The Search for Spin Liquid Ground States in Real Materials





 $\mathbf{H} = 2\mathbf{J} \ \Sigma_{ij} \ \mathbf{S_i} \cdot \mathbf{S_j}$





Detection of Antiferromagnetism by Neutron Diffraction*

C. G. SHULL Oak Ridge National Laboratory, Oak Ridge, Tennessee

AND J. SAMUEL SMART Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland August 29, 1949



Clifford G. Shull, MIT, Camebridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.

tiferromagnet



NEW YORK, N. Y.



[CONTRIBUTION FROM THE GATES CHEMICAL LABORATORY, CALIFORNIA INSTITUTE OF TECHNOLOGY, NO. 506]

The Structure and Entropy of Ice and of Other Crystals with Some Randomness of Atomic Arrangement

By LINUS PAULING



Exotic magnetism was of

interest from the beginning!



PHYSICAL REVIEW

VOLUME 102, NUMBER 4

MAY 15, 1956

Ordering and Antiferromagnetism in Ferrites

1956

P. W. ANDERSON Bell Telephone Laboratories, Murray Hill, New Jersey (Received January 9, 1956)

The octahedral sites in the spinel structure form one of the anomalous lattices in which it is possible to achieve essentially perfect short-range order while maintaining a finite entropy. In such a lattice nearestneighbor forces alone can never lead to long-range order, while calculations indicate that even the longrange Coulomb forces are only 5% effective in creating long-range order. This is shown to have many possible consequences both for antiferromagnetism in "normal" ferrites and for ordering in "inverse" ferrites.

PHYSICAL REVIEW

VOLUME 79, NUMBER 2

Antiferromagnetism. The Triangular Ising Net

G. H. WANNIER Bell Telephone Laboratories, Murray Hill, New Jersey (Received February 11, 1950)

1950

JULY 15, 1950

Z. Physik B 33, 31-42 (1979)



1979

Département de Recherche Fondamentale, Laboratoire de Diffraction Neutronique, Centre d'Etudes Nucléaires de Grenoble, Grenoble, France

Received September 21, 1978



Jacques Villain

Physics of Frustration in 2D on Triangles





Triangular

Antiferromagetism on a triangle: ↑↓ alignment energetically preferred, but 1 of 3 "bonds" must be ↑↑



Physics of Frustration in 3D on Tetrahedra



Cubic Pyrochlore Lattice

Spin Ice: Ferromagnetic interactions combined with local Ising anisotropy leads to 6-fold degeneracy on a single tetrahedron - macroscopic degeneracy on 3D crystal

M.J. Harris, S.T. Bramwell, D.F. McMorrow, T. Zeiske, and K.W. Godfrey, PRL 79 2554 (1997)

P.W. Anderson and the Resonating Valence Bond Model 1973





 $\mathcal{H}=JS_i, S_j, J>0$

Mat. Res. Bull. Vol. 8, pp. 153-160, 1973. Pergamon Press, Inc. Printed in the United States.

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR ?*

P. W. Anderson Bell Laboratories, Murray Hill, New Jersey 07974 and Cavendish Laboratory, Cambridge, England

P.W. Anderson and the Resonating Valence Bond Model 1987

Science

The Resonating Valence Bond State in La_2CuO_4 and Superconductivity

P. W. Anderson

The oxide superconductors, particularly those recently discovered that are based on La_2CuO_4 , have a set of peculiarities that suggest a common, unique mechanism: they tend in every case to occur near a metal-insulator transition into an odd-electron insulator with peculiar magnetic properties. This insulating phase is proposed to be the long-sought "resonating-valence-bond" state or "quantum spin liquid" hypothesized in 1973. This insulating magnetic phase is favored by low spin, low dimensionality, and magnetic frustration. The preexisting magnetic singlet pairs of the insulating state become charged superconducting pairs when the insulator is doped sufficiently strongly. The mechanism for superconductivity is hence predominantly electronic and magnetic, although weak phonon interactions may favor the state. Many unusual properties are predicted, especially of the insulating state.





 La_2CuO_4

- Cu^{2+} • O^{2-} • O^{2-}
 - La ³⁺

2D Heisenberg S=1/2 Kagome AF

(b) $ZnCu_3(OH)_6Cl_2$



A World of Kagome Minerals



Christo Kapellas

Haydeeite 2006, Chile Haydee copper mine

2D Heisenberg S=1/2 Kagome AF





 $E_{classical} = zJ\mathbf{S_i} \cdot \mathbf{S_j}/2$

 $\mathcal{H}=JS_i, S_j, J>0$



2D Heisenberg S=1/2 Kagome AF



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*



Cu Zn Zn Cl

PHYSICAL REVIEW B 88, 075106 (2013)

First-principles determination of Heisenberg Hamiltonian parameters for the spin- $\frac{1}{2}$ kagome antiferromagnet ZnCu₃(OH)₆Cl₂

Harald O. Jeschke,* Francesc Salvat-Pujol, and Roser Valentí

TABLE II. Exchange coupling constants for $ZnCu_3(OH)_6Cl_2$ (herbertsmithite) determined from total energies of nine different spin configurations. Energies were calculated with GGA + U functional at U = 6 eV, J = 1 eV and with atomic-limit double-counting correction.

Name	$d_{\rm Cu-Cu}$	Туре	$J_i (\mathbf{K}) U = 6 \text{ eV}$
	Kago	me laver couplings	
J_1	3.4171	Kagome nn	182.4
J_3	5.91859	Kagome 2nd nn	3.4
J_5	6.8342	Kagome 3rd nn	-0.4
	Inte	erlayer couplings	
J_2	5.07638	Interlayer 1st nn	5.3
J_4	6.11933	Interlayer 2nd nn	-1.5
J_6	7.00876	Interlayer 3rd nn	-6.4
J_7	8.51328	Interlayer 4th nn	3.0
J_9	9.17347	Interlayer 6th nn	2.5

Olariu et al, PRL 100, 087202 (2008)

Best Understood Spin Liquid: S=1/2 Heisenberg AF chain

Deconfined spinons as elementary excitations in zero field Spin waves in field-polarized state



$CuSO_4 \cdot 5D_2O_4$

M. Mourigal et al, Nature Physics, 9, 435, 2013

2D Heisenberg S=1/2 Kagome AF

(a) $ZnCu_3(OH)_6Cl_2$





2D S=1/2 Kagome Antiferromagnet: Gapped or Ungapped in Herbertsmithite?



P. Khuntia et al., Nature Physics 16, 469 (2020)

> Barthélemy et al., Phys. Rev. X (2022)

Gap consistent with 0 K



Physics of Frustration in 3D on Tetrahedra



Cubic Pyrochlore Lattice

Spin Ice: Ferromagnetic interactions combined with local Ising anisotropy leads to 6-fold degeneracy on a single tetrahedron - macroscopic degeneracy on 3D crystal

M.J. Harris, S.T. Bramwell, D.F. McMorrow, T. Zeiske, and K.W. Godfrey, PRL 79 2554 (1997)



Spin Ice

• Classical macroscopic degeneracy

Supports monopole excitations

Rare example of deconfined excitations in 3D



C. Castelnovo, R. Moessner, and S.L. Sondi, Nature, 451, 43 (2007) L. Balents, Nature, 464, 199 (2010) **Pyrochlores have the quintessential** lattice for the phenomena of magnetic frustration in 3D.

(Dy ³⁺) ₂ (Ti ⁴⁺) ₂ O ₇ (Ho ³⁺) ₂ (Ti ⁴⁺) ₂ O ₇															/			
				4f ⁹	Э,	J =	= 1	5/2	2								2	
H	2 11A 2A			<u>Δ</u> .	f 10	_		8				13 IIIA 3A	14 IVA 4A	15 VA 5A	16 VIA 6A	17 VIIA 7A	He	
Li Magan 1941	4 Be Beryllium 9.01218											5 B Boron 10.011	6 Carbon 12.011	7 Nitropan 14.00074	8 Ovygen 15.8994	9 F Pluotine 10.998403	10 Ne Neon 20.1797	
la dun estre	12 Mg Magnashum 24.505	3 111B 3B	4 IVB	5 VB 58	6 VIB 6B	7 VIIB 7B	8 ,	9 VIII— 8	10	11 IB 1B	12 IIB 2B	13 Al Aluminum 26.961539	14 Silicon 28.0855	P Phosphorus 30.973782	16 Sutter 32.005	17 Cl Chlorine 35.4527	18 Ar 47944 28344	
K sesture costa	20 Calcium 40.078	21 Scandle 44,9559	22 Ti Tianium 47.88	V medium 50.9415	24 Cr Chrymlum 51,9961	25 Mn Manganese 54,938	26 Fe	27 Co Coball 56.5332	28 Ni 56.0934	29 Cu Cooper 63.546	30 Zn 2014 65.30	31 Gallum 68.732	32 Ge 0eemaalam 72.44	33 Ass 74.82159	34 See 58.96	Br Br 78.904	36 Kr Krypten 83.80	
Rb Million Antes	38 Sr 500x150m	39 Yitstum 88.90585	Zr Zirconium 91.224	41 Notem 92,90638	42 Mo Molytotenum 95.94	43 Tc Technedum 96.9072	HARU Ruthansium 191.07	45 Rh Phodum	46 Pd Patedum 108.42	47 Ag 58ver 107.6682	48 Cd Cadmium 112.411	49 In Indum	50 Sn 116.71	51 Sb Antimory 121.310	52 Te Telurium 127.4	53	54 Xe Xanon 131.29	
CS solum 200543	Ba Batum 137.327	57-71	72 Hf Hatolaum 178.40	73 Ta Tantalum 180.9479	74 W Tungatan 183.85	75 Re Rhenlum 186.207	76 Os Osmium 190.23	77 Ir 192.22	78 Pt Platinum 195.08	79 Au Guide 196, 5665	80 Hg Marcury 200.59	81 Ti 204.3833	82 Pb Lead 207.2	Bi Bi 206.94037	84 Polosium [208.9824]	85 At Astactions 200.9471	86 Rn Radon 222.0176	
nclum 1.0197	88 Ra Radum 220.0254	89-103	104 Rf Putherfordium [281]	Dubnium [202]	106 Sg Eesteorghum [286]	107 Bh Botelum [284]	108 Hassium [209]	109 Mt EMI	110 Ds Darmataditum [209]	111 Rg	112 Cn Copersiders (277)	Uut Uurthur	FI FI Flerovium [289]	Uup Januar	LV LV LVermotium [294]	Ununseptium unknown	University of the second	
La	nthanide Series	57 La		59 Pr	Notes		1 52 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5			1 ⁶⁵ T	66 Dy Dyspecial	HC		69 Tr				
4	ctinide Series	89 Activity 227.027	90 Th Thorise 222.038	91 Protection 201 000	han 92 Uuuuuuu Maalaa	93 Neptoria	94 PL Planet	95 Anarita 20.00	n 96 Cn Outle Strat	n 97 Bk Deckelle Data and	a Daliforni a Daliforni	Es Deusis	100 Fm Fm 257 00	101 MC Manadatan 201	102 No No No			

Gardner, Gingras, and Greedan, Rev. Mod. Phys **82**, 53 (2010) Hallas, Gaudet, and Gaulin, ARCMP, **9**, 105 (2018)



"disorder-free" spin glass classical spin ice

quantum spin ice metal-insulator transitions spin liquid order-by-disorder moment fragmentation

Missing Pauling entropy is a defining characteristic of classical spin ice



A. P. Ramirez, A. Hayashi, R. J. Cava, R. Siddharthan, Nature 399, 333 (1999) B. C. den Hertog, M. J. P. Gingras, Phys. Rev. Lett. 84, 3430 (2000)



T. Fennell et al., Science, 326 (5951): 415-417 (2009)

Quantum Spin Ice: A U(1) Quantum Spin Liquid

$$\mathcal{H}_{\mathrm{U}(1)} = \frac{\mathcal{U}}{2} \sum_{\langle \mathbf{rr}' \rangle} \left[(\nabla_{\mathrm{O}} \times \mathcal{A})_{\mathbf{rr}'} \right]^2 + \frac{\mathcal{K}}{2} \sum_{\langle \mathbf{ss}' \rangle} \mathcal{E}_{\mathbf{ss}'}^2$$

$$J_{zz}$$
Magnetic Monopoles
Energy
$$J_{zz}^{3}/J_{zz}^{2}$$

$$J_{\perp}^{3}/J_{zz}^{2}$$
Visons
Photon Excitations

- QSI possesses an emergent QED
- Can tunnel between ice rules states
- Introduces fluctuations in the gauge field Hermele et al, PRB 69, 064404 (2004) Banerjee et al, PRL, 100, 047208 (2008) Shannon et al, PRL 100, 047201 (2012) Benton et al, PRB 86, 075154 (2012)
 Gingras and McClarty, RPP,77, 056501 (2014).



Pyrochlores have the quintessential lattice for the phenomena of magnetic frustration in 3D.

$(Ce^{3+})_2(Zr^{4+})_2O_7$ $(Ce^{3+})_2(Sn^{4+})_2O_7$

1 H Hydrogen 1.0079	2 IIA 2A			4	4f1	J=	=5/	2				13 IIIA 3A	14 IVA 4A	15 VA 5A	16 VIA 6A	17 VIIA 7A	² He Helun 4.00200
3 Li Litter 6.941	Be Beryllum 9.01218											5 B Boron 16411	6 Carbon 12.011	7 N Nitropen 14.00074	8 Orygen 15.8664	9 F	10 Neo 20.1797
Na Bodum 22.000708	Magnesium 24.305	3 111B 3B	4 IVB 4B	5 VB 58	6 VIB 6B	7 VIIB 7B	8 •		10	11 IB 1B	12 IIB 2B	Al Al Alminum 26.961539	14 Si 580000 28.0000	Phosphorus 30.972762	16 Sultur 32.005	17 Cl Chlorine 35.4527	18 Arpor 20.340
19 K Potasalum 38.0083	20 Ca Calithum 40.078	Sc Sc Brandhum 44.99391	22 Ti	23 Vanadium 50.9415	24 Cr Chromium 51,9901	25 Mn Manganese 54.938	26 Fe	27 Cobuit 58.5052	28 Ni Michael 55.0004	29 Cu Cooper 63.546	30 Zn 2011	Ga Gallum 68.732	Ge	33 As Armenia 74.02150	34 Se ⁵⁴⁰⁰⁰⁰	Branine 78.904	36 Kr 53.60
37 Rb Rubicium 85.4678	38 Sr Bityontium 87.42	39 Yindur 88.9058	40 Zr 201224	Nb Pastores Random	42 Mo Molybdanum 95.94	43 Tc Technedium 94.9072	44 Ruthenlum 191.07	45 Rh Phodum 102,9005	46 Pd Palladum 106.42	47 Ag 58ver 107.6662	48 Cd Cadmium 112.411	49 In Indust	50 Sn 111.71	Bb Filmony Tati 1960	52 Telutum 127.6	53 Iodine 125.90447	54 Xe Xanon 11120
55 Cs Cestum 132,00543	56 Batun 137.327	57-71	Hf Hatslum 178.49	73 Ta Tantalun 180.9479	74 W Tungatan 183.85	75 Re Rhandum 186.207	76 Os Osandum 190.23	77 Ir 192.32	78 Patinum 195.08	79 Au 196.9665	80 Hg 200.59	81 Ti 204.3833	PD Lead 2072	83 Bi 206.94037	84 Po Polocium (206.9624)	Astatine 200.9671	Radon 222.0176
87 Fr Prancium 223.0197	88 Radum 225.0254	89-103	104 Rf Ruthertordium	105 Db Dubnium [202]	106 Sg Eestoophen [266]	107 Bh Botelum [254]	108 Hss Hassalum [2009]	109 Mt Methodum [264]	110 Ds Dermetedium [209]	111 Rg Rosentration	Coperticium	Unutrium Ununtrium unknown	Fil Fil Filerovium [289]	Uup Uutoen	Lv Lv [294]	University of the second	
La	nthanide Series	57 La Landhan 138-900	58 Cee 56 540.11		60 Nd Necessaria	61 Promethic 144,911	62 Samadu 7 150.36	63 Europher 151.965	64 Gadedada 157,25	65 Tb Terster 158.925	66 Dysprese H 162.50	67 Hotelau 104.930	68 Erster 187.3	69 Tn Toda	70 Yesta 173.0	71 Lu	
4	ctinide Series	89 Activate 227 A2	There are a second	91 Patentes	92 U U U U U U U U U U U U U U	93 Nepture 237.64	94 Put Put 241 000	95 Am American 243.001	96 Contection	1 97 Bk Derhelle Def 207	98 Cf California 201.079	99 Es					

Gardner, Gingras, and Greedan, Rev. Mod. Phys **82**, 53 (2010) Hallas, Gaudet, and Gaulin, ARCMP, **9**, 105 (2018)



"disorder-free" spin glass classical spin ice **quantum spin ice** metal-insulator transitions

spin liquid order-by-disorder moment fragmentation

Ce₂Zr₂O₇: No obvious phase transition for T > 0.06 K



$$\chi_{CEF} = \frac{N_A g_J^2 \mu_B^2 X}{k_B Z} \sum_{\alpha} \left(\sum_n \frac{|\langle n | J_\alpha | n \rangle|^2 e^{-E_n/T}}{T} + \sum_n \sum_{m \neq n} \frac{|\langle m | J_\alpha | n \rangle|^2 (e^{-E_n/T} - e^{-E_m/T})}{E_m - E_n} \right)$$



Rare Earth-based Insulators often display A separation of Energy Scales: SOC >> CEF >> Exchange



Different Types of CEF Ground State Doublets

Define pseudospin-1/2 operator acting within states of CEF doublet



Behavior of this operator under time reversal and local D_{3d} symmetry divides $R_2M_2O_7$ pyrochlores into three groups

	# of f electrons	Symmetry of ground state doublet
Dy, Yb, Er	Odd: Kramer's (dipolar)	S ^z , S ^x , S ^y transform like dipoles
Ce, Sm, Nd	Odd: Kramer's (dipole-octupole)	S ^z , S ^x transform like dipoles S ^y transforms like an octupole
Ho, Tb, Pr	Even: non-Kramers	S ^z transforms like dipoles S ^x , S ^y tranform like quadrupoles

High Energy Physics: CEF excitations and Dipole-Octupole Ground State in Ce₂Zr₂O₇



Moment in the GS doublet is small

$$\boldsymbol{\mu}_{GS} = 1.286 \boldsymbol{\mu}_{B}$$



J=7/2 excited state multiplet is ~ 200 meV above the J=5/2 GS, as expected

J. Gaudet et al, PRL 122, 187201 (2019)

Two components of S_{eff}=1/2 for Ce³⁺ in Ce₂Zr₂O₇ transform as Dipoles; One component transforms as an Octupole



Smith et al, PRX 12, 021015 (2022)

Low energy neutron scattering from Ce₂Zr₂O₇



T=0.06 K - T=2 K

Classical spin ice

Quantum spin ice



Inelastic scattering shows no static moment at any T > 0.06 K

J. Gaudet et al, PRL 122, 187201 (2019)

Polarized diffraction from single crystal Ce₂Zr₂O₇ resembles classical spin ice Ho₂Ti₂O₇



$Ce_2Zr_2O_7$

Quantum Spin Ice/Liquid Smith et al, PRX 12, 021015 (2022)

$Ho_2Ti_2O_7$

Classical Spin Ice Fennel et al, Science, 326, 415 (2009)

Heat Capacity and Neutron Scattering Place Ce₂Zr₂O₇ within its Generalized Hamiltonian Phase Diagram



 4f¹ Ce₂X₂O₇ with X=Zr, Sn: No LRO, freezing for T>0.08 K

 Use NLC to Fit high T Cp and estimate Hamiltonian

• Strong evidence for U(1) π QSL ground state for Ce₂Zr₂O₇



Kitaev Physics on a 2D Honeycomb Lattice



PRL 102, 017205 (2009)

PHYSICAL REVIEWZZETEQ

week ending 9 JANUARY 2009

XX

 \mathbf{J}_{ij} 3 × 3

 $\mathcal{H}_{ij} = J_{ij} \,\mathbf{S}_i \cdot \mathbf{S}_j + K_{ij} \,S_i^{\gamma} S_j^{\gamma} + \Gamma_{ij} \left(S_i^{\alpha} S_j^{\beta} + S_i^{\beta} S_j^{\alpha} \right)$

 $+ \Gamma_{ij}^{\prime} \left(S_i^{\gamma} S_j^{\alpha} + S_i^{\gamma} S_j^{\beta} + S_i^{\alpha} \$_j^{\gamma} + S_i^{\beta} S_j^{\gamma} \right)$

77

Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models

G. Jackeli^{1,*} and G. Khaliullin¹ ¹Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany (Received 21 August 2008; published 6 January 2009)

(b)

 $\not\leftarrow$

 t^2/U

900

 $\Gamma_{ii} p_z$

 $\mathbf{D}_{ij} \propto \mathbf{D}_{ij} \otimes \mathbf{D}_{ij}$

́ΧΖ

Kitaev Physics on a 2D Honeycomb ^{t_{2g}} Lattice

 $\mathcal{H}_{ij} \neq \mathbf{S}_i \cdot \mathbf{J}_{ij} \cdot \mathbf{S}_j$

Kitaev Physics on a 2D Honeycomb Lattice



ARCMP 7, 195 (2016)

Kitaev Physics on a 2D Honeycomb Lattice: RuCl₃

(c) α -RuCl₃





The honeycomb lattice is a bipartite lattice; geometric frustration is not relevant

Kitaev Physics t^2/U $\Gamma_{ij} \propto \mathbf{D}_{ij} \otimes \mathbf{D}_{ij}$ \mathbf{A} \mathbf{A} \mathbf{A} $\Gamma_{ij} \propto \mathbf{D}_{ij} \otimes \mathbf{D}_{ij}$ \mathbf{A} </t

$$\mathcal{H}_{ij} = J_{ij} \mathbf{S}_{i} \cdot \mathbf{S}_{j} + K_{ij} S_{i}^{\gamma} S_{j}^{\gamma} + \Gamma_{ij} \left(S_{i}^{\alpha} S_{j}^{\beta} + S_{i}^{\beta} S_{j}^{\alpha} \right)$$

$$\mathbf{F}_{ij}$$

t^2/U Kitaev Physics $\Gamma_{ij} \propto D_{ij} \otimes D_{ij} \otimes D_{ij}$ A 2D Honeycom b Lattice



$\frac{-6.7}{t^2/U-5.1}$ Kitaev Physics $\Gamma_{ij} \propto D_{ij} \otimes \Omega_{ij}$ a 2D Honeycom Lattice



Kitaev Physics on a 2D Honeycomb Lattice



Table 2. Summary of magnetic parameters for honeycomb Na₂IrO₃, α -Li₂IrO₃, Li₂RhO₃, and α -RuCl₃. The latter material is discussed in section 2.3.2. See text for relevant references.

Property	Na ₂ IrO ₃	α -Li ₂ IrO ₃	Li ₂ RhO ₃	α -RuCl ₃
$\mu_{\mathrm{eff}}~(\mu_{\mathrm{B}})$	1.79	1.83	2.03	2.0 to 2.7
Θ_{iso} (K)	~ -120	-33 to -100	~ -50	$\sim +40$
Θ_{ab} (K)	-176	$\Theta_{ab} > \Theta_c$		+38 to +68
$\Theta_{c}(\mathbf{K})$	-40			-100 to -150
$T_N(\mathbf{K})$	13 - 18	~15	(6)	7 to 14
Order	Zigzag	Spiral	Glassy	Zigzag
k -vector	$(0, 1, \frac{1}{2})$	(0.32, 0, 0)	—	$(0, 1, \frac{1}{2})$

Winter S M et al., J.Phys.: Condens. Matter 29 493002 (2017)

$$(J_1, K_1, \Gamma_1)$$

$$J_3 = 0 \qquad \qquad J_1 = \cos \phi \sin \theta, K_1 = \sin \phi \sin \theta$$

 t_{2g}

Kitaev Physics on a 2D Honeycomb Lattice: RuCl₃

 $\mathcal{H}_{ij} = J_{ij} \,\mathbf{S}_i \cdot \mathbf{S}_j + K_{ij} \,S_i^{\gamma} S_j^{\gamma} + \Gamma_{ij} \left(S_i^{\alpha} S_j^{\beta} + S_i^{\beta} S_j^{\alpha}\right)$

Table 5. Bond-averaged values of the largest magnetic interactions (in units of meV) within the plane for α -RuCl₃ obtained from various methods. For [149], the two numbers represent the range of values found in various relaxed structures. 'Pert. Theo.' refers to second order perturbation theory, 'QC' = quantum chemistry methods, 'ED' = exact diagonalization, 'DFT' = density functional theory total energy, 'Exp. An.' = experimental analysis. See also figure 19.

Method	Structure	$y, \lambda, \lambda, j, \lambda, \lambda, y$	K_1	Γ_1	J_3	
Exp. An. [166]		-4.6	+7.0			
Pert. Theo. [149]	P3 ₁ 12	-3.5	+4.6	+6.4		
QC (2-site) [41]	<i>P</i> 3 ₁ 12	-1.2	-0.5	+1.0		_
ED (6-site) [45]	<i>P</i> 3 ₁ 12	-5.5	+7.6	+8.4	+2.3	
Pert. Theo. [149]	Relaxed	-2.8/-0.7	D $-9.5 - 3.0$	(-1) +3.7/+7.3		
ED (6-site) [45]	C2/m	-1.7	$\mathbf{D}_{ij} = 0.7 i$	$(-5)_{J})_{+6.6}$	+2.7	-(
QC (2-site) [41]	C2/m	+0.7	-5.1	+1.2		-
DFT [180]	C2/m	-1.8	-10.6	+3.8	+1.3	
Exp. An. [181]		-0.5	-5.0	+2.5	+0.5	



 $B_c \sim 7$

Kitaev Physics on a 2D Honeycomb Lattice: RuCl₃



Search for Spin Liquid Candidate Ground States in Real Materials

- Different routes to spin liquid ground states: geometry and competing interactions.
- Materials issues are present and somewhat uncontrolled in all candidate materials.
- Of the 4 examples discussed, 2 are relatively simple: Herbertsmithite (2D Kagome AF) is relatively simple due to its spin-only S=1/2. The character of its gap (gapped or gapless) remains an outstanding issue. Classical spin ice pyrochlores are relatively simple as they can be successfully modelled classically.
- Both 3D pyrochlore candidates and 2D Kitaev candidates require moderate to strong SOC, and anisotropic exchange as a consequence.