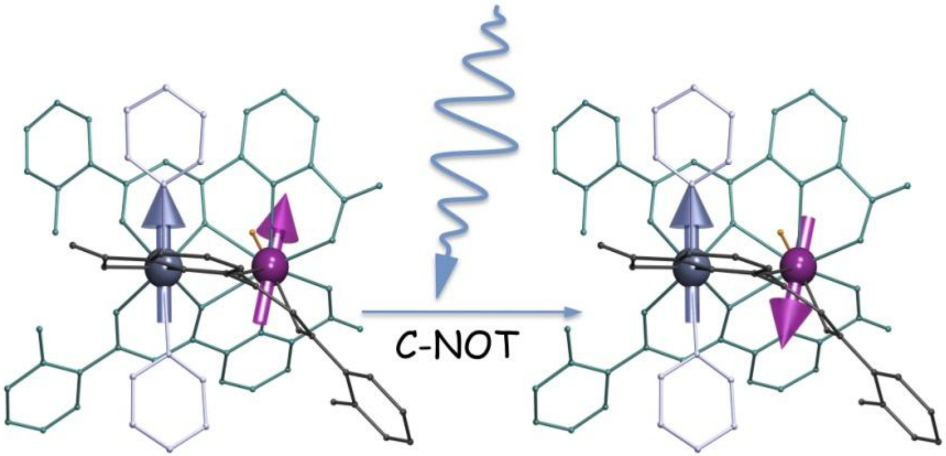
Reunión del Club Magnético
Zaragoza, 13th December 2012

Molecular prototypes for spin-based quantum logic gates



Fernando LUIS

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Universidad Zaragoza
1542
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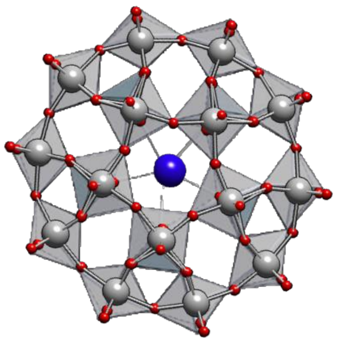
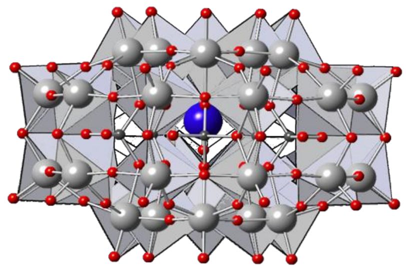
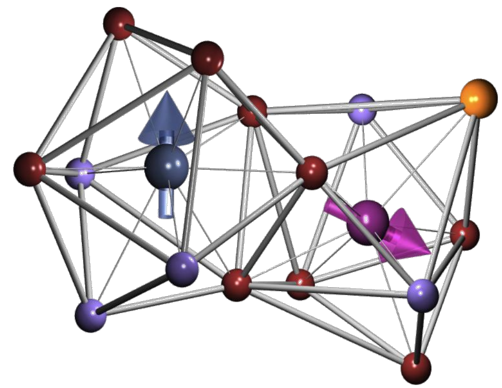
Outline

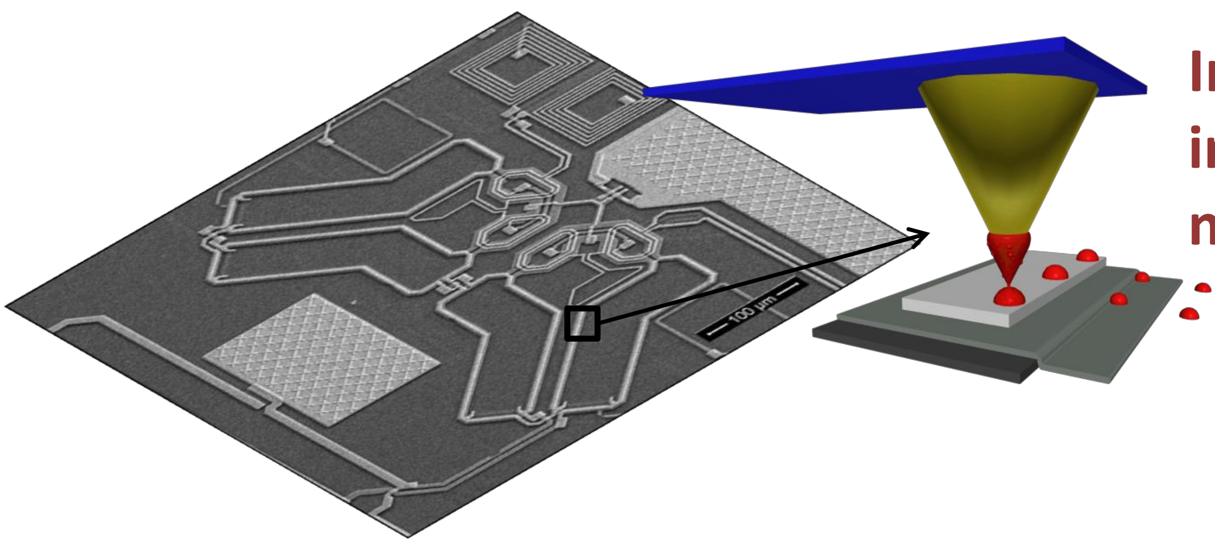
Molecular qubits





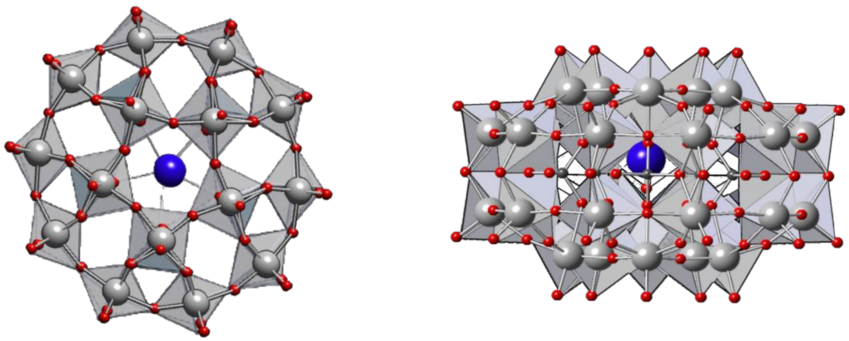
Molecular quantum gates



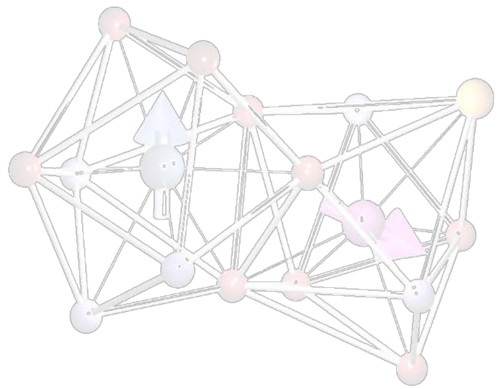


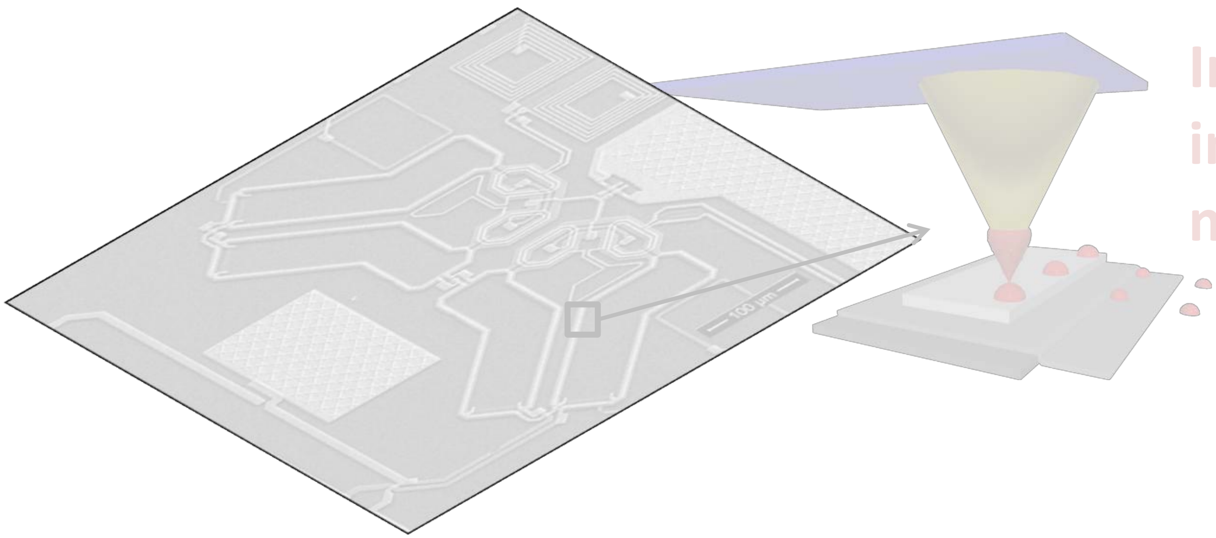
Integration of SMM into superconducting microdevices

Molecular qubits



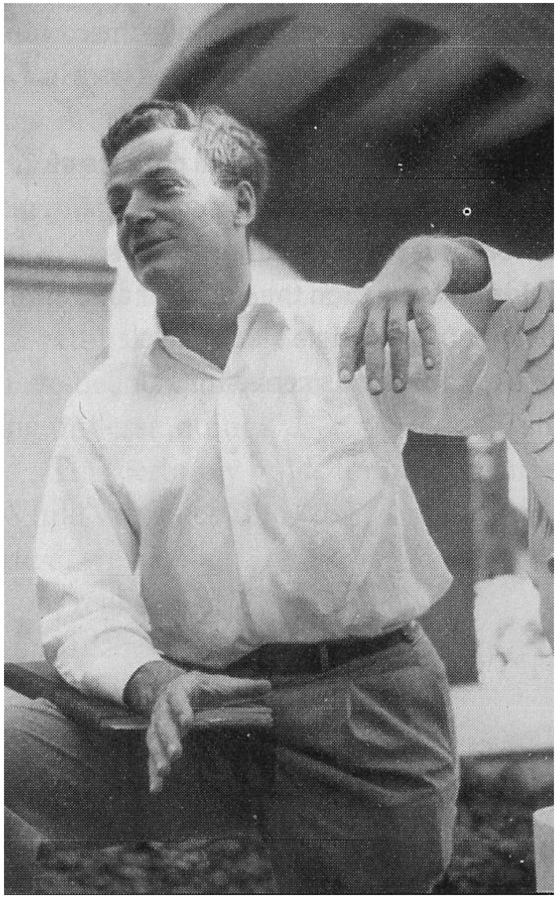
Molecular quantum gates





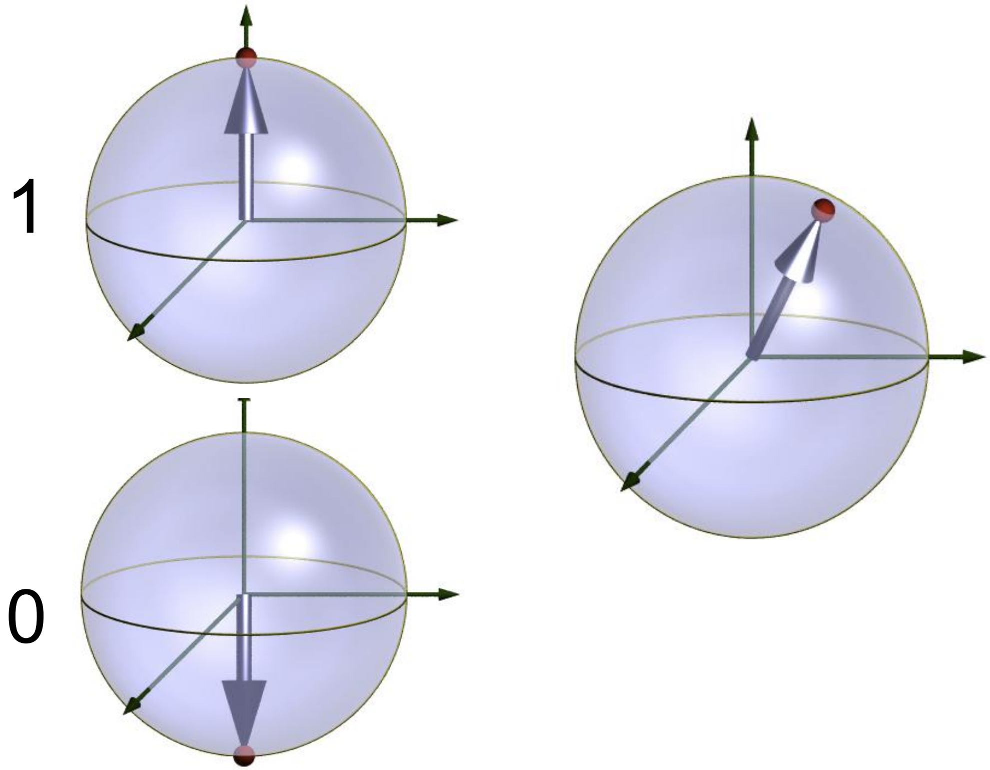
Integration of SMM into superconducting microdevices

Quantum computers

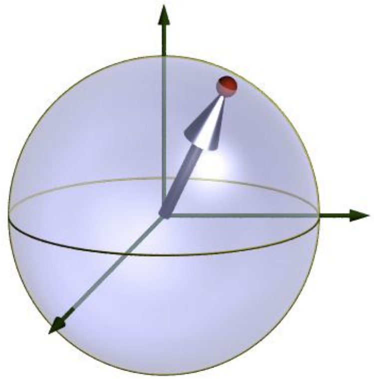


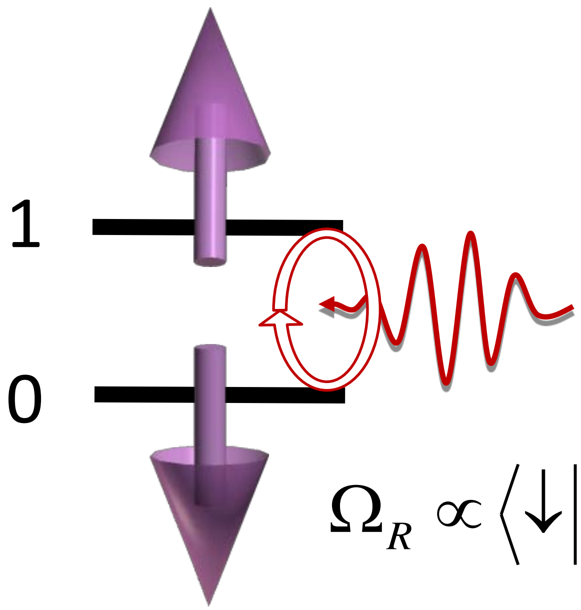
Richard Feynman, 1982

- Process information using quantum laws
- Bit $\rightarrow$ Qubit



Single qubits



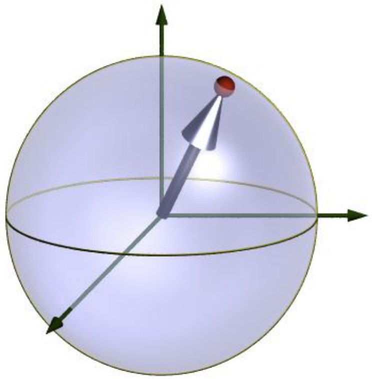


$$\Omega_R \propto \langle \downarrow | \vec{b}_{rf} \vec{S} | \uparrow \rangle$$

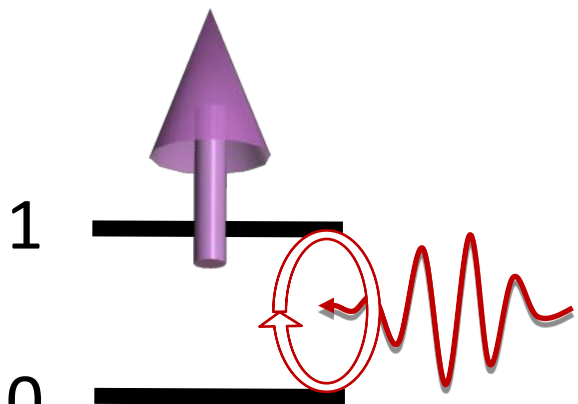
$$|\uparrow\rangle \rightarrow \alpha|\uparrow\rangle + \beta|\downarrow\rangle$$

Qubits

Single qubits



- Two well defined states
- High quantum coherence
- Integration into a scalable architecture



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Qubits

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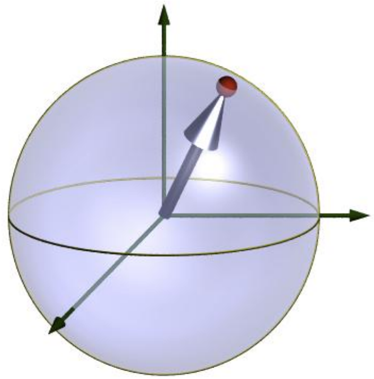
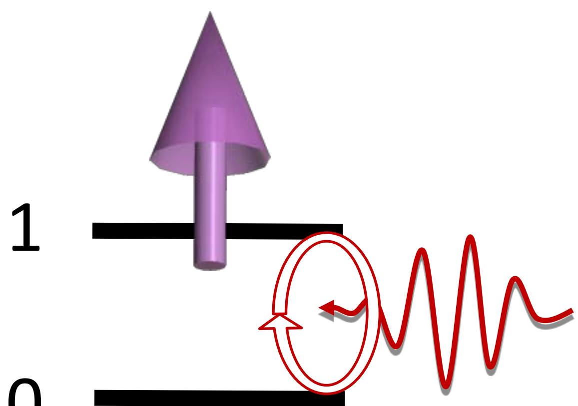
Single qubits





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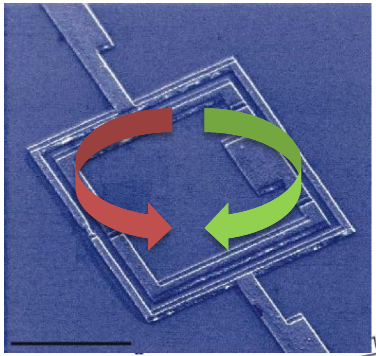
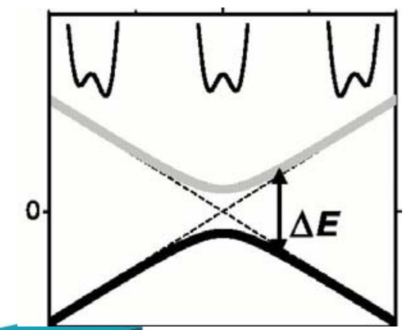
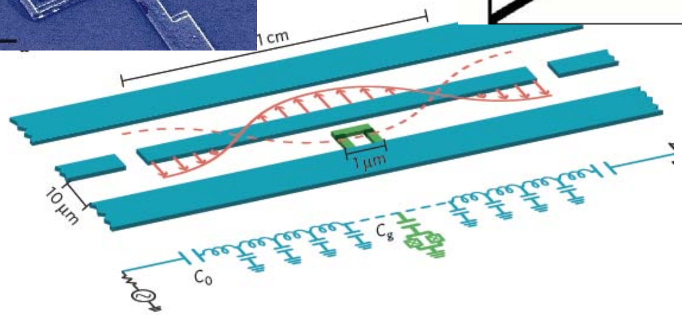
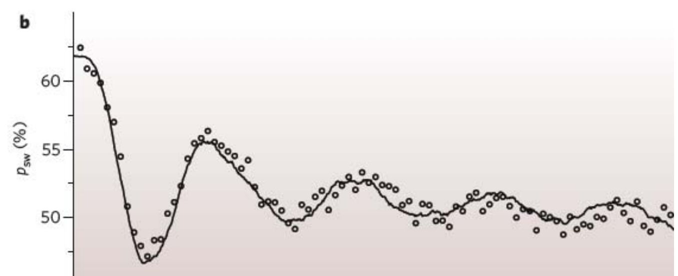




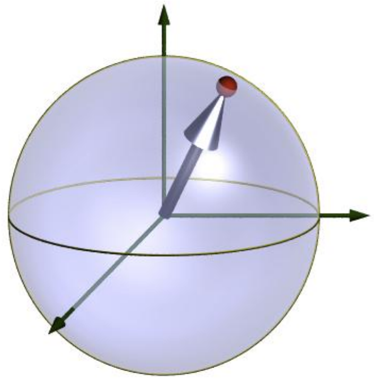


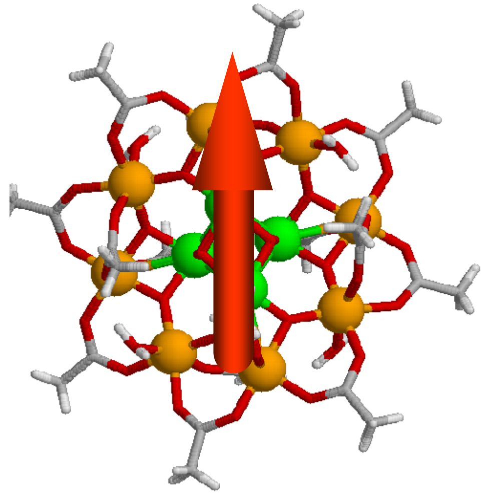


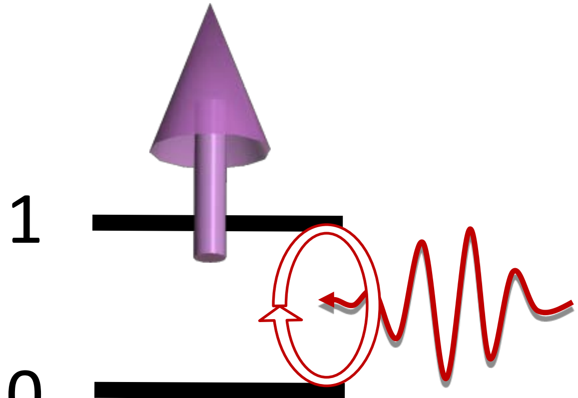
Molecular spin qubits

Leuenberger and Loss, Nature **410**, 789 (2001)

Single qubits







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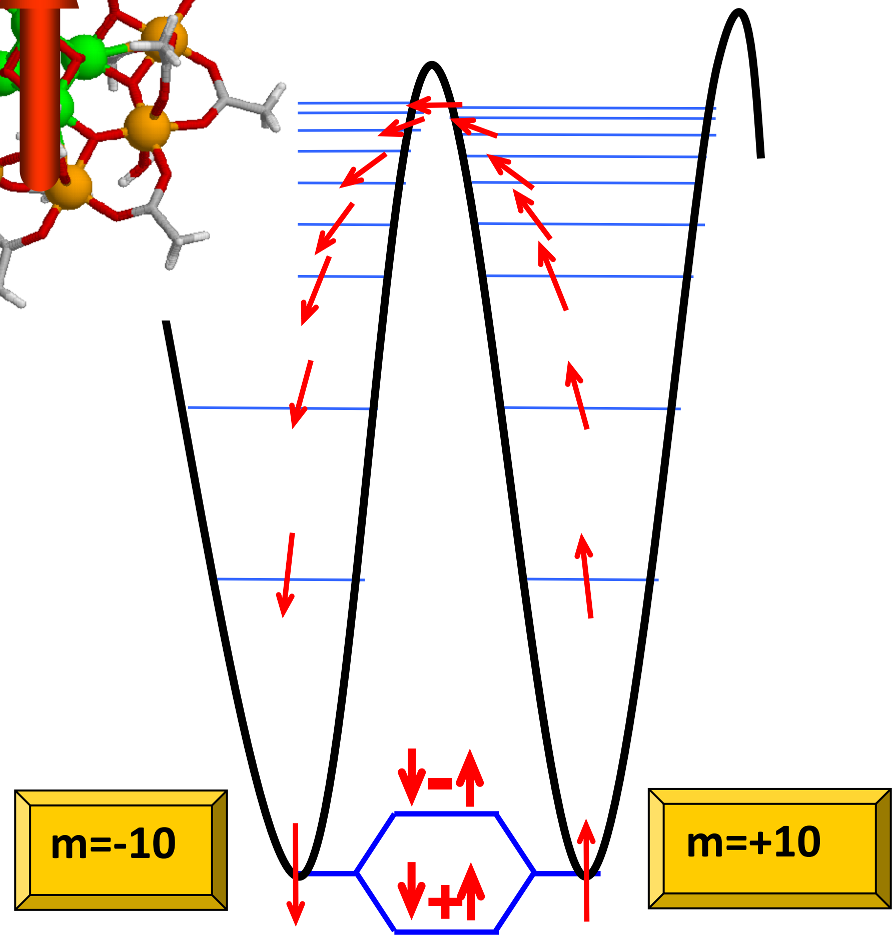

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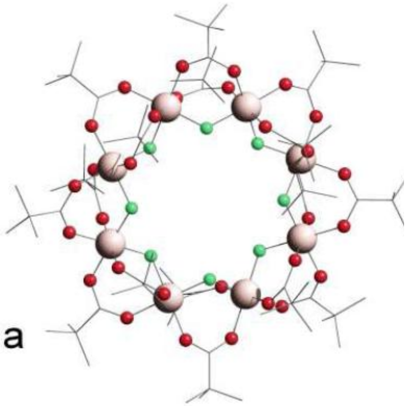
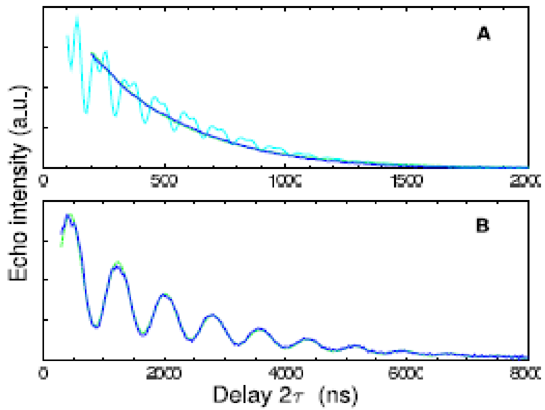
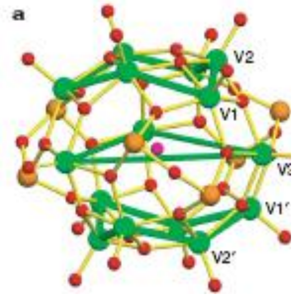
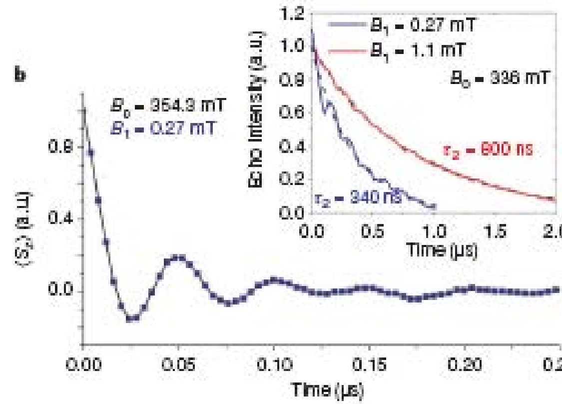


Cr_7Ni, S = 1/2



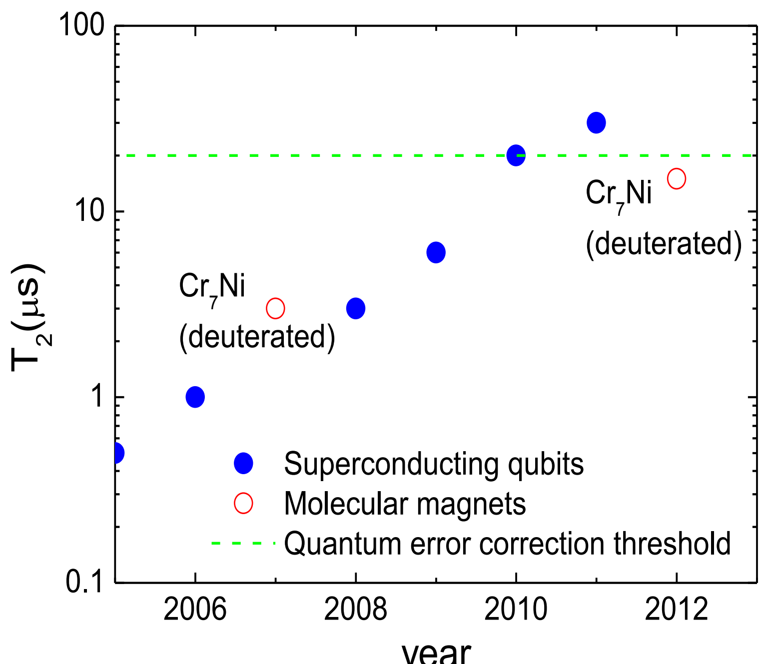


V_{15}, S = 1/2

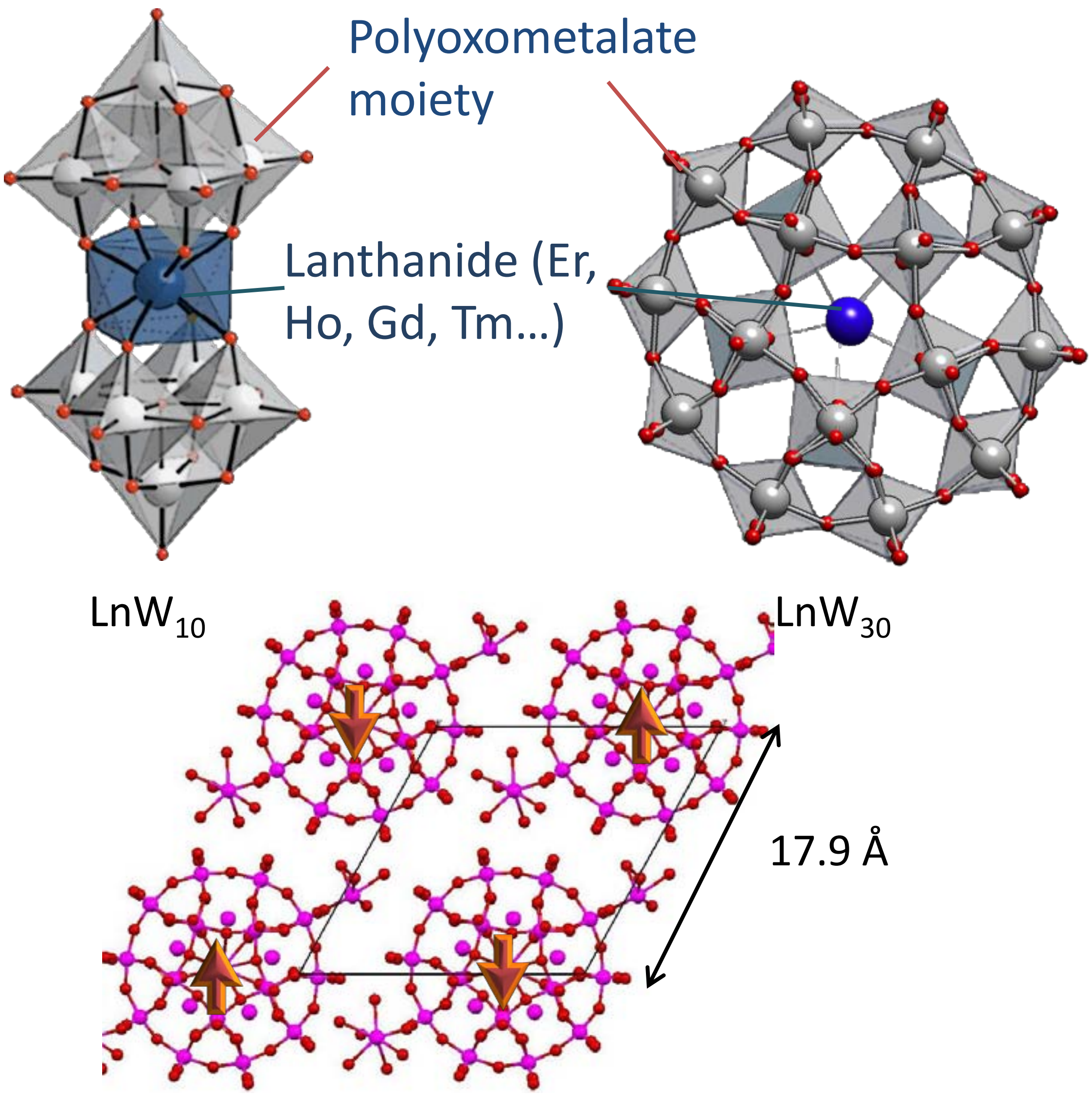


A. Ardavan et al. Phys. Rev. Lett. **98**, 057201 (2007); ibid (2012).

S. Bertaina et al. Nature **453** (2008)



Single-ion magnets

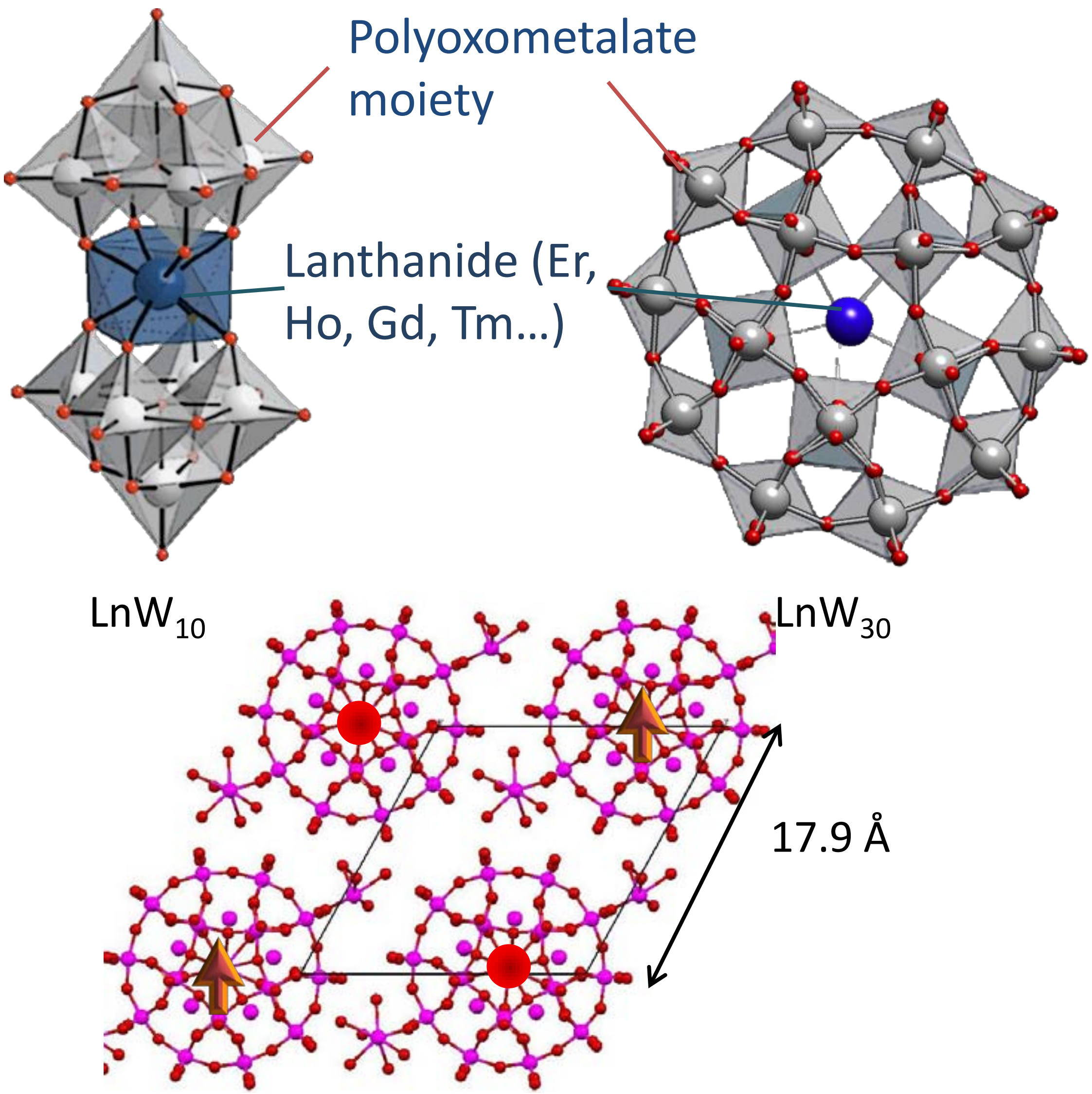


Some outstanding characteristics…

- Simple (just 1 magnetic atom)
- Weak interactions
- Magnetic solubility
- Nuclear-spin free systems
- Control over parameters

M. A. AlDamen *et al*, J. Am. Chem. Soc. **130**, 8874 (2008); M. A. Aldamen *et al*, Inorg. Chem. **48**, 3467 (2009)

Single-ion magnets



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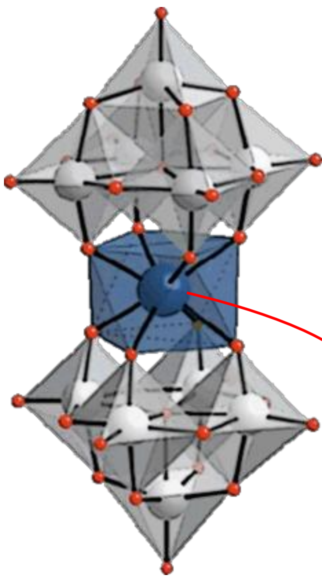
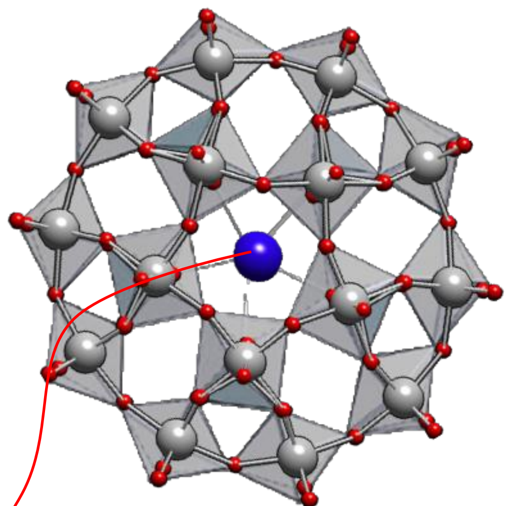
Tailoring the energy-level structure: the case of Gd

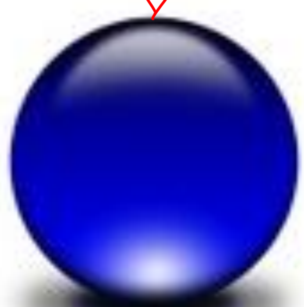


?



GdW_{10}

GdW_{30}



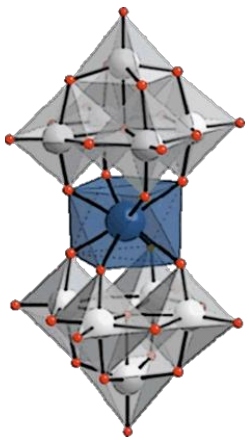
Gd: [Xe]$4f^7$

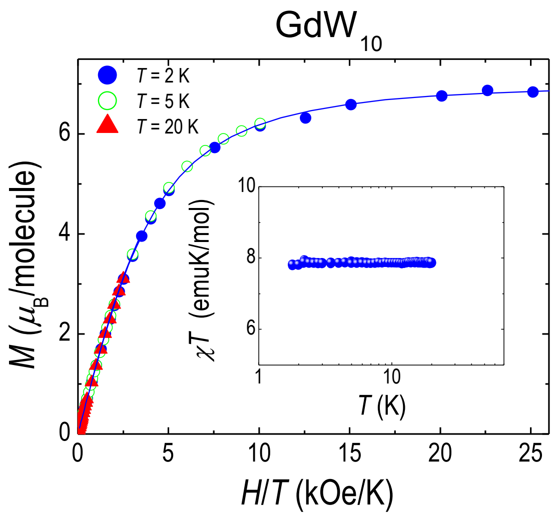
$L = 0$, $S = 7/2$

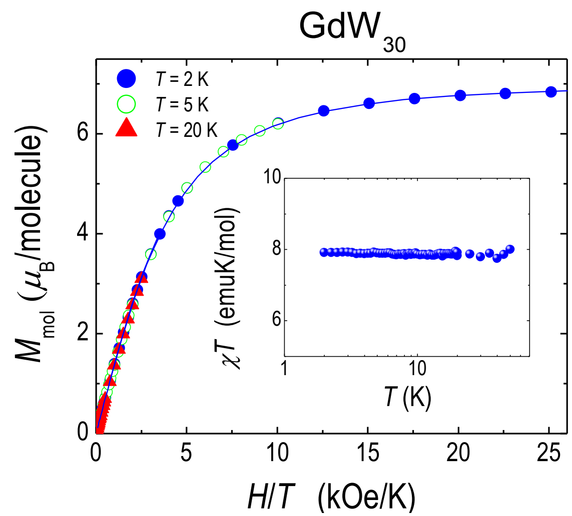
➡ Model crystal-field probe

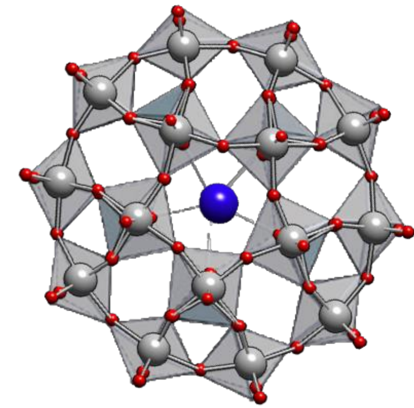
M. J. Martínez, S. Cardona, C. Schlegel, F. Moro, P. J. Alonso, H. Prima-García, J. M. Clemente, M. Evangelisti, A. Gaita, J. Sesé, J. van Slageren, E. Coronado, and F. Luis, Phys. Rev. Lett. **108**, 247213 (2012).

Magnetization curves: spin values









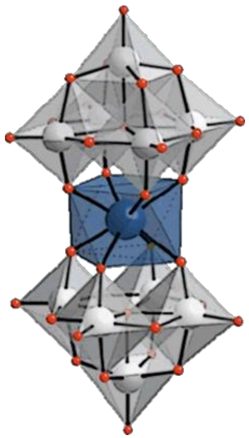
GdW_{10}

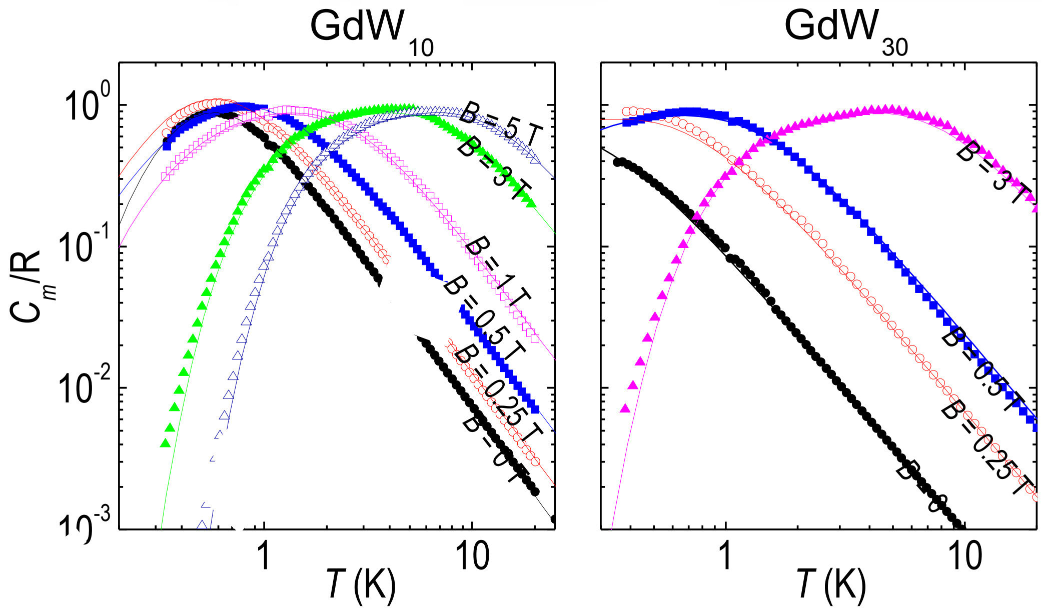
$S = 7/2$
$g = 2$

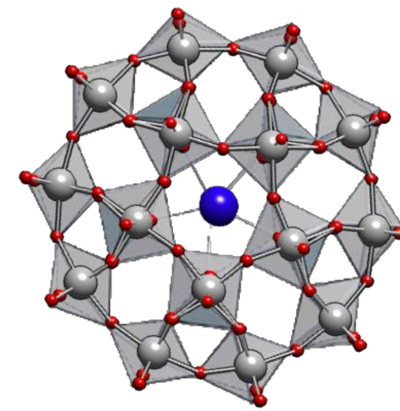
GdW_{30}

$S = 7/2$
$g = 2$

Heat capacity & EPR: zero-field splitting

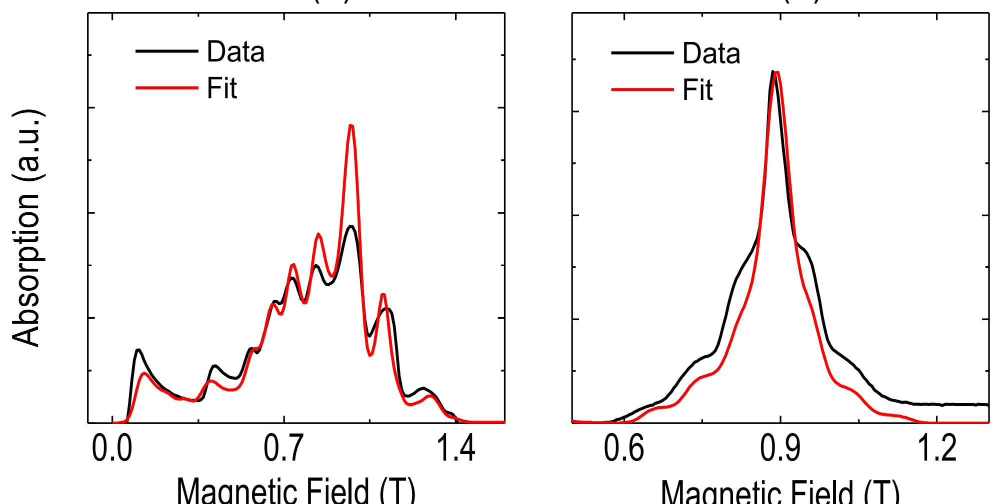






GdW_{10}

S = 7/2
g = 2



GdW_{30}

S = 7/2
g = 2

B_{20}/k_B = -0.059 K

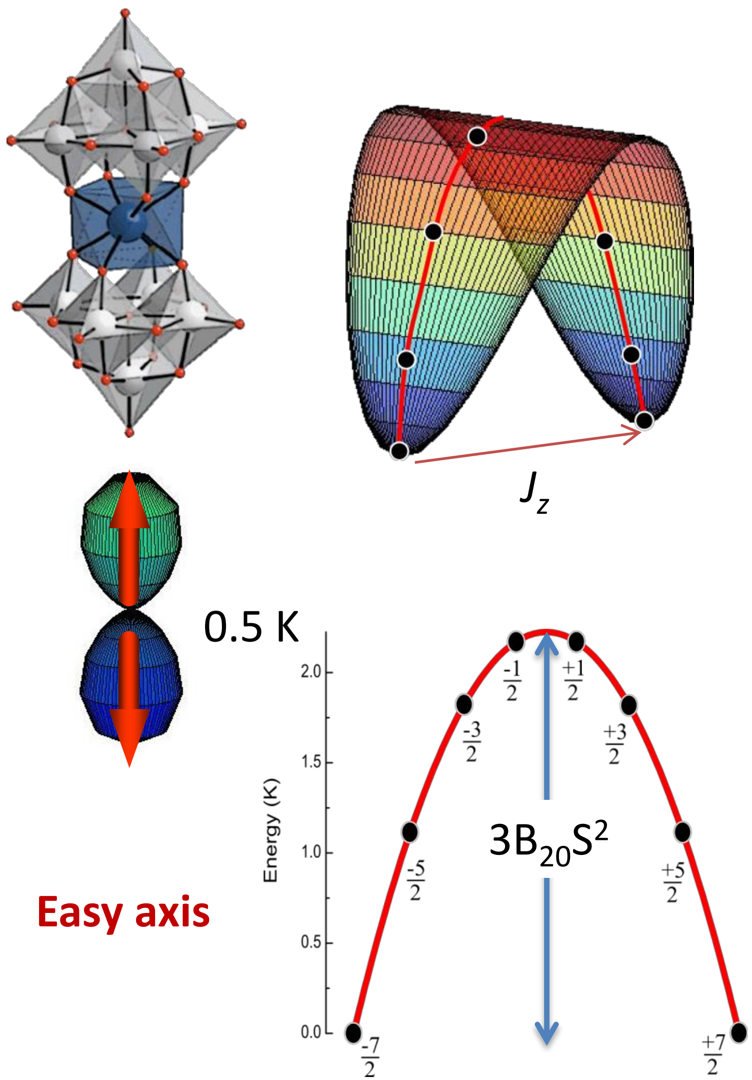
$B_{44}/k_B \approx 4 \times 10^{-4}$ K

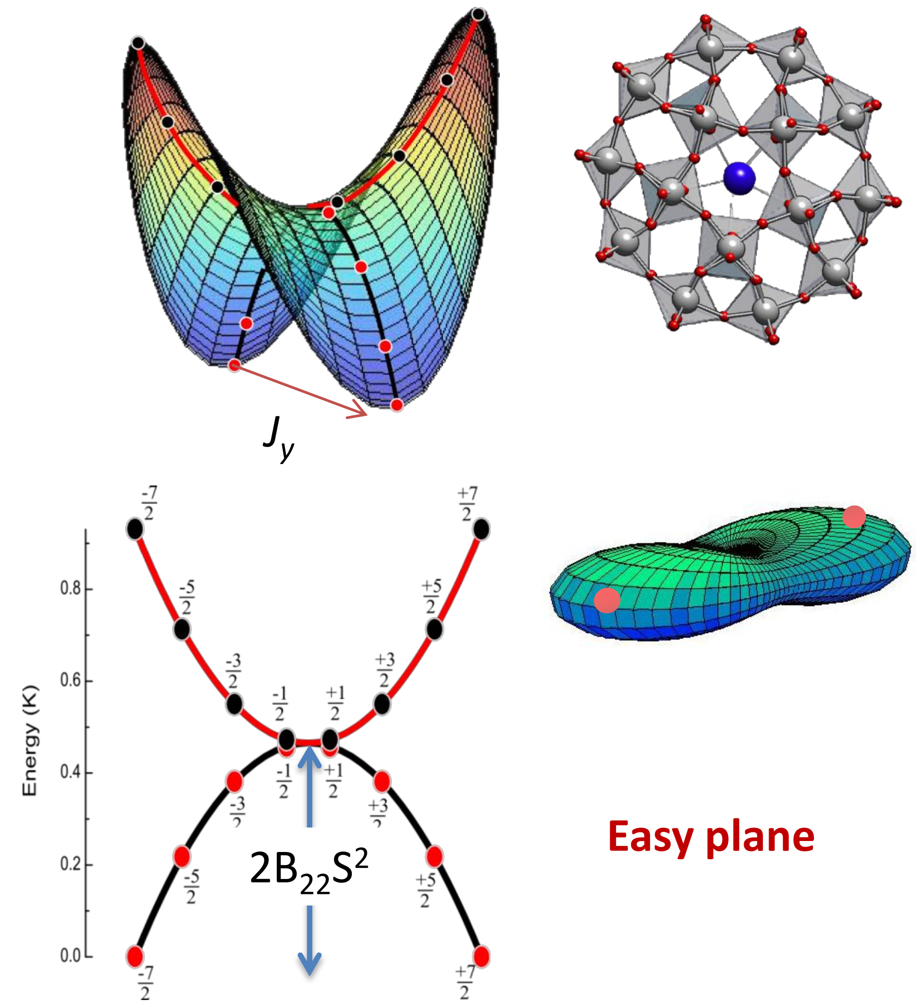
$$\mathcal{H} = B_{20}O_2^0 + B_{22}O_2^2$$

B_{20}/k_B = +0.019 K

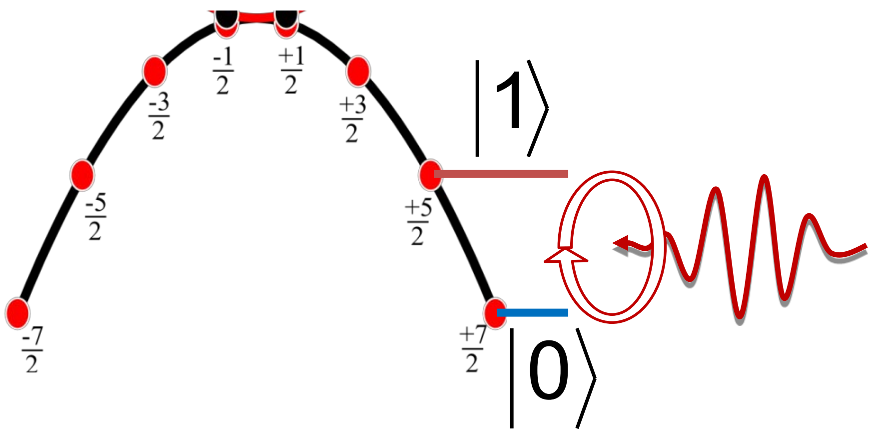
$B_{22}/k_B \approx$ +0.019 K

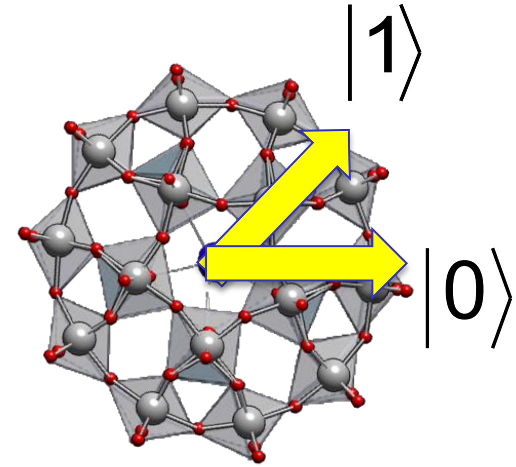
Molecular design of the spin Hamiltonian





Quantum coherence: pulsed EPR





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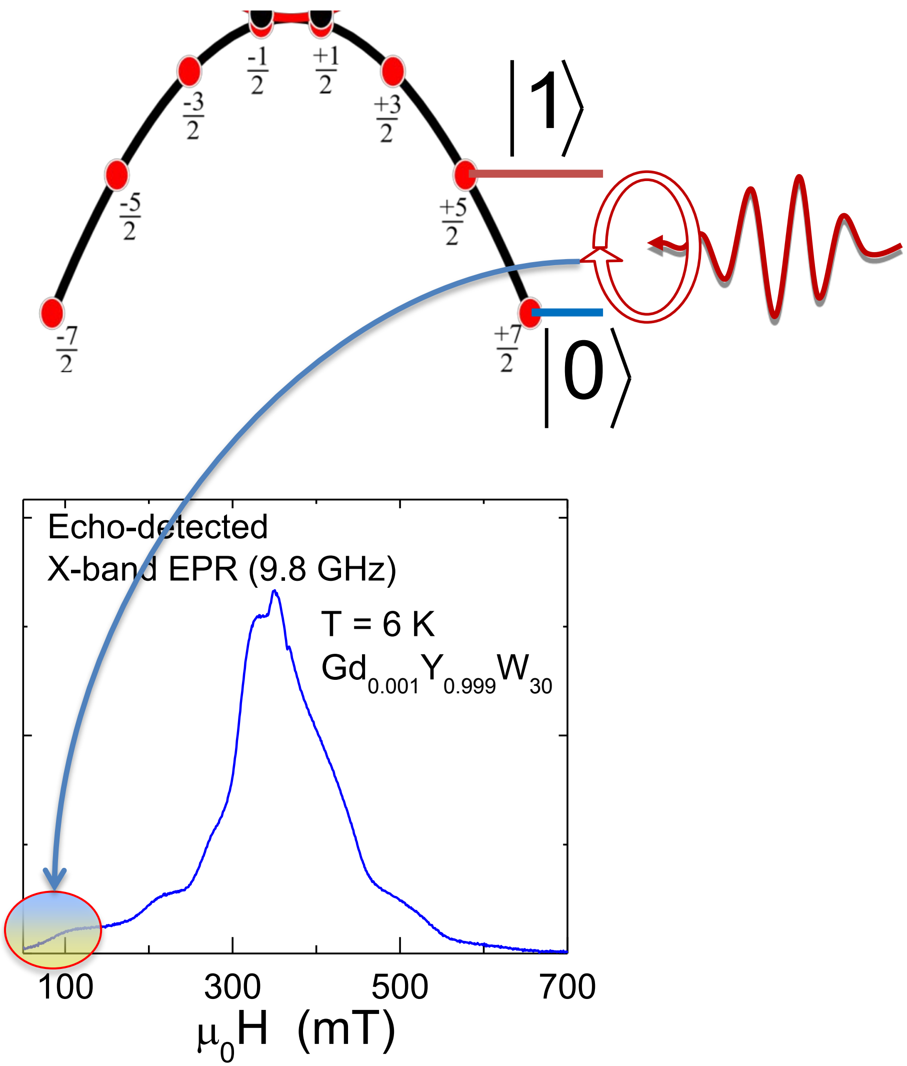

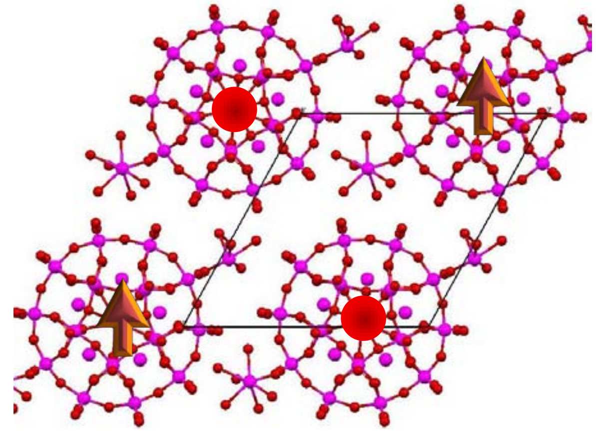

Magnetically diluted samples: $Gd_xY_{1-x}W_{30}$

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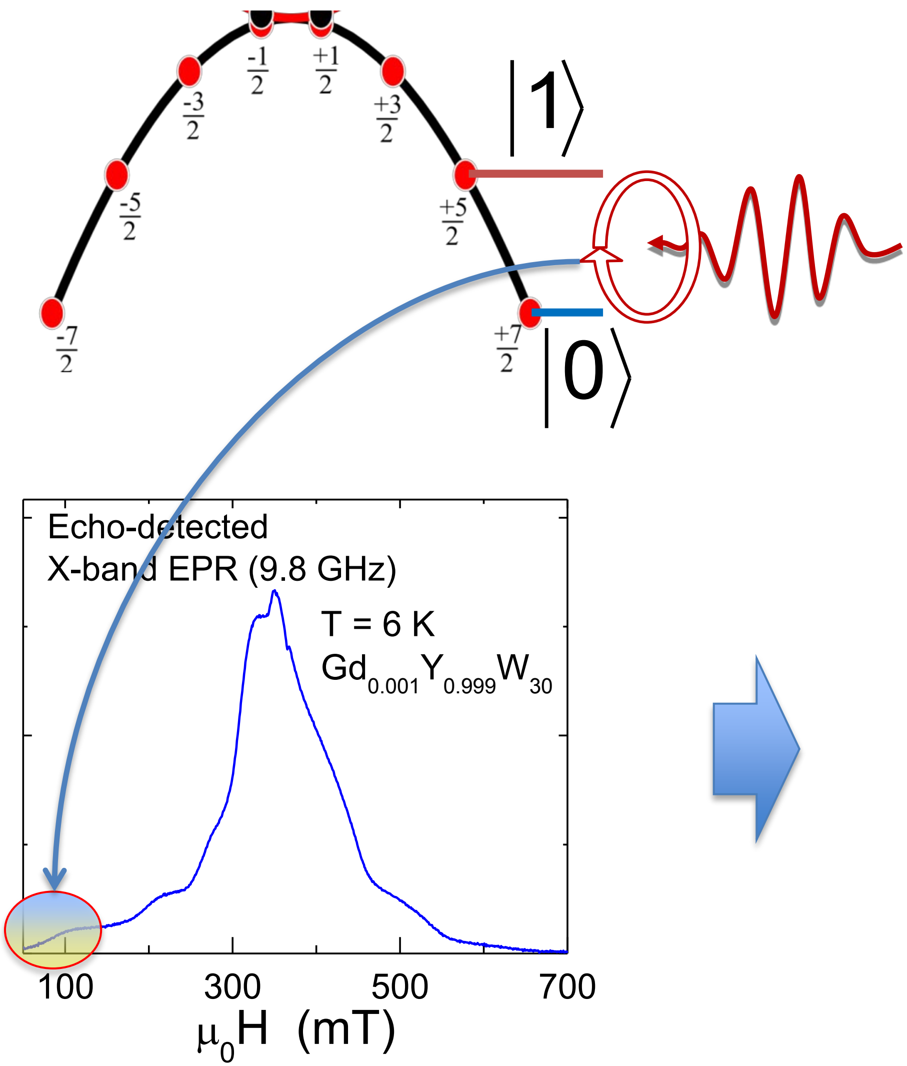
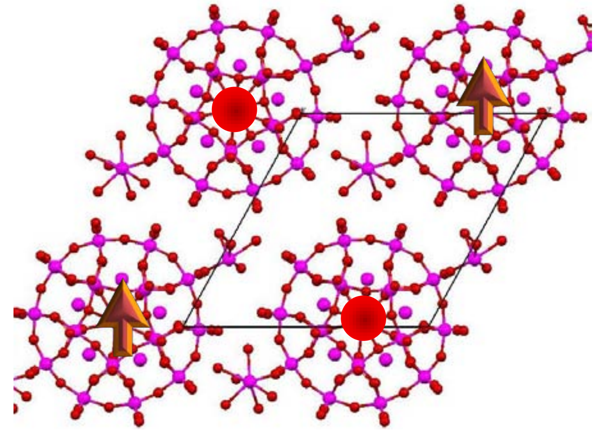
Quantum coherence: pulsed EPR





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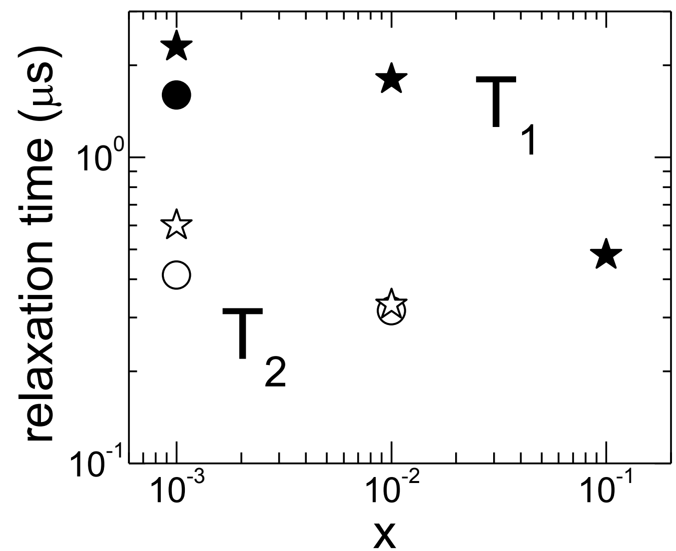
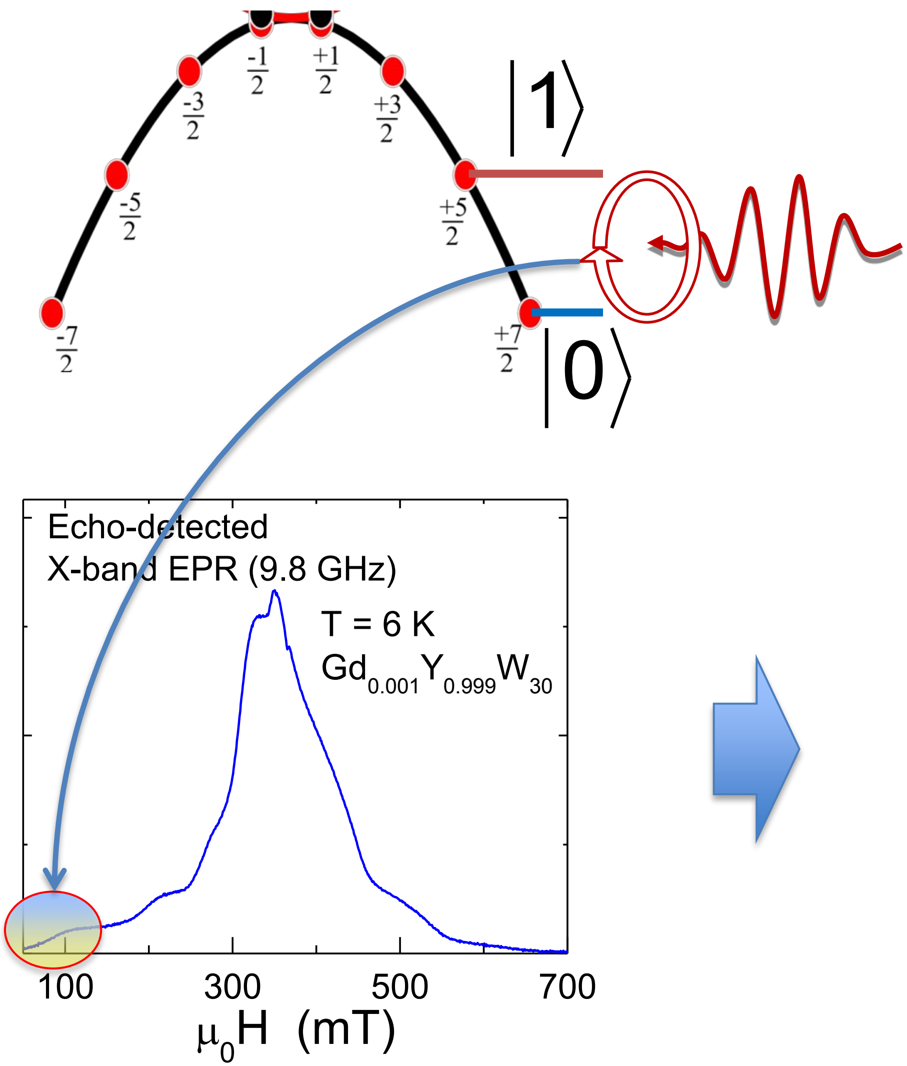
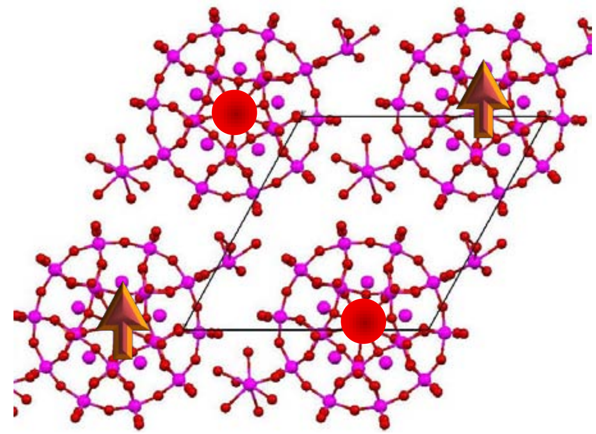


T_2 and T_1 increase with decreasing x

$T_2 \approx 450$ ns for $x = 0.001$

Quantum coherence: pulsed EPR





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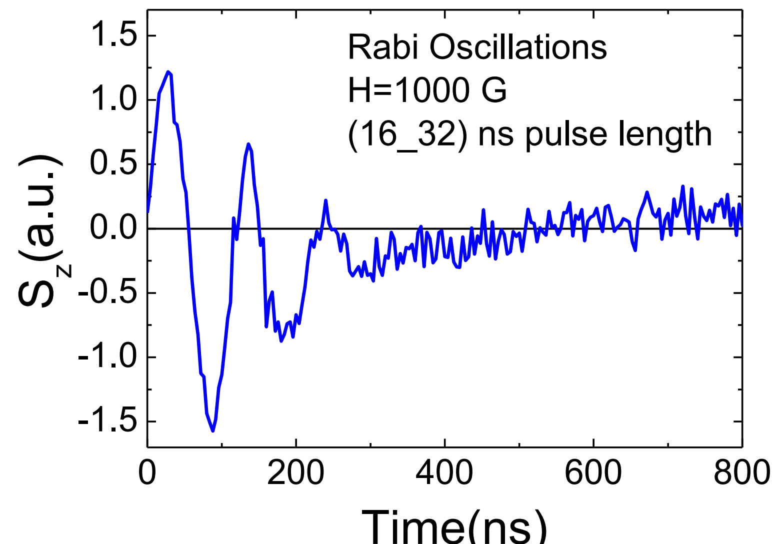
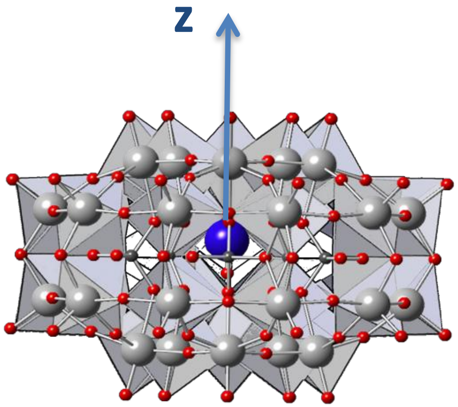
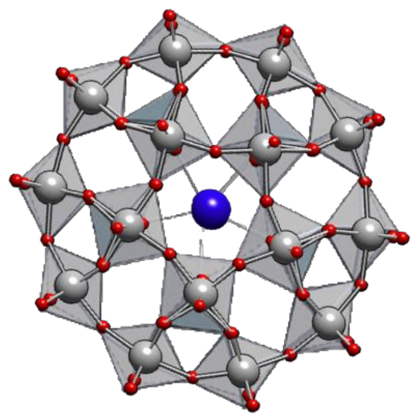
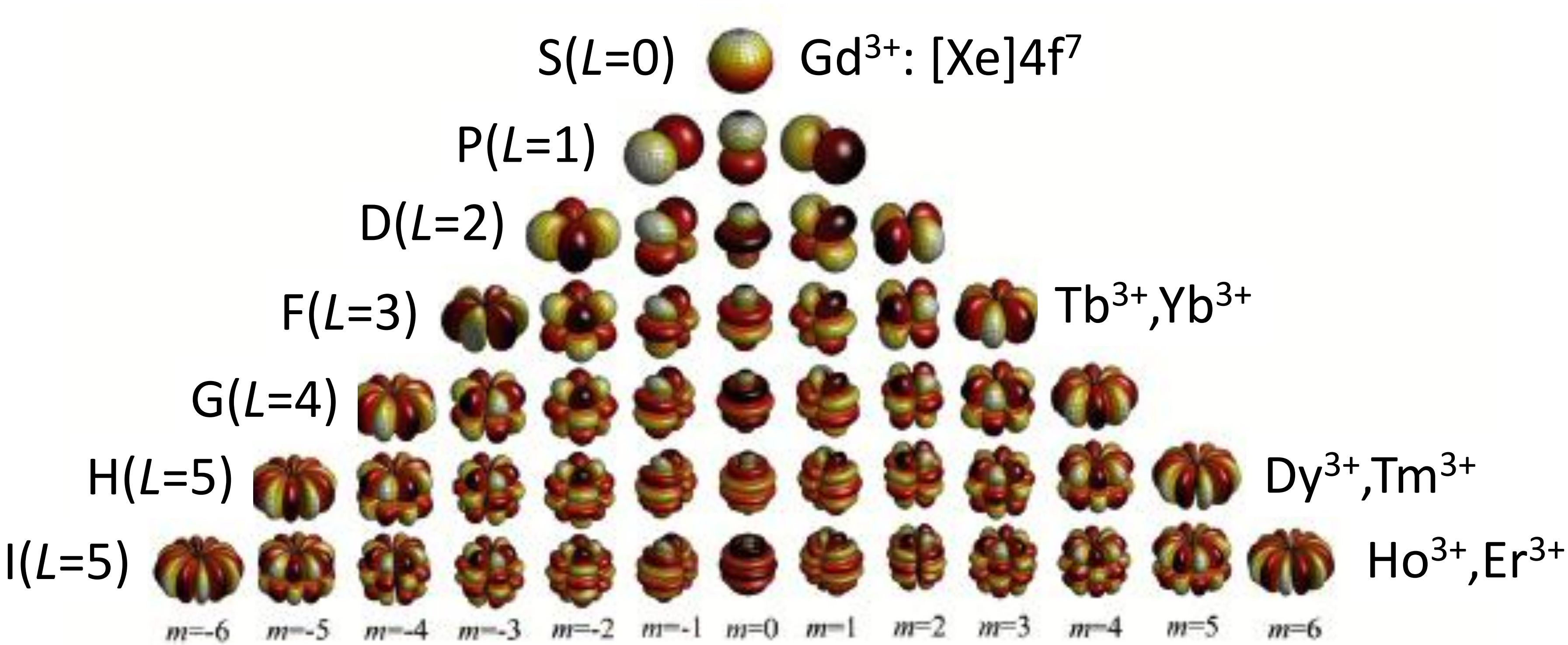


Direct observation of coherent Rabi oscillations

Extension to other lanthanides: SIM with fivefold symmetry







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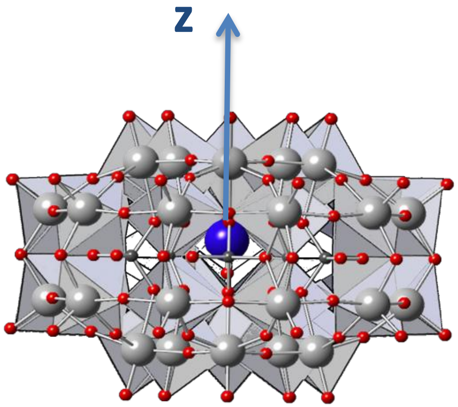
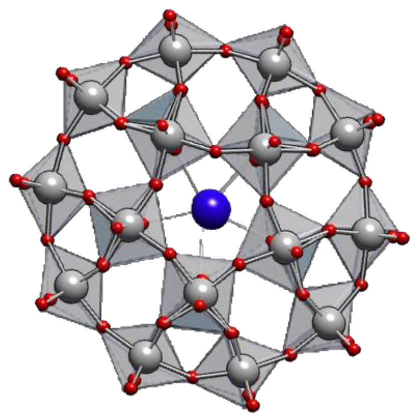
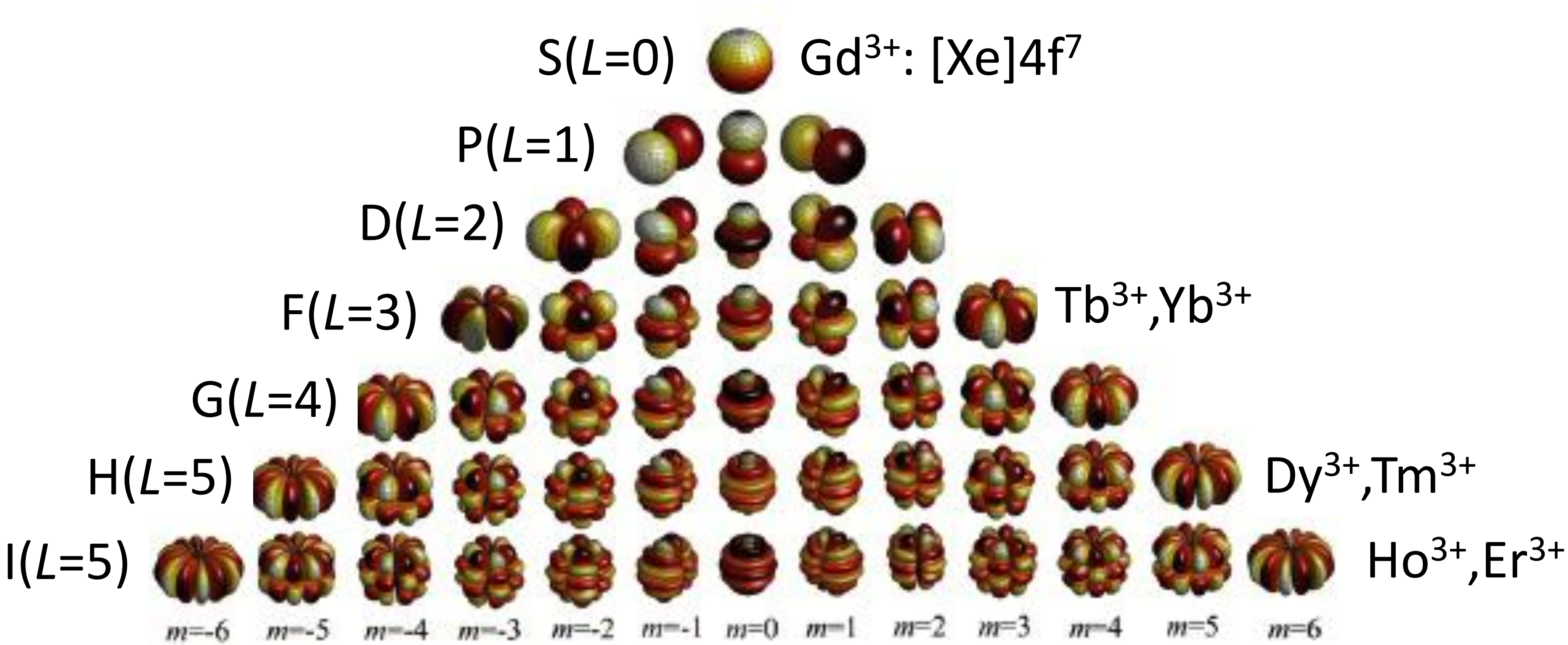




$$\mathcal{H} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^5 O_6^5$$



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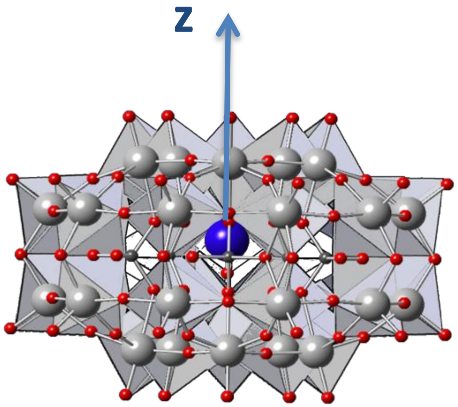

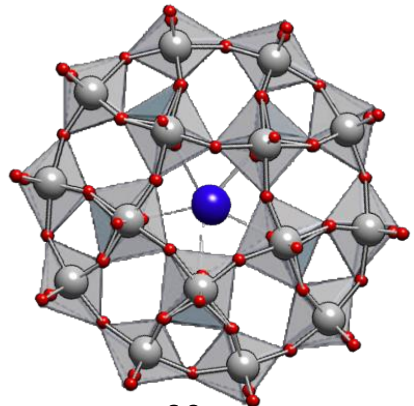

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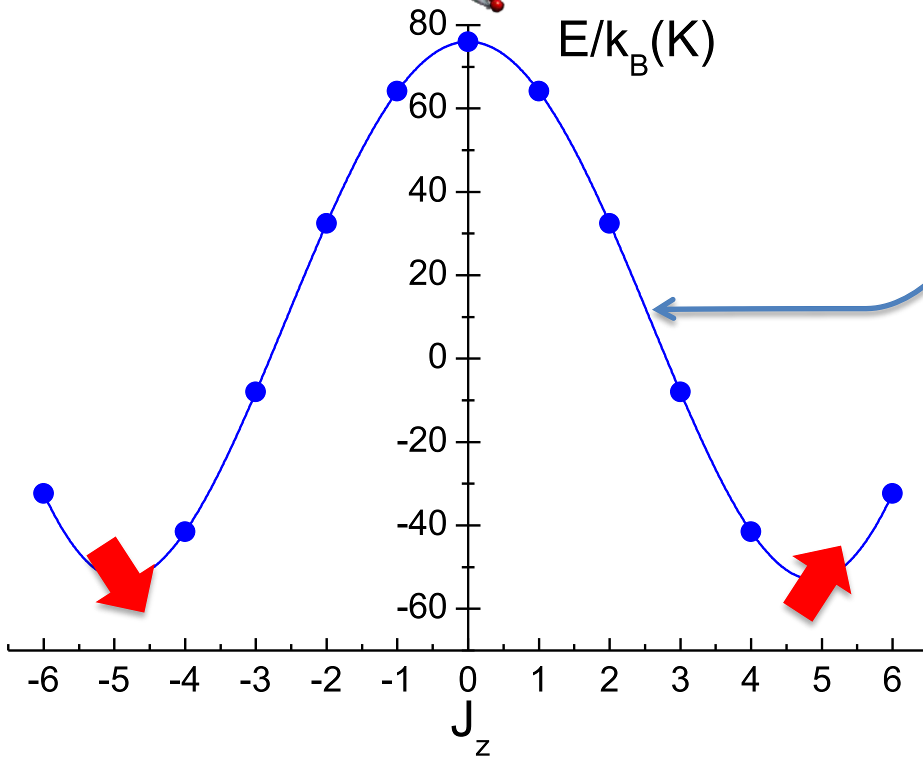
Diagonal terms

$$\left[J_z, O_m^0\right] = 0$$



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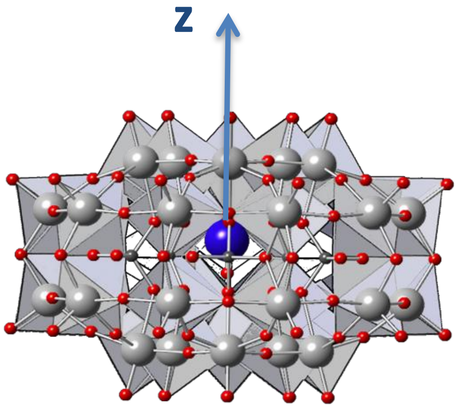
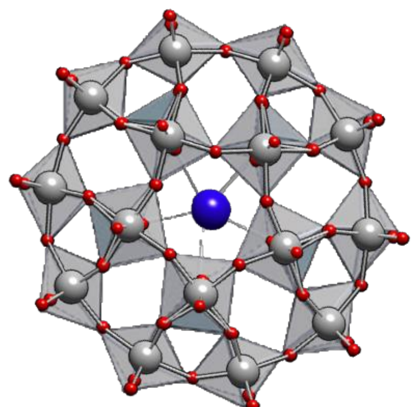
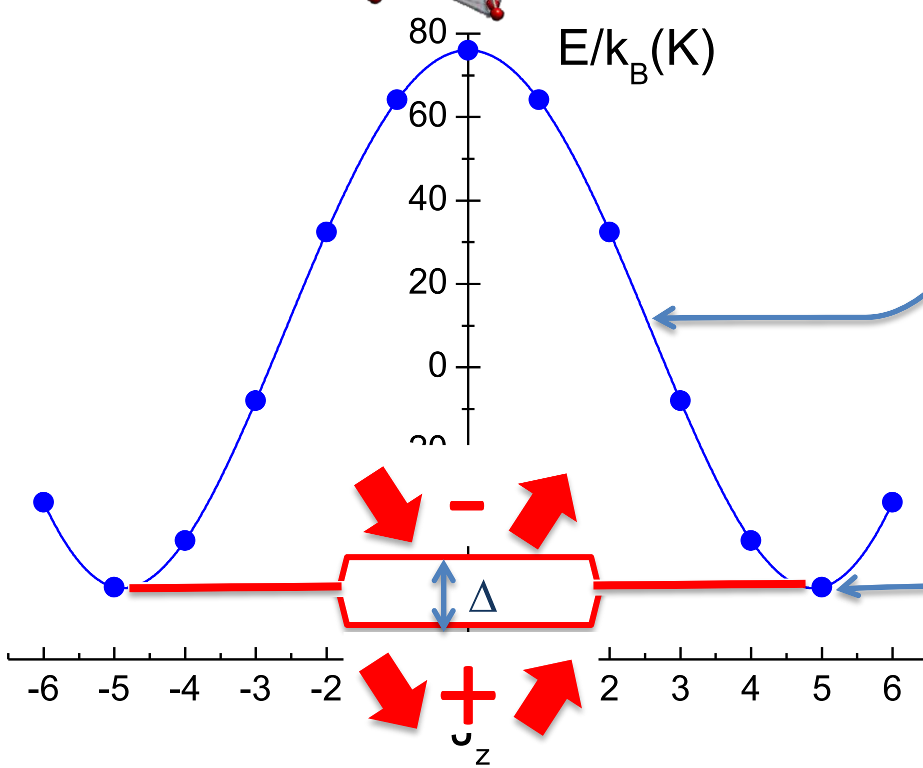




$$\mathcal{H} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^5 O_6^5$$

Diagonal terms

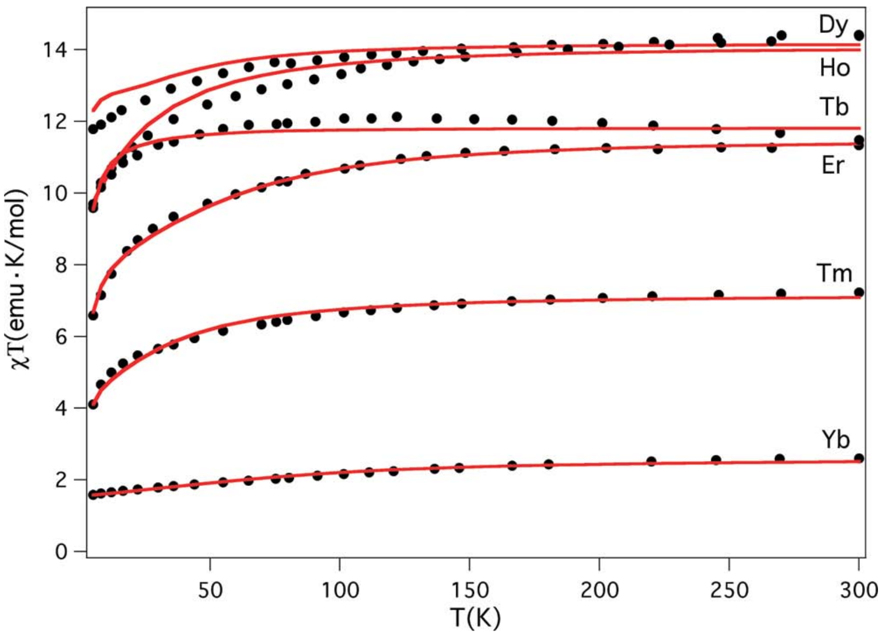
$$\left[J_z, O_m^0 \right] = 0$$

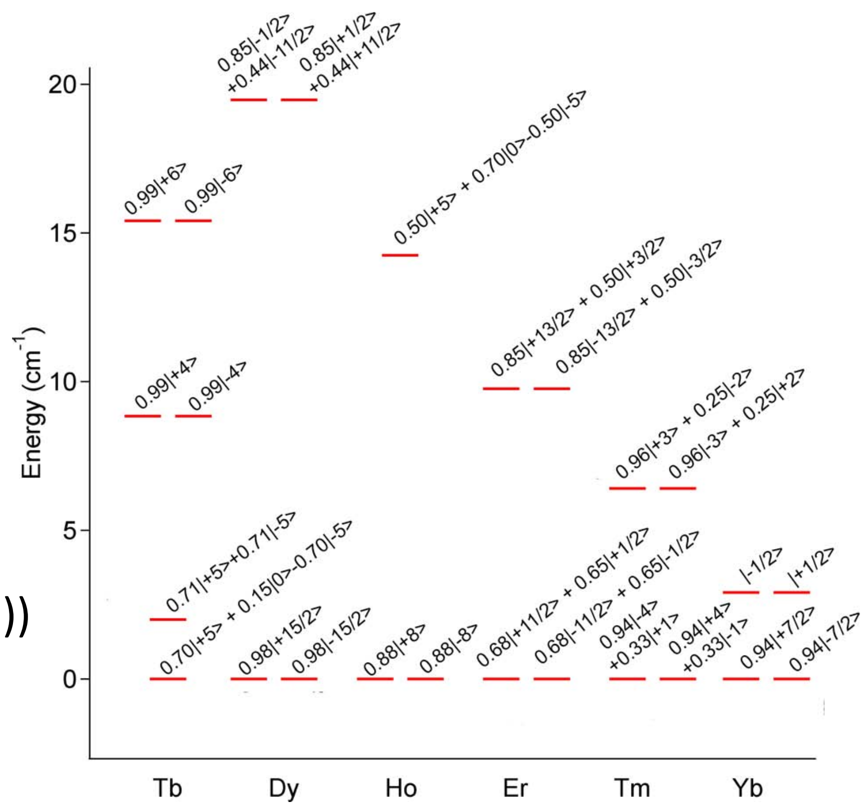


Off-diagonal terms

$$\left[J_z, O_m^0 \right] \neq 0$$

Determination of anisotropy parameters





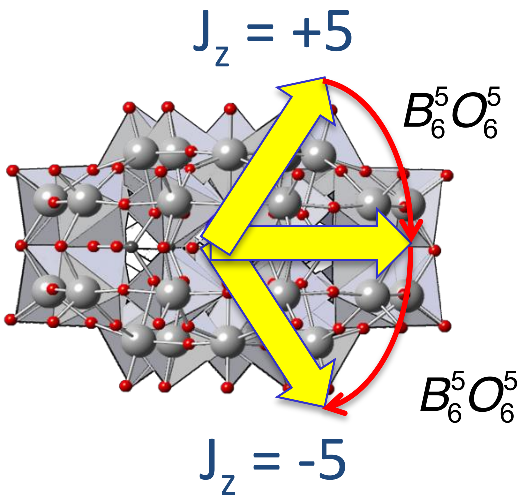
Ishikawa et al method (Inorg. Chem. **42**, 2440 (2003))

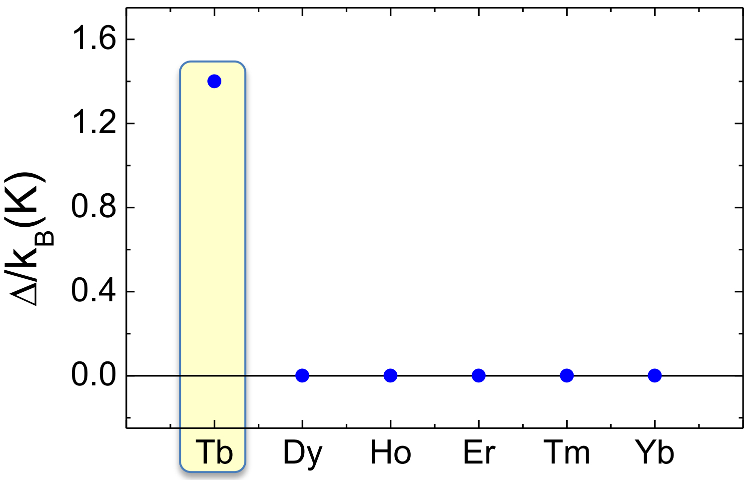
$J_z =$ ±5 ±15/2 ±8 ±11/2 ±4 ±7/2

- Simultaneous fit of all χT curves
- Constraint: $B_n^m = a_n^m + b_n^m(n_f - 7)$

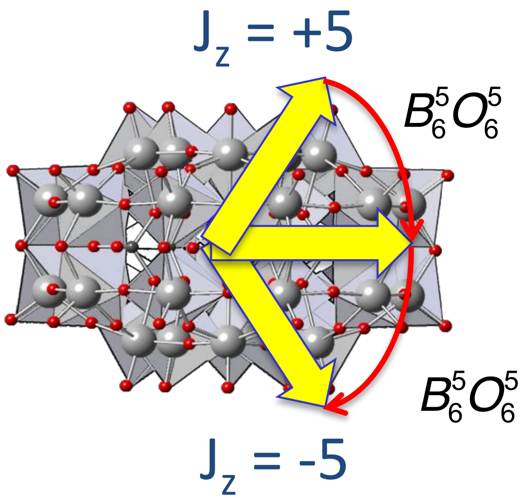
S. Cardona-Serra, J. M. Clemente-Juan, E. Coronado, A. Gaita-Ariño, A. Camón, M. Evangelisti, F. Luis, M. J. Martínez-Pérez, and J. Sesé, JACS **134**, 14982 (2012).

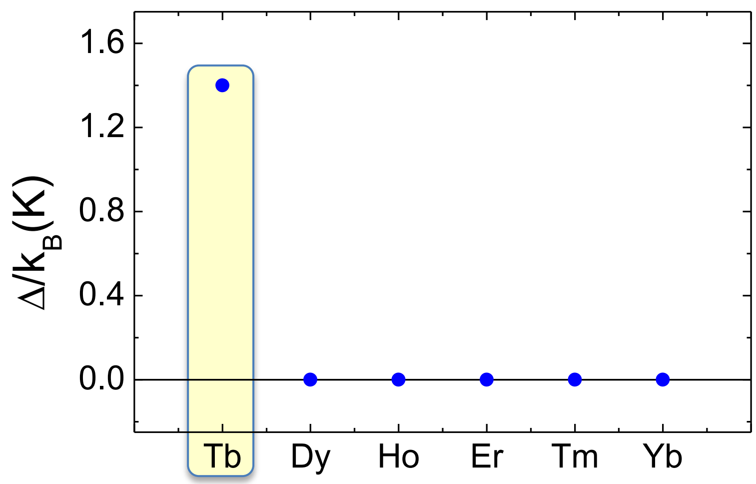
Quantum tunnel splitting



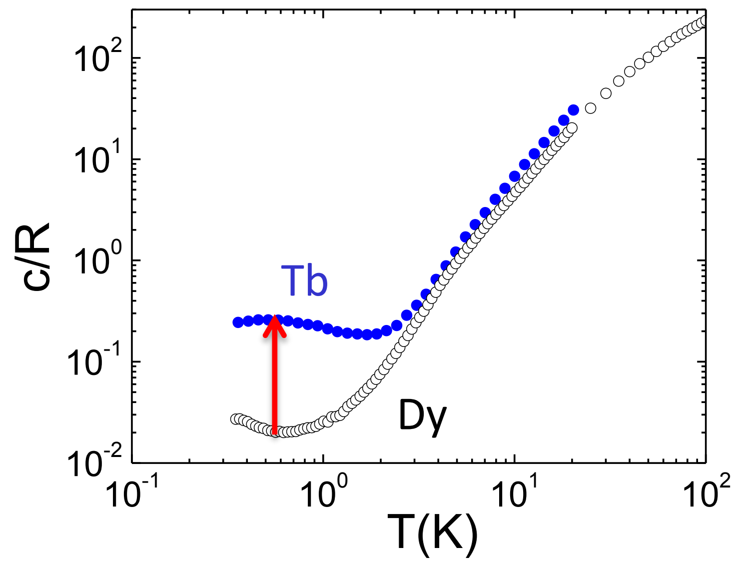


Quantum tunnel splitting

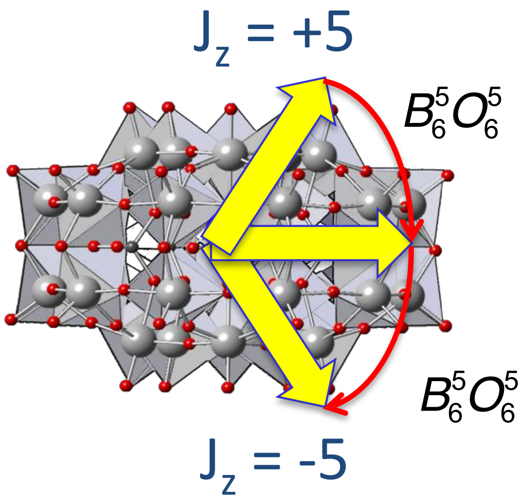


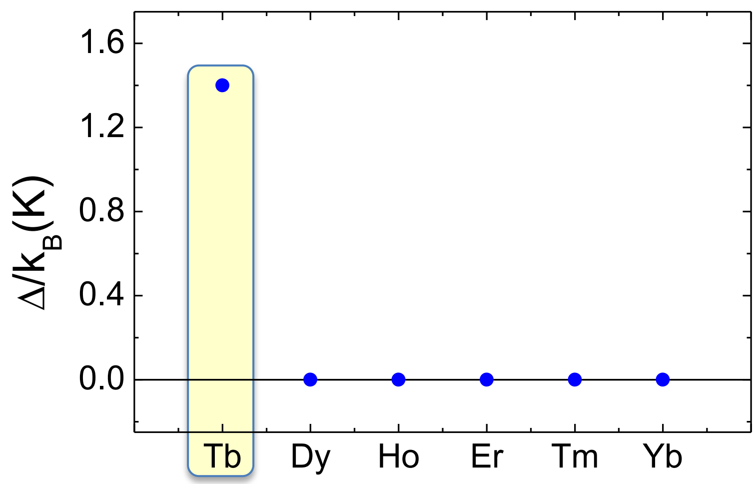


Direct detection of Δ by heat capacity measurements

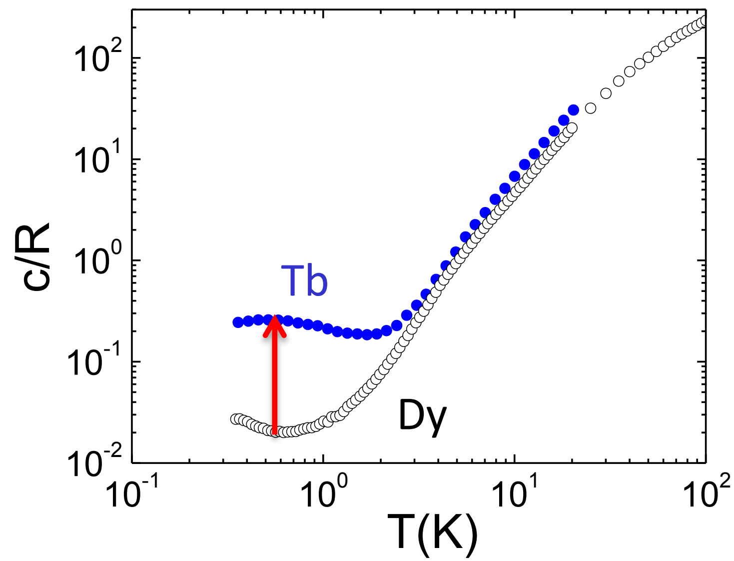


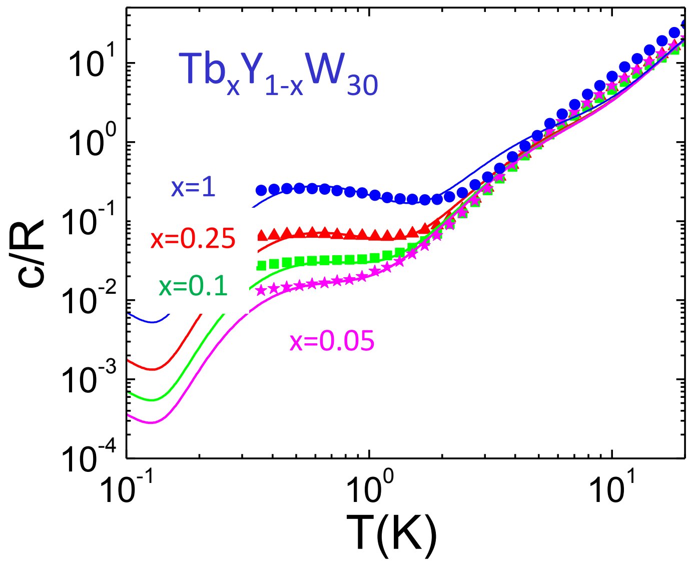
Quantum tunnel splitting



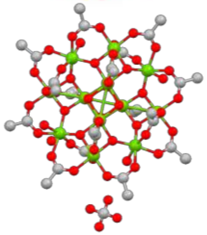


Direct detection of Δ by heat capacity measurements



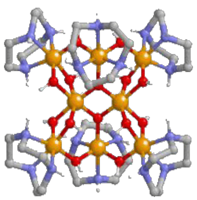


Strong quantum regime



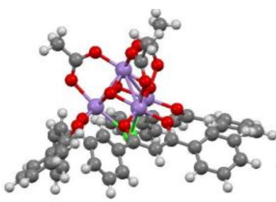
Mn_{12}

$\Delta/\hbar \approx 2$ Hz (10^{-10} K)



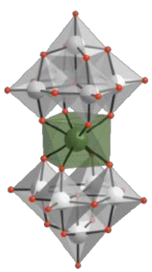
Fe_8

$\Delta/\hbar \approx 200$ Hz (10^{-8} K)



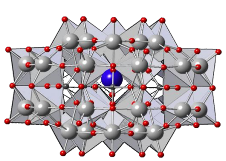
Mn_4Cl

$\Delta/\hbar \approx 21$ kHz (10^{-6} K)



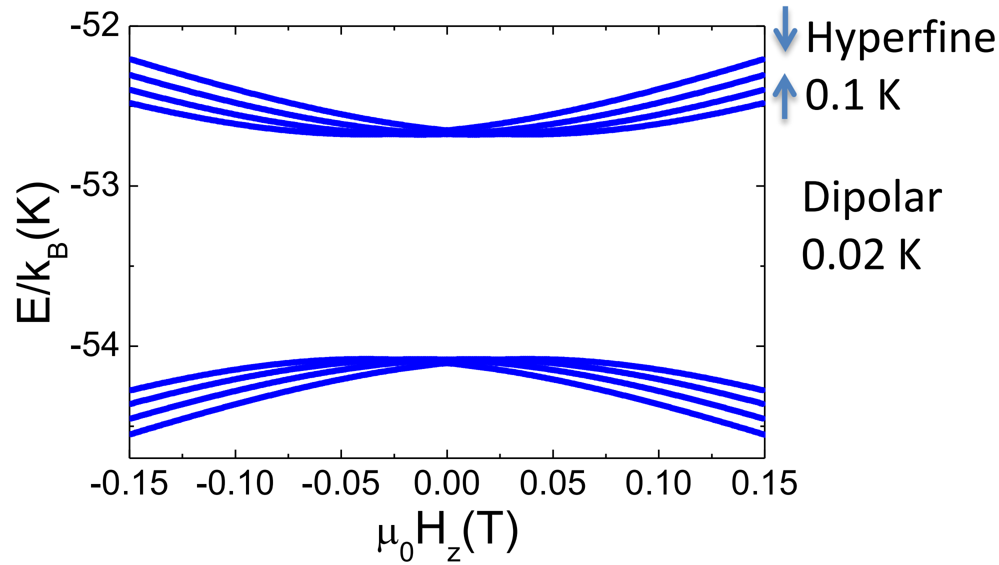
ErW_{10}

$\Delta/\hbar \approx 2$ MHz (10^{-4} K)

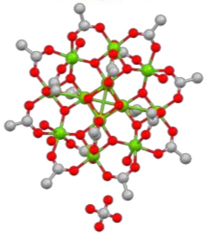


TbW_{30}

$\Delta/\hbar \approx 28$ GHz!! (1 K)

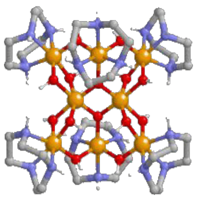


Strong quantum regime



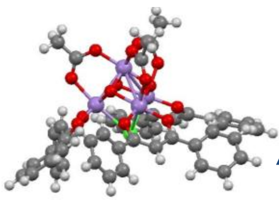
Mn_{12}

$\Delta/\hbar \approx 2$ Hz (10^{-10} K)



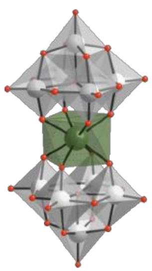
Fe_8

$\Delta/\hbar \approx 200$ Hz (10^{-8} K)



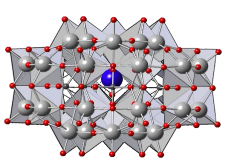
Mn_4Cl

$\Delta/\hbar \approx 21$ kHz (10^{-6} K)



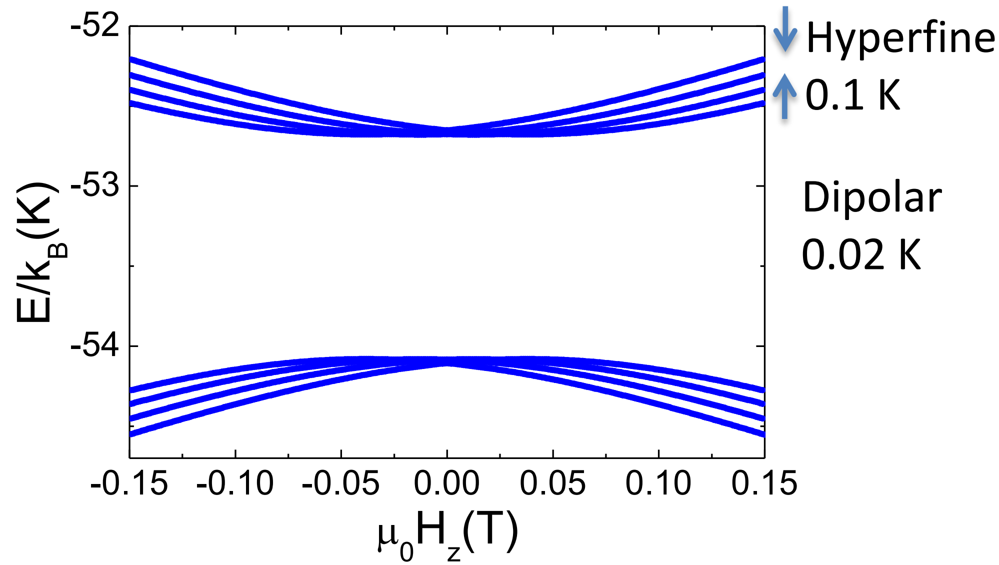
ErW_{10}

$\Delta/\hbar \approx 2$ MHz (10^{-4} K)

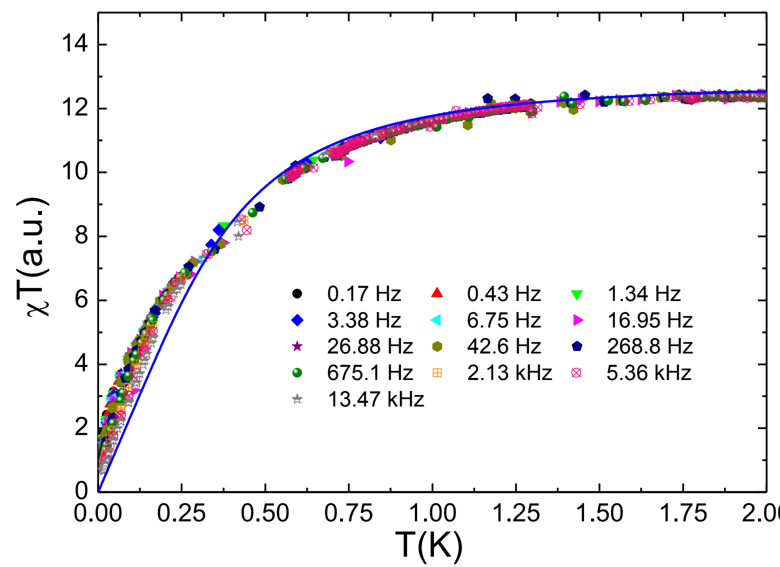


TbW_{30}

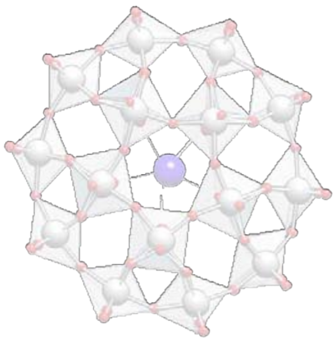
$\Delta/\hbar \approx 28$ GHz!! (1 K)

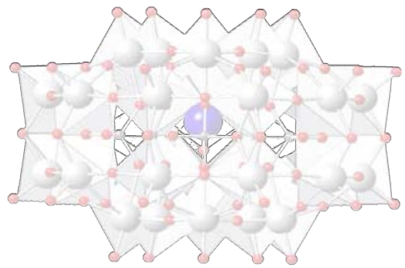


Suppression of μ_{eff} by quantum fluctuations

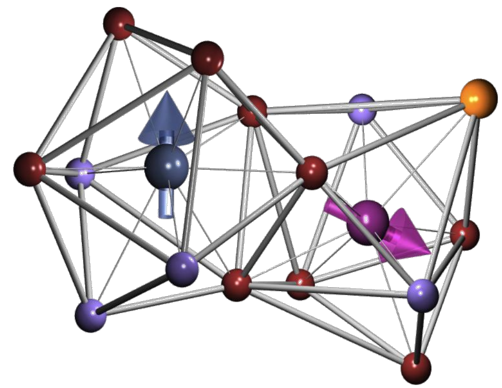


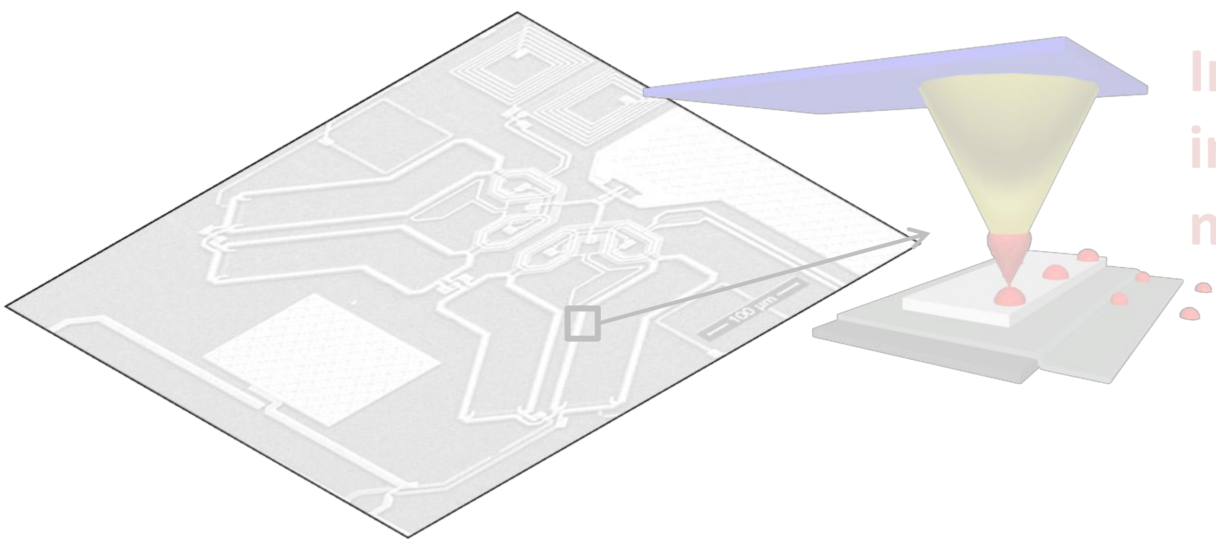
Molecular qubits





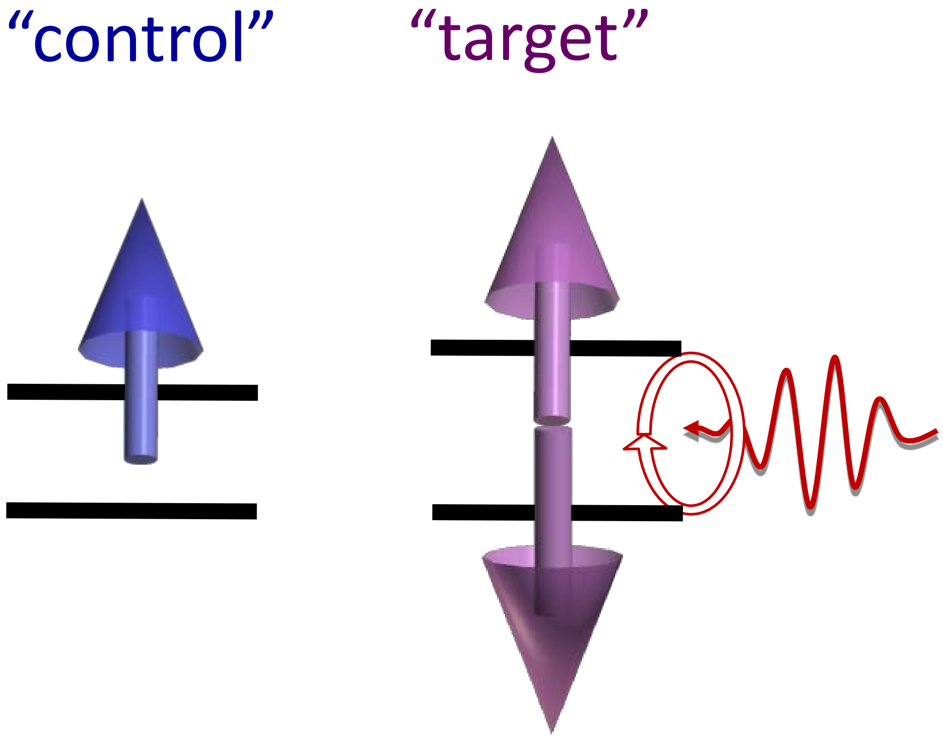
Molecular quantum gates



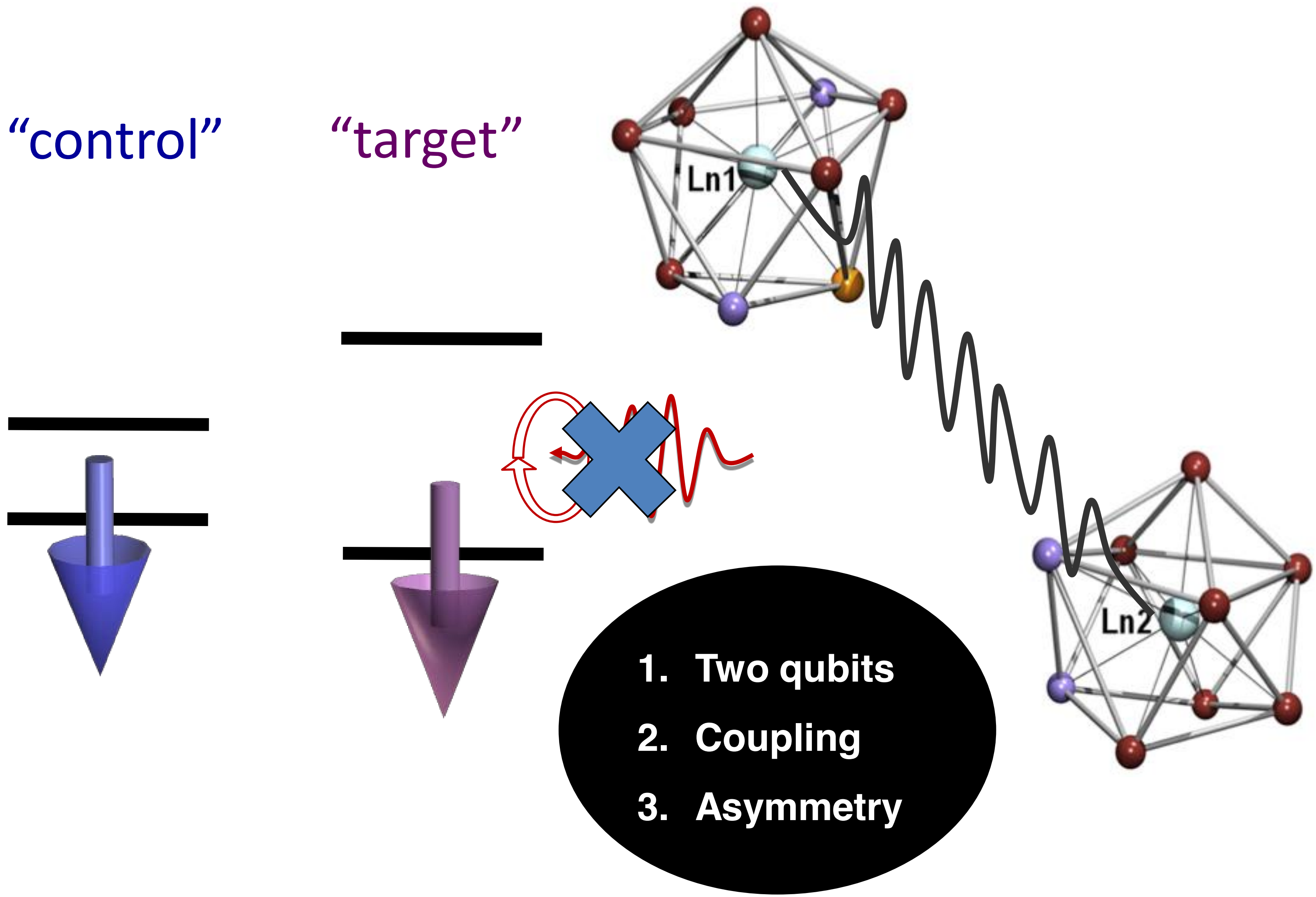


Integration of SMM into superconducting microdevices

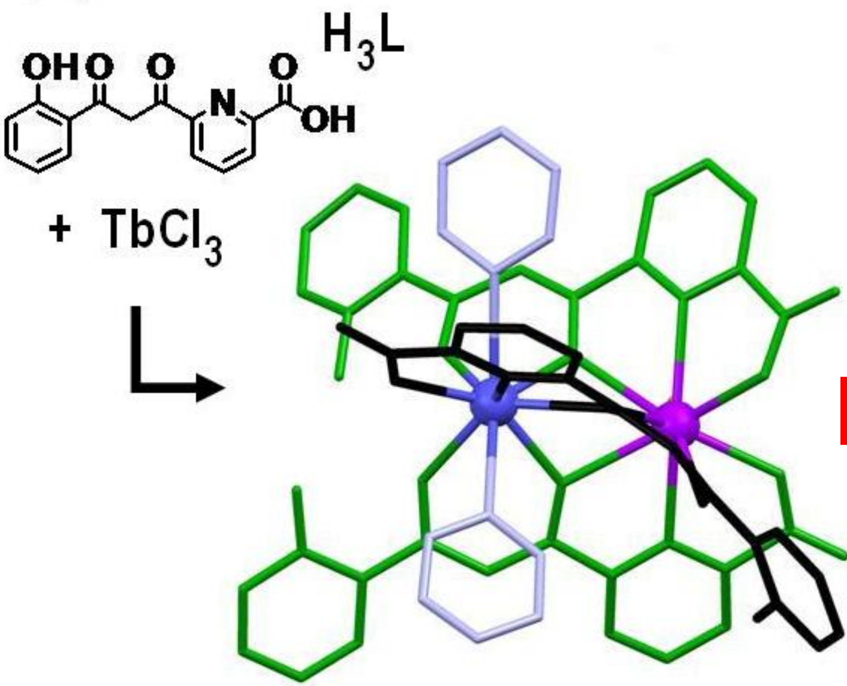
Universal CNOT quantum gate

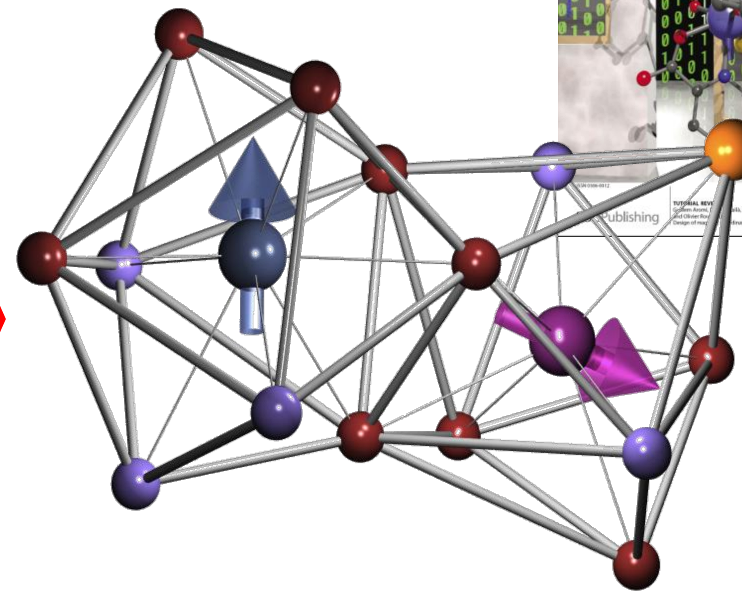


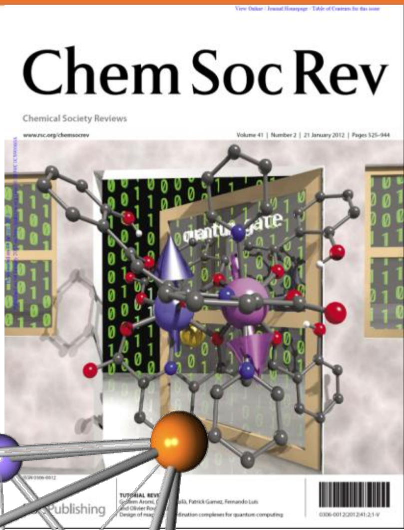
Universal CNOT quantum gate



Molecular design







Dinuclear $[Tb]_2$ complex

Linked to three asymmetric H_3L ligands

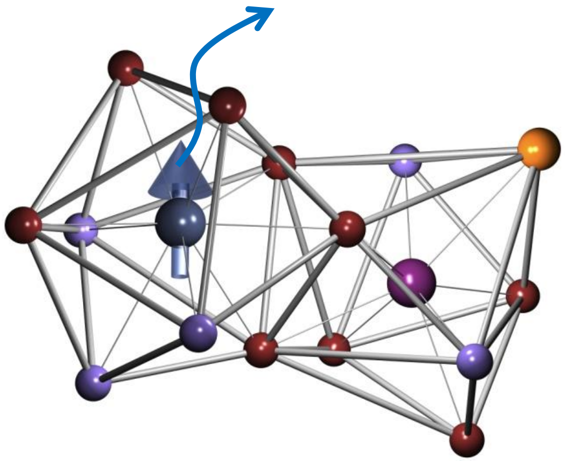
Two anisotropic spins in different coordinations

D. Aguilà *et al*, Inorg. Chem. **49** (2010) 6784

G. Aromí, D. Aguilà, P. Gámez, F. Luis, and O. Roubeau, Chem. Soc. Rev. **41**, 537-546 (2012).

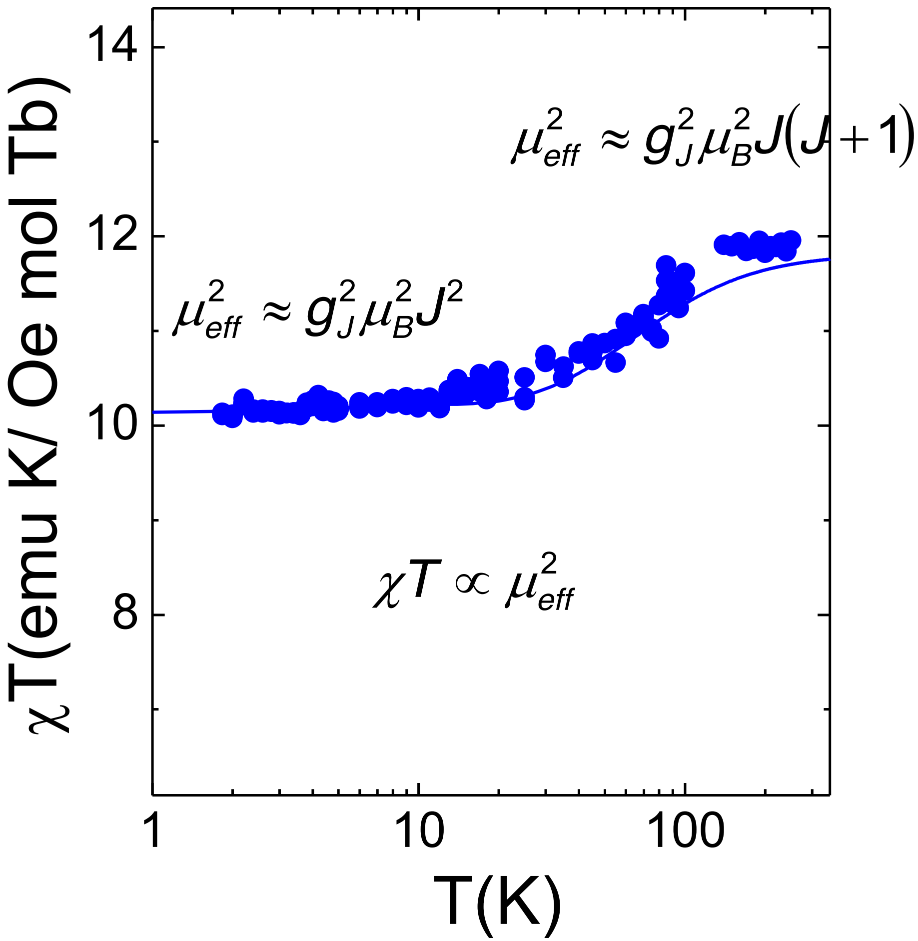
Definition of qubit states

[LaTb] $J = 6$, $g_J = 3/2$

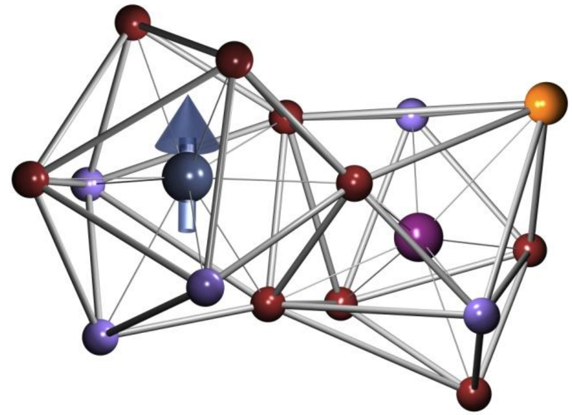


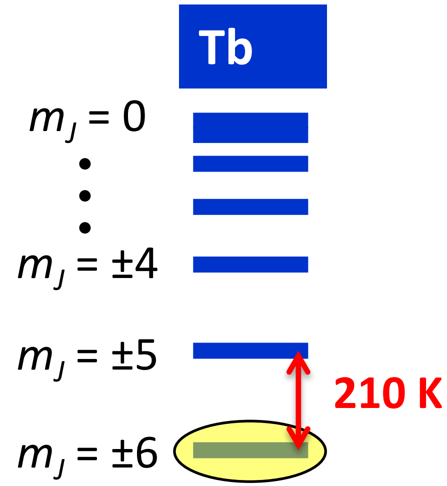
Definition of qubit states

[LaTb] $J = 6$, $g_J = 3/2$



$$\mathcal{H}_{anis} = -DS_z^2$$

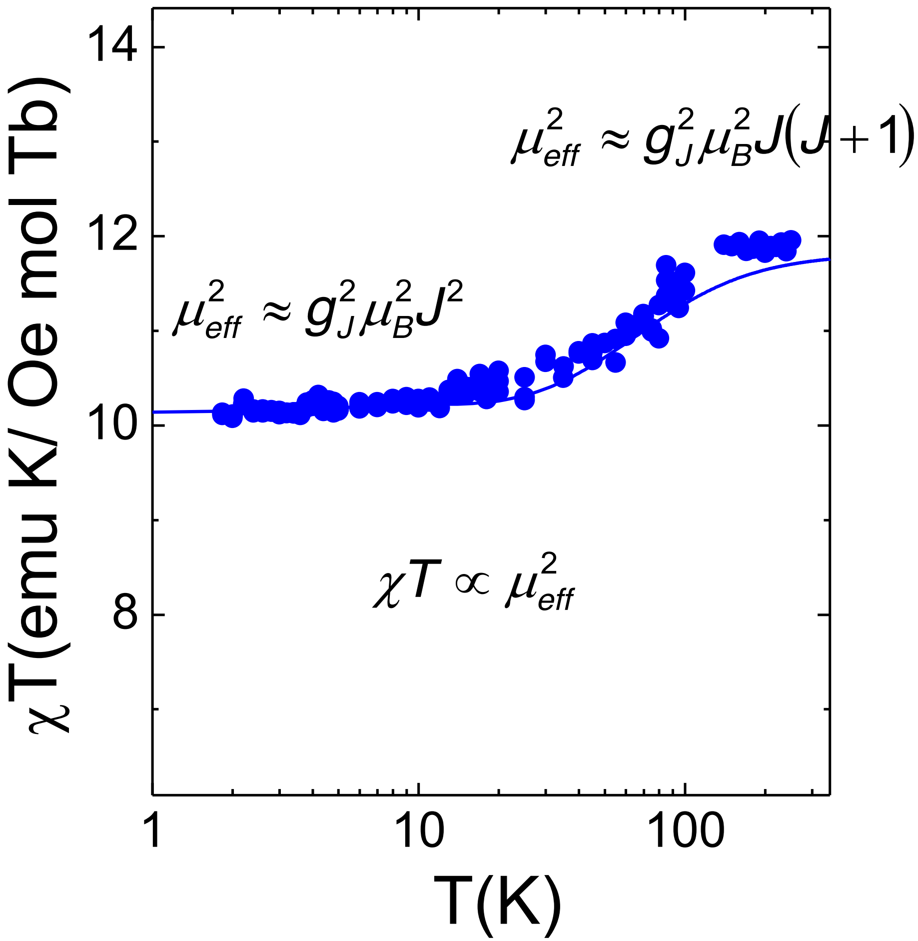


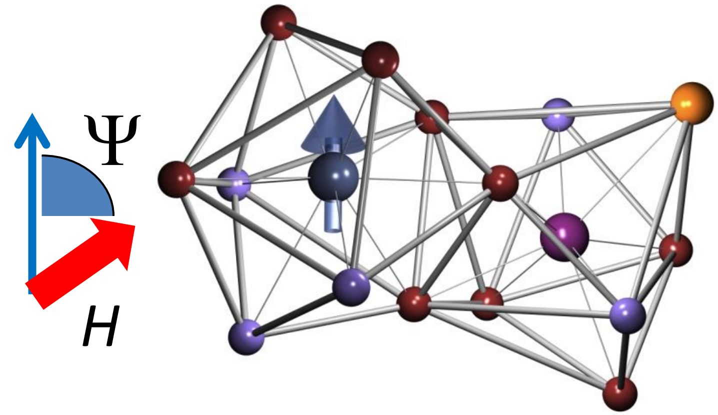


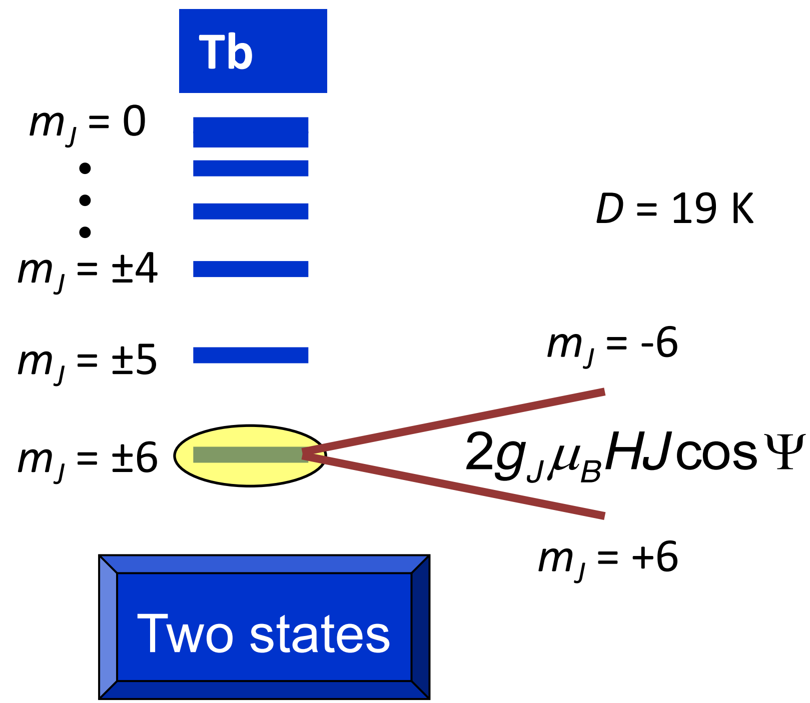
Two states

Definition of qubit states

[LaTb] $J = 6,\ g_J = 3/2$

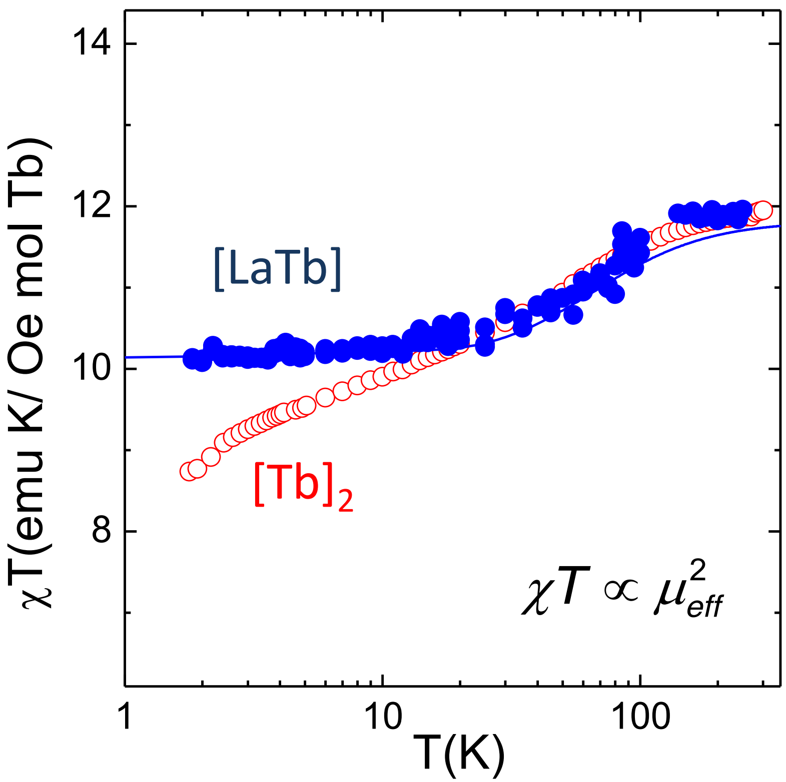




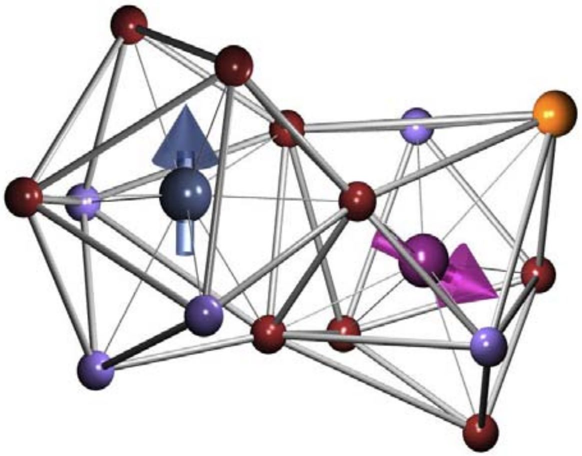


$$\mathcal{H}_{anis} = -DS_z^2 - g_J \mu_B H_z J_z$$

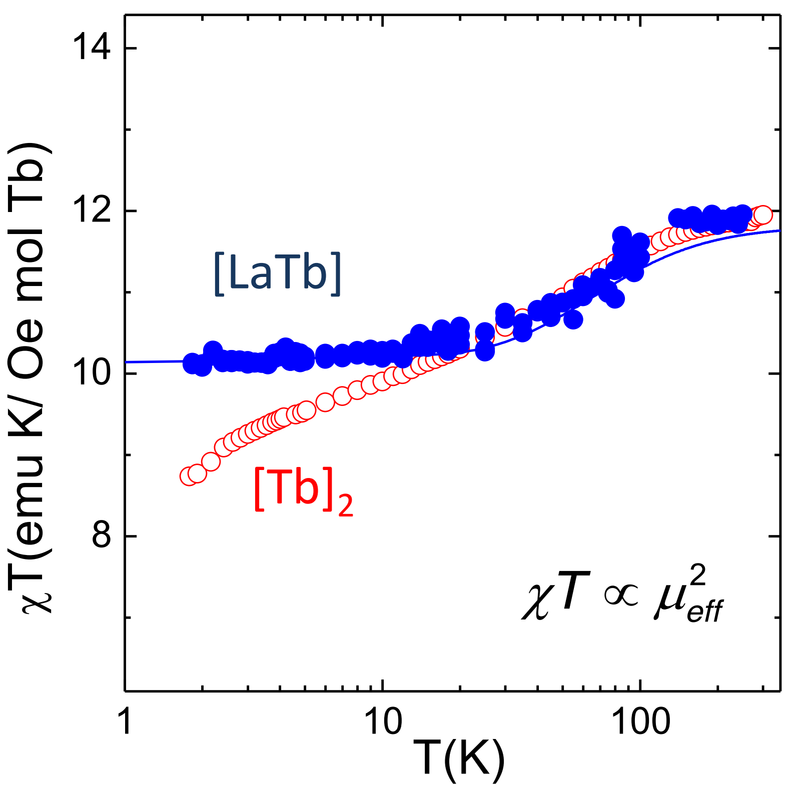
Coupling between the Tb^{3+} qubits

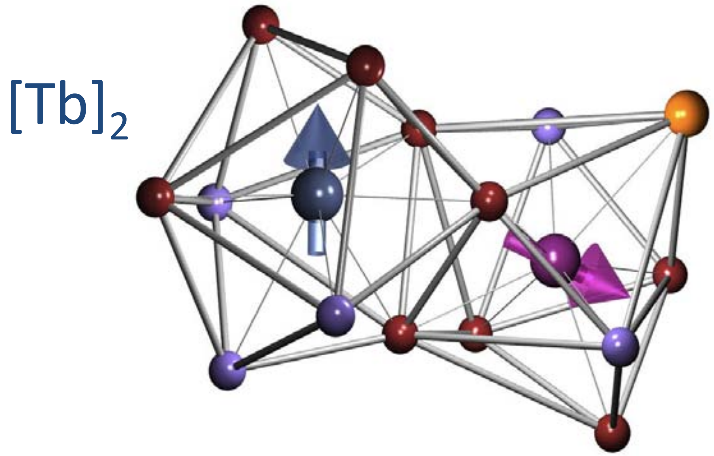


$[Tb]_2$

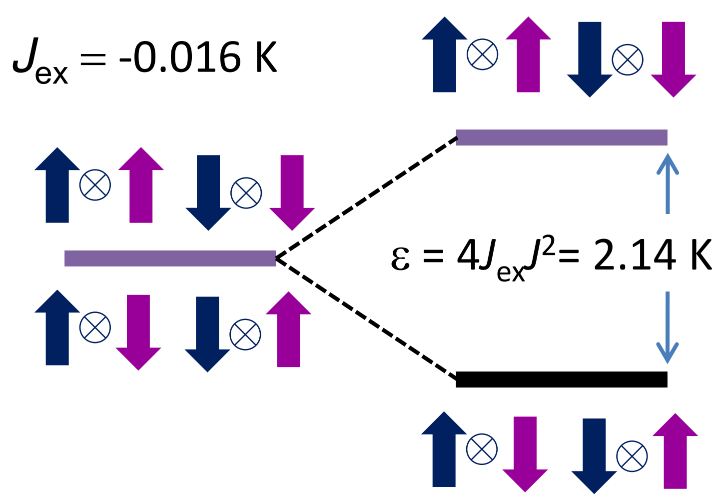


Coupling between the Tb^{3+} qubits



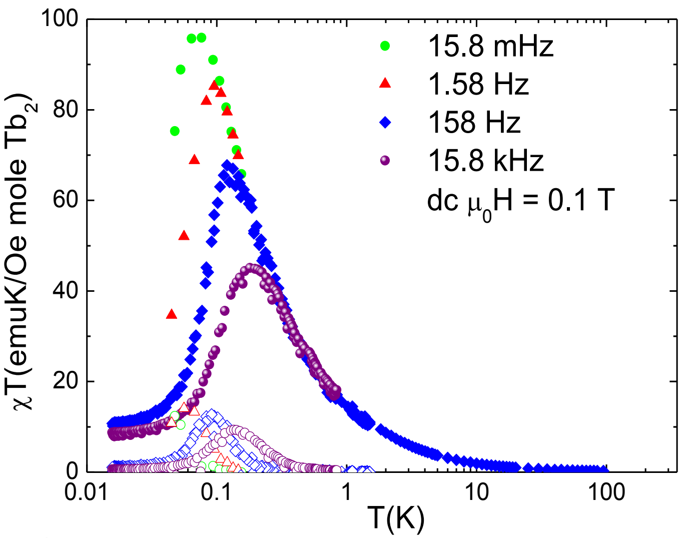


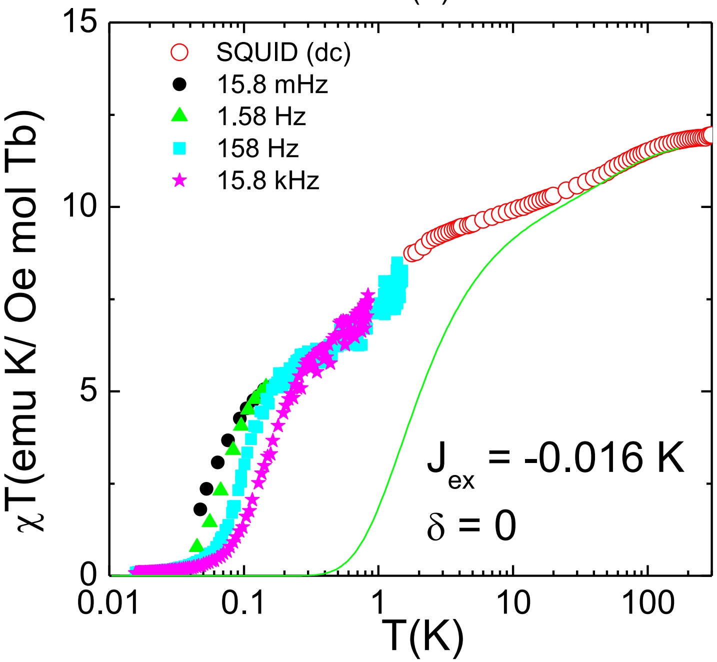
AF coupling



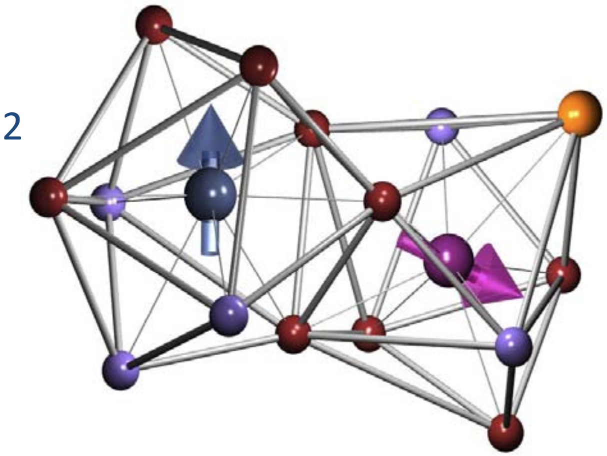
$$\mathcal{H}_{exch} = -J_{ex}J_{z1}J_{z2}$$

Magnetic asymmetry

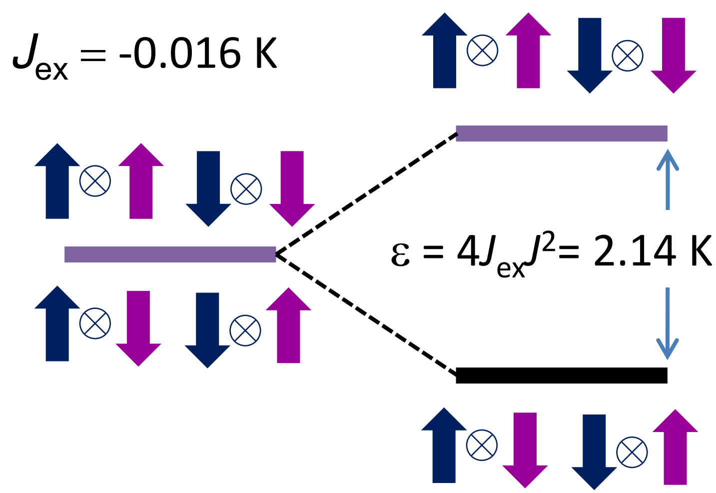




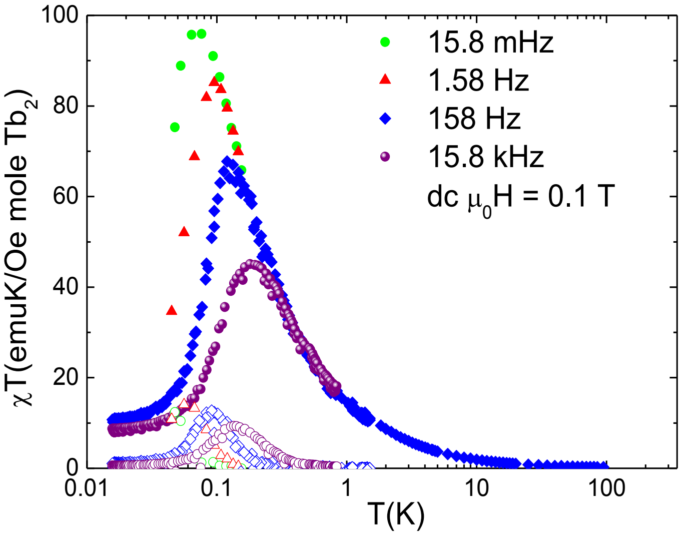
$[Tb]_2$

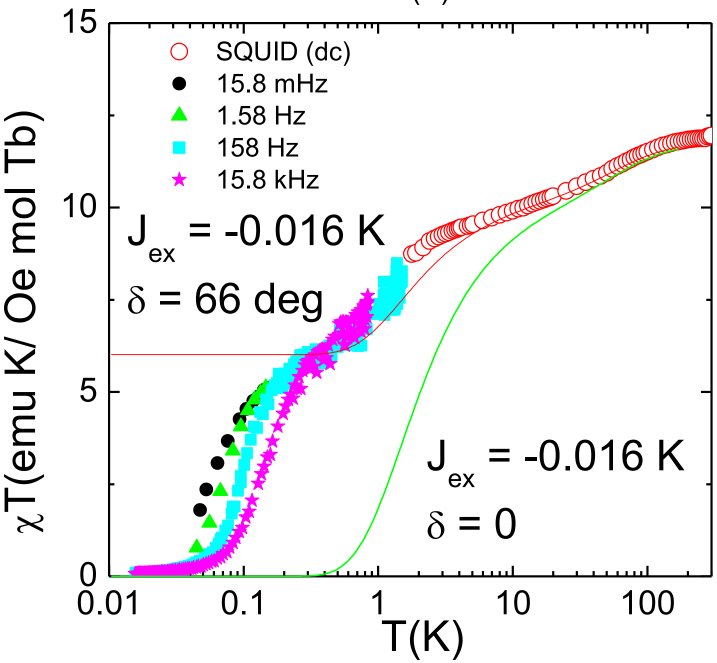


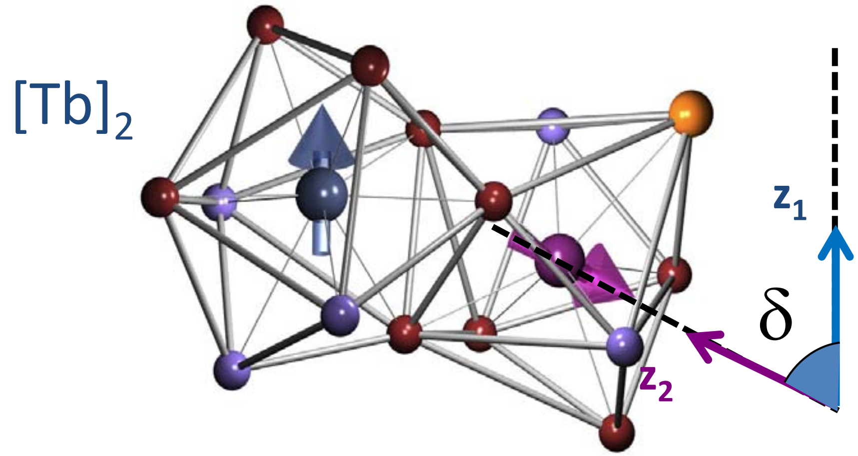
AF coupling



Magnetic asymmetry

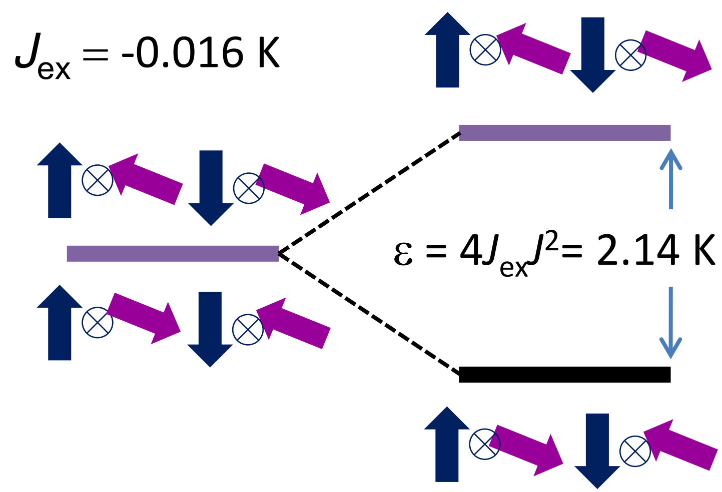




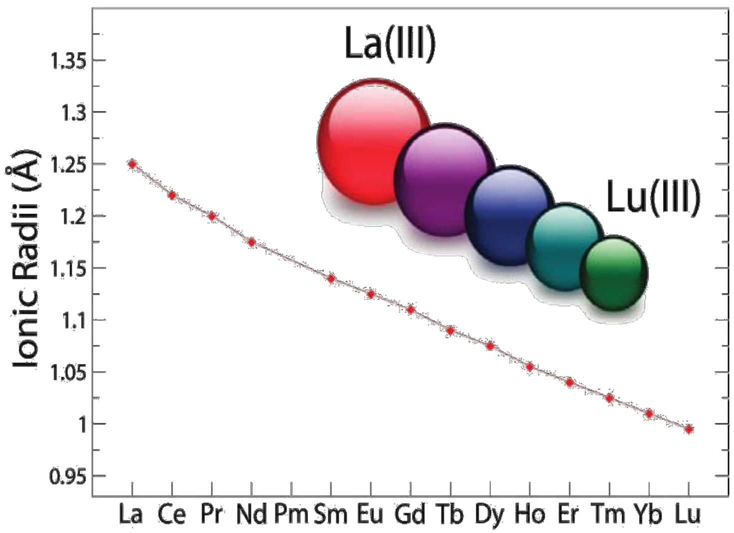


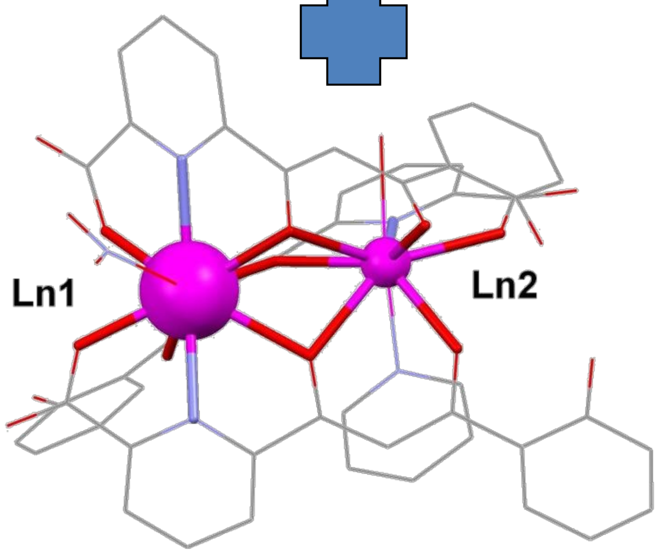
δ = 66 degrees

Noncollinear anisotropy axes

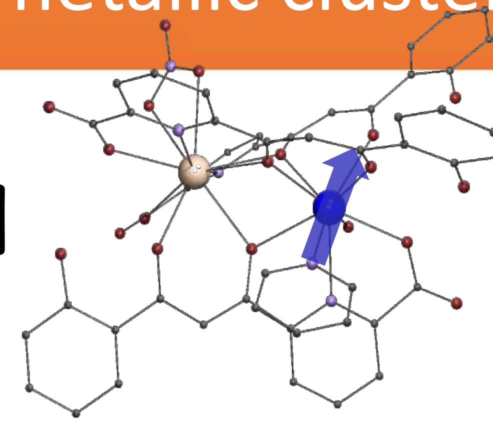


Heterometallic clusters



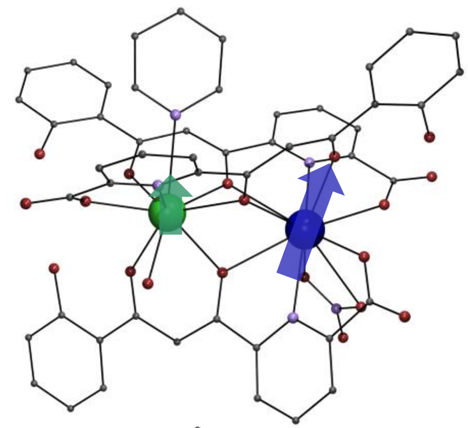


[LaEr]

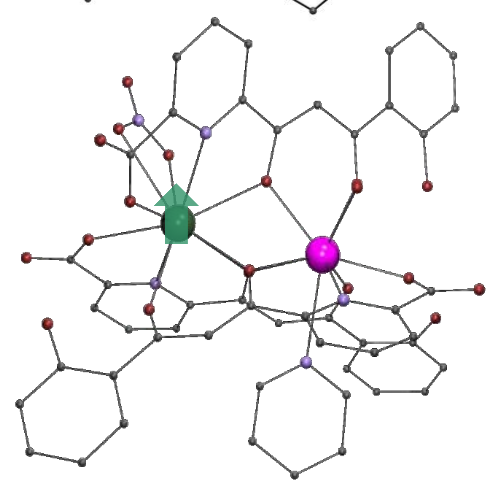


Er^{3+}
$J = 15/2$,
$g_J = 6/5$

[CeEr]

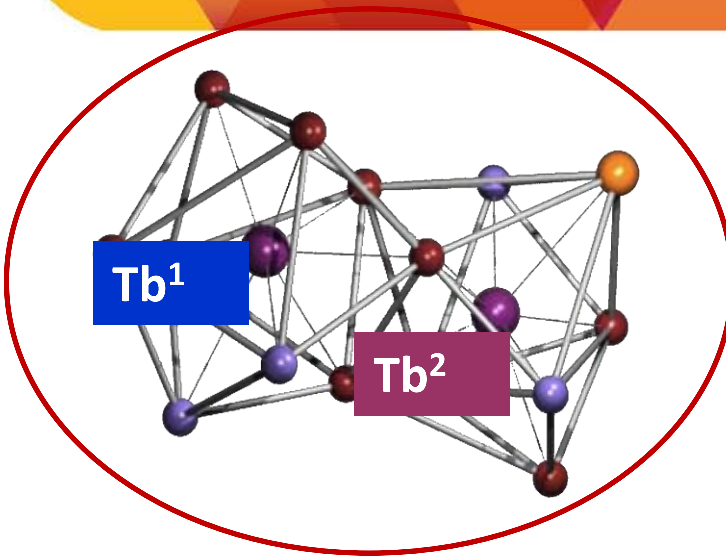


Ce^{3+}
$J = 5/2$,
$g_J = 6/7$

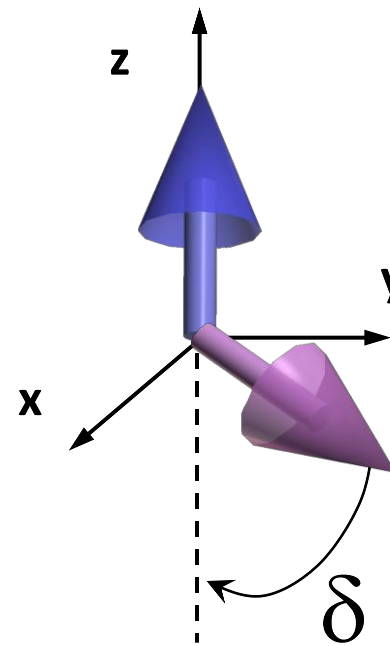


[CeY]

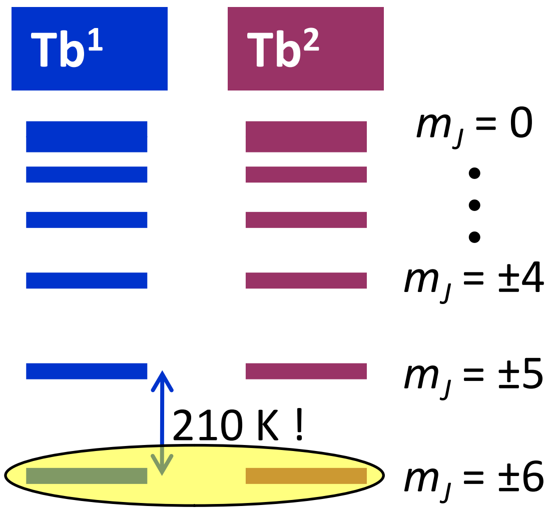
All ingredients are met!



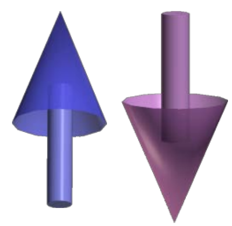
Non-collinear easy axes or different ions



99.99% lie in the ground state below 20 K



Antiferromagnetic coupling below 3 K



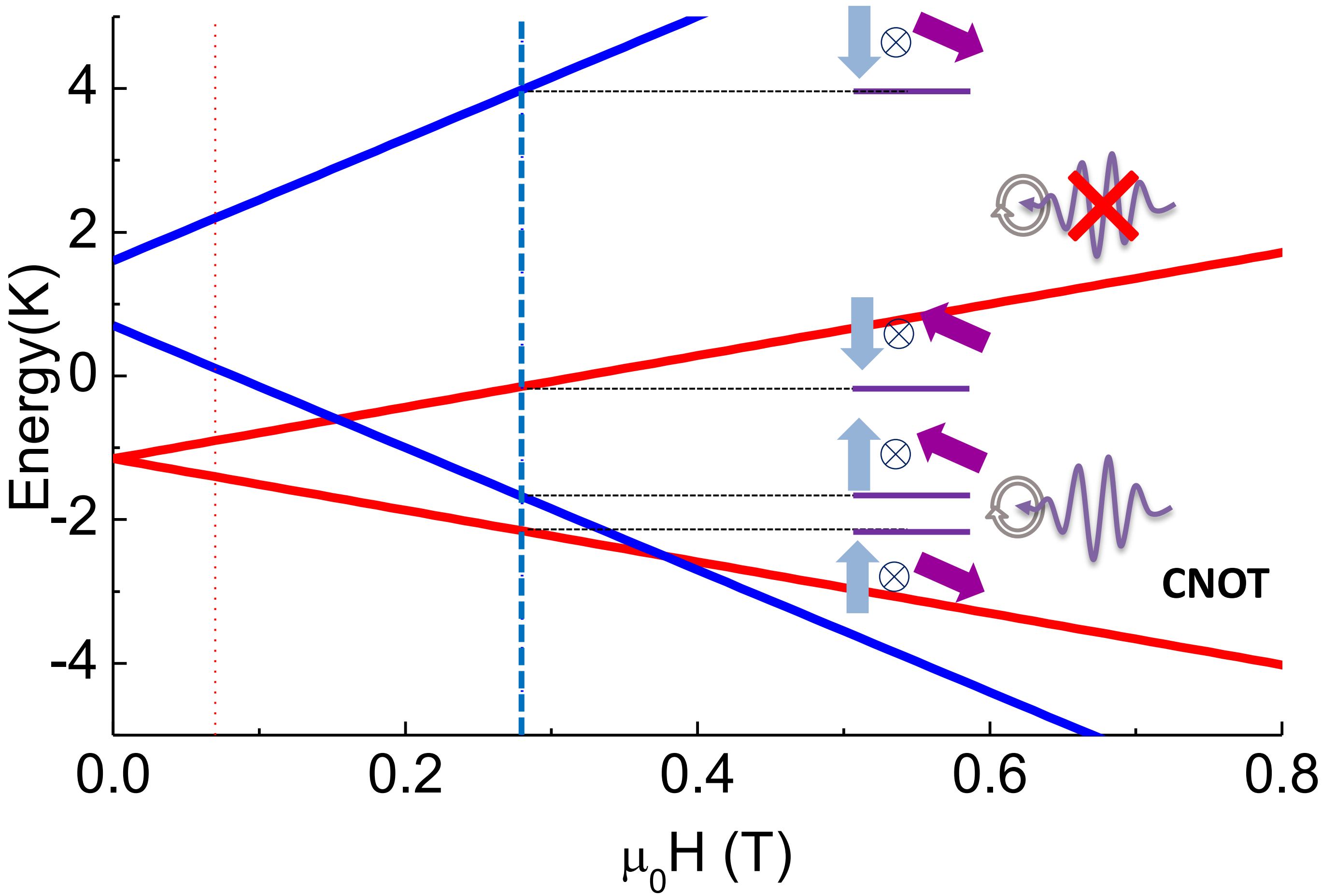
two qubits √

interaction √

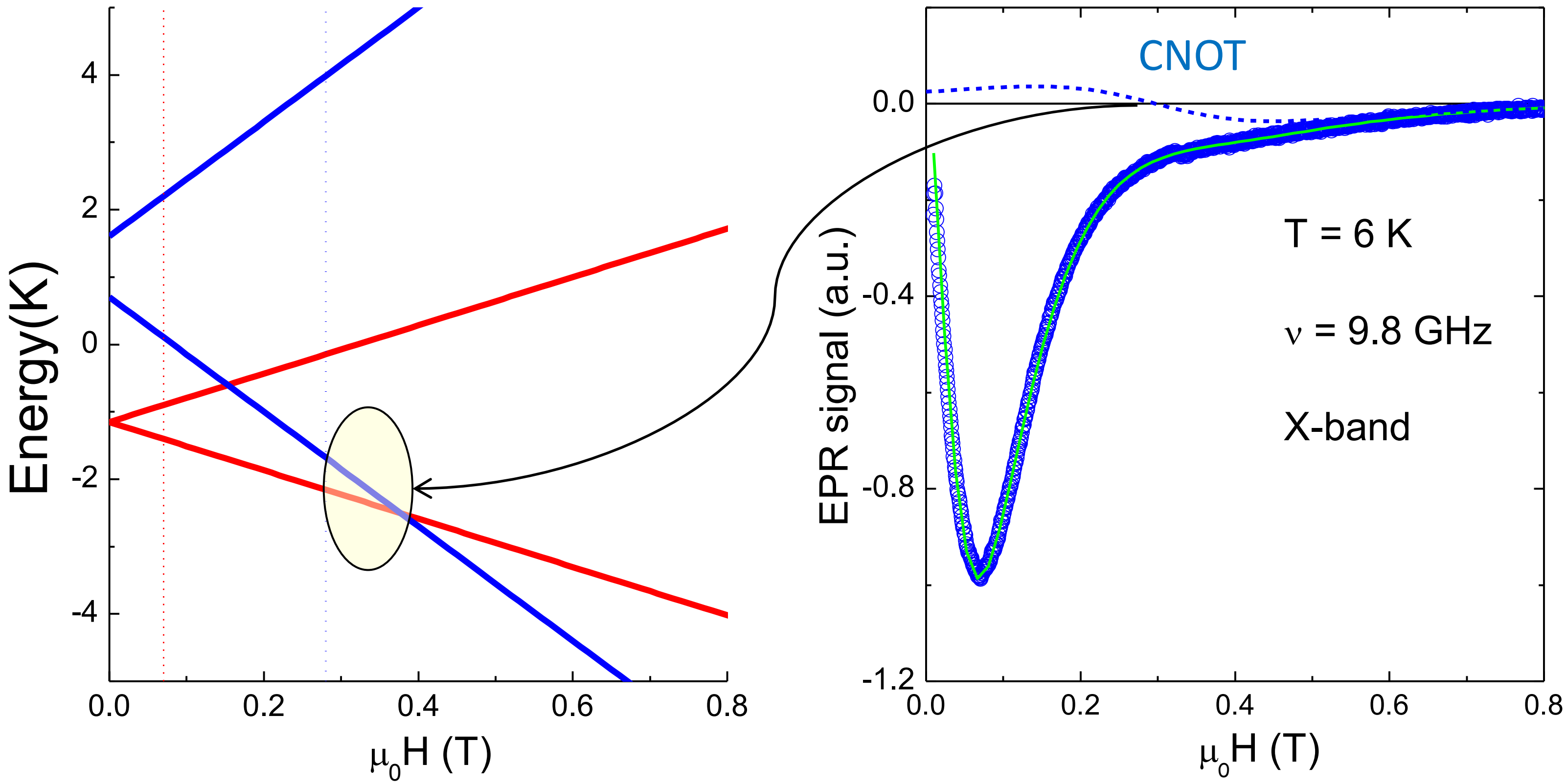
δ = 66 deg √

[Tb]$_2$ as a CNOT gate

$$\mathcal{H}_{m=\pm 6} = -2J_{ex}J_{z1}J_{z2} - g_J\mu_B\left(H_{z1}J_{z1} + H_{z2}J_{z2}\right) + A_{hf}\left(J_{z1}I_{z1} + J_{z2}I_{z2}\right)$$

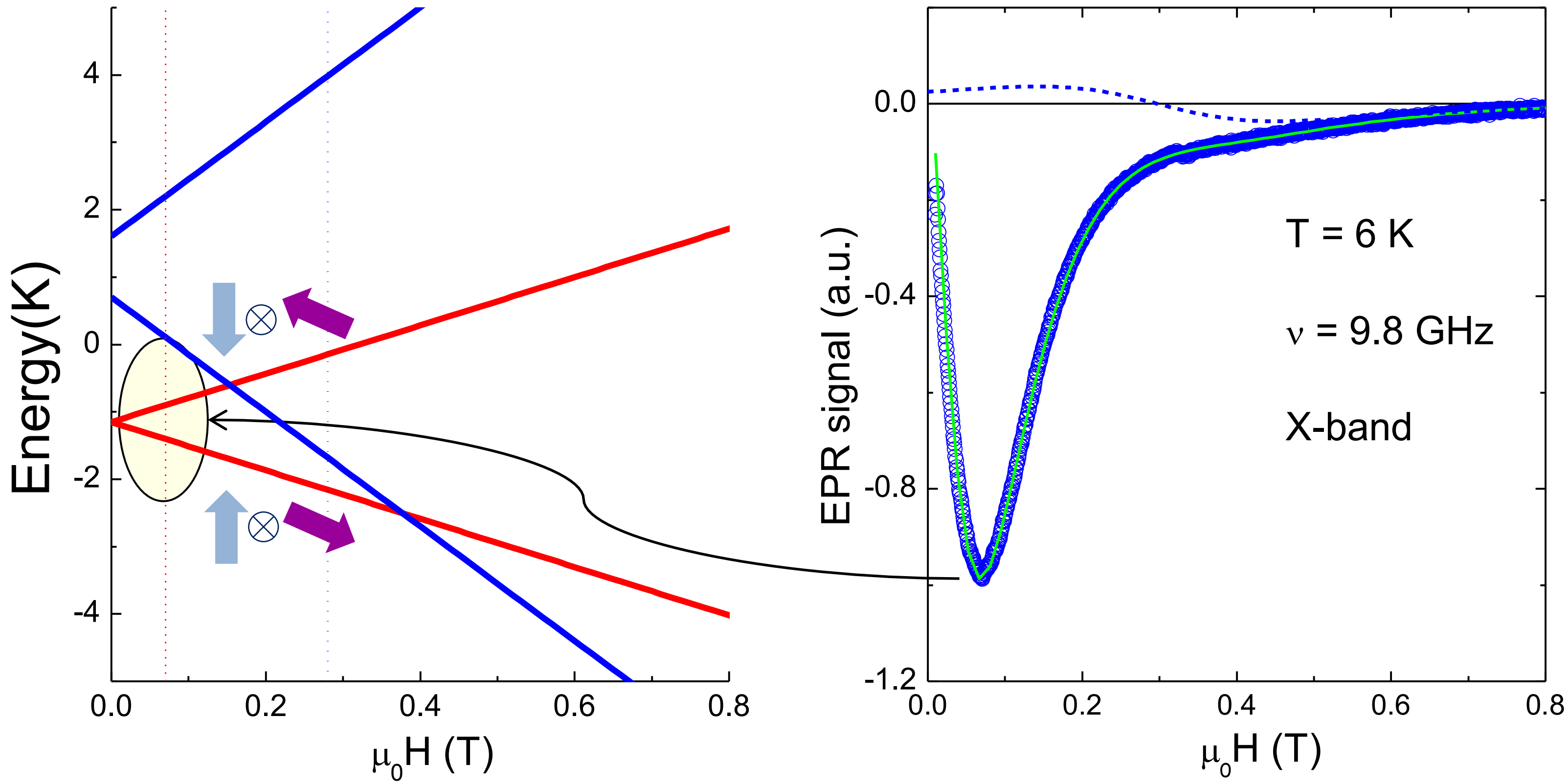


Implementation by EPR



CNOT transitions are not forbidden

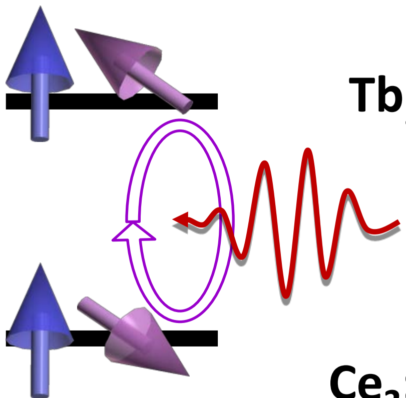
Implementation by EPR



SWAP gate operations are also possible!

F. Luis *et al*, Phys. Rev. Lett. **107**, 117203 (2011).

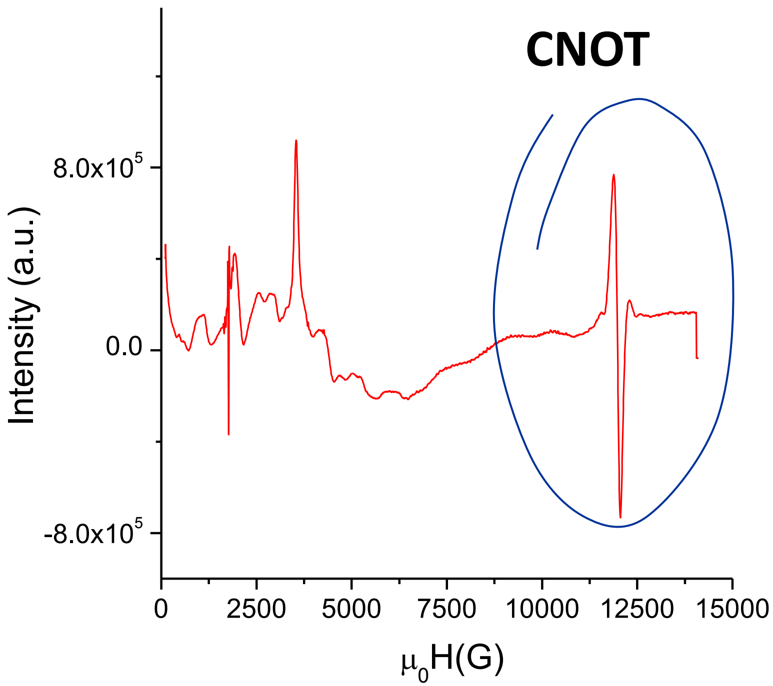
Quantum coherence? (X-band pulsed EPR)

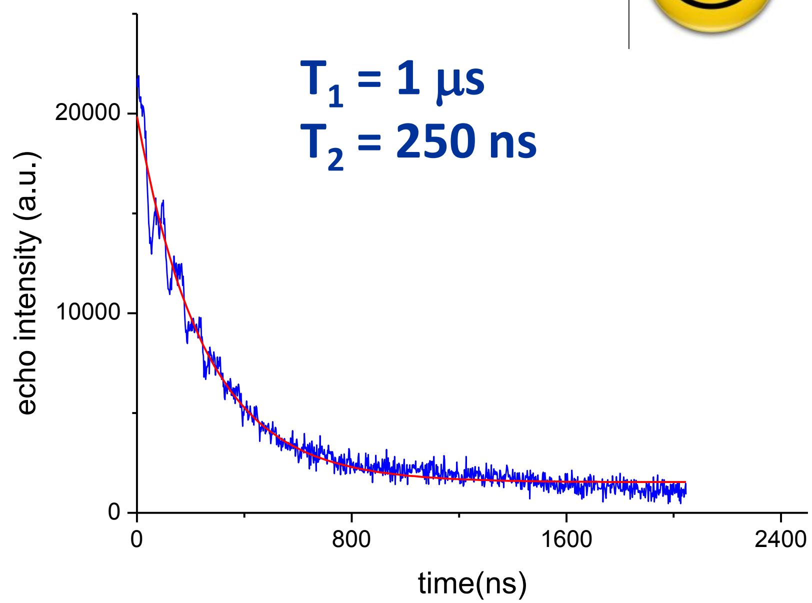


Tb_2: *m* = -6 → *m* = +6 ECHO?
NOT OBSERVED

Ce_2: *m* = -1/2 → *m* = +1/2
OBSERVED!!

CNOT

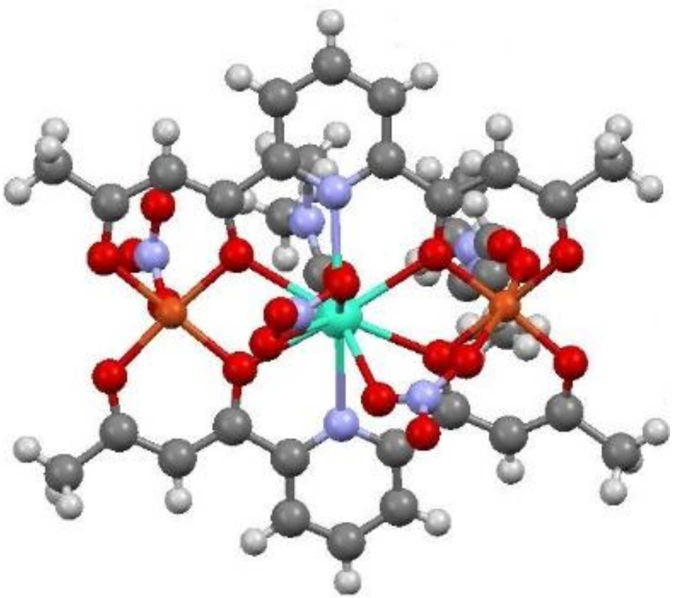


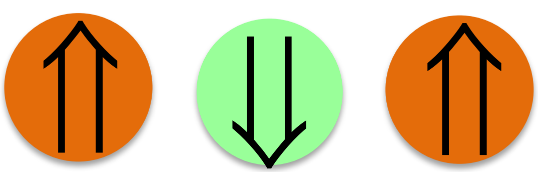


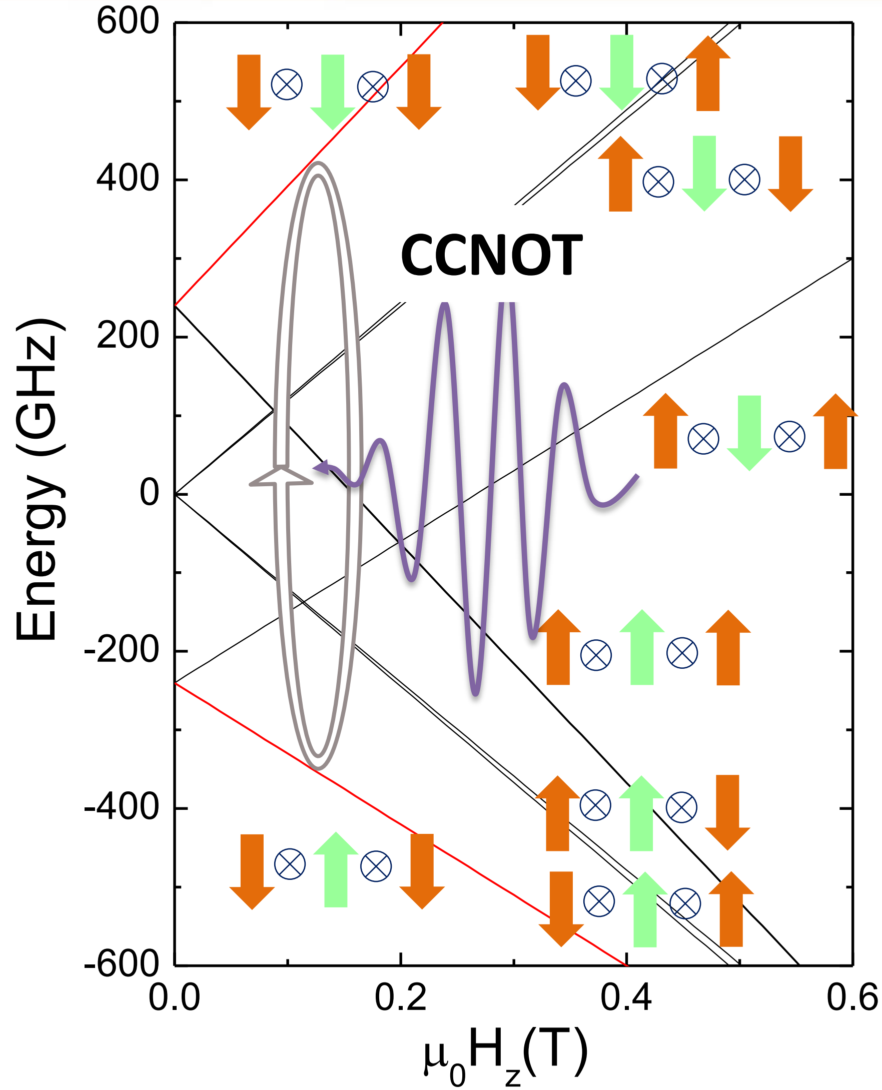
Three-qubit gates (in progress)

New molecular prototypes

CuTbCu Toffoli





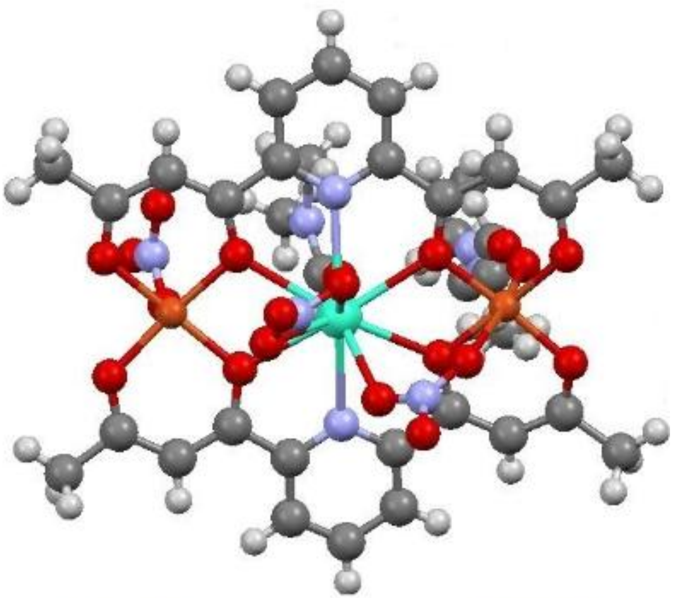


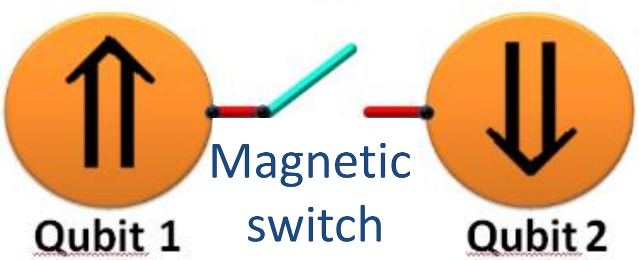
$$\mathcal{H} = -J_{ex,1}J_{z1}J_{z2} + J_{ex,2}J_{z2}J_{z3} - \mu_B H\left(g_1 J_{z1} + g_2 J_{z2} + g_3 J_{z3}\right)$$

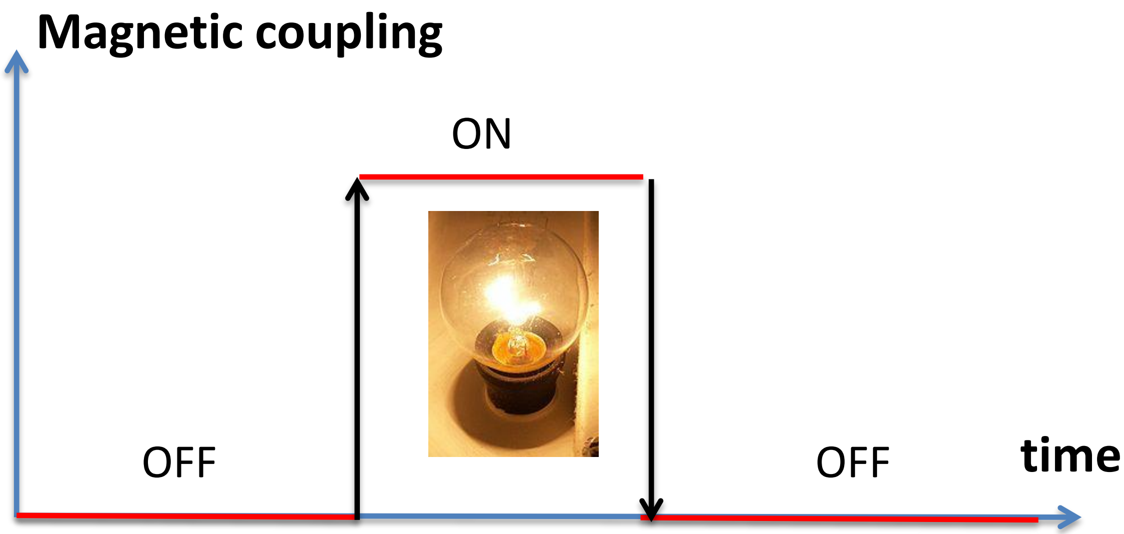
Three-qubit gates (in progress)

New molecular prototypes

CuEuCu optically controlled √SWAP





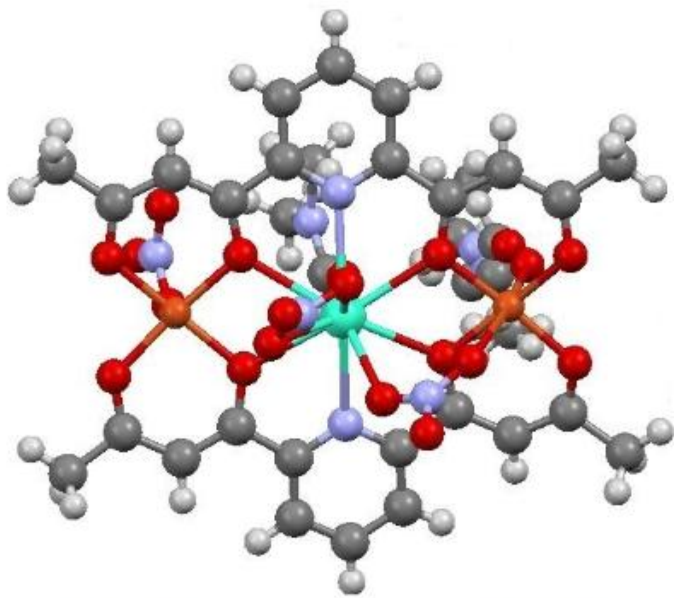


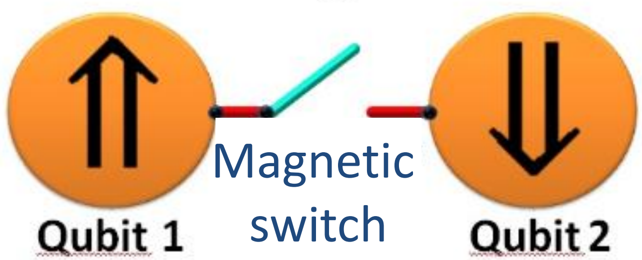
$$\left(|1\rangle_{qubit1} \otimes |0\rangle_{qubit2}\right)_{inicial} \xrightarrow{\sqrt{SWAP}} \left[\frac{\left(|1\rangle \otimes |0\rangle + i\,|0\rangle \otimes |1\rangle\right)}{(1+i)}\right]_{final}$$

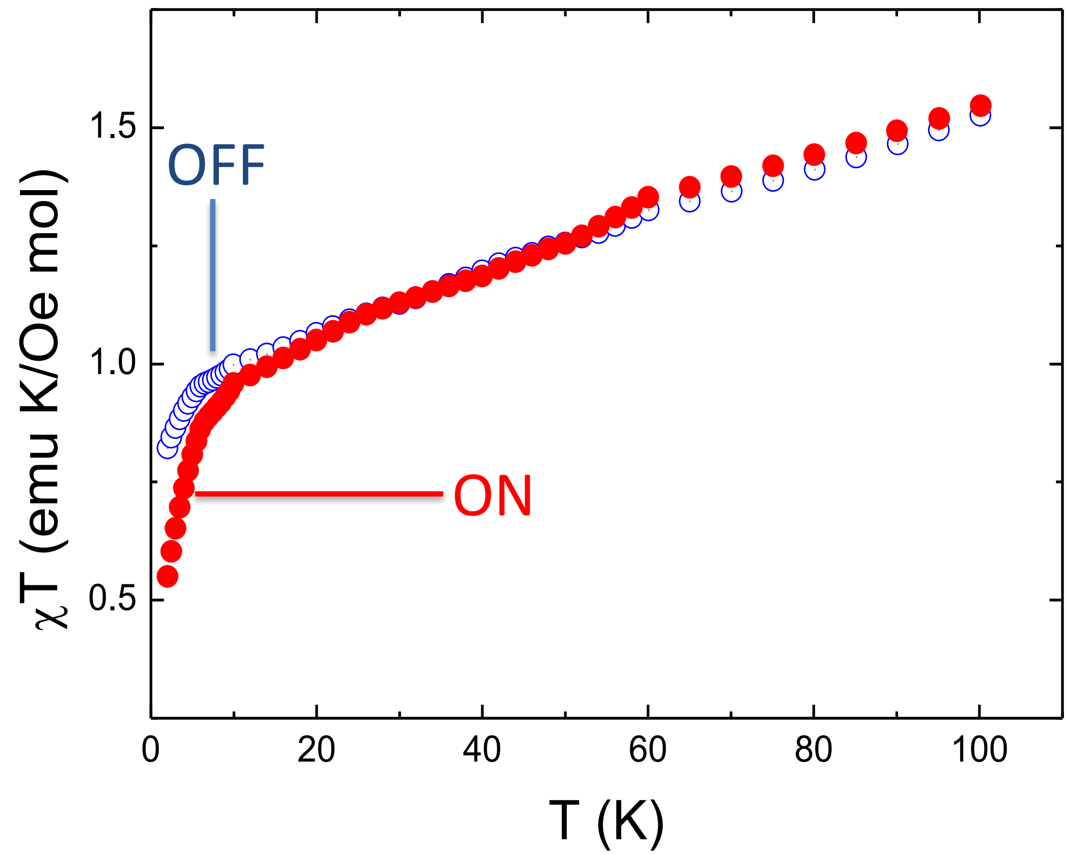
Three-qubit gates (in progress)

New molecular prototypes

CuEuCu optically controlled √SWAP

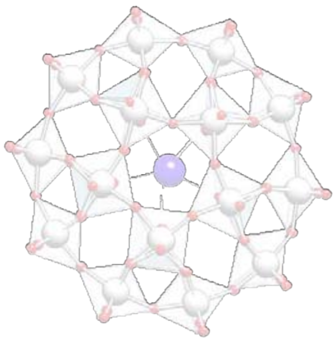


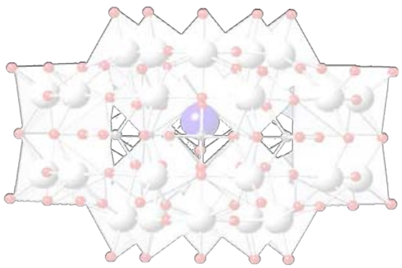




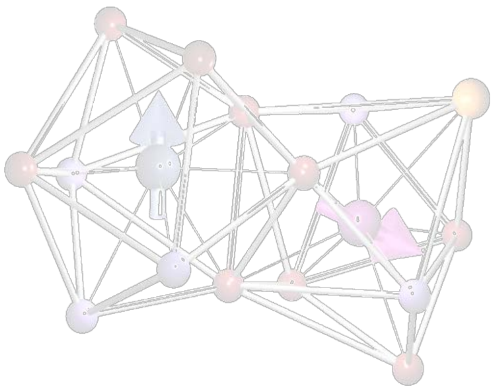
Outline

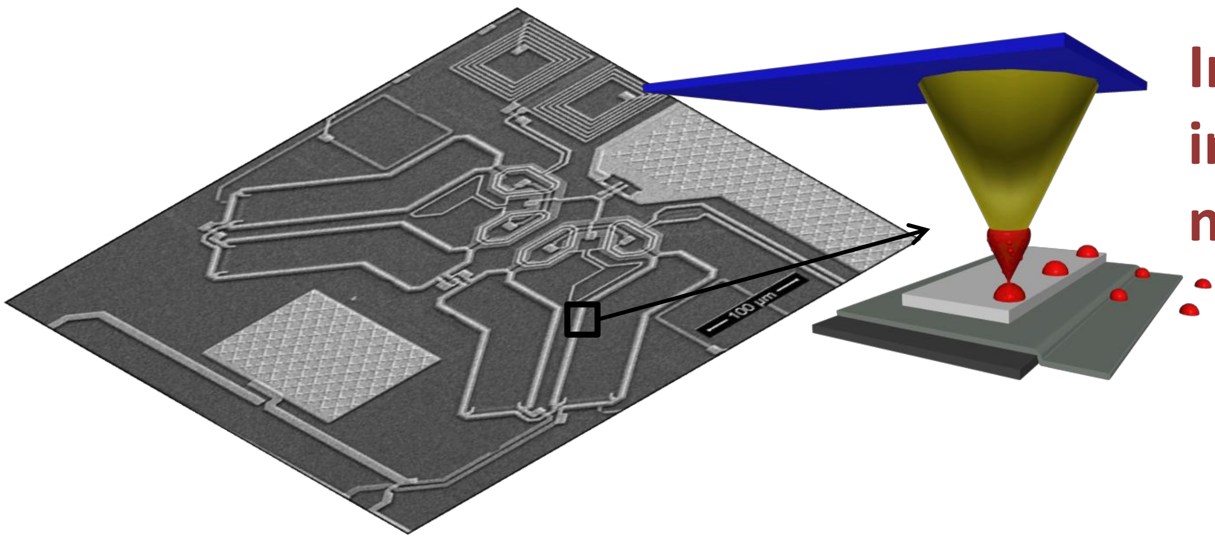
Molecular qubits





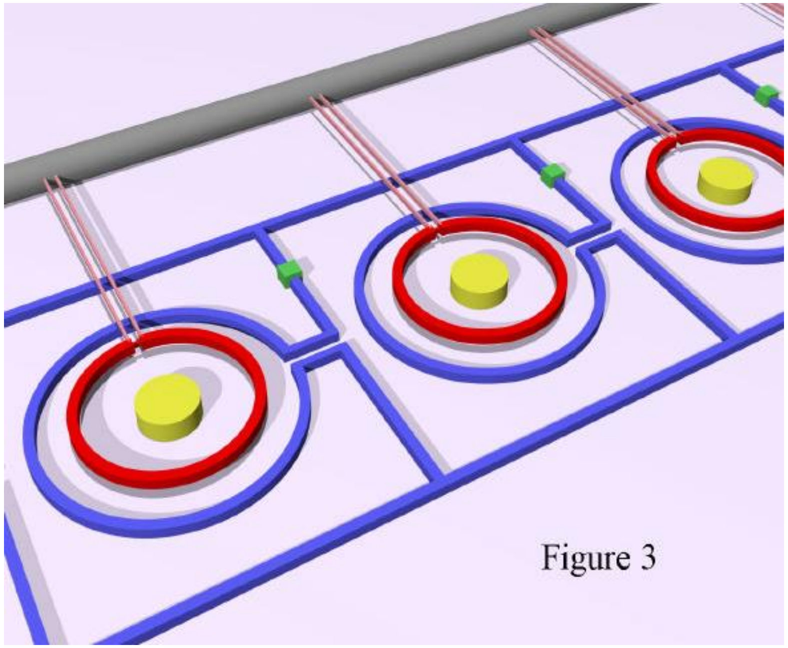
Molecular quantum gates



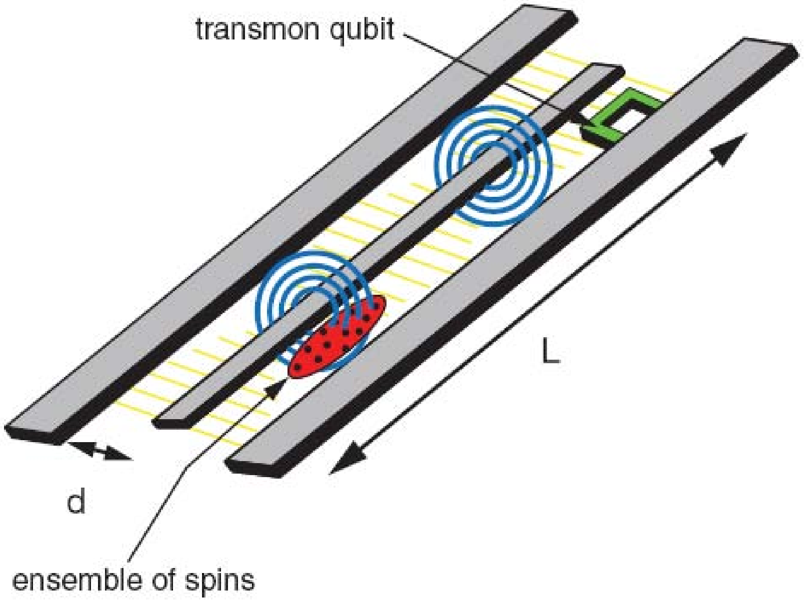


Integration of SMM into superconducting microdevices

Hybrid quantum computation architectures



Magnetic qubits as hardware for quantum computers. J. Tejada, E. M. Chudnovsky, E. del Barco, J. M. Hernandez and T. P. Spiller, Nanotechnology **12** (2001) 181–186



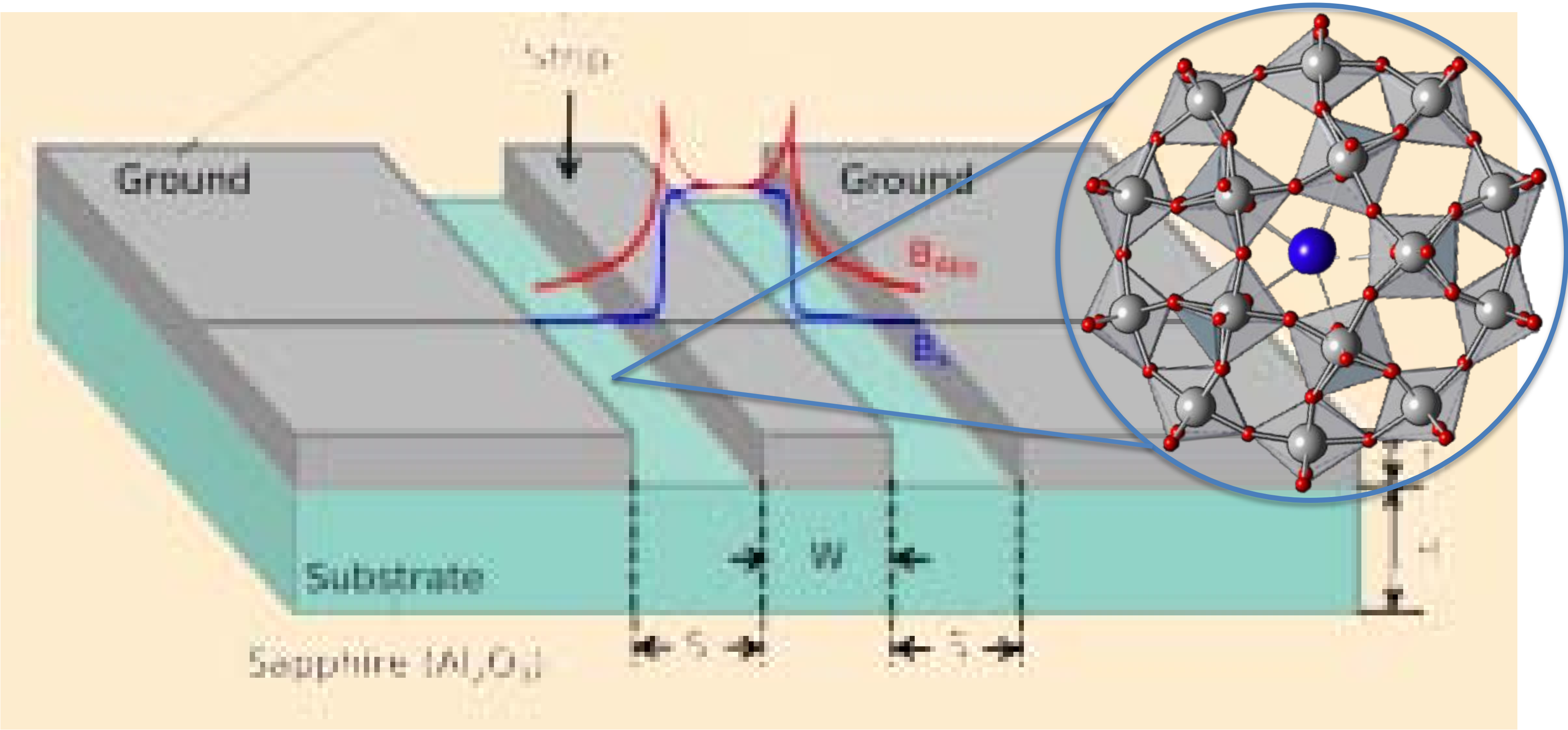
Cavity QED Based on Collective Magnetic Dipole Coupling:
Spin Ensembles as Hybrid Two-Level Systems.
Atac Imamoglu, PRL **102**, 083602 (2009)

Molecule-based qubits and qugates

+

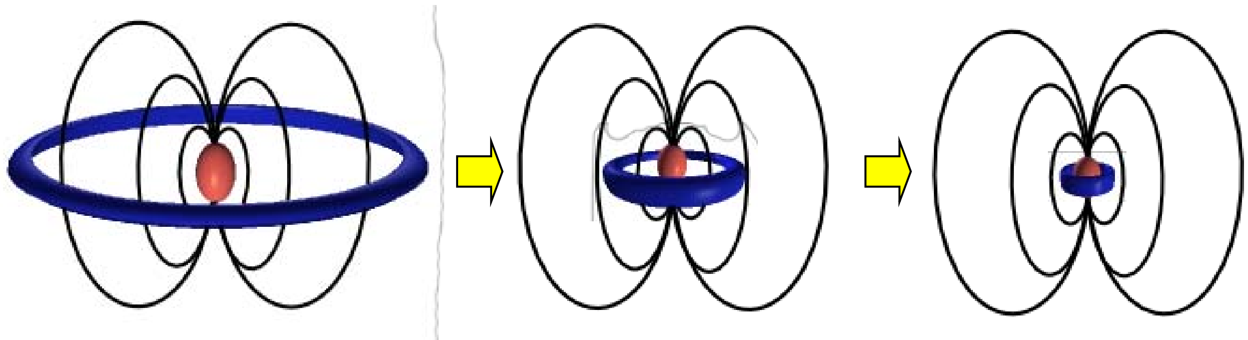
Superconducting μcircuits

The challenge: magnetic coupling

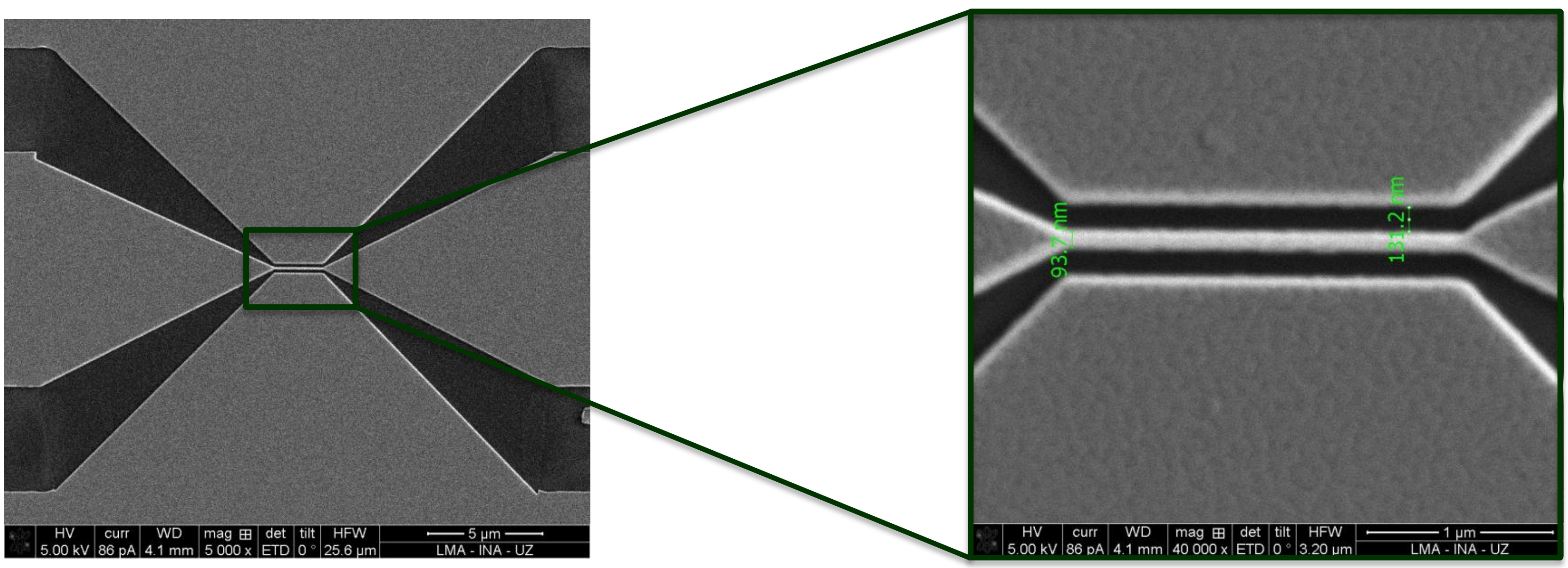


$$g = \frac{2 g_J \mu_B J h_{rf}}{h} \approx 100\,\mathrm{Hz} << \mathrm{T}_2^{-1}$$

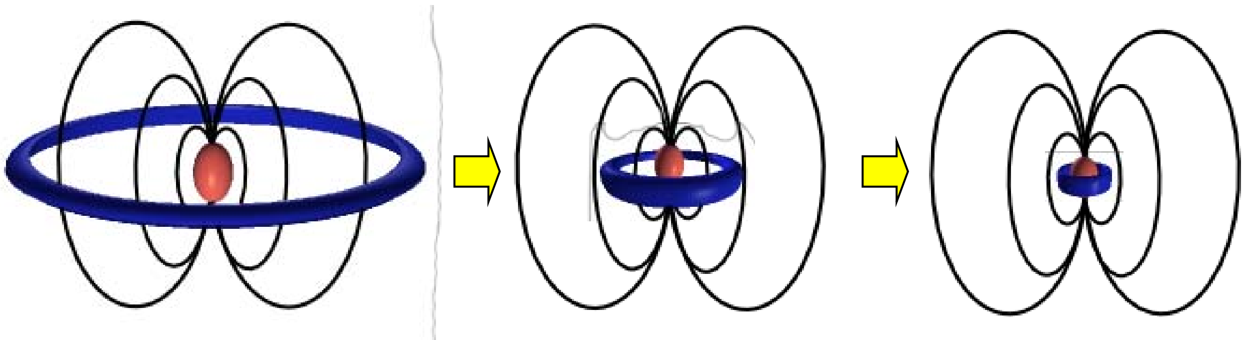
1. Scaling down the dimensions of the device



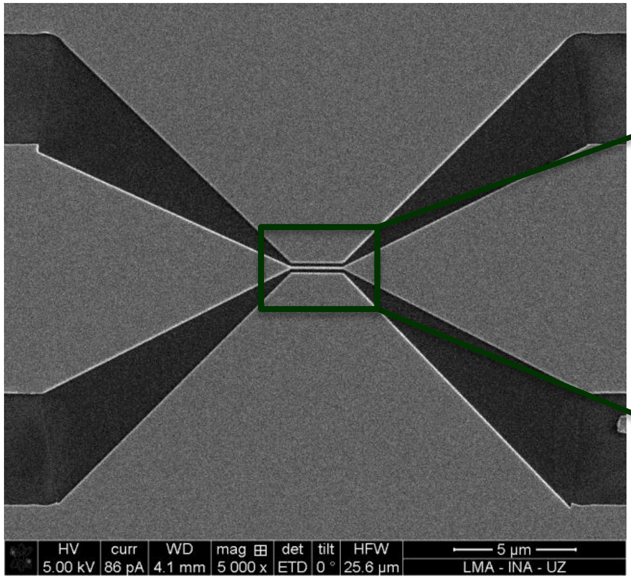
Nanoscopic coplanar transmission lines and resonators

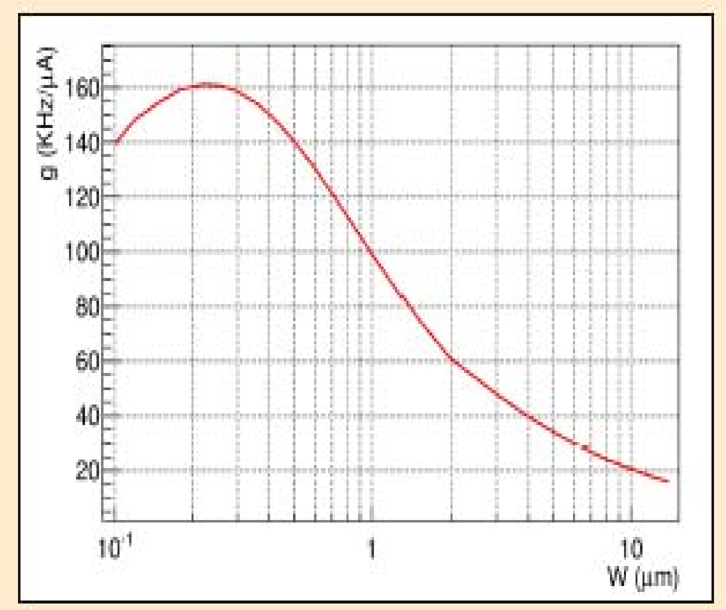


1. Scaling down the dimensions of the device

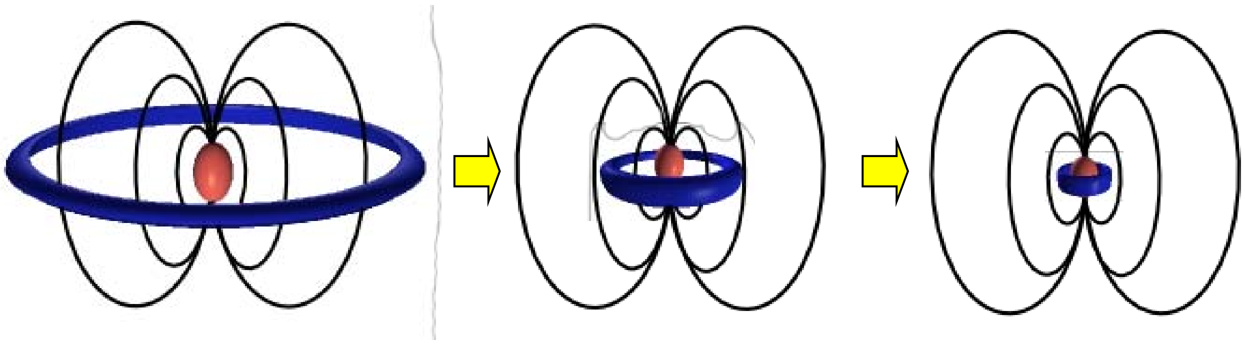


Nanoscopic coplanar transmission lines and resonators

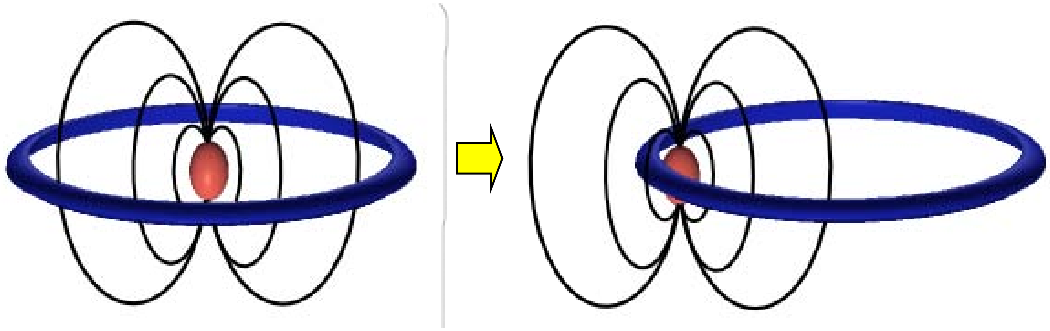




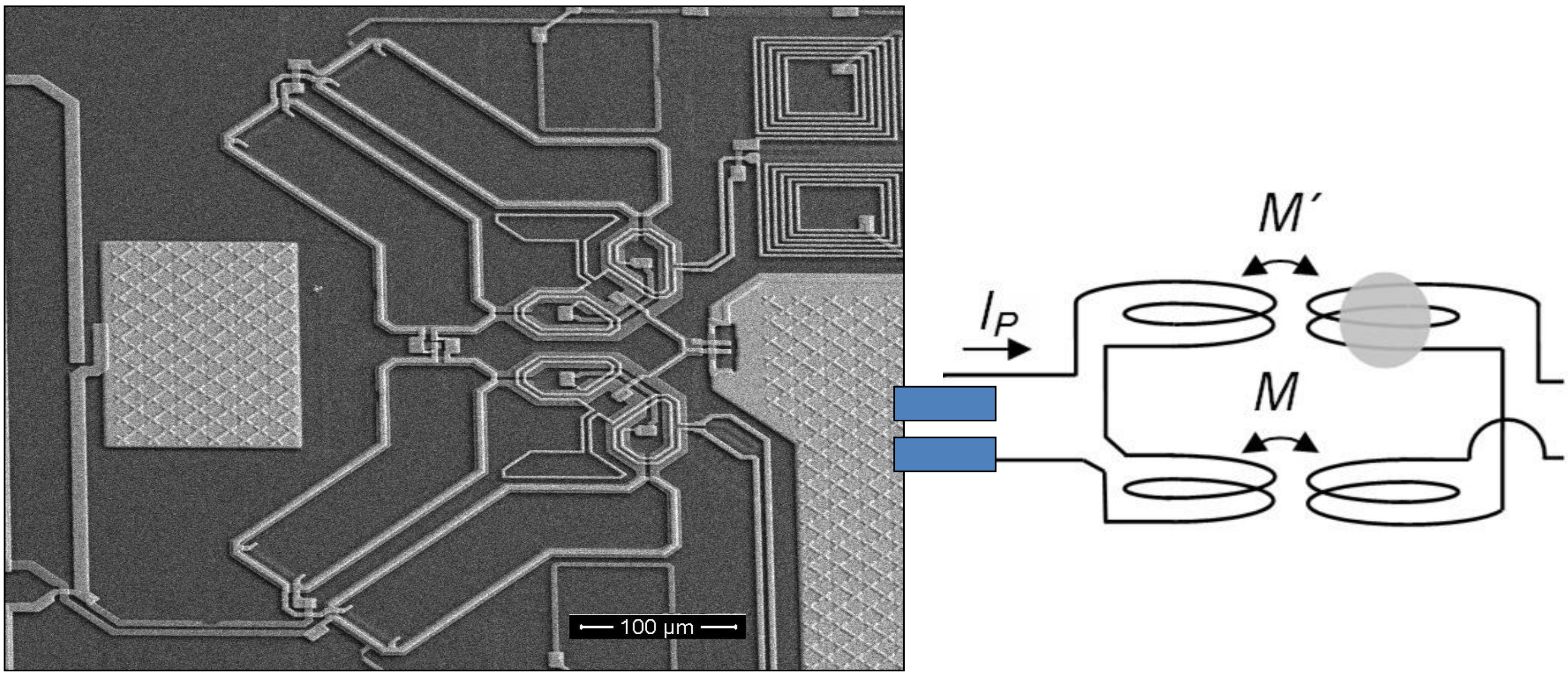
1. Scaling down the dimensions of the device



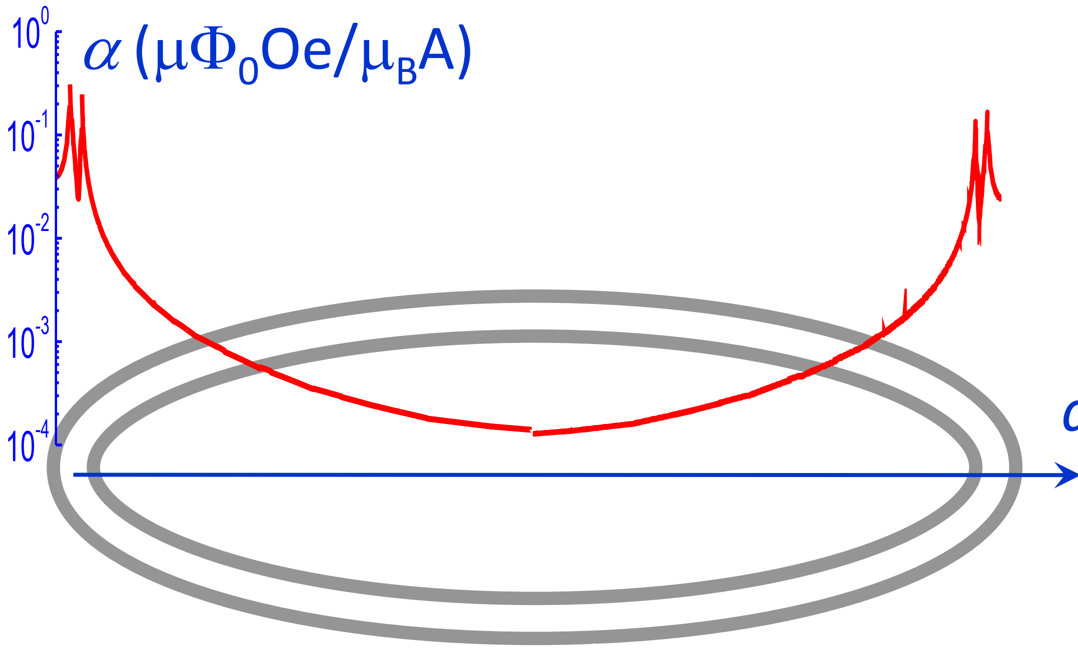
2. Playing with the sample position !!!



The device: microSQUID ac susceptometer

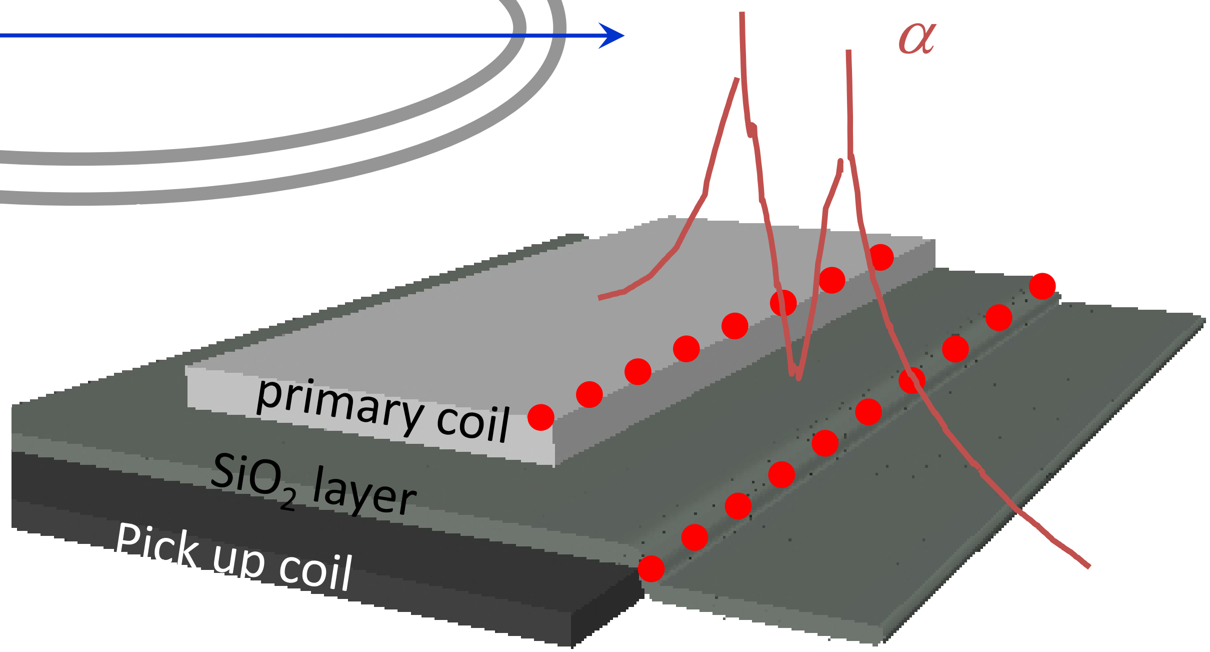


MJ Martínez-Pérez, J. Sesé, F. Luis, D. Drung and T. Schurig
Rev. Sci. Instrum. **81**, 016108 (2010)

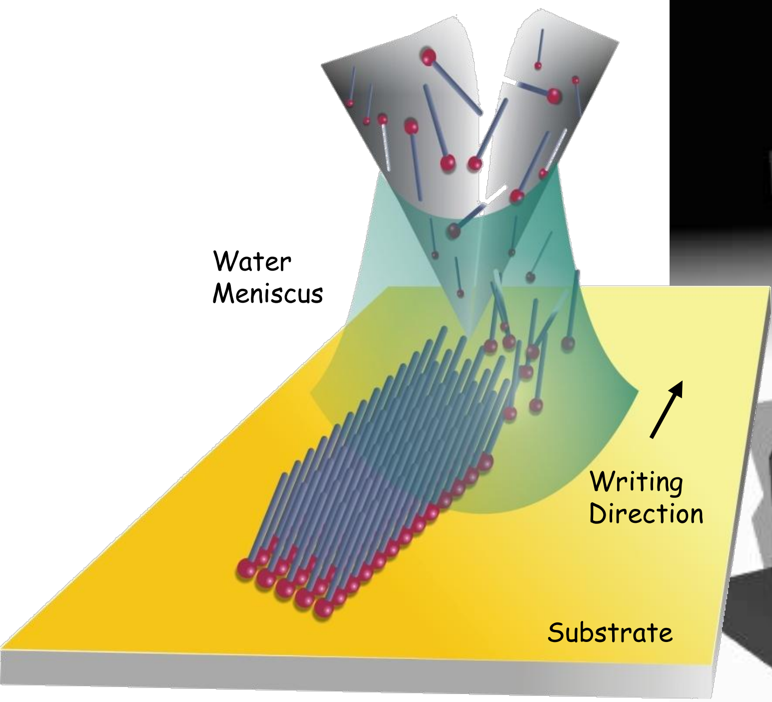


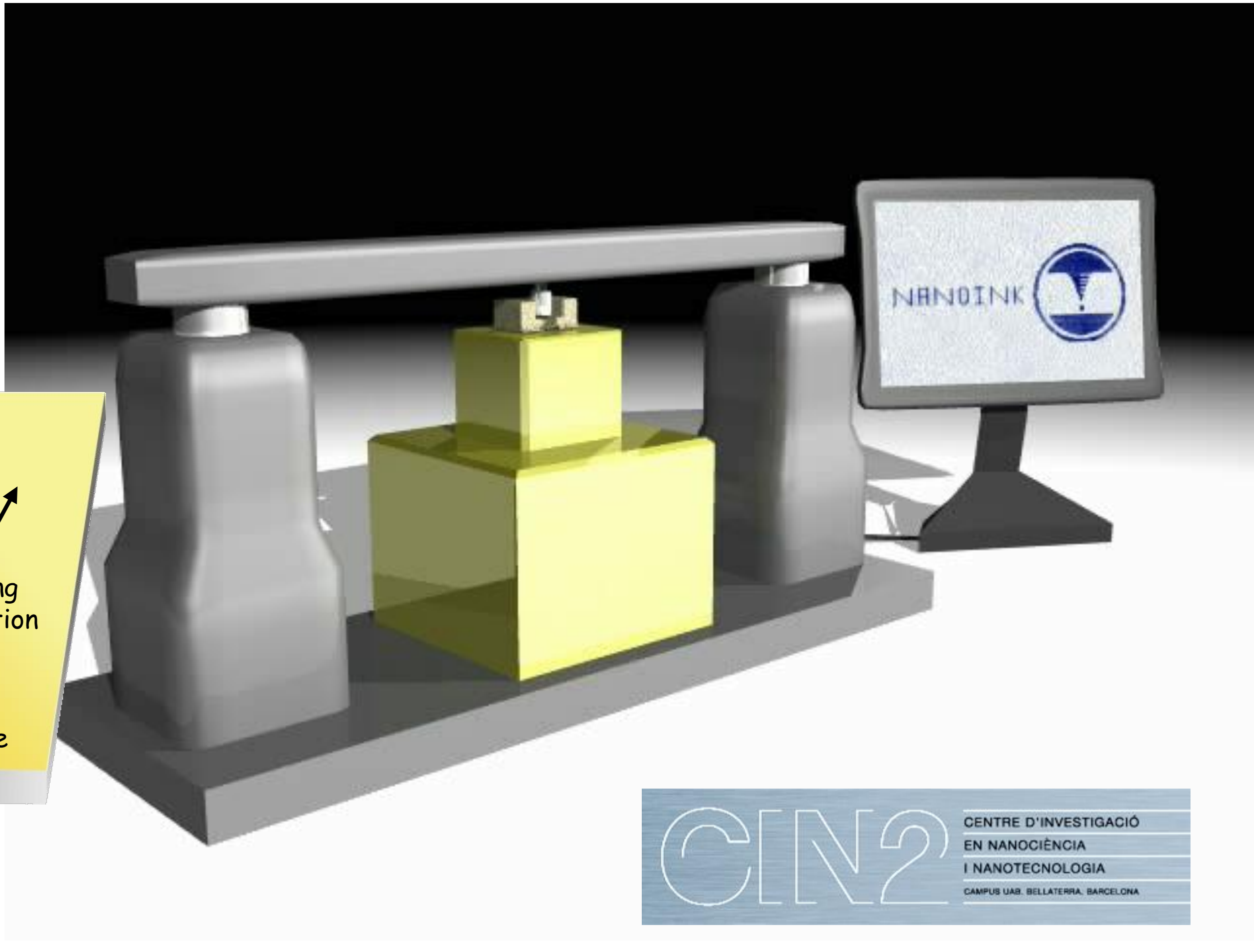
COMSOL MULTIPHYSICS®

$$\alpha = \frac{\phi_{coupled}}{n\mu_i}\frac{B_P}{i_P}$$

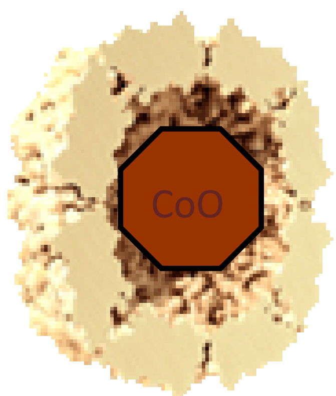


The tool: Dip pen nanolithography

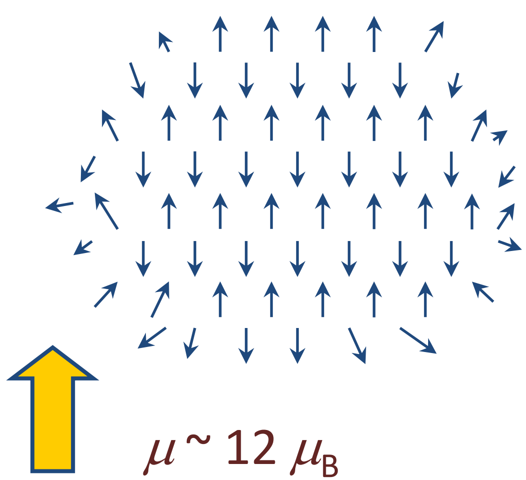




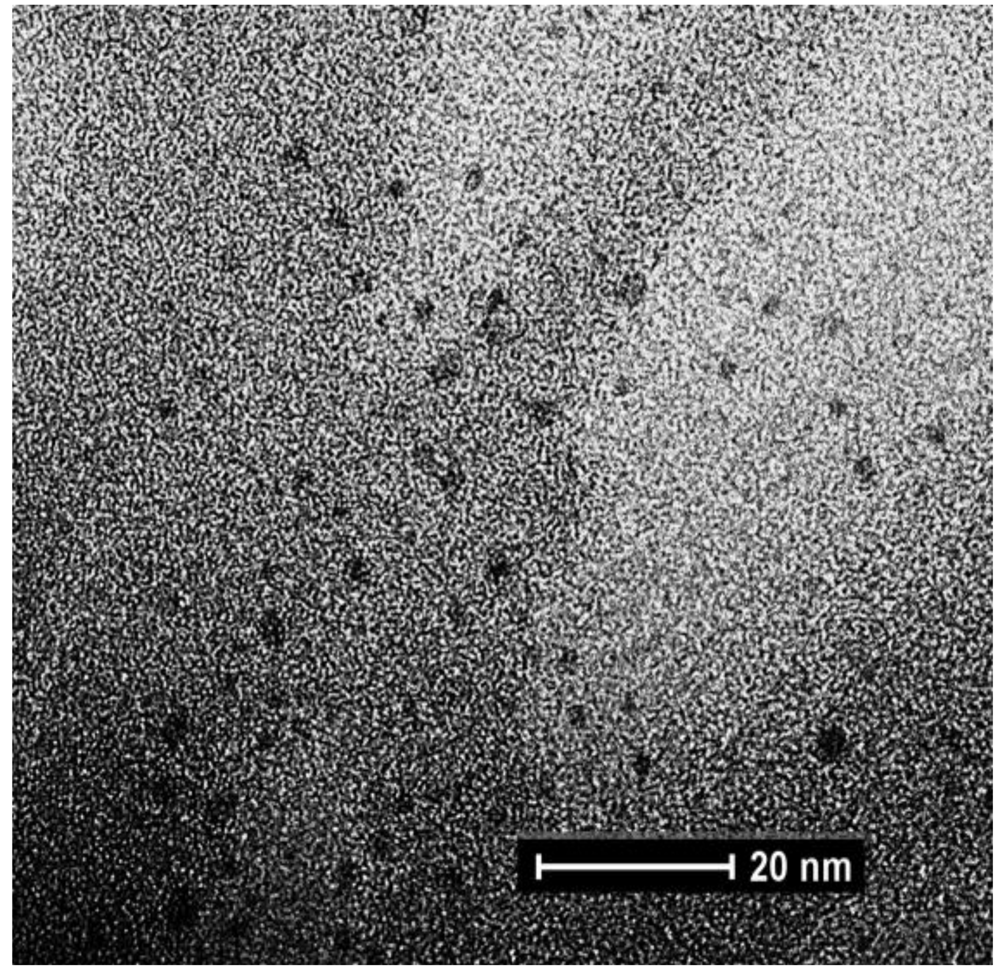
The sample: ferritin-based nanomagnets (CoO)

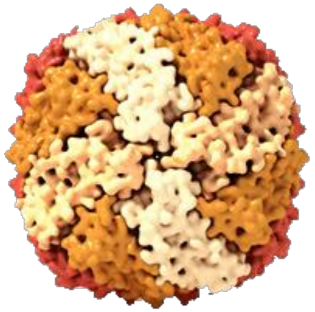


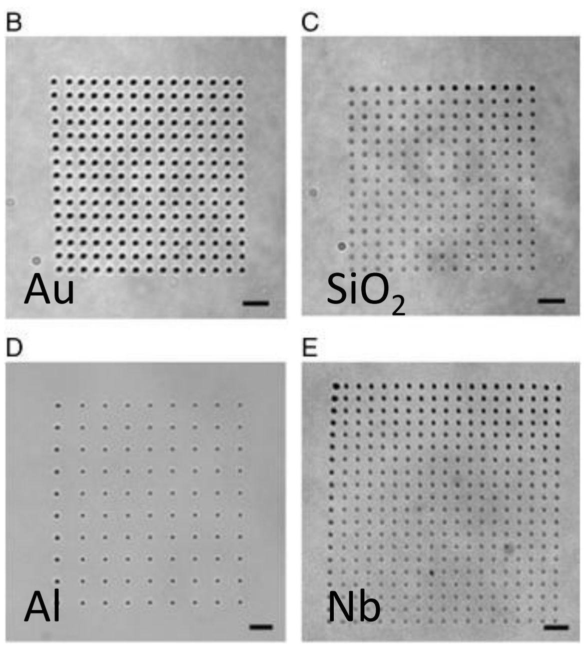
2 nm sized
Antiferromagnetic particle

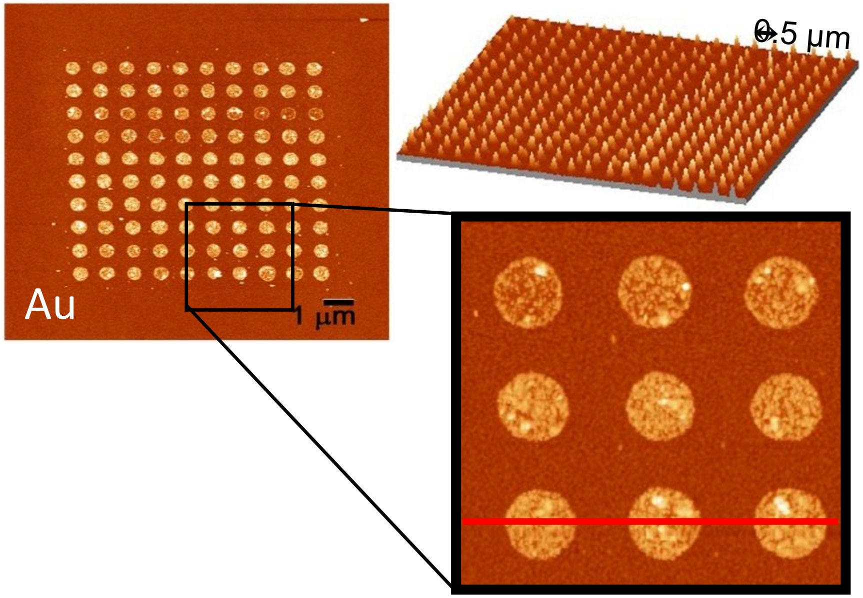


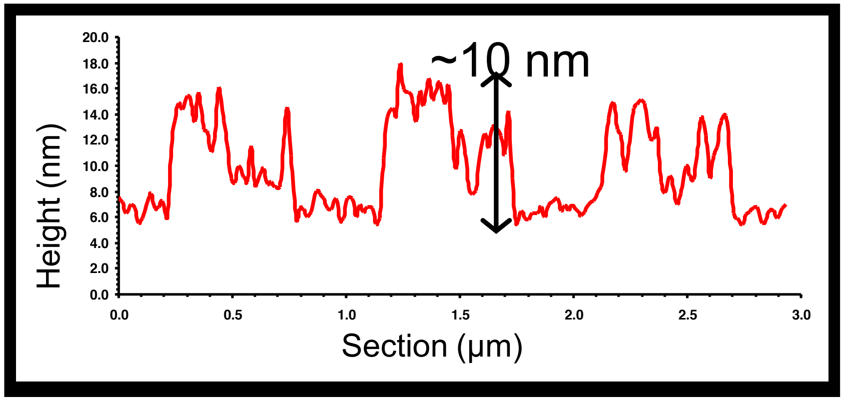
μ ~ 12 μ_B





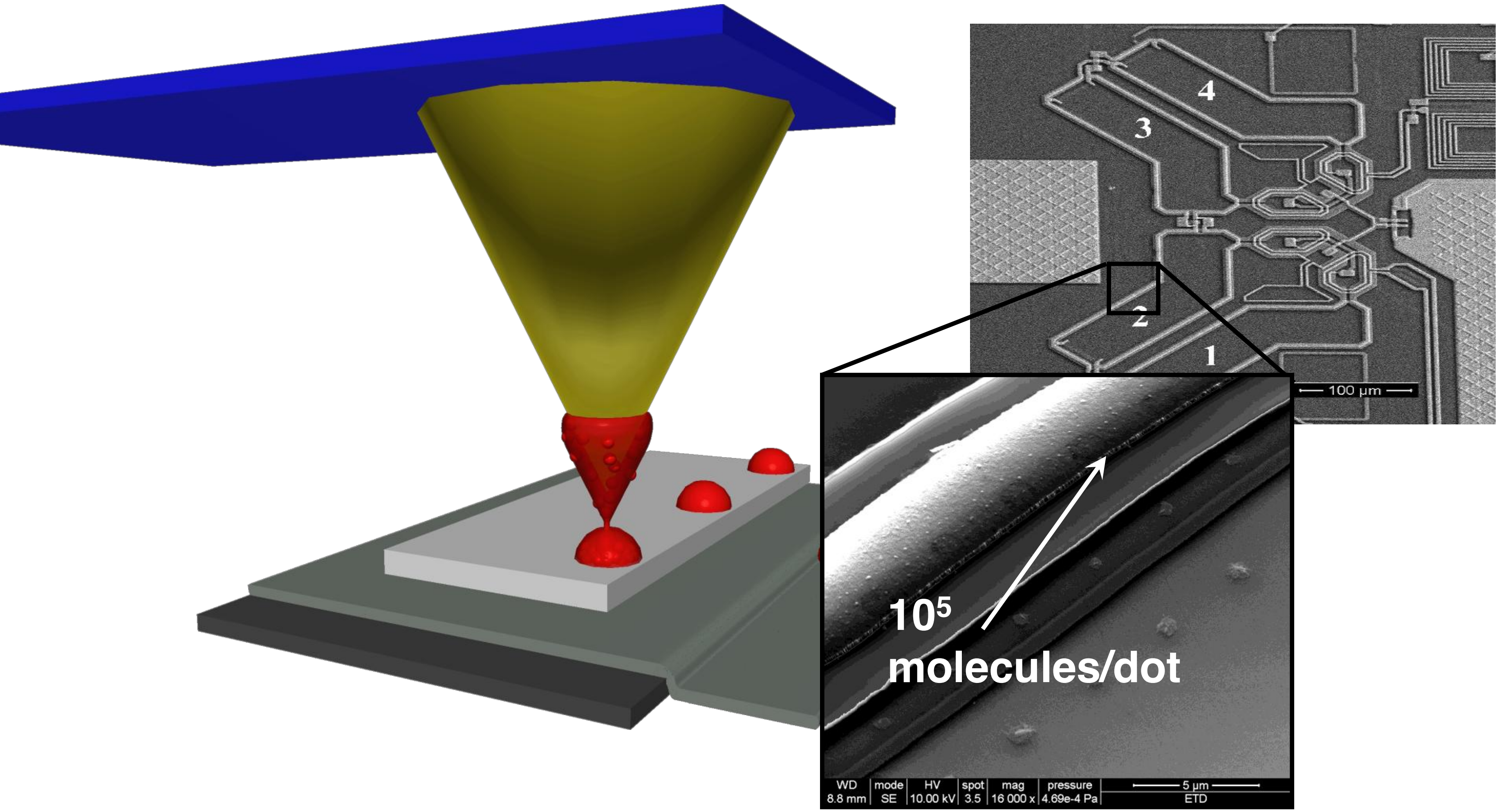

B
C
Au
SiO2
D
E
Al
Nb


0.5 µm
Au
1 µm

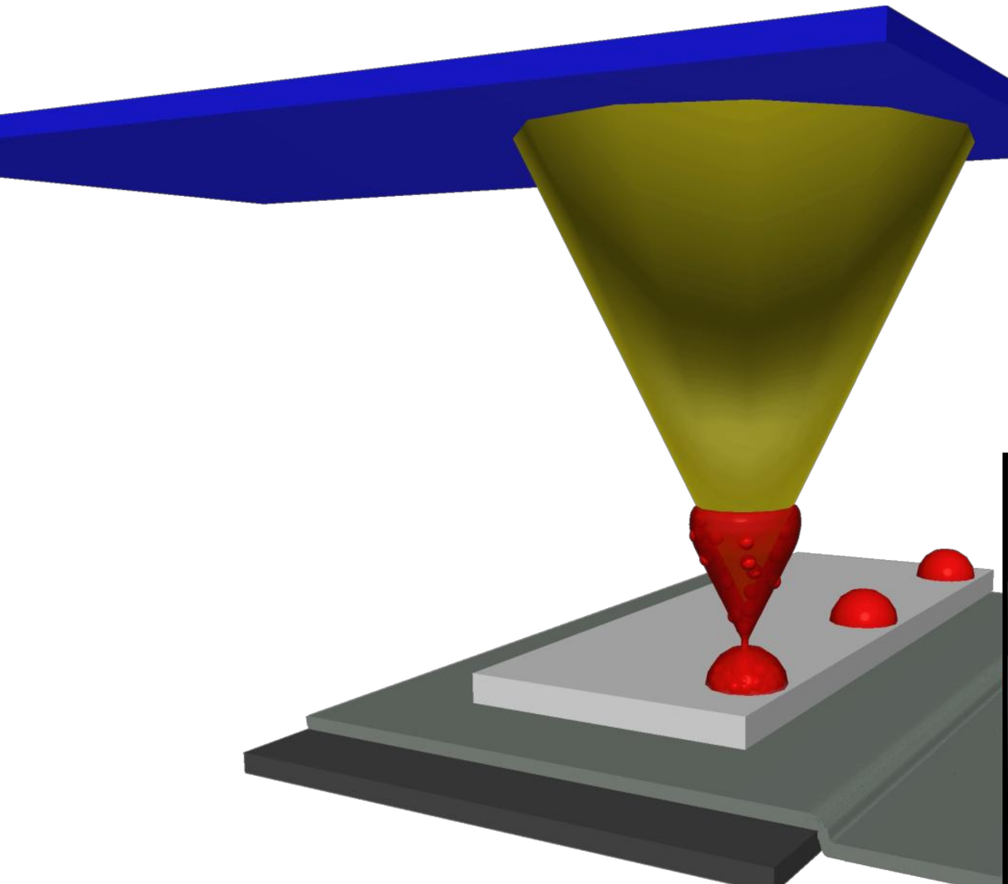

~10 nm
Height (nm)
Section (µm)

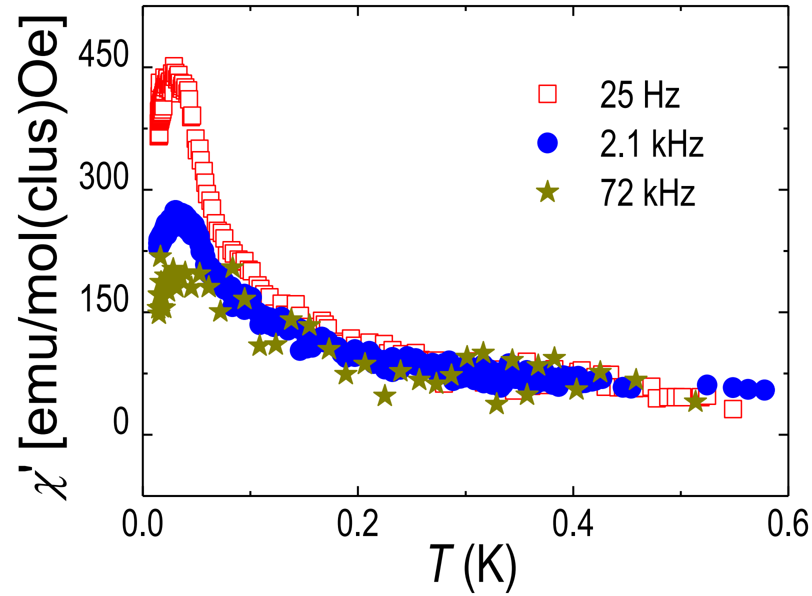
icma

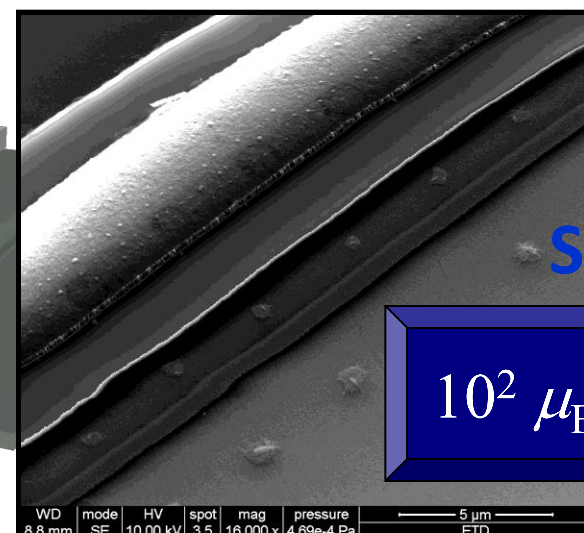
Direct deposition on the most sensitive areas



Detection of the linear response of a SMM monolayer





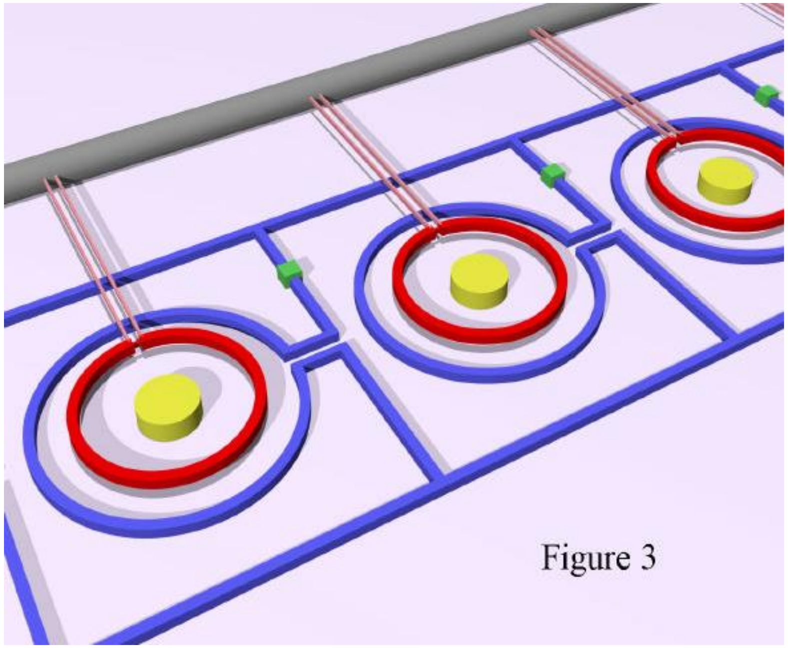


Sensitivity

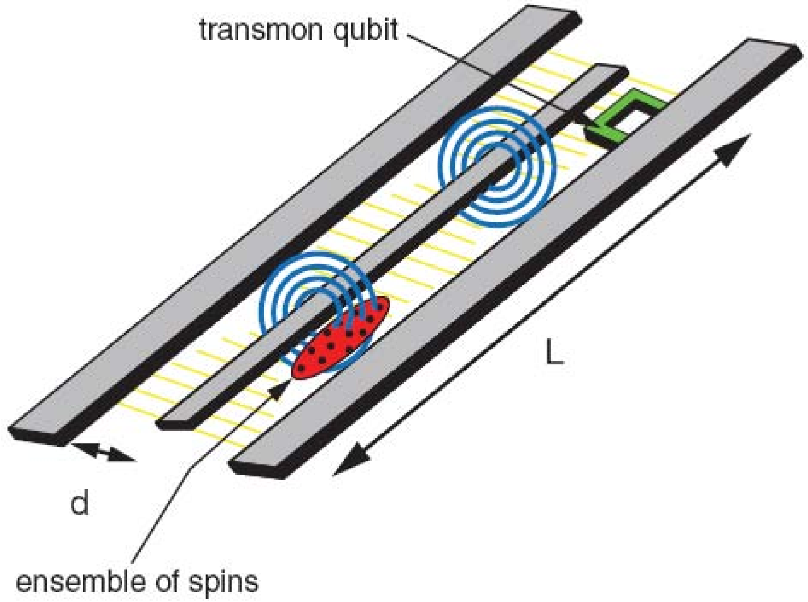
$10^2\ \mu_B/\mathrm{Hz}^{1/2}$

M. J. Martínez-Pérez, E. Bellido, R.. De Miguel, J. Sesé, A. Lostao, C. Gómez-Moreno, D. Drung, T. Schurig, D. Ruiz-Molina, and F. Luis, APL. **99**, 032504 (2011)

Hybrid quantum computation architectures



Magnetic qubits as hardware for quantum computers. J. Tejada, E. M. Chudnovsky, E. del Barco, J. M. Hernandez and T. P. Spiller, Nanotechnology **12** (2001) 181–186



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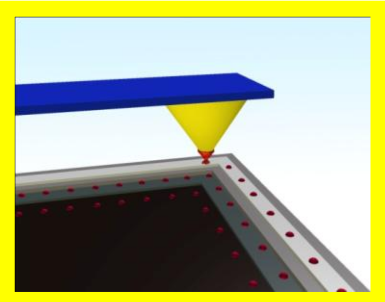
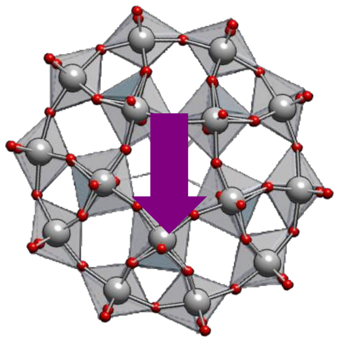
Molecule-based qubits and qugates



Superconducting μcircuits

Conclusions

- LnW_{10} and LnW_{30} are solid candidates to act as spin qubits

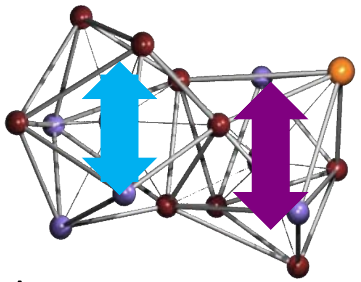


- [LnLn'] clusters, designed and synthesized via coordination chemistry, meet the following ingredients

    - weak AF coupling between qubits

    - magnetic asymmetry

      molecular prototypes for
      CNOT quantum gates



    - SWAP gate operations can be performed in the same molecule

- Dip pen nanolithography enables integrating molecular qubits into superconducting microdevices: towards the implementation of quantum architectures

icma

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Guillem Aromí,
David Aguilá (et al.)

ICMol
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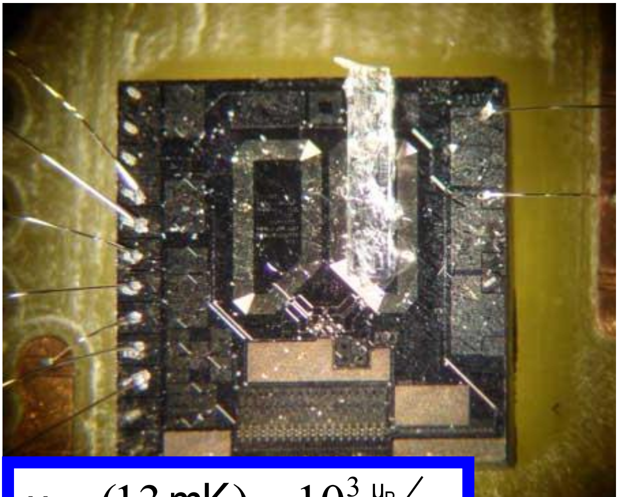
CAMPUS UAB. BELLATERRA. BARCELONA


Elena Bellido


Daniel Ruiz

Micro-SQUID ac susceptibility: single-ion magnet behaviour

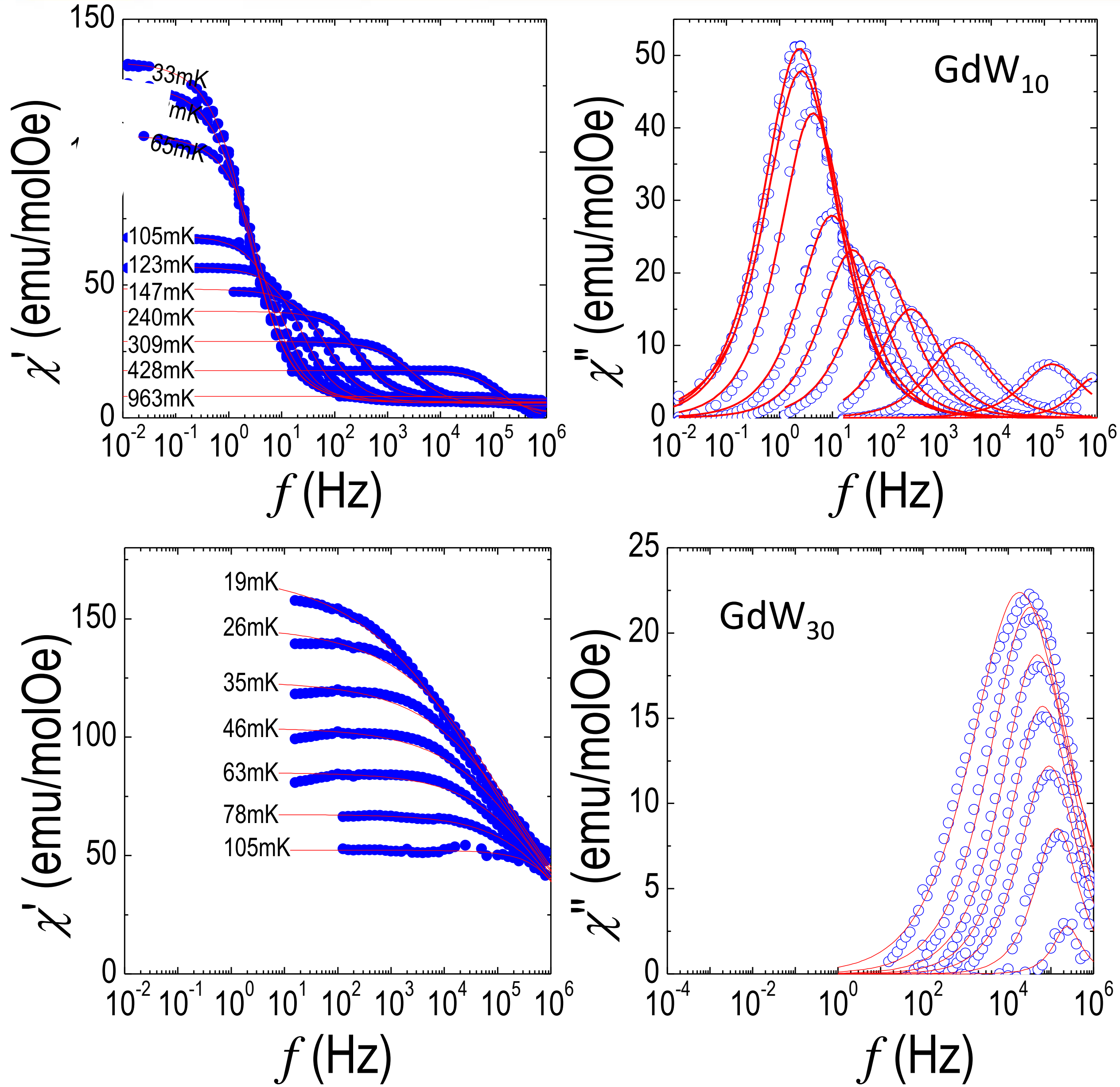


$$\mu_{min}\,(13\,\text{mK}) \approx 10^3\ \mu_B/\text{Hz}^{1/2}$$

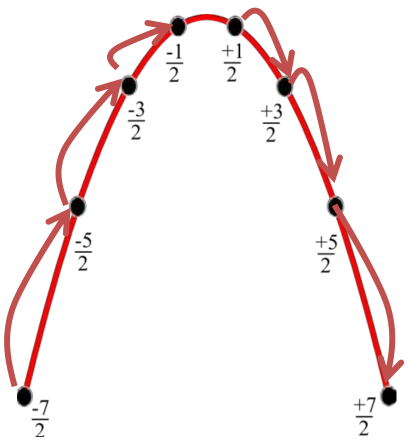
$$1\,\text{mHz} \leq f \leq 1\,\text{MHz}$$

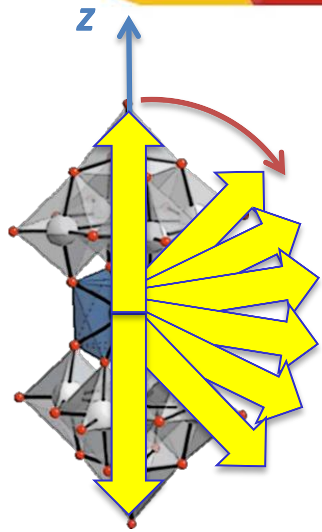
M. J. Martíinez-Pérez, J. Sesé, F. Luis, D. Drung, T. Schurig, Rev. Sci. Instrum. **81**, 016108 (2010).

$$\chi = \chi_S + \frac{\chi_T - \chi_S}{1 + (i\omega\tau)^{1-\beta}}$$



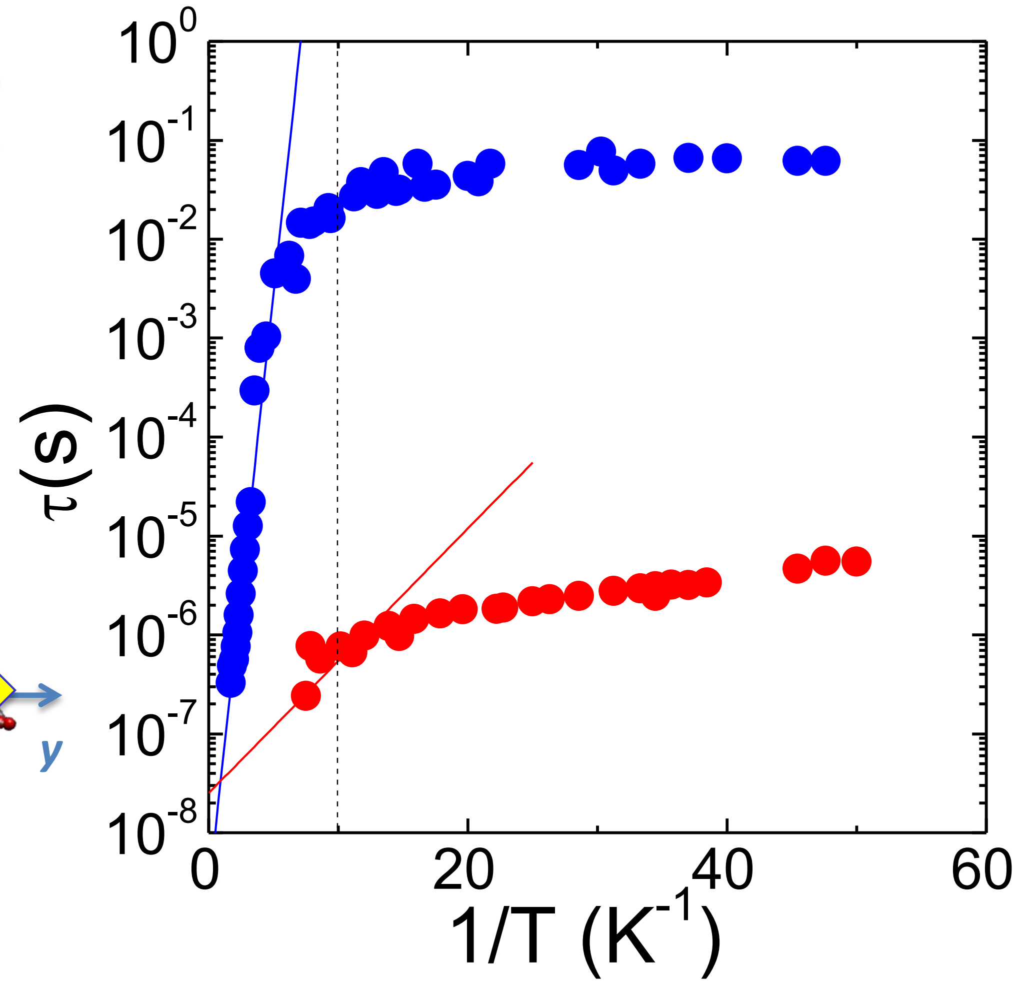
Spin-lattice relaxation

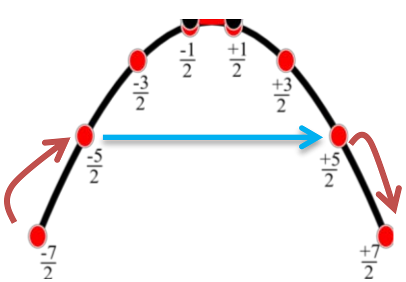


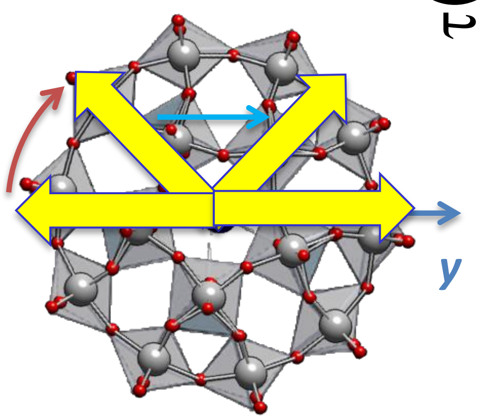


T > 0.1 K:

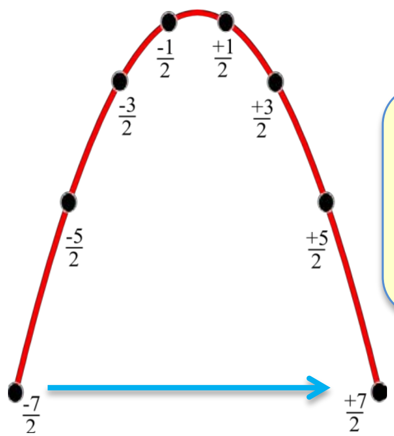
Thermally activated relaxation agrees with master equation calculations







Spin-lattice relaxation



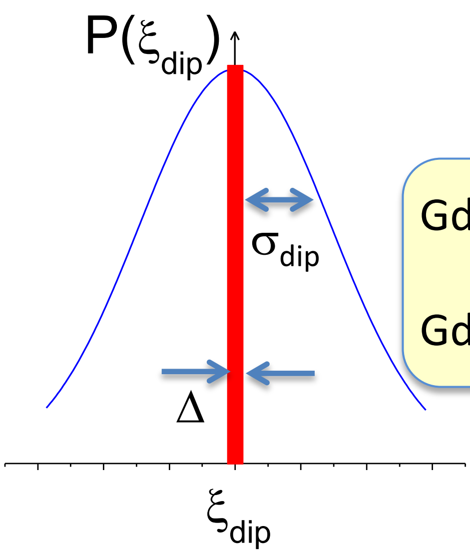
GdW$_{10}$: $\Delta/k_B \approx 3.7\ \mu K$

GdW$_{30}$: $\Delta/k_B \approx 74\ \mu K$

$$\Gamma_{tun} \approx \frac{\Delta^2}{\hbar \sigma_{dip}}$$

GdW$_{10}$: $\sigma_{dip}/k_B \approx 36$ mK

GdW$_{30}$: $\sigma_{dip}/k_B \approx 5$ mK



T < 0.1 K:

Pure quantum tunneling: agrees with prediction of Prokof'ev and Stamp (PS), PRL **80**, 5794 (1998).