

# Future shocks: opportunities in magnetism with Super-MuSR

Tom Lancaster  
Durham University

# (Mega) Events, dear boy, (mega) events

Tom Lancaster  
Durham University

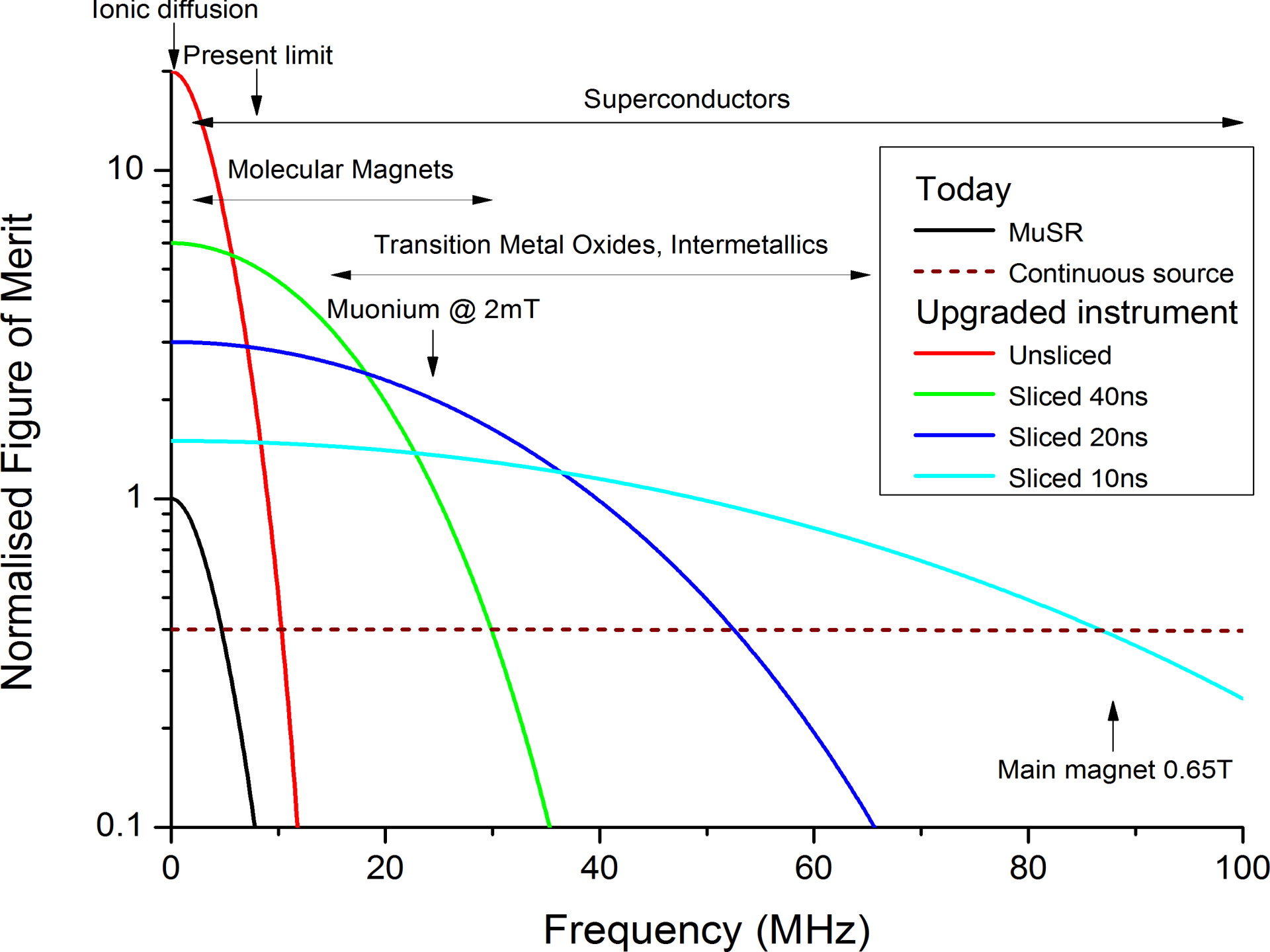
# Opportunities from Super-MuSR

- Data rate and quality
- Enhanced frequency response

# Opportunities from Super-MuSR

- Data rate and quality
- Enhanced frequency response

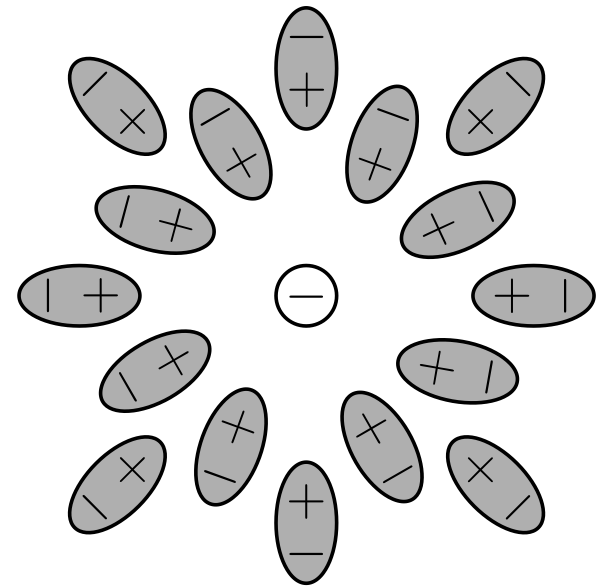
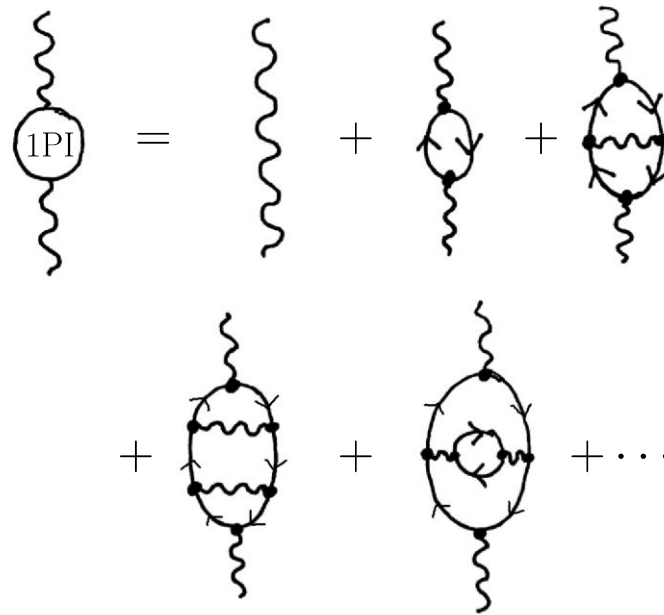
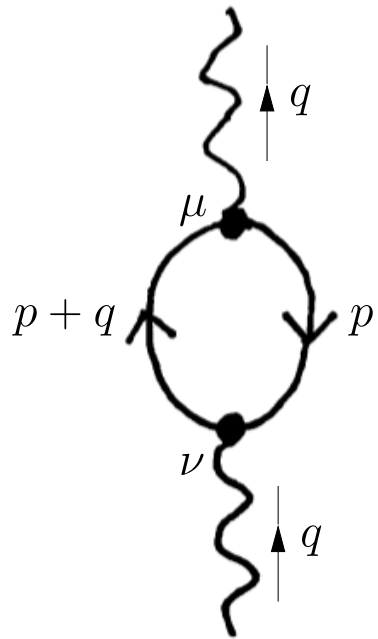
“Super!”



# Low moment systems

(reduced dimensionality, molecular systems, heavy fermion magnets...)

# An explanation of screening



# Low moment systems

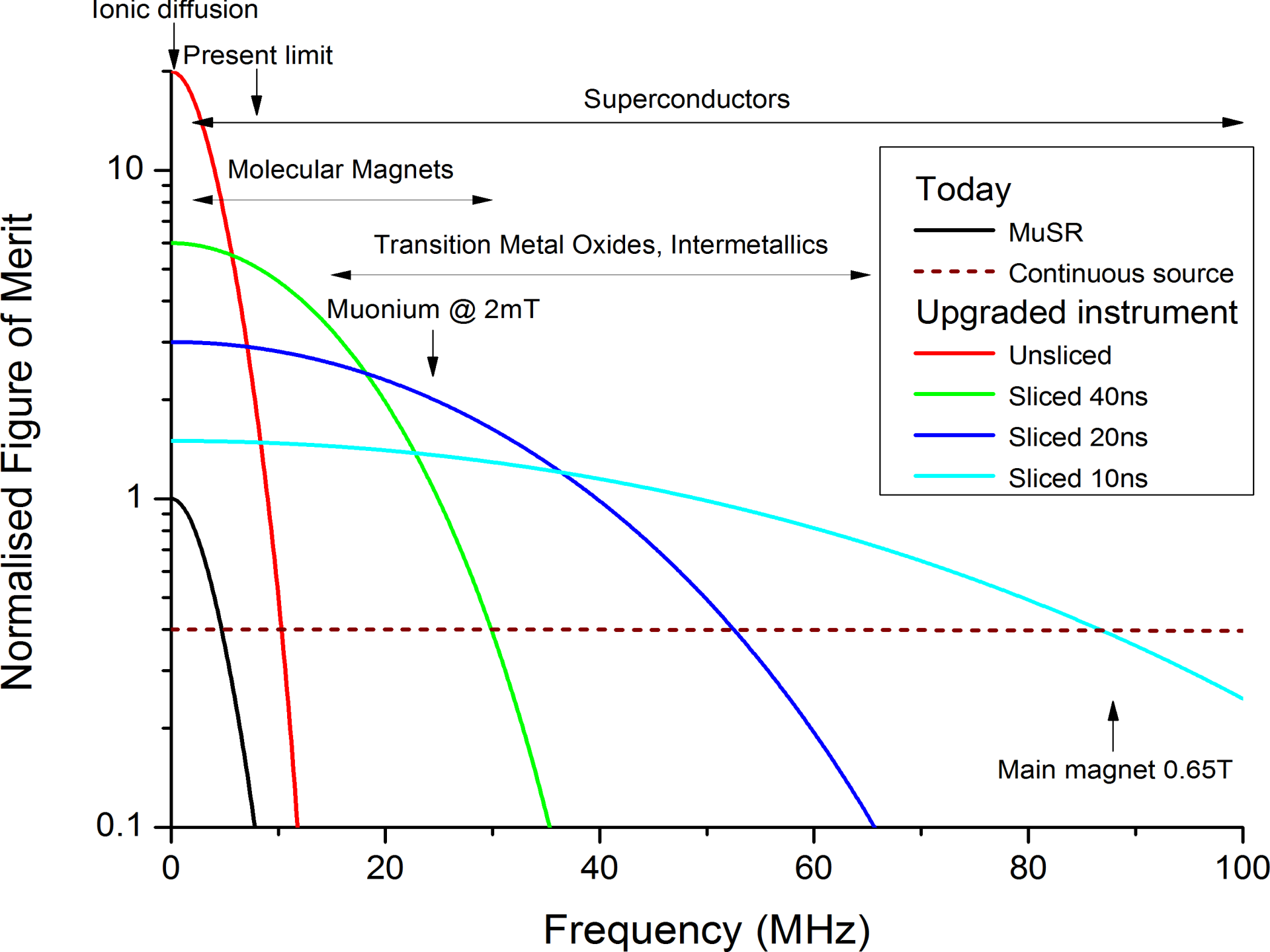
(reduced dimensionality, molecular systems, heavy fermion magnets...)

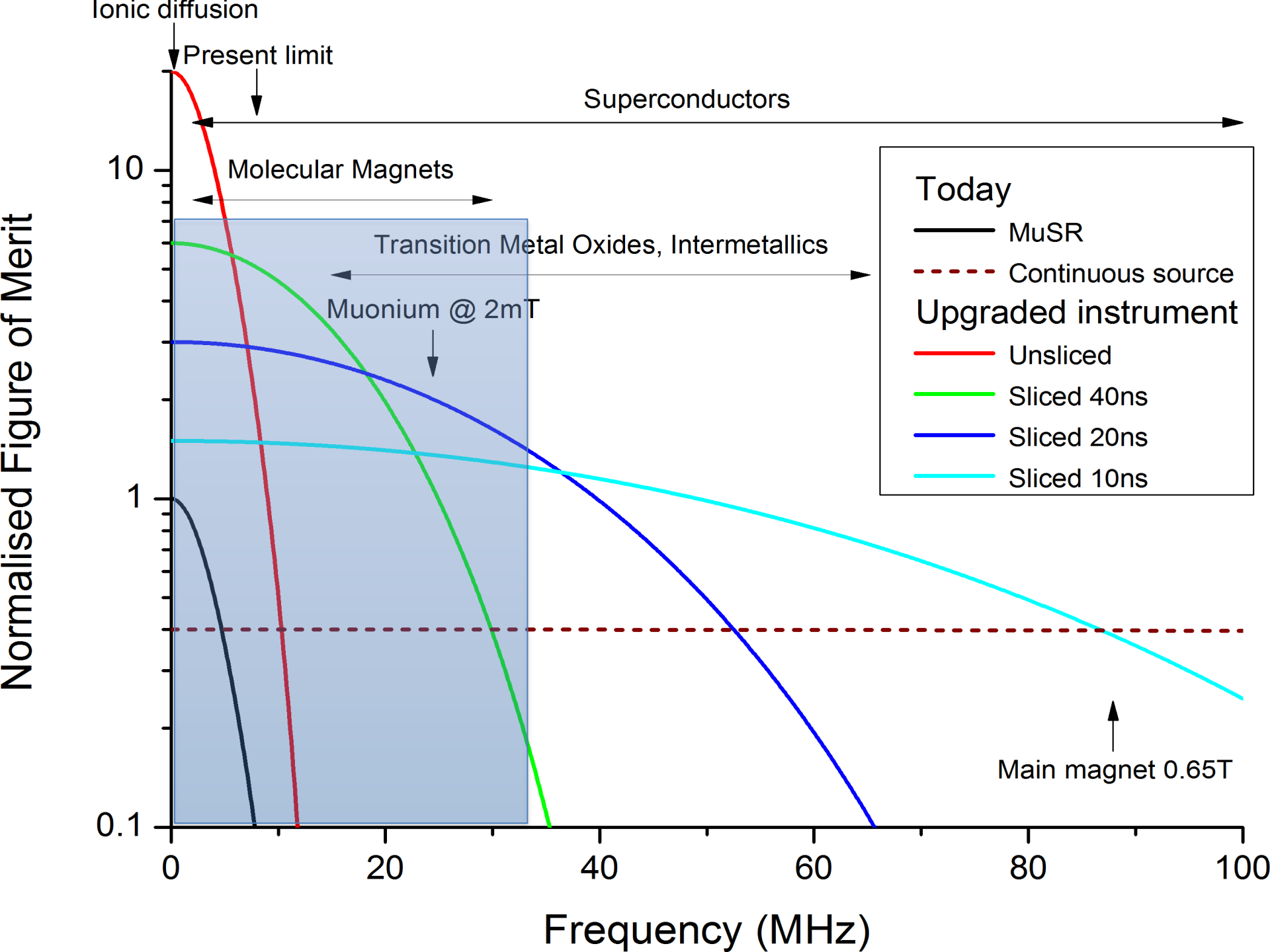
Count rate increased by a factor of 20

More data

Better quality data







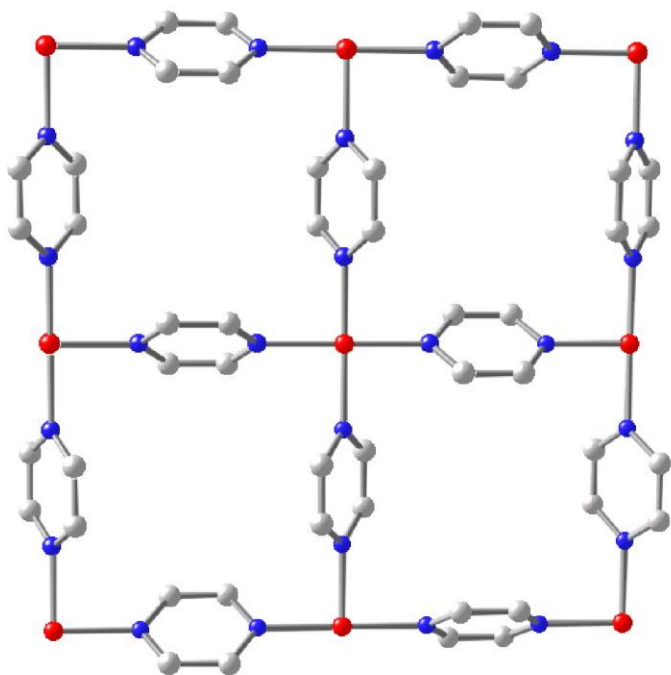
# Case study: molecular magnets



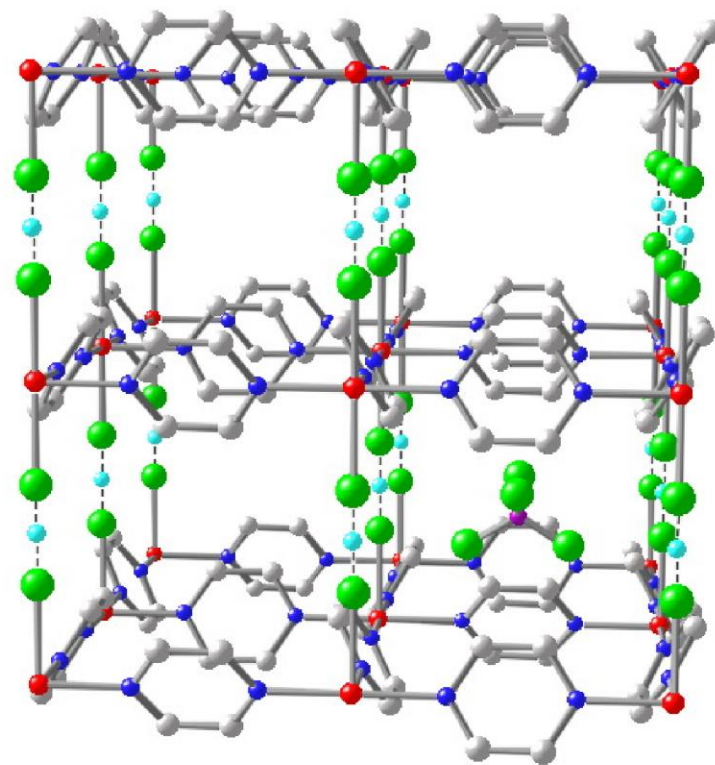
Highly tunable, self-assembled nanostructures with 2D character

First coordination polymer containing the  $\text{HF}_2^-$  ion

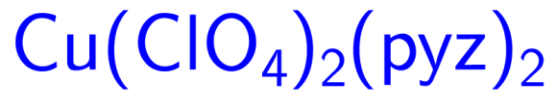
(strongest known hydrogen bond!)



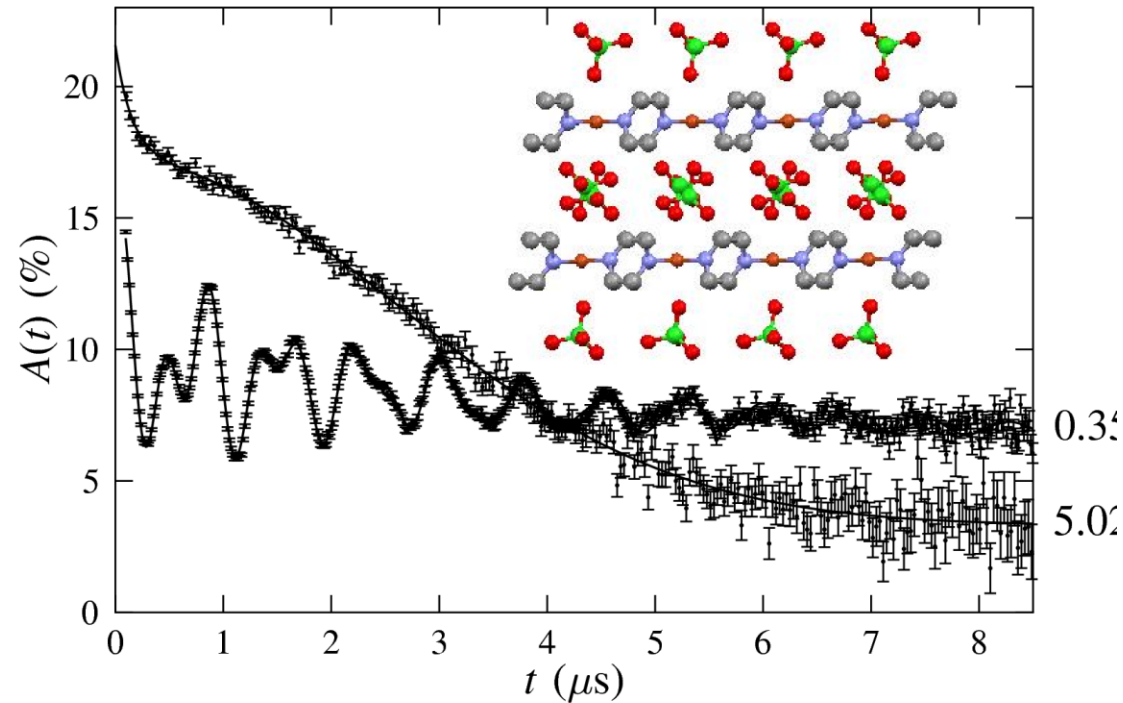
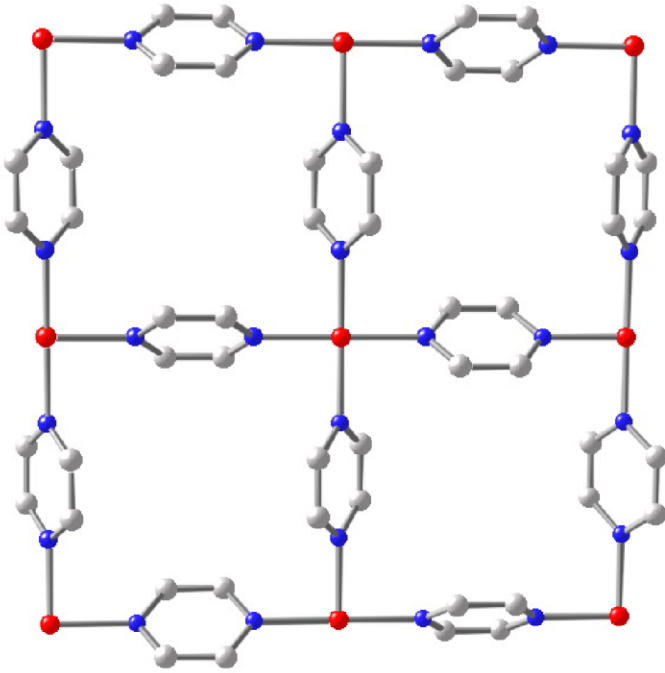
2D square lattice of  $\text{Cu}^{2+}$   $S=1/2$  spins



Linked by  $\text{HF}_2^-$  to form 3D structure  
(with  $X$  anions in the cubes)



$\mu^+$  SR results

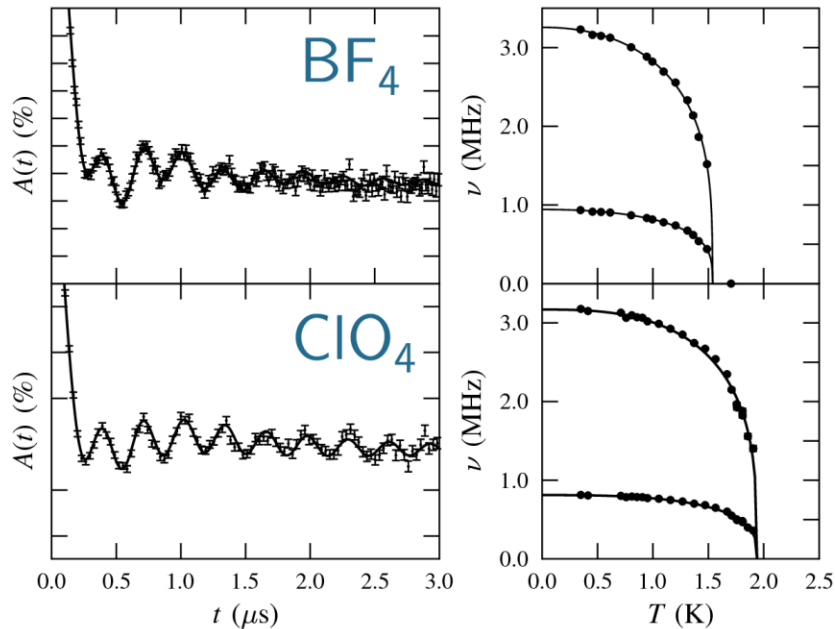
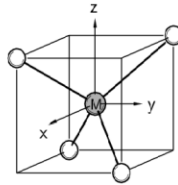


Another 2D square lattice Heisenberg system with  $J=17.8$  K  
Magnetic order detected with oscillations at three frequencies  
 $T_N=4.2(1)$  K

# $[\text{Cu}(\text{pyz})_2\text{HF}_2]\text{X}_2$ : trends across the series

Tetrahedral vs. octahedral anion in the cubes

Tetrahedral anion



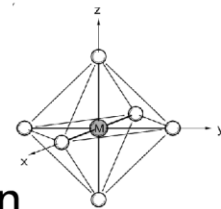
$$T_N \approx 1.5 \text{ K}$$

$$J \approx 7 \text{ K}$$

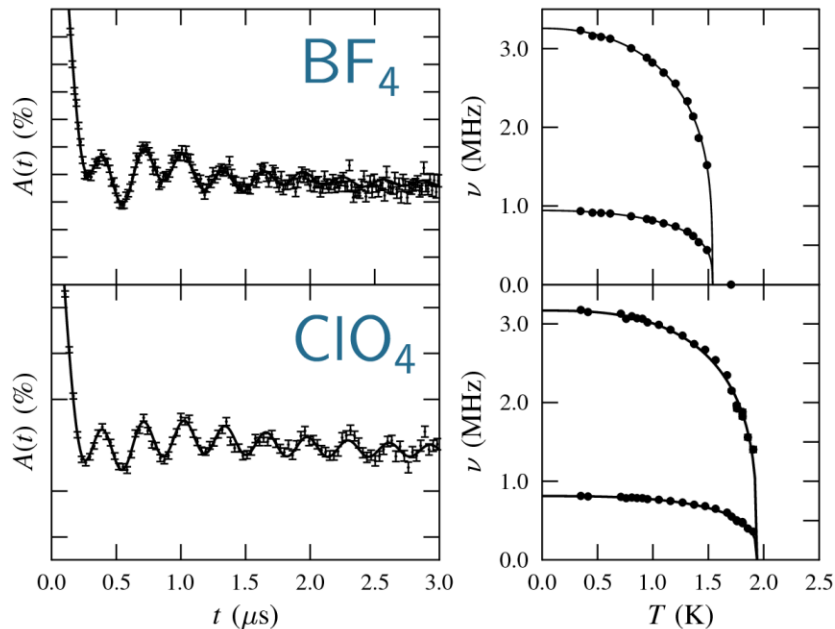
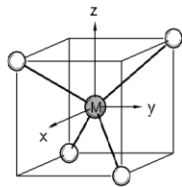
$$|J'/J| \approx 10^{-3}$$

# [Cu(pyz)<sub>2</sub>HF<sub>2</sub>]<sub>2</sub>X<sub>2</sub>: trends across the series

Tetrahedral vs. octahedral anion in the cubes



Tetrahedral anion

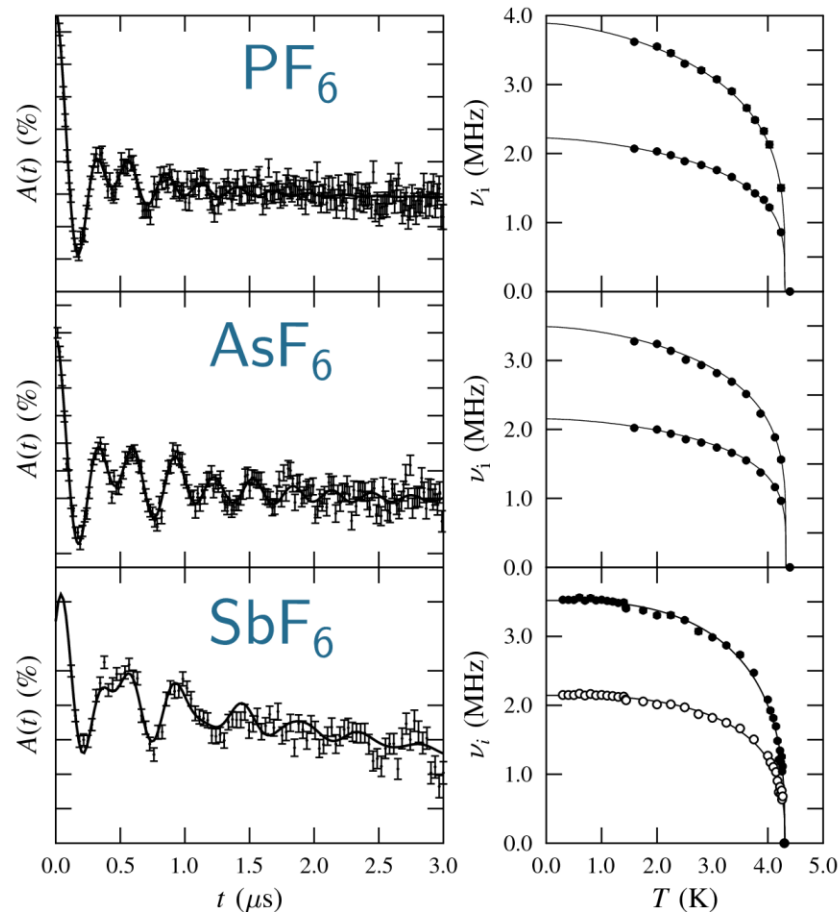


$$T_N \approx 1.5 \text{ K}$$

$$J \approx 7 \text{ K}$$

$$|J'/J| \approx 10^{-3}$$

Octahedral anion



$$T_N \approx 4.2 \text{ K}$$

$$J \approx 13 \text{ K}$$

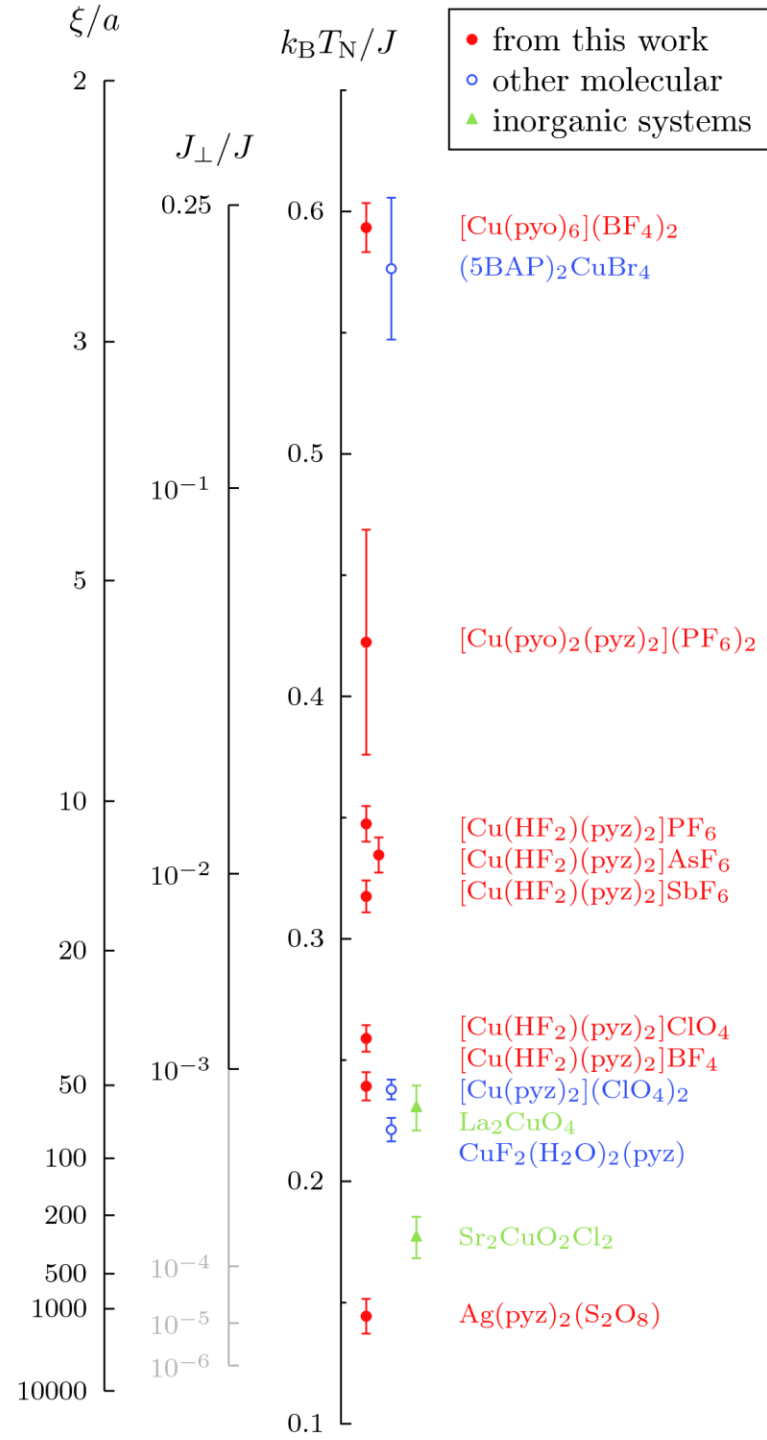
$$|J'/J| \approx 10^{-2}$$

Goddard *et al.* New J. Phys. **10** 083025 (2008)

# 2D molecular magnets

Evaluation of the separation of several systems

Steele et al., PRB (2011)





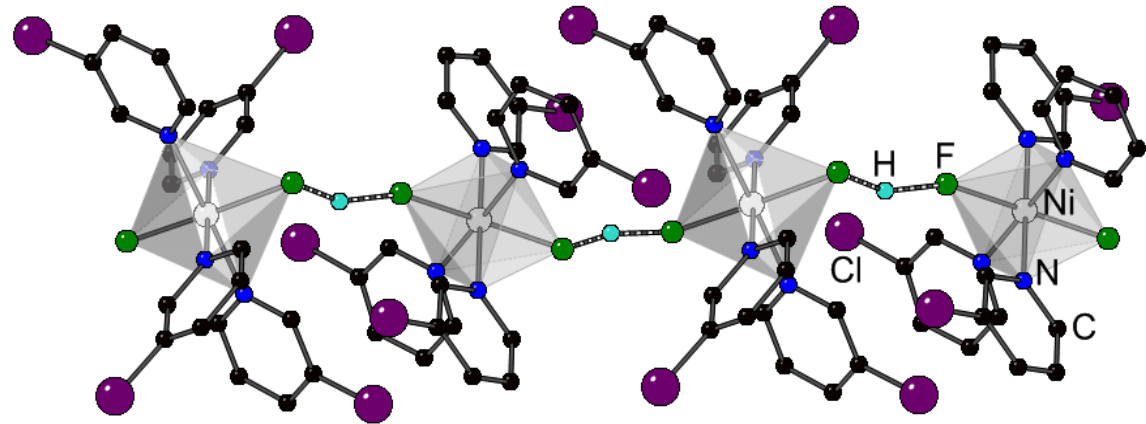
# With Super-MuSR

- High statistics results
- More throughput

# With Super-MuSR

- High statistics results
- More throughput
- Extension to other systems...

# $[\text{Ni}(\text{HF}_2)(3\text{-Clpy})_4]\text{BF}_4$ : the hunt for Haldane chains

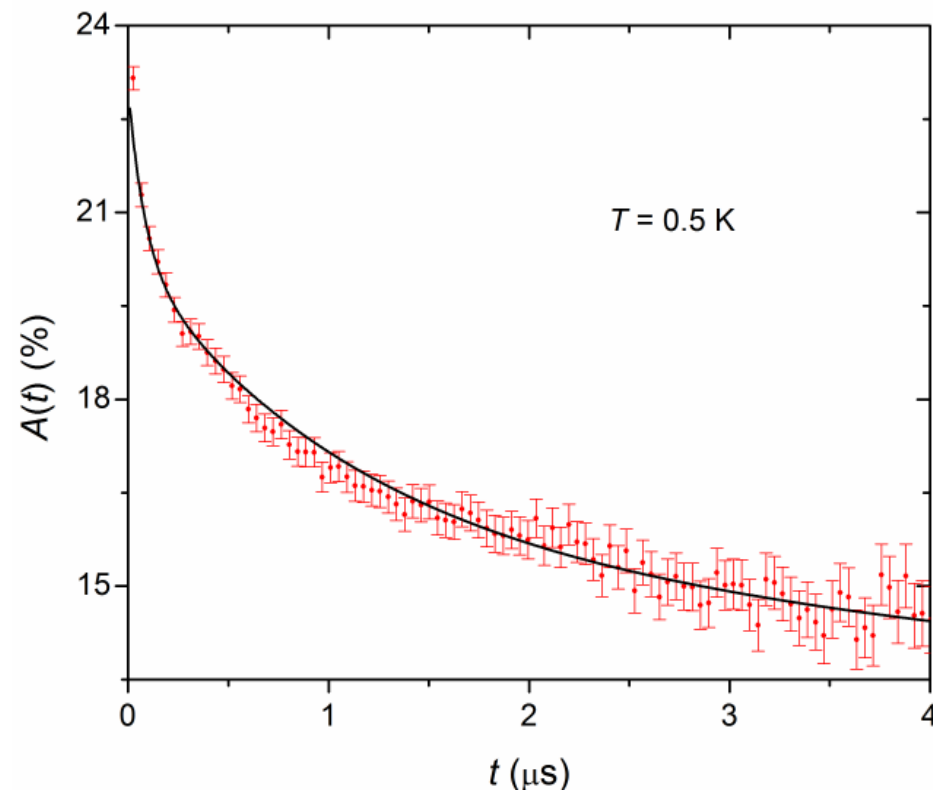


Well separated  $S=1$  chains

$$J=4.9 \text{ K}, D \approx 4 \text{ K}$$

Good evidence for gapless,  
disordered ground state

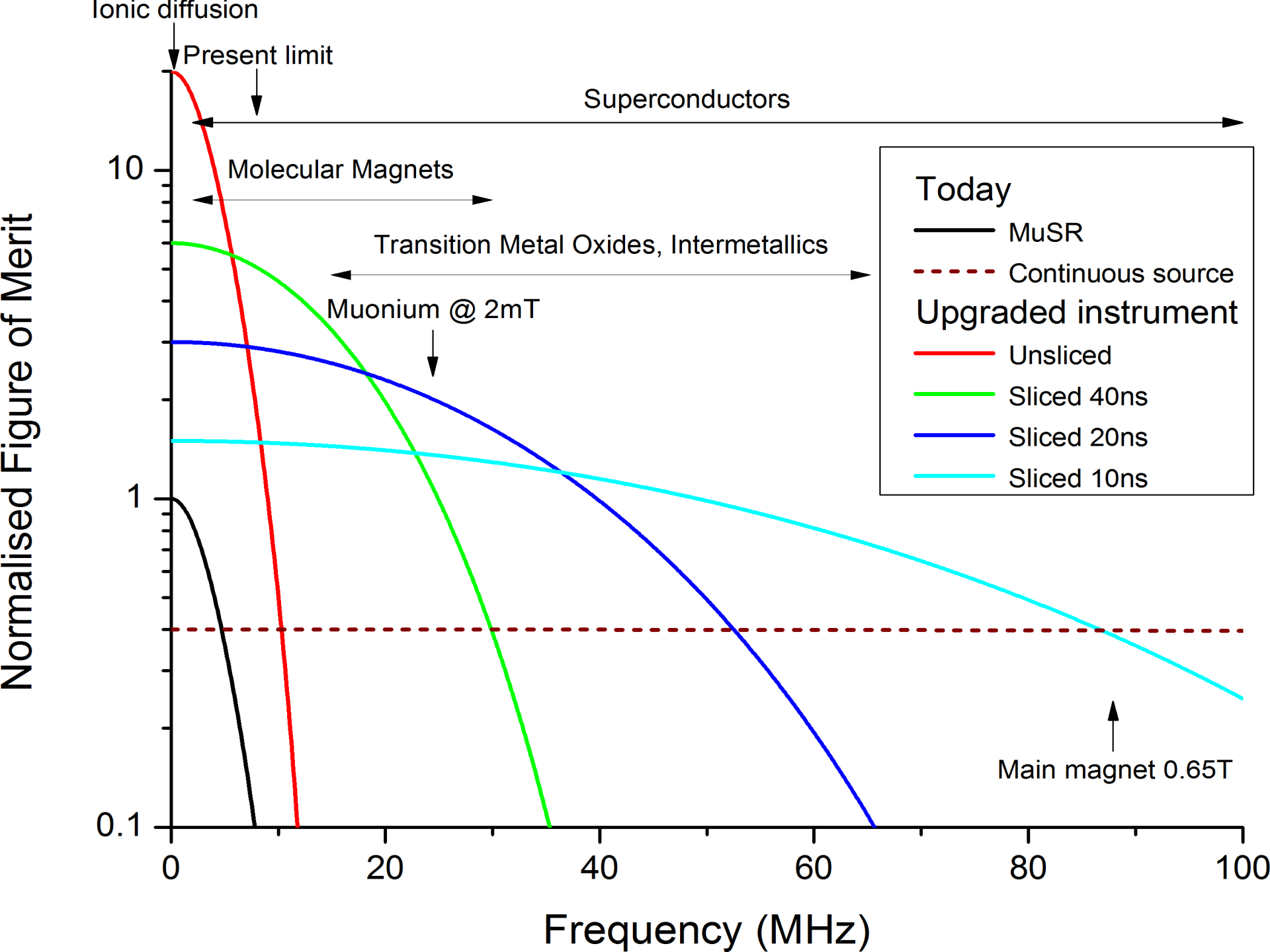
Manson et al., (2012)

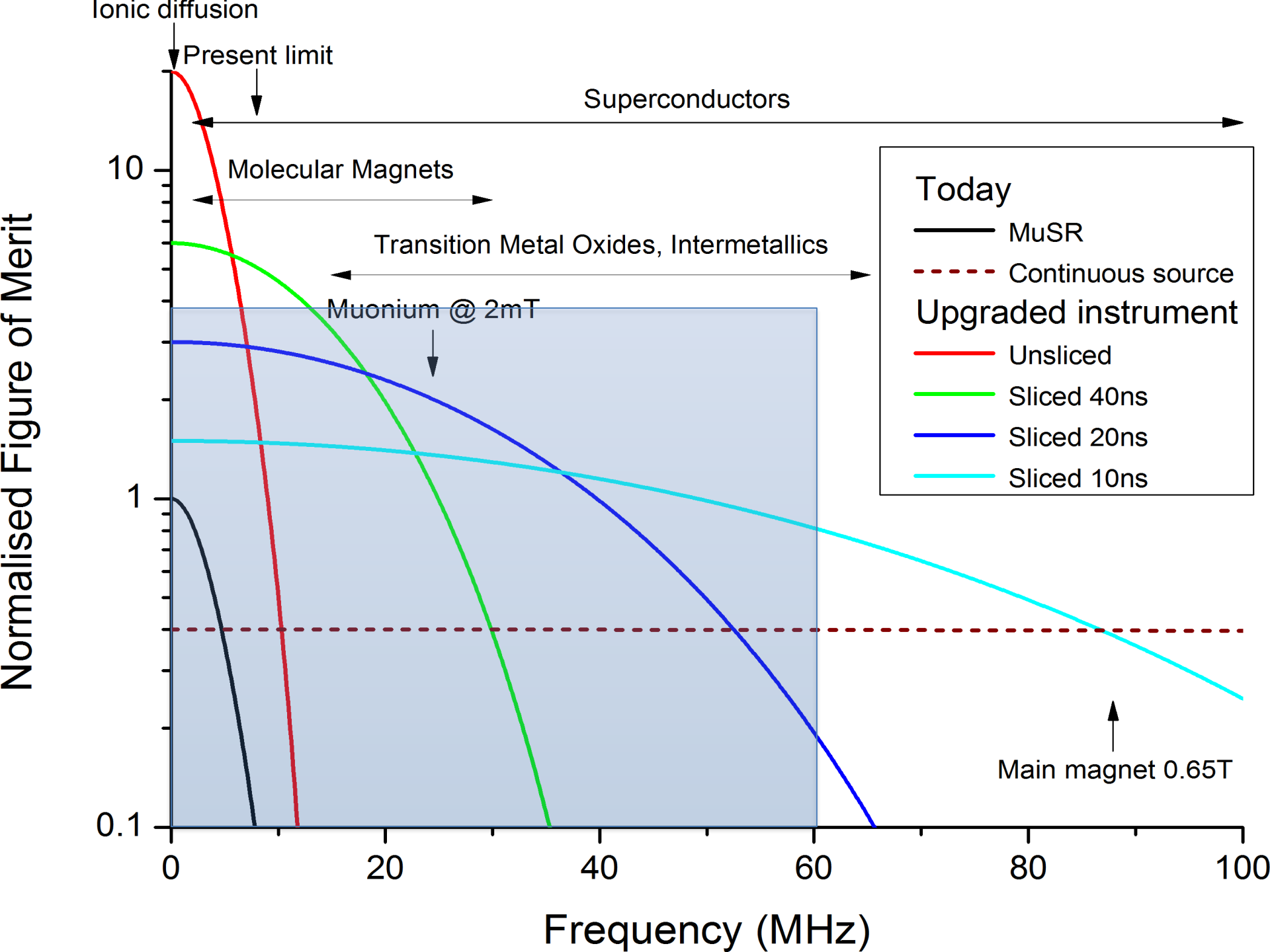


$[\text{Ni}(\text{HF}_2)(3\text{-Clpy})_4]\text{BF}_4$ :  
the hunt for  
Haldane chains

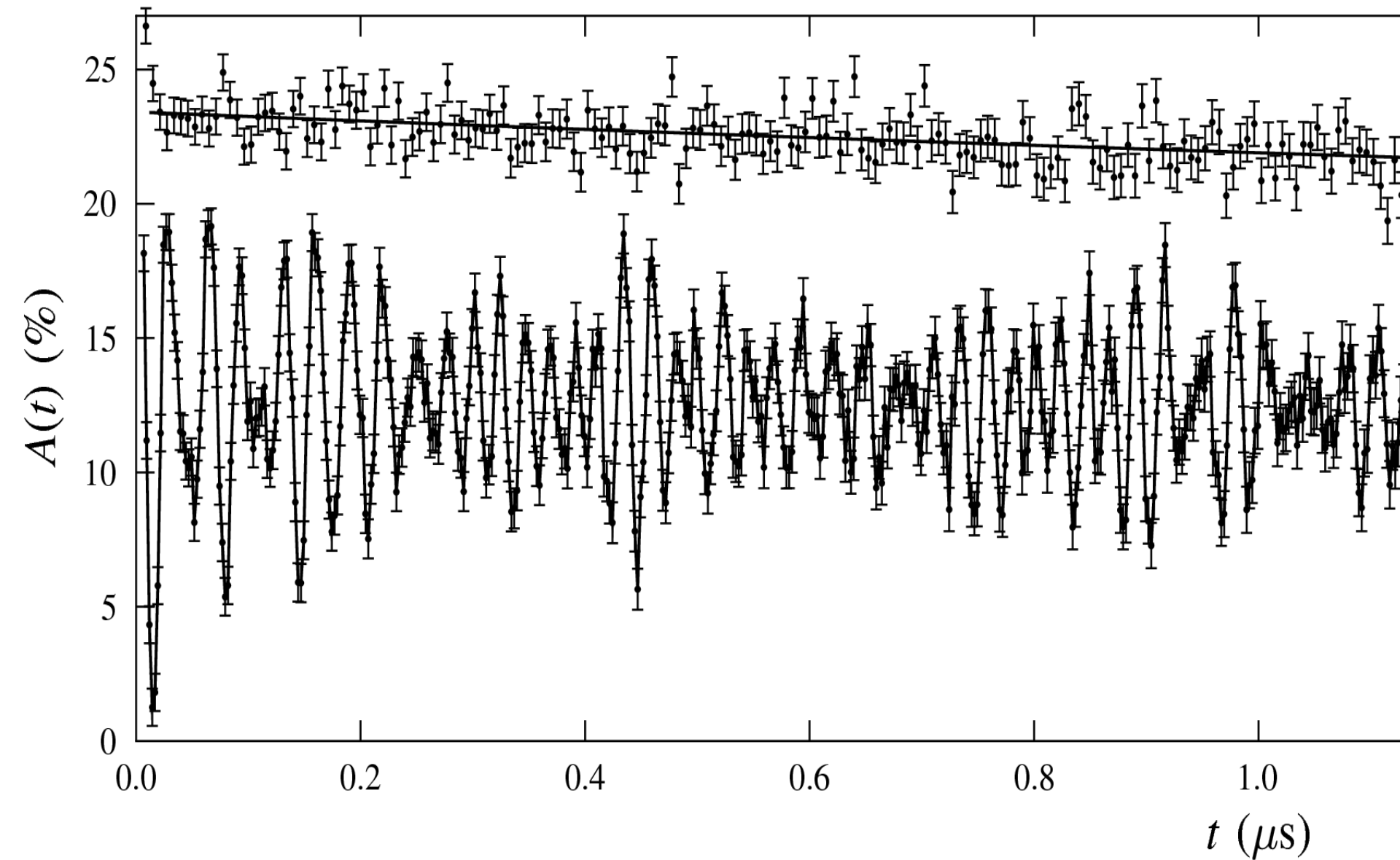
Bring these  $S=1$  systems into the  
ISIS time window

Larger moment systems

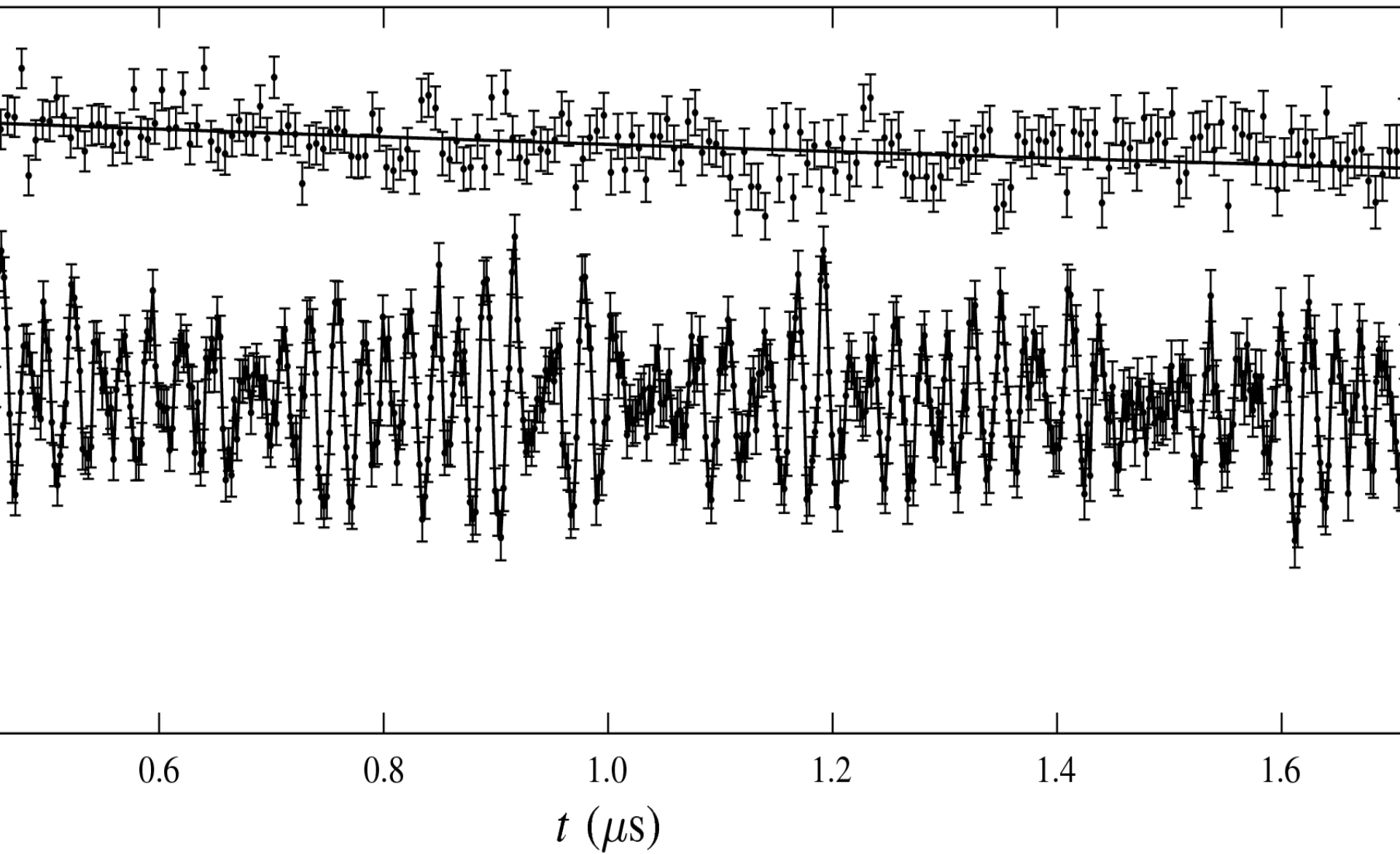


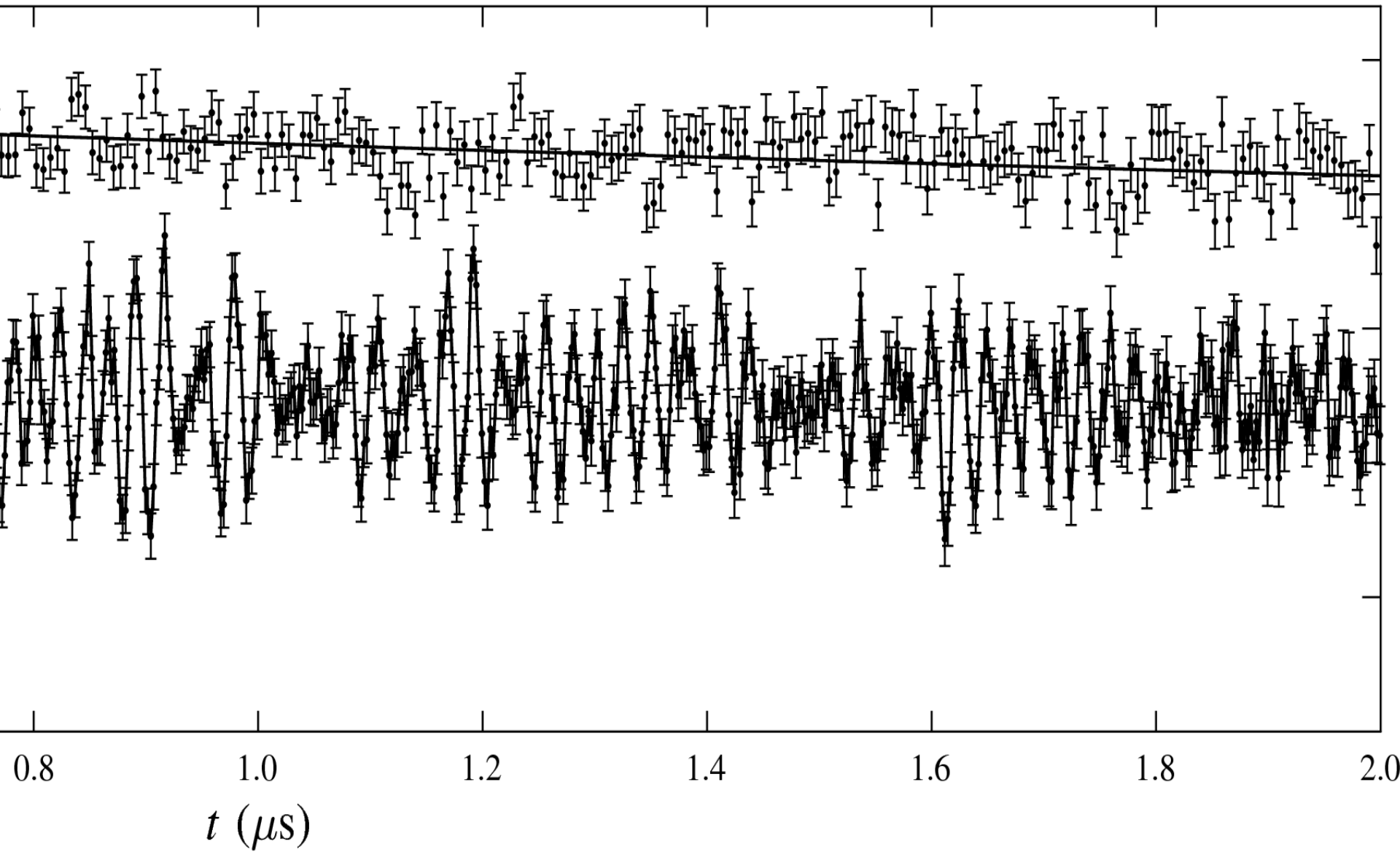


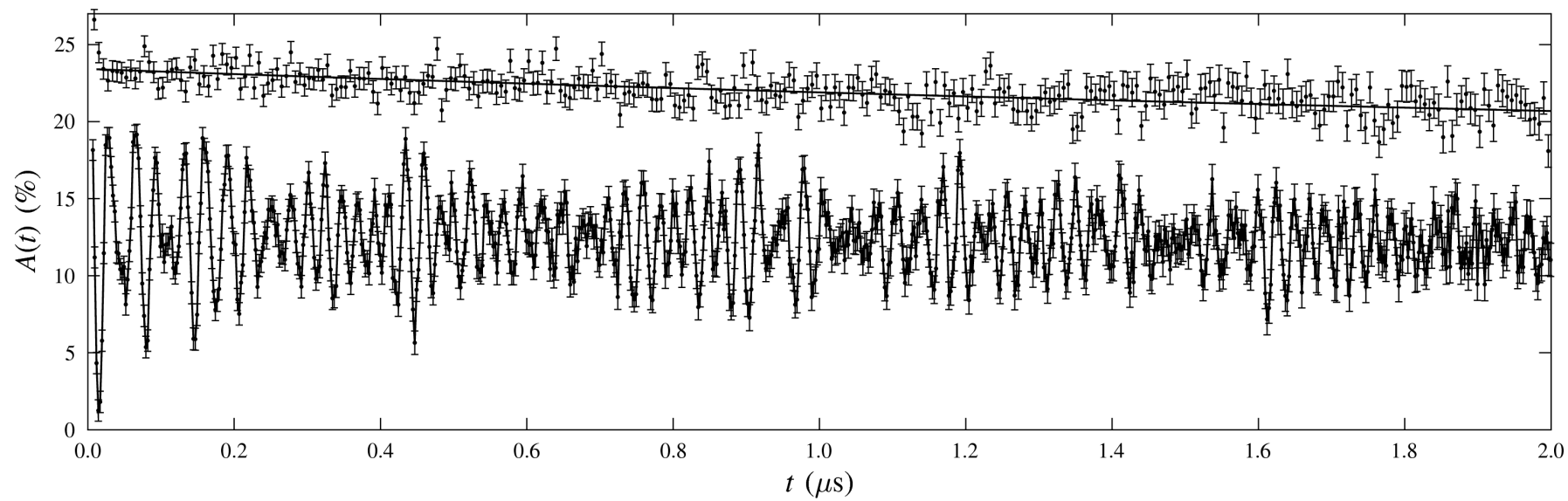
The most beautiful magnetic spectrum ever measured?

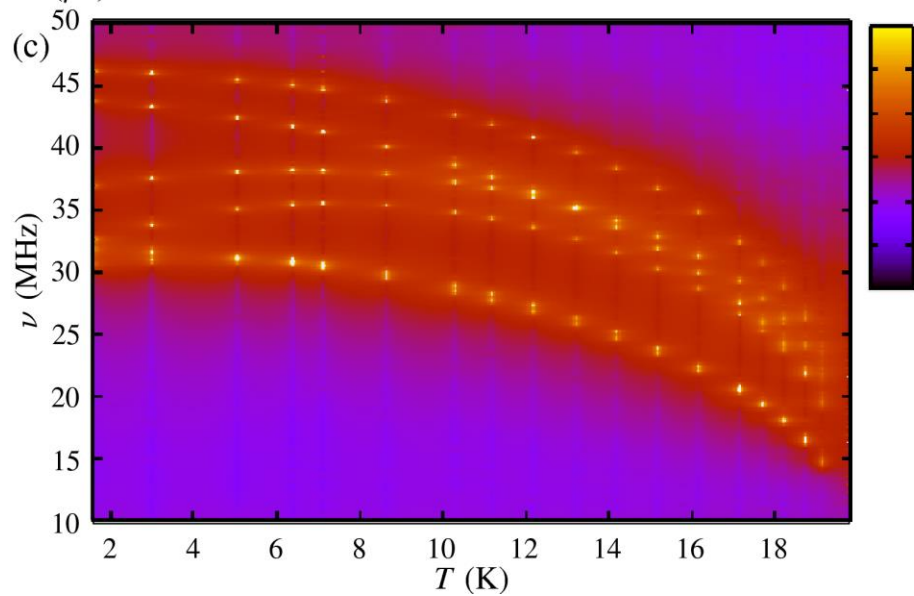
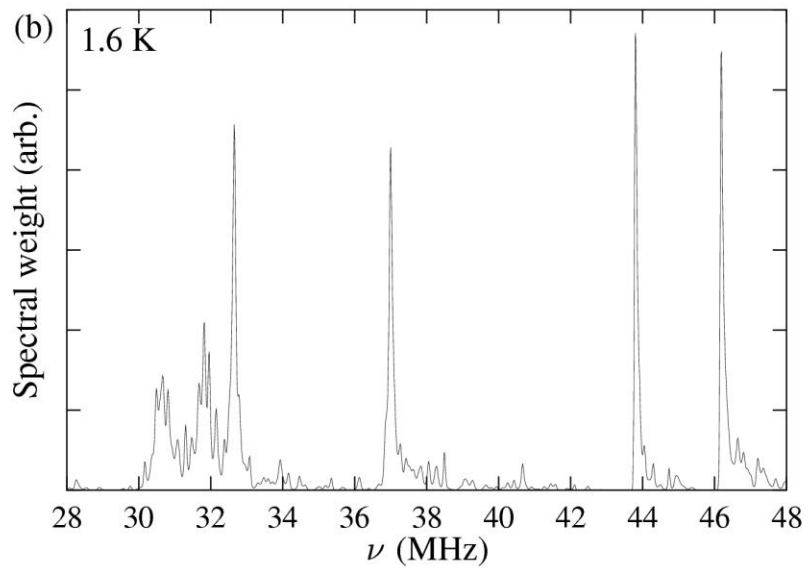
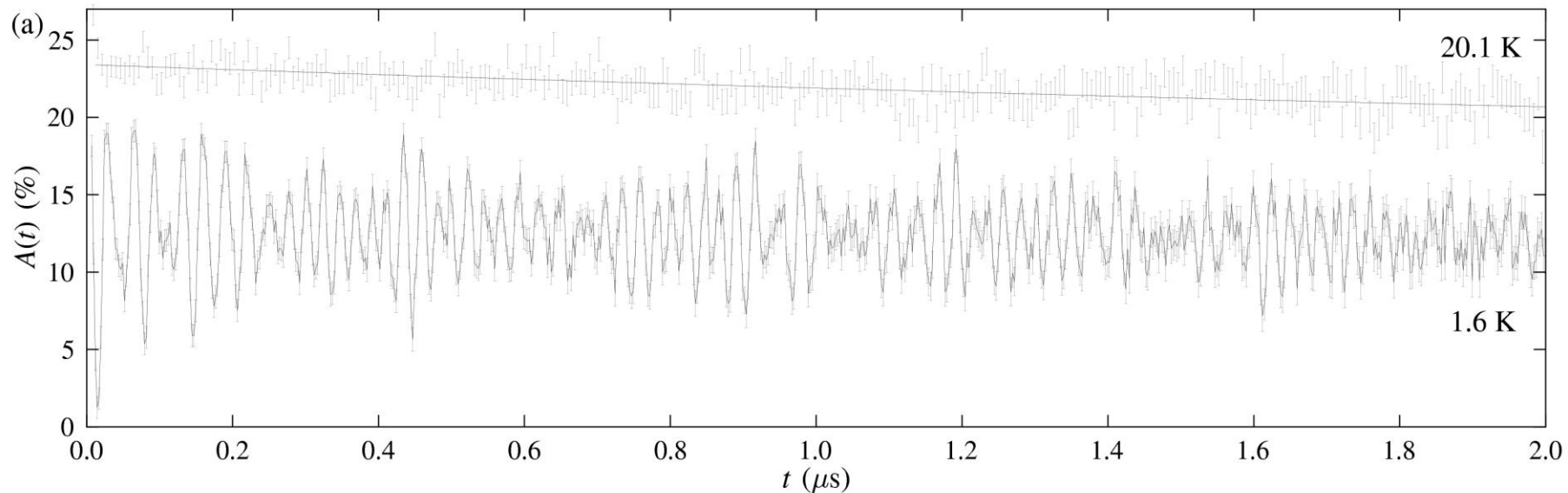




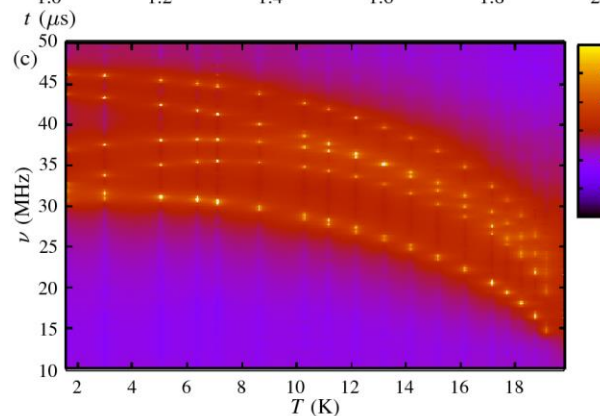
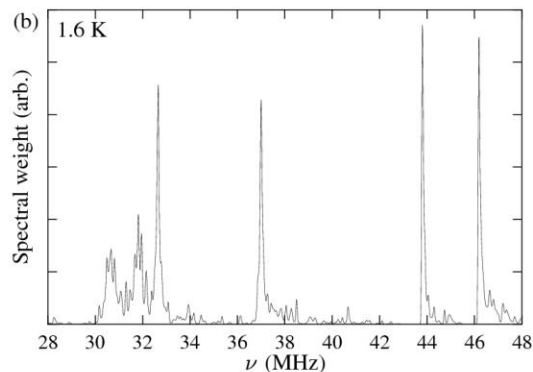
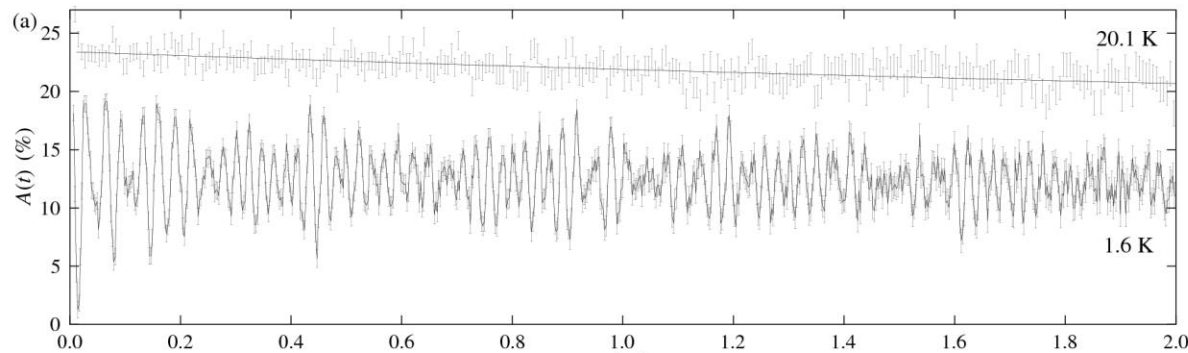
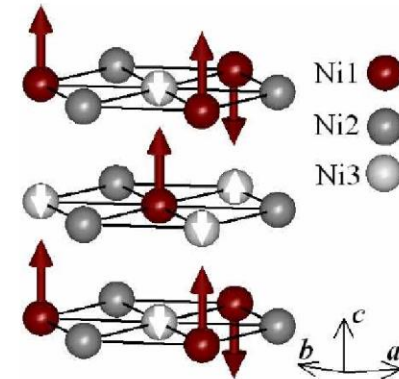
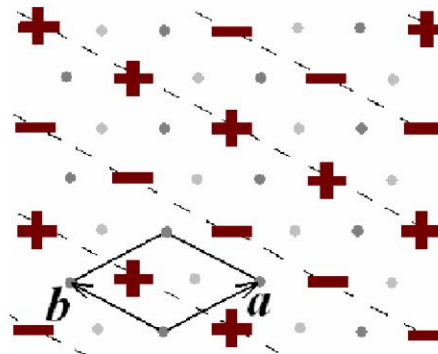
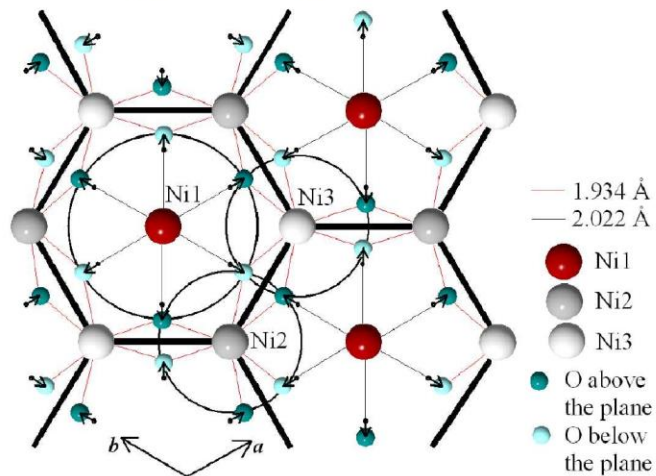








# AgNiO<sub>2</sub>: a new charge ordered state of matter?

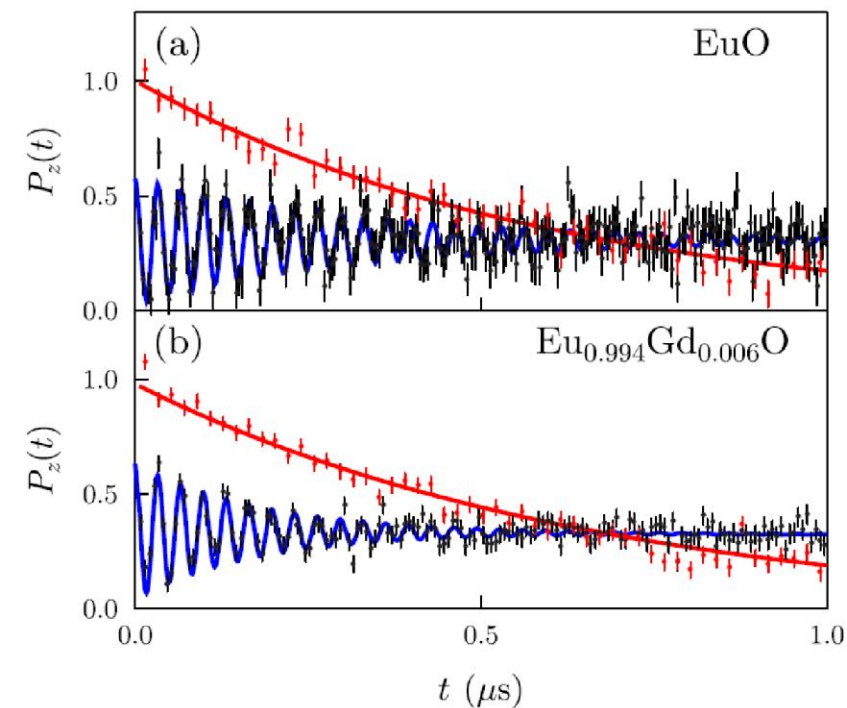


Orbital degeneracy lifted via a charge ordering mechanism

This gives rise to a well defined magnetic structure

Muons see this, but show an anomalous  $T$  dependence

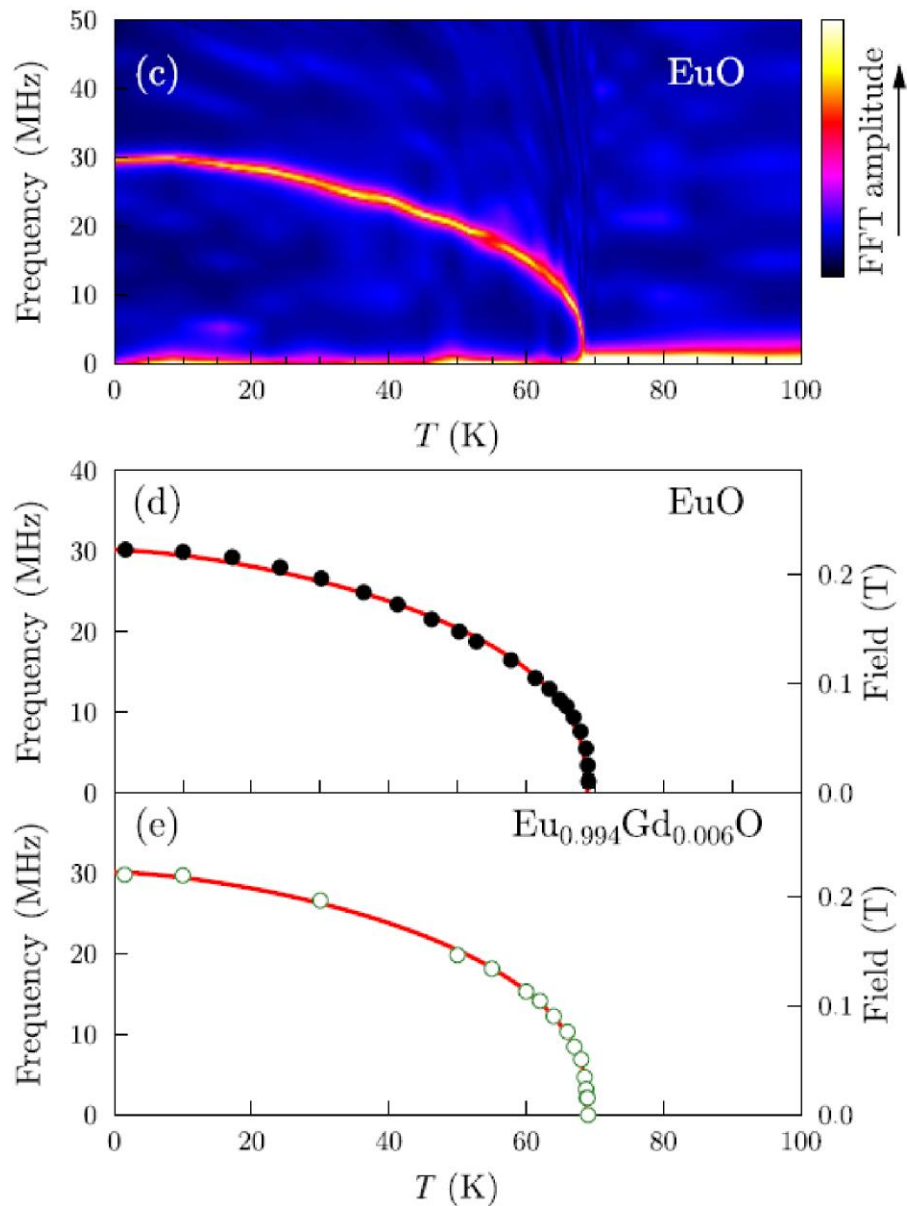
# EuO is THE localized ferromagnet



$$\nu(T) = \nu(0) [1 - (T/T_c)^\alpha]^\beta$$

$$T_c = 69.01(1) \text{ K}$$

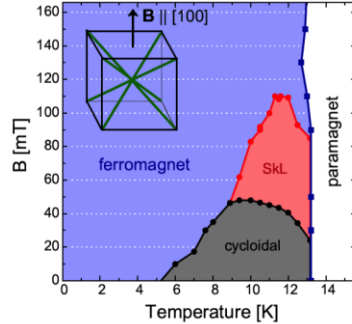
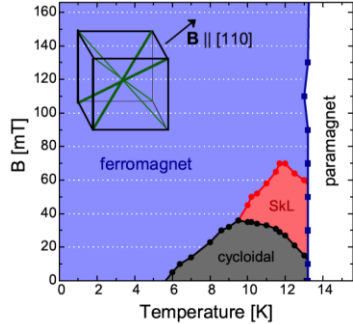
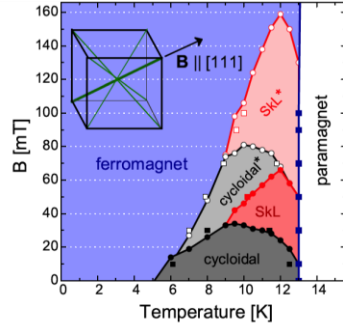
$$\beta = 0.32(1)$$



Dynamic and static  
magnetism combined

# Case study: the skyrmion lattice

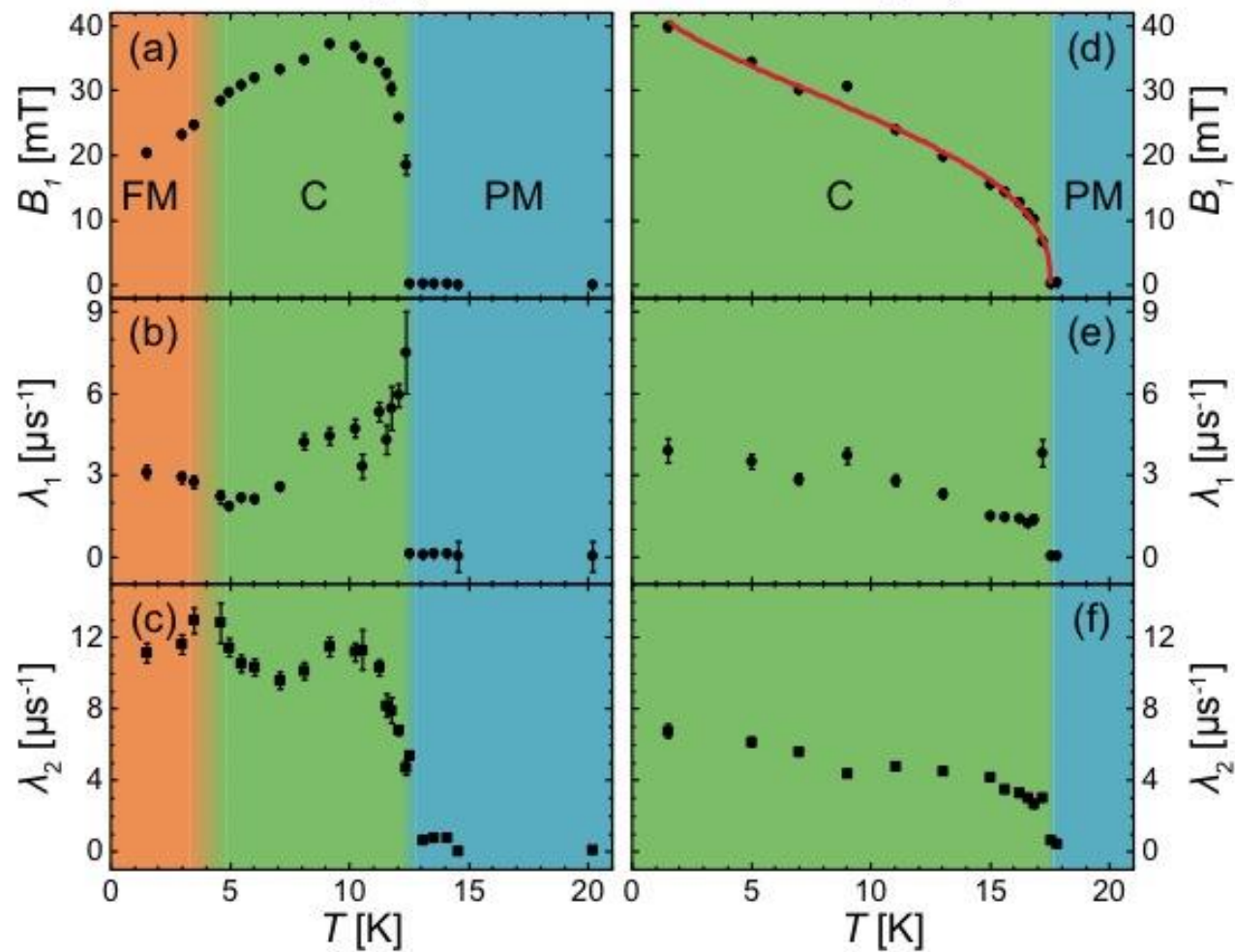


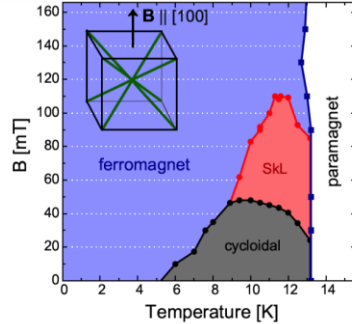
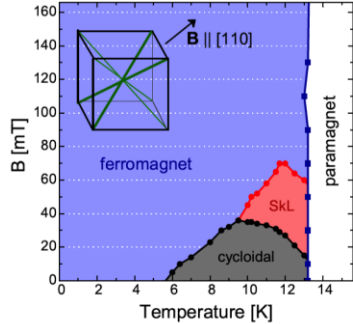
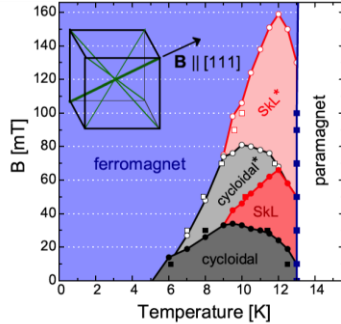


# GaV<sub>4</sub>S<sub>8</sub> and GaV<sub>4</sub>Se<sub>8</sub>

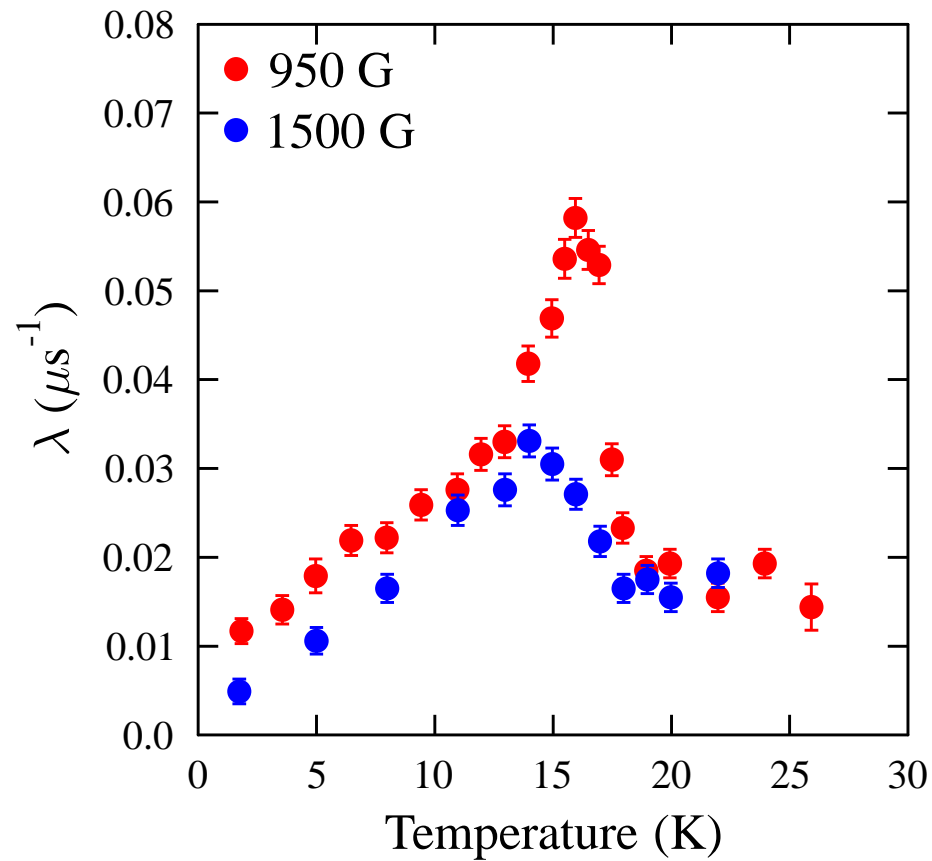
GaV<sub>4</sub>S<sub>8</sub>

GaV<sub>4</sub>Se<sub>8</sub>



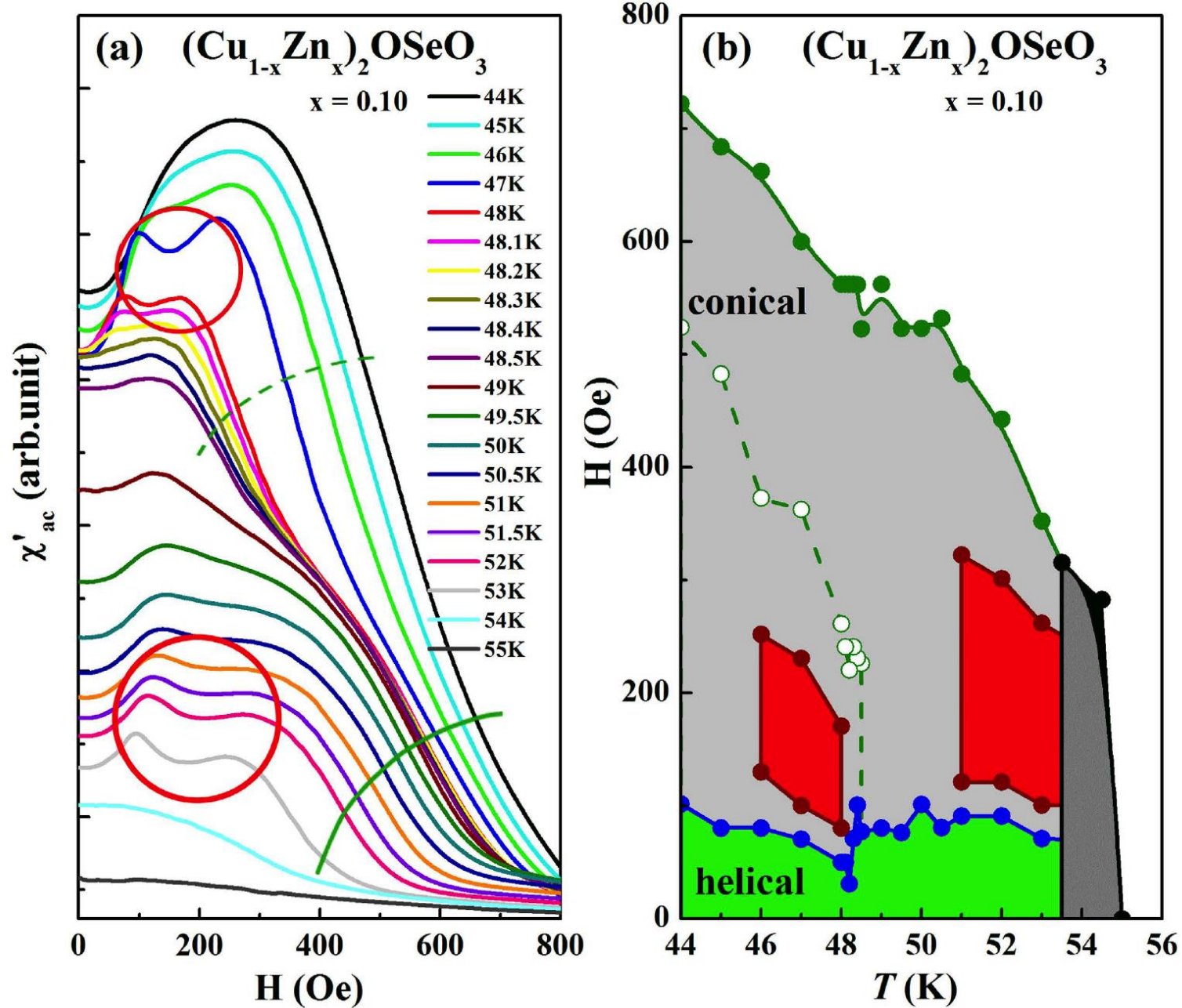


$\text{GaV}_4\text{S}_8$

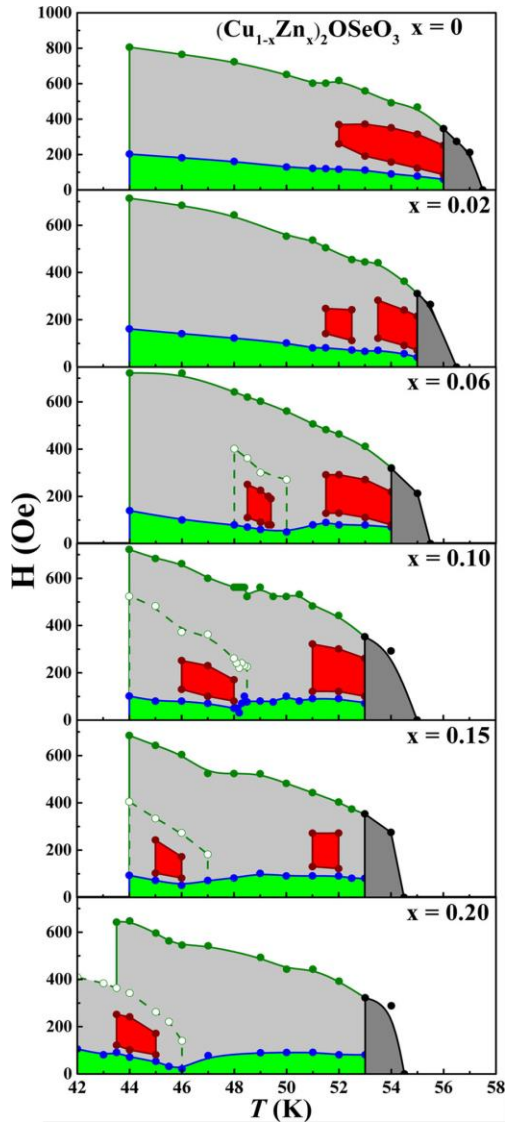


Large enhancement  
in dynamic response  
in the SkL

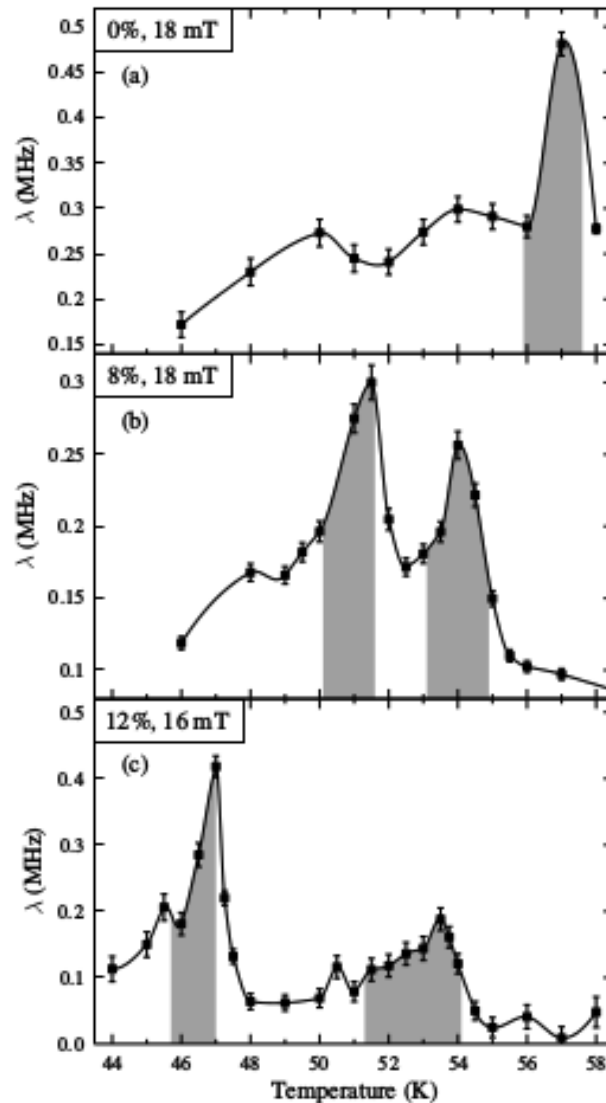
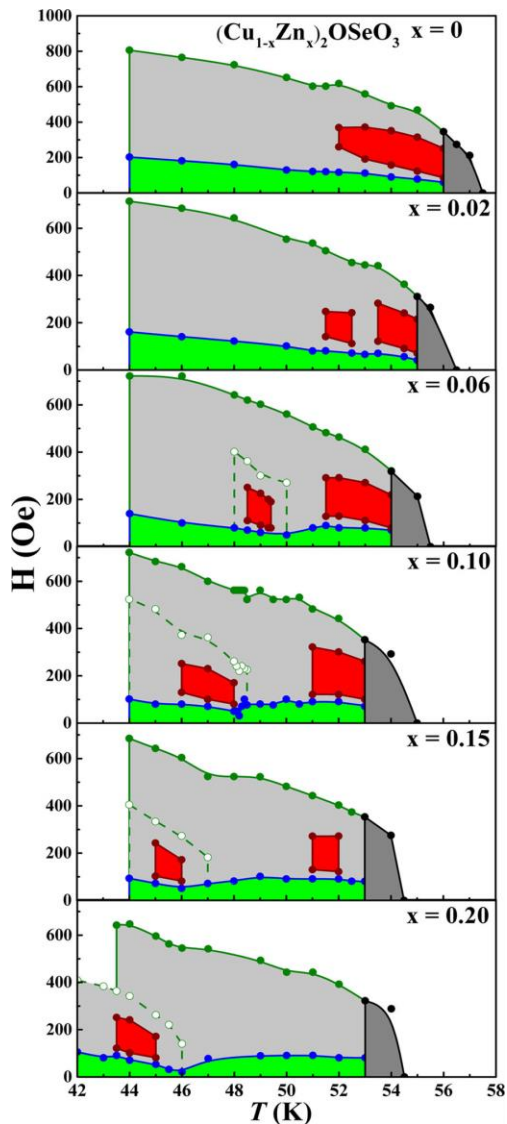
# Reentrant skyrmion phases in Zn-doped $\text{Cu}_2\text{OSeO}_3$



# Introducing disorder



# Introducing disorder

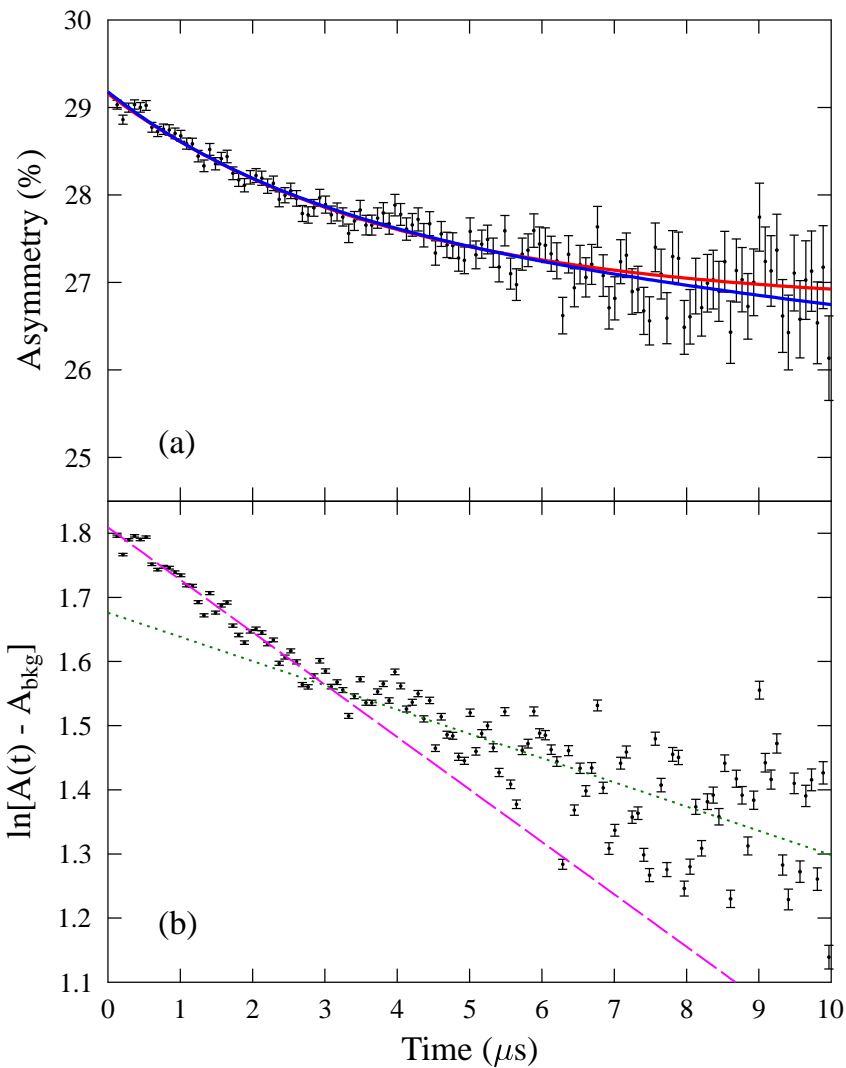
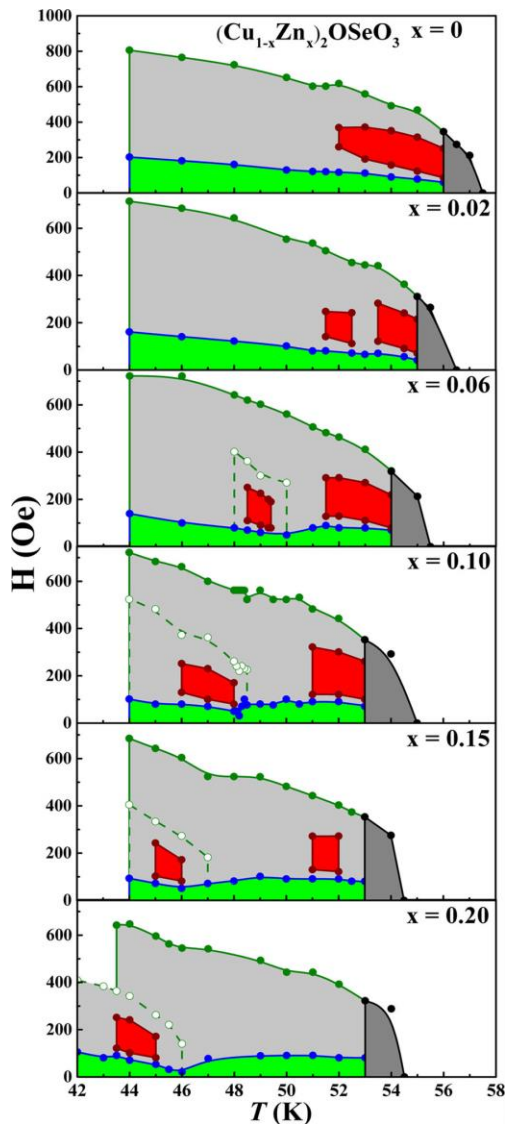


$x=0\%$

$x=8\%$

$x=12\%$

# Introducing disorder



2 exponentials,  
implying  
2 muon sites

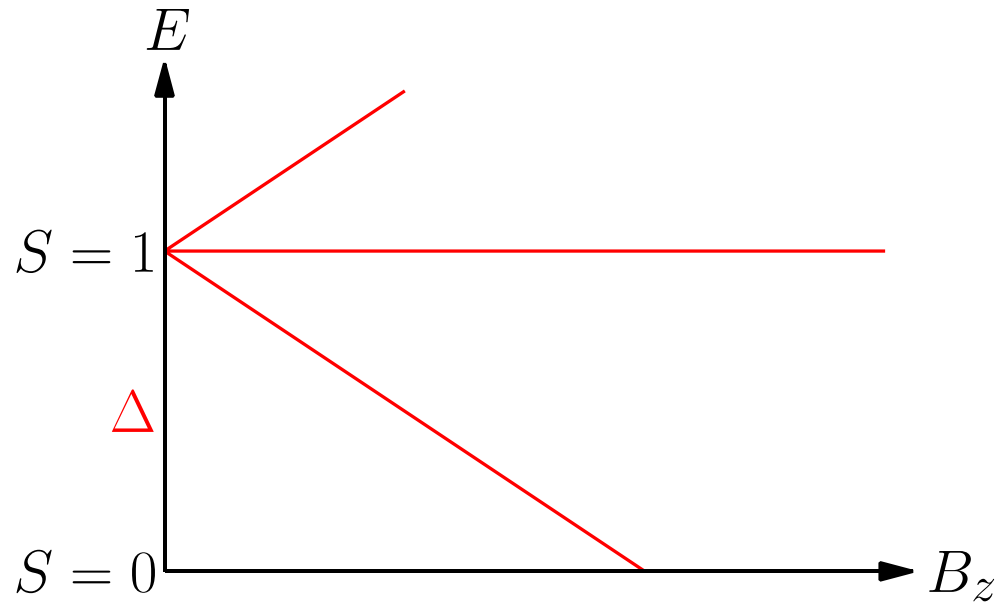
Evidence for  
Phase separation

Future directions in quantum  
magnetism:

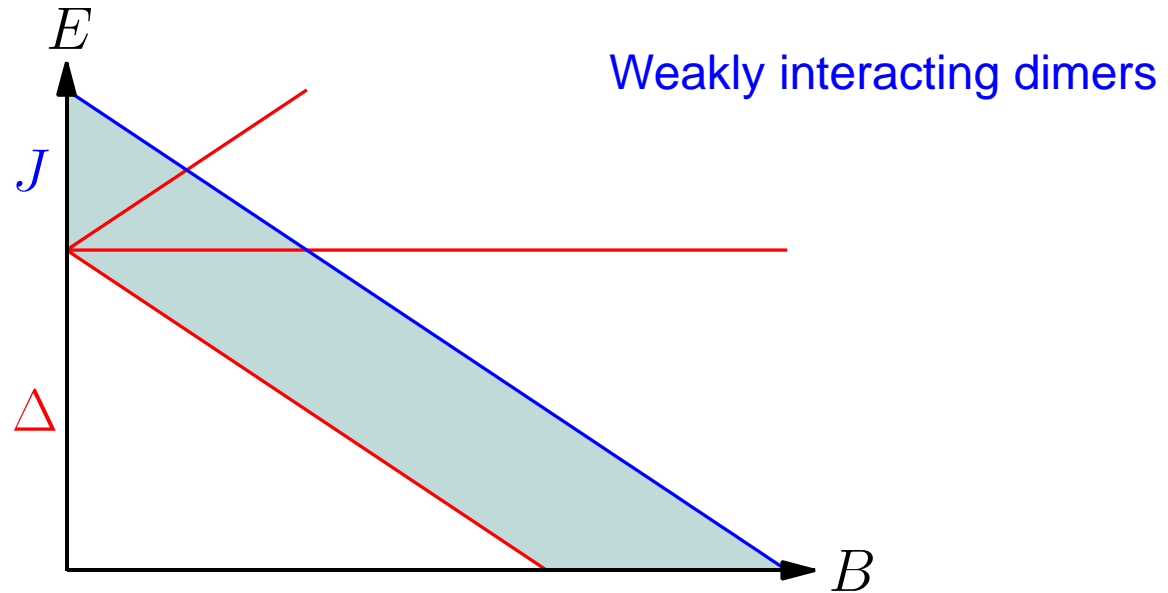
Systems built from dimers



# Isolated dimers

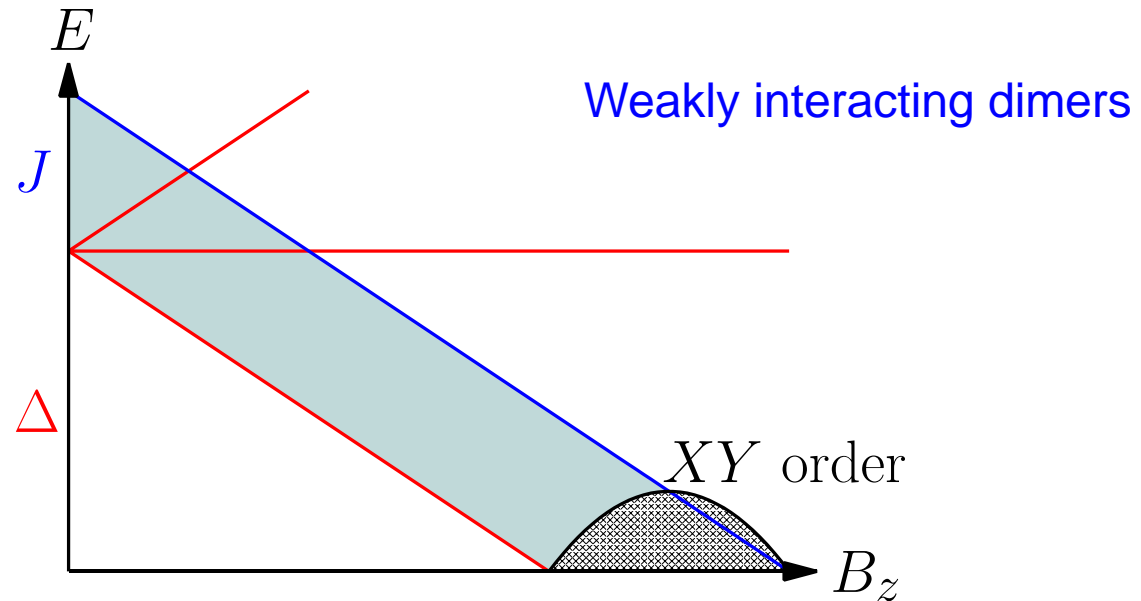


# Weakly coupled dimers



In an idealized case we expect a quantum phase transition to  $XY$  magnetic order

# Weakly coupled dimers

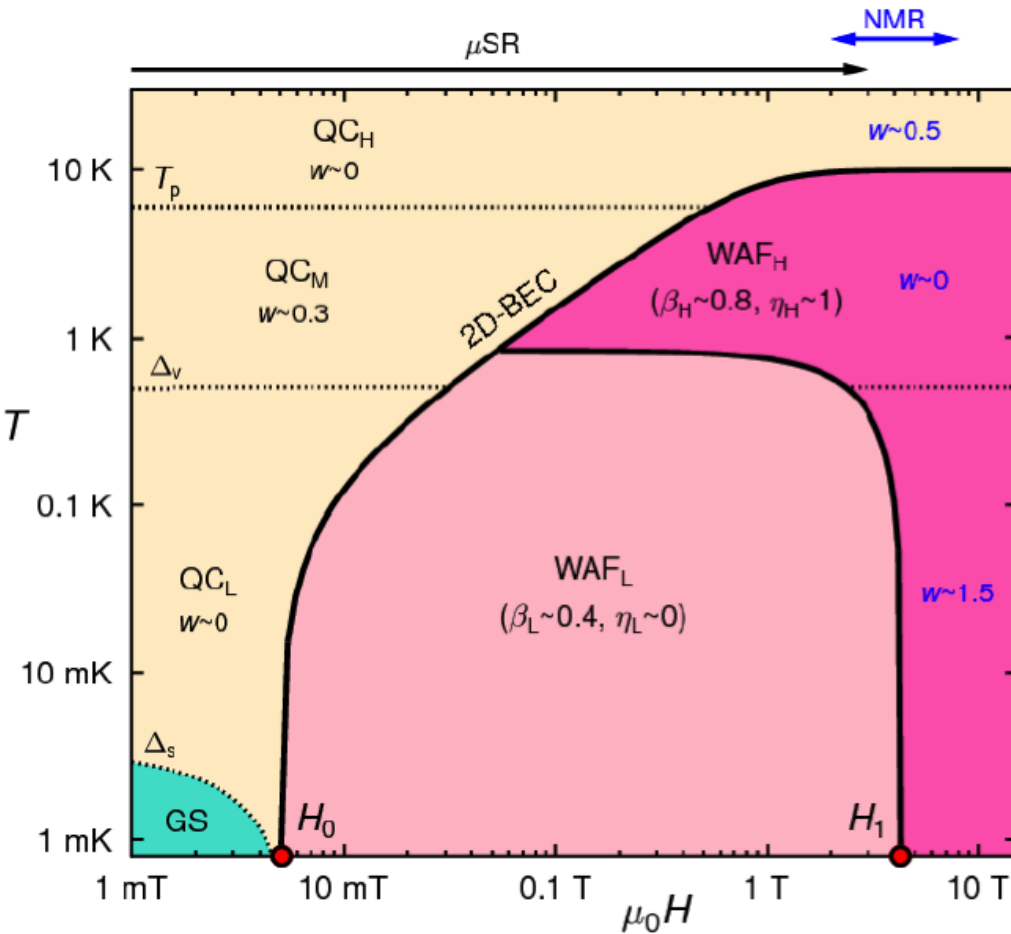


In an idealized case we expect a quantum phase transition to  $XY$  magnetic order

# Spin liquids

Frustration and fluctuations lead  
to an exotic ground state

# Spin liquid state in $\kappa\text{-ET}_2\text{Cu}_2(\text{CN})_3$



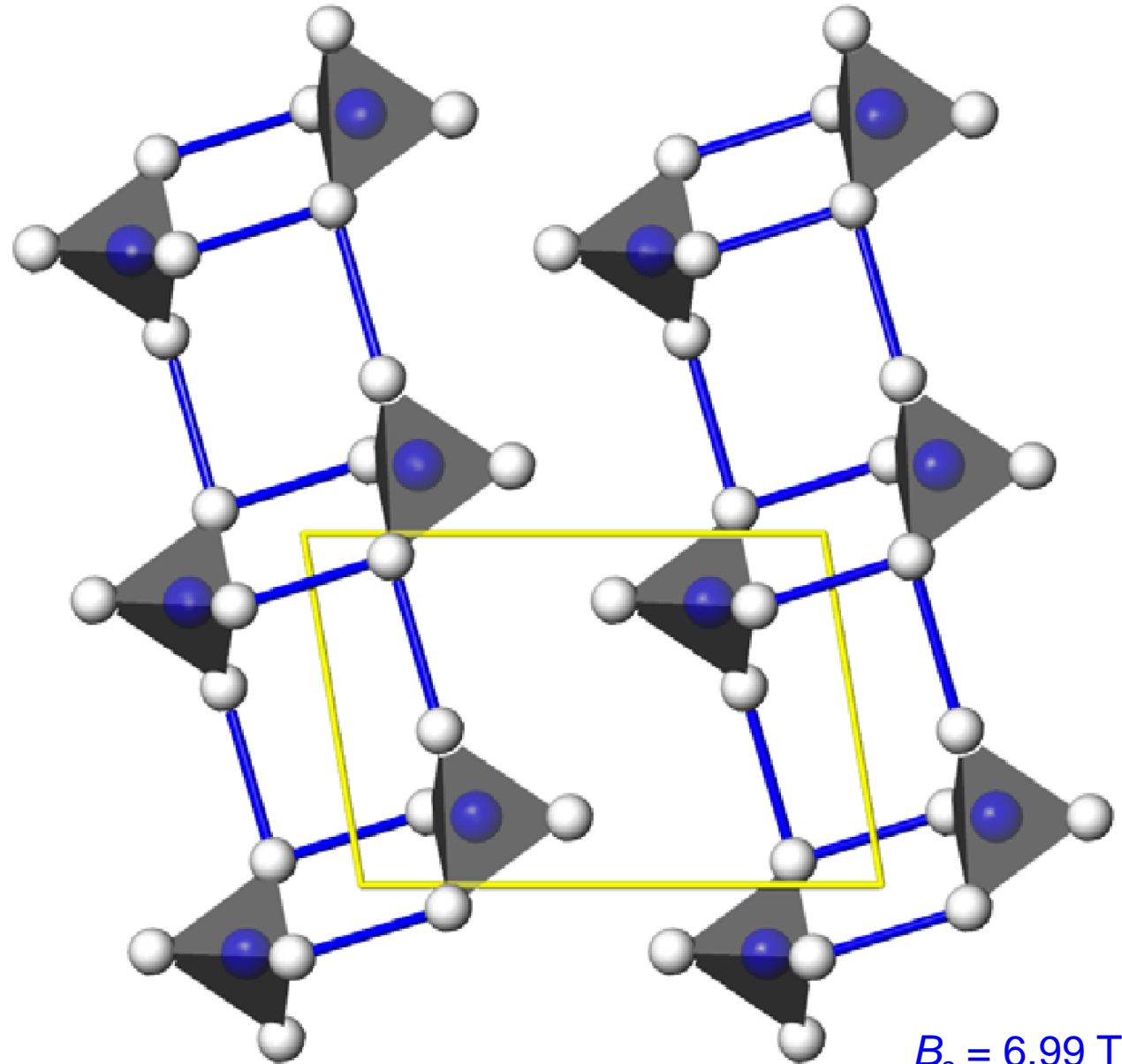
Ground state order suppressed by QM fluctuations

Field induced quantum phase transition

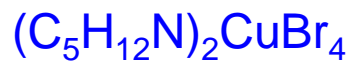
Critical behaviour consistent with bosonic or fermionic excitations

# Spin ladders as intermediate systems

# $(\text{Hpip})_2\text{CuBr}_4$ : strong run spin ladder

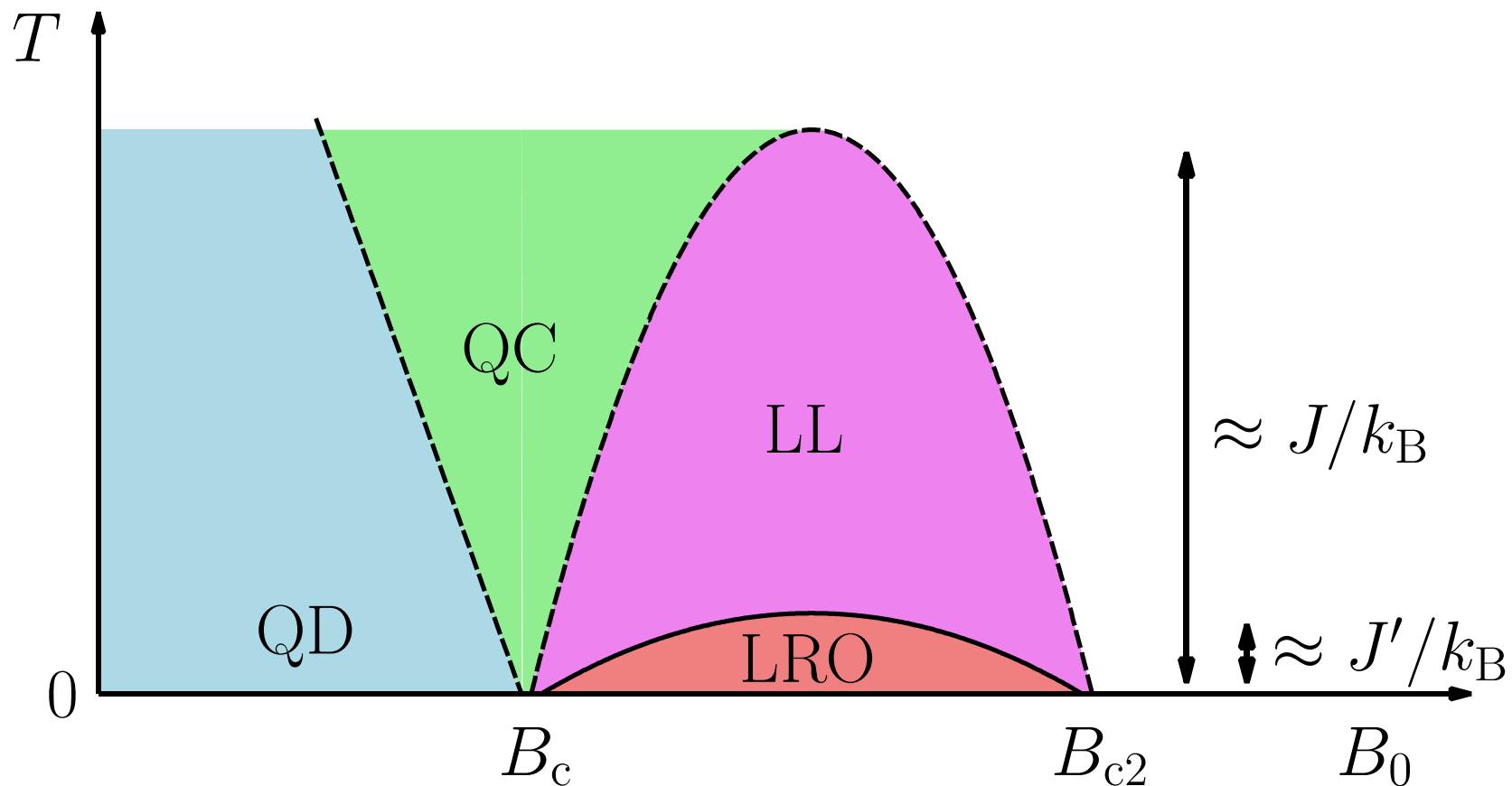


$$J_{\text{leg}}/J_{\text{rung}} = 0.25$$



$$B_c = 6.99 \text{ T}$$

# Generic phase diagram





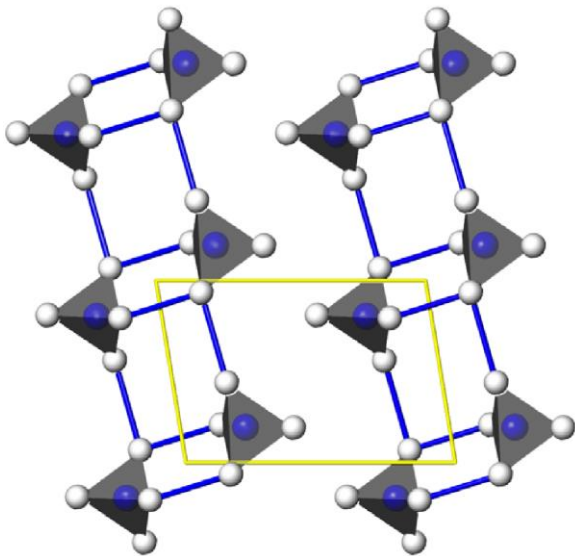
# Diffusion and dynamics

arXiv:1806.09402

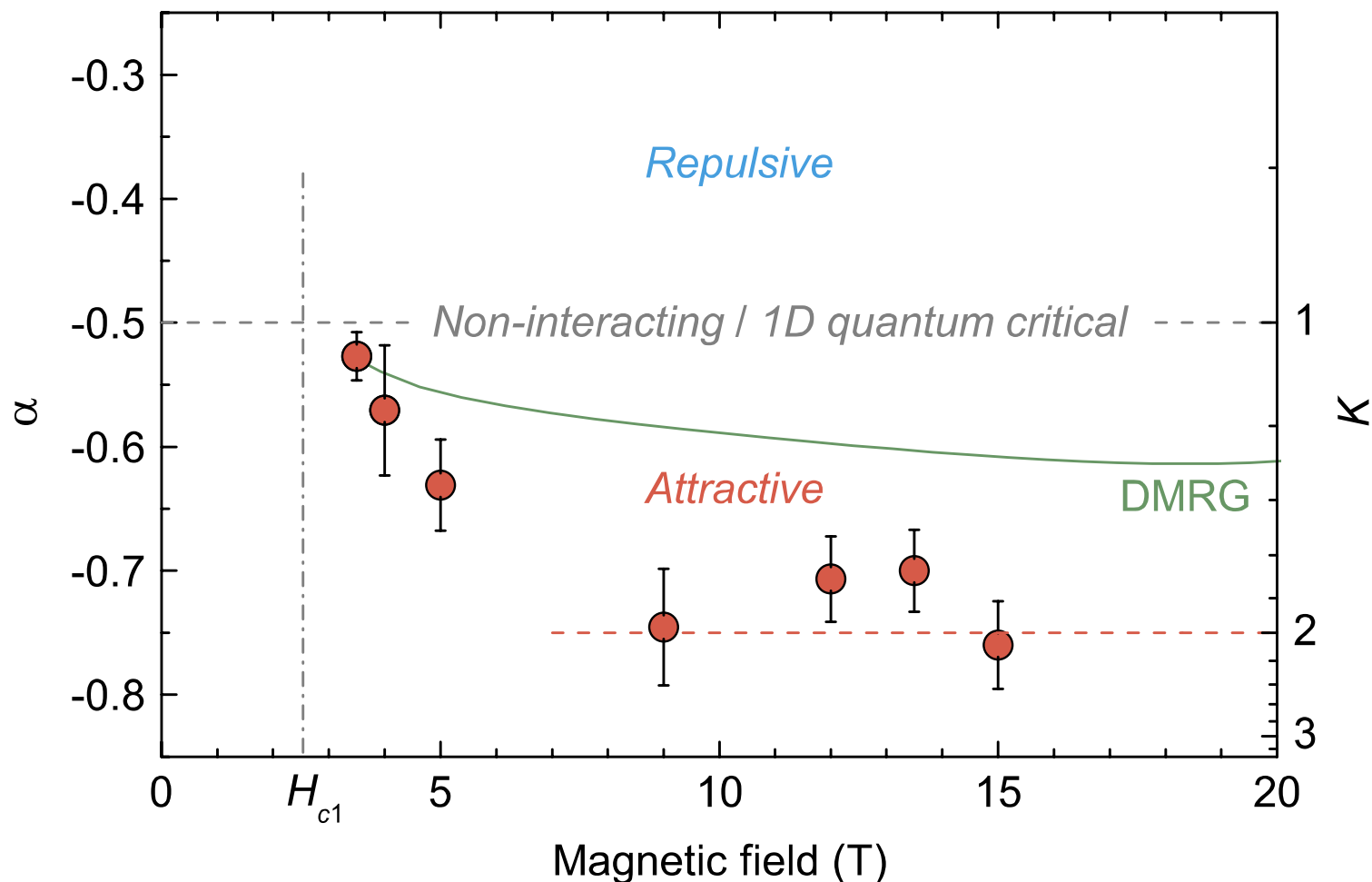
# Two predicted regimes of spin ladder behaviour:

Strong rung: repulsive interactions,  
 $1/T_1 \sim T^\alpha, \alpha > -0.5$

Strong leg: attractive interactions,  
 $1/T_1 \sim T^\alpha, \alpha < -0.5$



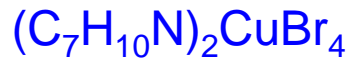
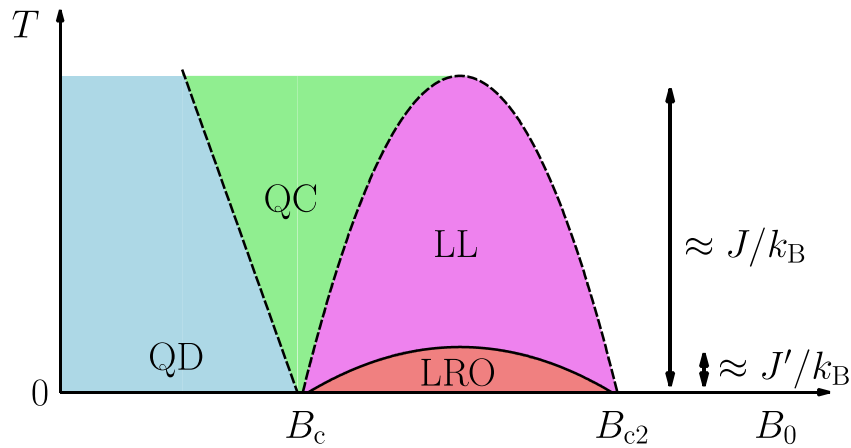
# DIMPY: a strong-leg spin ladder



Power law in spin relaxation  $1/T_1$  gives an insight into interactions

M. Jeong, *et al.*, Phys. Rev. Lett. **111**, 106404 (2013).

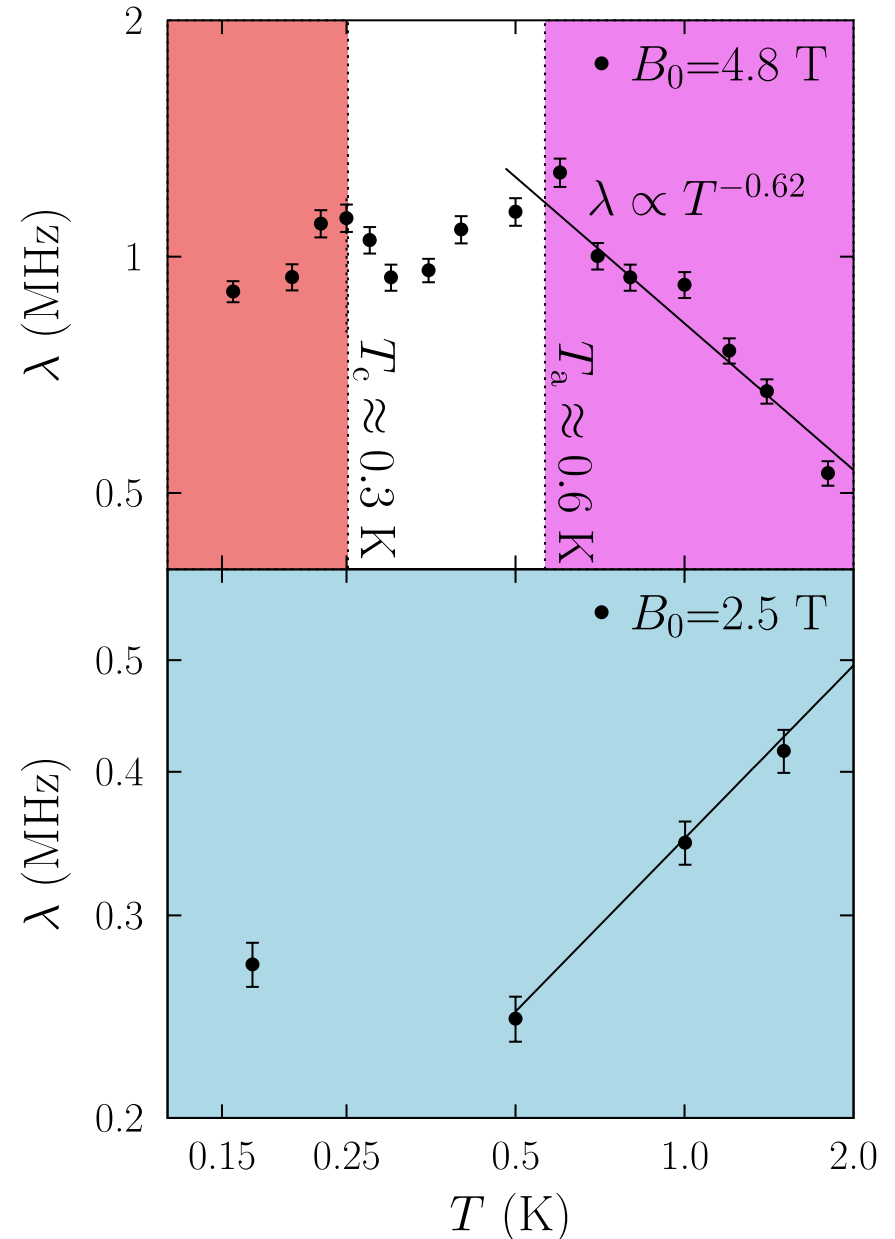
# DIMPY: a strong-leg spin ladder



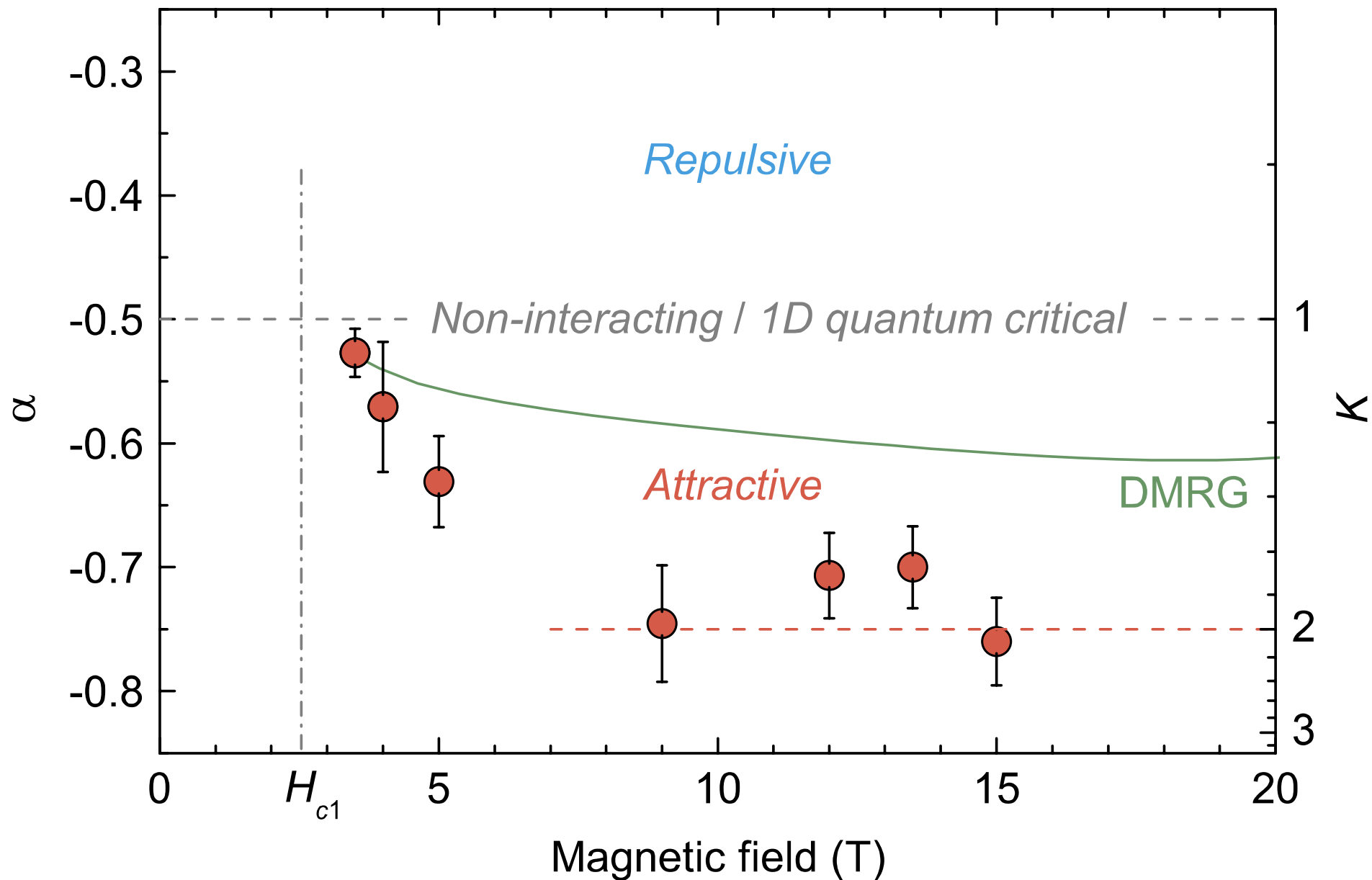
$J_{leg}/J_{rung}=2.3$

$B_c = 3.0 \text{ T}$

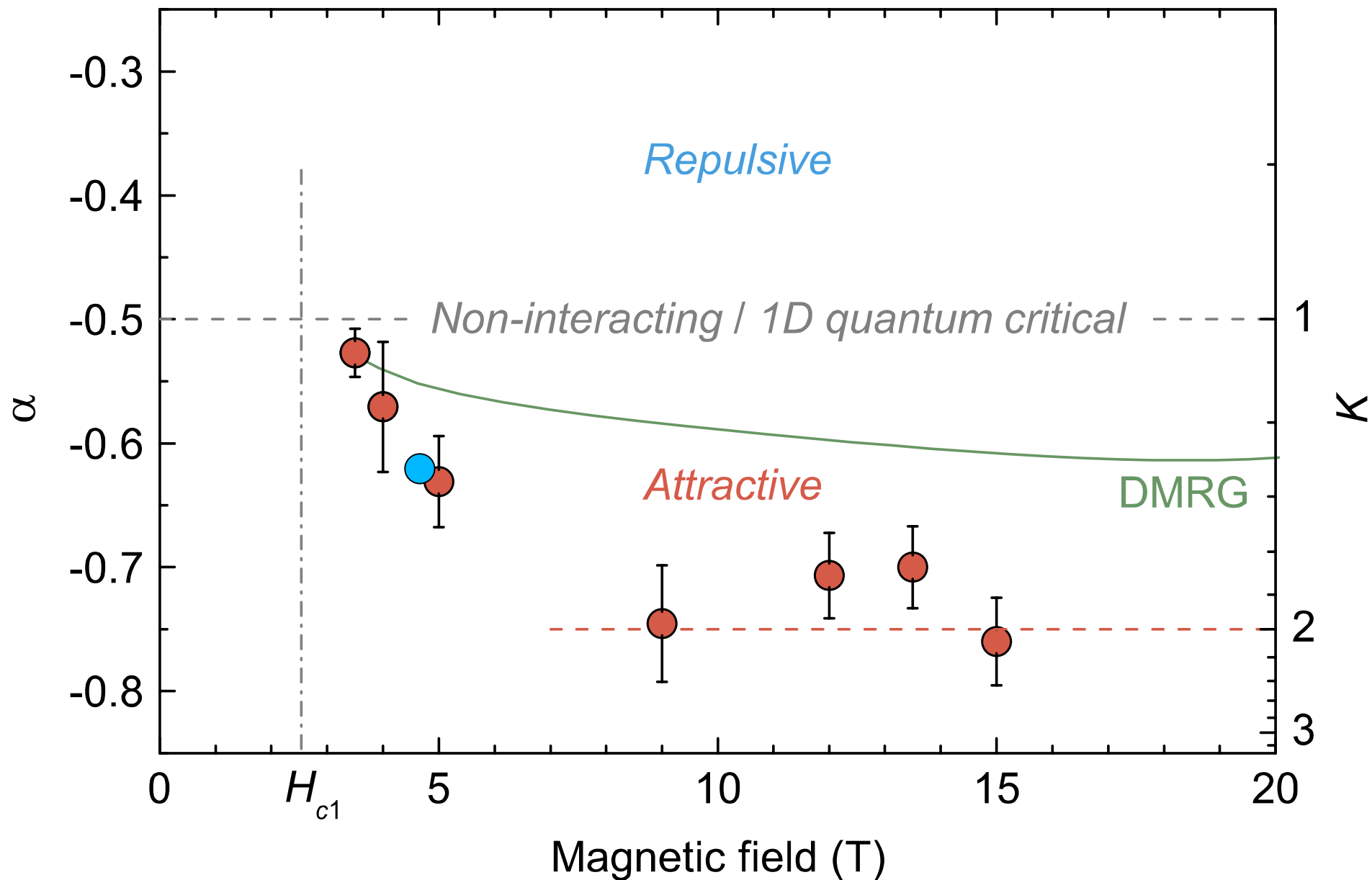
J.S. Möller *et al.*, Phys. Rev. B  
**95** 020402(R) (2017)



# DIMPY: a strong-leg spin ladder

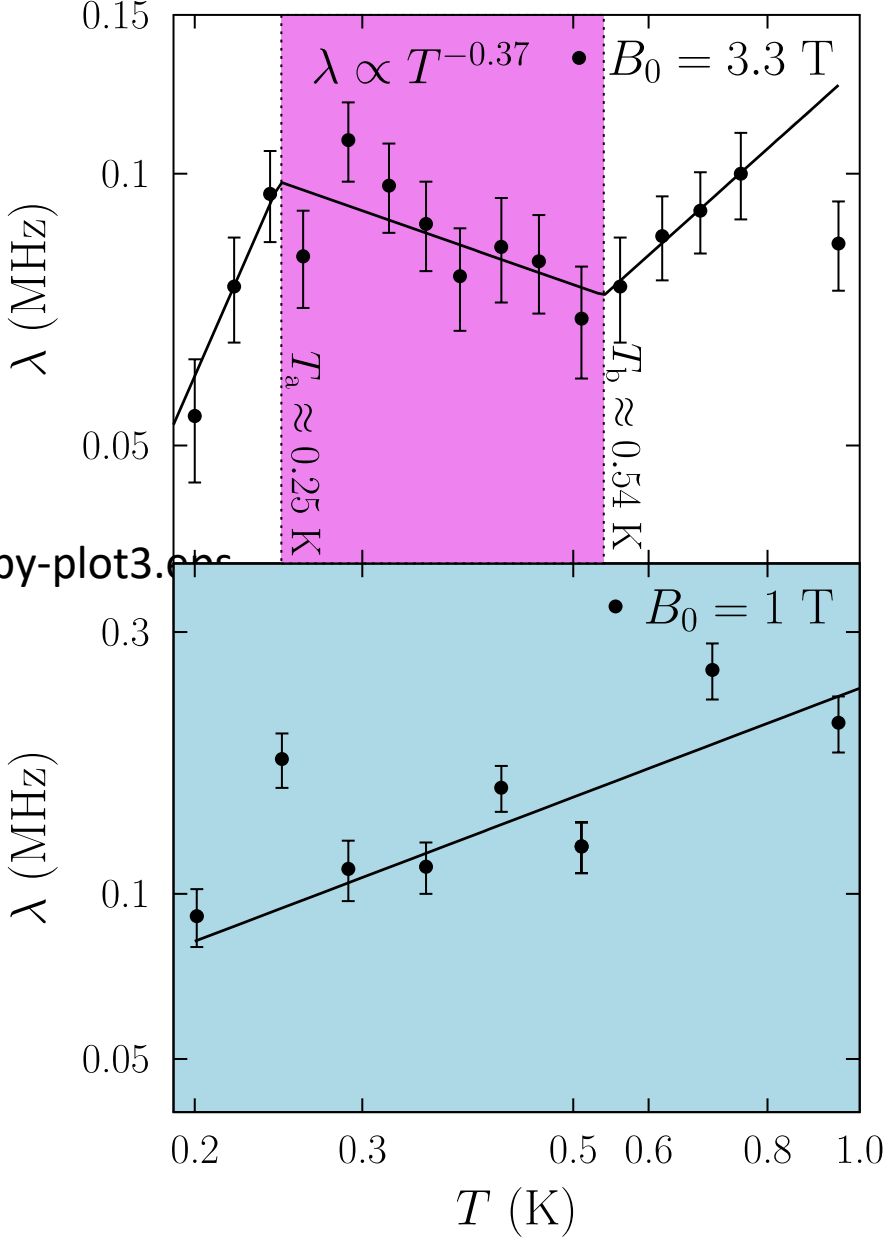
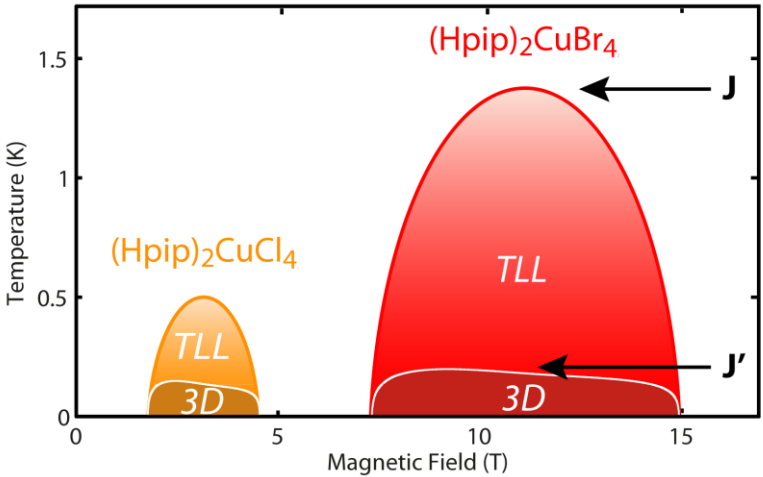


# DIMPY: a strong-leg spin ladder

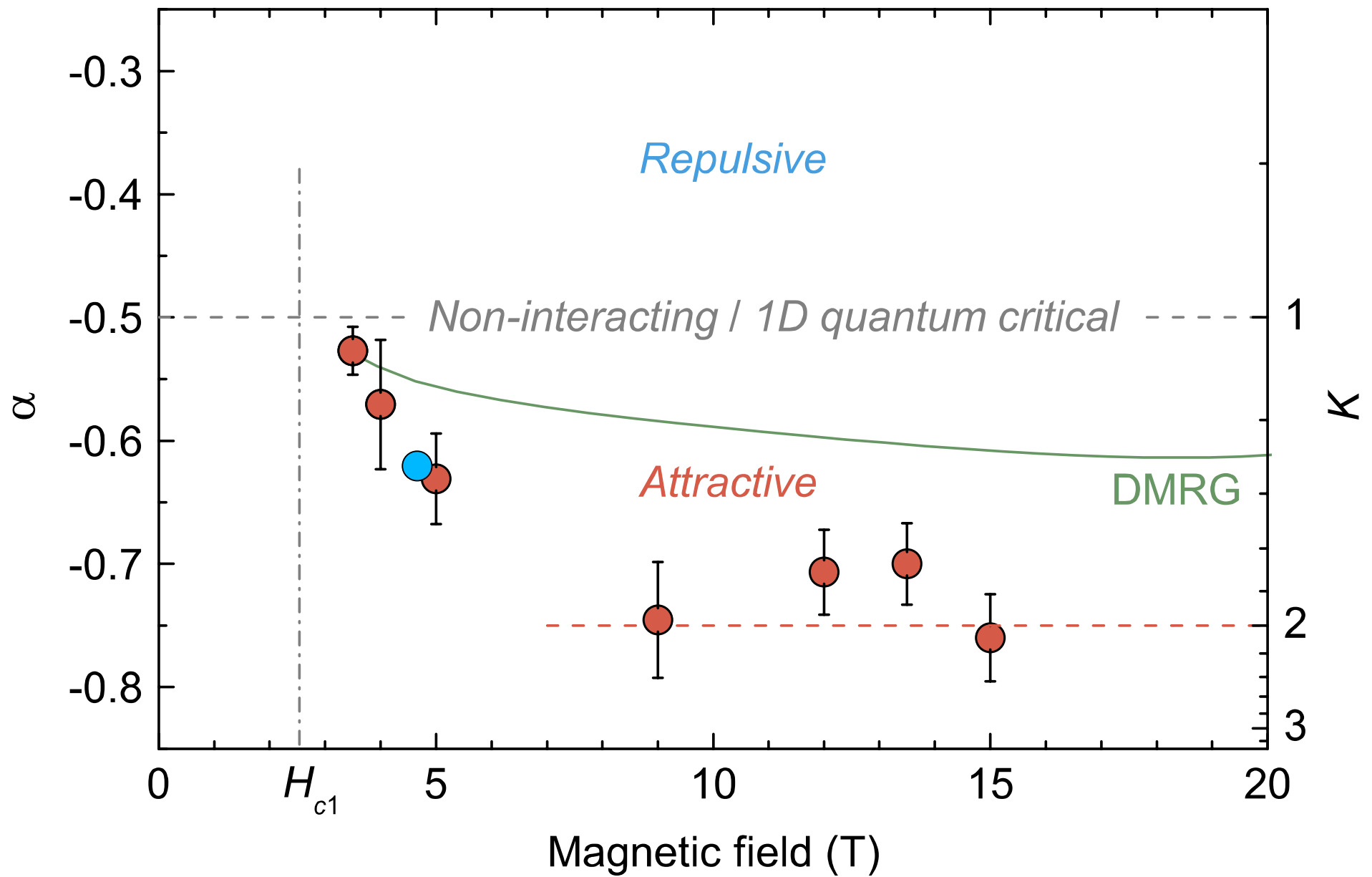


# (Hpip)<sub>2</sub>CuCl<sub>4</sub>: a strong rung ladder

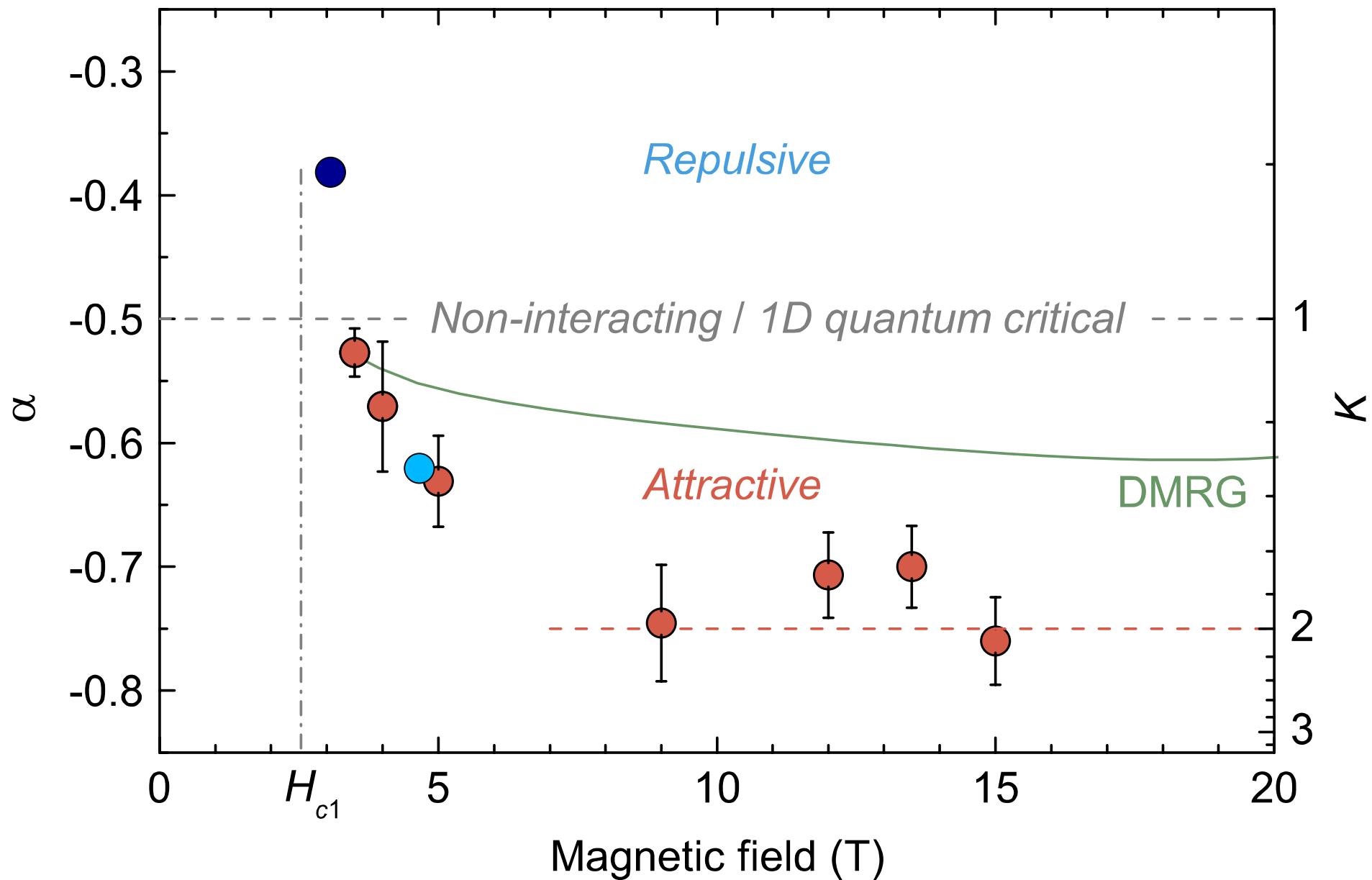
This strong-*rung* ladder has  $\alpha = -0.37$



dimpy-plot3.eps





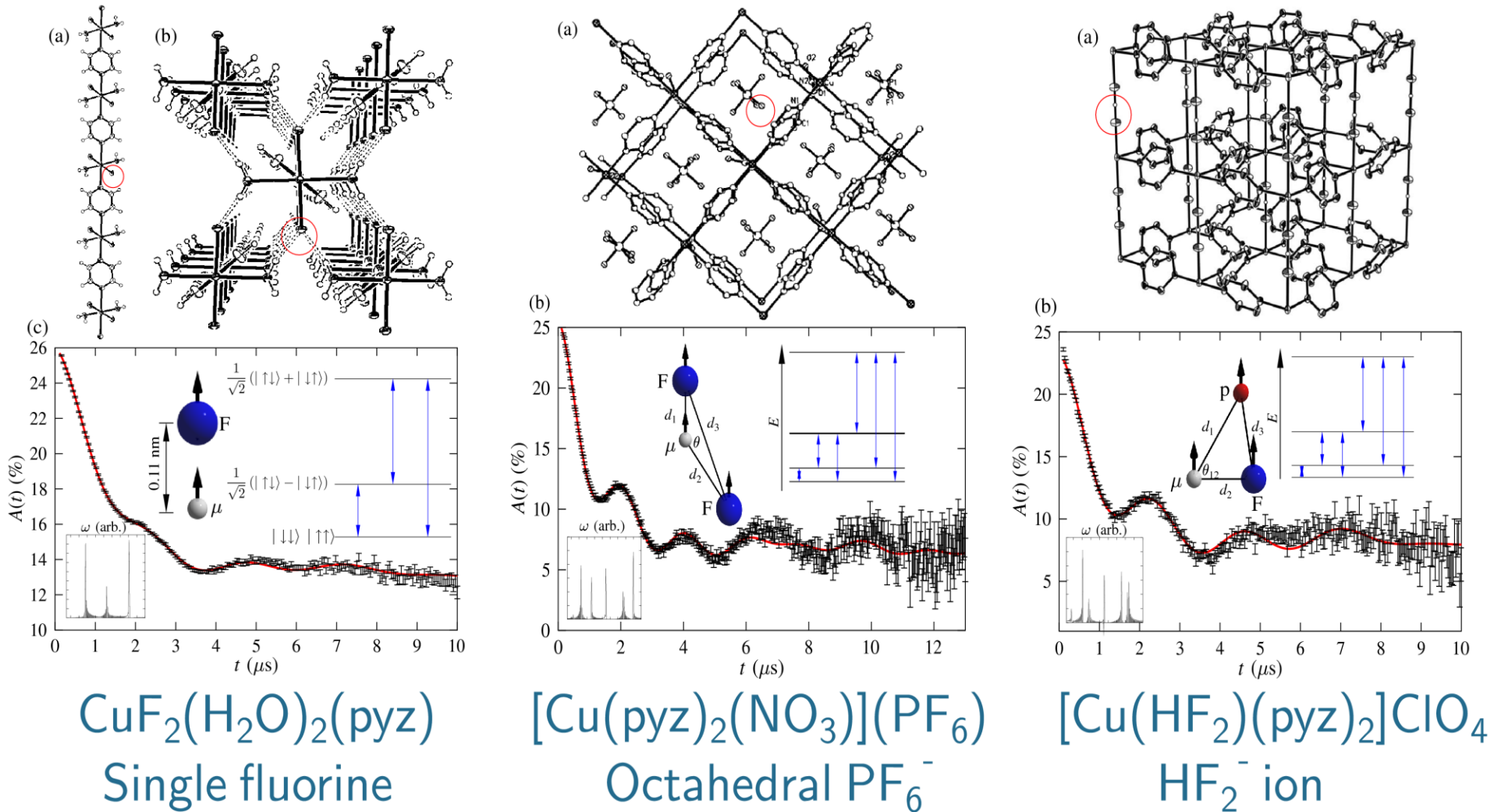


And finally...

Muon sites and DFT

# Muon-fluorine entangled states in molecular magnets

Above  $T_N$  entanglement allows us to locate the muon site

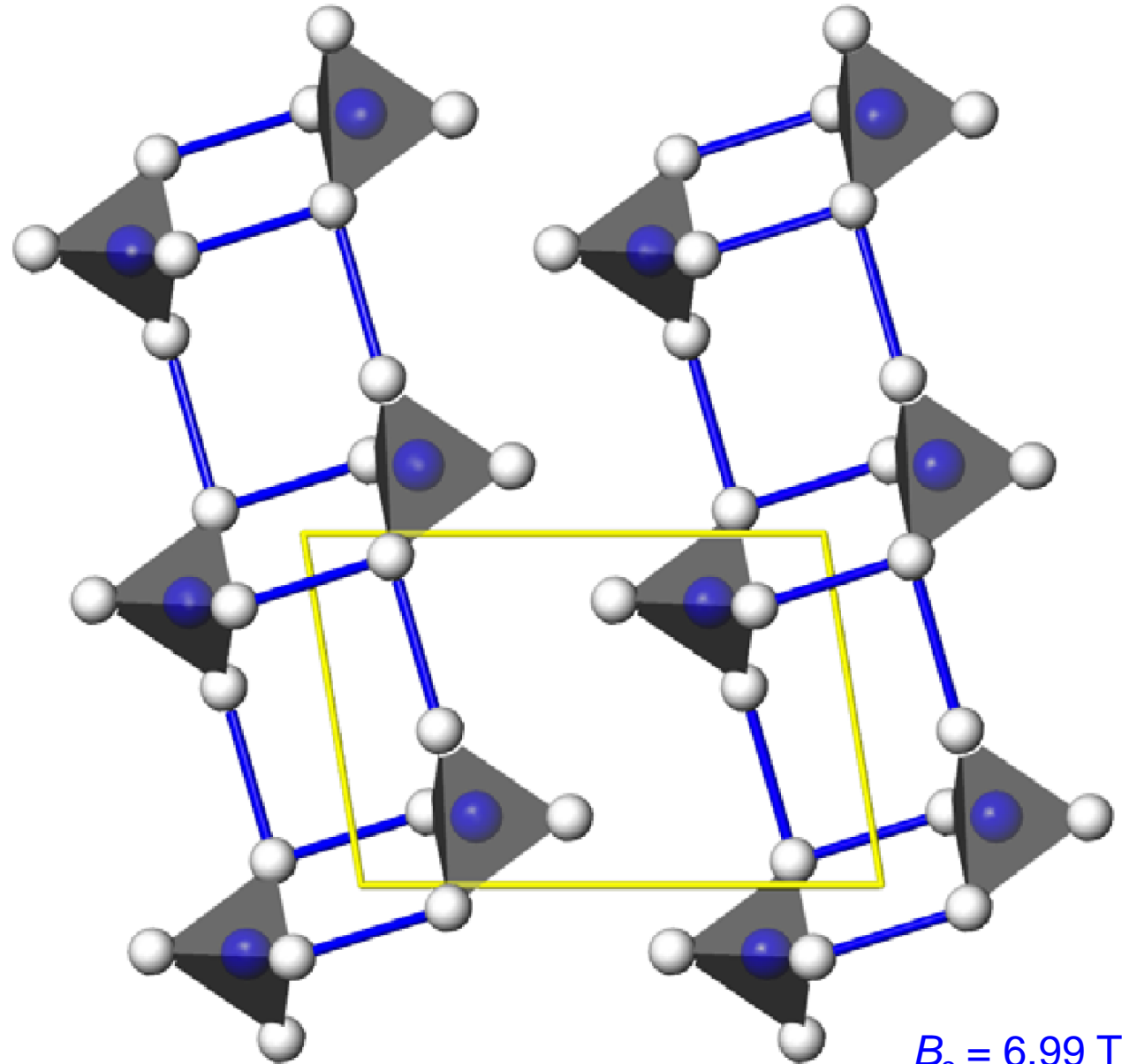


$\text{CuF}_2(\text{H}_2\text{O})_2(\text{pyz})$   
Single fluorine

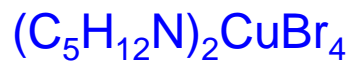
$[\text{Cu}(\text{pyz})_2(\text{NO}_3)](\text{PF}_6)$   
Octahedral  $\text{PF}_6^-$

$[\text{Cu}(\text{HF}_2)(\text{pyz})_2]\text{ClO}_4$   
 $\text{HF}_2^-$  ion

# $(\text{Hpip})_2\text{CuBr}_4$ : strong run spin ladder



$$J_{\text{leg}}/J_{\text{rung}} = 0.25$$



$$B_c = 6.99 \text{ T}$$

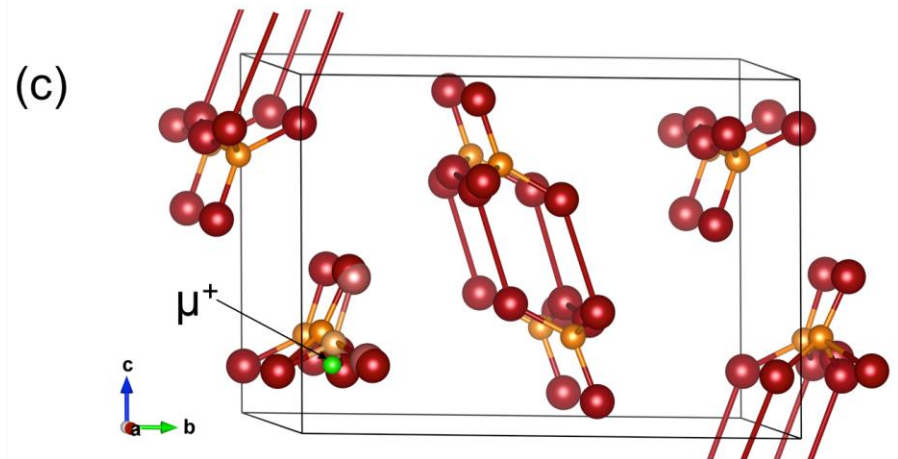
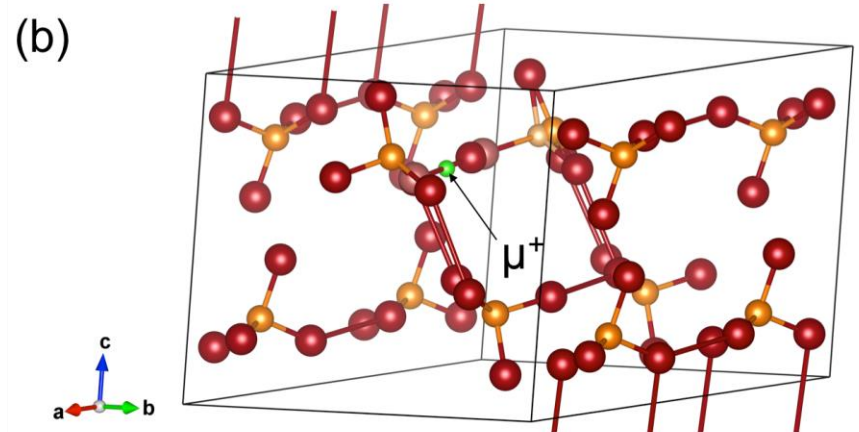
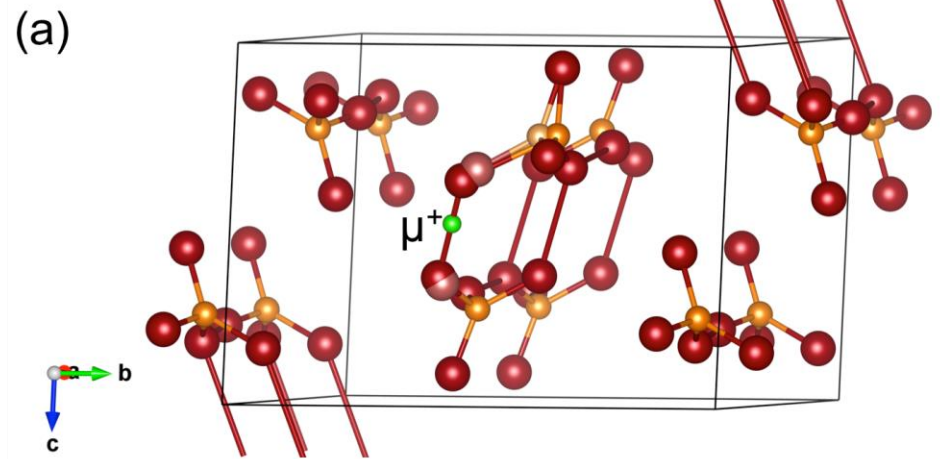
# Muon sites from DFT

1) Along the ladder rungs

2) Along the ladder legs

3) Inside the  $\text{CuBr}_4$  tetrahedra

In each case the muon forms a  $\text{Br} - \mu^+ - \text{Br}$  state



# Muon sites DFT analysis

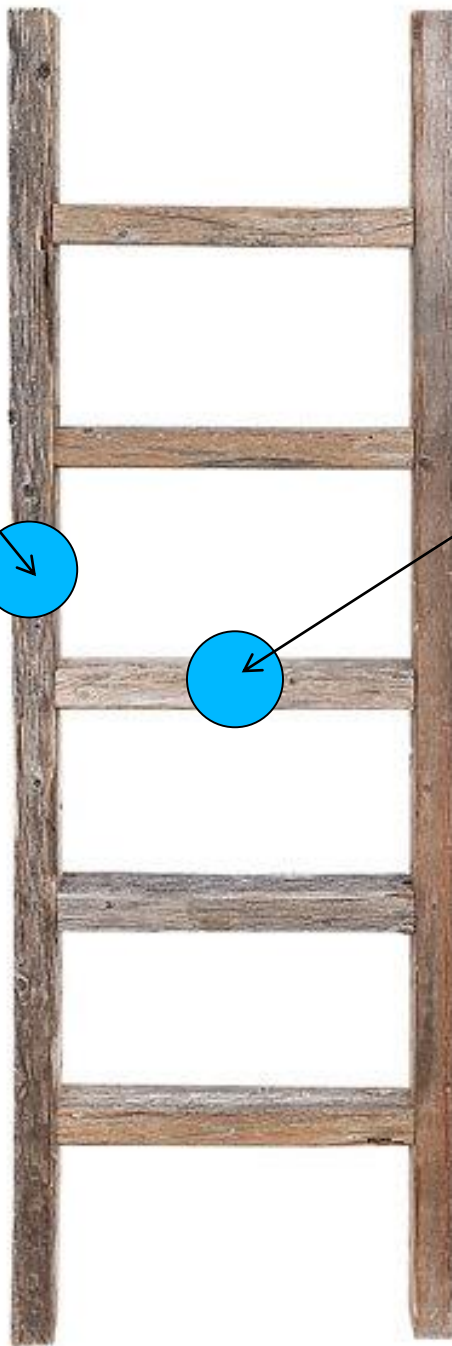
Muon site 2

Muon site 1

Making local singlets?

Hyperfine enhancement?

Breaking dimers?



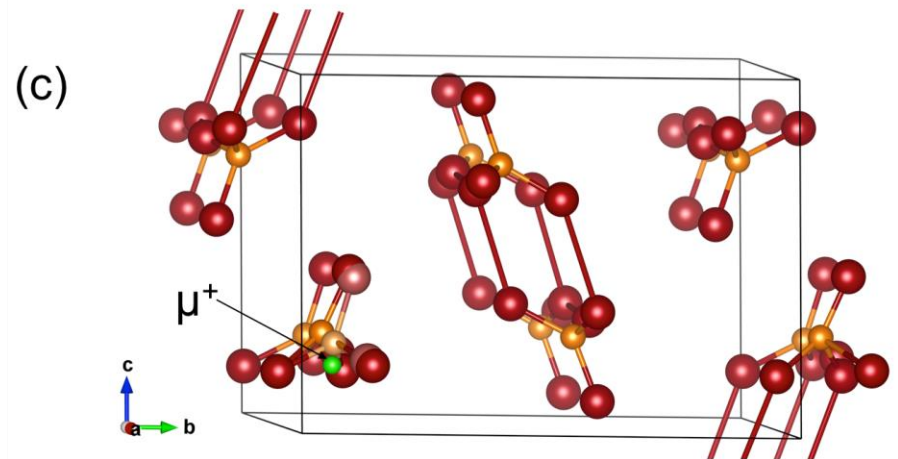
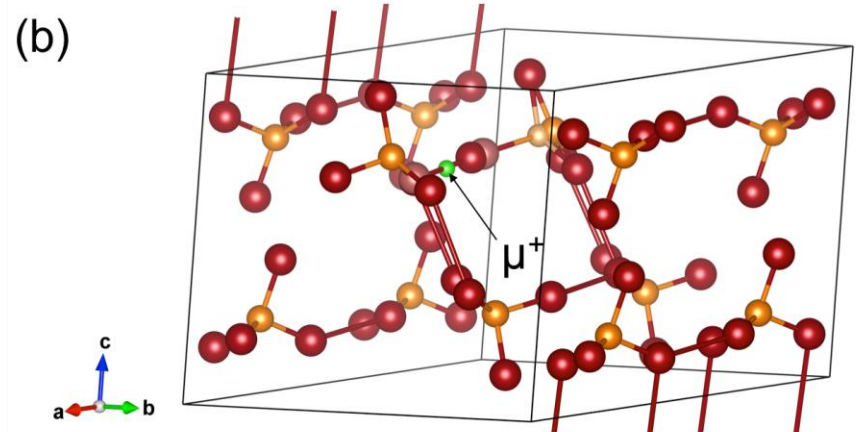
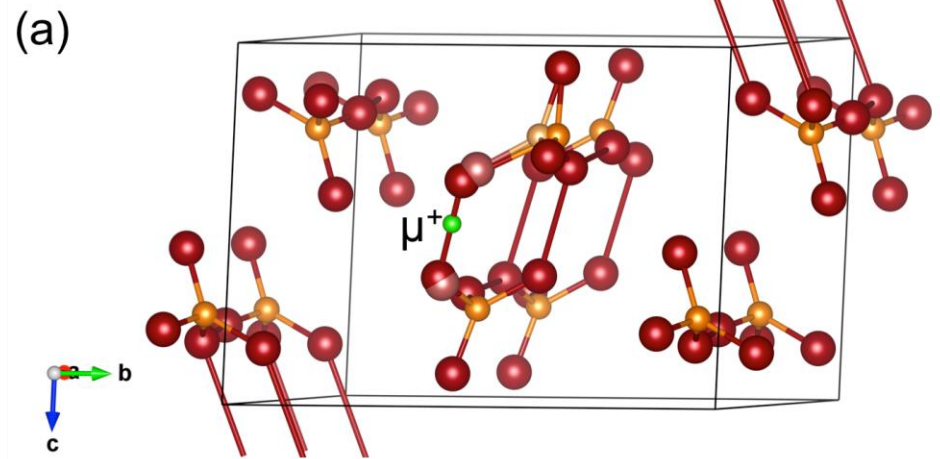
# Muon sites from DFT

1) Along the ladder rungs

2) Along the ladder legs

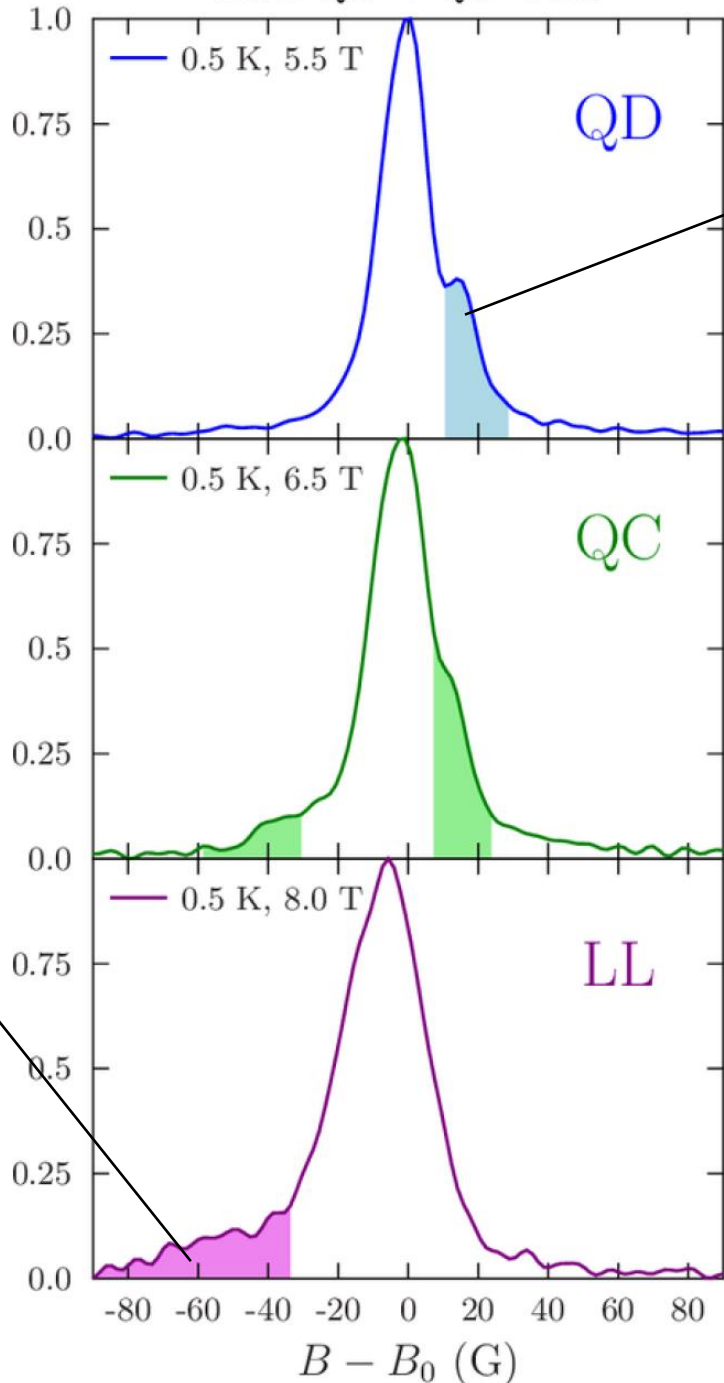
3) Inside the  $\text{CuBr}_4$  tetrahedra

In each case the muon forms a  $\text{Br} - \mu^+ - \text{Br}$  state



# TF results from PSI

Scan: QD  $\rightarrow$  QC  $\rightarrow$  LL



Rung site disrupting a dimer

Leg site disrupting a chain



# Conclusions

Super-MuSR will greatly expand the ISIS capability in magnetism

Data quality and quantity will be enhanced in small-moment systems

Increased frequency resolution will open up the variety of magnets

Data rate will transform studies of dynamics

Muon site determination frequently rests on high statistics data sets

# Acknowledgements

Durham: Rob Williams, Fan Xiao, Peter Hatton, Thomas Hicken, Ben Huddart, Kévin Franke

ISIS: Francis Pratt

Oxford: Stephen Blundell, Thorsten Hesjedal, Shilei Zhang

PSI: Christian Rüegg, Simon Ward, Tatsuo Goko, Robert Scheuermann, Zaher Salman, Thomas Prokscha, Andreas Suter.

Warwick: Geetha Balakrishnan, Monica Ciomaga Hatnean, Ales Stafancic

Eastern Washington: Jamie Manson