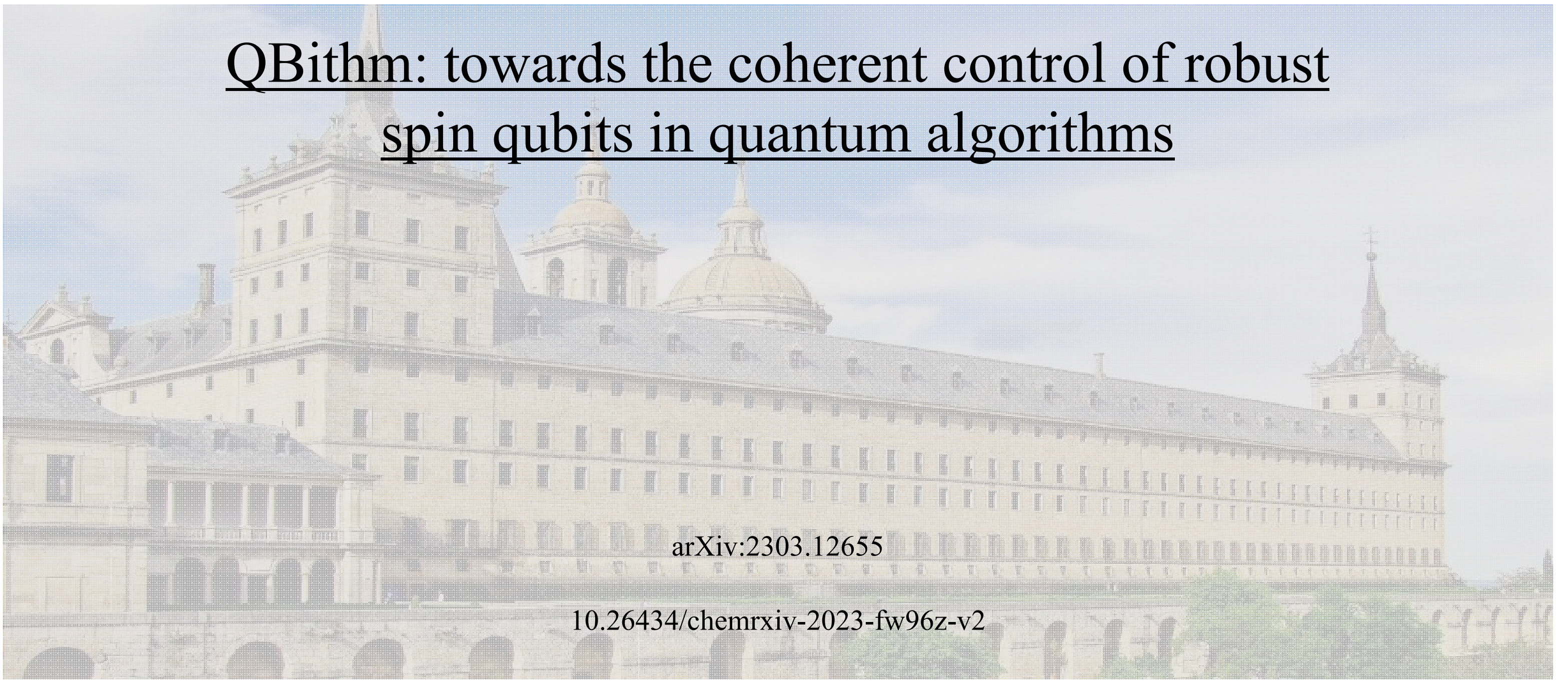


# QBithm: towards the coherent control of robust spin qubits in quantum algorithms

arXiv:2303.12655

10.26434/chemrxiv-2023-fw96z-v2

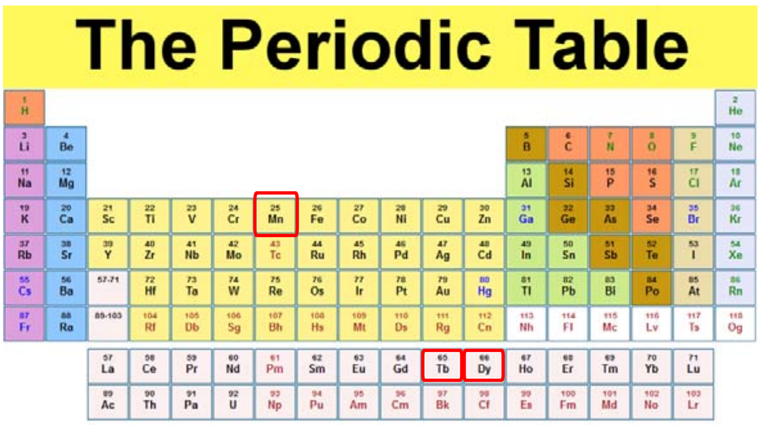
Dr. Luis Escalera Moreno

May 11th

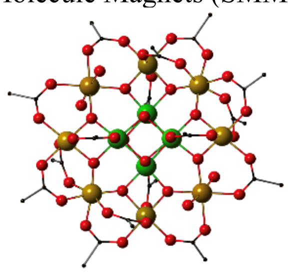
luis.escalera.moreno@mpq.mpg.de

Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Nanomagnets

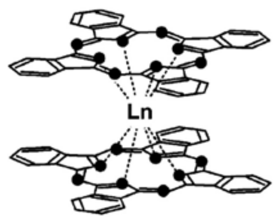



Polynuclear: Single-Molecule Magnets (SMMs)



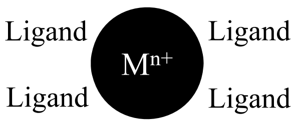
$Mn_{12}$ (R. Sessoli et al., 1993)

Mononuclear: Single-Ion Magnets (SIMs)




$[Ln^{3+}Pc]^-$ , Ln = Tb, Dy (N. Ishikawa et al., 2004)

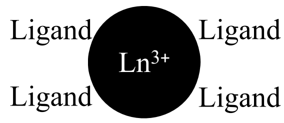
Molecular Nanomagnet ⇄ Magnetic Coordination Compound



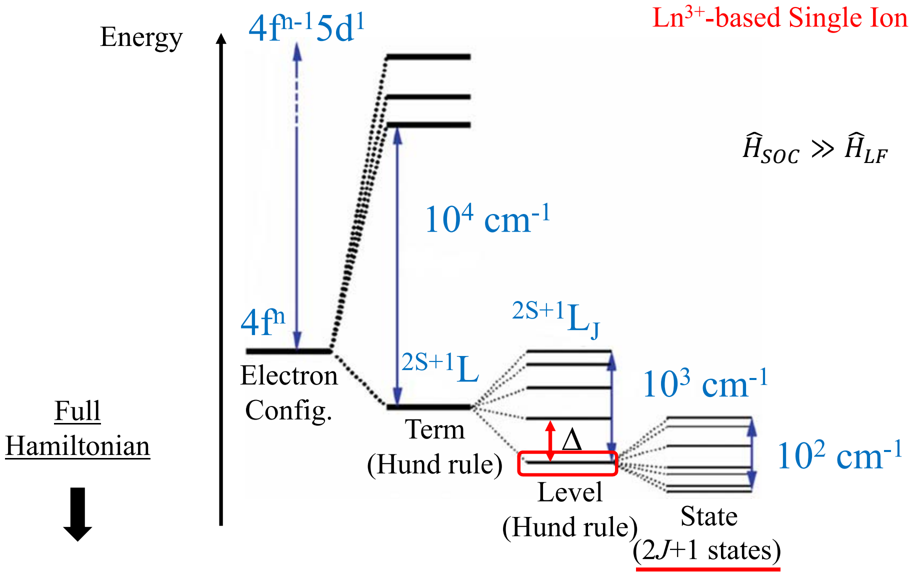
## Systems ➔ Applications ➔ Characterization ➔ Modeling

### Molecular Nanomagnets

$Ln^{3+}$-based Single Ion Magnets (SIMs)




$\hat{H}_{SOC} \gg \hat{H}_{LF}$



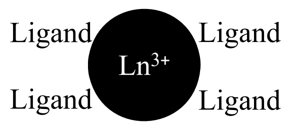

$\hat{H} = \hat{H}_{kin} + \hat{H}_{e-n} + \hat{H}_{e-e} + \hat{H}_{SOC} +$ Ligand Field $\hat{H}_{LF}$



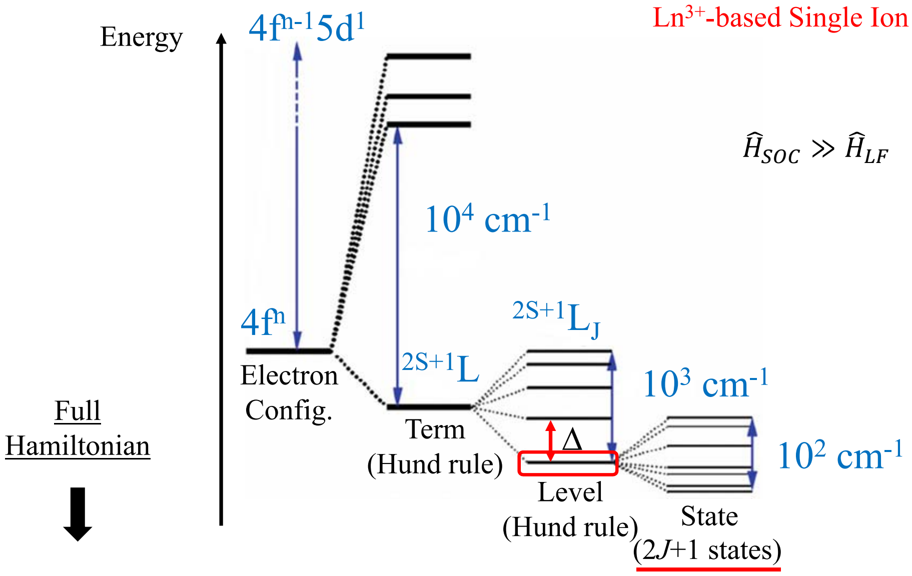
## Systems ➔ Applications ➔ Characterization ➔ Modeling

### Molecular Nanomagnets

$Ln^{3+}$-based Single Ion Magnets (SIMs)



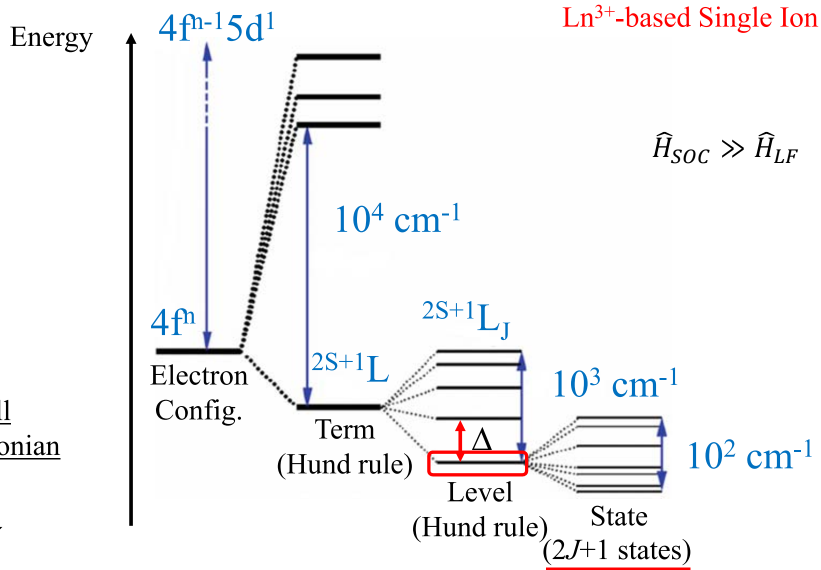

$\hat{H}_{SOC} \gg \hat{H}_{LF}$  $\Delta \gg k_B T$ ⟶ Only ground $J$ multiplet



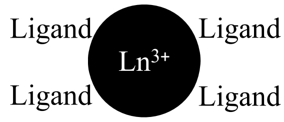

$\hat{H} = \hat{H}_{kin} + \hat{H}_{e-n} + \hat{H}_{e-e} + \hat{H}_{SOC} +$ Ligand Field $\hat{H}_{LF}$

## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Nanomagnets




$Ln^{3+}$-based Single Ion Magnets (SIMs)




$\hat{H}_{SOC} \gg \hat{H}_{LF}$ $\quad \Delta \gg k_B T \longrightarrow$ Only ground $J$ multiplet

Crystal Field Hamiltonian

$$\{|m_J\rangle\}_{-J}^{+J} \longrightarrow \hat{H}_{CF} = \sum_{k=2,4,6} \sum_{q=-k}^{+k} B_k^q \hat{O}_k^q$$

$B_k^q$ Crystal Field Parameters (27)

$\hat{O}_k^q$ Extended Stevens Operators (27)

Full Hamiltonian

⬇

$$\hat{H} = \hat{H}_{kin} + \hat{H}_{e-n} + \hat{H}_{e-e} + \hat{H}_{SOC} + \text{Ligand Field } \hat{H}_{LF}$$



## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Nanomagnets

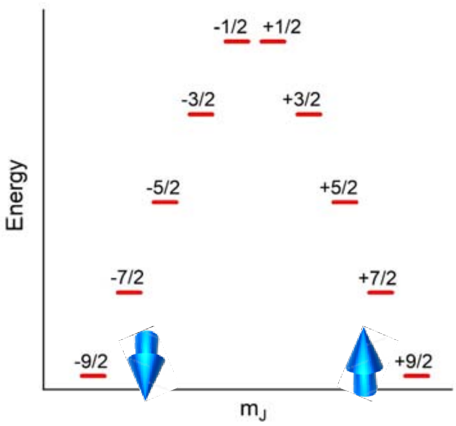
$Nd^{3+}$

[Xe] $4f^3$

$^4I_{9/2}$

$J_{ground} = 9/2$

10 states




**Ground Doublet** ✔

**Classical magnetic memory: BIT = {0,1}**

Systems ➡ **Applications** ➡ Characterization ➡ Modeling

## Molecular Nanomagnets

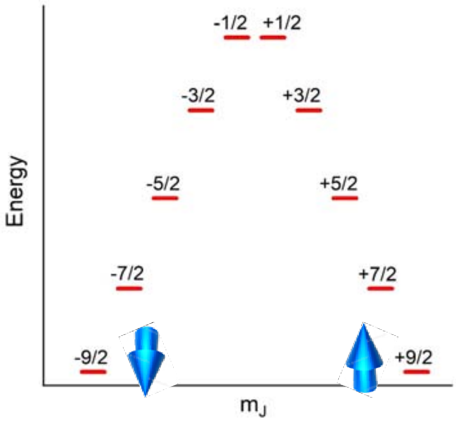
$Nd^{3+}$

[Xe] $4f^3$

$^4I_{9/2}$

$J_{ground} = 9/2$

10 states




**Ground Doublet**

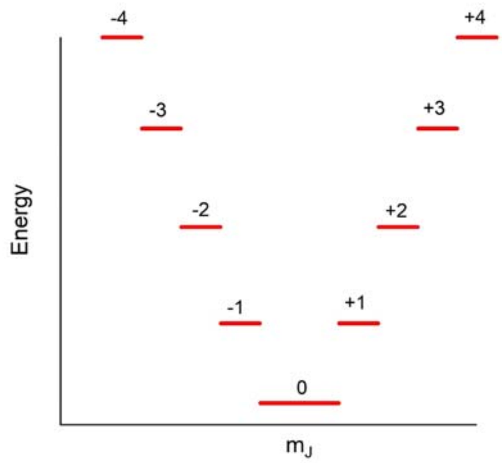
$Pr^{3+}$

[Xe] $4f^2$

$^3H_4$

$J_{ground} = 4$

9 states




**Ground Singlet**

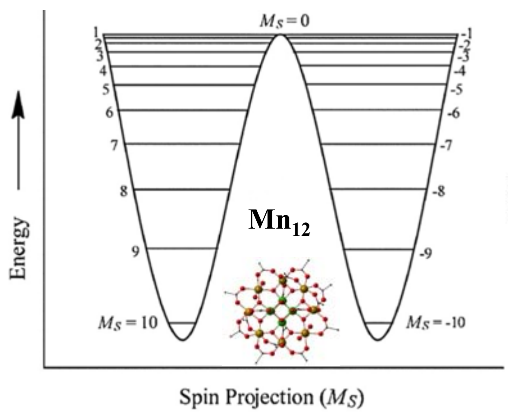
**Classical magnetic memory: BIT = {0,1}**



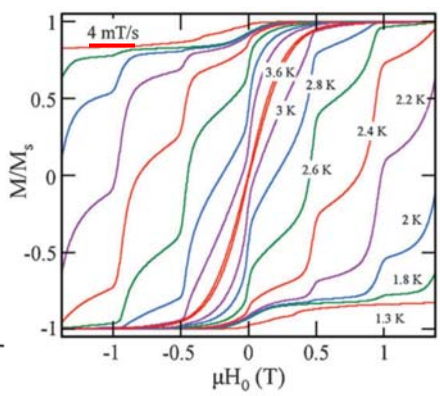
Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Nanomagnets

**Relaxation under user-driven evolution**




For a given sweep rate…




*Hysteresis Loop*

→ **Memory effect**

→ **Bistability at zero magnetic field**

→ **Classical magnetic memory**

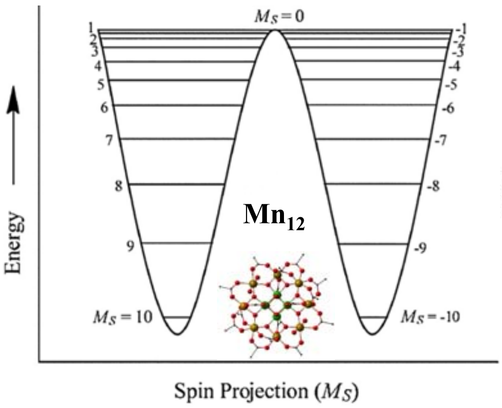
**BIT = {0,1}**

$S_{ground} = 10$ R. Bagai, G. Christou, Chem. Soc. Rev., 38, 1011-1026 (2009)

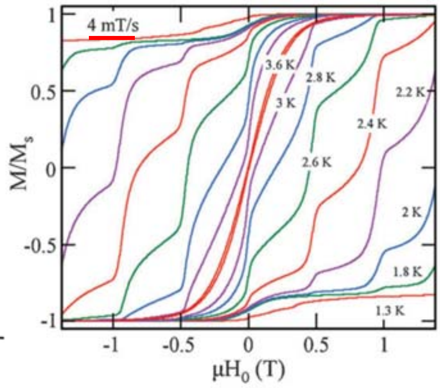
Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Nanomagnets

**Relaxation under user-driven evolution**




For a given sweep rate…




$S_{ground} = 10$ R. Bagai, G. Christou, Chem. Soc. Rev., 38, 1011-1026 (2009)

*Hysteresis Loop*

→ **Memory effect**

→ **Bistability at zero magnetic field**

→ **Classical magnetic memory**

**BIT = {0,1}**

…up to a maximum temperature called:

Blocking Temperature $T_b$

Take-home message:
To increase blocking temperature

**High temperature operativity**



Systems ➡ Applications ➡ **Characterization** ➡ Modeling



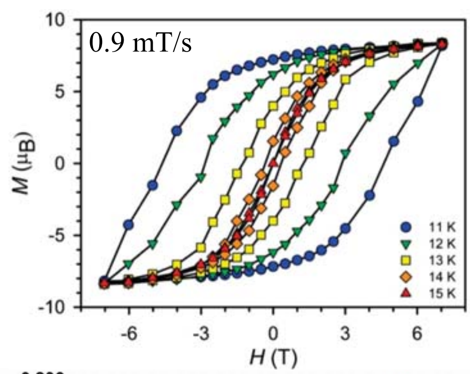
## Molecular Nanomagnets



**1993, $Mn_{12}$** → **2004, $[Ln^{3+}Pc]^-$** → **2011, $Tb^{3+}$-based**

$T_b \sim 4K$ — $T_b \sim 2K$ — $T_b \sim 14K$

Sessoli et al. — Ishikawa et al. — Long et al.

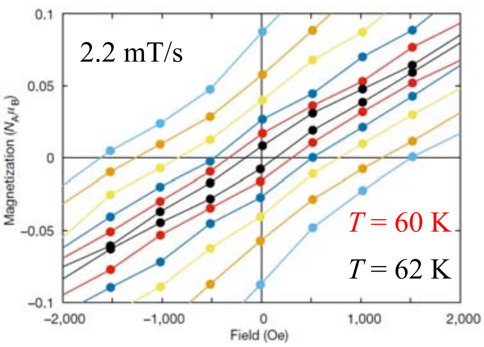



Chilton et al.

→

**2017, $[Dy^{3+}Cp^{ttt}]^+$**

$T_b \sim 60K$

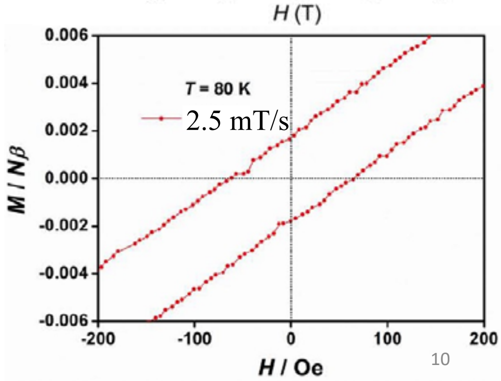



Layfield et al.

→

**2018, $Dy^{3+}$-5***

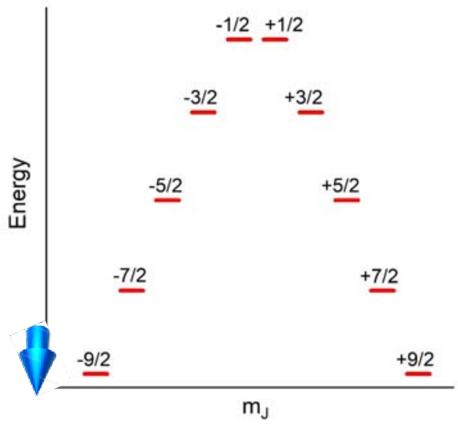
$T_b \sim 80K$



Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Nanomagnets

**Relaxation under free evolution**




**Sample magnetized**

Apply an external magnetic field ➡ $p_{-9/2} = 1$ $p_i = 0$ $i \neq -9/2$

**Not stable anymore**

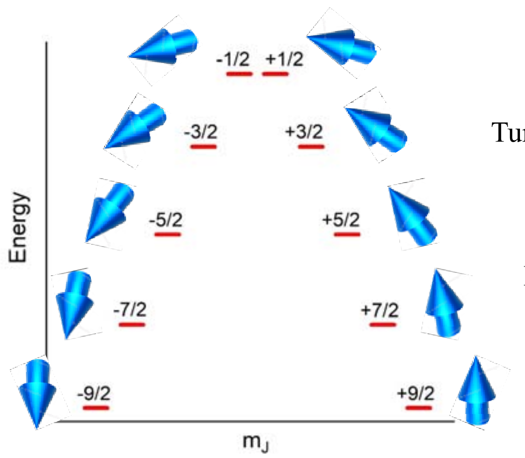
Turn the magnetic field off ➡ $p_{-9/2} = 1$ $p_i = 0$ $i \neq -9/2$



Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Nanomagnets

**Relaxation under free evolution**




**Sample magnetized**

Apply an external magnetic field ➡ $p_{-9/2} = 1$ $p_i = 0$ $i \neq -9/2$

**Not stable anymore**

Turn the magnetic field off ➡ $p_{-9/2} = 1$ $p_i = 0$ $i \neq -9/2$

For a long enough time… ➡ Boltzmann Population $p_i = \frac{\exp(-E_i/k_B T)}{Z} > 0$

$T > 0$

$\tau = \tau(T)$

Take-home message:
To increase relaxation time

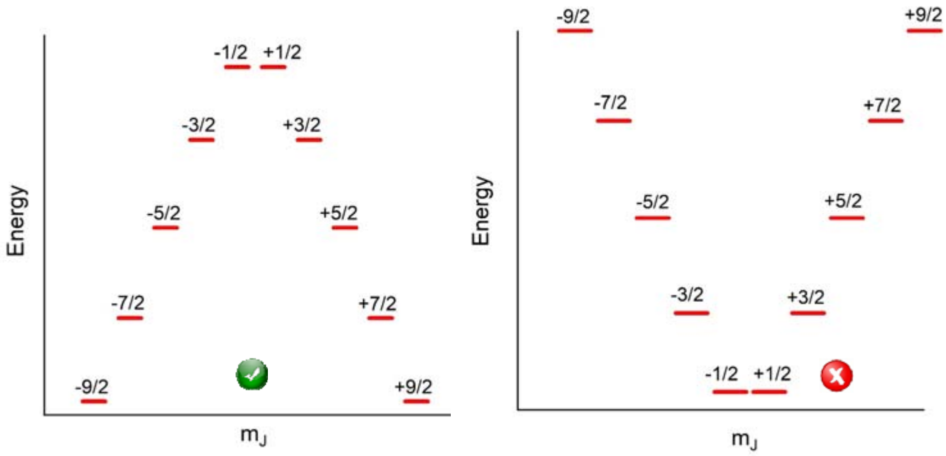
**Long-lived classical memory**

Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Nanomagnets

$$probability(|-J_g\rangle \rightarrow |+J_g\rangle) \propto \langle -J_g|\hat{H}_{int}|+J_g\rangle$$

$J_{ground} = 9/2$





For a small transition probability…

Try the two $m_J = \pm J$ states as the ground doublet

Try to use large $J_{ground}$

Pre $Gd^{3+}$: $J_{ground}$ = |L - S|

Post $Gd^{3+}$: $J_{ground}$ = |L + S|

$Dy^{3+}$ [Xe] $4f^9$ $^6H_{15/2}$

$J_{ground} = 15/2$ 16 states

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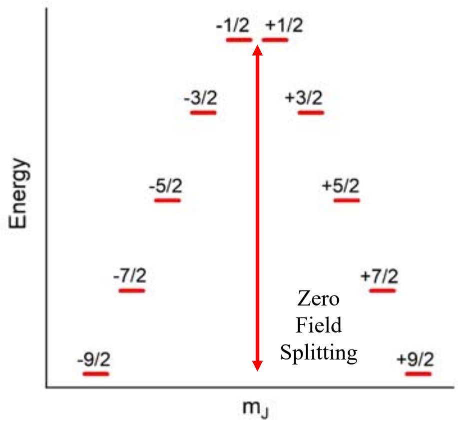
Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Nanomagnets

$$probability(|m_J\rangle \rightarrow |m'_J\rangle) \propto 1/\left(E_{m_J} - E_{m'_J}\right)$$

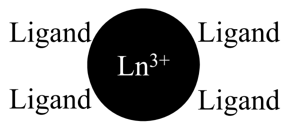
$J_{ground} = 9/2$




For a small transition probability…

Try a large Zero Field Splitting →

**Fine choice of the ligands**




**1993, $Mn_{12}$** $Z.F.S. \sim 10\ cm^{-1}$

**2004, $[Ln^{3+}Pc]^-$** $Z.F.S. \sim 100\ cm^{-1}$

**2017, $[Dy^{3+}Cp^{ttt}]^+$**

$Z.F.S. \sim 1400 - 1900\ cm^{-1}$

**2018, $Dy^{3+}$-5***

14

Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Molecular Nanomagnets

**Electronic Structure Engineering**

- The two $m_J = \pm J$ states as the ground doublet
- Large $J_{ground}$
- Large Zero Field Splitting

Goal: To increase $T_b, \tau$

➡ As working temperature is increased…

**Insufficient description!**

**Key Role of Spin-Vibration Coupling**

Important source of **magnetic relaxation**



Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Molecular Nanomagnets

**Spin-Vibration Coupling Engineering**

**Relaxation under free evolution**

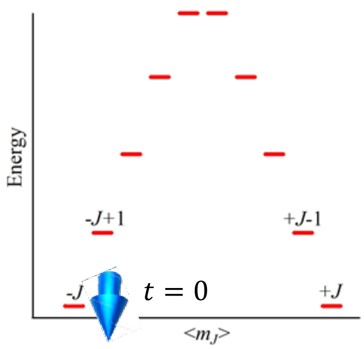
**Pauli Master Equation**

$$\frac{dp_i(t)}{dt} = \sum_{f=1, f\neq i}^{2J+1} \left[ \gamma_{f\to i} p_f(t) - \gamma_{i\to f} p_i(t) \right] \quad i = 1, \ldots, 2J+1$$

F. Luis, J. Bartolomé, J. F. Fernández, *Phys. Rev. B*, (1998), **57**(1), 505-513

J. F. Fernández, F. Luis, J. Bartolomé, *Phys. Rev. Lett.*, (1998), **80**(25), 5659-5662

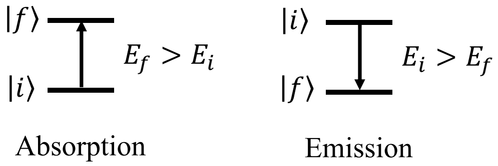
Gómez-Coca et al., *Nat. Commun.*, (2014), **5**, 4300



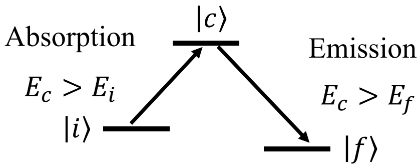

Spin Populations $\quad 0 \le p_i(t) \le 1 \quad \sum_i p_i(t) = 1$

Transition Rates ⟵ the spin absorbs and emits phonons from and to the vibration bath

**Direct between real states**




**Indirect (through real or virtual state)**



# Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Nanomagnets

**L. Escalera-Moreno**, N. Suaud, A. Gaita-Ariño, E. Coronado, *J. Phys. Chem. Lett.* **8**(7), 1695-1700 (2017)

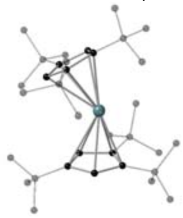
The Journal of Physical Chemistry Letters — **arXiv: 1512.05690 (2015)** — Letter — pubs.acs.org/JPCL

Determining Key Local Vibrations in the Relaxation of Molecular Spin Qubits and Single-Molecule Magnets

*ab initio* rates, $\tau(T)$, $p_i(t)$ efficient methods prediction

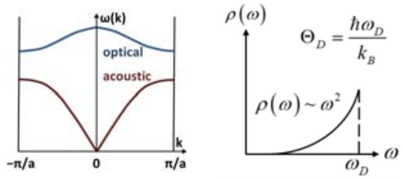
**L. Escalera-Moreno**, J. J. Baldoví, A. Gaita-Ariño, E. Coronado, *Chem. Sci.* **11**, 1593-1598 (2020)

PICK OF THE WEEK



**L. Escalera-Moreno**, J. J. Baldoví, A. Gaita-Ariño, E. Coronado, *Chem. Sci.* **9**(13), 3265-3275 (2018)

**Collection: Most impactful nanoscience articles**




review articles

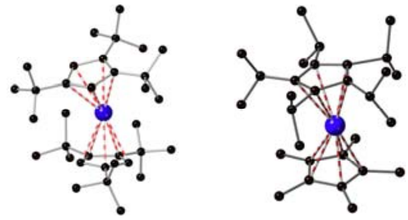
DOI: 10.1039/C7SC05464E (Perspective) Chem. Sci., 2018, 9, 3265-3275

Spin states, vibrations and spin relaxation in molecular nanomagnets and spin qubits: a critical perspective

Luis Escalera-Moreno, José J. Baldoví, Alejandro Gaita-Ariño and Eugenio Coronado

**L. Escalera-Moreno**, J. J. Baldoví, A. Gaita-Ariño, E. Coronado, *Inorg. Chem.* **58**(18), 11883-11892 (2019)

**Article Forum**



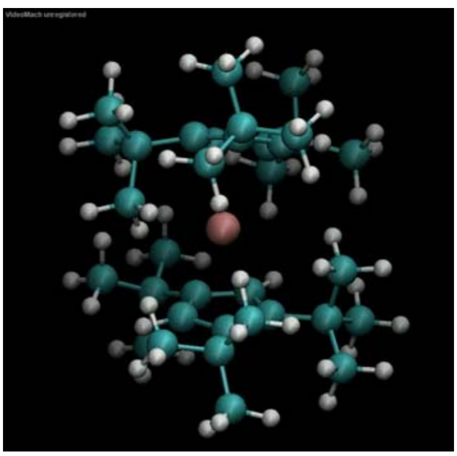


# Systems ➡ Applications ➡ Characterization ➡ Modeling

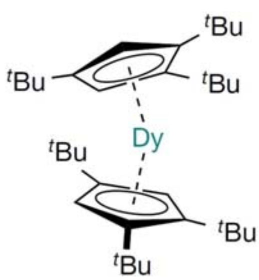
## Molecular Nanomagnets

**To identify the driving vibrations** ➡ **Re-design the molecule to suppress them** — **To guide synthetic efforts through a Rational Design**

**Rocking-like movements in carbon ring H atoms of $[Dy^{3+}(Cp^{ttt})_2]^+$ ⇒ not present in $[Dy^{3+}(Cp^*)(Cp^{iPr5})]^+$**

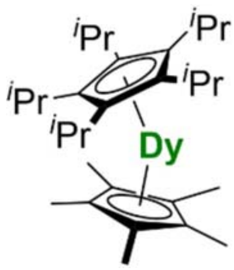


$\nu = 135.0\ \mathrm{cm}^{-1}$




C.A.P. Goodwin et al., Nature, (**2017**), 548, 439-442

➡




Fu-S. Guo et al., Science, (**2018**), 362, 1400-1403

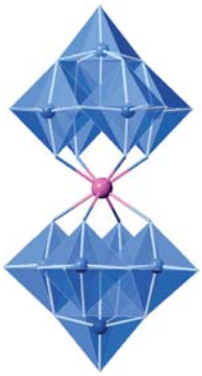
**It worked!**

$T_b \sim 60\mathrm{K} \rightarrow 80\mathrm{K}$

**Key Role of Spin-Vibration Coupling**

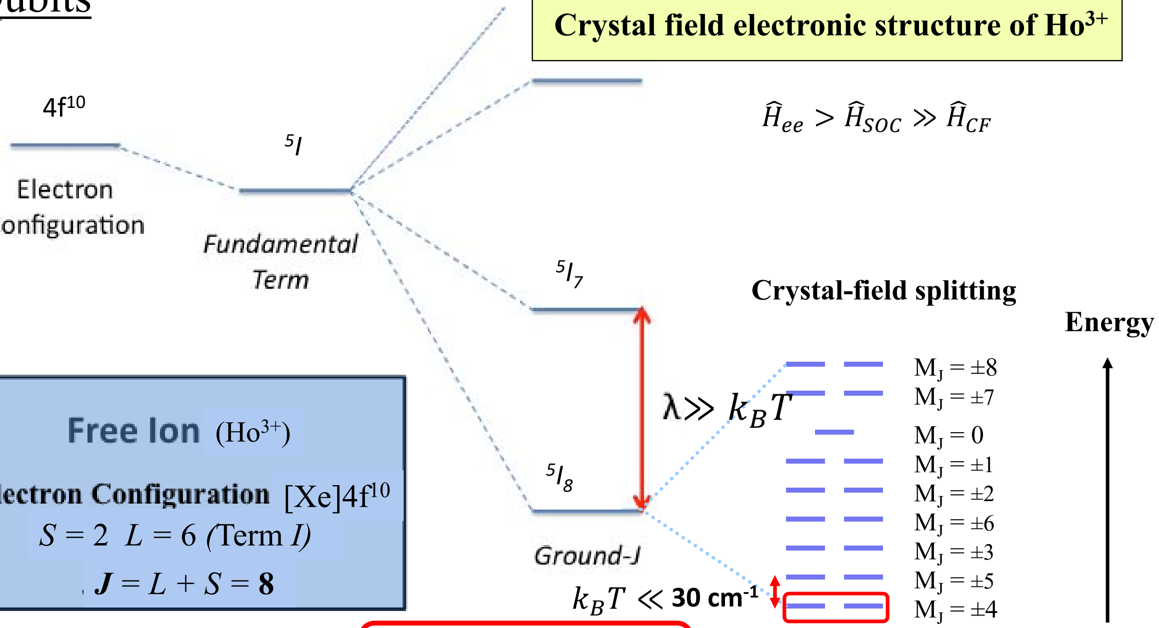
## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Spin Qubits



**$[Ho^{3+}(W_5O_{18})_2]^{9-}$**
**$(HoW_{10})$**

**Crystal field electronic structure of $Ho^{3+}$**




$\hat{H}_{ee} > \hat{H}_{SOC} \gg \hat{H}_{CF}$

**Free Ion** ($Ho^{3+}$)

**Electron Configuration** $[Xe]4f^{10}$

$S = 2 \;\; L = 6$ (Term $I$)

$\boldsymbol{J = L + S = 8}$

**Well-isolated Ground Doublet** $m_J = \pm 4$

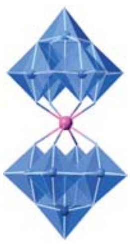
$\hat{H} = B_2^0\hat{O}_2^0 + B_4^0\hat{O}_4^0 + B_6^0\hat{O}_6^0$



## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Spin Qubits

Slight deviation from $D_{4d}$ symmetry

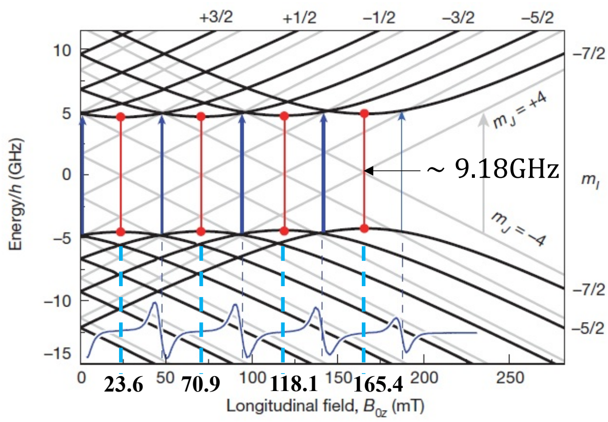


$B_2^0, B_4^0, B_6^0, B_4^4 \rightarrow$


$\hat{H} = B_2^0\hat{O}_2^0 + B_4^0\hat{O}_4^0 + B_6^0\hat{O}_6^0 + B_4^4\hat{O}_4^4 + A_{\parallel}\hat{J}_z\hat{I}_z + \mu_B g_J B_{0z}\hat{J}_z$

$[Ho^{3+}(W_5O_{18})_2]^{9-}$
$(HoW_{10})$

$(m_J = \pm 4) + (I = 7/2) \rightarrow$ **16 low-lying states**



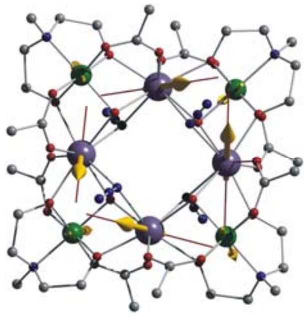

LETTER Nature **531**, 348-351 (2016)
doi:10.1038/nature16984

**Enhancing coherence in molecular spin qubits via atomic clock transitions**

Muhandis Shiddiq[1]*, Dorsa Komijani[1]*, Yan Duan[2], Alejandro Gaita-Ariño[2], Eugenio Coronado[2] & Stephen Hill[1]

## Systems ➡ Applications ➡ Characterization ➡ Modeling

Molecular Spin Qubits



**Physical platform: qubits encoded in Spin States of Magnetic Molecules**

**Non-zero Spin State (Unpaired Electrons)**
**$0 \neq S \geq 1/2$**

↓

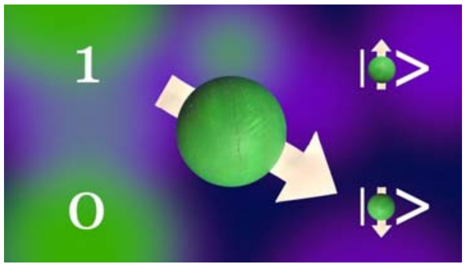
**$2S+1 \geq 2$ Spin Projections are now the energy levels**

**S = 1/2**
**$2^1 = 2$ Spin States**
**1 Qubit**

$|1\rangle$

$|0\rangle$

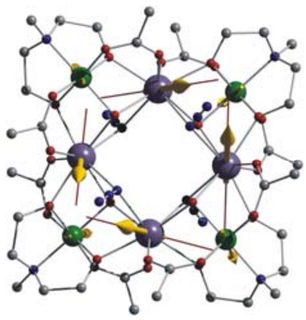
Quantum Two-Level System






## Systems ➡ Applications ➡ Characterization ➡ Modeling

Molecular Spin Qubits



**Physical platform: qubits encoded in Spin States of Magnetic Molecules**

**Basic requirements**

**$2^n$ Spin States**
**n Qubits**

$|1_1 \ldots 1_n\rangle$

$\vdots$

$|1_1 \ldots 0_n\rangle$

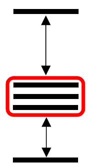
$|0_1 \ldots 0_n\rangle$

$2^n$ Labels

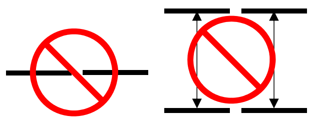
$$|\Psi(t)\rangle = \sum_{j=1}^{2^n} C_{1_j,\ldots,n_j} \left| x_{1_j} \ldots x_{n_j} \right\rangle$$

$$x_{k_j} \in \{0_{k_j}, 1_{k_j}\}$$

- Well-isolated states



- Non – degenerate states and distinguishable transitions



- To be able to drive all transitions between any pair of states

## Systems → Applications → Characterization → Modeling

Molecular Spin Qubits

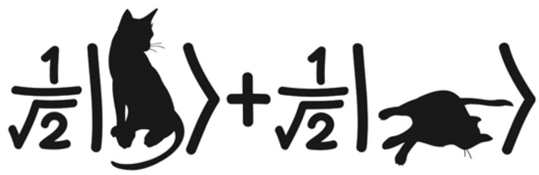
### Classical Computing

**BIT**

$|0\rangle$ $|1\rangle$

### Quantum Computing

R. P. Feynman, Int. J. Theo. Phys. 21, 467 (1982)



**QUBIT**

$|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle \quad \alpha, \beta \in \mathbb{C}$



## Systems → Applications → Characterization → Modeling

Molecular Spin Qubits

### Advantage

↓

**Quantum Algorithms can provide a better scaling with the problem size**

Faster Database Searching

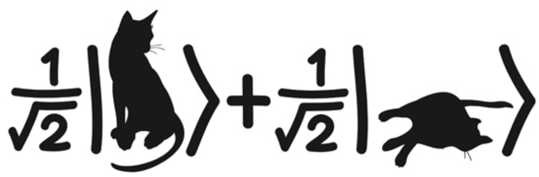
Classical: O(n)

Quantum: $O(n^{1/2})$

(Grover's algorithm)

### Quantum Computing

R. P. Feynman, Int. J. Theo. Phys. 21, 467 (1982)



**QUBIT**

$|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle \quad \alpha, \beta \in \mathbb{C}$

Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Spin Qubits

### Gate-based Quantum Computing

Input

$|\psi_1^i\rangle = \alpha_1^i|0_1\rangle + \beta_1^i|1_1\rangle$

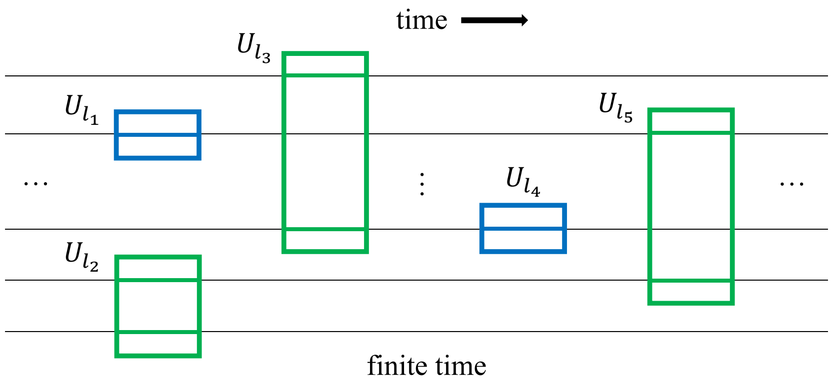
$|\psi_2^i\rangle = \alpha_2^i|0_1\rangle + \beta_2^i|1_1\rangle$

$\vdots$

$|\psi_{n-2}^i\rangle = \alpha_{n-2}^i|0_1\rangle + \beta_{n-2}^i|1_1\rangle$

$|\psi_{n-1}^i\rangle = \alpha_{n-1}^i|0_1\rangle + \beta_{n-1}^i|1_1\rangle$

$|\psi_n^i\rangle = \alpha_n^i|0_1\rangle + \beta_n^i|1_1\rangle$




Output measurement

$|\psi_1^o\rangle = 0_1$ or $1_1$

$|\psi_2^o\rangle = 0_2$ or $1_2$

$\vdots$

$|\psi_{n-2}^o\rangle = 0_{n-2}$ or $1_{n-2}$

$|\psi_{n-1}^o\rangle = 0_{n-1}$ or $1_{n-1}$

$|\psi_n^o\rangle = 0_n$ or $1_n$

$$|\Psi^i\rangle = |\psi_1^i\rangle \otimes \cdots \otimes |\psi_n^i\rangle$$

n qubits

Gate-based Quantum Algorithm

$$|\Psi^o\rangle = \overbrace{U_L U_{L-1} \cdots U_2 U_1}^{\text{finite time}} |\Psi^i\rangle$$

1-qubit gate (blue) 2-qubit gate (green)

$$|\Psi^o\rangle = \sum_{j=1}^{2^n} C_{1_j,\ldots,n_j}^o \left|x_{1_j} \ldots x_{n_j}\right\rangle$$

$$x_{k_j} \in \left\{0_{k_j}, 1_{k_j}\right\}$$

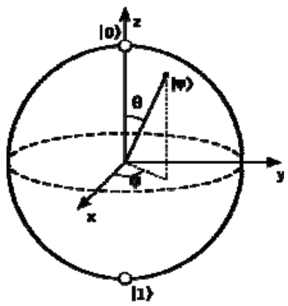


Systems ➡ Applications ➡ Characterization ➡ Modeling

Molecular Spin Qubits

Gate-based Quantum Computing

Bloch Sphere




$$|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle \qquad \alpha, \beta \in \mathbb{C}, |\alpha|^2 + |\beta|^2 = 1$$

⬇

$$|\Psi\rangle = \cos\left(\frac{\theta}{2}\right)|0\rangle + e^{i\varphi}\sin\left(\frac{\theta}{2}\right)|1\rangle \qquad \theta \in [0,\pi] \quad \varphi \in [0,2\pi]$$

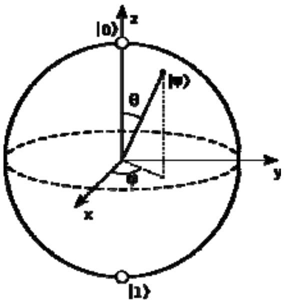
Systems ➡ **Applications** ➡ Characterization ➡ Modeling

## Molecular Spin Qubits

### Gate-based Quantum Computing

Bloch

Sphere




$|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle \qquad \alpha, \beta \in \mathbb{C}, |\alpha|^2 + |\beta|^2 = 1$

⬇

$|\Psi\rangle = \cos\left(\frac{\theta}{2}\right)|0\rangle + e^{i\varphi}\sin\left(\frac{\theta}{2}\right)|1\rangle \qquad \theta \in [0, \pi], \ \varphi \in [0, 2\pi]$

1-qubit gates

Pauli X: $X$

$|0\rangle \to |1\rangle$

$|1\rangle \to |0\rangle$

$|\Delta\theta| = \pi, \varphi = \pi$

Pauli Y: $Y$

$|0\rangle \to \mathbb{i}|1\rangle$

$|1\rangle \to -\mathbb{i}|0\rangle$

$|\Delta\theta| = \pi, \varphi = \pi/2$

Hadamard: $H$

$|0\rangle \to \frac{1}{\sqrt{2}}(|0\rangle + e^{\mathbb{i}\varphi}|1\rangle)$

$|1\rangle \to \frac{1}{\sqrt{2}}(|0\rangle - e^{\mathbb{i}\varphi}|1\rangle)$

$|\Delta\theta| = \pi/2$

Phase: $P$

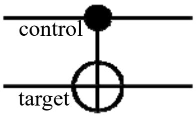
$|0\rangle \to |0\rangle$

$|1\rangle \to e^{\mathbb{i}\Delta\varphi}|1\rangle$

$\theta$ fixed, $\Delta\varphi = \varphi_f - \varphi_i$

2-qubit gates

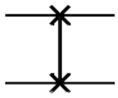
CNOT




$c: |0\rangle \begin{cases} t: |0\rangle \to |0\rangle \\ t: |1\rangle \to |1\rangle \end{cases}$

$c: |1\rangle \begin{cases} t: |0\rangle \to |1\rangle \\ t: |1\rangle \to |0\rangle \end{cases}$

SWAP





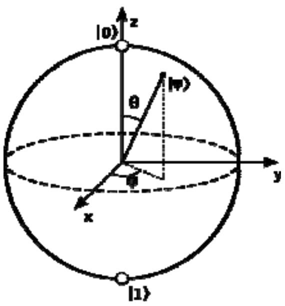

Systems ➡ Applications ➡ Characterization ➡ Modeling

## Molecular Spin Qubits

### Gate-based Quantum Computing: implementation of 1-qubit gates

Bloch

Sphere




$|u_+\rangle \equiv |1\rangle$

$\omega_{+-} = (u_+ - u_-)/\hbar$

Larmor frequency

$|u_-\rangle \equiv |0\rangle$

Phase: $P$

$|0\rangle \to |0\rangle$

$|1\rangle \to e^{\mathbb{i}\Delta\varphi}|1\rangle$

$\theta$ fixed, $\Delta\varphi = \varphi_f - \varphi_i$

**Larmor precession**

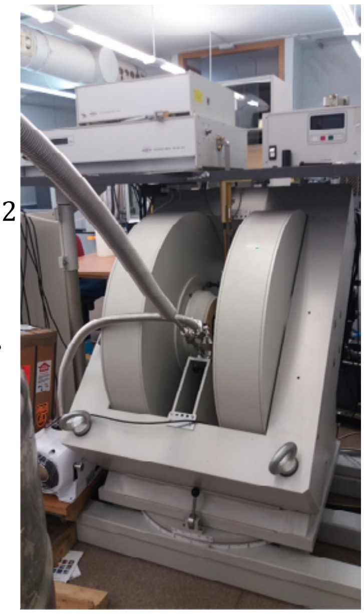
+

**Proper waiting time (free evolution)**

(Pulse) EPR Spectroscopy

## Molecular Spin Qubits

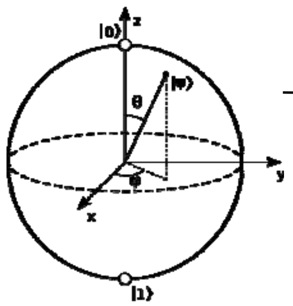
(Pulse) EPR Spectroscopy



Gate-based Quantum Computing: implementation of 1-qubit gates

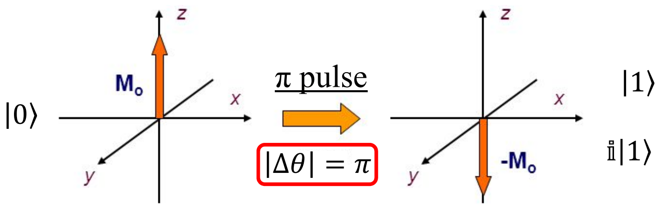
Bloch

Sphere




Pauli X: $X$ — $|0\rangle \rightarrow |1\rangle$, $|1\rangle \rightarrow |0\rangle$, $|\Delta\theta| = \pi, \varphi = \pi$

Pauli Y: $Y$ — $|0\rangle \rightarrow \mathbb{i}|1\rangle$, $|1\rangle \rightarrow -\mathbb{i}|0\rangle$, $|\Delta\theta| = \pi, \varphi = \pi/2$




Oscillating magnetic field

$\Omega_g$ Generalized Rabi frequency

$|u_+\rangle \equiv |1\rangle$

$\omega_{+-} = (u_+ - u_-)/\hbar$ Larmor frequency

$|u_-\rangle \equiv |0\rangle$

$\Omega_g = \sqrt{|\Omega_R|^2 + \delta^2}$

**Resonant radiation pulse (10-100 ns)**
+
**Proper rotation time (driven evolution)**





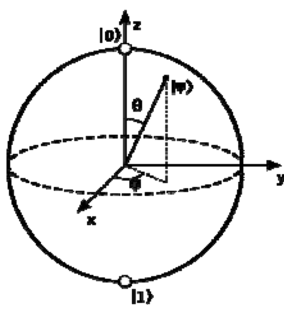
## Molecular Spin Qubits

(Pulse) EPR Spectroscopy

Gate-based Quantum Computing: implementation of 1-qubit gates

Bloch

Sphere

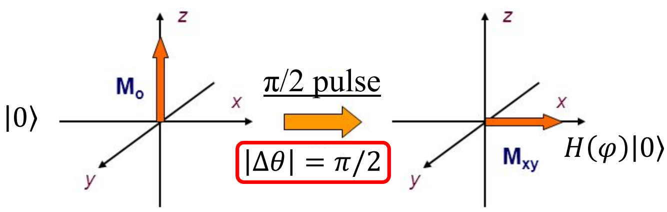



Hadamard: $H$

$|0\rangle \rightarrow \frac{1}{\sqrt{2}}(|0\rangle + e^{\mathbb{i}\varphi}|1\rangle)$

$|1\rangle \rightarrow \frac{1}{\sqrt{2}}(|0\rangle - e^{\mathbb{i}\varphi}|1\rangle)$

$|\Delta\theta| = \pi/2$




Oscillating magnetic field

$\Omega_g$ Generalized Rabi frequency

$|u_+\rangle \equiv |1\rangle$

$\omega_{+-} = (u_+ - u_-)/\hbar$ Larmor frequency

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$\Omega_g = \sqrt{|\Omega_R|^2 + \delta^2}$

**Resonant radiation pulse (10-100 ns)**
+
**Proper rotation time (driven evolution)**

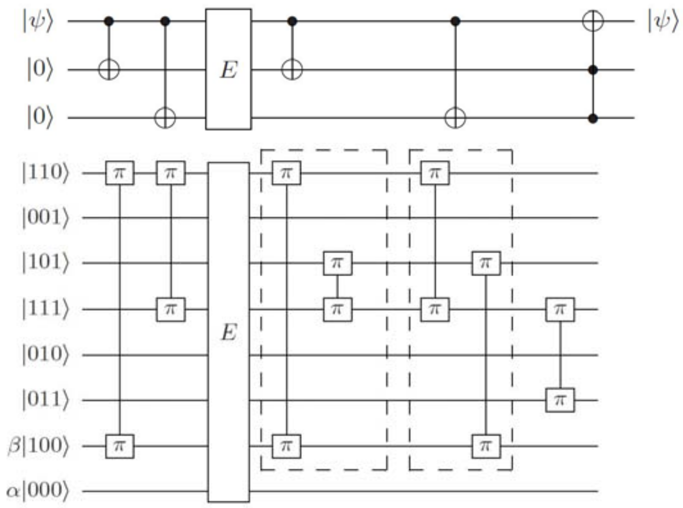
## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Spin Qubits

Gate-based Quantum Computing

**Sequence of Quantum Gates** ➡ **Sequence of EPR Pulses**

Shor's flip error correction code




3 qubits, $2^3$ = spin states

Computational Basis Set:

$|000\rangle, |001\rangle, |010\rangle, |011\rangle$

$|100\rangle, |101\rangle, |110\rangle, |111\rangle$

(Pulse) EPR Spectroscopy



## Systems ➡ Applications ➡ Characterization ➡ Modeling

### Molecular Spin Qubits

Gate-based Quantum Computing

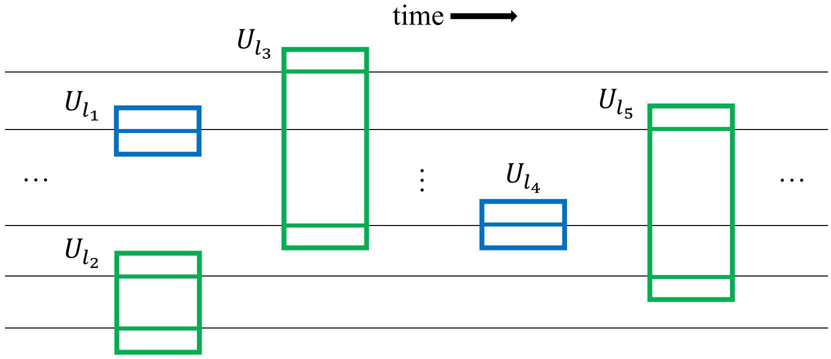
Input

time ⟶

Output measurement

$|\Psi^i\rangle = |\psi_1^i\rangle \otimes \cdots \otimes |\psi_n^i\rangle$

n qubits




$|\Psi^o\rangle = \sum_{j=1}^{2^n} C^o_{1_j,\ldots,n_j} \left|x_{1_j} \ldots x_{n_j}\right\rangle$

$x_{k_j} \in \{0_{k_j}, 1_{k_j}\}$

Gate-based Quantum Algorithm

$|\Psi^o\rangle = \overbrace{U_L U_{L-1} \cdots U_2 U_1}^{\text{finite time}} |\Psi^i\rangle$

1-qubit gate — 2-qubit gate

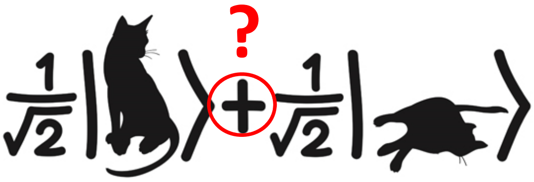
no computation errors: **unitary** dynamics

$\varrho^i = |\Psi^i\rangle\langle\Psi^i| \rightarrow i\hbar \frac{\partial \varrho}{\partial t} = [\widehat{H}_l, \varrho] \quad U_l = e^{-i\widehat{H}_l \Delta t_l/\hbar} \rightarrow \varrho^o \quad \mathrm{Tr}[\hat{O}\varrho^o]$

Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

Gate-based Quantum Computing




**PROBLEM:** Qubits are open systems: uncontrolled interactions qubit-environment → Quantum Superposition destroyed by Decoherence

$$|\Psi\rangle = \alpha(t/T)|0\rangle + \beta(t/T)|1\rangle \quad \longrightarrow \quad |\Psi\rangle \to |0\rangle \ \textbf{or} \ |\Psi\rangle \to |1\rangle \quad t \to +\infty$$

$T$ = characteristic timescale of the decoherence process

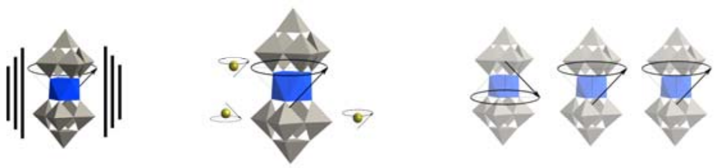


Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

Gate-based Quantum Computing

**Important decoherence mechanisms**



**Vibration bath**

**(molecular and lattice vibrations)**

**spin-orbit interaction**

**Spin bath**

**(non-excited spins)**

**magnetic dipolar interaction**

$\omega_{+-} \approx \omega_{MW}$

$\omega_{+-} \ll \omega_{MW}$ | $\Delta\omega_{MW}$ | $\omega_{+-} \gg \omega_{MW}$

non-excited | excited | non-excited $\quad \hbar\omega$

$\Delta\omega_{MW} \lesssim 100$ MHz

$\Delta t_{pulse} \gtrsim 10$ ns

Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

### Gate-based Quantum Computing

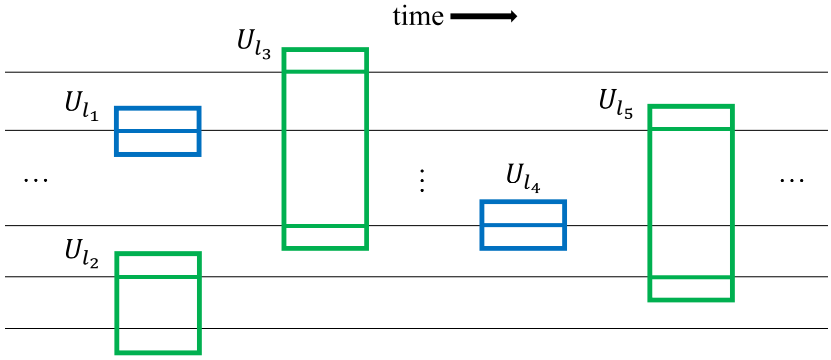
Input

$|\Psi^i\rangle = |\psi_1^i\rangle \otimes \cdots \otimes |\psi_n^i\rangle$

$\varrho^i = |\Psi^i\rangle\langle\Psi^i|$

n qubits

time ⟶




Output measurement

$|\Psi^o\rangle = U_L U_{L-1} \cdots U_2 U_1 \, |\Psi^i\rangle$

Expected: $\varrho^o = |\Psi^o\rangle\langle\Psi^o|$

Measured: $\varrho^m, \neq \varrho^o, \neq |\Psi\rangle\langle\Psi|$

**Real experiments** { decoherence, imperfections } ➡

computation errors: **non-unitary** dynamics

$\varrho^i = |\Psi^i\rangle\langle\Psi^i|$ ➡ $i\hbar \frac{\partial \varrho}{\partial t} = [\widehat{H}_l, \varrho] + \cdots$ $\quad U_l \neq e^{-i\widehat{H}_l \Delta t_l/\hbar}$ ➡ $\varrho^c \approx \varrho^m$? $\mathrm{Tr}[\widehat{O}\varrho^c]$



Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

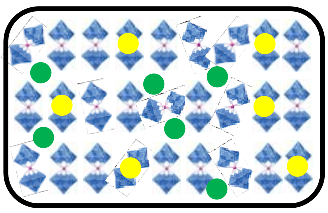
### Gate-based Quantum Computing

**Real experiments** { decoherence ➡ Try lengthening decoherence timescale $T$; imperfections }

**Shorter timescale:** phase memory time $T_m$

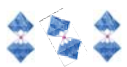
$|\Psi\rangle = \cos\left(\frac{\theta}{2}\right)|0\rangle + e^{i\varphi}\sin\left(\frac{\theta}{2}\right)|1\rangle$ How long $\varphi$ is ≈ the expected one?

Each qubit feels a different: $\delta\vec{B}_i$ ⟶ **unknown extra phase**

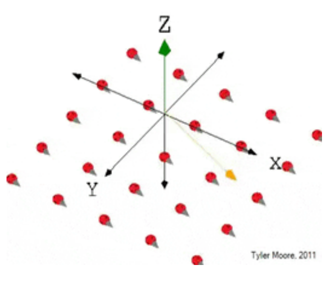


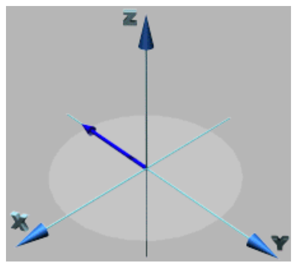
-Spin bath ● ●

-Different local environments



$|\Psi_i\rangle = (|0\rangle + e^{i\varphi}|1\rangle)/\sqrt{2} \xrightarrow{t \gtrsim T_m} |\Psi_i\rangle = (|0\rangle + e^{i\varphi + \delta\varphi_i}|1\rangle)/\sqrt{2}$




Each qubit has a different:

$|u_+^i\rangle \equiv |1\rangle$

$\omega_{+-}^i = (u_+^i - u_-^i)/\hbar$ ➡ **spin dephasing**

$|u_-^i\rangle \equiv |0\rangle$

## Systems ➡ Applications ➡ Characterization ➡ Modeling

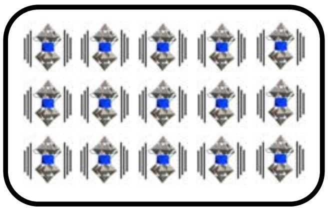
## Molecular Spin Qubits

Gate-based Quantum Computing

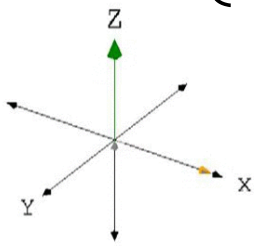
**Real experiments** { decoherence ➡ Try lengthening decoherence timescale $T$; imperfections }

**Longer timescale:** spin-lattice relaxation time $T_1$  $|\Psi\rangle = \cos\left(\frac{\theta}{2}\right)|0\rangle + e^{i\varphi}\sin\left(\frac{\theta}{2}\right)|1\rangle$  How long $\theta$ is ≈ the expected one?

Each qubit tries to reach the thermodynamic equilibrium $|\Psi_i\rangle = \left(|0\rangle + e^{i\varphi}|1\rangle\right)/\sqrt{2}$ $\xrightarrow{t \gtrsim T_1}$ { $|\Psi_i\rangle = |1\rangle$ or $|\Psi_i\rangle = |0\rangle$ }



e.g. by exchanging energy with the vibration bath




Tyler Moore, 2011

➡ **spin thermalization**





## Systems ➡ Applications ➡ Characterization ➡ Modeling

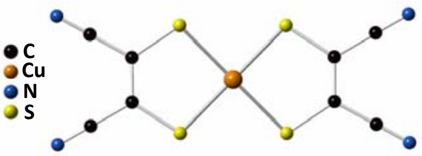
## Molecular Spin Qubits

Gate-based Quantum Computing

**Real experiments** { decoherence ➡ Try lengthening decoherence timescale $T$; imperfections }

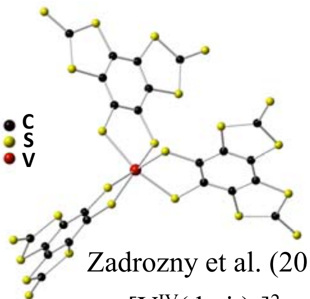


Removal of surrounding spin bath




Bader et al. (2014) $[Cu^{II}(mnt)_2]^{2-}$

$T_m(7K) = 68\ \mu s$



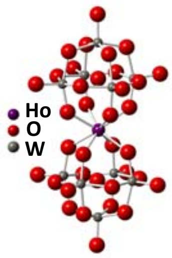

Zadrozny et al. (2015)

$[V^{IV}(dmit)_3]^{2-}$

$T_m(10K) = 675\ \mu s$

**no H, N atoms, change H by D**

Clock Transitions

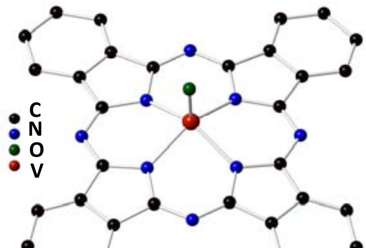



Shiddiq et al. (2016)

$[Ho^{III}_x Y_{1-x}(W_5O_{18})_2]^{9-}$

$T_m(5K, x = 0.1) = 0.7\ \mu s$

Key Role of Molecular Rigidity




$[V^{IV}OPc]$

Atzori et al. (2016)

$T_m(300K) = 1\ \mu s$

Systems ➡ Applications ➡ Characterization ➡ Modeling

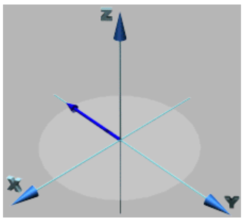
## Molecular Spin Qubits

Gate-based Quantum Computing

**Real experiments** { decoherence ➡ Try lengthening decoherence timescale $T$ ; imperfections

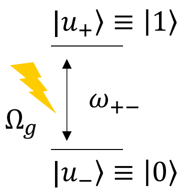
Take-home message:
To increase $T_m$ $T_1$ ?

Application? {
free evolution?
free + driven evolution? (gate-based algorithms)



to store quantum information

figure of merit: $T_m$ $T_1$




to compute quantum information

figure of merit: $T_m$ $T_1$ and…?



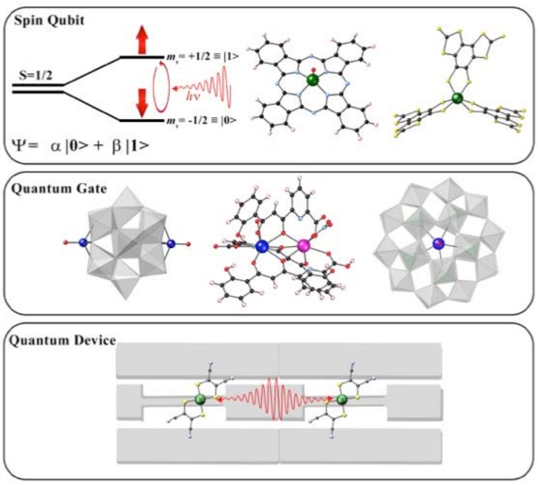
Systems ➡ Applications ➡ Characterization ➡ Modeling

Molecular Spin Qubits

Gate-based Quantum Computing

**Real experiments** { decoherence ➡ Try lengthening decoherence timescale $T$ ; imperfections

Take-home message:
To increase $T_m$ $T_1$ ?




E. Coronado, *Nat. Rev. Mater.*, **5**, 87-104 (2020)

(single) Spin **coherent manipulation**

Implementation of **quantum gates** and **few-qubit algorithms**

**Scalable architecture projects**
SUMO (Scaling Up with Molecular qubits, QuantERA Call 2017)
FATMOLS (FAult Tolerant MOLecular Spin processor, Horizon 2020)

Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

Gate-based Quantum Computing

**Real experiments**
- decoherence ➡ Try lengthening decoherence timescale $T$
- imperfections

Take-home message:
To increase $T_m$ $T_1$ ?

Test of qubits for computation:

-as a memory for info. storage / waiting times ($T_m$ $T_1$)
-implementing the desired gate-based algorithm
-within the quantum device

⬇

free and driven evolution + decoherence/imperfections

Error characterization protocols in algorithms: gates, state preparation, measurement; **Figure of merit**: fidelity (density matrix)

Certification protocols of quantum devices: without running algorithms, e.g. validation of state preparation; **Figure of merit**: success probability

Optimal control protocols



Systems ➡ Applications ➡ **Characterization** ➡ Modeling

## Molecular Spin Qubits

Gate-based Quantum Computing

**Real experiments**
- decoherence ➡ Try lengthening decoherence timescale $T$
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Take-home message:
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-implementing the desired gate-based algorithm
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⬇

free and driven evolution + decoherence/imperfections

Error characterization protocols in algorithms: gates, state preparation, measurement; **Figure of merit**: fidelity (density matrix)

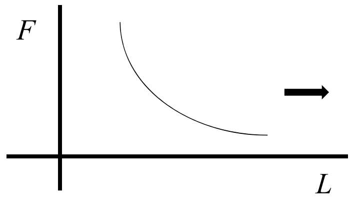
Certification protocols of quantum devices: without running algorithms, e.g. validation of state preparation; **Figure of merit**: success probability

Optimal control protocols

Randomized benchmarking

Random sequence of gates

$U_1^{-1}U_2^{-1}\cdots U_{L-1}^{1}U_L^{-1}U_L U_{L-1}\cdots U_2 U_1$ { Expected: $F = 1$; Observed: $F < 1$




➡ Fit to a mathematical model: info. on errors

Efficient scaling with the number of qubits

Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

Gate-based Quantum Computing

**Real experiments** { decoherence, imperfections } ➡ Try increasing **fidelity**

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

one spin-qubit dynamics:

$\varrho_{11}$ $|u_+\rangle \equiv |1\rangle$

$\varrho_{22}$ $|u_-\rangle \equiv |0\rangle$

$$\dot{\hat{\rho}} = \frac{1}{i\hbar}\left[\hat{H}_{eff}, \hat{\rho}\right] + \Gamma_{ab}\mathcal{L}[L]\hat{\rho} + \Gamma_{em}\mathcal{L}\left[L^\dagger\right]\hat{\rho} + \Gamma_{mag}\mathcal{L}_{mag}\hat{\rho}$$

Lindblad master equation

$$\Gamma_{ab} = \Gamma^{ab,1p}_{|u_-\rangle\to|u_+\rangle} + \Gamma^{ab,2p}_{|u_-\rangle\to|u_+\rangle} \qquad \Gamma_{em} = \Gamma^{em,1p}_{|u_+\rangle\to|u_-\rangle} + \Gamma^{em,2p}_{|u_+\rangle\to|u_-\rangle} \qquad \Gamma_{mag} = \frac{1}{T_n} + \frac{1}{T_e}$$

decoherence rates

$$\hat{H}_{eff} = \hat{H}_0 + \hat{H}_1 = \frac{1}{2}(u_+ - u_-)\left(|u_+\rangle\langle u_+| - |u_-\rangle\langle u_-|\right) + \frac{\mu_B g_I}{\hbar}\sum_{\gamma=x,y,z}\left(\langle u_-|\hat{J}_\gamma|u_+\rangle|u_+\rangle\langle u_-| + c.c.\right)B_{1\gamma}\cos\left(\omega_{\mathrm{MW}}(t-t_0)\right)$$

driving field

analytical resolution (no numerical methods)



Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

Gate-based Quantum Computing

**Real experiments** { decoherence, imperfections } ➡ Try increasing **fidelity**

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

Hamiltonian and decoherence rates:

$(2J_g+1)(2I_g+1)$ states

$$J_g, I_g \quad \hat{H} = \sum_{k=2,4,6}\sum_{q=-k}^{+k} B_k^q \hat{O}_k^q + \mu_B \sum_{\alpha=x,y,z} g_\alpha B_\alpha \hat{J}_\alpha + A_\perp\left(\hat{J}_x\hat{I}_x + \hat{J}_y\hat{I}_y\right) + A_\parallel \hat{J}_z\hat{I}_z + P\hat{I}_z^2$$

➡ $|u_+\rangle \equiv |1\rangle$, $|u_-\rangle \equiv |0\rangle$ Select two proper states as a qubit

vibration bath:

$$\Gamma_{ab} = \Gamma^{ab,1p}_{|u_-\rangle\to|u_+\rangle} + \Gamma^{ab,2p}_{|u_-\rangle\to|u_+\rangle}$$

$$\Gamma_{em} = \Gamma^{em,1p}_{|u_+\rangle\to|u_-\rangle} + \Gamma^{em,2p}_{|u_+\rangle\to|u_-\rangle}$$

**L. Escalera-Moreno**, N. Suaud, A. Gaita-Ariño, E. Coronado, *J. Phys. Chem. Lett.* **8**(7), 1695-1700 (2017)

A. Lunghi, S. Sanvito, *J. Phys. Chem. Lett.* **11**(15), 6273-6278 (2020)

A. Lunghi, *Sci. Adv.* **8**(31), (2022)

spin bath:

$$\Gamma_{mag} = \frac{1}{T_n} + \frac{1}{T_e}$$

P. C. E. Stamp, I. S. Tupitsyn, *Phys. Rev. B* **69**, 014401 (2004)

S. J. Balian et al., *Phys. Rev. B* **89**, 045403 (2014) M. Warnet et al., *Nature* **503**, 504-508 (2013)

**L. Escalera-Moreno**, A. Gaita-Ariño, E. Coronado, *Phys. Rev. B* **100**(6), 064405 (2019)

**taking up the torch of *ab initio* calculations**

Systems ➡ Applications ➡ Characterization ➡ **Modeling**

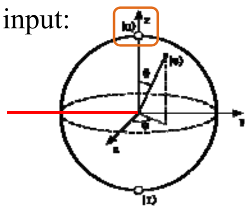
## Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

software: **QBithm** **one-qubit gates and algorithms (decoherence rates = 0, no imperfections)**

input:



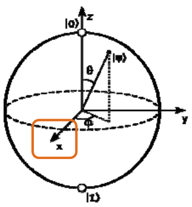
$S = 1/2$ $\varrho_{11}$ $|+1/2\rangle \equiv |1\rangle$, $\varrho_{22}$ $|-1/2\rangle \equiv |0\rangle$

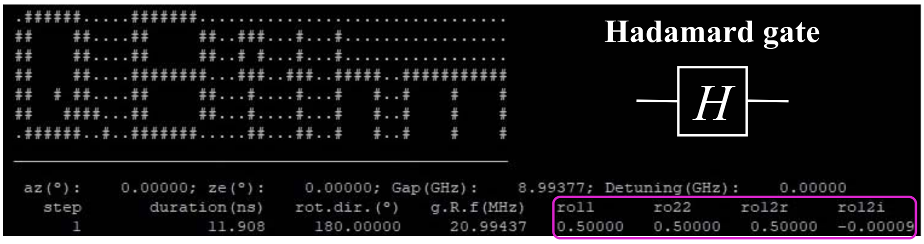
$|\Psi(t=0)\rangle = |0\rangle$ $\varrho(t=0) = |\Psi\rangle\langle\Psi|(t=0) = \begin{pmatrix} \varrho_{11} & \varrho_{12,r} + i\varrho_{12,i} \\ \varrho_{12,r} - i\varrho_{12,i} & \varrho_{22} \end{pmatrix} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}$

```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
2    11.908           180.0
```

$\omega_{MW}, |\vec{B_1}|$ rotation time: $\Delta t = \frac{2\pi + \Delta\theta}{\Omega_g}$ if $\Delta\theta < 0$ $\Delta t = \frac{\Delta\theta}{\Omega_g}$ if $\Delta\theta > 0$

output:






$H|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$

$\varrho = H|0\rangle(H|0\rangle)^\dagger = \begin{pmatrix} \frac{1}{2} & \frac{1}{2} \\ \frac{1}{2} & \frac{1}{2} \end{pmatrix}$



Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

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**free and driven evolution + decoherence/imperfections**

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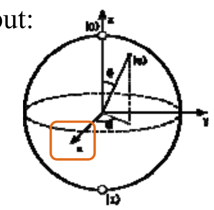
input:



$S = 1/2$ $\varrho_{11}$ $|+1/2\rangle \equiv |1\rangle$, $\varrho_{22}$ $|-1/2\rangle \equiv |0\rangle$

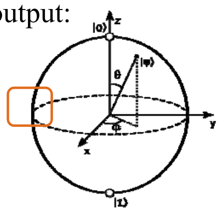
$H|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ $\varrho = H|0\rangle(H|0\rangle)^\dagger = \begin{pmatrix} \frac{1}{2} & \frac{1}{2} \\ \frac{1}{2} & \frac{1}{2} \end{pmatrix}$

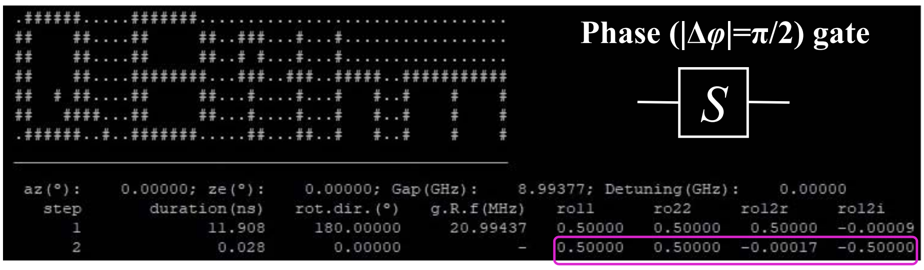
```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
2    11.908           180.0
1    0.0278           0.0
```

→ **not used**

$\omega_{MW} = 0, |\vec{B_1}| = 0$ waiting time: $\Delta t = -\frac{\Delta\varphi}{\omega_{+-}}$ if $\Delta\varphi < 0$ $\Delta t = \frac{2\pi - \Delta\varphi}{\omega_{+-}}$ if $\Delta\varphi > 0$

output:






$SH|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle - i|1\rangle)$

$\varrho = SH|0\rangle(SH|0\rangle)^\dagger = \begin{pmatrix} \frac{1}{2} & \frac{-i}{2} \\ \frac{i}{2} & \frac{1}{2} \end{pmatrix}$

Systems ➡ Applications ➡ Characterization ➡ **Modeling**

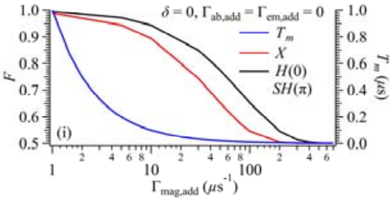
## Spin Qubits

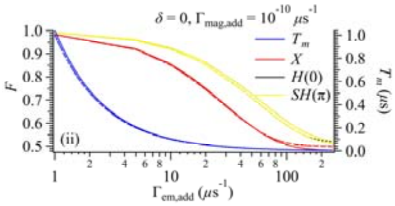
arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

software: **QBithm** **one-qubit gates and algorithms (decoherence rates ≠ 0, imperfections)**

only decoherence rates




expected

calculated

$$F(\varrho^o, \varrho^c) = \mathrm{Tr}(\varrho^o \varrho^c) + 2\sqrt{\det(\varrho^o)\det(\varrho^c)}$$

2x2 matrices

translation $T_m \leftrightarrow F$: $T_m \geq T_m^{min} \rightarrow F \geq F^{min}$

alternative to $Q = T_m/\text{gate time}$: number of gates while $F \geq F^{min}$



Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

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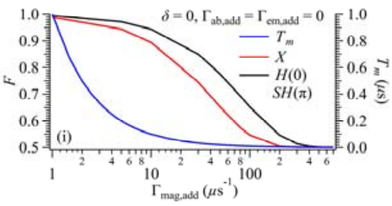
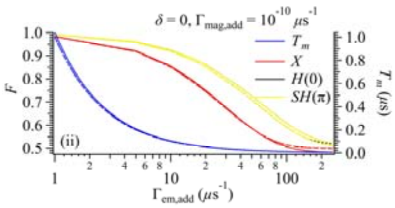
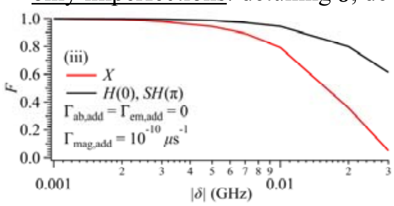
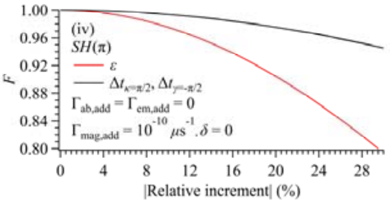


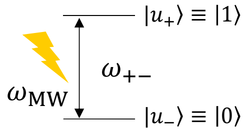


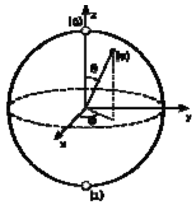
only imperfections: detuning $\delta$, deviations in rotation direction $\varepsilon$, rotation/waiting times $\Delta t$




$\delta = \omega_{+-} - \omega_{MW}$



expected

calculated

$$F(\varrho^o, \varrho^c) = \mathrm{Tr}(\varrho^o \varrho^c) + 2\sqrt{\det(\varrho^o)\det(\varrho^c)}$$

2x2 matrices

translation $T_m \leftrightarrow F$: $T_m \geq T_m^{min} \rightarrow F \geq F^{min}$

alternative to $Q = T_m/\text{gate time}$: number of gates while $F \geq F^{min}$

## Systems ➡ Applications ➡ Characterization ➡ Modeling

# Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

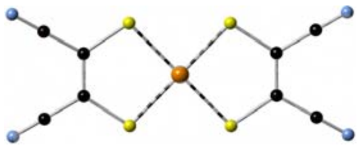
**free and driven evolution + decoherence/imperfections**

software: **QBithm** **determination of Rabi osc., $T_m$, $T_1$, $T_{dd}$**

two operation modes: $\Gamma_{ab}, \Gamma_{em}, \Gamma_{mag}$ { *ab initio* rates / semi-empirical

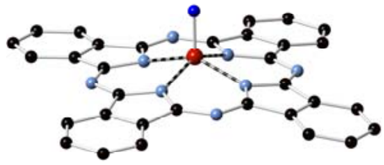
**benchmarking**

$[Cu^{II}(mnt)_2]^{2-}$
$S = 1/2$
$I = 3/2$



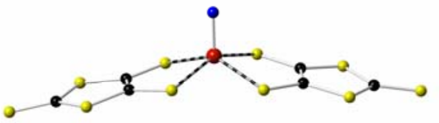
Bader et al., *Nat. Commun.*, **5**, 5304, (2014)

$V^{IV}OPc$
$S = 1/2$
$I = 7/2$



Atzori et al., *J. Am. Chem. Soc.*, **138**(7), 2154-2157, (2016)

$[V^{IV}(dmit)_3]^{2-}$
$S = 1/2$
$I = 7/2$



$[V^{IV}O(dmit)_2]^{2-}$
$S = 1/2$
$I = 7/2$

Atzori et al., *J. Am. Chem. Soc.*, **138**(35), 11234-11244, (2016)



## Systems ➡ Applications ➡ Characterization ➡ Modeling

# Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

software: **QBithm** **determination of Rabi osc., $T_m$, $T_1$, $T_{dd}$**

two operation modes: $\Gamma_{ab}, \Gamma_{em}, \Gamma_{mag}$ { *ab initio* rates / semi-empirical

**combined**

longitudinal magnetization $M_z = \mathrm{Tr}[\widehat{\sigma_z}\varrho^c]$ Rabi oscillations

input:

```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
3    0.0 ──►variable 0.0
```

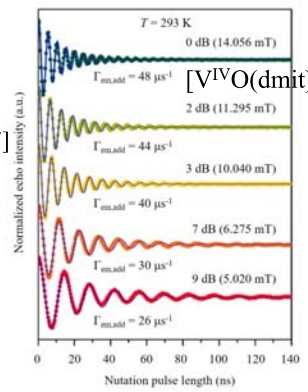
adjusted: $|\overrightarrow{B_1}|$ fitted: $\Gamma_{em}$ calculated: $\Gamma_{mag}, \Gamma_{ab} = \Gamma_{em}\exp(-(u_+ - u_-)/k_B T)$

output:

$\varrho^c$ time evolution

```
-magnetization:      1

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
   step      duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11       ro22       ro12r      ro12i
     1            0.000       0.00000      20.99437   -0.00000    1.00000    0.00000   -0.00000

-magnetization:      2

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
   step      duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11       ro22       ro12r      ro12i
     1            2.500       0.00000      20.99437    0.03104    0.96896   -0.16074    0.00000

-magnetization:      3

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
   step      duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11       ro22       ro12r      ro12i
     1            5.000       0.00000      20.99437    0.11181    0.88819   -0.30197    0.00002
```

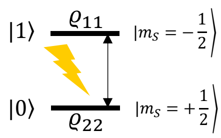
$M_z = \mathrm{Tr}[\widehat{\sigma_z}\varrho^c]$
vs
rotation time




$[V^{IV}O(dmit)_2]^{2-}$

$t \gtrsim \Gamma_{ab}^{-1}, \Gamma_{em}^{-1}, \Gamma_{mag}^{-1} \rightarrow$ { $0.5 \approx \varrho_{22} \gtrsim \varrho_{11} \approx 0.5$ ⟶ spin thermalization; $\varrho_{12} = \varrho_{12,r} + \mathbb{i}\varrho_{12,i} \approx 0$ ⟶ spin dephasing

$[Cu^{II}(mnt)_2]^{2-}$ $V^{IV}OPc$
$[V^{IV}(dmit)_3]^{2-}$
$[V^{IV}O(dmit)_2]^{2-}$




$|\Psi(t=0)\rangle = |0\rangle$

**only one free parameter**
$\Gamma_{em}$

Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

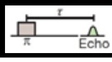
software: **QBithm** **determination of Rabi osc., $T_m$, $T_1$, $T_{dd}$**

two operation modes: $\Gamma_{ab}, \Gamma_{em}, \Gamma_{mag}$ { *ab initio* rates / semi-empirical } **combined**

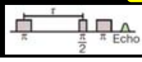
longitudinal magnetization $M_z = Tr[\widehat{\sigma_z}\varrho^c]$ spin-lattice relaxation time $T_1$

input:

```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
2   32.0     0.0
0   0.0 → variable   0.0 → not used
```




or

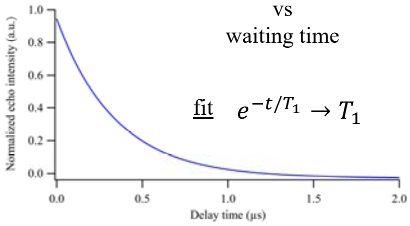



given: $t_\pi = 32$ ns from Rabi osc.: $|\overrightarrow{B_1}|$ fitted: $\Gamma_{em}$ calculated: $\Gamma_{mag}, \Gamma_{ab} = \Gamma_{em}\exp(-(u_+ - u_-)/k_BT)$

output: $\varrho^c$ time evolution

```
-magnetization:       1

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
  step       duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11      ro22      ro12r      ro12i
    1             32.000       0.00000     20.99437   0.97179   0.02821  -0.00061   0.00017
    2              0.000       0.00000            -   0.97179   0.02821  -0.00061   0.00017

-magnetization:       2

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
  step       duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11      ro22      ro12r      ro12i
    1             32.000       0.00000     20.99437   0.97179   0.02821  -0.00061   0.00017
    2             20.000       0.00000            -   0.94424   0.05576  -0.00053  -0.00030

-magnetization:       3

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
  step       duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11      ro22      ro12r      ro12i
    1             32.000       0.00000     20.99437   0.97179   0.02821  -0.00061   0.00017
    2             40.000       0.00000            -   0.91826   0.08174  -0.00016  -0.00057
```

$M_z = Tr[\widehat{\sigma_z}\varrho^c]$ vs waiting time



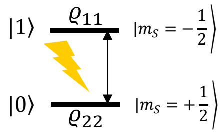

fit $e^{-t/T_1} \rightarrow T_1$

$t \gtrsim \Gamma_{ab}^{-1}, \Gamma_{em}^{-1}, \Gamma_{mag}^{-1} \rightarrow 0.5 \approx \varrho_{22} \gtrsim \varrho_{11} \approx 0.5$ ⟶ spin thermalization

$[Cu^{II}(mnt)_2]^{2-}$ $V^{IV}OPc$

$[V^{IV}(dmit)_3]^{2-}$

$[V^{IV}O(dmit)_2]^{2-}$




$|\Psi(t=0)\rangle = |0\rangle$

**only one free parameter**

$\Gamma_{em}$



Systems ➡ Applications ➡ Characterization ➡ **Modeling**

## Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

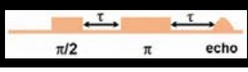
software: **QBithm** **determination of Rabi osc., $T_m$, $T_1$, $T_{dd}$**

two operation modes: $\Gamma_{ab}, \Gamma_{em}, \Gamma_{mag}$ { *ab initio* rates / semi-empirical } **combined**

in-plane magnetization $M_{xy} = Tr[(\widehat{\sigma_x} + \mathbb{i}\widehat{\sigma_z})\varrho^c]$ phase memory time $T_m$

input:

```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
2   16.0     0.0
0   0.0 → variable   0.0 → not used
2   32.0     0.0
0   0.0 → variable   0.0 → not used
```



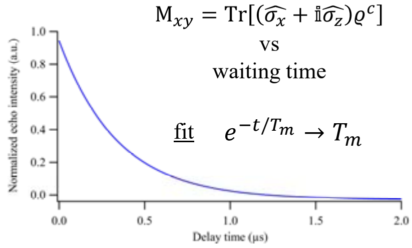

**2-pulse Hahn sequence**

given: $t_\pi = 32$ ns from Rabi osc.: $|\overrightarrow{B_1}|$ fitted: $\Gamma_{em}$ calculated: $\Gamma_{mag}, \Gamma_{ab} = \Gamma_{em}\exp(-(u_+ - u_-)/k_BT)$

output: $\varrho^c$ time evolution

```
-magnetization:       1

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
  step       duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11      ro22      ro12r      ro12i
    1             16.000       0.00000     20.99437   0.50145   0.49855  -0.48600   0.00009
    2              0.000       0.00000            -   0.50145   0.49855  -0.48600   0.00009
    3             32.000       0.00000     20.99437   0.49862   0.50138   0.45798   0.00008
    4              0.000       0.00000            -   0.49862   0.50138   0.45798   0.00008

-magnetization:       2

 az(°):    0.00000; ze(°):    0.00000; Gap(GHz):    8.99377; Detuning(GHz):    0.00000
  step       duration(ns)   rot.dir.(°)   g.R.f(MHz)    ro11      ro22      ro12r      ro12i
    1             16.000       0.00000     20.99437   0.50145   0.49855  -0.48600   0.00009
    2             20.000       0.00000            -   0.50057   0.49943  -0.33150  -0.32941
    3             32.000       0.00000     20.99437   0.49957   0.50043   0.31219  -0.31439
    4             20.000       0.00000            -   0.49879   0.50121   0.42603  -0.00277
```

$M_{xy} = Tr[(\widehat{\sigma_x} + \mathbb{i}\widehat{\sigma_z})\varrho^c]$ vs waiting time



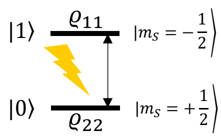

fit $e^{-t/T_m} \rightarrow T_m$

$t \gtrsim \Gamma_{ab}^{-1}, \Gamma_{em}^{-1}, \Gamma_{mag}^{-1} \rightarrow \varrho_{12} = \varrho_{12,r} + \mathbb{i}\varrho_{12,i} \approx 0$ ⟶ spin dephasing

$[Cu^{II}(mnt)_2]^{2-}$ $V^{IV}OPc$

$[V^{IV}(dmit)_3]^{2-}$

$[V^{IV}O(dmit)_2]^{2-}$




$|\Psi(t=0)\rangle = |0\rangle$

**only one free parameter**

$\Gamma_{em}$

## Systems ➔ Applications ➔ Characterization ➔ Modeling

# Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

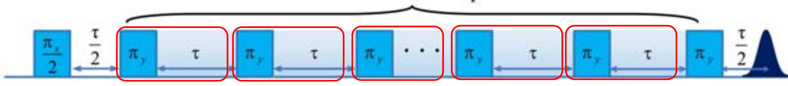
**free and driven evolution + decoherence/imperfections**

software: **QBithm** **determination of Rabi osc., $T_m$, $T_1$, $T_{dd}$**

two operation modes: $\Gamma_{ab}, \Gamma_{em}, \Gamma_{mag}$ { *ab initio* rates / semi-empirical } **combined**

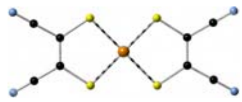
in-plane magnetization $M_{xy} = \mathrm{Tr}[(\widehat{\sigma_x} + i\widehat{\sigma_z})\varrho^c]$ dynamical decoupling $T_{dd}$

n = total number of π pulses



arXiv:1706.09259 Dai et al. (2017) $T_{dd} = 1.4$ ms, $n = 2048$

Bader et al., *Nat. Commun.*, **5**, 5304, (2014) $T_m = 68$ μs

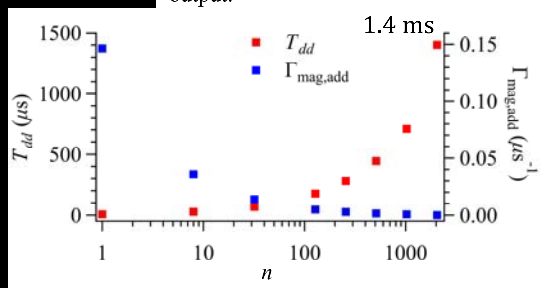


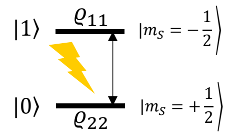
$[Cu^{II}(mnt)_2]^{2-}$

input: $\Gamma_{em}$ fixed, fits $T_{dd} = 1.4$ ms $n = 2048$ with $\Gamma_{mag} = 0$

```
Sequence of the algorithm (0 = variable free evolution / 1 = fixed free evolution / 2 = rotation / 3 = variable rotation)
Step Duration(0<=,ns) Rotation direction (0<=,<360,°)
2    24.0             0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
0    0.0              0.0
2    48.0             90.0
0    0.0              0.0
```

output:




$|\Psi(t = 0)\rangle = |0\rangle$

**only one free parameter** $\Gamma_{mag}$



## Systems ➔ Applications ➔ Characterization ➔ Modeling

# Spin Qubits

arXiv:2303.12655
10.26434/chemrxiv-2023-fw96z-v2

**free and driven evolution + decoherence/imperfections**

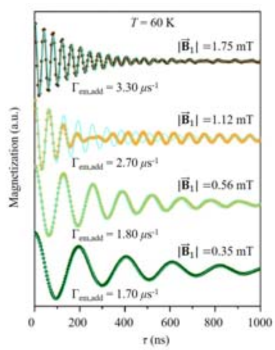
software: **QBithm** **what else?**

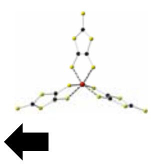
$$\dot{\hat{\rho}} = \frac{1}{i\hbar}[\hat{H}_{eff}, \hat{\rho}] + \Gamma_{ab}\mathcal{L}[L]\hat{\rho} + \Gamma_{em}\mathcal{L}[L^\dagger]\hat{\rho} + \Gamma_{mag}\mathcal{L}_{mag}\hat{\rho} + \cdots ?$$

- stretched magnetization $e^{-(t/T)^k}, k \neq 1$
- leakage imperfection $\varrho_{11} + \varrho_{22} \neq 1$

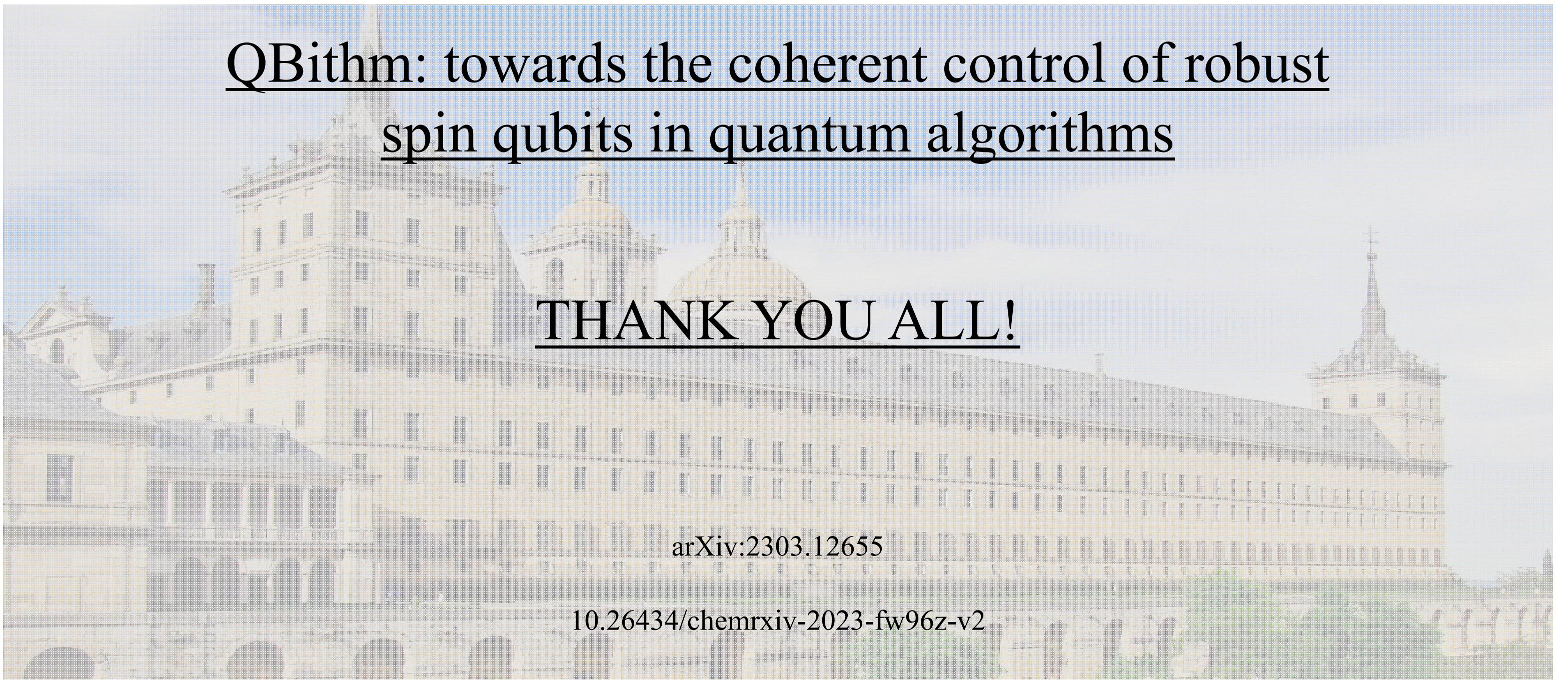
$\varrho_{11}$ |1⟩
$\varrho_{22}$ |0⟩

enlarge Hilbert space
- coupling to nearby nuclear spins { Larmor oscillations / Hartman-Hahn condition in Rabi osc }
- two-qubit gates ($2^2$ x $2^2$ density matrix)
- many-qubits with efficient scaling (alternative to density matrix $2^n$ x $2^n$)



$[V^{IV}(dmit)_3]^{2-}$



# QBithm: towards the coherent control of robust spin qubits in quantum algorithms

## THANK YOU ALL!

arXiv:2303.12655

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May 11th

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ESMolNa European School on Molecular Nanoscience

MAX-PLANCK-INSTITUT FÜR QUANTENOPTIK