Electrical two-qubit gates within a pair of clock-qubit magnetic molecules

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Outline

1. Introduction and objectives

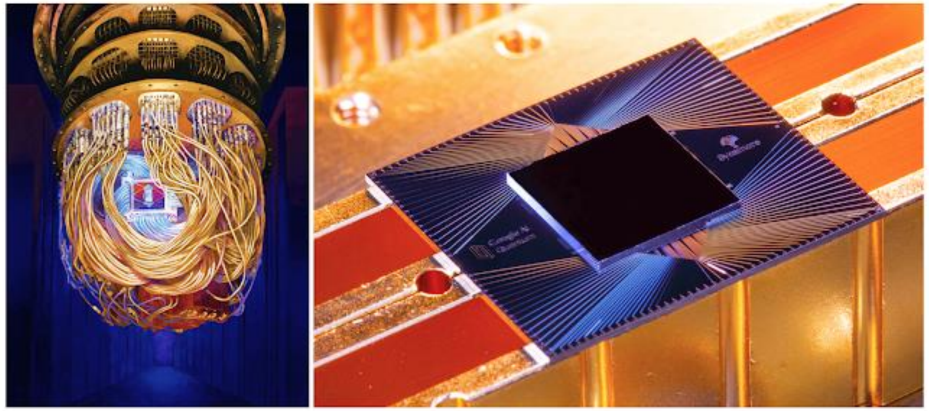
2. Spin electric coupling (SEC) and coherent control over spins

3. Entangling two-qubit gates acting on molecular spin-qubits

4. Conclusions

Introduction and objectives

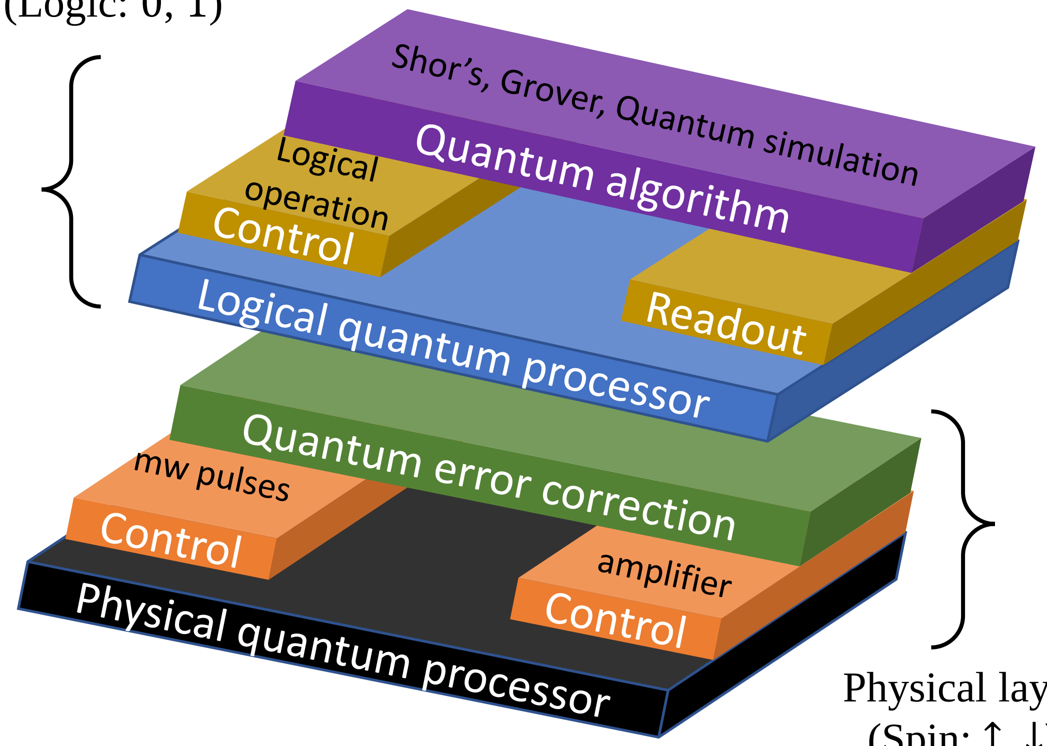
- ✓ A classical computer uses classical bits: either “0” or “1”
- ✓ A quantum computer uses quantum superpositions of “0” and “1”, qubit: $|\varphi\rangle = \alpha|0\rangle + \beta|1\rangle$
- ✓ Non-polynomial (NP) problems such as prime-factorization can be solved using quantum computers
- ✓ Superconducting qubits, nitrogen vacancies in diamond, semiconductor spin-qubits, molecular spin-qubits



Sycamore quantum processor mounted in the cryostat.

Molecular spin-qubit based quantum computing

Logical layer
(Logic: 0, 1)



Physical layer
(Spin: ↑, ↓)

Advantages

- ✓ Solid state approach, embodied on integrated chips
- ✓ Long coherence times (T_m) in 'μs' to 'ms'
- ✓ Rich Hilbert space for molecular spin-qubits
- ✓ Tunable

Challenges

A. Physical realization of qubit operations:

  i. Addressing of individual molecular spin-qubits and entangling 2-qubits

  ii. A path for scalability

B. A precise control of molecular positioning and quantum circuits

General objectives

Addressing of individual molecular spin-qubits and entangling 2-qubits

1. **Quantum coherent control** of molecular spin-qubit using E-field: $|\varphi\rangle = \alpha|0\rangle + \beta|1\rangle$

2. **Entangling two qubits**:

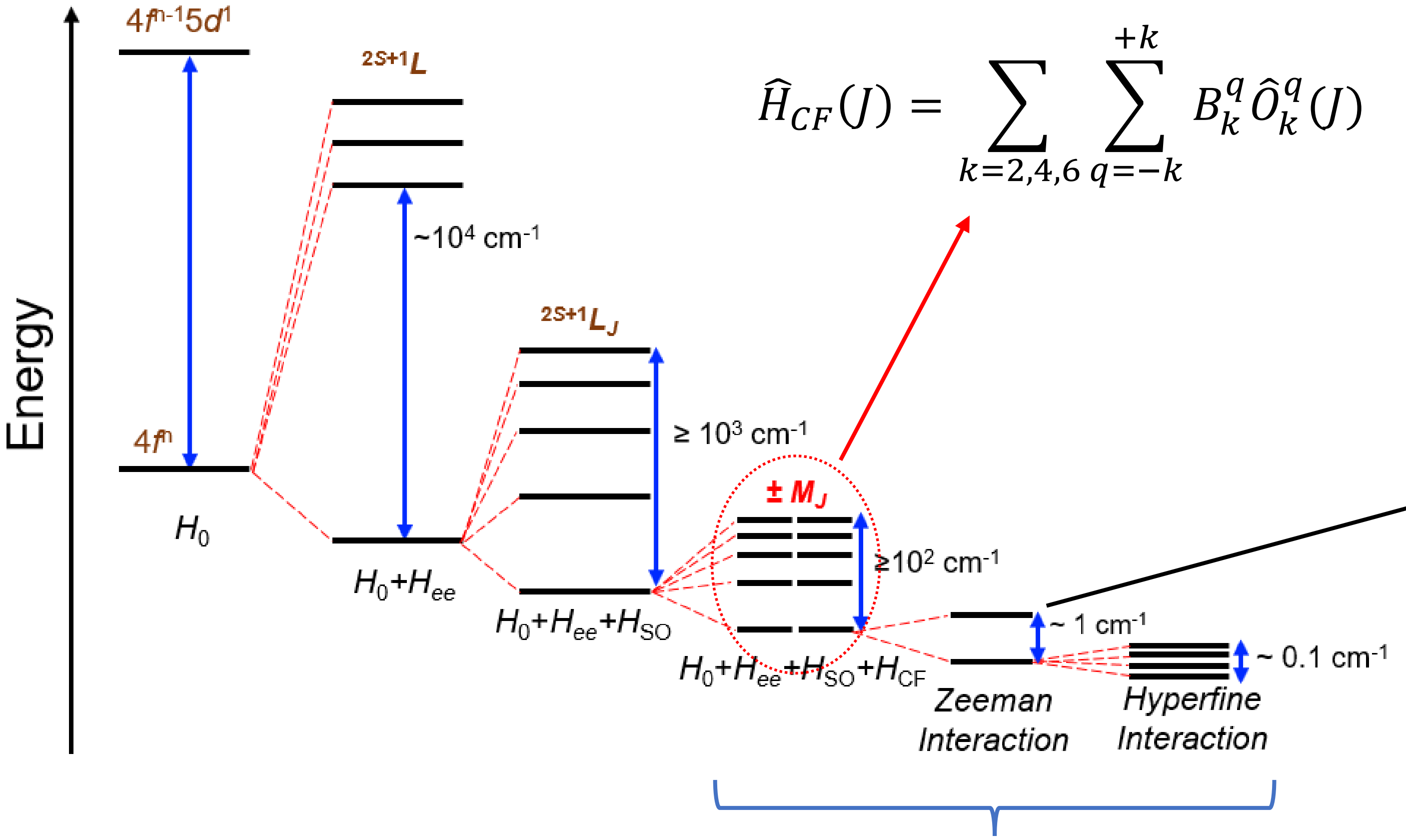
   $|\varphi\rangle_1 = \alpha_1|0\rangle + \beta_1|1\rangle$ & $|\varphi\rangle_2 = \alpha_2|0\rangle + \beta_2|1\rangle$,

   Entangled state: $|\psi^{\pm}\rangle = \alpha_1\alpha_2|00\rangle \pm \beta_1\beta_2|11\rangle$

Molecular spin-qubit

Electronic structure

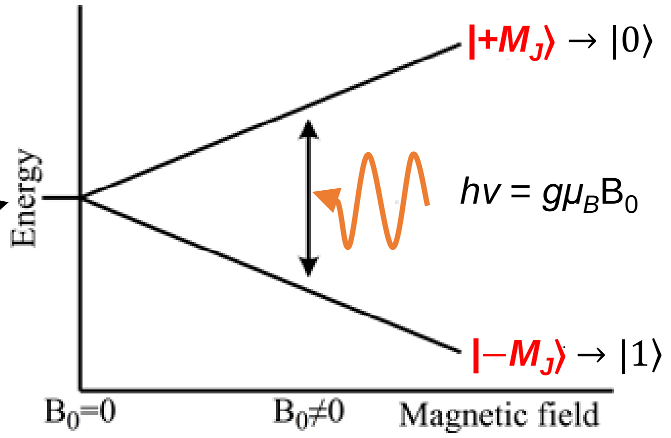
$$\hat{H}_{CF}(J) = \sum_{k=2,4,6} \sum_{q=-k}^{+k} B_k^q \hat{O}_k^q(J)$$

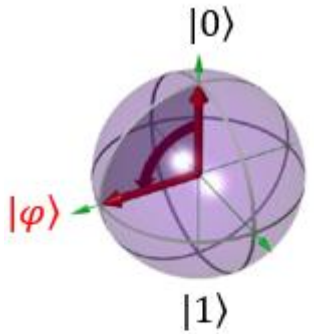


$$\hat{H}_S = \hat{H}_{CF} + \hat{H}_{Zeeman} + \hat{H}_{hyp} + \hat{H}_{nuc.\,Zeeman}$$

Molecular spin-qubit

- ✓ Ground M_J is well isolated from Excited M_J
- ✓ 0.3 cm^{-1} for X band



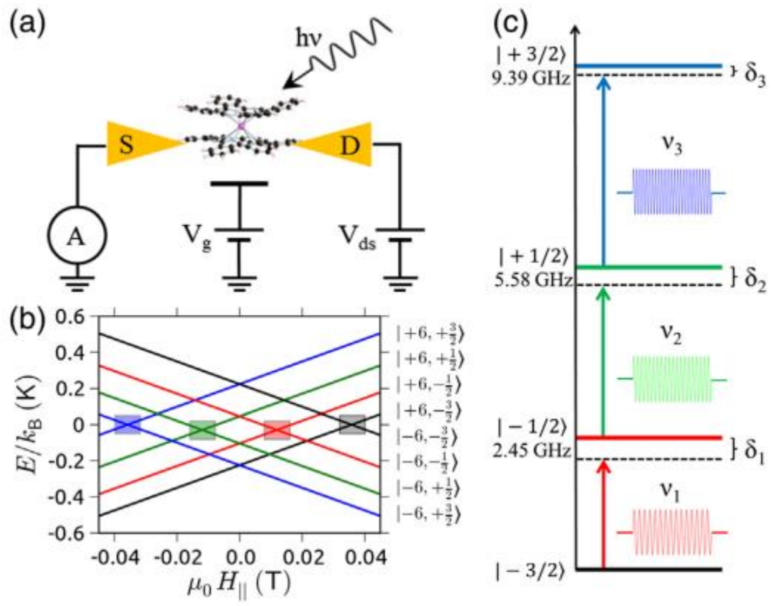


$|\varphi\rangle = \alpha|0\rangle + \beta|1\rangle$

QIP using molecular spin-qubits

✓ Grover's Quantum Algorithm

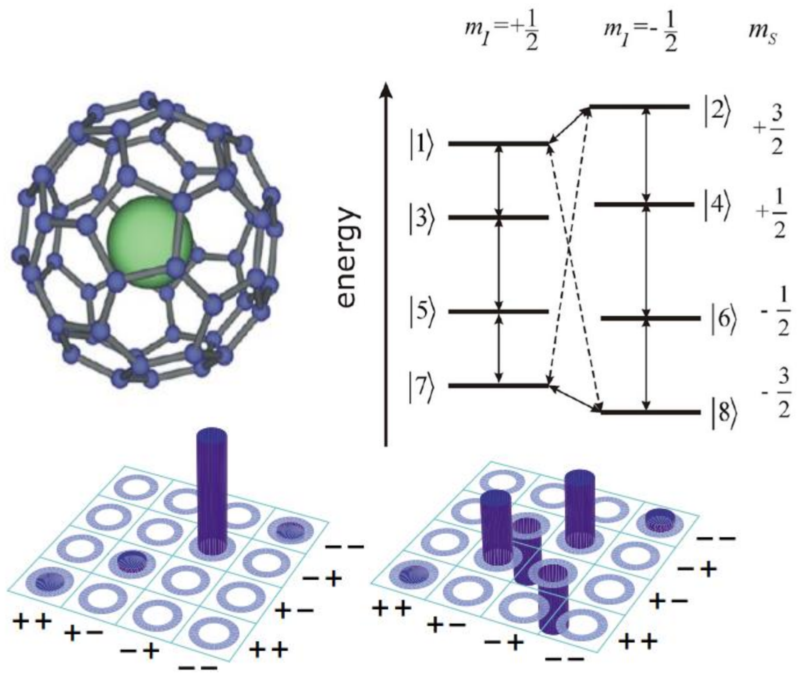
$TbPc_2$



Godfrin, C., et al., *Phy. Rev. Lett.* 119.18 (2017): 187702.

✓ Quantum entanglement

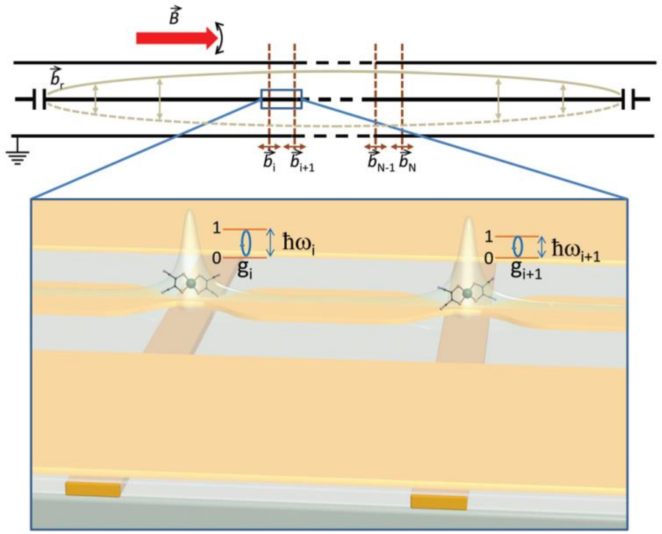
$^{15}N@C_{60}$



Mehring, M., et al., *Phy. Rev. Lett.* 93, 20 (2004): 206603.

➢ A quantum processor

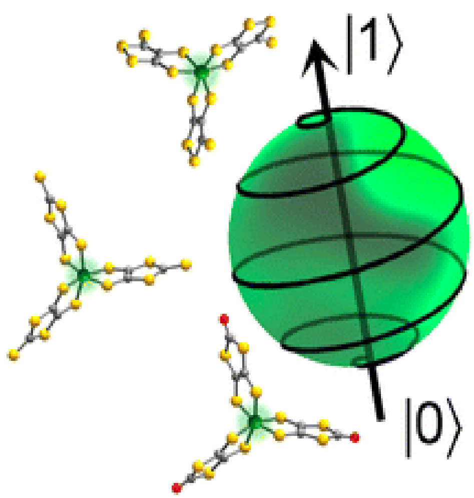
Superconducting resonator



Gaita-Ariño, A., et al., *Nat. Chem.* 11.4 (2019): 301-309.

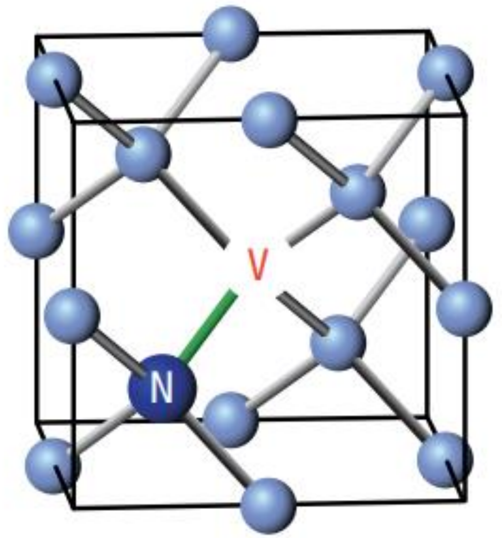
Quantum coherences in spin-qubits

- ✓ $T_m \sim$ 1 ms @10K
- ✓ $(d_{20}\text{-}Ph_4P)_2[V(C_8S_8)_3]$

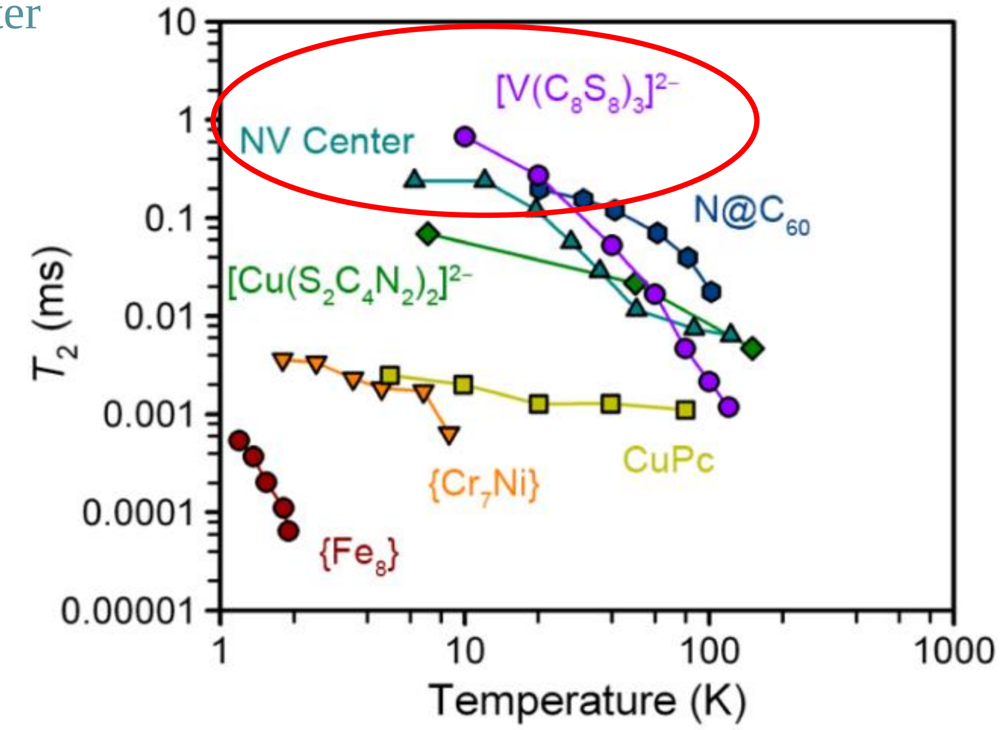


Zadrozny, Joseph M., et al., *ACS Cent. Sci.* 1.9 (2015): 488-492.

- ✓ $T_m \sim$ 0.2 ms @10K
- ✓ Nitrogen Vacancy (NV) center



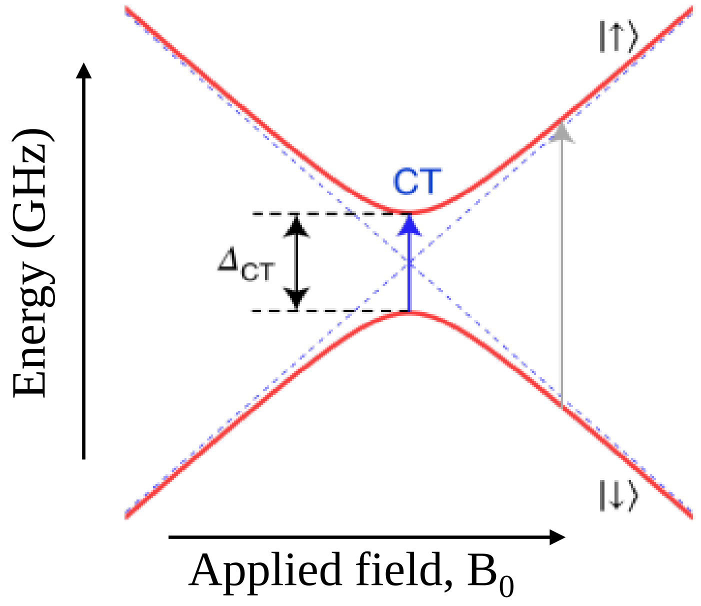
Takahashi, Susumu, et al. *Phy. Rev. Lett.* 101.4 (2008): 047601.



Molecular clock-transition (CT) spin-qubits

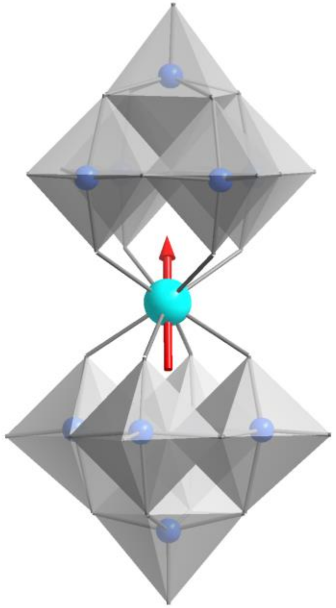
✓ At CT field, $\partial E/\partial B = 0$

✓ CT frequency = Δ_{CT}

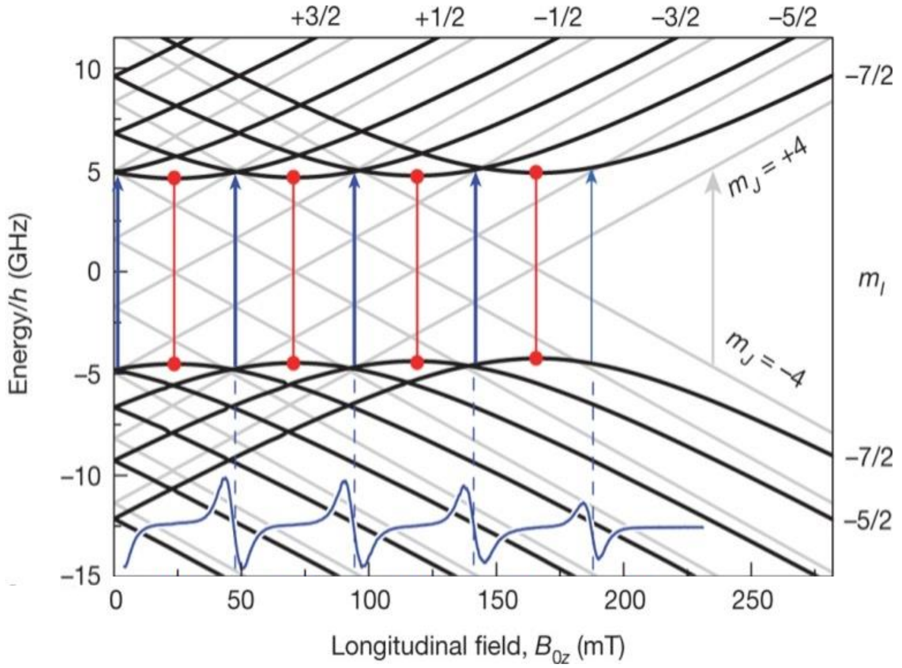


✓ $T_m \sim 8.4\ \mu s$ @ 5K

✓ Extra-diagonal CF B_4^4= 94.3 MHz



$[Ho(W_5O_{18})_2]^{9-}$



Shiddiq, Muhandis, et al., *Nature* 531.7594 (2016): 348-351.

Molecular clock-transition (CT) spin-qubits

✓ Ho^{3+} is a Lanthanide non-Kramer ion with J = 8 and I = 7/2

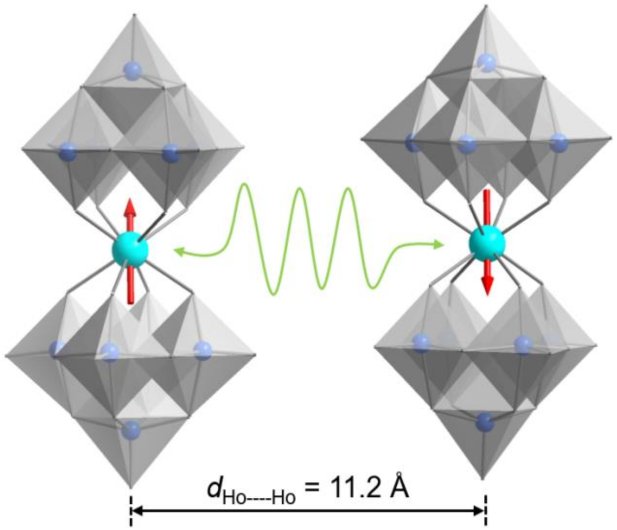
✓A unit cell contains two inversion related $[Ho(W_5O_{18})_2]^{9-}$ (in short $\mathbf{HoW_{10}}$)

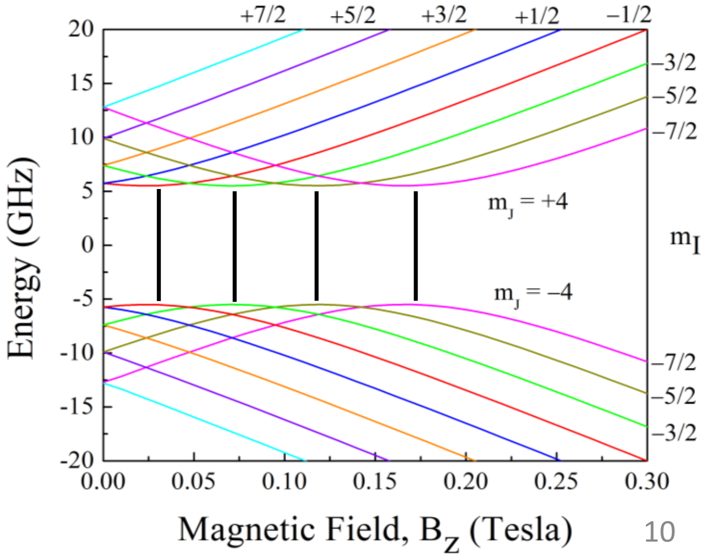
✓ Symmetry: near to D_{4d}

✓ Spin spectrum:

$$\hat{H}_S = \hat{H}_{CF} + \hat{H}_{Zeeman} + \hat{H}_{hyp} + \hat{H}_{nuc.\,Zeeman}$$

✓ Spin-qubit: $|\varphi\rangle = (\alpha|\pm 4\,\rangle \pm \beta|\mp 4\rangle)\;|-1/2\rangle$





General objectives

Addressing of individual molecular spin-qubits and entangling 2-qubits

1. **Quantum coherent control** of molecular spin-qubit using E-field: $|\varphi\rangle = \alpha|0\rangle + \beta|1\rangle$

2. **Entangling two qubits**:

$$|\varphi\rangle_1 = \alpha_1|0\rangle + \beta_1|1\rangle \quad \& \quad |\varphi\rangle_2 = \alpha_2|0\rangle + \beta_2|1\rangle,$$

Entangled state: $|\psi^{\pm}\rangle = \alpha_1\alpha_2|00\rangle \pm \beta_1\beta_2|11\rangle$

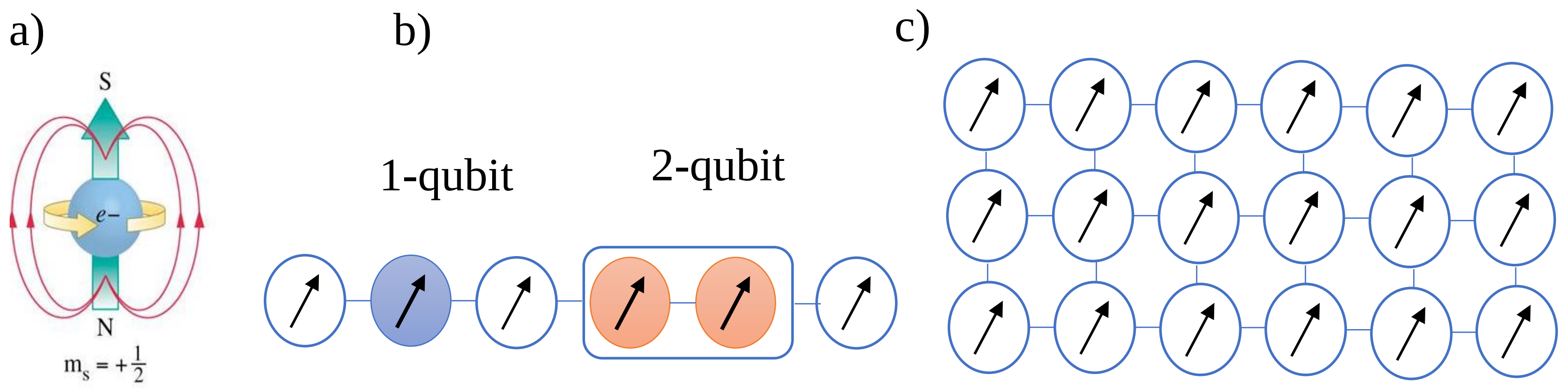
Coherent control of spin using electric field

- ✓ Maxwell´s equations:

$$\nabla \cdot E = \frac{\rho}{\varepsilon_0} \quad (1) \qquad \nabla \times E = -\frac{\partial B}{\partial t} \quad (3)$$

$$\nabla \cdot B = 0 \quad (2) \qquad \nabla \times B = \mu_0 j + \frac{1}{c^2}\frac{\partial E}{\partial t} \quad (4)$$

- ✓ We can generate a directional or controlled **E**-field but we do not have magnetic monopoles (eq. 1 and 2)
- ✓ We have more freedom in designing the geometry of quantum device using E-Field rather than B-Field
- ✓ For scalable quantum computing we need coherent control over spins (Objective)

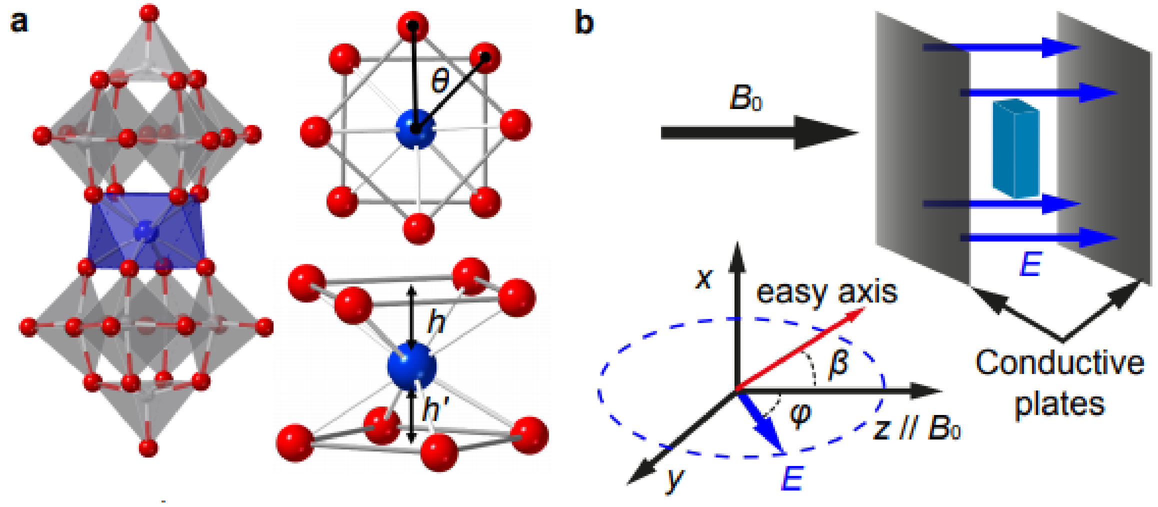


Requirement

Presence of strong spin-electric couplings (SECs).

Coherent control of spin using electric field

Experimental configuration



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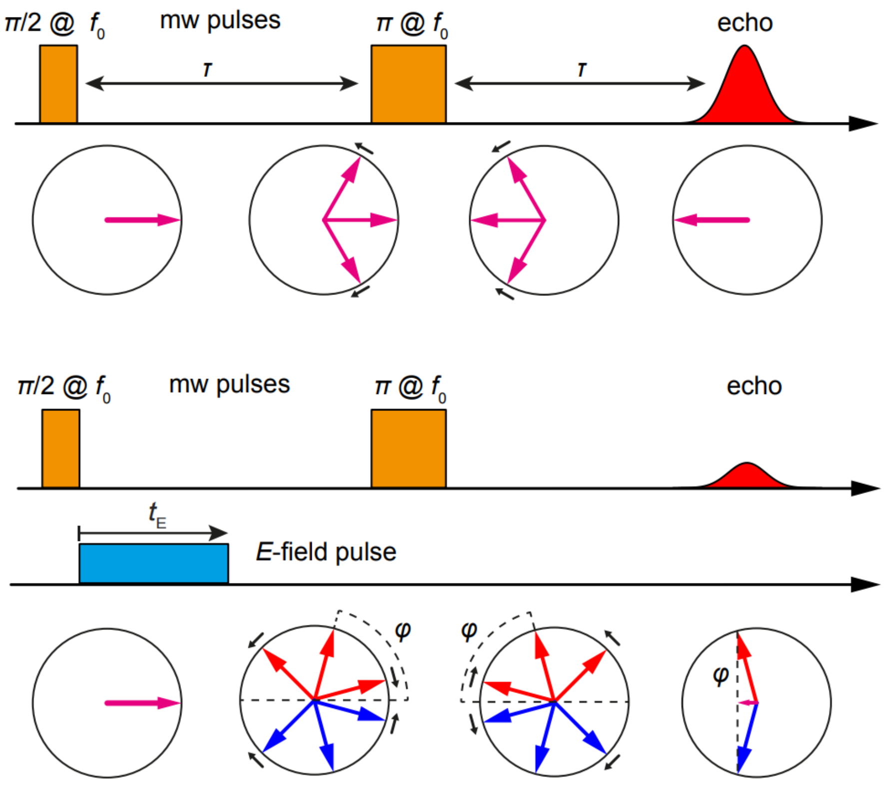
Quantum coherent spin–electric control in a molecular nanomagnet at clock transitions

Junjie Liu[1], Jakub Mrozek[1], Aman Ullah[2], Yan Duan[2], José J. Baldoví[2], Eugenio Coronado[2], Alejandro Gaita-Ariño[2] and Arzhang Ardavan[1]

Microwave Pulse sequence in EPR



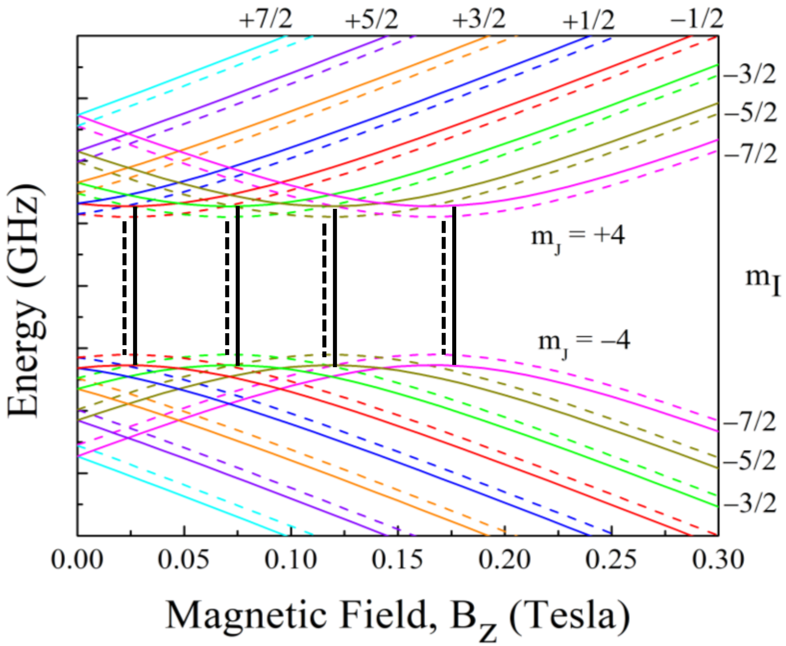
Experiment: Arzhang Ardavan, Junjie Liu at CAESR, Department of Physics, University of Oxford

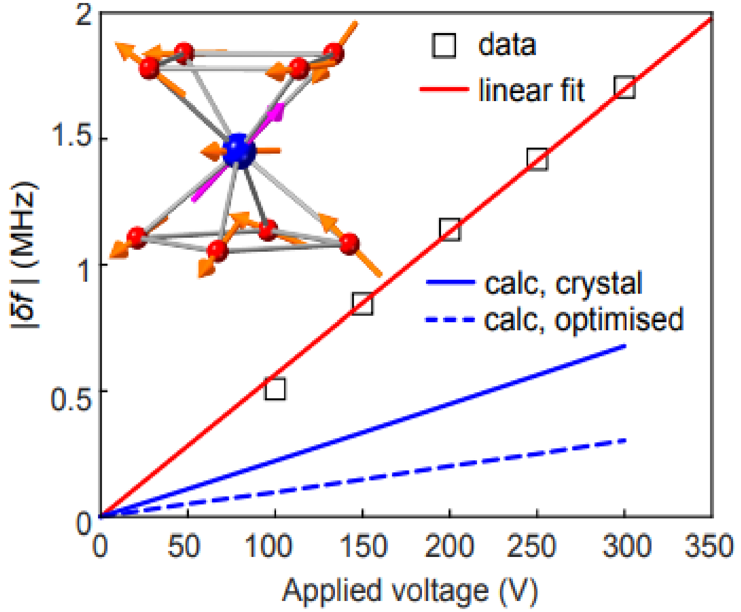
Coherent control of spin using electric field

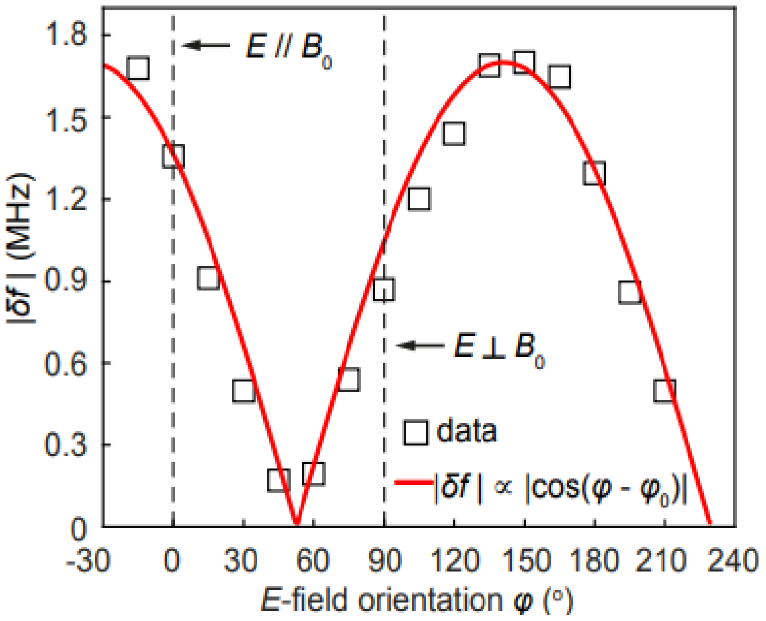
✓ **Applied Voltage(V) = E · d**, where \`d´ (=2mm) is the distance between two-plates

✓ A linear SECs is observed, SECs of order **11.4 ± 0.3 Hz/Vm[-1]**

✓ Linear response is further confirmed through rotating E-field in y-z plane

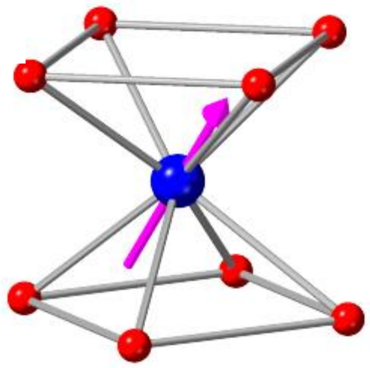






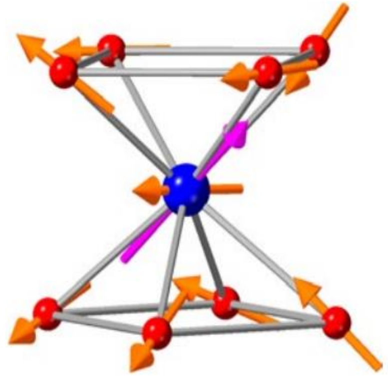
Liu, J., Mrozek, J., **Ullah, A.**, et al., *Nat. Physics*, *17*.11 (2021): 1205-1209.

Coherent control of spin using electric field



- ✓ **E-Field** will modify the spin-Hamiltonian, $\hat{H}_{CF}(J, \vec{\boldsymbol{R}}) \rightarrow \hat{H}_{CF}\left(J, \vec{\boldsymbol{R}}(\boldsymbol{E})\right)$

$$\hat{H}_{eff}\left(J, \vec{\boldsymbol{R}}(\boldsymbol{E})\right) = \sum_{k} \sum_{k=-q}^{+q} B_k^q(E)\widehat{O_k^q}(J)$$



- ✓ **E-Field** will modify the electronic dipole-moment ($\boldsymbol{\delta p}$)
- ✓ $\vec{\boldsymbol{R}}(\boldsymbol{E})$ can be modeled by decomposing the $\boldsymbol{\delta p}$ into vibrational basis (orthonormal basis)
- ✓ From linear combination we can obtain $\vec{\boldsymbol{R}}(\boldsymbol{E}) = \sum_{i=1}^{3N-6} r_i$ and compute $\hat{H}_{CF}\left(J, \vec{\boldsymbol{R}}(\boldsymbol{E})\right)$
- ✓ $\boldsymbol{\delta p}$ is determined for each normal mode at DFT level and $\hat{H}_{CF}\left(J, \vec{\boldsymbol{R}}(\boldsymbol{E})\right)$ is determined at *ab initio* level (CASSCF-SO)

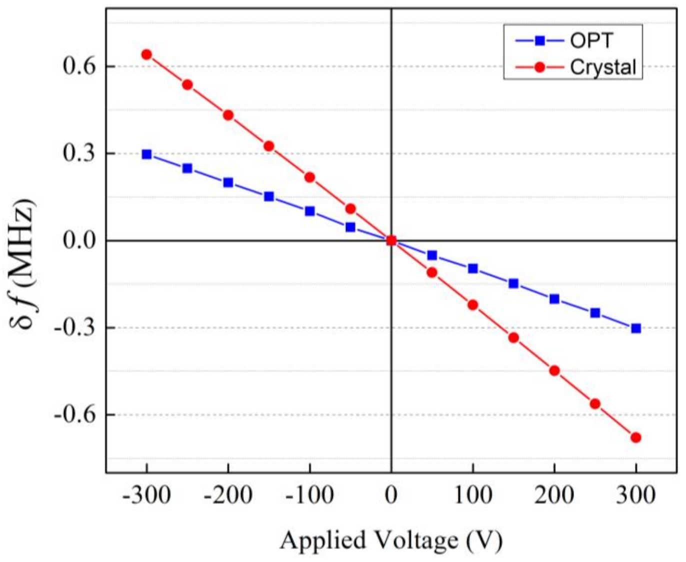
Coherent control of spin using electric field

✓ Spin-Hamiltonian,

$$\widehat{H}_{eff}\left(J,\vec{R}(E)\right)=\sum_{k}\sum_{k=-q}^{+q}B_k^q(E)\widehat{O_k^q}(J)$$

✓ Theory: SECs of order 4.7 Hz/Vm^{-1}.

✓ Experimental value: 11.4 ± 0.3 Hz/Vm^{-1}.

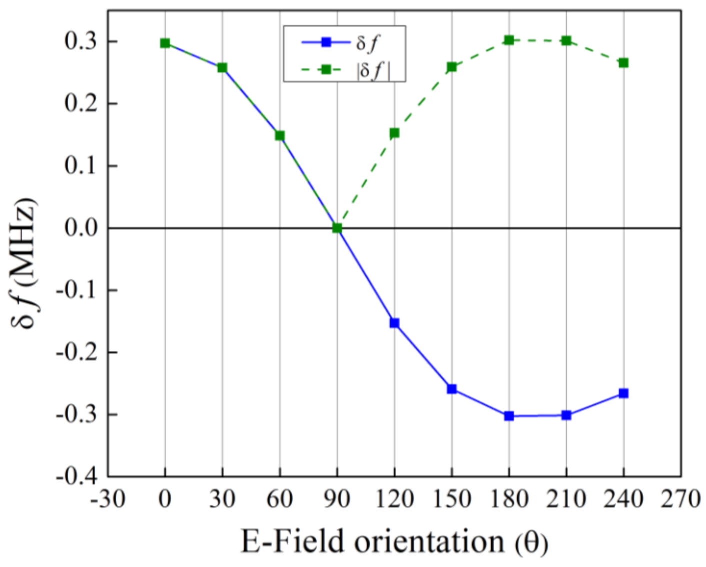


Angular Dependency

$$U_i=U_E=-\delta pE\cos(\theta)$$

$$\vec{R}(E,\theta)=\sum_{i=1}^{3N-6}r_i(E(\theta))$$

$$\widehat{H}_{CF}\left(J,\vec{R}(E,\theta)\right)=\sum_{k}\sum_{k=-q}^{+q}B_k^q(E,\theta)\widehat{O_k^q}(J)$$



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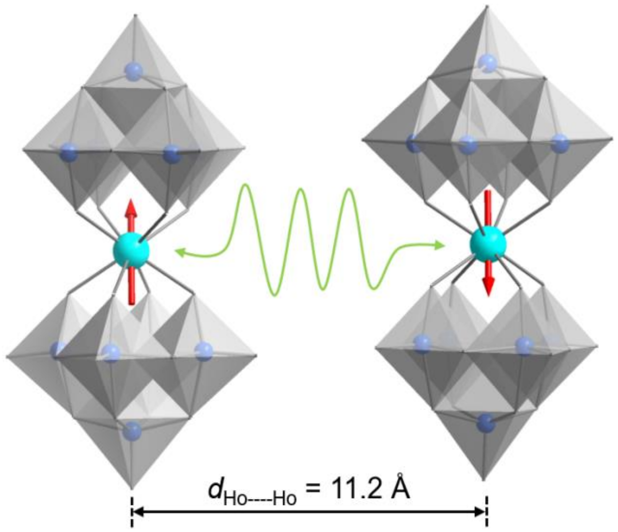
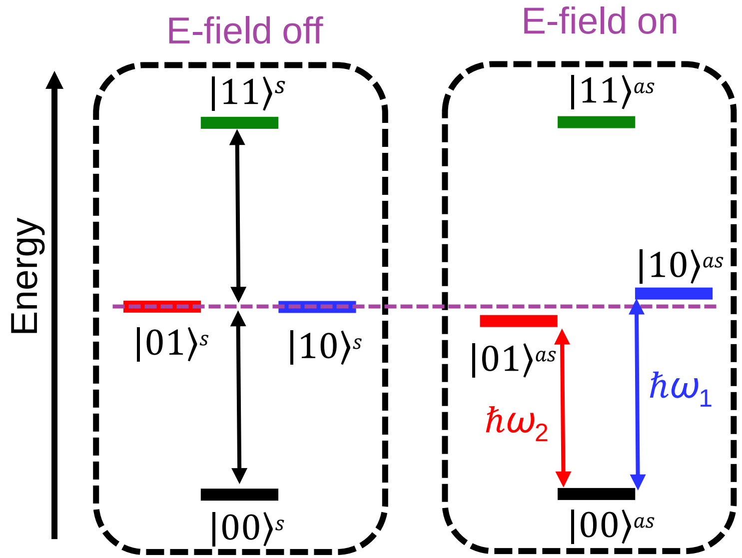
Entangling two-qubit gates within a pair of clock-qubit magnetic molecules





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Electrical two-qubit gates within a pair of clock-qubit magnetic molecules

Aman Ullah[1,2], Ziqi Hu[1,2], Jesús Cerdá[1], Juan Aragó[1] and Alejandro Gaita-Ariño[1]

QIP using molecular spin-qubit: Requirements

1. Well defined **Hilbert space**, physical qubit to logical qubit mapping: $\{00, 01, 10, 11\} \rightarrow \{E_1, E_2, E_3, E_4\}$

2. **Enabled interaction** , **differentiable transitions** & **coherent control**

3. **Coherence times** (T_1 & T_2) in the presence of control fields, i.e., **E**-field and **B**-Field.

4. **Physical implementation**, **a)** initialization, **b)** preparation & **c)** readout

Single-qubit, $|\varphi\rangle_1 = \alpha_1|0\rangle \pm \beta_1|1\rangle \rightarrow \{E_1, E_2\}$ & $|\varphi\rangle_2 = \alpha_2|0\rangle \pm \beta_2|1\rangle \rightarrow \{E_3, E_4\}$

Entangled states, $|\psi^{\pm}\rangle = \alpha_1\alpha_2|00\rangle \pm \beta_1\beta_2|11\rangle$ & $|\varphi^{\pm}\rangle = \alpha_1\beta_2|01\rangle \pm \beta_1\alpha_2|10\rangle$

1- Two-qubit Hilbert space

- ✓ Spin spectrum for the dipolar coupled HoW_{10} dimers:

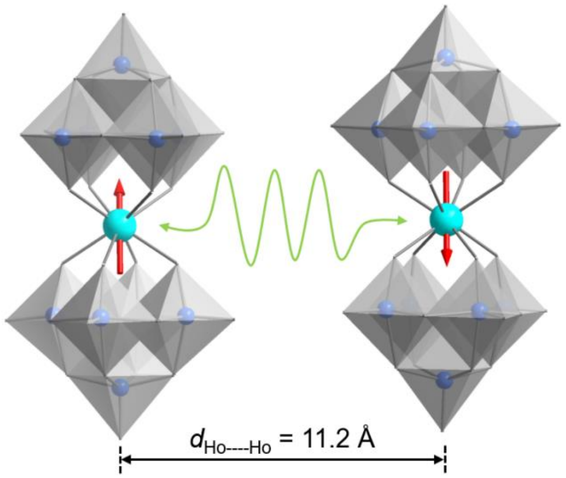
$$\hat{H}_S^{tot} = \hat{H}_S^a + \hat{H}_S^b + \hat{H}_{a,b}^{ex} = \hat{H}_S^a \otimes \mathbb{I}_b + \mathbb{I}_a \otimes \hat{H}_S^b + j_{a,b}^{\mathrm{dip}} J_a \otimes J_b$$

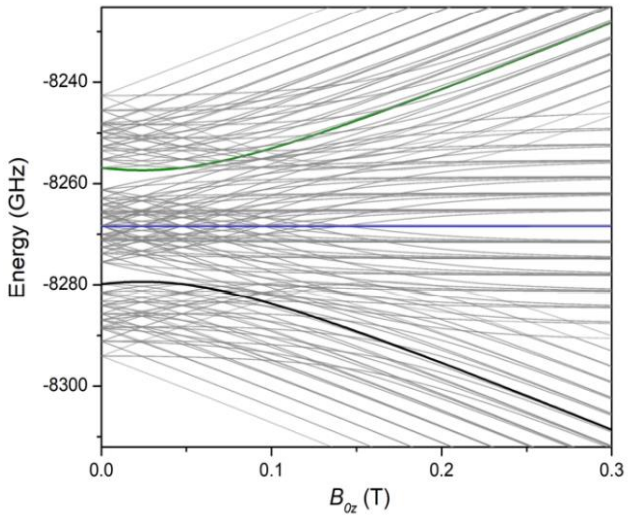
- ✓ Electro-nuclear spin levels: $M_J = \pm 4$ and $I = 7/2$:

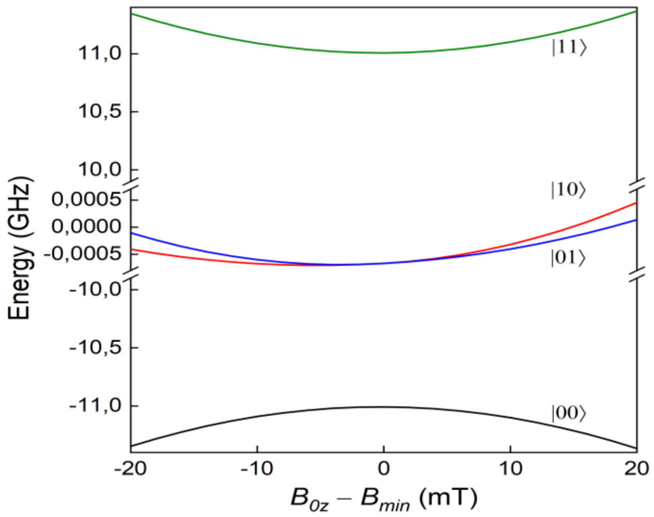
$$|\pm 4, \pm 7/2\rangle^a \otimes |\pm 4, \pm 7/2\rangle^b = 256.$$

- ✓ Near first CT-field ($\mathrm{B_{min}}$):

$$|\pm 4, -1/2\rangle^a \otimes |\pm 4, -1/2\rangle^b \;\; = |00\rangle, |01\rangle, |10\rangle, |11\rangle.$$

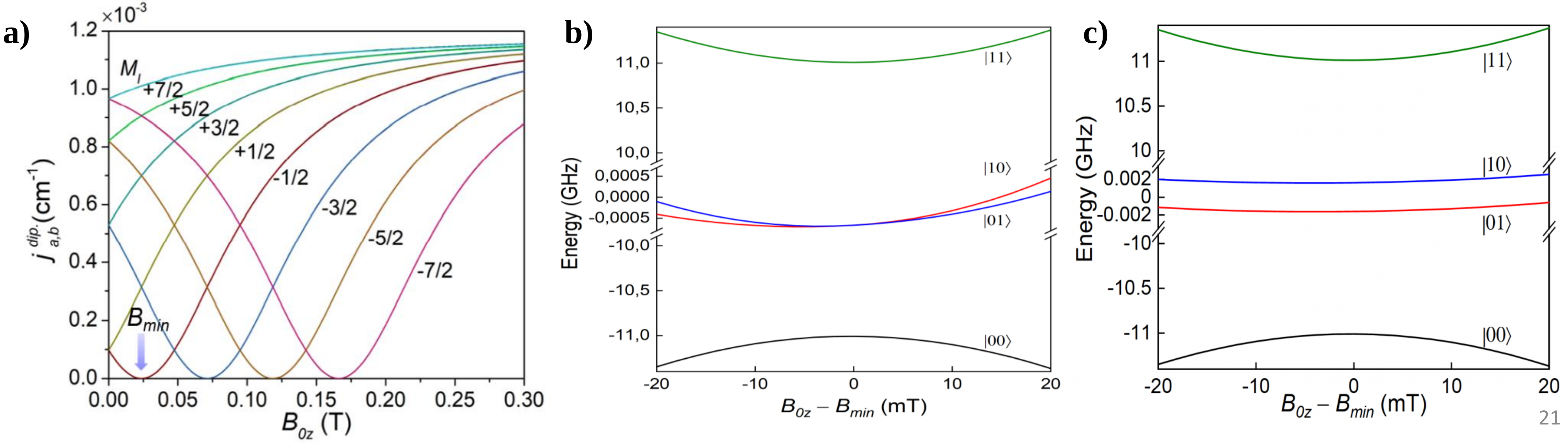






2-Enabled interaction, differentiable transitions & coherent control

- ✓ **Enabled interaction**: moving away from B_{min} will enable interaction between qubit states
- ✓ **Differentiable transitions:** @12 mT away from B_{min}, $\delta f = |10\rangle - |01\rangle$
- ✓ **Coherent control** : an E-field of 300 V/2mm is enough to coherently control the qubit states



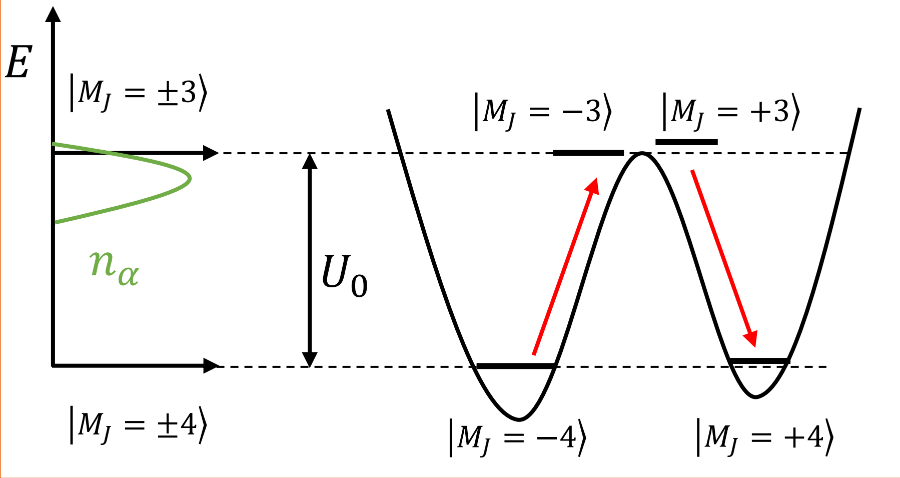
3- Coherence times (T_1 & T_2), Redfield theory

- ✓ Dynamics of entire system by time evolution of density matrix:

$$\dot{\hat{\rho}} = -\frac{i}{\hbar}\left[\hat{H}_{S-ph}, \hat{\rho}(t)\right], \quad where, \quad \hat{H}_{S-ph} = \sum_{\alpha}\left(\frac{\partial \hat{H}_S}{\partial \hat{q}}\right)\hat{q} = V_{ab}^{\alpha}\hat{q}$$

1. $\hat{\rho}(t) \approx \hat{\rho}^s(t) \otimes \hat{\rho}^{ph}$ (weak-couplings, Born-approximation)
2. $\hat{\rho}^s(s) \rightarrow \hat{\rho}^s(t),\ t' = t - s$ (Markov-approximation)

Phonon induced spin relaxation



- ✓ Time-evolution of reduced density matrix:

$$\frac{d\hat{\rho}_{ab}^s(t)}{dt} = i\omega_{ab}\hat{\rho}_{ab}^s - \sum_{c,d} R_{ab,cd}\hat{\rho}_{cd}^s(t), \quad where, \quad R_{ab,cd} = \delta_{bd}\sum_j \Gamma_{aj,jc} - \Gamma_{db,ac} - \Gamma_{ca,bd}^* + \delta_{ca}\sum_j \Gamma_{bj,jd}^*$$

- ✓ Where, $R_{ab,cd}$ is tetradic Redfield relaxation operation with $\Gamma_{db,ac}$ are the rate constants:

$$\Gamma_{db,ac} = \sum_{\alpha} V_{db}^{\alpha} V_{ac}^{\alpha} G(\omega_{db}, \omega_\alpha), \qquad G(\omega_{ij}, \omega_\alpha) = \delta(\omega_{ij} - \omega_\alpha)\bar{n}_\alpha + \delta(\omega_{ij} + \omega_\alpha)(\bar{n}_\alpha + 1)$$

Here, V_{db}^{α} are spin-phonon couplings and $G(\omega_{db}, \omega_\alpha)$ is a spectral function.

3- Coherence times (T_1 & T_2)

- ✓ Eigenstate decay profile in terms of magnetization expectation value:

$$\langle \vec{M}(t) \rangle = \sum_a \langle a | \hat{\rho}^s(t) \vec{M} | a \rangle$$

- ✓ Longitudinal relaxation time, T_1, (**Fig. a**):

$$\hat{\rho}^s(t=0) = |0\rangle\langle 0|$$

$$M_z(t) = \left(M_z(0) - M_z(\infty)\right)e^{-t/\tau} + M_z(\infty)$$

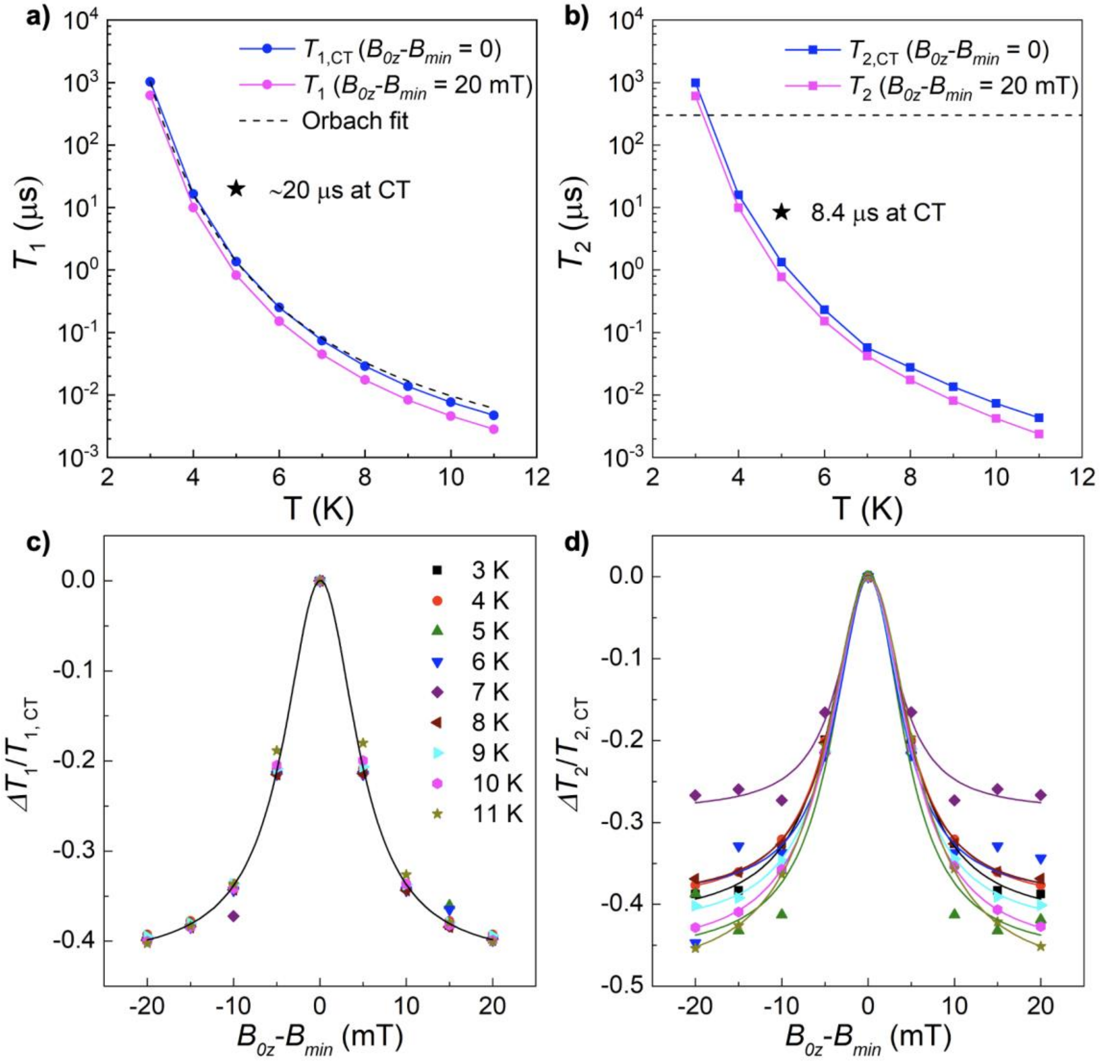
- ✓ Transverse relaxation time, T_2, (**Fig. b**):

$$\hat{\rho}^s(t=0) = |0\rangle\langle 1|$$

$$M_{x,y}(t) = \left(M_{x,y}(0) - M_{x,y}(\infty)\right)e^{-t/\tau} + M_{x,y}(\infty)$$

- ✓ T_1 & T_2 divergence around CT B-field, (**Fig. c, d**):

$$\frac{\Delta T_{1\&2}}{T_{1\&2,CT}} = \frac{T_{1\&2,Bz\neq 0} - T_{1\&2,Bz=CT}}{T_{1\&2,Bz=CT}}$$



4- Physical Implementation

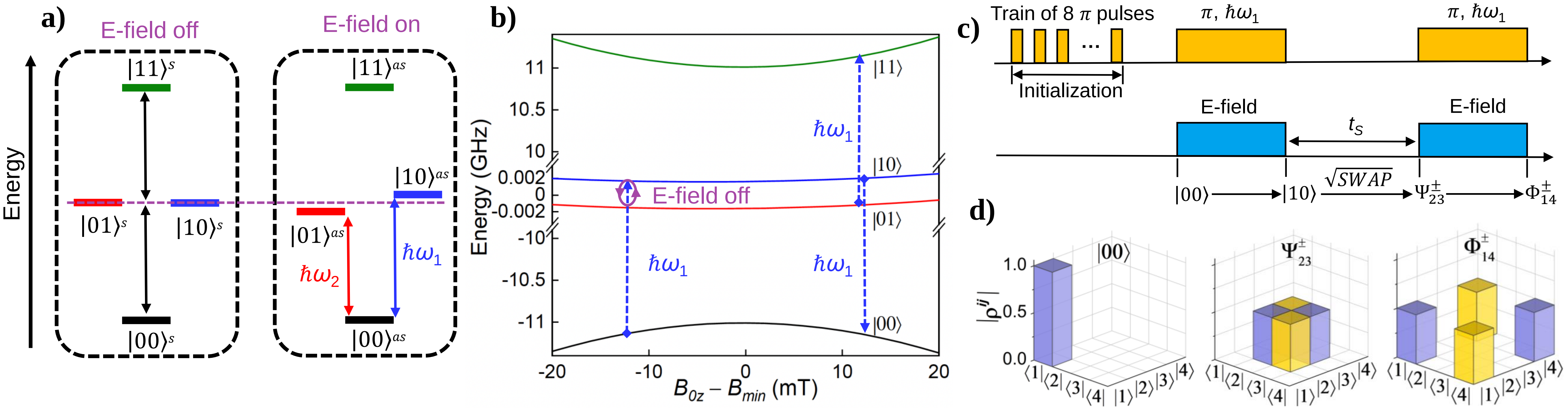
✓ E-field will modify the qubit states from symmetric to asymmetric state. (Op. conditions: B=12 mT & E=300 V/2mm)

1. We initialize the system at $|00\rangle$,
2. E-field is turned on,
3. A π-pulse is applied,
4. E-field is turned off.

$$|\psi^{\pm}\rangle = \alpha_1\alpha_2|01\rangle \pm \beta_1\beta_2|10\rangle \;\; \&$$

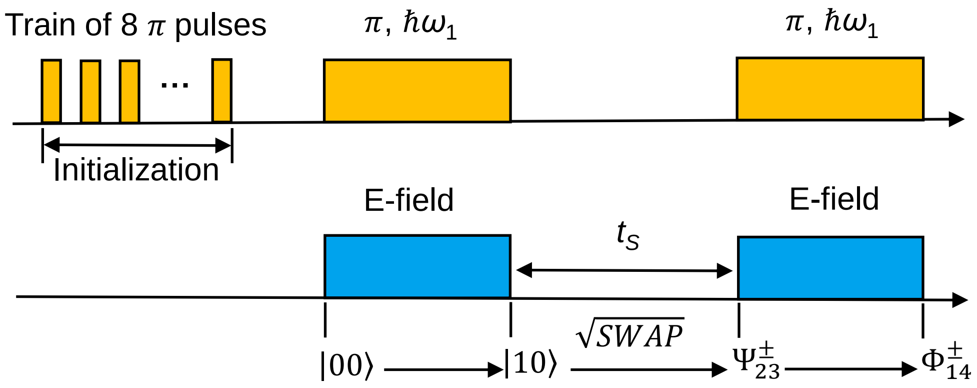
$$|\varphi^{\pm}\rangle = \alpha_1\beta_2|00\rangle \pm \beta_1\alpha_2|11\rangle$$

Operation	pulses
$\sigma_x(1)$	$\hbar\omega_1(\pi)$
$\sigma_x(2)$	$\hbar\omega_2(\pi)$
$SWAP$	E-Field (τ)



4- Physical Implementation & gating time

✓ Entangled-state generation time or gating time: $t_{gating} \propto \frac{1}{\delta f}$



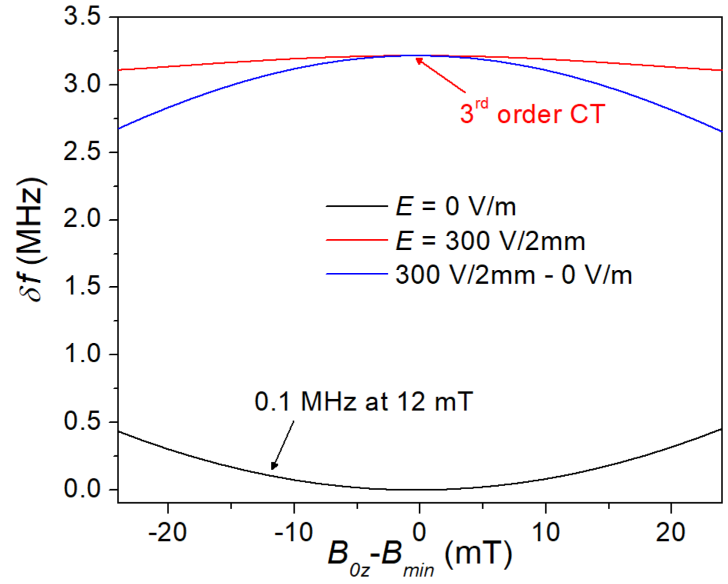
Initialization, 8 π pulses	0.3 µs
Preparation, for π-pulse (ℏω)	0.8 µs
Creation of Entanglement, for SWAP gate	5.0 µs
Creation, for π-pulse (ℏω)	0.8 µs
Total	7.0 µs

Phase memory time	5 K @ 12 mT	4 K @ 12 mT
T_2	2 µs	10 µs

✓ Presence of highly protected qubit (3rd order CT)

✓ Flip-flop qubit:

$$\varphi = |10\rangle \pm |01\rangle$$

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Electrical two-qubit gates within a pair of clock-qubit magnetic molecules

Aman Ullah[1,2], Ziqi Hu[1,2], Jesús Cerdá[1], Juan Aragó[1] and Alejandro Gaita-Ariño[1]

Conclusions & outlook

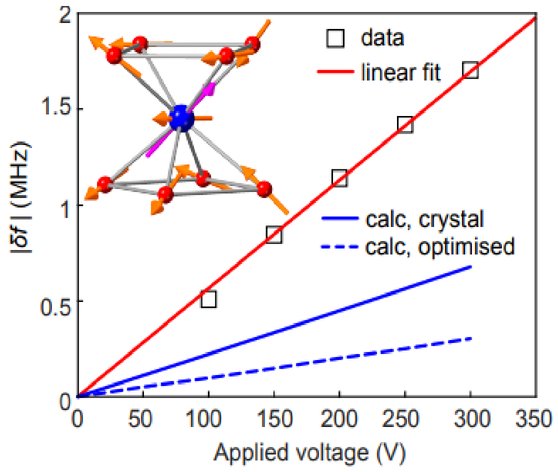
1- Enhancing spin sensitivity to the E-Field for coherent manipulation of spin information:

i) Large unquenched angular momentum ($\hat{L}$)

ii) Broken symmetry

iii) Polarizable environment (ligands)

iv) Spin-sensitivity to molecular distortion



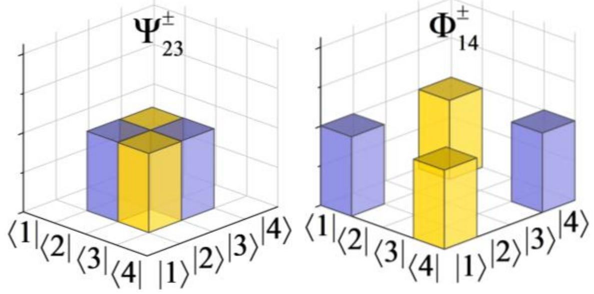
2- These findings demonstrate a relation between:

i) Spin states, lattice coordinate (optical or acoustic phonons) & electric charge

3- Entanglement generation within pair of HoW_{10}--HoW_{10}:

i) Coherent manipulation of each qubit due to the presence of strong E-field effect

ii) One-to-one correspondence between physical and logical qubits.



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  Arzhang Ardavan, Junjie Liu *et al.* for Coherent control over spin-qubit and SECs.

Thank you for your time.