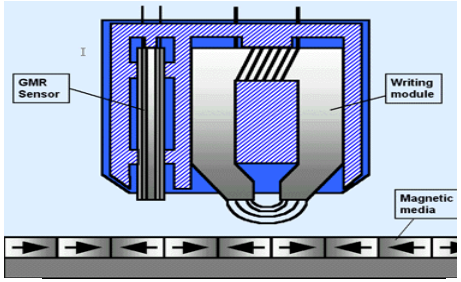
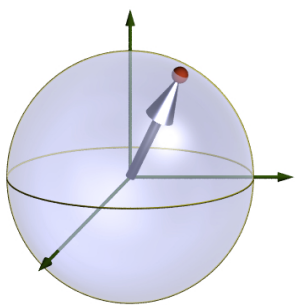
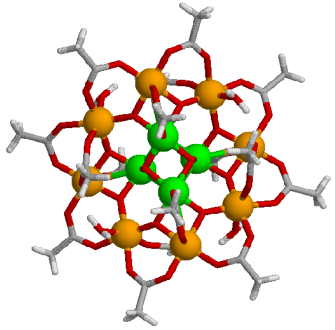
2021 European School on Magnetism

Cluj Napoca
14th September 2021

Molecular magnets: physics and applications







Fernando LUIS

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INMA
Instituto de Nanociencia y Materiales de Aragón

Universidad Zaragoza
1542

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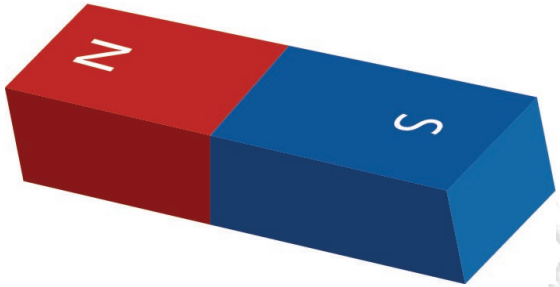
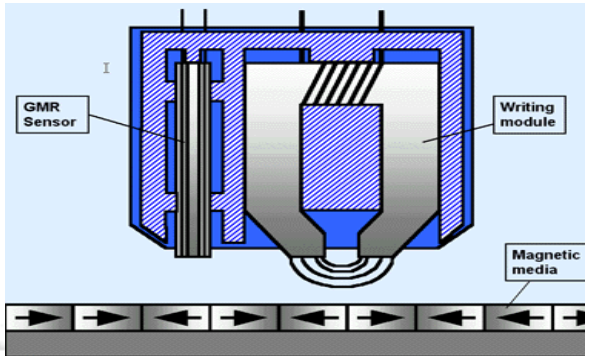
Outline



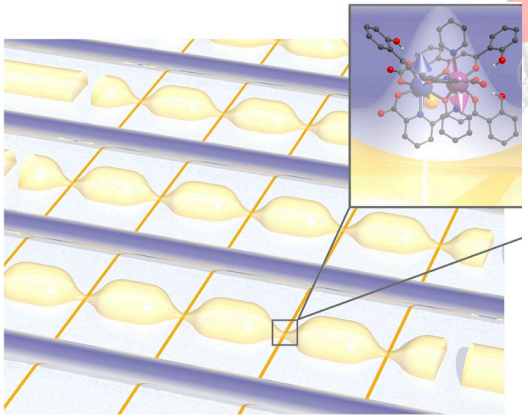
2. Magnets



3. Magnetic recording

1. Physics

4. Quantum information



5. Magnetic refrigeration

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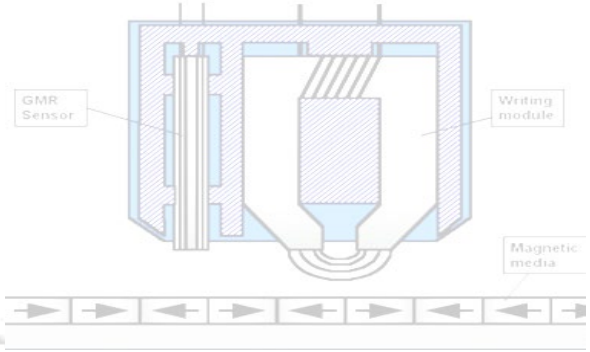
Outline

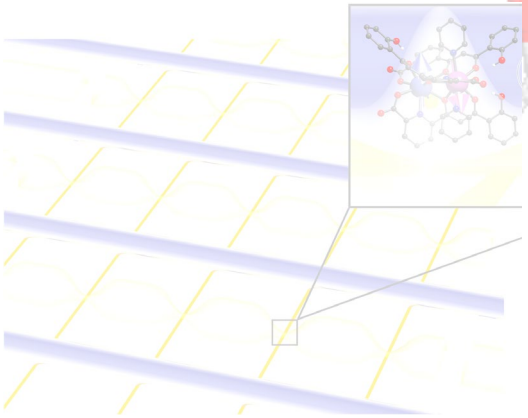
2. Magnets



3. Magnetic recording

1. Physics

4. Quantum information

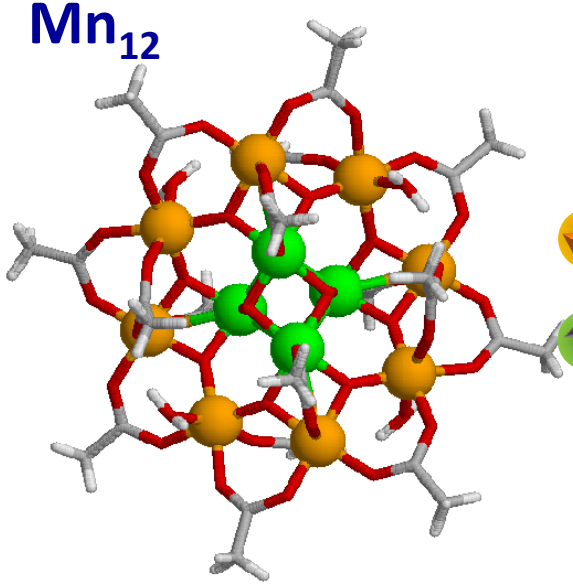


5. Magnetic refrigeration

Molecular magnets

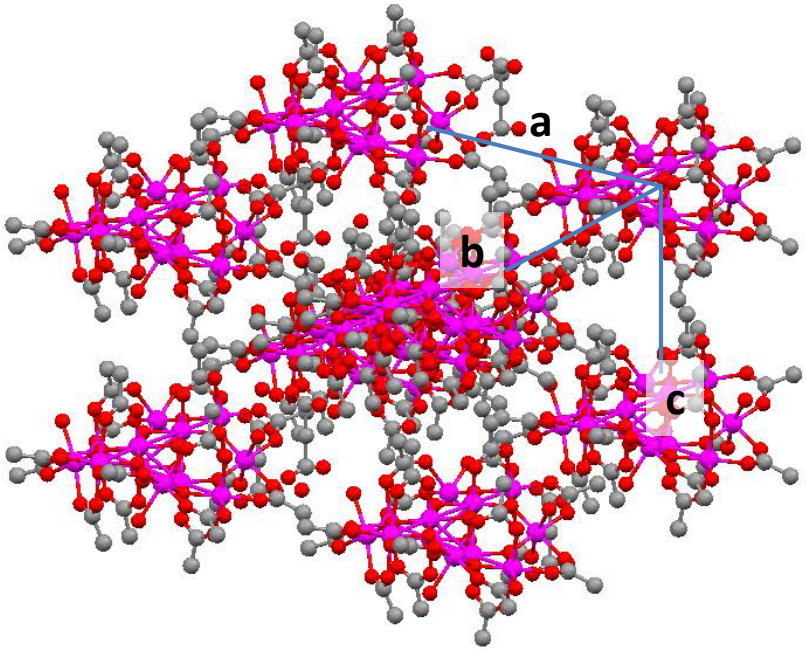
T. Lis, Acta Crystalogr. Sect. B **36**, 2042 (1980)

Mn_{12}



Mn^{3+} $s = 2$

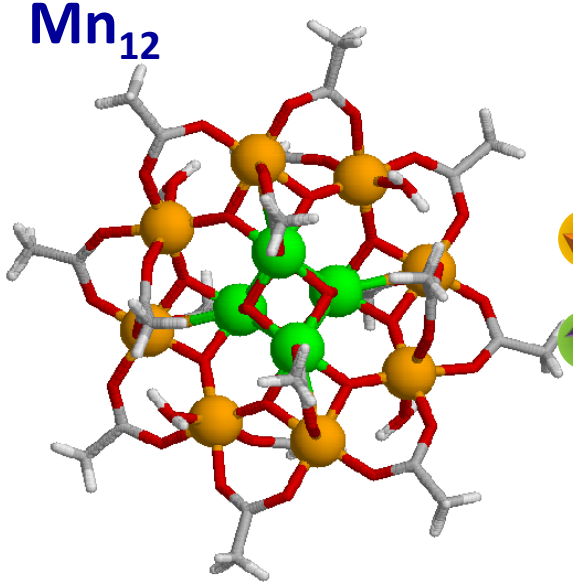
Mn^{4+} $s = 3/2$



Molecular magnets

T. Lis, Acta Crystalogr. Sect. B **36**, 2042 (1980)

Mn_{12}



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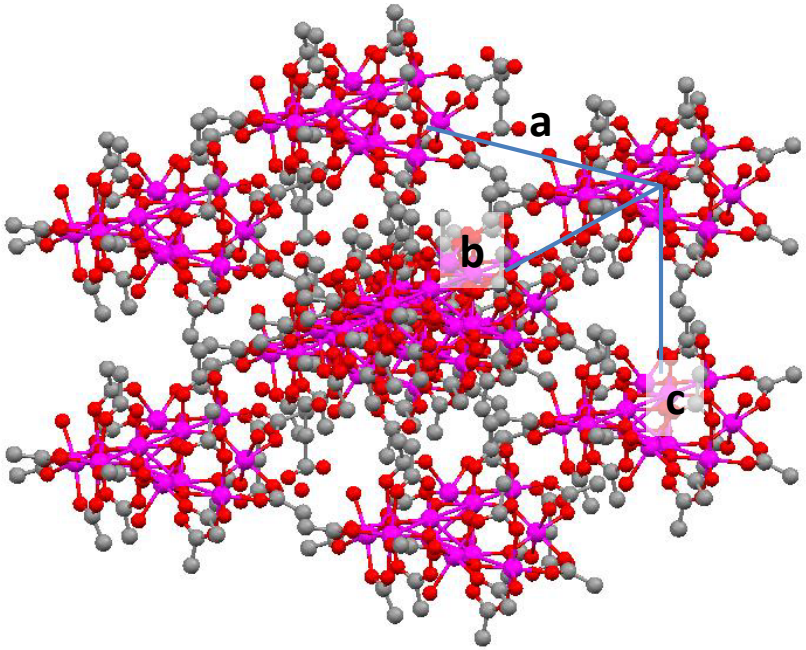
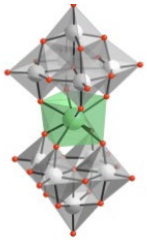
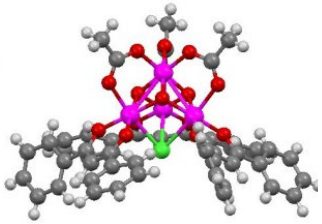
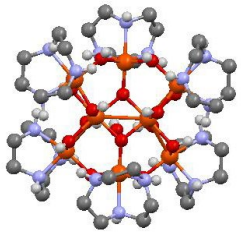
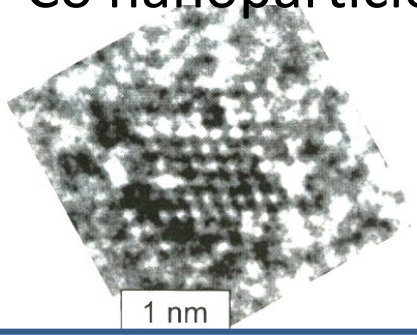


ErW_{10} Mn_4Cl Fe_8 Co nanoparticle Magnet









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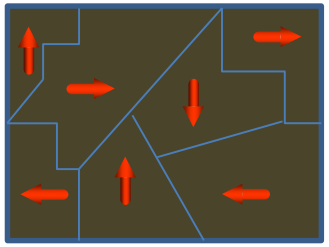

0.1 nm 1 nm 10 nm > 100 nm

Molecular Magnets: Physics and Applications, edited by J. Bartolomé, F. Luis, and J. F. Fernández, Springer Verlag (Berlín, Heidelberg, 2014)

Molecular magnets

T. Lis, Acta Crystalogr. Sect. B **36**, 2042 (1980)

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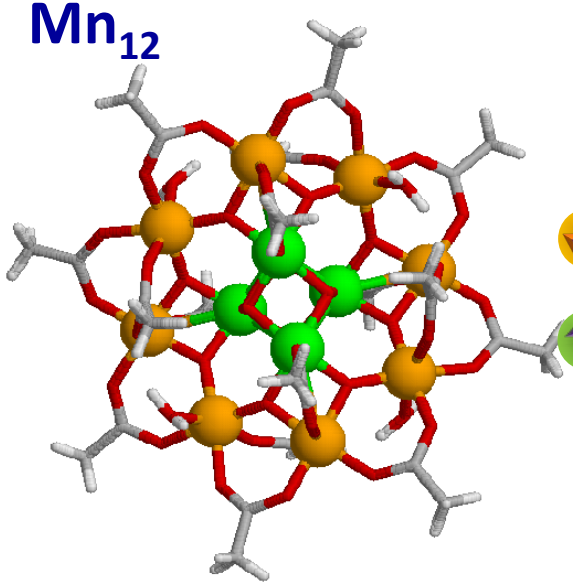
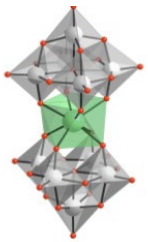


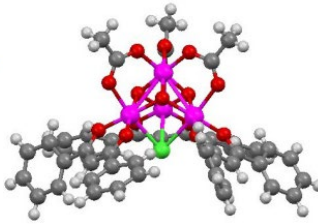
Mn^{3+} $s = 2$

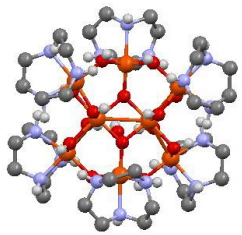
Mn^{4+} $s = 3/2$

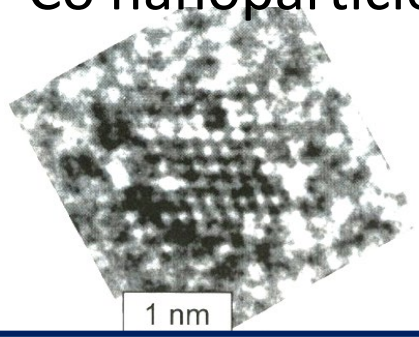
- Mesoscopic size
- Monodisperse
- Chemical design

ErW_{10} Mn_4Cl Fe_8 Co nanoparticle Magnet









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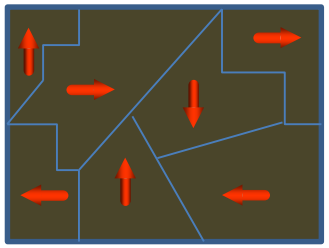

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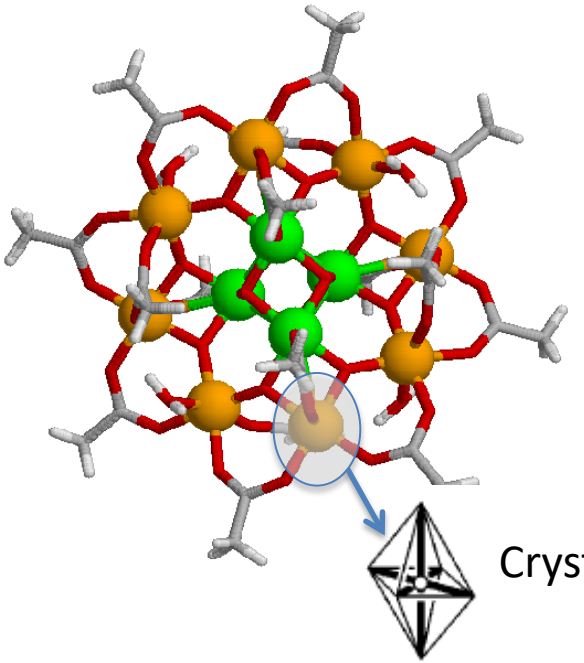
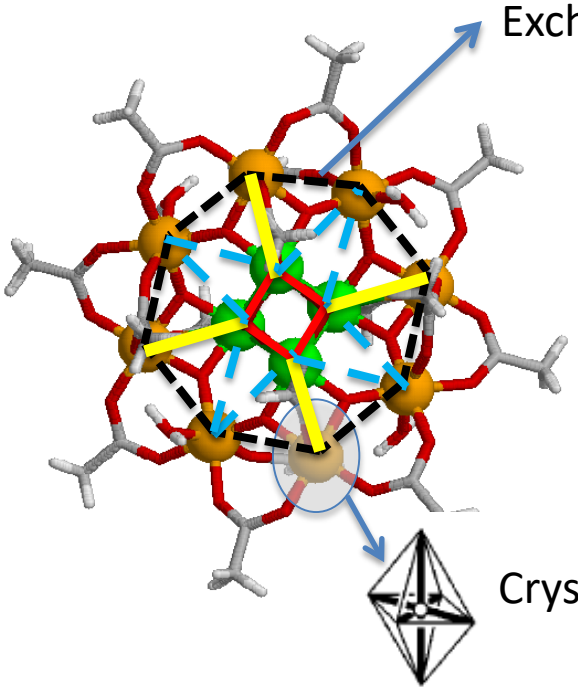
Molecular spin states



Crystal field: anisotropy
D_i

$$\mathcal{H} = \sum_{i=1}^{n} \overrightarrow{s_i} \widehat{D_i} \overrightarrow{s_i}$$

Molecular spin states

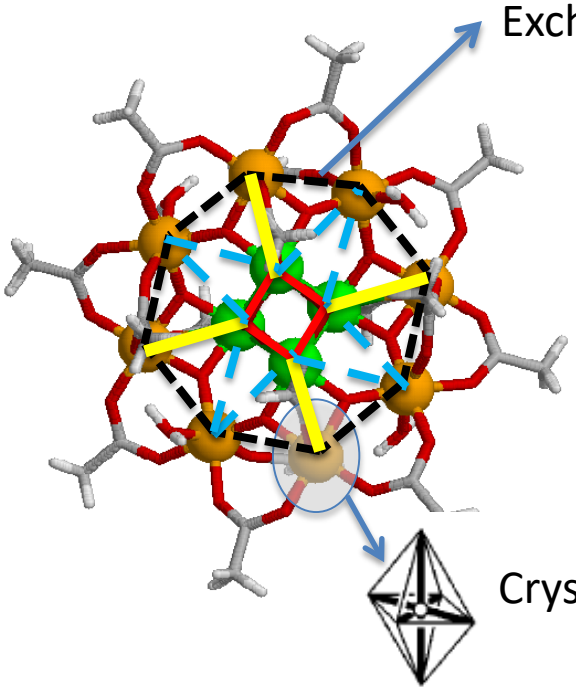


Exchange couplings J_{ij}

Crystal field: anisotropy
D_i

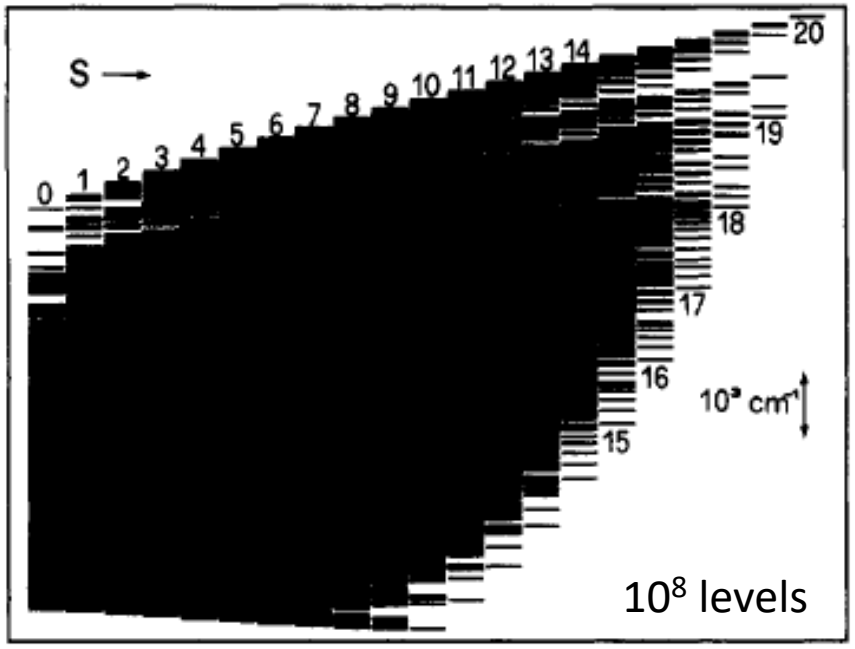
$$\mathcal{H} = \sum_{i=1}^{n} \overrightarrow{s_i}\widehat{D_i}\overrightarrow{s_i} + \sum_{i \neq j}^{n} \overrightarrow{s_i}\widehat{J_{ij}}\overrightarrow{s_j}$$

Molecular spin states



Exchange couplings J_{ij}

Crystal field: anisotropy D_i



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R. Sessoli, H.-L. Tsai, A.R. Shake, S. Wang, J.B. Vincent, K. Folting, D. Gatteschi, G. Christou, D.N. Hendrickson, J. Am. Chem. Soc. **115**, 1804 (1993).

M.I. Katsnelson, V.V. Dobrovitski, B.N. Harmon, Phys. Rev. B **59**, 6919 (1999).

V. V. Mazurenko, Y. O. Kvashnin, Fengping Jin, H. A. De Raedt, A. I. Lichtenstein, and M. I. Katsnelson, Phys. Rev. B **89**, 214422 (2014)

O. Hanebaum and J. Schnack, Phys. Rev. B **92**, 064424 (2015)

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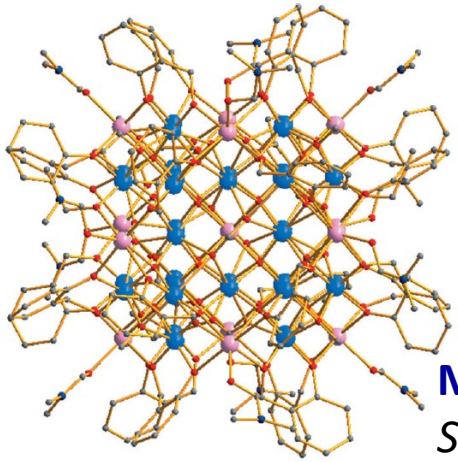
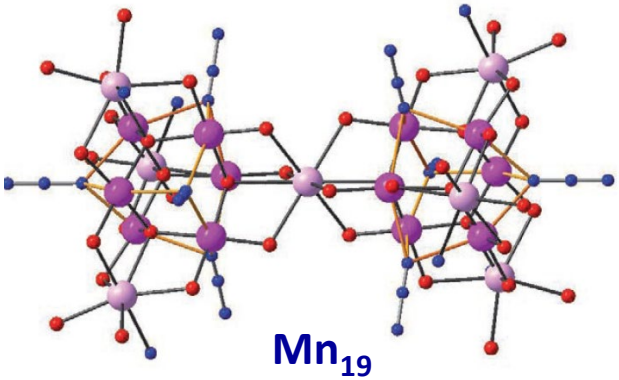
Record spin values



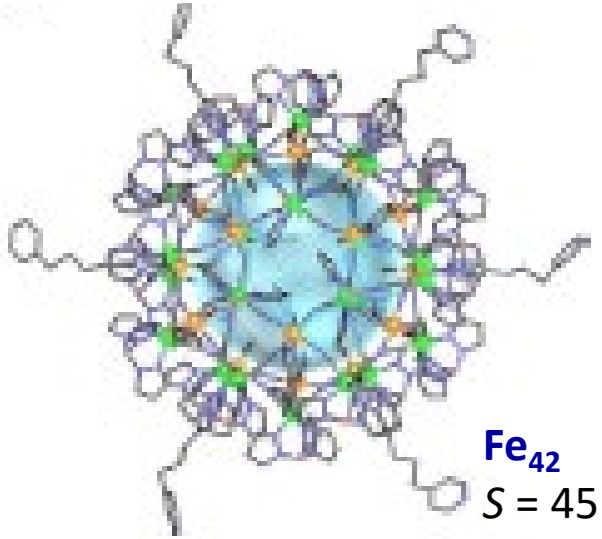
Mn_{49}
$S = 61/2$

M. Manoli et al, Angew. Chem. **55**, 679 (2016)



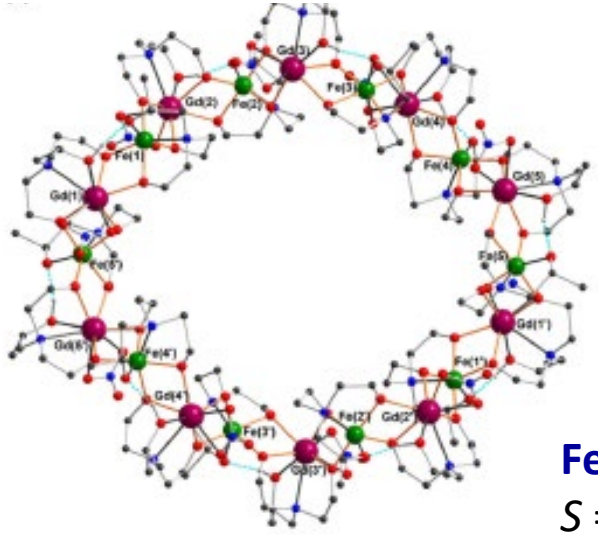
Mn_{19}
$S = 83/2$

A. M. Ako et al, Angew. Chem. **45**, 4926 (2006)



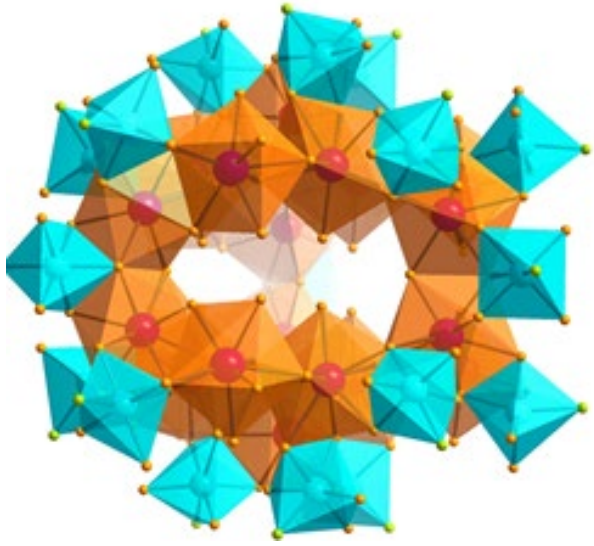
Fe_{42}
$S = 45$

A. M. Ako et al, Nat. Commun. **6**, 5955 (2015)



$Fe_{10}Gd_{10}$
$S = 60$

A. Baniodeh et al, npj Quant. Mater. **3**, 10 (2018)



$Ni_{21}Gd_{20}$
$S = 91$

W. P. Chen et al, Nat. Commun. **9**, 2107 (2018)

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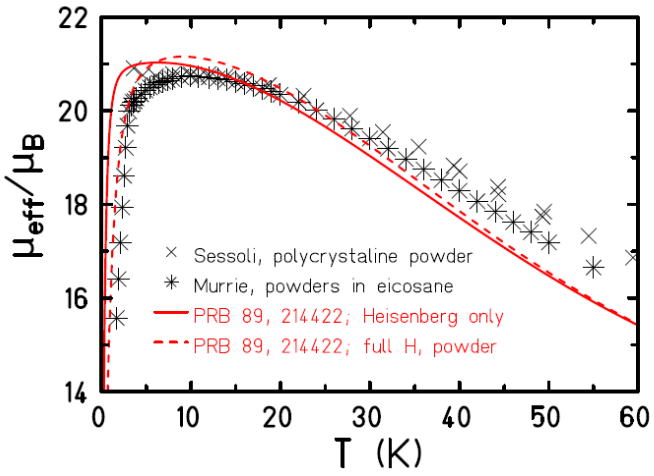
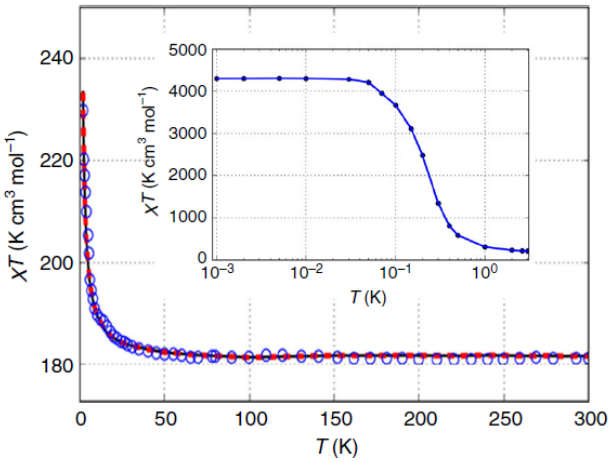
Test bed for quantum simulation methods

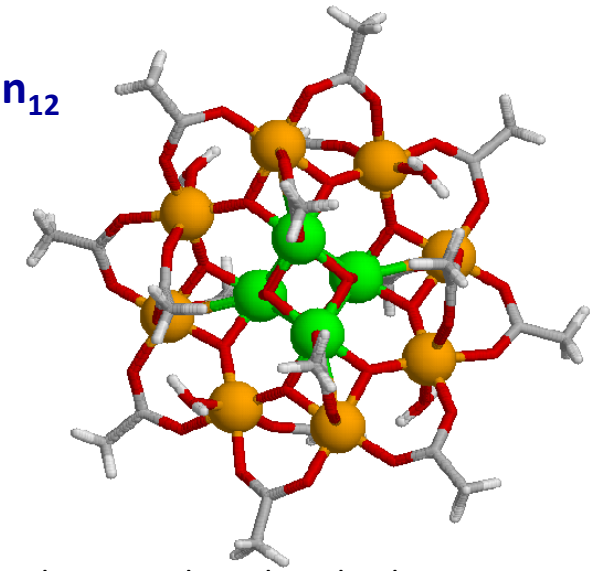


Lanczos approximate diagonalization method

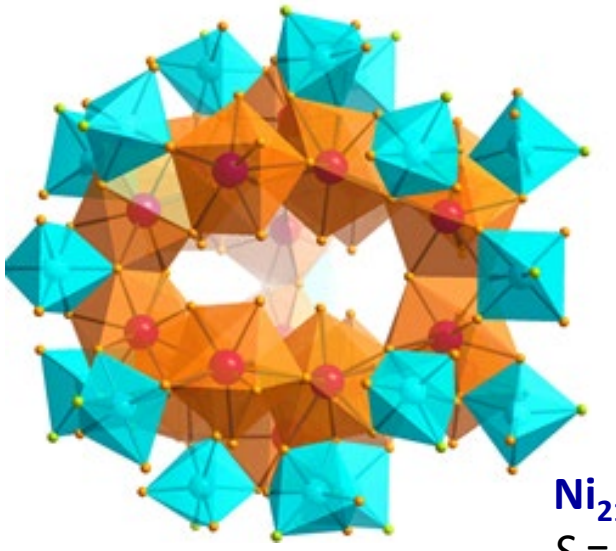


Quantum Monte Carlo simulations

Mn_{12}



O. Hanebaum and J. Schnack, Phys. Rev. B **92**, 064424 (2015)



$Ni_{21}Gd_{20}$
$S = 91$

W. P. Chen et al, Nat. Commun. **9**, 2107 (2018)

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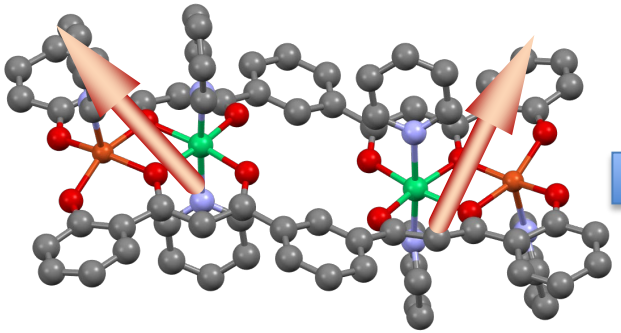
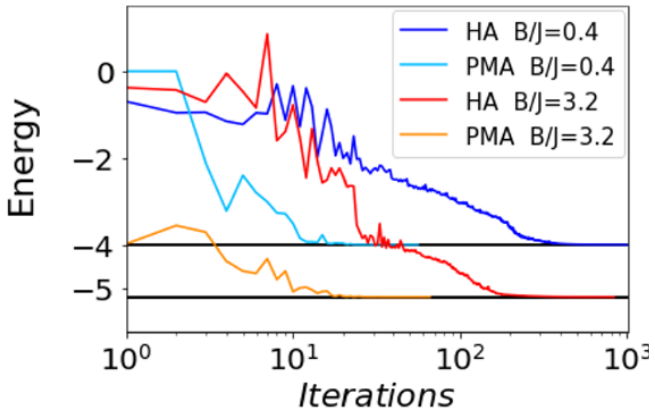
Quantum hardware simulations

Molecular dimer of two coupled $S = ½$ spins

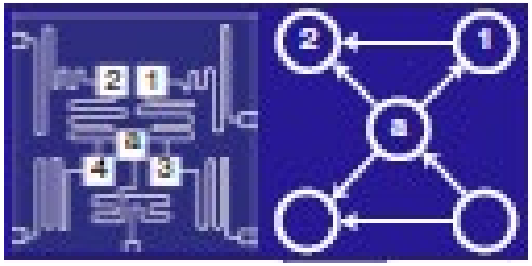


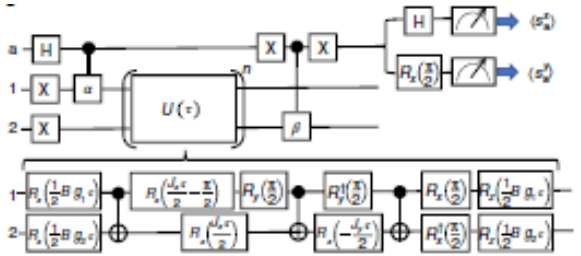
$$\mathcal{H} = \overrightarrow{s_1}\hat{J}\overrightarrow{s_2}$$

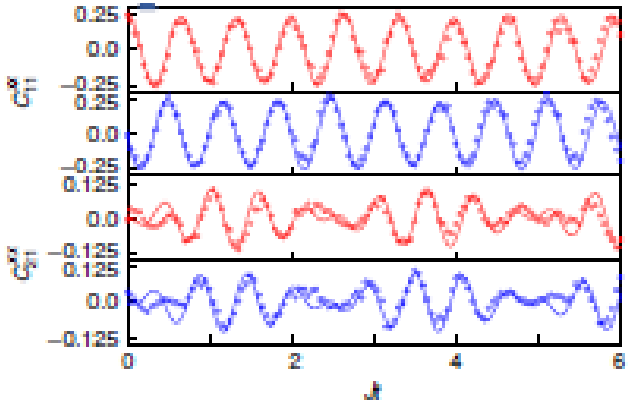


A. Chiesa et al, Nature Phys. **15**, 455 (2019)

Ibmqx4 processor

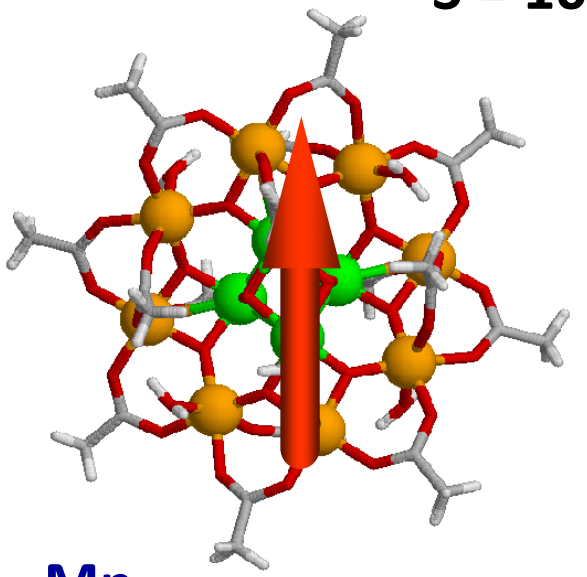






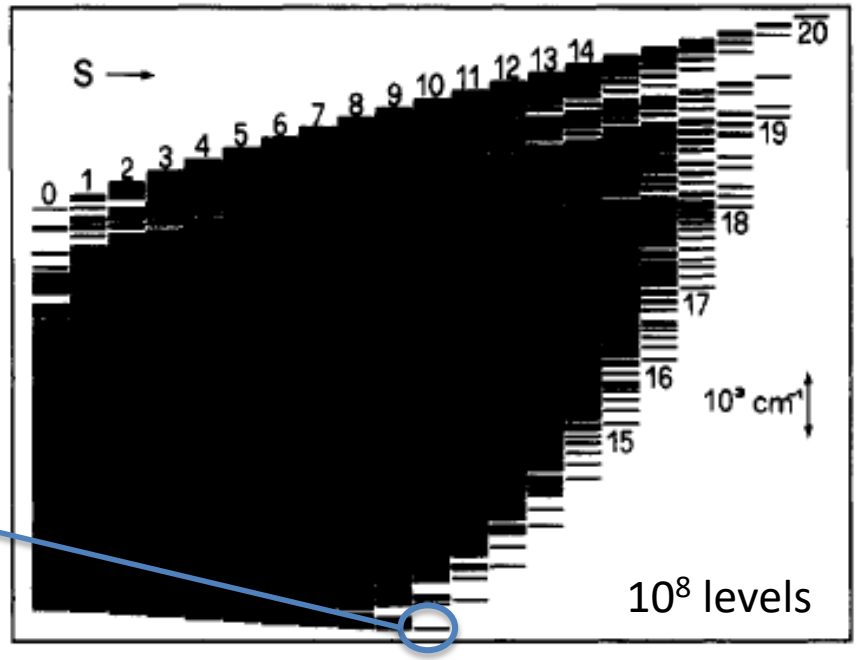
L. Crippa et al, Magnetochem. **7**, 117 (2021)

“Giant spin” approximation



S = 10

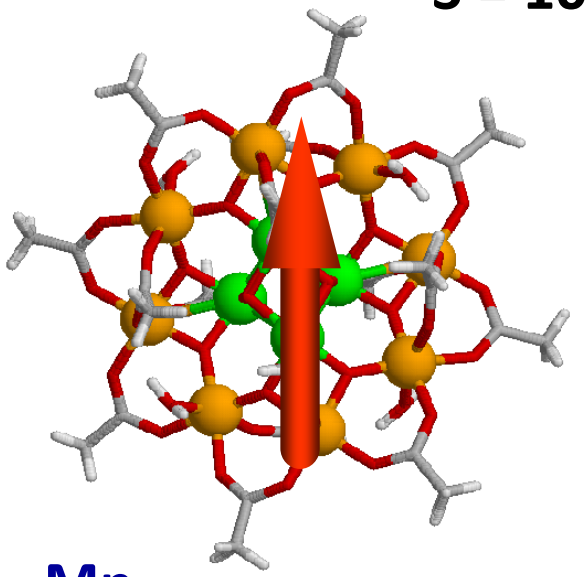
Mn_{12}



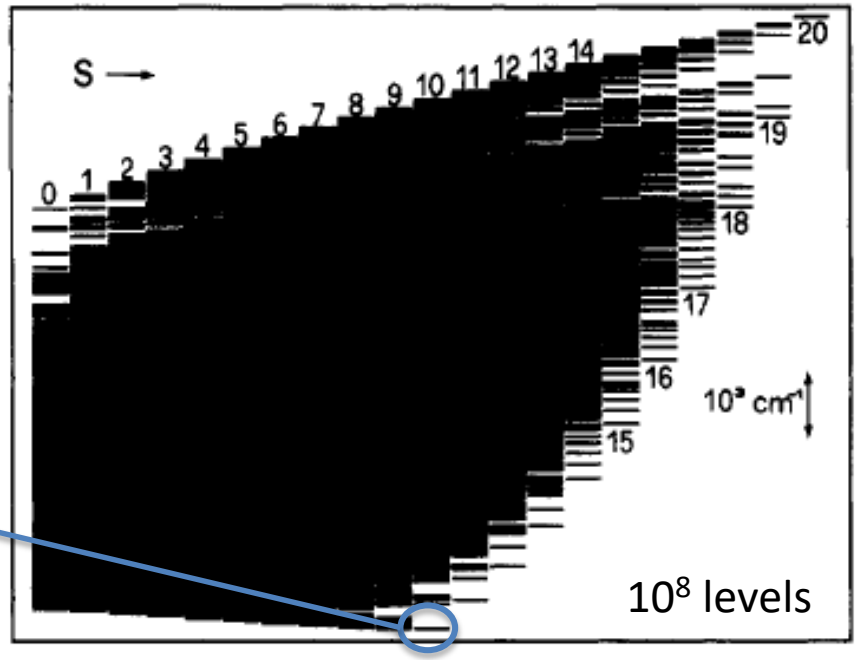
$$\mathcal{H} = \sum_{i=1}^{n} \vec{s_i} \hat{D_i} \vec{s_i} + \sum_{i \neq j}^{n} \vec{s_i} \vec{s_j}$$

Magnetic anisotropy

S = 10

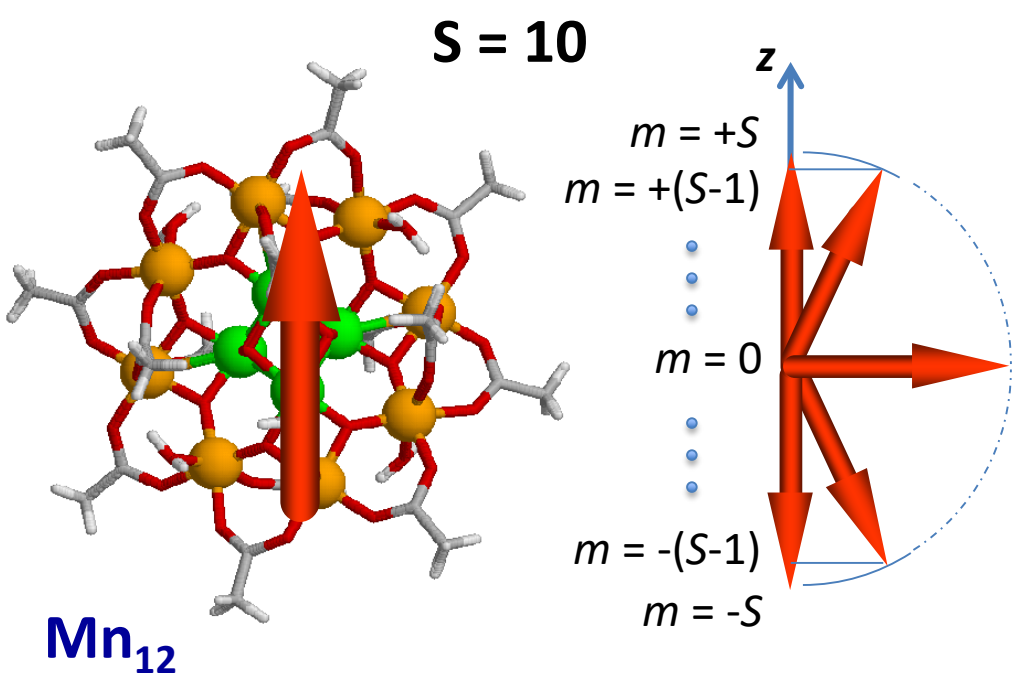


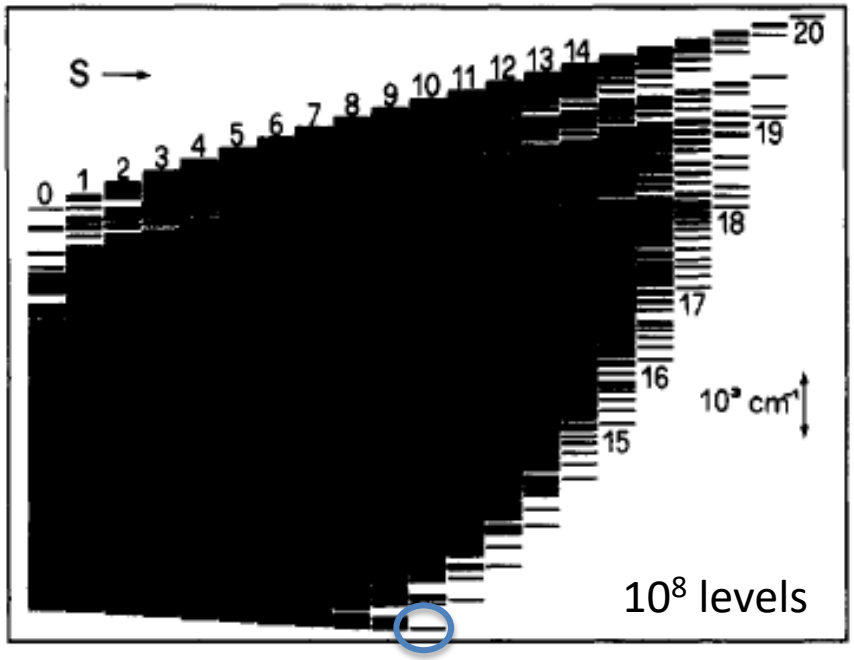
Mn_{12}



$$\mathcal{H} \approx -DS_z^2 + AS_z^4 + C\left(S_x^4 + S_y^4\right)$$

Magnetic anisotropy





$$\mathcal{H} \approx \boxed{-DS_z^2 + AS_z^4} + C\left(S_x^4 + S_y^4\right)$$

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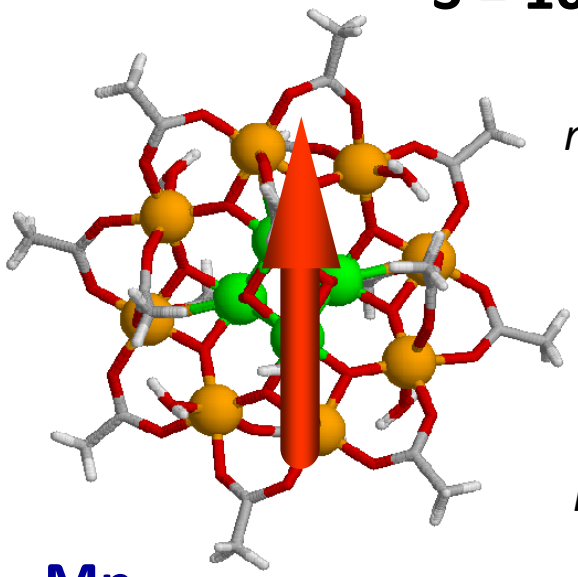
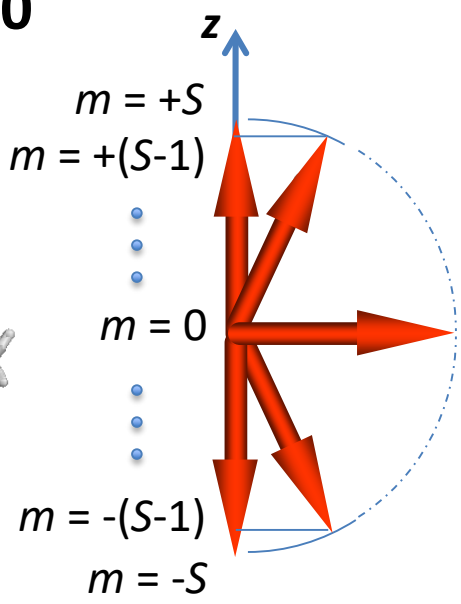
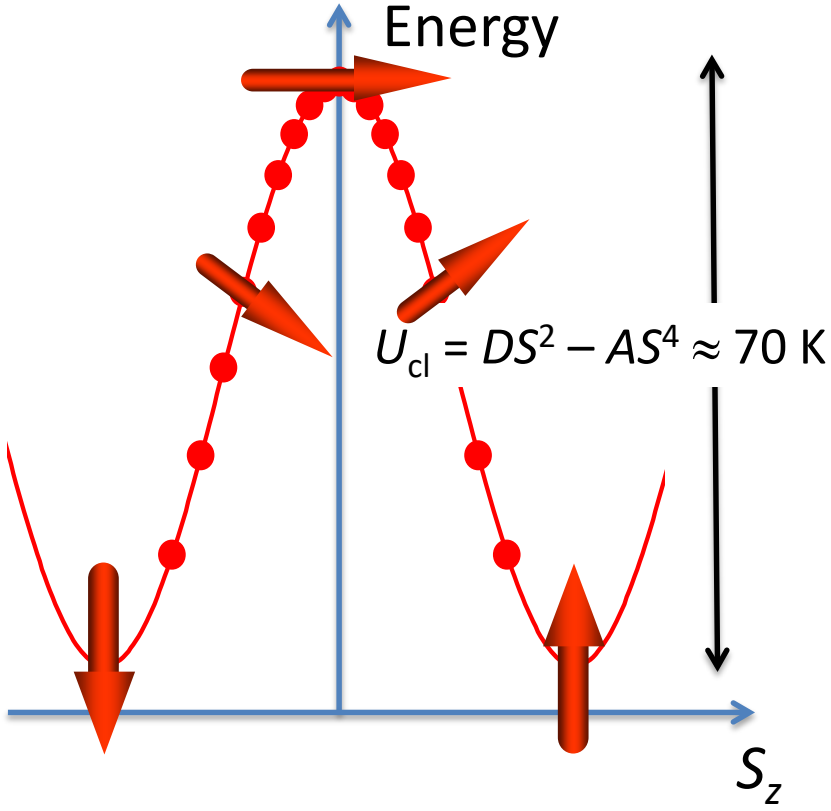
Magnetic anisotropy: diagonal terms



S = 10

Mn_{12}

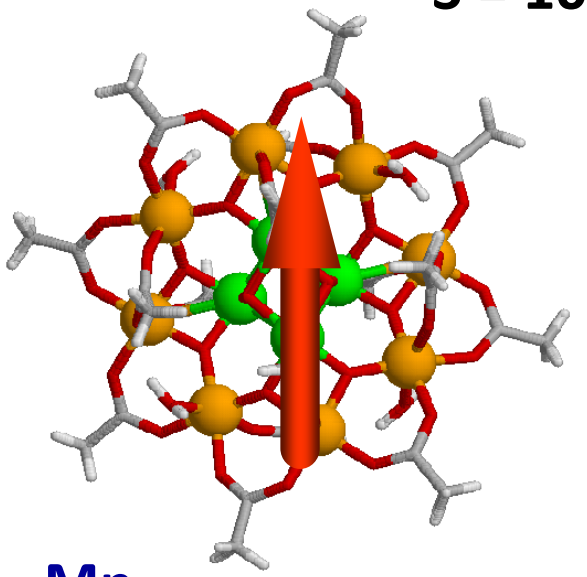




$$\mathcal{H} \approx \boxed{-DS_z^2 + AS_z^4} + C\left(S_x^4 + S_y^4\right)$$

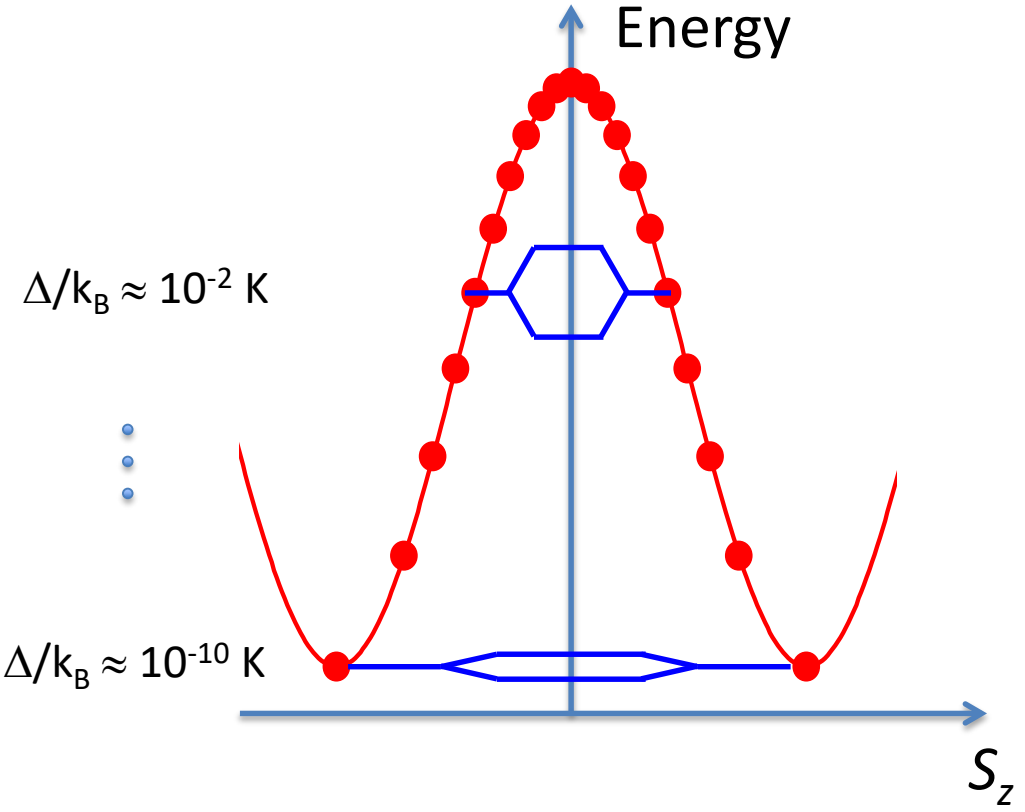
R. Sessoli, D. Gatteschi, A. Caneschi and M. A. Novak, Nature **365**, 141 (1993); F. Hartmann-Boutron, P. Politi and J. Villain, Int- J. Mod. Phys. **10**, 2577 (1996).

Magnetic anisotropy: off-diagonal terms



S = 10

Mn_{12}



$$\mathcal{H} \approx -DS_z^2 + AS_z^4 + C\left(S_x^4 + S_y^4\right)$$

R. Sessoli, D. Gatteschi, A. Caneschi and M. A. Novak, Nature **365**, 141 (1993); F. Hartmann-Boutron, P. Politi and J. Villain, Int- J. Mod. Phys. **10**, 2577 (1996).

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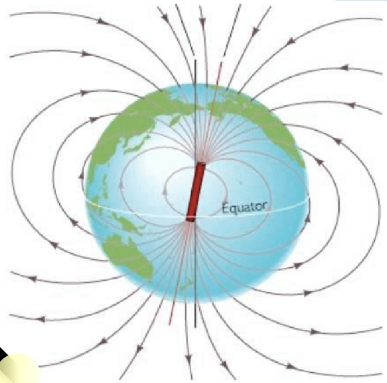
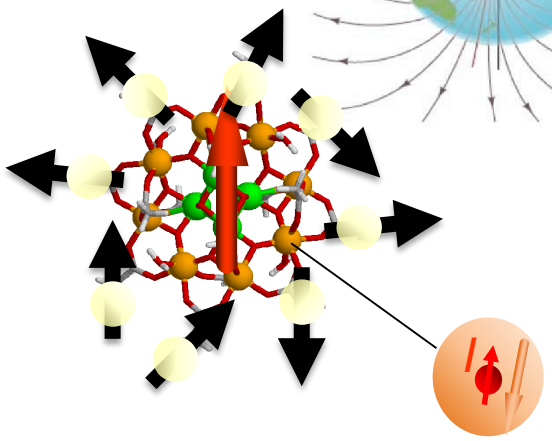
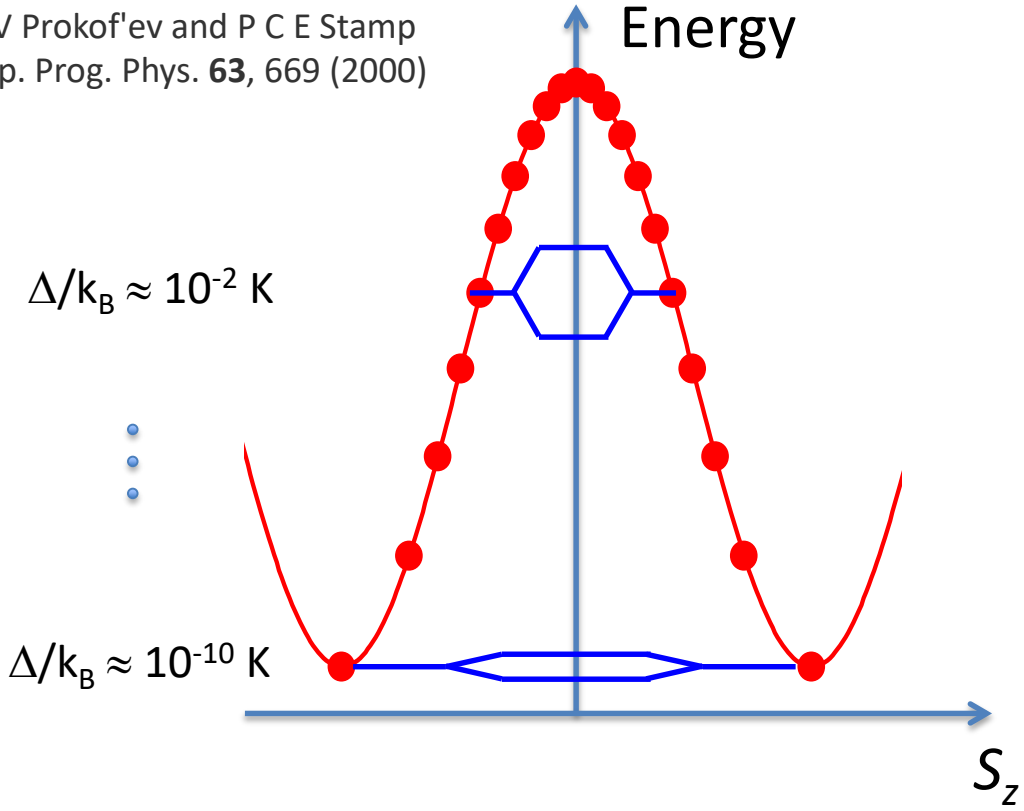
Magnetic anisotropy: off-diagonal terms

Environmental fields





N V Prokof'ev and P C E Stamp
Rep. Prog. Phys. **63**, 669 (2000)



Magnetic anisotropy: off-diagonal terms

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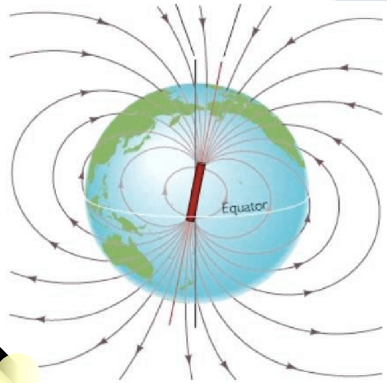
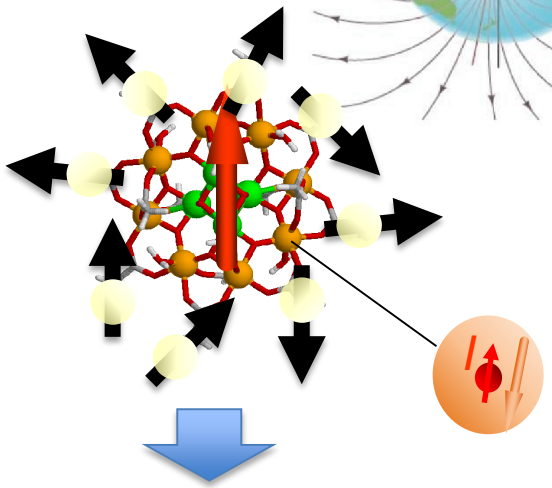
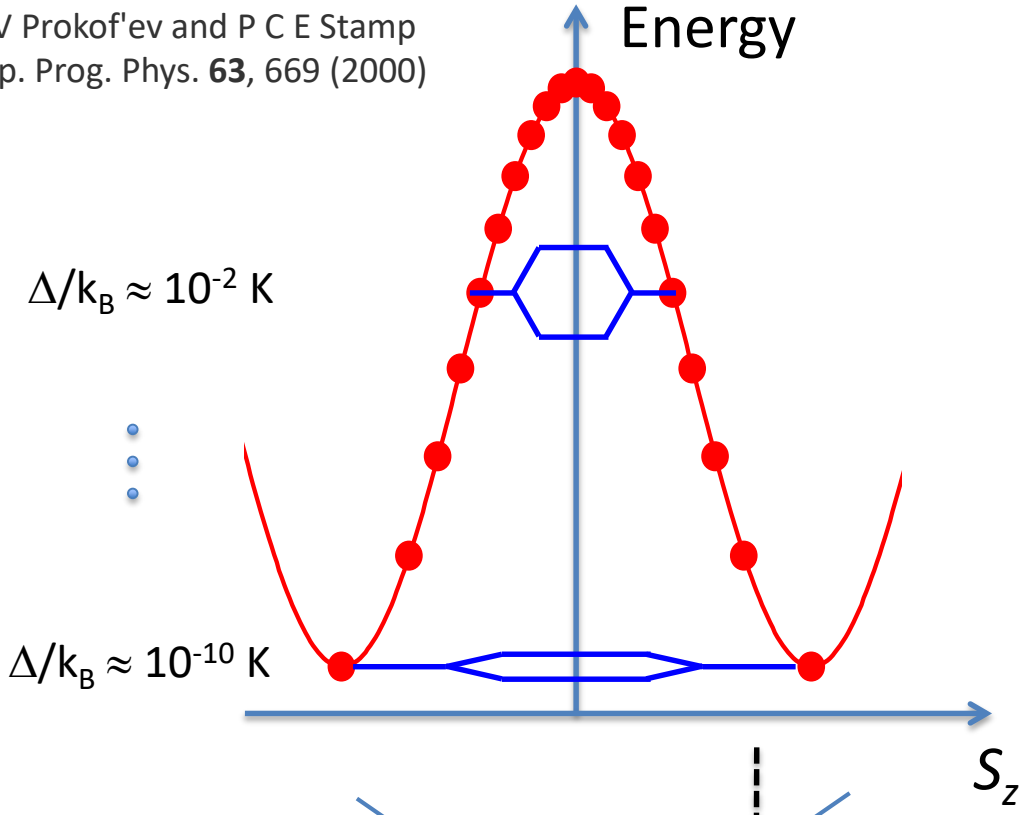
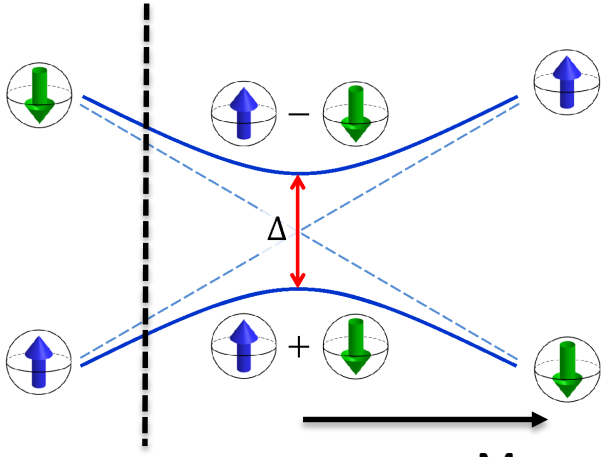
Environmental fields

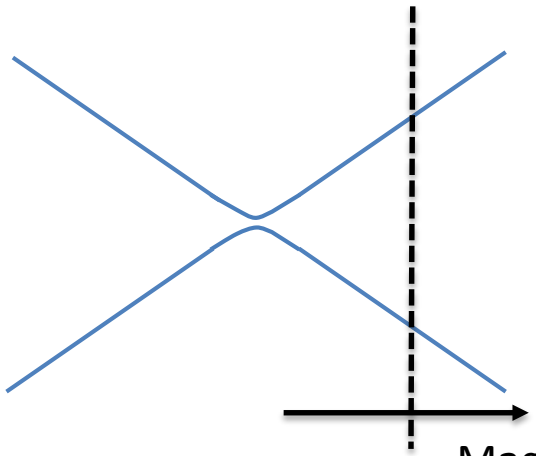




N V Prokof'ev and P C E Stamp
Rep. Prog. Phys. **63**, 669 (2000)







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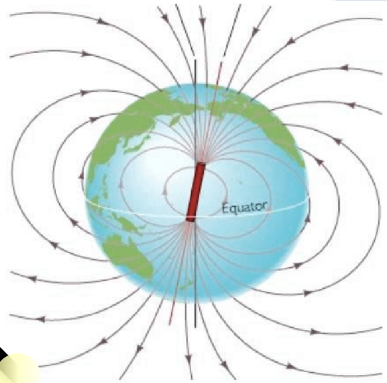
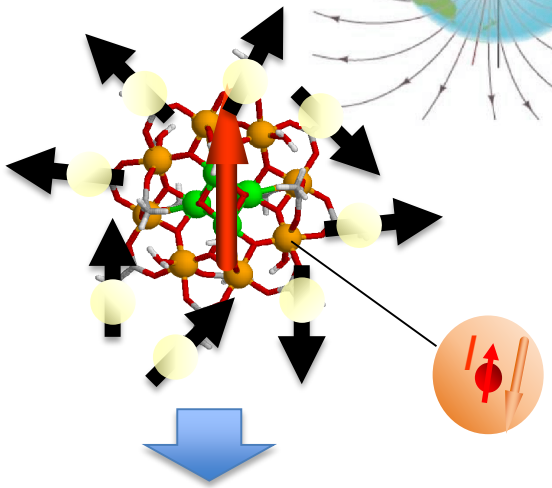
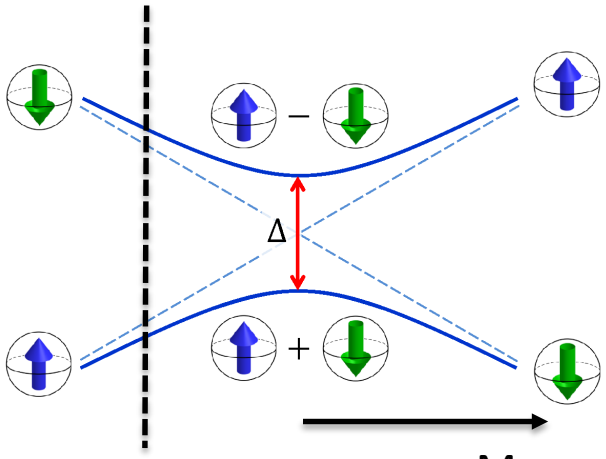
Magnetic anisotropy: off-diagonal terms

N V Prokof'ev and P C E Stamp
Rep. Prog. Phys. **63**, 669 (2000)

Environmental fields



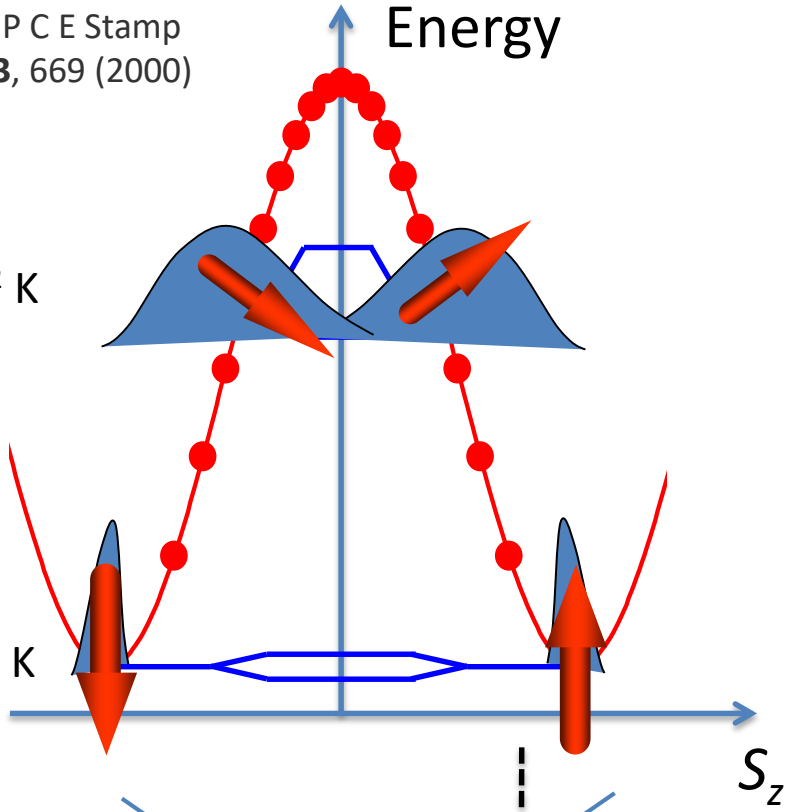


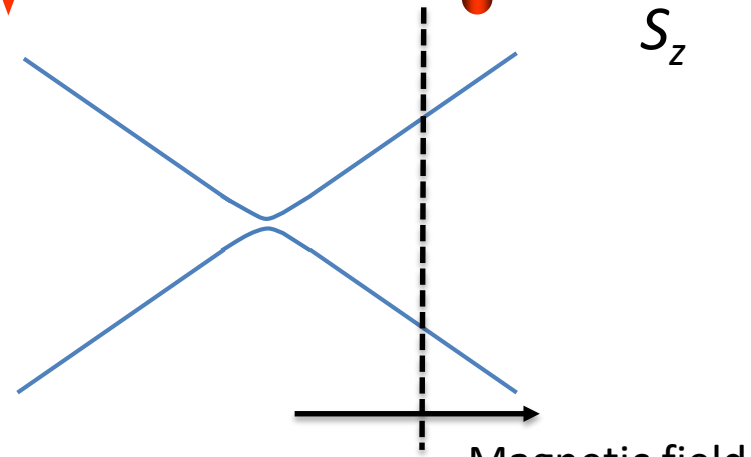


Energy

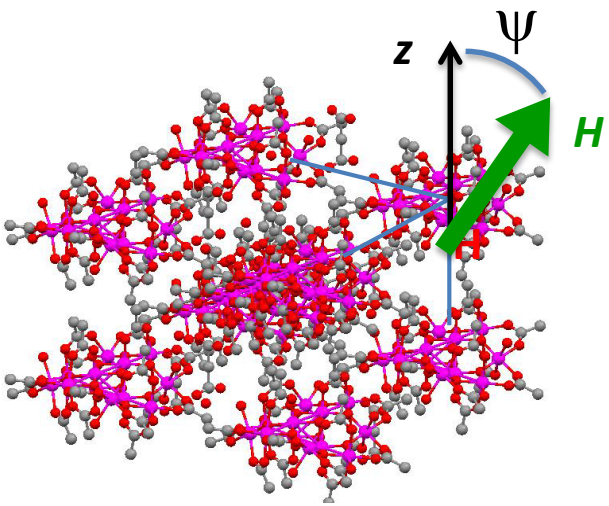
$\Delta/k_B \approx 10^{-2}$ K

$\Delta/k_B \approx 10^{-10}$ K

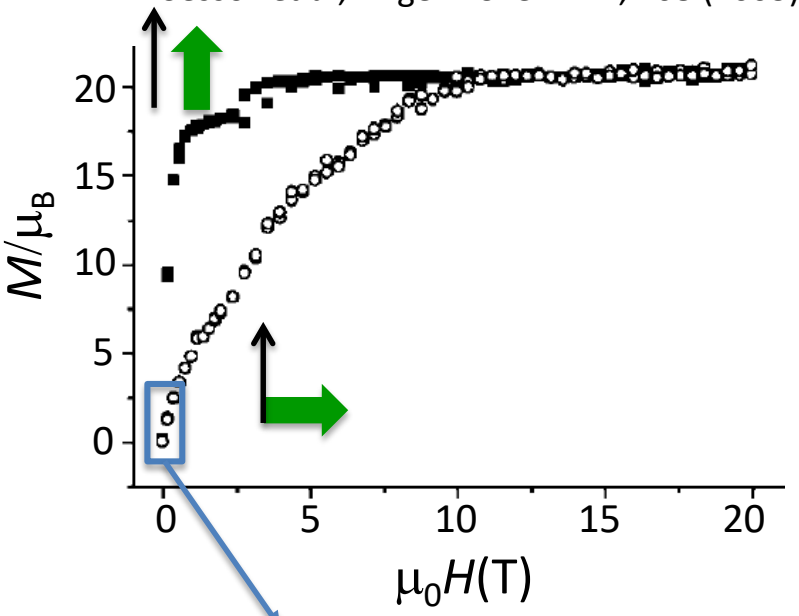




Angle-dependent response



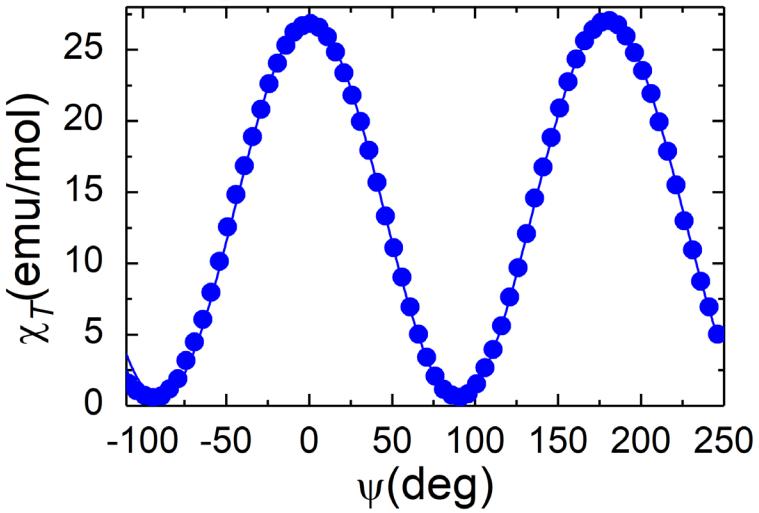
R. Sessoli *et al*, Angew. Chem. **42**, 268 (2003)



$$\mathcal{H} \approx \mathcal{H}_{\text{anis}} - g\mu_B \vec{H}\vec{S}$$

$$\chi_{\perp} \simeq \frac{N g \mu_B S}{H_k}$$

$$\chi_{zz} \simeq \frac{N (g\mu_B S)^2}{k_B T}$$



E. Burzurí *et al*, Phys. Rev. B **80**, 224428 (2009)

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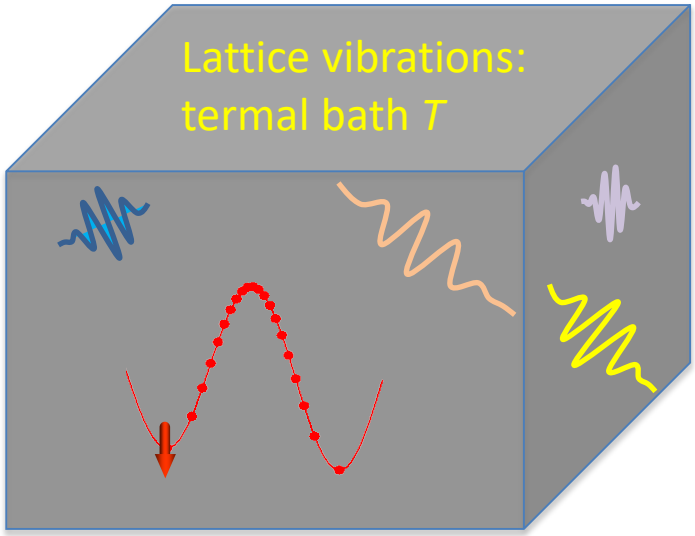
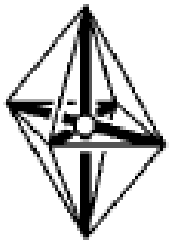
Magnetic relaxation





Modulation of the magnetic anisotropy

↓

Spin-phonon interaction

Master equation for spin level populations P_i

$$\frac{dP_i}{dt} = \sum_{j\neq i}^{2S+1} \Gamma_{i\leftarrow j} P_j - \sum_{j\neq i}^{2S+1} \Gamma_{j\leftarrow i} P_i$$

↓

$$M \simeq M_0 + (M_T - M_0)\left(1 - e^{-t_{exp}/\tau}\right)$$

Characteristic time scale $\tau(T,\boldsymbol{H})$

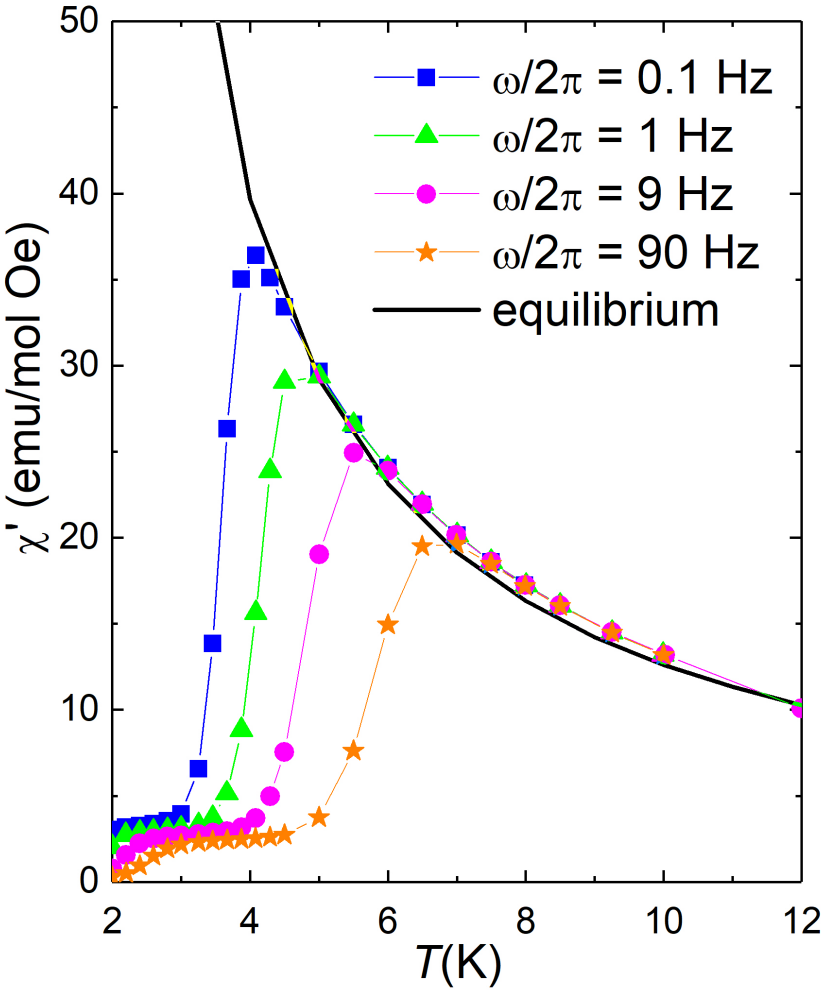
↓

Off-equilibrium for $\tau > t_{exp}$

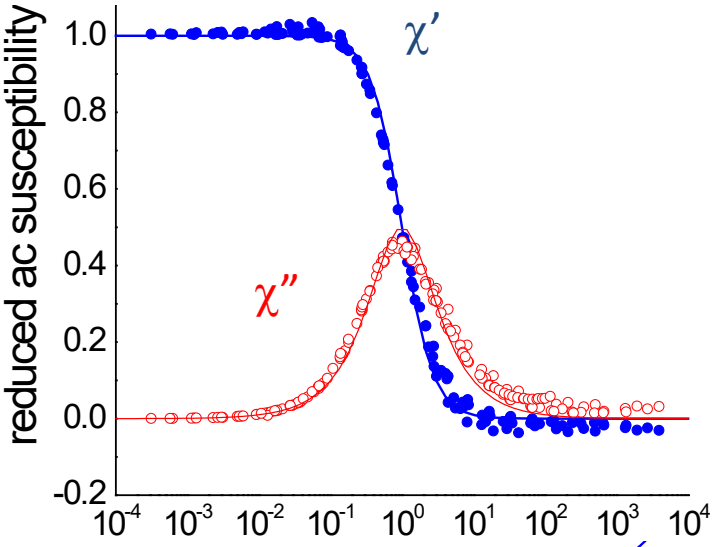
P. Ehrenfest, Leiden Commun. Suppl. **4b** (1919)
H. A. Kramers, Physica **7**, 284 (1940)
J. Villain, F. Hartman-Boutron, R. Sessoli and A. Rettori, EPL **27**, 159 (1994)
F. Luis, J. Bartolomé and J. F. Fernández, Phys. Rev. B **57**, 505 (1998)

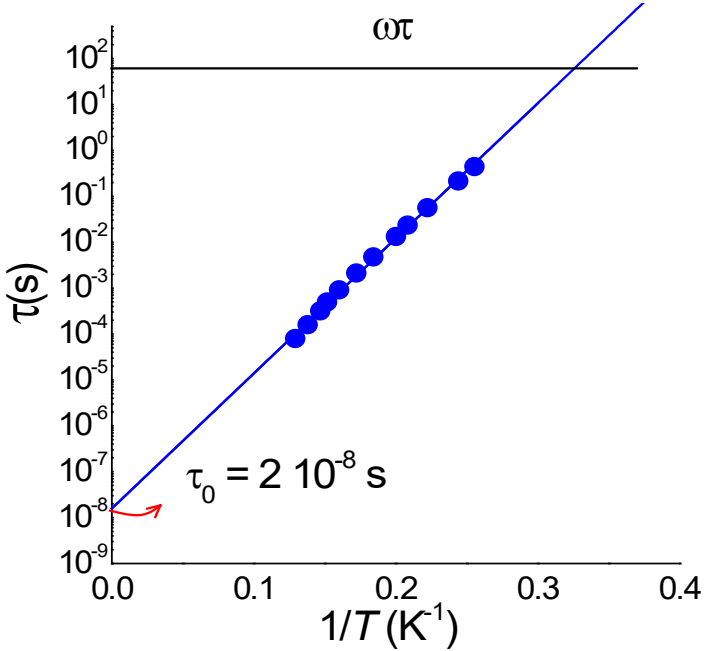
Single-molecule magnets

R. Sessoli et al, Nature **365**, 141 (1993)



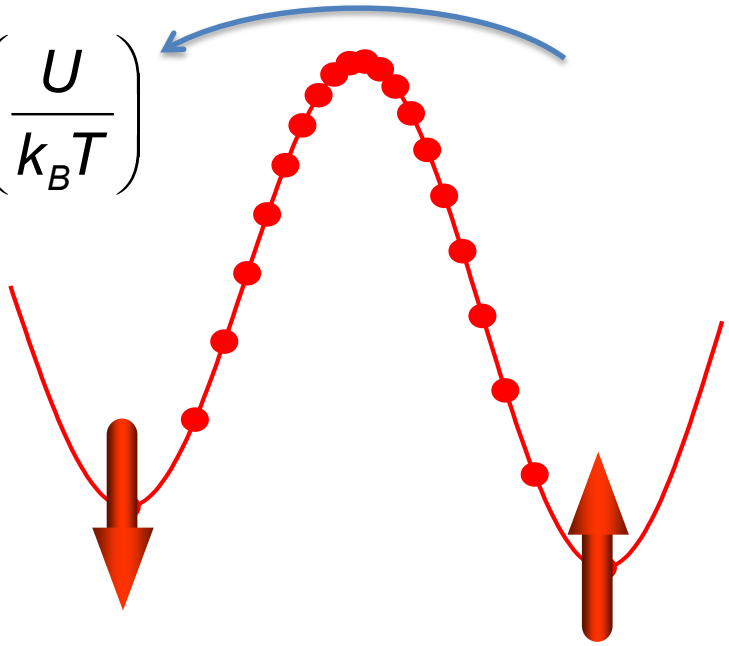
$$\chi_{zz} \equiv \chi'_{zz} - i\chi''_{zz} \simeq \frac{\chi_T}{1 + i\omega\tau}$$

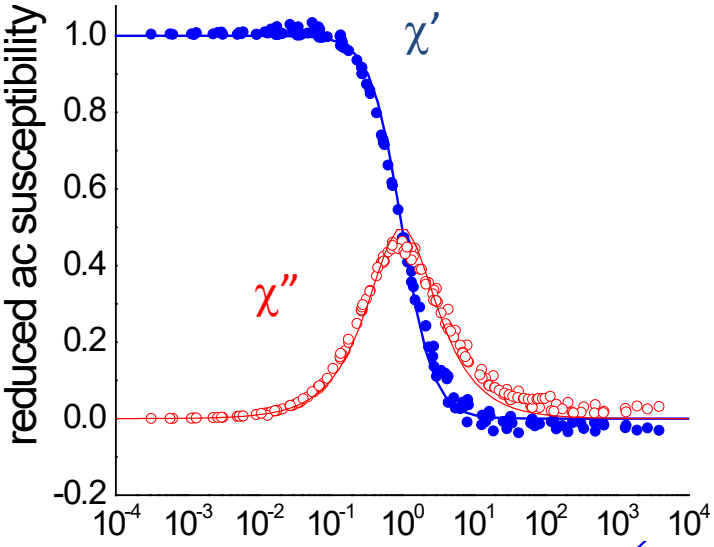


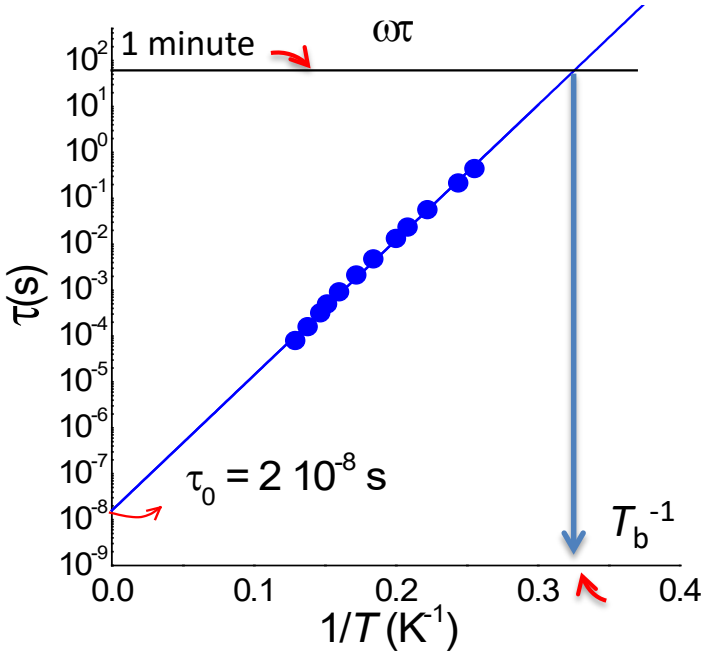


Single-molecule magnets

$$\tau = \tau_0 \exp\left(\frac{U}{k_B T}\right)$$



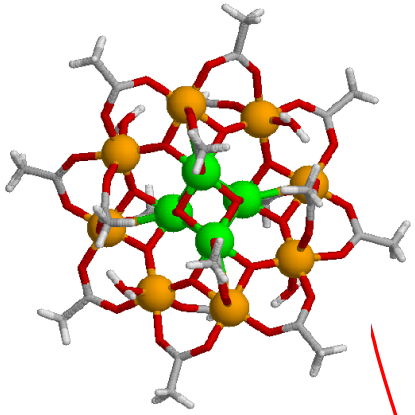


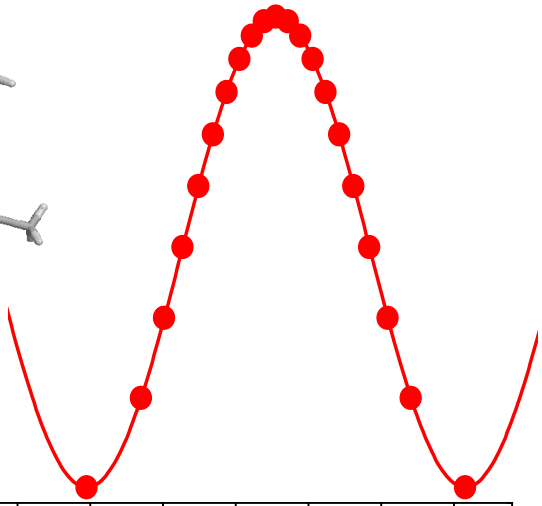


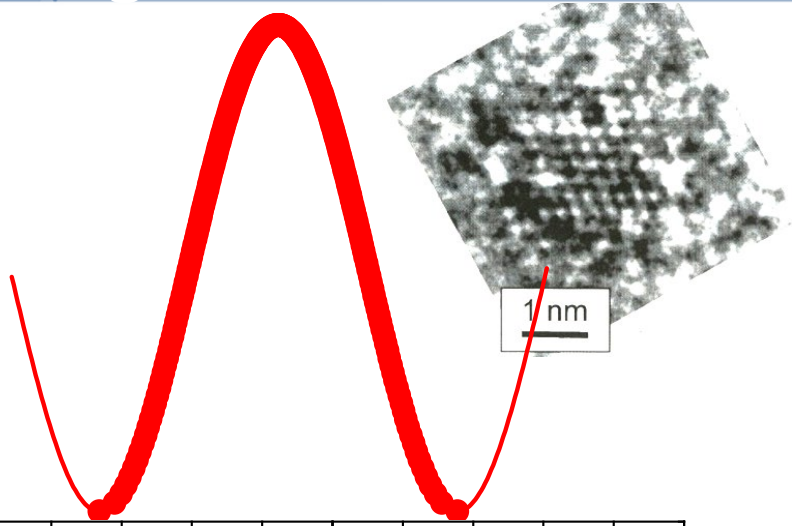
R. Orbach, Proc. Roy. Soc. (London) **A264**, 456 (1961)

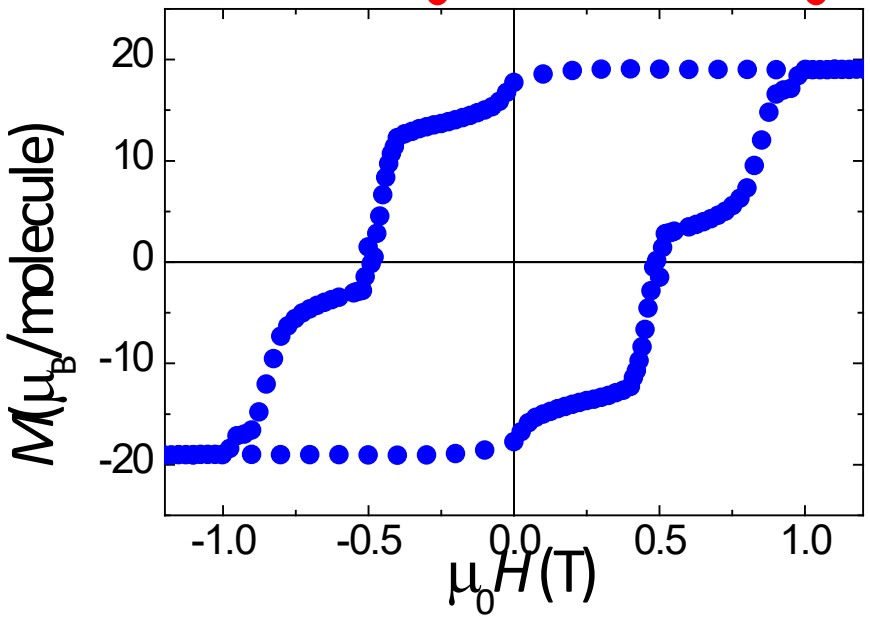
J. Villain, F. Hartman-Boutron, R. Sessoli and A. Rettori, EPL **27**, 159 (1994)

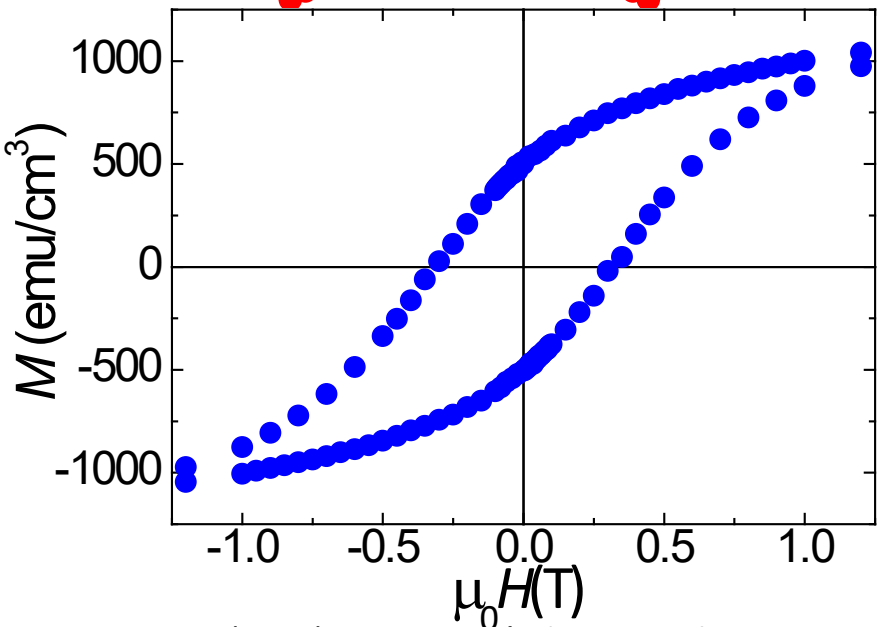
Magnetic memory and spin tunneling





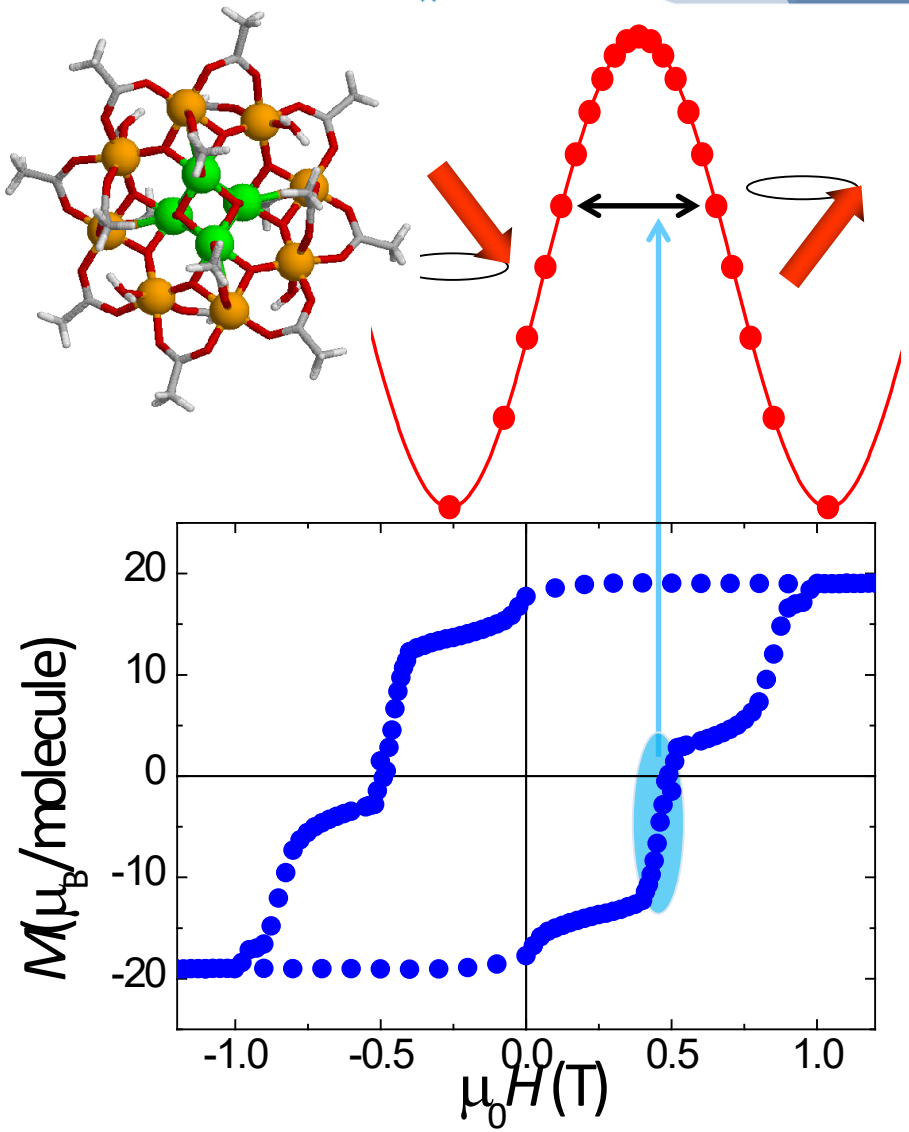


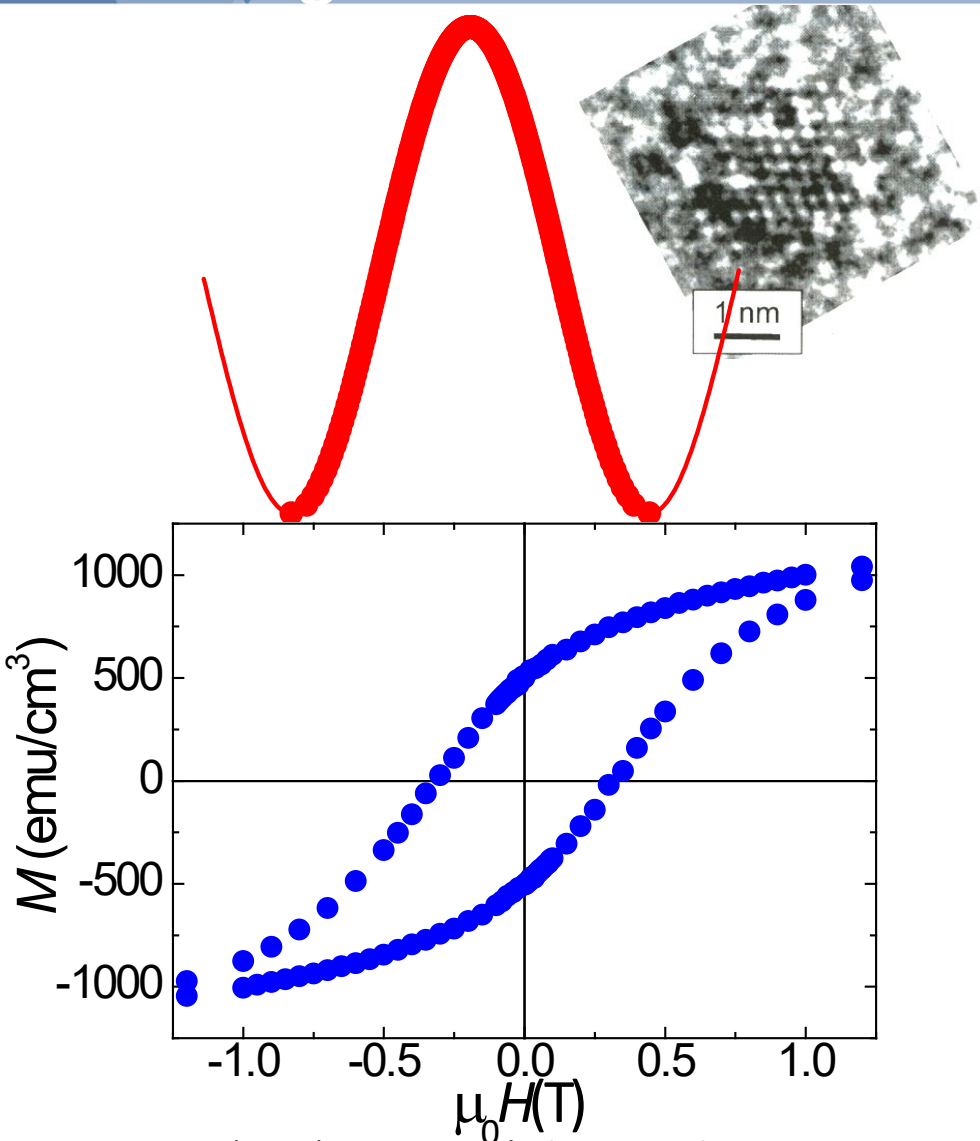




J. R. Friedman, M. P. Sarachik, J. Tejada and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996); J. M. Hernández, X. X. Zhang, F. Luis, J. Bartolomé, J. Tejada and R. Ziolo, Europhys. Lett. **35**, 301 (1996); L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli and B. Barbara, Nature **383**, 145 (1996)

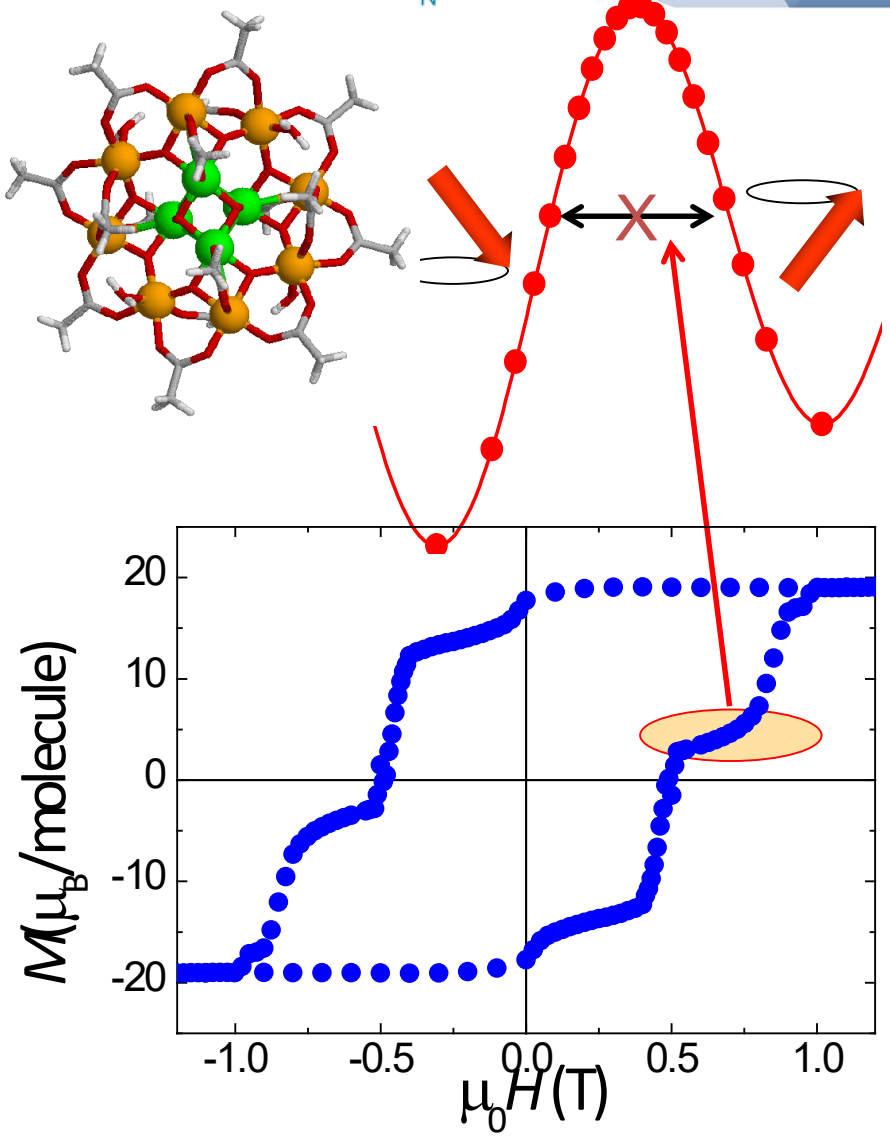
Magnetic memory and spin tunneling

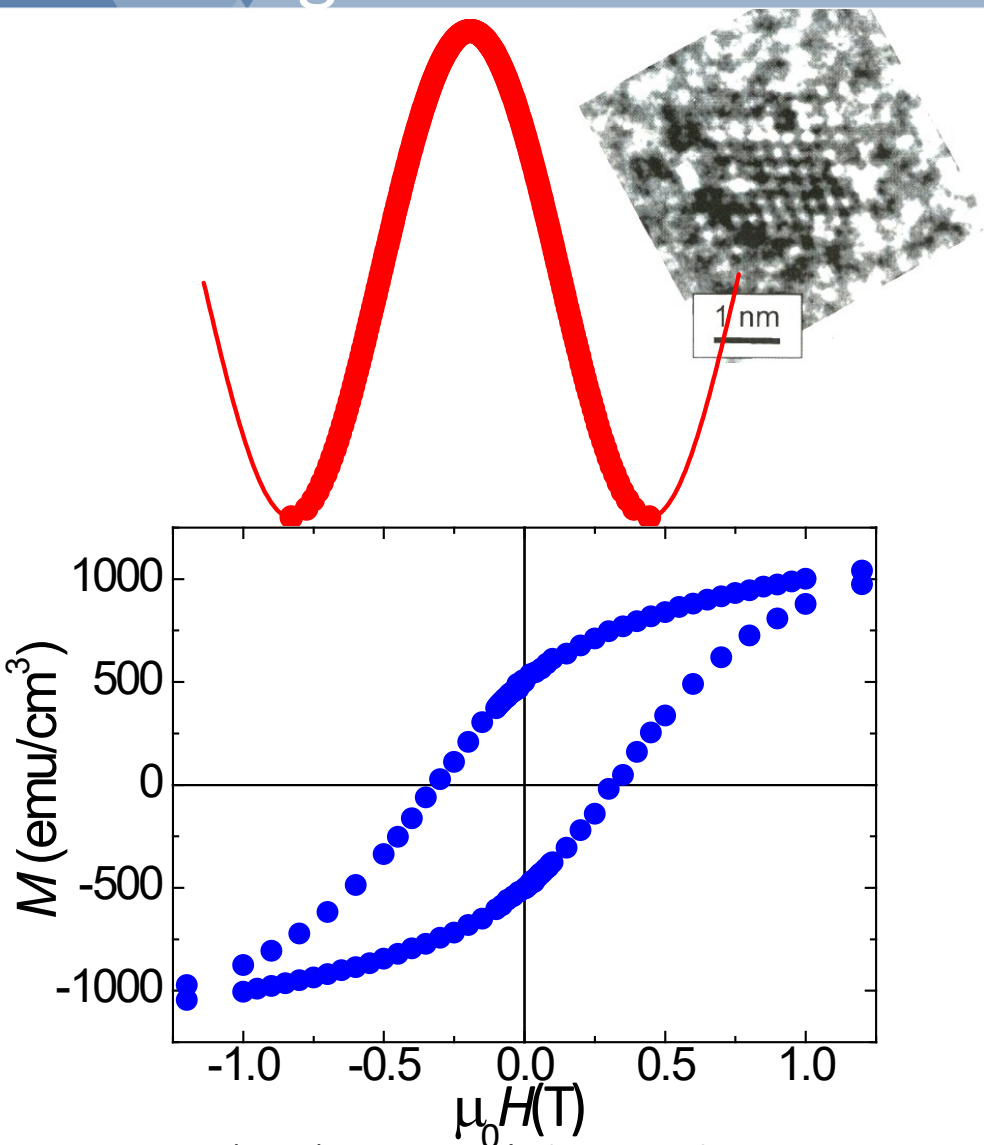




J. R. Friedman, M. P. Sarachik, J. Tejada and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996); J. M. Hernández, X. X. Zhang, F. Luis, J. Bartolomé, J. Tejada and R. Ziolo, Europhys. Lett. **35**, 301 (1996); L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli and B. Barbara, Nature **383**, 145 (1996)

Magnetic memory and spin tunneling

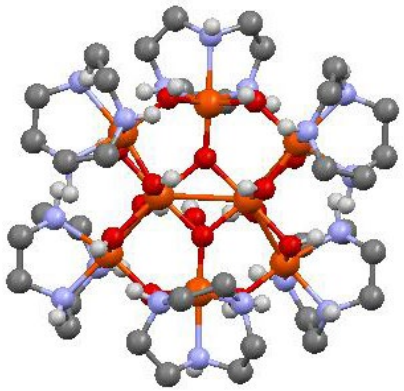




J. R. Friedman, M. P. Sarachik, J. Tejada and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996); J. M. Hernández, X. X. Zhang, F. Luis, J. Bartolomé, J. Tejada and R. Ziolo, Europhys. Lett. **35**, 301 (1996); L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli and B. Barbara, Nature **383**, 145 (1996)

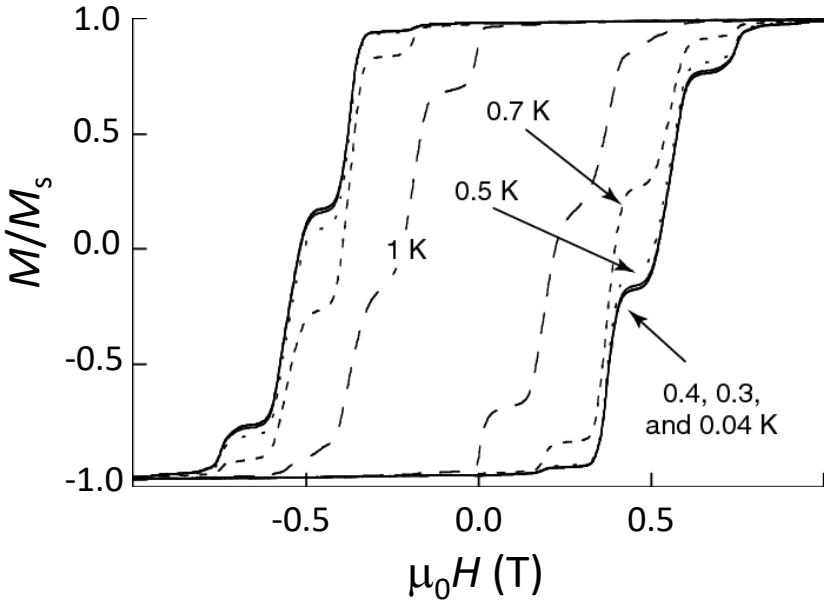
Ground state tunneling Landau-Zener experiments

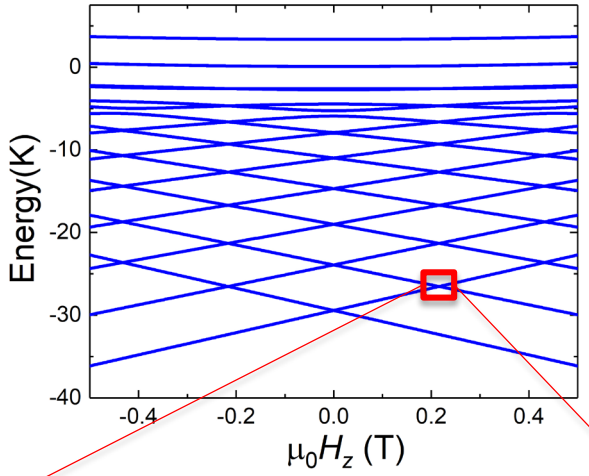
$$\mathcal{H} \approx -DS_z^2 + E\left(S_x^2 - S_y^2\right) - g\mu_B \vec{H}\vec{S}$$

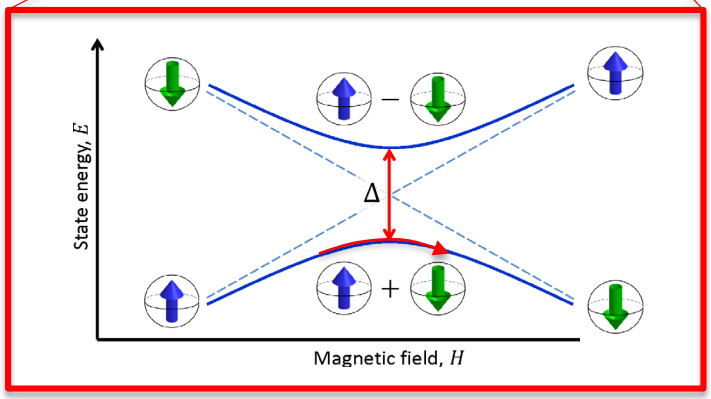


Fe_8

S = 10



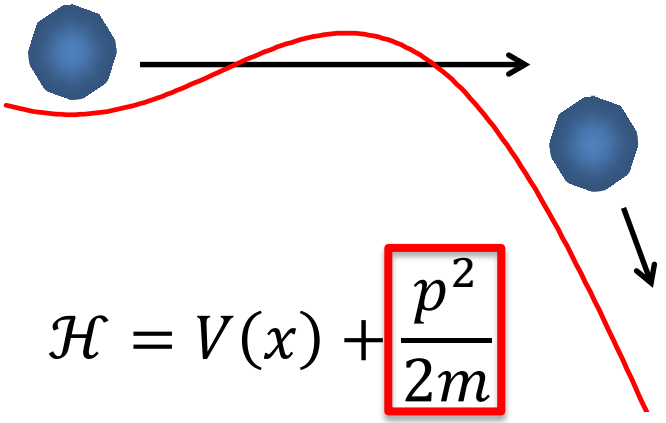




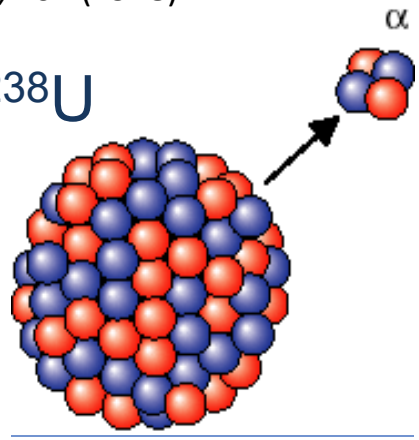
$$P_{LZ} = 1 - exp\left(-\pi\Delta^2 \Big/ 4\hbar g\mu_B \frac{dH}{dt}\right)$$

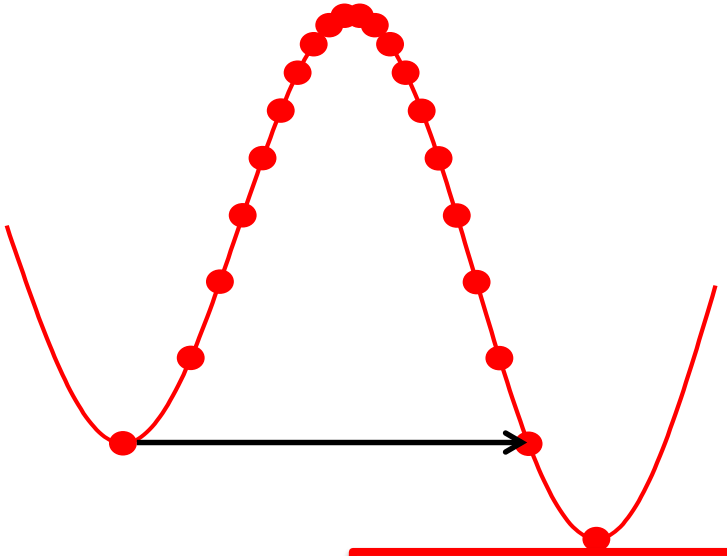
Sangregorio C., Ohm T., Paulsen C, Sessoli R. and Gatteschi D., *Phys. Rev. Lett.*, **78** 4645 (1997) ; W. Wernsdorfer *et al*, Europhys. Lett. **50** (4), 552 (2000)

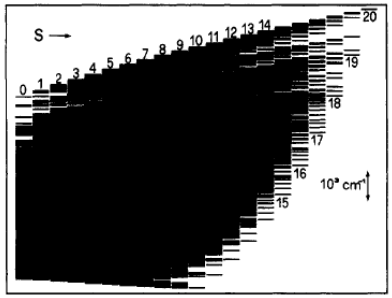
G. Gamow, Z. Physik **51**, 204 (1928)

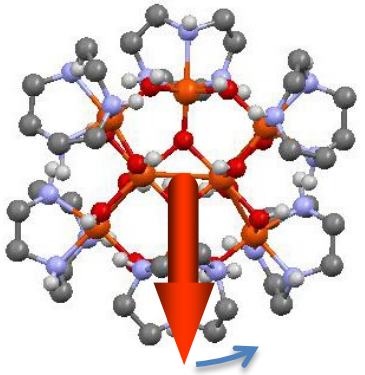


$$\mathcal{H} = V(x) + \frac{p^2}{2m}$$



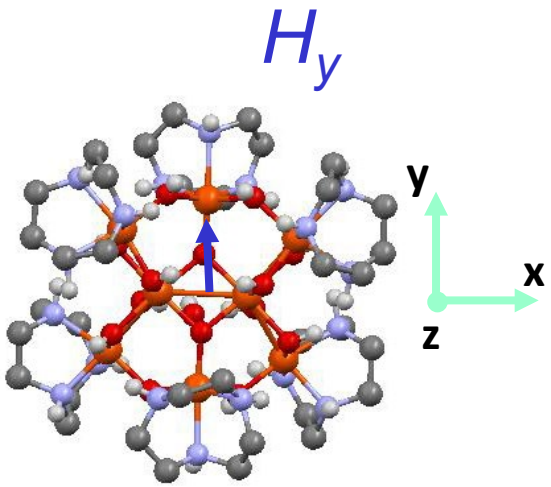


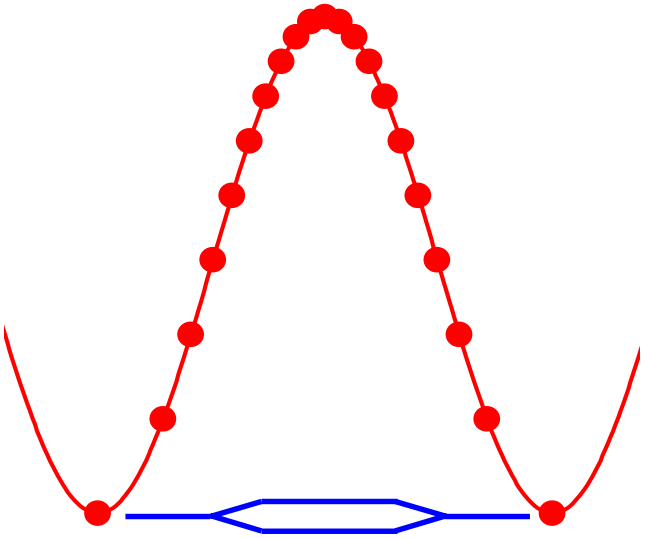


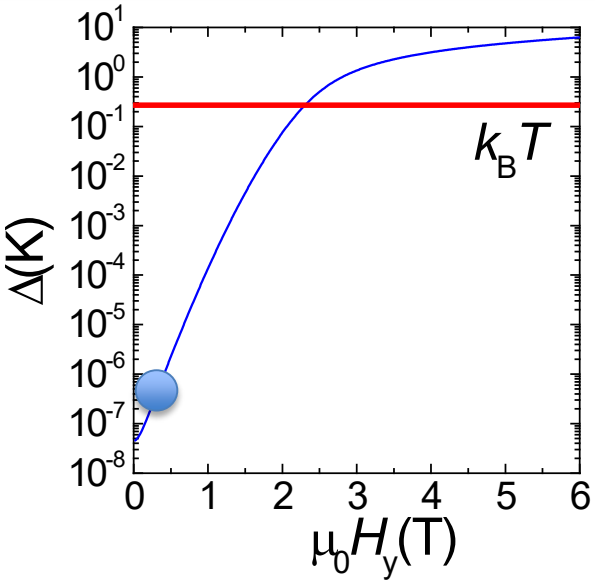


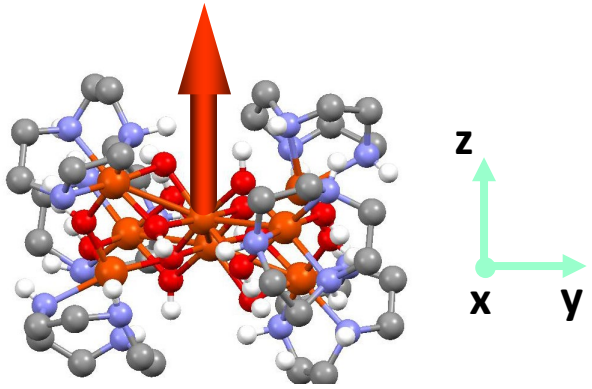
$$\mathcal{H} = -DS_z^2 + E\left(S_x^2 - S_y^2\right) - g\mu_B\left(H_xS_x + H_yS_y\right)$$

A magnetic Schrödinger cat





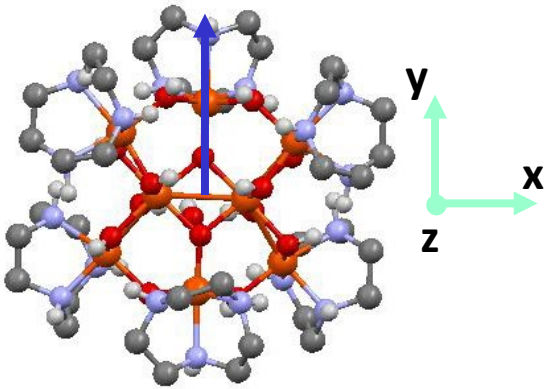


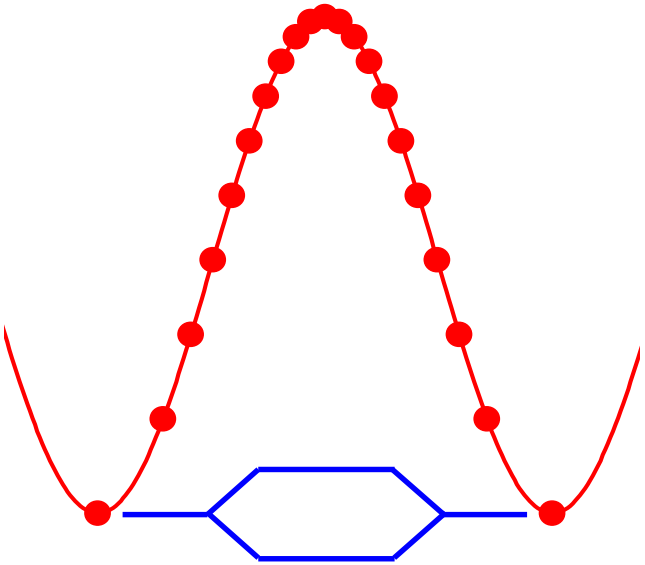


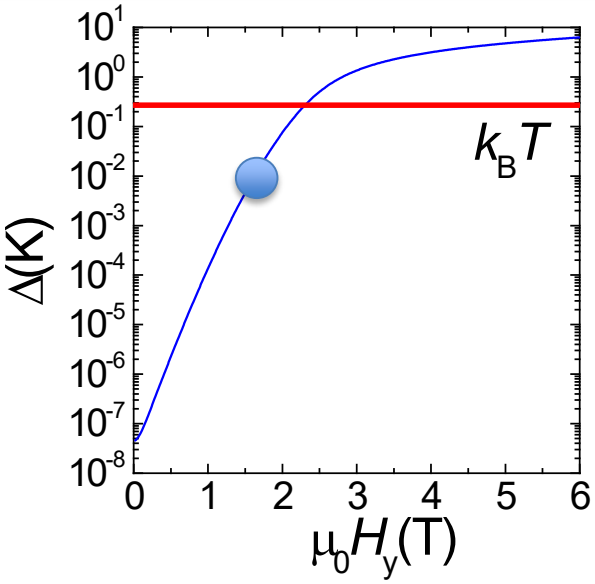
F. Luis, F. L. Mettes, J. Tejada, D. Gatteschi, and L. J. de Jongh, Phys. Rev. Lett. **85**, 4377 (2000)

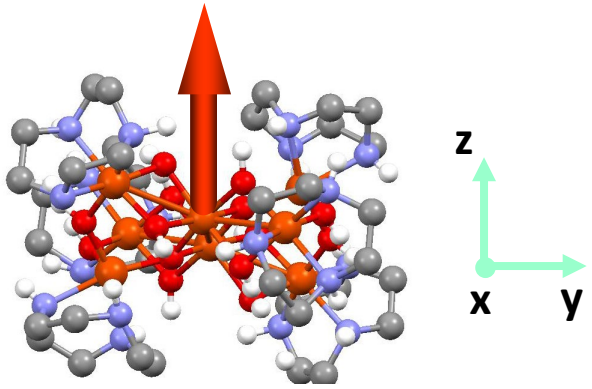
A magnetic Schrödinger cat

H_y



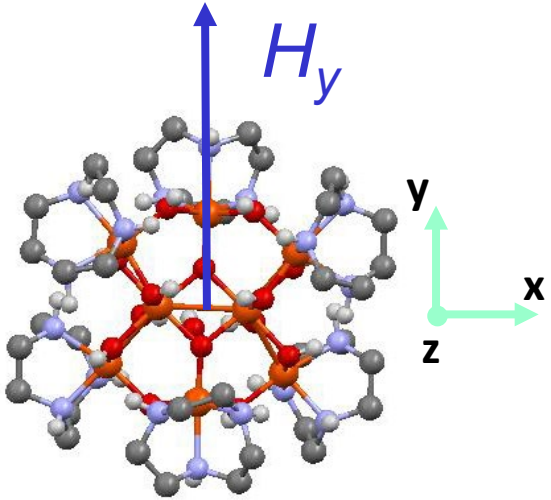


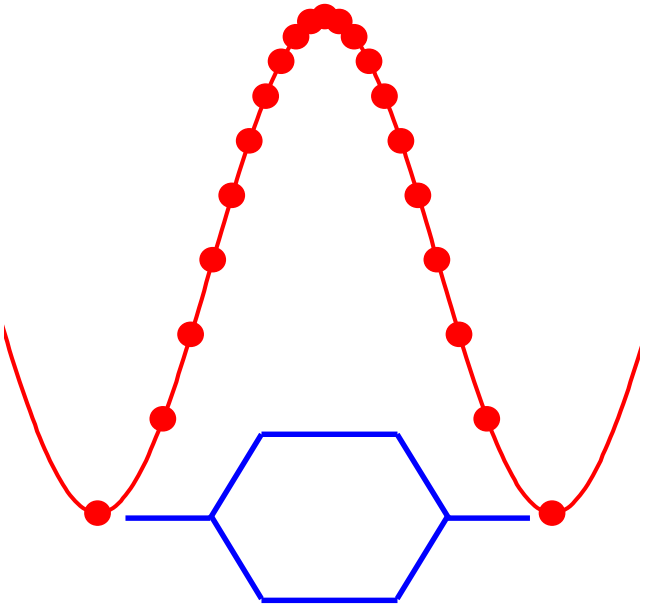


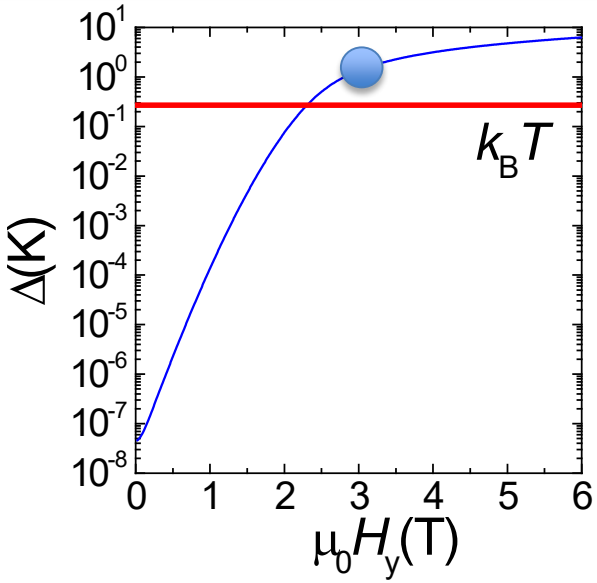


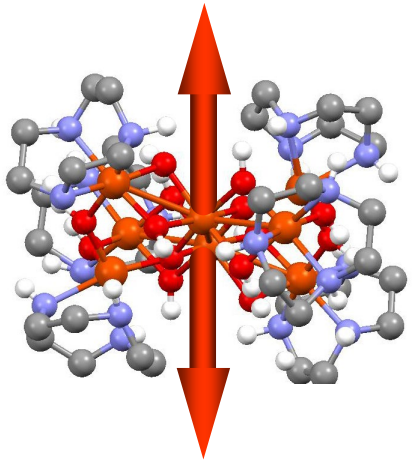
F. Luis, F. L. Mettes, J. Tejada, D. Gatteschi, and L. J. de Jongh, Phys. Rev. Lett. **85**, 4377 (2000)

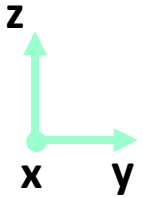
A magnetic Schrödinger cat



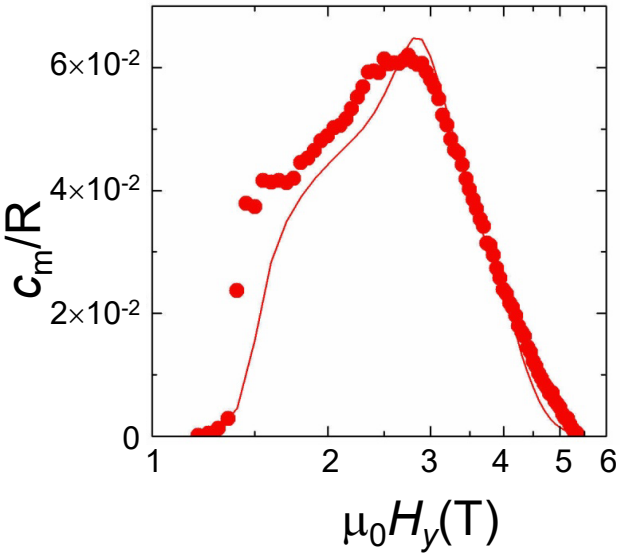








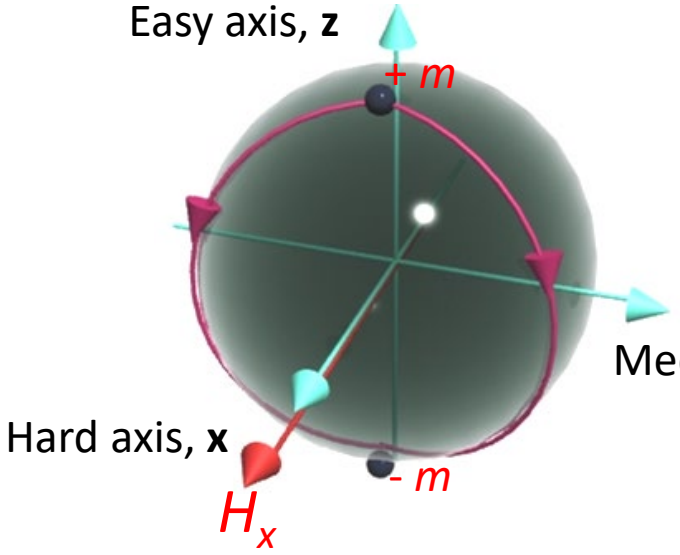
$$\frac{c_m}{R} = \left(\frac{\Delta/k_B T}{e^{\Delta/2k_B T} + e^{-\Delta/2k_B T}} \right)^2$$

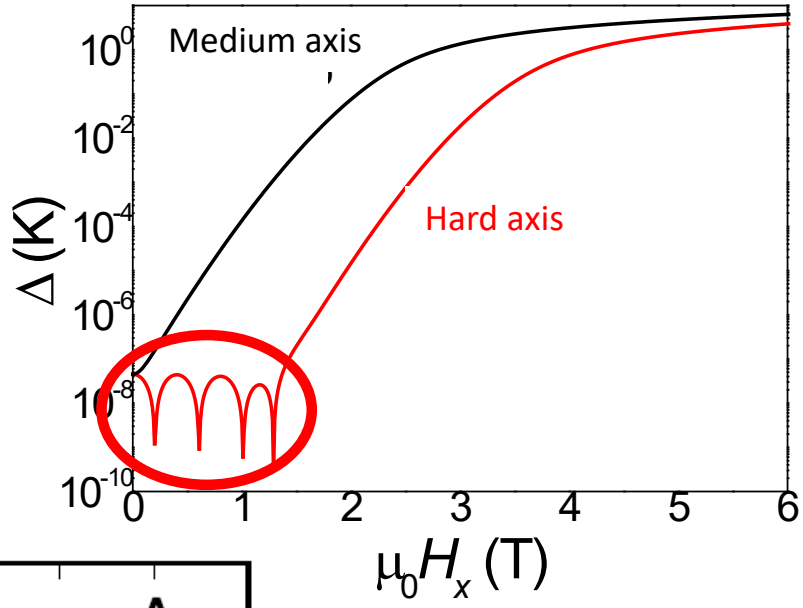


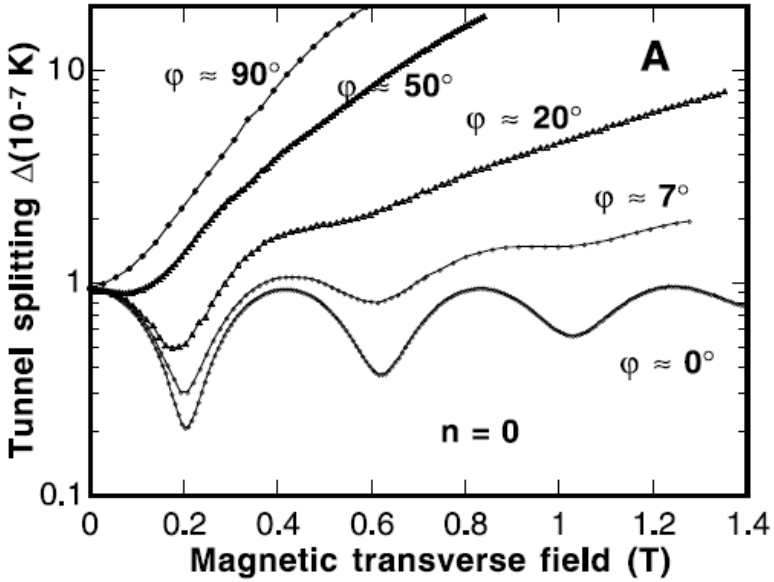
Robust superposition states

F. Luis, F. L. Mettes, J. Tejada, D. Gatteschi, and L. J. de Jongh, Phys. Rev. Lett. **85**, 4377 (2000)

Quantum spin interference

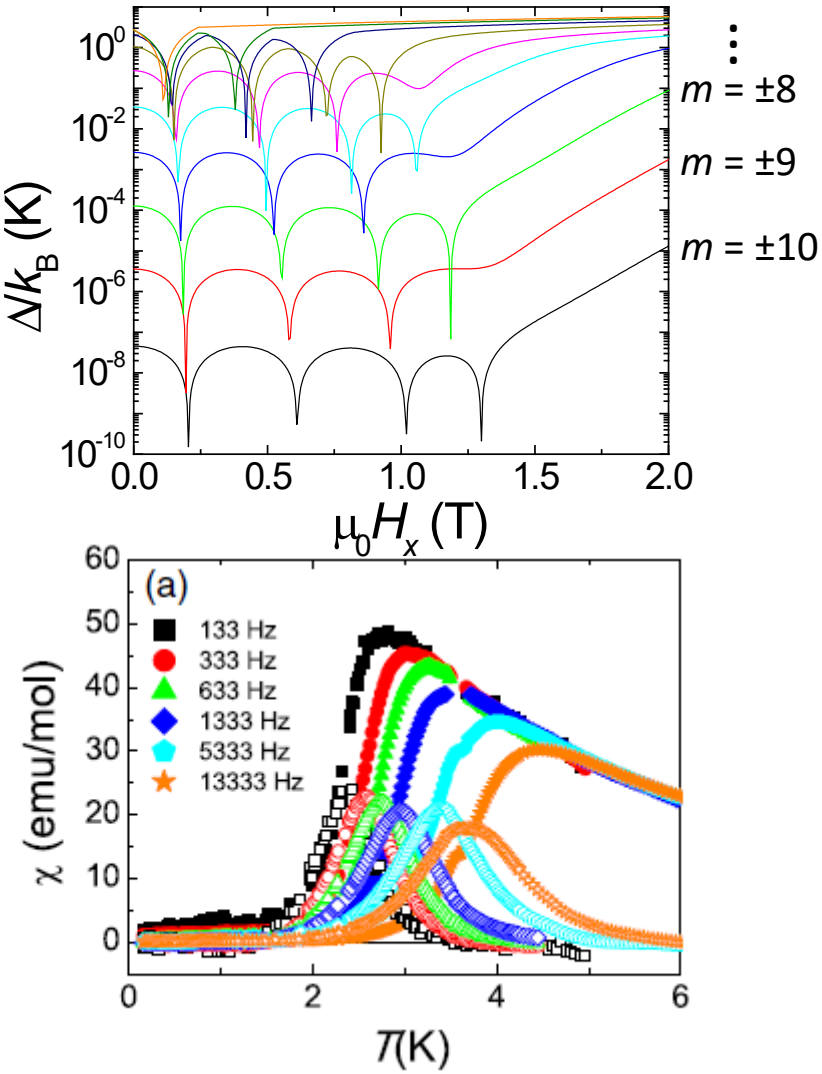






W. Wernsdorfer and R. Sessoli , Science **284**, 133 (1999)

Effect on superparamagnetic blocking & magnetic memory



E. Burzurí *et al*, Phys. Rev. Lett. **111**, 057201 (2013)

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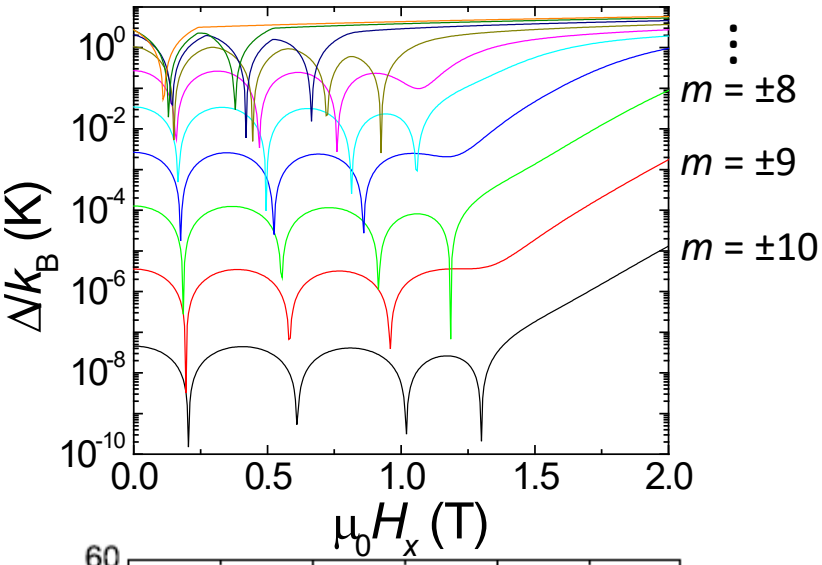
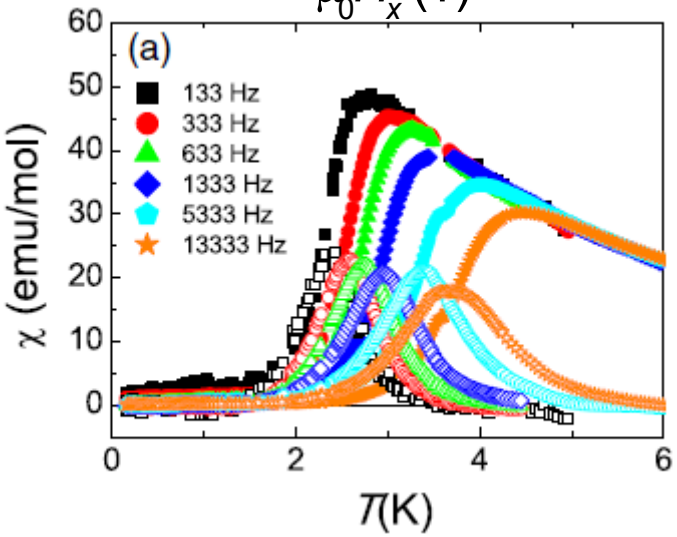
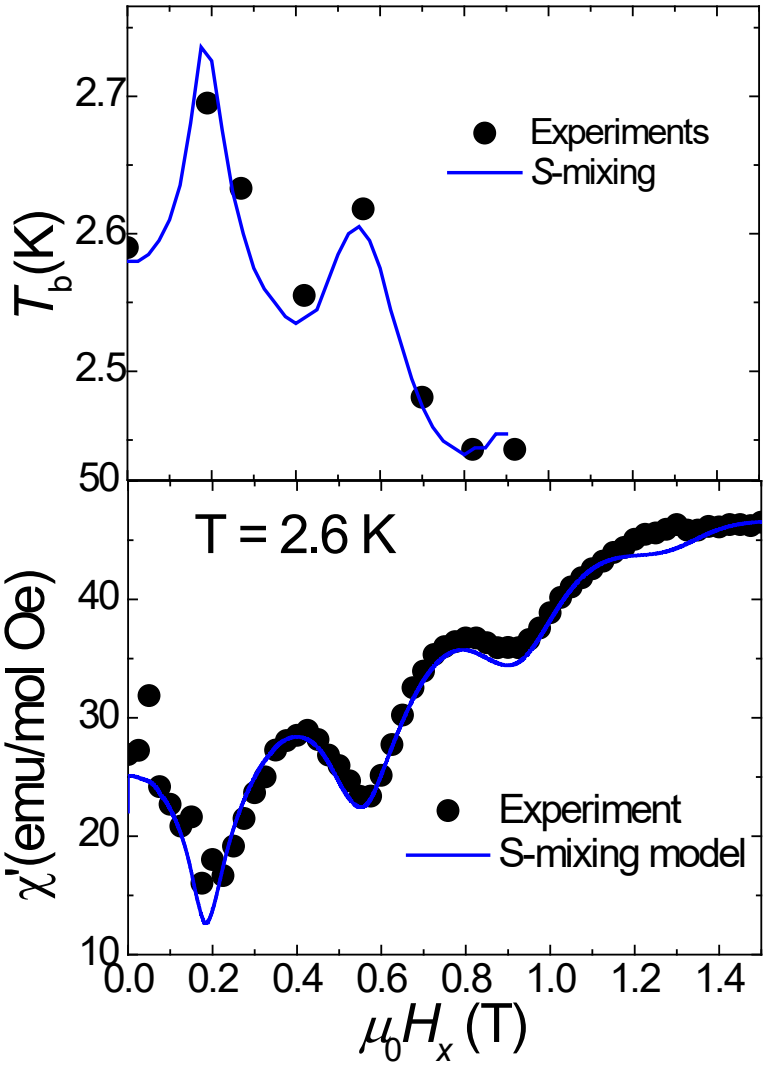
Effect on superparamagnetic blocking & magnetic memory

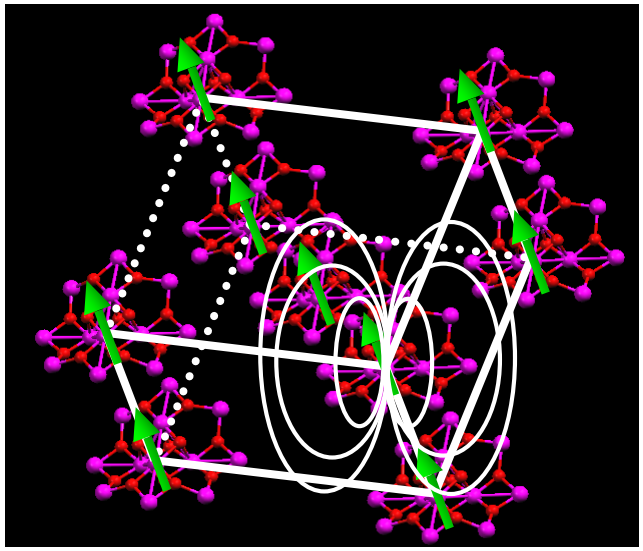






E. Burzurí *et al*, Phys. Rev. Lett. **111**, 057201 (2013)

Pure dipolar magnetic order

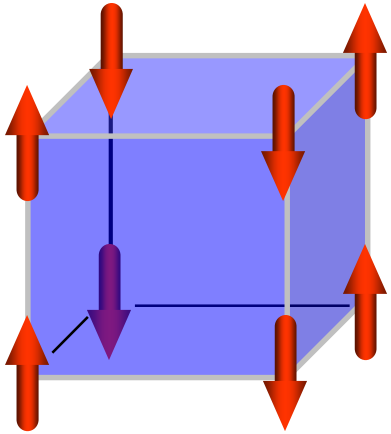


$$\overrightarrow{B_{dip}}(\vec{r}) = \frac{\vec{\mu}}{r^3} - \frac{3(\vec{\mu}\vec{r})}{r^5}$$

Ground state can be predicted from symmetry considerations

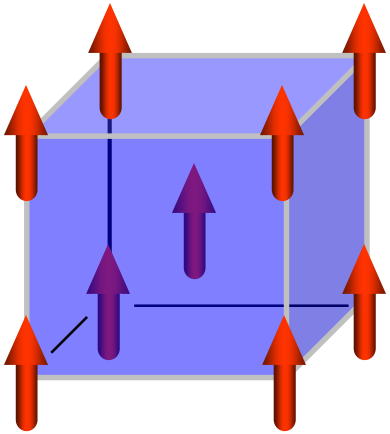
J. M. Luttinger and L. Tisza, Phys. Rev. **70**, 954 (1946)

AF



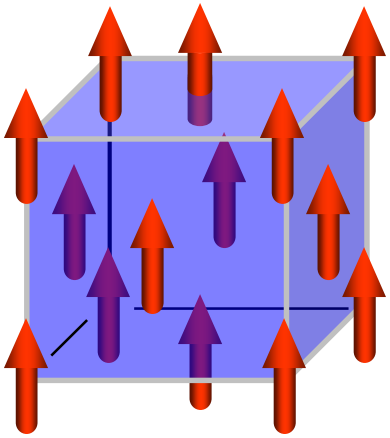
Simple cubic, tetragonal, hexagonal

F



Body centered lattices

F

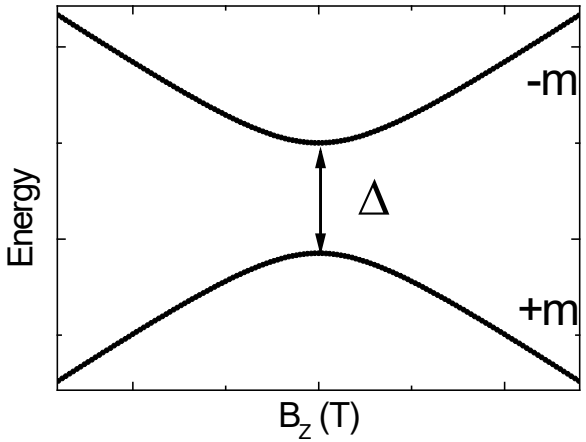


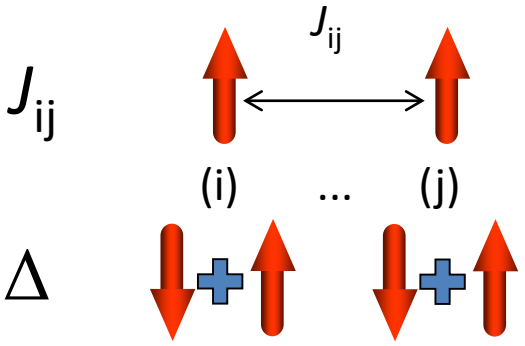
Face centered lattices

Field-driven quantum phase transitions

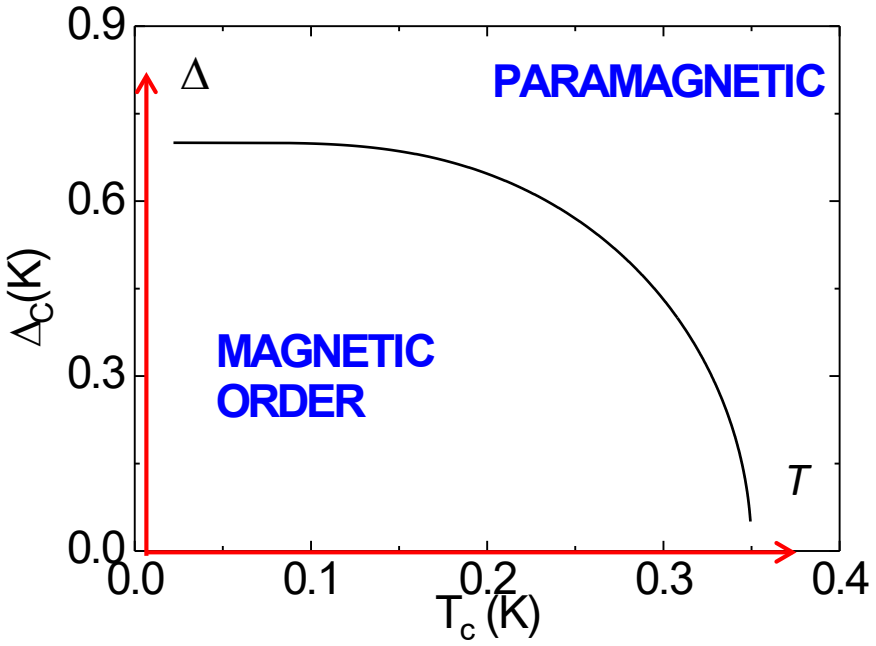
Ising model under transverse magnetic field

$$\mathcal{H} = 2S^2 \sum_{ij} J_{ij}\sigma_{iz}\sigma_{jz} - \Delta \sum_{i} \sigma_{ix}$$



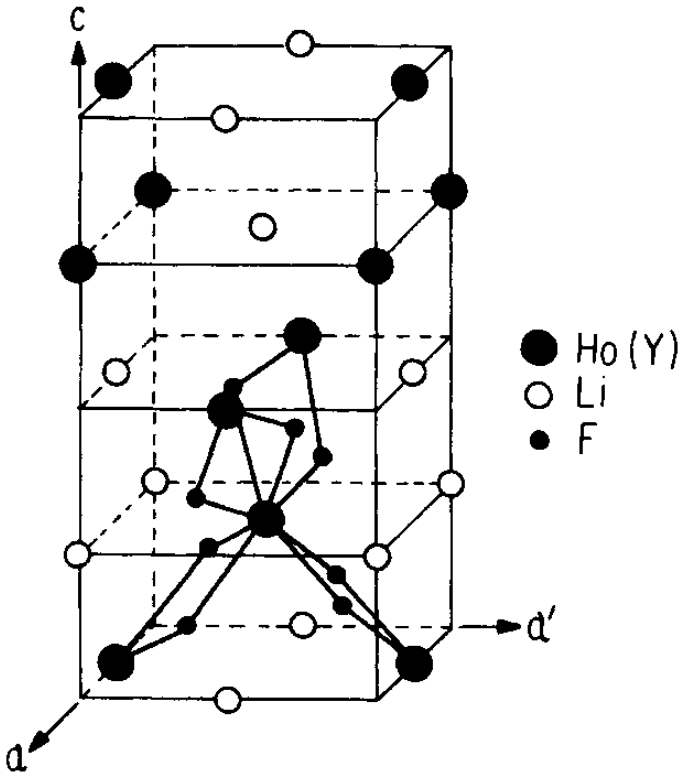


$$\coth\left(\frac{\Delta}{2kT_c}\right) = \frac{J}{\Delta}$$

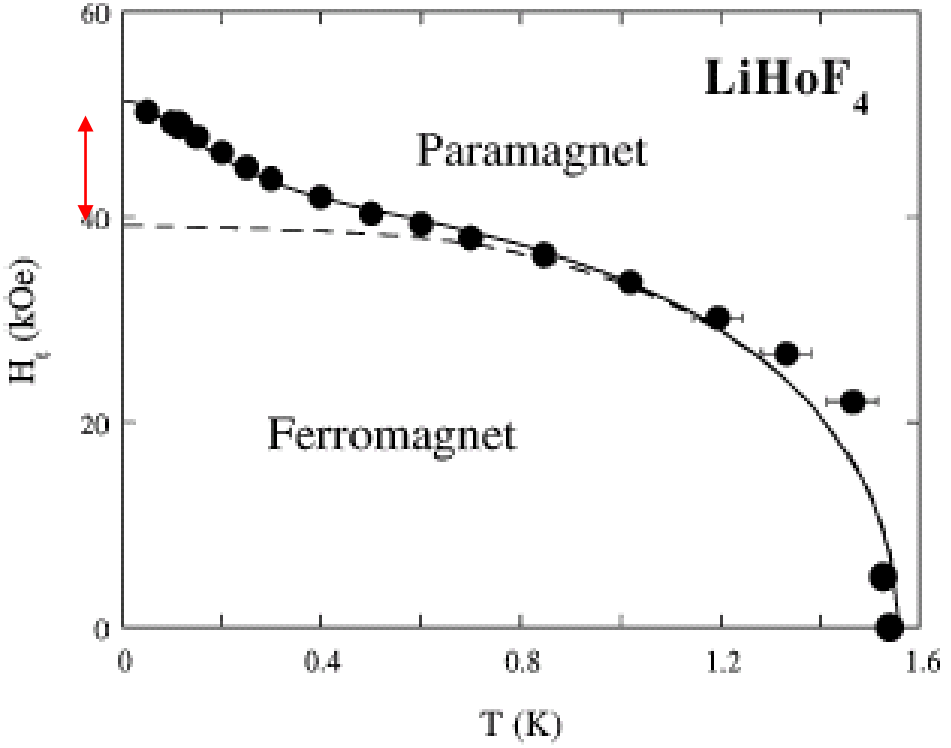


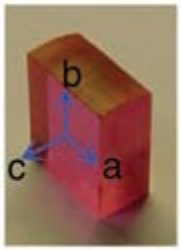
R. B. Stinchcombe, J. Phys. C **6**, 2459 (1973); S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, England, 1999)

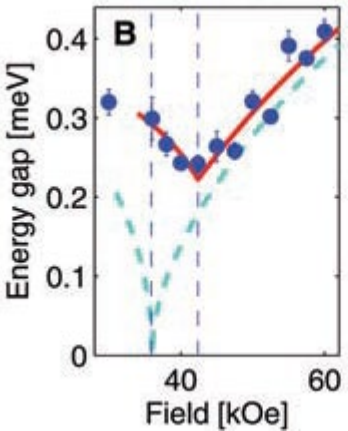
Lanthanide-based salts

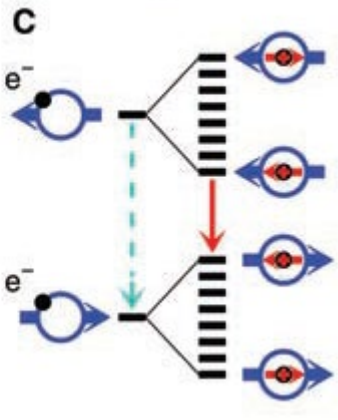


D.Bitko,T.F. Rosenbaum, G. Aeppli, PRL **77**,940 (1996)









- Significant exchange interactions
- High hyperfine interactions

H. M Rønnow et al, Science **308**, 389 (2005)

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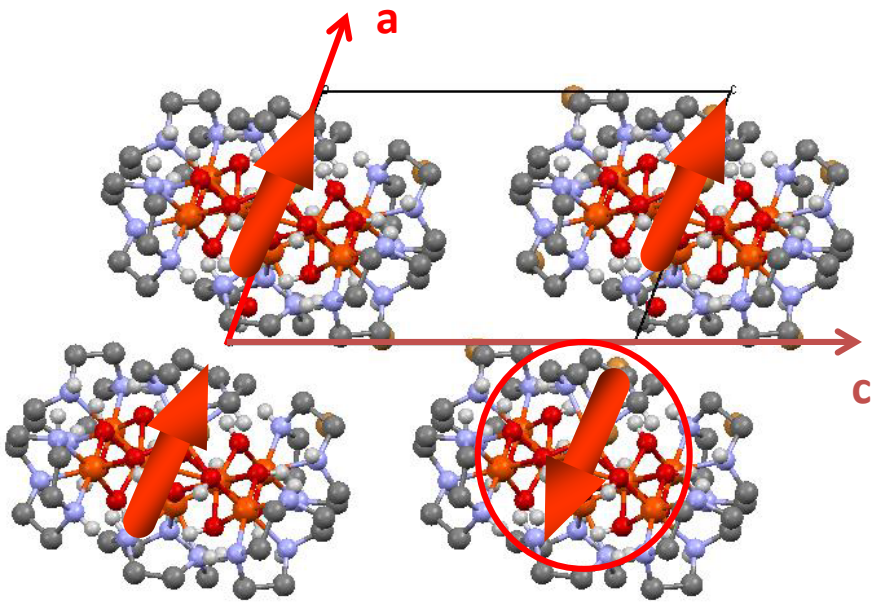
Dipolar ferromagnetic order in Fe_8



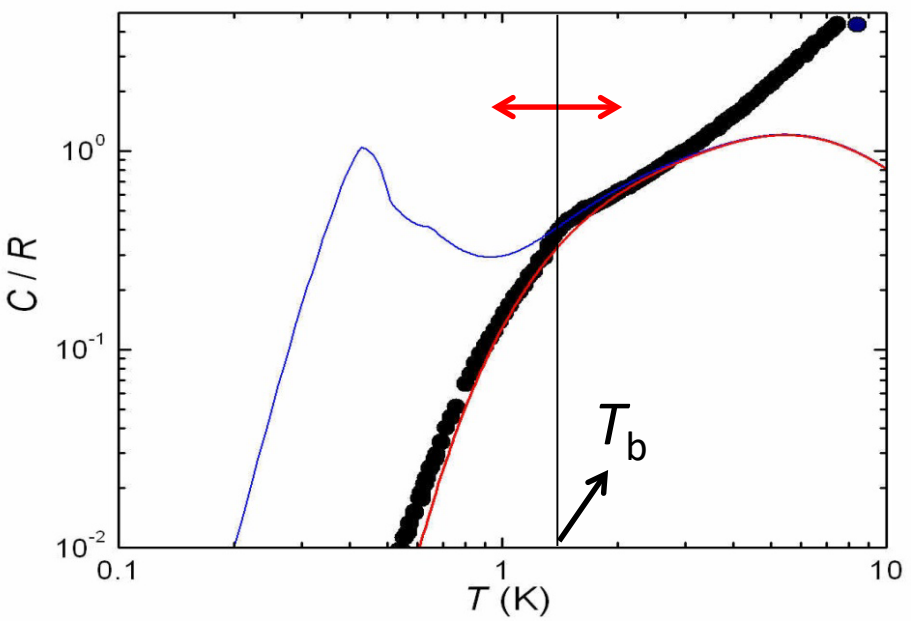
Monte Carlo Simulations

Ferromagnetic ground state

$T_C \simeq 540$ mK

J.F. Fernández et al, PRB **62**, 55 (2000)
X. Martínez-Hidalgo, E. M. Chudnovsky, and A. Aharony, Europhys. Lett. **55**, 273 (2001)

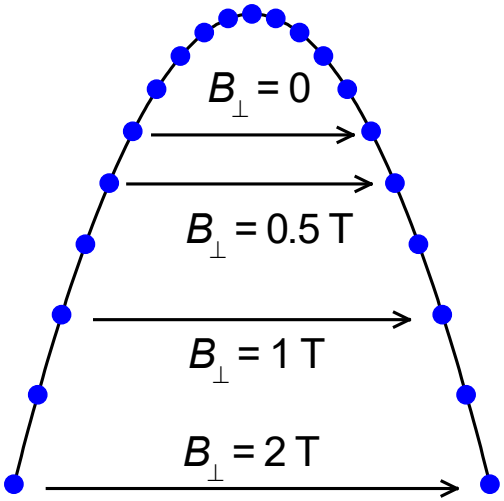
Off-equilibrium equilibrium

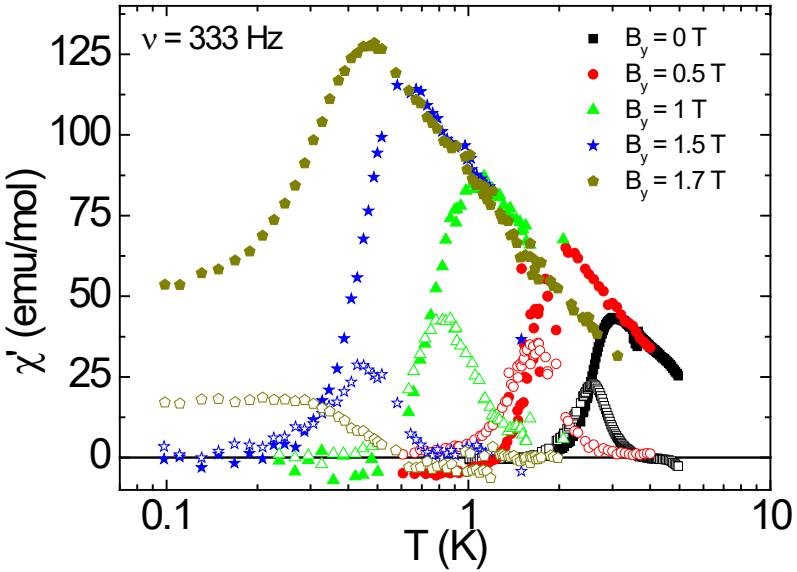


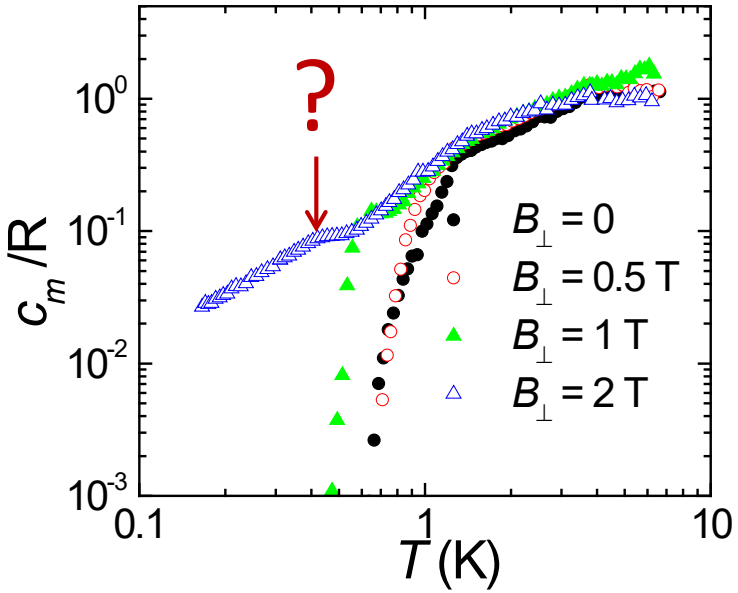
High anisotropy barriers: $U \approx 20$ K

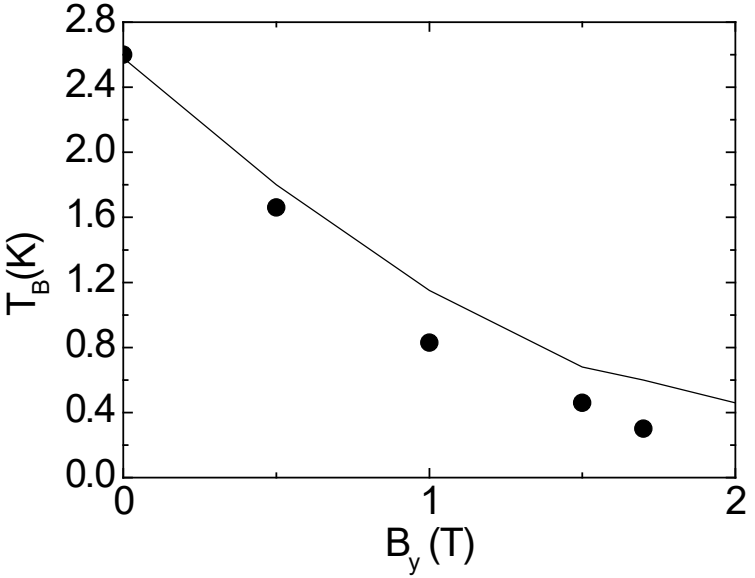
No equilibrium for $T_b \approx U/12 > T_c$

Quantum annealing

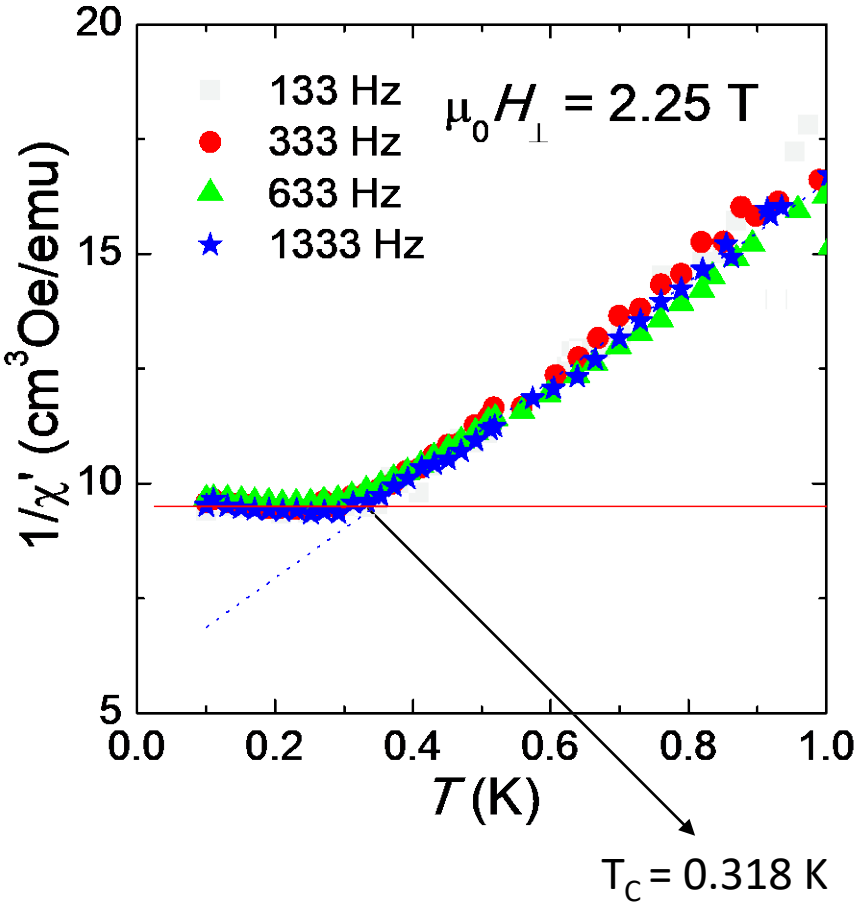




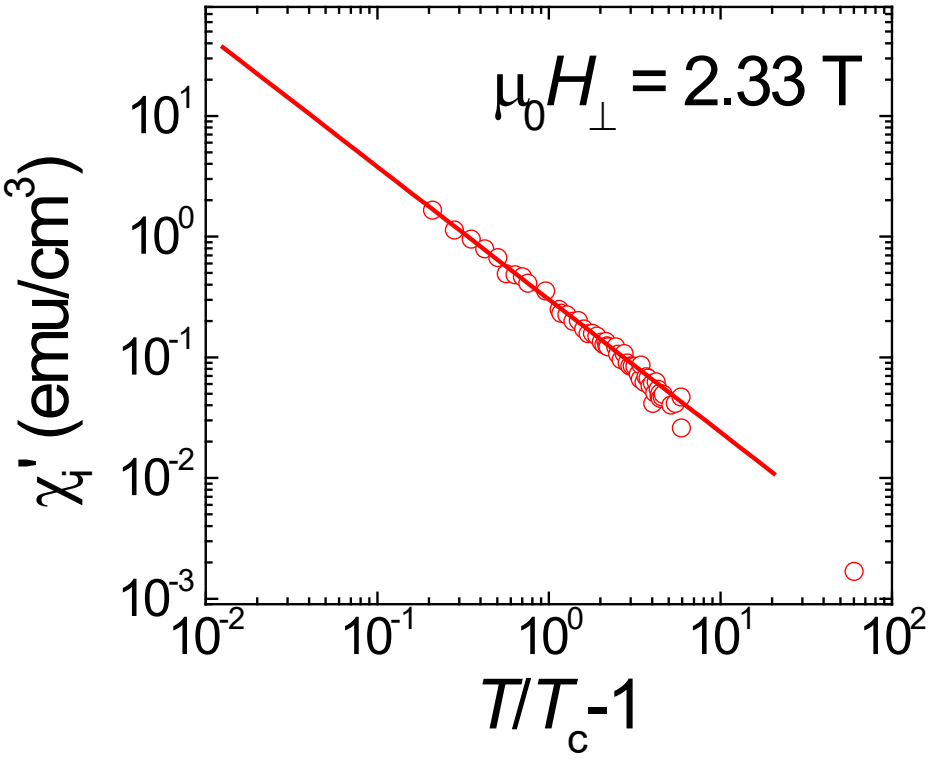




Equilibrium magnetic order



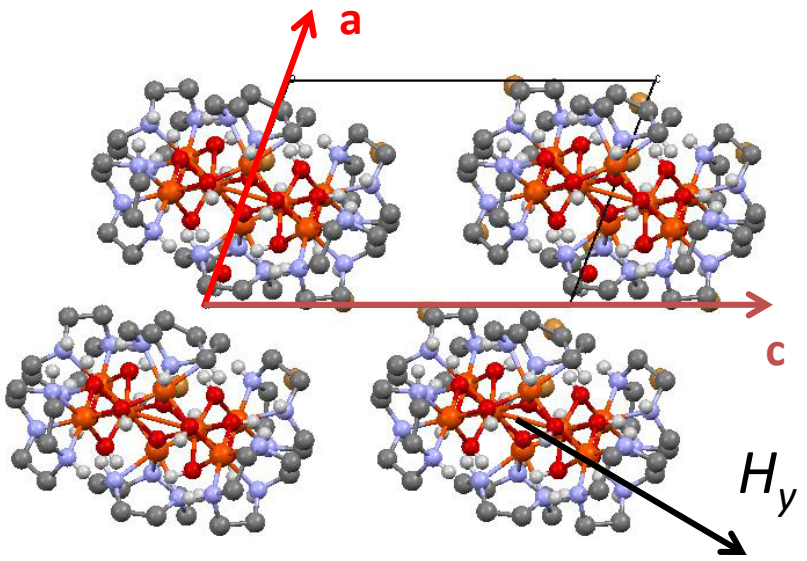
$$\chi_i \propto \left| T/T_C - 1 \right|^{-\gamma}$$

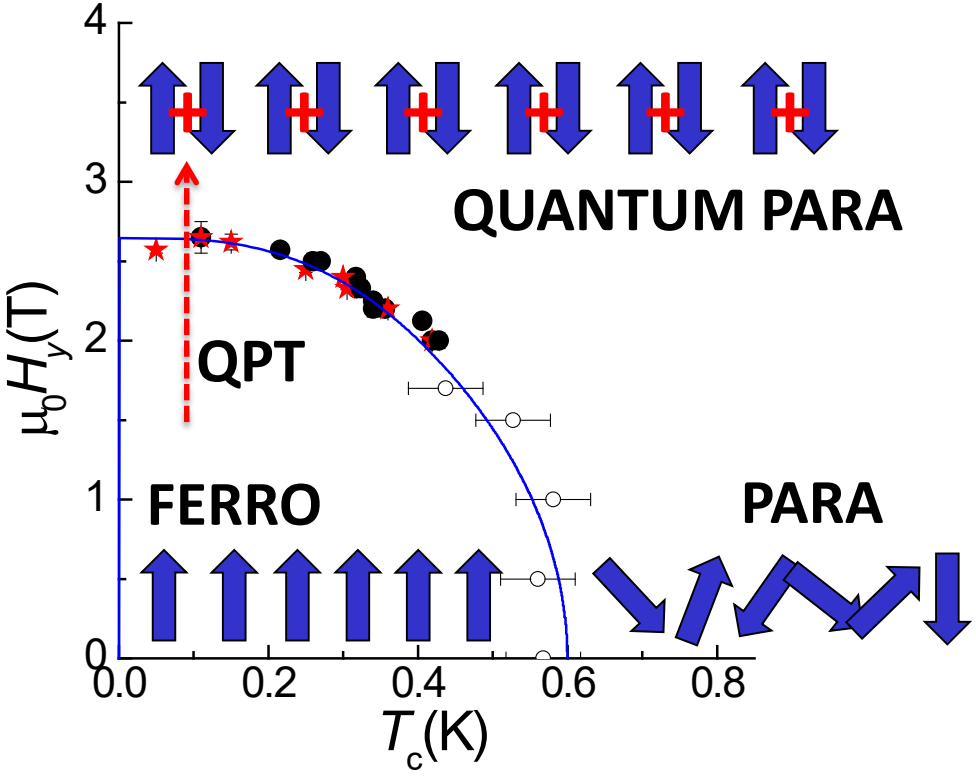


$$\frac{1}{\chi_{max}} \approx N$$ → **FERRO**

$$\gamma \sim 1$$ → **Mean field dipolar ferromagnet**

Quantum phase transition





Model realization of the quantum Ising model

$$\mathcal{H} = 2S^2 \sum_{ij} J_{ij} \sigma_{iz} \sigma_{jz} - \Delta \sum_{i} \sigma_{ix}$$

E. Burzurí, F. Luis, B. Barbara, R. Ballou, E. Ressouche, O. Montero, J. Campo, and S. Maegawa, Phys. Rev. Lett. **107**, 097203 (2011)

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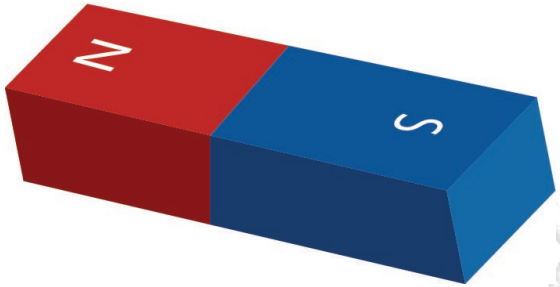
Outline

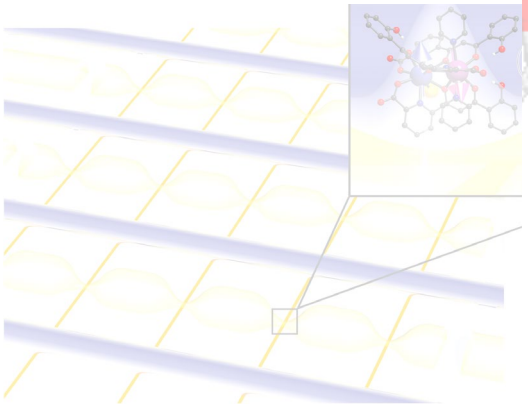


2. Magnets

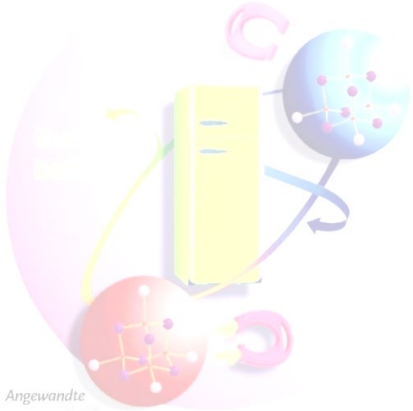
3. Magnetic recording

1. Physics

4. Quantum information



5. Magnetic coolers



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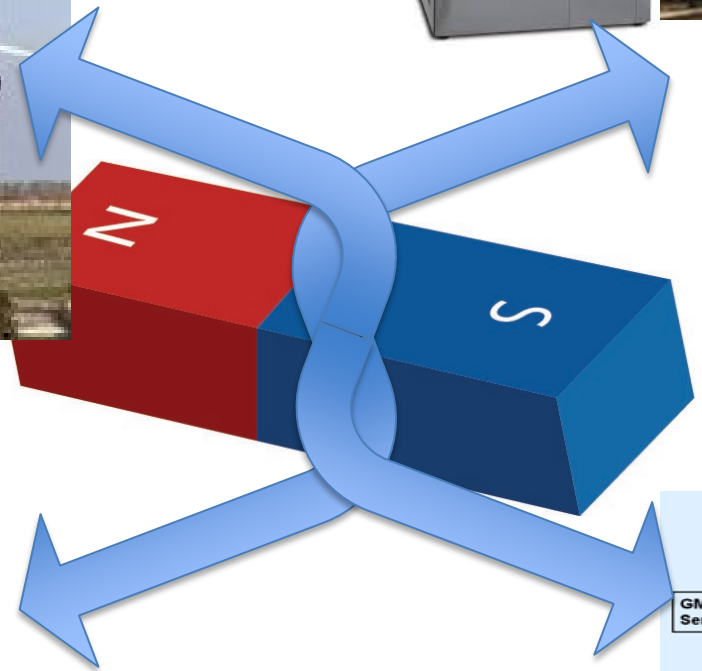
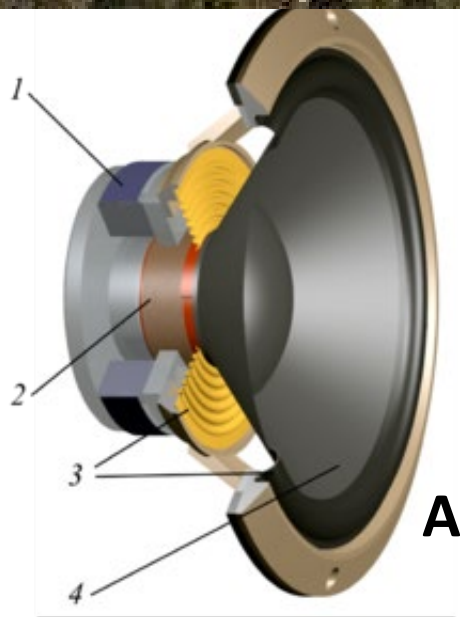
Magnets are everywhere

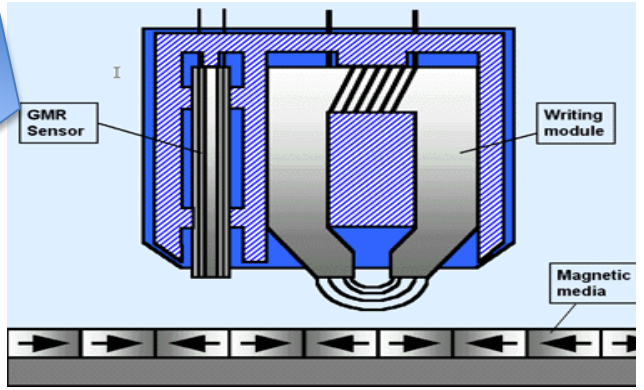


Motors



Actuators

Data storage



1 0 1 0 1 0 0…

Figure I
Development of Permanent Magnets in the 1900's

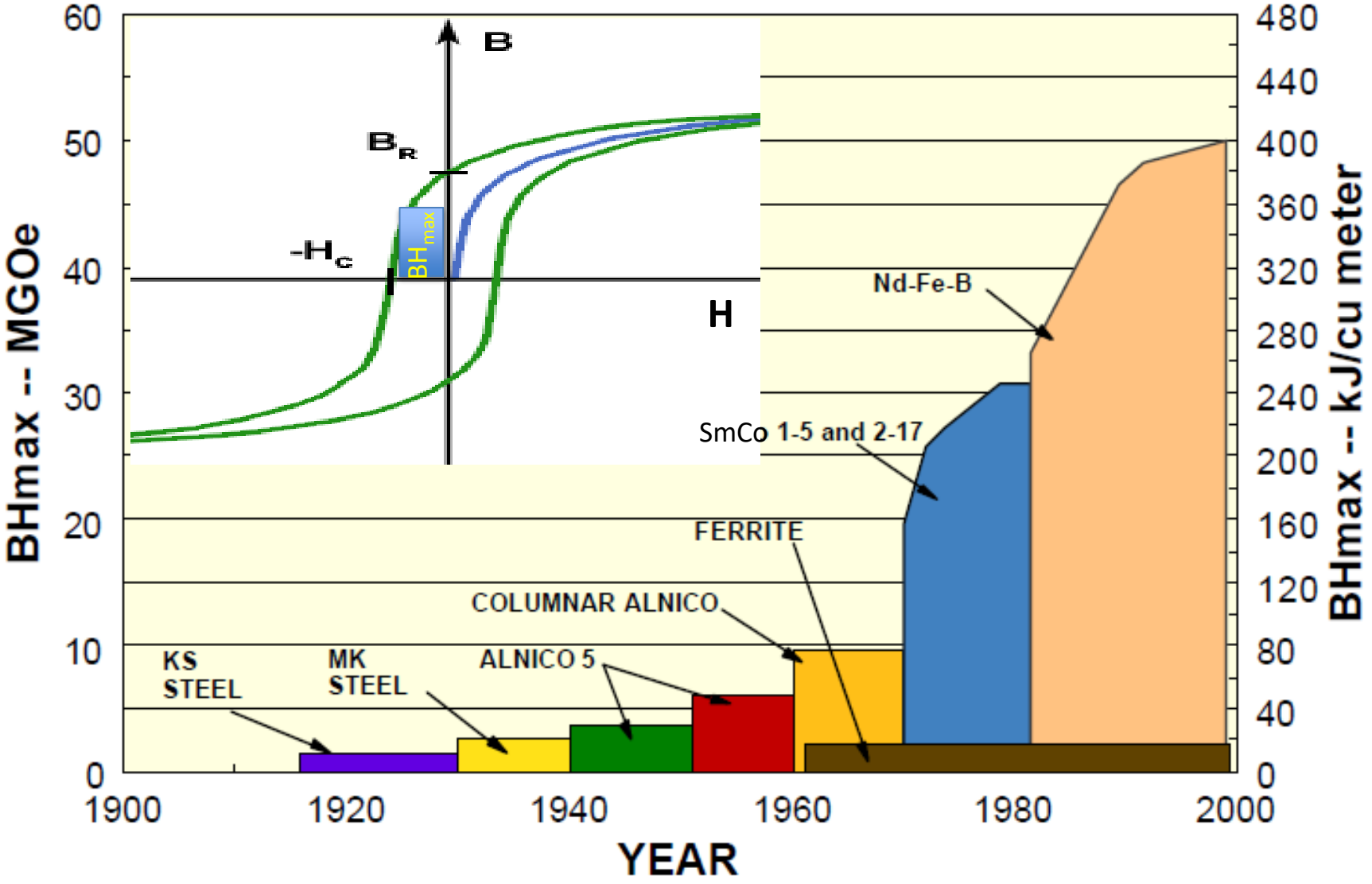
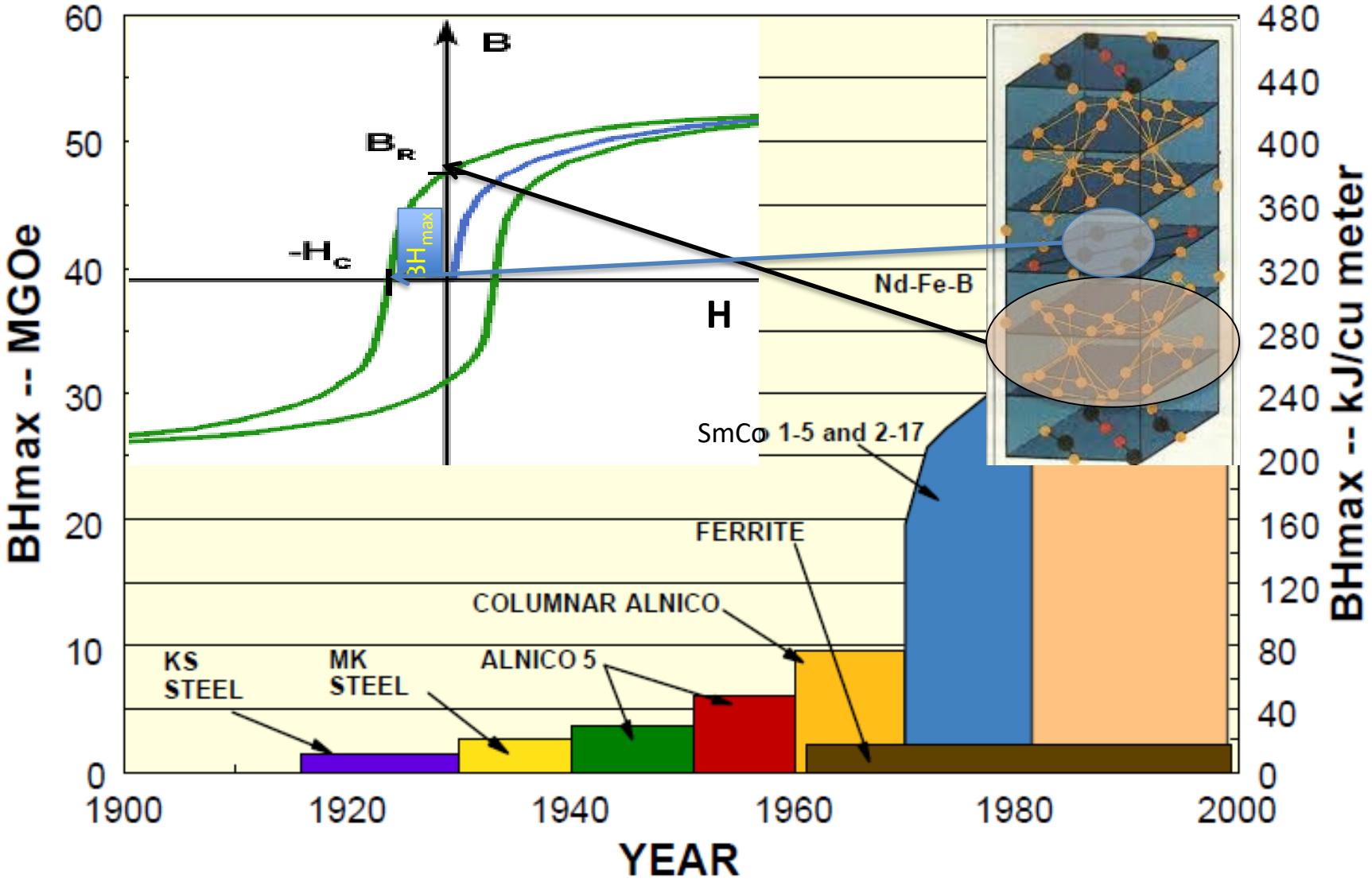
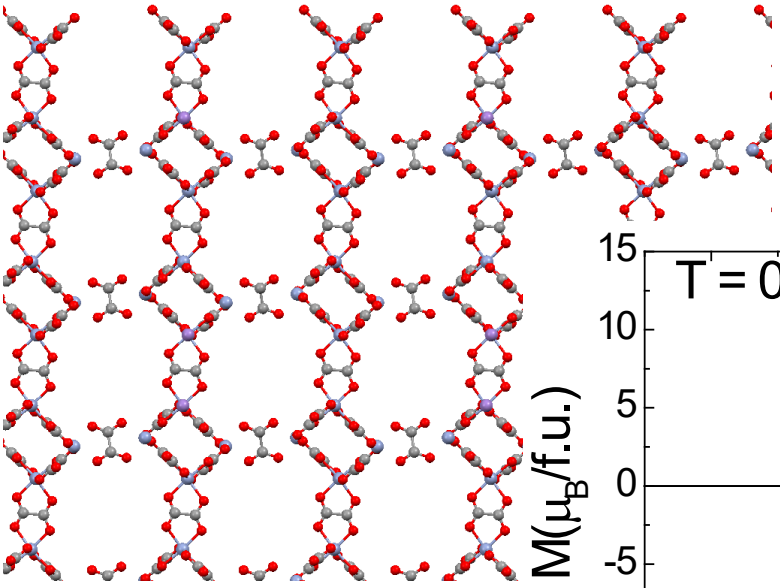


Figure I
Development of Permanent Magnets in the 1900's

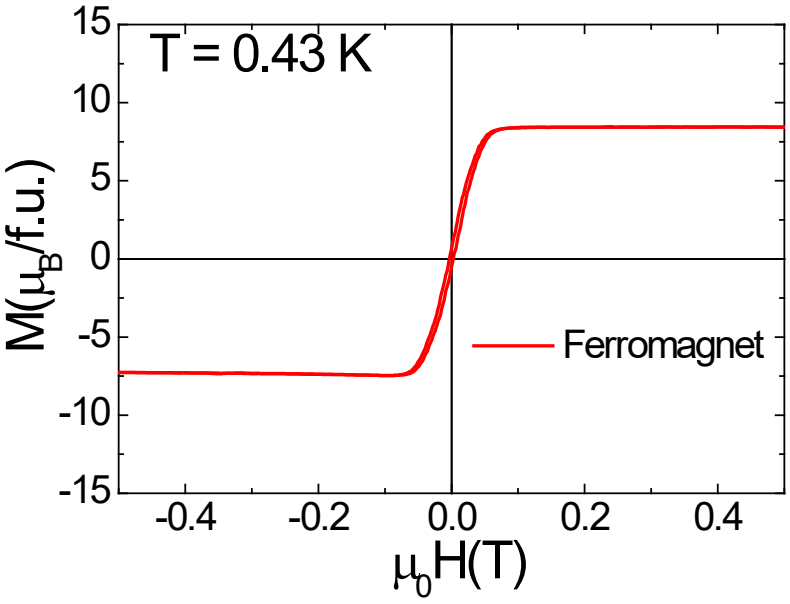


Molecular analogues

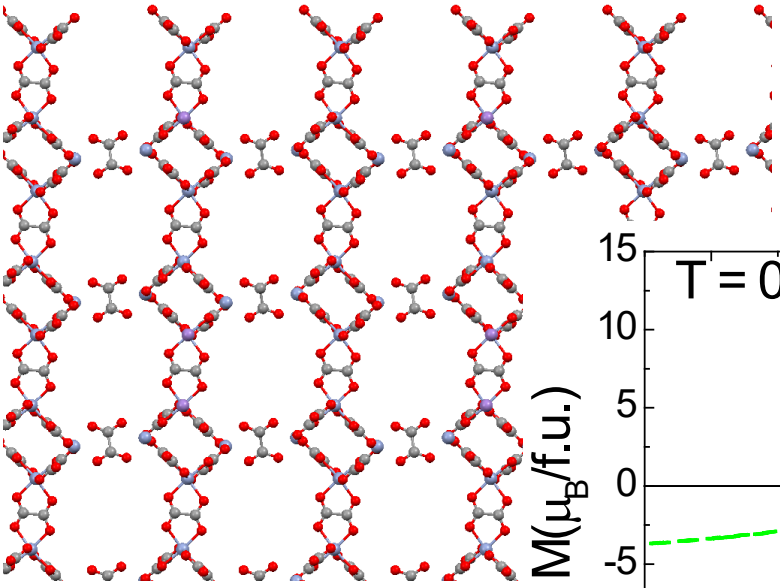


MnCr-oxalate

T_c = 5 K

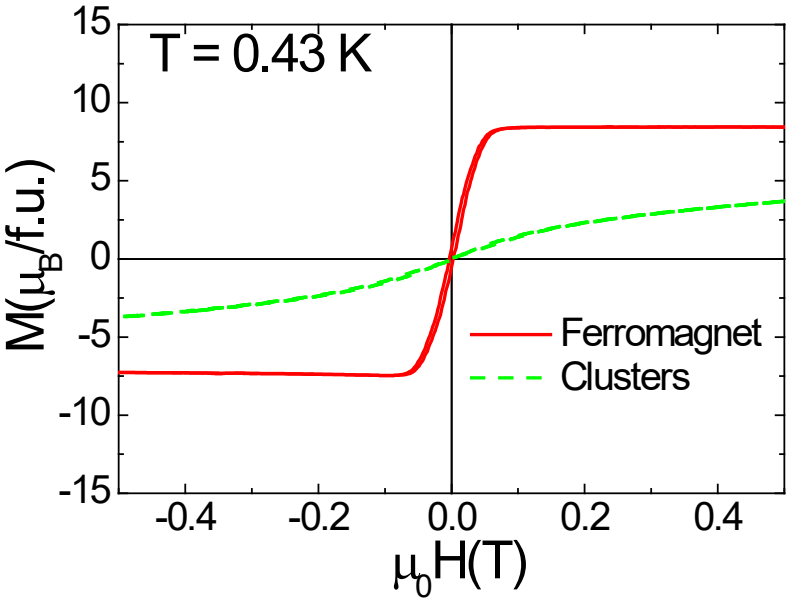


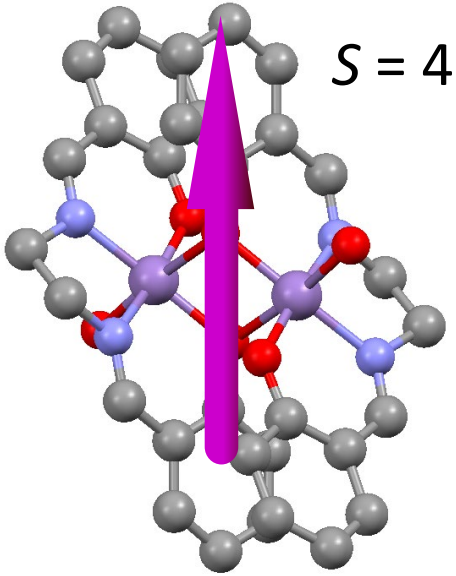
Molecular analogues



MnCr-oxalate

T_c = 5 K





Mn_2 cluster

T_b < 0.4 K

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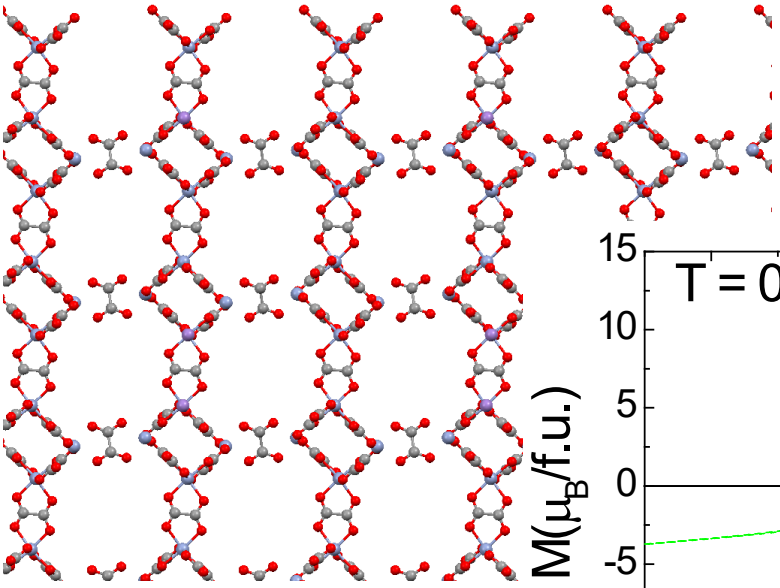
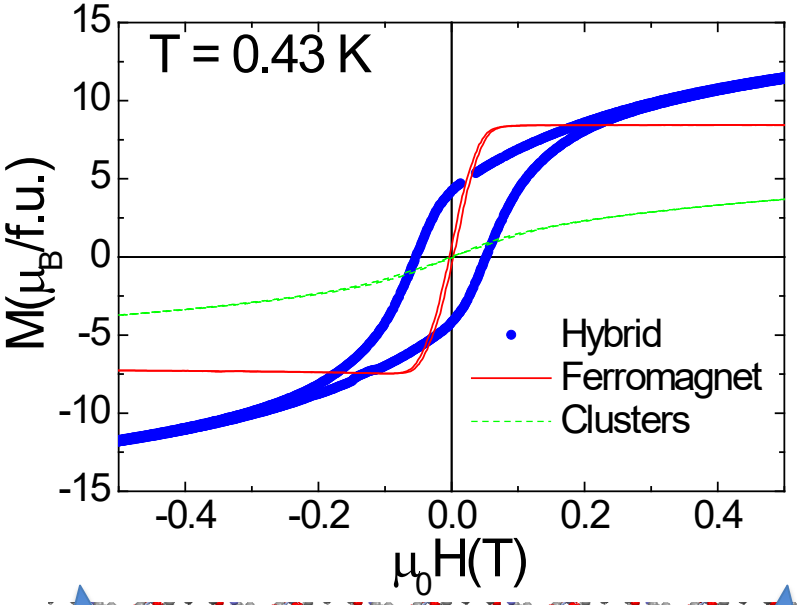
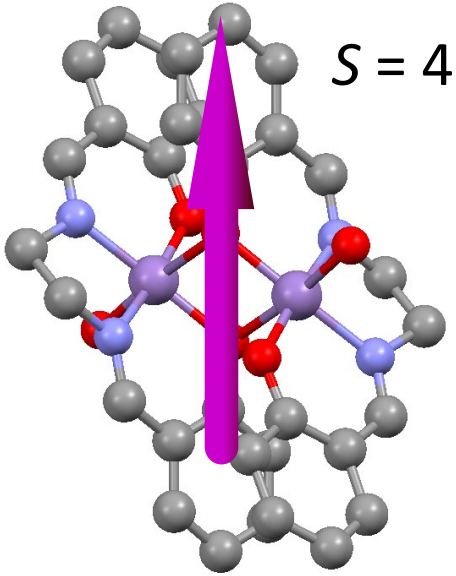
Molecular analogues

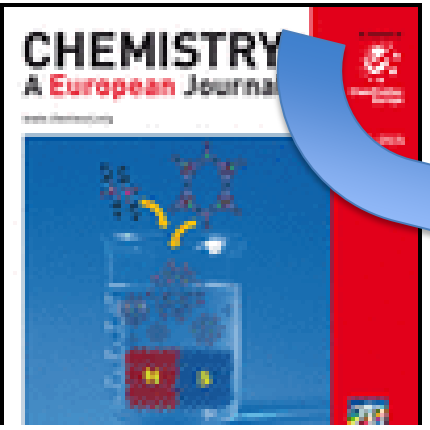


MnCr-oxalate

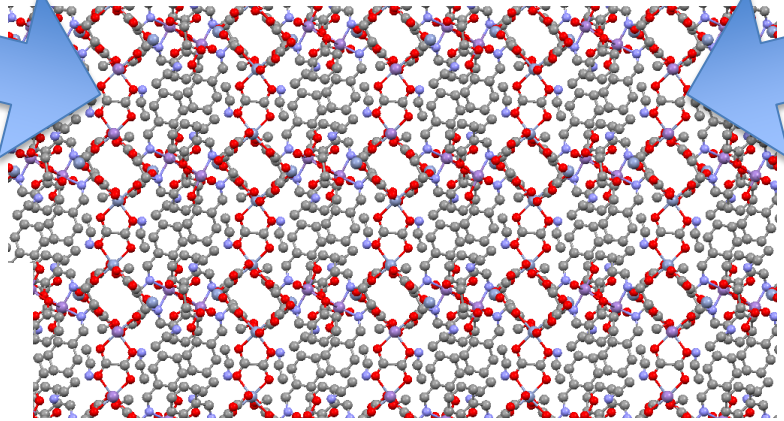




Mn_2 cluster



M. Clemente, E. Coronado, C. Gómez, A. Camón, A. Repollés, F. Luis, Chem. A Eur. J. **20**, 1669 (2014)



Challenge

$T_c \approx 5\ K \rightarrow T_c > 400\ K$

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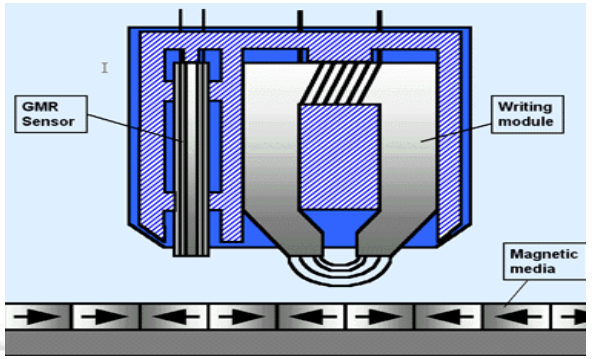
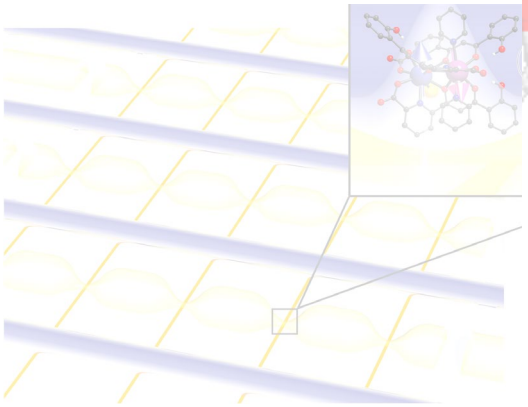
Outline

2. Magnets



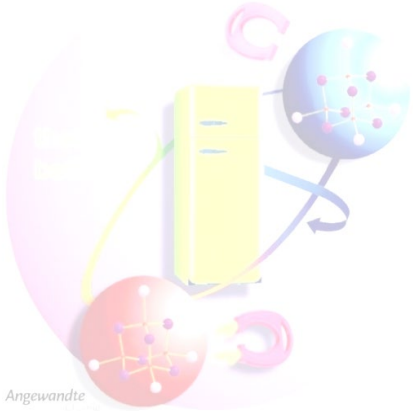
3. Magnetic recording

4. Quantum information



1. Physics

5. Magnetic coolers



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A century of magnetic recording

Telegraphone
Denmark
1898

RAMAC
IBM
1956

Magnetic hard disk

2021

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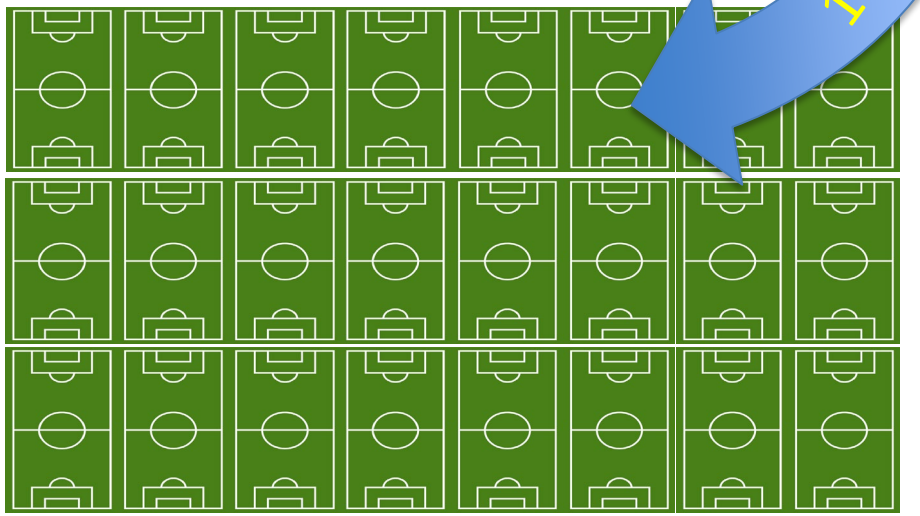
A century of magnetic recording

Telegraphone
Denmark
1898

RAMAC
IBM
1956

Ma ic hard disk
2021

1 Tbyte



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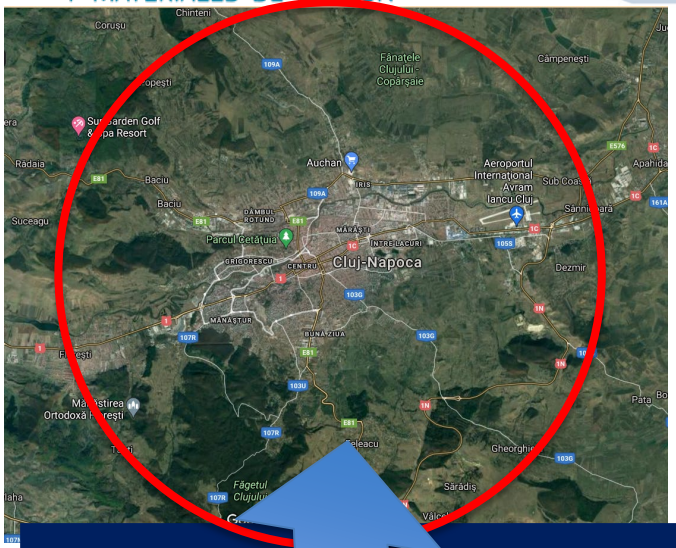
A century of magnetic recording



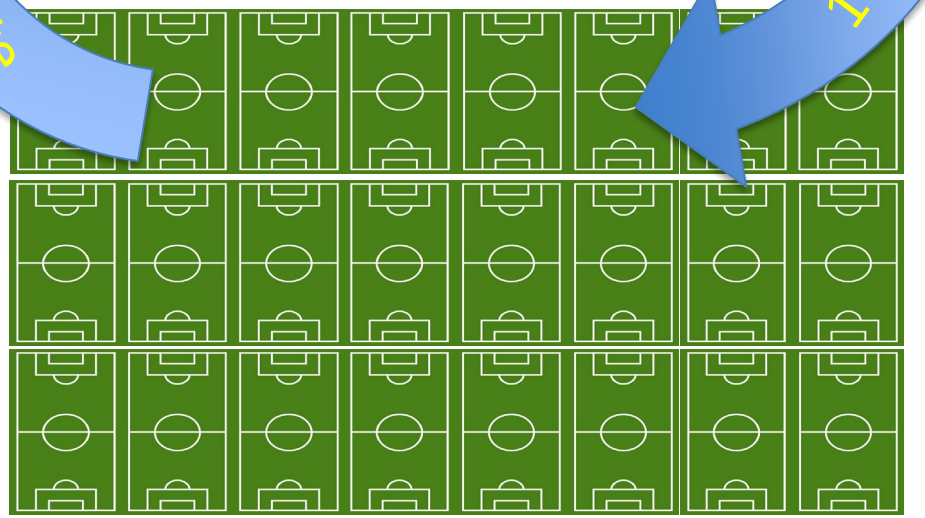
Telegraphor
Denmark
1898

RAMAC
IBM
1956

Ma ic hard disk
2021

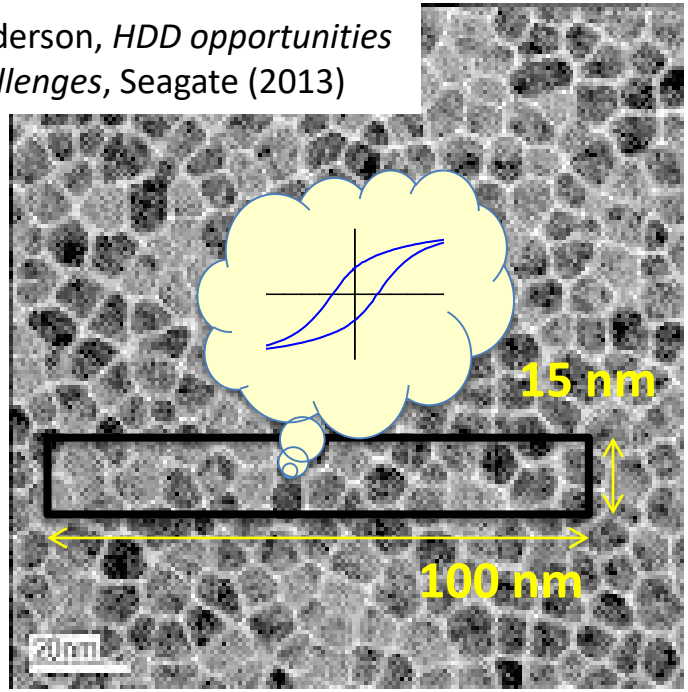
1 Tbyte

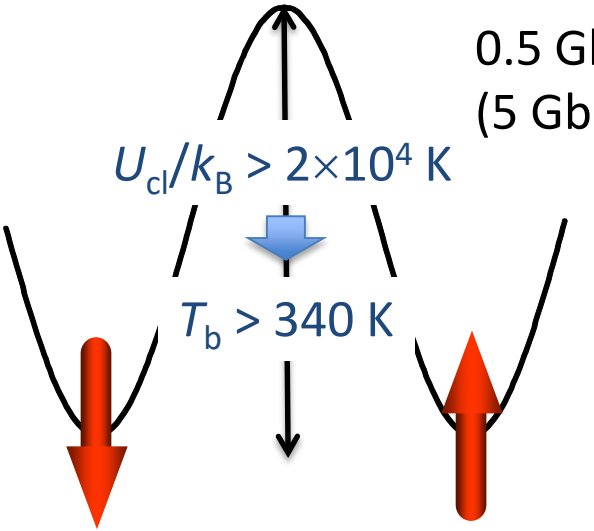
1 Tbyte



Magnetic bits

D. Anderson, *HDD opportunities & challenges*, Seagate (2013)

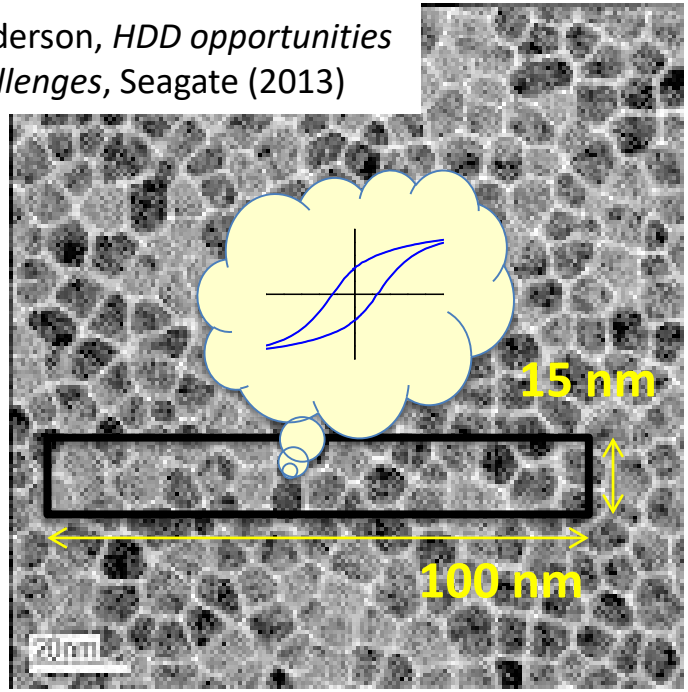


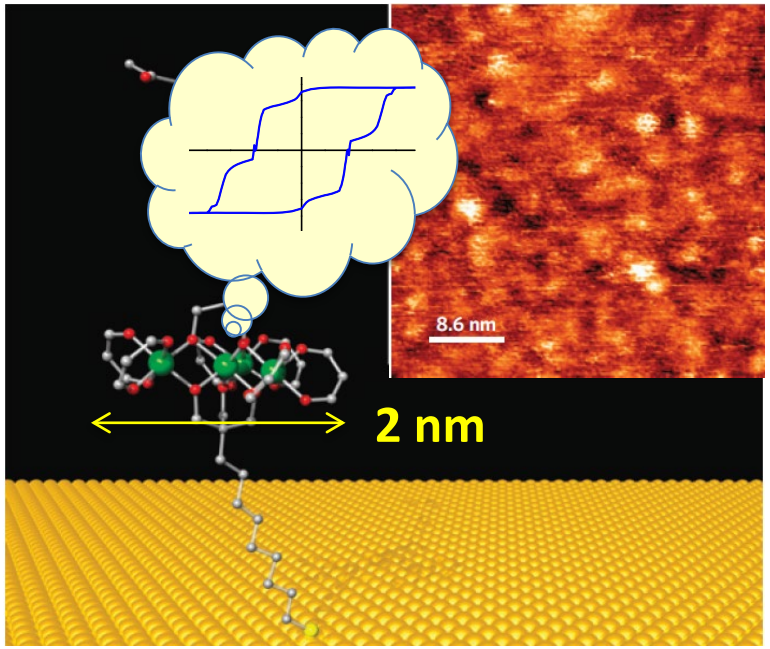


0.5 Gbit/in^2
(5 Gbit/in^2 expected)

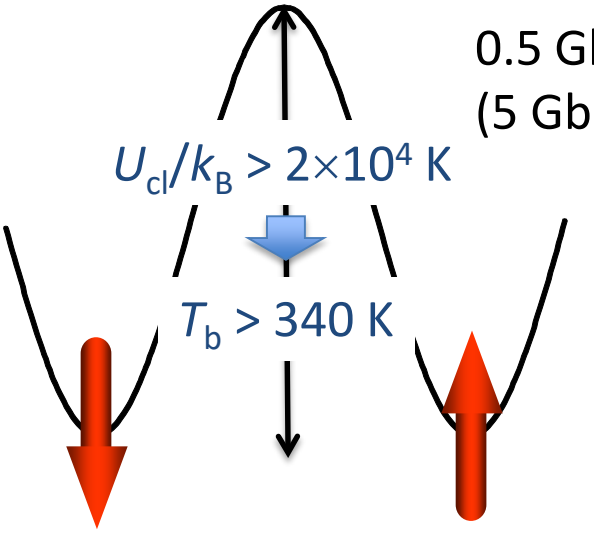
Magnetic bits

D. Anderson, *HDD opportunities & challenges*, Seagate (2013)





M. Mannini *et al*, Nature **468**, 417 (2010)



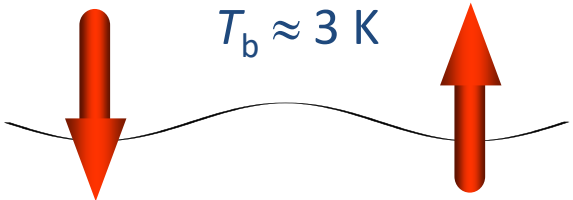
0.5 Gbit/in^2
(5 Gbit/in^2 expected)

5 Gbit/in^2 – 50 Gbit/in^2

BUT

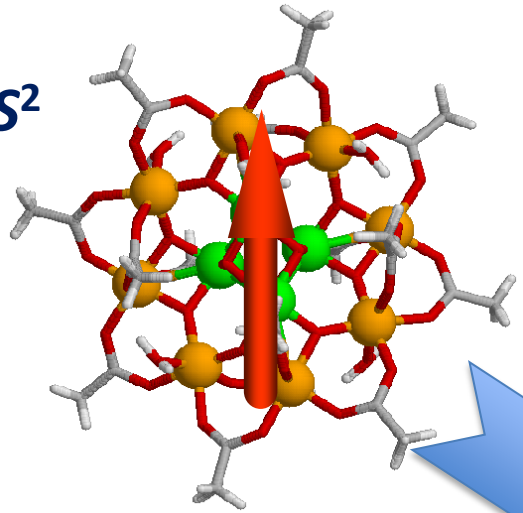
$U_{cl}/k_B \approx 70$ K

$T_b \approx 3$ K



The challenge: enhance T_b

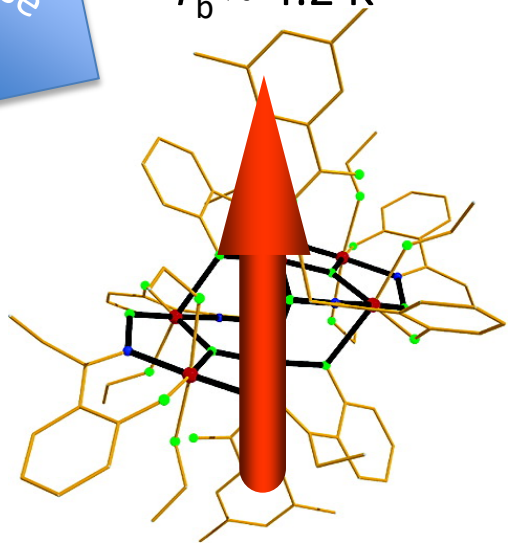
$U_{cl} \approx DS^2$



Mn_{12} (1993)
$S = 10$
$T_b \approx 3$ K

Increase S

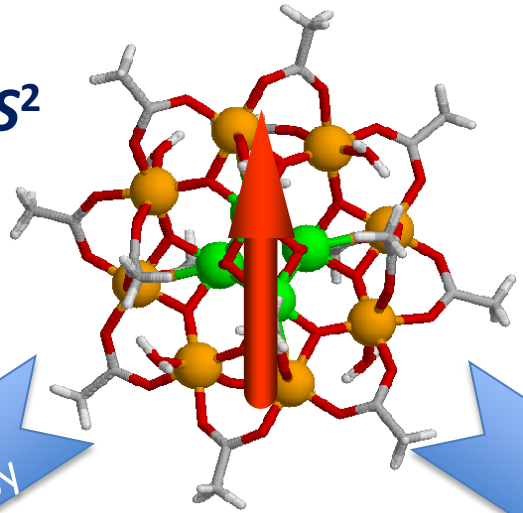
Ferromagnetic
Mn_6 cluster (2007)
$S = 12$
$T_b \approx 4.2$ K



C. Milios *et al*, J. Am. Chem. Soc. **129**, 2754 (2007)

The challenge: enhance T_b

$U_{cl} \approx DS^2$

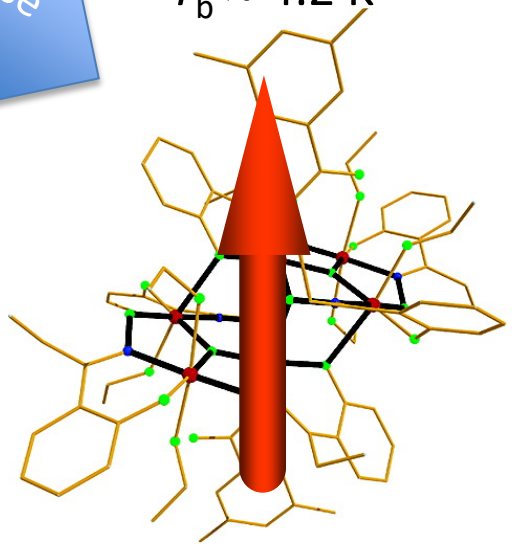


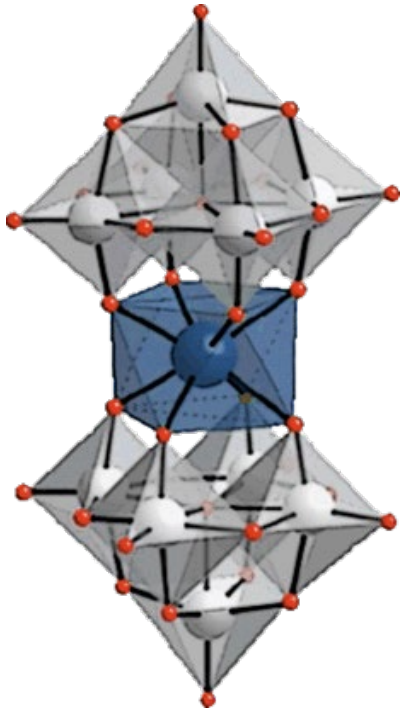
Increase anisotropy

Increase S

Mn_{12} (1993)
$S = 10$
$T_b \approx 3$ K

Ferromagnetic
Mn_6 cluster (2007)
$S = 12$
$T_b \approx 4.2$ K





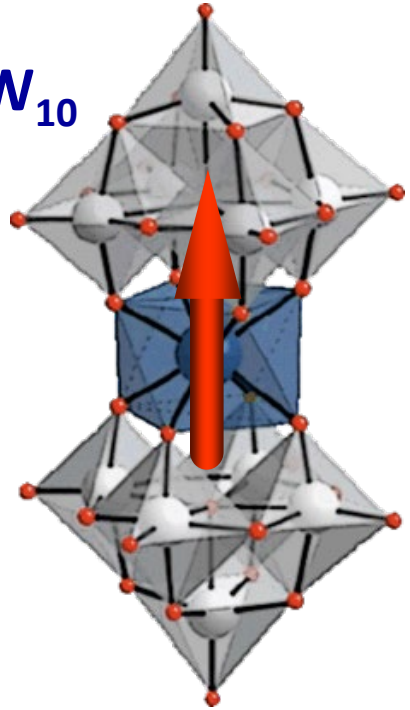
Lanthanide single ion magnets

J. van den Broek and L. C. van der Marel, Physica **29**, 948 (1963); N. Ishikawa et al, J. Am. Chem. Soc. **125**, 8694 (2003); M. AlDamen et al, J. Am. Chem. Soc. **130**, 8874 (2008); ...

C. Milios et al, J. Am. Chem. Soc. **129**, 2754 (2007)

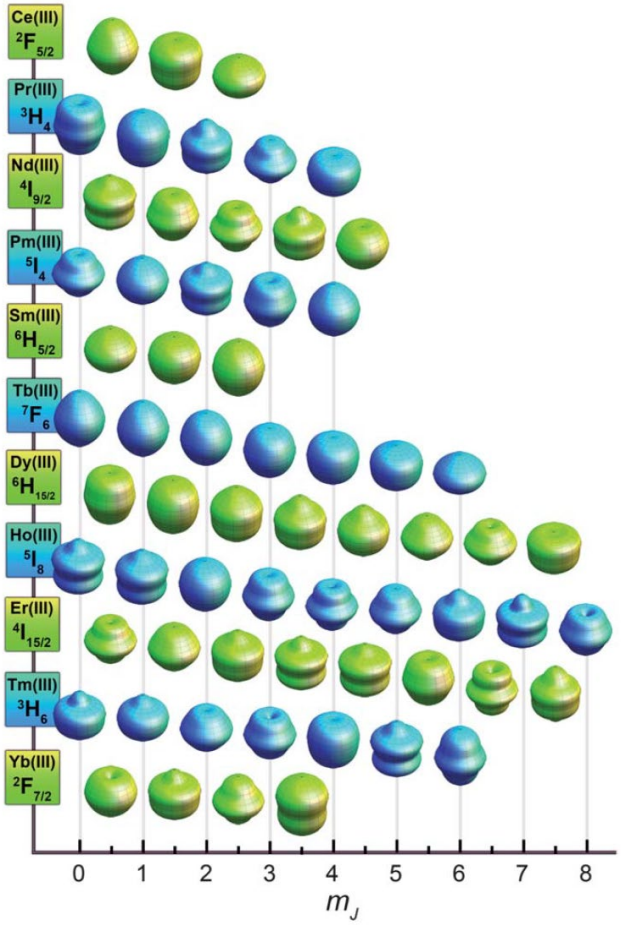
Lanthanide single-ion magnets

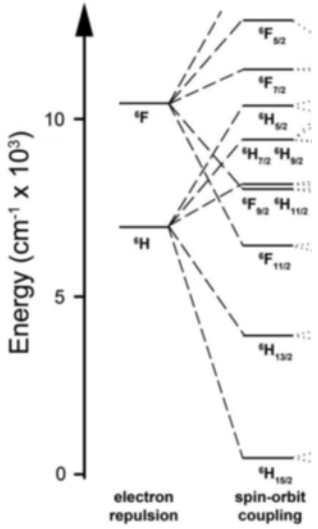
LnW_{10}



High magnetic moment

$$\vec{\mu} = g_J \mu_B \vec{J}$$





A. Abragam and B. Bleaney, *EPR of transition ions*, Dover (New York 1970); J. D. Rinehart and J. R. Long, Chem. Sci. **2**, 2078 (2011)

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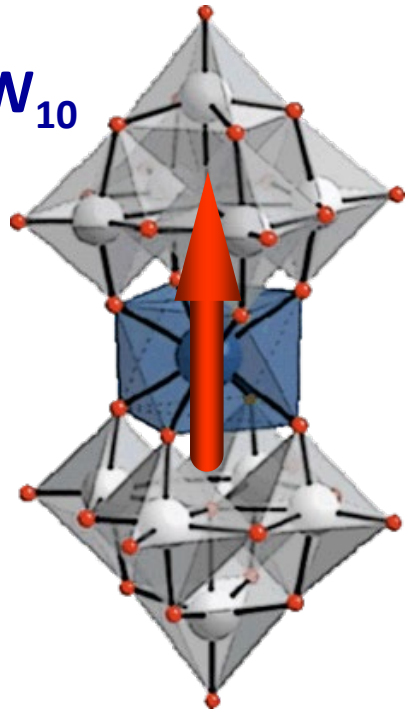
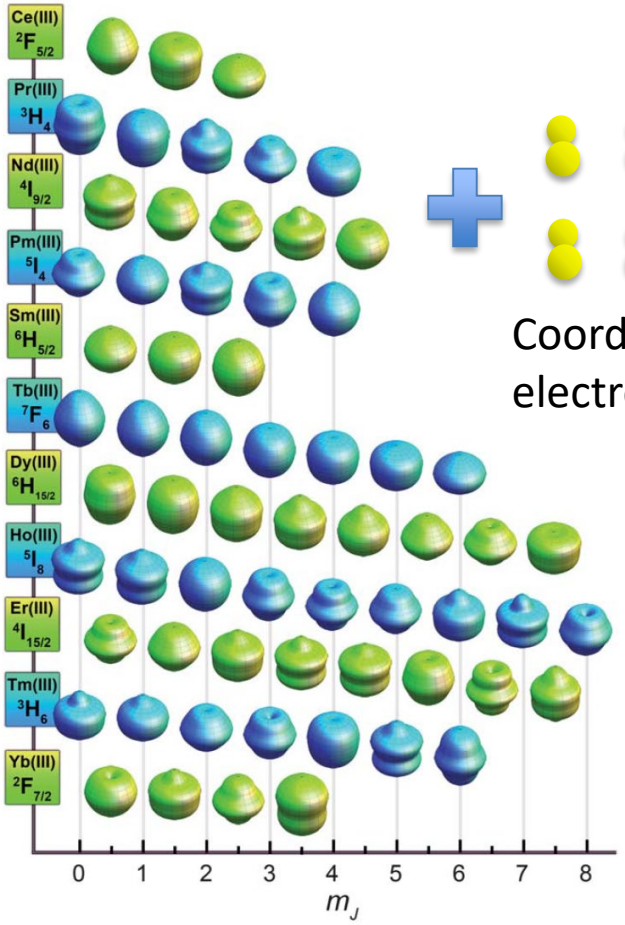
Lanthanide single-ion magnets

LnW_{10}



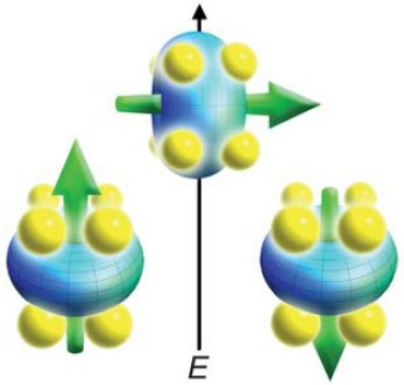
High magnetic moment

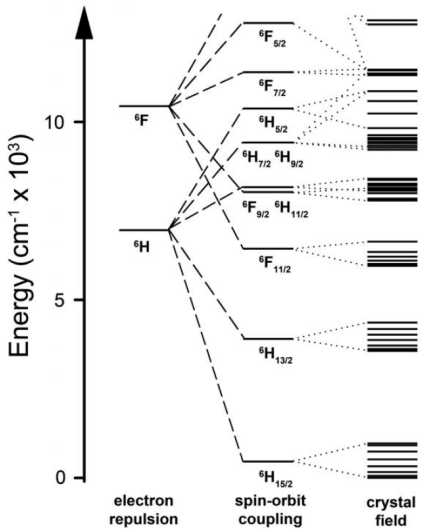
$$\vec{\mu} = g_J \mu_B \vec{J}$$



Coordination electrons

Strong magnetic anisotropy

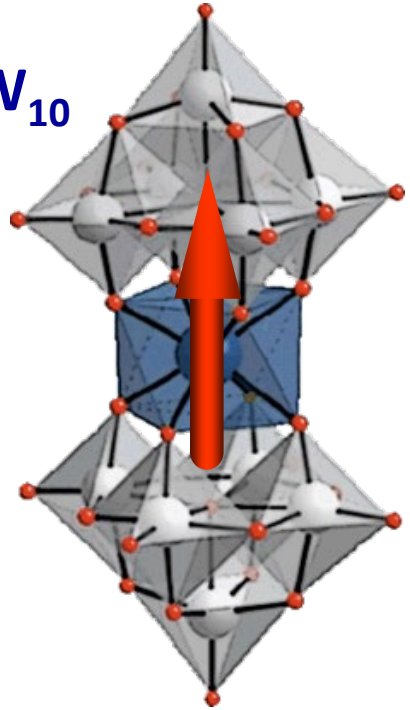




A. Abragam and B. Bleaney, *EPR of transition ions*, Dover (New York 1970); J. D. Rinehart and J. R. Long, Chem. Sci. **2**, 2078 (2011)

Lanthanide single-ion magnets

ErW_{10}

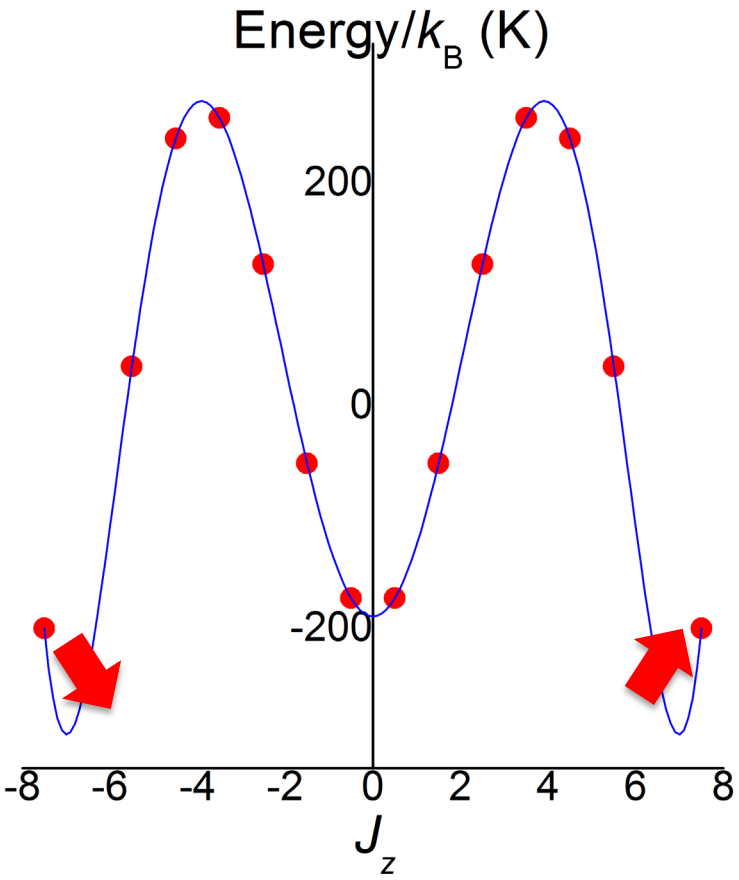


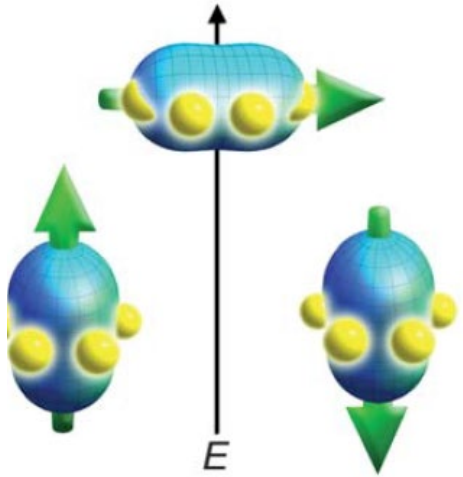
High magnetic moment

$$\vec{\mu} = g_J \mu_B \vec{J}$$

Strong magnetic anisotropy

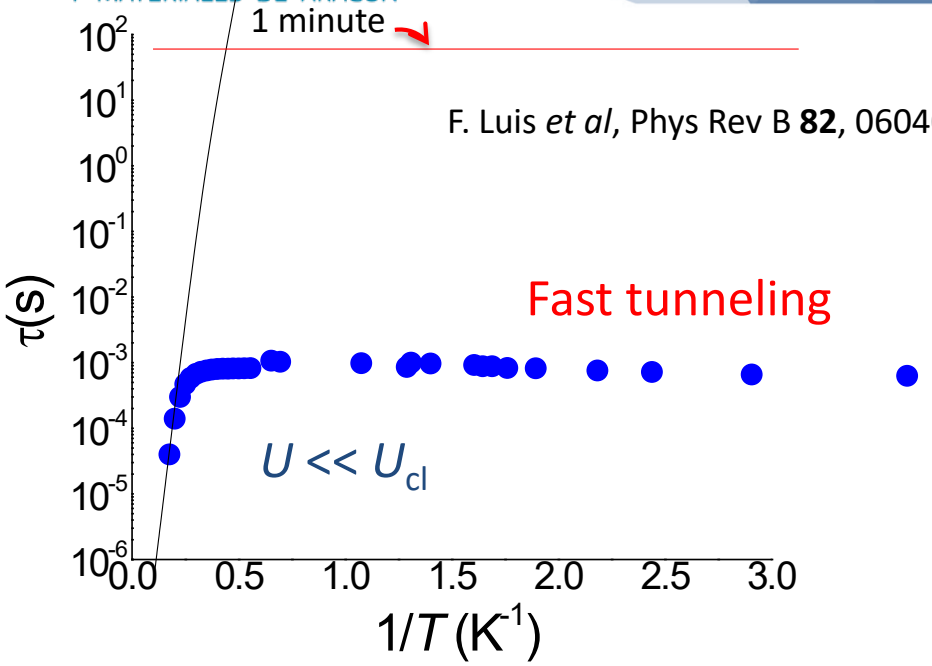
$$\mathcal{H} = \sum_{n,\text{even}} B_n^0 O_n^0 + \sum_{\substack{n,\text{even} \\ m \le n}} B_n^m O_n^m$$



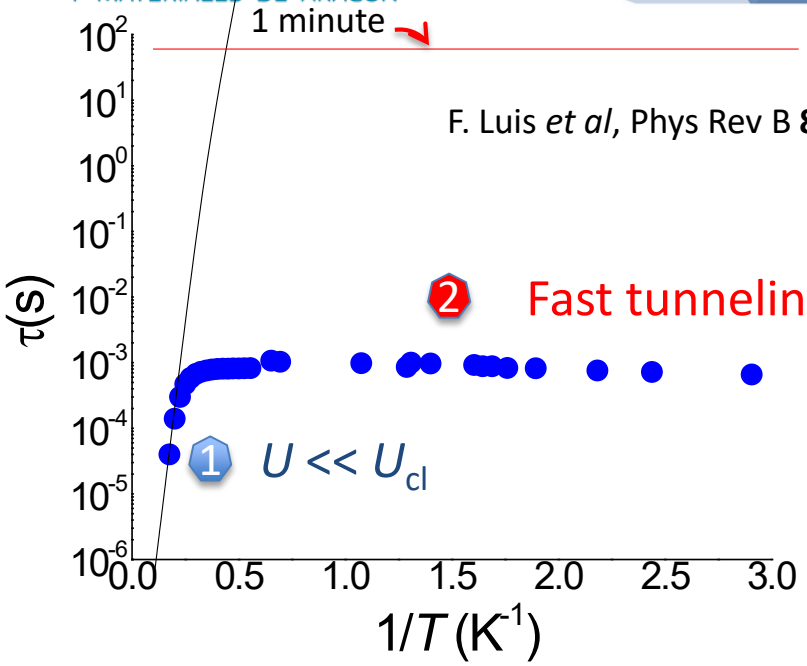


K. W. H. Stevens Proc. Phys. Soc. London A **65**, 209 (1952)

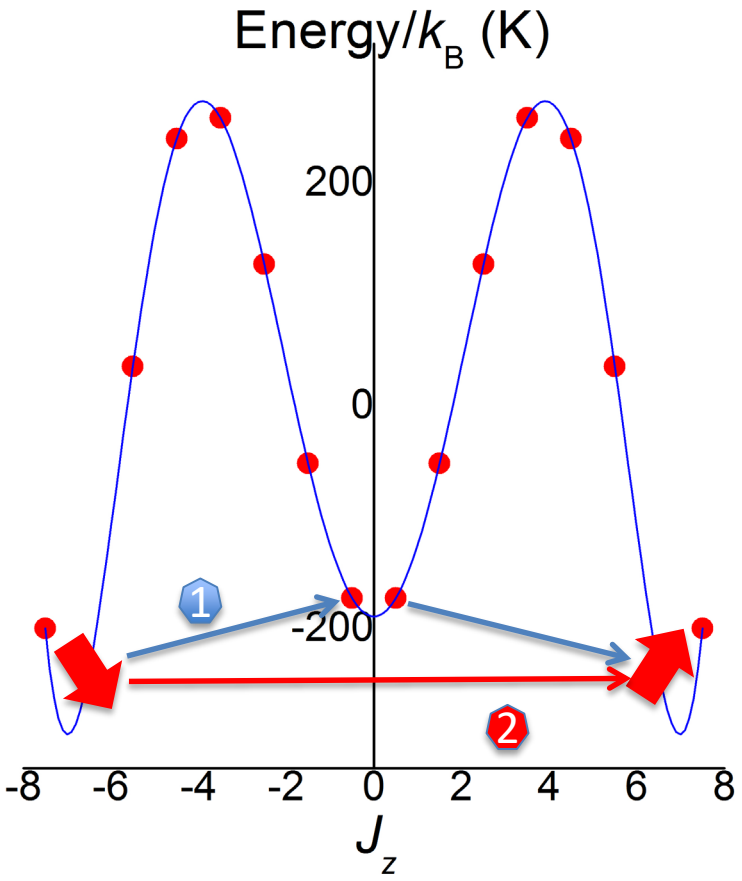
Spin dynamics

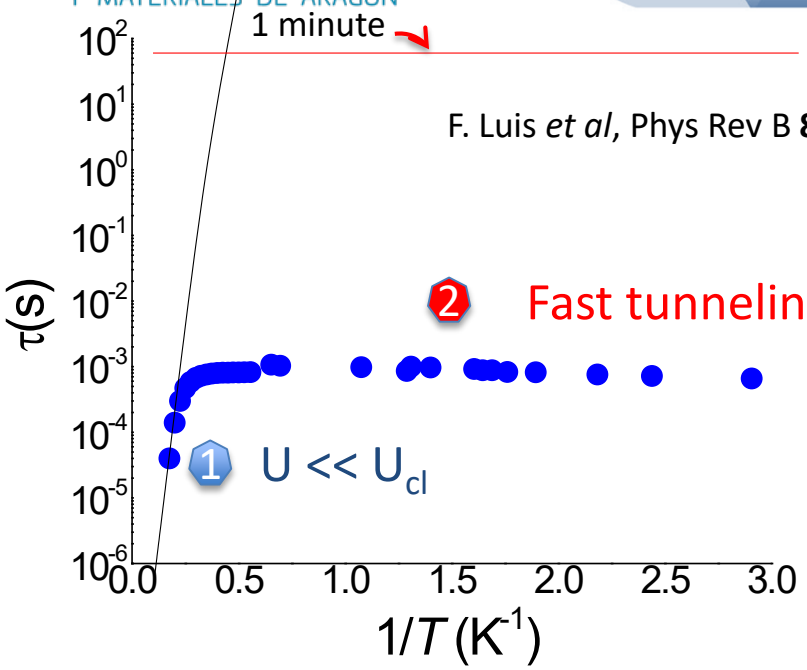


F. Luis *et al*, Phys Rev B **82**, 060403 (2010)

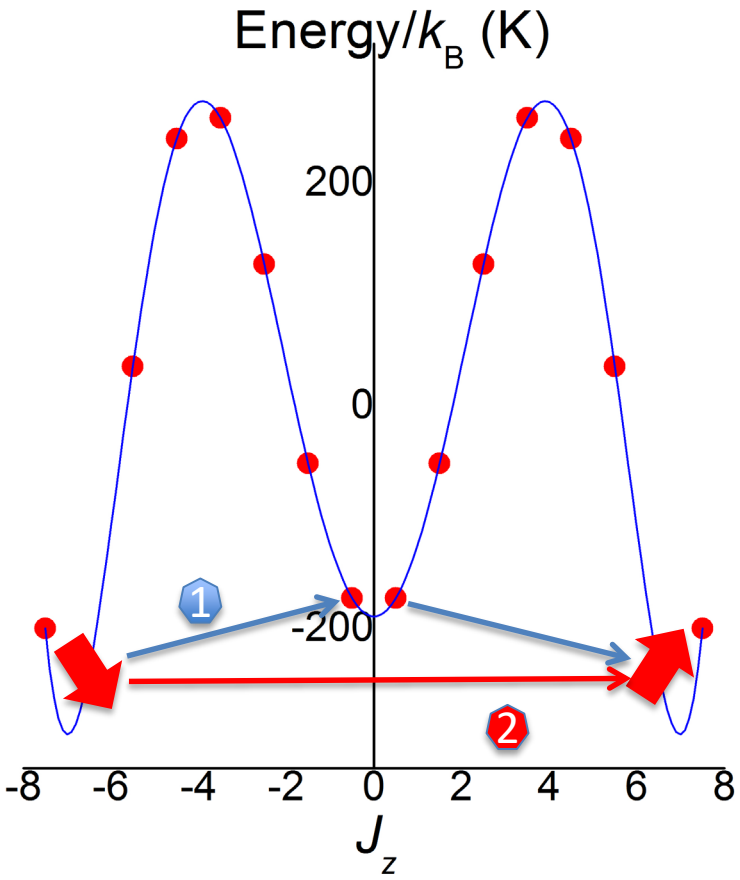


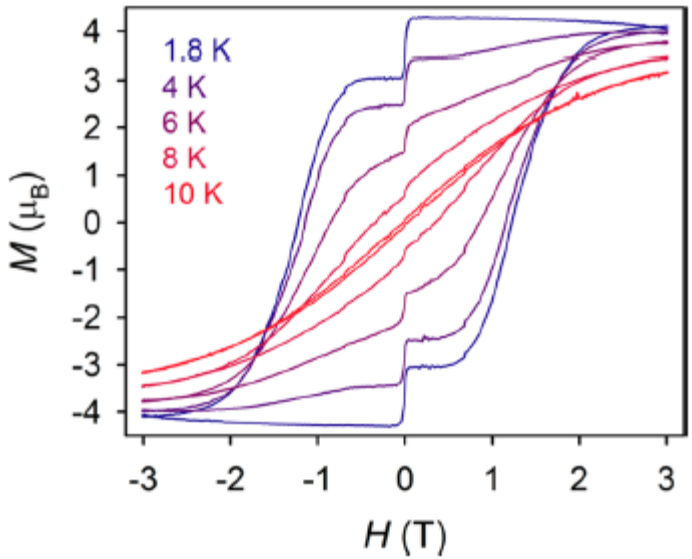
F. Luis *et al*, Phys Rev B **82**, 060403 (2010)





F. Luis *et al*, Phys Rev B **82**, 060403 (2010)





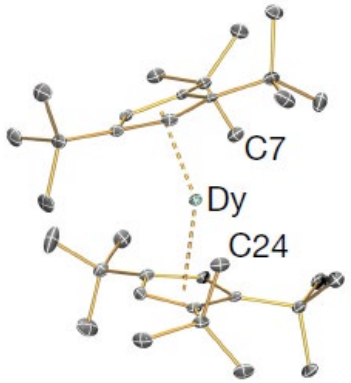
$T_b < 10$ K

$U < 150$ K

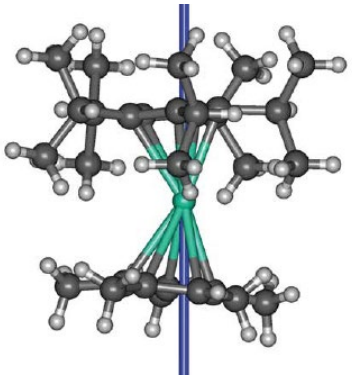
K. R. Meihaus and J. R. Long, J. Am. Chem. Soc. **135**, 17952 (2013)

Coordination design

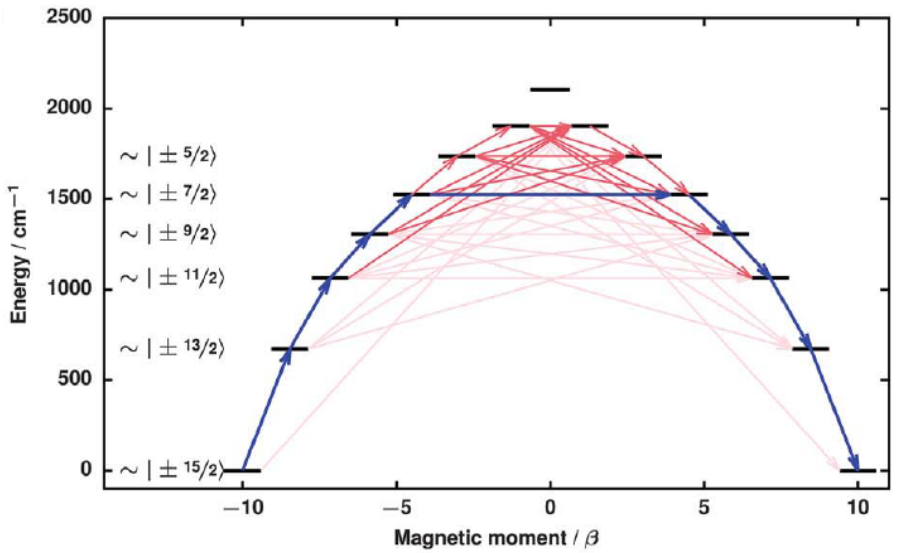
Linear coordination with “oblate” magnetic orbitals (e.g. Dy^{3+})



Dysprocenium

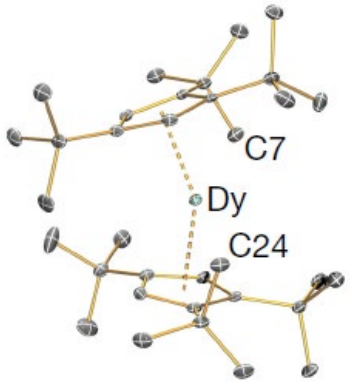


Cp-Dy-Cp*

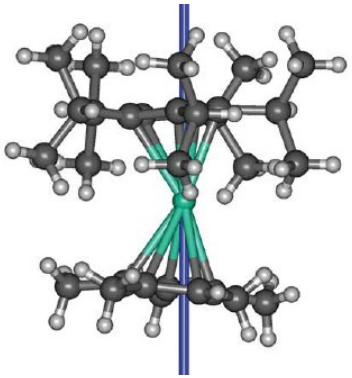


Coordination design

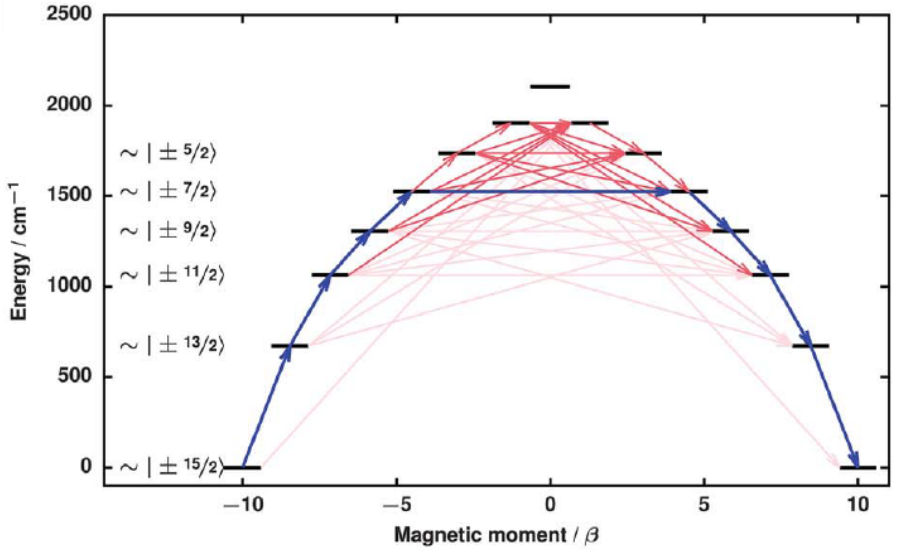
Linear coordination with “oblate” magnetic orbitals (e.g. Dy^{3+})

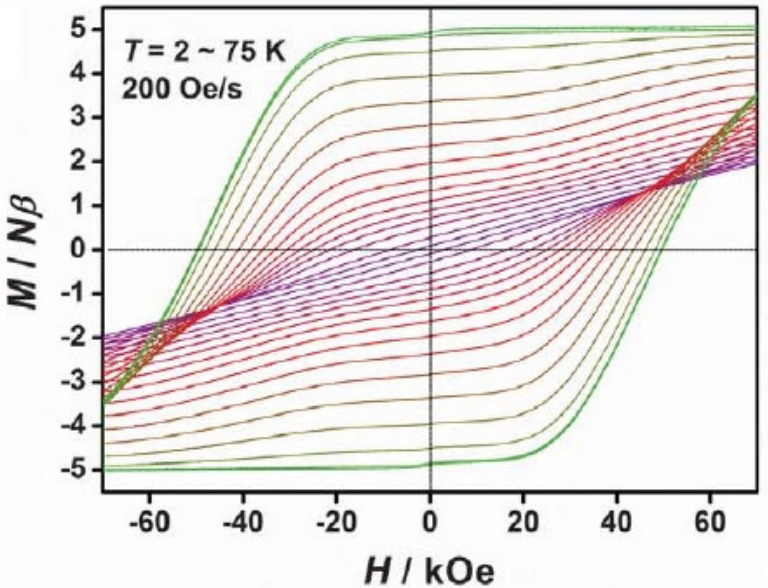


Dysprocenium



Cp-Dy-Cp*

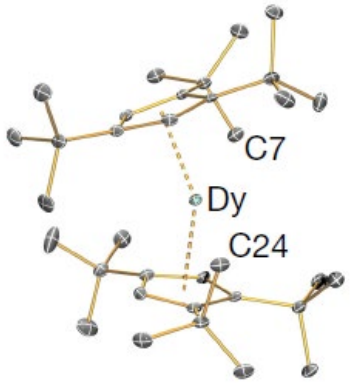




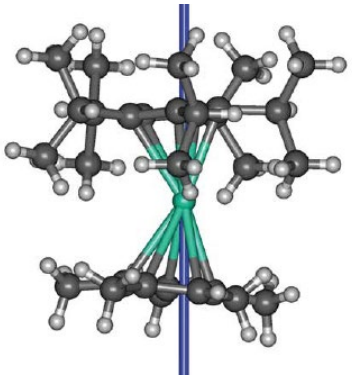
Hysteresis above liquid nitrogen temperatures

Coordination design

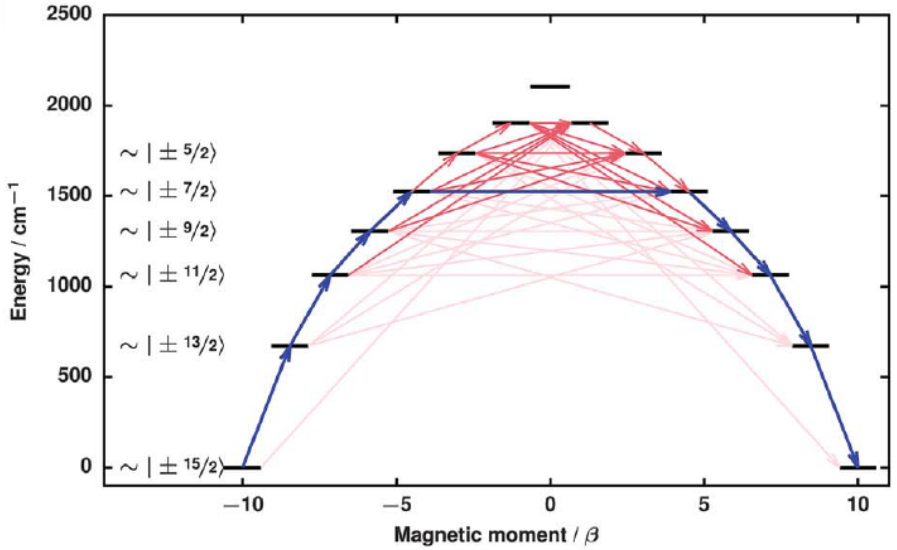
Linear coordination with “oblate” magnetic orbitals (e.g. Dy^{3+})

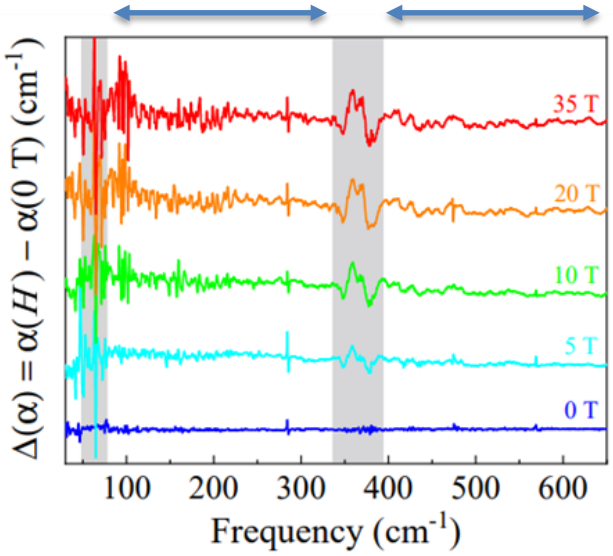


Dysprocenium



Cp-Dy-Cp*





Model and tune key molecular vibrations

Vibrational “windows”

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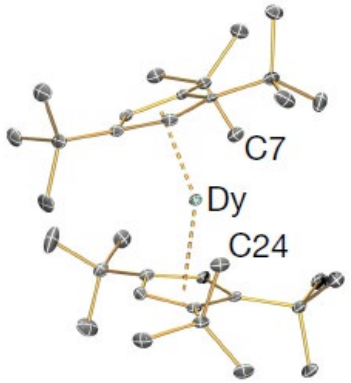
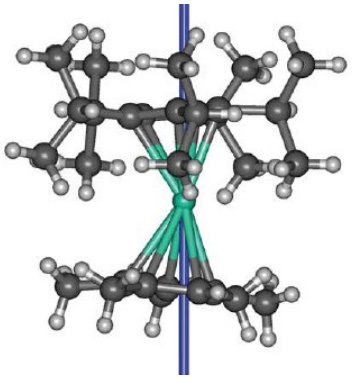
Coordination design

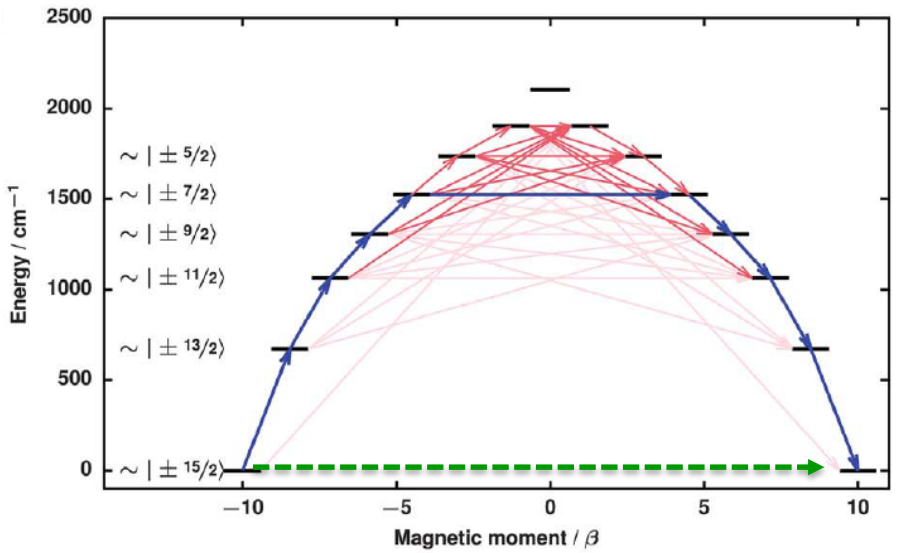
Linear coordination with “oblate” magnetic orbitals (e.g. Dy^{3+})

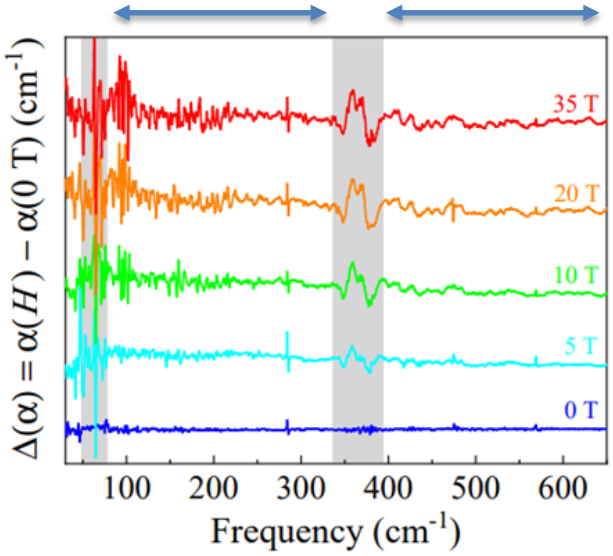


Dysprocenium



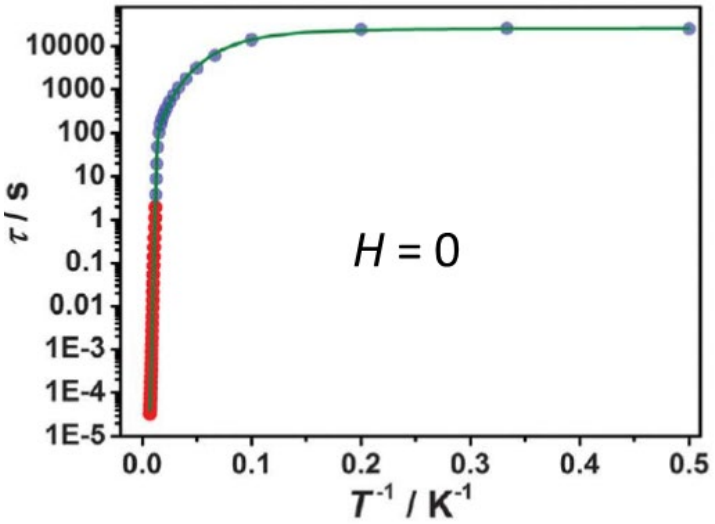
Cp-Dy-Cp*





Model and tune key molecular vibrations

Vibrational “windows”



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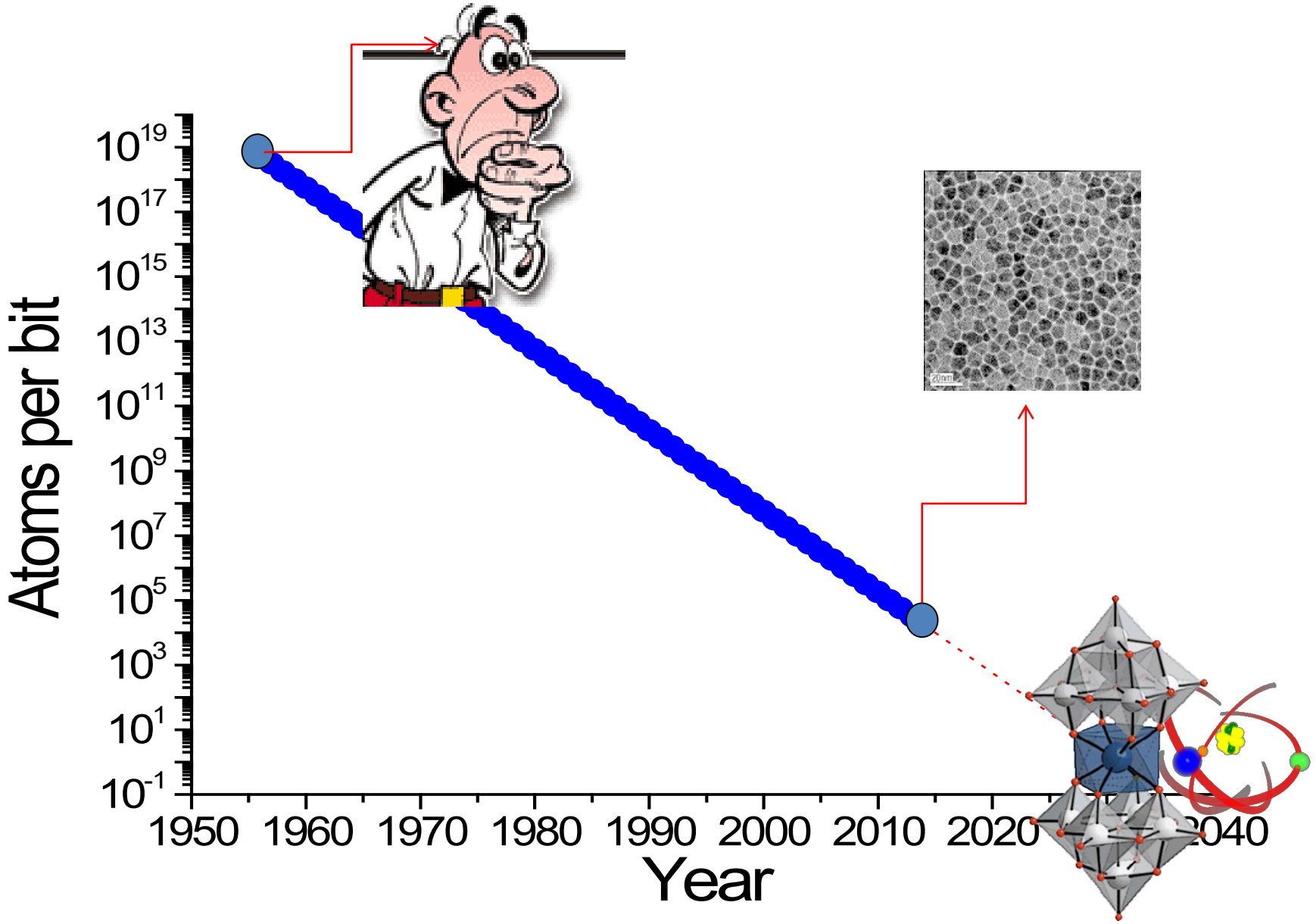
Quantum roadblock



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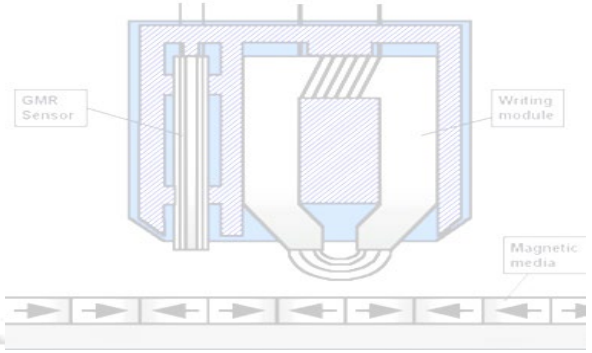
Outline

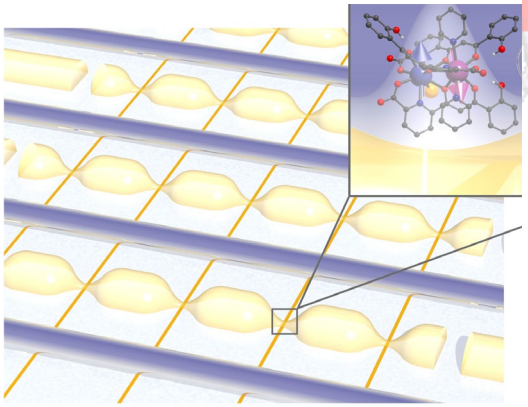
2. Magnets



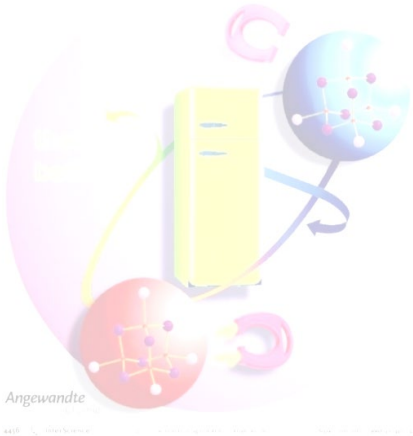
3. Magnetic recording

1. Physics

4. Quantum information



5. Magnetic coolers



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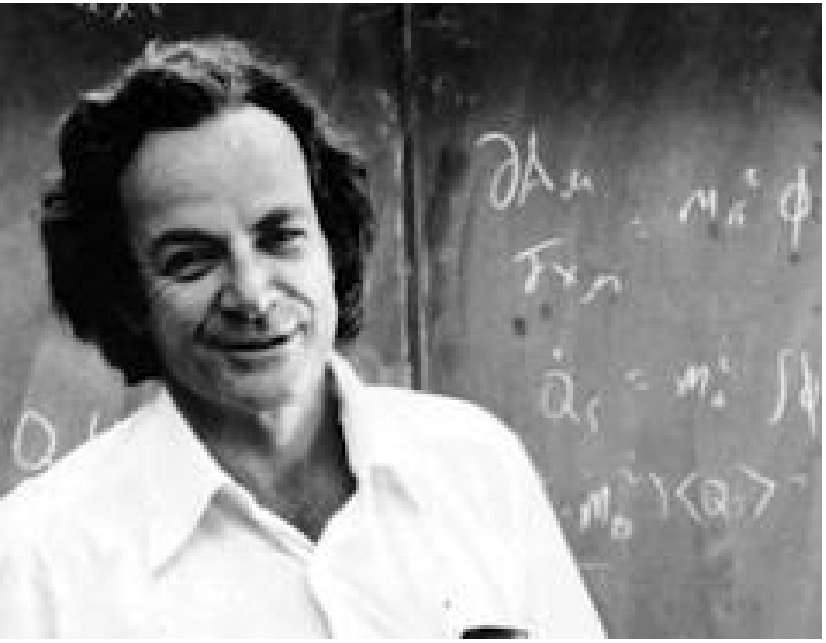
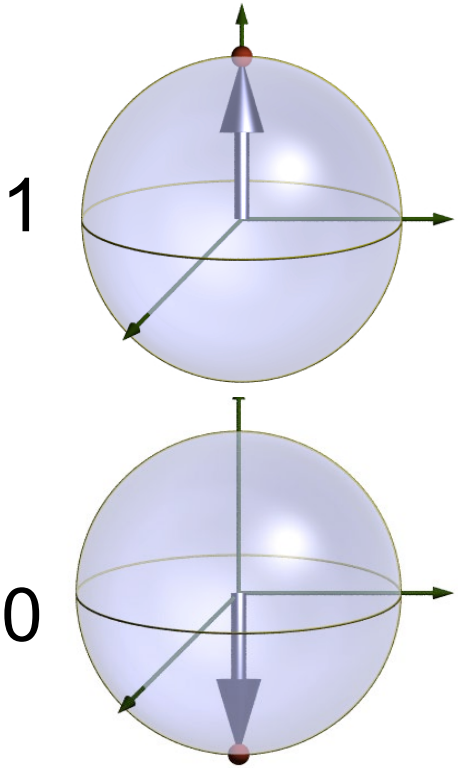
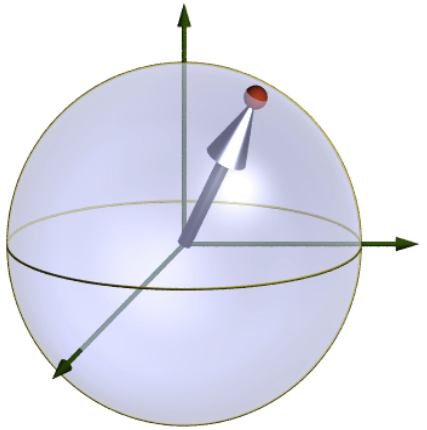
Quantum computers



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

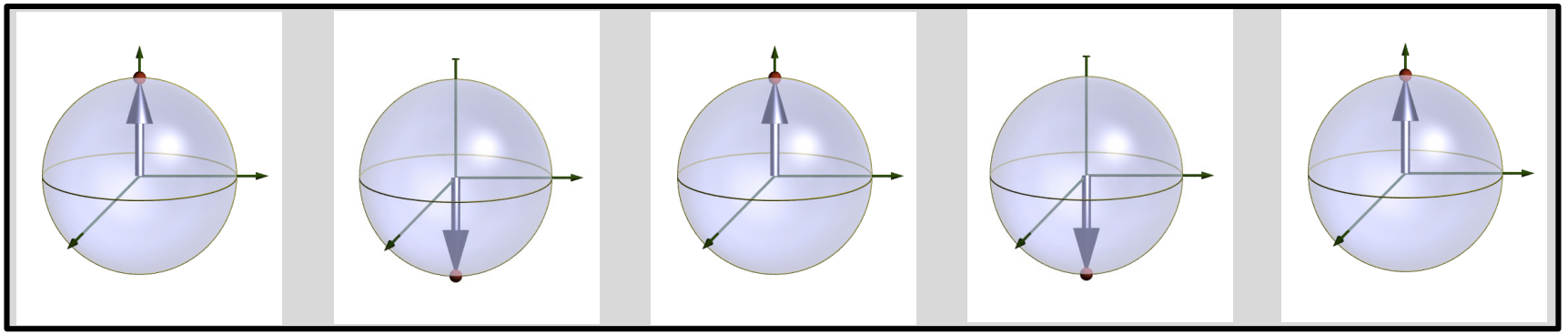




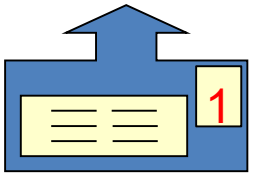
R. P. Feynman, Int. J. Theoret. Phys. **21**, 467 (1982)

D. Deutsch, Proc. Royal Soc. A **400**, 97 (1985)

Quantum paralelism

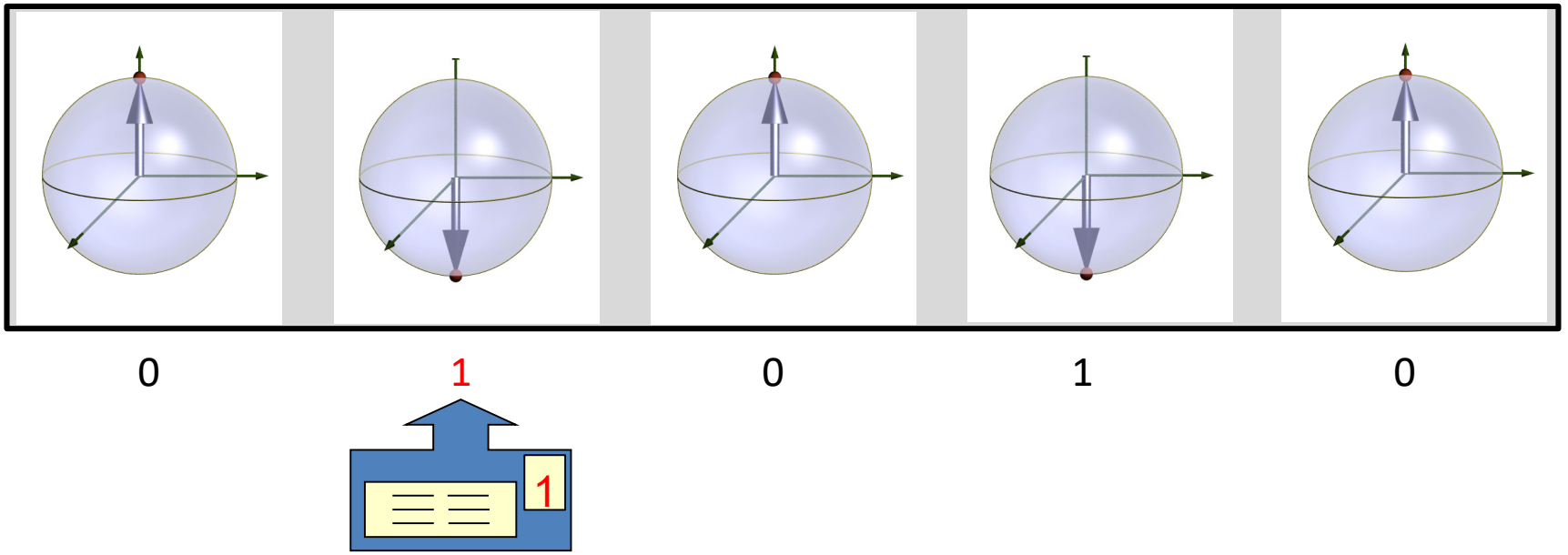


0 1 0 1 0



D. Deutsch, Proc. R. Soc. Lond. A **400,** 97 (1985).

Quantum paralelism



D. Deutsch, Proc. R. Soc. Lond. A **400,** 97 (1985).

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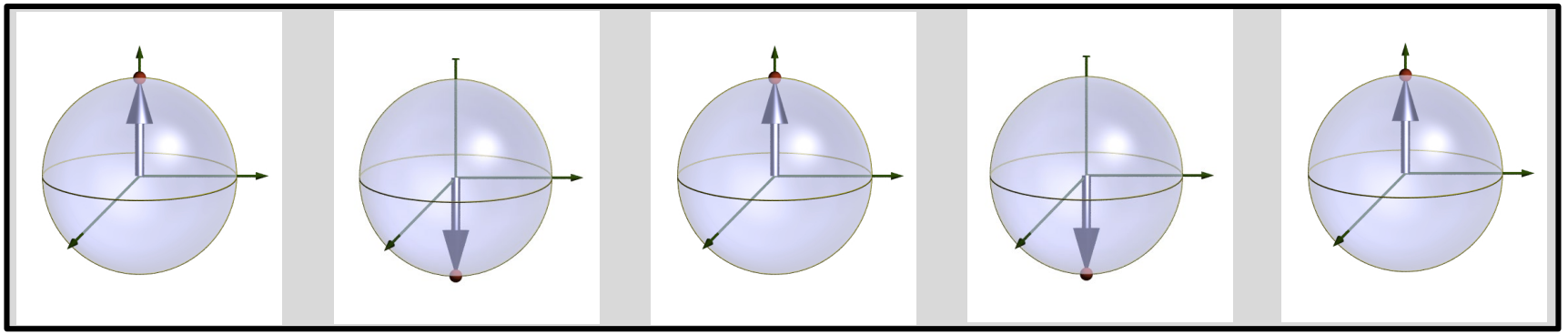
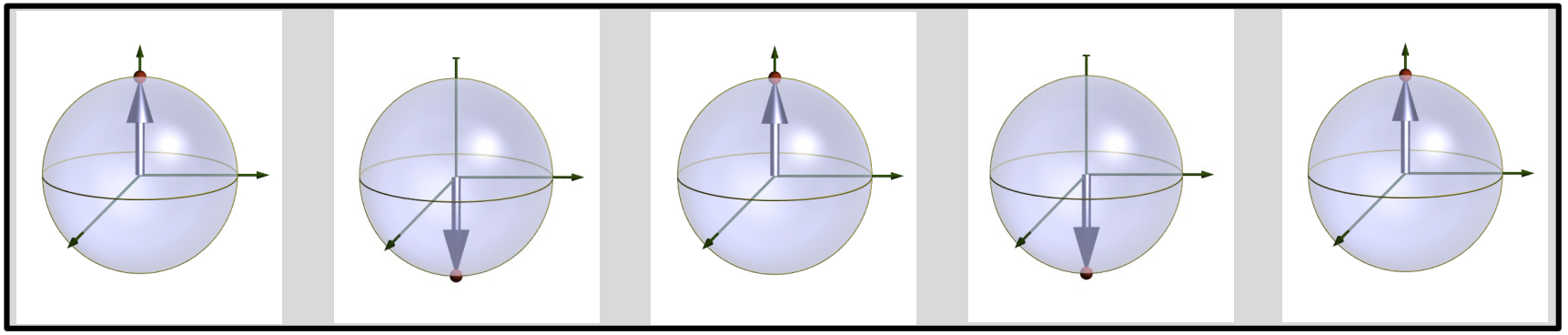
Quantum paralelism

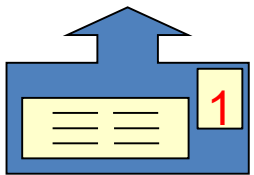


0 1 0 1 0

+



0 0 0 1 0



D. Deutsch, Proc. R. Soc. Lond. A **400,** 97 (1985).

Algorithms

Prime number factorization

Classical $t \propto e^N \Rightarrow$ Quantum $t \propto N^p$

P. W. Shor, in *Proc. Symp. Foundations of Computer Science, IEEE Computer Society Press,* 124 (1994)

Fast database searching

Phone number $\Rightarrow$ Suscriber?

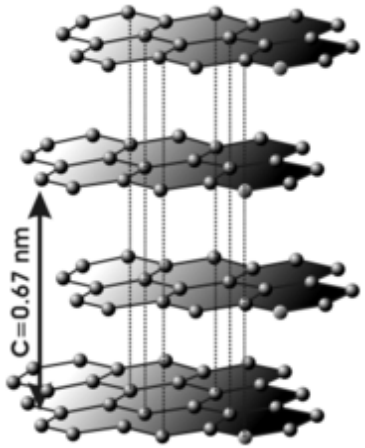
Classical $t \propto N$

$\Downarrow$

Quantum $t \propto \sqrt{N}$

L. K. Grover,Phys. Rev. Lett. **79**, 325 (1997)

Quantum digital simulators



Classical $t \propto e^N$

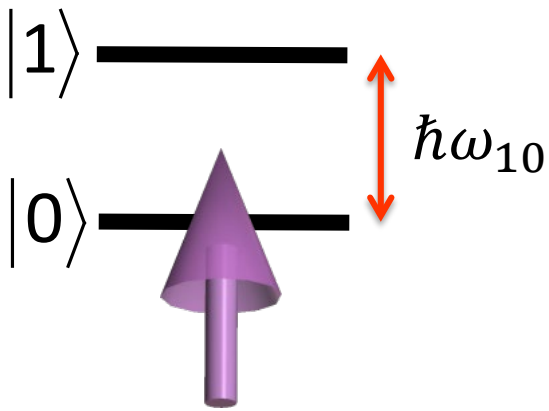
$\Downarrow$

Quantum $t \propto N$

J. I. Cirac & P. Zoller, Nature Phys. **8** 264 (2012)

Material qubits

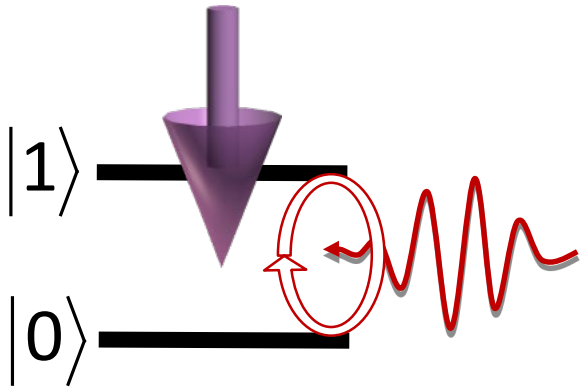
D. P. Di Vincenzo, Fortschr. Phys. **48**, 771 (2000)



- Two well-defined states + initialization

Material qubits

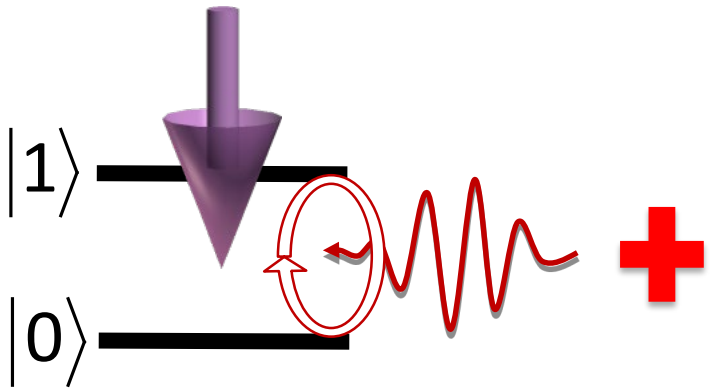
D. P. Di Vincenzo, Fortschr. Phys. **48**, 771 (2000)



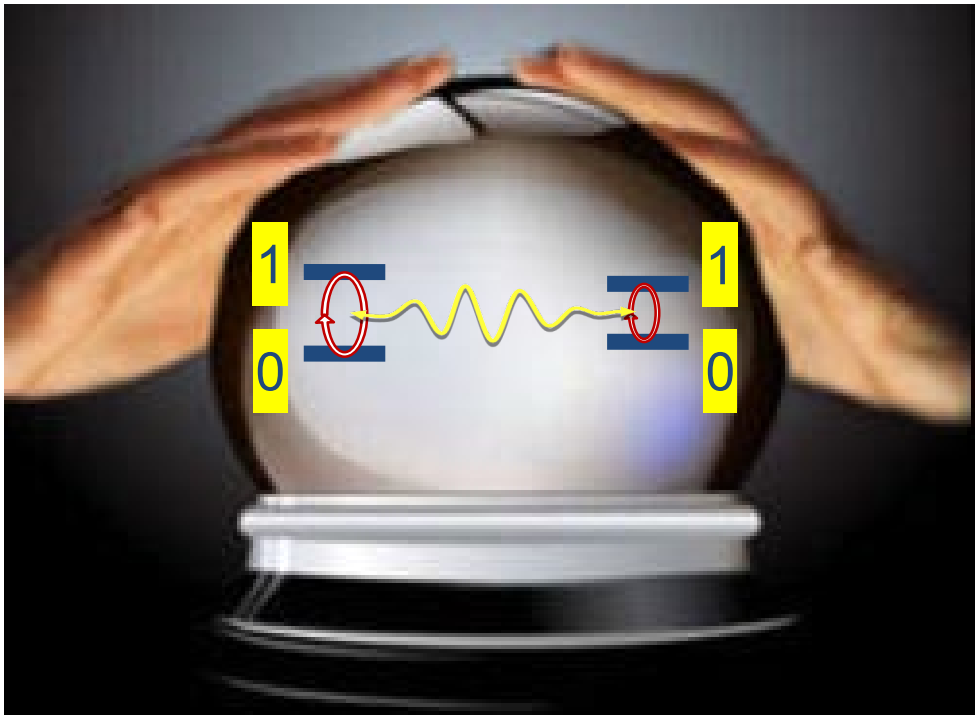
- Two well-defined states + initialization
- External control of the wave function
- Sufficiently high quantum coherence $T_2\Omega_R >> 1$ (≥ 100)

Material qubits

D. P. Di Vincenzo, Fortschr. Phys. **48**, 771 (2000)



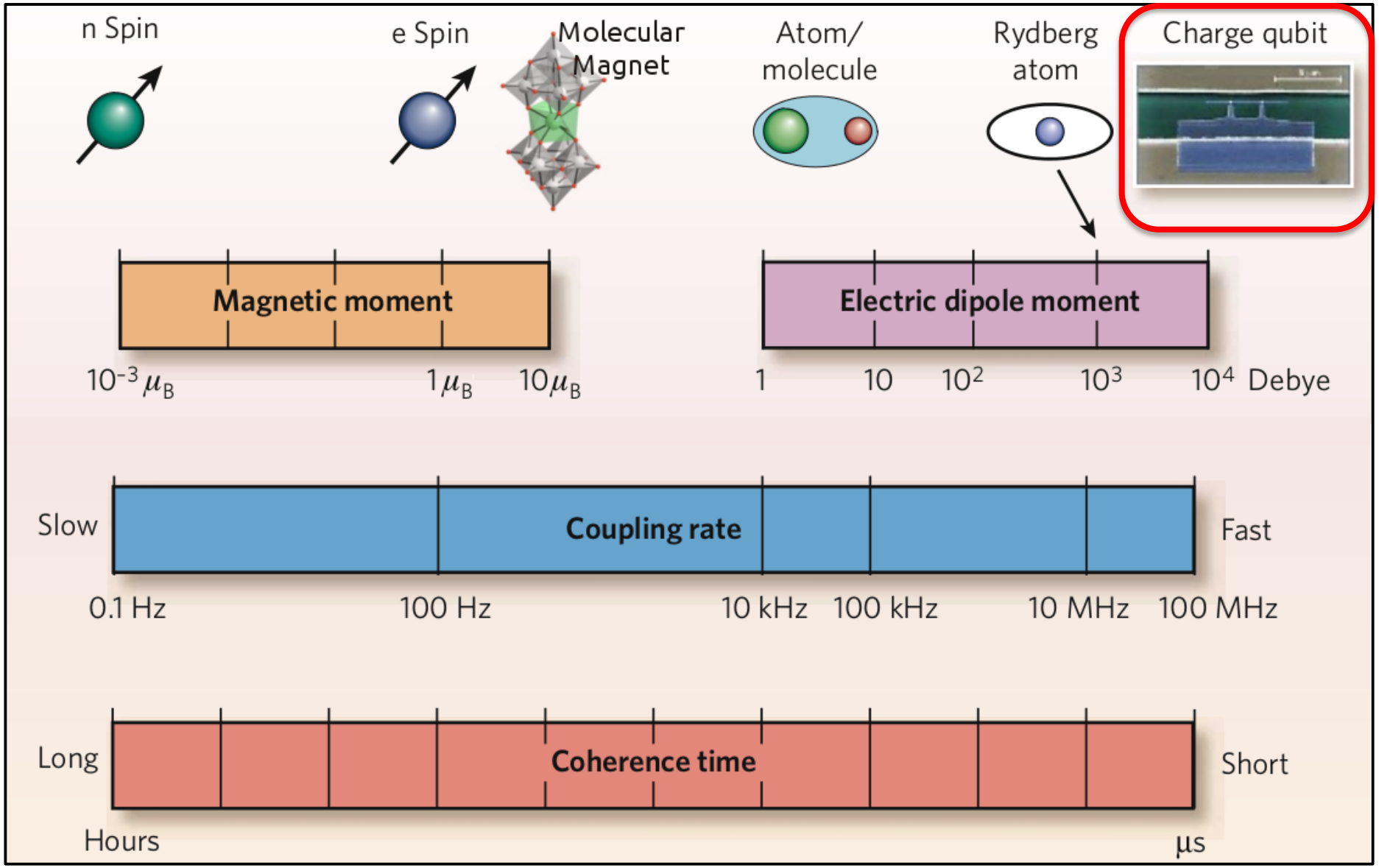
D. P. Di Vincenzo, Phys. Rev. A **51**, 1015 (1995)



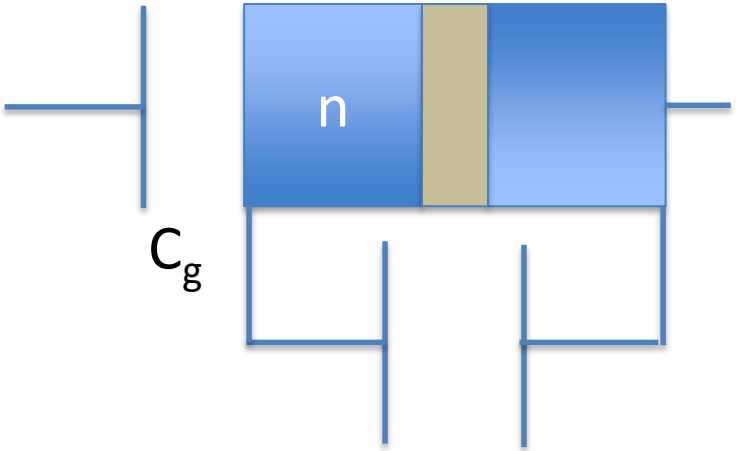
- Two well-defined states + initialization
- External control of the wave function
- Sufficiently high quantum coherence $T_2\Omega_R >> 1$ (≥ 100)
- Scalability: wire up ≥ 2 qubits

Material qubits

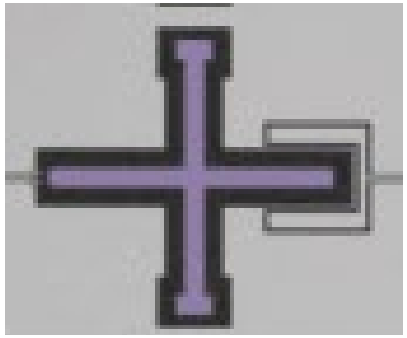
R. J. Schoelkopf & S. M. Girvin, Nature **451**, 664–669 (2008)

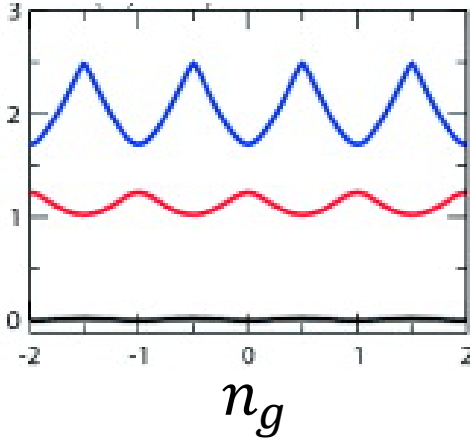


Quantum supremacy: NISQs era



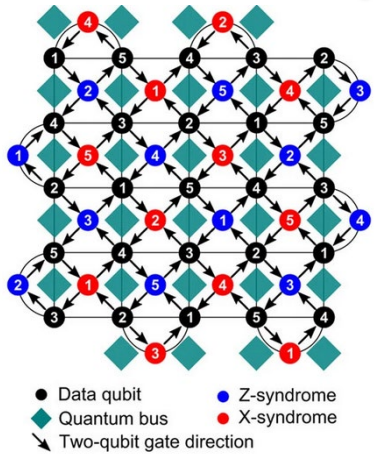
F. Arute, *et al. Nature* **574**, 505–510 (2019)





Transmon

J. A. Schreier *et al*, Phys. Rev. B **77**, 180502(R) (2008)



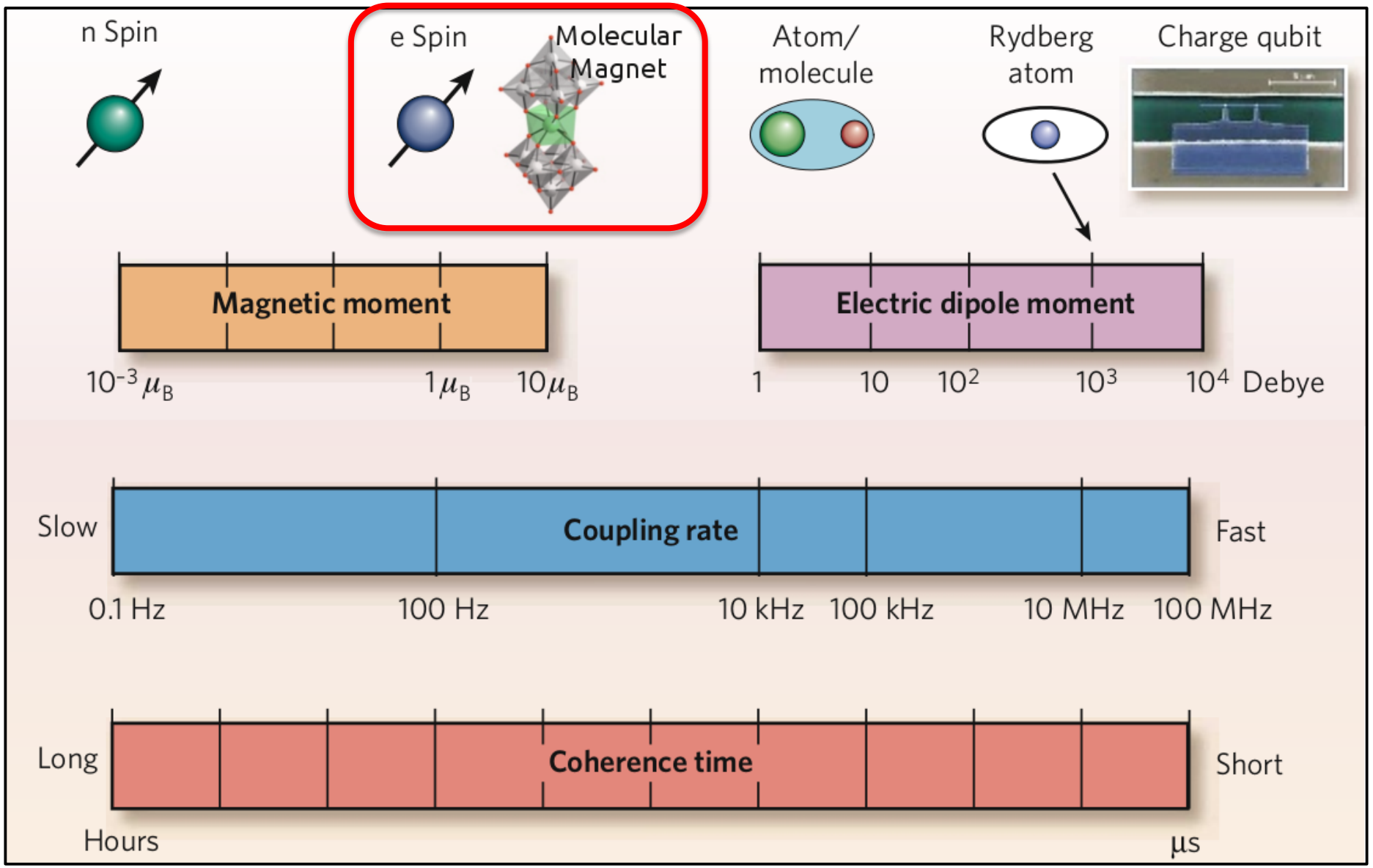
Surface QEC code (>25 qubits per logical qubit)

Shor code: 10^8 qubits

A. Fowler, Phys. Rev. A **86**, 032324 (2012)

Alternatives?

R. J. Schoelkopf & S. M. Girvin, Nature **451**, 664–669 (2008)

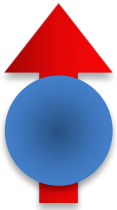


Spin qubits

Electron in a magnetic field

$S = ½$

$g = 2$



$|1\rangle \equiv |\downarrow\rangle$

$\hbar\omega_{10} = g\mu_B B_{dc}$

$|0\rangle \equiv |\uparrow\rangle$

26 GHz/T

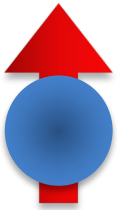
1.3 K/T

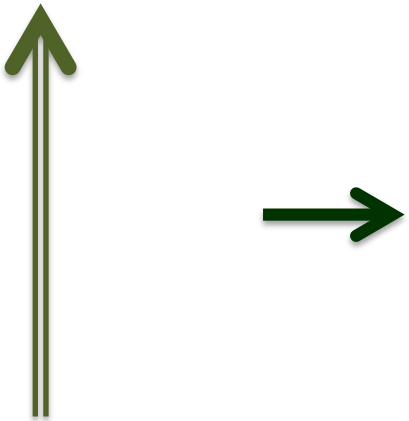
Spin qubits

Electron in a magnetic field

$S = ½$

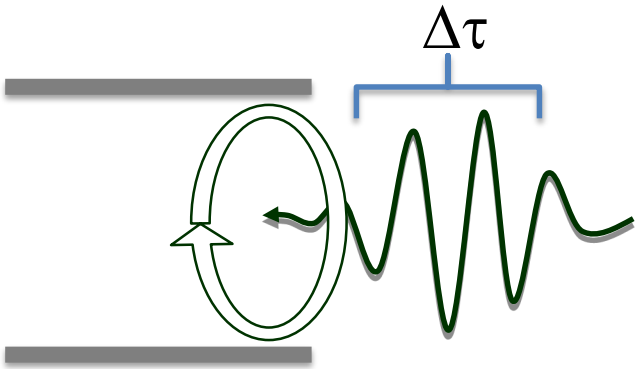
$g = 2$



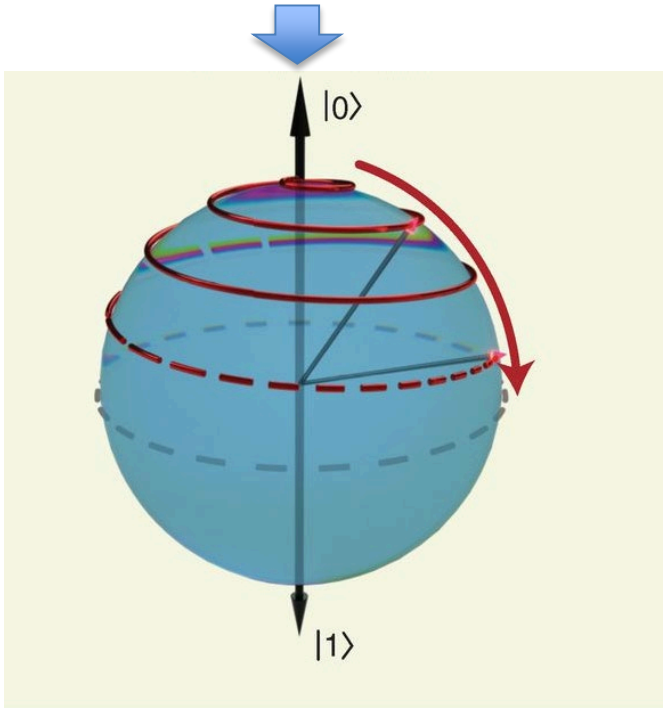


$$|1\rangle \equiv |\downarrow\rangle$$

$$|0\rangle \equiv |\uparrow\rangle$$



In resonance: $\omega = \omega_{10}$



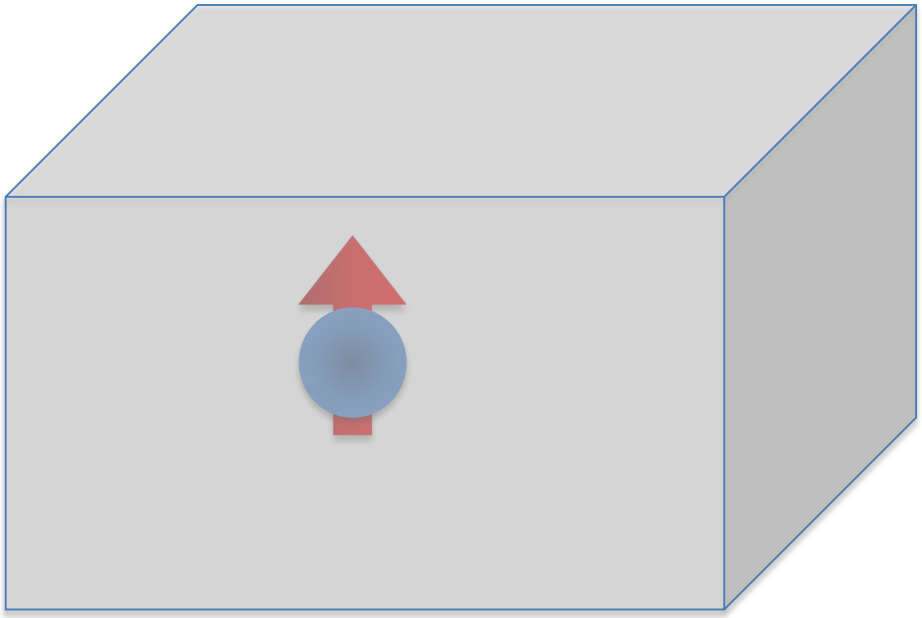
$$|0\rangle \rightarrow \cos\left(\frac{\Omega_R t}{2}\right)|0\rangle + e^{i\phi(t)} \sin\left(\frac{\Omega_R t}{2}\right)|1\rangle$$

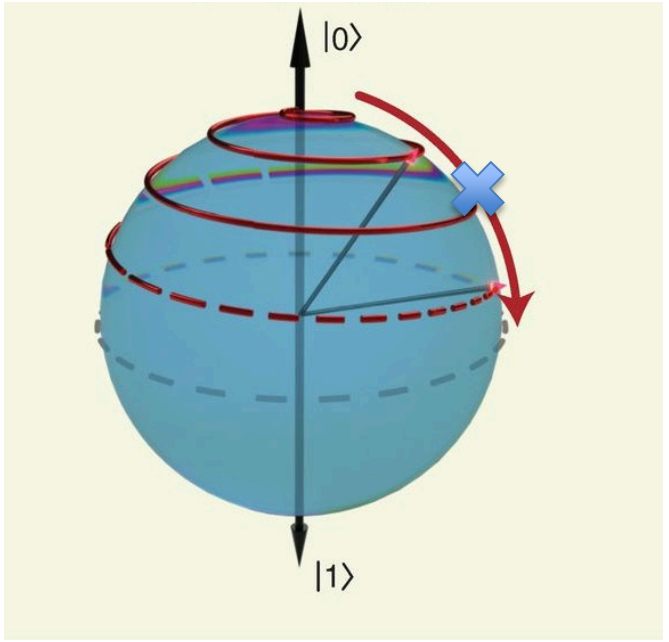
$$\frac{\Omega_R}{2\pi} = \frac{g\mu_B \langle 1|\vec{S}\vec{B}_1|0\rangle}{h} \approx 13\ \text{MHz/mT}$$

$$\phi(t) = \frac{g\mu_B B_{dc}}{\hbar} t$$

I. I. Rabi, Phys. Rev. B **51**, 652 (1937)

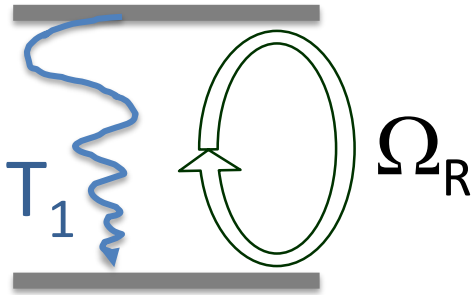
Relaxation





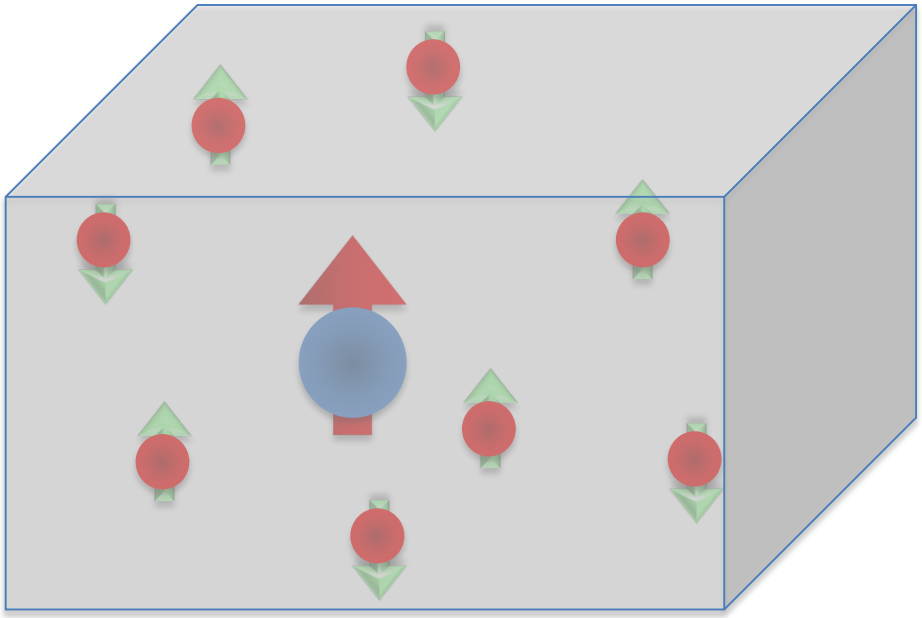
$$|1\rangle \equiv |\downarrow\rangle$$

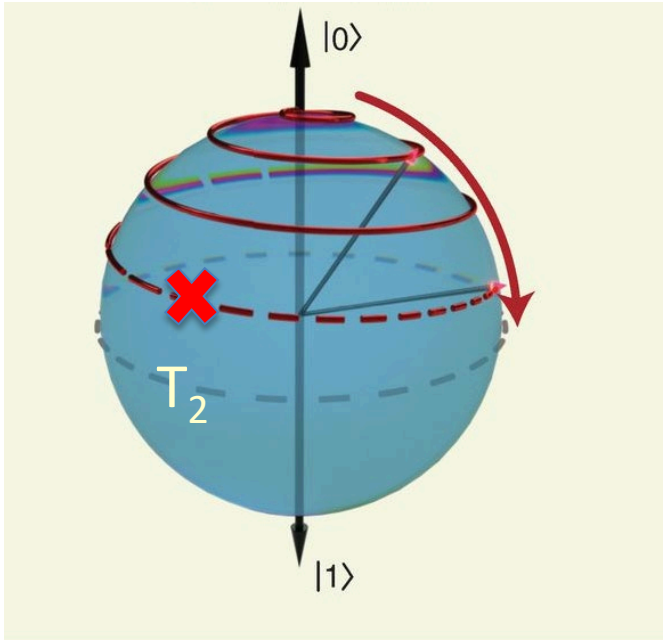
$$|0\rangle \equiv |\uparrow\rangle$$



$$|0\rangle \rightarrow \cos\left(\frac{\Omega t}{2}\right)|0\rangle + e^{i\phi(t)}\sin\left(\frac{\Omega t}{2}\right)|1\rangle$$

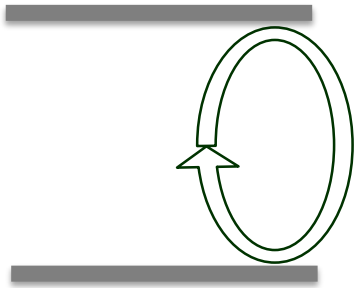
Dephasing





$$|1\rangle \equiv |\downarrow\rangle$$

$$|0\rangle \equiv |\uparrow\rangle$$



$$\Omega_R$$

$$|0\rangle \rightarrow \cos\left(\frac{\theta(t)}{2}\right)|0\rangle + e^{i\varphi(t)}\sin\left(\frac{\theta(t)}{2}\right)|1\rangle$$

$$\tau^{-1}_{\text{decoherence}} = \frac{1}{2T_1} + \frac{1}{T_2} \approx \frac{1}{T_2}$$

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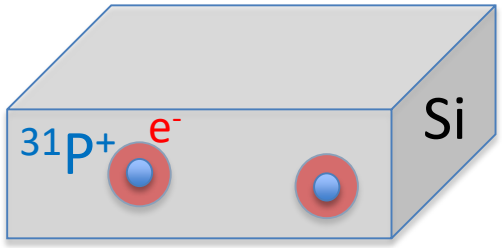
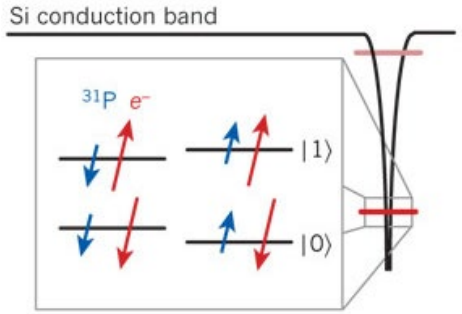
Impurity spins in semiconductors

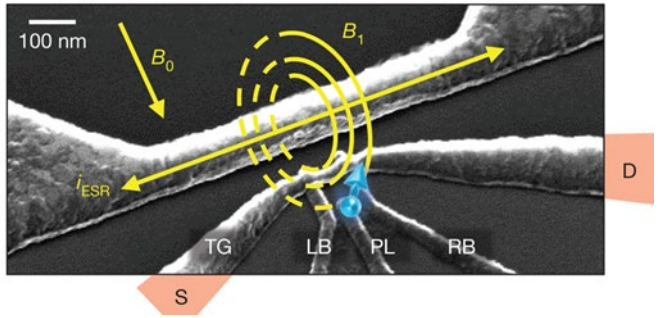
$^{31}P^{+}$ in Si

B. E. Kane, Nature **393**, 133 (1998)





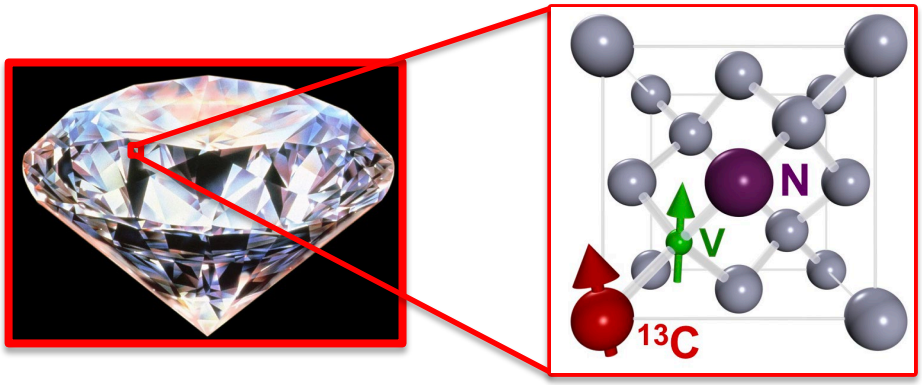
A. Morello et al., Nature **467**, 687 (2010)

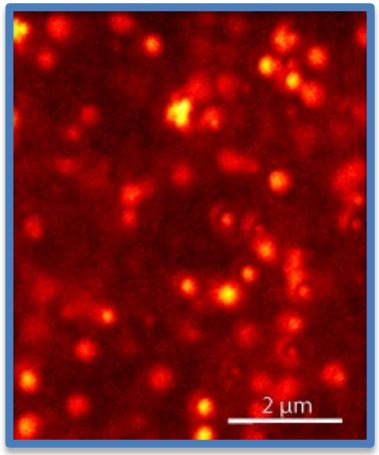


T_{2e} = 500 ms
T_{2n} = 30 s

NV^{-} in diamond

F. Jelezko et al., Phys. Rev. Lett. **92**, 076401 (2004)
G. De Lange et al., Science **330**, 60 (2010)

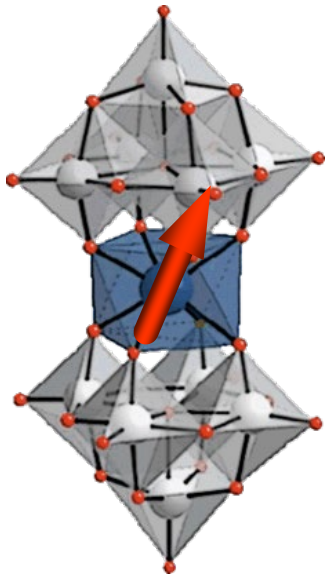




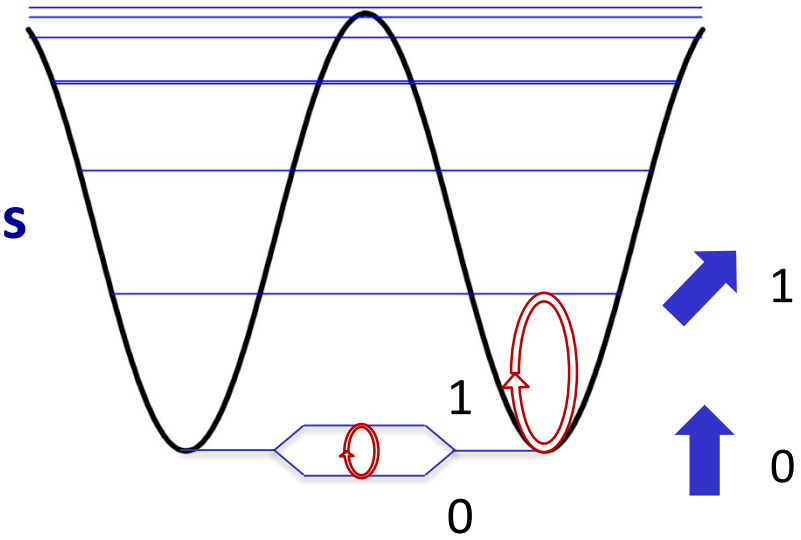
$T_2 \approx$ 20-30 µs

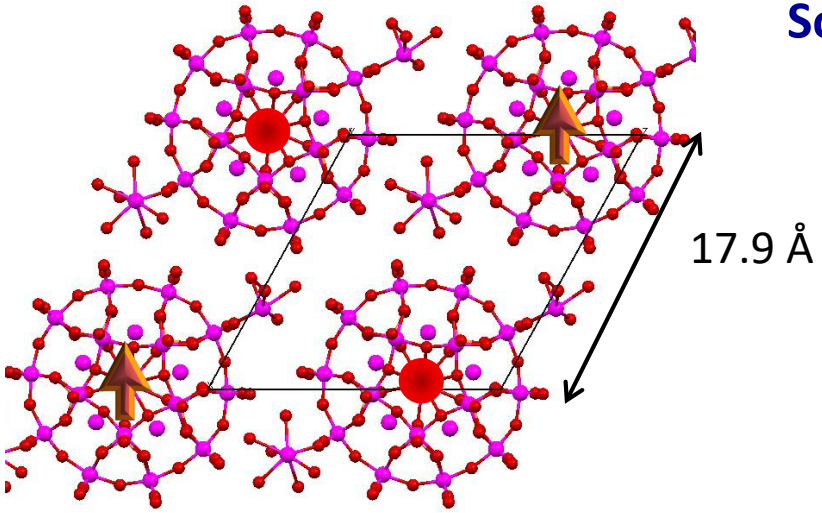
Molecular spin qubits

A. Gaita-Ariño, F. Luis, S. Hill, E. Coronado, Nature Chem. **11**, 301–309 (2019).



Single ion magnets





Some appealing properties

- Simples (1 magnetic ion)
- Weak spin-spin interactions
- Magnetically soluble

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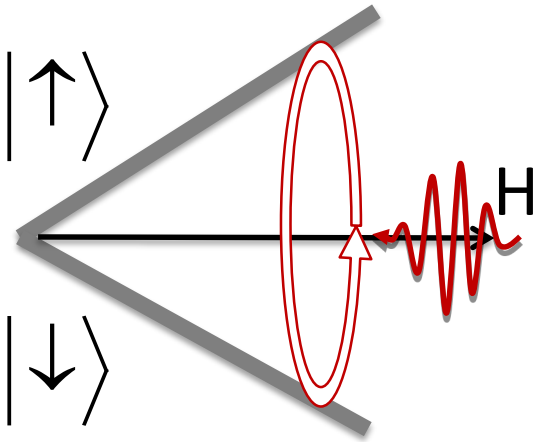
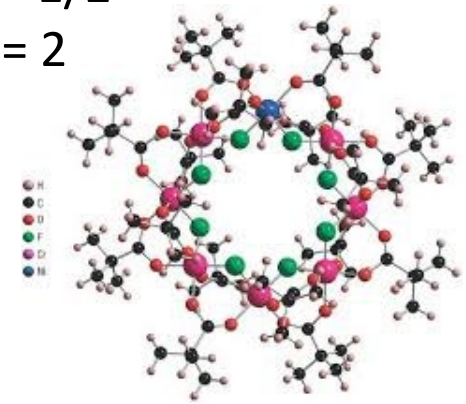
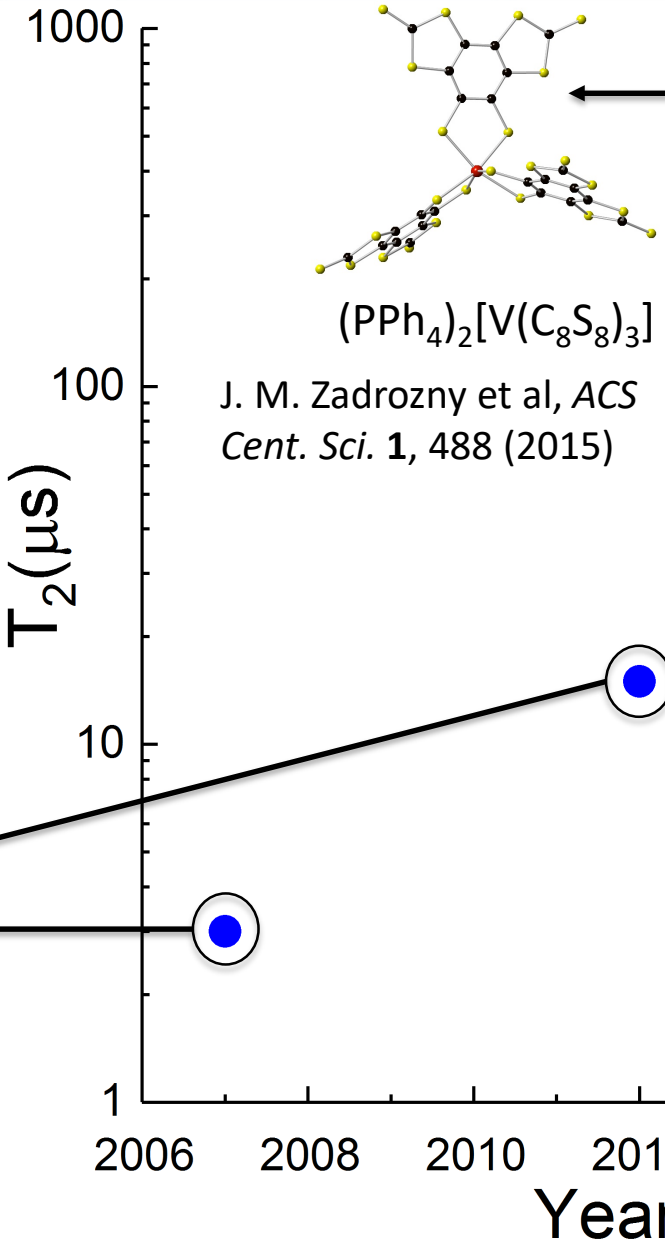
Beating decoherence



Cr_7Ni
$S = 1/2$
$g = 2$

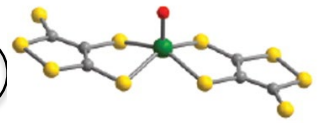


A. Ardavan et al, *Phys. Rev. Lett.* **98**, 057201 (2007); C. J. Wedge et al. *Ph Rev. Lett.* **108**, 107204 (2012).



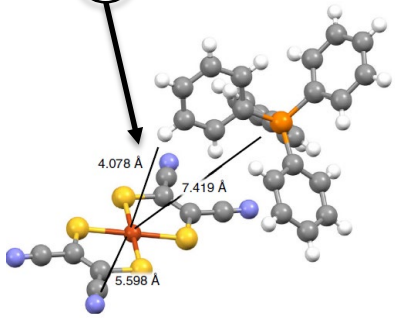
$(PPh_4)_2[V(C_8S_8)_3]$

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



2
$[VO(\beta\text{-}C_3S_5)_2]^{2-}$

C.-J. Yu et al, *JACS* **138**, 14678 (2016)

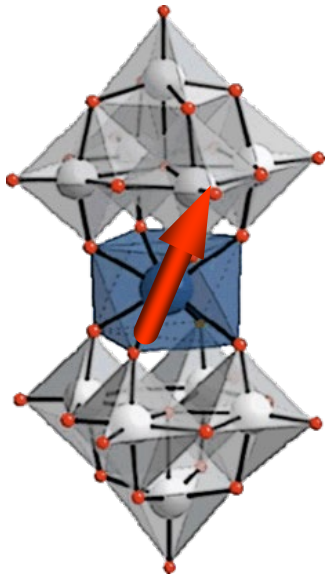


$(PPh_4)_2[Cu(mnt)_2]$

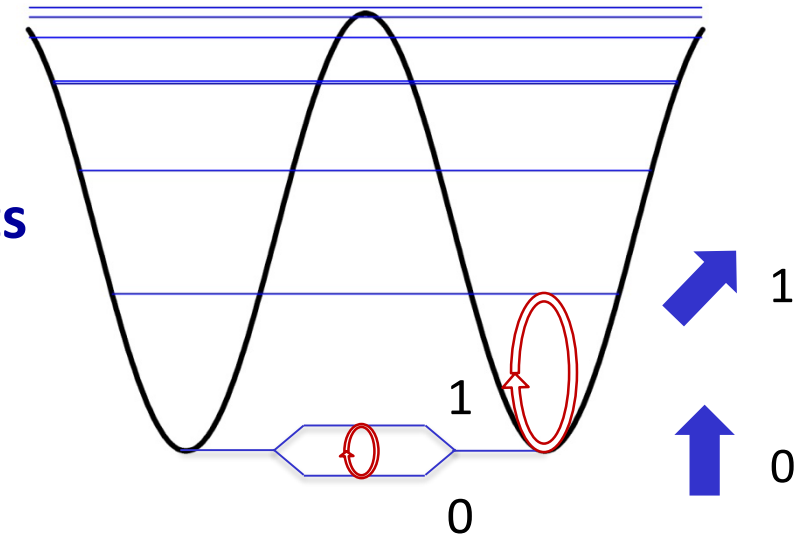
K. Bader et al, *Nat. Commun.* **5**, 5304 (2014).

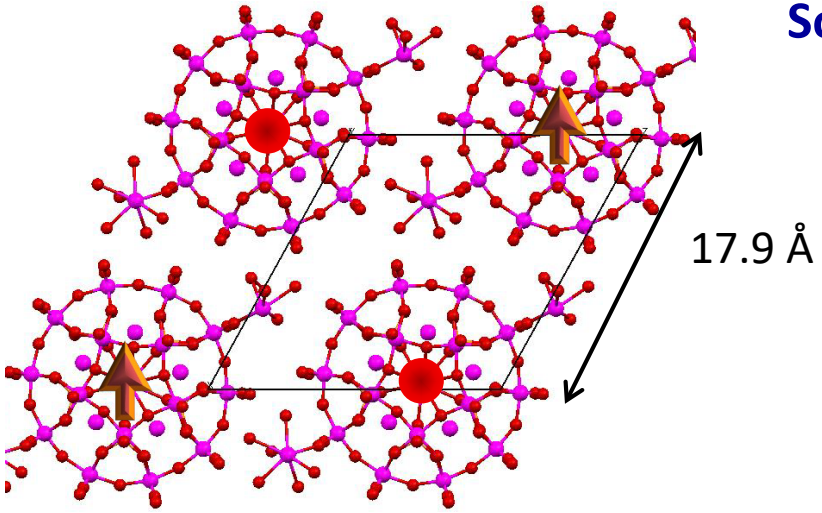
Molecular spin qubits

A. Gaita-Ariño, F. Luis, S. Hill, E. Coronado, Nature Chem. **11**, 301–309 (2019).



Single ion magnets





Some appealing properties

- Simples (1 magnetic ion)
- Weak spin-spin interactions
- Magnetically soluble
- Tunable

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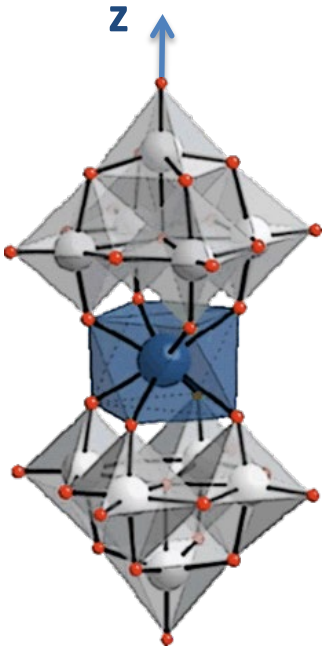
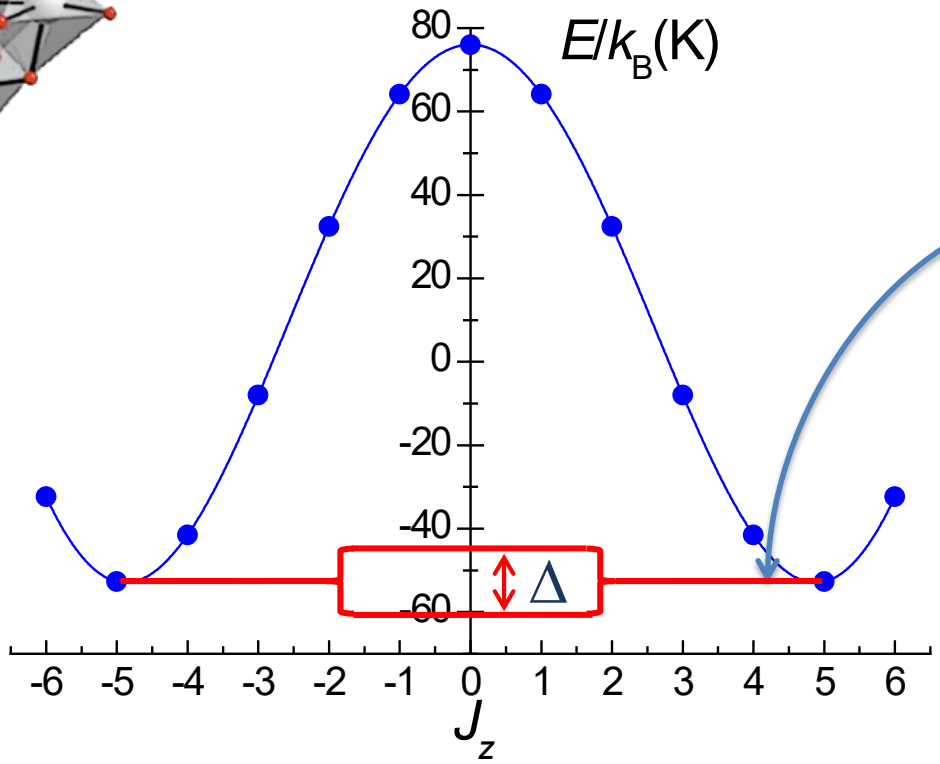
Molecular design: qubit parameters



$$\mathrm{H} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_n^m O_n^m$$

Off-diagonal terms

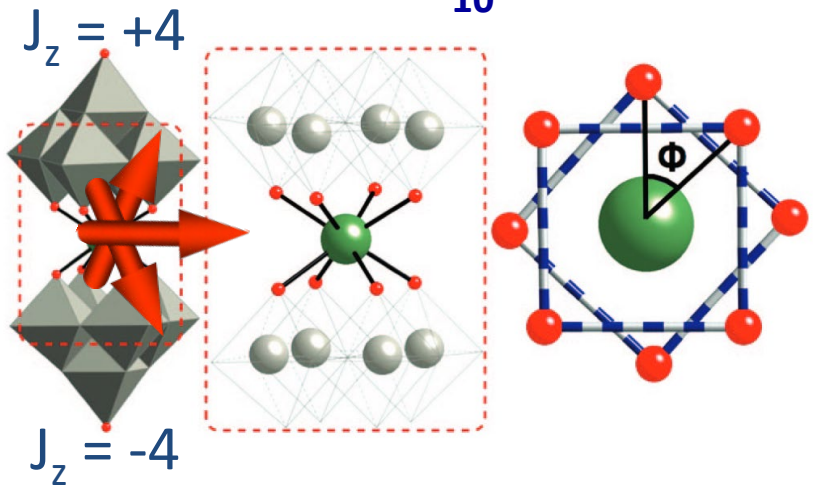
$$\left[J_z, O_m^0\right] \neq 0$$



Quantum tunnel splitting

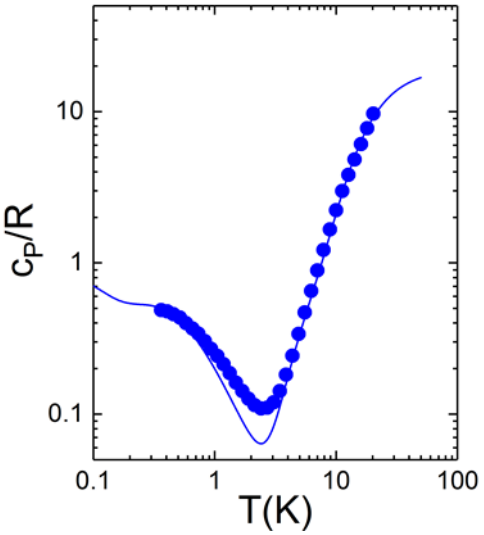
Spin "clock transitions"

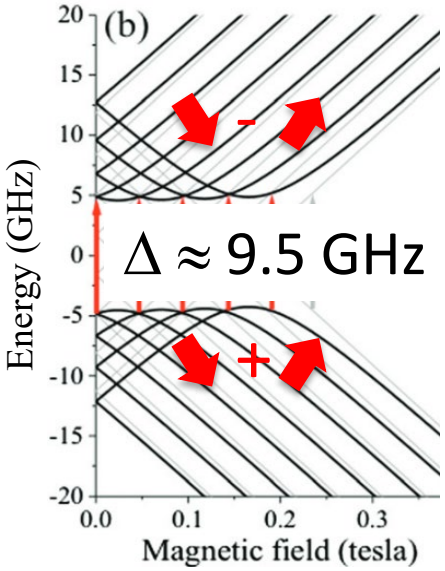
HoW_{10}



$$H = BO_n^0 + B_4^4 O_4^4$$

M. AlDamen et al, Inorg. Chem. **48**, 3467 (2009)





Decoherence-free qubits

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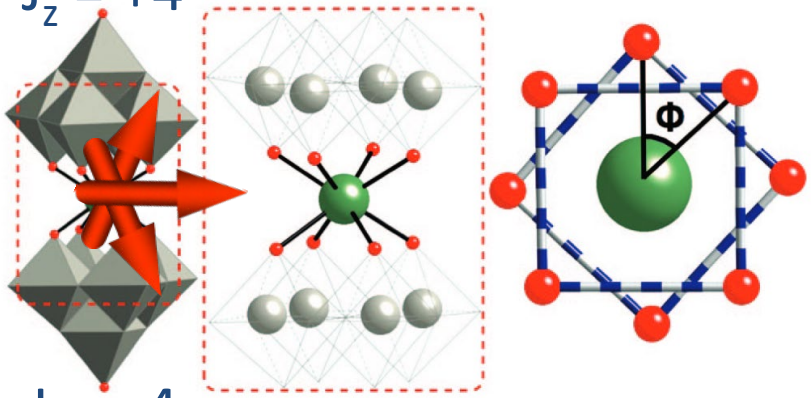
HoW_{10}

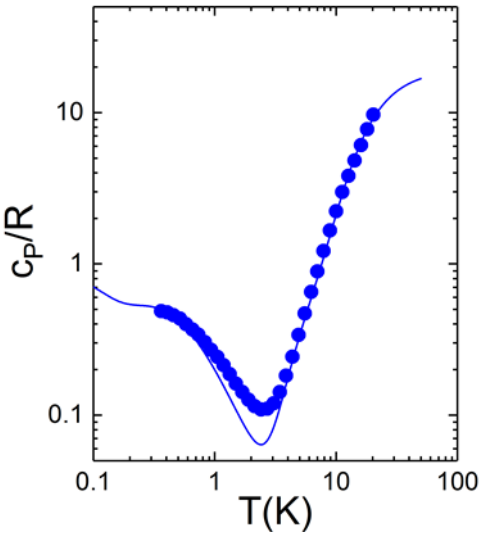
M. AlDamen et al, Inorg. Chem. **48**, 3467 (2009)

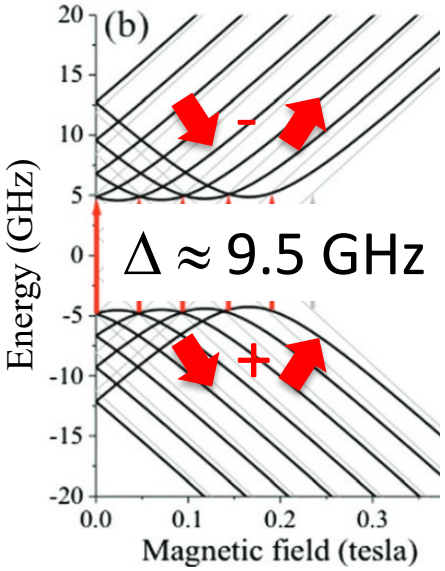
$J_z = +4$



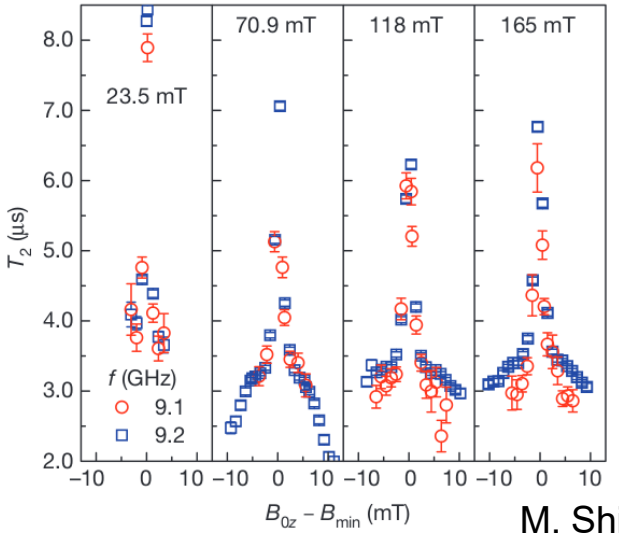
$J_z = -4$

$$H = BO_n^0 + B_4^4 O_4^4$$





$$\frac{1}{T_2} \propto \frac{\partial \omega_{10}}{\partial B}$$



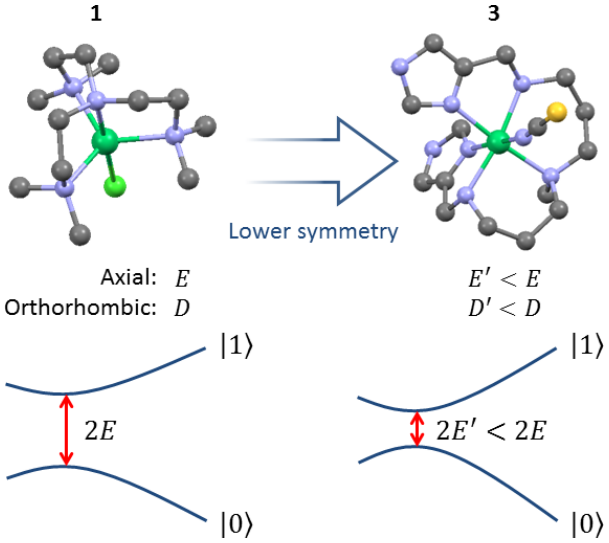
M. Shiddiq et al, Nature **531**, 348 (2016)

Tuning the gap: proper coordination

[Ni(Me$_6$tren)Cl](ClO$_4$)

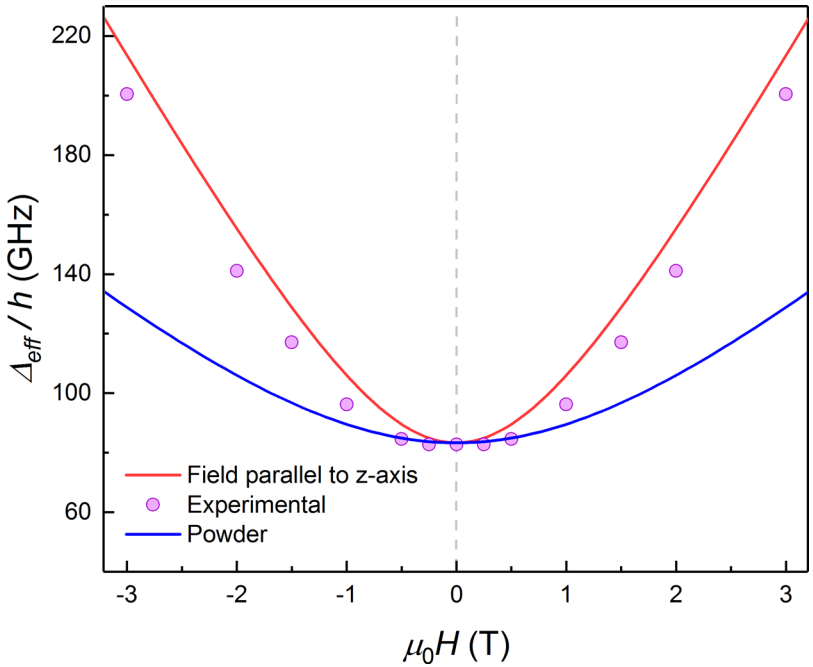
Ni(II) $S = 1$

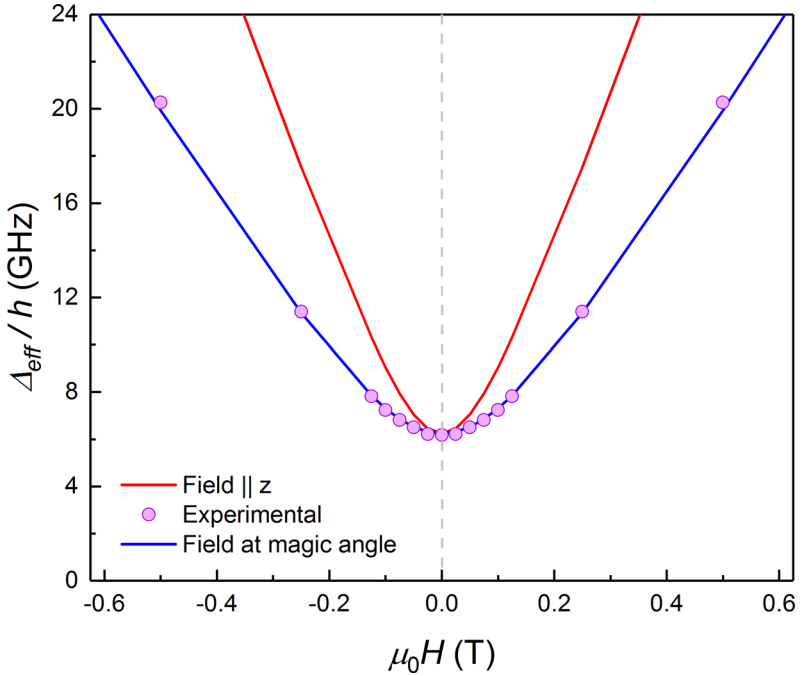
$$\mathrm{H} = BO_2^0 + B_2^2O_2^2$$



[Ni-Imdipa]-(NCS)

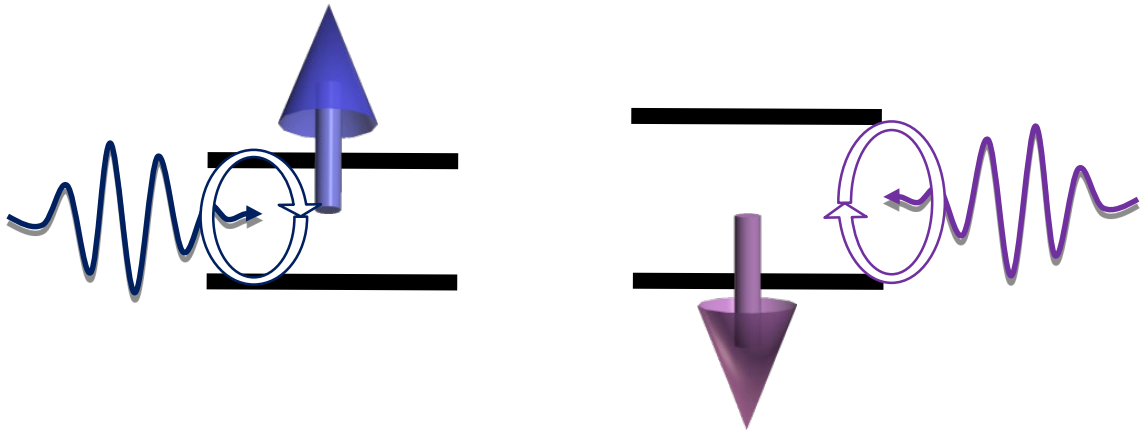
M. Rubín et al, Chem. Sci. **12**, 5123 (2021)

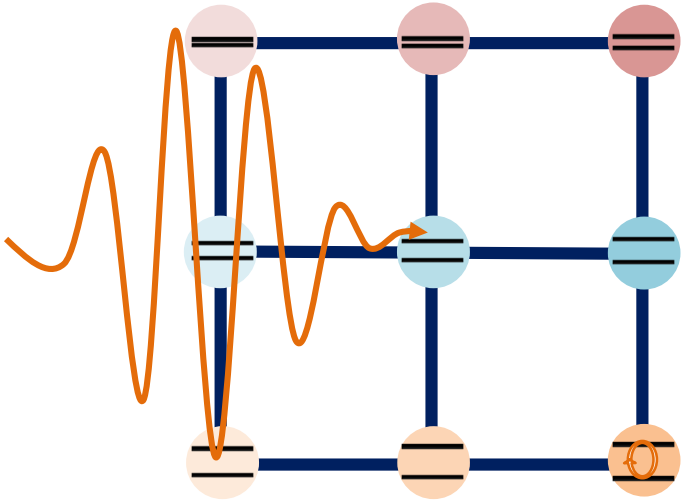




Scaling-up by molecular design

Spectroscopic addressing





S. Carretta et al, Appl. Phys. Lett. **118**, 240501 (2021)

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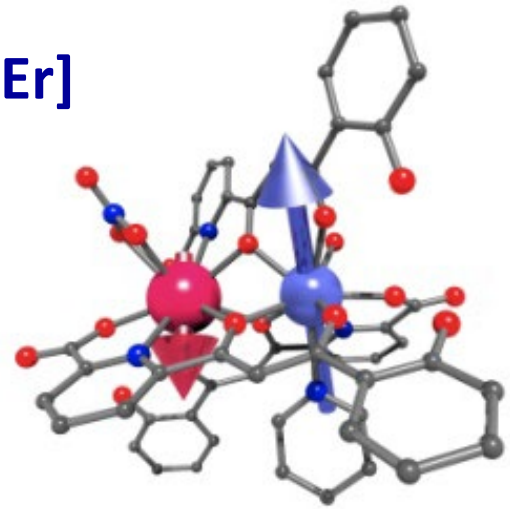
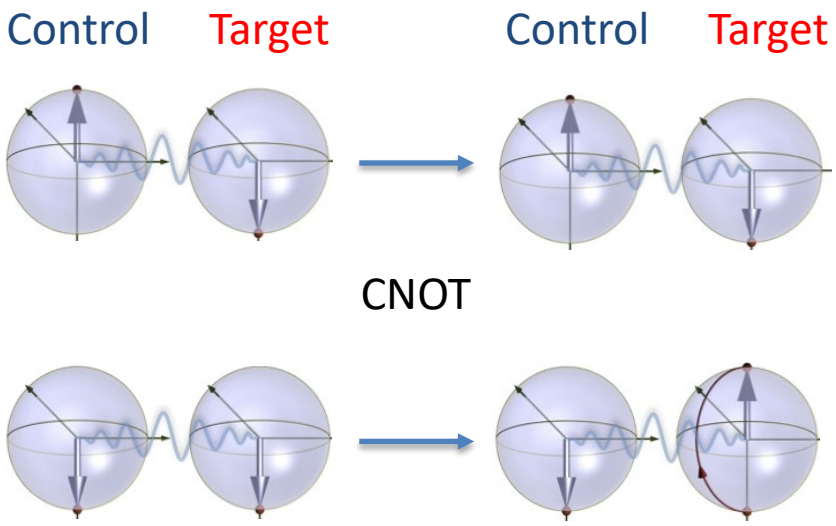
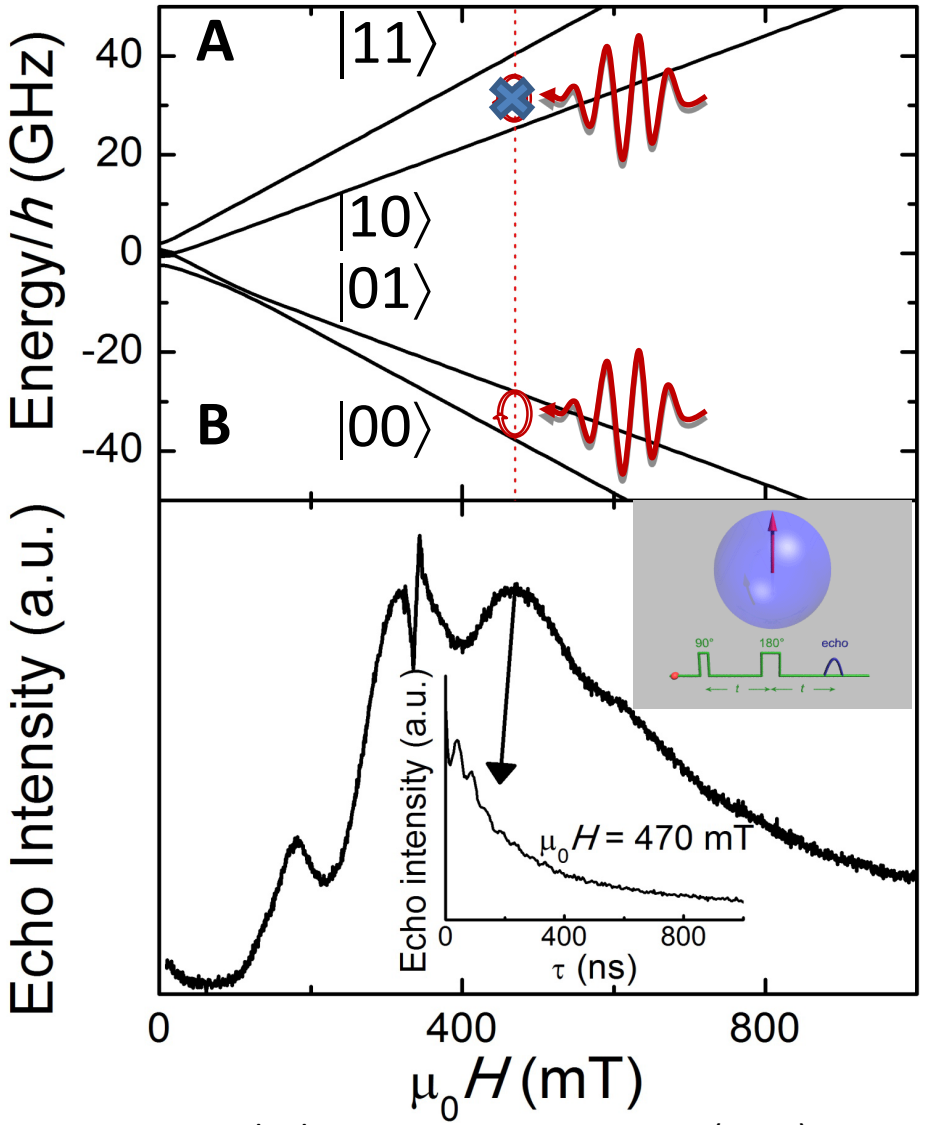
[CeEr]: CNOT quantum gate

[CeEr]



$$\mathcal{H} = g_1\mu_B\vec{H}\vec{S_1} + g_2\mu_B\vec{H}\vec{S_2} - J\vec{S_1}\vec{S_2}$$





F. Luis et al, Phys. Rev. Lett. **107**, 117203 (2011);
D. Aguilà et al., JACS **136**, 14215 (2014)

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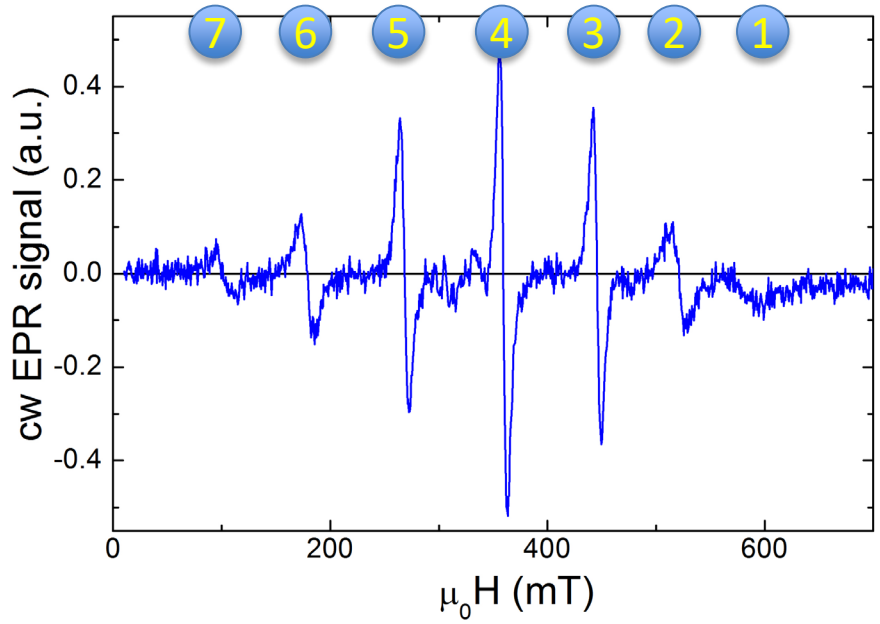
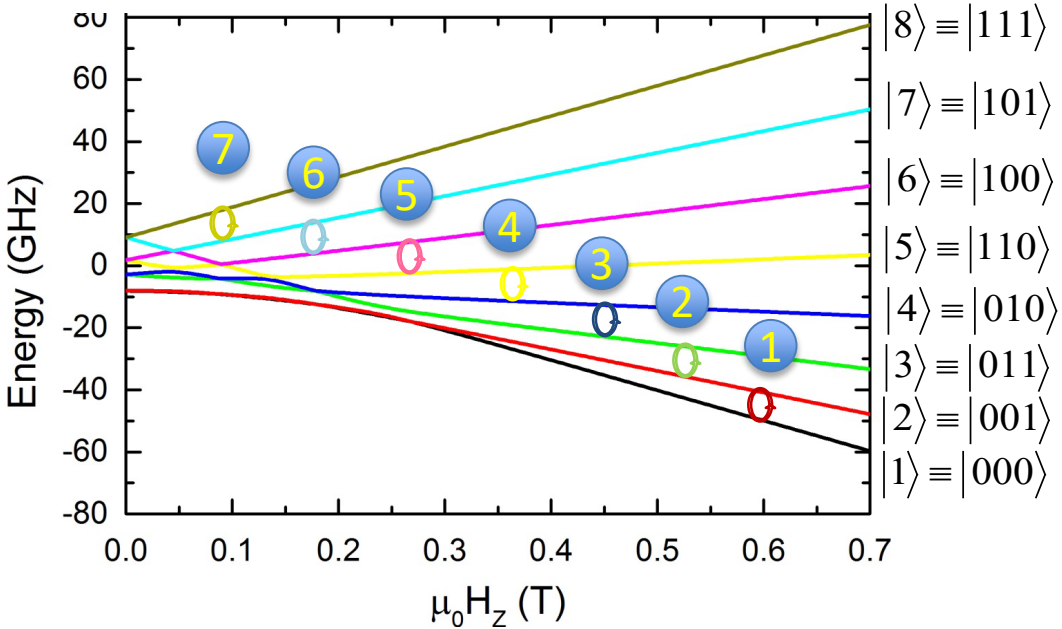
Internal states: qubits ⇒ qudits

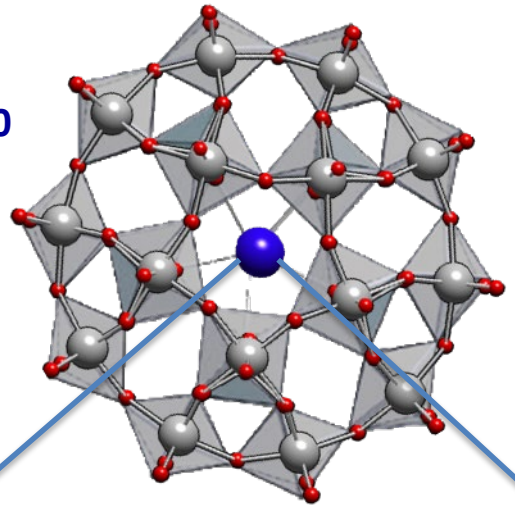




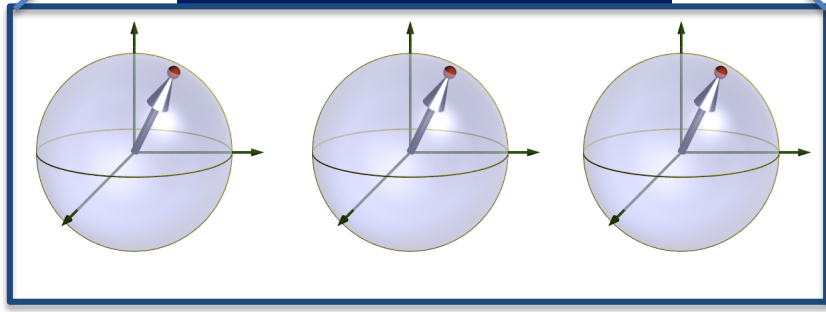
M. Jenkins et al, *Phys. Rev. B* **95**, 064423 (2017)

GdW_{30}

$S = 7/2$



$2S + 1 = 8 = 2^3$ states



Three qubits

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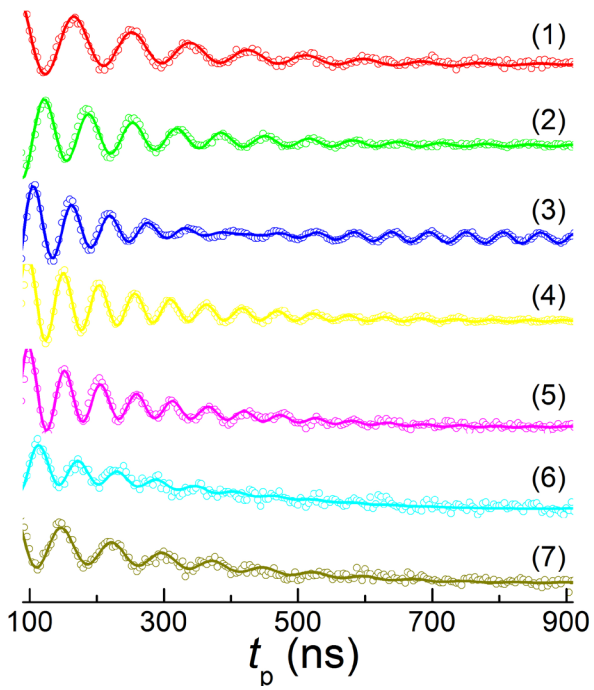
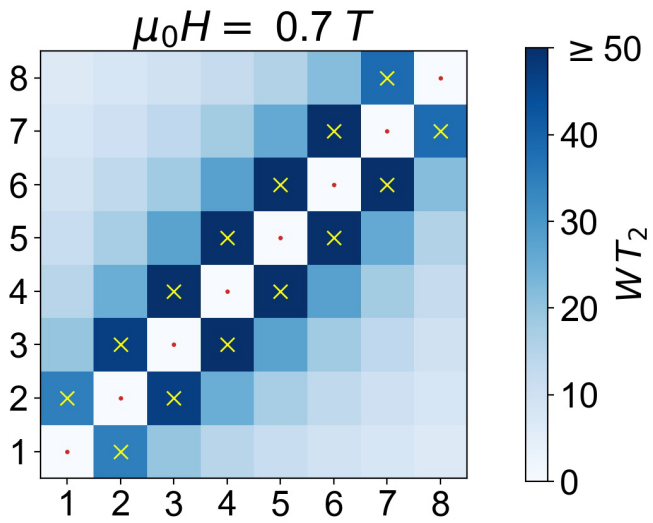
Universality: a molecular NISQ

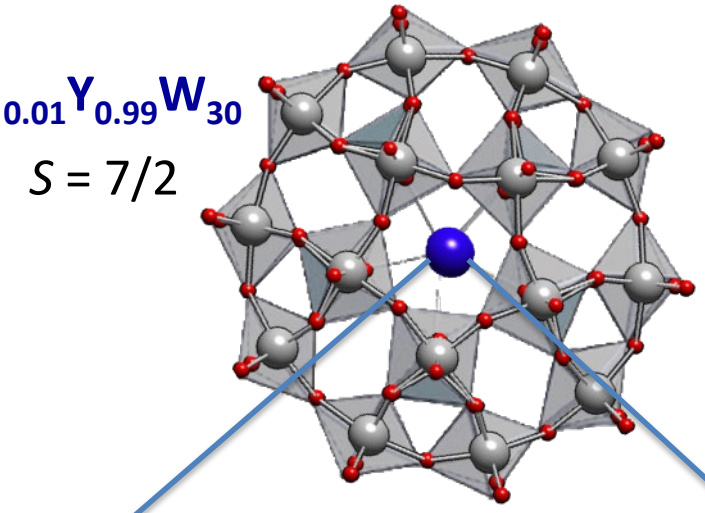




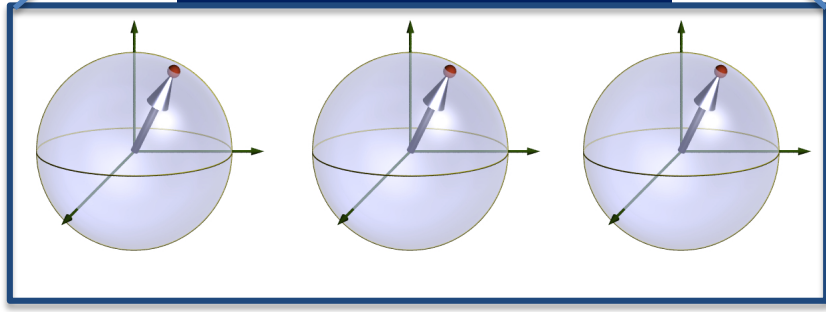
M. Jenkins et al, *Phys. Rev. B* **95**, 064423 (2017)

$Gd_{0.01}Y_{0.99}W_{30}$

$S = 7/2$



$2S + 1 = 8 = 2^3$ states



Three qubits

+

Universal set of operations

Quantum error correction in a molecular NISQ

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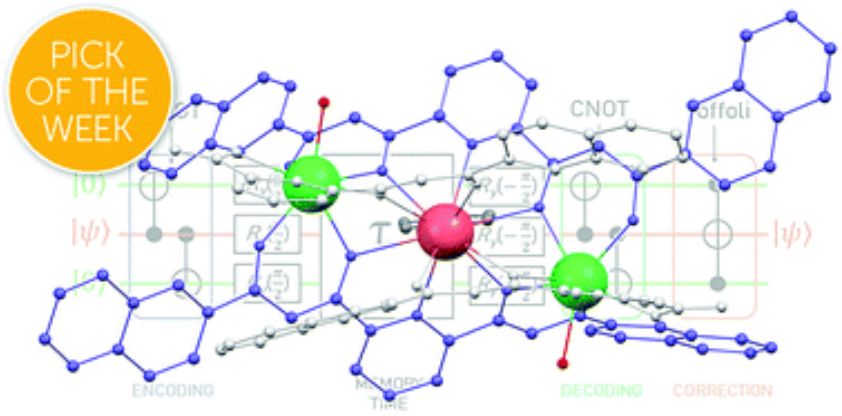
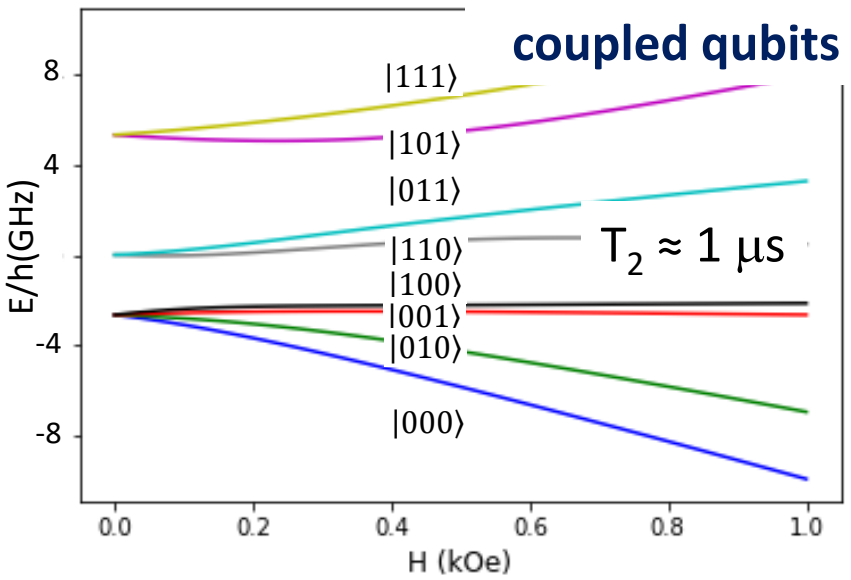
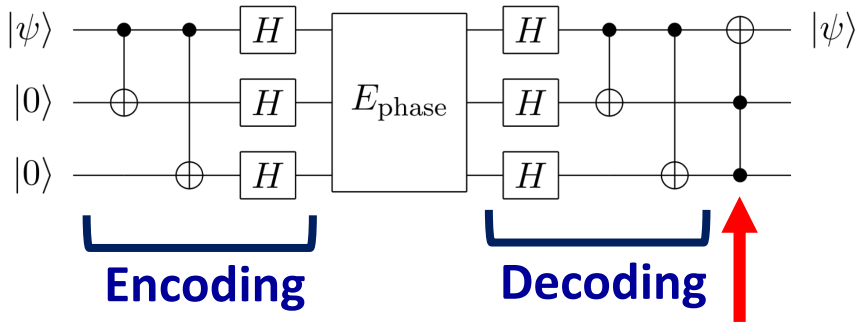
[ErCeEr]



Three different & coupled qubits

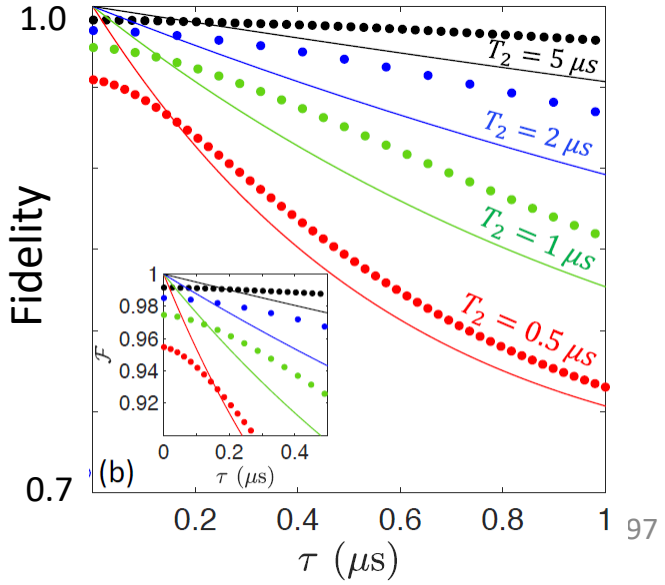


Three-qubit repetition code



E. Macaluso *et al*, Chem. Sci. **11**, 10337 (2020)

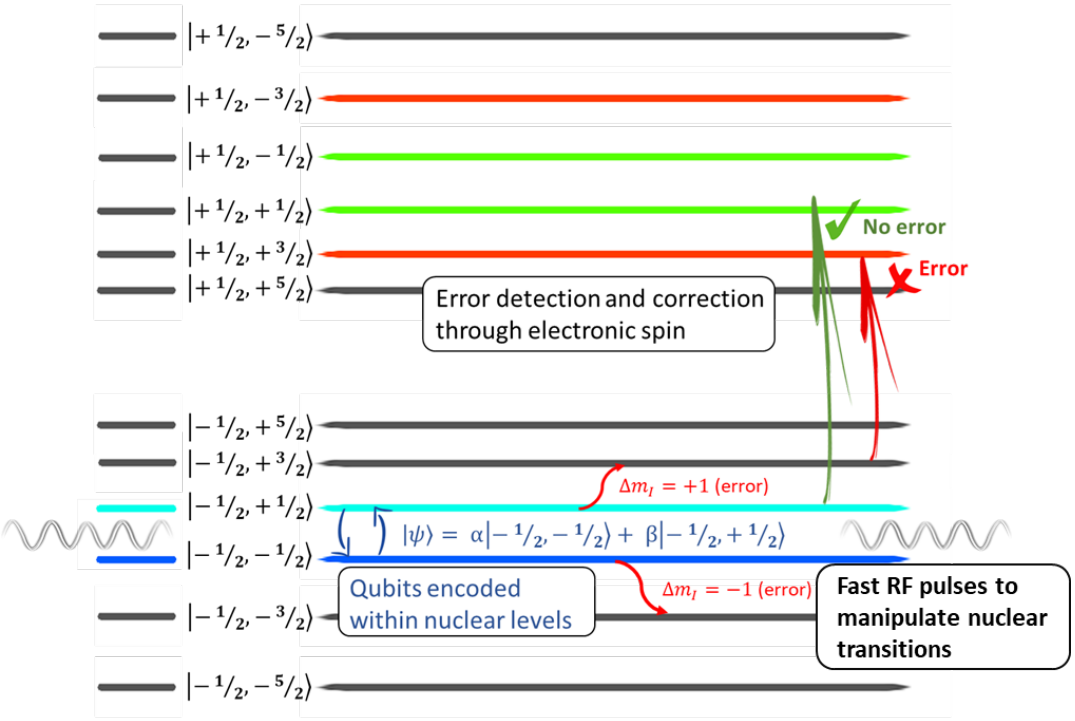
Error correction

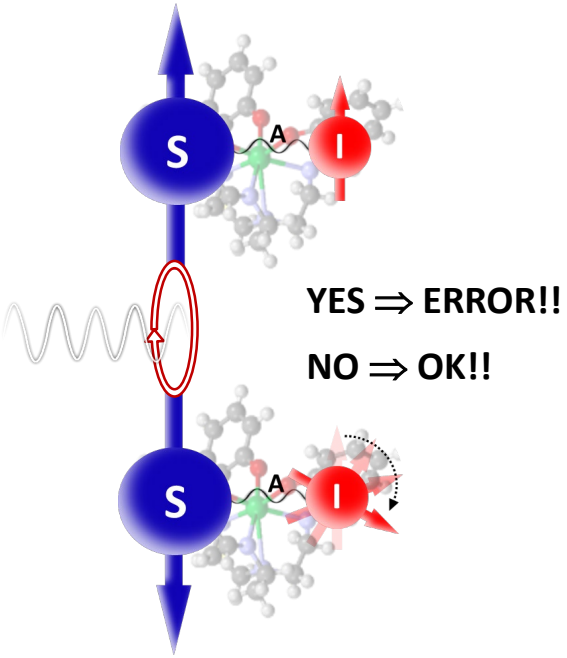


Qudit codes

NUCLEAR SPIN → **ENCODES LOGIC QUBIT STATES**

ELECTRONIC SPIN → **DETECTS ERRORS**





A single molecule can perform as a logical qubit with embedded error correction

R. Hussain *et al. J. Am. Chem. Soc.* **140** (2018)

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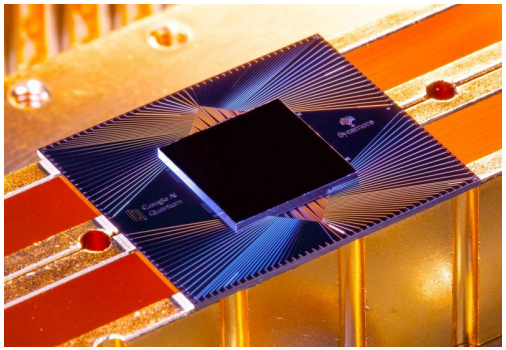
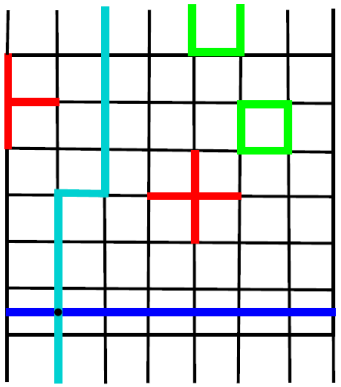
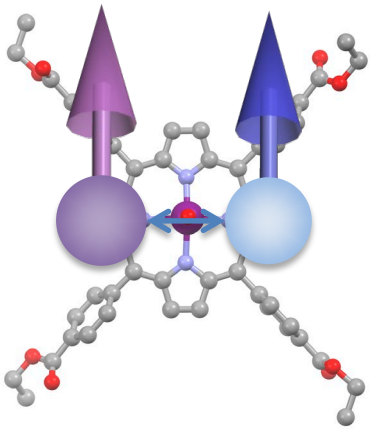
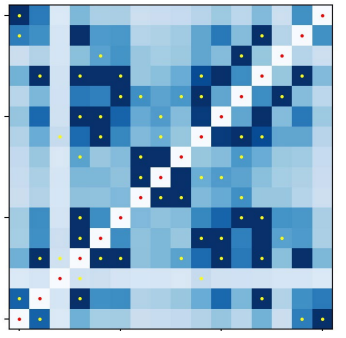
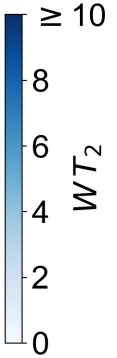
Competitive advantages & challenges











- Tuneable and interconectable
- Well-established protocols and technologies for control & read-out
- Challenging for large-scale computing

- Tuneable small and reproducible
- Multiply connected qubits/qudits in each molecular unit
- Specific algorithms save resources
- Control, read-out & wire up multiple molecules

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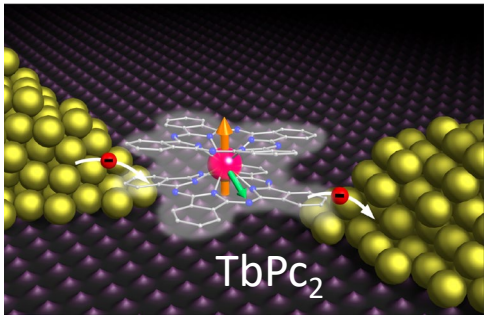
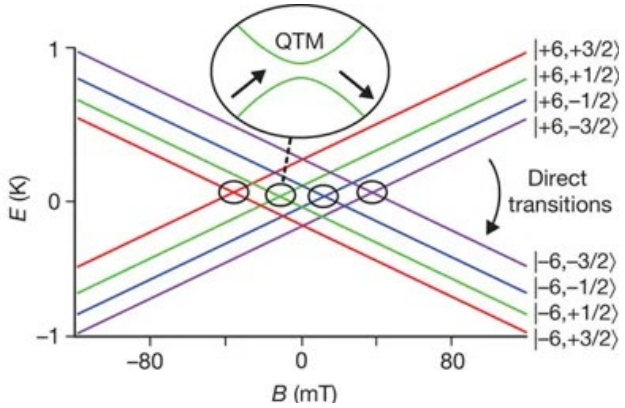
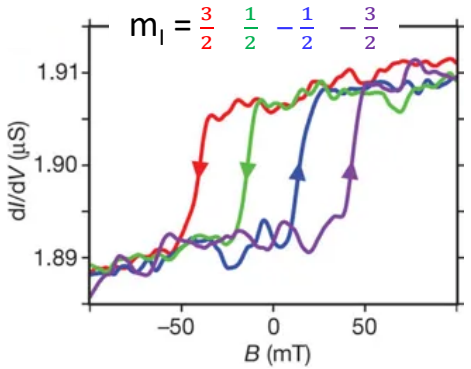
Read-out and coherent control of individual nuclear spins

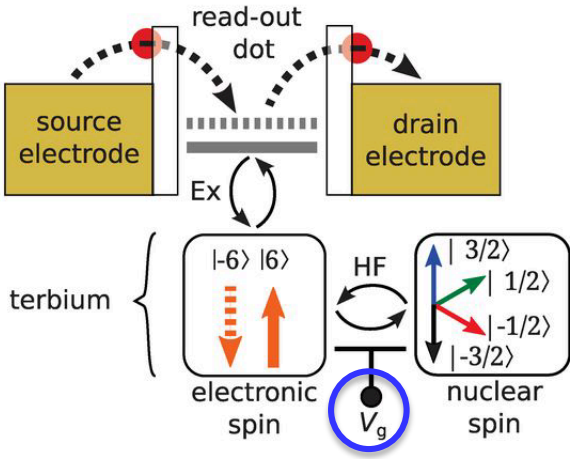




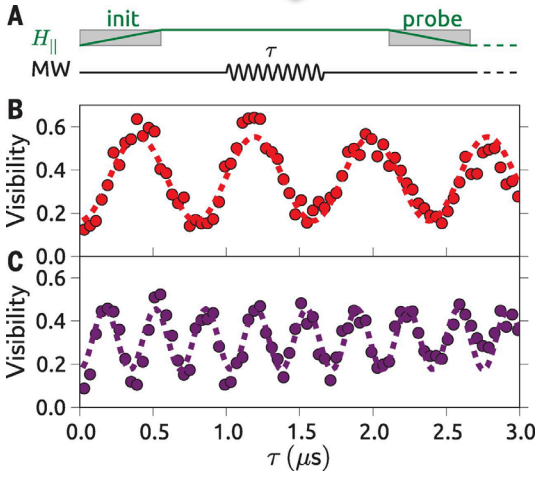


Nuclear spin read-out

R. Vincent et al, *Nature* **488**, 357 (2012)



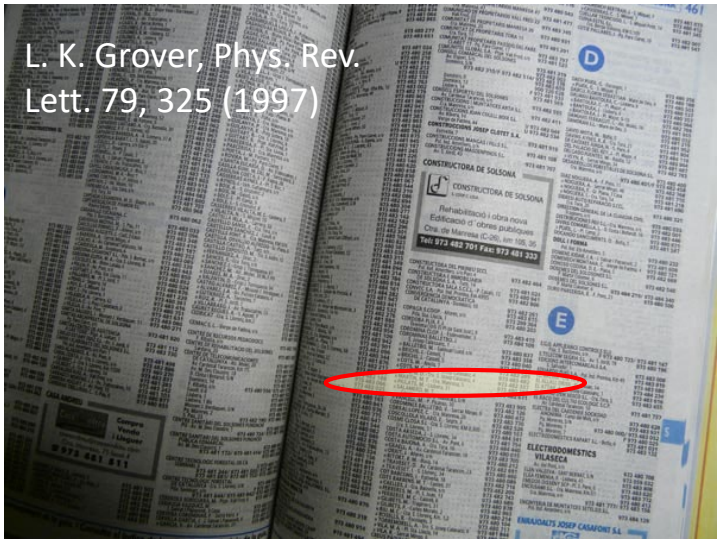
Electrical modulation of the hyperfine interaction



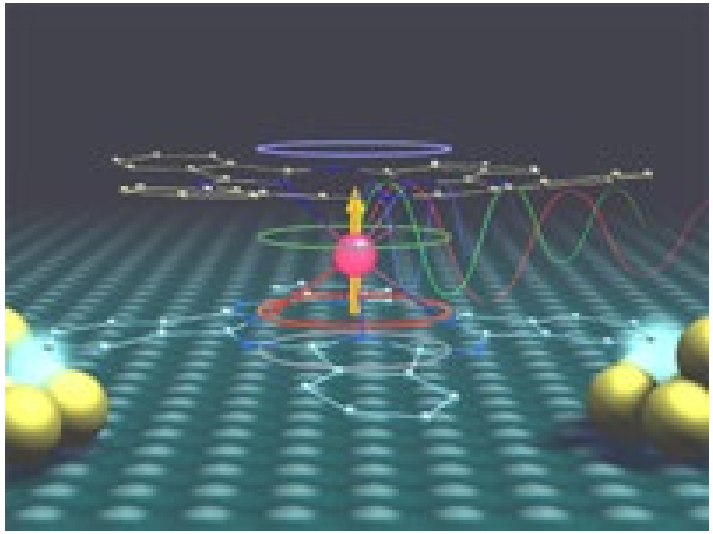
Coherent control

S. Thiele et al, *Science* **344**, 1135 (2014)

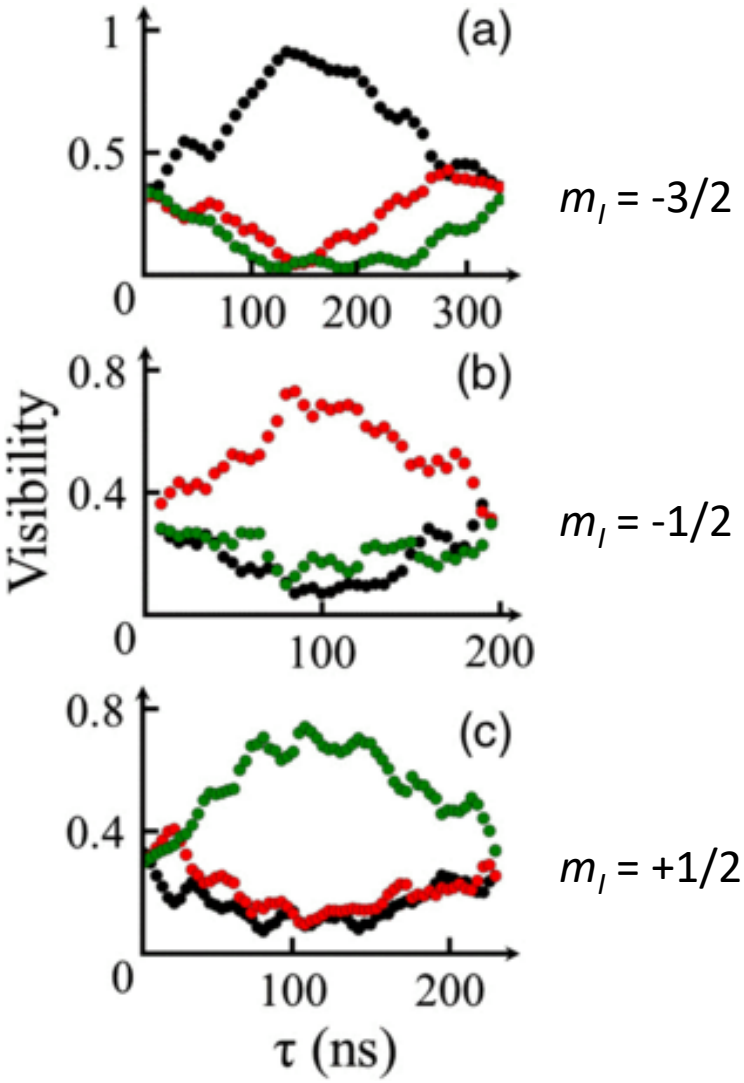
Grover search algorithm in a single molecule



Phone number ⇒ name??



Nuclear spin ⇒ state??

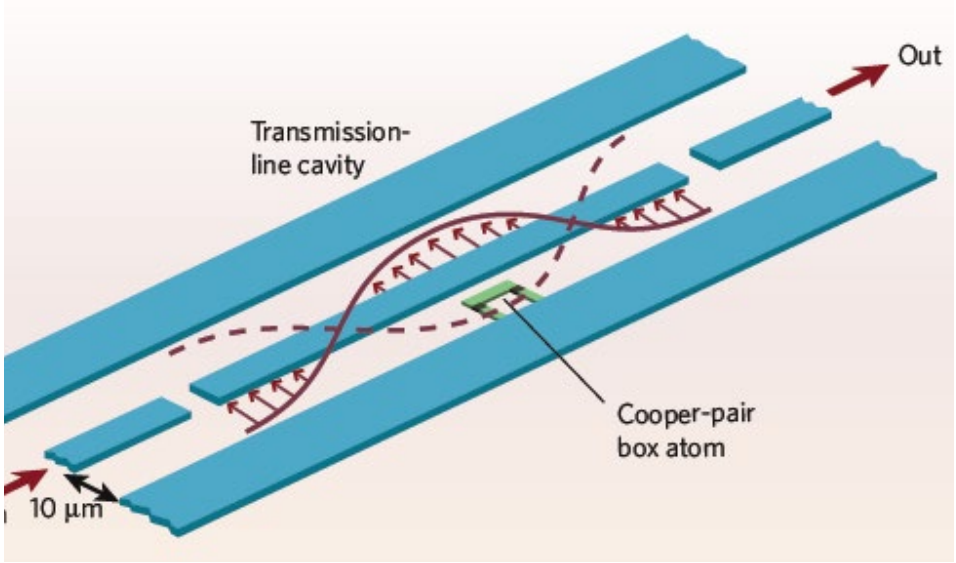


C. Godfrin et al, *Phys. Rev. Lett.* **119**, 187702 (2017)

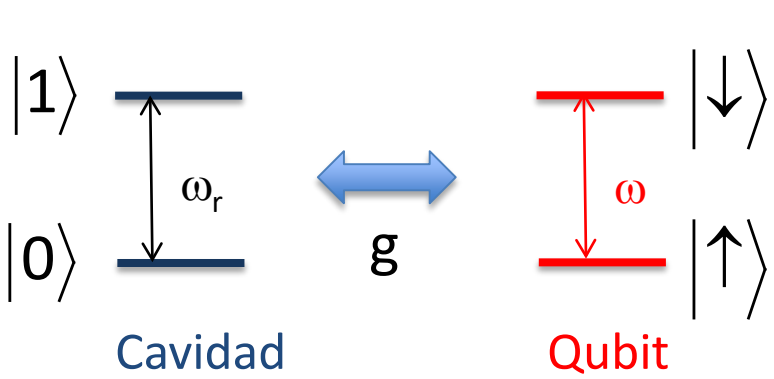
Circuit QED: "wiring-up" qubits

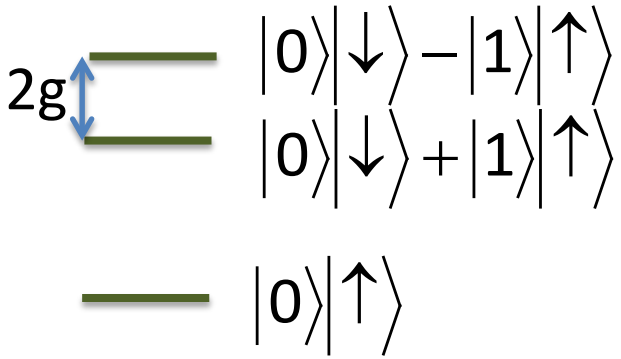
A. Blais, R.-S. Huang, A. Wallraff, S. Girvin, & R. Schoelkopf, Phys. Rev. A **69**, 062320 (2004)
A. Wallraff et al., Nature (London) **431**, 162 (2004).

Light-matter interaction in a chip



$$\mathcal{H} = \hbar\omega_r(n + 1/2) + \frac{\hbar\omega}{2}\sigma_z + \hbar g(a^+\sigma^- + a^-\sigma^+)$$





$g\ T_2 >> 1$ "Strong coupling"

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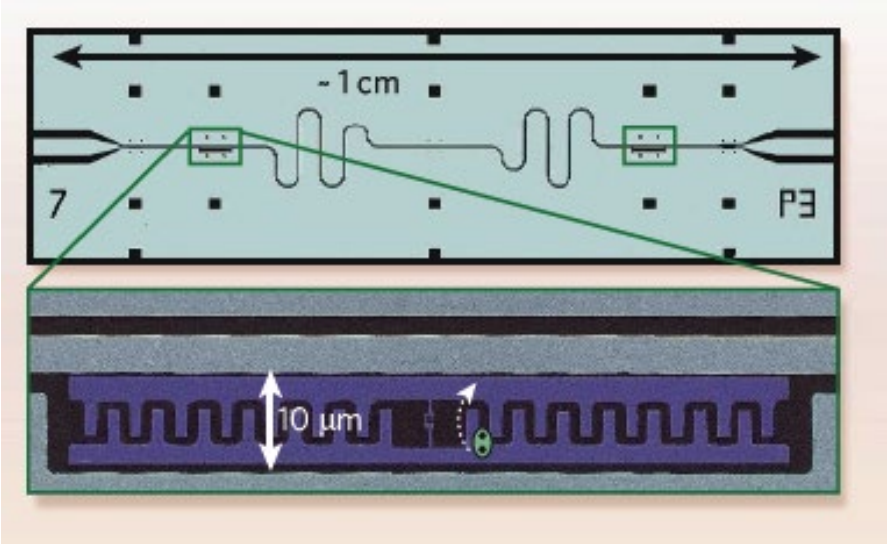
Circuit QED: "wiring-up" qubits

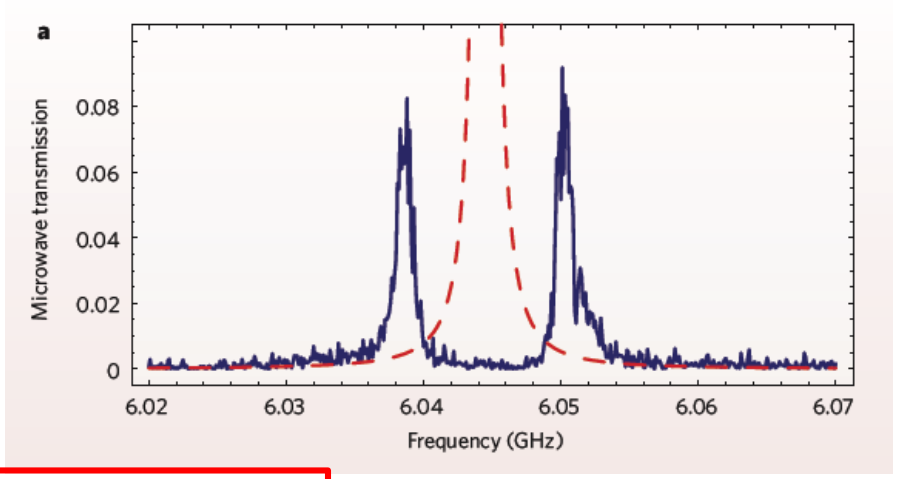
A. Blais, R.-S. Huang, A. Wallraff, S. Girvin, & R. Schoelkopf, Phys. Rev. A **69**, 062320 (2004)
A. Wallraff et al., Nature (London) **431**, 162 (2004).



Light-matter interaction in a chip

$$\mathcal{H} = \hbar\omega_r(n + 1/2) + \frac{\hbar\omega}{2}\sigma_z + \hbar g(a^+\sigma^- + a^-\sigma^+)$$

- Read-out
- Effective couplings



g T_2>>1 "Strong coupling"

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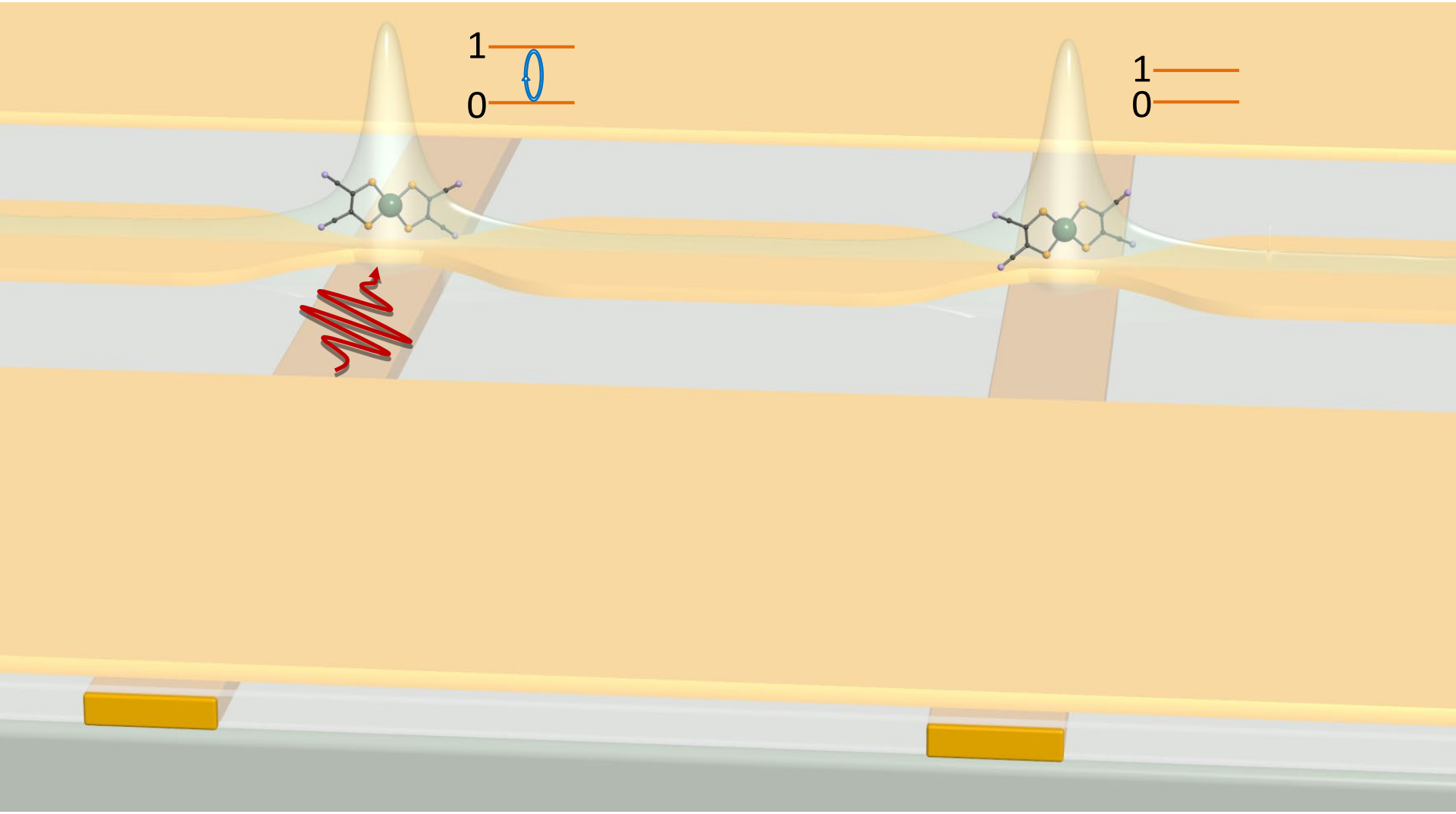
A scalable architecture for QC with molecular spins



M. Jenkins et al., Dalton Transactions **45**, 16682 (2016)

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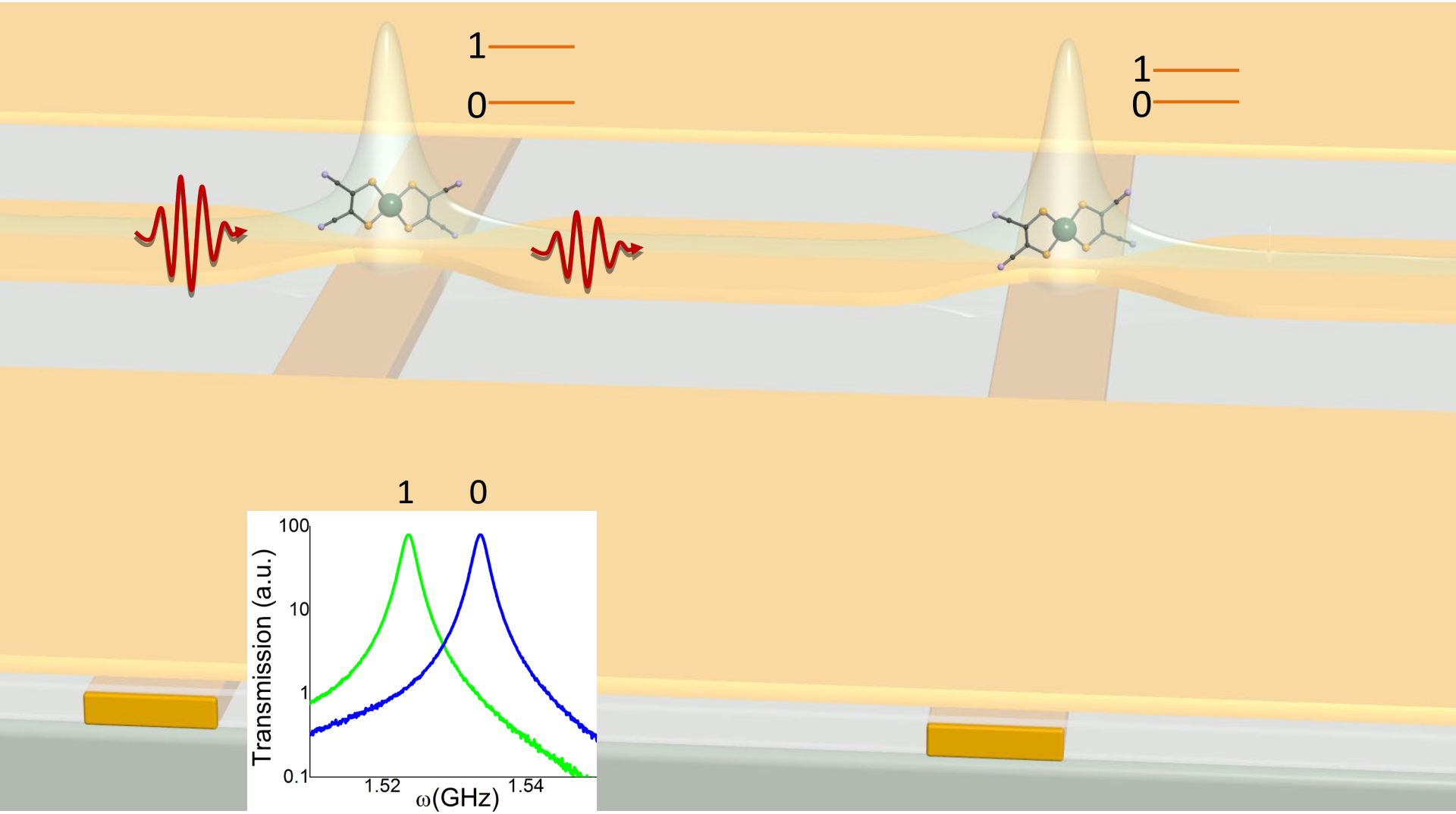

M. Jenkins et al., Dalton Transactions **45**, 16682 (2016)

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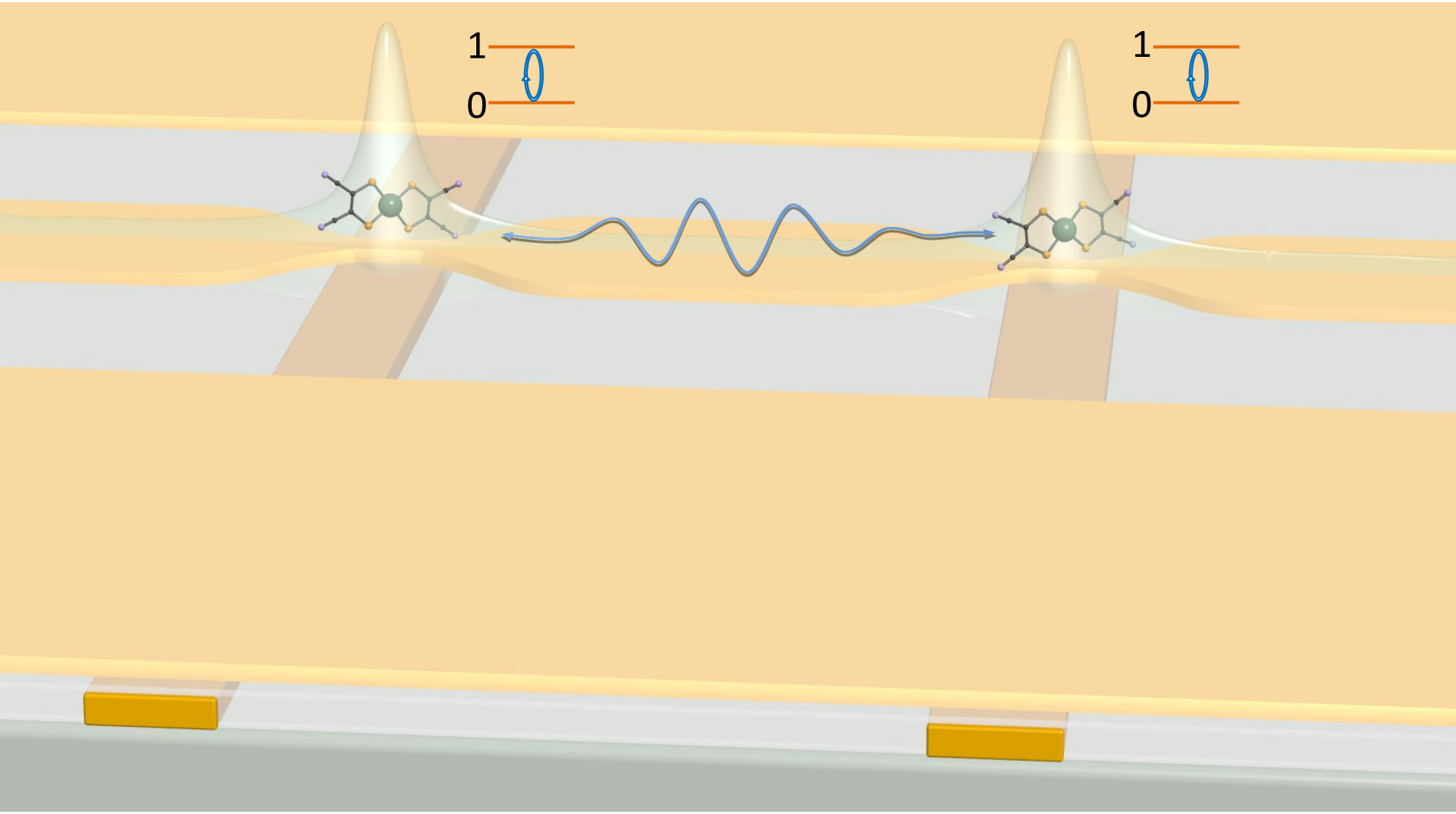

M. Jenkins et al., Dalton Transactions **45**, 16682 (2016)

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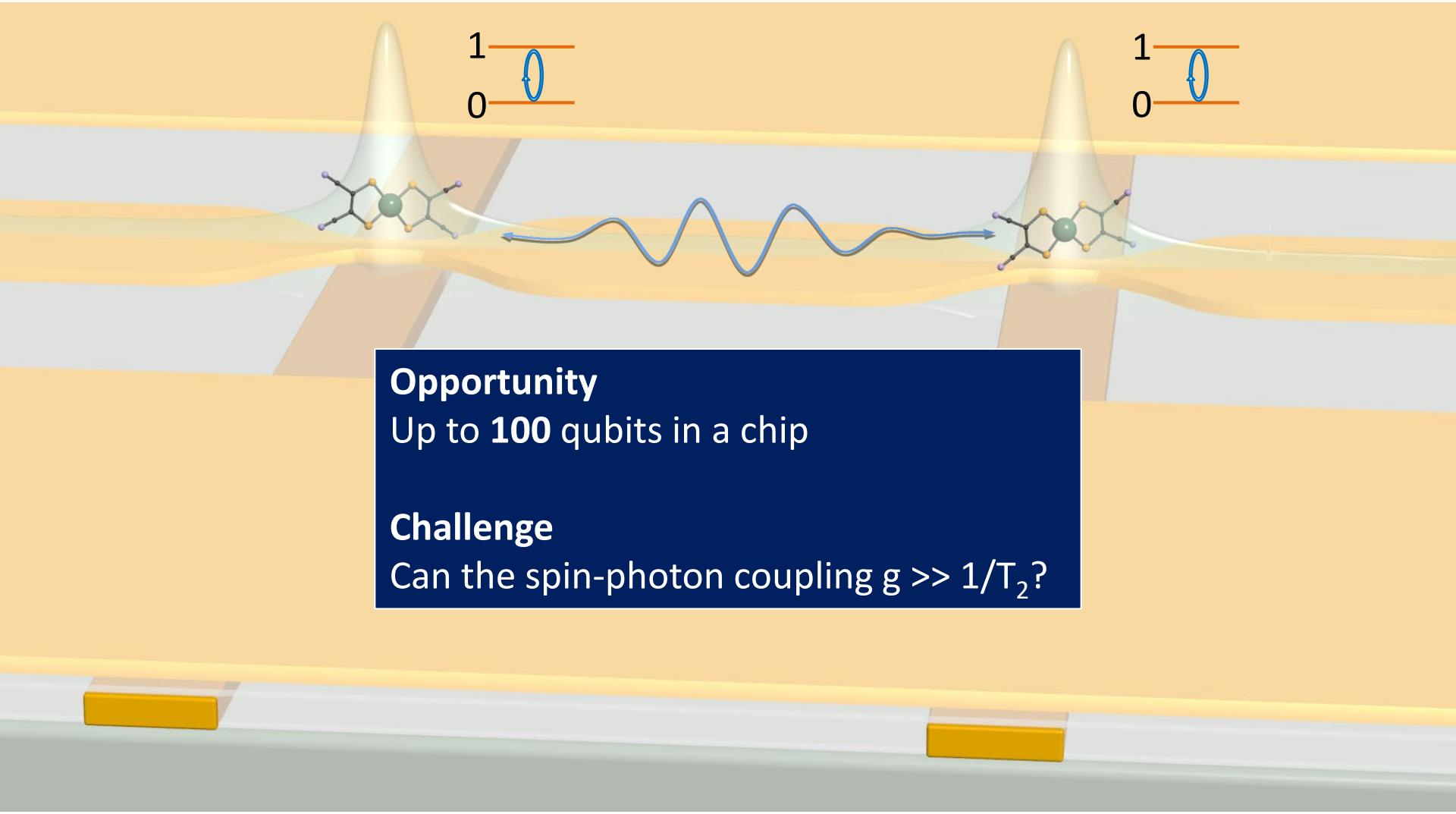
A scalable architecture for QC with molecular spins



Opportunity
Up to **100** qubits in a chip

Challenge
Can the spin-photon coupling $g >> 1/T_2$?

M. Jenkins et al., Dalton Transactions **45**, 16682 (2016)

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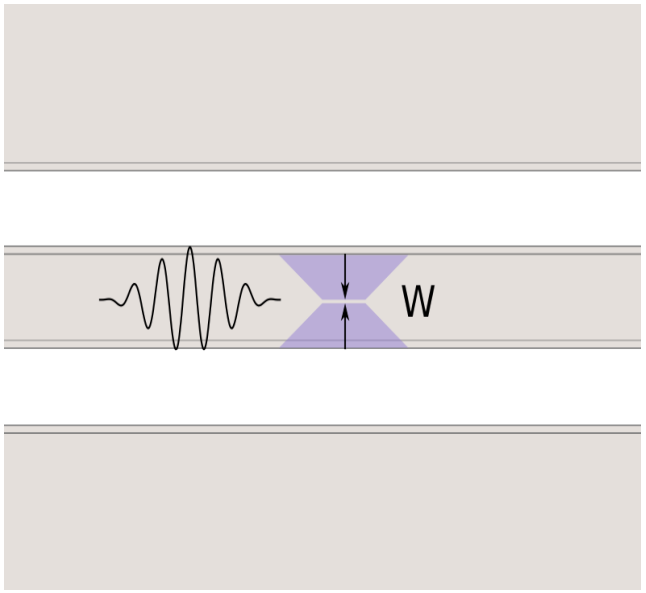
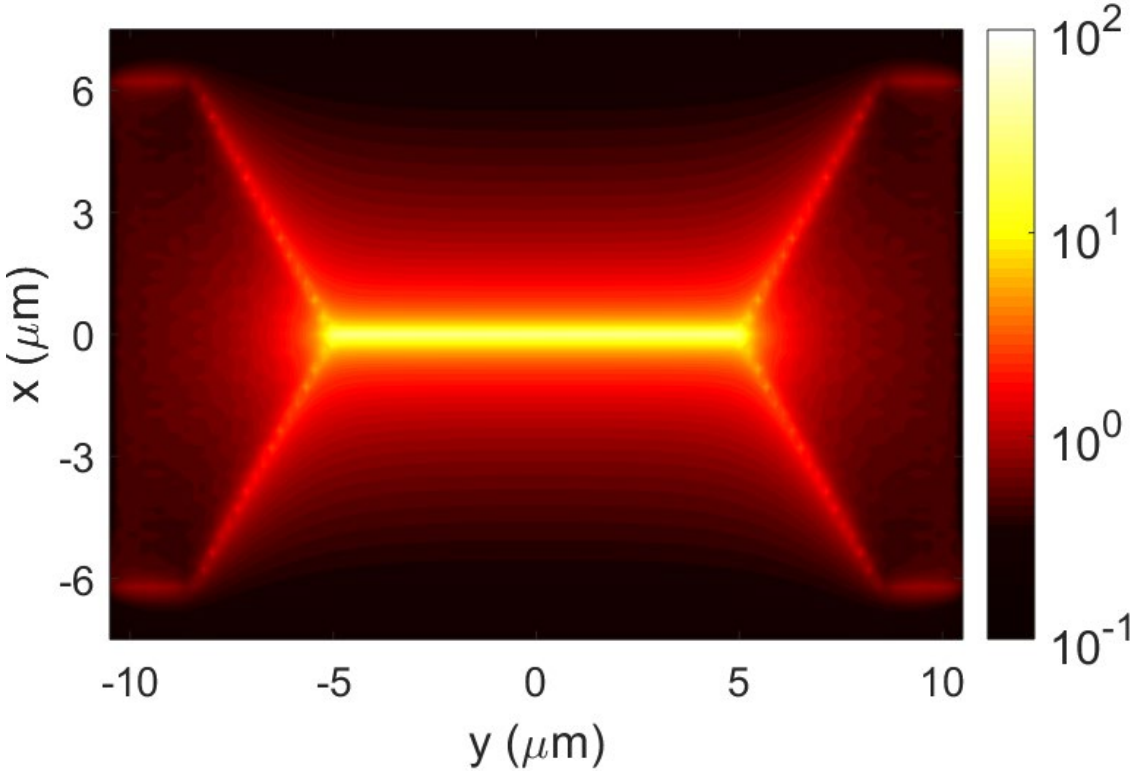
Brute-force approximation: nanoresonators

Nanoscale constriction



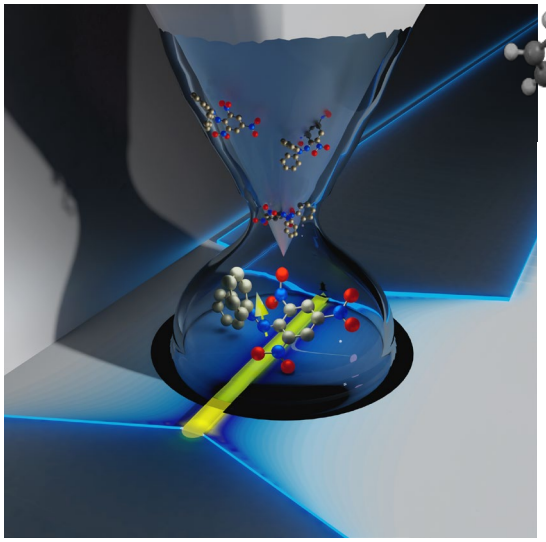
Microwave magnetic field (simulation)

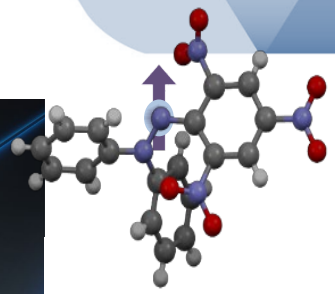


Enhances current density and microwave magnetic field

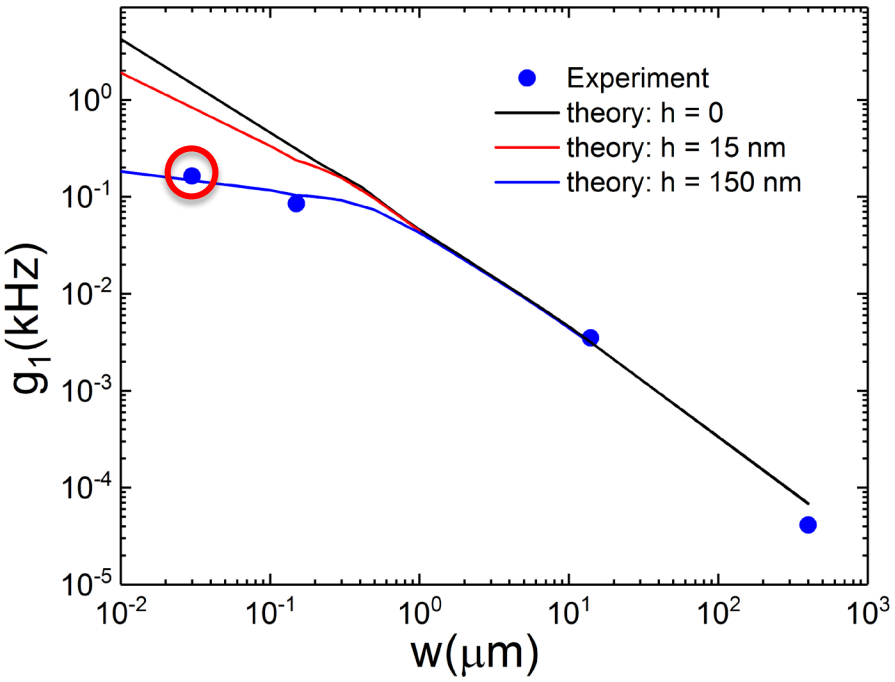
M. Jenkins et al, New J. Phys. **15**, 095007 (2013)

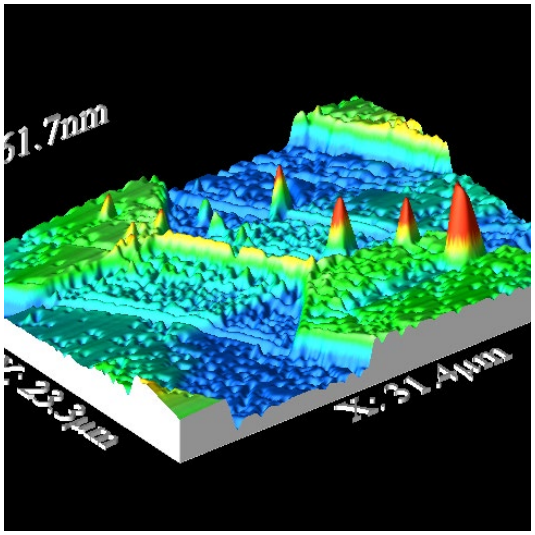
Spin ensemble integration: Dip-pen nanolithography





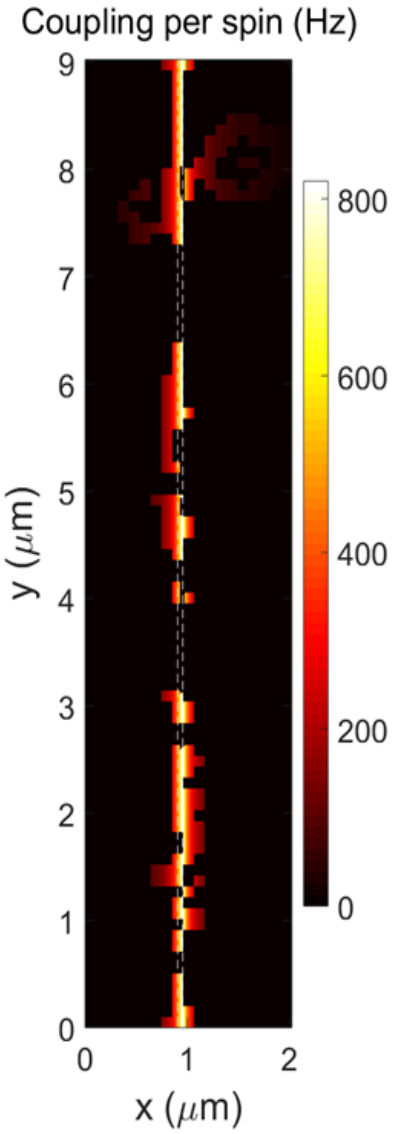
DPPH, $S = 1/2$





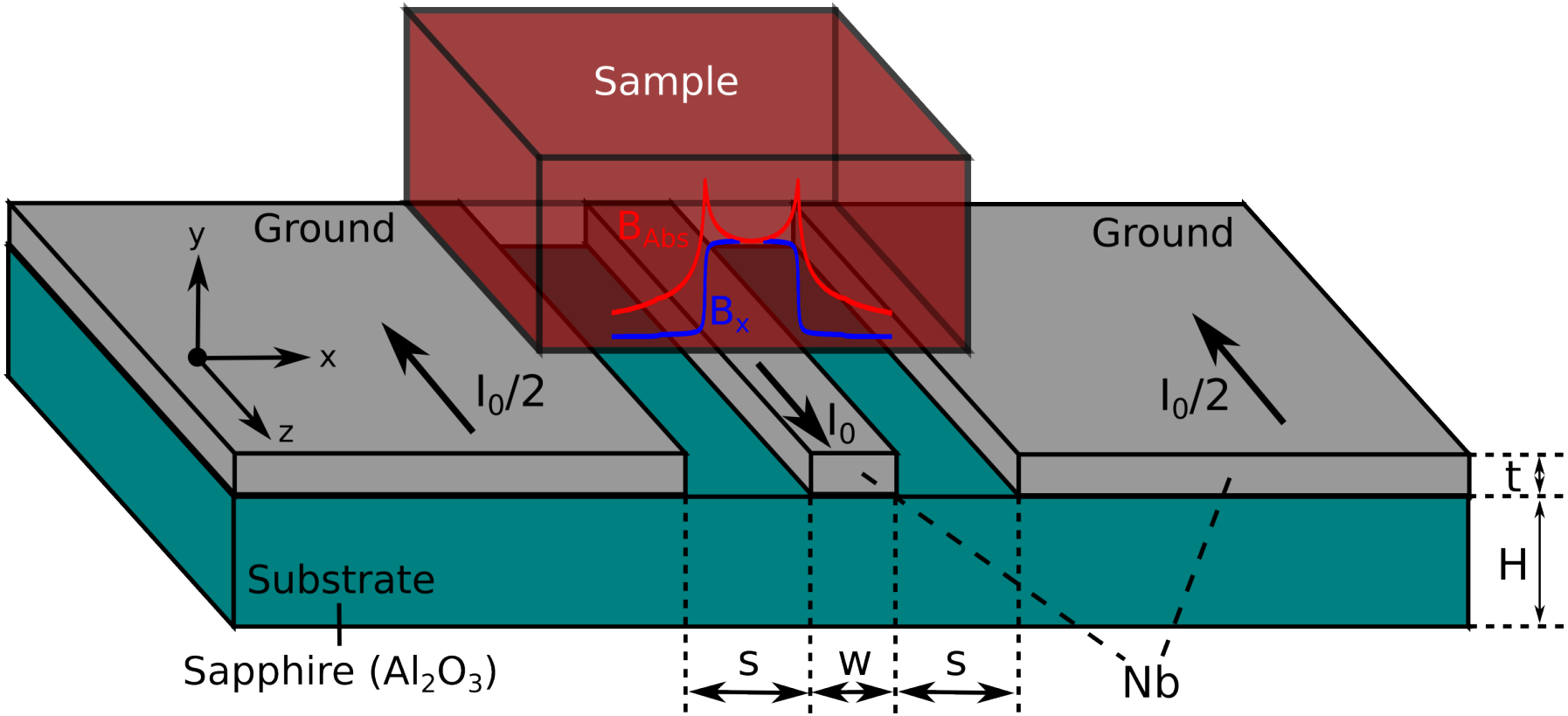
10^3 enhanced coupling near nanoconstrictions

g_1T_2 ~ 0.001-0.01



I. Gimeno *et al*, ACS Nano **14**, 8707 (2020)

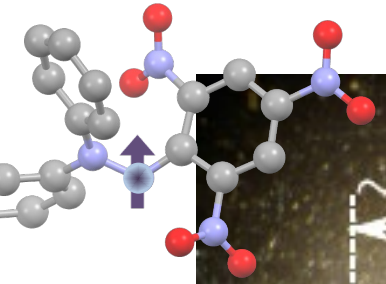
Collective coupling: ensembles



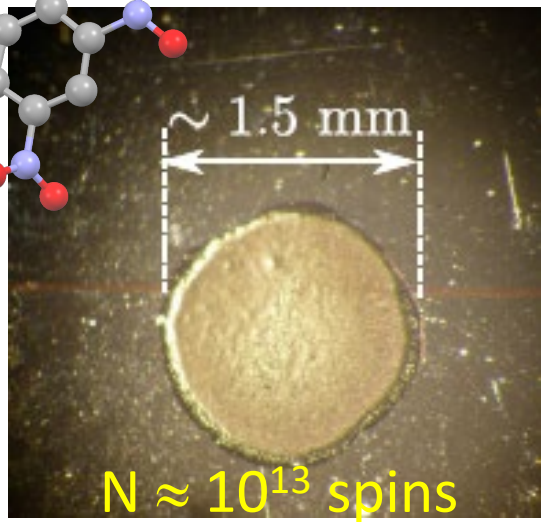
$$g_N \approx \sqrt{N}\,\frac{2g_J\mu_B J b_{rf}}{h}$$

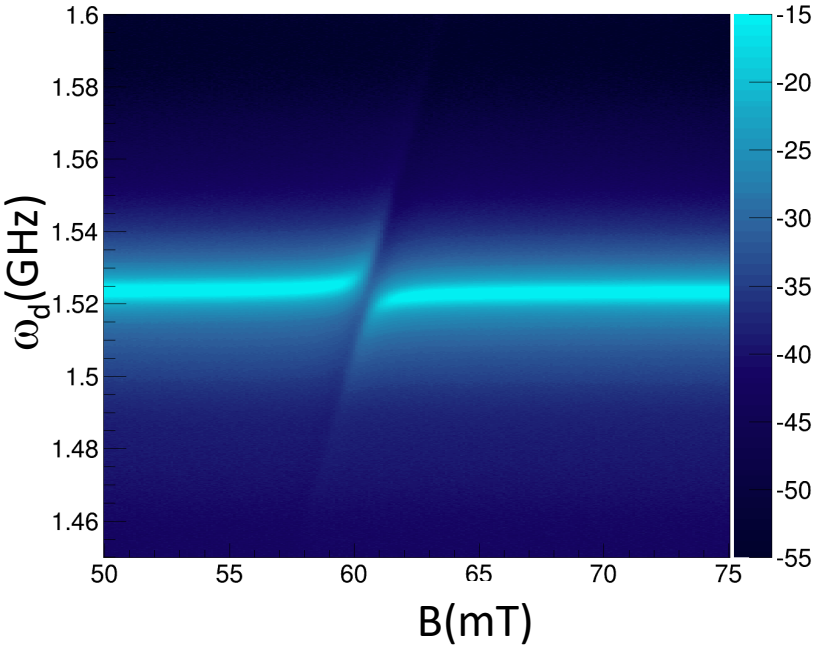
R. H. Dicke, Phys. Rev. **93**, 99 (1954)

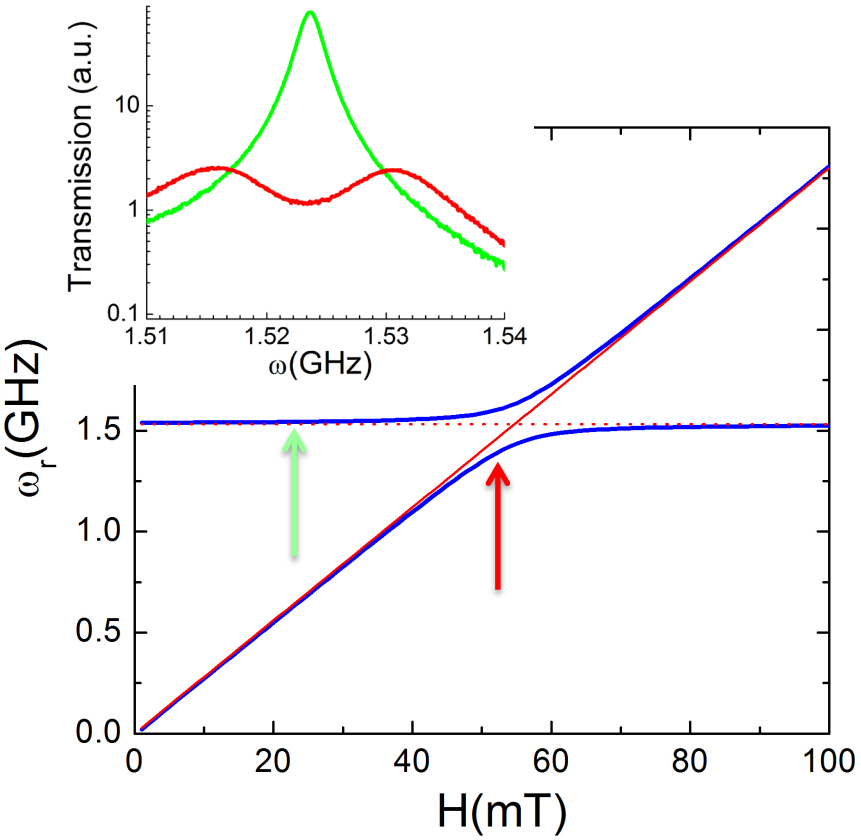
Strong collective coupling



DPPH
S=1/2
g=2



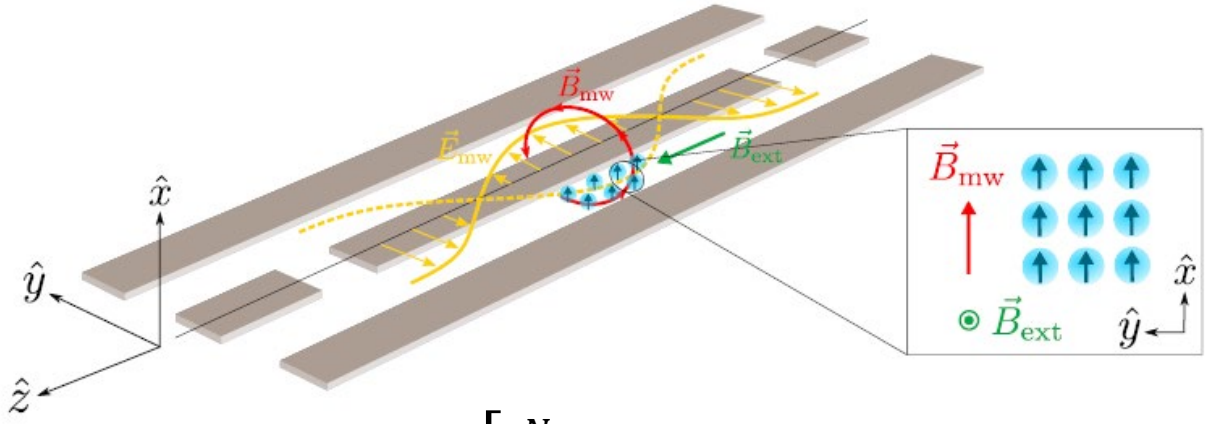




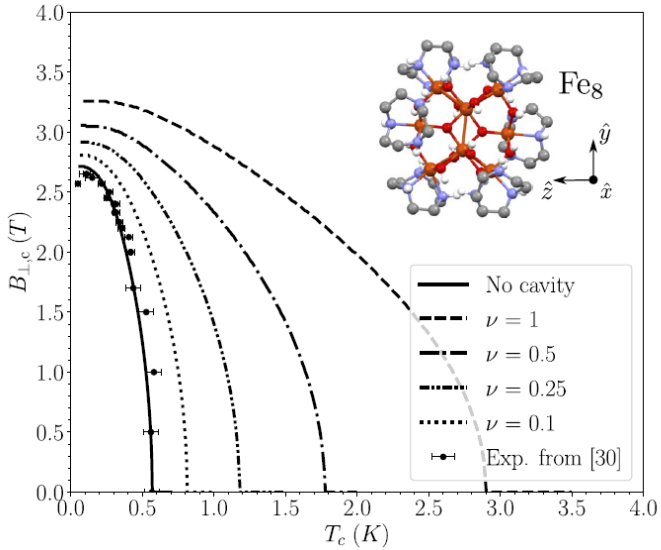
M. Mergenthaler et al, Phys. Rev. Lett. **119**, 147701 (2017).

Control of magnetism by quantum light

Coupling to cavity photons introduces effective ferromagnetic correlations



$$\mathcal{H} \xrightarrow{\text{cavity}} \mathcal{H}_{\text{eff}} = \mathcal{H} - \frac{1}{\hbar\omega_r}\left[\sum_{j=1}^{N}\frac{g\mu_B}{2}B_{mw}(\vec{r_j})\left(e^{i\theta_j}S_j^{+} + e^{-i\theta_j}S_j^{-}\right)\right]^2$$



J. Román, F. Luis and D. Zueco,
Phys Rev Lett (2021), in press

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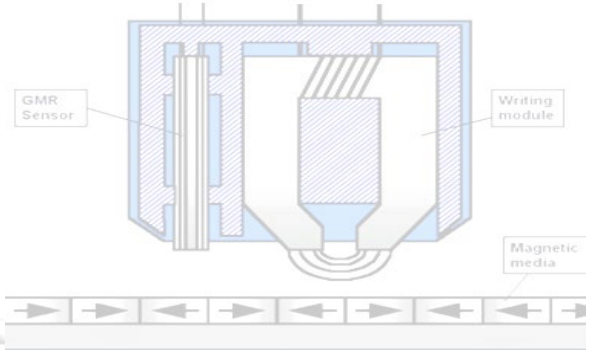
Outline

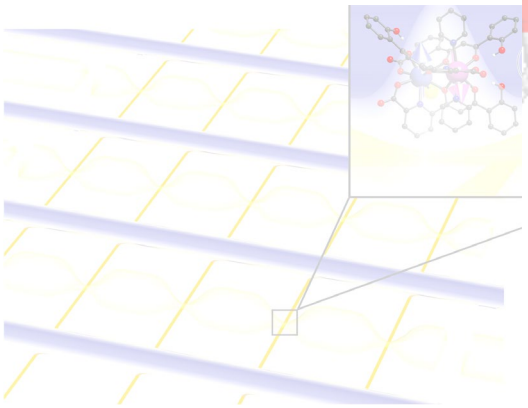
2. Magnets



3. Magnetic recording

1. Physics

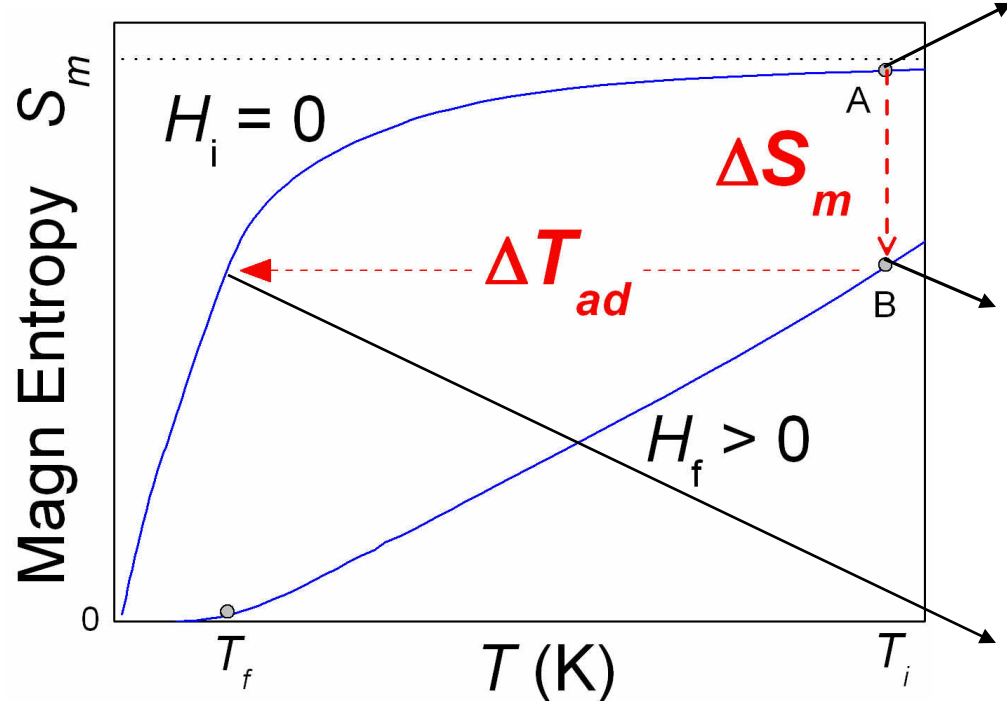
4. Quantum information



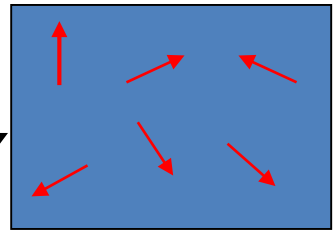
5. Magnetic coolers

Magnetocaloric effect (MCE)

W. F. Giauque and D. P. MacDougall, Phys Rev **43**, 768 (1933)



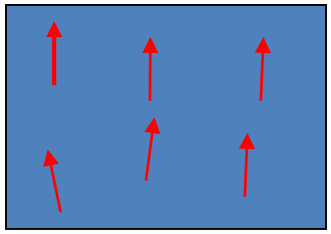
Isotropic paramagnet



$H = 0$

$S_m = R \ln(2S+1)$

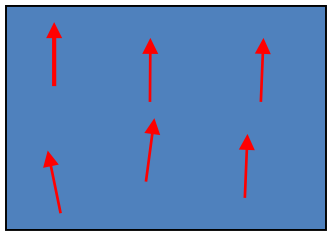
Isothermal magnetization



$H > 0$

$S_m << R \ln(2S+1)$

Adiabatic demagnetization



S_m constant

$T_f << T_i$

Magnetic coolers for very low temperatures

Gd: low anisotropy & high spin

$$\Delta S_m = R(\ln 8)$$

Large entropy content per spin: magnetic cooler

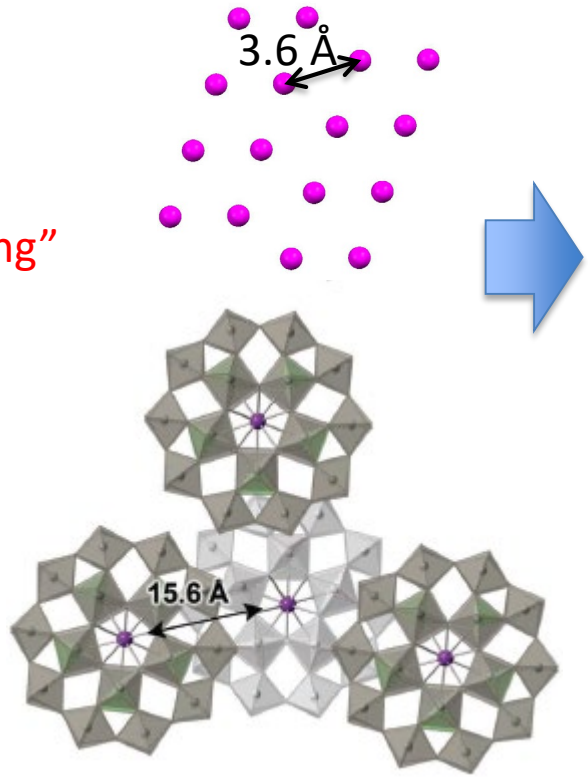
Pure Gd

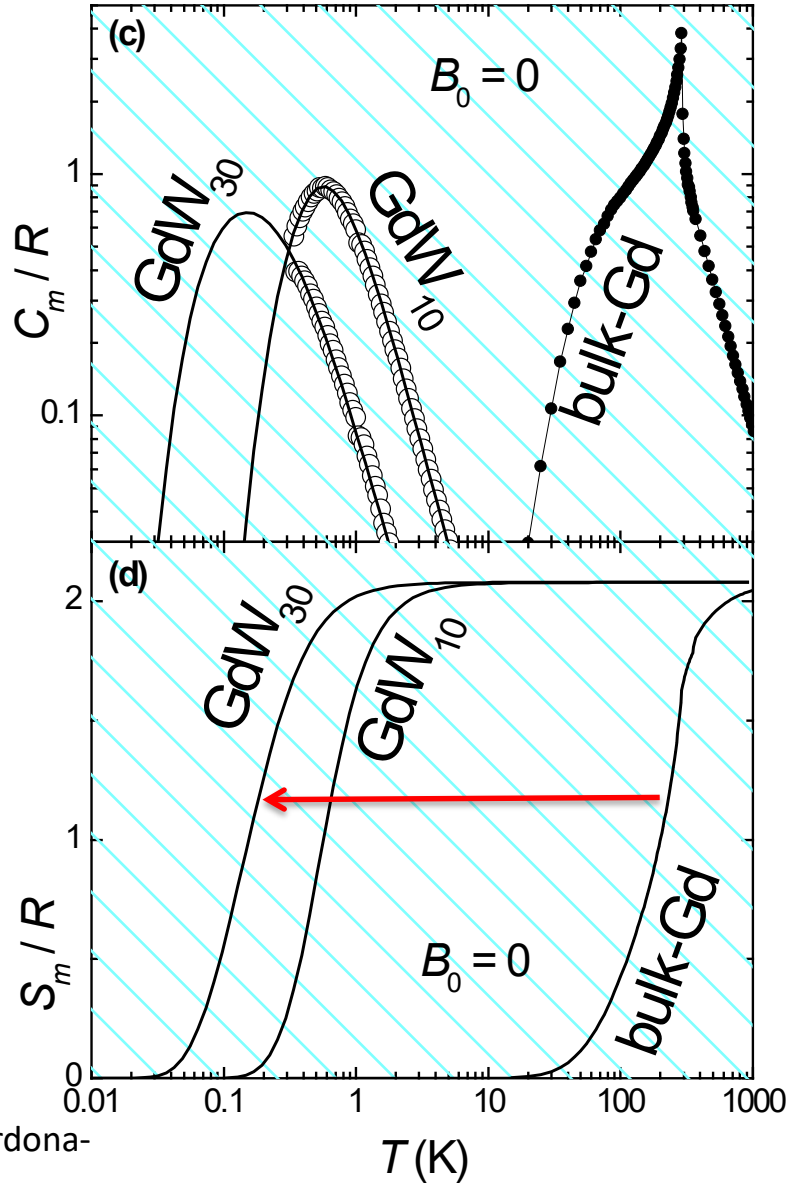
$T_c \approx 300$ K

"Fragmenting" Gadolinium

GdW_{30}

$T_c < 0.02$ K

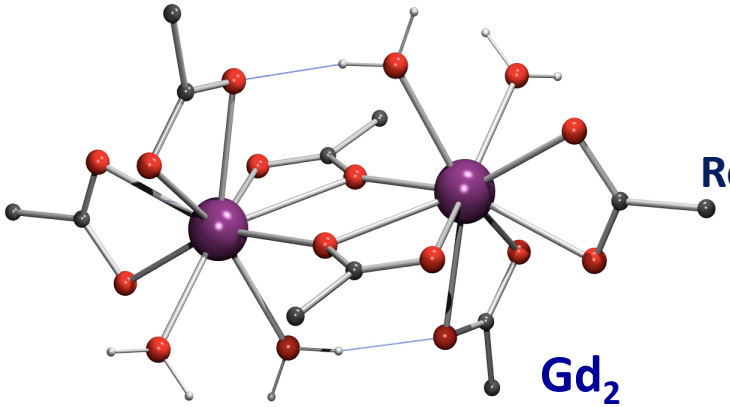




M. J. Martínez-Pérez O. Montero , M. Evangelisti , F. Luis , J. Sesé , S. Cardona-Serra , and E. Coronado, Adv. Mater. **24**, 4301 (2012)

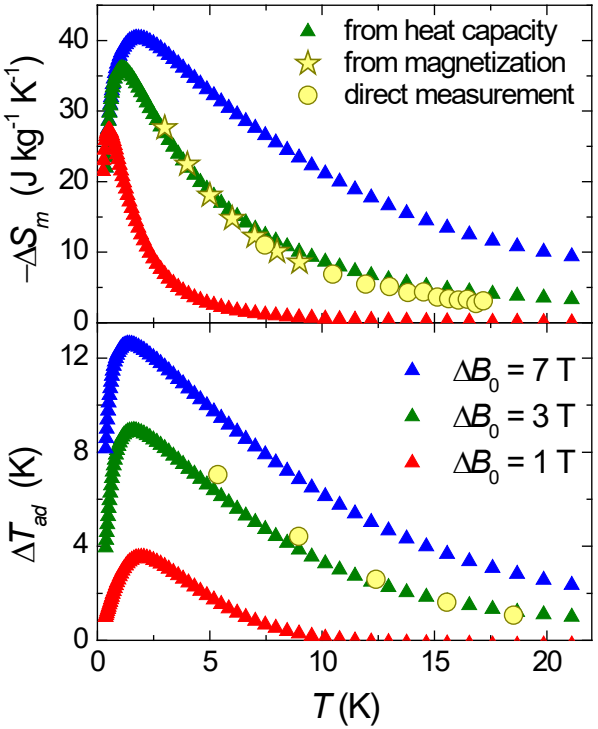
On chip refrigeration

M. Evangelisti et al., Angew. Chem. Int. Ed. **50**, 6606 (2011); G. Lorusso, M. Evangelisti *et al*, Adv. Mater. **25**, 2984 (2013); L. X. Chang et al, Chem. Commun. 49, 1055 (2013).



Gd_2

Record MCE for $T < 4.2$ K



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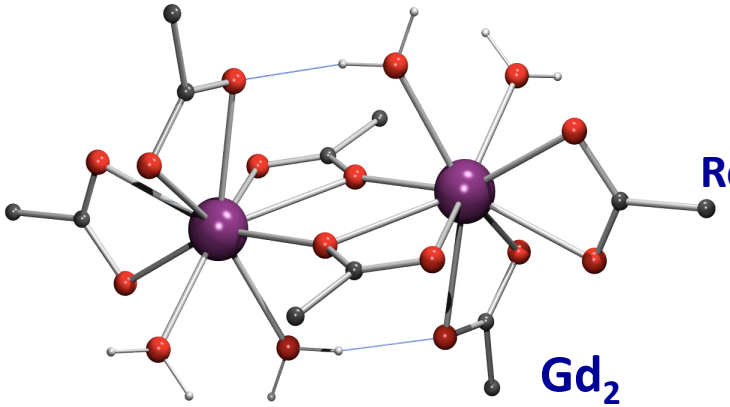

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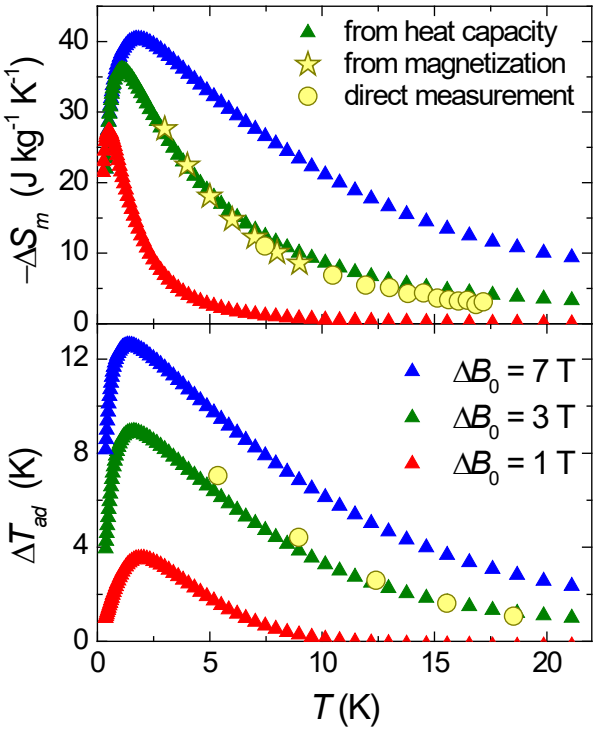
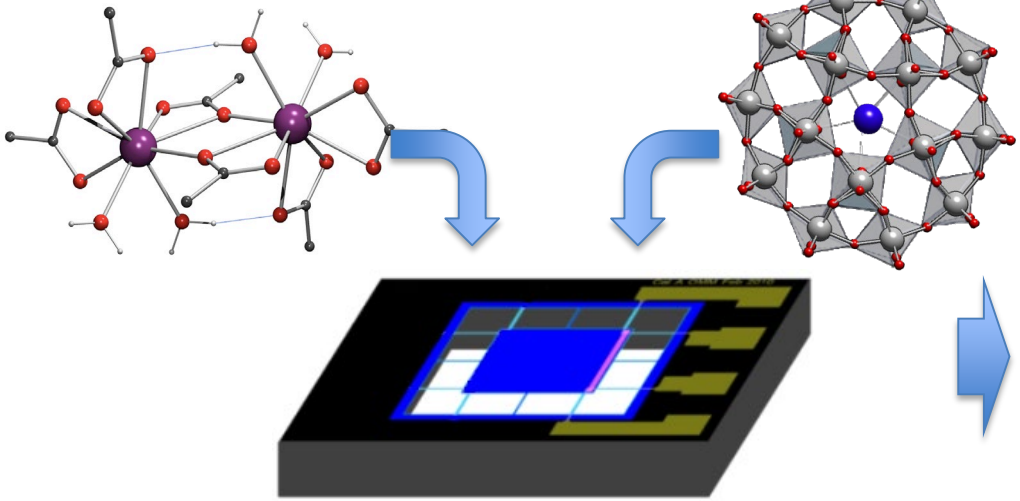
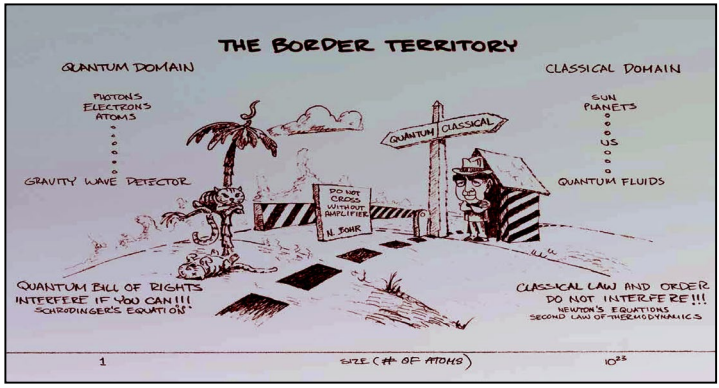


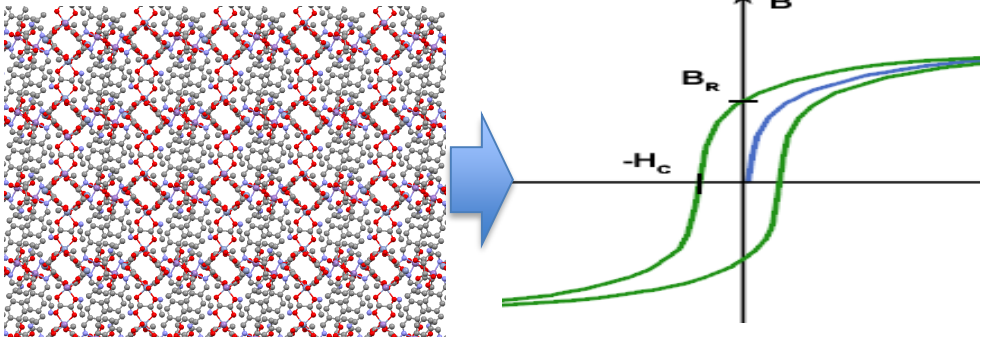


4.2 K — 500 mK — 10 mK

Conclusions

Quantum physics at the mesoscopic scale
⇒ quantum relaxation, multipartite entanglement , non-equilibrium QPTs, control of magnetic order by q-light, …





Lanthanide-free permanent magnets ⇒ enhance T_c

Magnetic recording: High potential of miniaturization ⇒ enhance T_b

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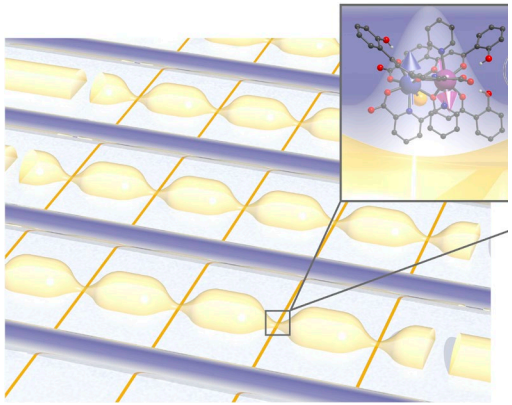
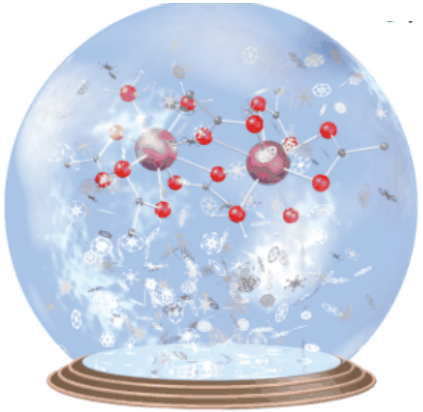
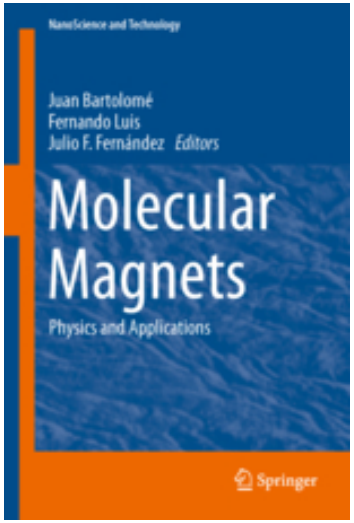
Conclusions



Solid candidates to realice basic units of a quantum computer + potential for scaling up ⇒ proof-of-concept implementations



Best coolers in the liquid Helium region and below ⇒ devices



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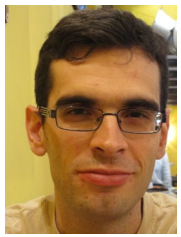
Quantum Materials and Devices


Mark Jenkins


Ignacio Gimeno


Marcos Rubín


David Zueco


Olivier Roubeau


Pablo Alonso


María José Martínez


Javier Sesé


Pablo Hermosilla


Ana Isabel Lostao

IFF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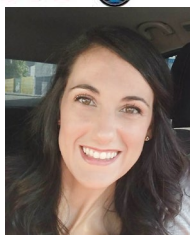
Enrique Burzurí

CENTRO DE ASTROBIOLOGÍA
ASOCIADO AL NASA ASTROBIOLOGY INSTITUTE
CSIC INTA

Marina Calero


Alicia Gómez

TU WIEN

Andreas Angerer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Yan Duan


Eugenio Coronado

GOBIERNO DE ESPAÑA MINISTERIO DE CIENCIA E INNOVACIÓN

QUANTERA

FATMOLS

European Commission

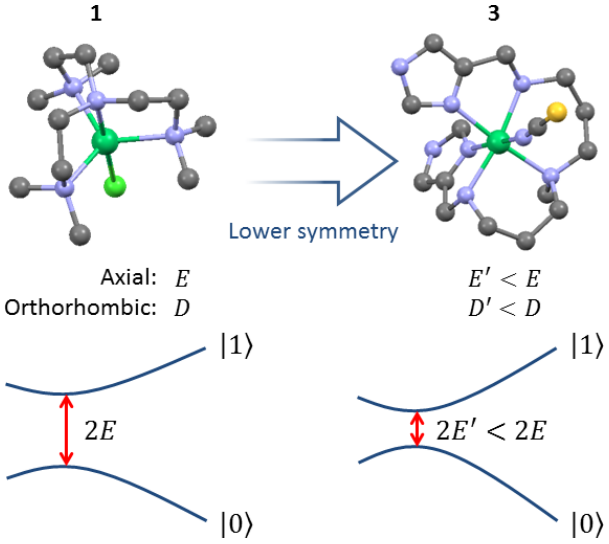
This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 862893 (FATMOLs).

Tuning the gap: proper coordination

[Ni(Me$_6$tren)Cl](ClO$_4$)

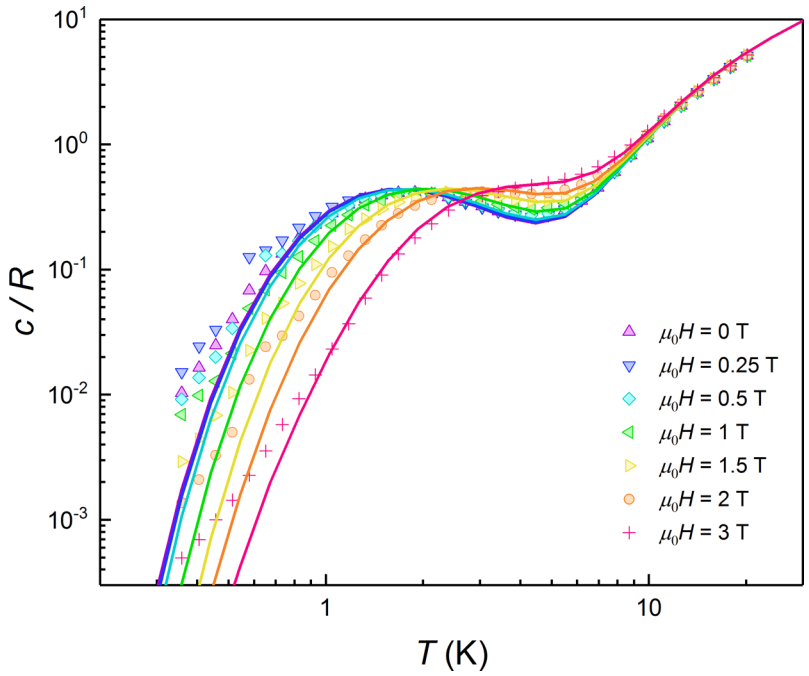
Ni(II) $S = 1$

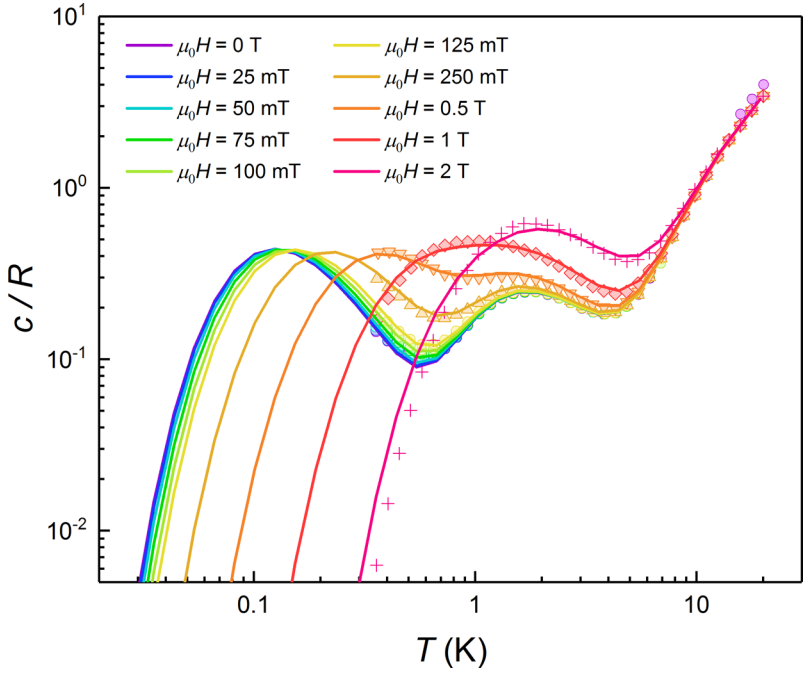
$$\mathrm{H} = BO_2^0 + B_2^2O_2^2$$



[Ni-Imdipa]-(NCS)

M. Rubín et al, Chem. Sci. **12**, 5123 (2021)





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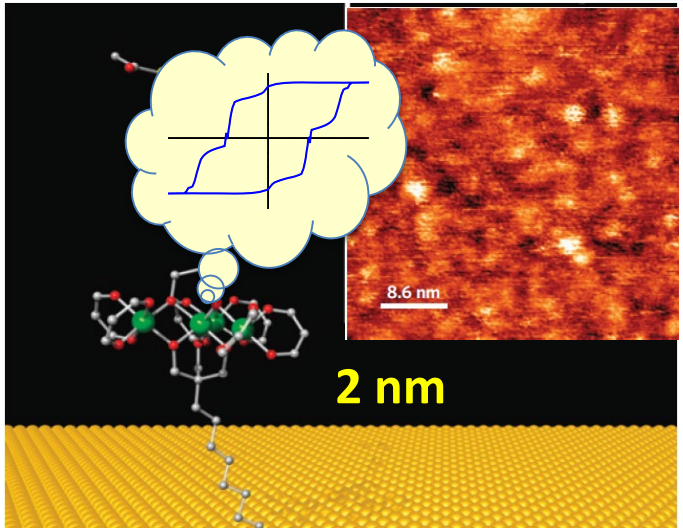
From the bulk to single molecules



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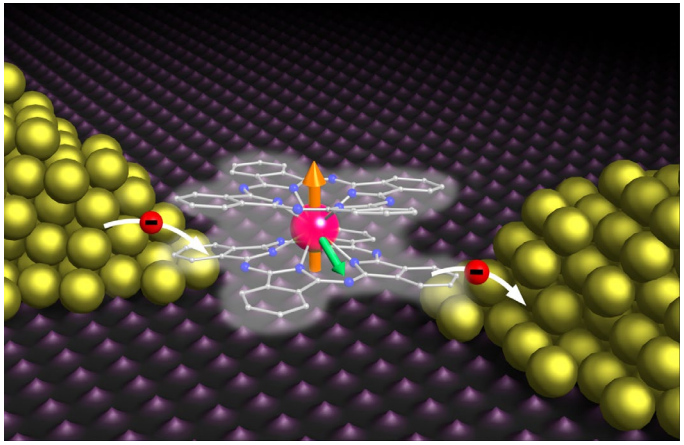

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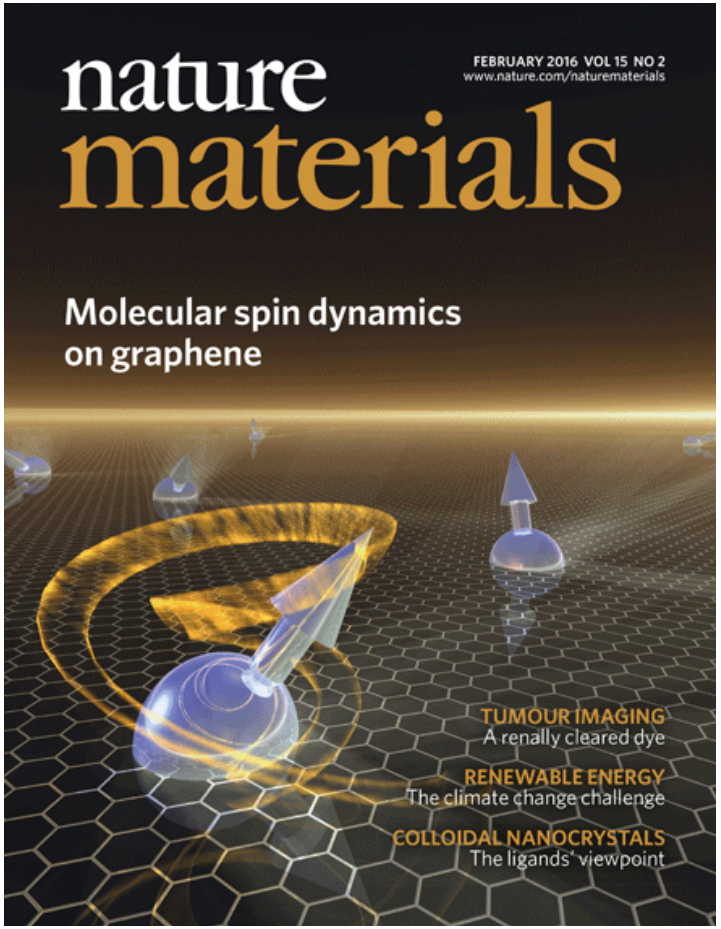

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