Domain wall spin dynamics in Kagome antiferromagnets

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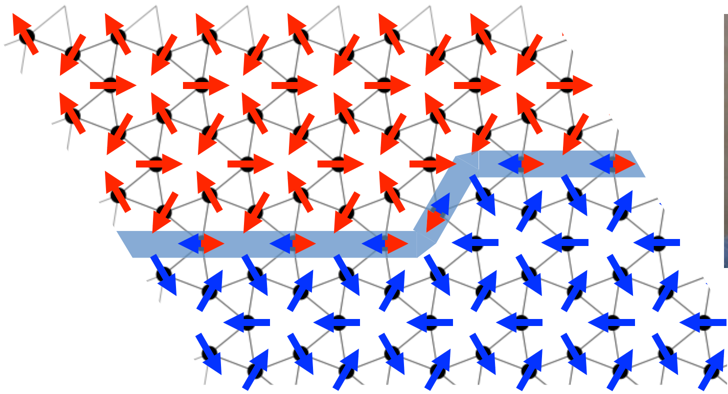
[3]Institut Laue Langevin, Grenoble, France
[4]Univ Glasgow, School of Chemistry, Scotland
[5]Univ Cambridge, Chem Lab, England
[6]Institute of Inorganic Chemistry, Karlsruhe Institute of Technology, Germany





FM Brazil, 2011

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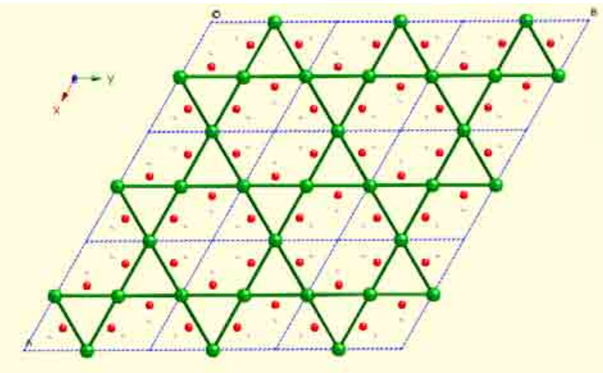
Outline

- **A new family of kagomé compounds**: the quinternary oxalate family
- **Magnetic properties close to T_N**: realization of a "q=0" kagomé antiferromagnet with strong anisotropy
- **Magnetic properties at low temperature**: domain walls dynamics associated to quasi-Ising free spins in the ordered phase
- **Discussion**

Introduction

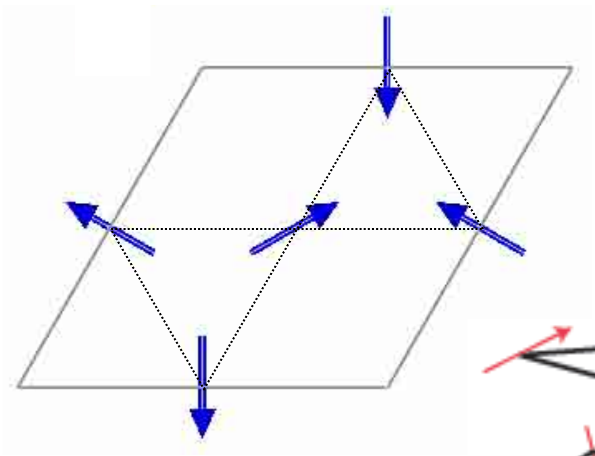
Geometric frustration : highly degenerate ground states with short-range spin-spin correlations, spin liquids and spin ices, or exotic ordered states



Example: Fe jarosites forming a kagomé lattice

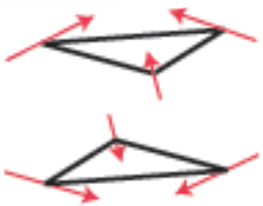
$KFe_3(OH)_6(SO_4)_2$ $S = 5/2$

Antiferromagnetic order: T_N=65 K <<|θ|=800K



"**q=0" umbrella magnetic structure** :
120° moment on each triangle
+ weak out of plane component

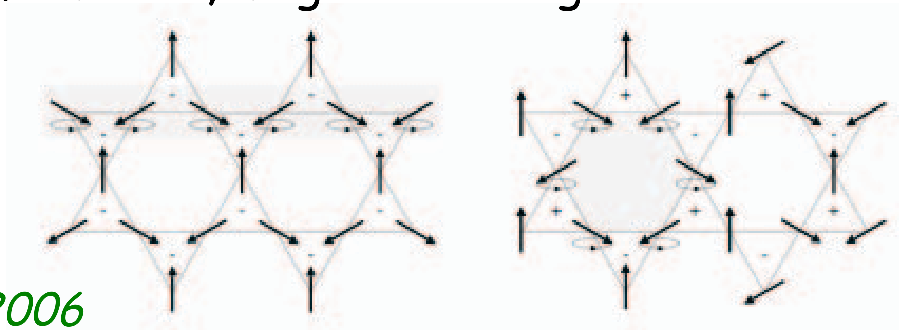
=> Frustration released by 2nd order perturbation:
attributed to **Dzyaloshinsky-Moriya interaction**



Matan et al., PRB 2011

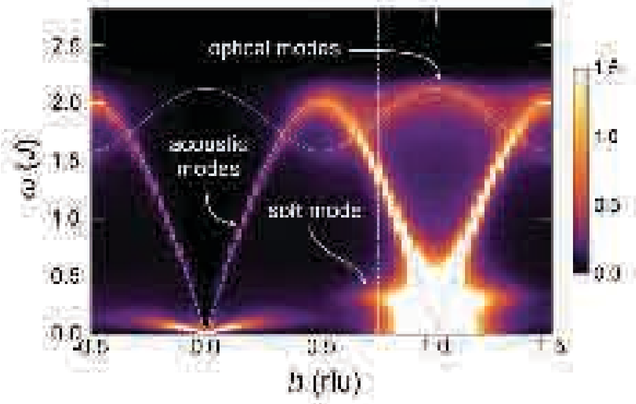
Introduction

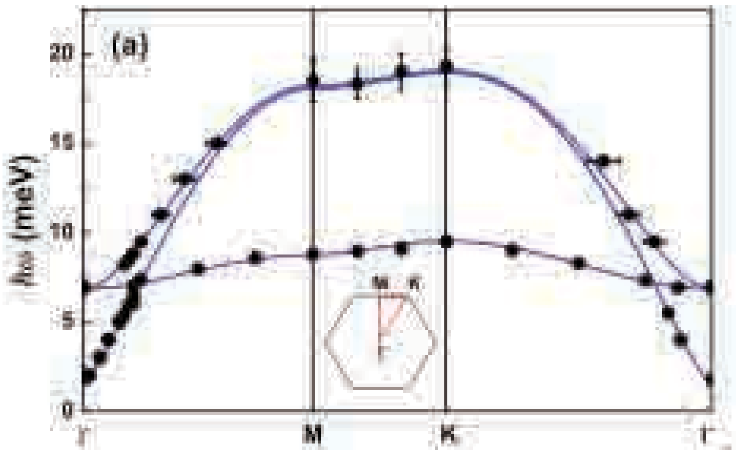
Excitations and dynamics associated to these states:
propagative spin-waves like modes,
weathervane soft modes, magnetic charges



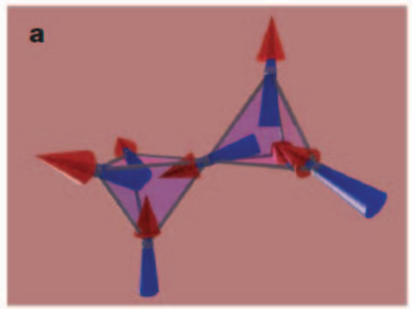
Matan et al. PRL 2006

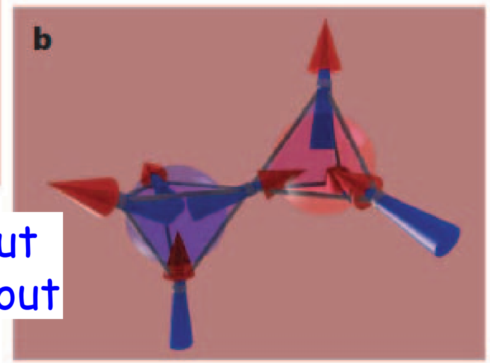
Robert et al., PRL 2008





Castelnovo et al. Nature 2007



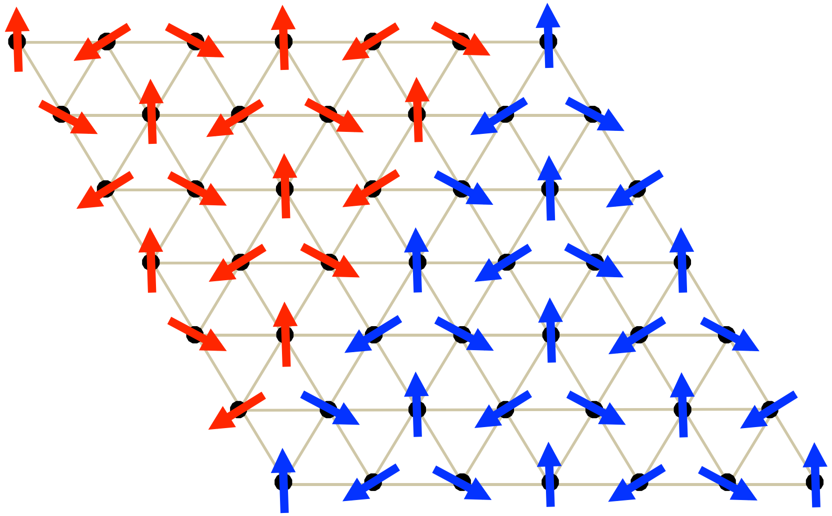


3 spins in, 1 spin out
or 1 spin in, 3 spins out

Introduction

Magnetic domains always present in conventional magnets

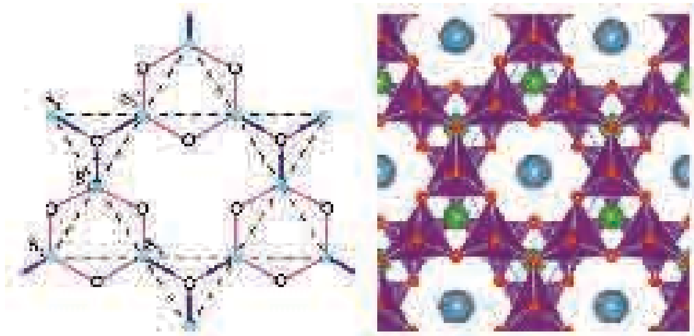
180° antiferromagnetic domains:
all spins reversed



Magnetic domains and frustration

Importance in multiferroics (handling via the ME effect, electronic properties of domain walls etc.)

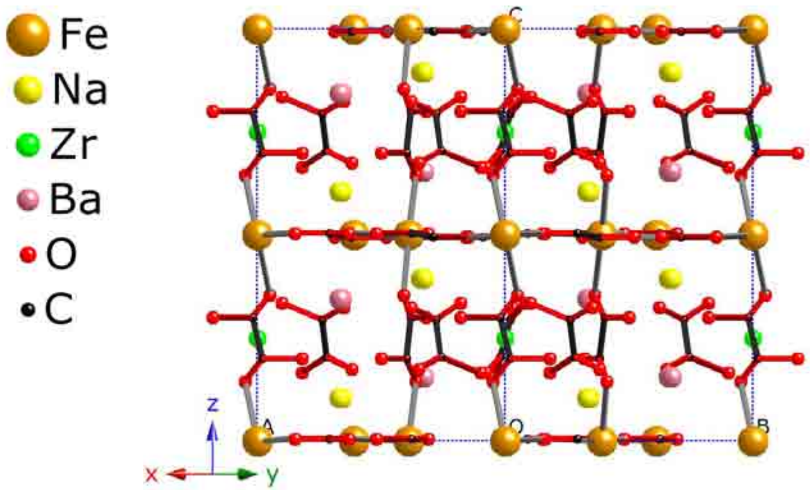
Delaney et al, PRL 2009

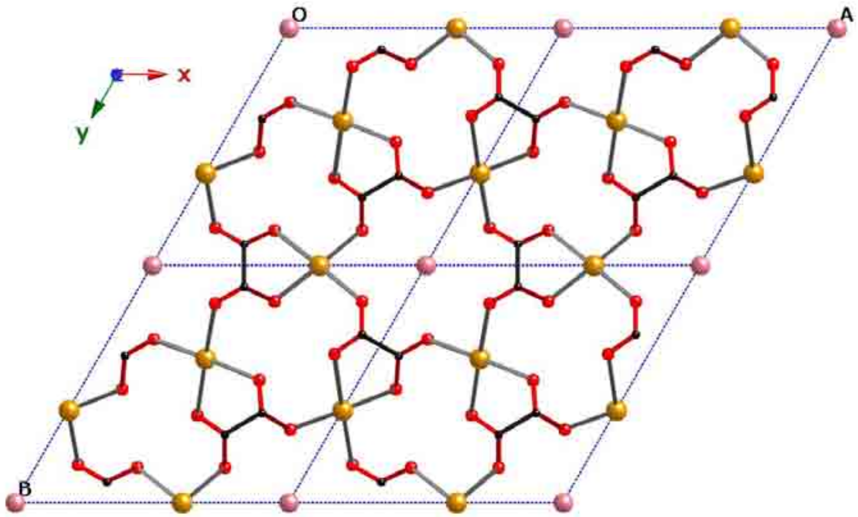


cf. talk of G.J. MacDougall

A new family of kagomé compounds

Fe-oxalate compounds with generic formula:
$Na_2Ba_3M_3(C_2O_4)_6X$ where for instance M = Fe^{2+}
and X = $A^{IV}(C_2O_4)_3$ with A^{IV} = Zr, Sn
or X = $[A^{III}(C_2O_4)_3]_{0.5}[A^{III}(C_2O_4)_2(H_2O)_4]_{0.5}$ with A^{III} = Fe, Al



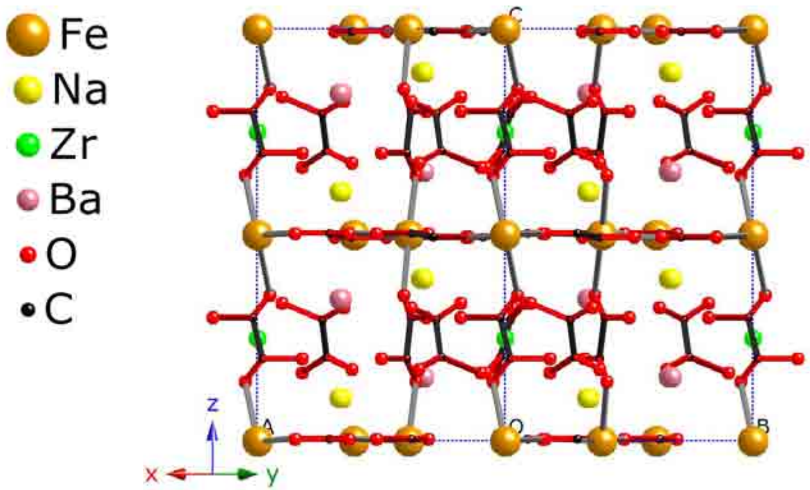


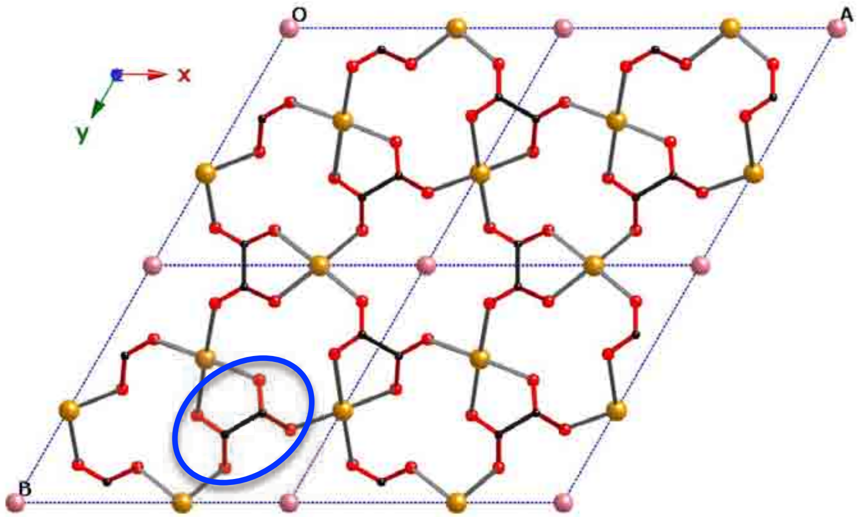
$Na_2Ba_3Fe_3(C_2O_4)_6\ Zr(C_2O_4)_3$

Trigonal non-centrosymmetric space group P321: a=b=10.45 Å, c=7.54 Å

A new family of kagomé compounds

Fe^{2+} bridged by oxalate organic ligands $C_2O_4^{2-}$



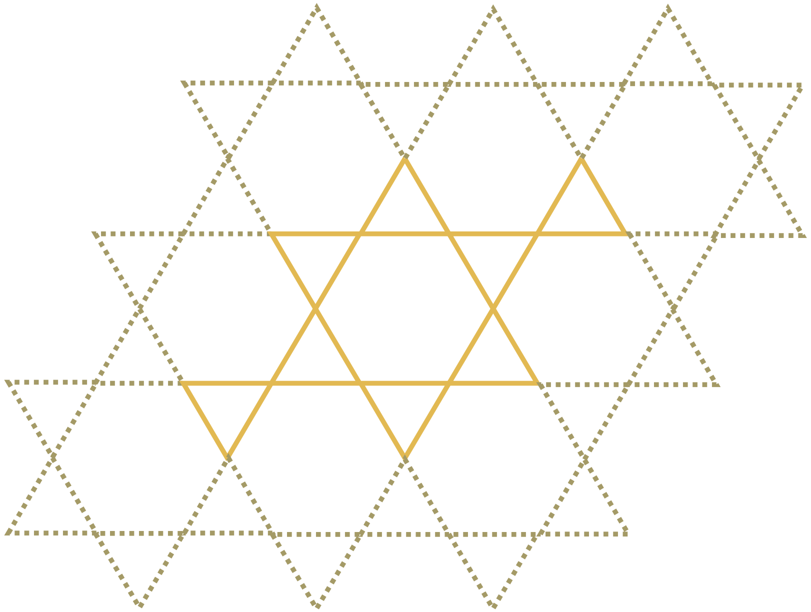


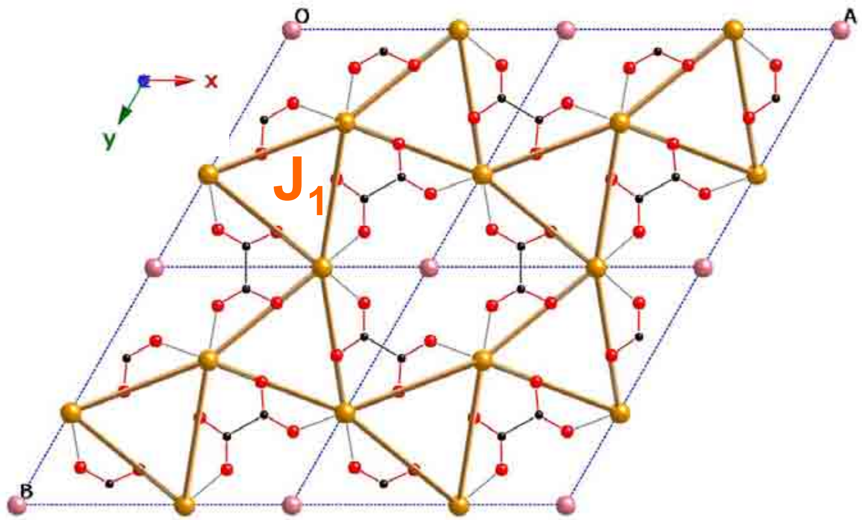
$Na_2Ba_3Fe_3(C_2O_4)_6\ Zr(C_2O_4)_3$

A new family of kagomé compounds

Network of Fe^{2+} in the ab plane:

- **same topology as the kagomé for nearest-neighbor interactions J_1**
- identical in all compounds





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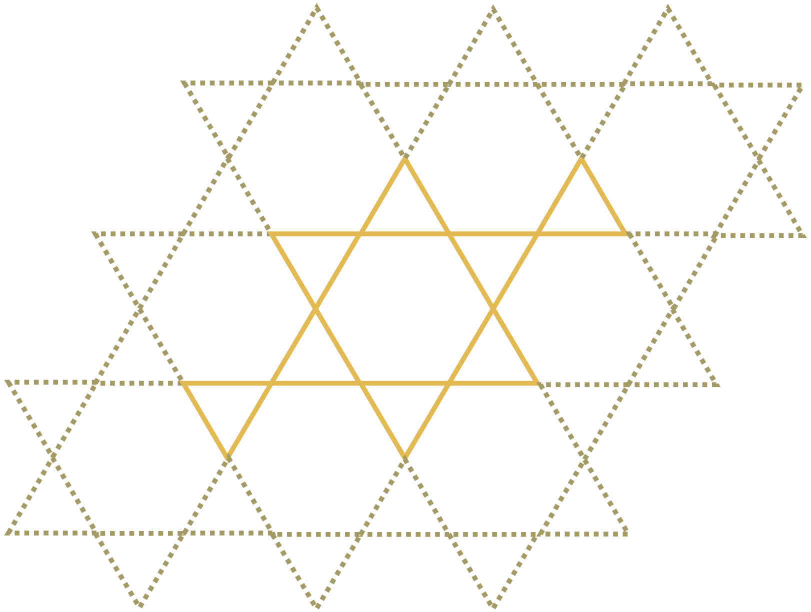
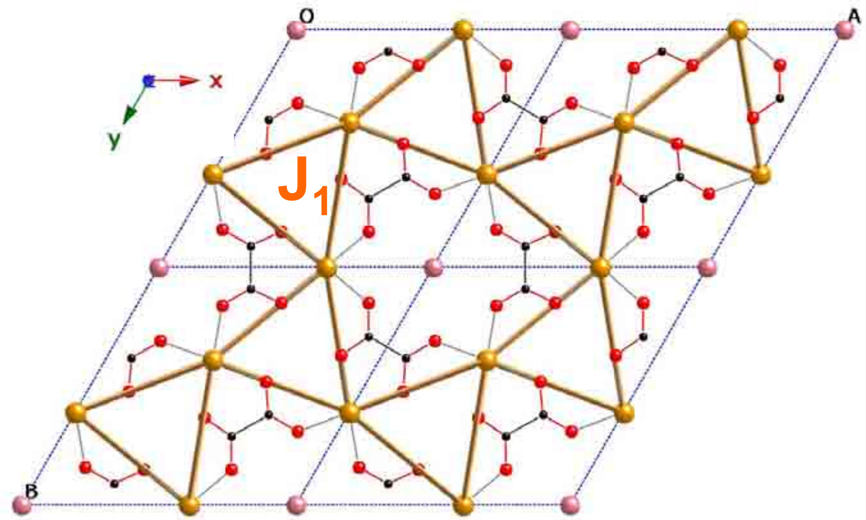
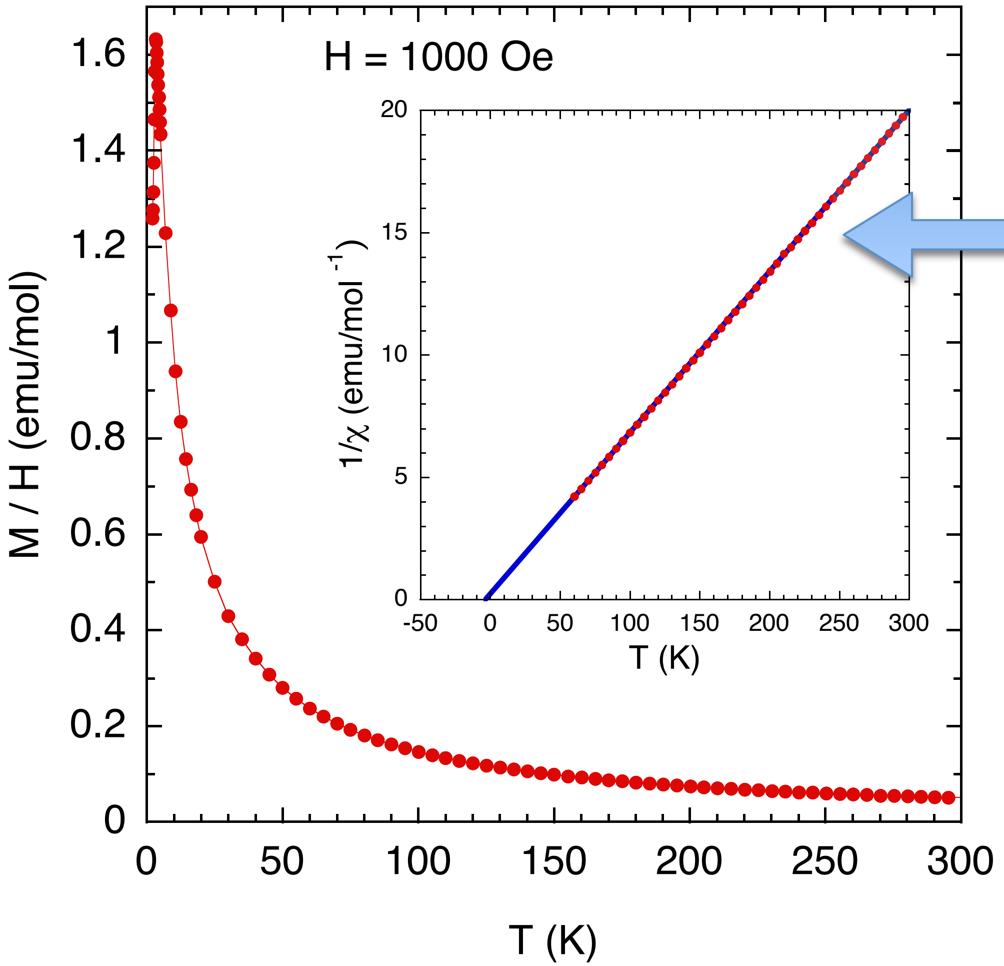




➔Magnetization, DC & AC susceptibility and neutron diffraction down to 70 mK same results on all FeFe-oxalate, FeZr-oxalate, FeSn-oxalate powder samples

Magnetic properties close to T_N

Magnetization measurements



Curie-Weiss model above 50 K

$$\chi = \frac{C}{T - \theta}$$

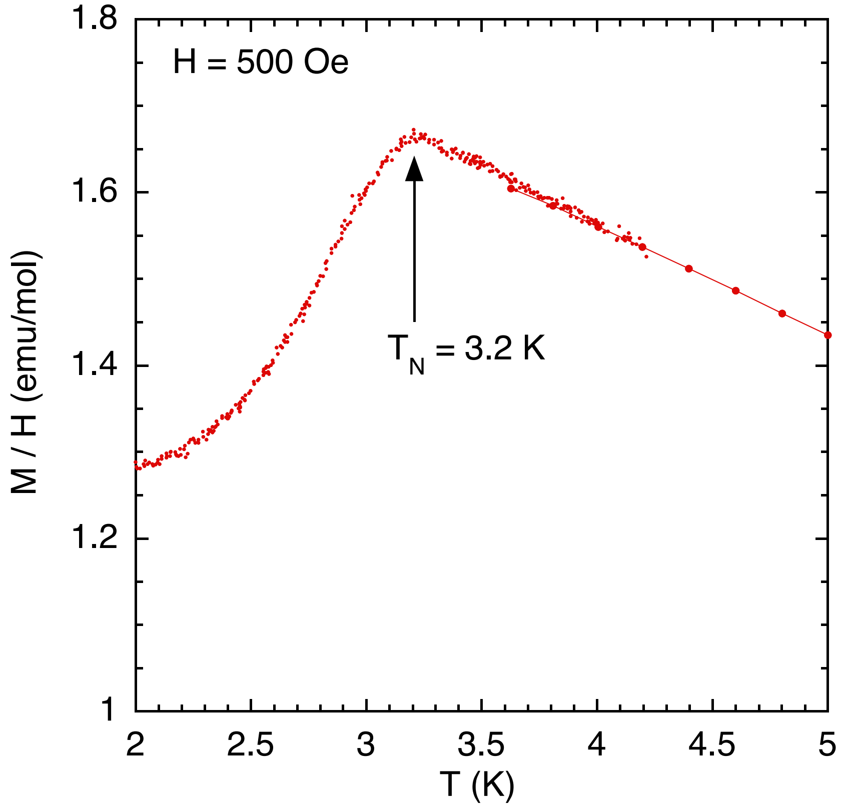
where $C = \frac{N\mu_{Fe}^2}{3k_B}$

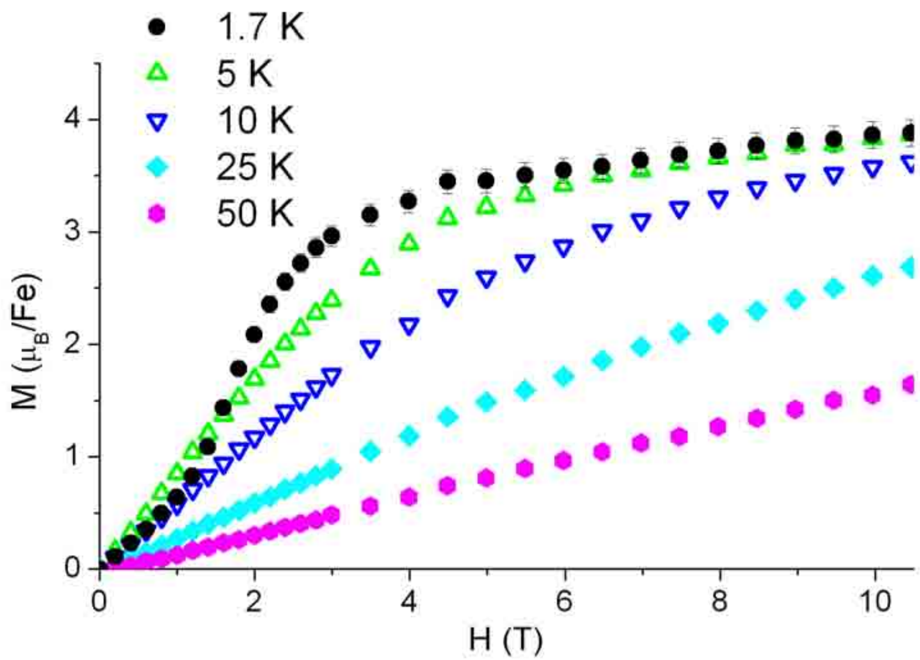
with $\theta \sim$ -5 K and μ_{Fe} = 6.4 μ_B

$\Rightarrow$ **S=2, L=2**

Magnetic properties close to T_N

Magnetization measurements





Below T_N,
metamagnetic process at ~ 1 T

Antiferromagnetic transition
at T_N ~ 3 K in all compounds

➔ θ close to T_N: low degree of frustration

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Magnetic properties close to T_N

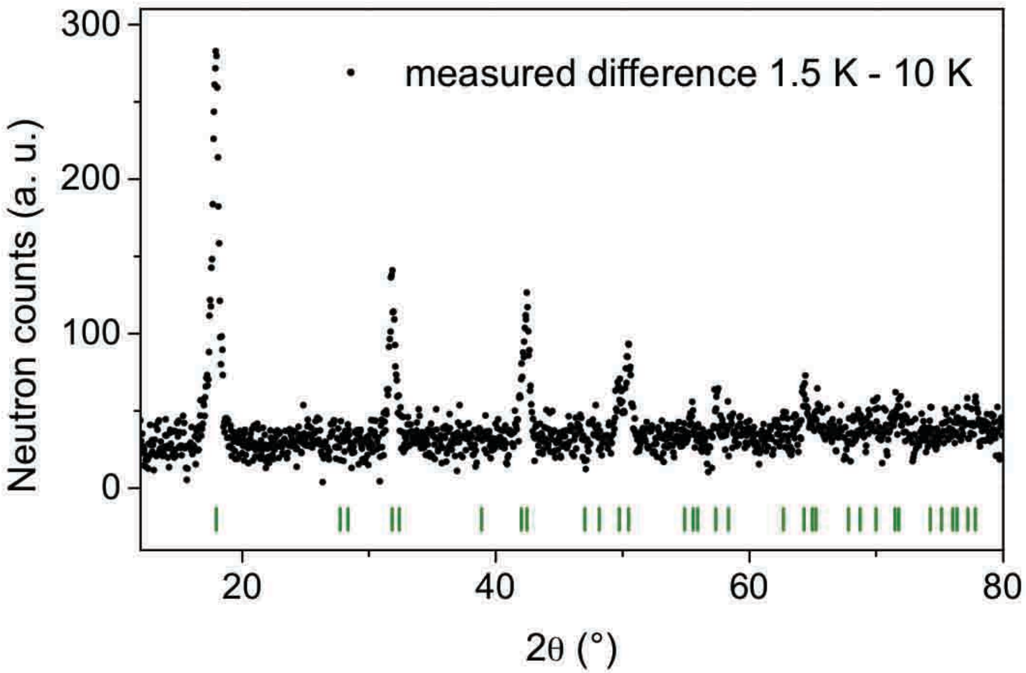
ILL: D20 - D2B, λ =2.4 Å

Powder Neutron Diffraction

Indexation of neutron diffractogram

⇒ propagation vector : k=(0,0,1/2)

⇒ Antiferromagnetic stacking along c

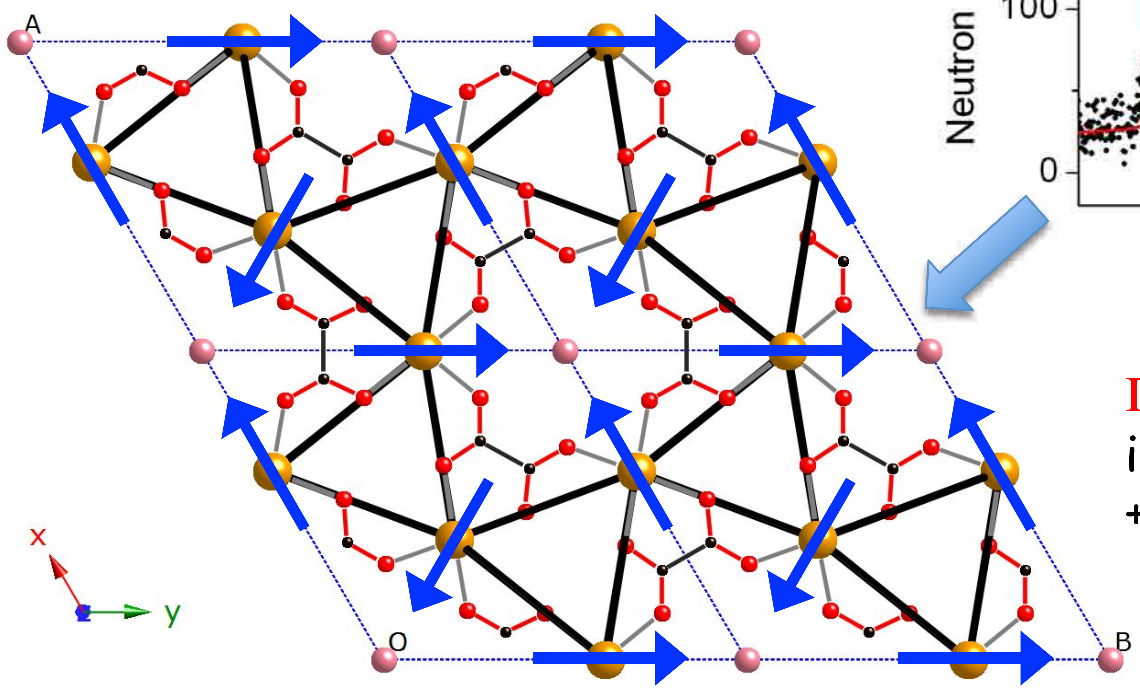


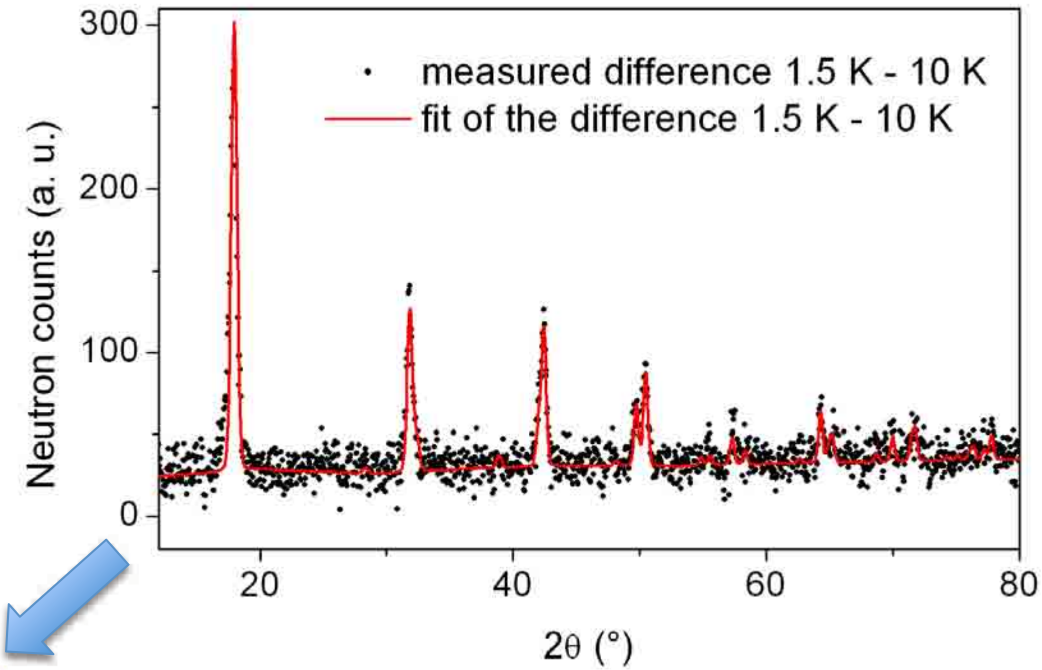
Magnetic properties close to T_N

Powder Neutron Diffraction

ILL: D20 - D2B, λ =2.4 Å

Antiferromagnetic stacking along c
"q=0" arrangement in the ab plane



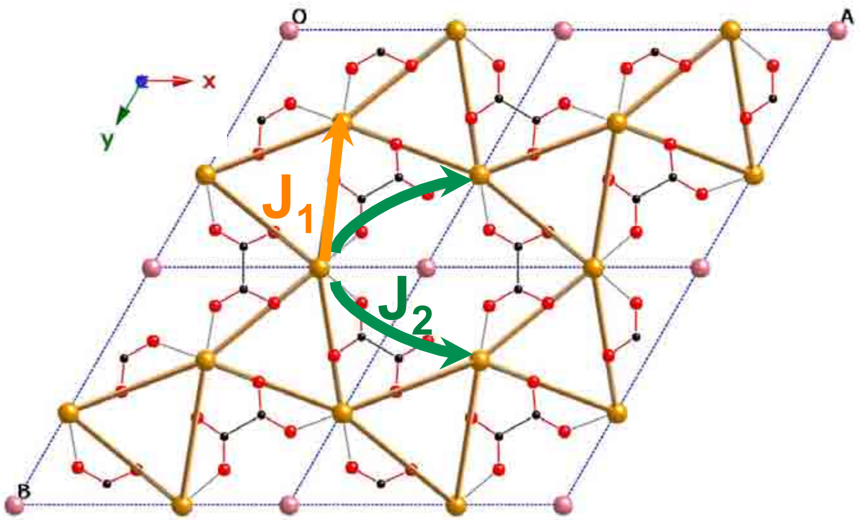


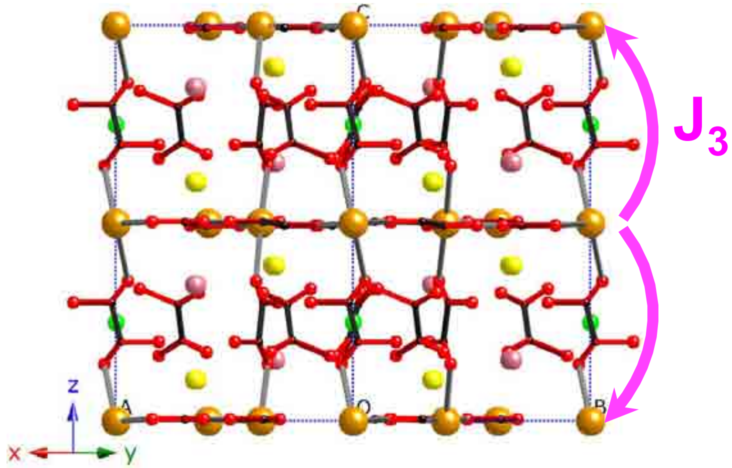
Γ_1: 3 magnetic moments at 120° in the Kagomé plane along a, b, -a-b + chirality

Magnetic properties close to T_N: Interpretation

Antiferromagnetic interactions:
J_1 ~ a few kelvins >> J_2, J_3

3rd inter plane neighbors: J_3
via O-C-C-O bond = 7.54 Å





1st Neighbors: J_1 via O-C-O bond = 5.54 Å
2nd in plane Neighbors: J_2 via O-C-C-O bond = 7.15 Å

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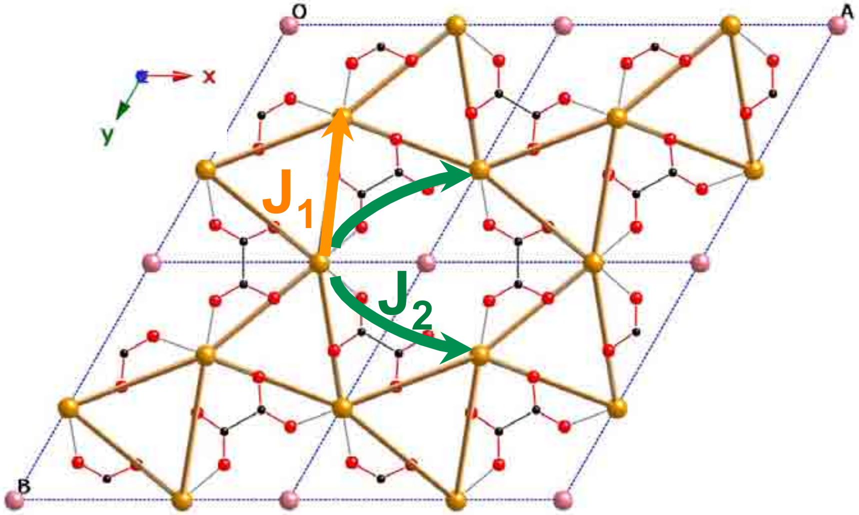

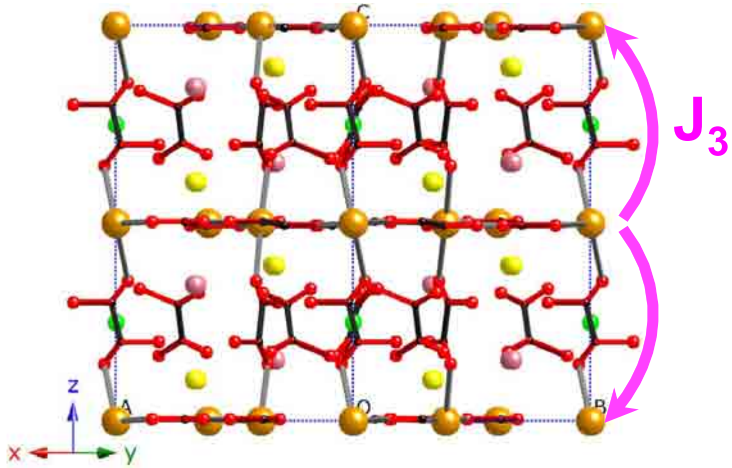

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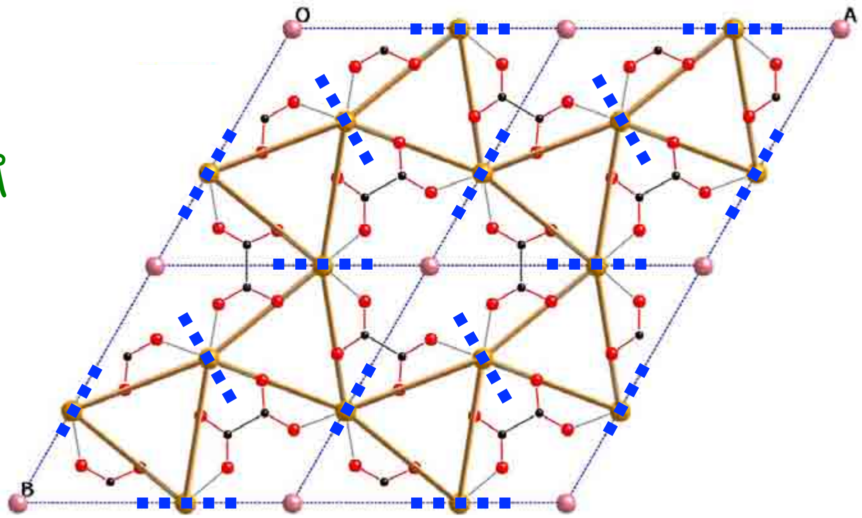
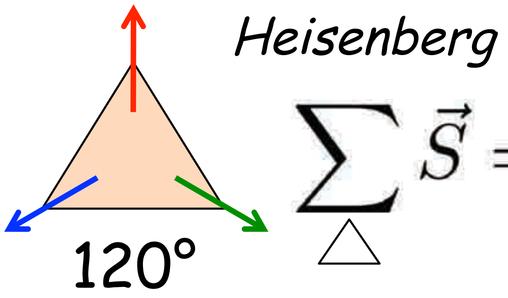


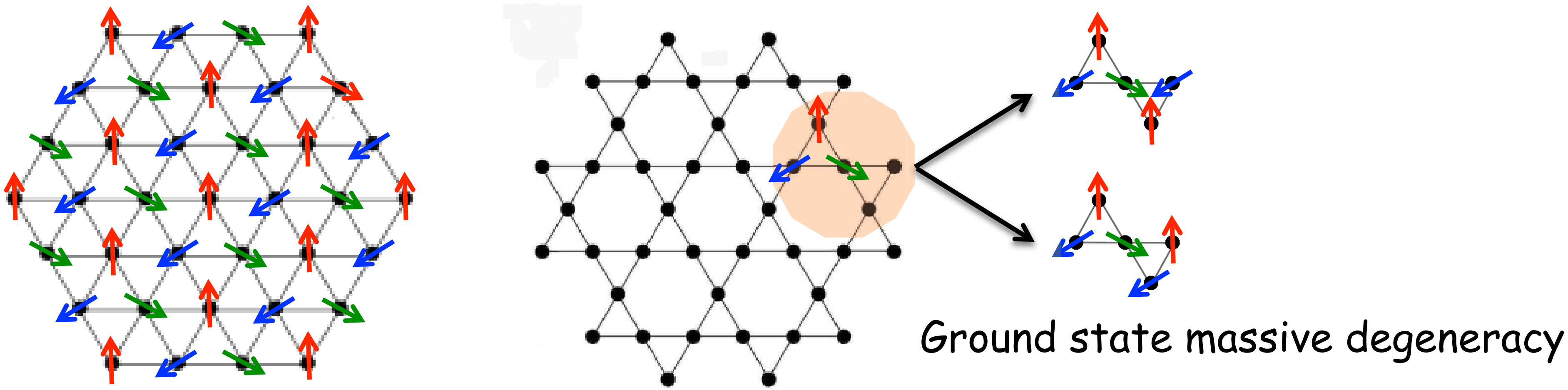
Multiaxial in plane anisotropy:
along the local 2-fold axis
~ 10 kelvins

Magnetic properties close to T_N: Interpretation

2D triangle-based lattices with antiferromagnetic interactions



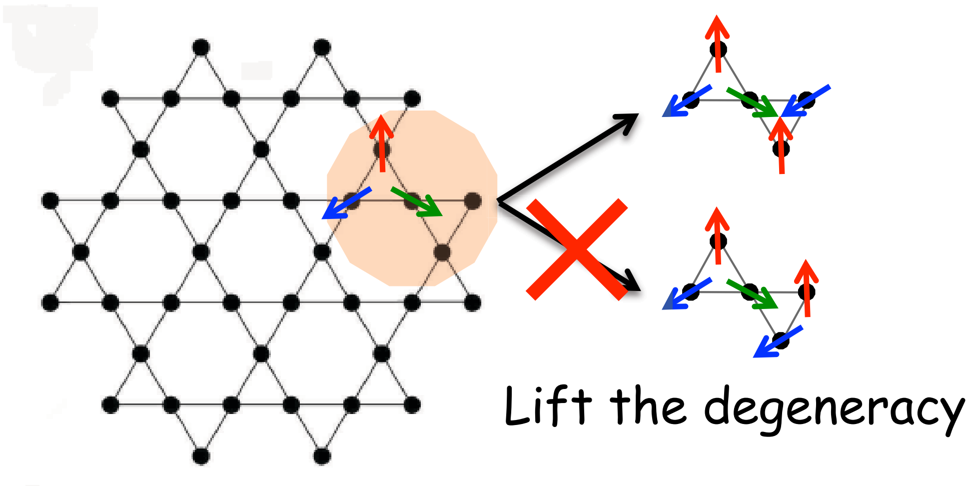
$$\sum_{\triangle} \vec{S} = 0$$



triangular (Néel order) vs **kagomé** (spin liquid)

Magnetic properties close to T_N: Interpretation

Multiaxial anisotropy in the **kagomé** lattice: release of the frustration



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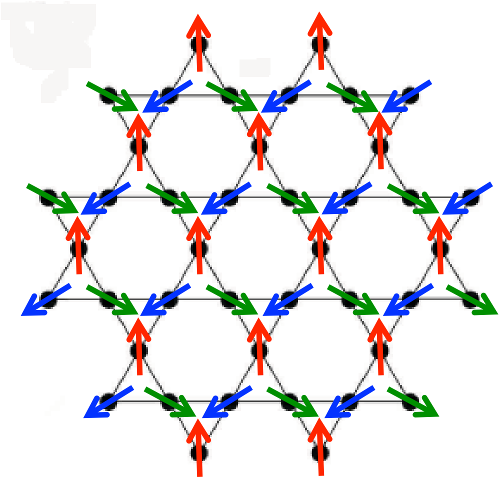
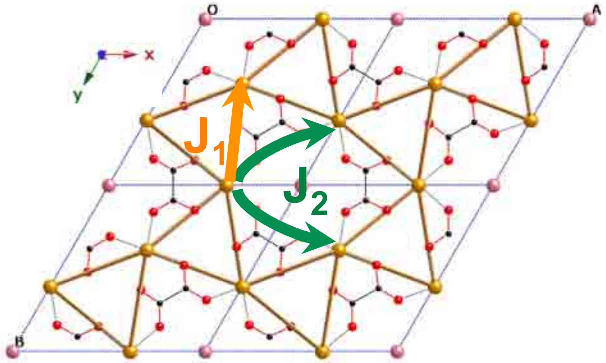
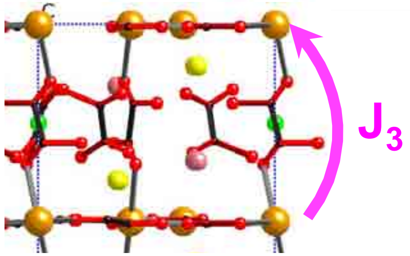
Multiaxial anisotropy in the **kagomé** lattice: release of the frustration



Lift the degeneracy ➔ magnetic order "q=0"

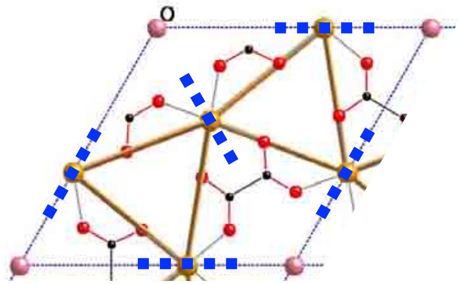
Magnetic properties close to T_N: Interpretation



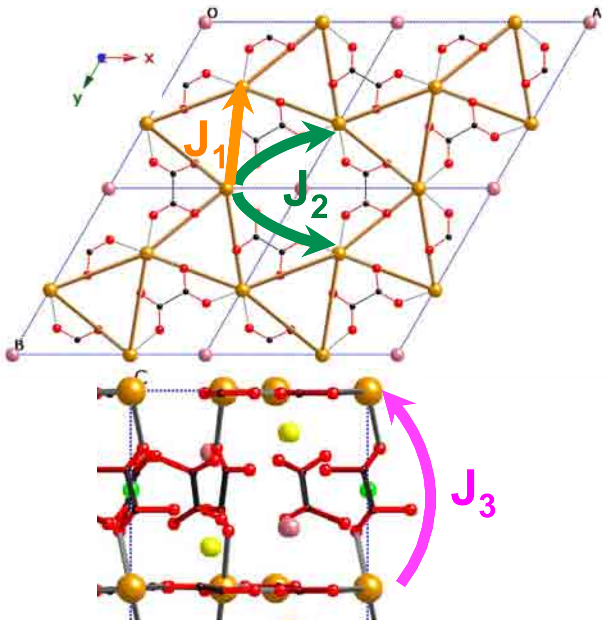


Calculation with the model:

- Antiferromagnetic interactions: $\mathbf{J_1}$ (3 K) and $\mathbf{J_2}$, $\mathbf{J_3}$ (0.3 K)
- Multiaxial anisotropy along the 2-fold axis (10 K)



Magnetic properties close to T_N: Interpretation



Calculation with the model:

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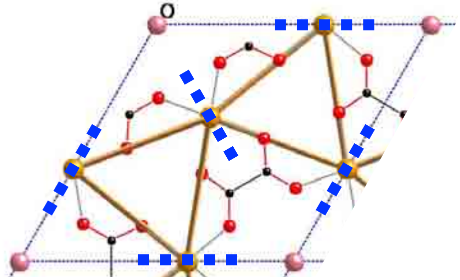
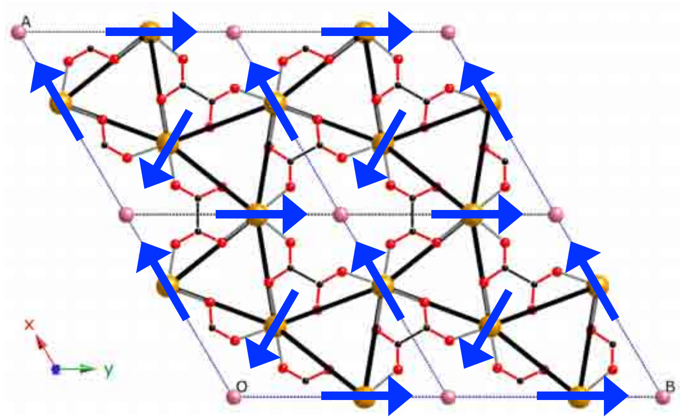
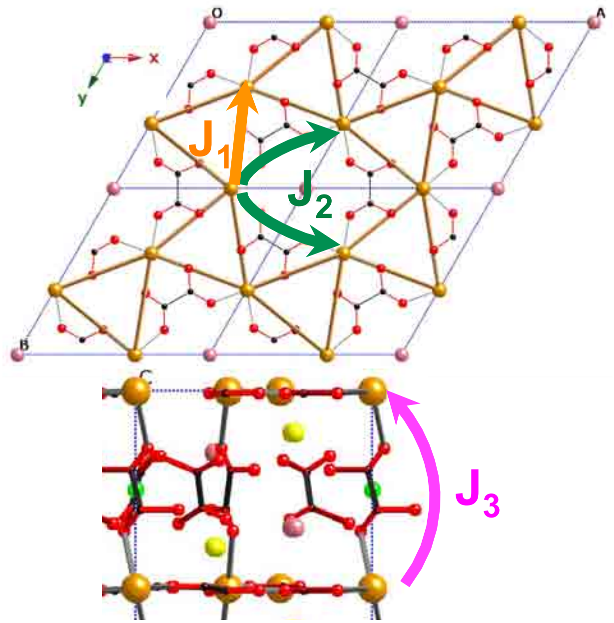


$\Rightarrow$ Explains **magnetic order**



Magnetic properties close to T_N: Interpretation



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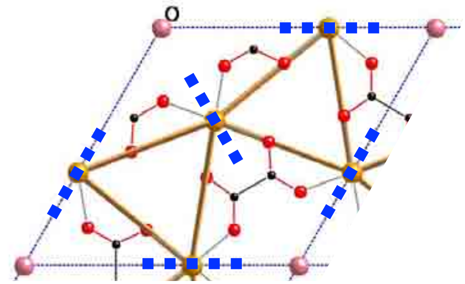
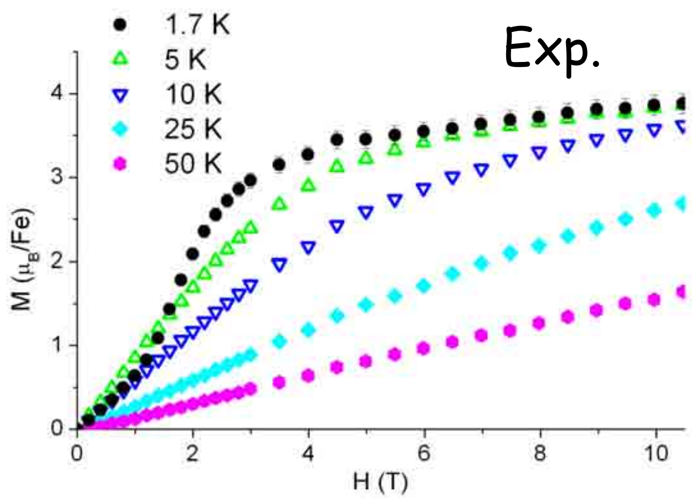
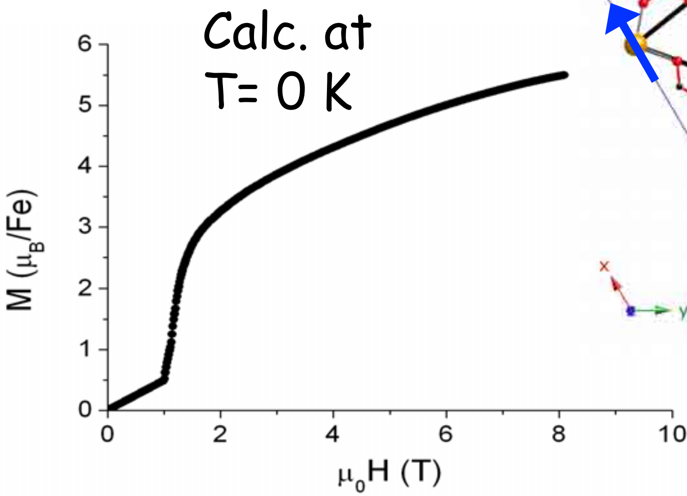
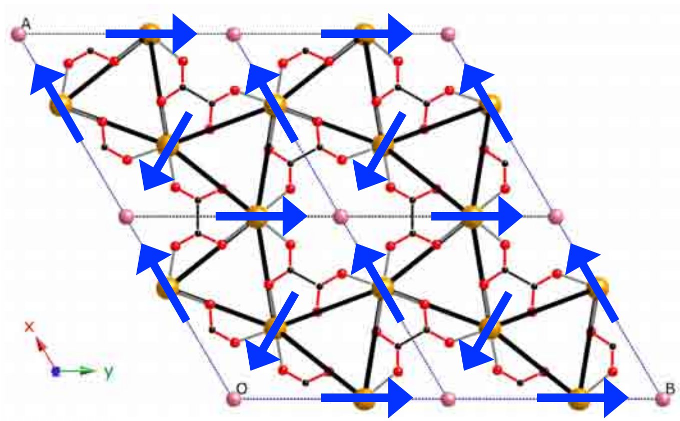


⇒ Explains **magnetic order**
⇒ Reproduces **magnetisation curves**





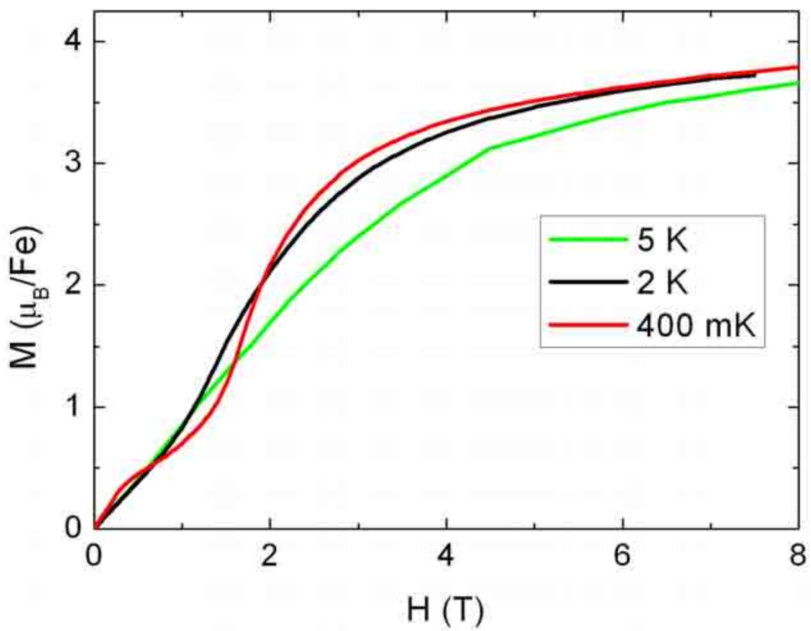


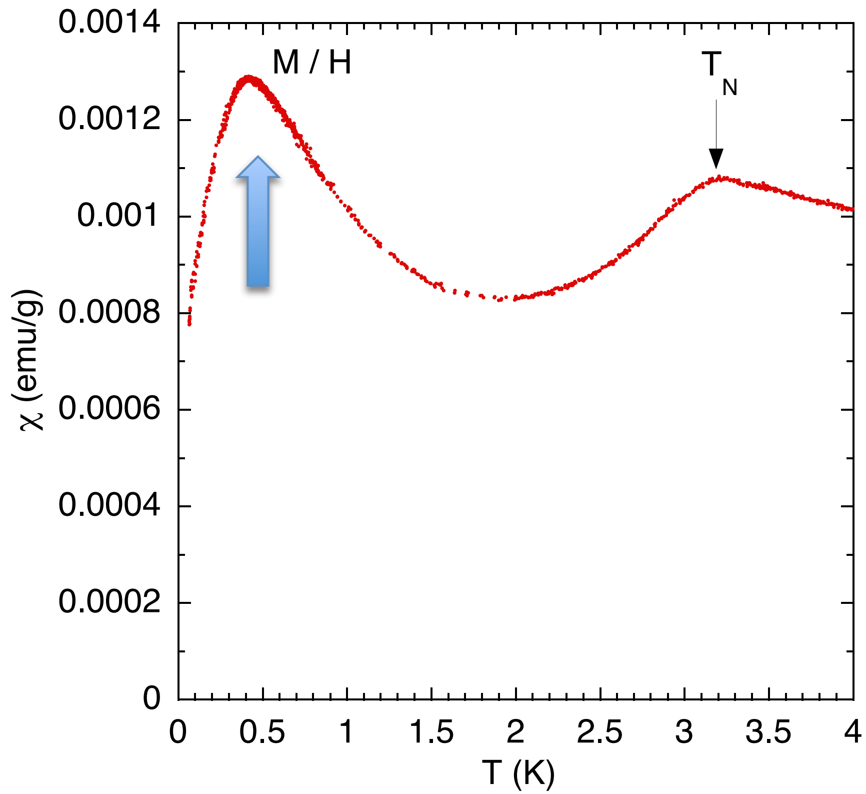
But this is not the end of the story…

Magnetic properties at low T

2nd maximum in the susceptibility at ~ **400 mK** in all compounds

Step-like feature in magnetization curves





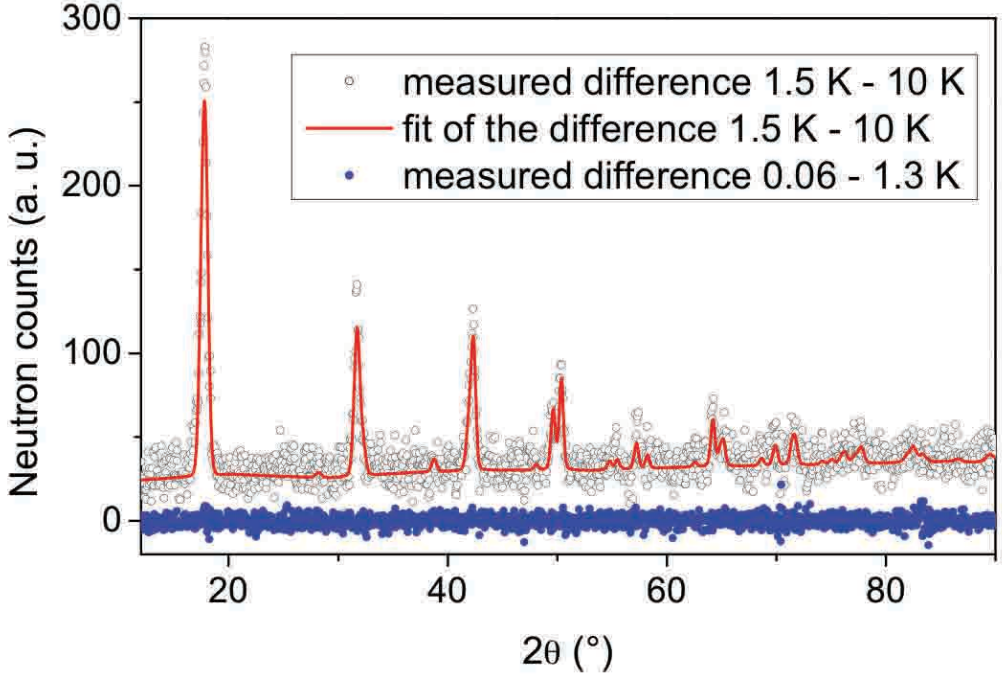
Magnetic properties at low T

2nd maximum in the susceptibility
at ~ **400 mK** in all compounds

Step-like feature in magnetization curves

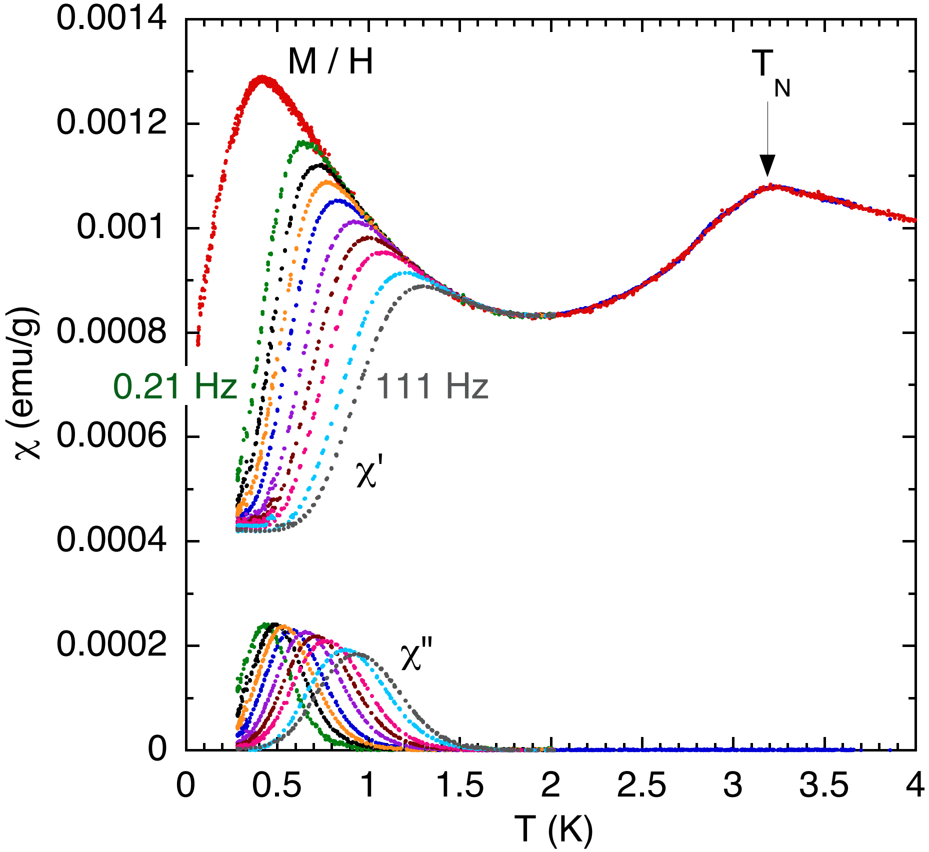
Origin?

Neutron diffraction : no change in the magnetic structure down to 60 mK



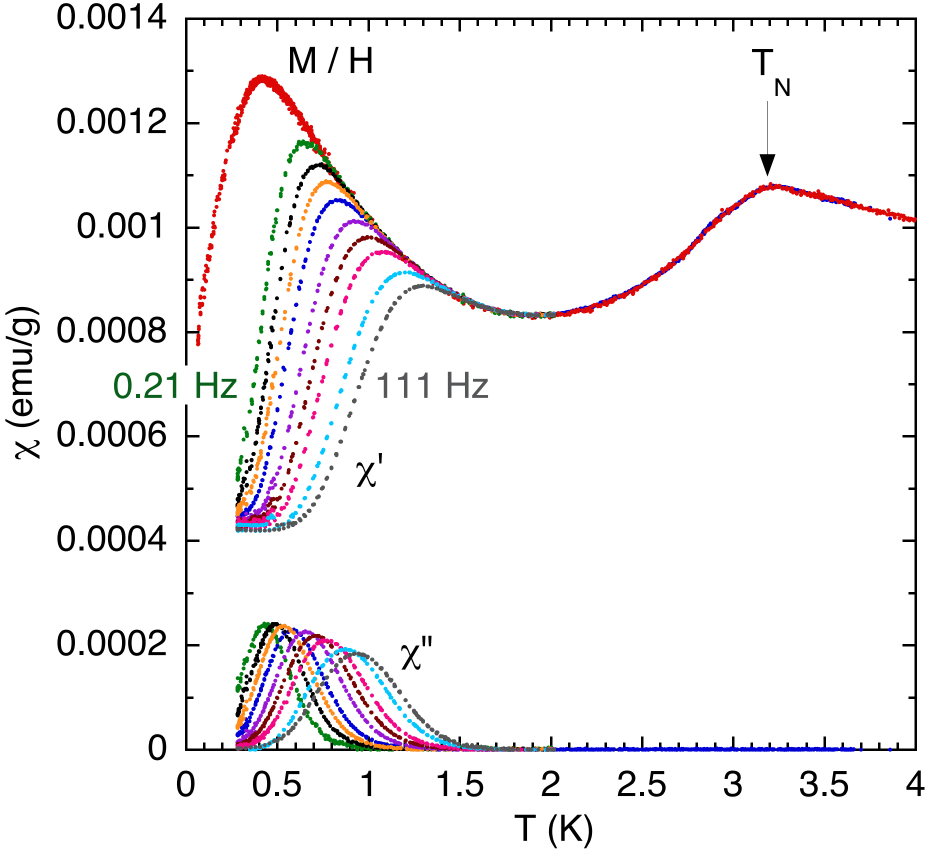
Magnetic properties at low T

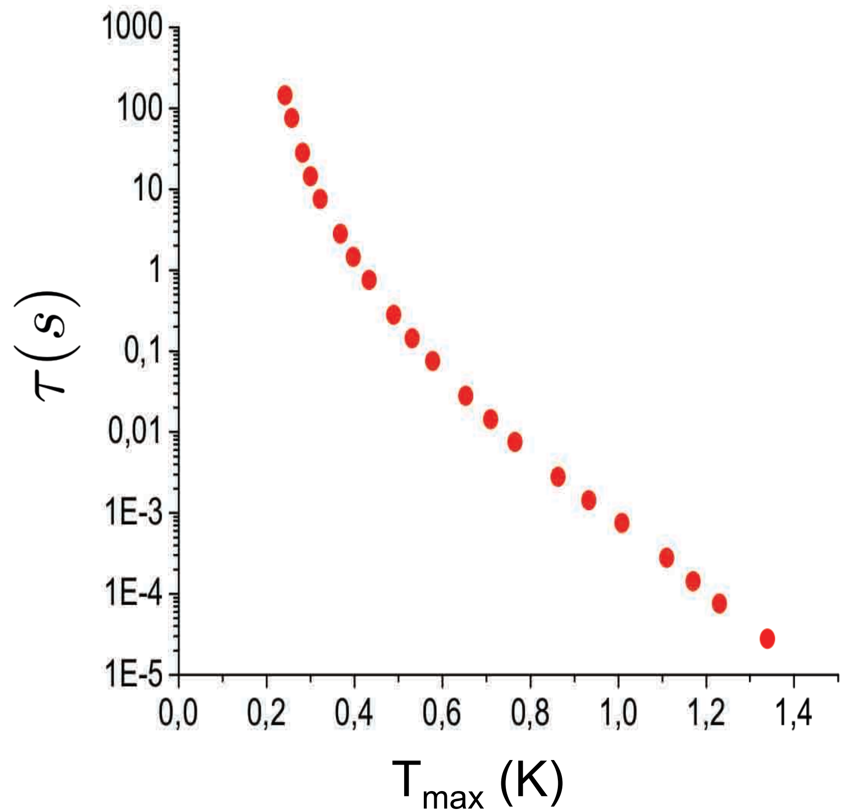
Spin dynamics probed by AC susceptibility down to 60 mK
H=0.5 Oe, f=1.1 mHz – 5.7 kHz



Magnetic properties at low T

Spin dynamics probed by AC susceptibility down to 70 mK
H=0.5 Oe, f=1.1 mHz – 5.7 kHz

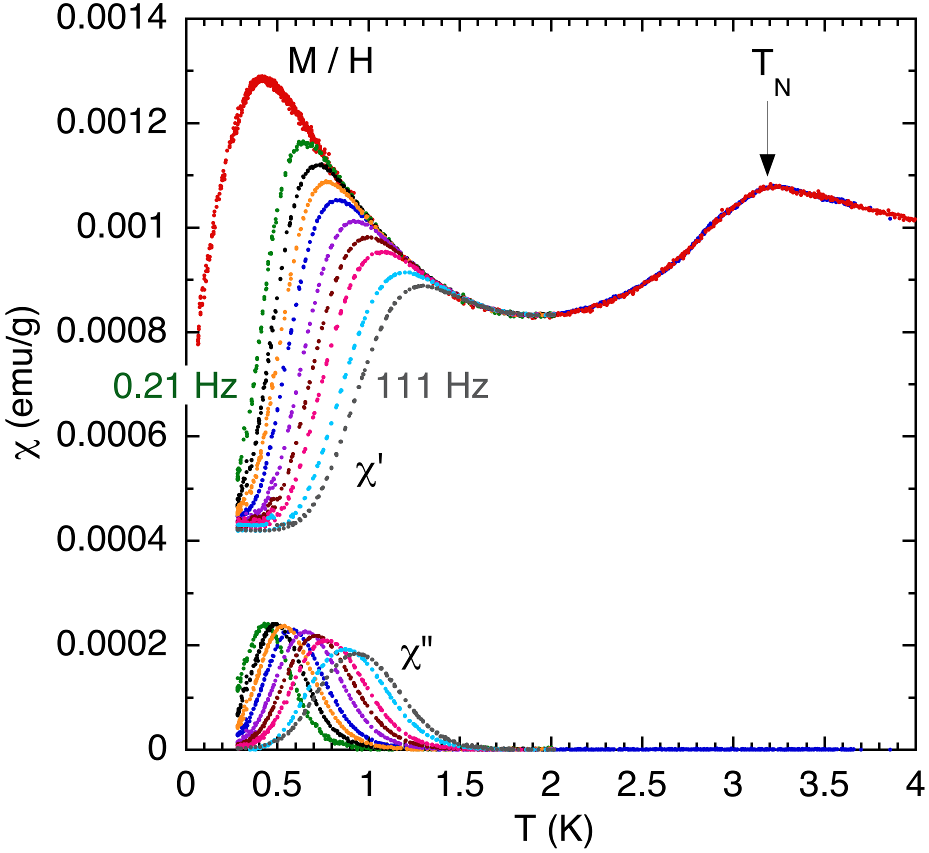


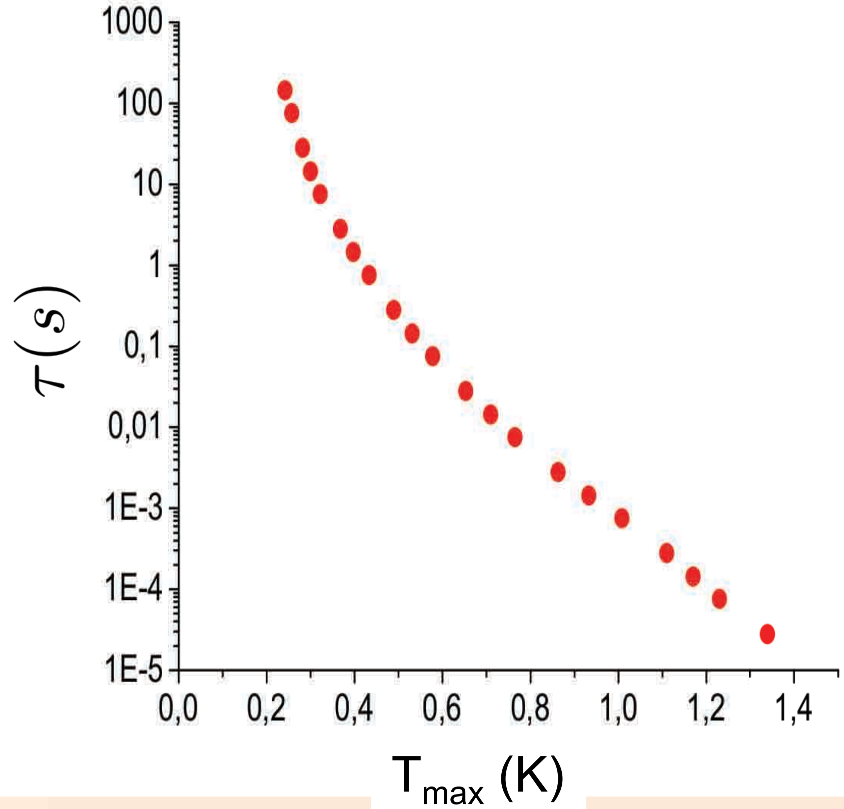


Assumption:
Process with a single relaxation time τ
$\Rightarrow \chi''(T)$ maximum when measurement time ($\sim 1/f$) equal to τ

Magnetic properties at low T

Spin dynamics probed by AC susceptibility down to 70 mK
H=0.5 Oe, f=1.1 mHz – 5.7 kHz





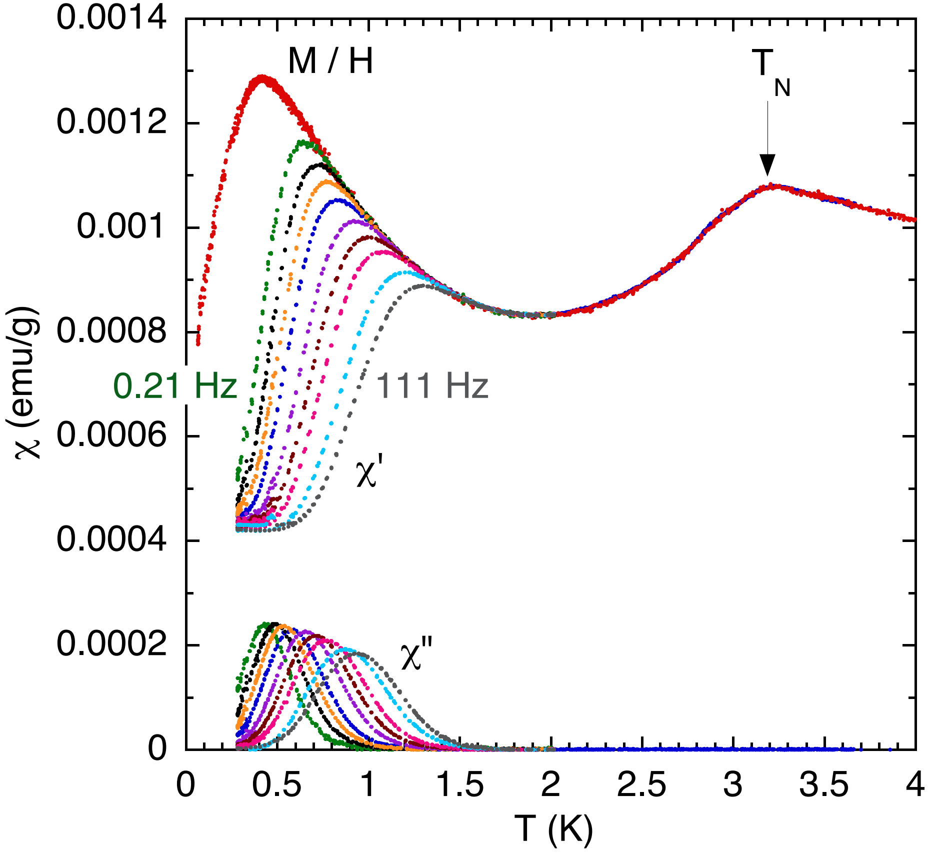
Thermally activated process: Arrhenius law

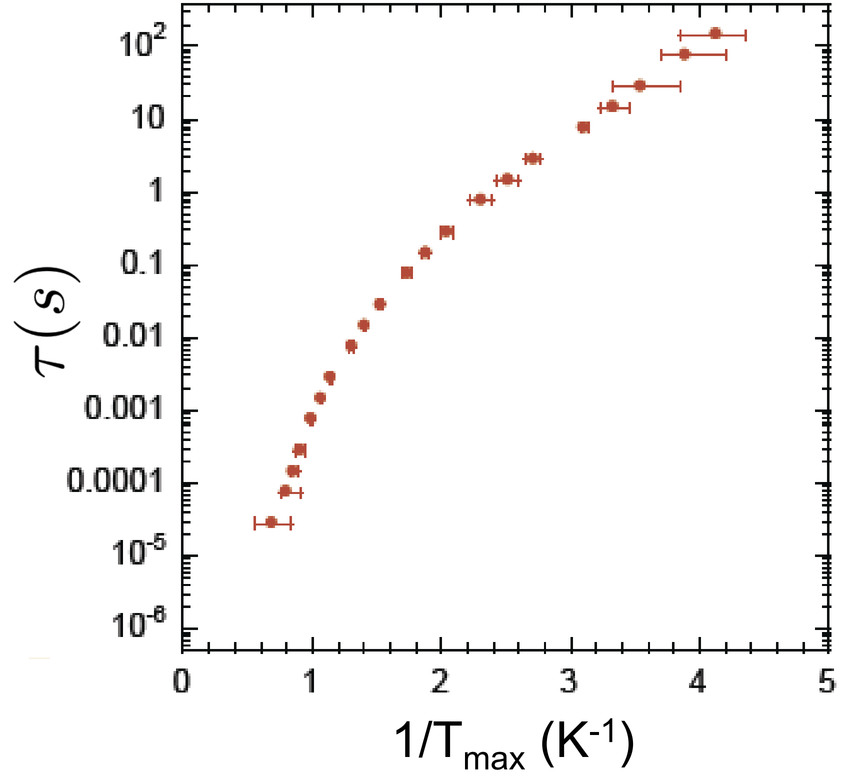
$$\tau = \tau_0 \exp(E/k_B T)$$

τ_0 : characteristic relaxation time
E : Energy barrier

Magnetic properties at low T

Spin dynamics probed by AC susceptibility down to 70 mK
H=0.5 Oe, f=1.1 mHz – 5.7 kHz





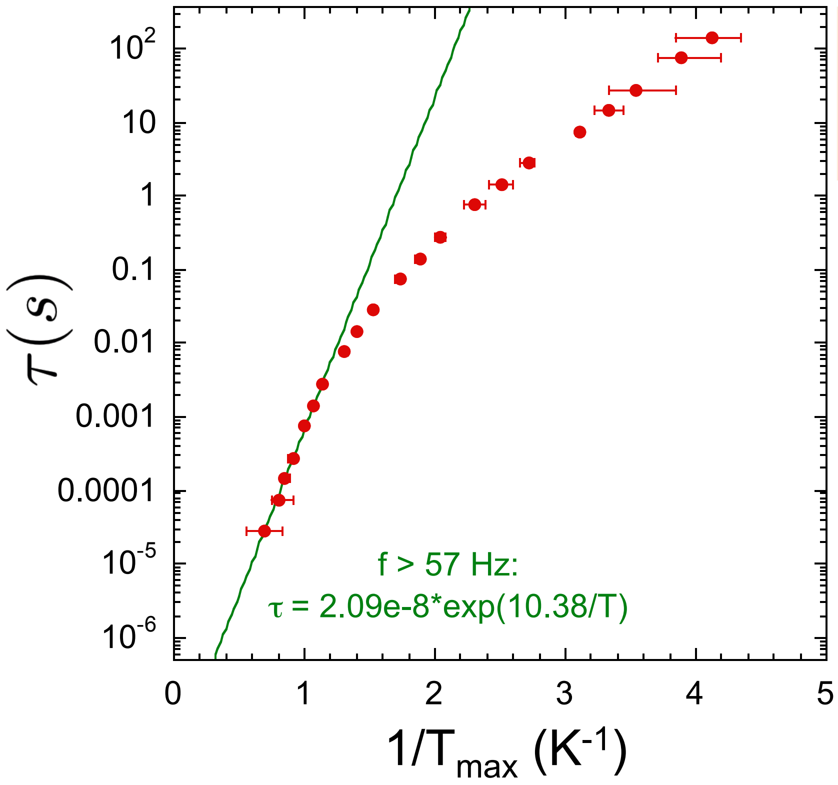
Thermally activated process: Arrhenius law

$$\tau = \tau_0 \exp(E/k_B T)$$

τ_0 : characteristic relaxation time
E : Energy barrier

Two dynamical regimes

Magnetic properties at low T



"Fast" high T regime : τ_0 =2x10^{-8} s, E =10 K

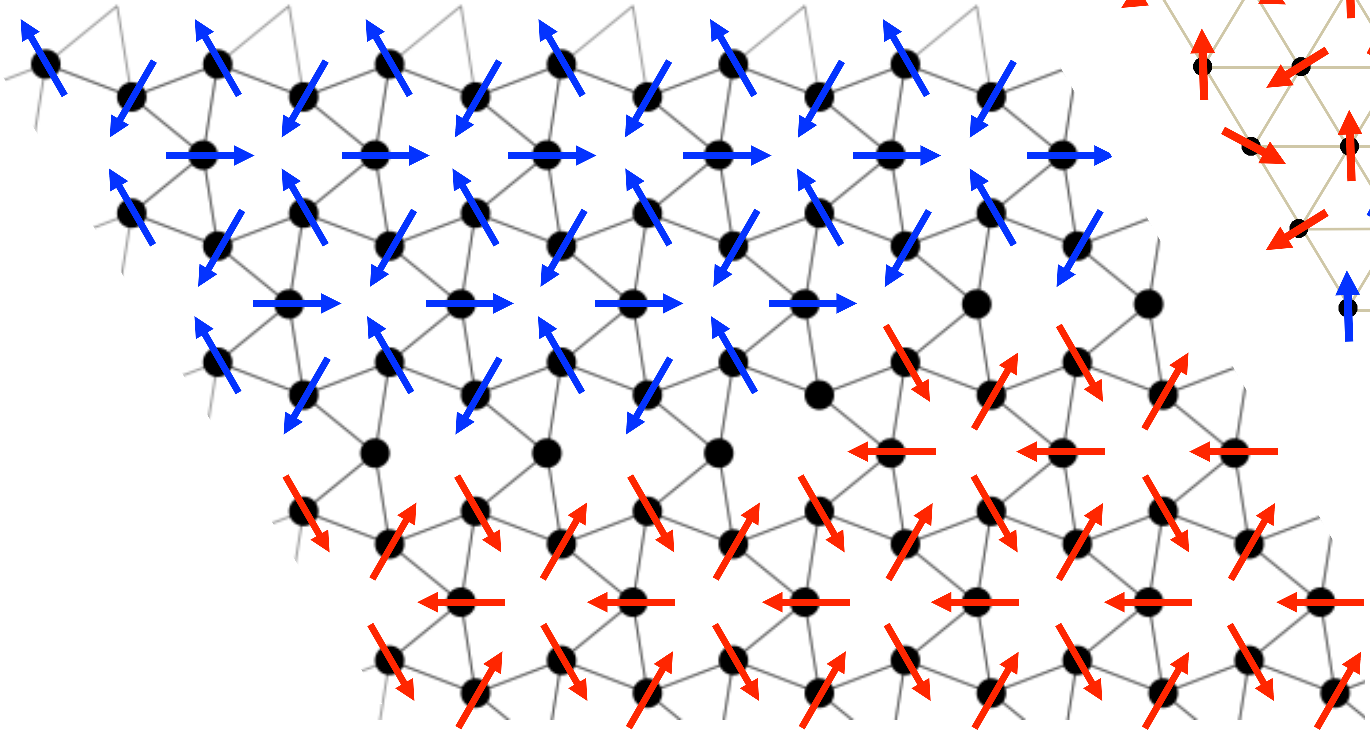
single spin-flips above an anisotropy barrier

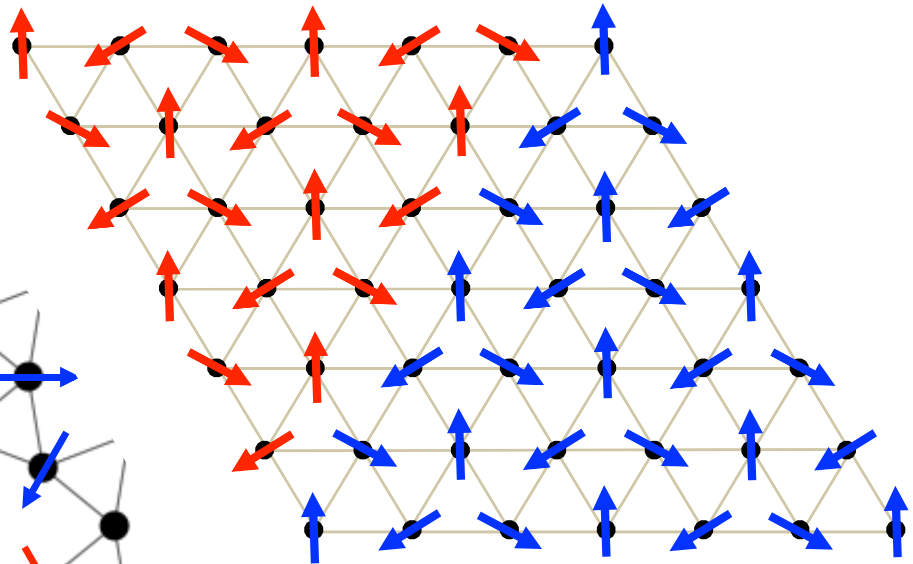
How single spin-flips can occur
in an ordered antiferromagnet ?

Magnetic properties at low T

180° antiferromagnetic domains: all spins reversed
Strong anisotropy: single atomic distance width of domain walls

Kagomé Fe-oxalate



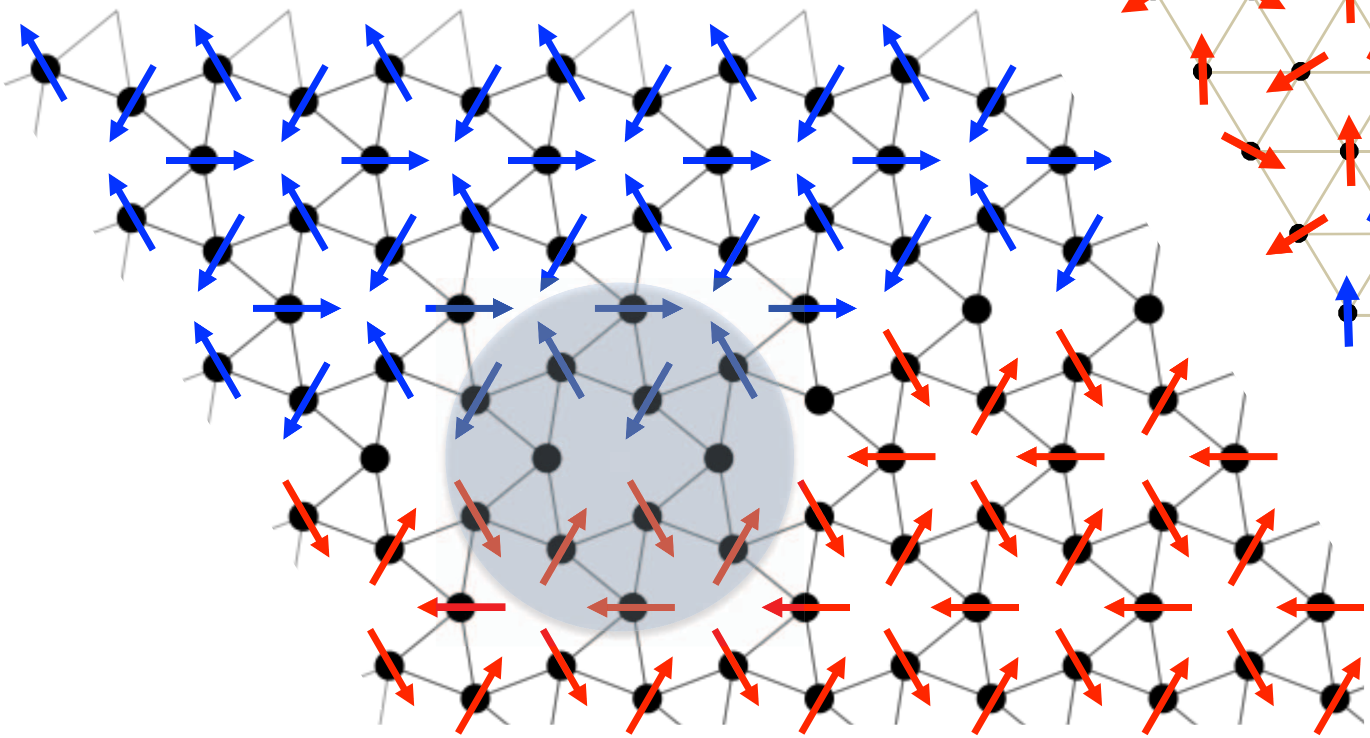


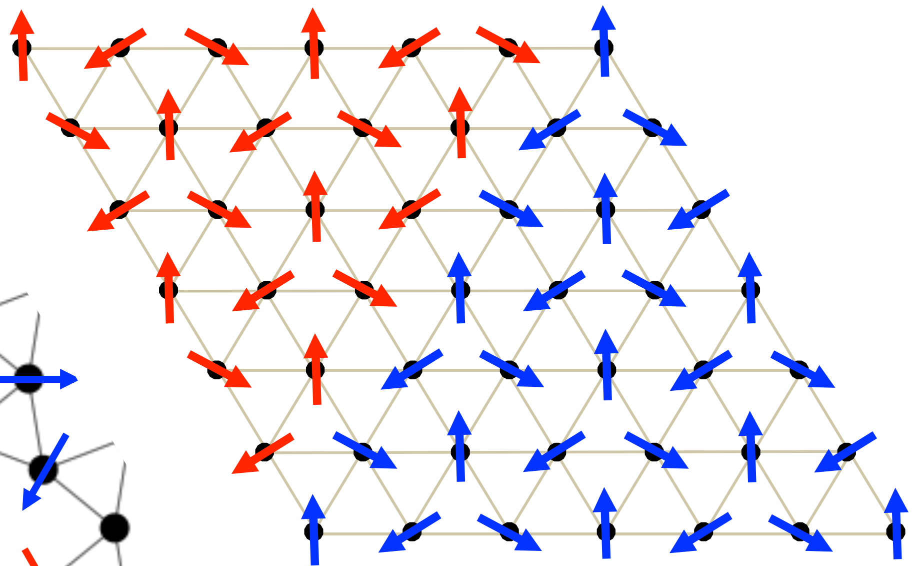
Triangular lattice

Magnetic properties at low T

180° antiferromagnetic domains: all spins reversed
Strong anisotropy: single atomic distance width of domain walls

Kagomé Fe-oxalate



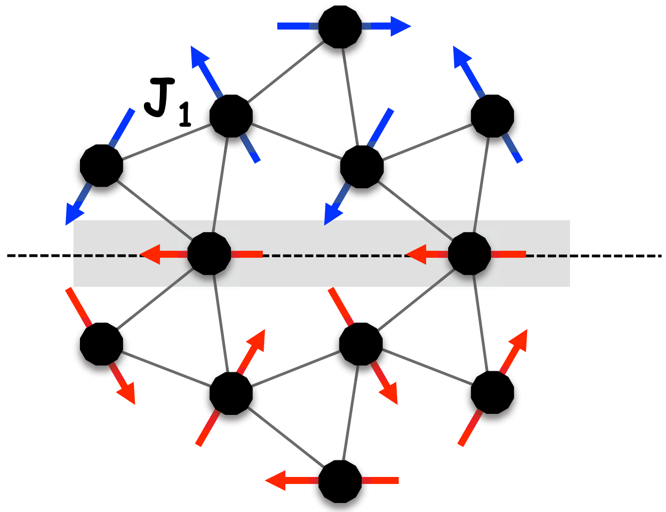


Triangular lattice

Magnetic properties at low T

Low kagomé lattice connectivity:

- Independent spins, blind to their neighbors, along the domain wall
- Same J_1 exchange energy cost in both orientations



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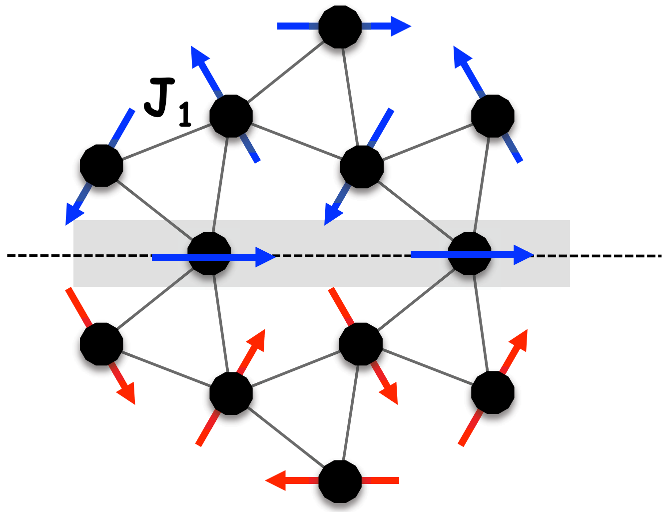

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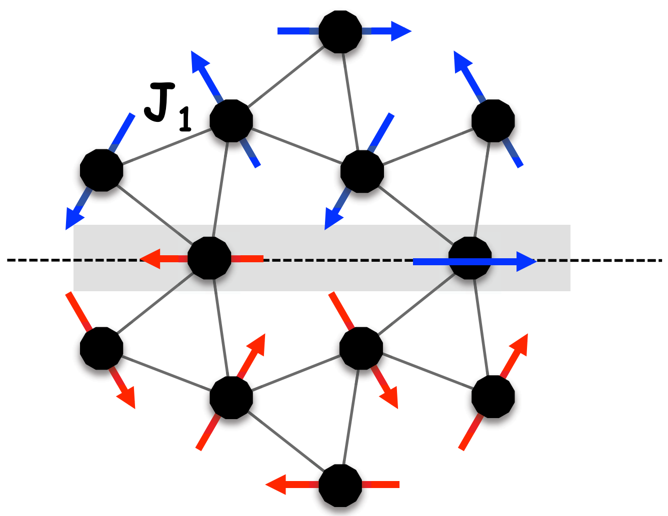

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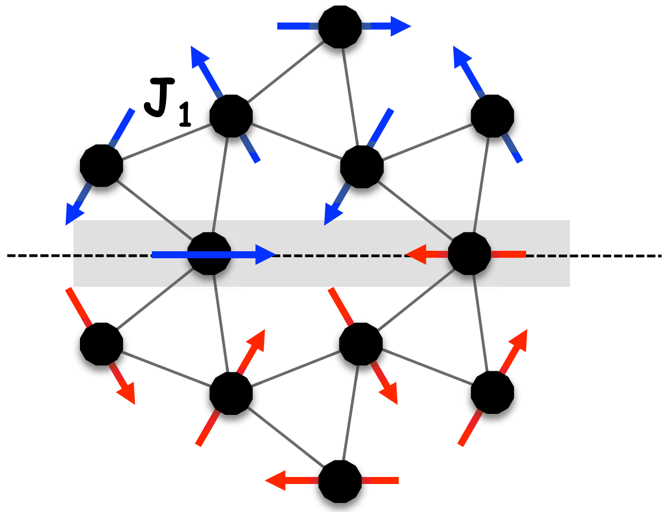
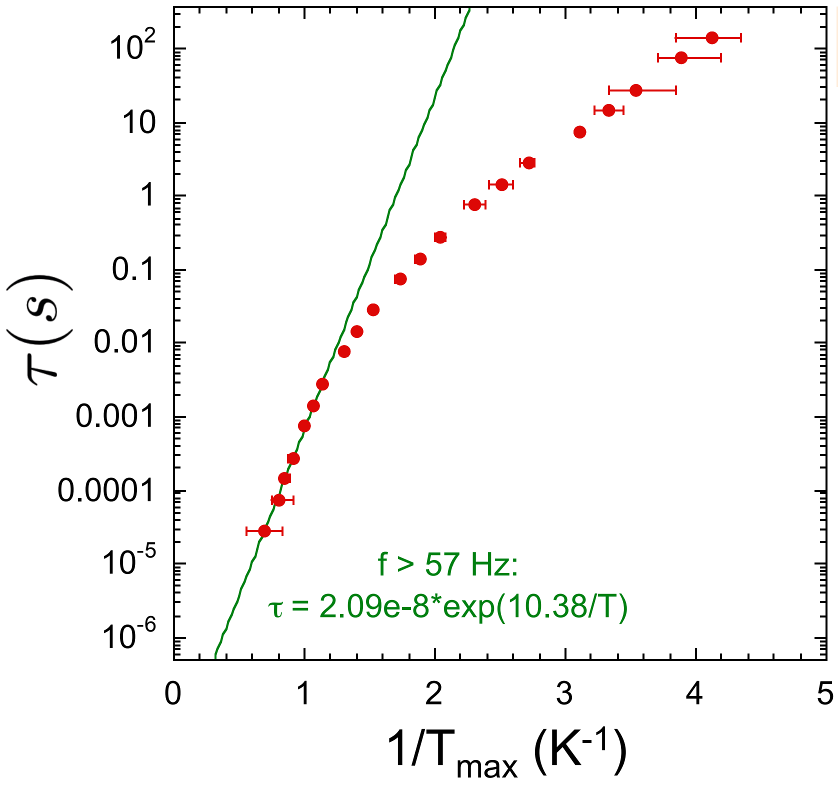


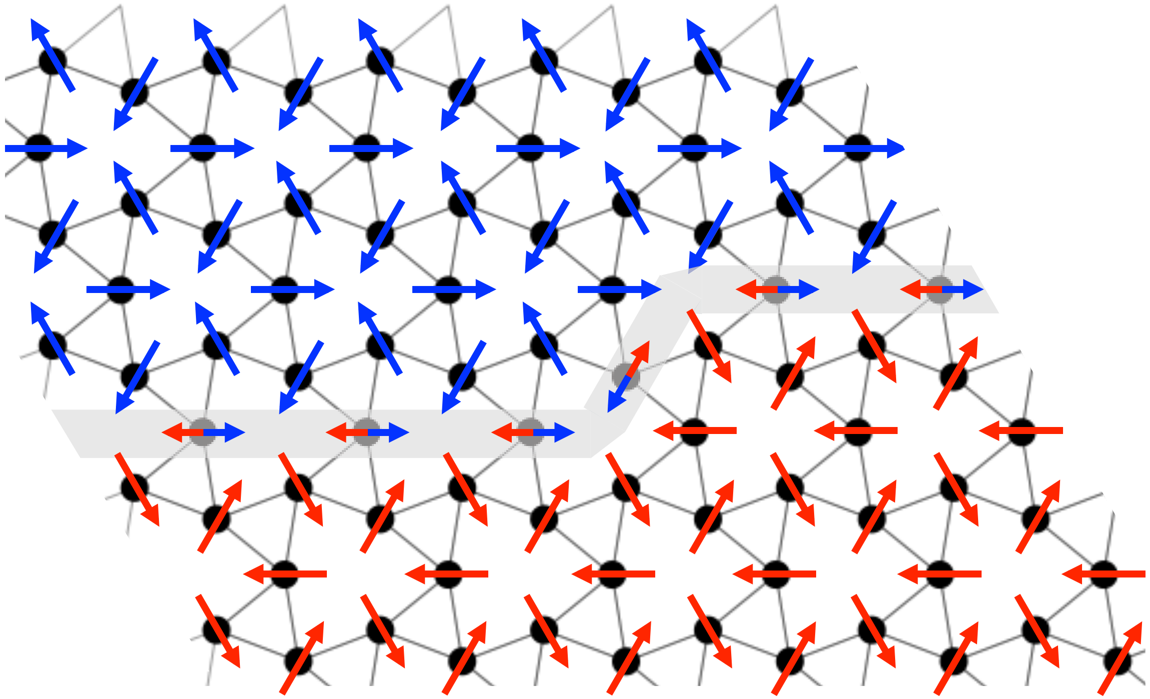
Magnetic properties at low T



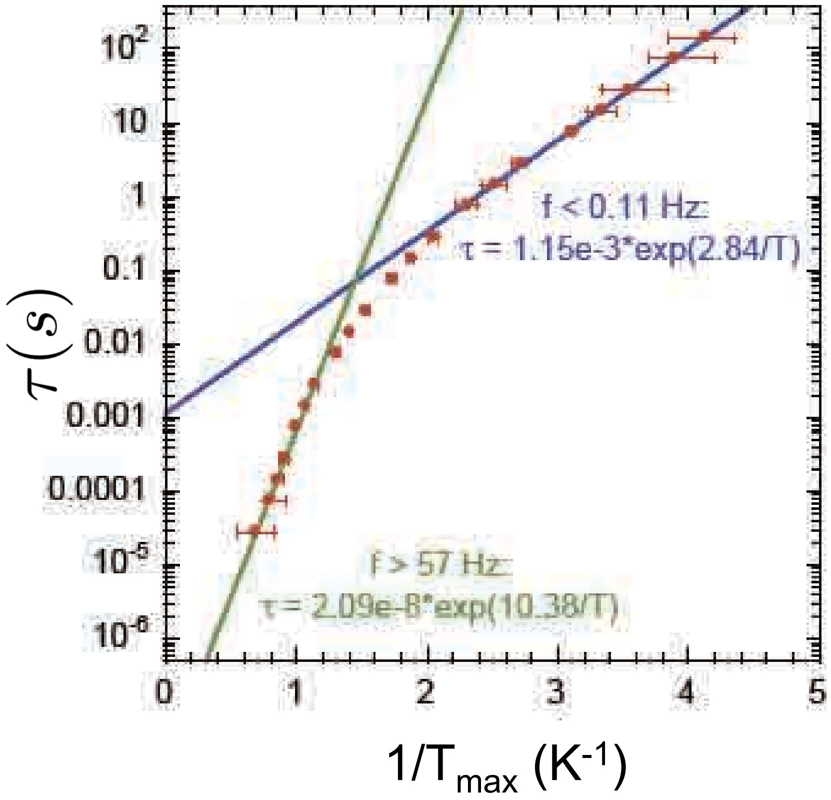
"Fast" high T regime : τ_0=2 10^{-8} s, E=10 K

➔ **String of 5% of quasi-Ising free spins between decoupled magnetic domains**

agrees with a magnetic domain size of ≈ 500 Å from the magnetic peaks FWHM in neutron.



Magnetic properties at low T



Cross-over toward a very slow low T regime :

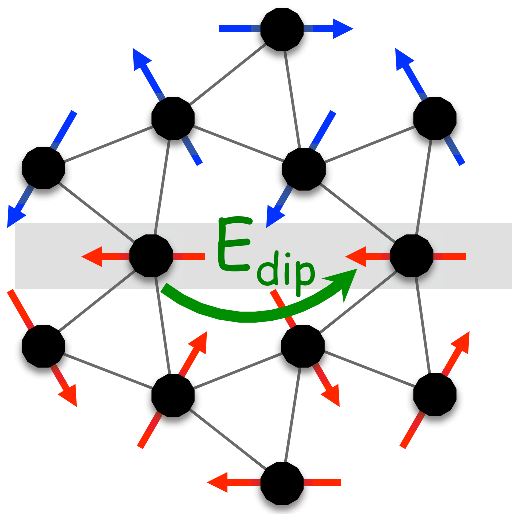
large τ_0 = 10^{-3} s => collective behavior ?

reduced E = 3 K

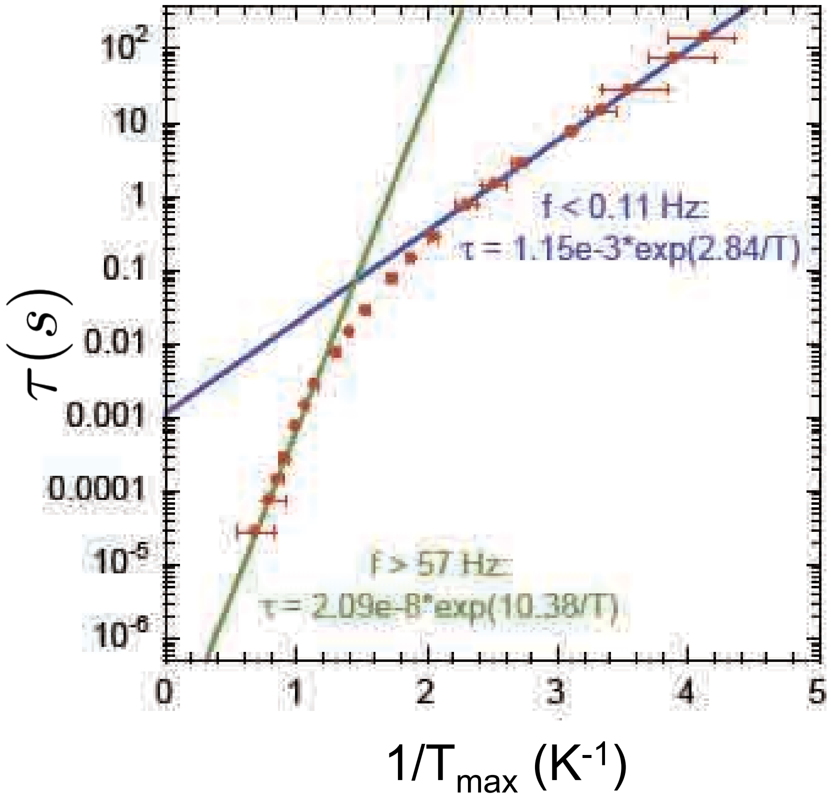
and broadening of the τ distribution

Long range dipolar interactions start to couple the free spins along the domain walls.

Calculated E_{dip}/spin ≈ 0.2 K



Magnetic properties at low T



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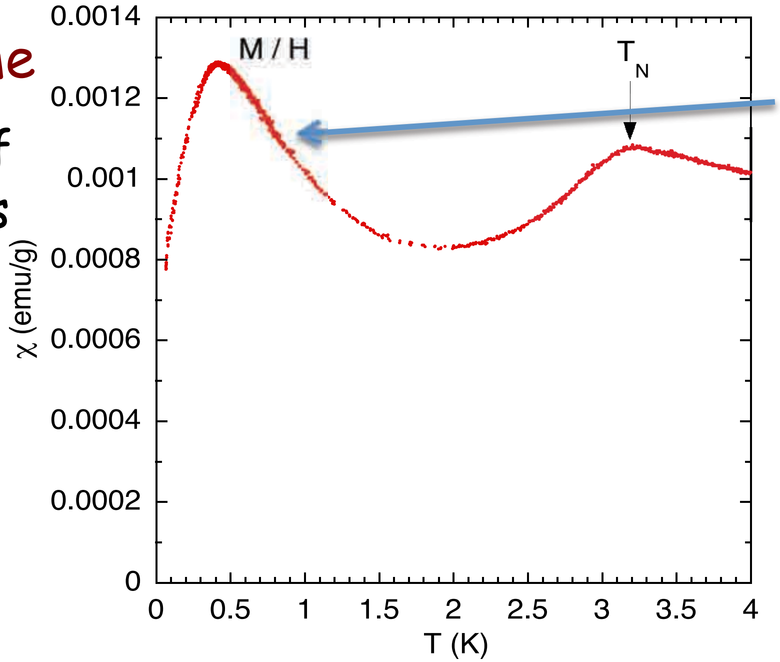
Jaubert & Holdsworth JPCM 2011

Analogy with dipolar pyrochlore spin ice dynamics:
pyrochlore spin-ice = low connectivity, strong multiaxial anisotropy
BUT disordered ground state
high temperature regime: single spin-flips above the anisotropy barrier
low temperature: magnetic excitations (monopoles) coupled by dipolar interactions

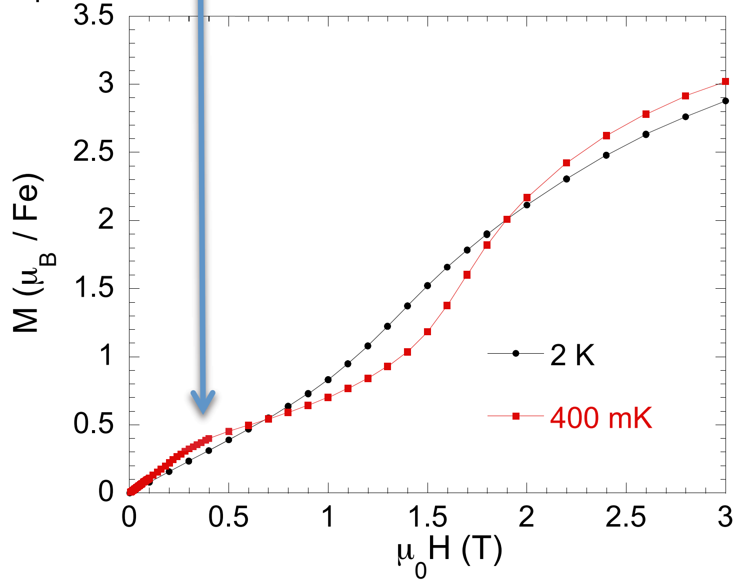
Magnetic properties at low T

High T regime

Further signatures of quasi-Ising free spins



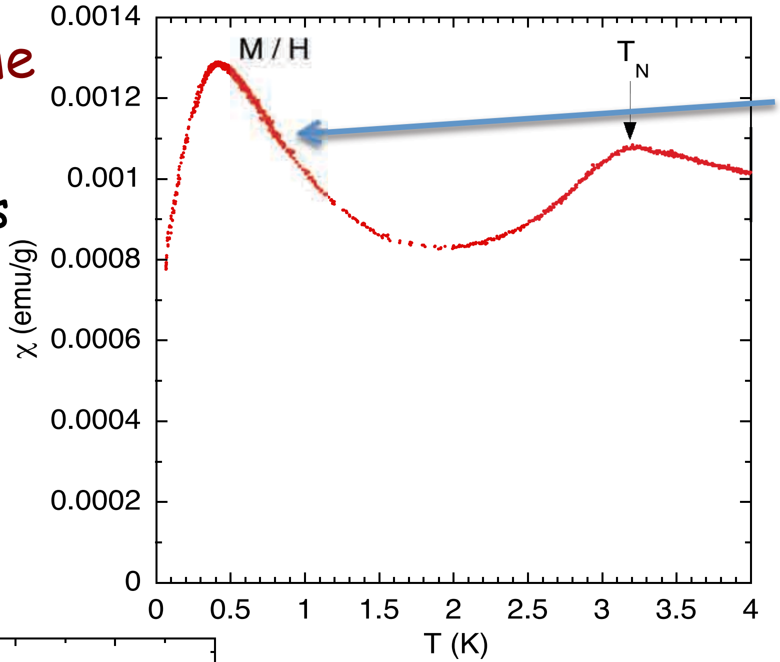
- **upturn of magnetization** *vs* T below 2 K
- **paramagnetic like** contribution in M *vs* H



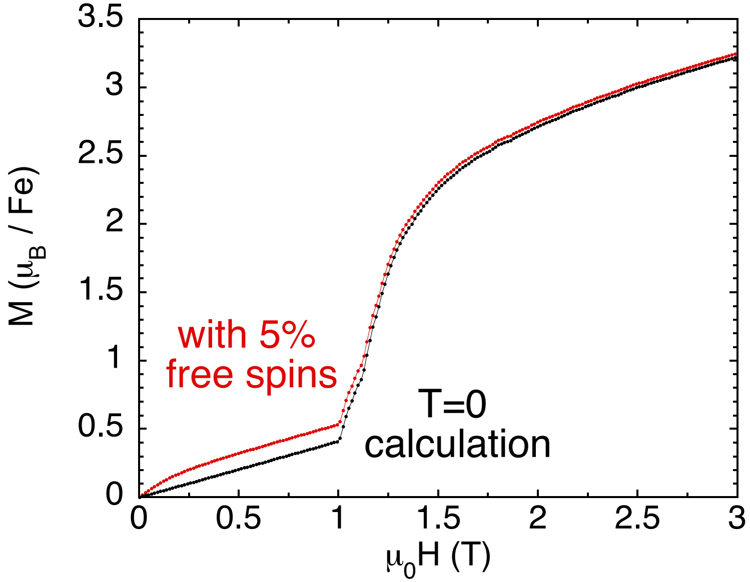
Magnetic properties at low T

High T regime

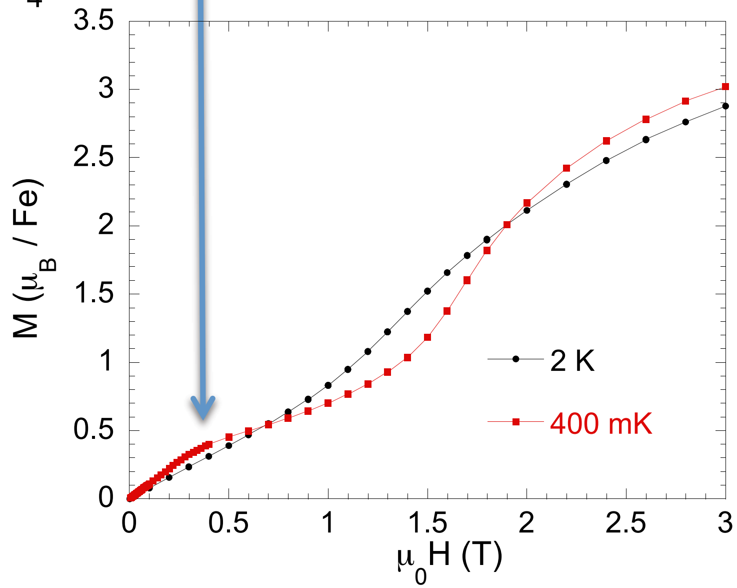
Signatures of the quasi-Ising free spins



- **upturn of magnetization** *vs* T below 1.5 K
- **paramagnetic like** contribution in M *vs* H

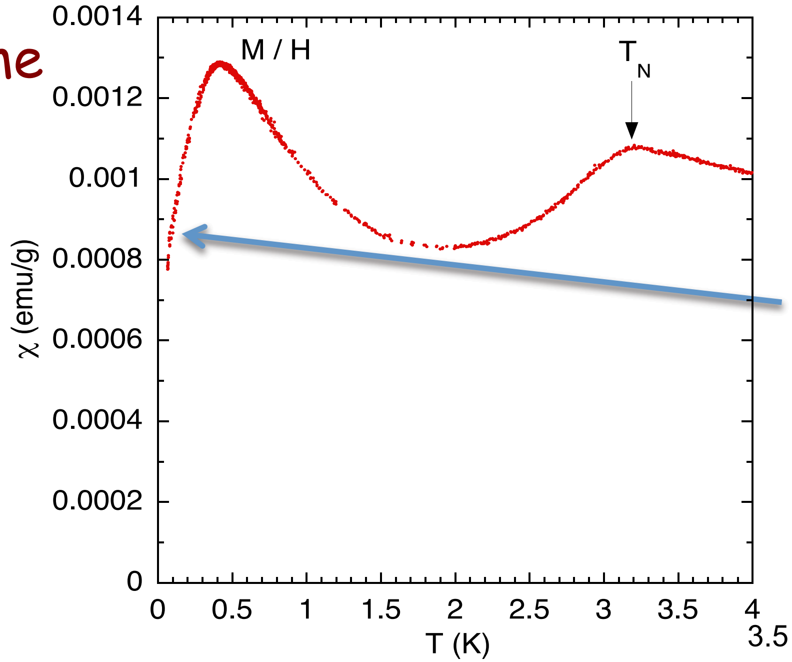


powder averaged T=0 calculation



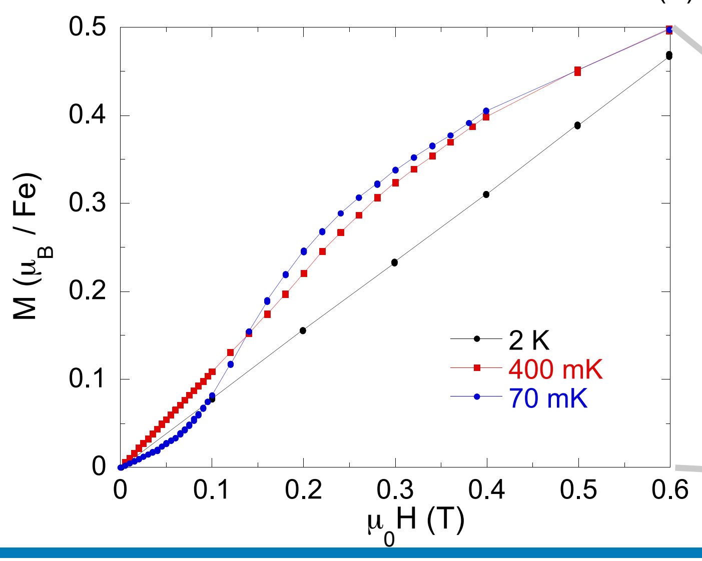
Magnetic properties at low T

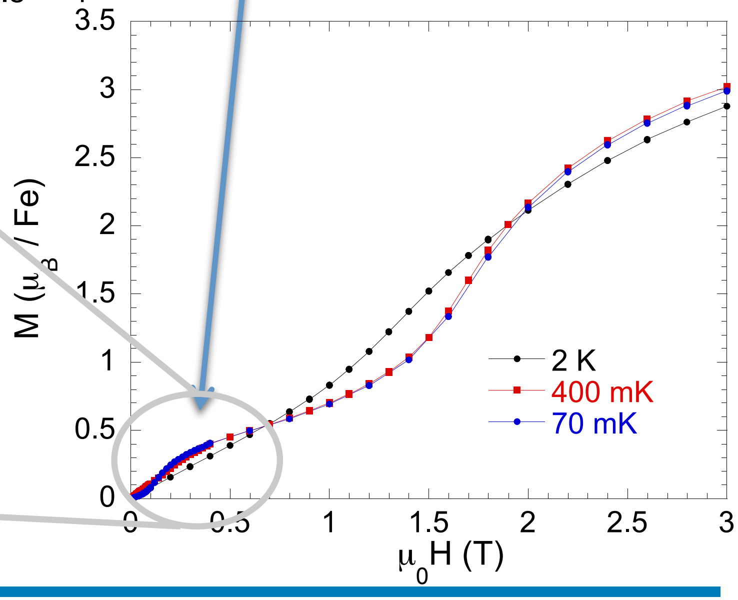
Low T regime



quasi-Ising free spins coupled through dipolar interactions

- **decrease of M** below 0.4 K
- **metamagnetic** contribution in M *vs* H





Discussion

Fe-oxalate compounds = model system of Kagomé antiferromagnets with strong multiaxial anisotropy, exchange and dipolar interactions

•Released frustration: "q=0" 120° magnetic order

•However, low lattice connectivity
➔ string of exchange free quasi-Ising spins along the domain walls decoupling the antiferromagnetic domains

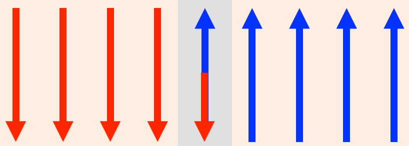
•At lower temperature: collective behavior induced by dipolar interactions

Discussion

General to different systems?

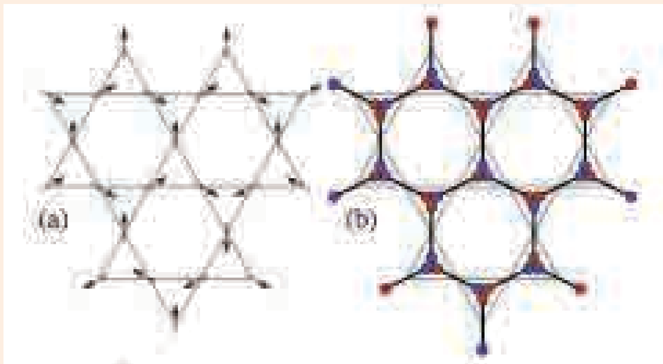
Coulon et al Struct Bond 122, 163 2006

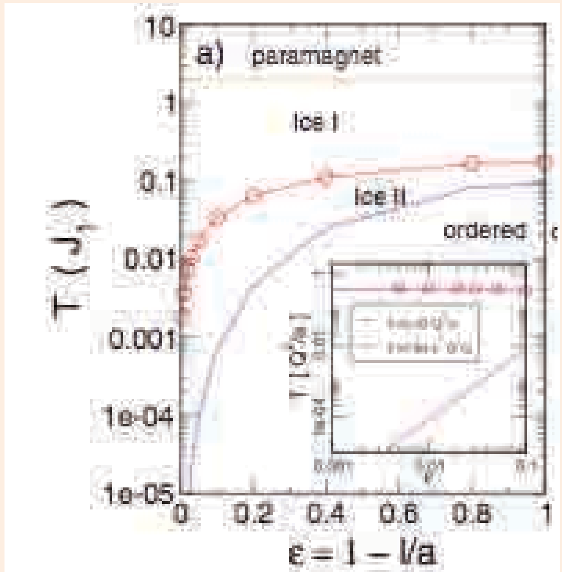
➔Ferromagnetic Single-chain magnets with anisotropy



➔Kagomé with AFM or FM (ordered spin ice) NN interactions and anisotropy

Wills et al PRB 2002, Moeller et al PRB 2009, Chern et al PRL 2011





➔other frustrated lattices with both ordered and fluctuating spins (ex. GGG…)?

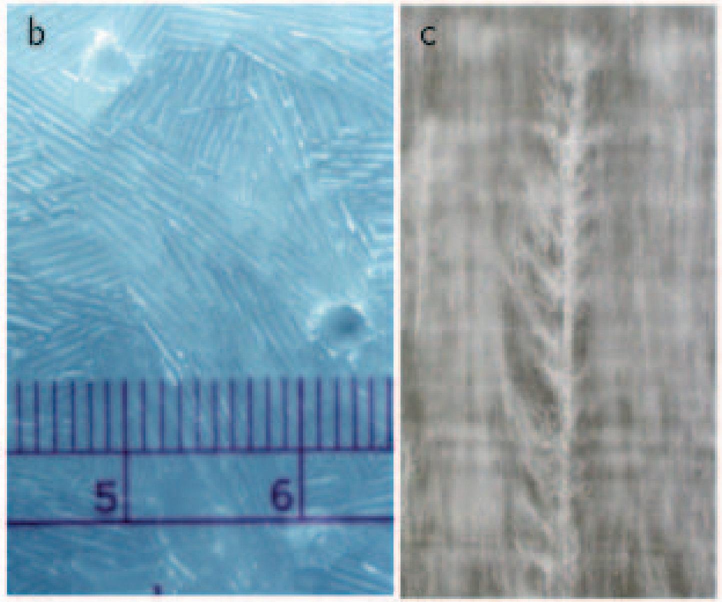
➔Pyrochlores?

Discussion

Analogy to Mushy sea ice phase :

Ice nanocrystals with fluid
flowing inside interstices,
and getting amorphously frozen when T ↘

from E. C. Hunke et al. TCD 2011



Domain wall spin dynamics in Kagome antiferromagnets

PRL 107, 257205 (2011)

E. Lhotel[1], **V. Simonet[1]**, R. Ballou[1], J. Ortloff[1,2], B. Canals[1], C. Paulsen[1]
E. Suard[3], T. Hansen[3], D. J. Price[4], P. T. Wood[5], A. K. Powell[6]

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