Overview Lecture on Basics of Frustrated Magnets : Experimental-I

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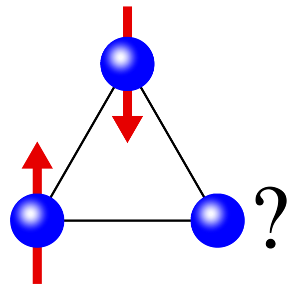
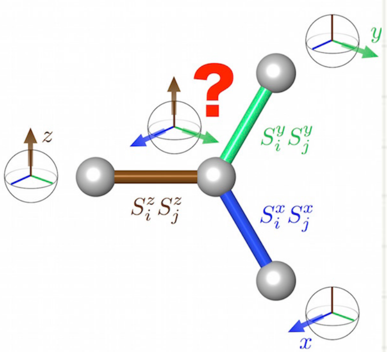
IISER Mohali



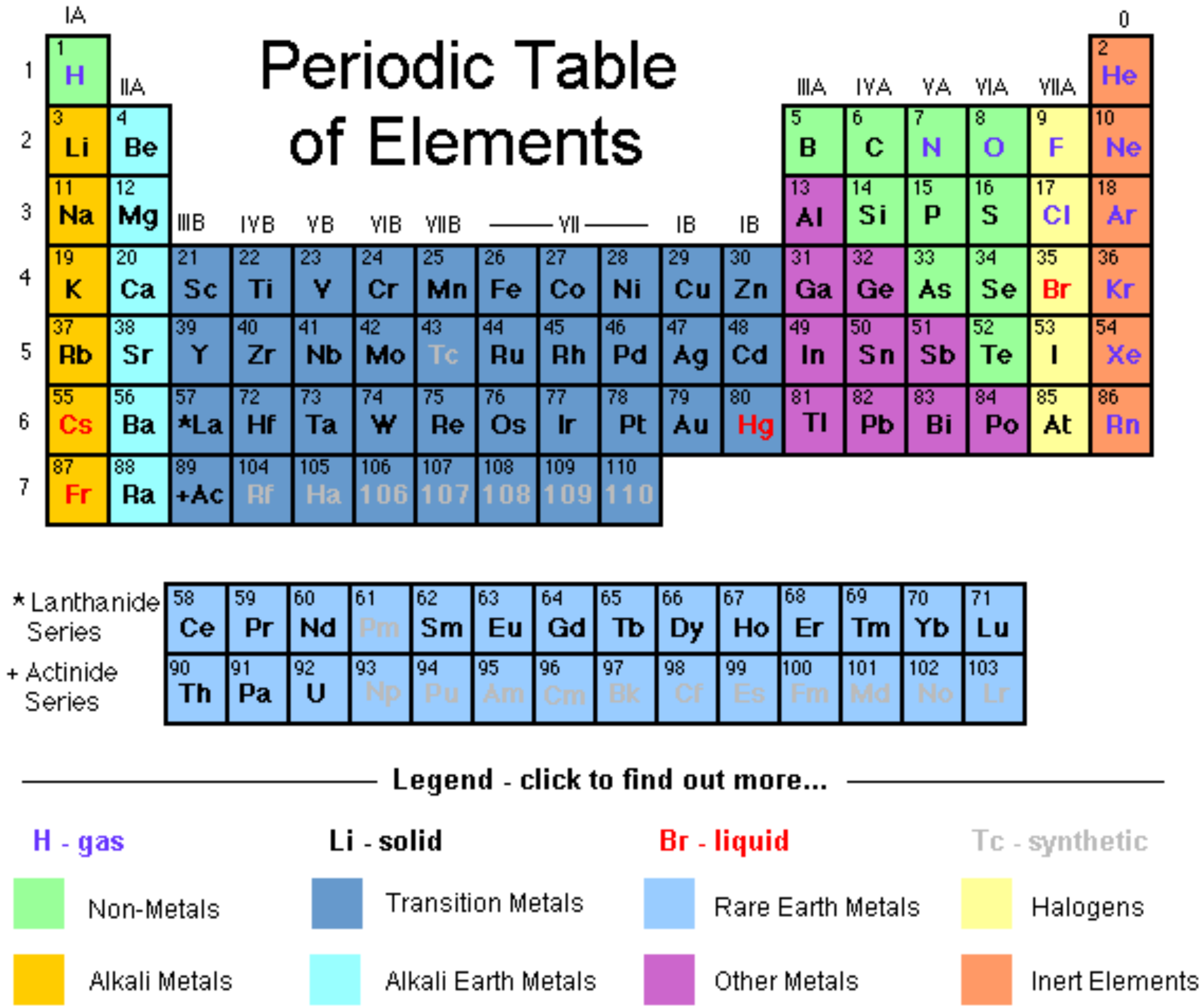


OUTLINE

- Materials to Models
- Geometric Frustration : consequences for magnetic and thermal properties
- Experimental Techniques to study GFM
- Examples :

I. Shastry-Sutherland Model : $SrCu_2(BO_3)_2$
II. Triangular Lattice : Organics κ-ET, dmit
III. Kagome : Herbertsmithite +
IV. Hyper-Kagome : $Na_4Ir_3O_8$
V. Bi-Layer Kagome : $Ca_{10}Cr_7O_{28}$

Where to look for Magnetism



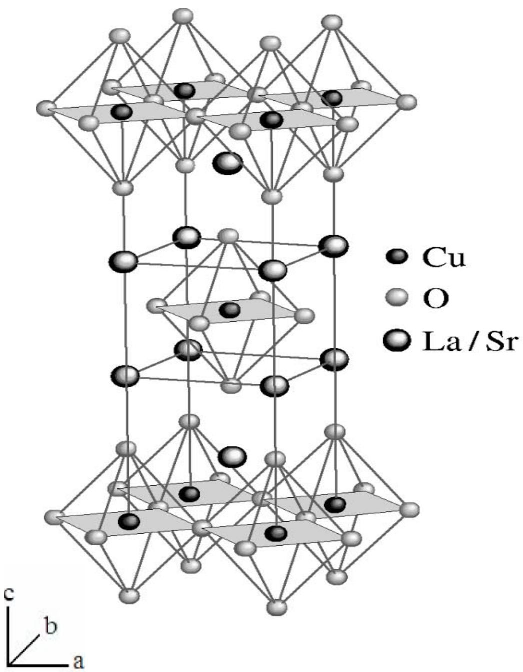
Materials to Models: A case study of La_2CuO_4

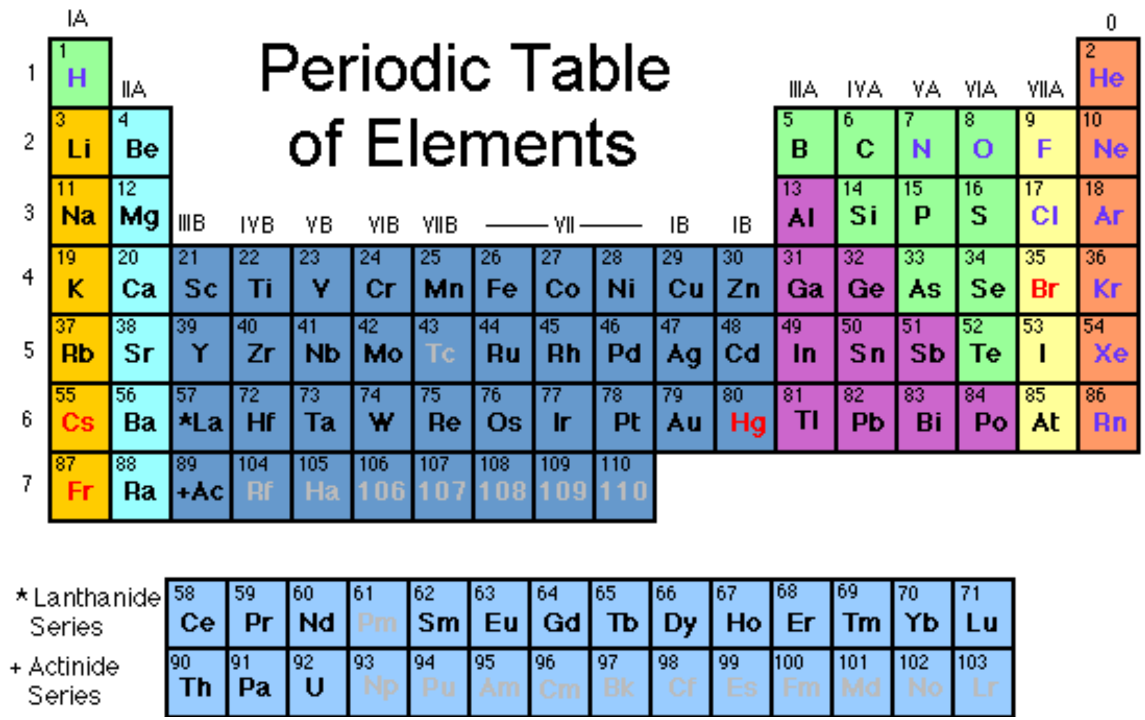
O - [He].$2s^2$.$2p^4$

La - [Xe].$5d^1$.$6s^2$

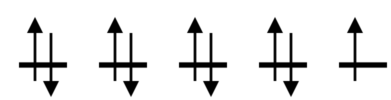
Cu - [Ar].$4s^1$.$3d^{10}$

$(La^{+3})_2Cu^{+2}(O^{-2})_4$



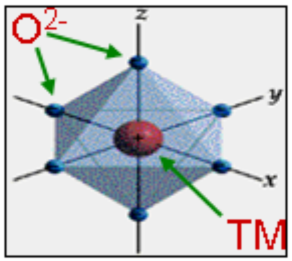


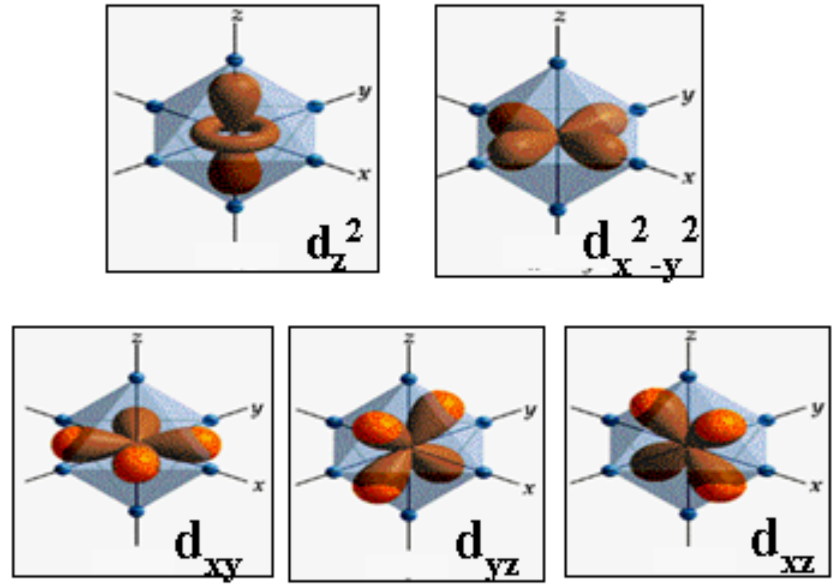
Cu^{+2} - [Ar].$3d^9$

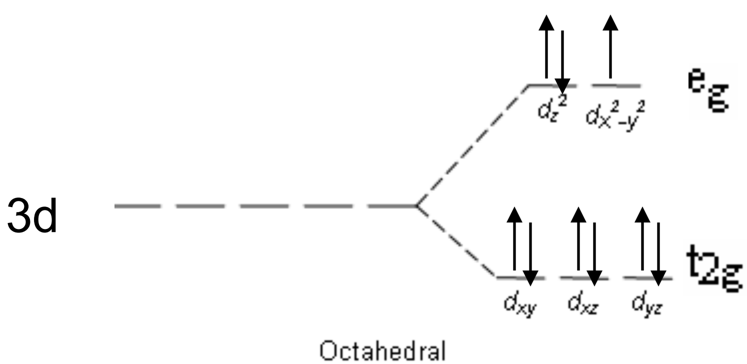


Splitting of d-orbitals in an octahedral crystal field

Crystal field around the d-orbitals







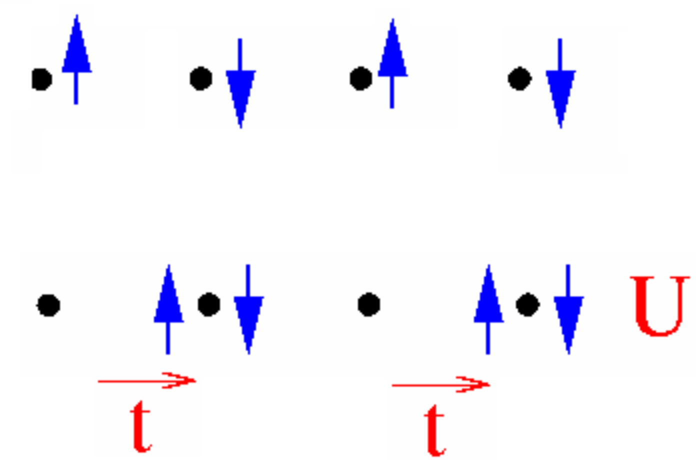
Expect a metal

But La_2CuO_4 is an insulator

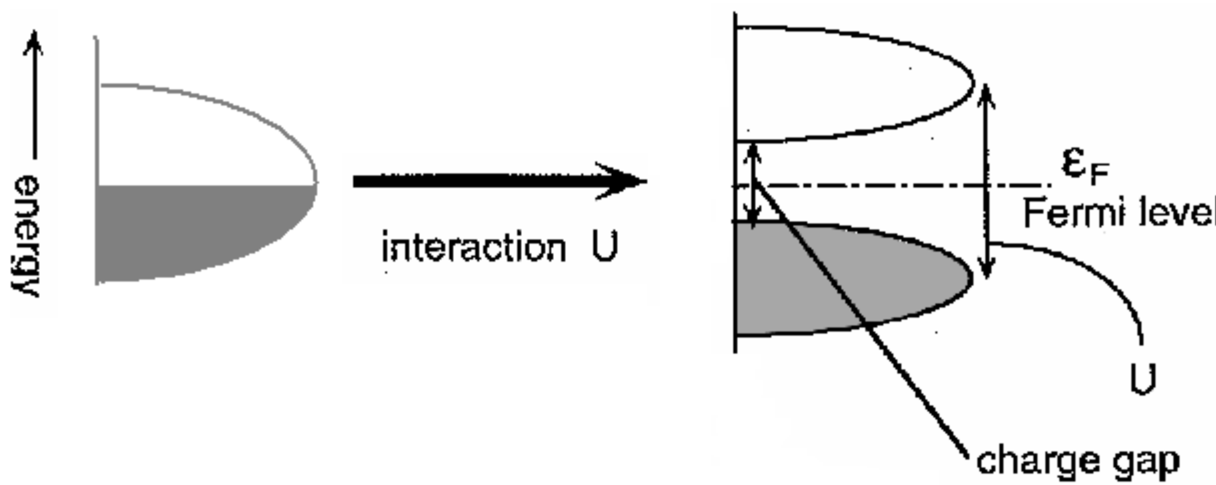
As are MnO, NiO, Fe_3O_4,….

The Mott (Hubbard) insulator

$$H = -t \sum_{(ij)\sigma} (c_{i\sigma}^{\dagger} c_{j\sigma} + c_{j\sigma}^{\dagger} c_{i\sigma}) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

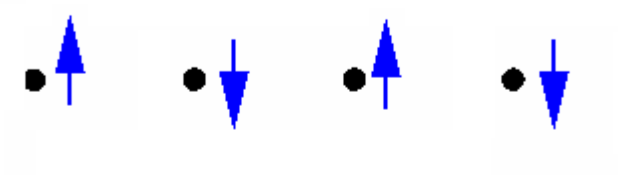


When U/t > 1



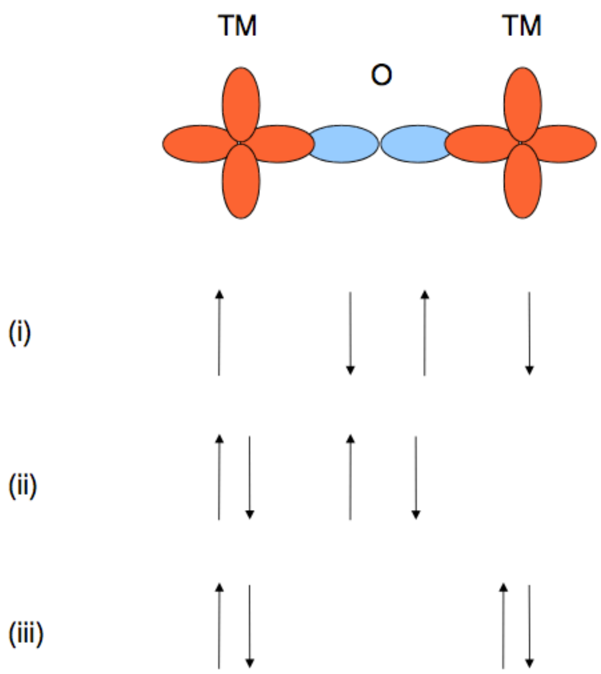
Mott-Hubbard Insulator

Effective Theory: Heisenberg Model



$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Superexchange between two magnetic ions mediated by a non-magnetic ion



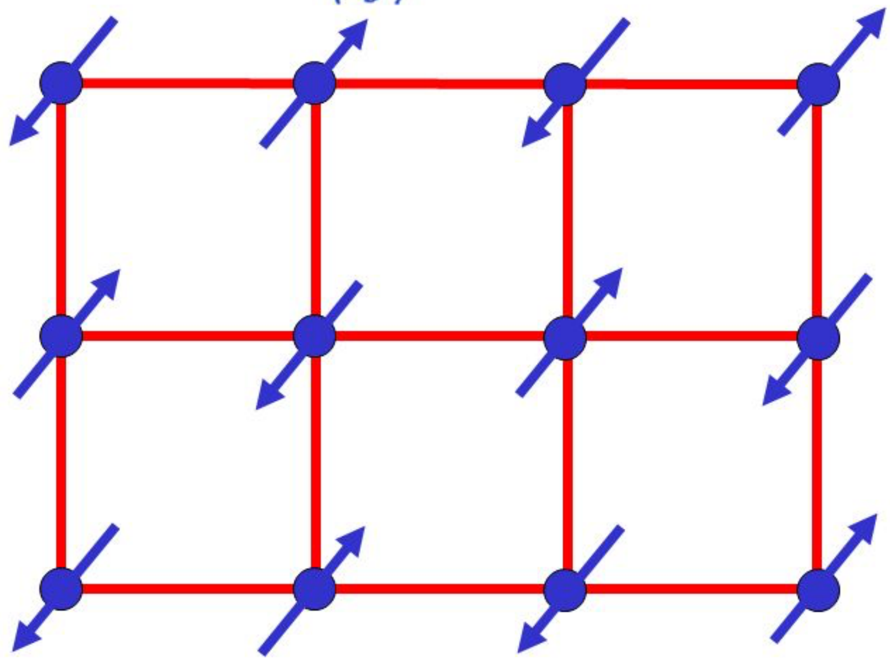
MnO [Mn^{2+} ($3d^5$) and

La_2CuO_4 [Cu^{2+} ($3d^9$)]

Superexchange is mostly antiferromagnetic

Square lattice antiferromagnet

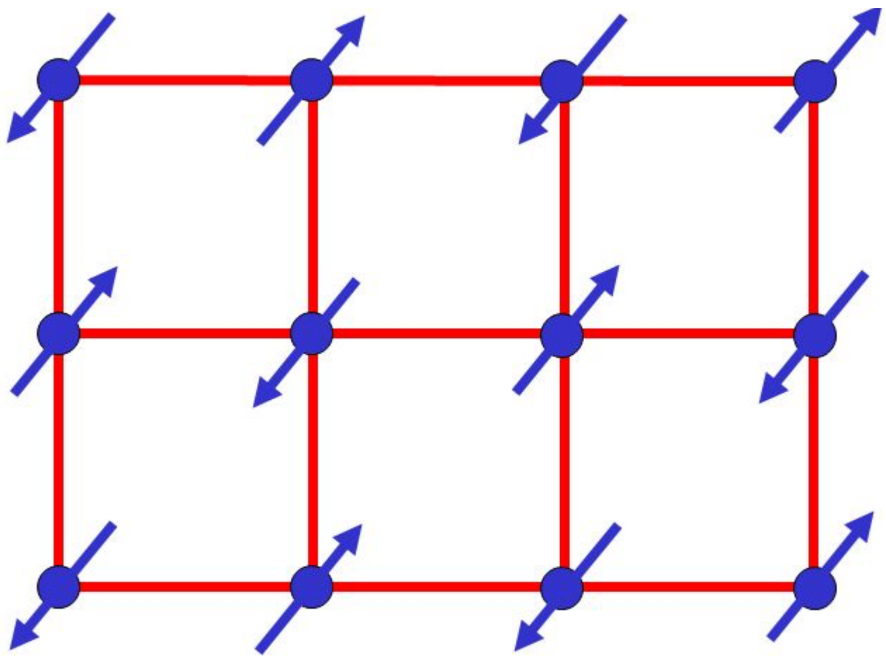
$$H = \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j$$



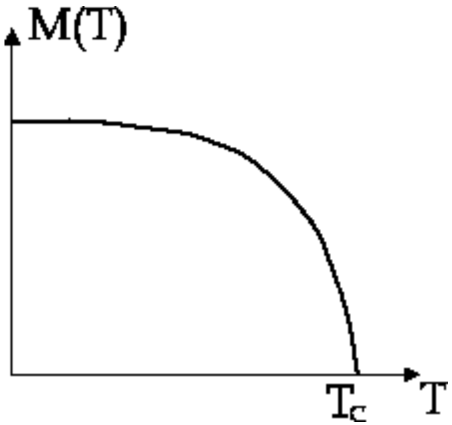
Ground state has long-range Néel order

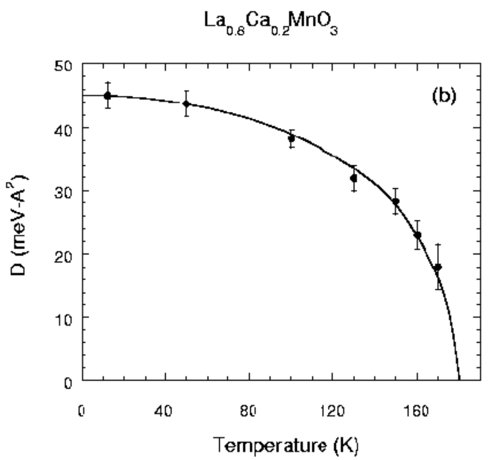
Ordering temperature ~ J

Broken Symmetry, Order Parameter



Ground state has long-range Néel order



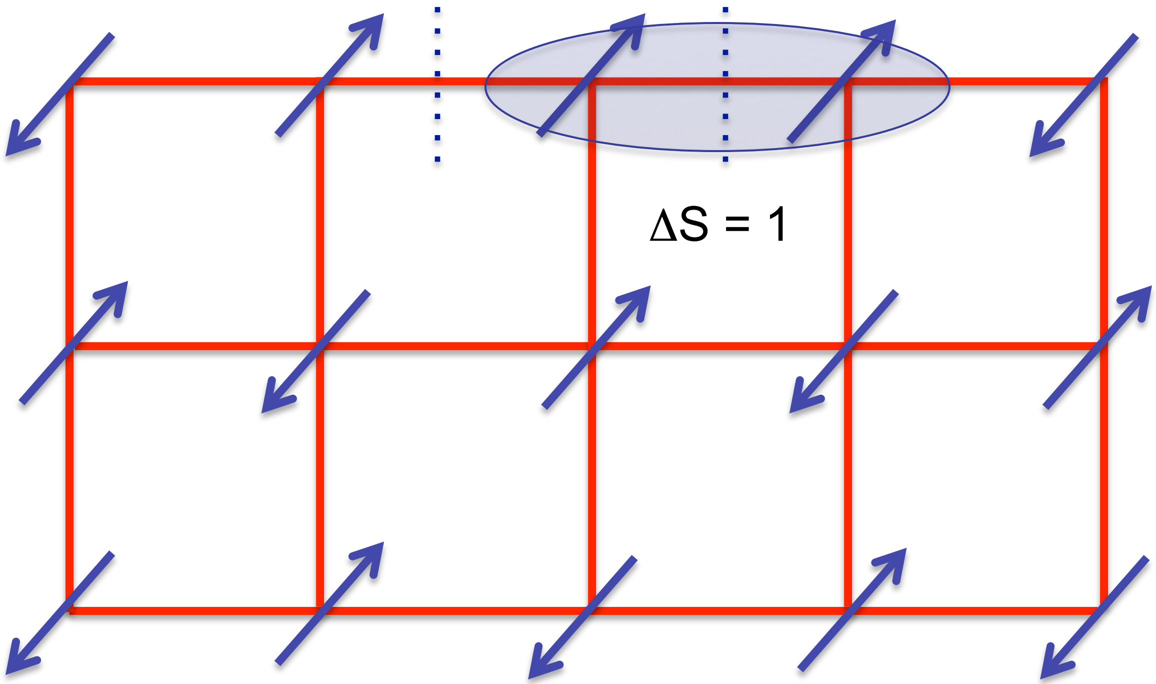


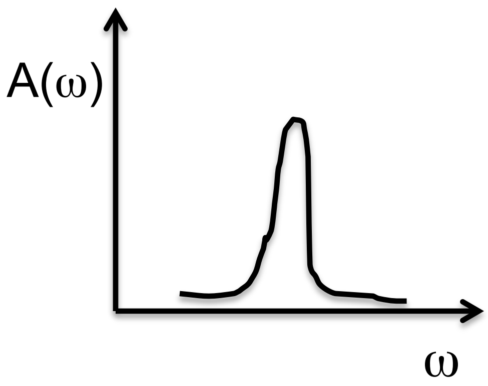
Order Parameter = Sublattice Magnetization

Can be understood within MF theory,

Ground state = product state (approximately)

Excitations of the Antiferomagnet

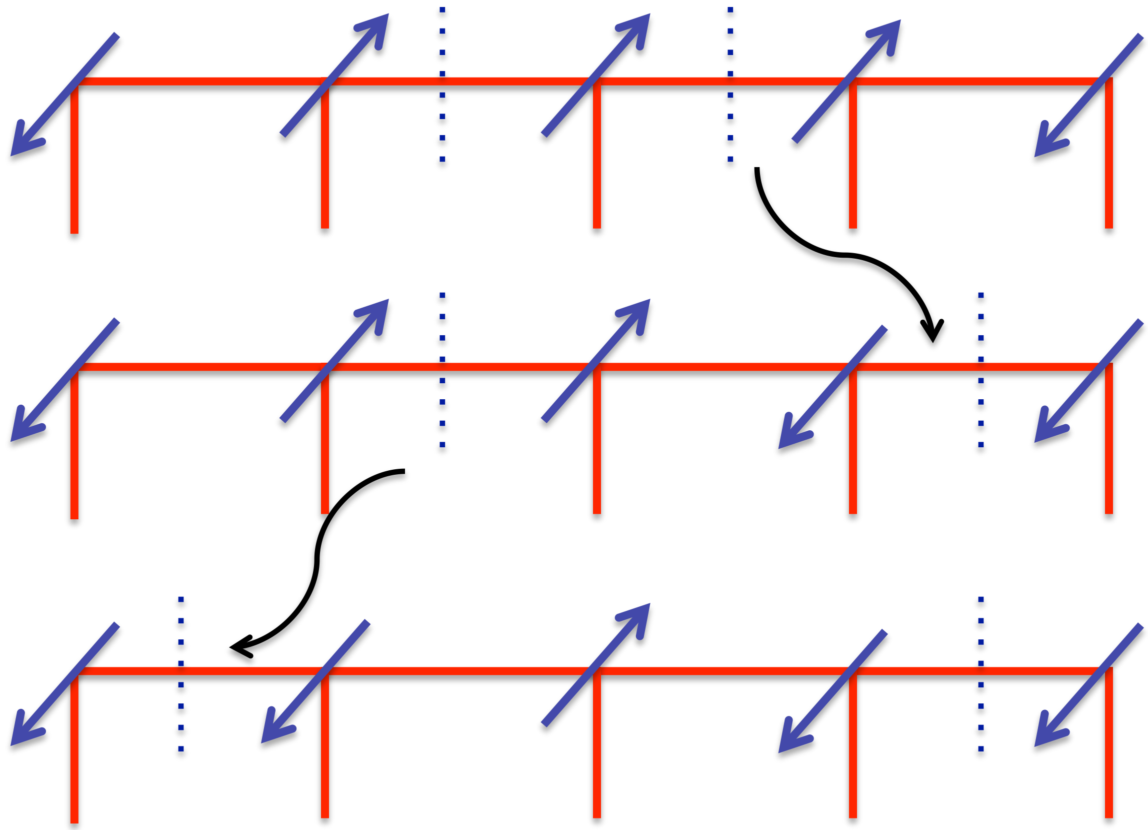




Sharply defined quasi-particles

Can we separate the defects ??

Confinement



Energy cost to pull defects apart ~ L
So excitations are bound $\Delta S = 1$ quasi-particles (magnons)

Conventional Magnets

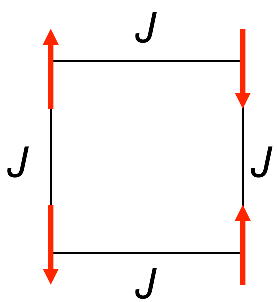
- Order Parameters
- Broken Symmetries
- Ground state ~ product state
- Locality
- Confinement of Quasi-Particles

Geometrically Frustrated Magnets

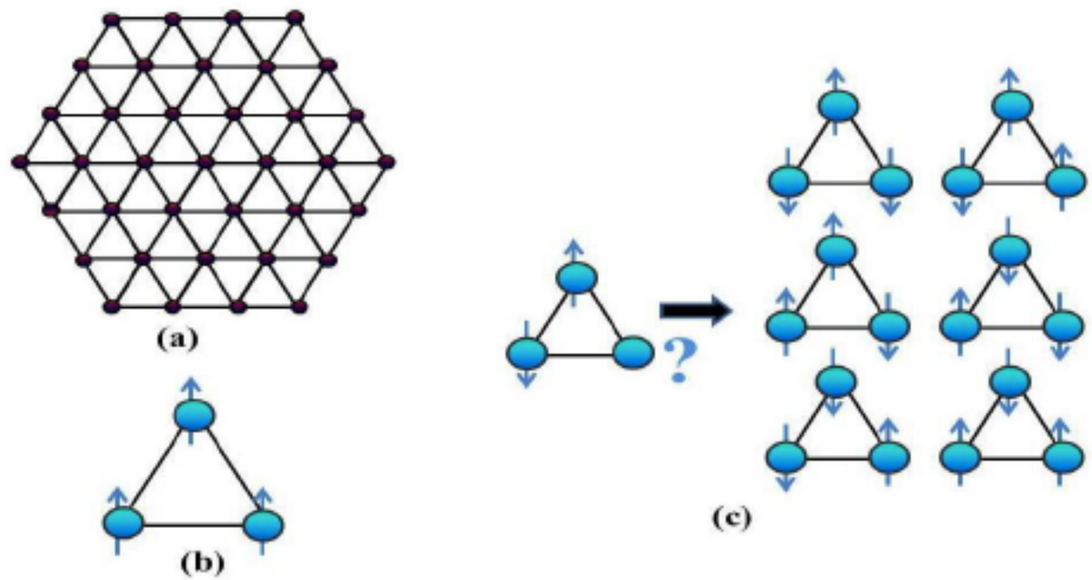
Geometric Magnetic Frustration

$H = JS_i.S_j$ $\equiv$ $J > 0$ *antiferromagnetic* (AFM)

$J < 0$ *Ferromagnetic (FM)*



J > 0 (AFM)

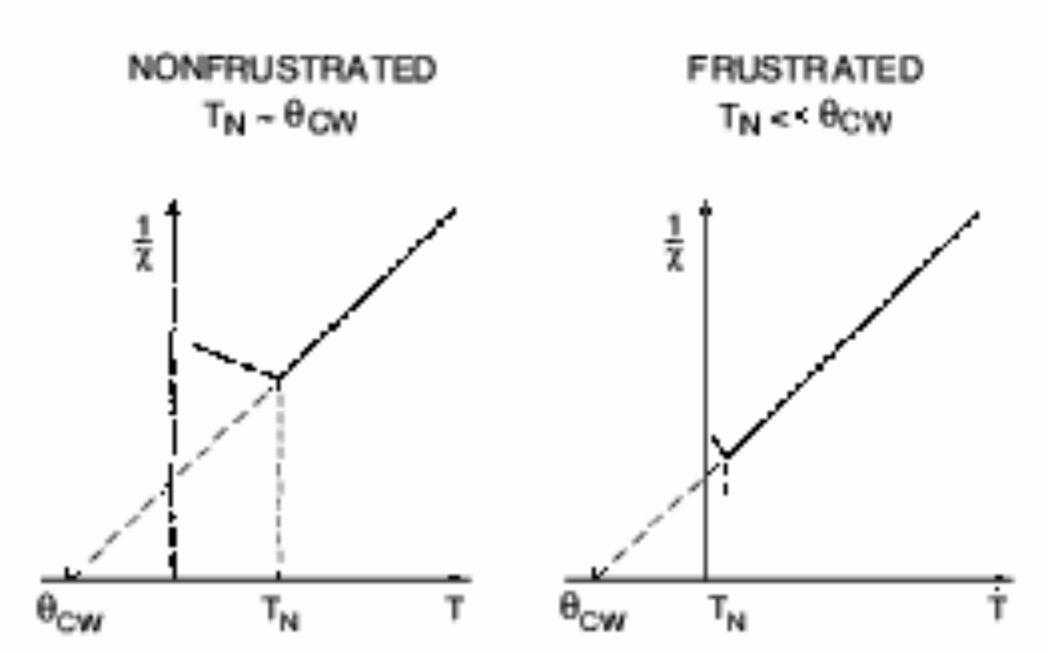


Large Accidental Degeneracy

Consequences of Frustration

$$\chi = \frac{C}{T - \theta_{CW}}$$

$$\frac{1}{\chi} = \frac{T}{C} - \frac{\theta_{CW}}{C}$$



- Frustration suppresses magnetic order

- Reduced ordered moment, unconventional order

- Small entropy at T_N

- Frustration index $f = |\theta|/T_N$

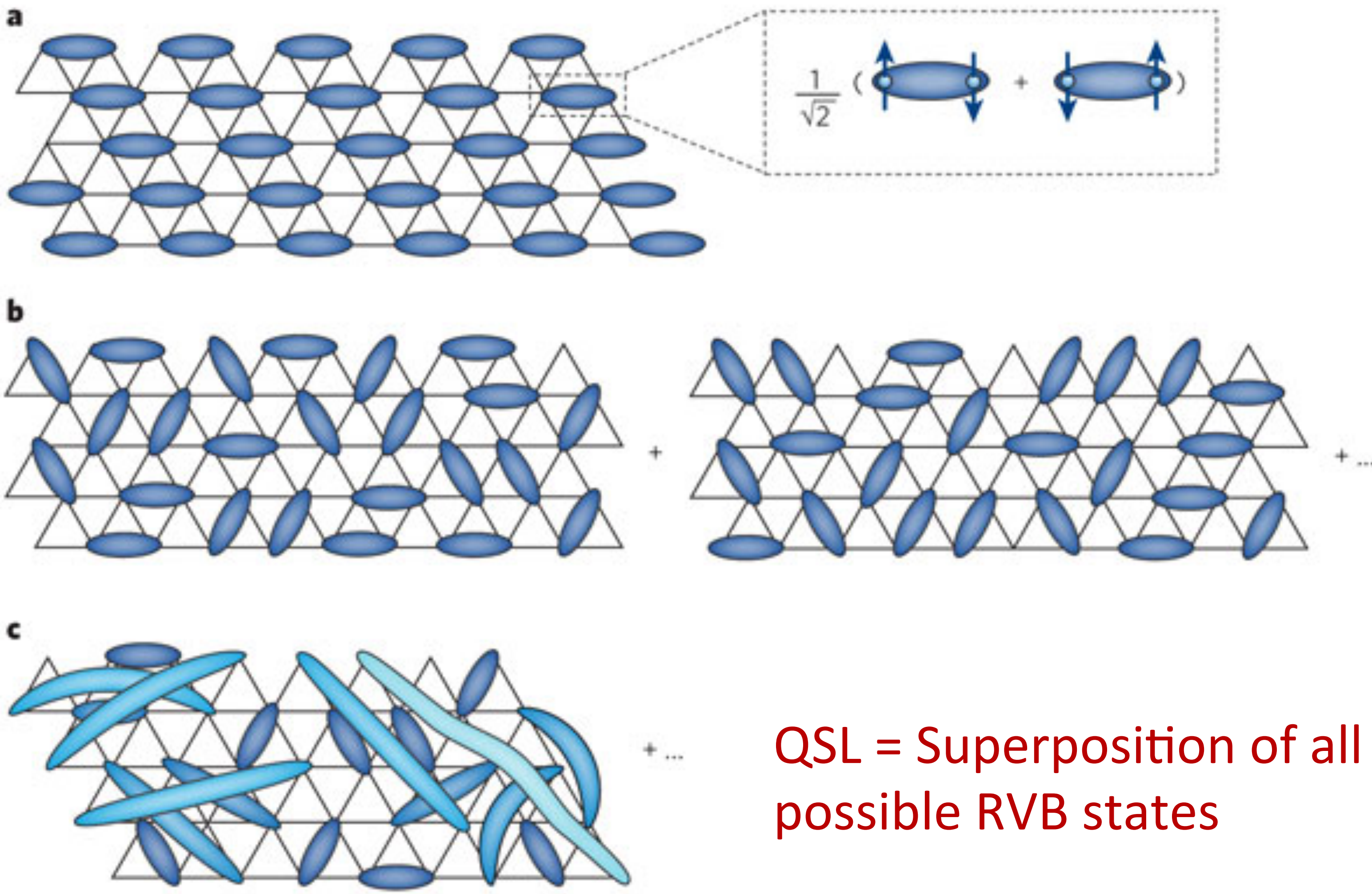
- For Spin–liquid, $f = \infty$

Valence Bond States

Valence Bond Solid

Resonating Valence Bond : Short Range

Resonating Valence Bond : Long Range

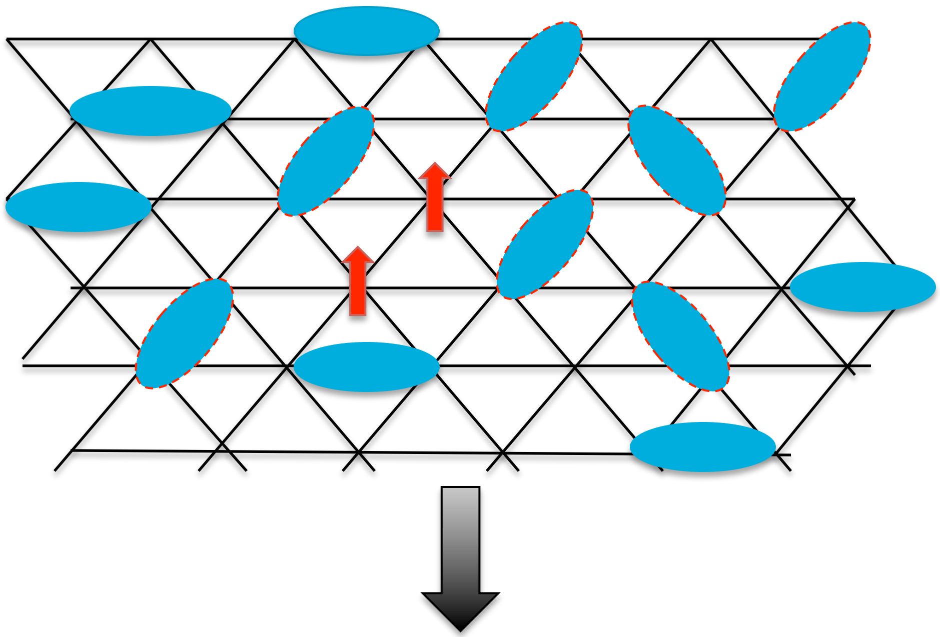


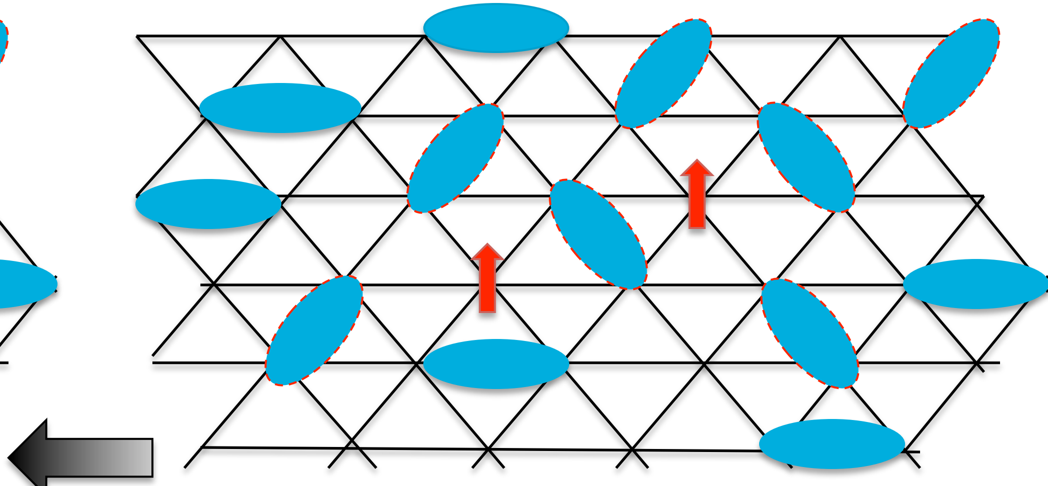
QSL = Superposition of all possible RVB states

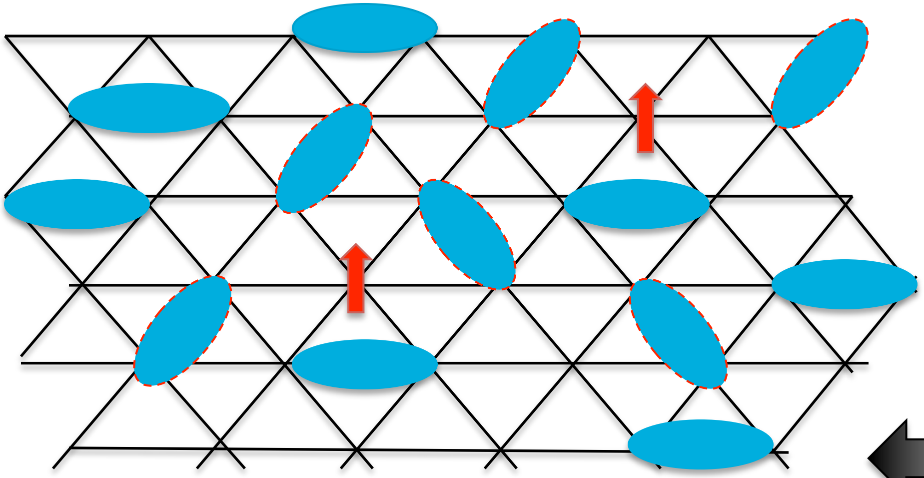
No Broken Symmetry, No Order Parameter, Massive Entanglement

Excitations in spin liquids: Fractional Deconfined Particles

- $\Delta S = 1$ excitation created
- Quasi-particles with $S = 1/2$ but no charge (= Spinons) "delocalize" with little or no energy cost







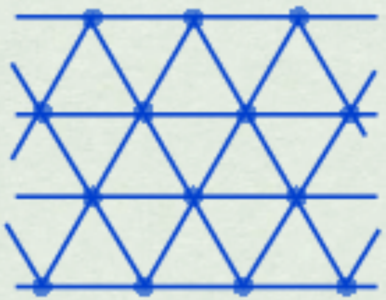
Quantum Spin Liquids

- No Order Parameters
- No Broken Symmetries
- Massive Entanglement
- Non-Locality
- De-confinement of Quasi-Particles
- Topological Order

Experimental Signatures for Spin Liquids

- Absence of LRO (χ, C)
- Persistent dynamics (NMR, μSR)
- Spinon = diffuse excitations in neutron scattering (INS)
- Mobile/deconfined Spinons (κ, C)

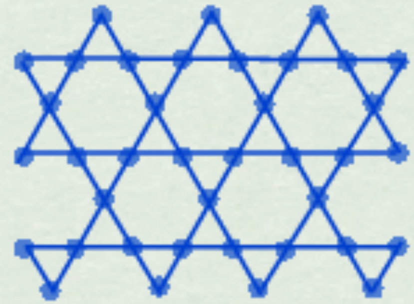
Frustrated lattices: examples

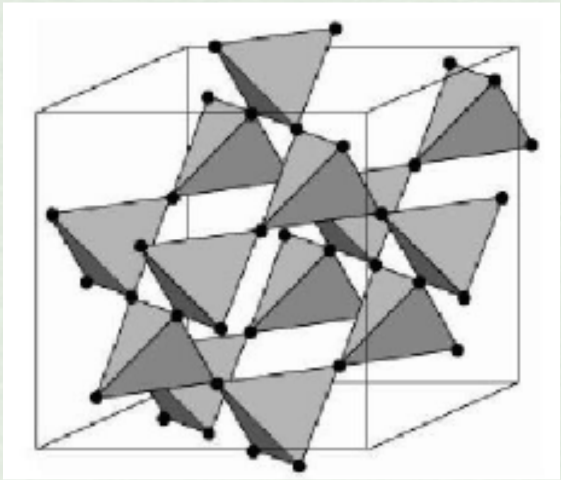

triangular

$NiGa_2S_4$,

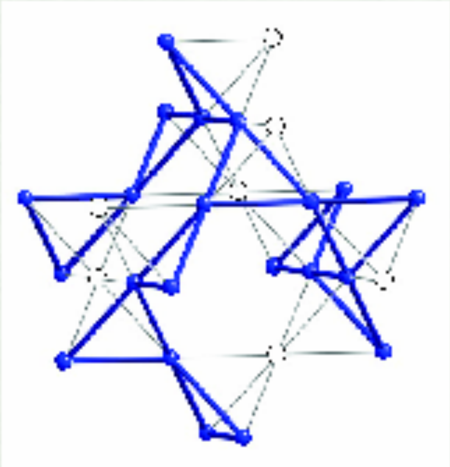
κ-ET, dmit

$ZnCu_3(OH)_6Cl_2$


kagome

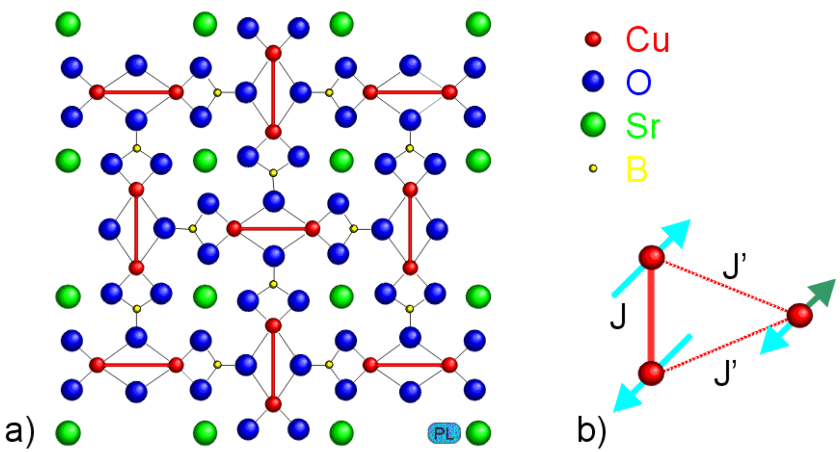

pyrochlore

$R_2Ti_2O_7$

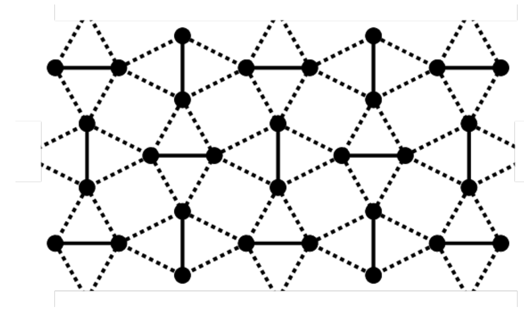

hyper-kagome

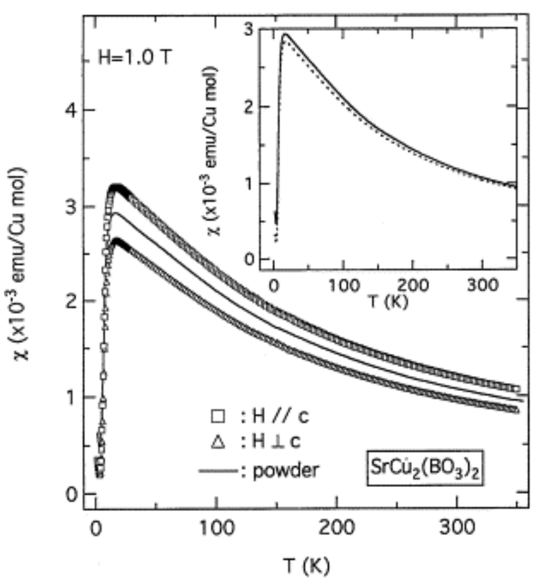
$Na_4Ir_3O_8$

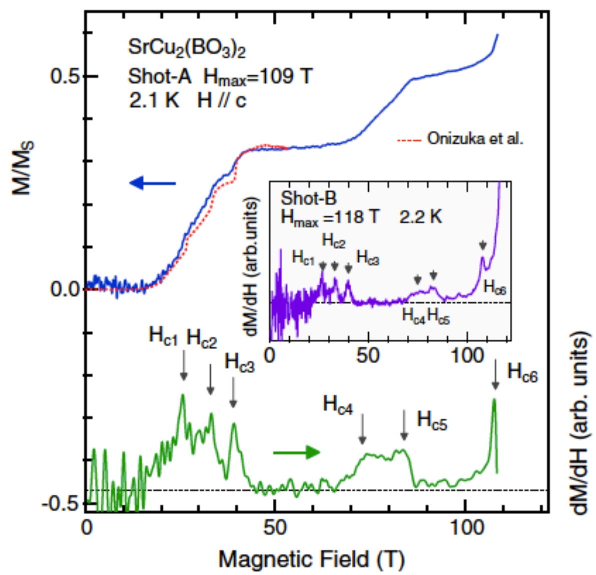
Shastry-Sutherland Lattice

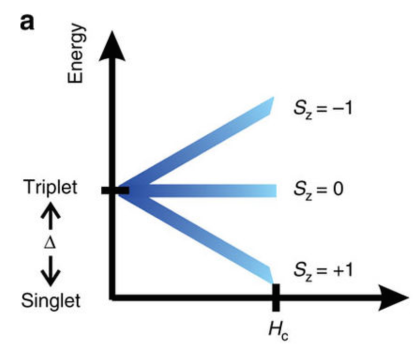


$Sr_2Cu_2(BO_3)_2$, TmB_4, YbB_4...

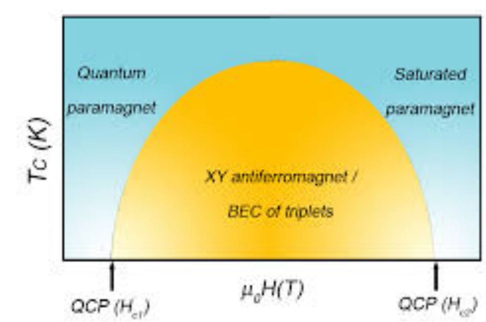




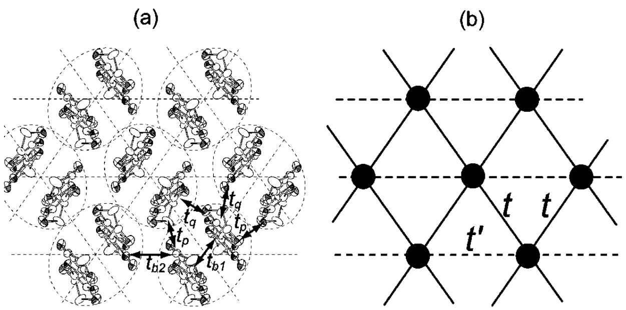




BEC of Triplons

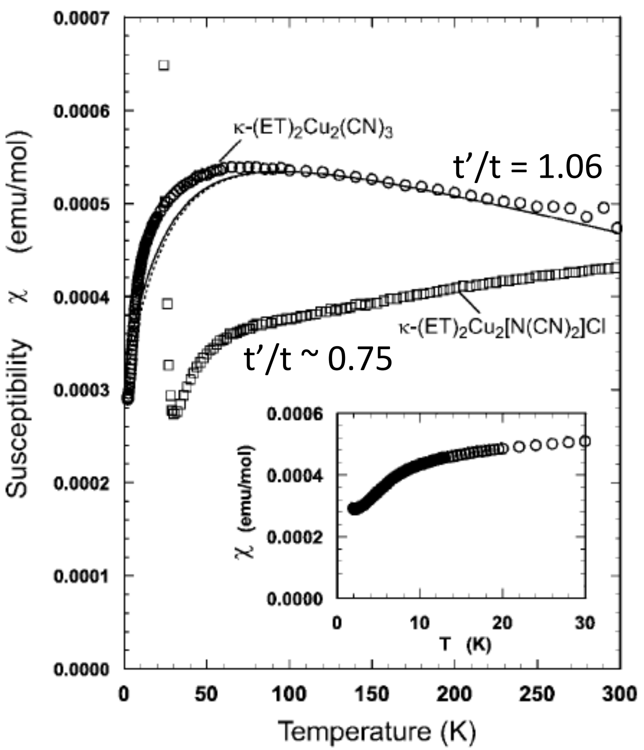


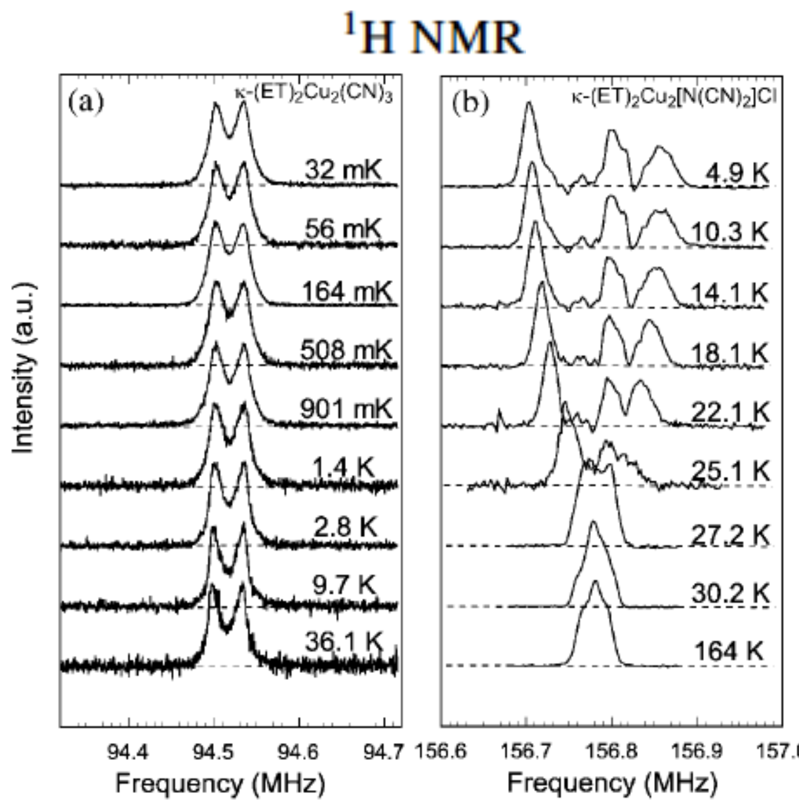
Triangular Lattice Organics



κ-(ET)$_2X$ family

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

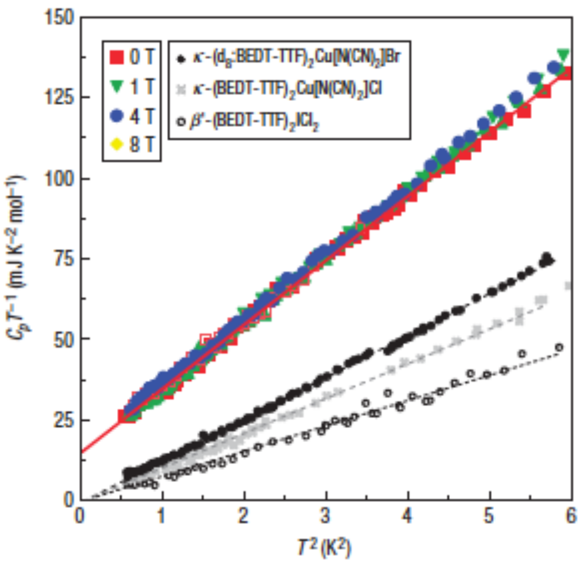


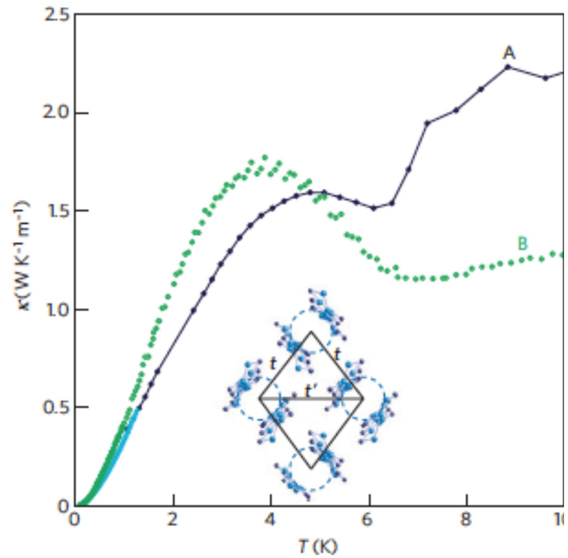


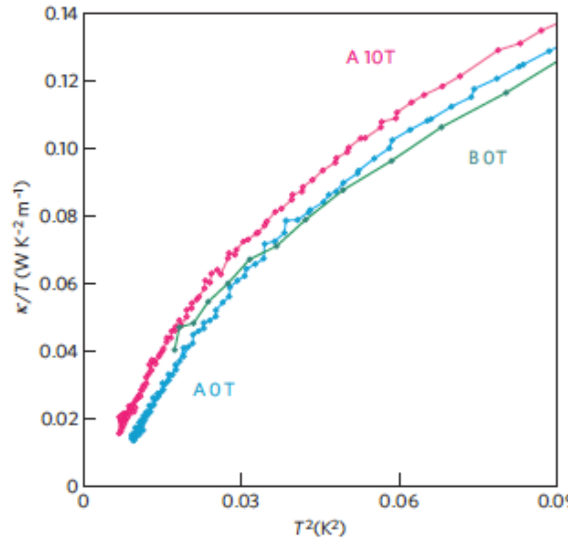
κ-ET : Heat Capacity + Thermal Conductivity

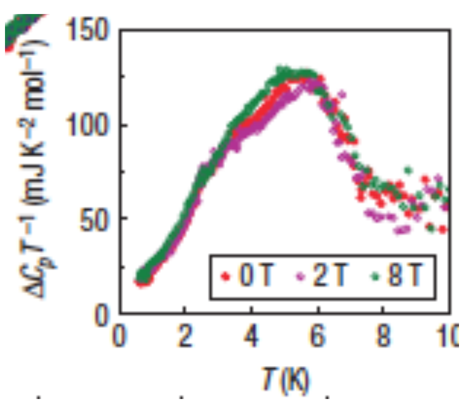
M. Yamashita, Nature Phys. **4**, 469 (2008).

M. Yamashita, Nature Phys. **5**, 44 (2009).









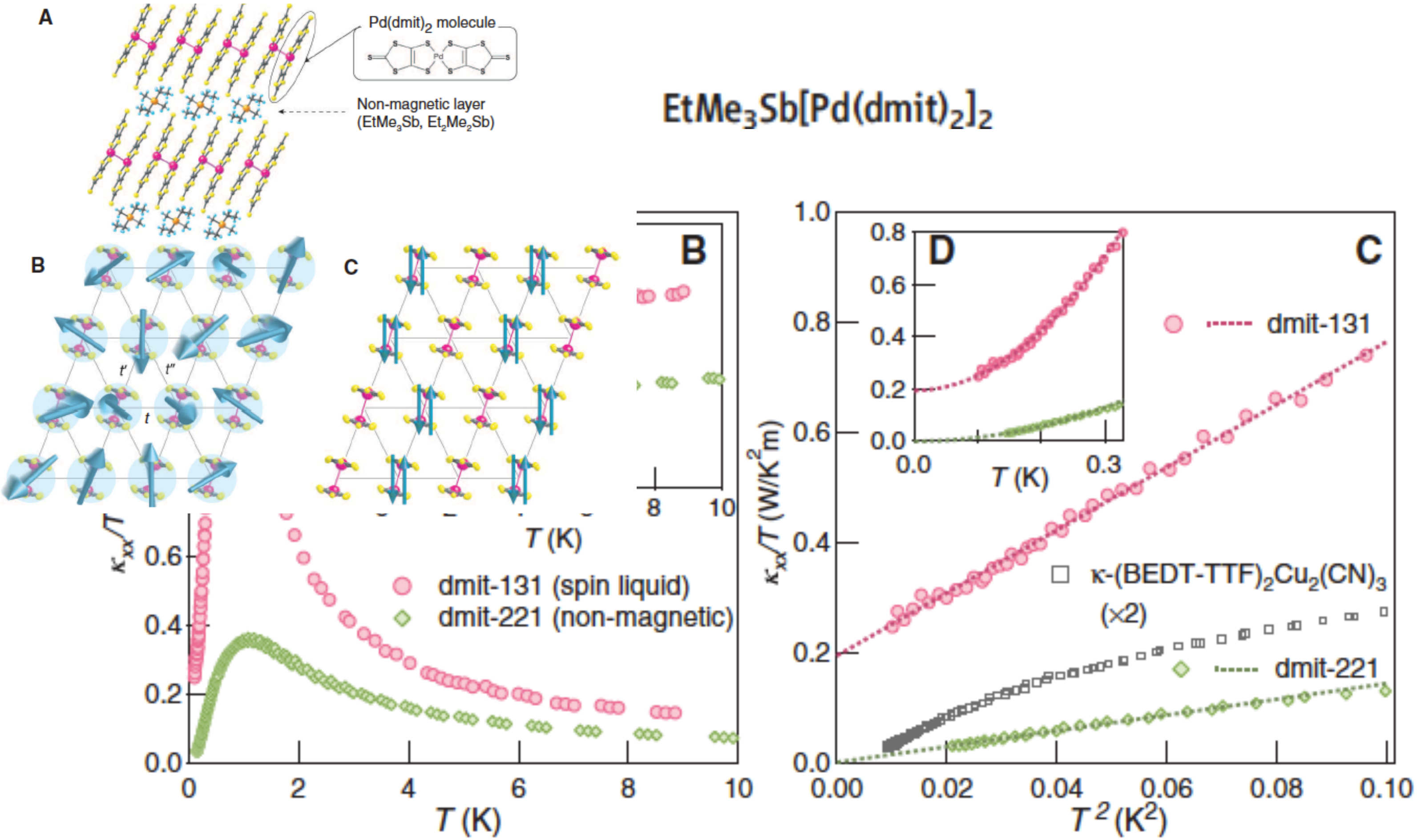
kappa- (BEDT-TTF)2Cu2(CN)3 (κ/T → 0, γ ≈ 20 mJ/mol K^2)

Dichotomy between χ and C/T (Gapless), and κ/T (Gapped)

Dichotomy Resolution : phase separation of gapless and gapped phases

S. Nakajima *et al.*, J. Phys. Soc. Jpn. **81**, 063706 (2012).

dmit: Heat Capacity + Thermal Conductivity



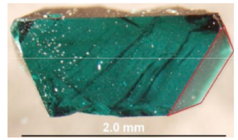
ET-dmit ($\kappa/T \approx 180$ mW/K^2 m, $\gamma \approx 20$ mJ/mol K^2)

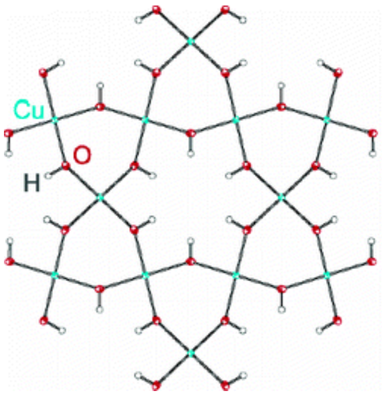
M. Yamashita, Science **328**, (2010).

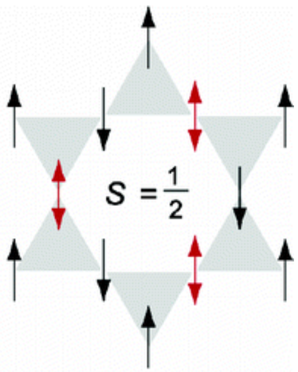
Gapless QSL

Kagome : Herbertsmithite + others

$Zn_xCu_{4-x}(OH)_6Cl_2$

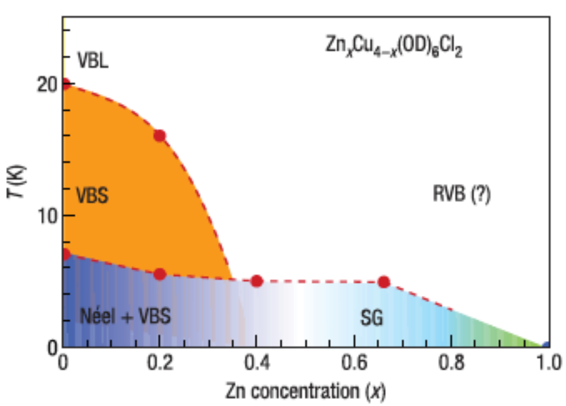


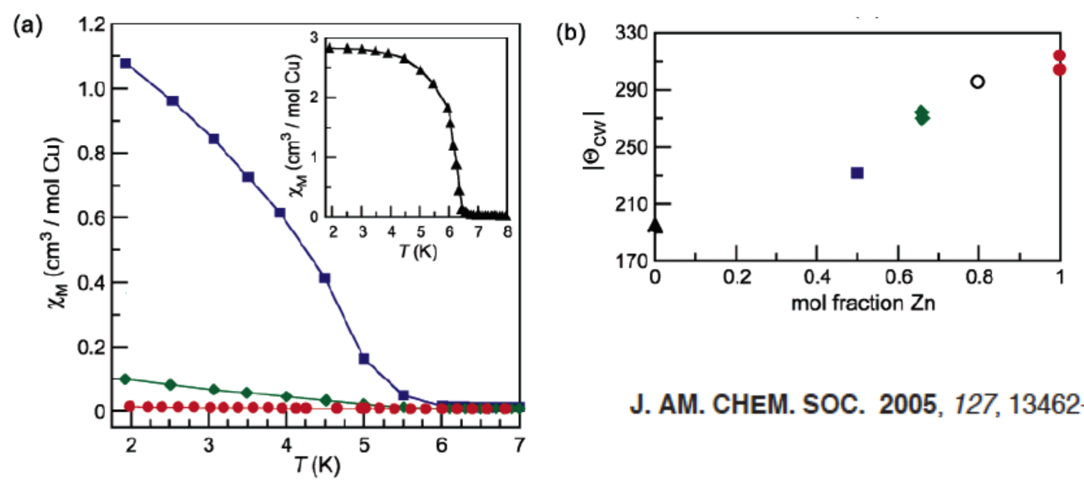




Name	Formula	Group	Lattice	Order
Botallackite	$Cu_4(OH)_6Cl_2$	$P2_1/m$	T	AF (7.2 K)
Atacamite	$Cu_4(OH)_6Cl_2$	$Pnma$	P	AF (9 K)
Clinoatacamite	$Cu_4(OH)_6Cl_2$	$P2_1/n$	P	AF (6.5 K)
Claringbullite	$Cu_4(OH)_6ClF$	$P6_3/mmc$	P	AF (17 K)
Barlowite	$Cu_4(OH)_6BrF$	$P6_3/mmc$	P	AF (15 K)
Bobkingite	$Cu_5(OH)_8Cl_2W_2$	$C2/m$	P	?
Herbertsmithite	$ZnCu_3(OH)_6Cl_2$	$R\bar{3}m$	K	AF $(\cdots)$
Tondiite	$MgCu_3(OH)_6Cl_2$	$R\bar{3}m$	K	AF $(\cdots)$
Kapellasite	$ZnCu_3(OH)_6Cl_2$	$P\bar{3}m1$	K	F $(\cdots)$
Haydeeite	$MgCu_3(OH)_6Cl_2$	$P\bar{3}m1$	K	F (4.2 K)
Zn-brochantite	$ZnCu_3(OH)_6SO_4$	$P2_1/a$	K*	AF $(\cdots)$

M. R. Norman, RMP 88, 041002, (2016).





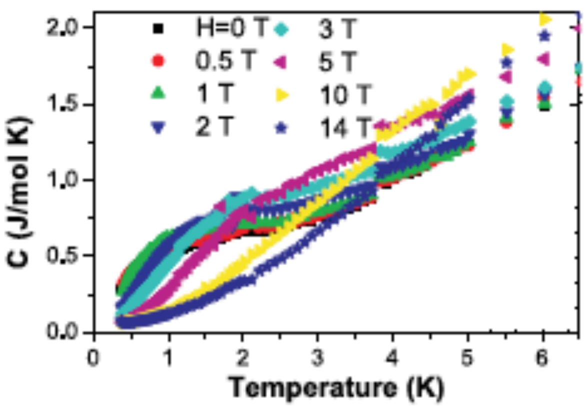
J. AM. CHEM. SOC. 2005, *127*, 13462

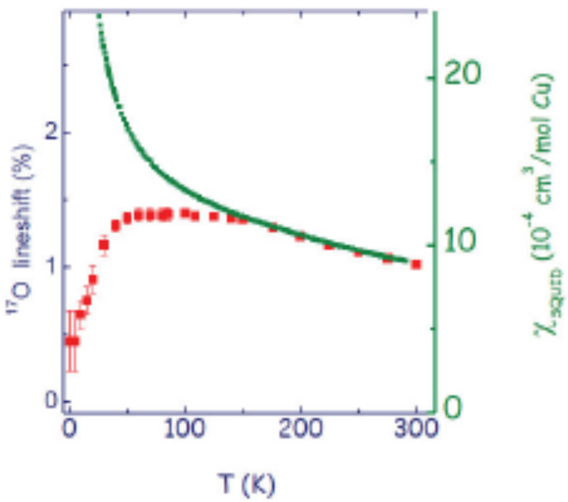
Herbertsmithite : Magnetic + Thermal

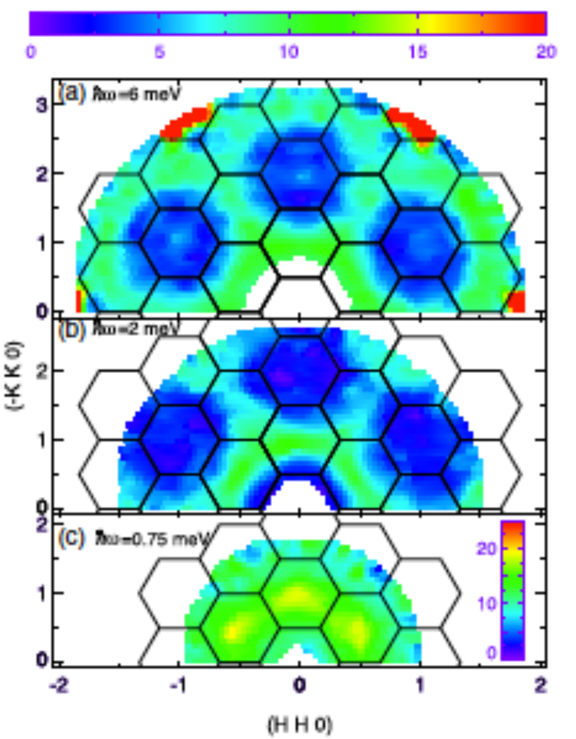
C(T) and χ(T) dominated by impurity Cu spins

NMR reveals intrinsic behaviour : gapless QSL!!

INS

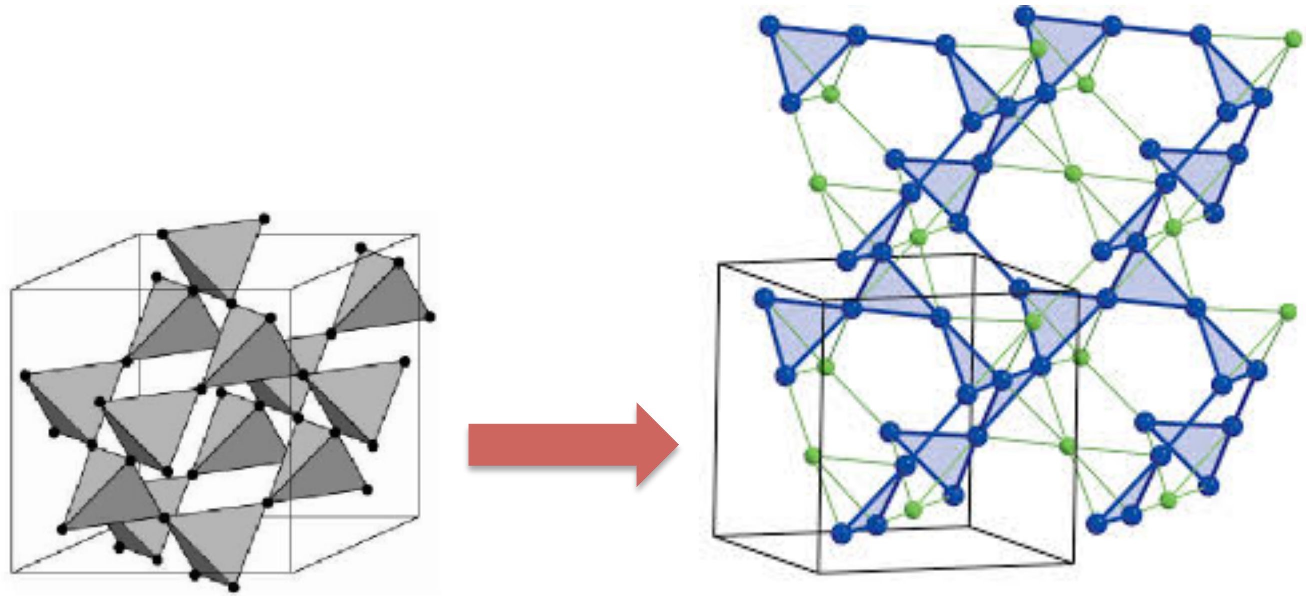






Hyper-Kagome : $Na_4Ir_3O_8$

The Hyperkagome magnetic Sub-Lattice is a depleted Pyrochlore

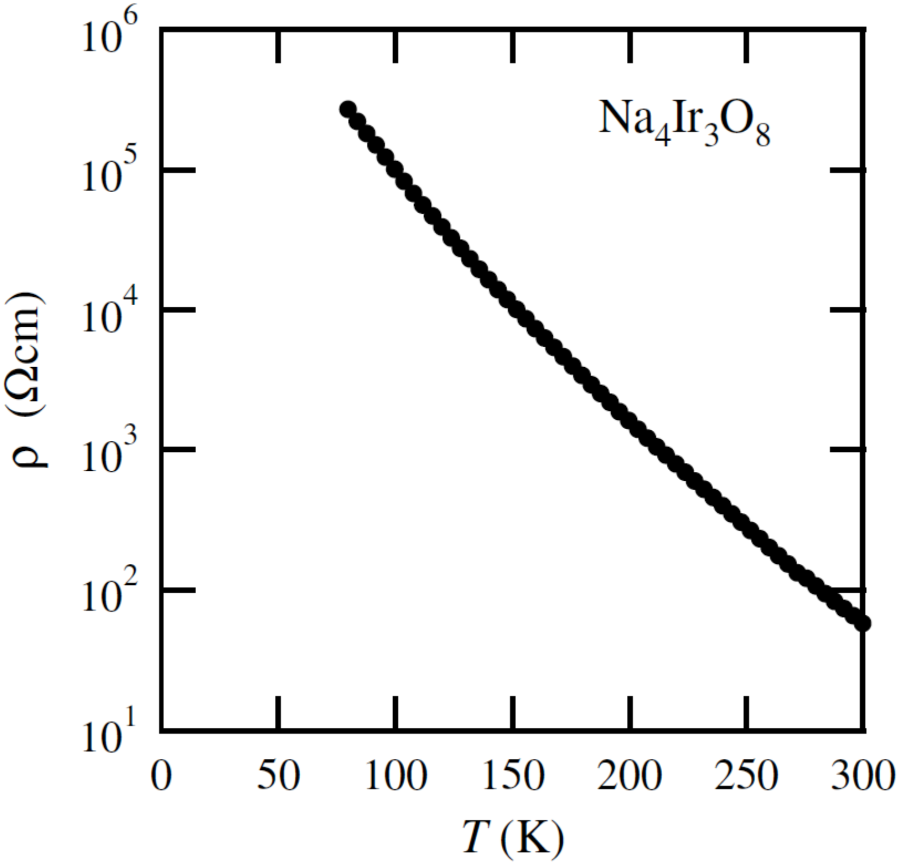


Ir4+ ions form a twisted Kagome network of corner shared triangles

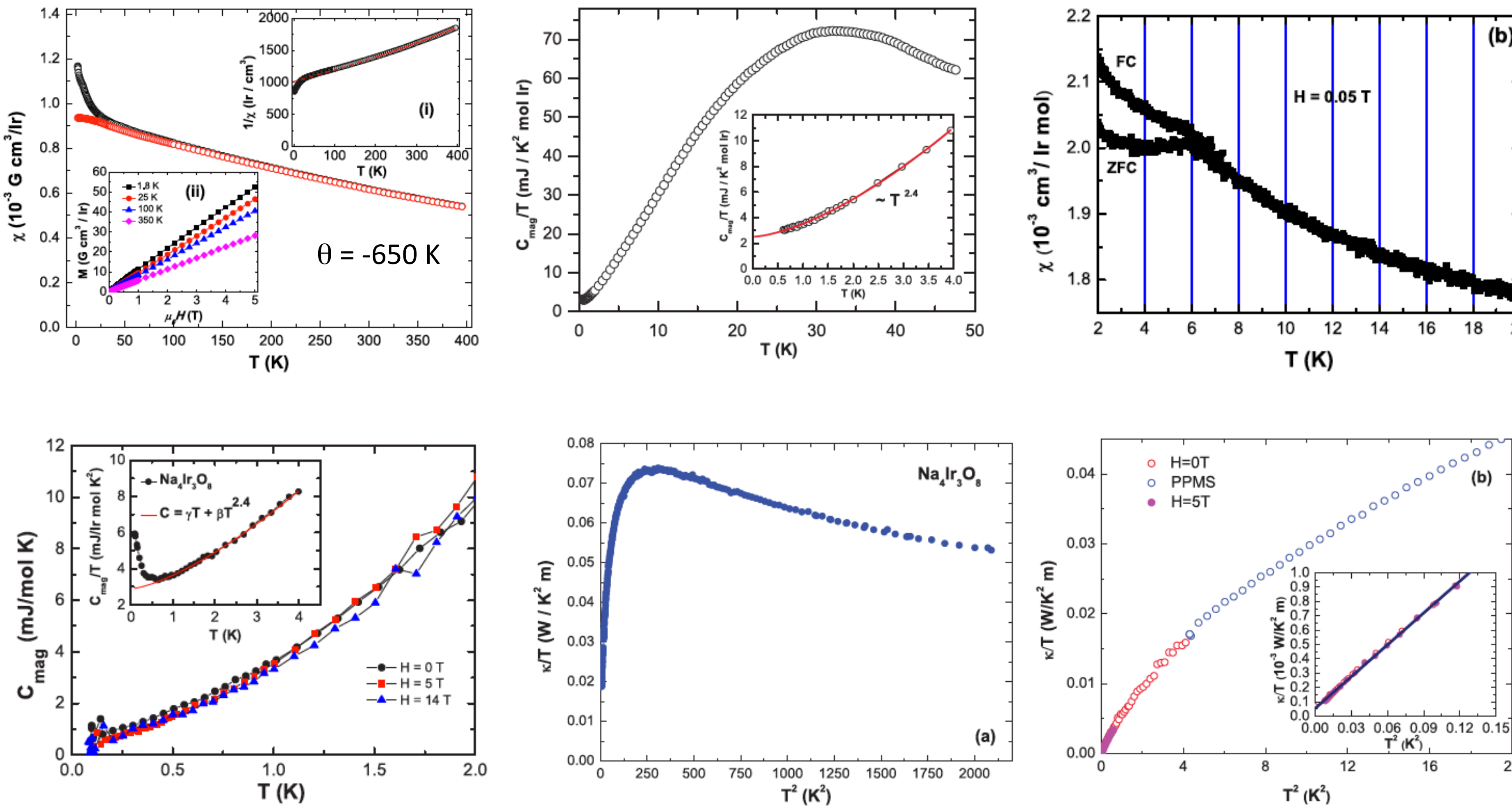
$Na_4Ir_3O_8$: Resistivity

Insulator but near MIT

$\Delta \approx 500 - 900$ K

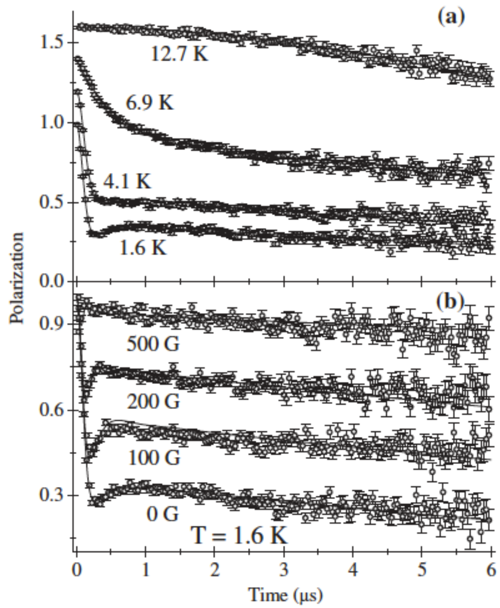


$Na_4Ir_3O_8$: Magnetic and Thermal Properties

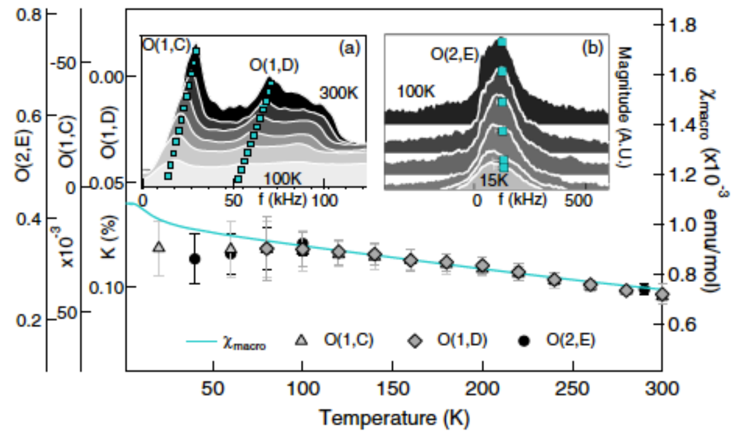


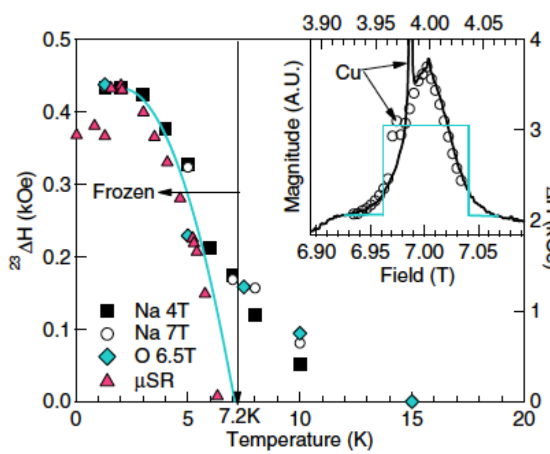
Dichotomy between χ and C/T (Gapless), and κ/T (Gapped)

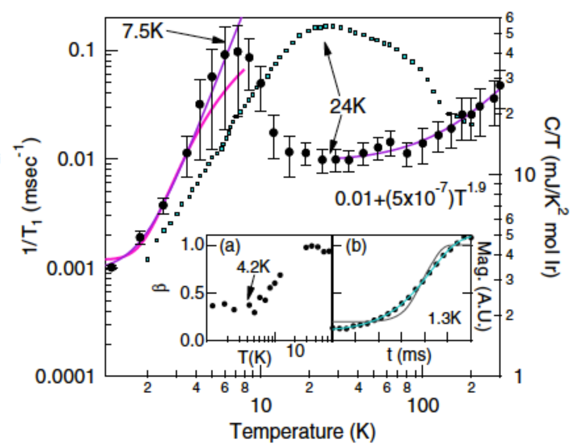
$Na_4Ir_3O_8$: Inhomogeneous Frozen state revealed by muSR and NMR



Dally *et al.* PRL **113**, 247601 (2014).







Shockley *et al.* PRL **115**, 047201 (2015).

Kagome bi-layer QSL $Ca_{10}Cr_7O_{28}$

C. Balz, B. Lake, J. Reuther, Y. Singh... Nature Physics **12**, 942 (2016)

C. Balz, B. Lake, Y. Singh…. Physical Review B **95**, 174414 (2017)

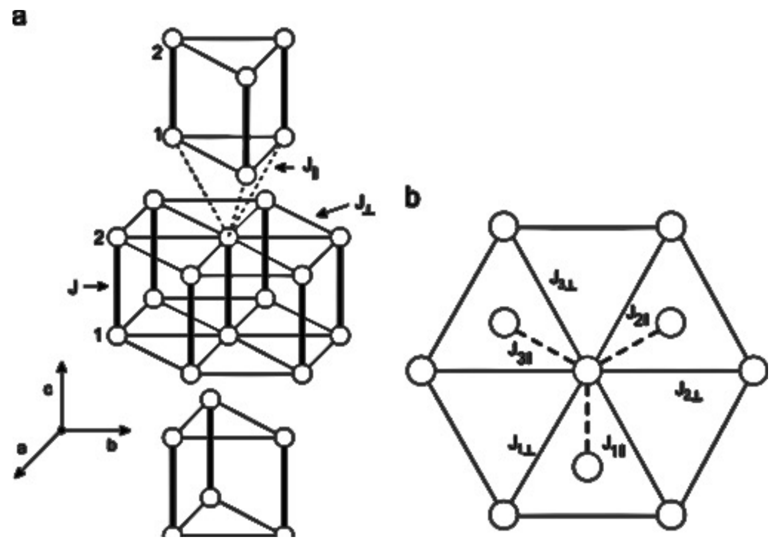
C. Balz, B. Lake, Y. Singh…. J. of Phys. Cond. Matt. **29**, 225802 (2017)

A. Balodhi and Y. Singh, Physical Review Materials **1**, 024407 (2017)

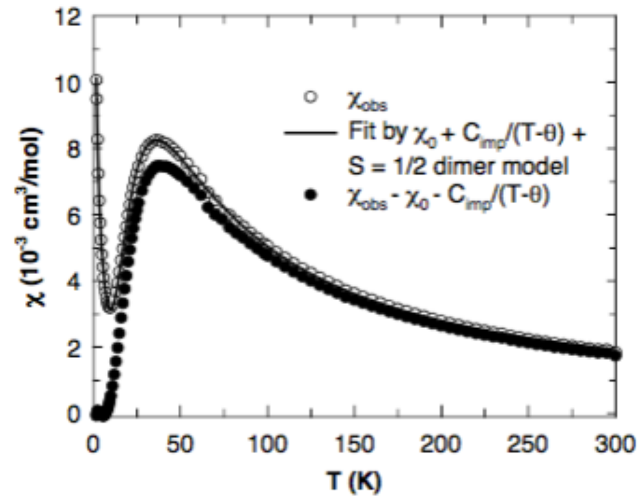
Route to the Material $Ca_{10}Cr_7O_{28}$

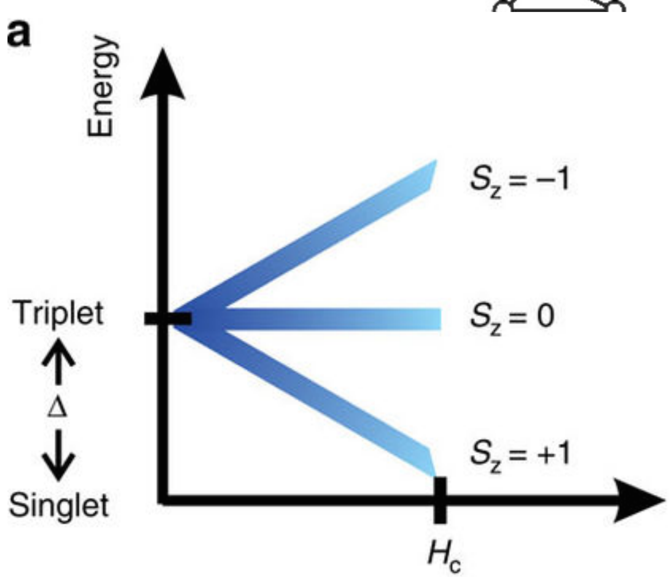
Family of bilayer triangular magnets $A_3T_2O_8$ (A = Ba, Sr, T = Mn, Cr)

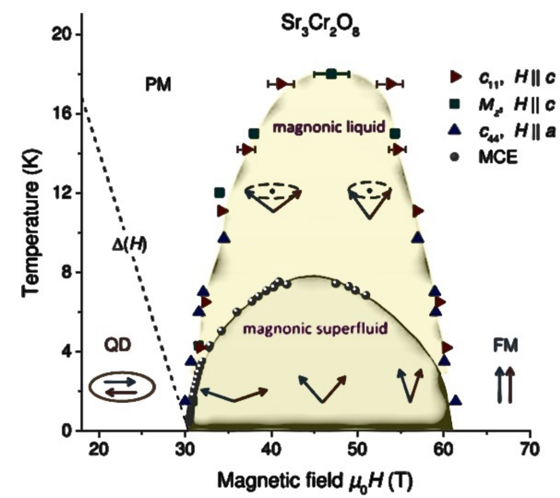
Cr^{5+}, S = ½



Yogesh Singh and D. C. Johnston, PRB 76, 012407 (2007)





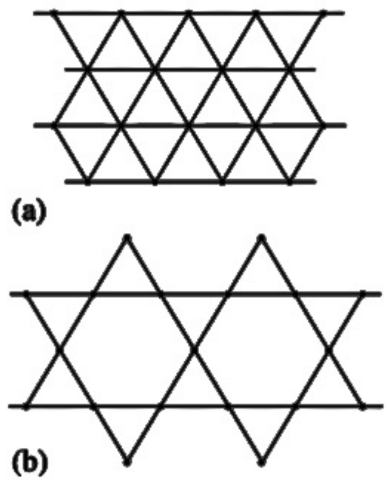


Route to the Material $Ca_{10}Cr_7O_{28}$

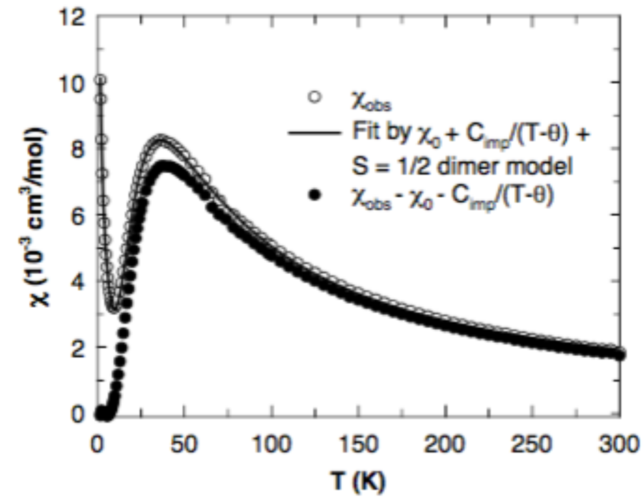
$Ca_3T_2O_8 = Ca_{10.5}Cr_7O_{28}$

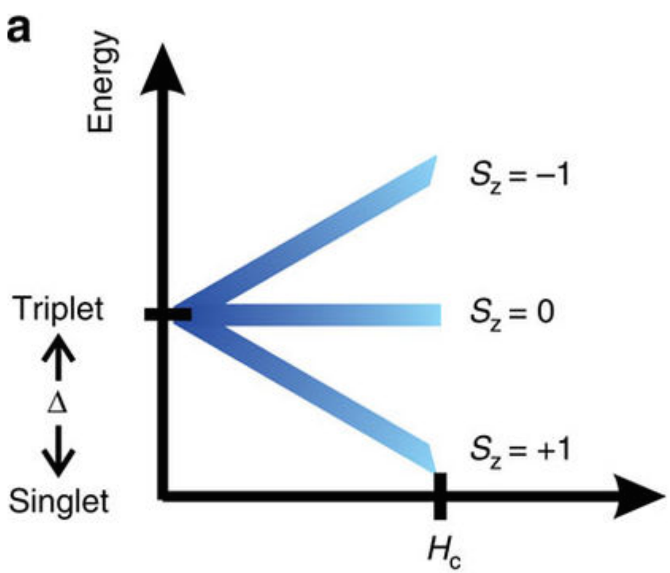
$Ca_{10}Cr_7O_{28}$

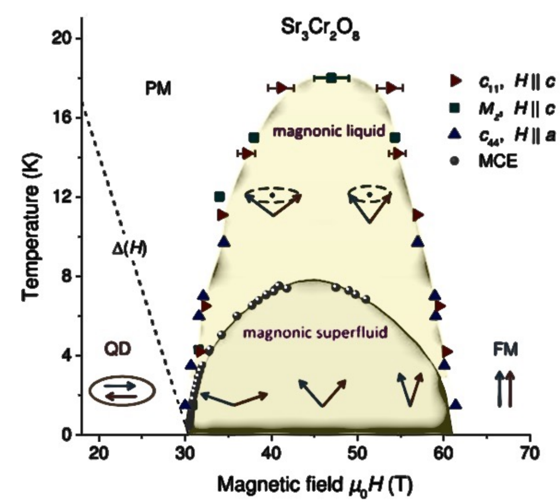
6 Cr^{5+} (S = ½) and 1 Cr^{6+} (S = 0)



Yogesh Singh and D. C. Johnston, PRB 76, 012407 (2007)

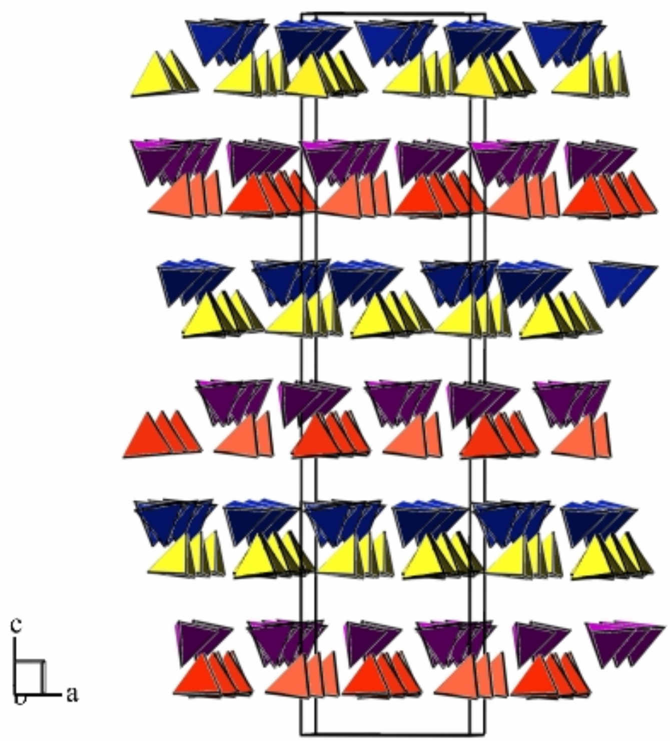


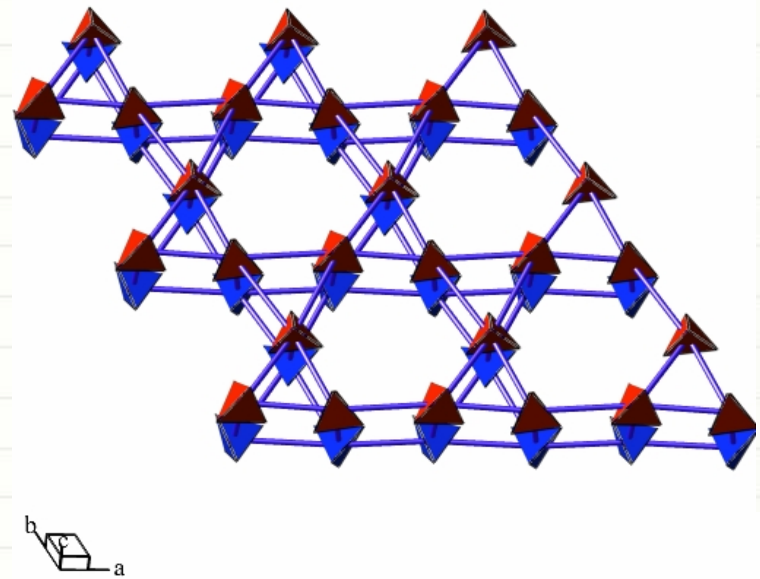


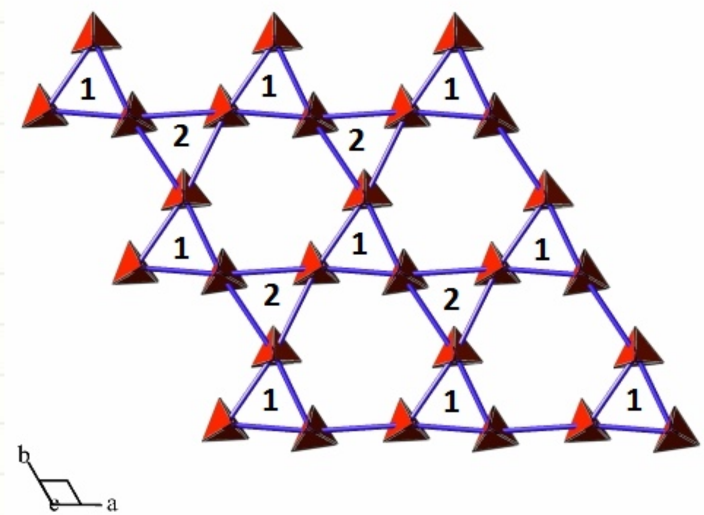


Crystal Structure of $Ca_{10}Cr_7O_{28}$

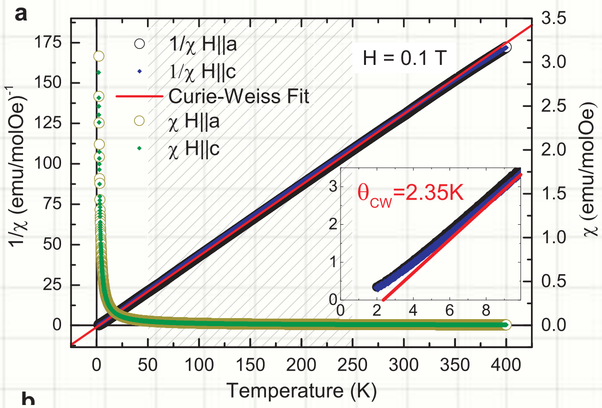
Lattice Parameters		Space Group
a	10.76892(3) Å	$R3c$
c	38.09646(12) Å	



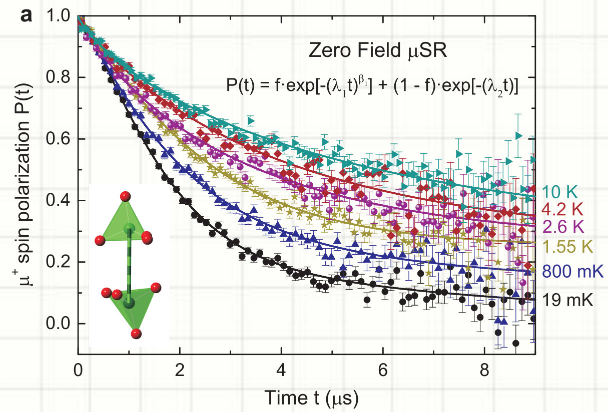


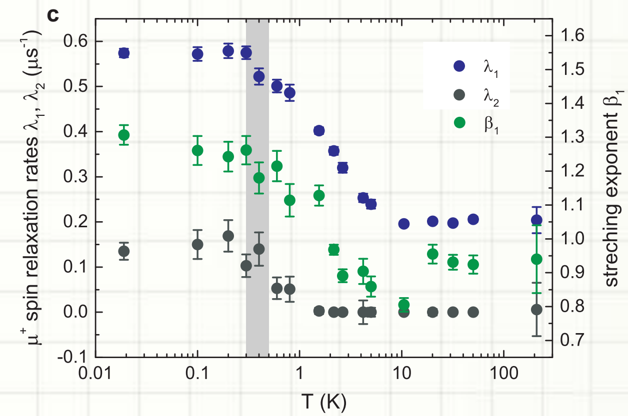


Gapless Spin Liquid State in $Ca_{10}Cr_7O_{28}$

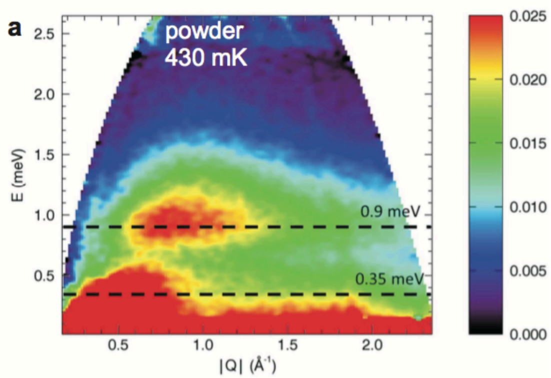


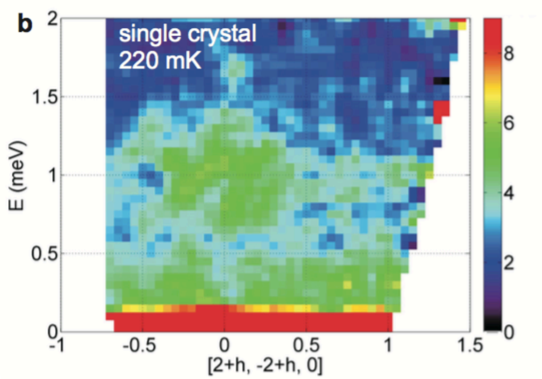
χ



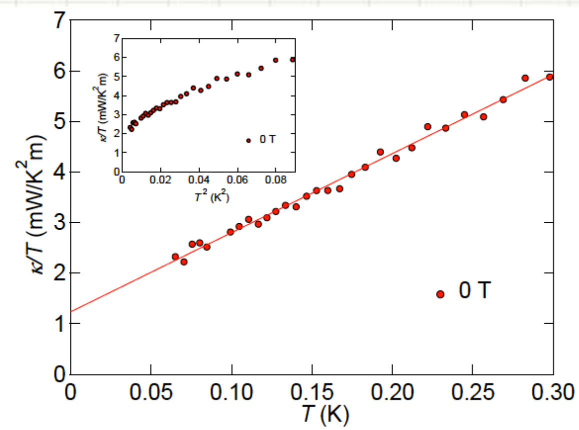


μSR



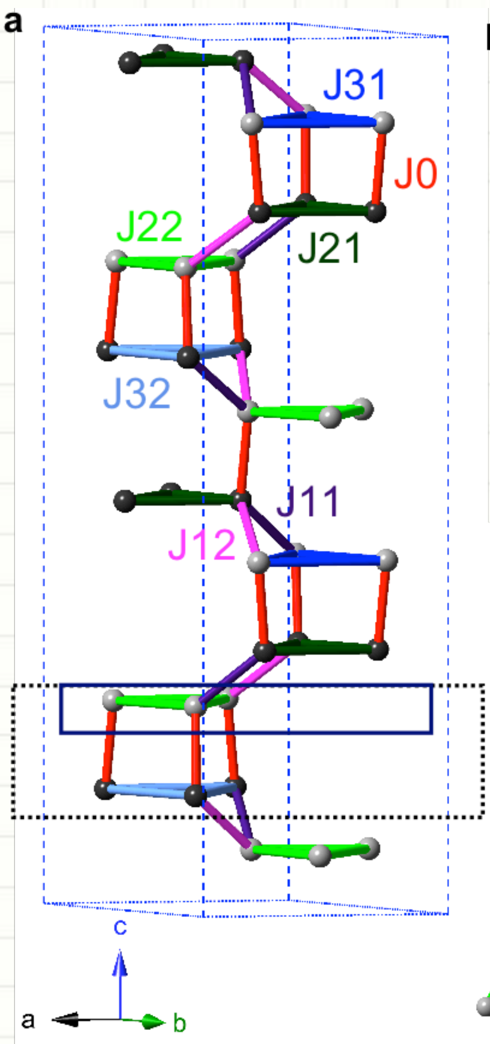


INS



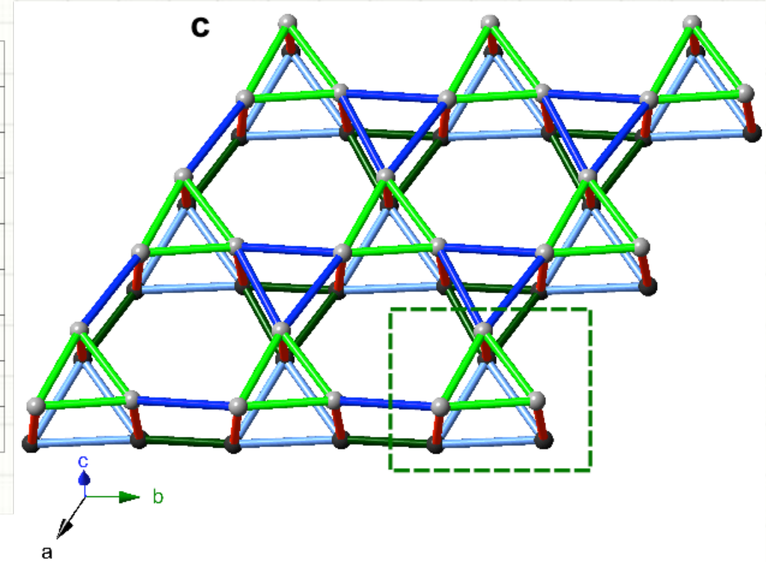
κ

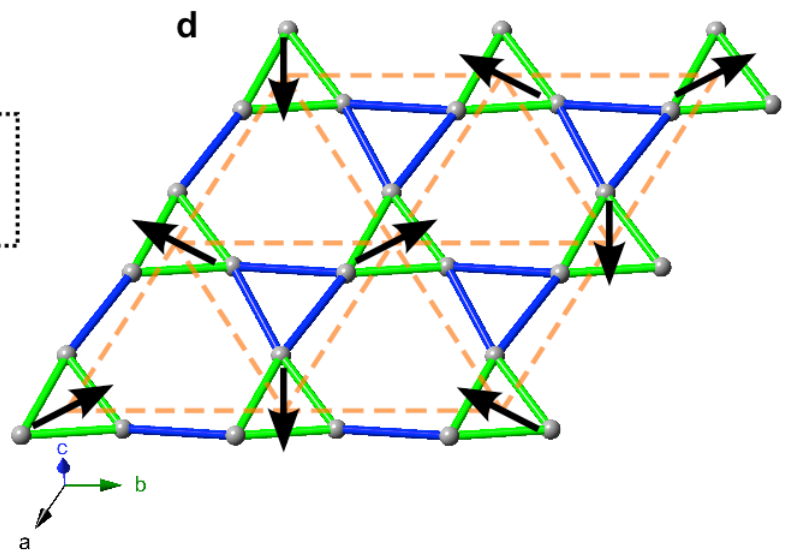
Magnetic Interactions : Novel Frustration Mechanism

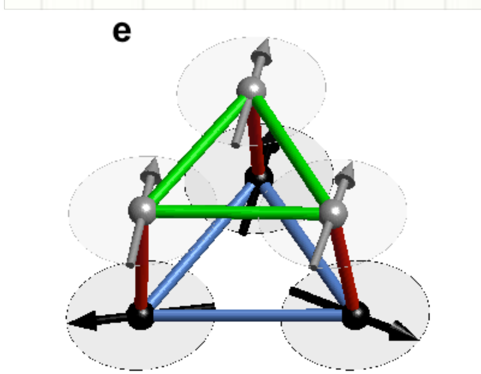


b

exchange	coupling [meV]	type
J0	-0.08(4)	FM
J11	0	
J12	0	
J21	-0.76(5)	FM
J22	-0.27(3)	FM
J31	0.09(2)	AFM
J32	0.11(3)	AFM
ΣJ	-0.91(17)	







Summary

- Geometrically Frustrated Magnets are interesting….
- Novel Magnetic states
- Magnetization Plateaus
- BEC of triplons
- QSLs
- Fractionlization
- Kitaev Magnets : Majorana Fermions
- ………..
- ………