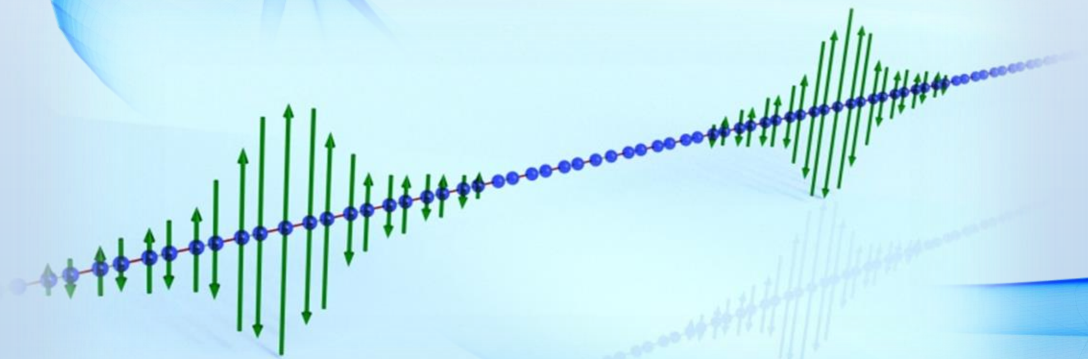
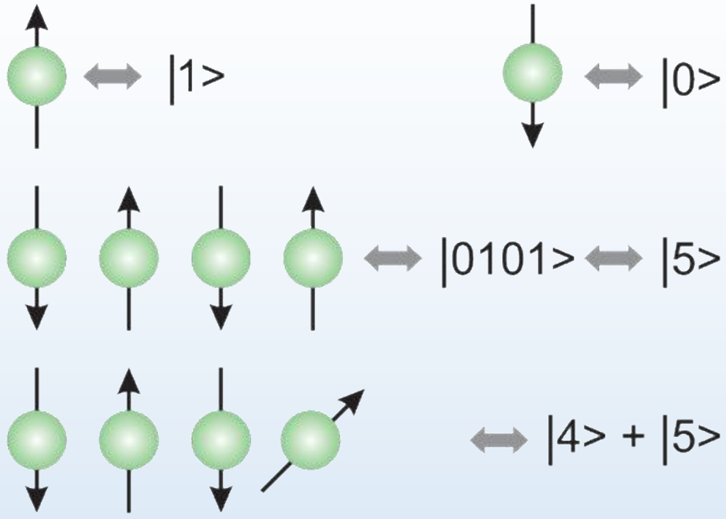
Defects in spin chains:

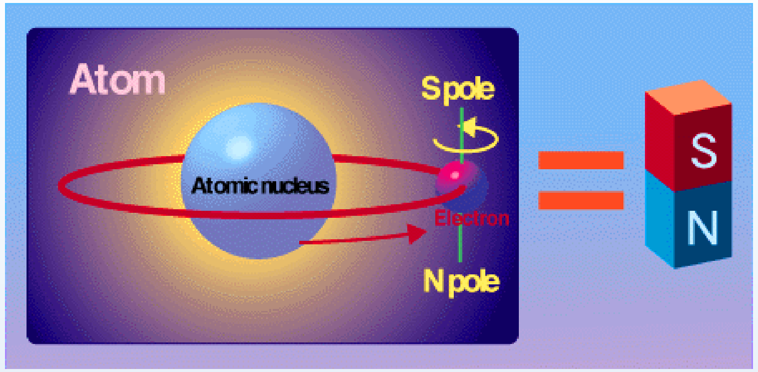
a virtual molecular magnet with quantum coherence properties.

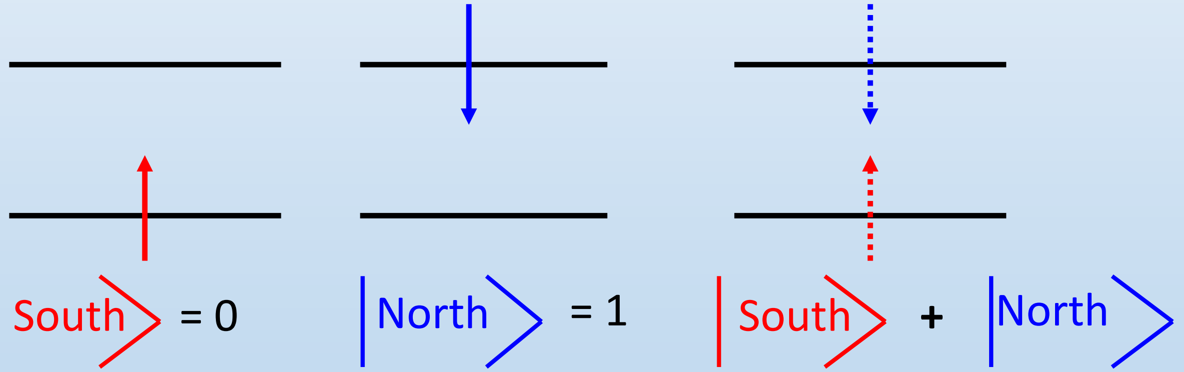
Sylvain Bertaina



Quantum Information – Quantum coherence

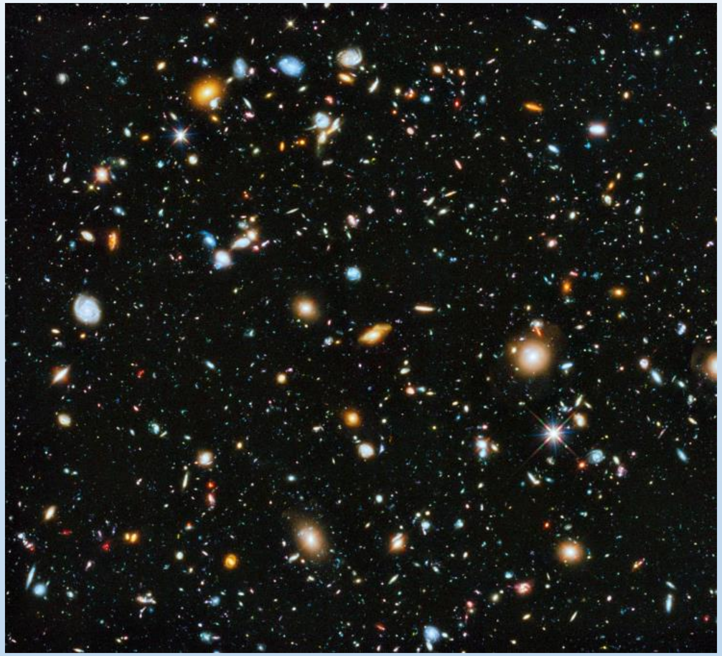






Quantum memory

2^{300} = 1 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 000 = 10^{90}

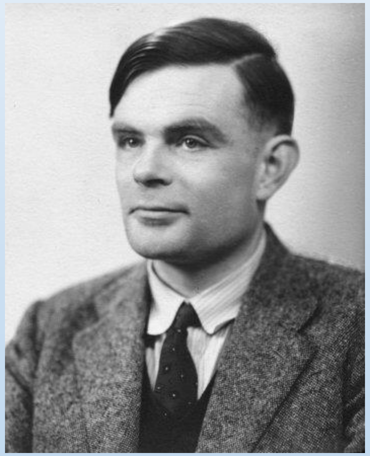


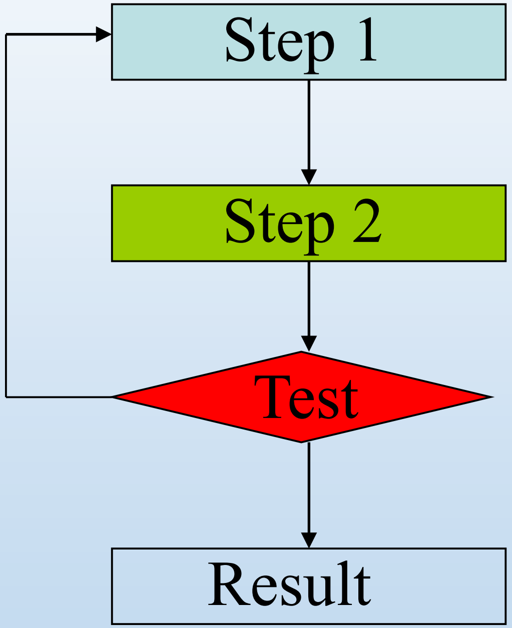
Classical Algorithm

Euclide ~ 300 BC : GCD calculation

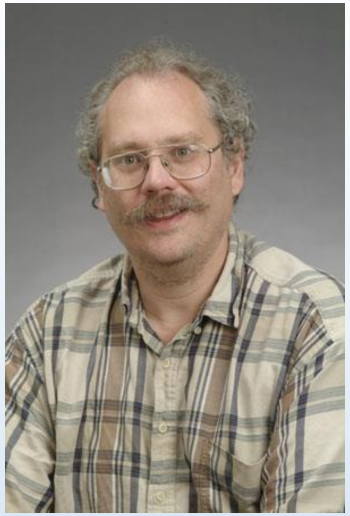
Turing 1936 :

1° computer



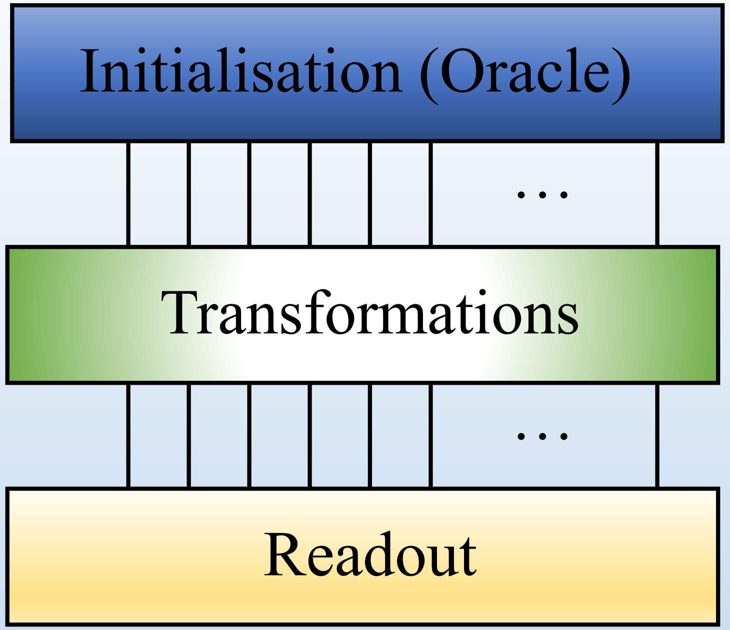


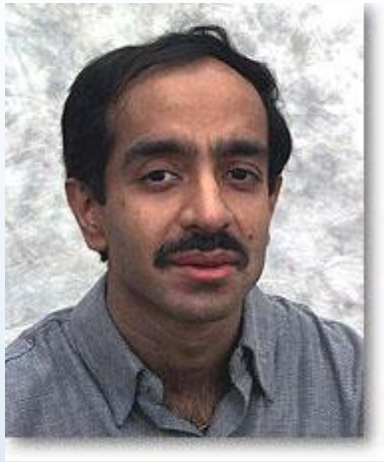
Quantum algorithm



Shor 1998

Factorisation

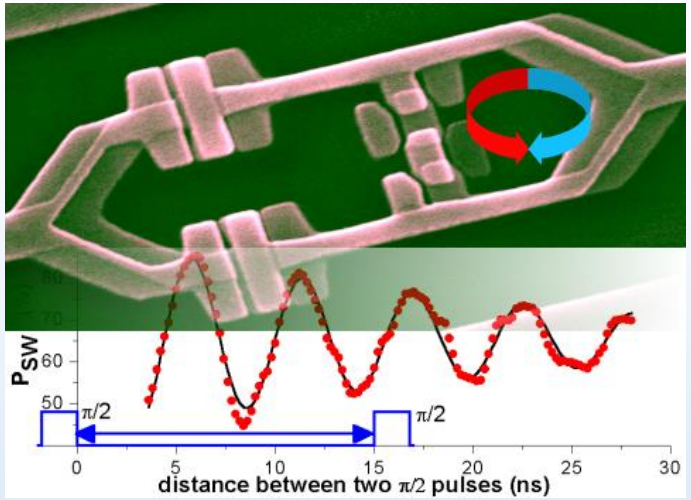




Grover 1996

Search in database

Examples of qubits

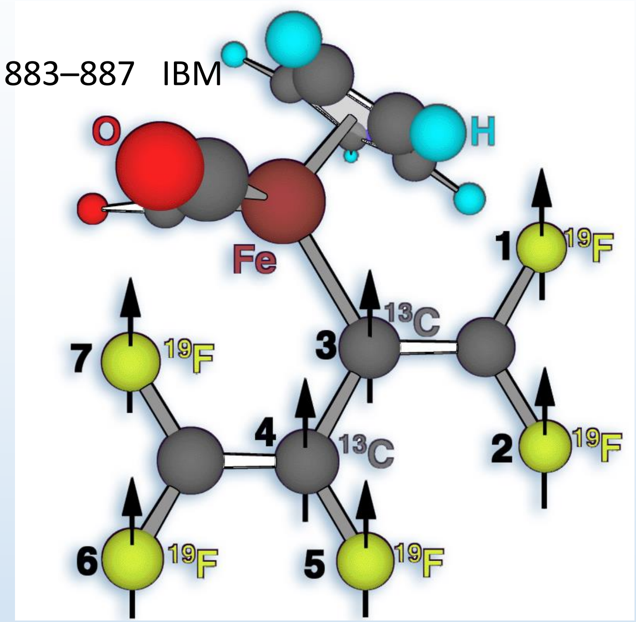


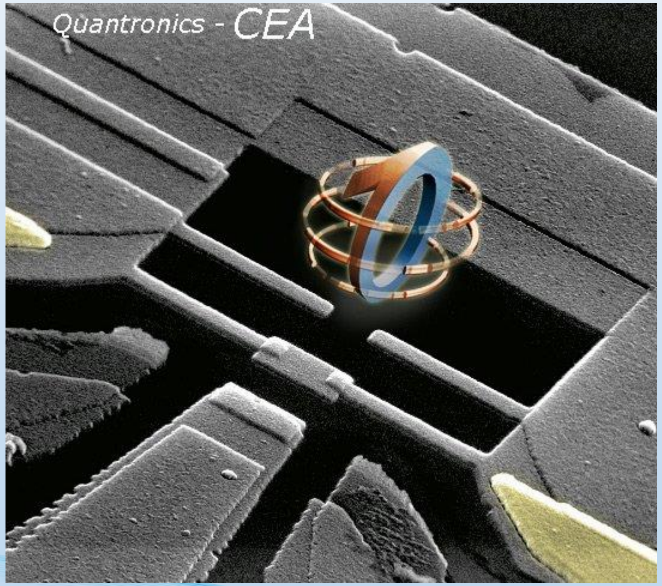
Superconducting flux qubit

Chiorescu *et al.* Science **299**, 1869 (2003)

Nuclear Spins

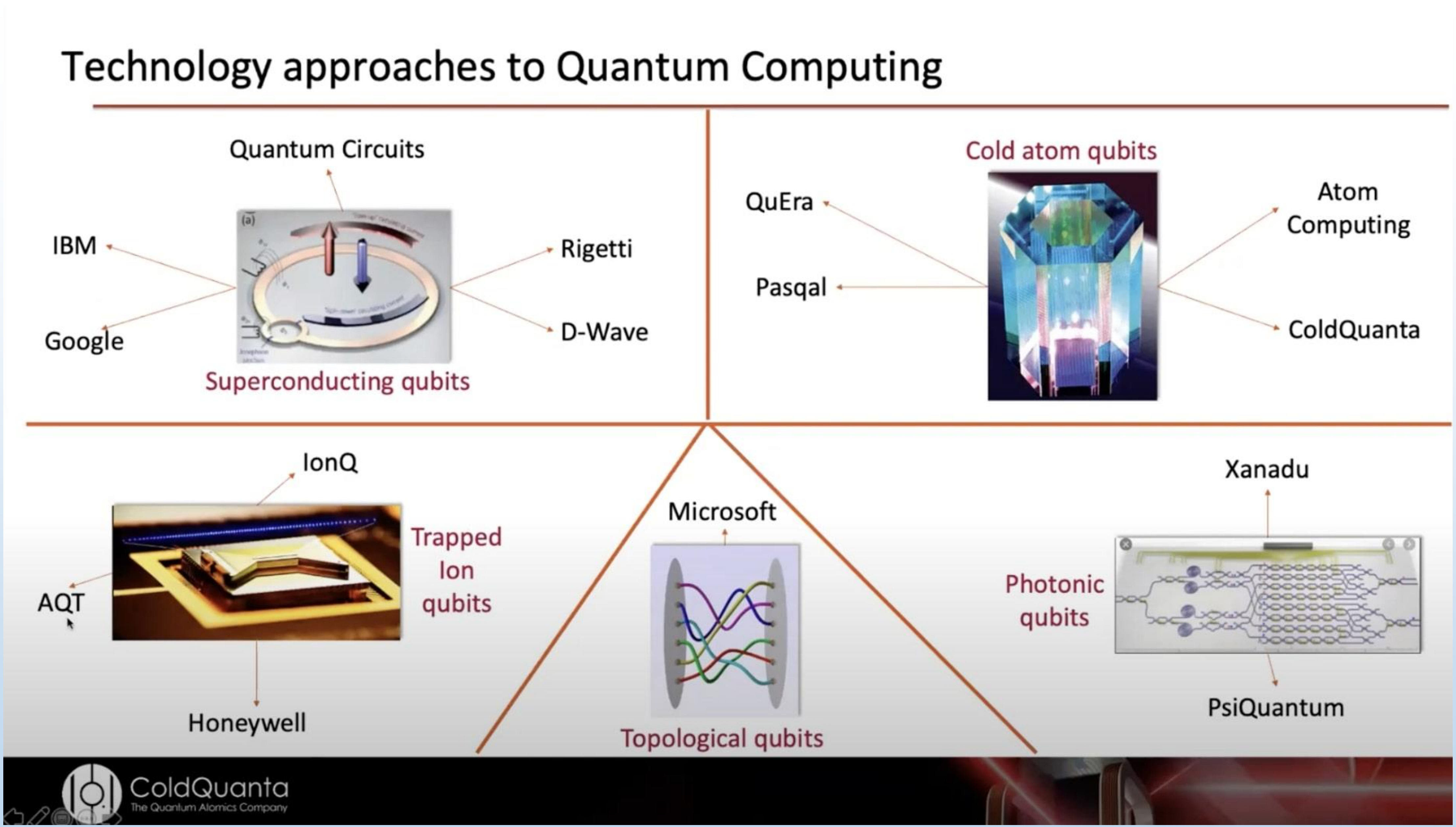
Lieven *et al. Nature* **414,** 883–887 IBM





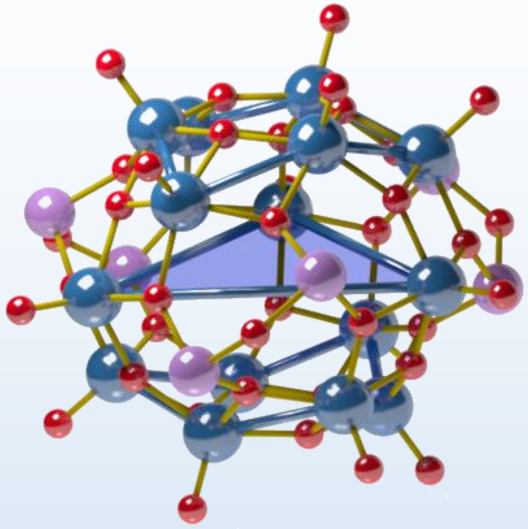
Quantronium

Bouchiat, CEA Patent



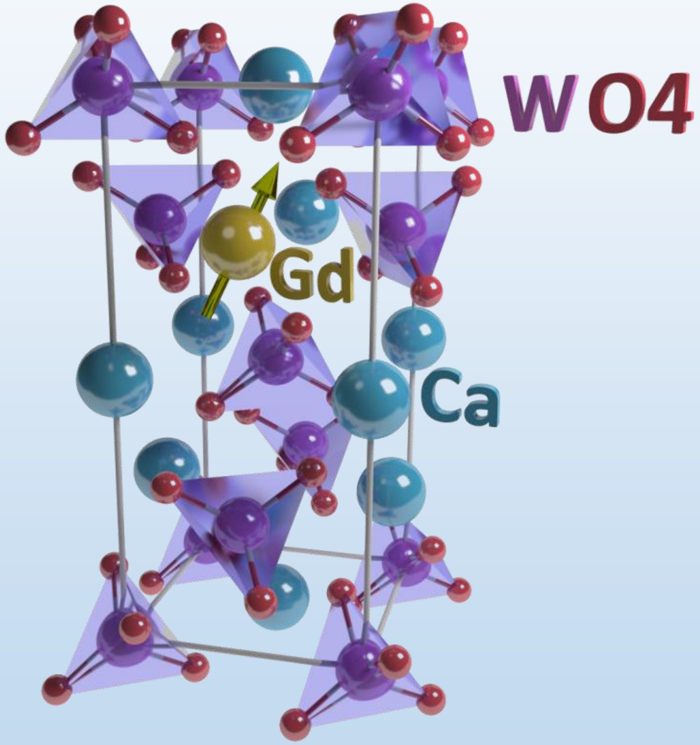
Technology approaches to Quantum Computing
Quantum Circuits
IBM
Google
Rigetti
D-Wave
Superconducting qubits
Cold atom qubits
QuEra
Pasqal
Atom Computing
ColdQuanta
IonQ
AQT
Trapped Ion qubits
Honeywell
Microsoft
Topological qubits
Xanadu
Photonic qubits
PsiQuantum
ColdQuanta
The Quantum Atomics Company

Non conventional electron spin qubits



Molecular Magnets : V15

Bertaina *et al.* Nature **453**-203(2008),
PRL **109** 050401 (2012)



Qubits multiphotons

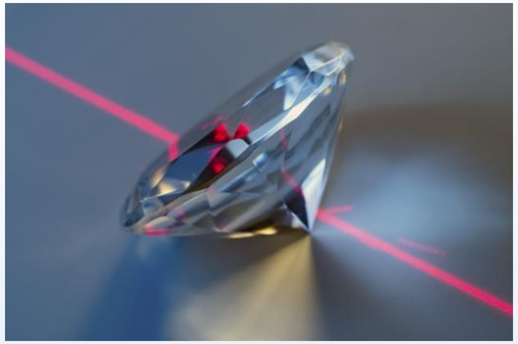
Bertaina *et al.* PRL **102** 050501(2009)
PRB **84** 114433 (2011), PRB **92** 024498 (2015)

Spin Orbit Qubit of Rare Earth

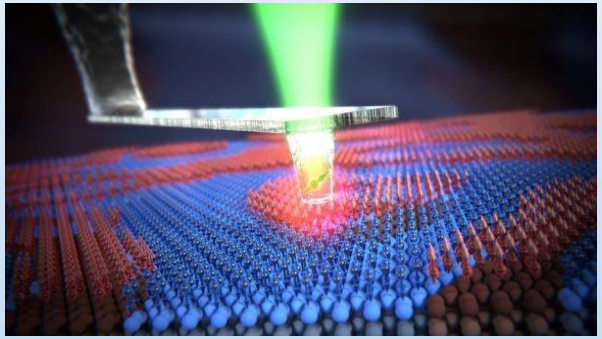
Bertaina *et al.* Nature Nano **2** 39 (2007), PRL **103** 226402 (2009), LeDantec *et al* Sci Adv (2022)

Examples of electron spin qubits – State of the art

NV center in diamond

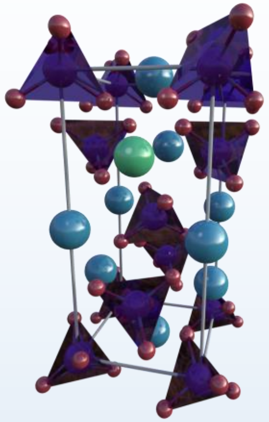


Quantum sensing

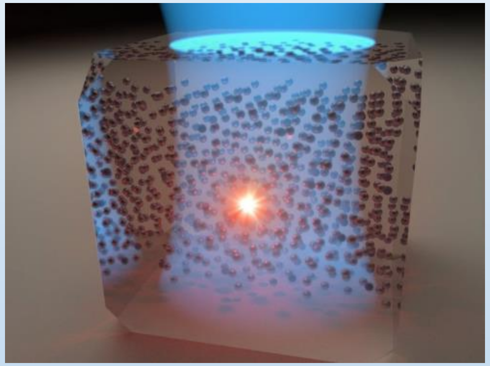


ASTERIQS

Rare earth ions

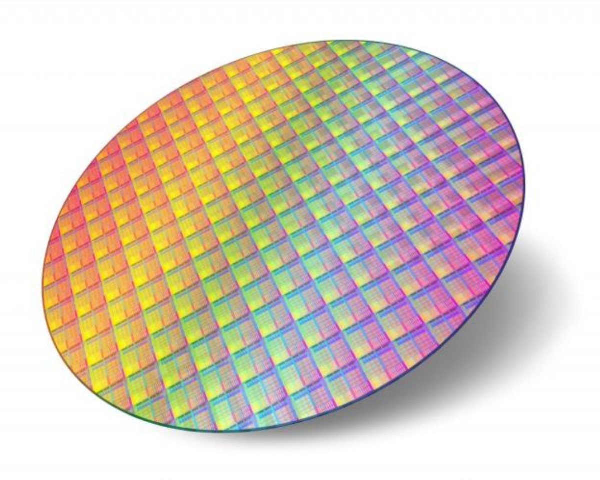


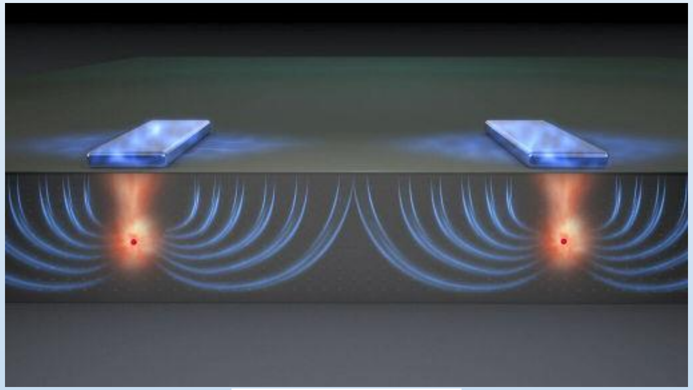
Quantum memory



SQUARE
Scalable Rare Earth Ion QUAntum Computing Nodes

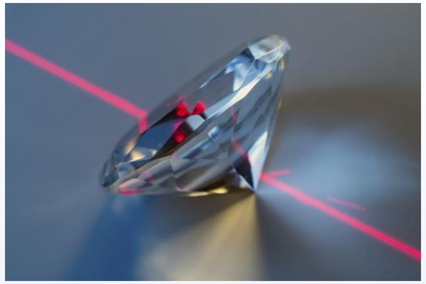
Silicon

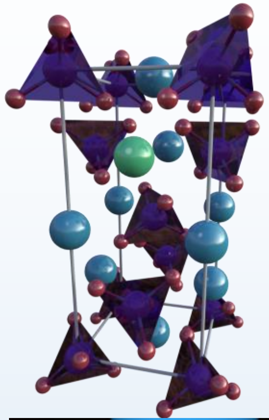


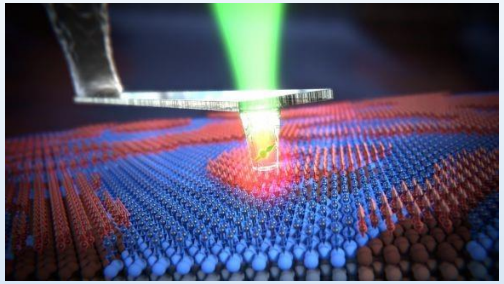


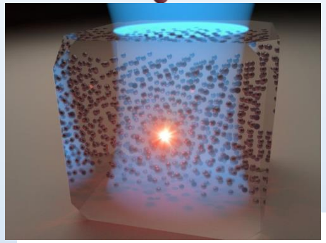
MOSQuito
MOS-based Quantum Information Technology

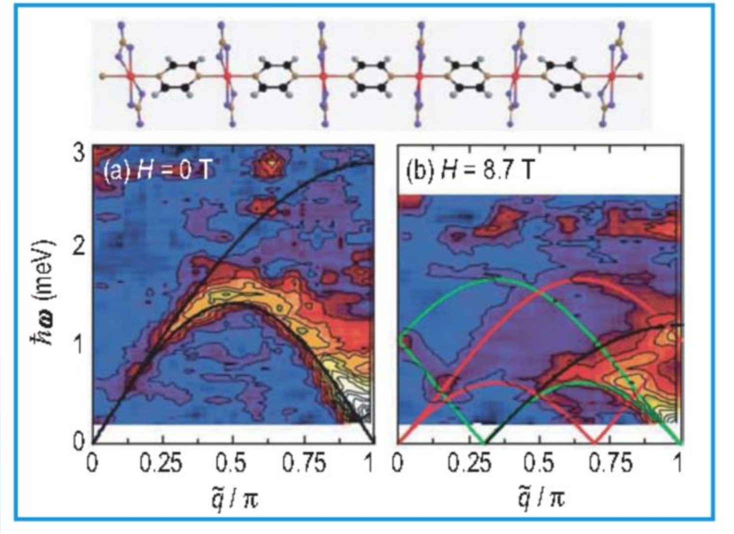
Motivations











Interaction

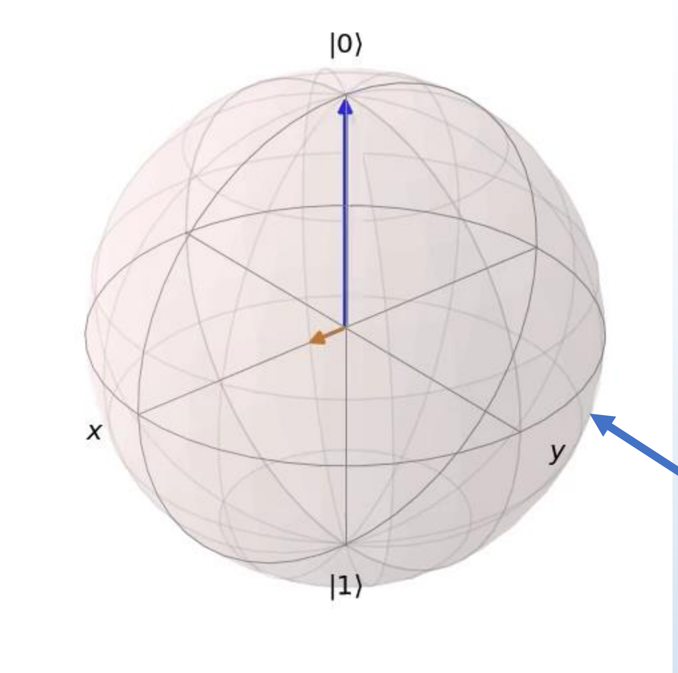
→

Electron spin Qubit : Diluted ions

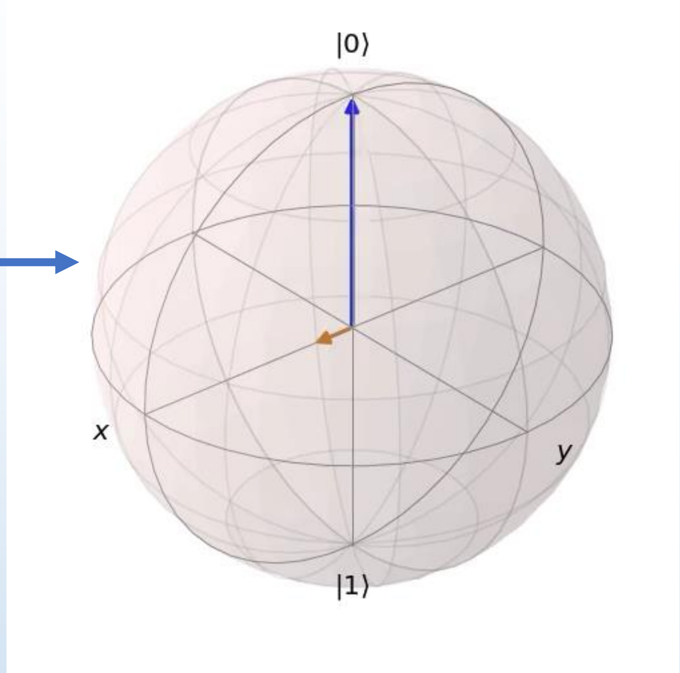
Strongly correlated magnets : Many body physics

Electron Spin Resonance

$$\frac{d\vec{S}}{dt} = \vec{S} \times (\vec{H} + \vec{h}\cos(2\pi f t)) - \Gamma\vec{S}$$



CW ESR :
Frequency fixed
Magnetic field swept



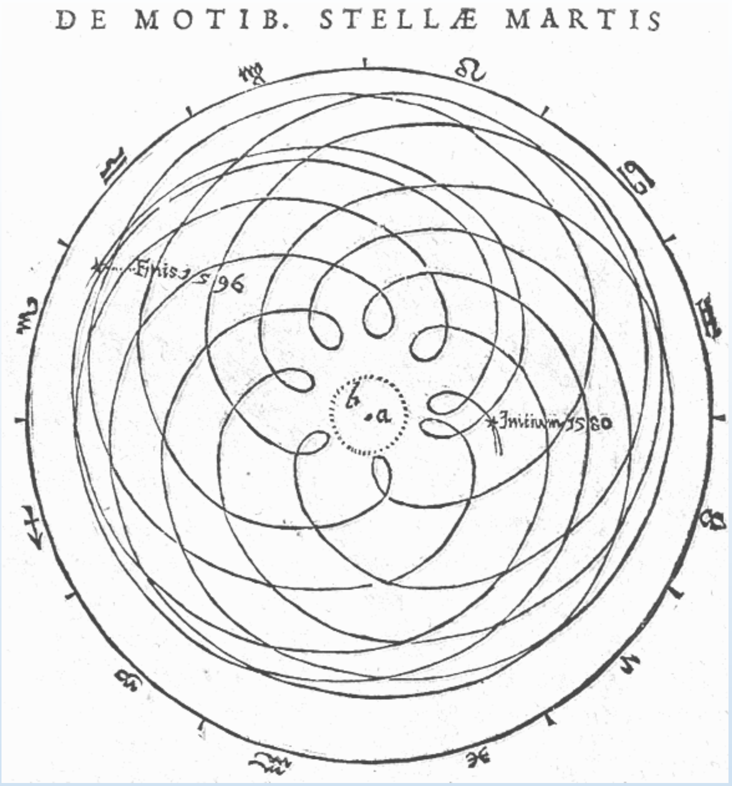
Pulsed ESR:
Frequency fixed
Magnetic field fixed
Recorded in time

Relaxation time << experiment time:
Transient regime or coherent

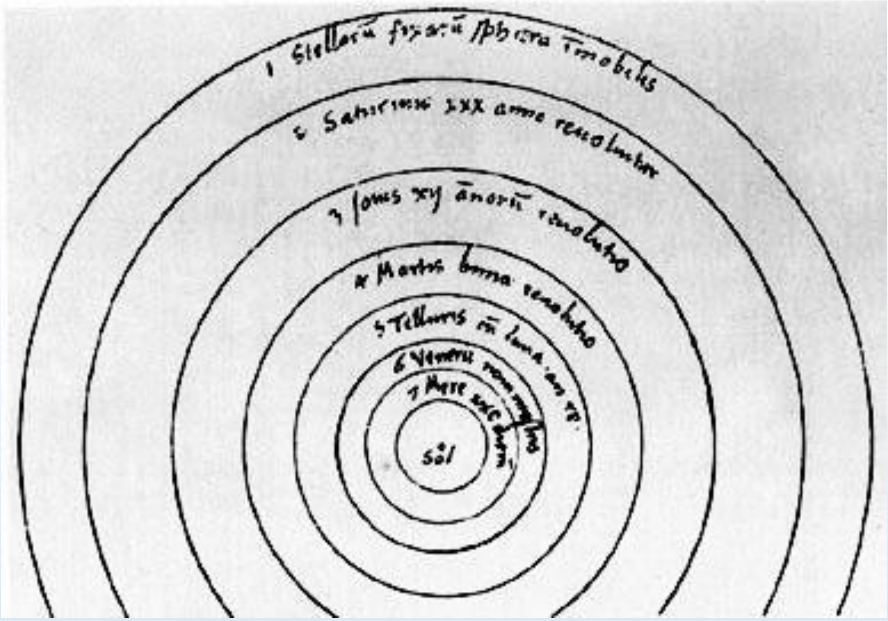
Relaxation time >> experiment time:
Steady state regime or incoherent

J. R. Johansson, et al. QuTiP 2: *Comput. Phys. Commun.* **184**, 1234–1240 (2013).

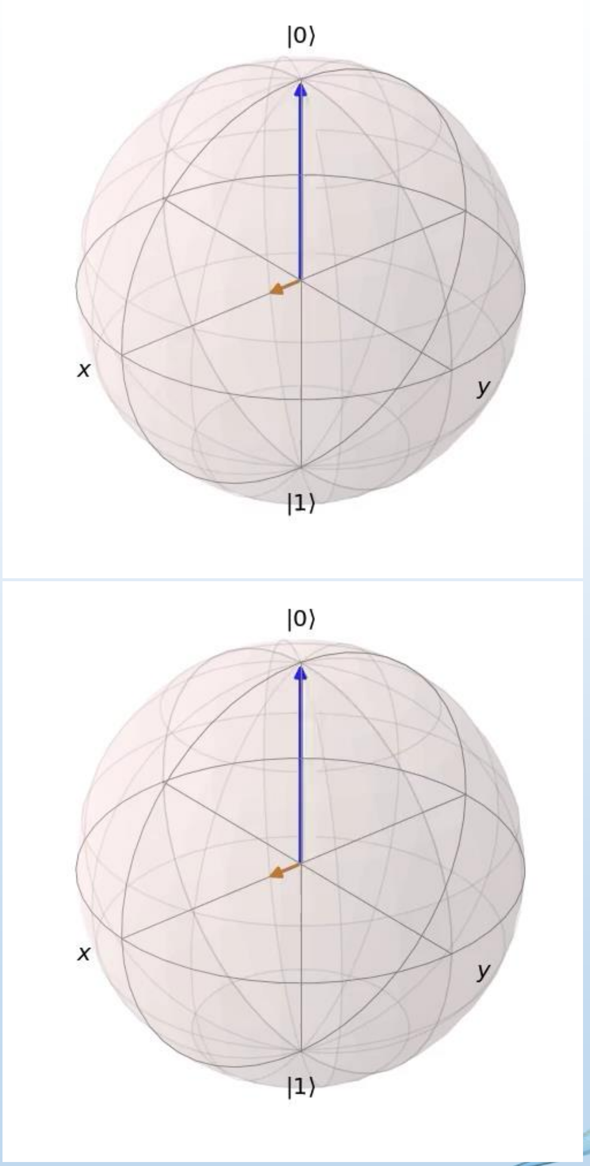
Choice of the frame



Brahe – Kepler (1609)
Astromia nova



Copernic (1530)
De Revolutionibus orbium coelestium



Rotating frame transformation

$$U = \exp(i2\pi f t Sz)$$

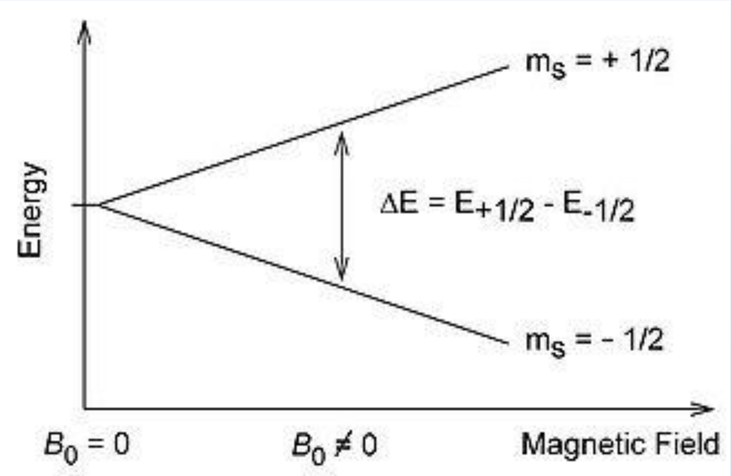
$$\hat{H} = \gamma H_0 Sz + 2hSx\cos(2\pi f t)$$

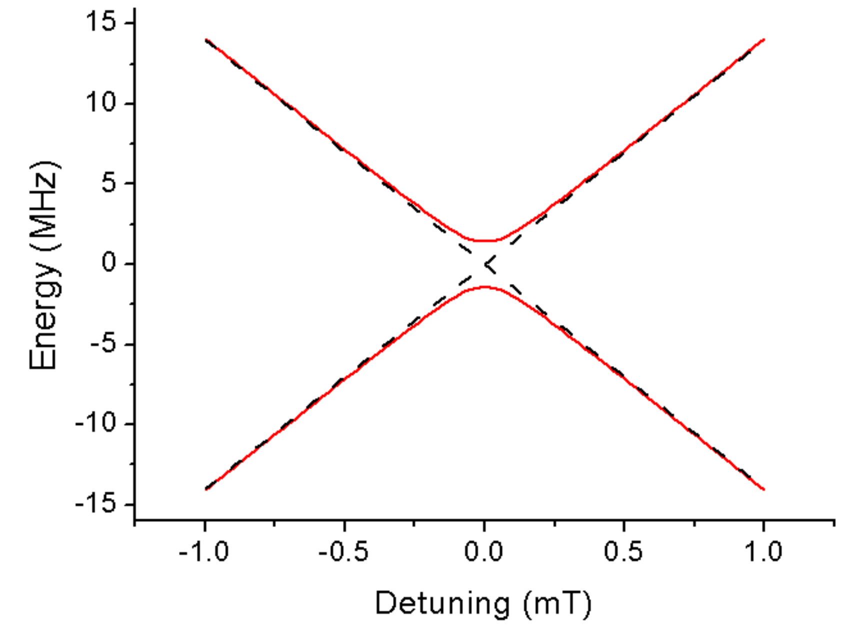
$$\hat{H}rot = \Delta Sz + h(1 + \exp(i4\pi f t))Sx$$

$$\Delta = \gamma H_0 - f$$

Time dependant

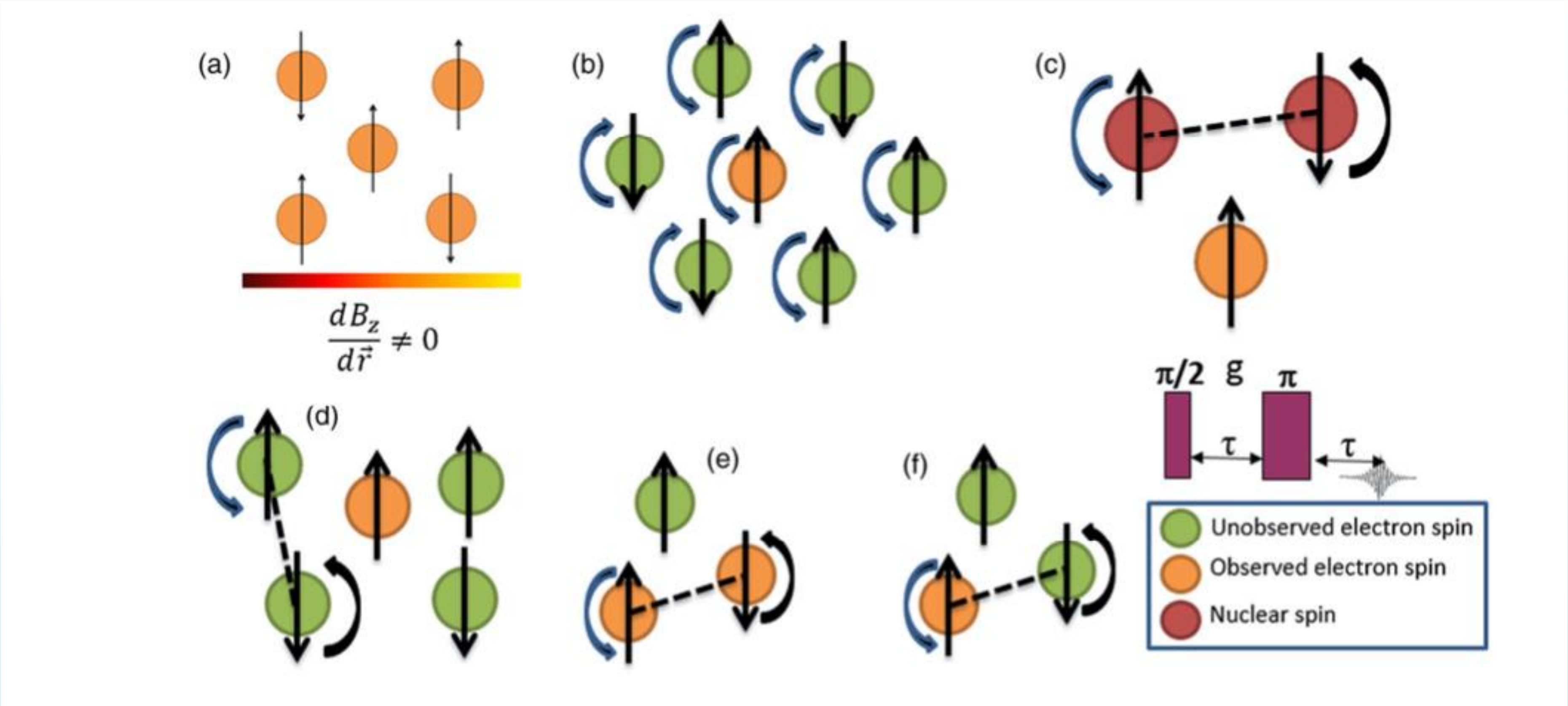
Time independant





Right unitary transformation → problem solved

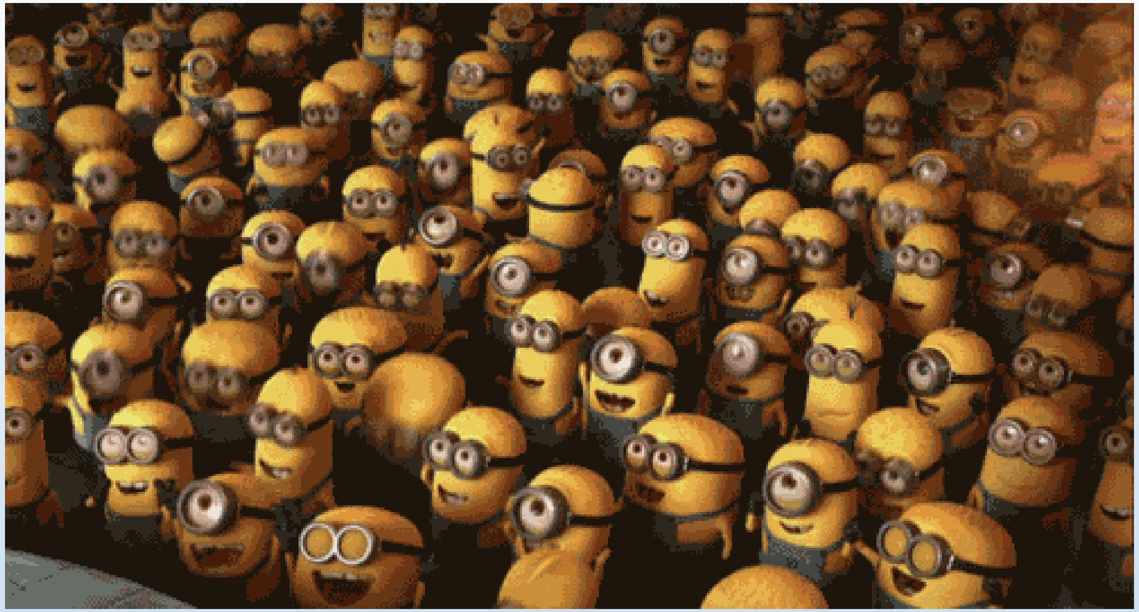
Sources of decoherence



DIKAROV PR Applied 6, 044001 (2016)

Dipole-dipole interaction

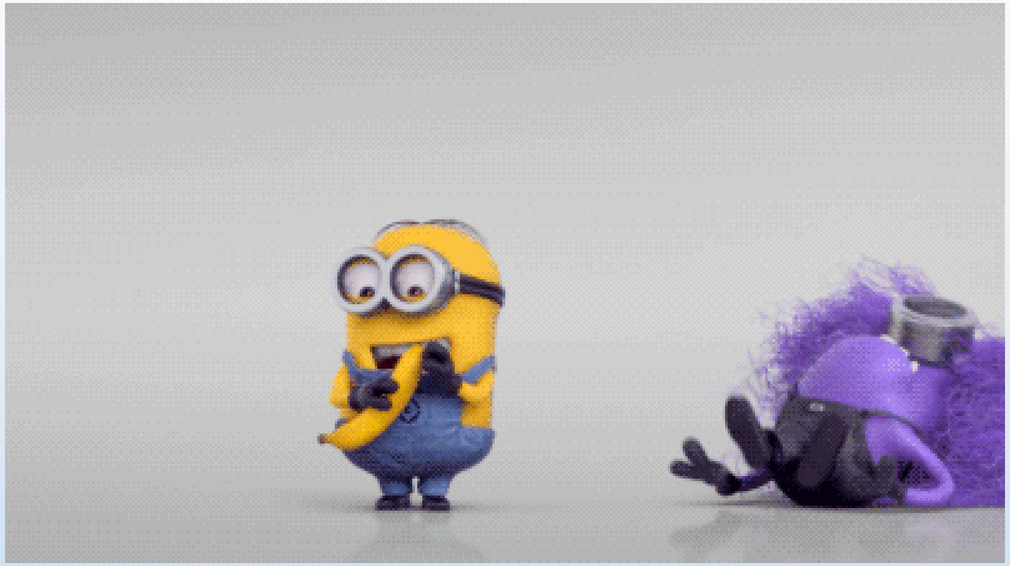
Too many spins and too close → increase the relaxation



Solution: separate them by magnetic dilution (doping material, frozen solution)

(Super)hyperfine interaction

The nuclear spins surrounding the electron spin perturb it



Solution: find systems with few nuclear spins or far from the electron spin / Isotopic enrichment

Effect of temperature : phonon interaction

Temperature too high increases the relaxation

Solution: decrease the temperature

Time to play

Sources of relaxations

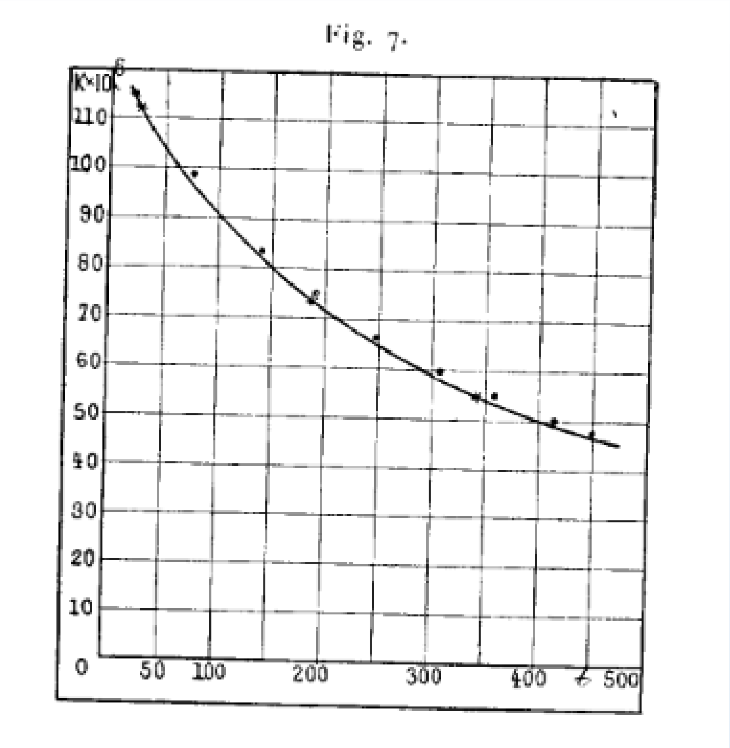
How to increase the coherence regime time ?

- **Dipole-dipole interaction → magnetic dilution**
- **Nuclear spin bath → isotopic enrichment**
- **Spin-phonon interaction → low temperature**

Change the paradigm → use strong correlation

Strongly Correlated Magnets

Diluted magnetism :



Susceptibility of oxygen:
P. Curie 1895 (Thesis)

1D correlation
No order

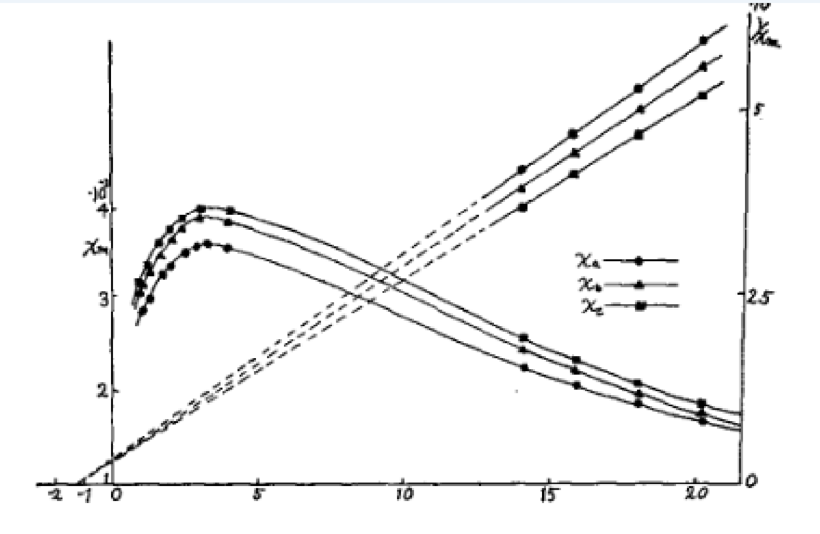
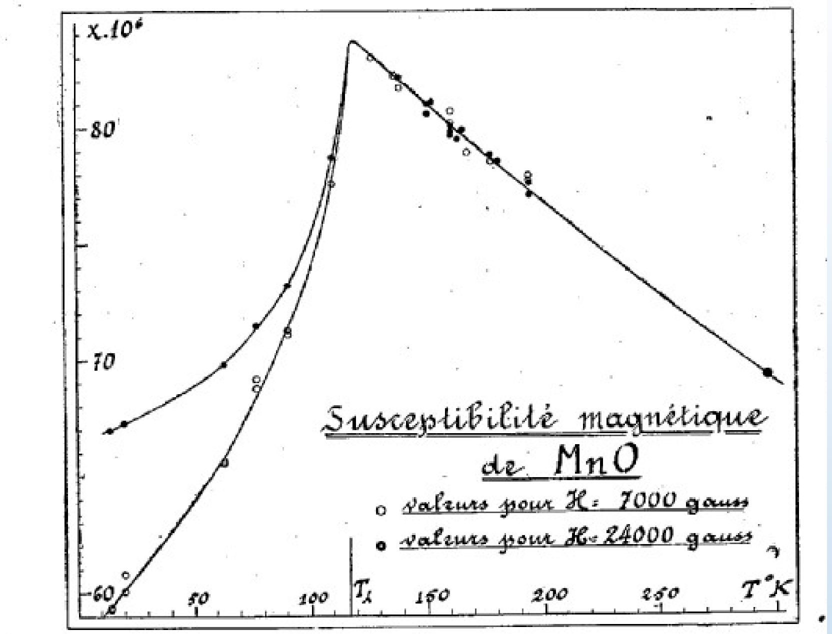


FIG. 1. Molar magnetic susceptibilities and reciprocal susceptibilities of $Cu(NH_3)_4SO_4 \cdot H_2O$ parallel to the a, b, and c axis of the crystal in the range of liquid helium and liquid hydrogen temperatures.

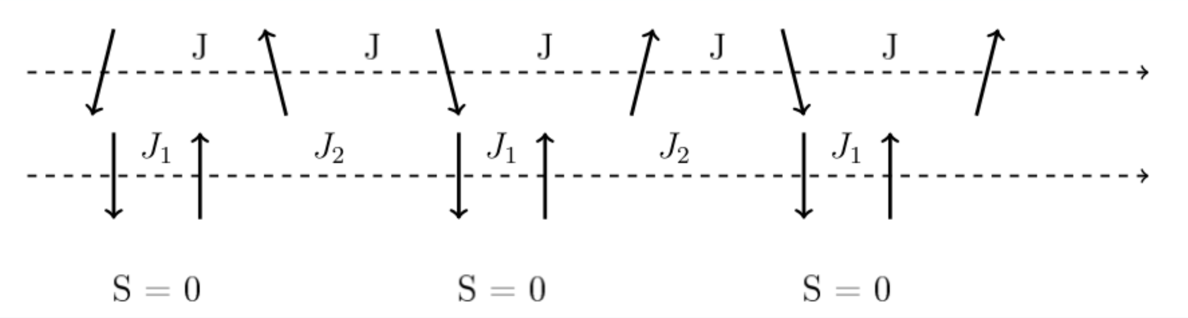
3D correlation
Néel order



MAGNÉTISME. — *Le point de transition λ de la susceptibilité magnétique du protoxyde de manganèse* MnO. Note (¹) de MM. Henri Bizette, Charles F. Squire et Belling Tsaï, transmise par M. Aimé Cotton.

Susceptibility of MnO:
Bizette 1938 (CR Acc. Sci)

Strongly correlated 1D spin systems

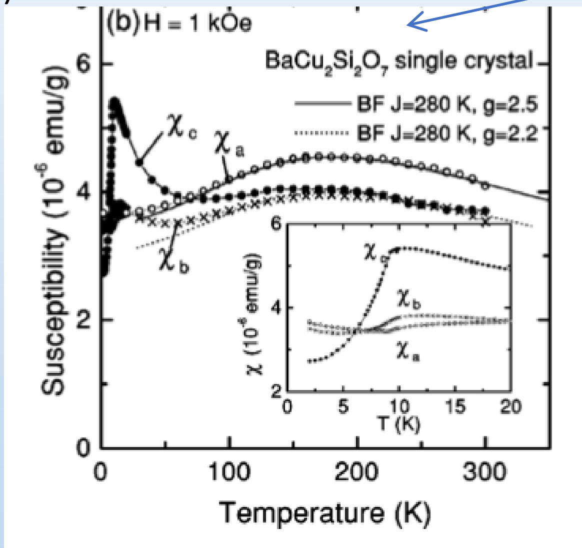


Uniform

Alternated

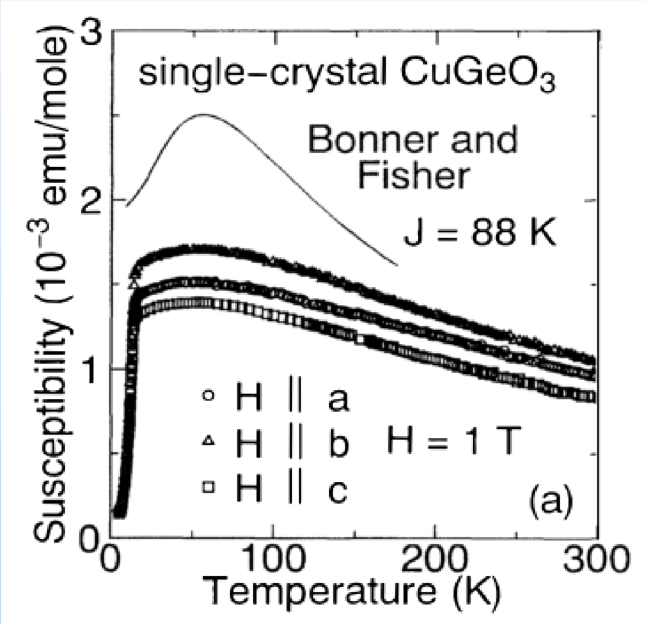
$$H = J \sum_i [(1-\delta) S_{2i-1} S_{2i} + (1+\delta) S_{2i} S_{2i+1}]$$

Uniform spin chain
$KCuF_3$, $BaCu_2Si_2O_7$
BaV_3O_8, Sr_2CuO_3

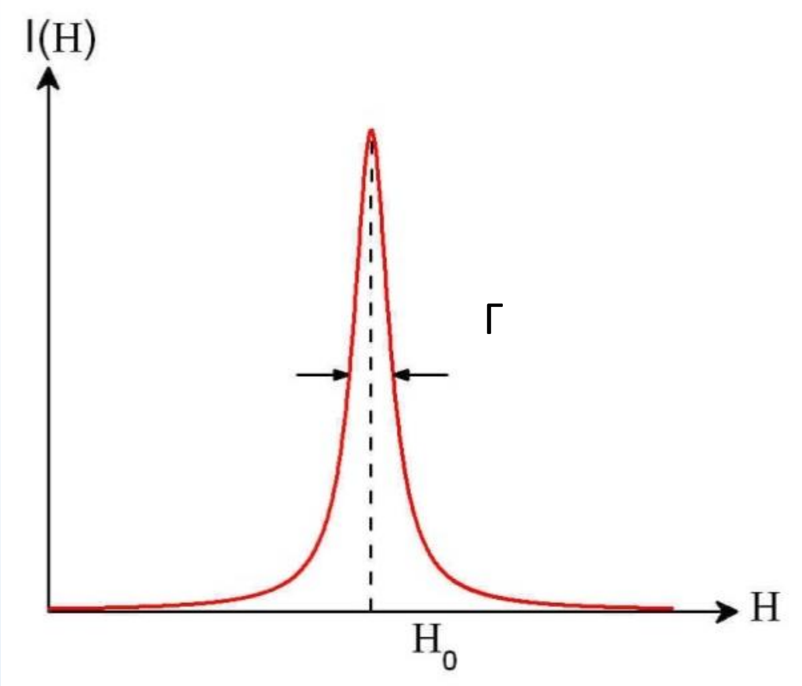


Tsukada PRB 1999

Dimerized spin chain
$CuGeO_3$, NaV_2O_5



1 line – 4 independent information



$\chi_s = \int I(H)dH$ Susceptibility - Kramer Kronig relation.

Γ Linewidth : Dynamic properties

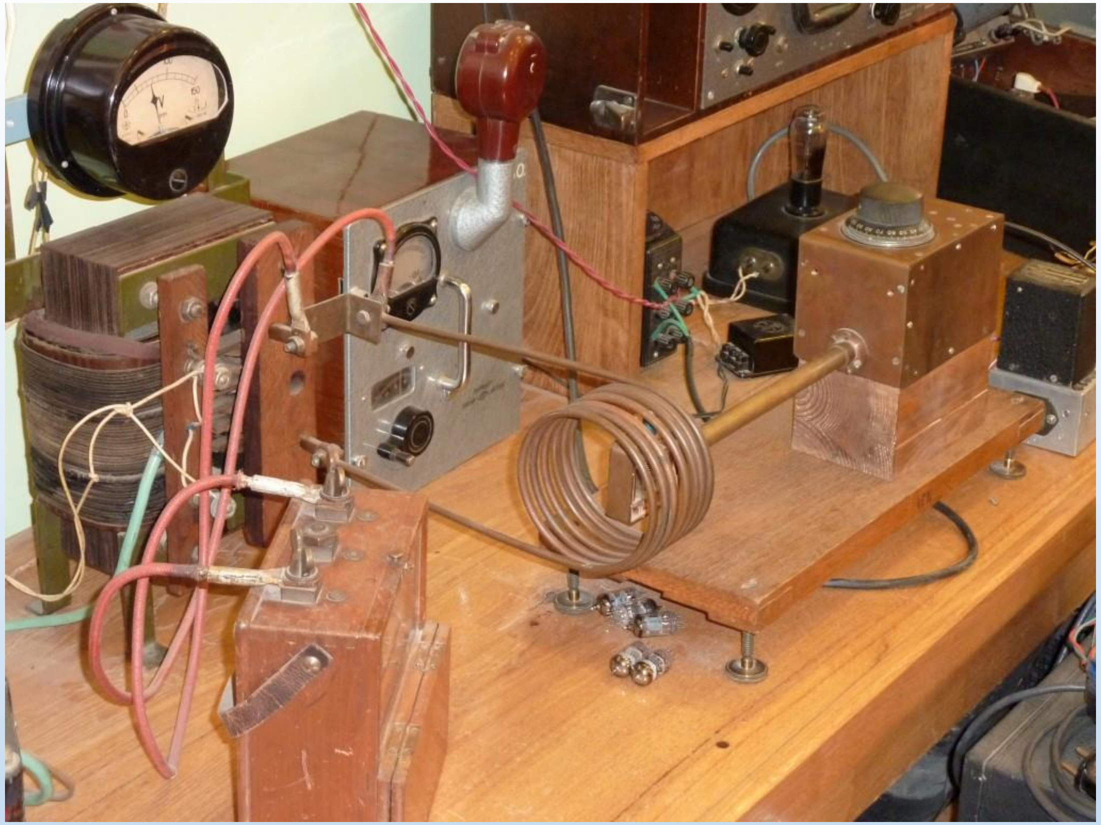
H_0 Resonance field : Local properties (crystal field)

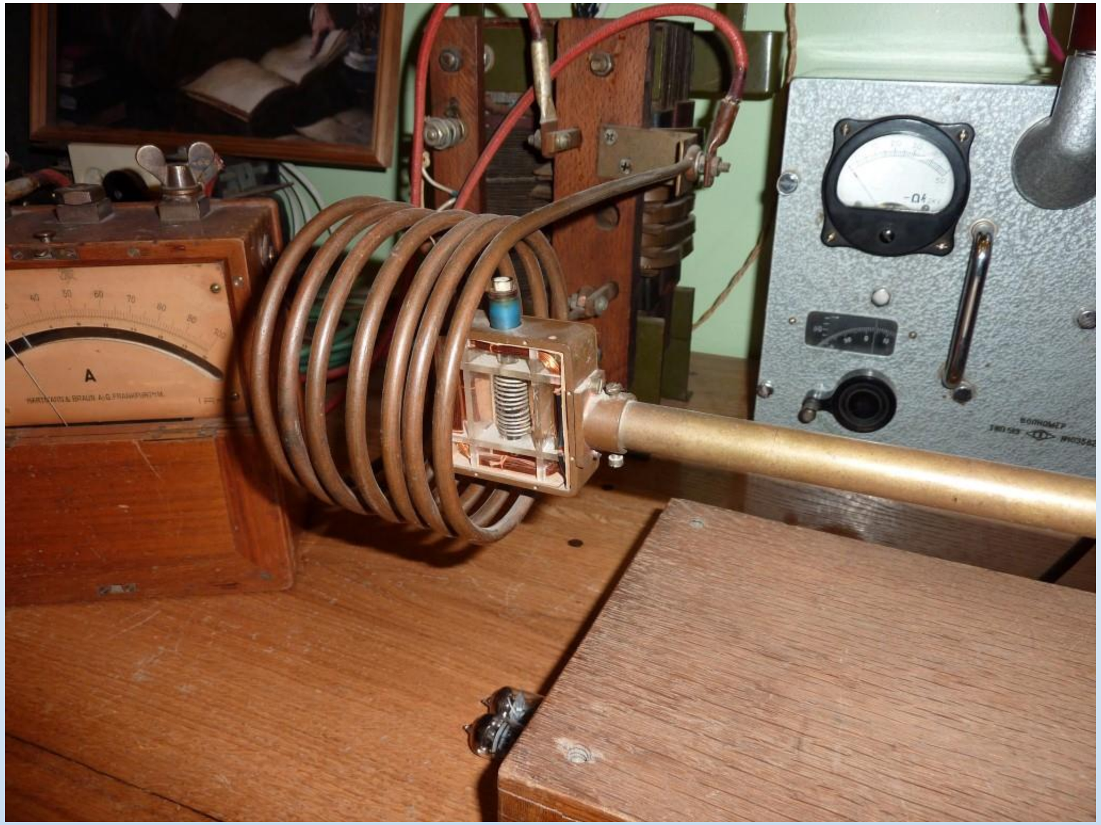
Asymmetry : Dispersion signal - conductivity

$$I(H) = \frac{\Gamma}{(H - H_0)^2 + \Gamma^2}$$

ESR spectrometer – the original one

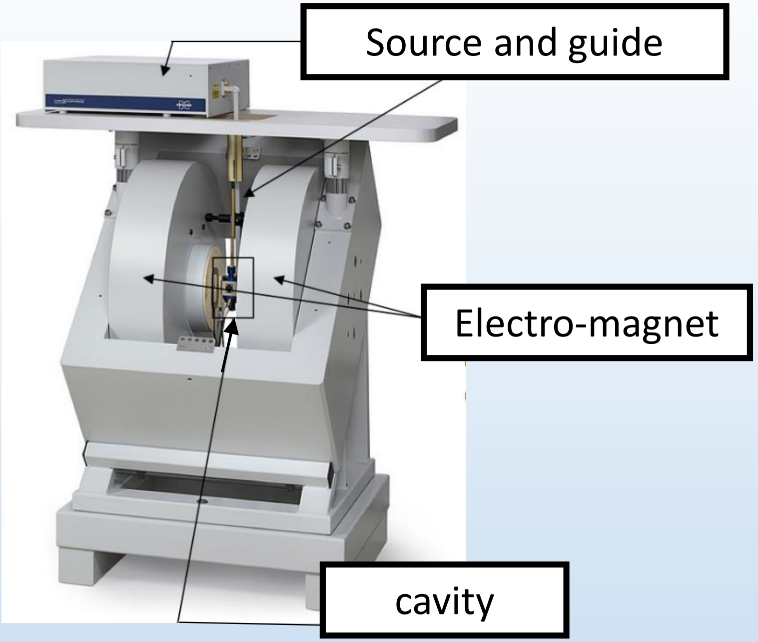
1944 – Zavoiski (Kazan State University)





Electron Paramagnetic Resonance: experiment

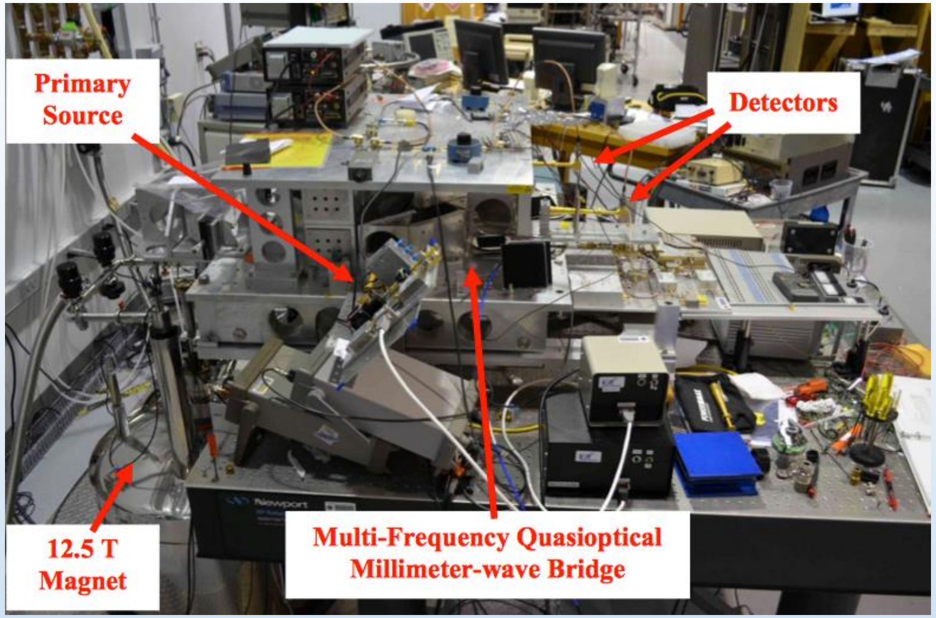
Low frequency



IM2NP, Marseille (France)

- Bruker
- X Band – 10GHz

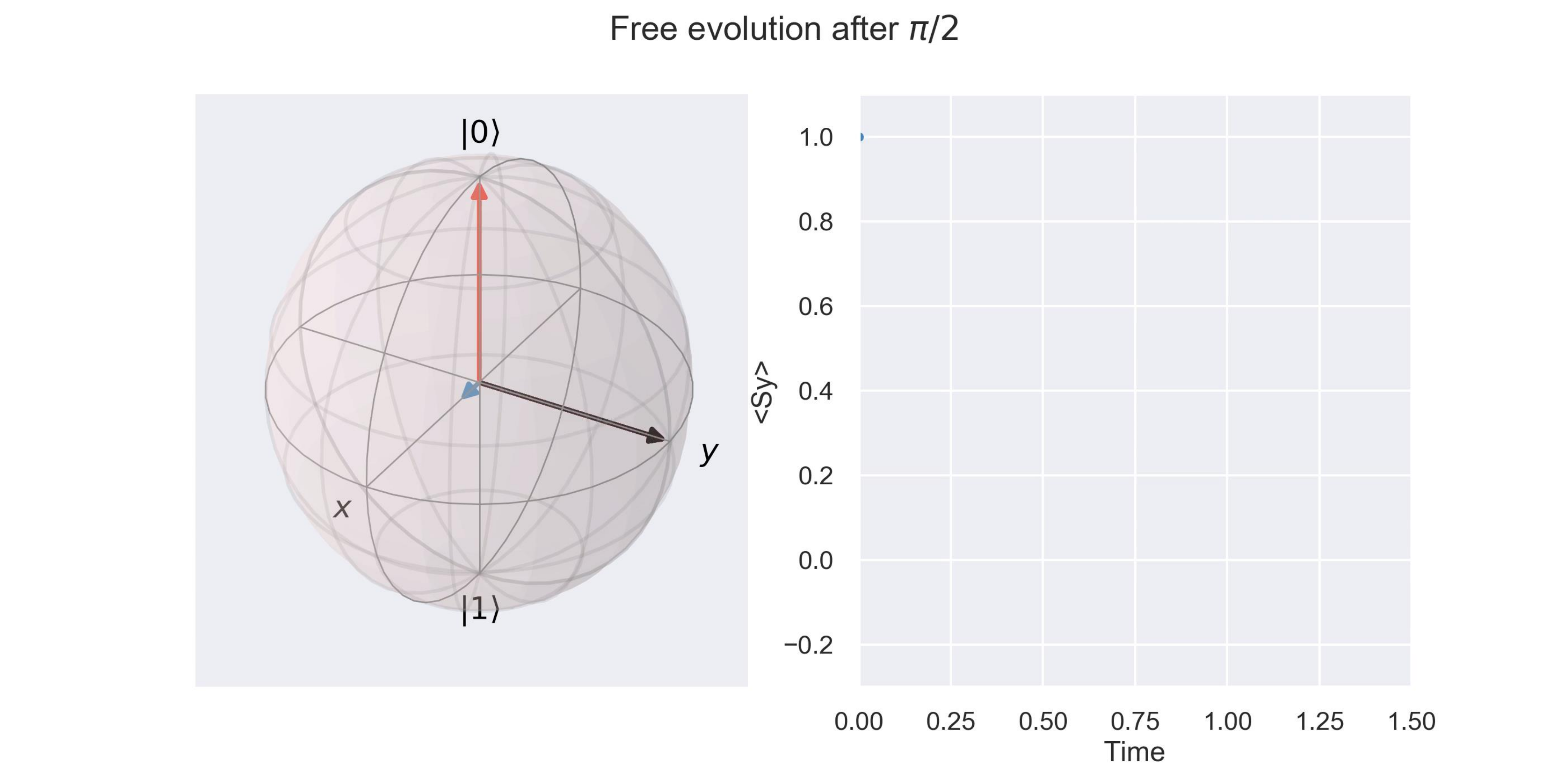
High frequency



NHMFL, Tallahassee (USA)

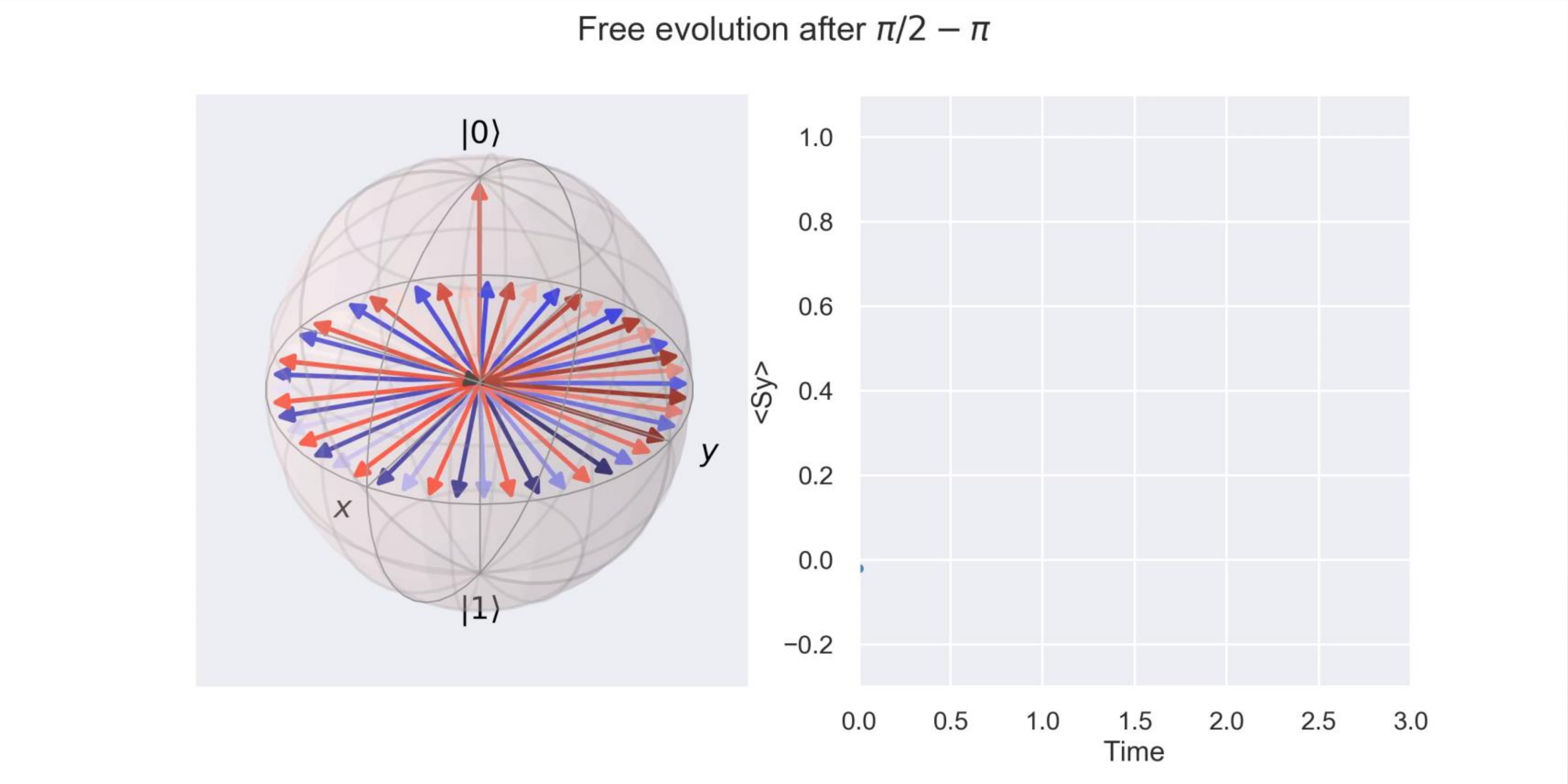
- Quasi-optical superheterodyn
- 120 GHz, 240GHz et 336 GHz
- Champ magnétique: -12.5T à 12.5T

Inhomogeneous line – in the rotating frame

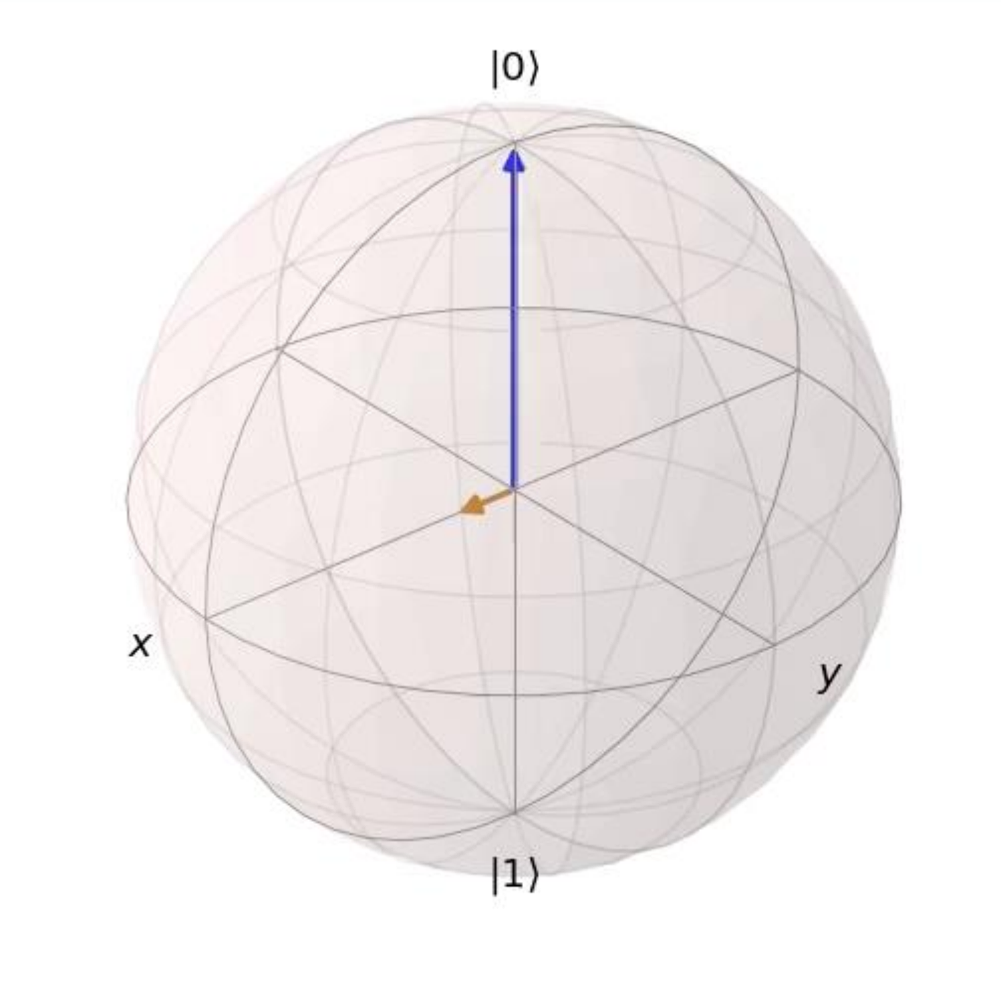


Spin - echo

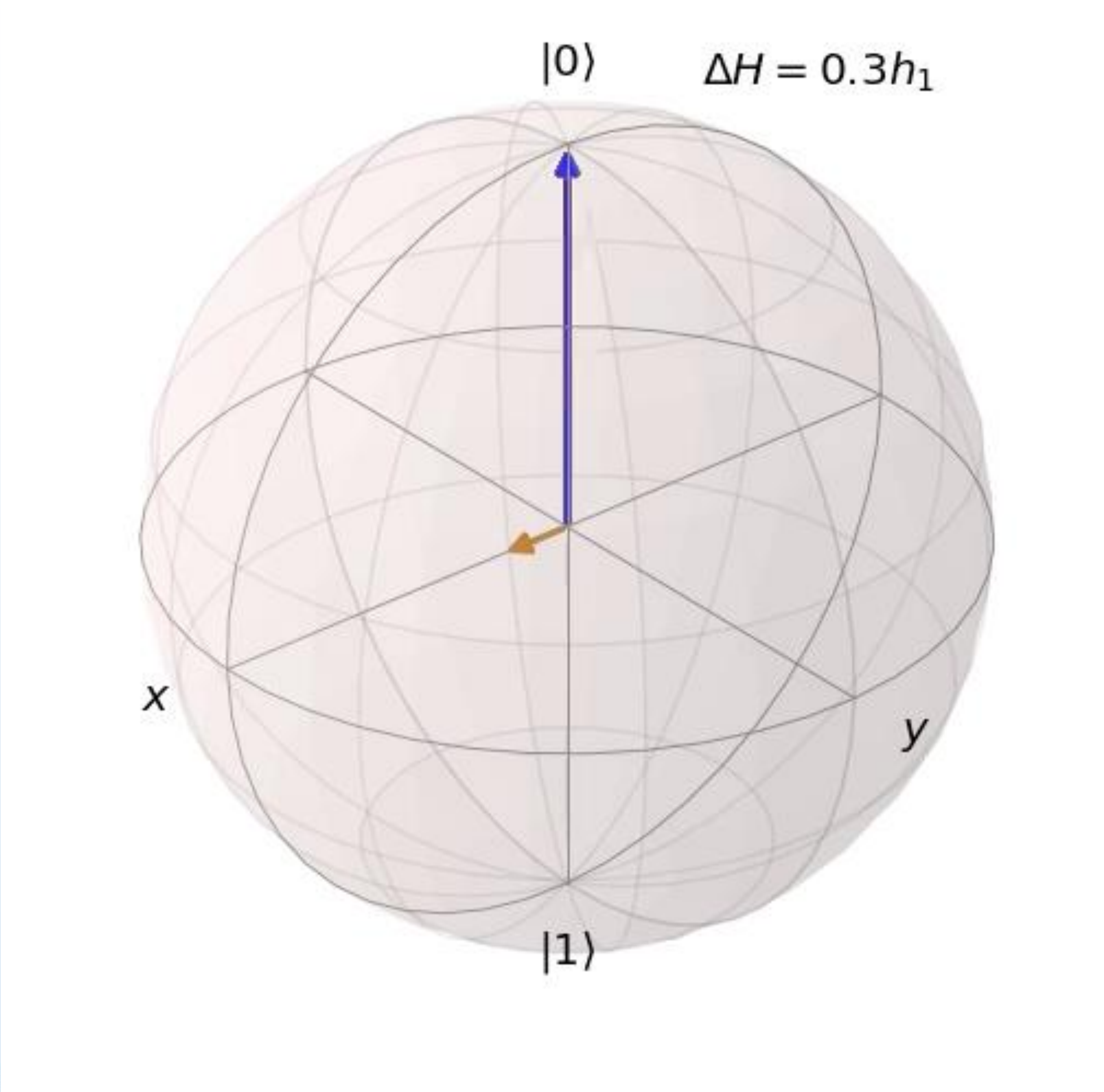
$$\frac{\pi}{2} - free\ evolution\ - \pi - free\ evolution.$$



Hahn echo sequence – Hard pulses

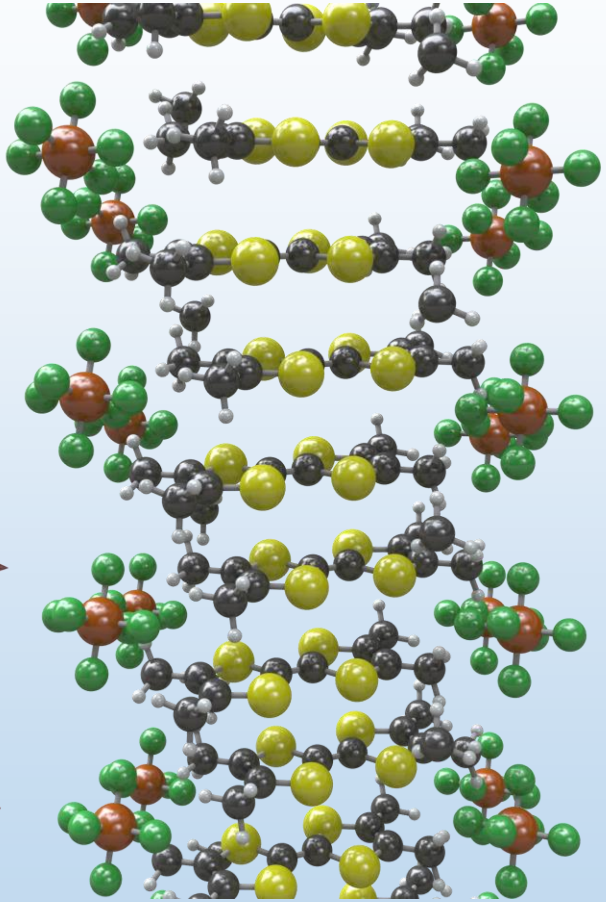


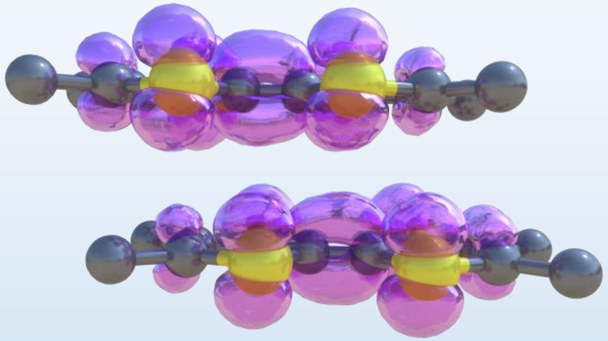
Hahn echo sequence – Soft pulses



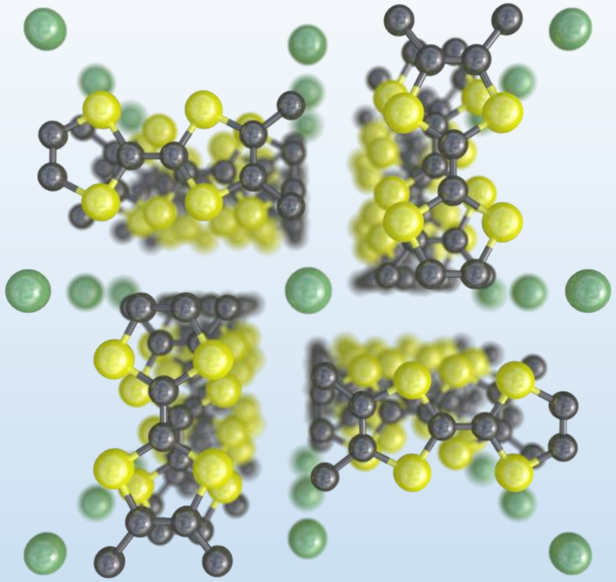
Presentation : organic spin chains

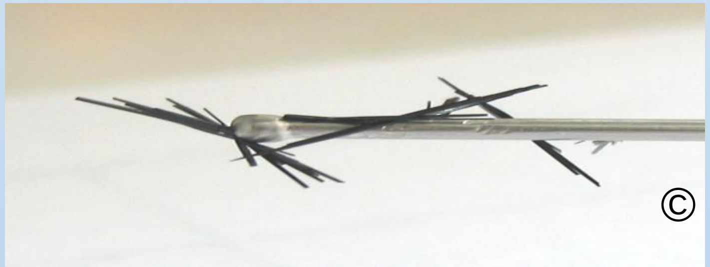
$(TMTTF)_2XF_6$ X=As,P,Sb



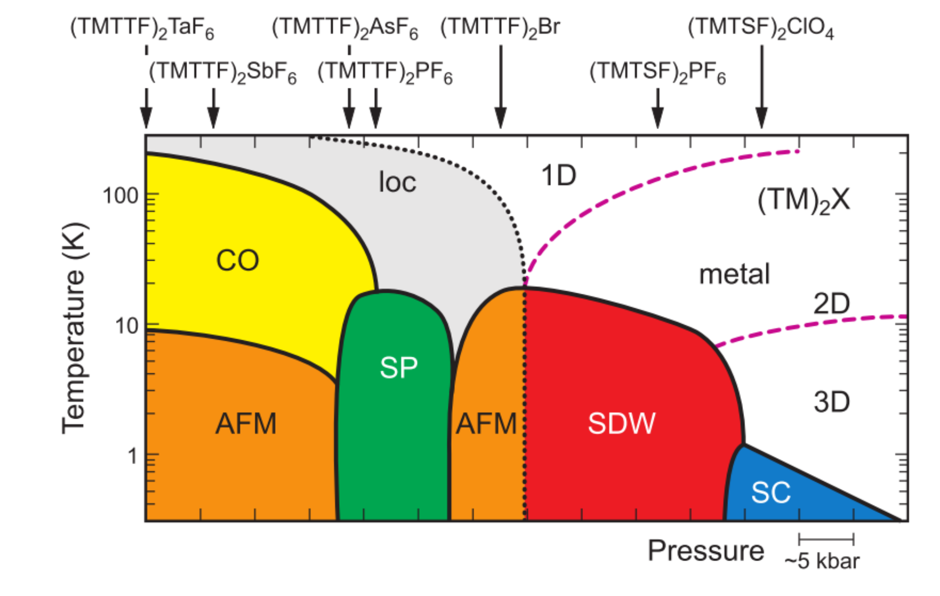


$o(DMTTF)_2X$ X=Cl,Br,I





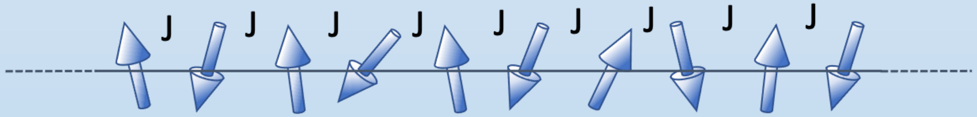
Presentation : organic spin chains

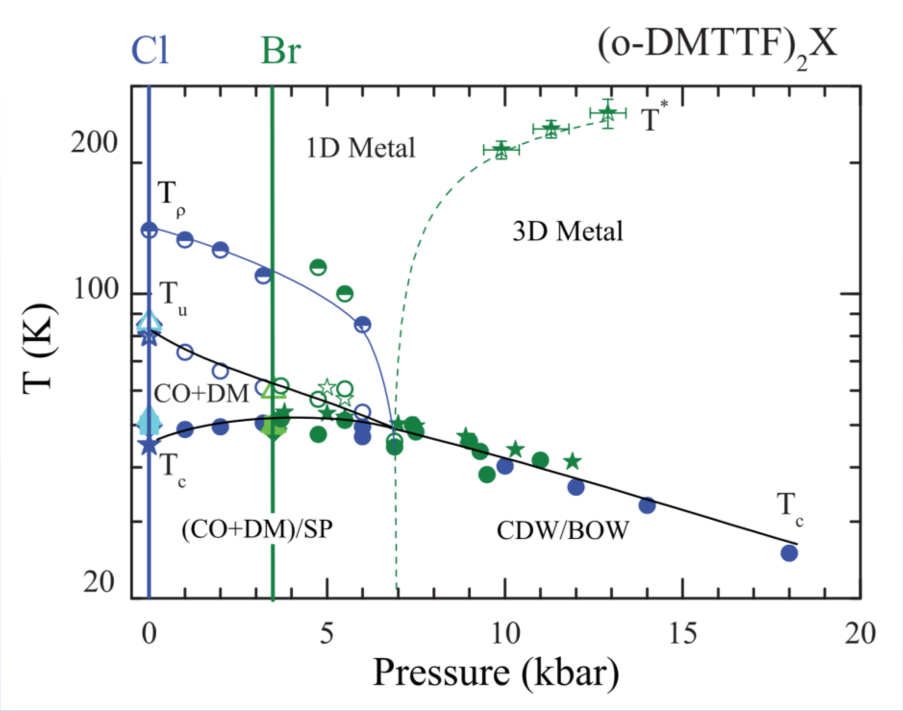


Dressel et al. Crysrallogrphy, **2**, 2012

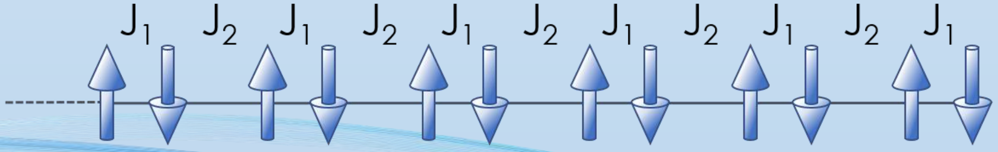
J ~ 400K, 300T, 300cm^{-1}

$$H_{Heisenberg} = J\sum_{i}^{N} S_i . S_{i+1}$$





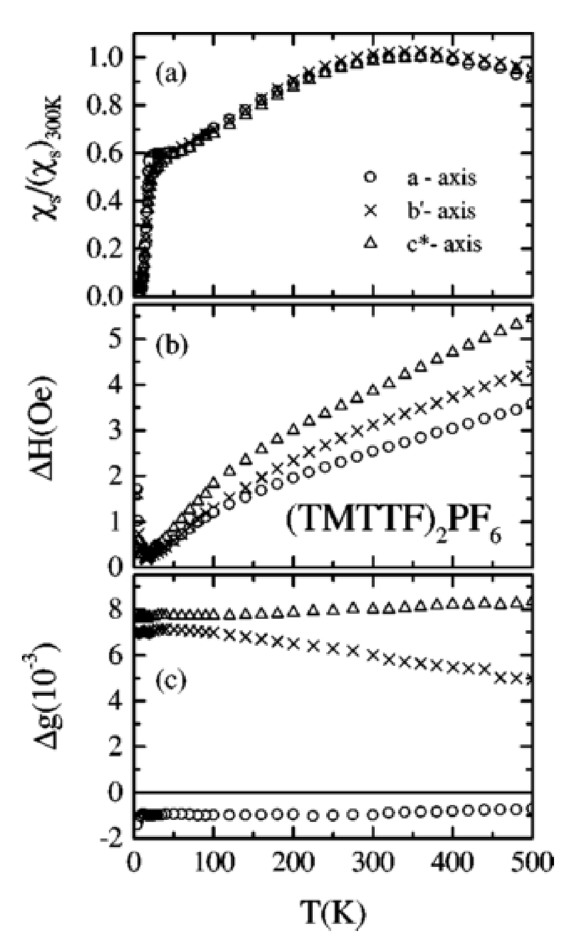
P. Foury-Leylekian, et al. *Phys. Rev. B*. **84**, 195134 (2011).



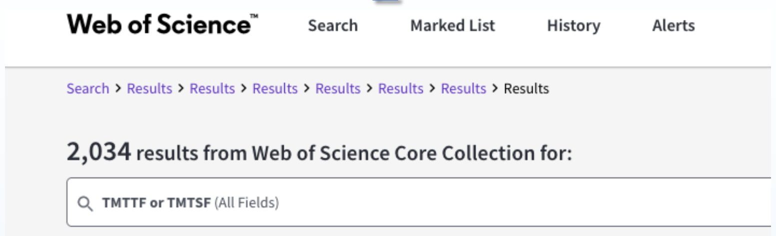
$$\boldsymbol{J_1 = 1 + \delta}$$
$$\boldsymbol{J_2 = 1 - \delta}$$

$$\boldsymbol{H_{SP} = \sum_{i}^{N} J_1 S_{2i-1}.S_{2i} + J_2 S_{2i}.S_{2i+1}}$$

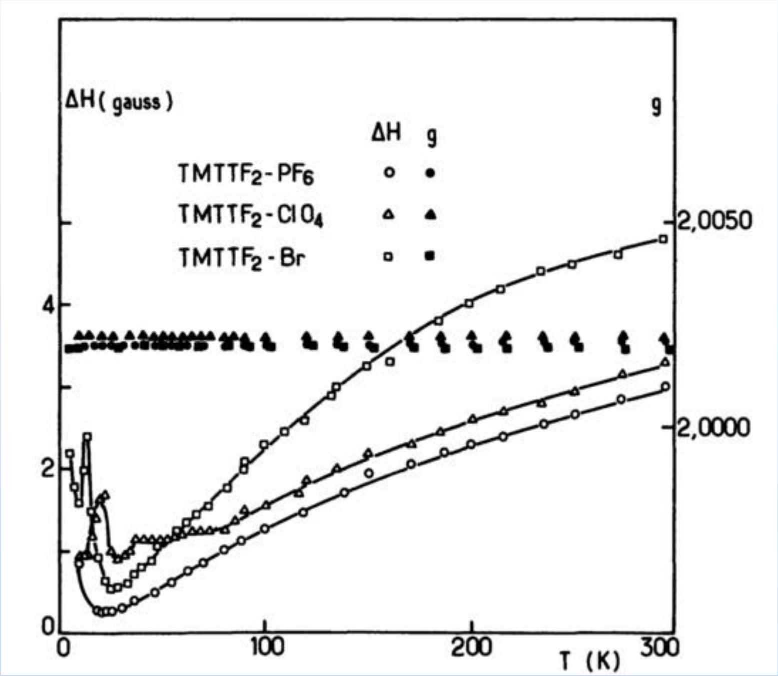
ESR of $(TMTTF)_2X$ – already done ?

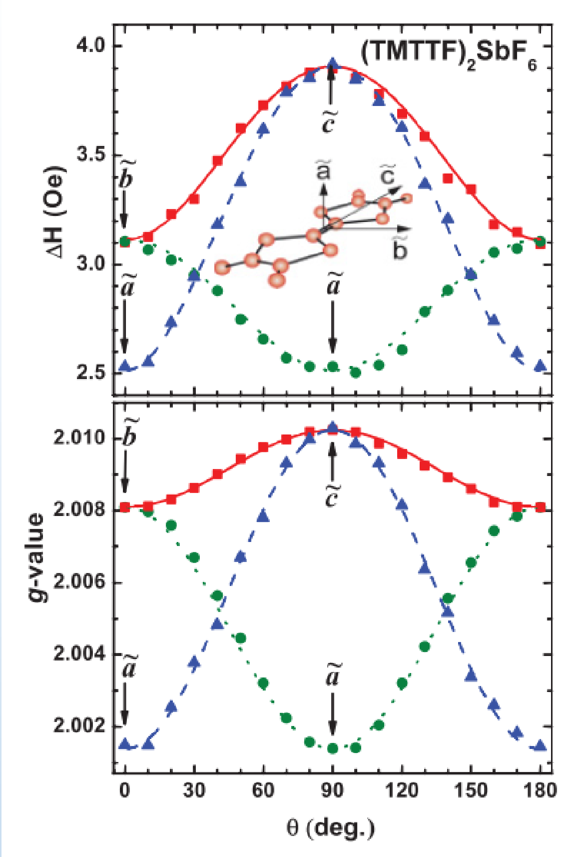


M. Dumm *et al.* Phys. Rev. B **61**, 511 (2000).



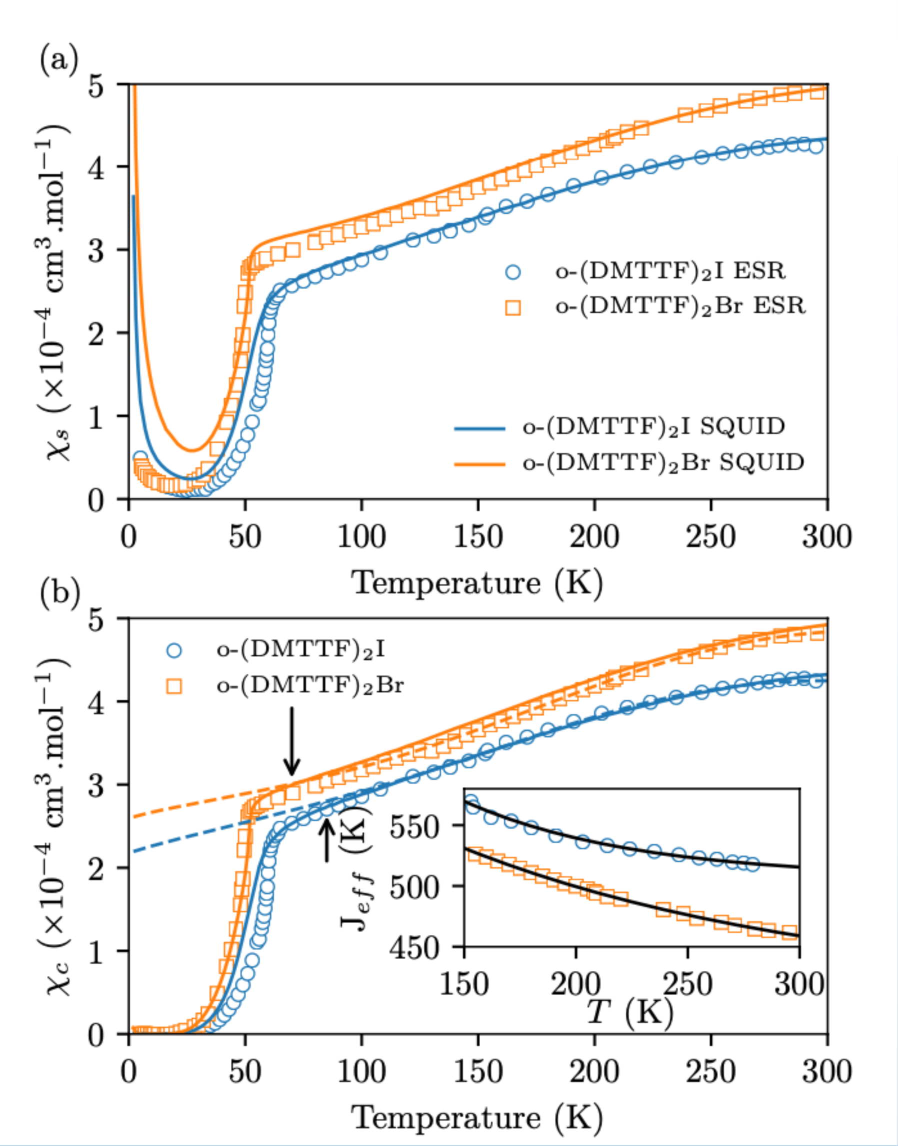
C. Coulon *et al.*, J. Phys. **43**, 1059 (1982).



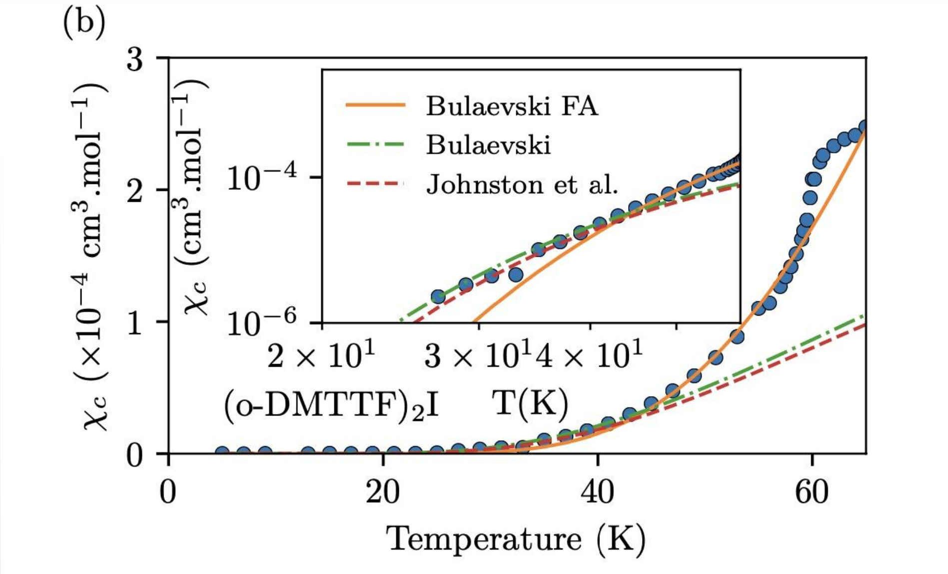


S. Yasin *et al.* Phys. Rev. B **85**, 144428 (2012).

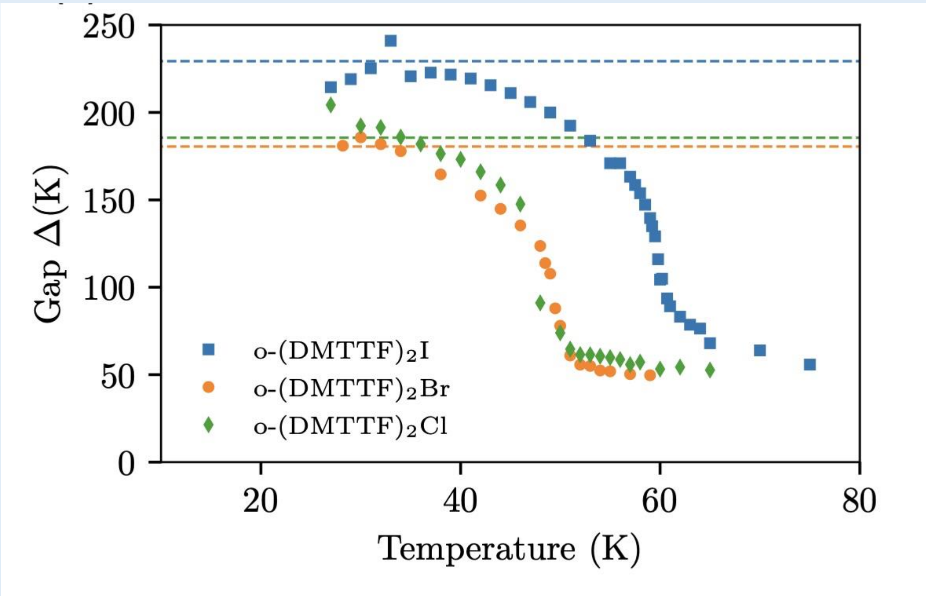
The chain



(a)
o-(DMTTF)2I ESR
o-(DMTTF)2Br ESR
o-(DMTTF)2I SQUID
o-(DMTTF)2Br SQUID
χs (×10−4 cm3.mol−1)
Temperature (K)
(b)
o-(DMTTF)2I
o-(DMTTF)2Br
χc (×10−4 cm3.mol−1)
Jeff (K)
T (K)
Temperature (K)

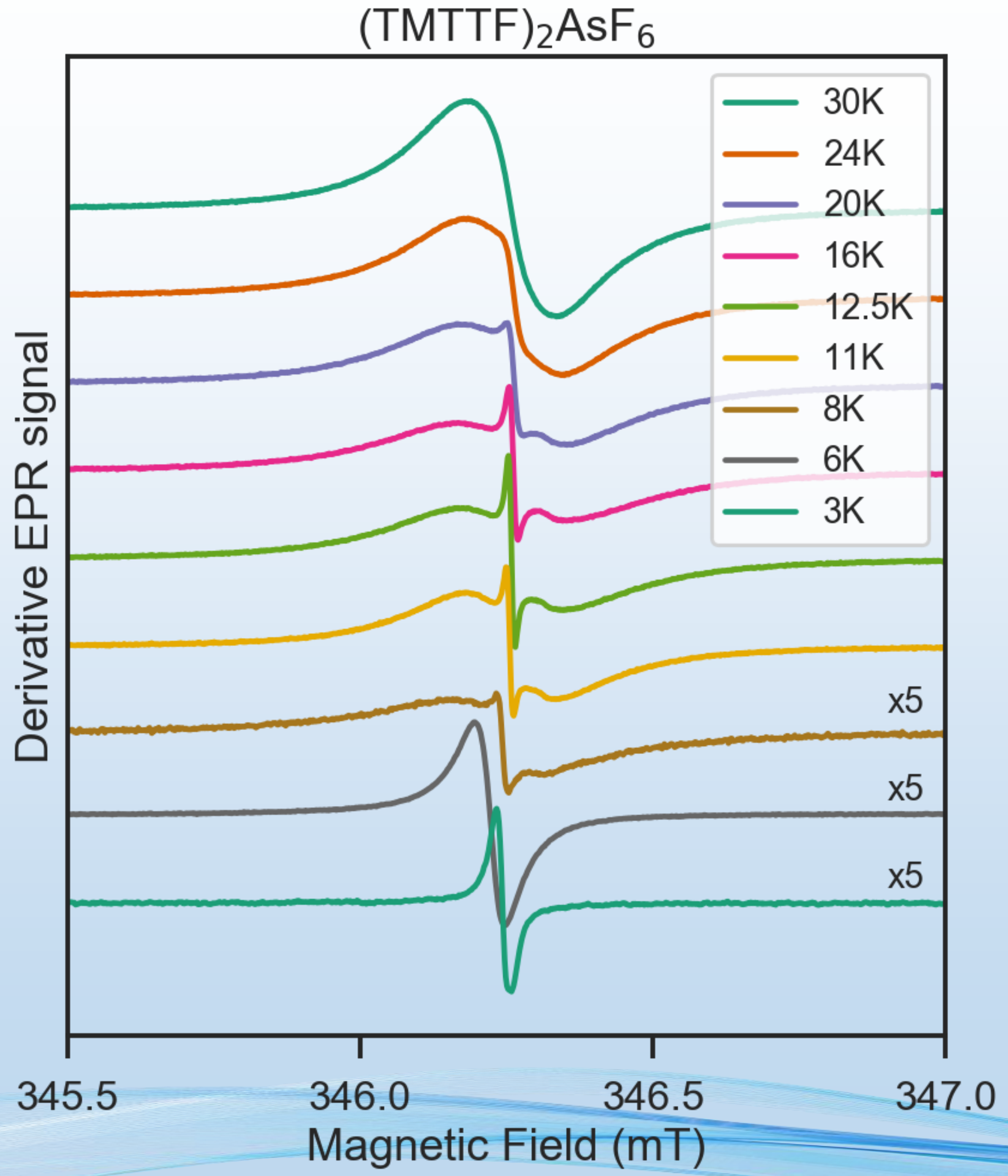


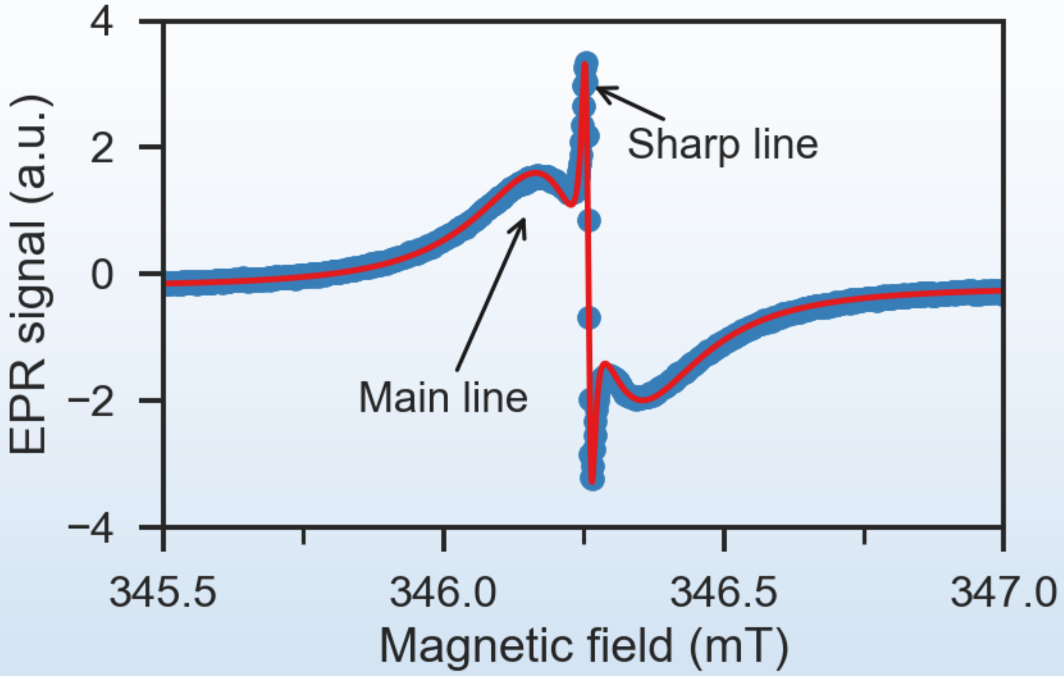
(b)
Bulaevski FA
Bulaevski
Johnston et al.
χc (cm3.mol−1)
(o-DMTTF)2I
T(K)
χc (×10−4 cm3.mol−1)
Temperature (K)



Gap Δ(K)
o-(DMTTF)2I
o-(DMTTF)2Br
o-(DMTTF)2Cl
Temperature (K)

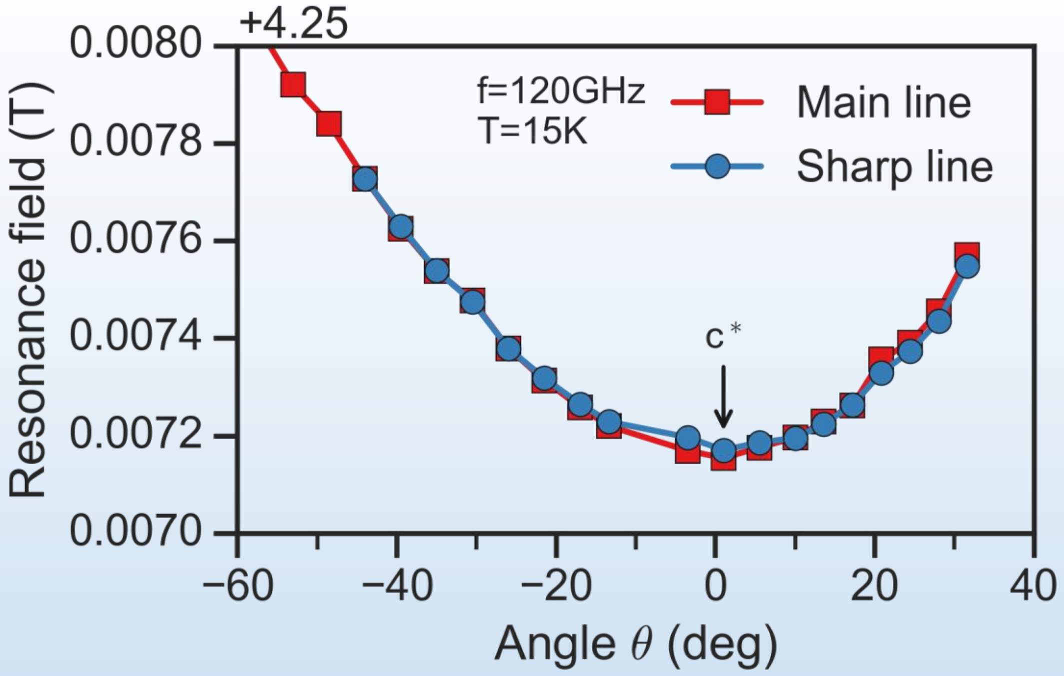
CW - ESR

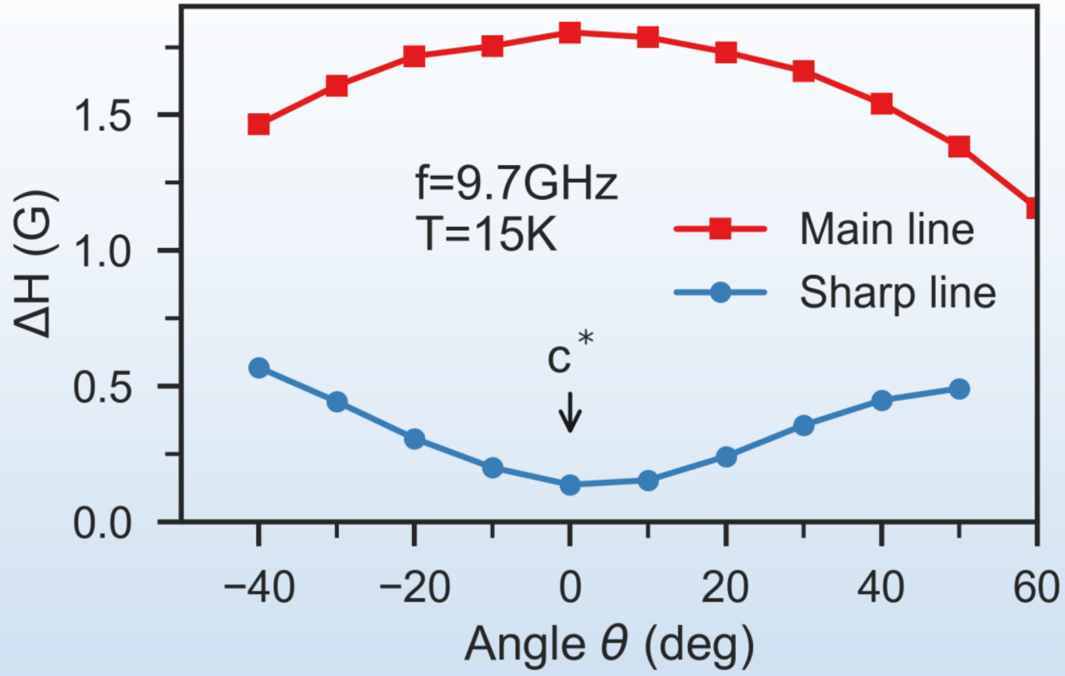




S. Bertaina, et al .*Phys. Procedia*. **75**, 23–28 (2015).

Properties of this new ESR line



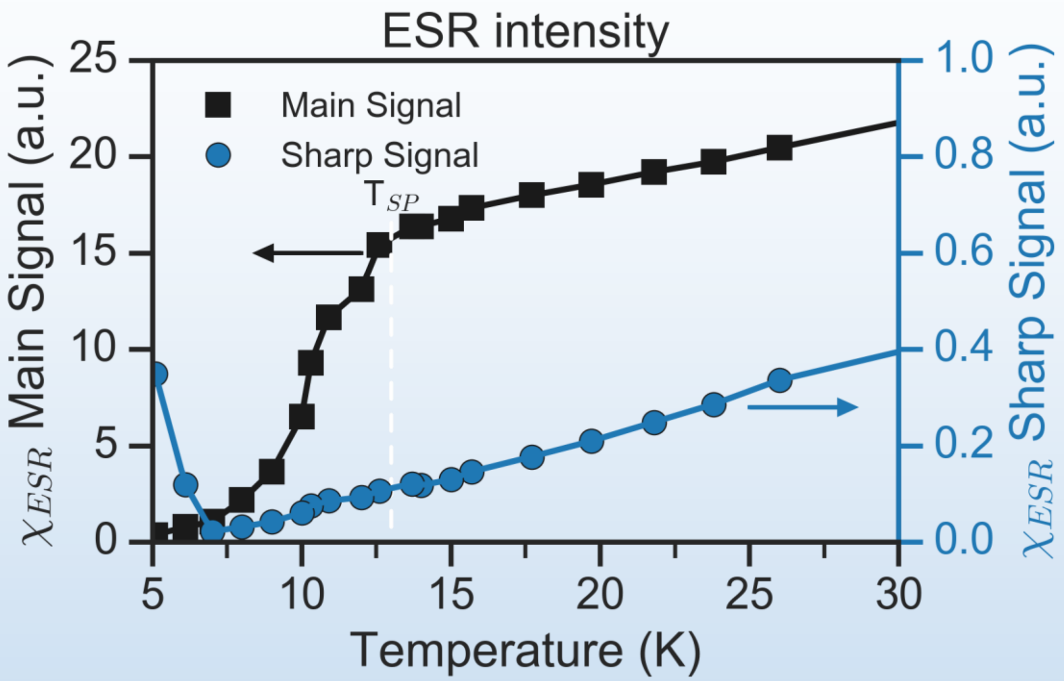


Sharp and Main lines feel the same local field : the “impurity” is related to the chain

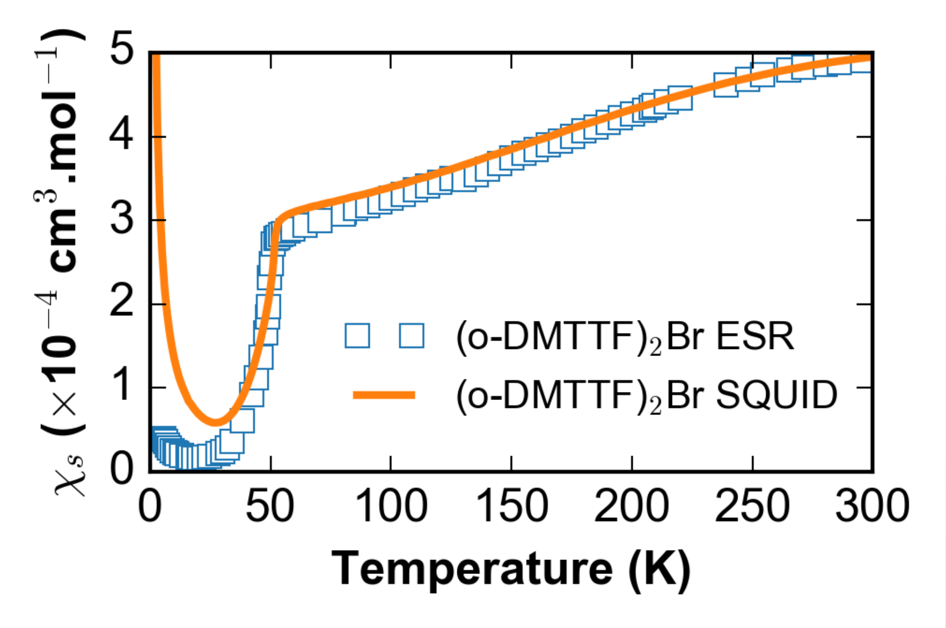
The dynamics of the chain and the “impurity” are different.

Susceptibility

$(TMTTF)_2AsF_6$



$o(DMTTF)_2X$



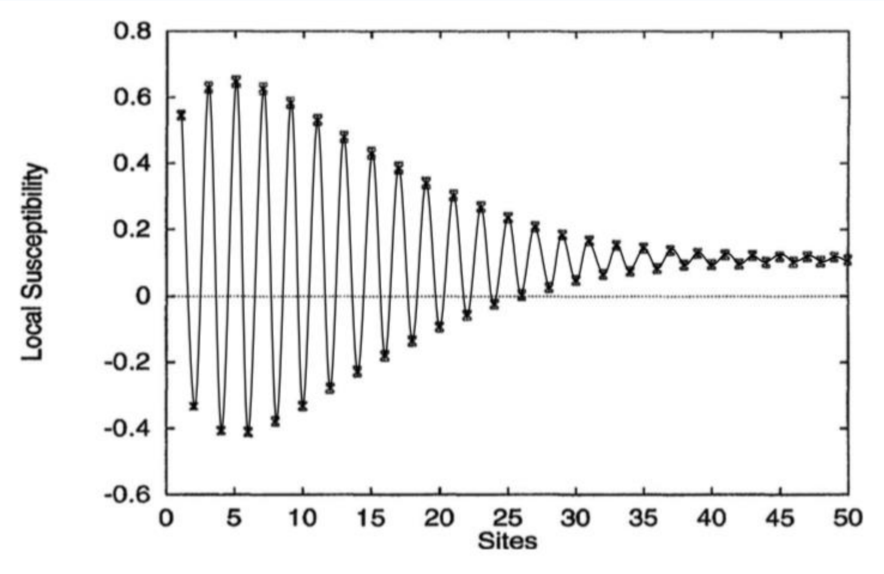
P. Foury-Leylekian, et al. Phys. Rev. B. 84, 195134 (2011).

J. Zeisner, et al. *Phys. Rev. B*. **100**, 224414 (2019).

Conclusion: the defect is inside the spin chain

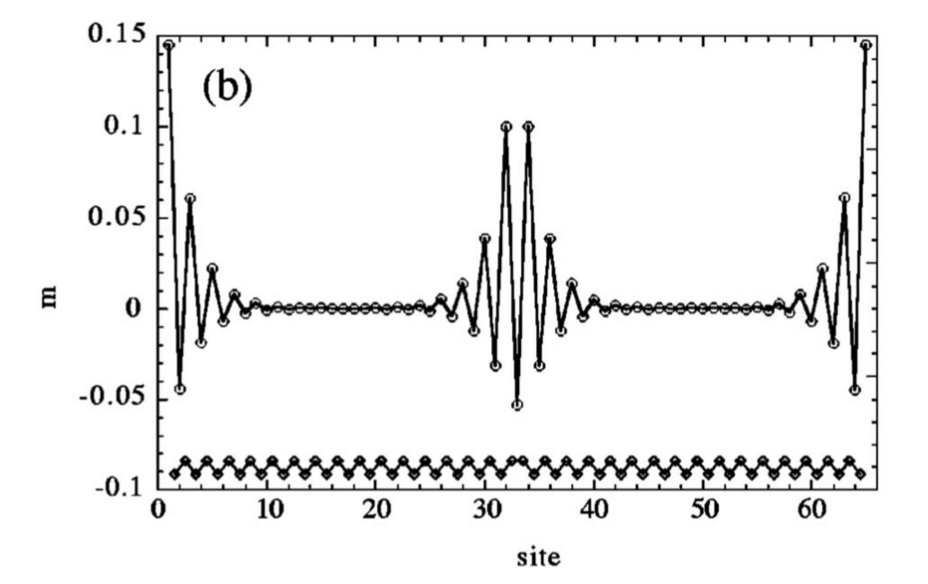
Break the perfection : cut the chain

End chain in uniform Heisenberg chain



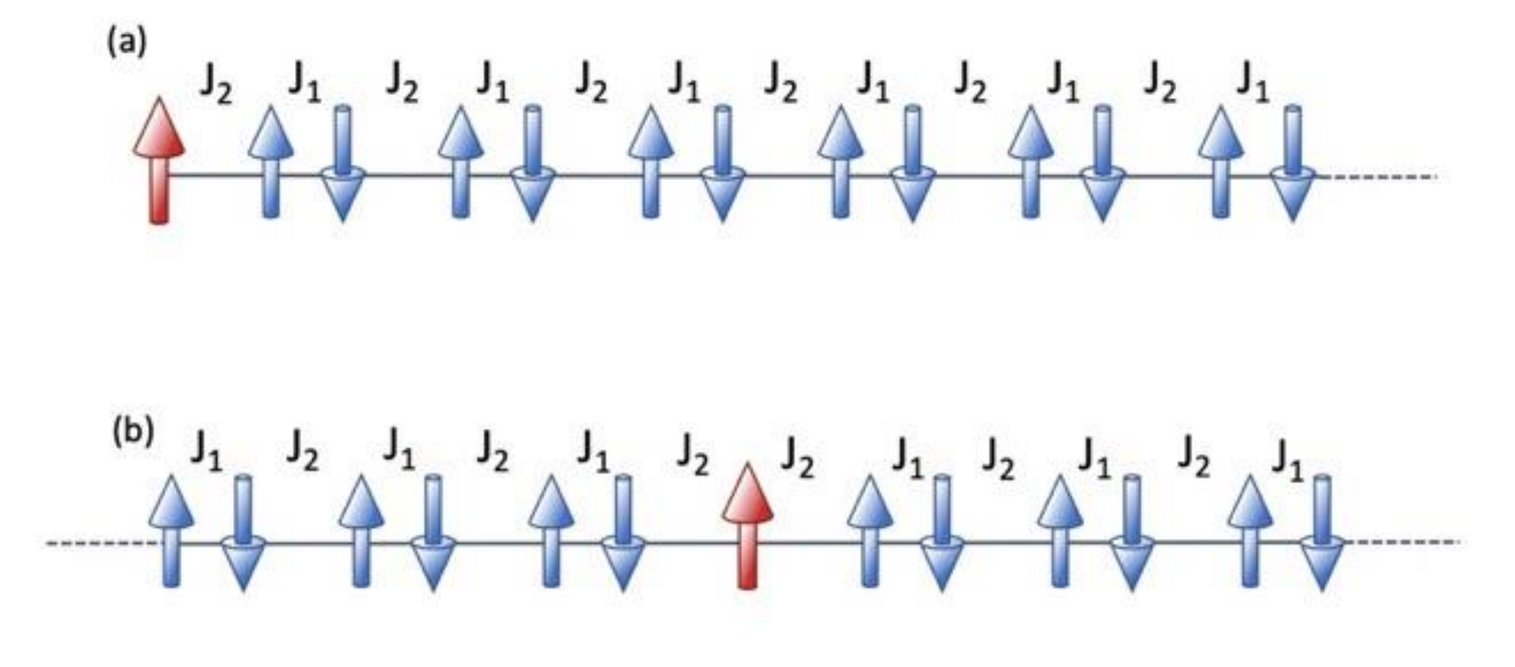
Eggert, Affleck *Physical Review Letters* 75, 934–937 (1995).

End chain and stacking fault in alternated Heisenberg chain

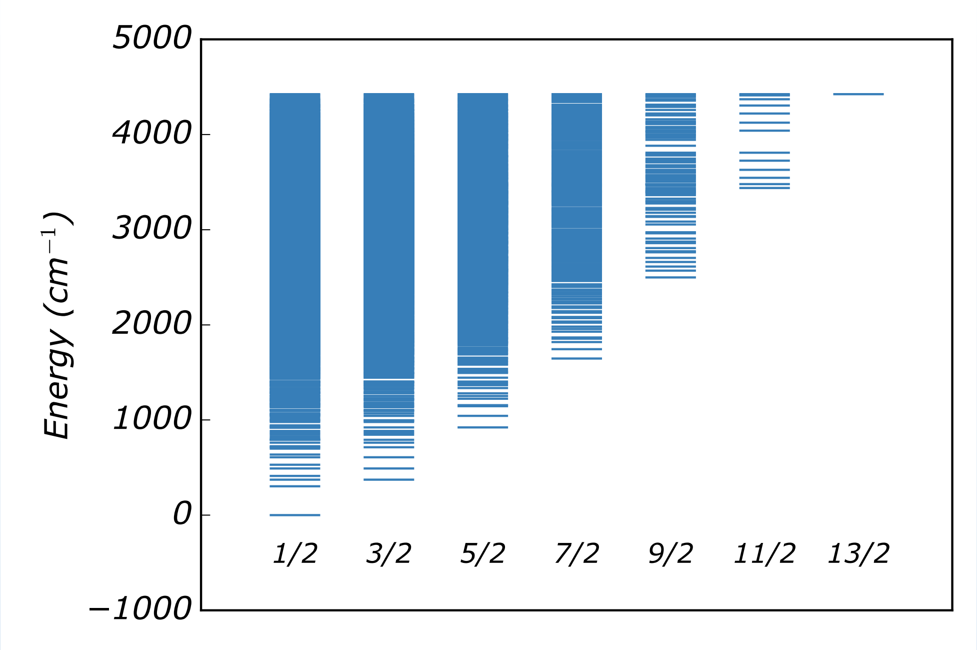


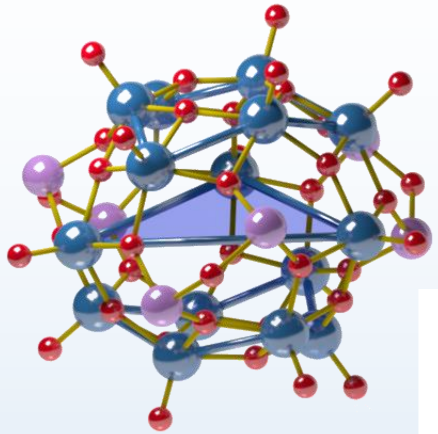
Nishino et al, *Physical Review B* 62, nº 14 (2000) 9463–9471.

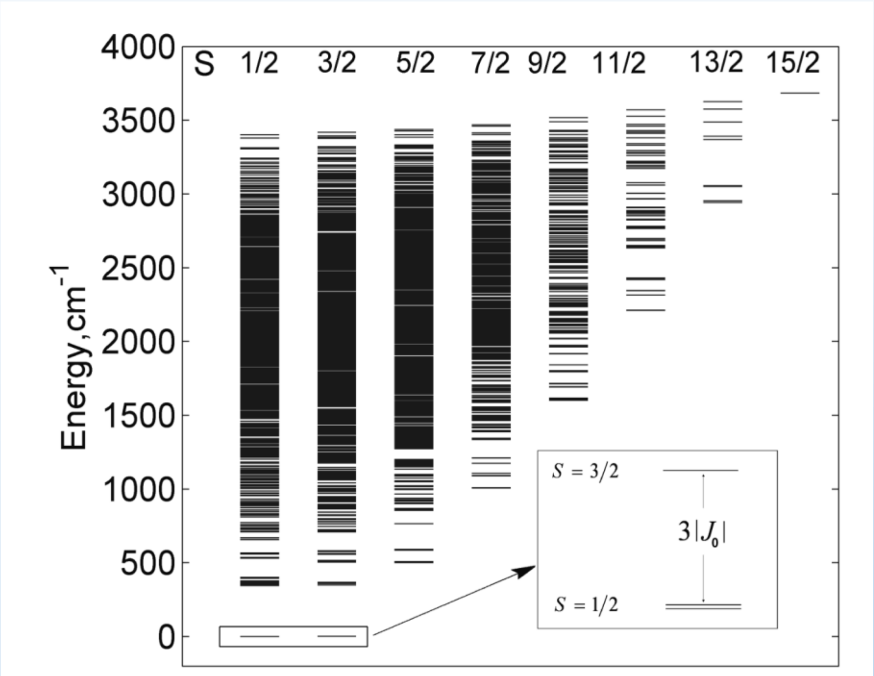
Impurity in a spin chain



Virtual Molecular Magnet

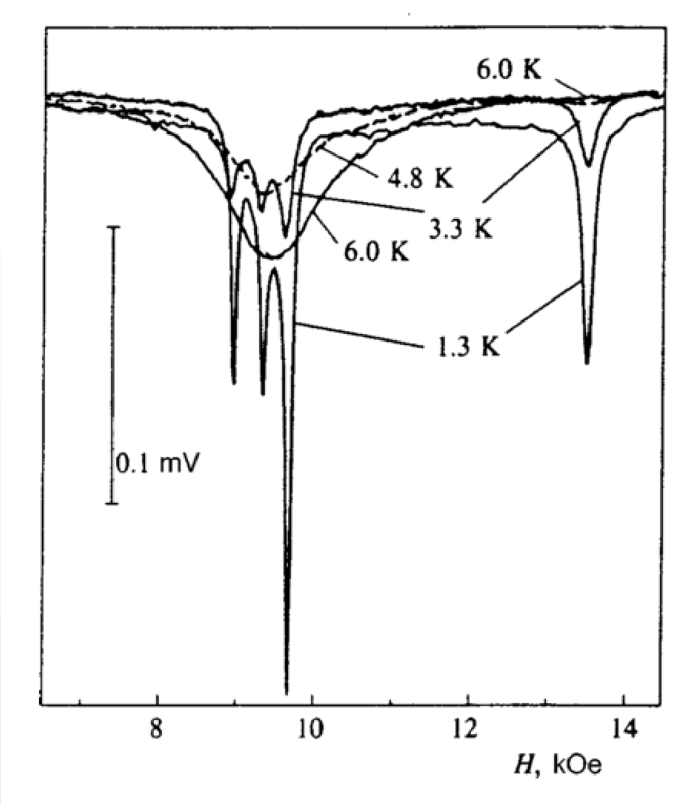






Soliton in $CuGeO_3$

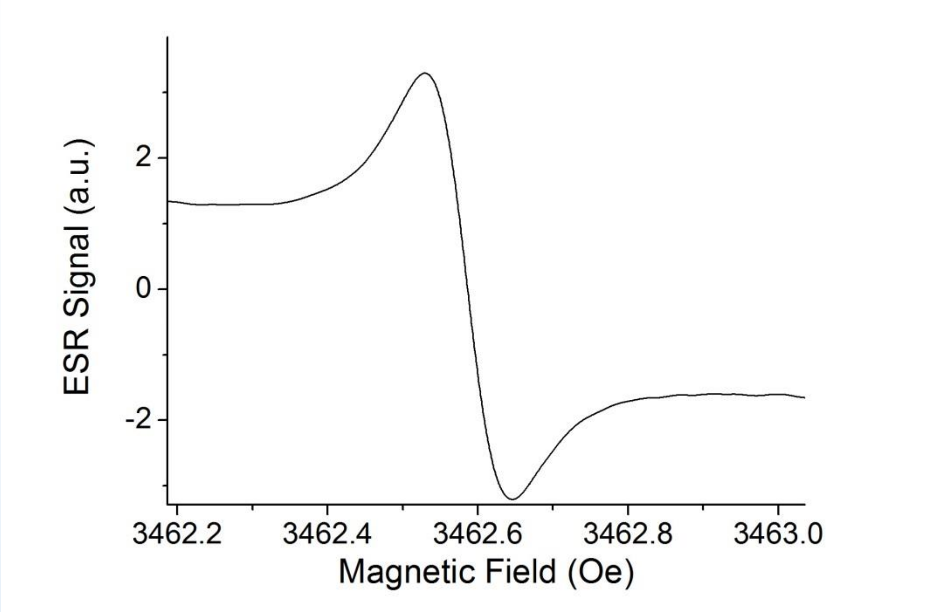
$CuGeO_3$



$\Delta H = 100$ G
$\tau = 0.5$ns

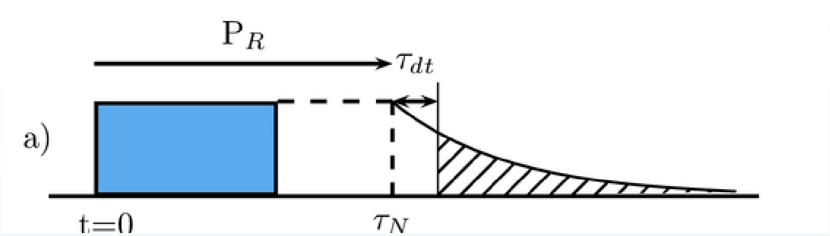
A. I. Smirnov *et al.*, J. Exp. Theor. Phys. **87**, 1019 (1998).

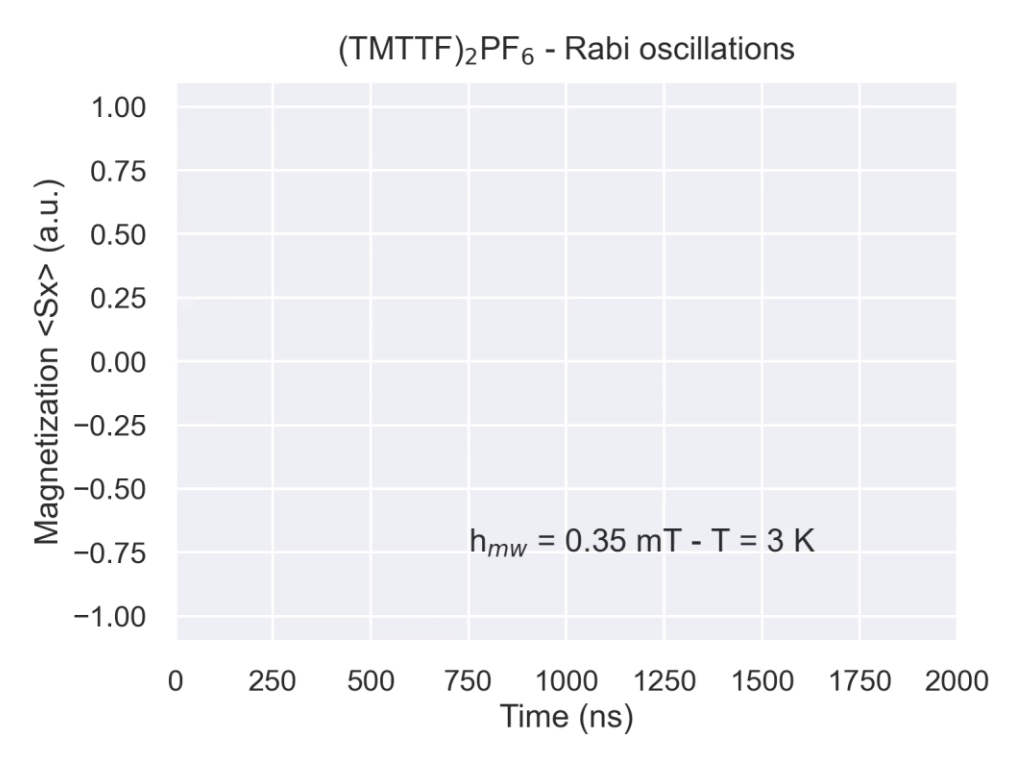
$(TMTTF)_2AsF_6$

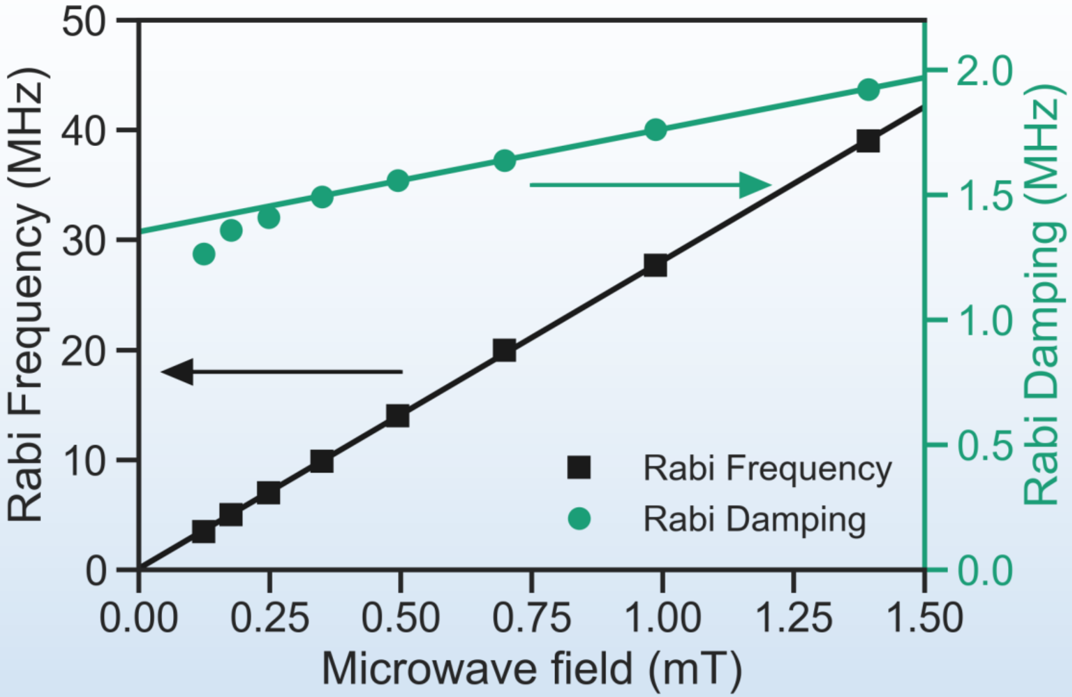


$\Delta H = 0.1$ G
$\tau = 500$ns

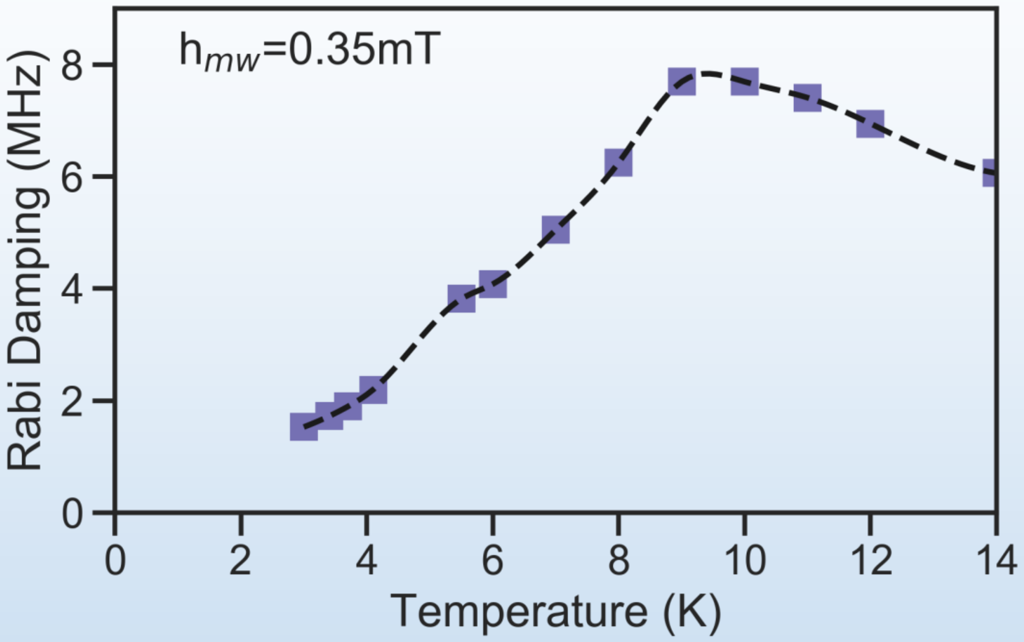
Rabi oscillations of the pinned soliton



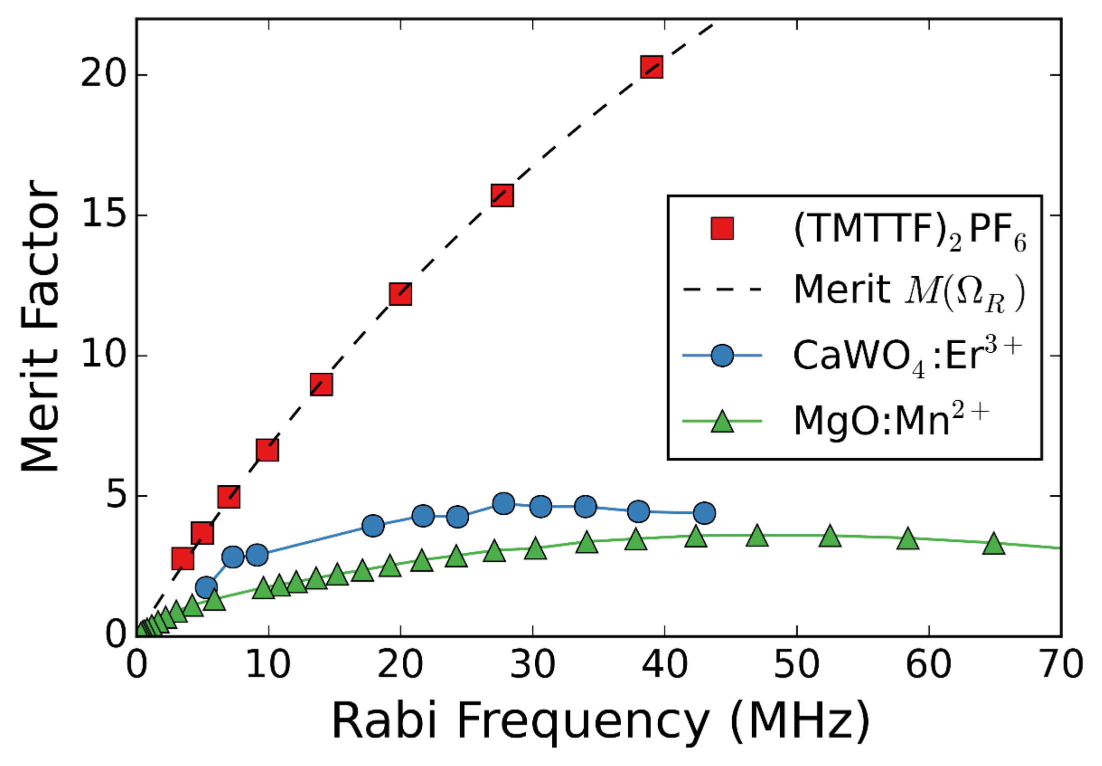




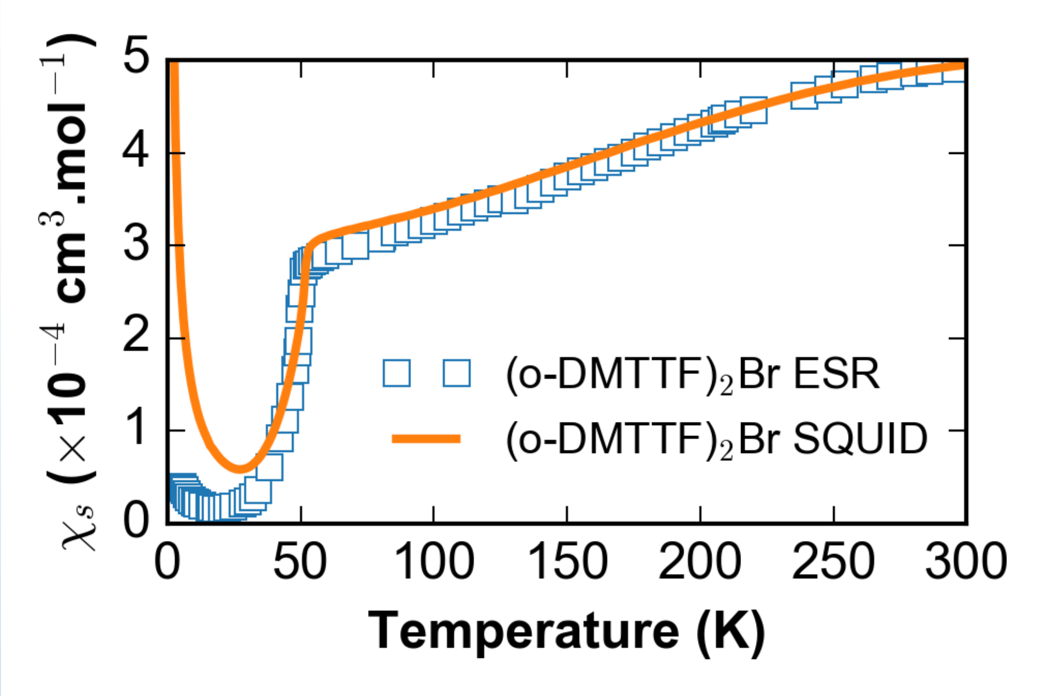
Low damping → long coherence

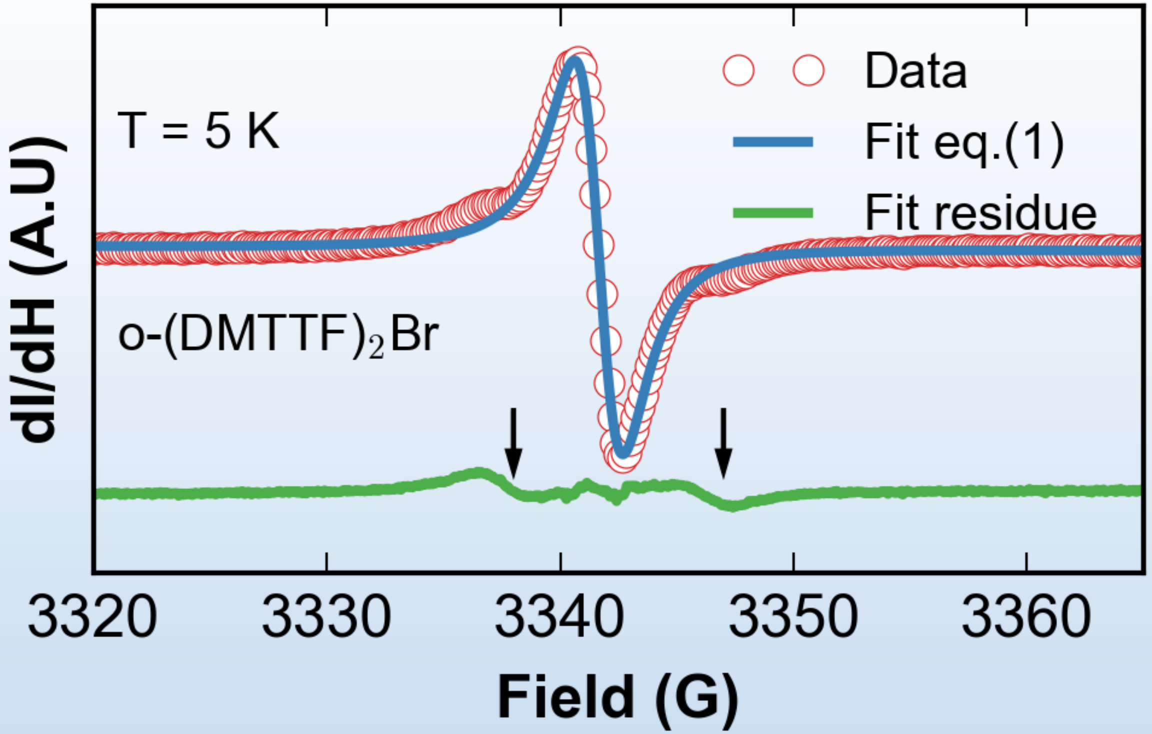


Spin Peierls transition Tsp=19K



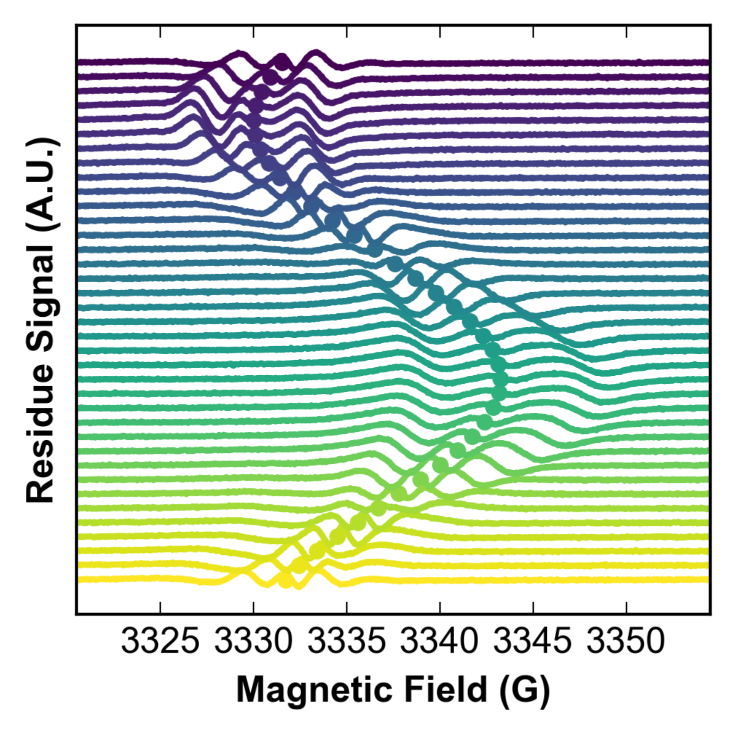
Low Temperature Behavior

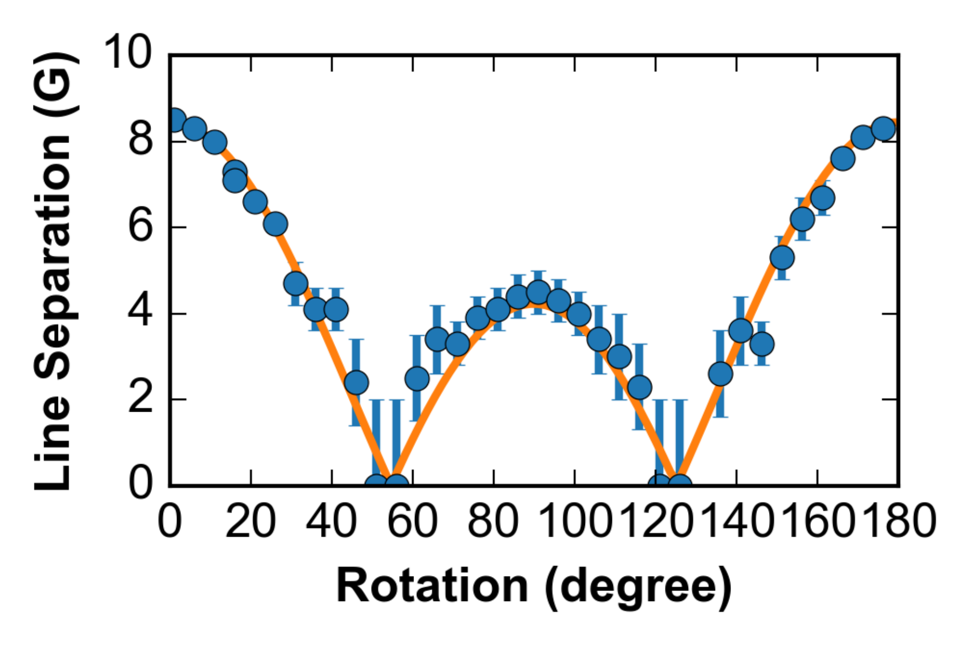




X =	Cl	Br	I	NO_2	NO_3
SQUID	25 10^{-4}	37 10^{-4}	14 10^{-4}	37 10^{-4}	66 10^{-4}
EPR	5.6 10^{-4}	6.6 10^{-4}	4 10^{-4}	7.9 10^{-4}	0.12 10^{-4}

Low Temperature Behavior

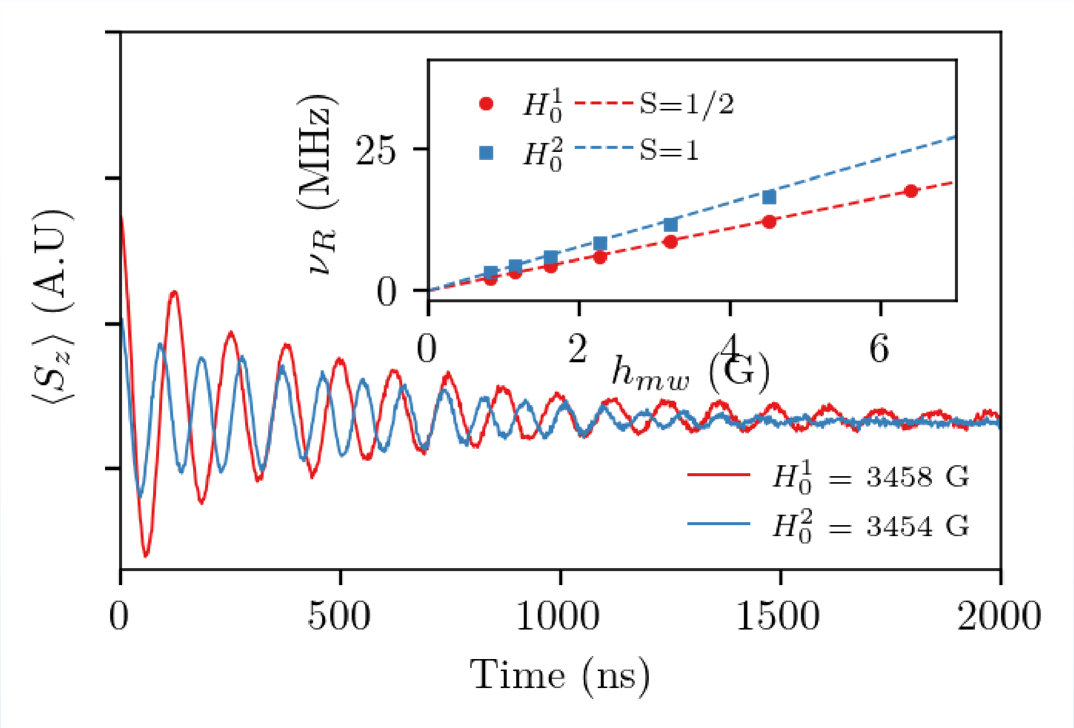


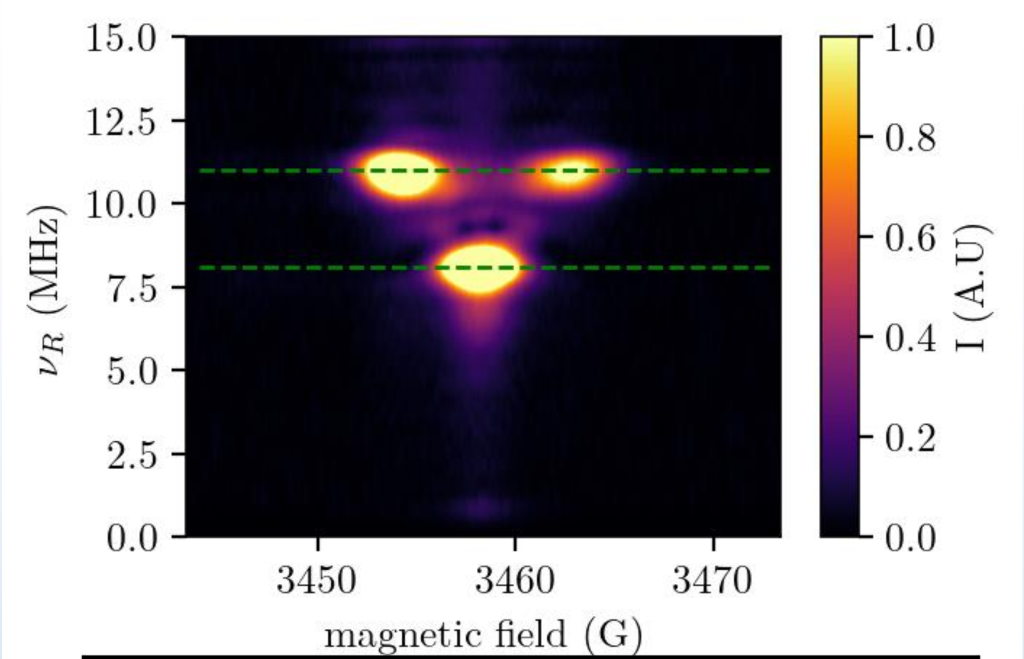


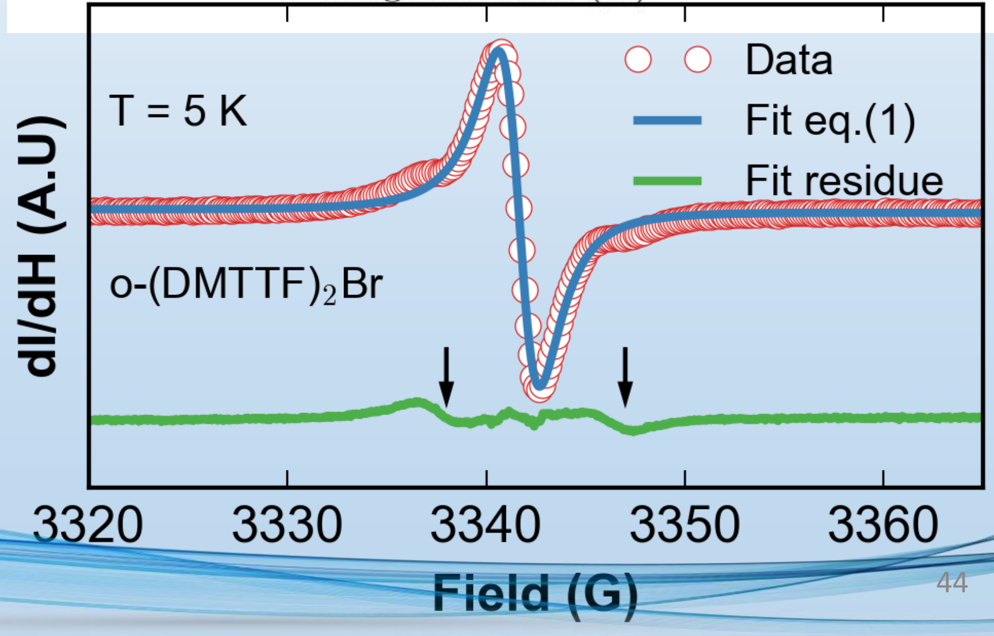
$$\mathbf{H}\ (\theta\ ) = H_{main}(\theta) + \mathbf{d}(3cos^2\theta - 1)$$

Satellites lines comes from the chains

Rabi oscillations



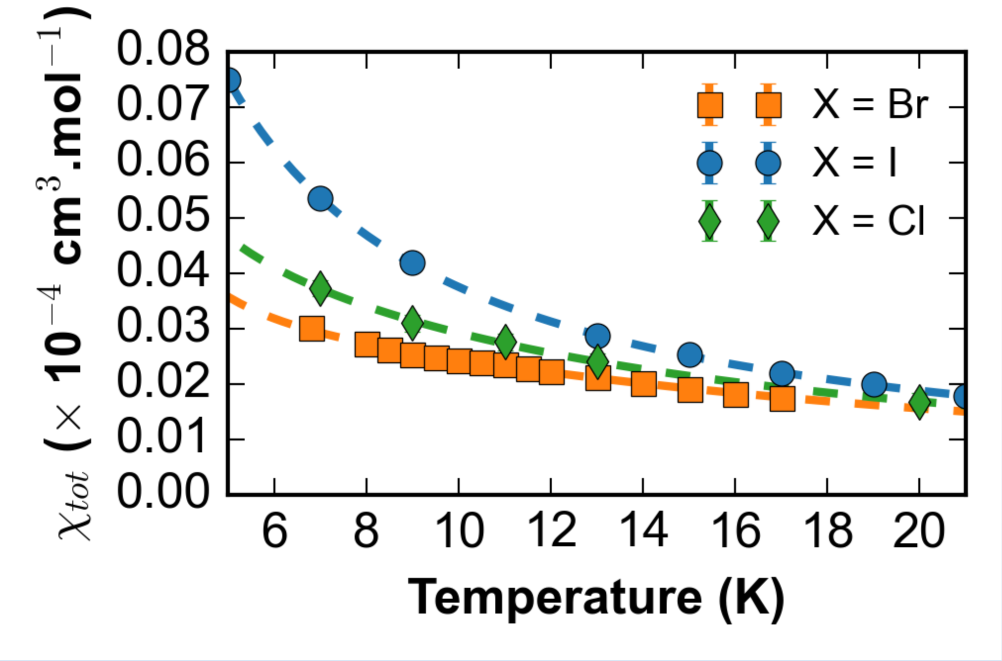


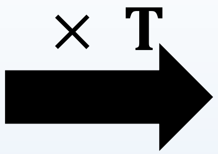


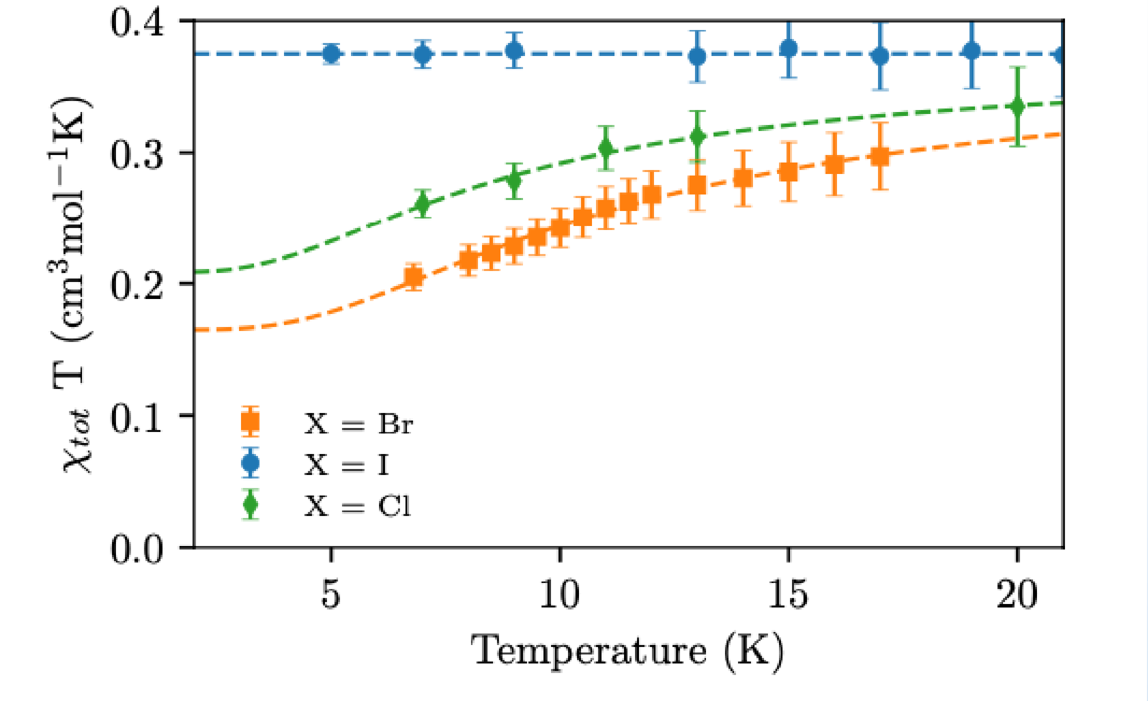
$$\nu_R{}^1 = \sqrt{S(S+1) - m_s(m_s+1)}\,\nu_R{}^{1/2}$$

Central line : S = ½ dynamics
Satellite lines : S = 1 dynamics

Non Curie of the SCD







$$\chi(T) = \frac{3}{8}\frac{(1-n)}{T} + n\frac{1}{2T(1+\frac{1}{3}\exp\left(\frac{\Delta_s}{T}\right))}$$

Cl : $\Delta_s = 16\ K$, n= 0.44

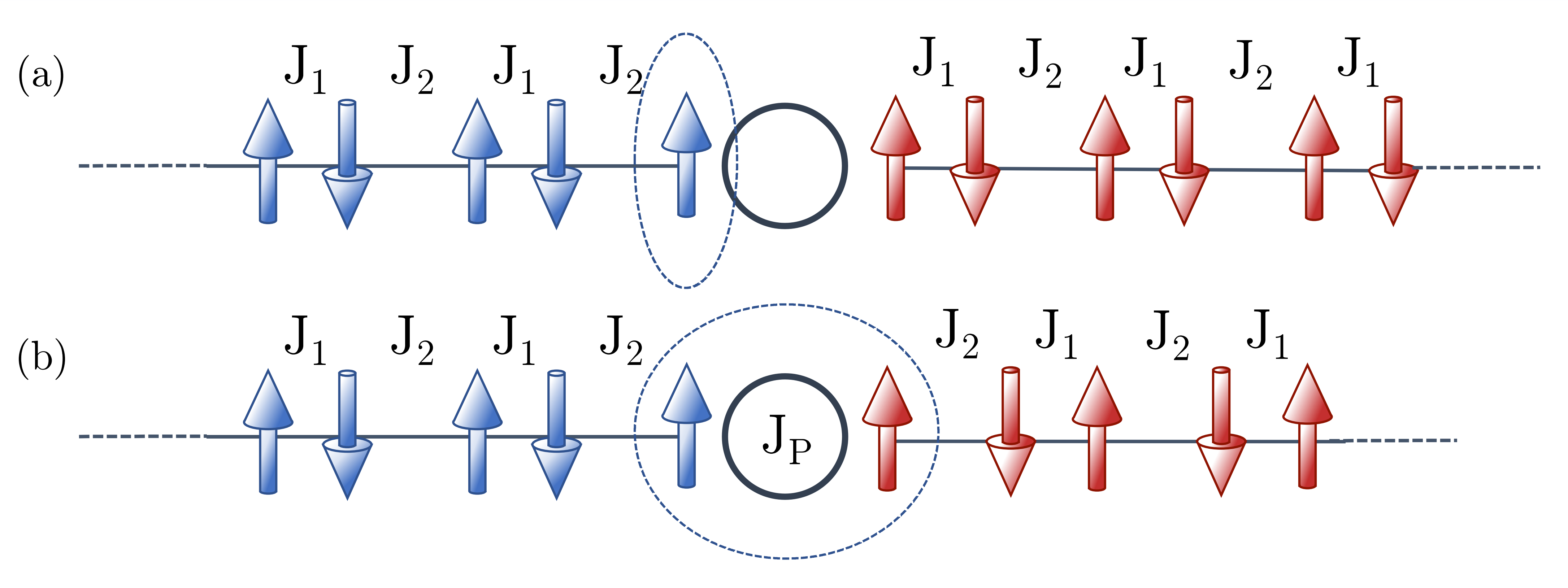
Br : $\Delta_s = 21\ K$, n= 0.56

Pb: Concentration of defects ~10^{-4} → pairs/singles ~ 10^{-4} in a 3D crystal

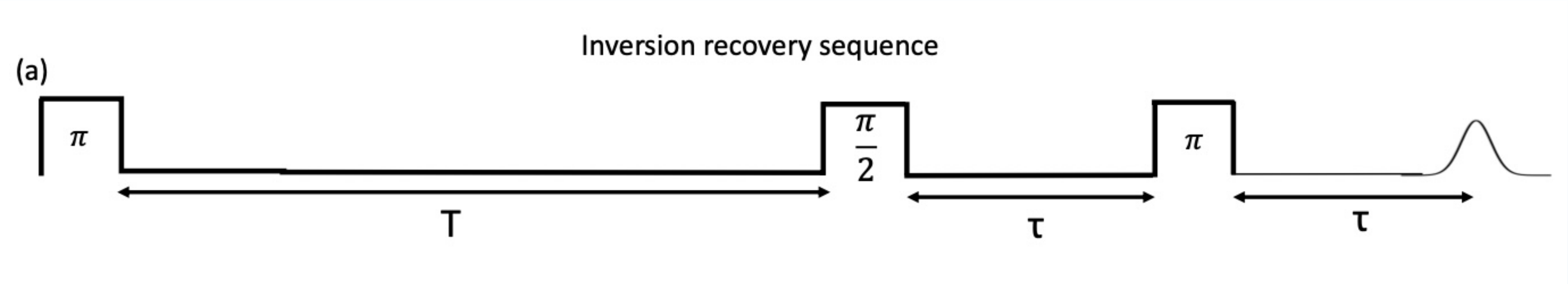
B. Bleaney and K. D. Bowers, Proc. R. Soc. Lond. Ser. 964 Math. Phys. Sci. 214, 451 (1952)

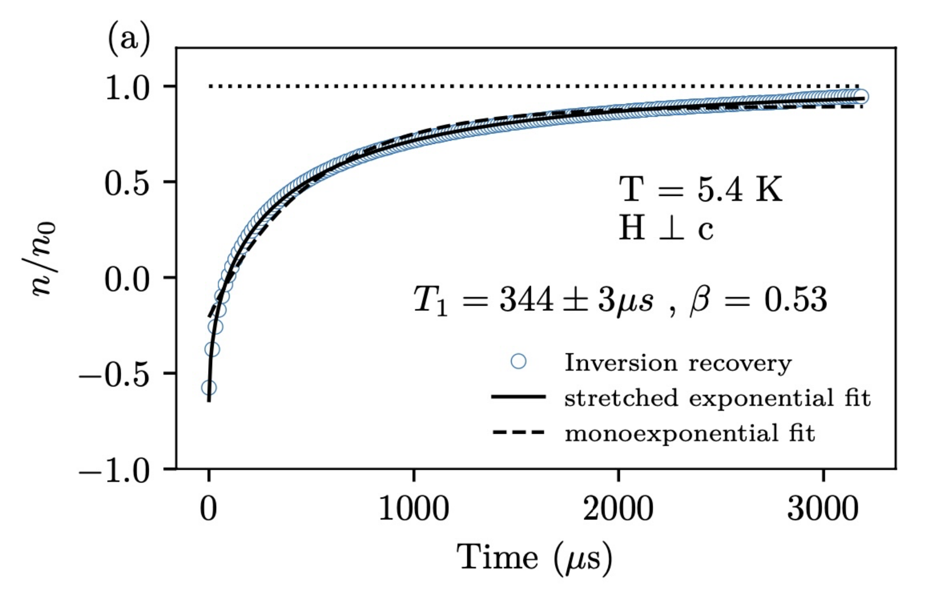
L.Soriano & al., Phys. Rev. B 105, 064434 (2022)

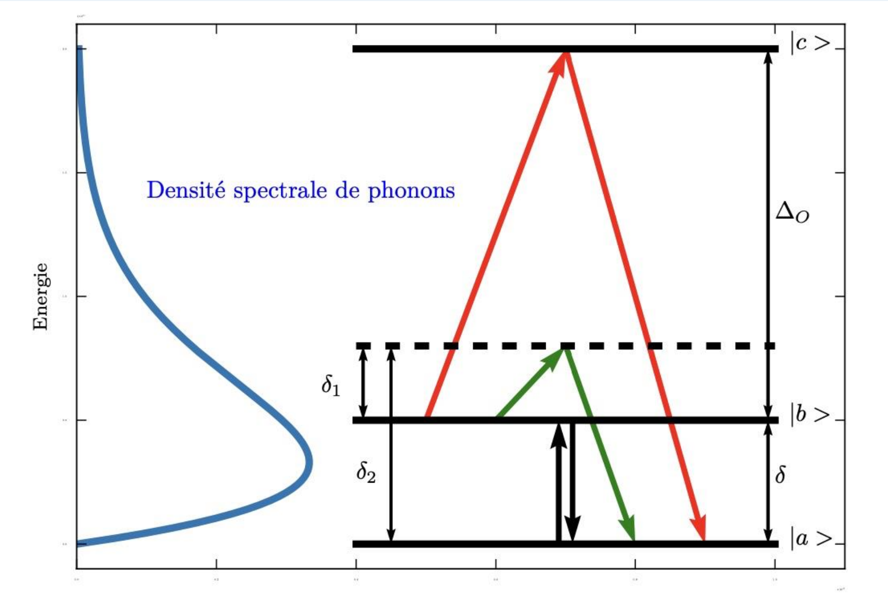
Pairs of solitons



Spin Relaxation



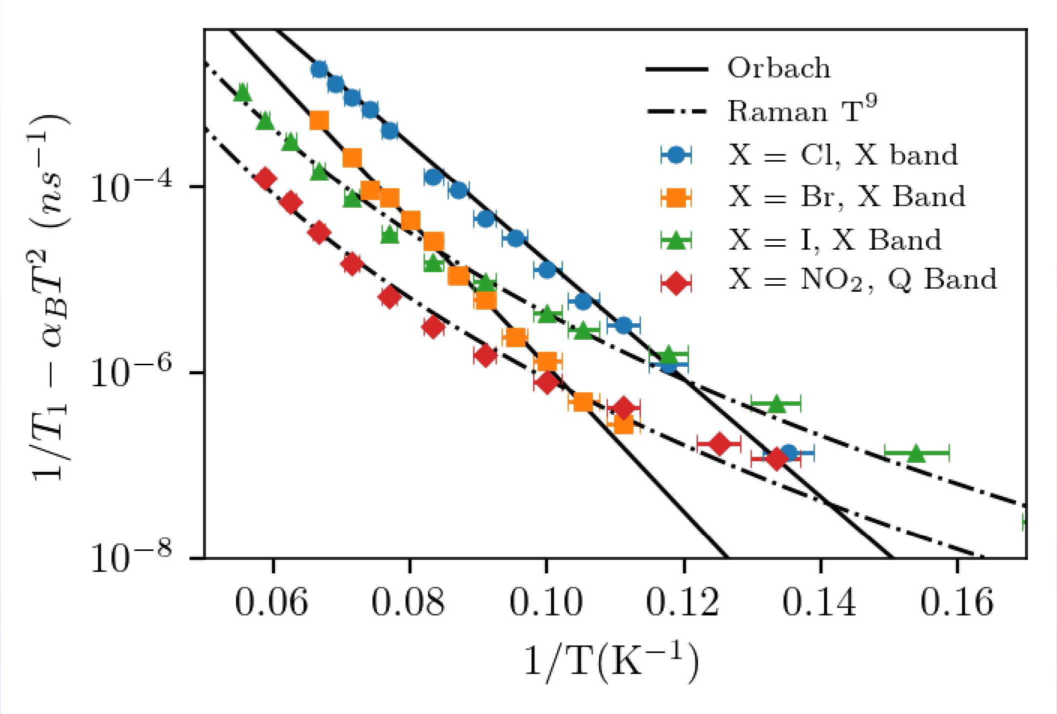


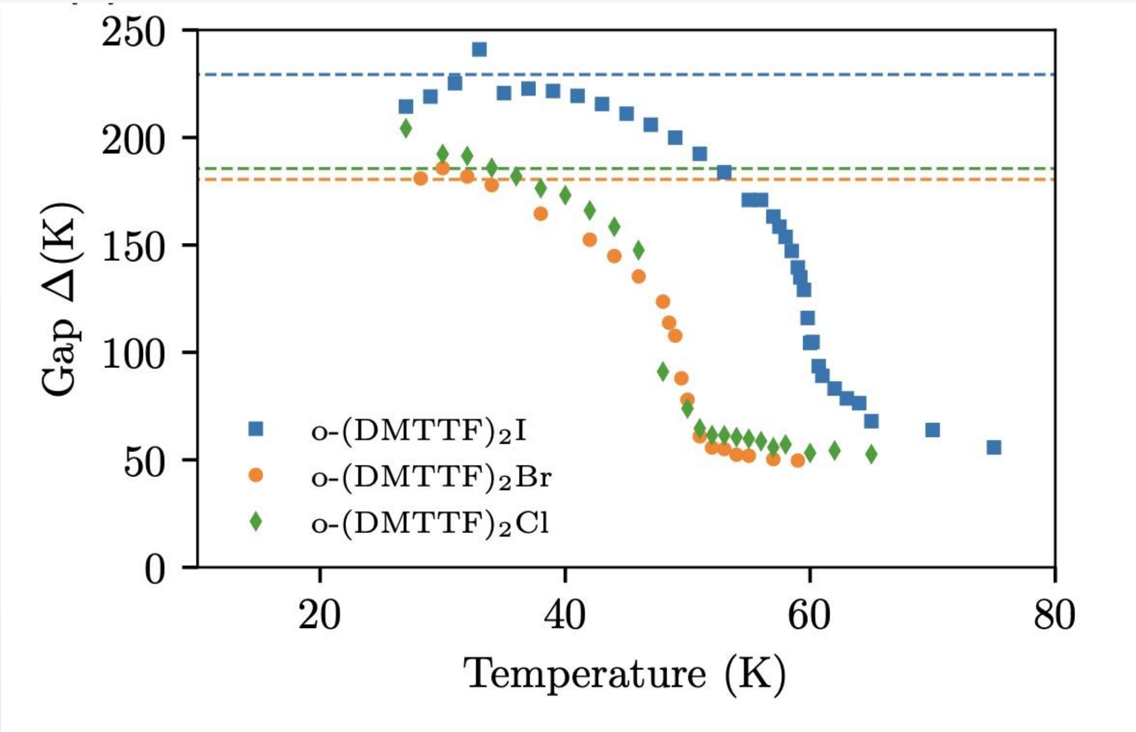


Spin Relaxation

$$\frac{1}{T_1} = \alpha_O \frac{\Delta_0{}^3}{e^{\Delta_0/k_B T} - 1}$$

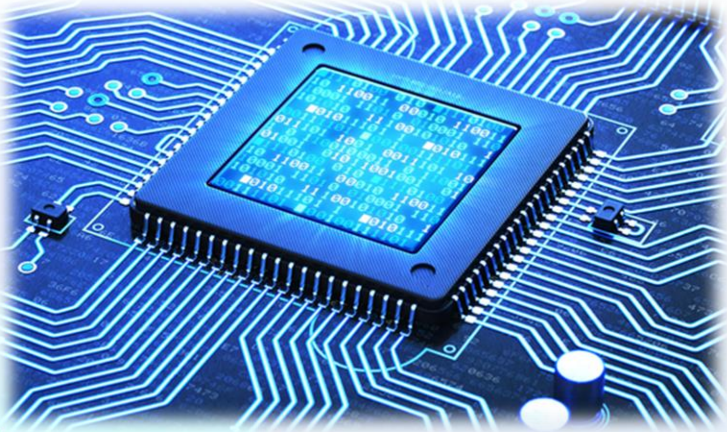
$$\log\left(\frac{1}{T_1}\right) = \log(\alpha_O \Delta_O{}^3) - \frac{\Delta_O}{k_B T}$$

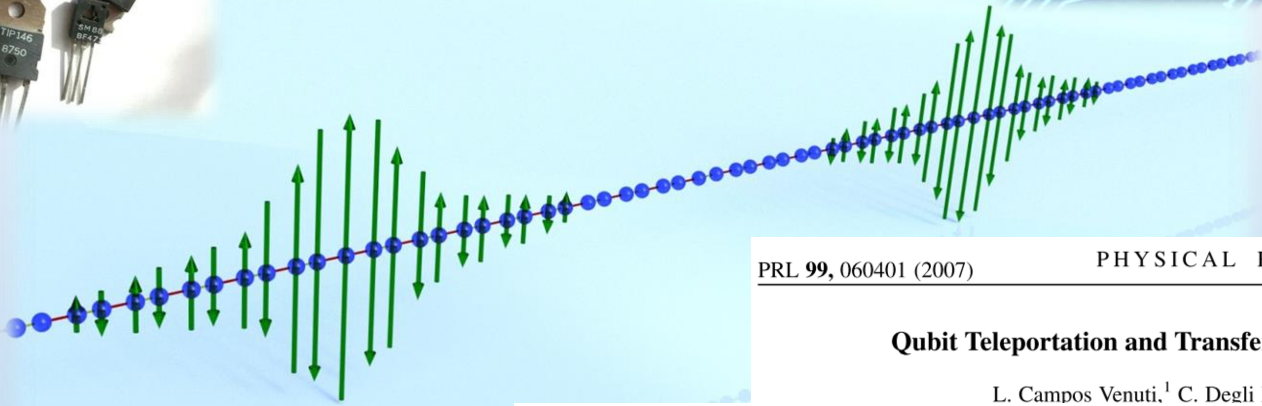




Cl : $\Delta_O = 150\ K$
Br : $\Delta_O = 180\ K$

Quantum communication ?





PRL **99,** 060401 (2007) PHYSICAL REVIEW LETTERS week ending 10 AUGUST 2007

Qubit Teleportation and Transfer across Antiferromagnetic Spin Chains

L. Campos Venuti,[1] C. Degli Esposti Boschi,[2,3] and M. Roncaglia[3,4]

VOLUME 91, NUMBER 20 PHYSICAL REVIEW LETTERS week ending 14 NOVEMBER 2003

Quantum Communication through an Unmodulated Spin Chain

Sougato Bose

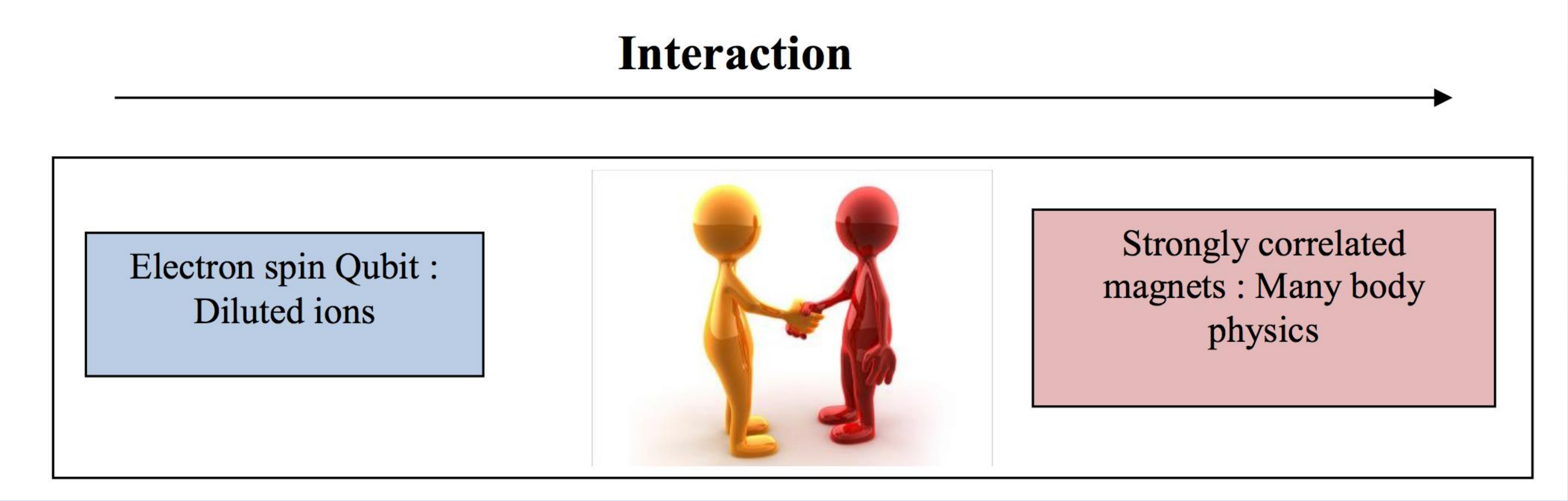
PRL **96,** 247206 (2006) PHYSICAL REVIEW LETTERS week ending 23 JUNE 2006

Long-Distance Entanglement in Spin Systems

L. Campos Venuti,[1,2] C. Degli Esposti Boschi,[1,3] and M. Roncaglia[1,2,3]

Conclusion

- New mechanism of coherence protected by quantum fluctuation
- 2 levels system made by hundreds of spins
- Use the chain as quantum communication wire ?



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