

Many levels → Many qubits and complicated gates

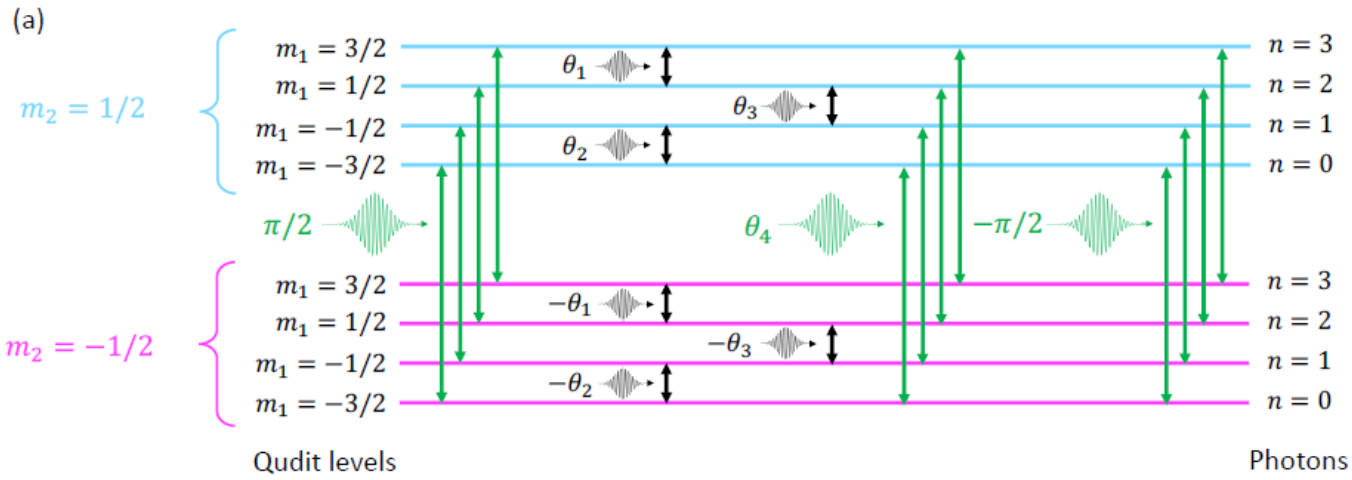
Mapping each boson mode to a single spin S qudit.



Great semplification by exploiting molecular qudits

F. Tacchino, A. Chiesa, R. Sessoli, I. Tavernelli, S. Carretta, Journal of Materials Chemistry C 9, 10266 (2021).

Ground state determination

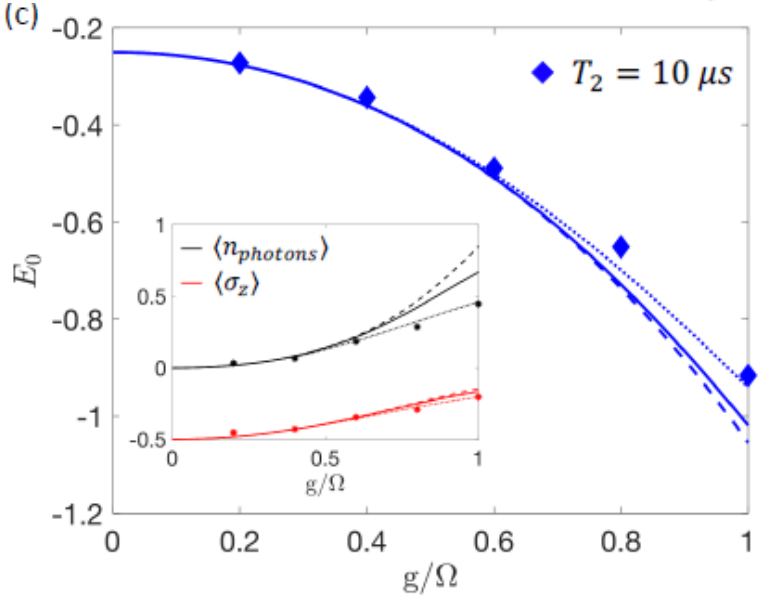


Cr-Cu dimer

S=3/2 – S=1/2

Target Hamiltonian:

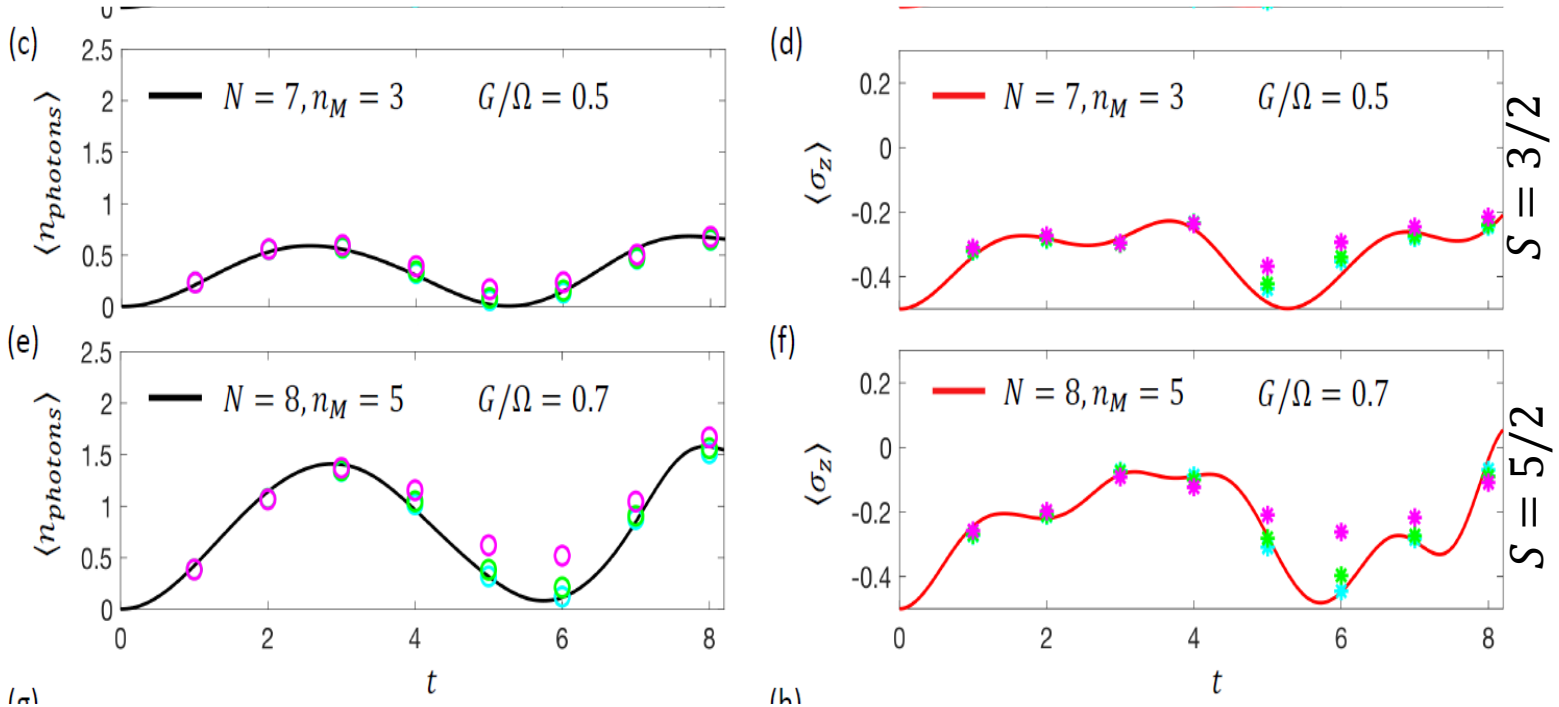
$$\mathcal{H}_S = \omega_a \sigma_z + \Omega a^\dagger a + 2G\sigma_x(a + a^\dagger)$$



- We design the microwave pulse sequence to achieve full control of the coupled qubit-qudit systems: qubit (green) and qudit (black) transitions.
- We can determine the target system **ground state**, by using the **variational** quantum eigensolver.

Time evolution

$$|\psi(0)\rangle \longrightarrow |\psi(t)\rangle = e^{-i\mathcal{H}st}|\psi(0)\rangle$$

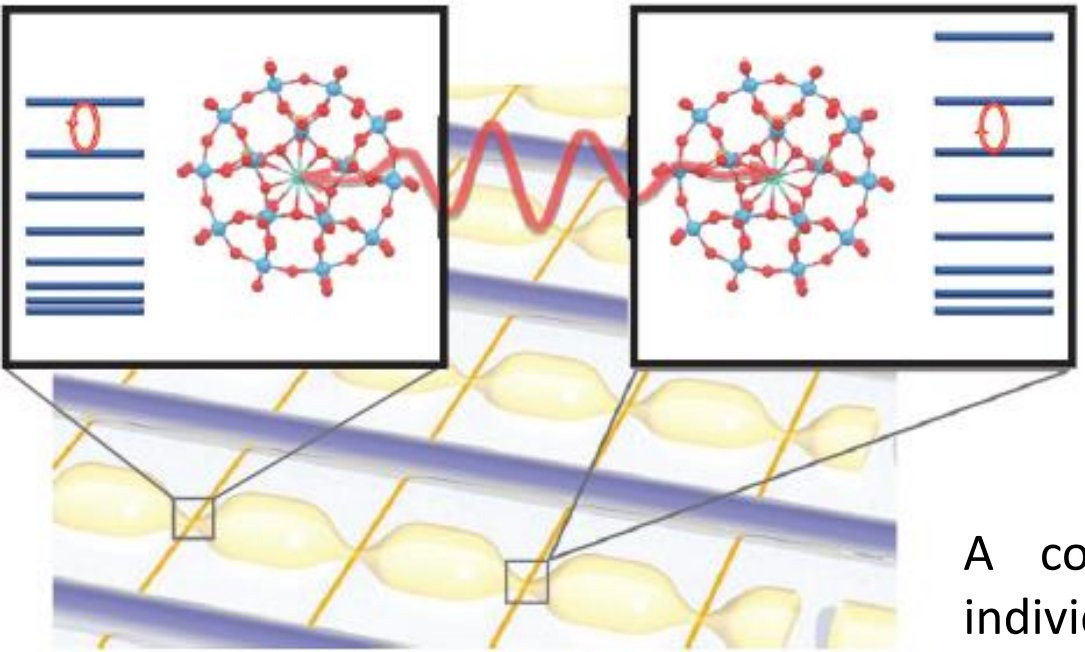


- The same pulses are used to simulate the **time evolution** of the system, by decomposing its evolution using Suzuki-Trotter approximation.

Oscillations are a direct signature of the ultra-strong coupling regime

F. Tacchino, A. Chiesa, R. Sessoli, I. Tavernelli, S. Carretta, Journal of Materials Chemistry C 9, 10266 (2021).

Perspective: the Magnetic Quantum Processor



M. D. Jenkins, et al., Dalton Trans., 2016, 45, 16682

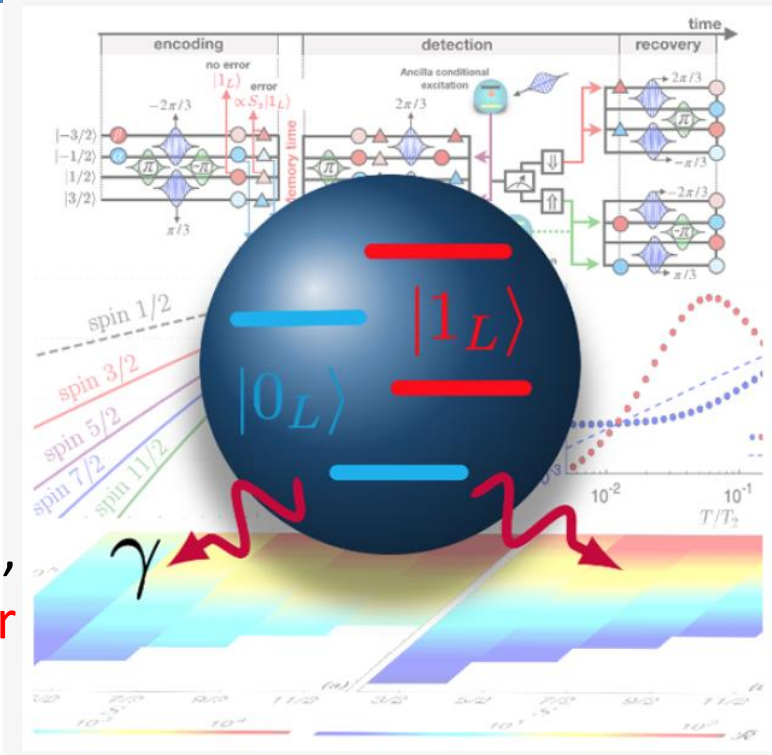
S. Carretta, D. Zueco, A. Chiesa, A. Gomez-Leon, F. Luis, *Appl. Phys. Lett.* 118, 240501 (2021)

A coplanar superconducting resonator, individual molecular nanomagnets placed on specific locations of its central line, and perpendicular auxiliary wave guides.

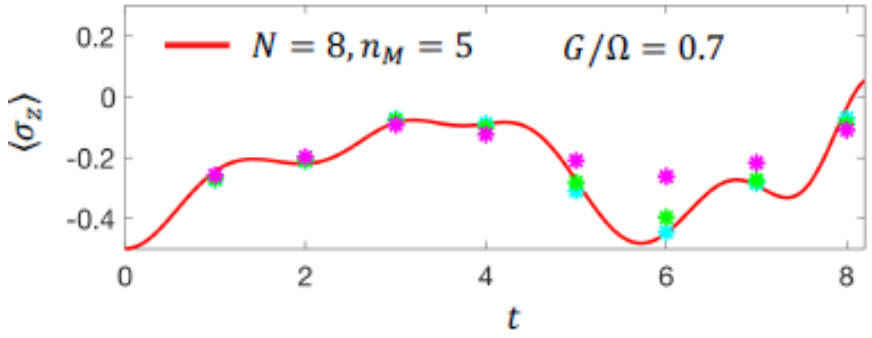
A switchable coupling between distant molecules can be obtained through virtual or real exchange of photons

Conclusions

- Molecular qudits can be used as qubits with embedded Quantum Error Correction.



- By modelling interaction with nuclear spins, we can design QEC codes optimized for specific molecules.



- Molecular qudits can simplify the relization of quantum simulation algorithms.

UNIVERSITÀ
DI PARMA

P. Santini, R. De Renzi,

G. Allodi, S. Wimberger,

A. Chiesa, E. Garlatti,

F. Petiziol, E. Macaluso,
S. Chicco

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 862893

FATMOLS

PRIN 2017

Progetti di ricerca di Rilevante Interesse Nazionale

HORIZON 2020

SUMO

QUANTERA

Involved people

R. Sessoli, L. Sorace, M. Atzori (now CNRS Grenoble)

F. Luis, D.Zueco, O. Roubeau, J.I. Martınez, P.J. Alonso,

G. Aromì, D. Aguilà, L.A. Barrios

R. Winpenny, E. McInnes, G. Timco, S. Lockyer

I. Tavernelli, F. Tacchino

D. Gerace, M. Grossi (IBM Italy)