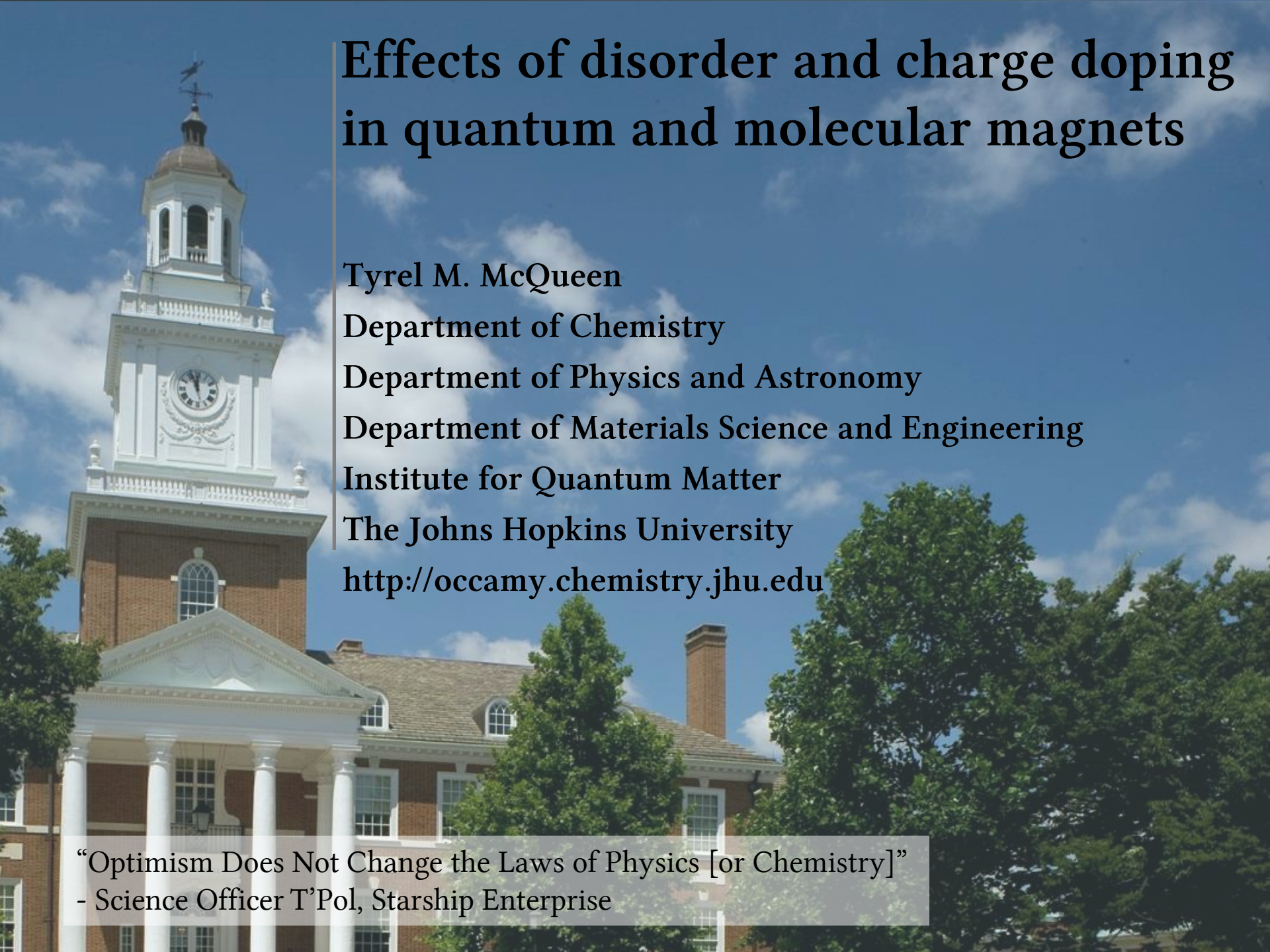

Effects of disorder and charge doping in quantum and molecular magnets

Tyrel M. McQueen

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http://occamy.chemistry.jhu.edu

"Optimism Does Not Change the Laws of Physics [or Chemistry]"
- Science Officer T'Pol, Starship Enterprise

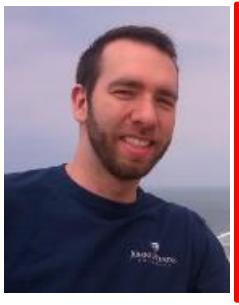
Outline

- The experimental history of layered frustrated magnets
- Three complementary examples:
  - $NaVO_2$ (Orbital ordering relieves frustration)
  - $Zn_{1-\delta}Cu_{3+\delta}(OH)_6Cl_2$ (Magnetic disorder between layers)
  - $LiZn_2Mo_3O_8$ (Non-magnetic disorder between layers)
- Effect of electron count on the properties: is there superconductivity?
- Last remarks

Acknowledgements



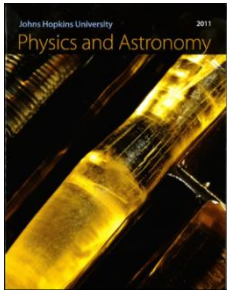


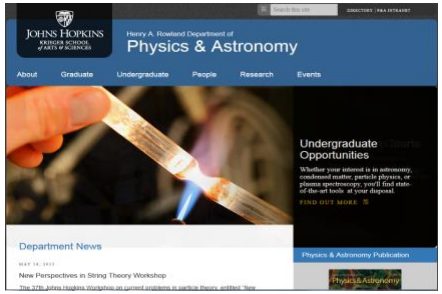



John Sheckelton







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- Prof. Takashi Imai (McMaster)
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- Dr. Matthew Suchomel (APS)
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- Prof. Z. Tesanovic (JHU/IQM)
- Dr. Natalia Drichko (JHU)
- All collaborators on other projects

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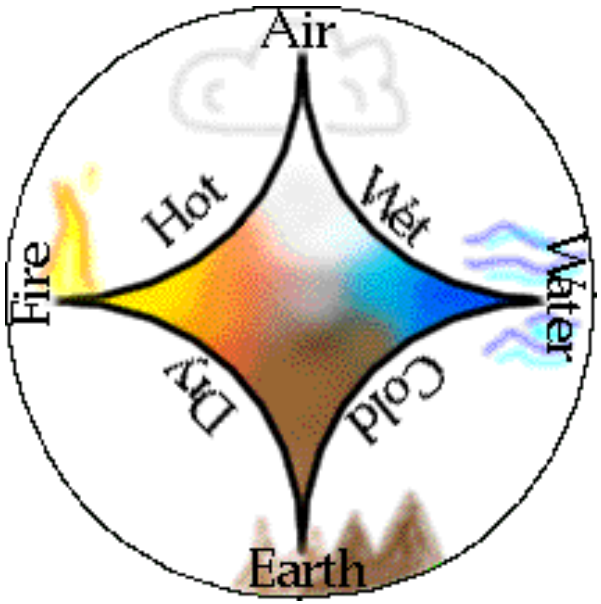
Magnets and Molecules go *way* back

Lodestone (circa 600 BC)

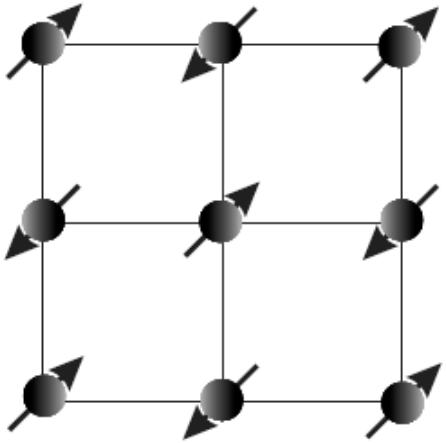


- 600 BC recorded that iron attracts lodestone: Aristotle. De Anima (On the Soul). Book I, part 2
- Modern view started: 1819, Oersted's experiment

- 450 BC Empedocles imagined fundamental elements (fire, earth, air, water)
- Modern view started: 1661, Robert Boyle's The Sceptical Chymist

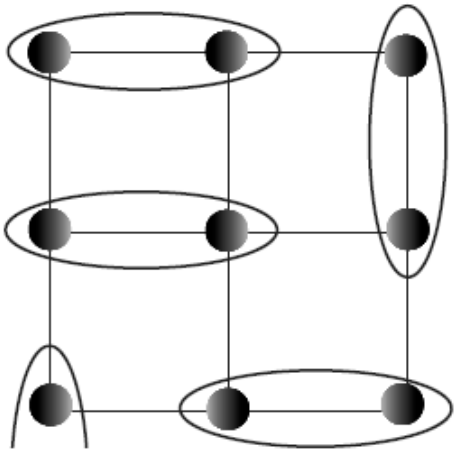
http://www.magnet.fsu.edu/education/tutorials/museum/lodestone.html
http://www-pdg.lbl.gov/cpep/four_elem_ans.html

Resonating Valence Bonds and Spin Liquids



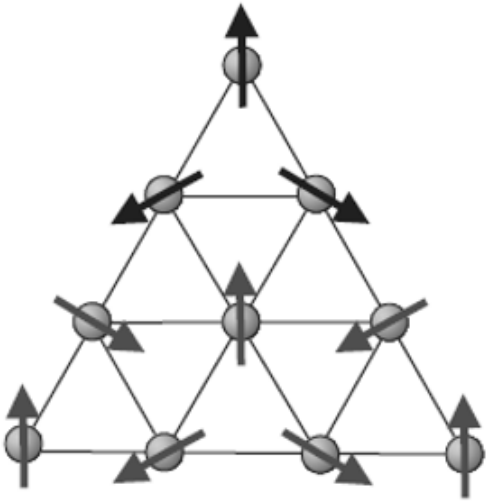
Neél order

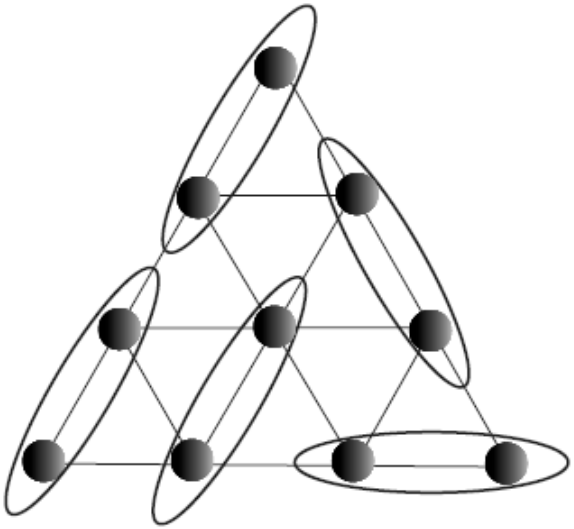
(static, classical)



Quantum spin-liquid state

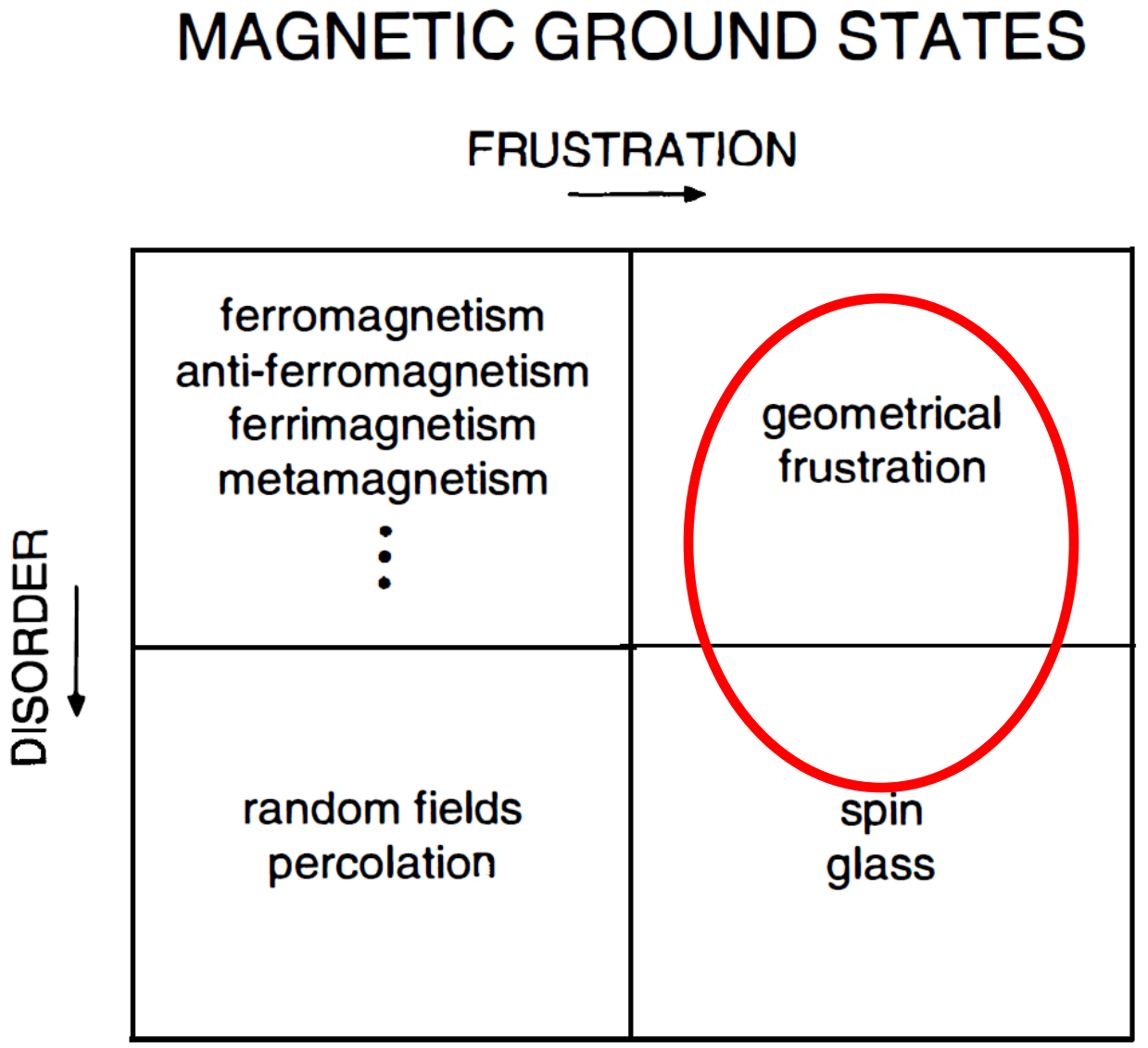
$$H = J \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j$$



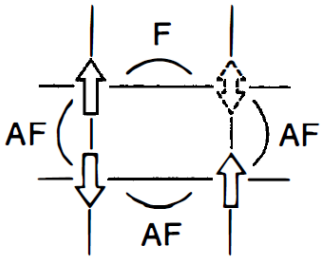


P.W. Anderson, "Resonating Valence Bonds: A New Kind of Insulator?", Mat. Res. Bull. 8, 153-160 (1973)
R. Moessner and S.L. Sondhi, "Resonating Valence Bond Liquid Physics on the Triangular Lattice,", Prog. Theor. Phys. (2002)

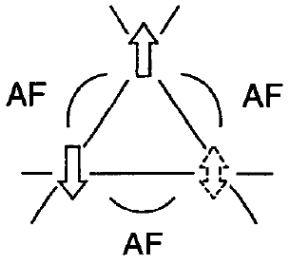
Magnetic Frustration



"SPIN GLASS" FRUSTRATION

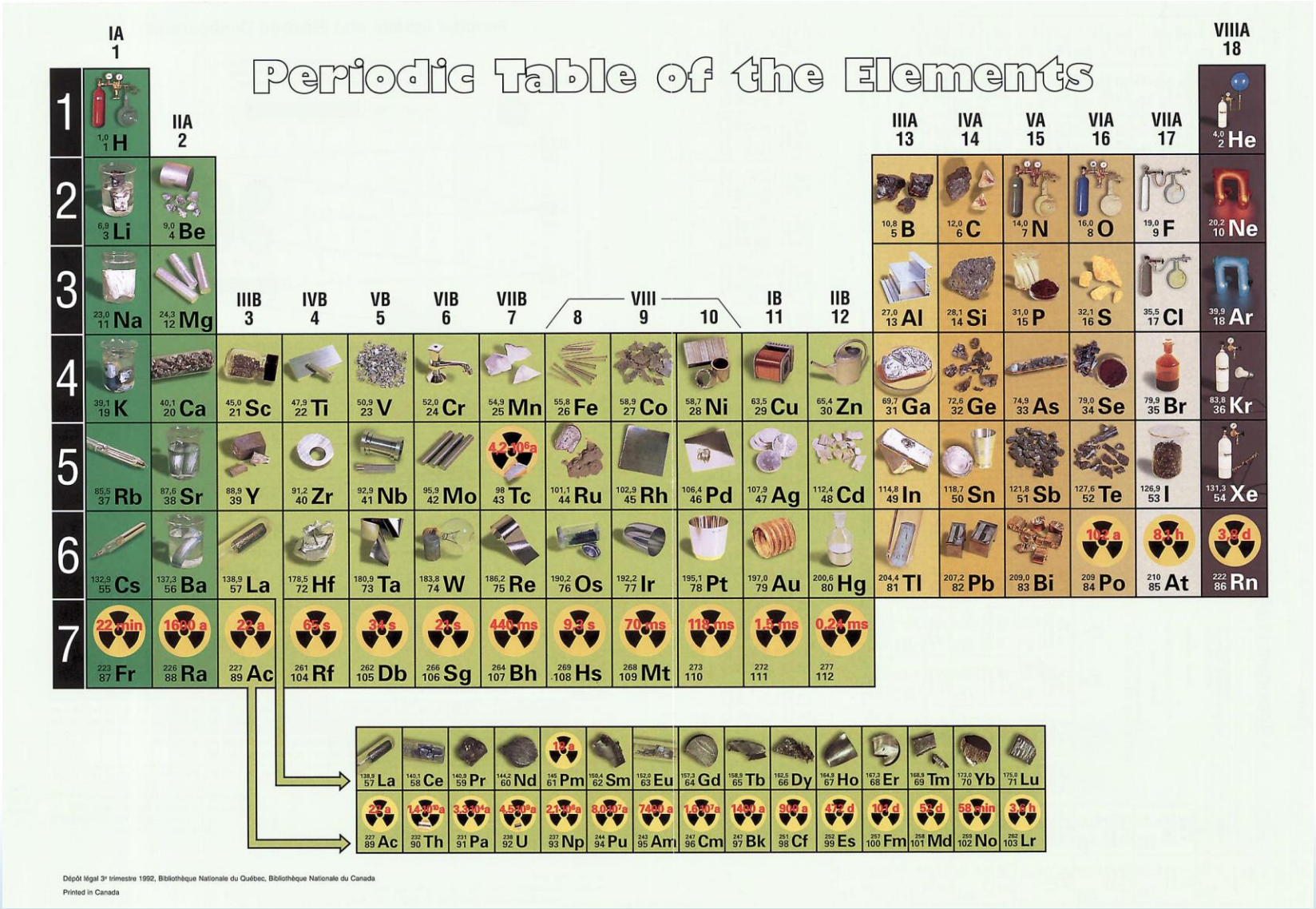


GEOMETRICAL FRUSTRATION



A.P. Ramirez, "Strongly Geometrically Frustrated Magnets," Annu. Rev. Mater. Sci. 24, 453-80 (1994).

The Experimentalist's Building Blocks



Bibliotheque Nationale du Quebec, Bibliotheque National du Canada (1992)

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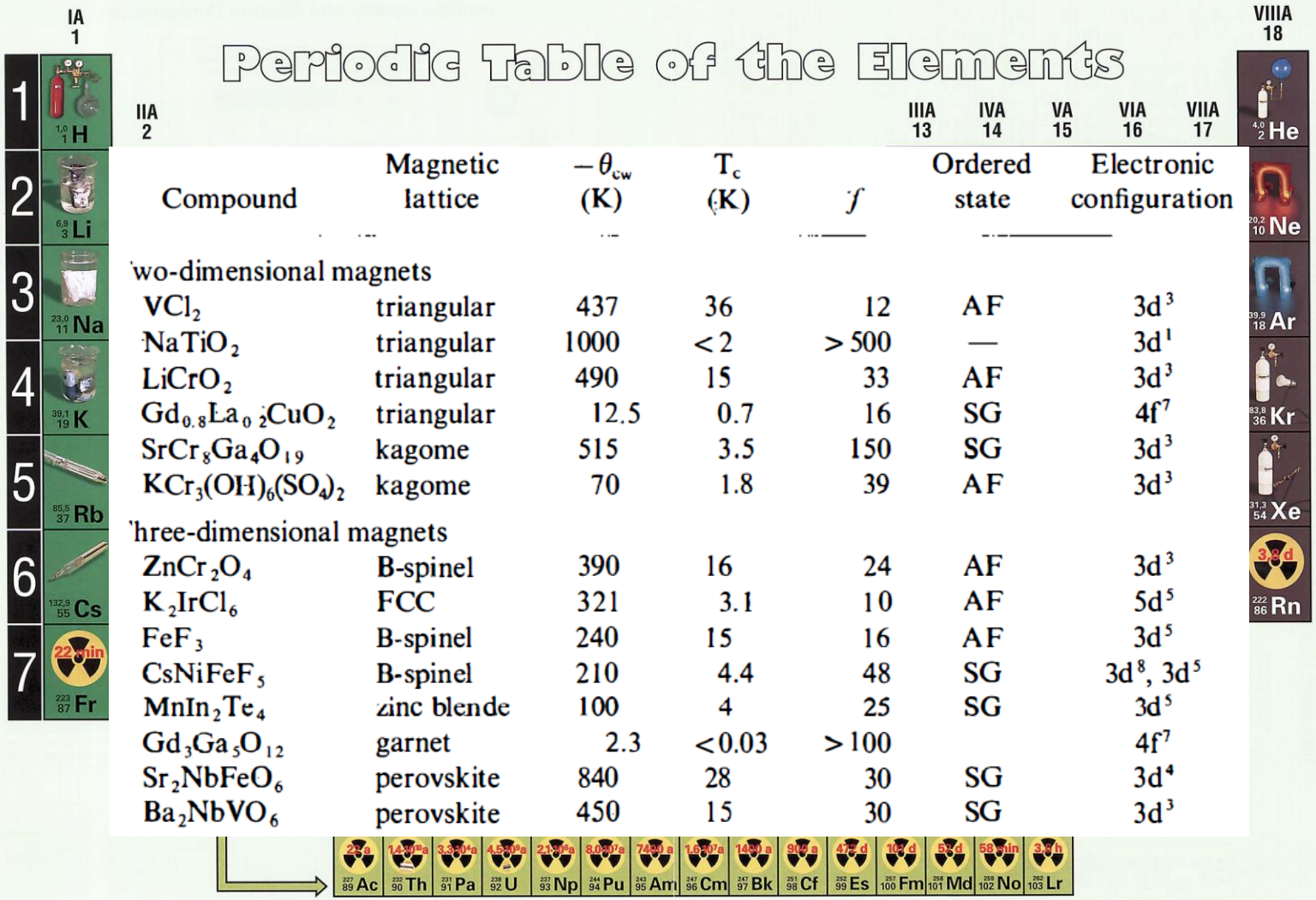
The Experimentalist's Building Blocks



Compound	Magnetic lattice	$-\theta_{cw}$ (K)	T_c (K)	f	Ordered state	Electronic configuration
Two-dimensional magnets						
VCl_2	triangular	437	36	12	AF	$3d^3$
$NaTiO_2$	triangular	1000	<2	>500	—	$3d^1$
$LiCrO_2$	triangular	490	15	33	AF	$3d^3$
$Gd_{0.8}La_{0.2}CuO_2$	triangular	12.5	0.7	16	SG	$4f^7$
$SrCr_8Ga_4O_{19}$	kagome	515	3.5	150	SG	$3d^3$
$KCr_3(OH)_6(SO_4)_2$	kagome	70	1.8	39	AF	$3d^3$
Three-dimensional magnets						
$ZnCr_2O_4$	B-spinel	390	16	24	AF	$3d^3$
K_2IrCl_6	FCC	321	3.1	10	AF	$5d^5$
FeF_3	B-spinel	240	15	16	AF	$3d^5$
$CsNiFeF_5$	B-spinel	210	4.4	48	SG	$3d^8$, $3d^5$
$MnIn_2Te_4$	zinc blende	100	4	25	SG	$3d^5$
$Gd_3Ga_5O_{12}$	garnet	2.3	<0.03	>100		$4f^7$
Sr_2NbFeO_6	perovskite	840	28	30	SG	$3d^4$
Ba_2NbVO_6	perovskite	450	15	30	SG	$3d^3$

Dépôt légal 3e trimestre 1992, Bibliothèque Nationale du Québec, Bibliothèque Nationale du Canada
Printed in Canada

Bibliotheque Nationale du Quebec, Bibliotheque National du Canada (1992)

A.P. Ramirez, "Strongly Geometrically Frustrated Magnets," Annu. Rev. Mater. Sci. 24, 453-80 (1994).

Zoo of Triangle-Based Magnets

Table I. Planar lattices based on Edge-sharing Equilateral triangles

Compound	Structure type	Stacking Type	M-M in-plane	M-M next plane
VCl_2	CdI_2	Eclipsed	3.60	5.84
$CuCoO_2$	Delafossite 3-layer	Staggered	2.85	5.95
$CuFeO_2$	Delafossite 2-layer	Eclipsed	3.04	5.73
$NaCrO_2$	α-$NaFeO_2$	Staggered	2.98	5.59
$Fe_{1/3}NbS_2$	intercalated dichalcogenide	Staggered	5.76	6.95
$Cr_{1/4}NbS_2$	intercalated dichalcogenide	Eclipsed	6.63	5.99
$CsNiCl_3$	$BaNiO_3$	Eclipsed	7.17	2.97
$RbFe(SO_4)_2$	anhydrous Alum	Eclipsed	4.82	8.22
$NiGa_2S_4$	$NiGa_2S_4$	Eclipsed	3.63	12.00

Table II. Planar lattices based on Edge-sharing of non-equilateral triangles

Compound	Structure Type	Triangle Type	Stacking Type	M-M in-plane	M-M next plane
Cs_2CuCl_4	Cs_2MCl_4	isosceles	Staggered	7.60, 7.26	6.81
$Cu_2(OH)_3Cl$	Botallackite	scalene	Eclipsed	3.06, 3.18, 3.23	5.72
$KFe(SO_4)_2$	anhydrous Alum	isosceles	Eclipsed	4.79, 5.14	7.88
$NaNiO_2$	distorted α-$NaFeO_2$	isosceles	Staggered	2.84, 3.02	5.47, 5.58

Table III. Planar lattices based on corner-sharing triangles

Compound	Structure Type	Layer Type	Stacking Type	M-M in plane	M-M next plane
$NH_4Fe_3(SO_4)_2(OH)_6$	Jarosite	Kagomé	Eclipsed	3.65	6.13
$ZnCu_3(OH)_6Cl_2$	Paratacamite	Kagomé	Staggered	3.39, 3.43	5.05, 5.15
$Cu_3V_2O_7(OH)_2.2H_2O$	Volborthite	Kagomé	Eclipsed	2.93, 3.03	7.21
$Ni_3V_2O_8$	$Co_3V_2O_8$	Kagomé Staircase	Staggered	2.94, 2.96	5.71
Mn_2SiO_4	Olivine	isosceles sawtooth	Staggered	3.12, 3.35	3.69, 3.76, 4.02
$ZnTm_2S_4$	Olivine	isosceles sawtooth	Staggered	3.89, 4.04	4.81 4.85, 5.02

Table IV. Planar Lattices based on Honeycombs

Compound	Layer Type	Stacking Type	MM in-plane	M-M next-plane
$BaNi_2V_2O_8$	regular	Staggered	2.91	7.99, 7.87
$Na_3Co_2SbO_6$	regular	Eclipsed	3.10	5.65
$Na_2Cu_2TeO_6$	triangular lattice of dimers	Staggered	2.86, 3.21	5.65

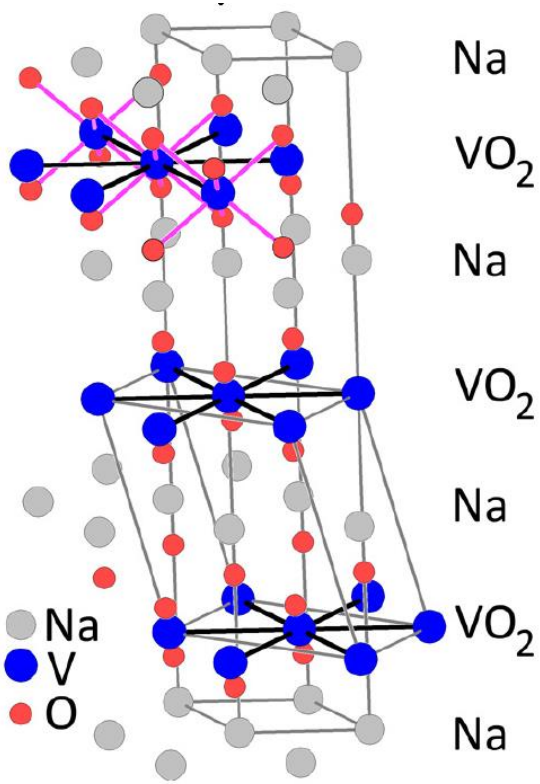
Table V. Three-dimensional Materials with triangle-based lattices

Compound	Structure Type	Lattice Type	M-M in-triangle	M-M next nearest
Ba_2HoSbO_6	Double Perovskite	edge-sharing tetrahedra (FCC)	5.93	
Sr_2MgIrO_6	Double Perovskite	edge-sharing tetrahedra (FCC)	5.58	
$Dy_2Ti_2O_7$	Pyrochlore A site	corner-sharing tetrahedra	3.58	
$Y_2Mo_2O_7$	Pyrochlore B site	corner-sharing tetrahedra	3.62	
$Dy_{2+x}Ti_{2-x}O_{7-x/2}$	Stuffed Pyrochlore	corner to edge-sharing tetrahedra	3.58-3.66	
$ZnCr_2O_4$	Spinel (B site)	corner-sharing tetrahedra	2.86	
$CdDy_2S_4$	Spinel (B site)	corner-sharing tetrahedra	3.98	
$MnSc_2S_4$	Spinel (A site)	Diamond	4.59	7.50 (2nd neighbor)
$SrGa_4Cr_8O_{19}$	M-type Ferrite	Kagomé + tetrahdera +pyramid connectors	2.86	2.99 (cap of tetrahedron) 3.44, 2.68 (to connectors)
$Ba_2Sn_2Ga_3ZnCr_7O_{22}$	QS ferrite	Kagomé +tetrahedra	2.87, 2.98	9.49, 9.86 (interlayer) 3.05 (cap of tetrahedron)
$SrCo_6O_{11}$	R-type Ferrite	Kagomé +connector	2.81	6.29 (interlayer)
$YBaCo_4O_7$	spinel-like	Kagomé + tetrahedra	3.03, 3.25	3.05 (cap of tetrahedron)
$Tb_3Ga_5O_{12}$	Garnet	interpenetrating lattices of corner-sharing triangles	3.78	5.78 (interlattice)

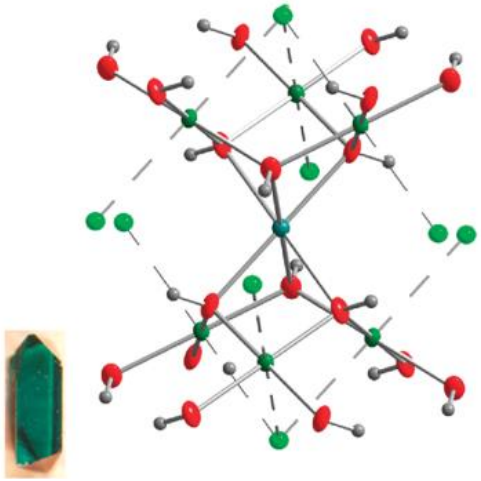
Cava, R.J., K.L. Holman, T.M. McQueen, E.J. Welch, D.V. West, and A.J. Williams, "The Geometries of Triangular Magnetic Lattices," in Introduction to Frustrated Magnetism, Springer: 2011. ISBN: 978-3-642-10588-3.

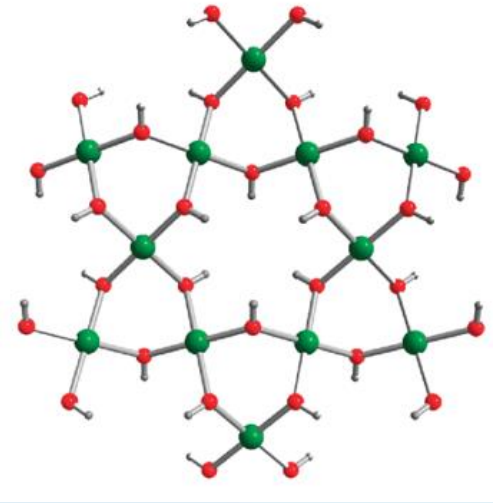
The Research Vignettes for Today

$NaVO_2$

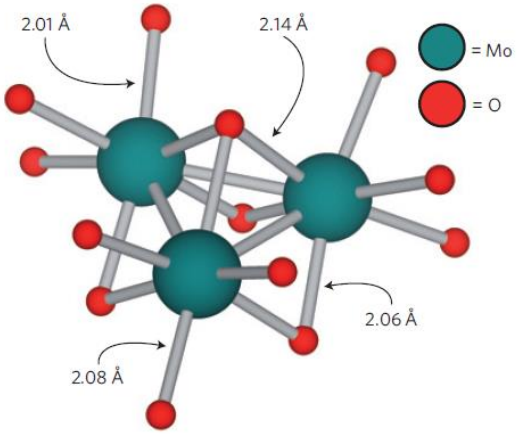


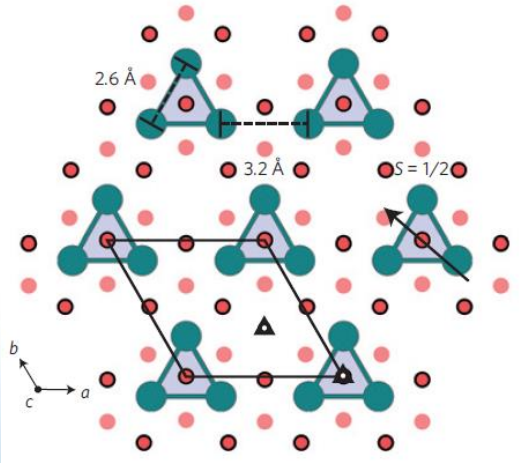
$ZnCu_3(OH)_6Cl_2$



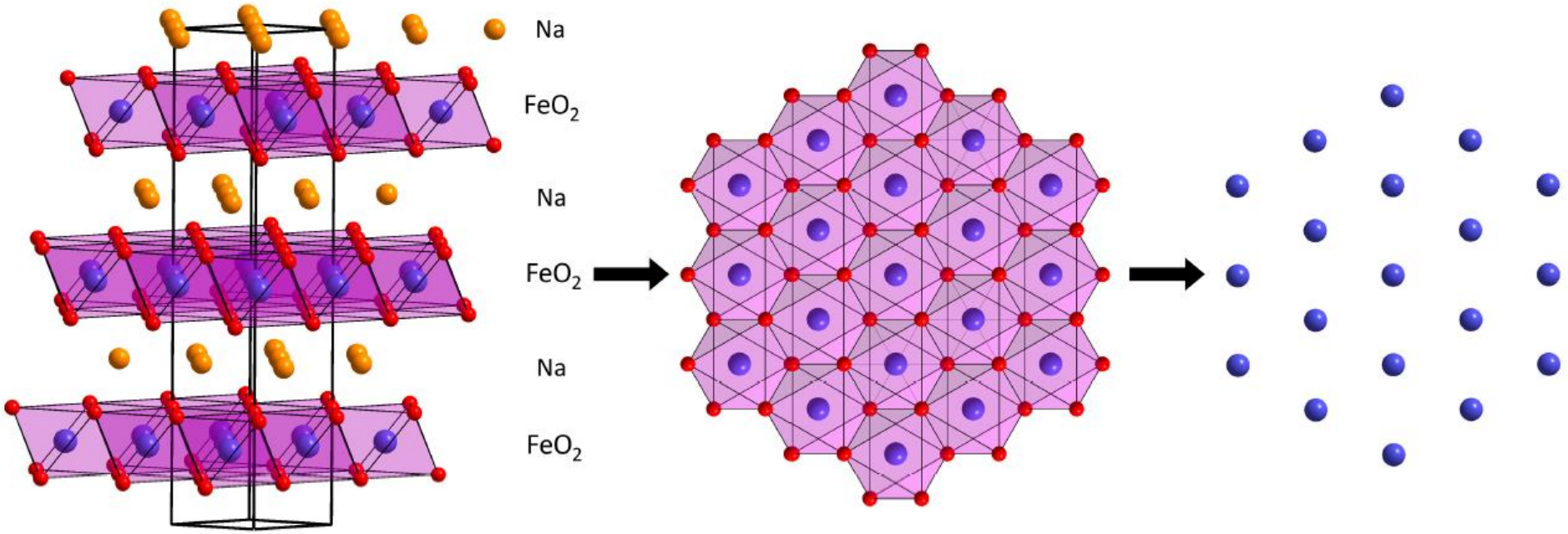


$LiZn_2Mo_3O_8$





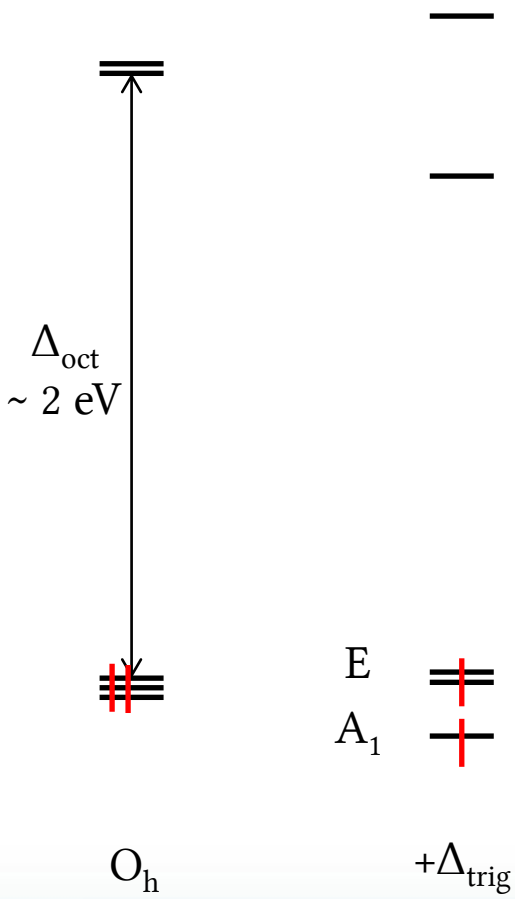
AMO_2 (A = Li,Na,…, M = Ti^{3+}, V^{3+},…)

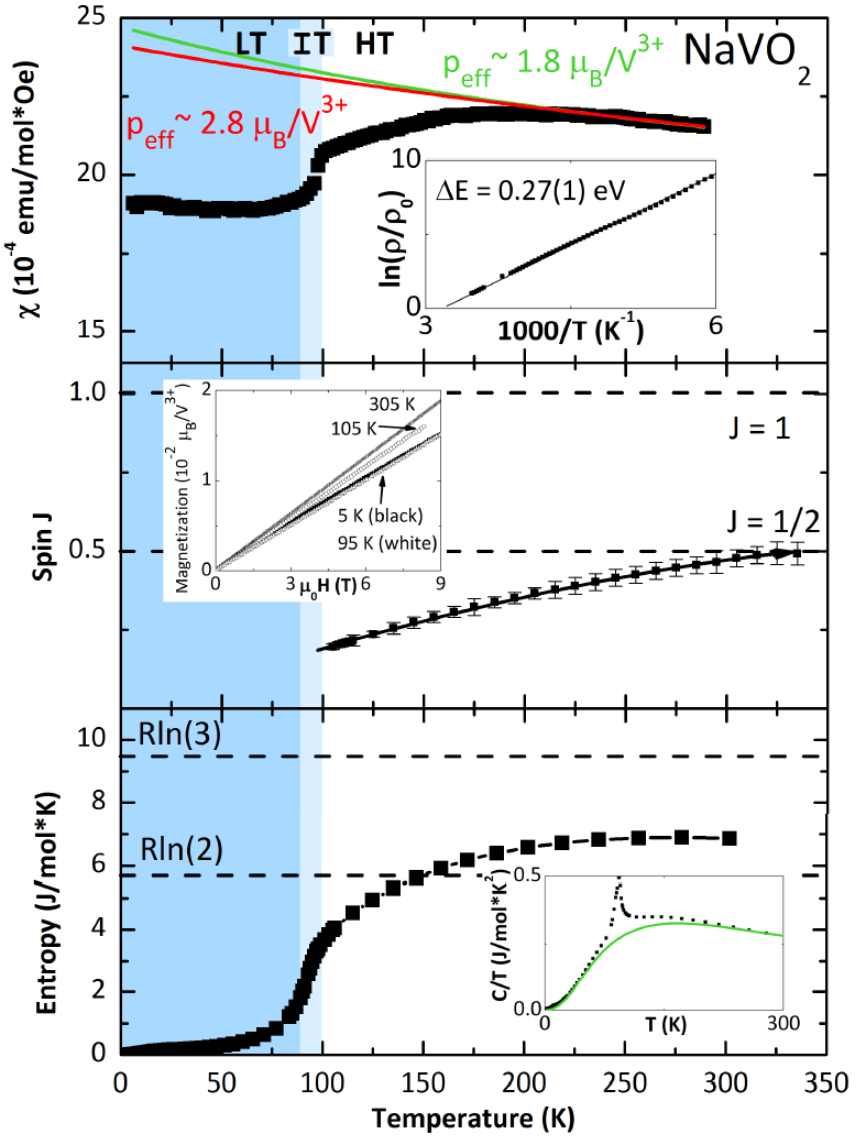


Compound	Magnetic Ordering	Obs. Mag. Moment	Expected Mag. Moment (spin-only)
$NaScO_2$	boring non-magnet	-	-
$NaTiO_2$	no long range order	-	1 μ_B/Ti^{3+}
$NaVO_2$	*3D order*	*1.0 μ_B/V^{3+}*	*2 μ_B/V^{3+}*
$NaCrO_2$	short range order	-[1]	3 μ_B/Cr^{3+}
$NaMnO_2$	3D order	3.0 μ_B/Mn^{3+}	4 μ_B/Mn^{3+} (HS)
$NaFeO_2$	*3D order*	*4.3 μ_B/Fe^{3+}*	*5 μ_B/Fe^{3+} (HS)*
$NaNiO_2$	3D order	1 μ_B/Ni^{3+}	1 μ_B/Ni^{3+} (LS)

Smaller than Expected Moment, Rich Phases

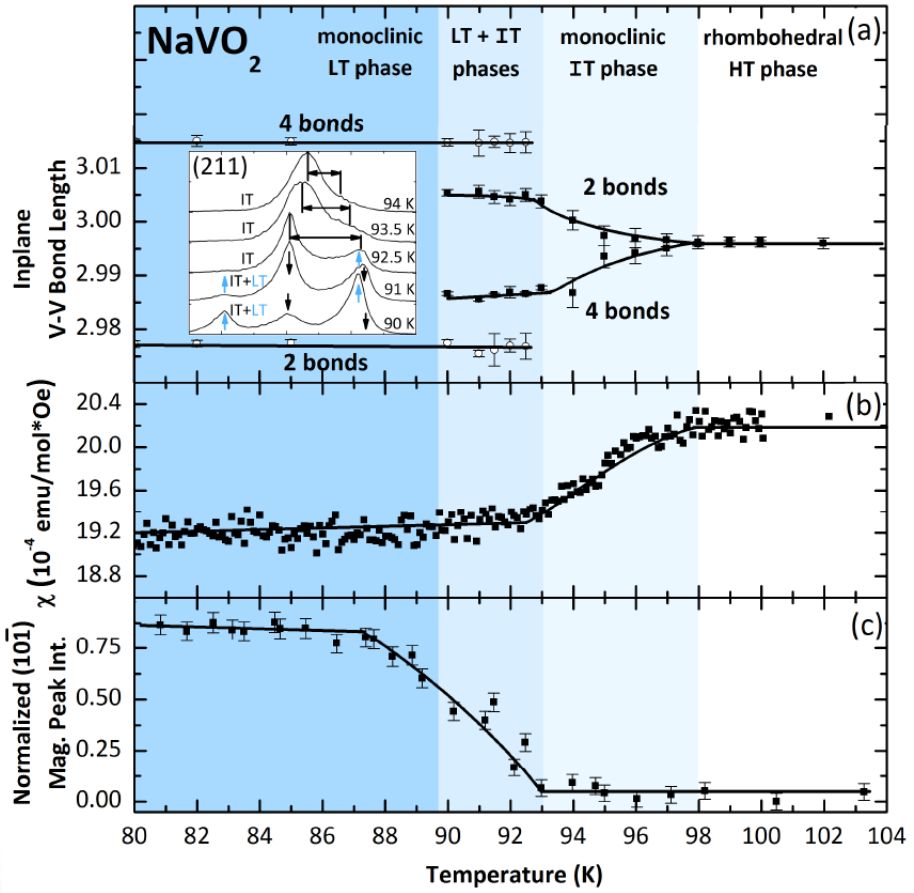
V^{3+}: $(t_{2g})^2$ (S = 1)

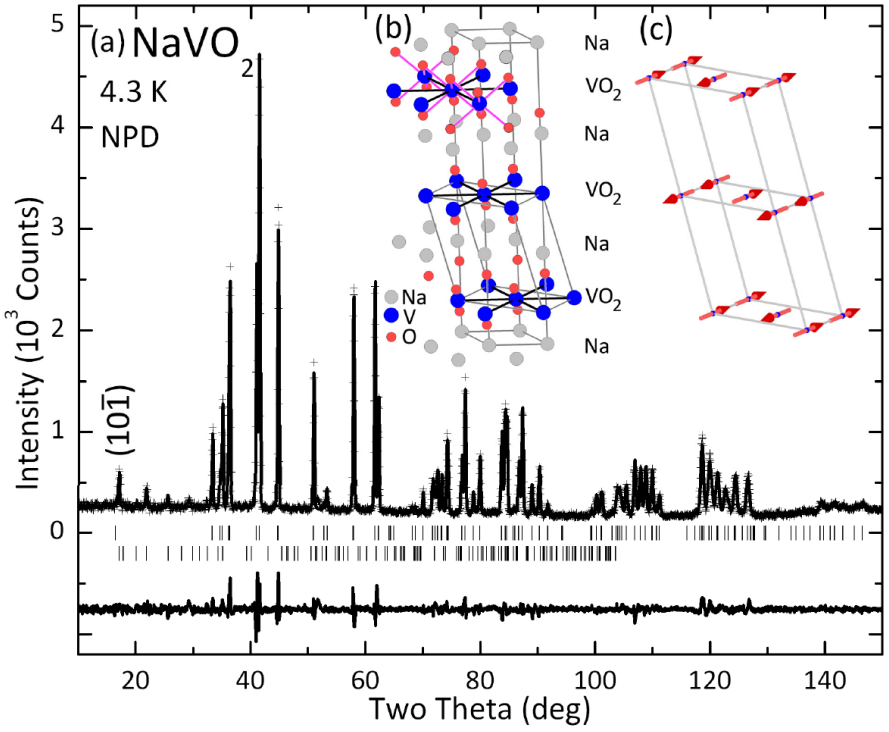




T.M. McQueen, et al. *Phys. Rev. Lett.* **101**, 166402 (2008)

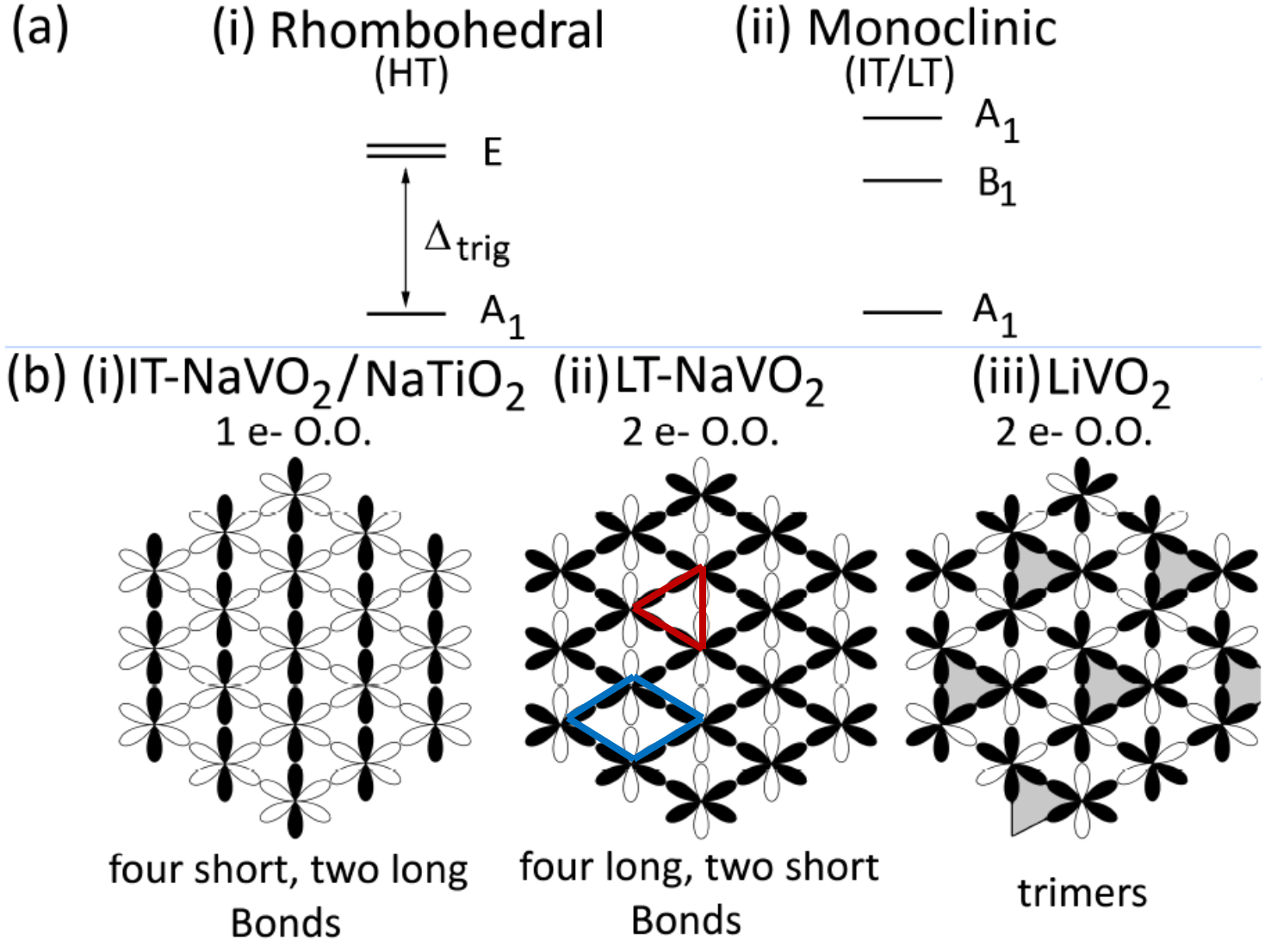
$NaVO_2$: Two Phase Transitions, Magnetic Order





T.M. McQueen, et al. *Phys. Rev. Lett.* **101**, 166402 (2008)

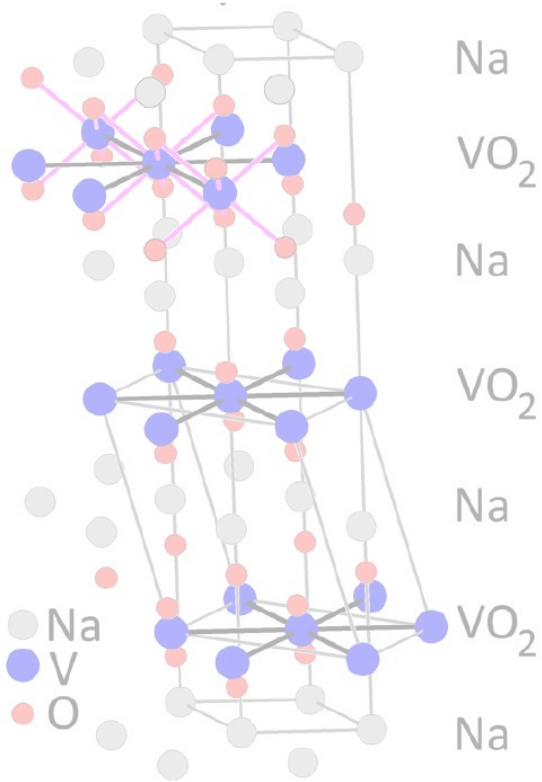
Orbital Ordering: Triangles to Squares



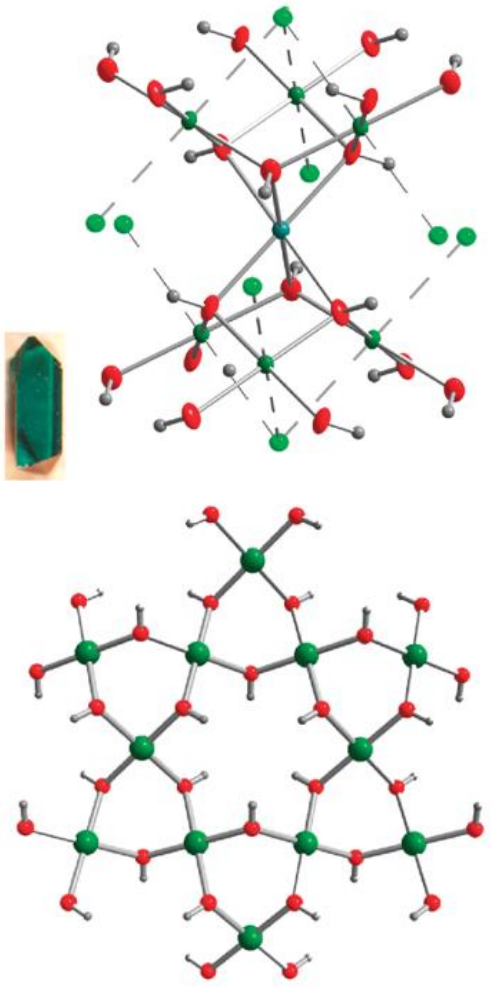
T.M. McQueen, et al. *Phys. Rev. Lett.* **101**, 166402 (2008)

The Research Vignettes for Today

$NaVO_2$



$ZnCu_3(OH)_6Cl_2$



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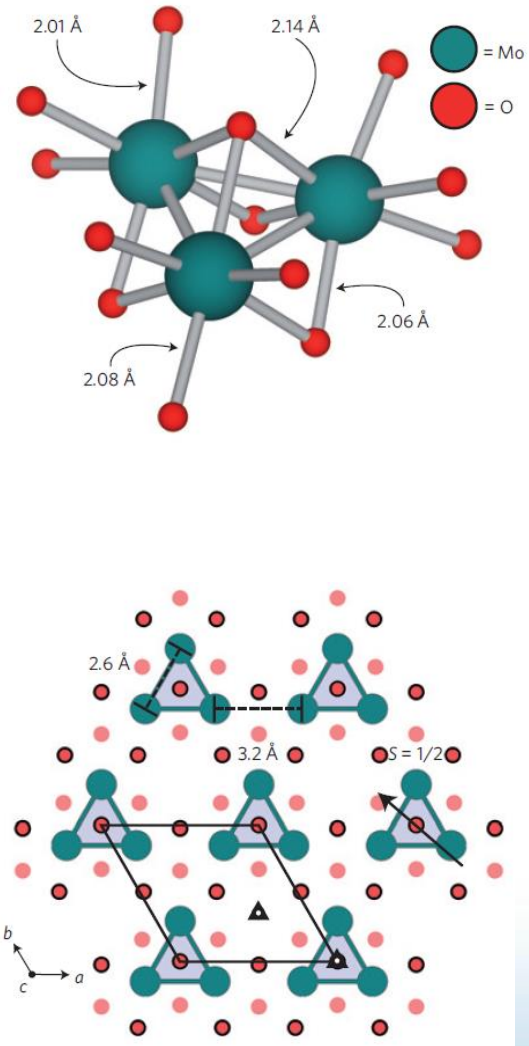


From Minerals to Kagomé Magnets

Mineralogical Magazine, June 2004, Vol. 68(3), pp. 527–539

Herbertsmithite, $Cu_3Zn(OH)_6Cl_2$, a new species, and the definition of paratacamite

R. S. W. Braithwaite[1], K. Mereiter[2], W. H. Paar[3,*] and A. M. Clark[4]

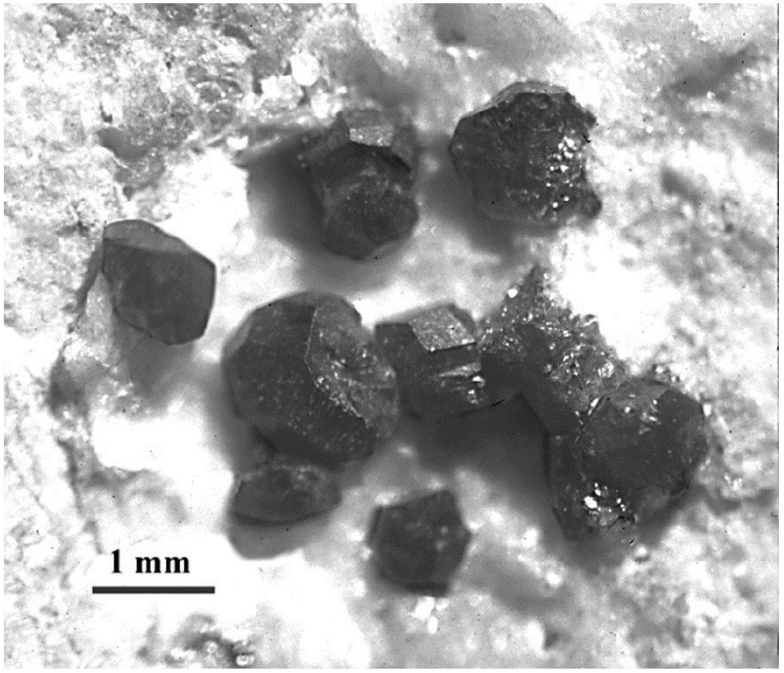
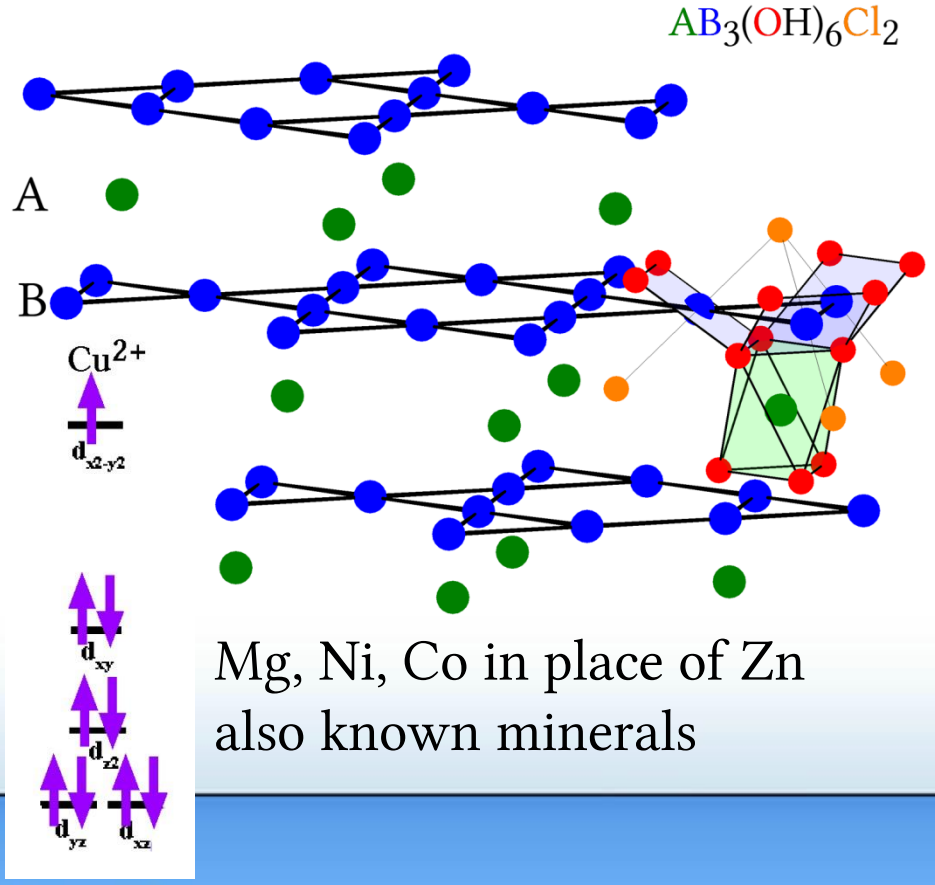
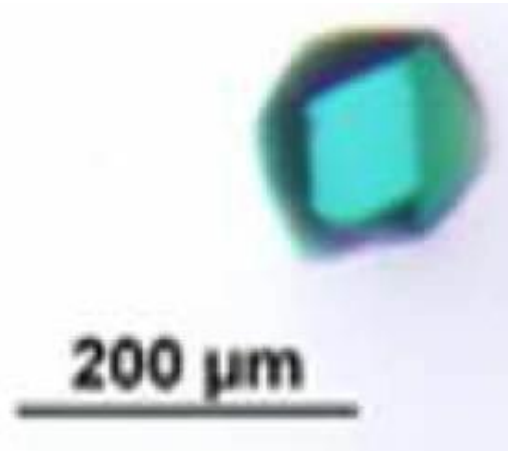


Fig. 1. Herbertsmithite, dark green crystals on pale buff matrix. From RSWB 84-27, Mina Los Tres Presidentes, Sierra Gorda, Chile.



Mg, Ni, Co in place of Zn also known minerals

Synthetic $Zn_{1-\delta}Cu_{3+\delta}(OH)_6Cl_2$



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COMMUNICATIONS

Published on Web 09/09/2005

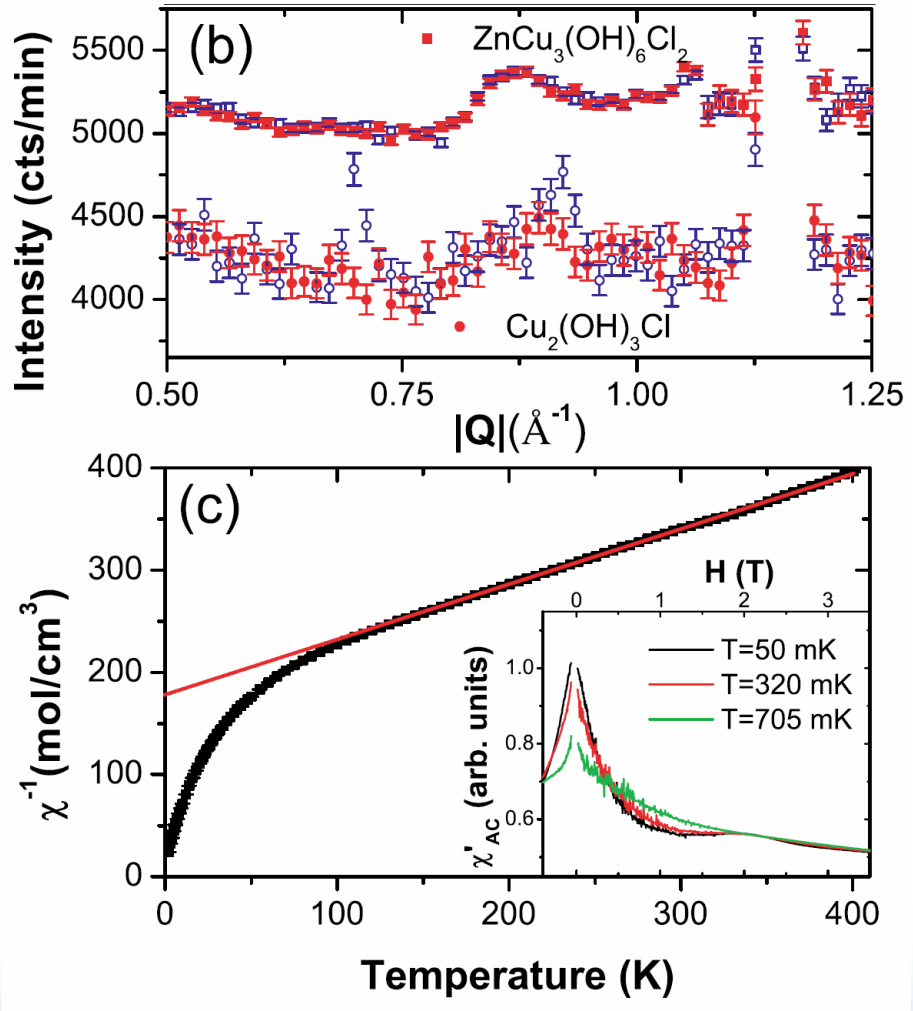
A Structurally Perfect $S = {}^1/_2$ Kagomé Antiferromagnet

Matthew P. Shores, Emily A. Nytko, Bart M. Bartlett, and Daniel G. Nocera*

Department of Chemistry, 6-335, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139-4307

M.P. Shores, et al. J. Am. Chem. Soc. 127, 13462-3 (2005)

Candidate Spin Liquid

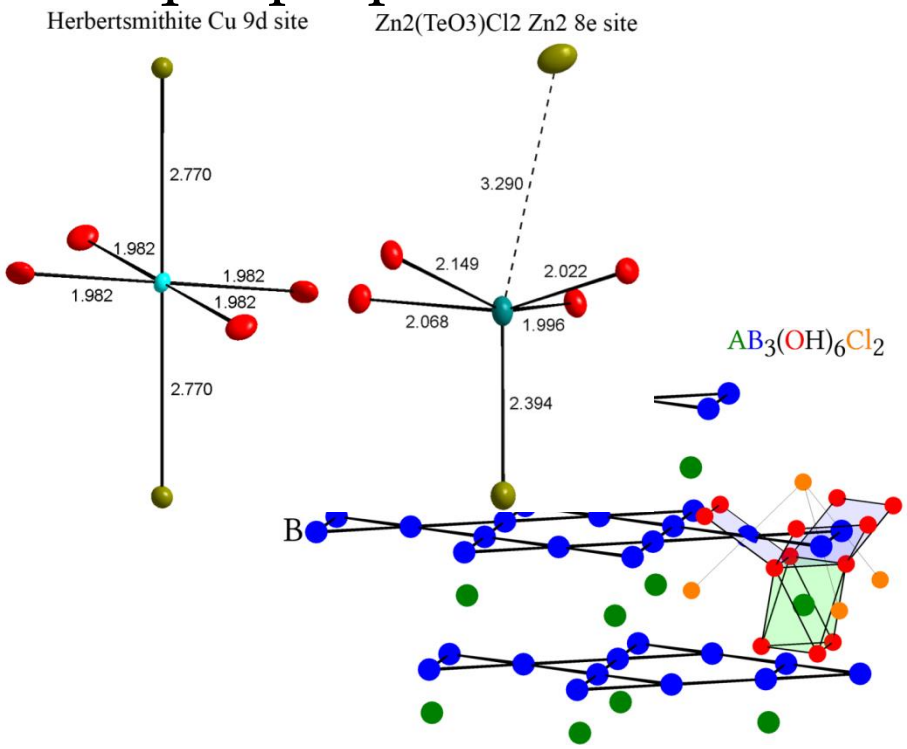


- $|\theta|$ ~ -300 K, indicative of strong AFM
- But, no magnetic order to 50 mK
- No sharp magnetic Bragg peaks and dispersionless magnetic excitations in neutron diffraction

J.S. Helton, *et al.*, Phys. Rev. Lett. **98**, 107204 (2007)

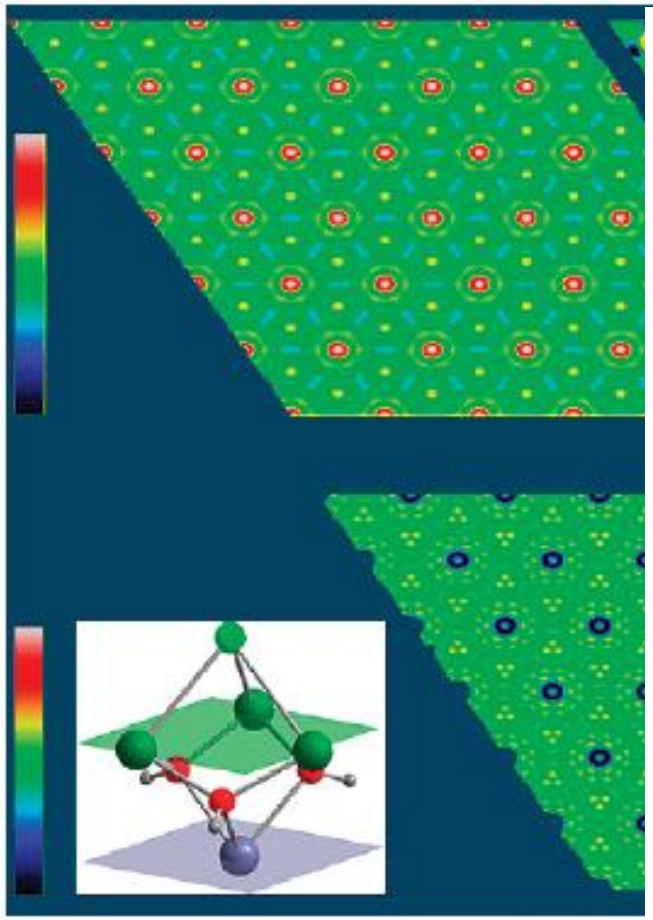
Disordered Mess?

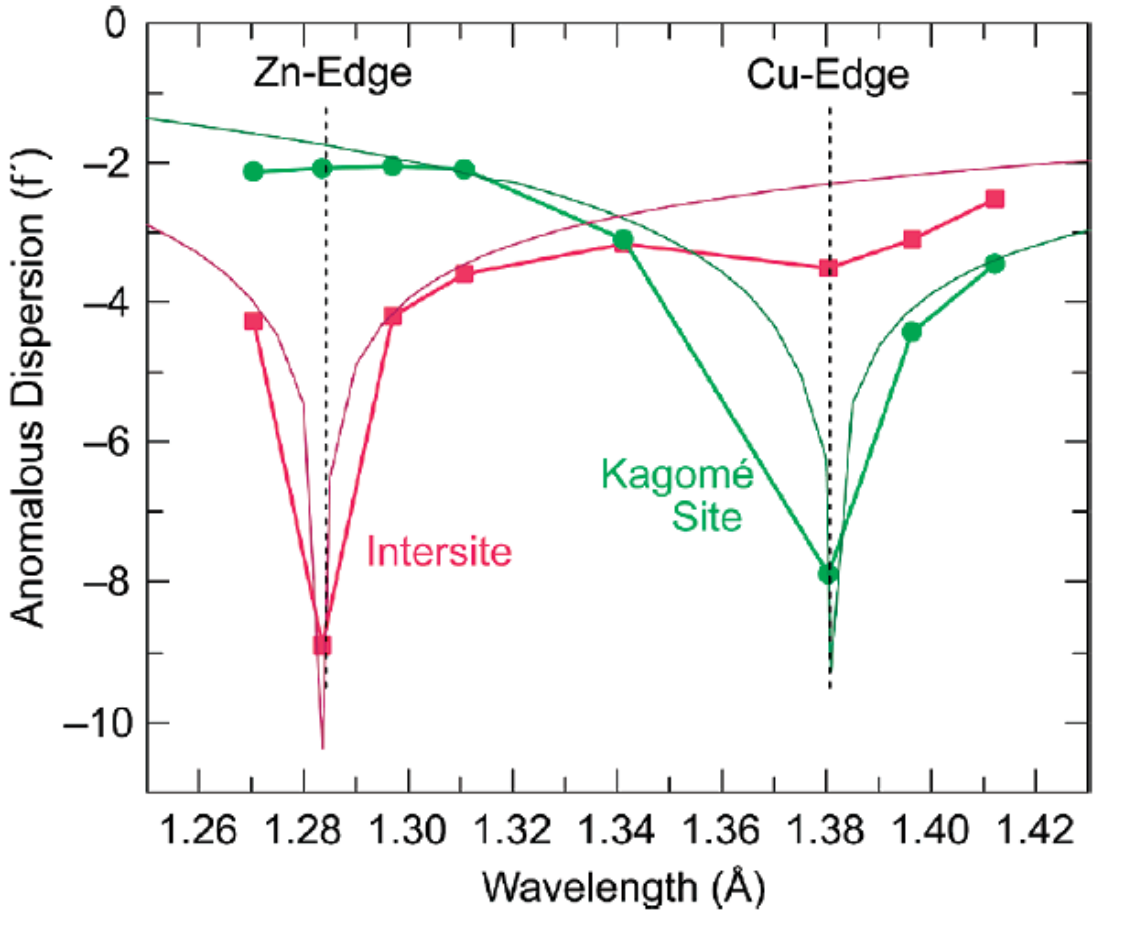
- Is "$ZnCu_3(OH)_6Cl_2$" really a 'perfect' kagomé antiferromagnet? Or are these unique properties the result of chemical disorder?
- Zn^{2+} and Cu^{2+} have the same charge and similar ionic radii (0.75 vs. 0.73 Å)
- Zn^{2+} is known to go in O_4Cl_2 coordination, and Cu^{2+} is known to go into O_6 coordination



- Consequently, Zn-Cu mixing is chemically plausible
- But quantifying this is non-trivial

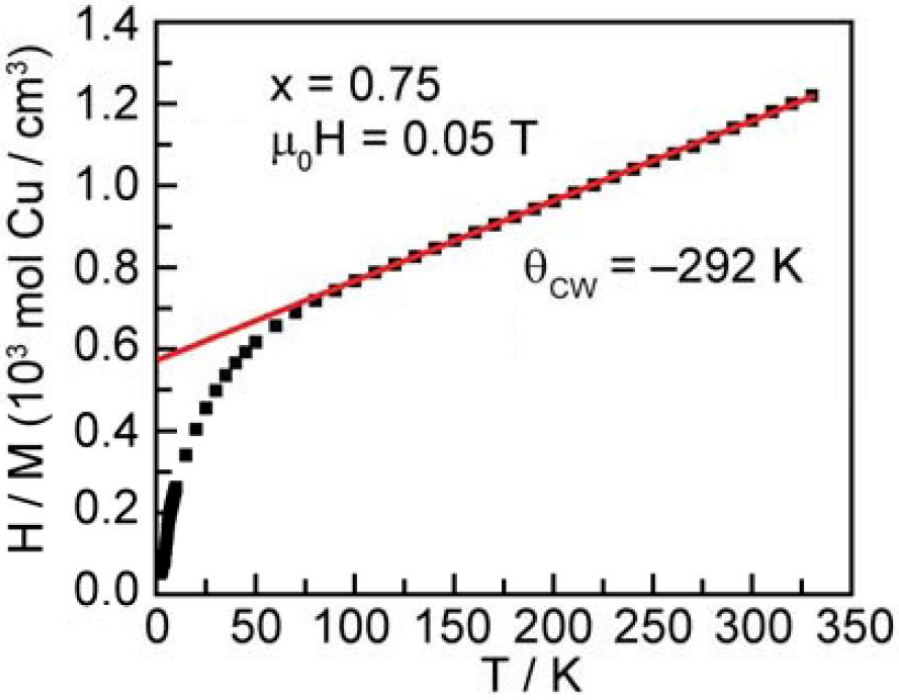
Kagomé Planes Low Defect Density, But…

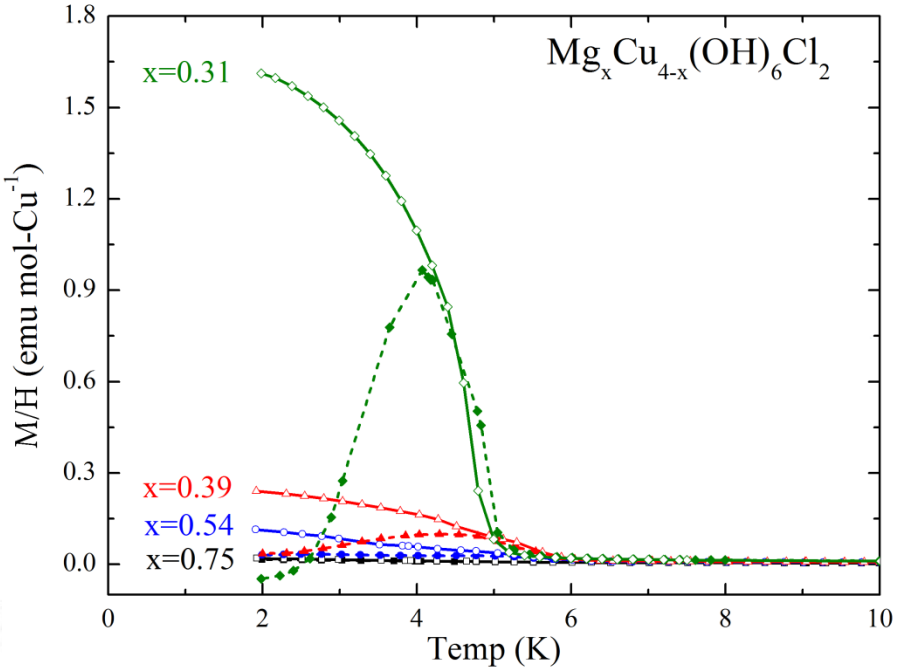




D.E. Freedman, et al. *J. Am. Chem. Soc.* **132**, 16185-90 (2010)

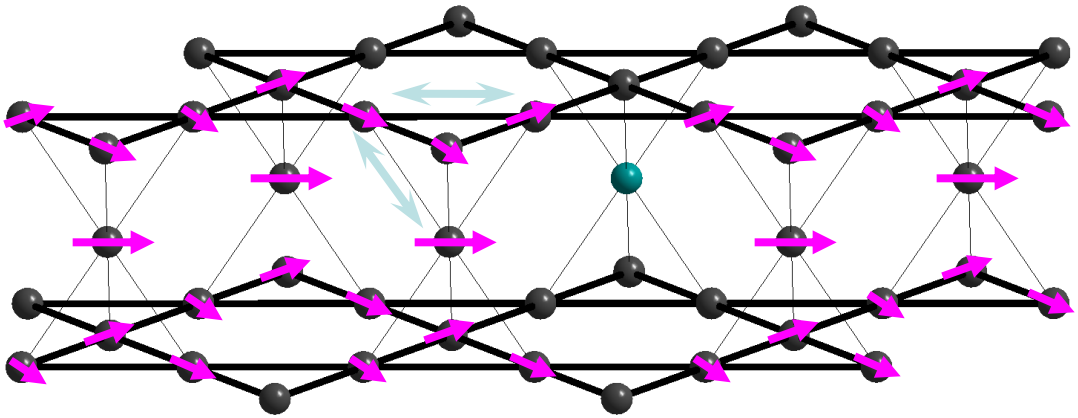
Interlayer Defects Modulate Low-T Physics



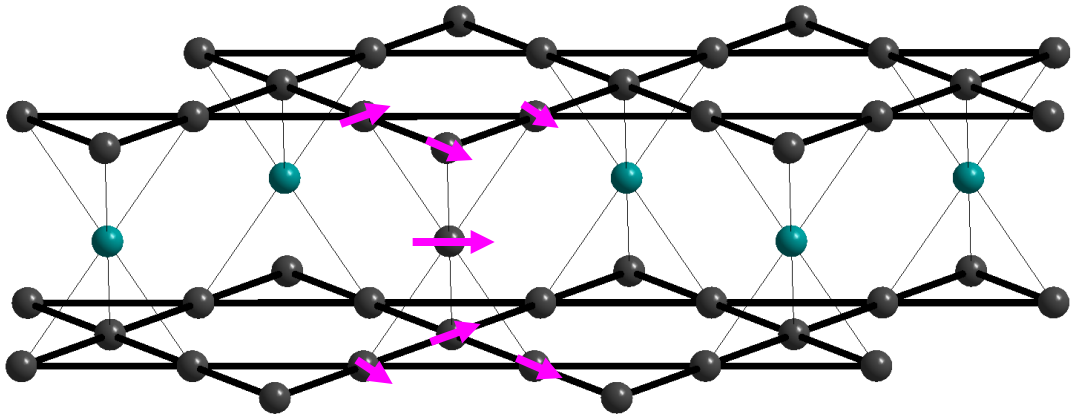


S. Chu, et al. J. Am. Chem. Soc. 132, 5570-1 (2010)

Pictorial Explanation

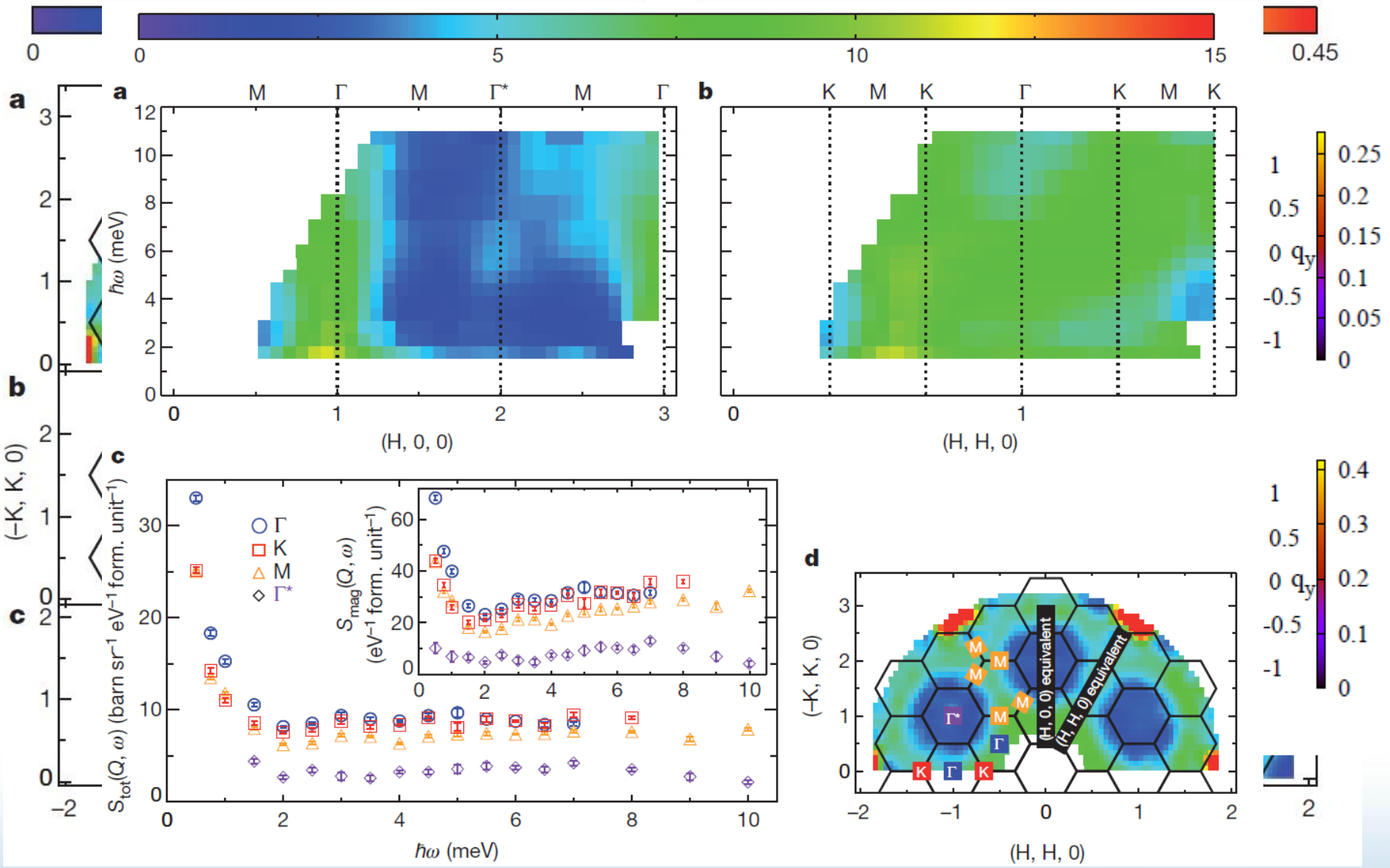


Spins are not meant to indicate actual magnetic structure



- Coupling within a layer strongly AFM
- Coupling between kagomé and interlayer sites makes domains
- Low x: Static magnetic domains
- High x: Domains get too small to freeze out
- Explains all the data
- Implies native state of 2D layers not just magnetic ordering…

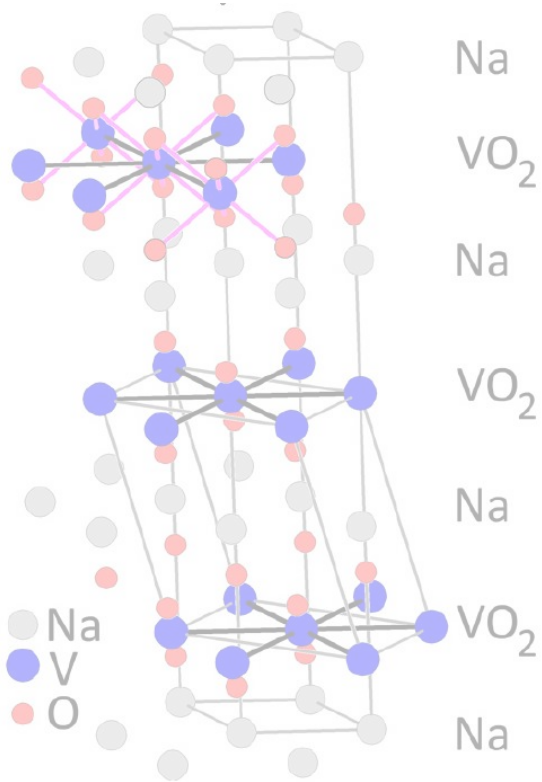
Fractionalized Excitations



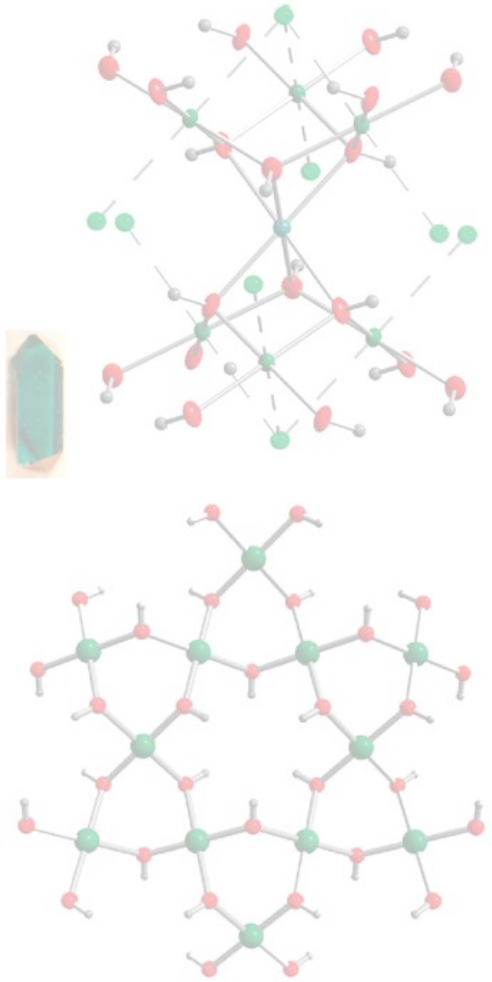
T.-H. Han, et al. Nature 492, 406 (2012)
Z. Hao and O. Tchernyshyov, *Phys. Rev. B* **81**, 214445 (2010)

The Research Vignettes for Today

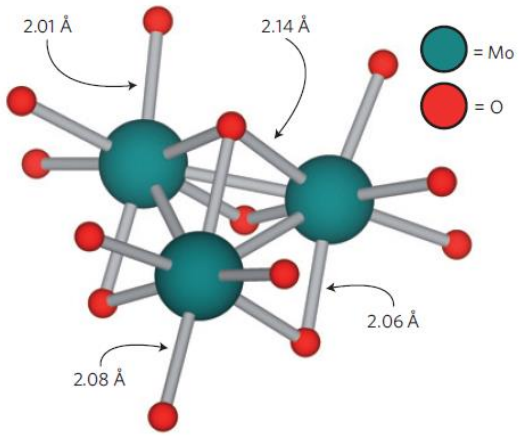
$NaVO_2$

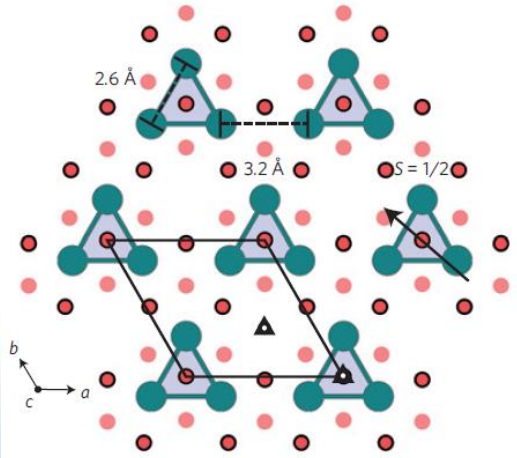


$ZnCu_3(OH)_6Cl_2$



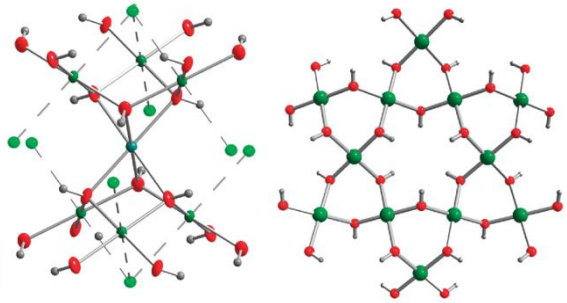
$LiZn_2Mo_3O_8$





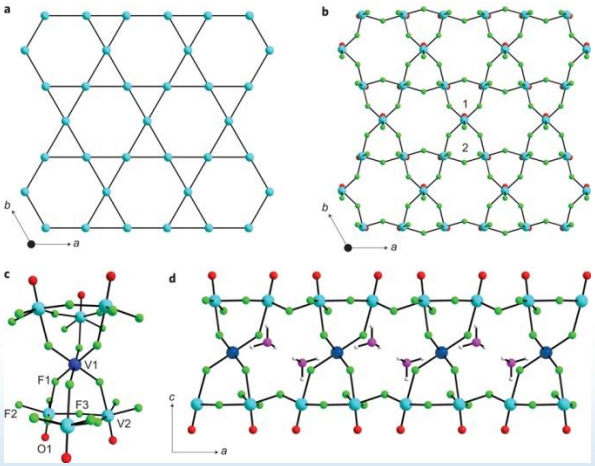
Classes of Two Dimensional S=1/2 Magnets

Single Ions

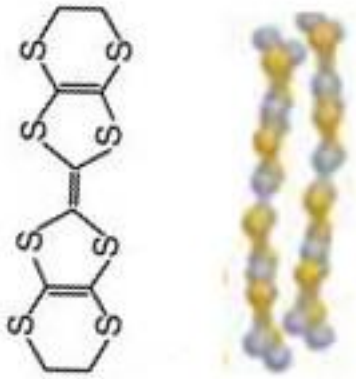


Cu^{2+} (d^9) Minerals

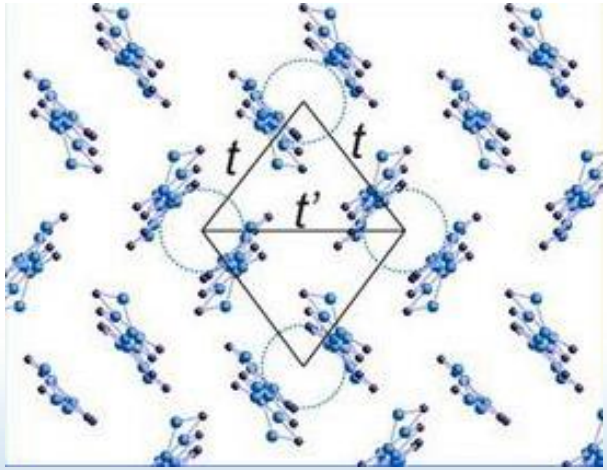
V^{4+} (d^1) Compounds



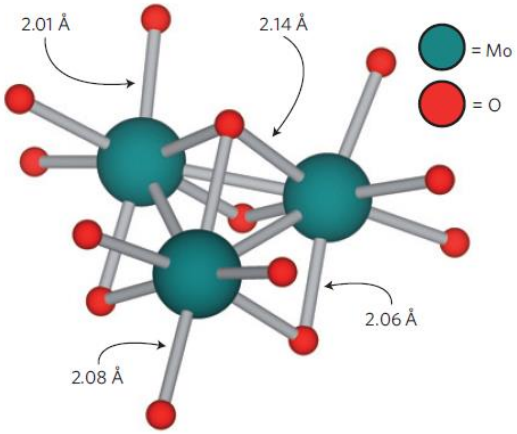
Organic Molecules



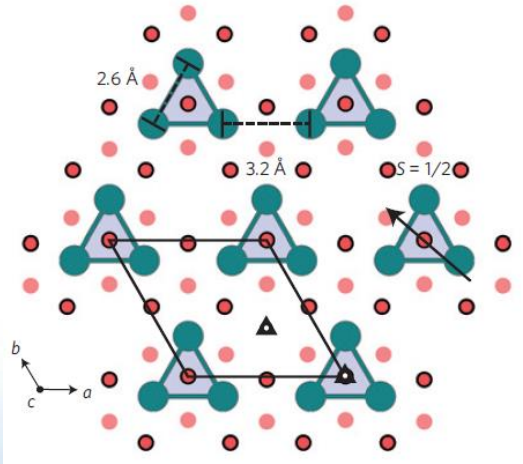
BEDT-TTF Dimers



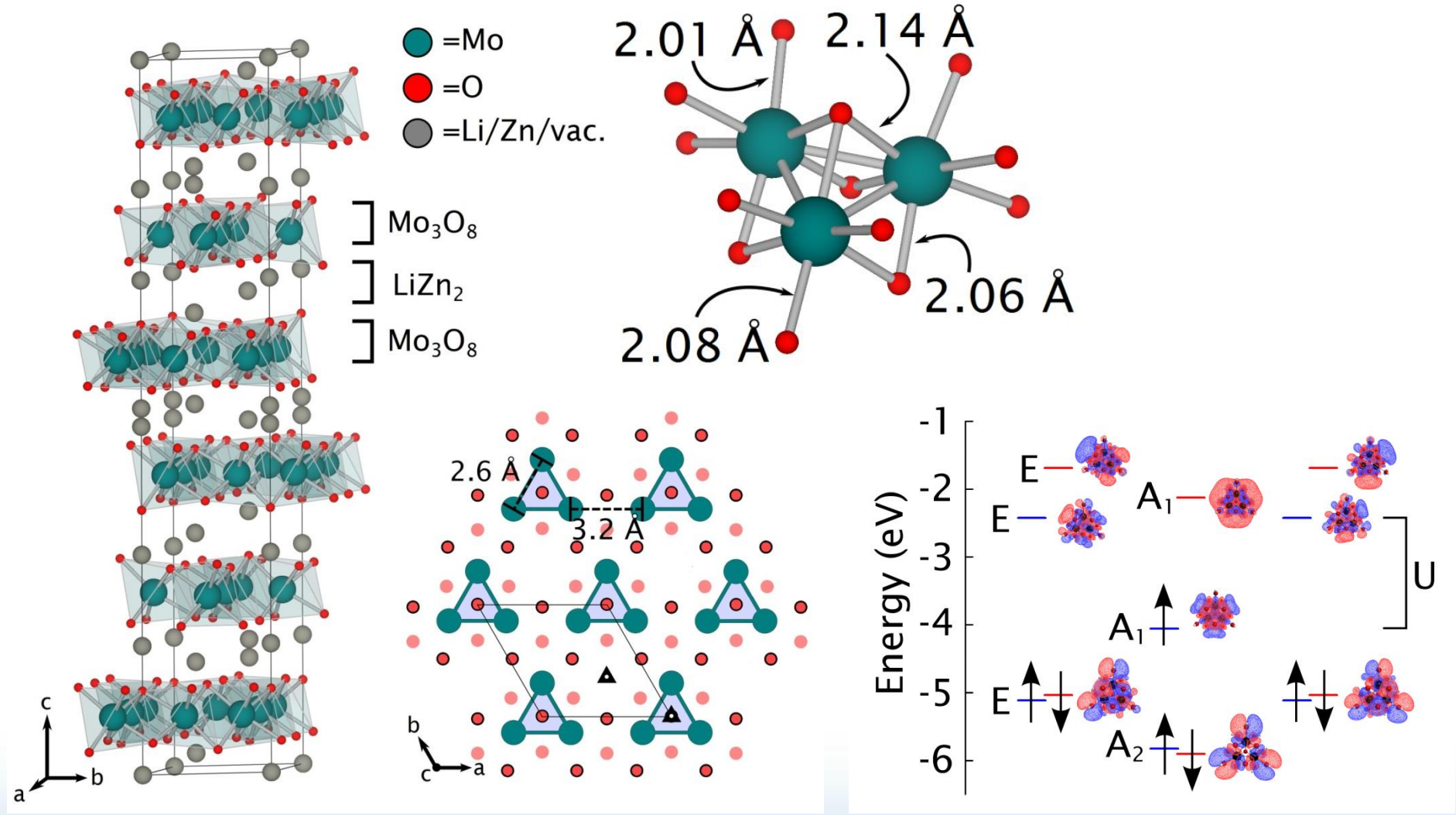
Inorganic "Molecules"



Mo_3O_{13} Cluster

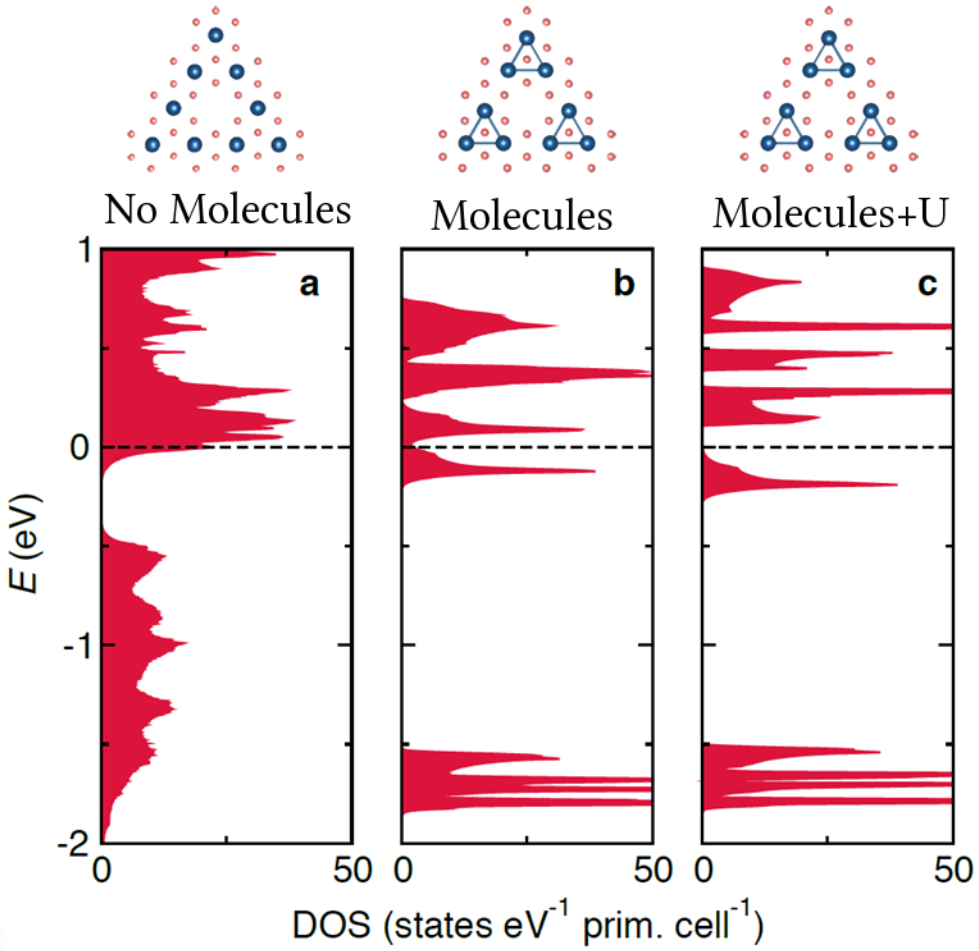


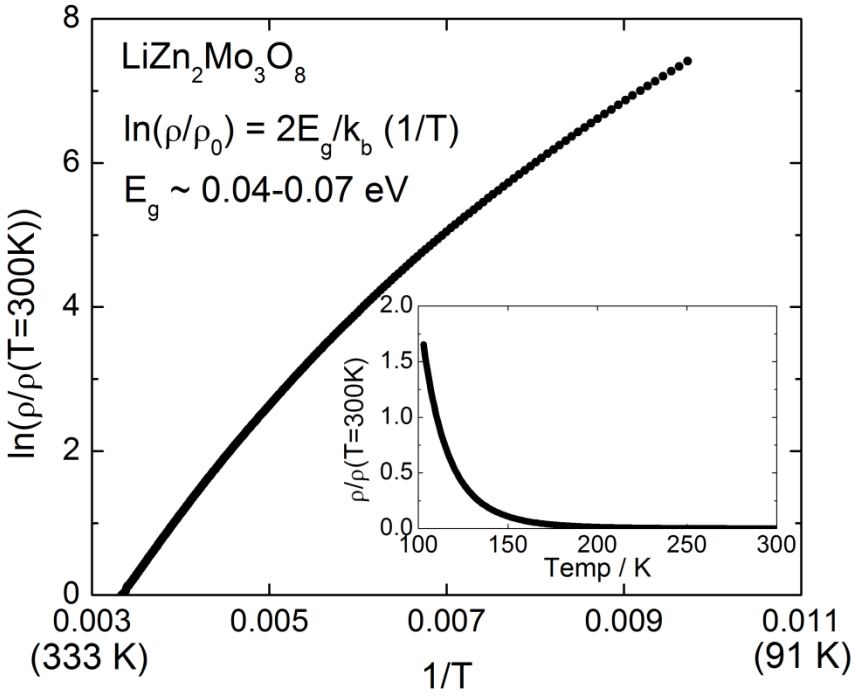
Structure of $LiZn_2Mo_3O_8$



J.P. Sheckelton, et al., *Nature Materials* **11**, 493-6 (2012)

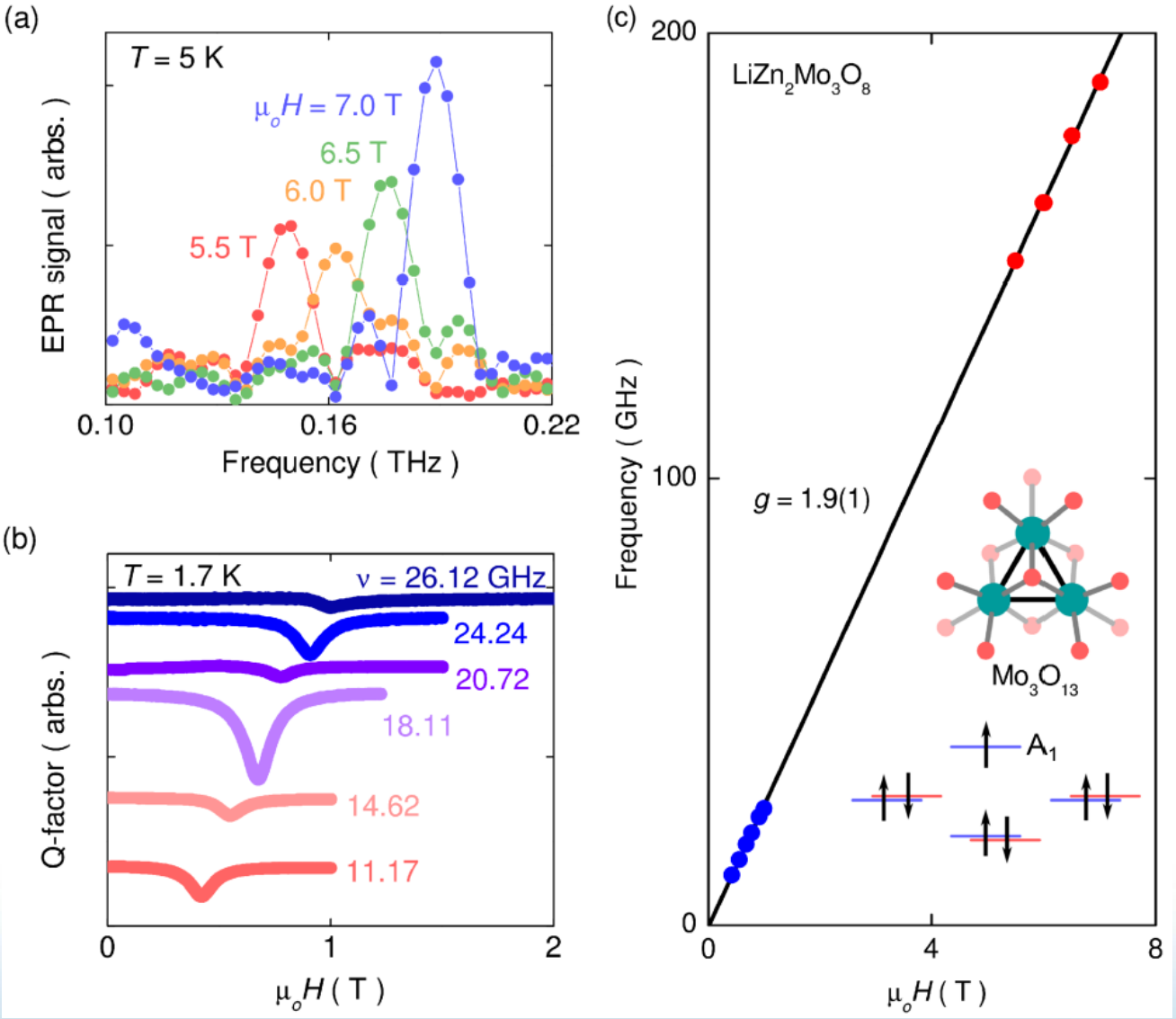
Likely Mott Insulator





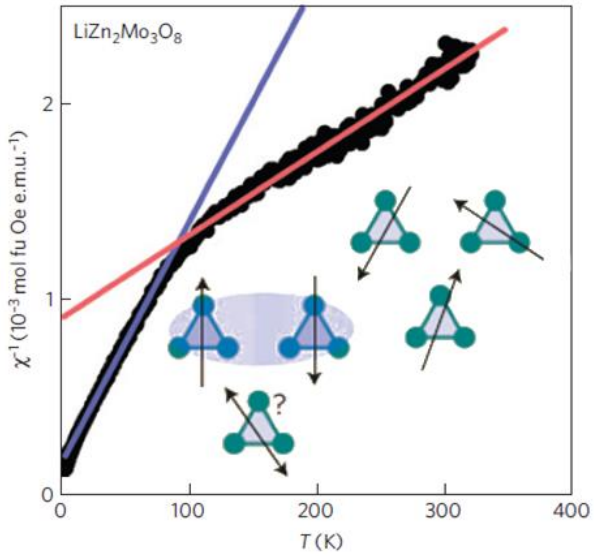
J.P. Sheckelton, et al., Nature Materials 11, 493-6 (2012)
J.P. Sheckelton, et al., Materials Horizons 2, 76-80 (2015)

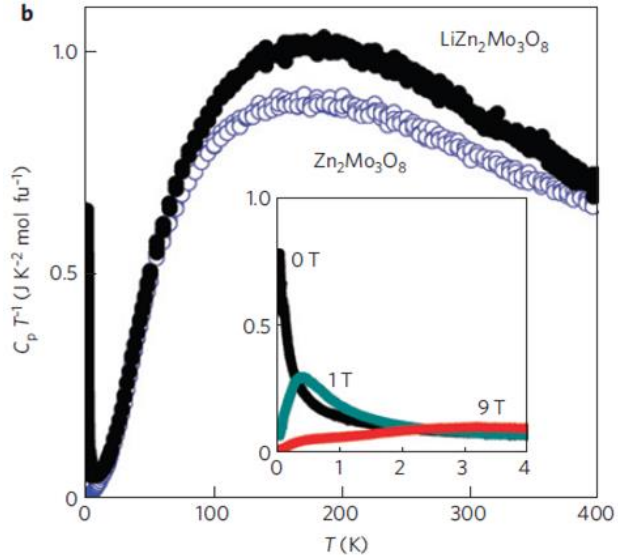
ESR Shows S=1/2

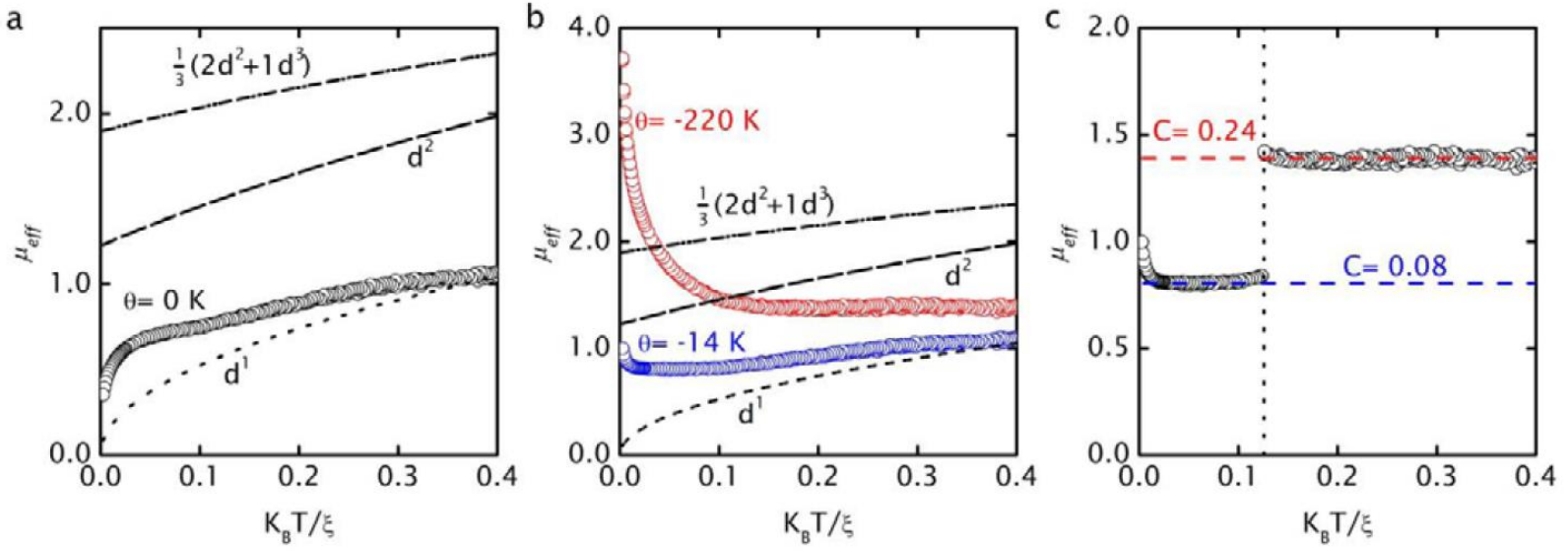


J.P. Sheckelton, et al., *Phys. Rev. B* **89**, 064407 (2014)

Valence Bonds?

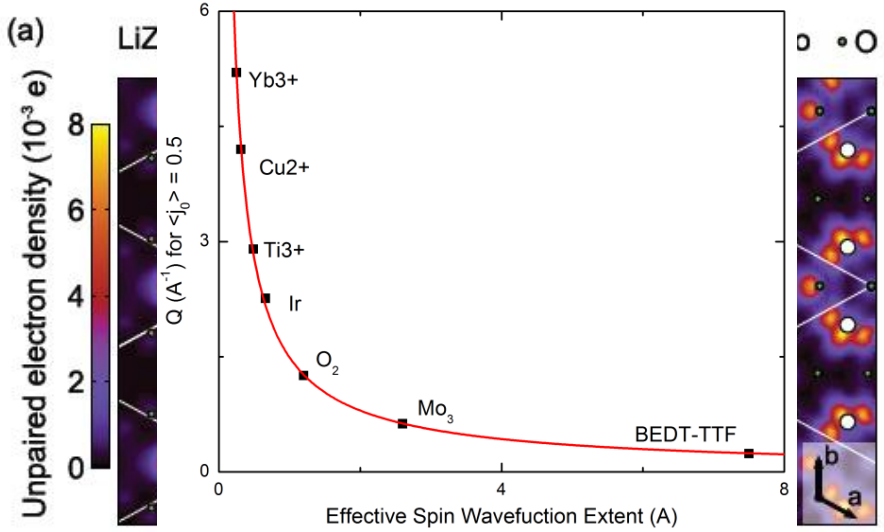


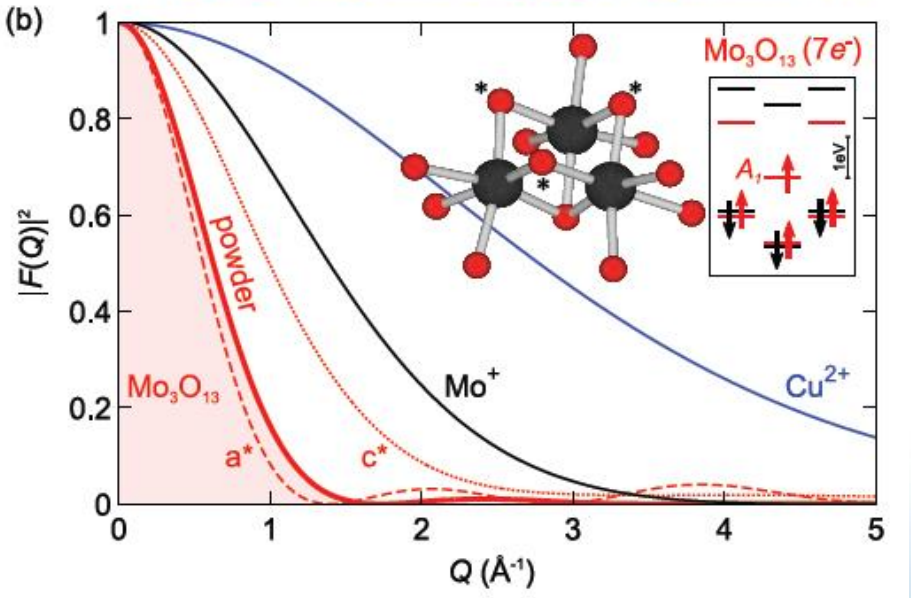


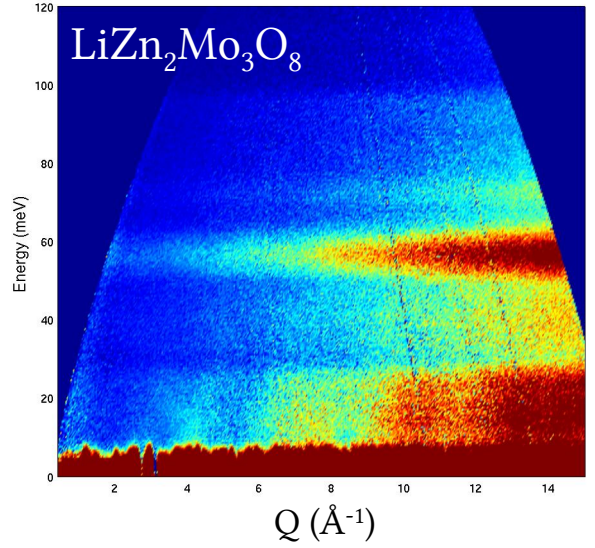


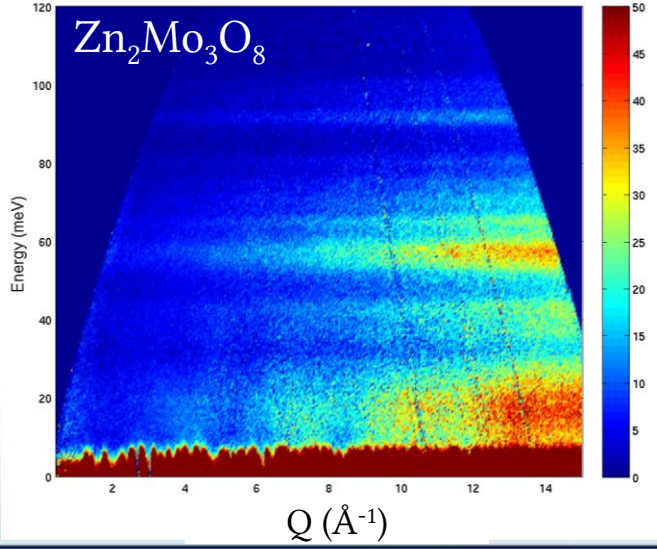
J.P. Sheckelton, et al., *Nature Materials* **11**, 493-6 (2012)
Model courtesy Oleg Tchernyshyov (unpublished)

Molecules, but where is the Magnetism?



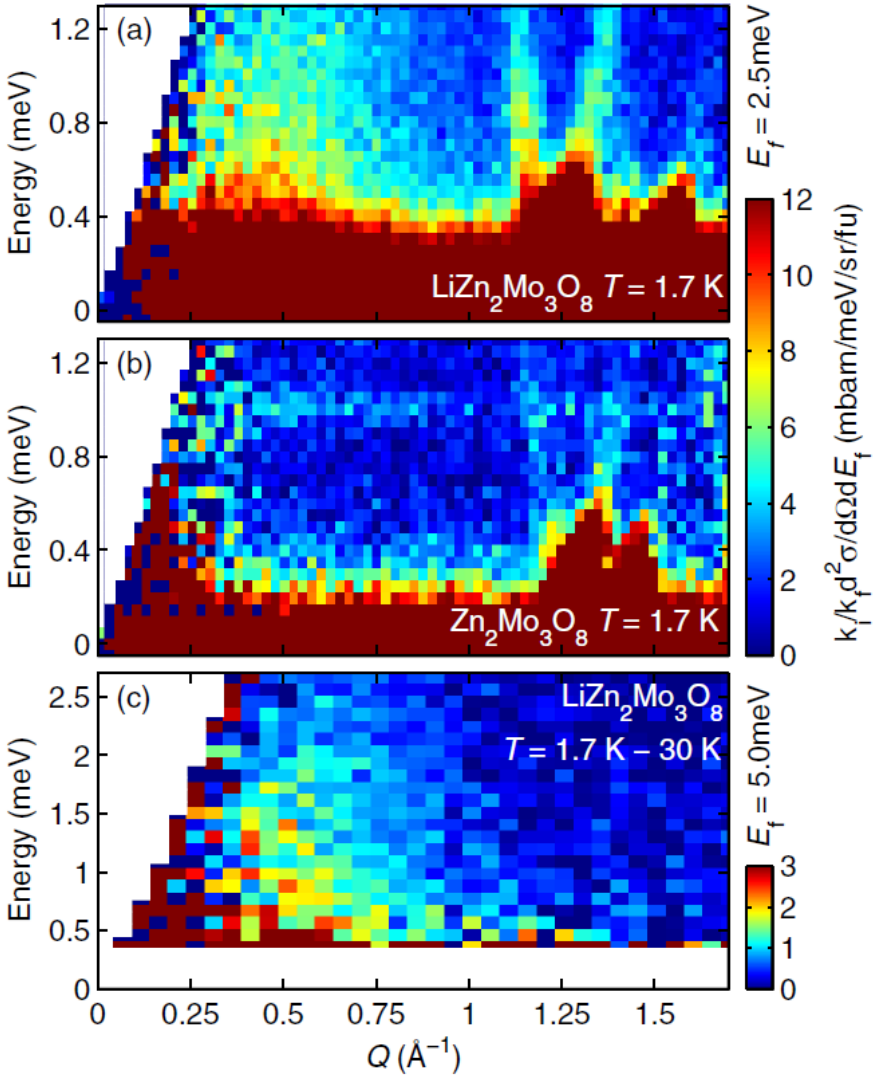


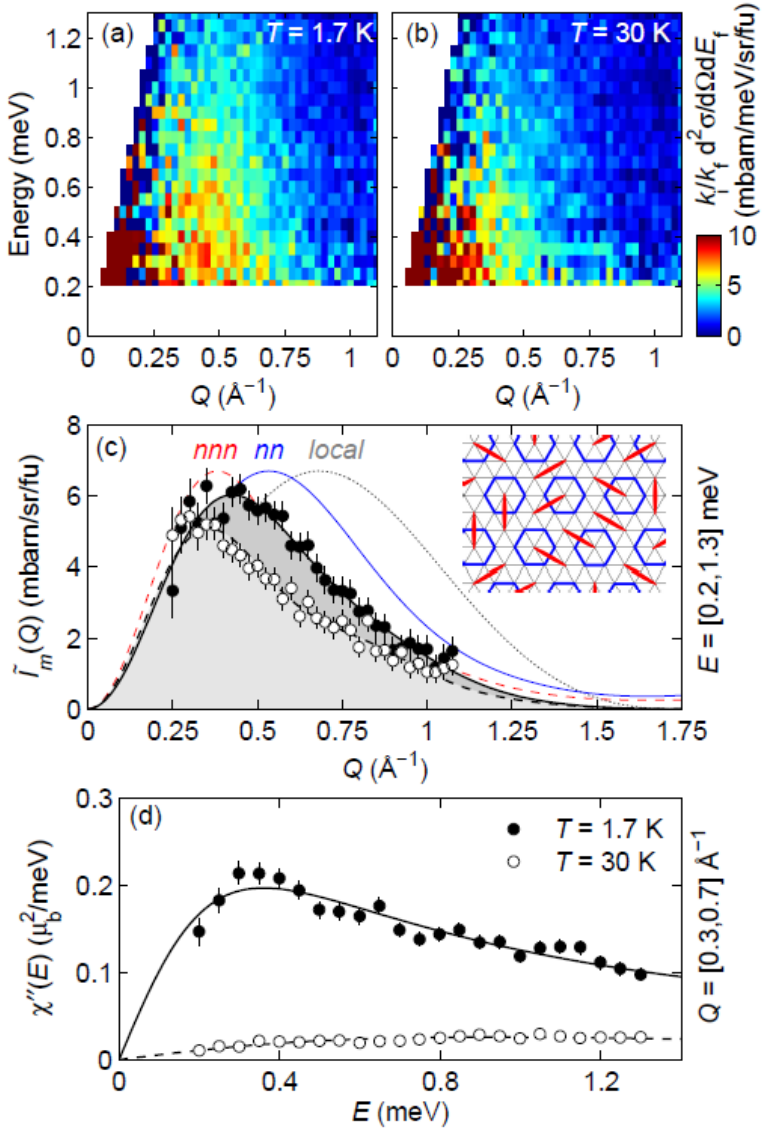




M. Mourigal, T.M. McQueen, C.L. Broholm, et al., *Phys. Rev. Lett.* **112**, 027202 (2014)

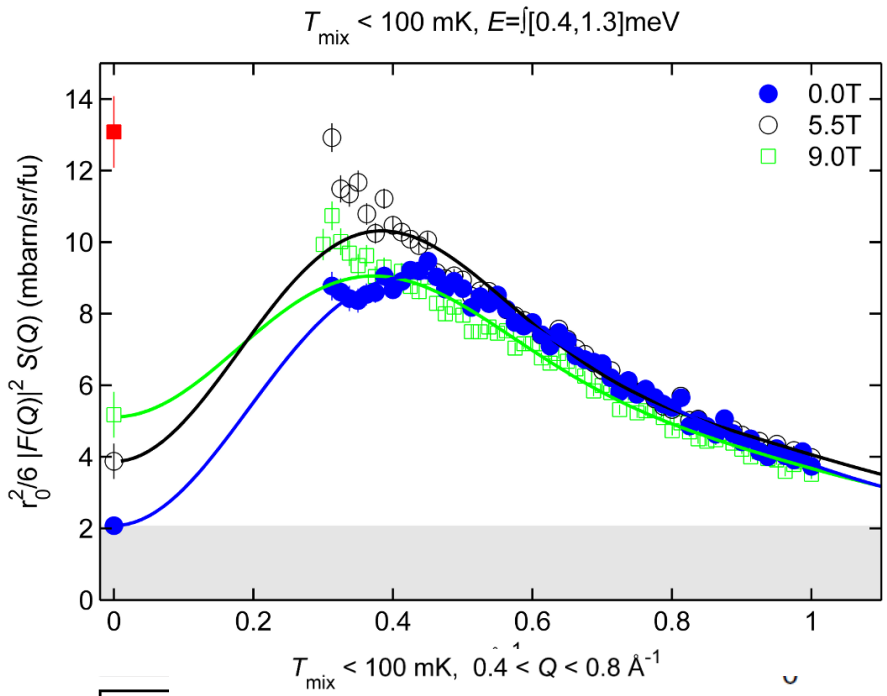
Valence Bonds!

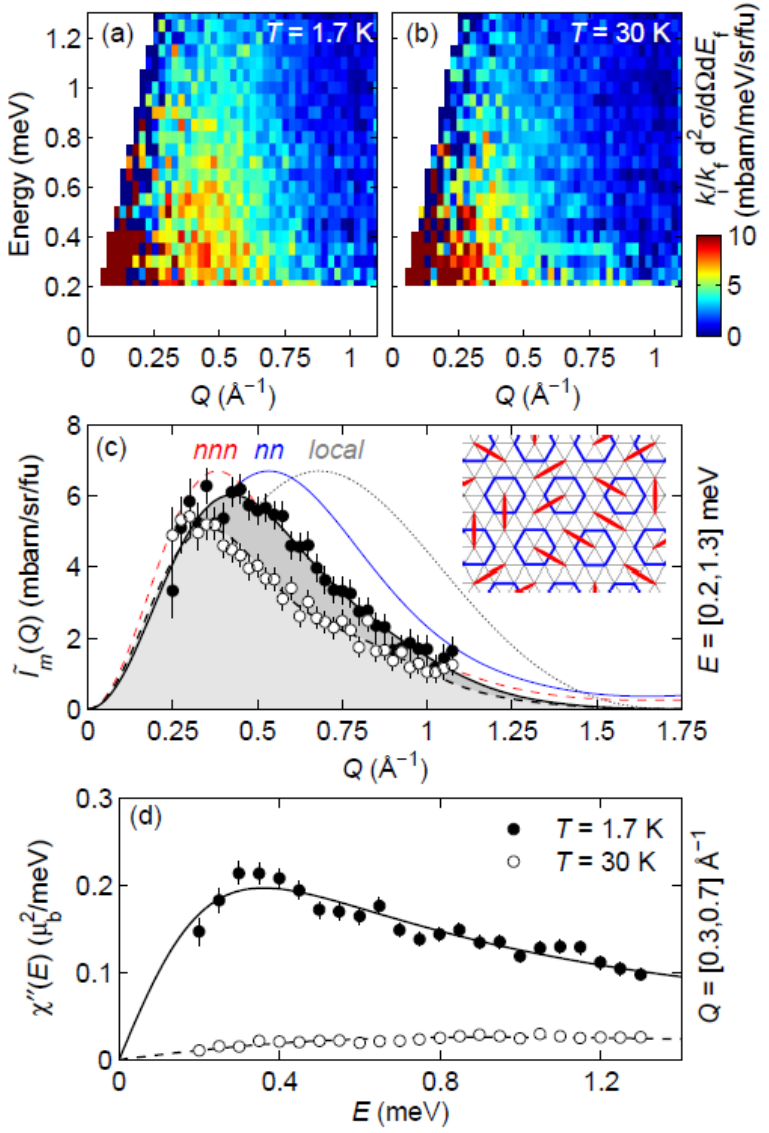




M. Mourigal, T.M. McQueen, C.L. Broholm, et al., *Phys. Rev. Lett.* **112**, 027202 (2014)

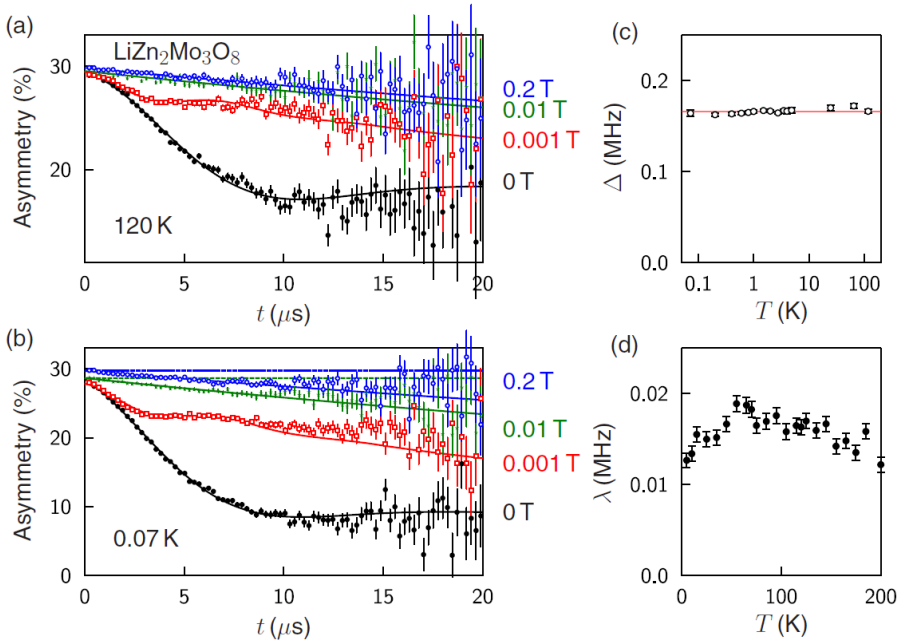
Valence Bonds!

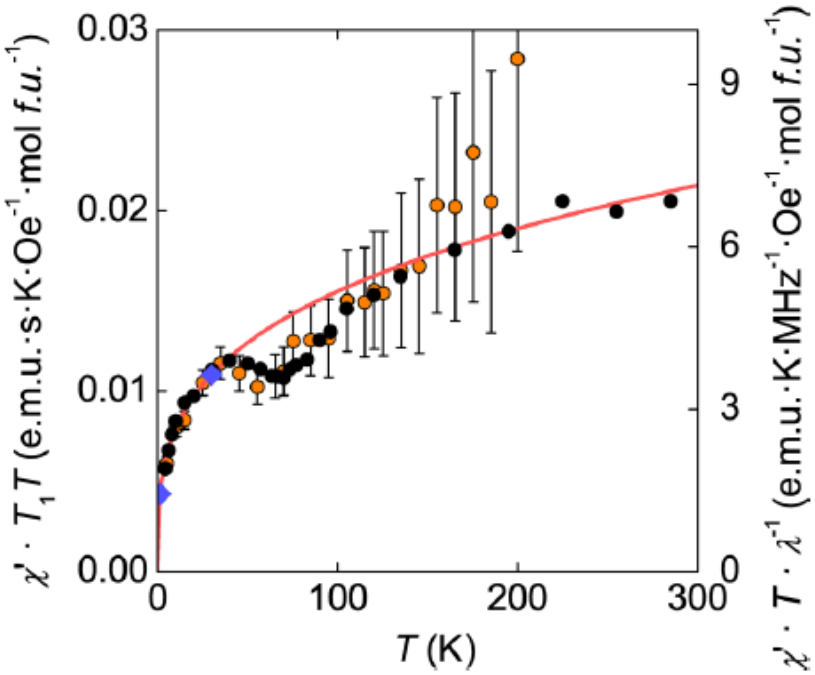




ueen, C.L. Broholm, et al., *Phys. Rev. Lett.* **112**, 027202 (2014)

No Magnetic Order to T = 0.07 K





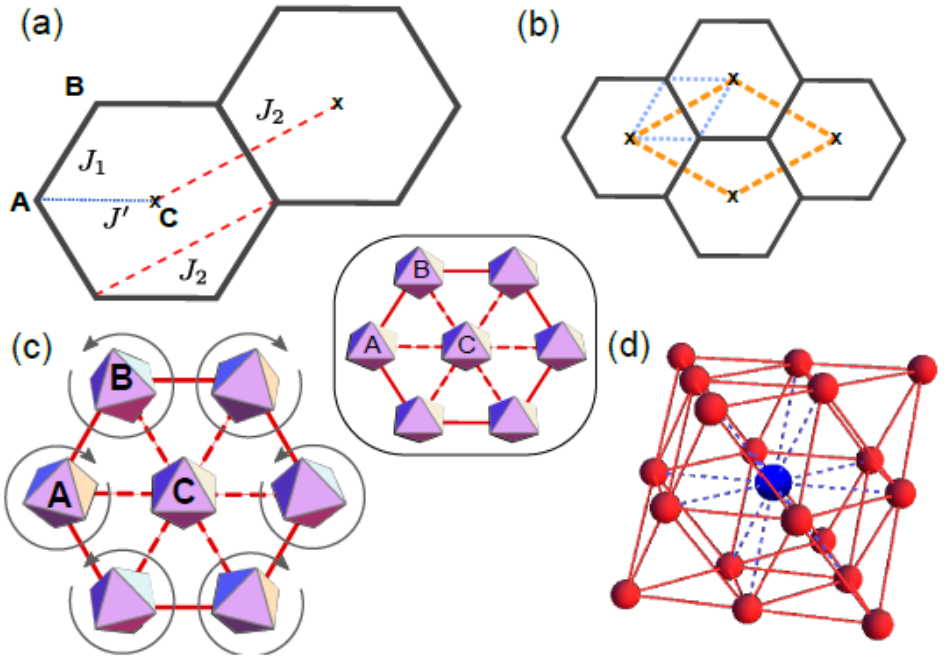
- Where are the other "2/3"rds of the spins?
- Why is it not a 120° ground state?

$$\frac{\lambda}{T} \simeq \frac{1}{T_1 T} \simeq \frac{\sum |A(\mathbf{q})|^2 \chi''(\mathbf{q},\omega_o)}{\omega_o},$$

$$\chi''(E) = \frac{\chi' E \Gamma}{E^2 + \Gamma^2}$$

J.P. Sheckelton, et al., *Phys. Rev. B* **89**, 064407 (2014)

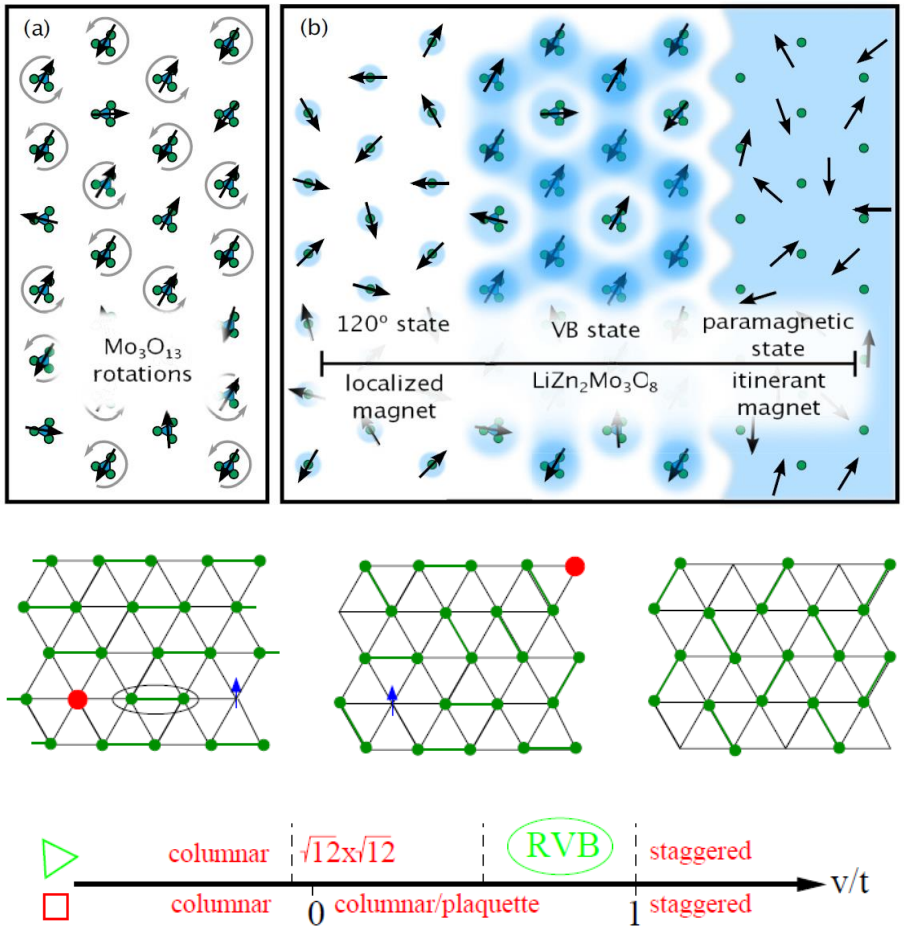
Possibilities



Emergent honeycomb lattice in $LiZn_2Mo_3O_8$

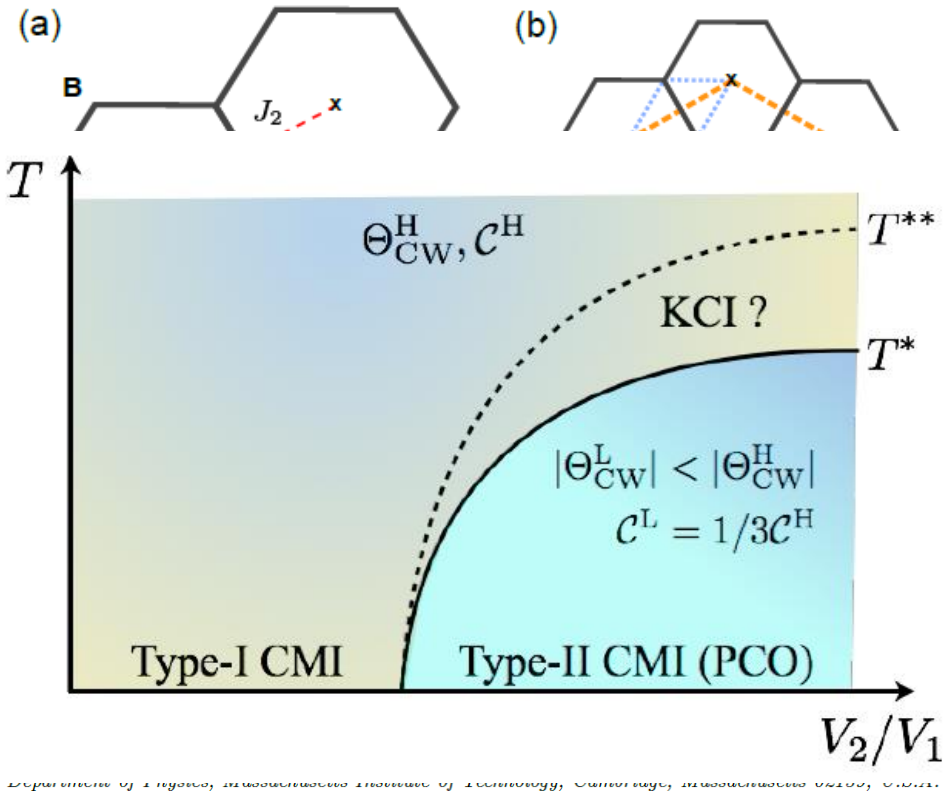
Rebecca Flint and Patrick A. Lee
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, U.S.A.

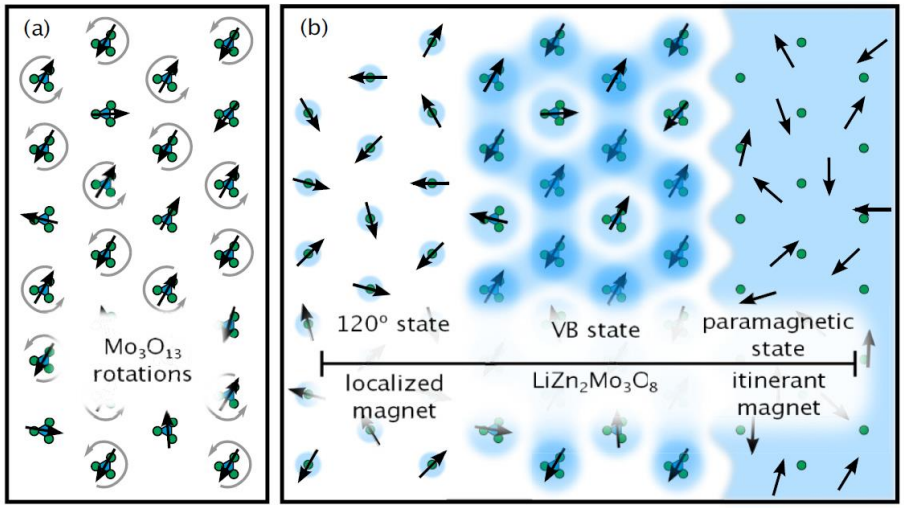
We introduce the idea of *emergent lattices*, where a simple lattice decouples into two weakly-coupled lattices as a way to stabilize spin liquids. In $LiZn_2Mo_3O_8$, the disappearance of 2/3rds of the spins at low temperatures suggests that its triangular lattice decouples into an emergent honeycomb lattice weakly coupled to the remaining spins, and we suggest several ways to test this proposal. We show that these orphan spins act to stabilize the spin-liquid in the $J_1 - J_2$ honeycomb model and also discuss a possible 3D analogue, Ba_2MoYO_6 that may form a "depleted fcc lattice."

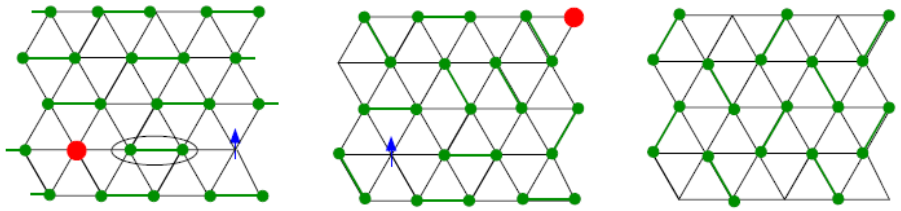


Flint and Lee, Phys. *Rev. Lett.* **111**, 217201 (2013)
R. Moessner and S.L. Sondhi, "Resonating Valence Bond Liquid Physics on the Triangular Lattice,", Prog. Theor. Phys. (2002)
J.P. Sheckelton, et al. *Phys. Rev. B* **89**, 064407 (2014)
Gang Chen

Possibilities

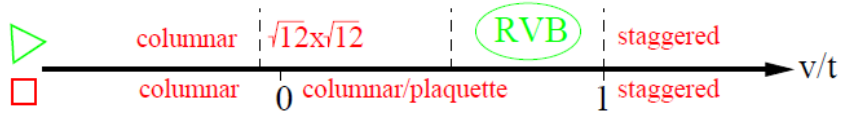






Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, U.S.A.

We introduce the idea of *emergent lattices*, where a simple lattice decouples into two weakly-coupled lattices as a way to stabilize spin liquids. In $LiZn_2Mo_3O_8$, the disappearance of 2/3rds of the spins at low temperatures suggests that its triangular lattice decouples into an emergent honeycomb lattice weakly coupled to the remaining spins, and we suggest several ways to test this proposal. We show that these orphan spins act to stabilize the spin-liquid in the $J_1 - J_2$ honeycomb model and also discuss a possible 3D analogue, Ba_2MoYO_6 that may form a "depleted fcc lattice."



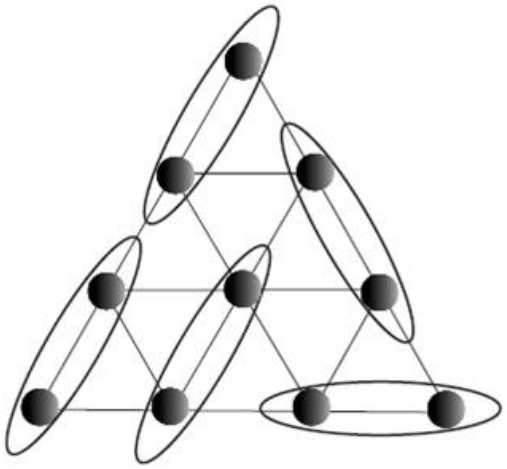
Flint and Lee, Phys. *Rev. Lett.* **111**, 217201 (2013)
R. Moessner and S.L. Sondhi, "Resonating Valence Bond Liquid Physics on the Triangular Lattice,", Prog. Theor. Phys. (2002)
J.P. Sheckelton, et al. *Phys. Rev. B* **89**, 064407 (2014)
Gang Chen

A breather…

- $LiZn_2Mo_3O_8$ is one of a handful of materials known to have a valence bond structure, and the gapless nature means it is a candidate spin liquid
- Strong interactions between isolated spins on magnetic clusters possible
- Avoid 1st order Jahn-Teller Effects
- Limit number of defects

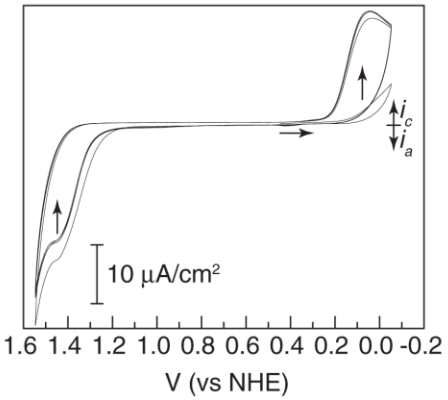
A Five Step Plan

Synthesize a resonating valence bond or spin liquid material



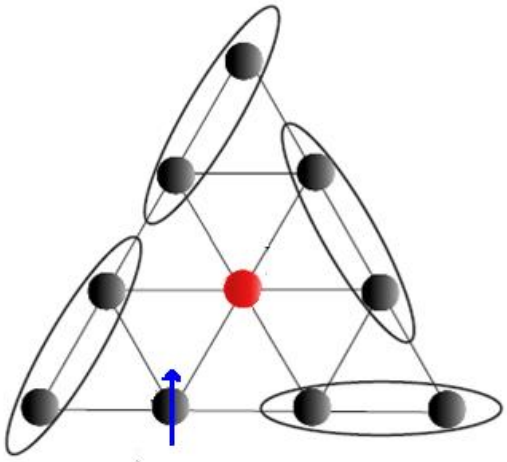
Step 1

Prove that oxidation or reduction is possible



Step 2

Dope spin liquid with holes (preferred) or electrons



Step 3

Demonstrate superconductivity

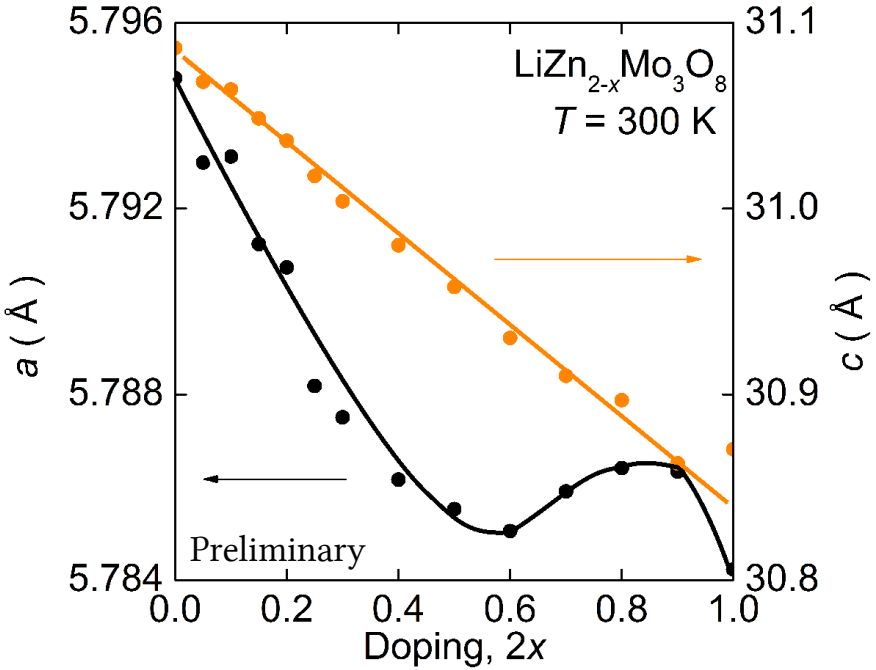
Step 4

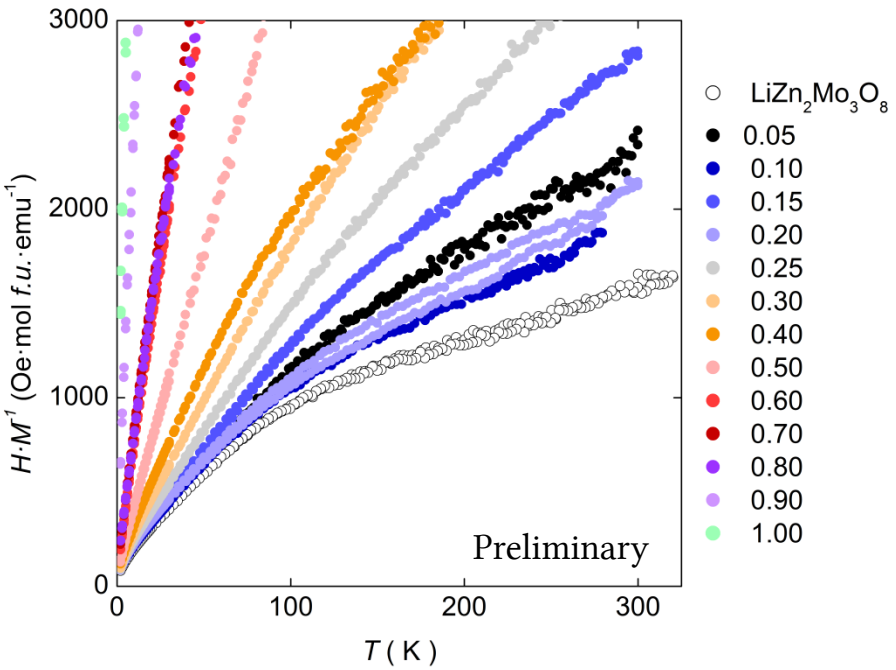
Nobel Prize in Physics (??)

Step 5

Charge Doping $LiZn_2Mo_3O_8$

Lets skip the messy chemistry, and just say $LiZn_{2-x}Mo_3O_8$ works



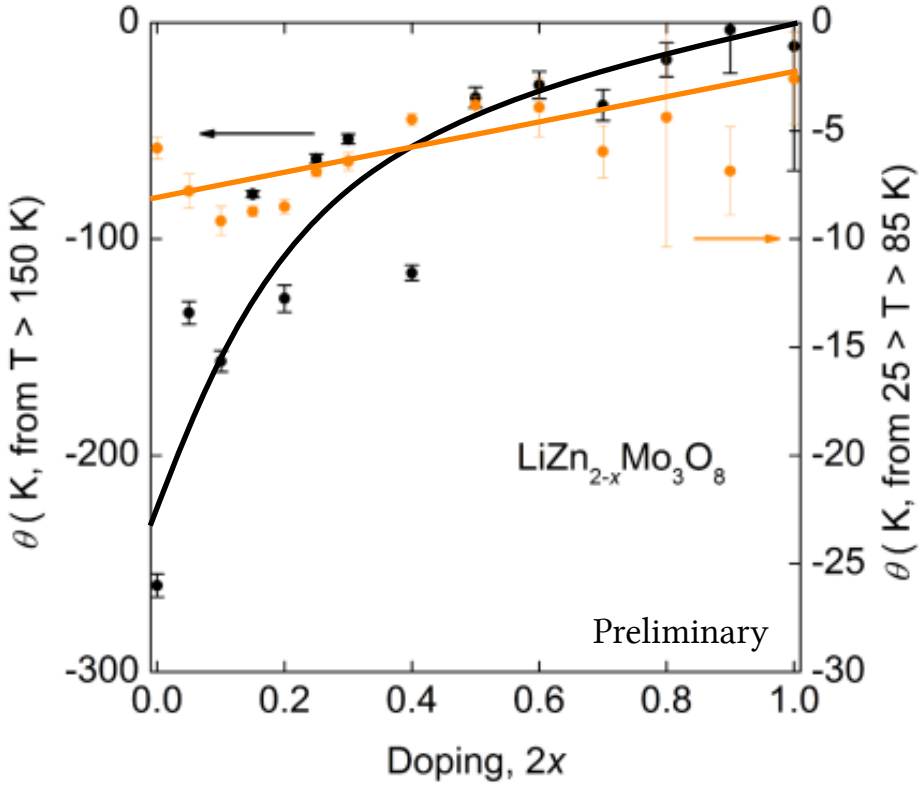


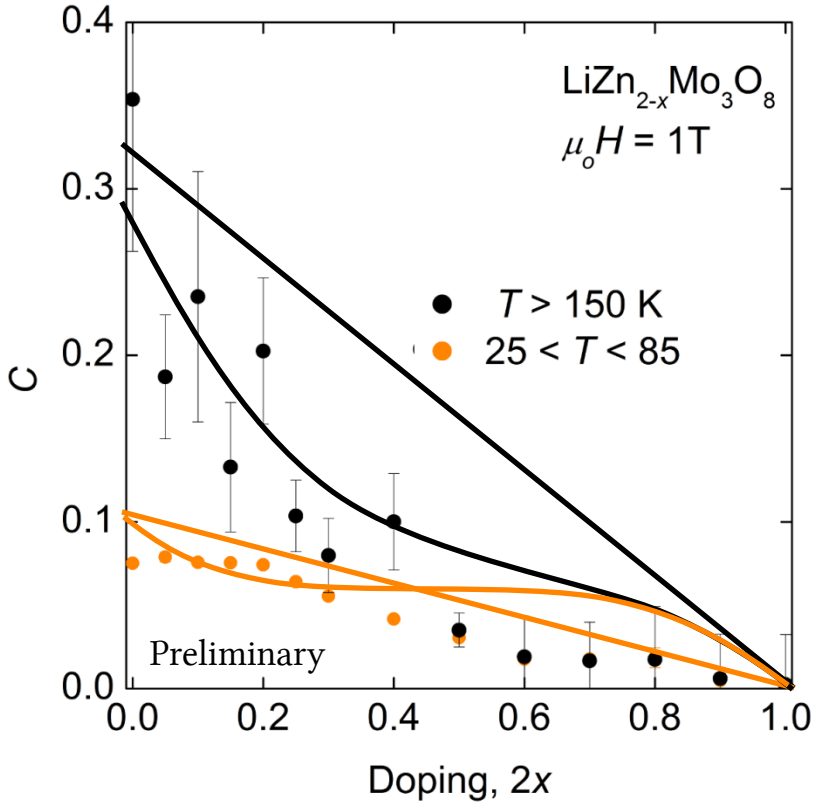
Can pull out at most 0.5 Zn (1 e^-/site)

Systematic loss of "kink" in susceptibility

J.P. Sheckelton, et al., Materials Horizons 2, 76-80 (2015)

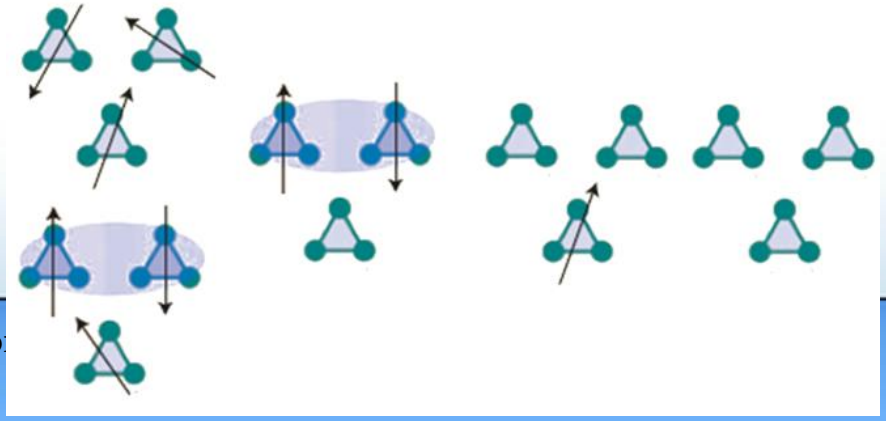
Magnetic Trends…



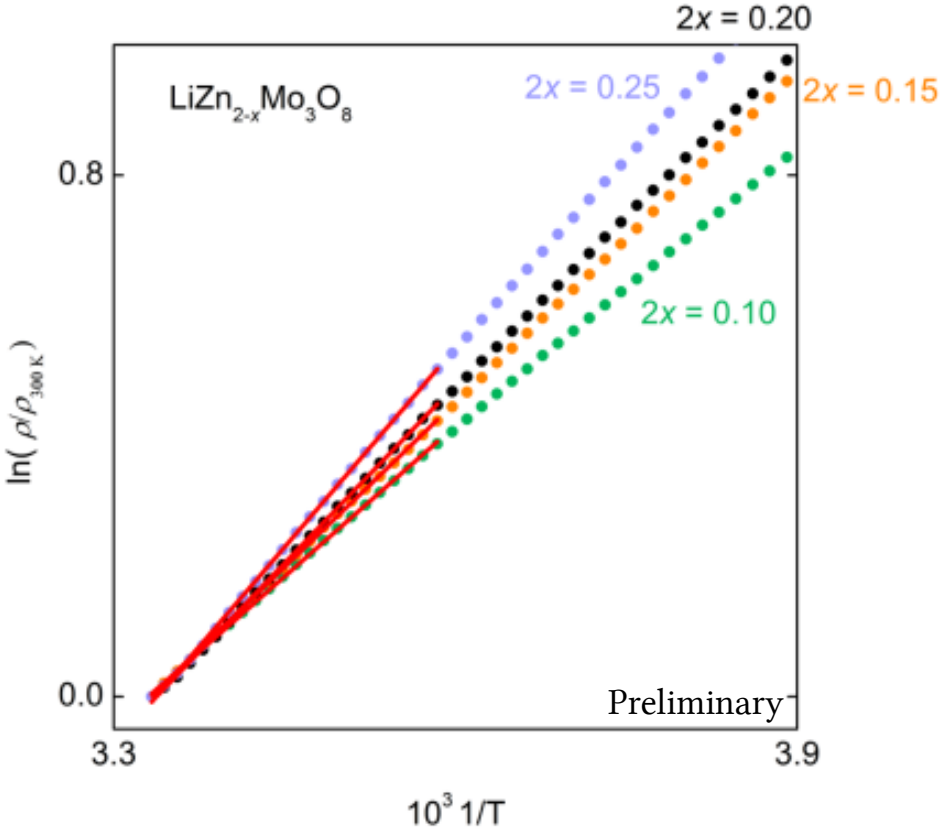


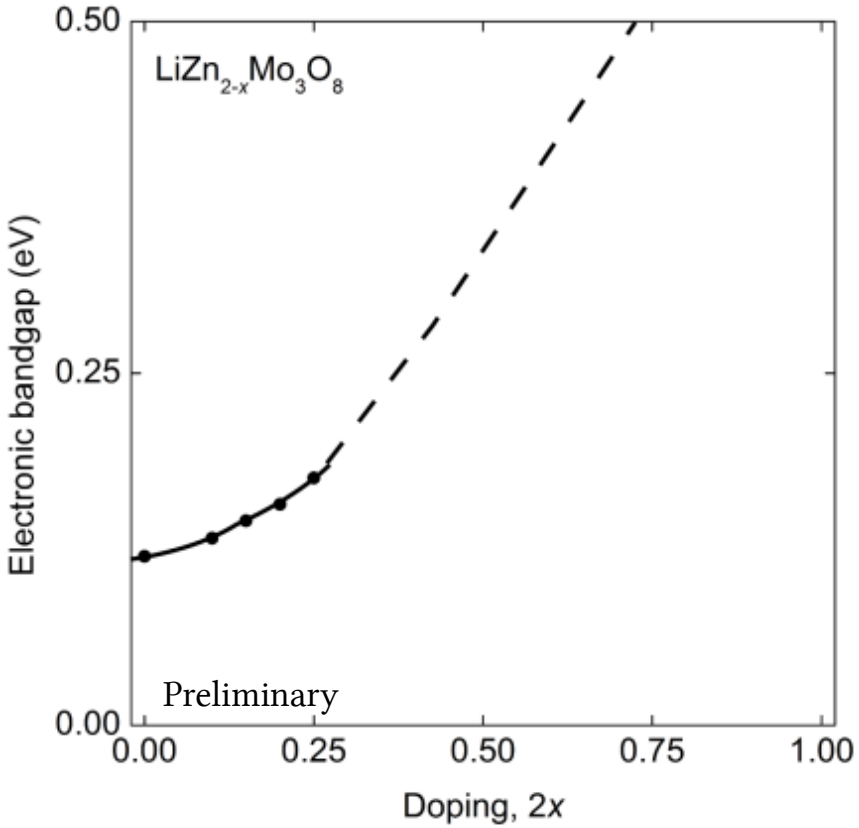
T > 150 K

25 < T < 85 K



Band gap increases on doping…

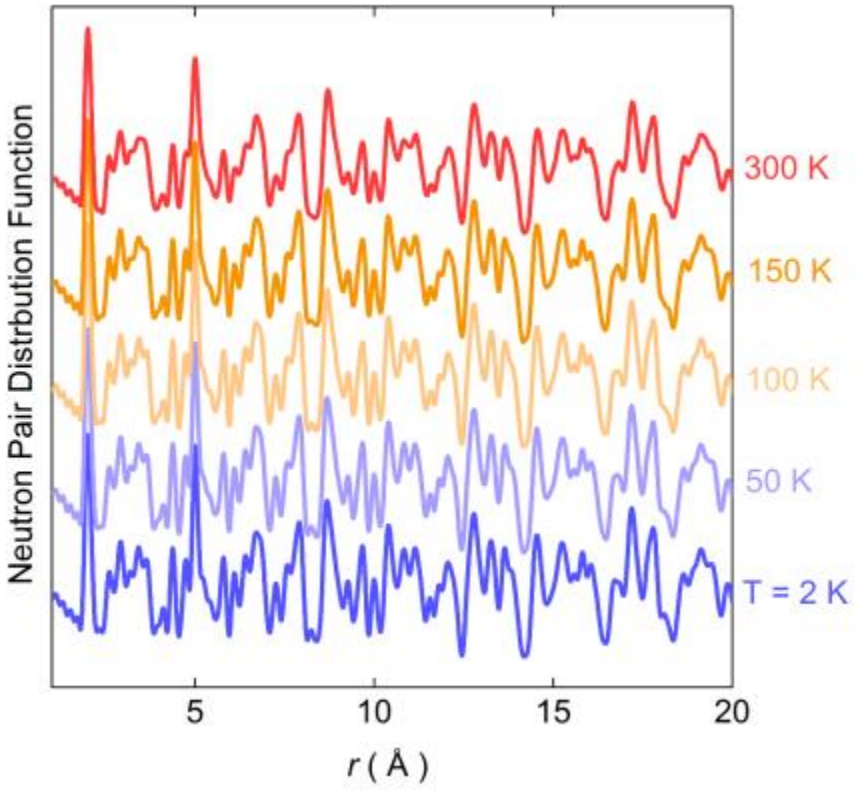


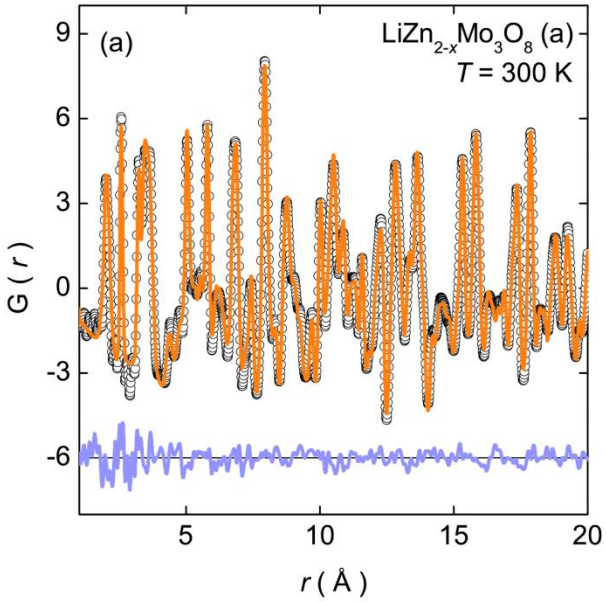


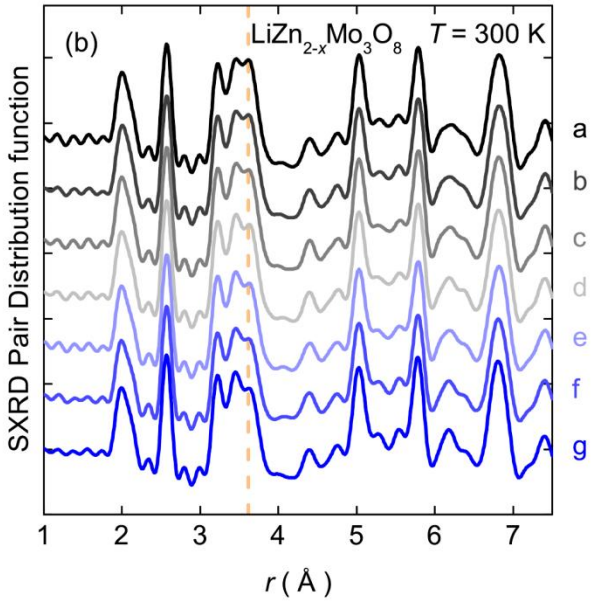
Maybe Anderson Localization Wins?

J.P. Sheckelton, et al., Materials Horizons 2, 76-80 (2015)

No (Static) Local Structural Distortions





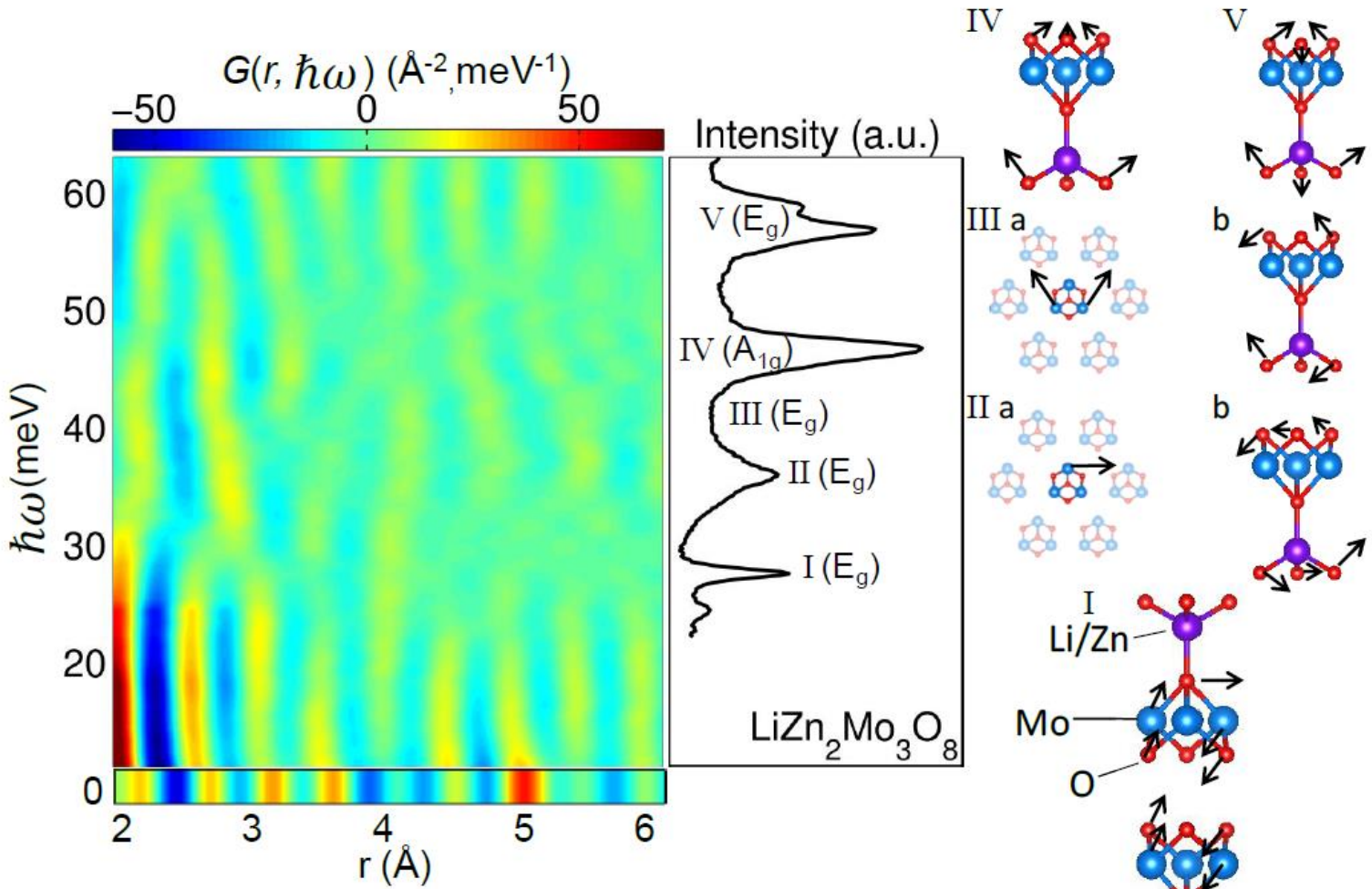


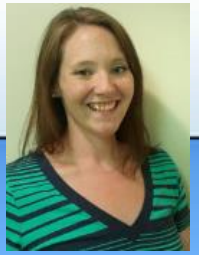
$$S(Q) = 1 + \frac{I^{\mathrm{coh}}(Q) - \sum c_i |f_i(Q)|^2}{|\sum c_i f_i(Q)^2|}$$

$$G(r) = 4\pi r[\rho(r) - \rho_0] = \frac{2}{\pi}\int_0^{Q_{\max}} Q[S(Q) - 1]\sin(Qr)dQ$$

J.P. Sheckelton, et al., Materials Horizons 2,

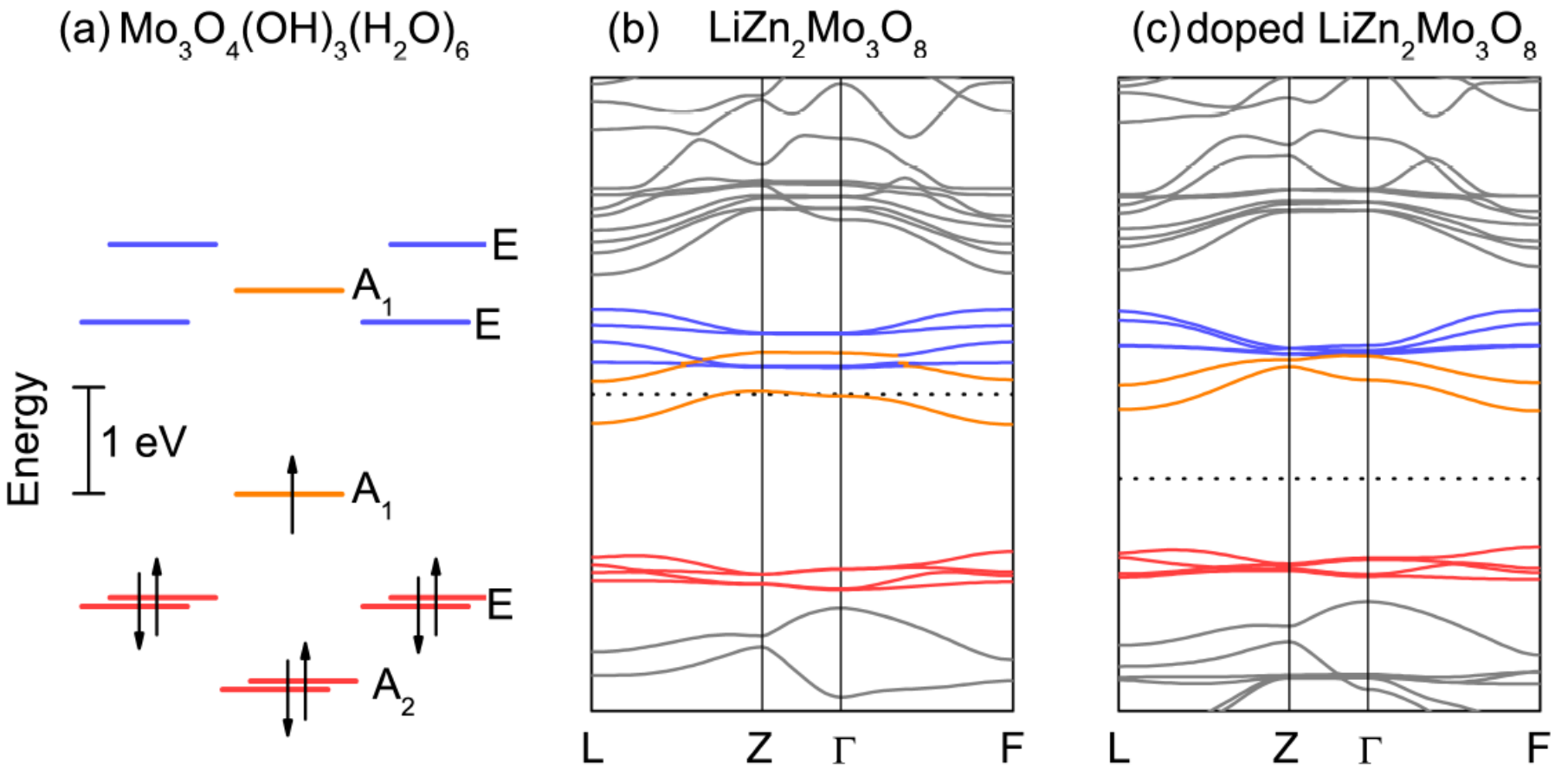
Role of Molecular Vibrations?





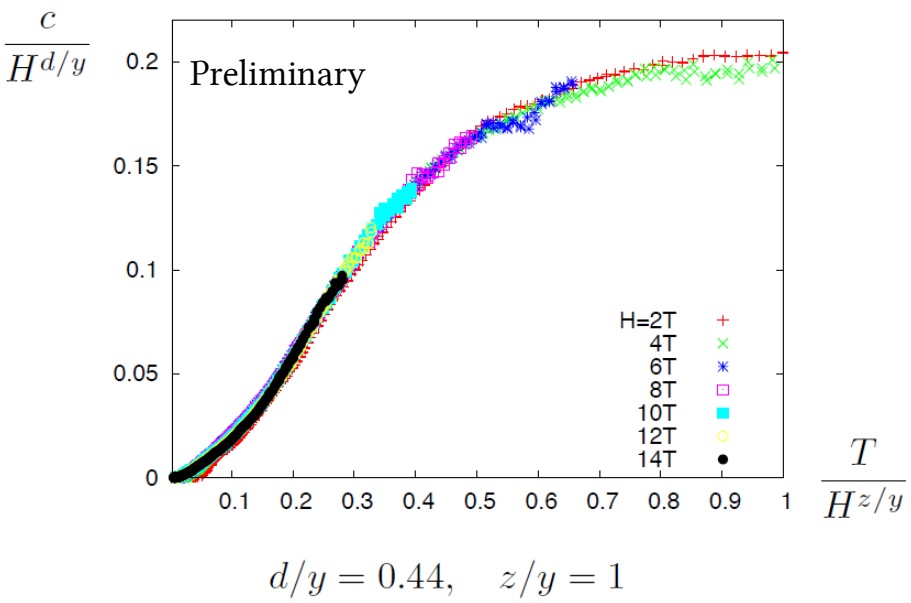
A. Fry, et al., Under Review

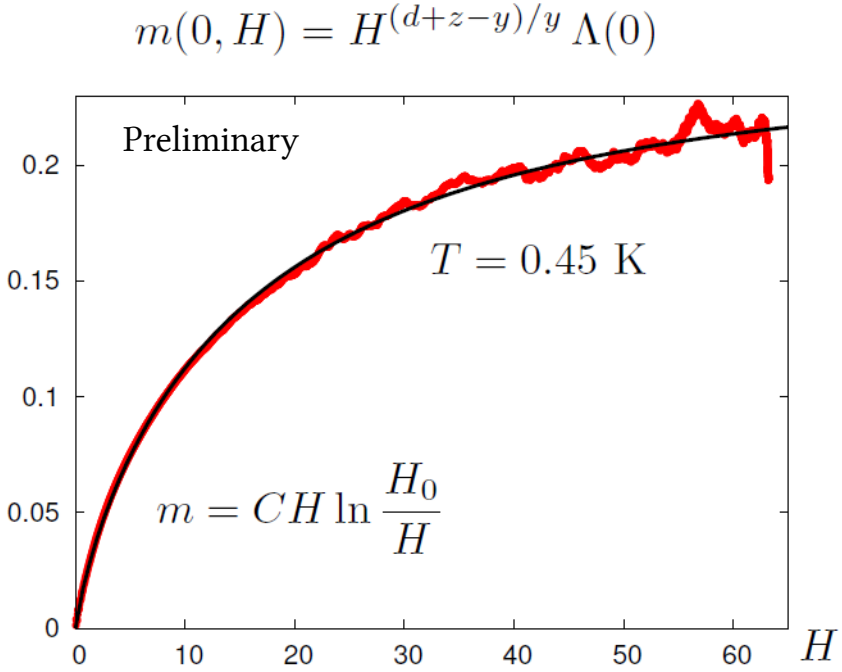
Insight from (simplistic) DFT+U



J.P. Sheckelton, et al., Materials Horizons 2, 76-80 (2015)

Critical Scaling?



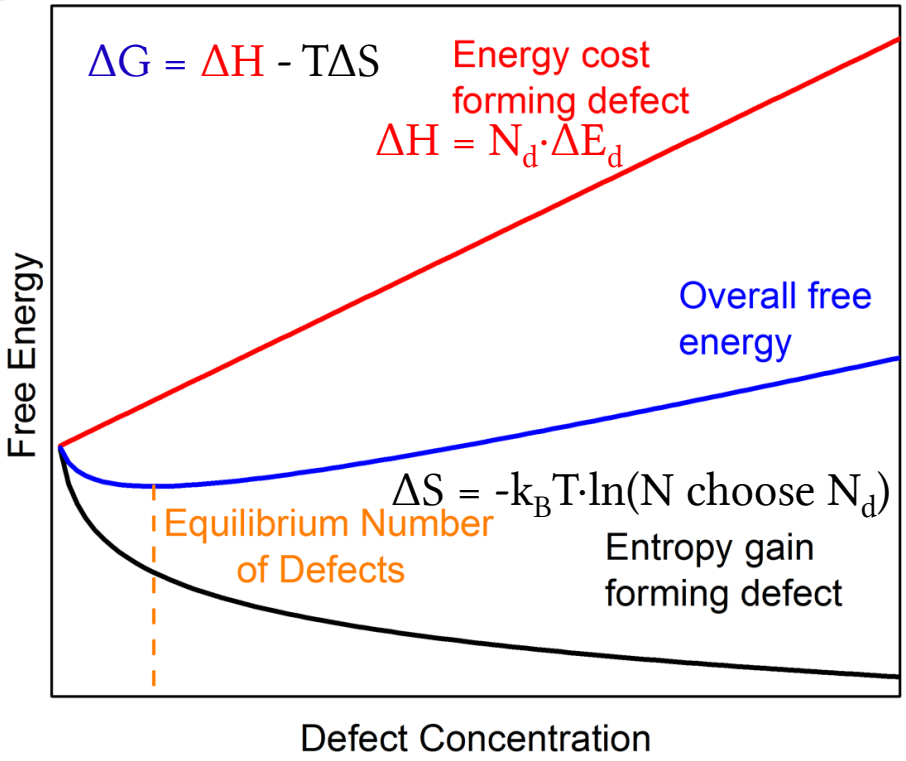


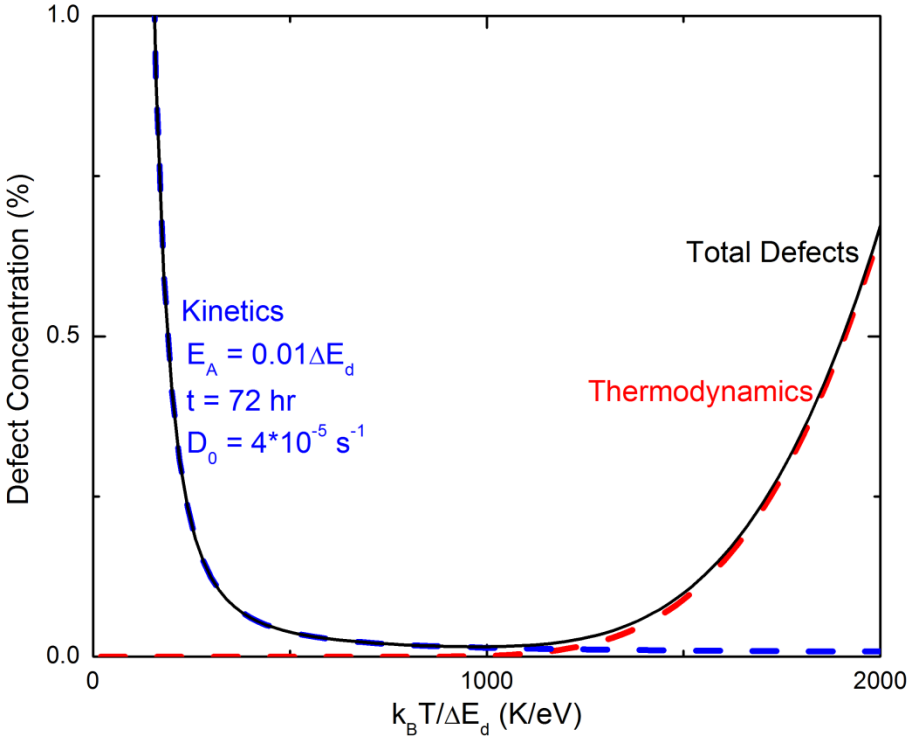
Not quite…

Conclusions

- $LiZn_2Mo_3O_8$ is one of a handful of materials known to have a valence bond structure, and the gapless nature means it is a candidate spin liquid
- Strong interactions between isolated spins on magnetic clusters possible
- Charge doping never induces metallicity, let alone superconductivity
  - Anderson localization too strong?
  - Charge gap too big? (unlikely…)
- Dynamic local structure measurements constrain role of the lattice
- Stay tuned… this cluster approach is not a one-time affair!

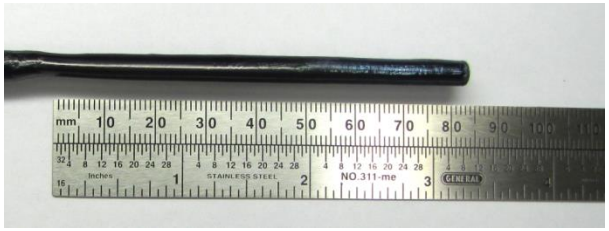
"Defects Rule, Physics Drools" (and Entropy Always Wins)





And now my challenge to you: we now have a variety of strong candidates for magnets with non-trivial magnetic ground states. What experimentally achievable devices, etc. should we build with these (e.g. how do I make a qubit?)

IQM Crystal Growth Successes

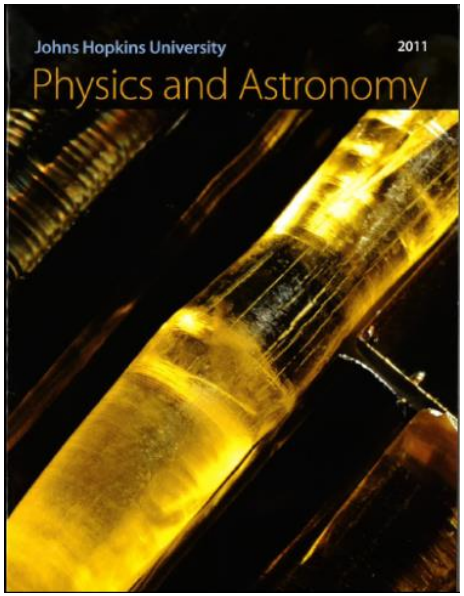


SmB_6

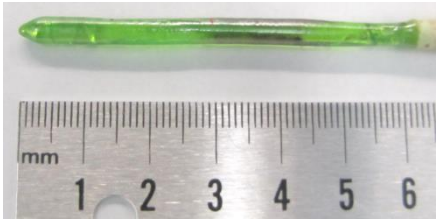
$SrCr_2O_4$

Tl_5Te_3

$MgCr_2O_4$



$SrHo_2O_4$



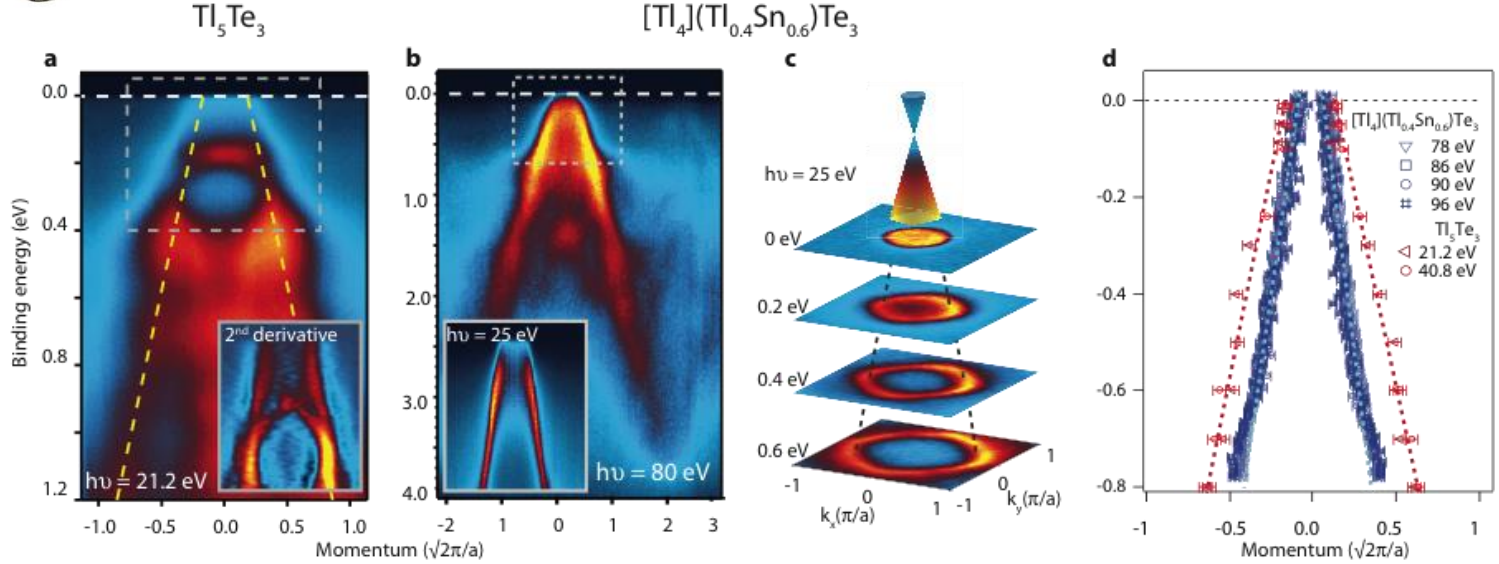
$Pr_2Zr_2O_7$

$CoNb_2O_6$, $NiNb_2O_6$

Not an exhaustive list!

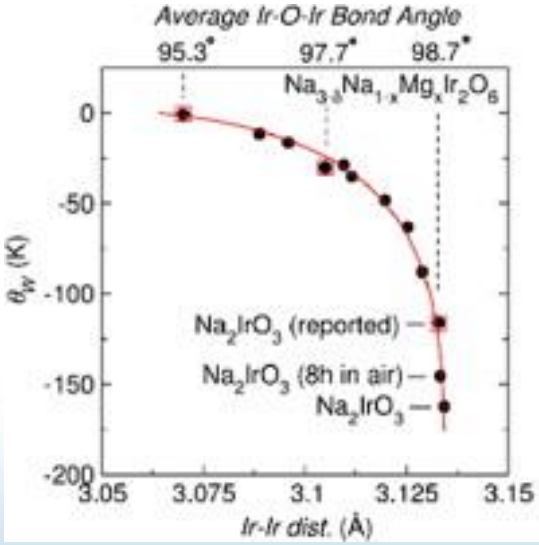
We are actively seeking external funding to turn this into an external user facility

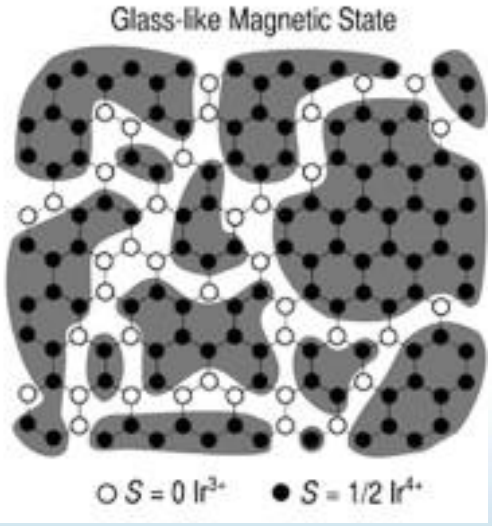
Only One Example of What We Do!



Dirac-like surface states plus superconductivity in perovskite Tl_5Te_3

Phys. Rev. Lett. 112, 017002 (2014)





$Na_{3-y}Ir_2(Na_{1-x}Mg_x)O_6$
$[Na_2IrO_3]$

J. Sol. St. Chem.
doi:10.1016/j.jssc.2014.03.013

Thank You!
https://occamy.chemistry.jhu.edu