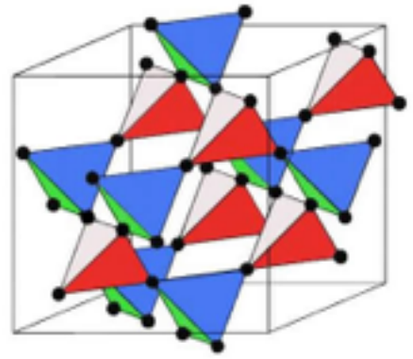
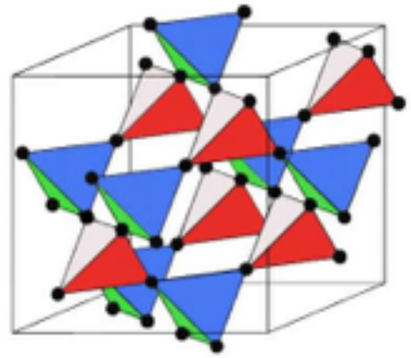
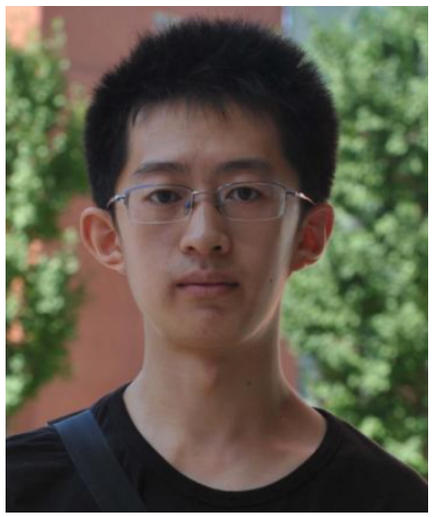


Competing orders and topological excitations in spin-1 pyrochlore antiferromagnets

Gang Chen
Department of Physics
Fudan University
(currently on a sabbatical leave to University of Hong Kong)





Fei-Ye Li
Fudan

PHYSICAL REVIEW B **98**, 045109 (2018)

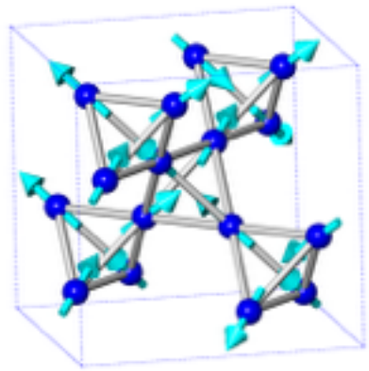
Competing phases and topological excitations of spin-1 pyrochlore antiferromagnets

Fei-Ye Li[1,2] and Gang Chen[1,2,3,4,*]

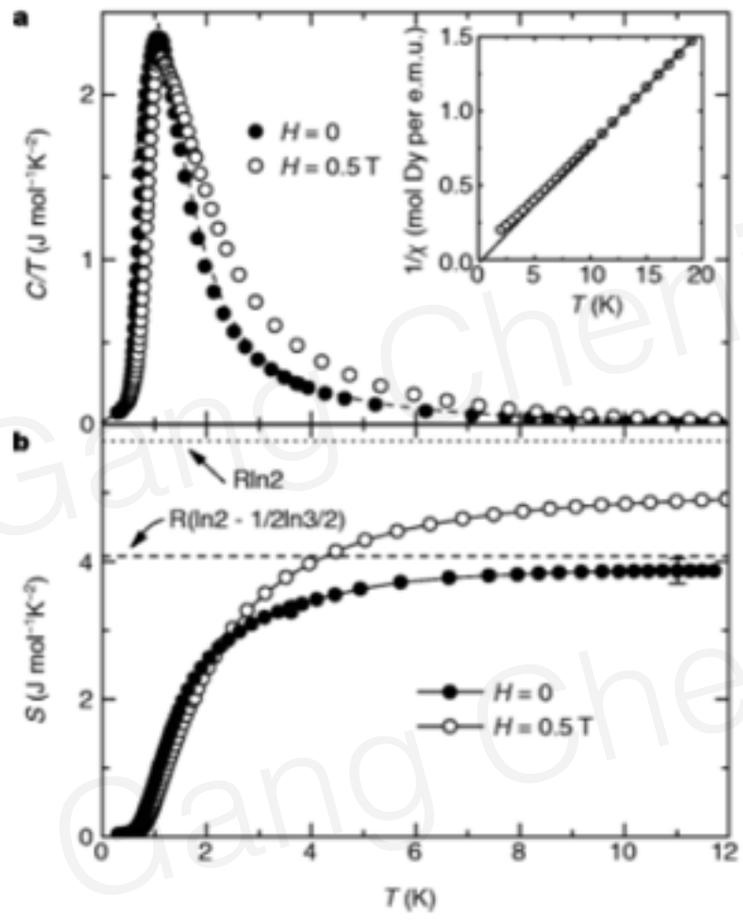
[1] *State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China*
[2] *Center for Field Theory and Particle Physics, Fudan University, Shanghai 200433, China*
[3] *Institute for Nanoelectronic Devices and Quantum Computing, Fudan University, Shanghai, 200433, China*
[4] *Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China*

Fei-Ye Li, GC, PhysRevB 98, 045109 (2018)

Everlasting success of spin-1/2 pyrochlores

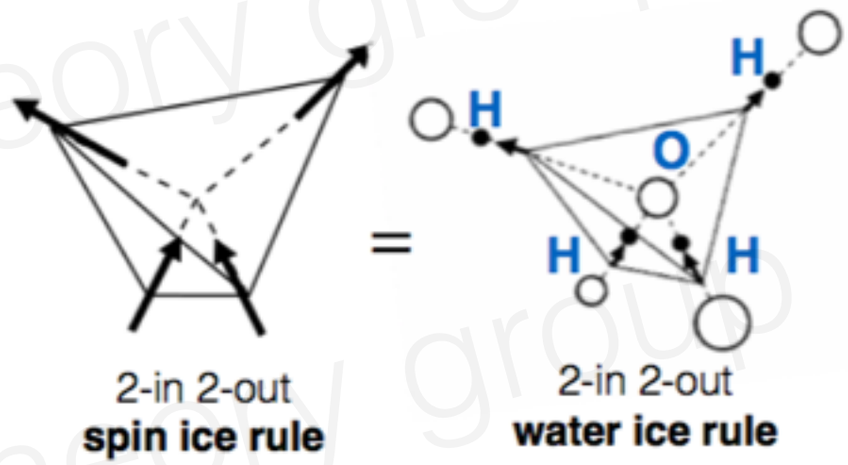


$Dy_2Ti_2O_7$



Pauling entropy in spin ice

Classical spin ice



2-in 2-out
spin ice rule

2-in 2-out
water ice rule

Ramirez, etc, Nature 1999,

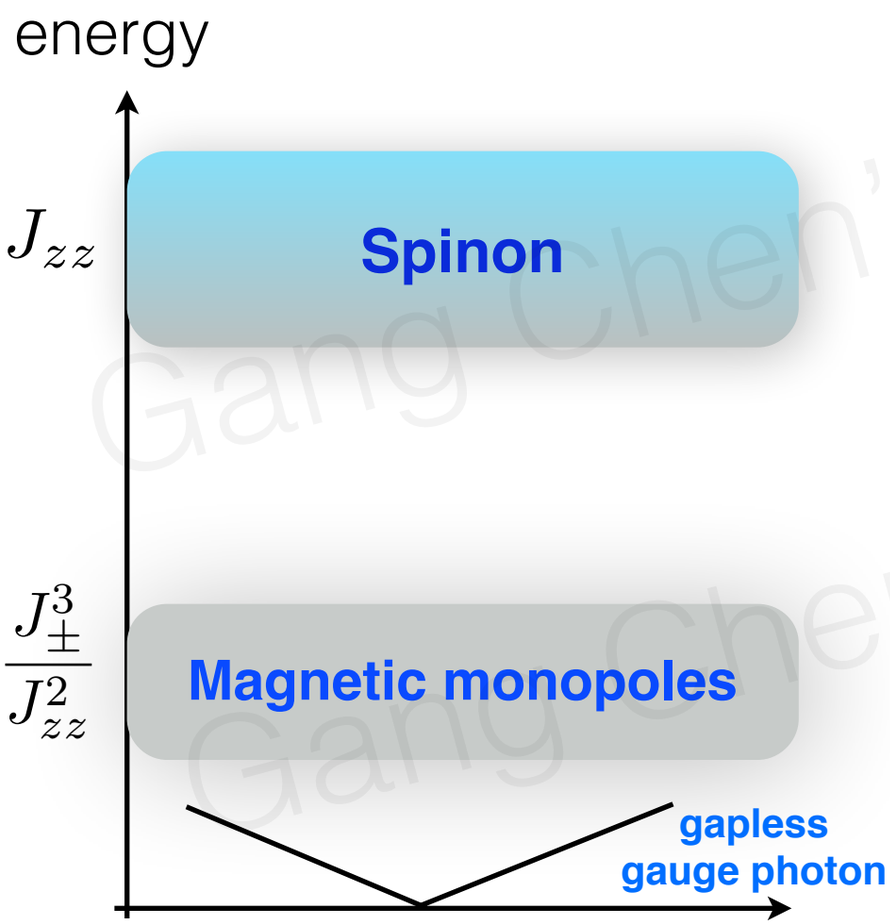
Gingras, etc, Science, 2009

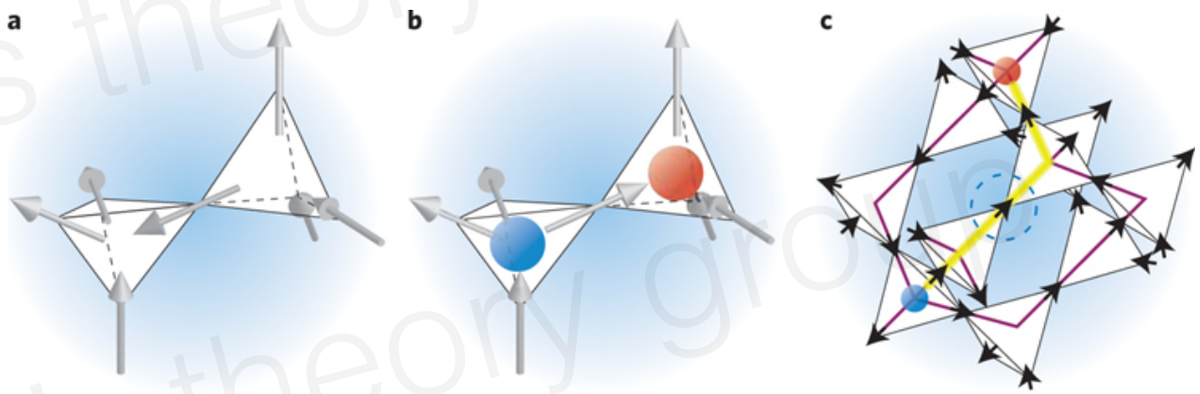
Castelnovo, Moessner, etc, Nature 2008

Gang Chen's theory group

Everlasting success of spin-1/2 pyrochlores

Quantum spin ice,
Quantum spin liquid





Figs from Moessner&Schiffer,2009

Spinon deconfinement

Gingras, Gaulin, Balents, Savary, SB Lee, GC, ….

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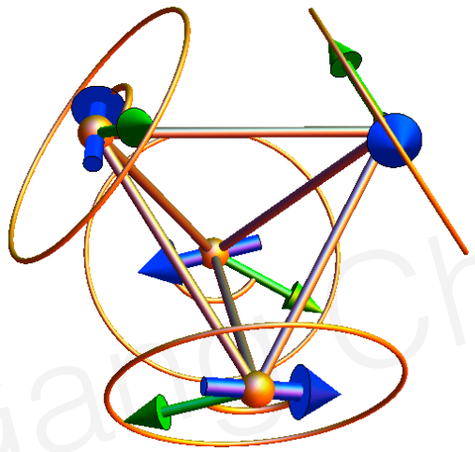
Everlasting success of spin-1/2 pyrochlores

Order by quantum disorder



XY pyrochlore

Kate Ross, Bruce Gaulin, etc

$Er_2Ti_2O_7$ Hamiltonian: The effective $S = 1/2$ description applies to $Er_2Ti_2O_7$ below about 74 K [2, 11]. Nearest-neighbor exchange dominates, for which the Hamiltonian is constrained by symmetry to the form [9]

$$H = \sum_{\langle ij \rangle} \Big[J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{\pm\pm} \left[\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right] + J_{z\pm} \left[\mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j \right] \Big], \qquad (6)$$

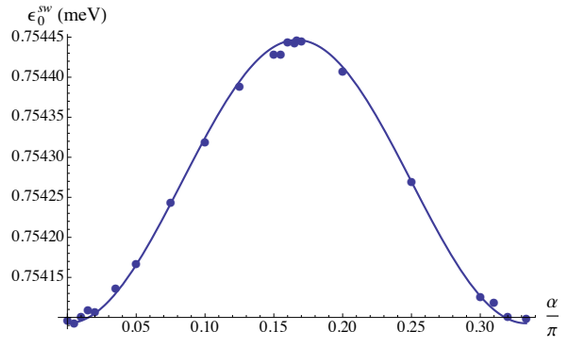


FIG. 2. Zero-point fluctuation energy ϵ_0^{sw} in the classically degenerate manifold parametrized by α. The peak-to-peak energy is $\lambda \approx 3.5\,10^{-4}$ meV.

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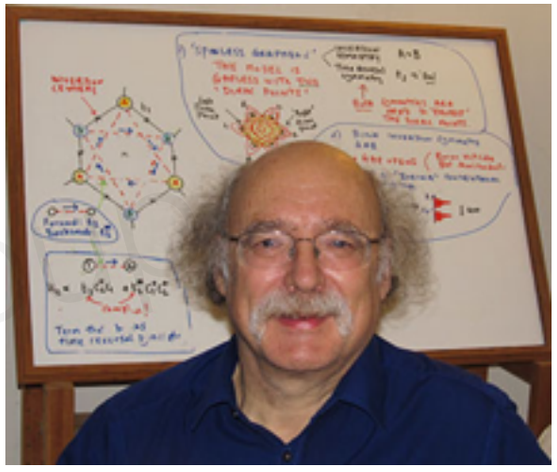
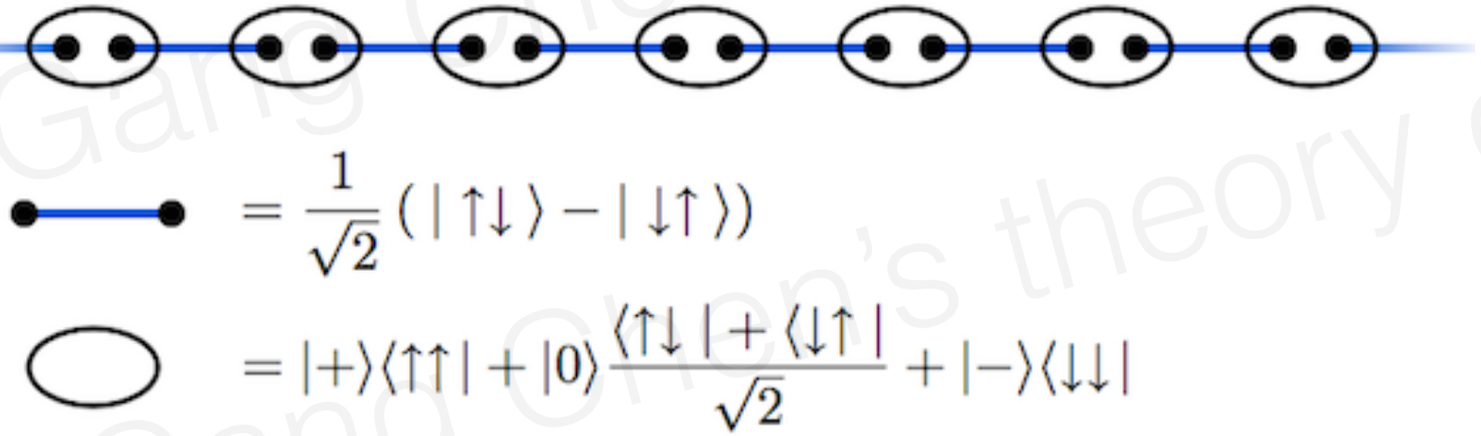
The difference between spin-1/2 and spin-1

Due to Berry phase effect, spin-1/2 chain is gapless, spin-1 Heisenberg chain is gapped.



Duncan Haldane

Nobel Prize 2016



S=1 chain

AKLT state

Building degree of freedom is S=1, but at there is S=1/2 edge state.

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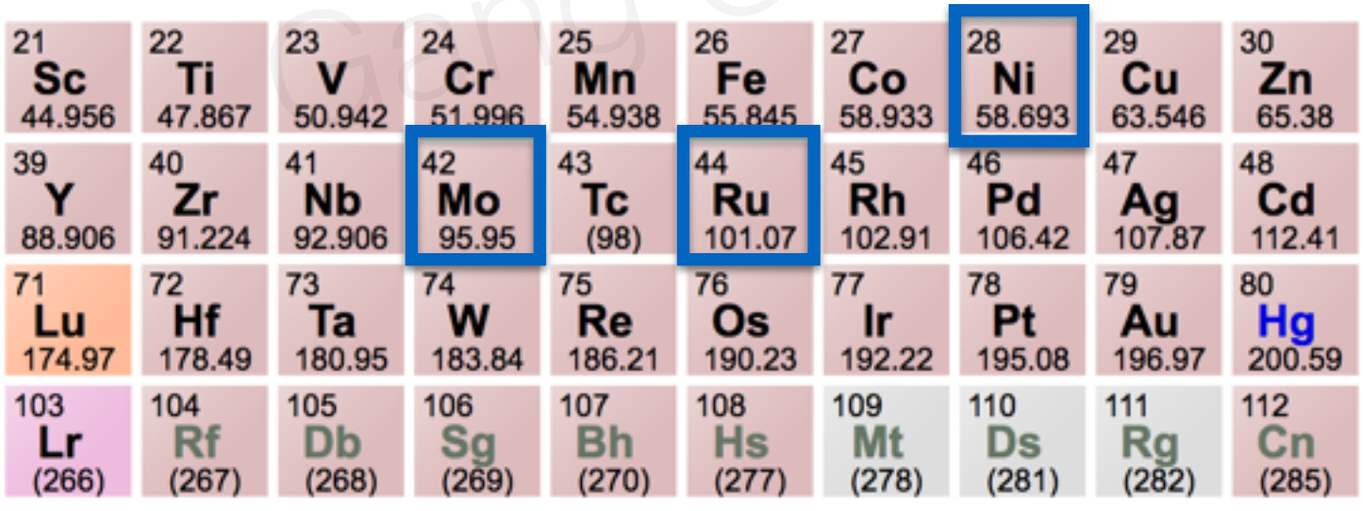
Spin-1 pyrochlores

materials	magnetic ions	Θ_{CW}	magnetic transitions	magnetic structure
$NaCaNi_2F_7$	$Ni^{2+}(3d^8)$	-129K	glassy transition at 3.6K	spin glass
$Y_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-1250K	AFM transition at 76K	canted AFM $\boldsymbol{Q=0}$
$Tl_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-956K	structure transition at 120K	gapped paramagnet
$Eu_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-	Ru order at 118K	Ru order
$Pr_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Pr^{3+}(4f^2)$	-224K	Ru AFM order at 162K	Ru AFM order
$Nd_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Nd^{3+}(4f^3)$	-168K	Ru AFM order at 143K	Ru AFM order
$Gd_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Gd^{3+}(4f^7)$	-10K	Ru AFM order at 114K	Ru AFM order $\boldsymbol{Q=0}$
$Tb_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Tb^{3+}(4f^8)$	-16K	Ru AFM order at 110K	Ru AFM order $\boldsymbol{Q=0}$
$Dy_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Dy^{3+}(4f^9)$	-10K	Ru AFM order at 100K	Ru AFM order
$Ho_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Ho^{3+}(4f^{10})$	-4K	Ru AFM order at 95K	Ru FM order $\boldsymbol{Q=0}$
$Er_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Er^{3+}(4f^{11})$	-16K	Ru AFM order at 92K	Ru AFM order $\boldsymbol{Q=0}$
$Yb_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Yb^{3+}(4f^{13})$	-	Ru AFM order at 83K	Ru AFM order
$Y_2Mo_2O_7$	$Mo^{4+}(4d^2)$	-200K	Mo spin glass at 22K	Mo spin glass
$Lu_2Mo_2O_7$	$Mo^{4+}(4d^2)$	-160K	Mo spin glass at 16K	Mo spin glass
$Tb_2Mo_2O_7$	$Mo^{4+}(4d^2)$, $Tb^{3+}(4f^8)$	20K	spin glass at 25K	spin glass

Candidate spin-1 pyrochlore materials

References are listed in PRB 98, 045109



www.ptable.com

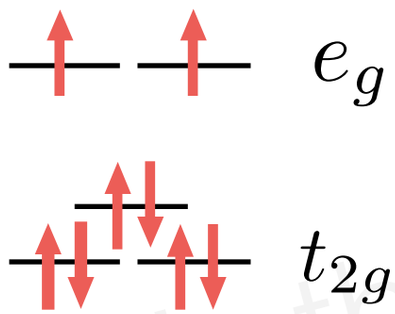
Gang Chen's theory group

Model Hamiltonian

Local moment physics of the Ni^{2+} ion in $\mathrm{NaCaNi_2F_7}$

J. W. Krizan and R. J. Cava
PRB 92, 014406 (2015)

Ni^{2+}: $3d^8$ → CEF



$S = 1$ local moment

No orbital degeneracy

Generic Model Hamiltonian

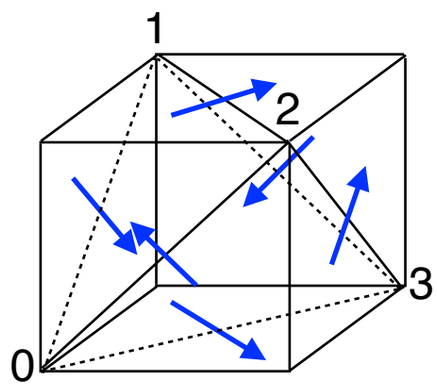
$$H = \sum_{\langle ij \rangle} \left[J \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j) \right] + \sum_i D_z (\boldsymbol{S}_i \cdot \hat{z}_i)^2$$

Heisenberg (J>0) **Dzyaloshinskii-Moriya** **single ion anisotropy**

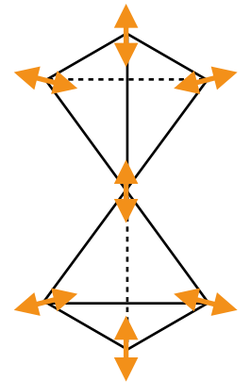
$|D_{ij}|/J \sim \mathcal{O}(\lambda/\Delta)$

$|D_z|/\Delta \sim \mathcal{O}(\lambda^2/\Delta^2)$

λ: SOC Δ: CEF splitting



$\boldsymbol{D}_{ij}$ vectors



local $\hat{z}$ axes

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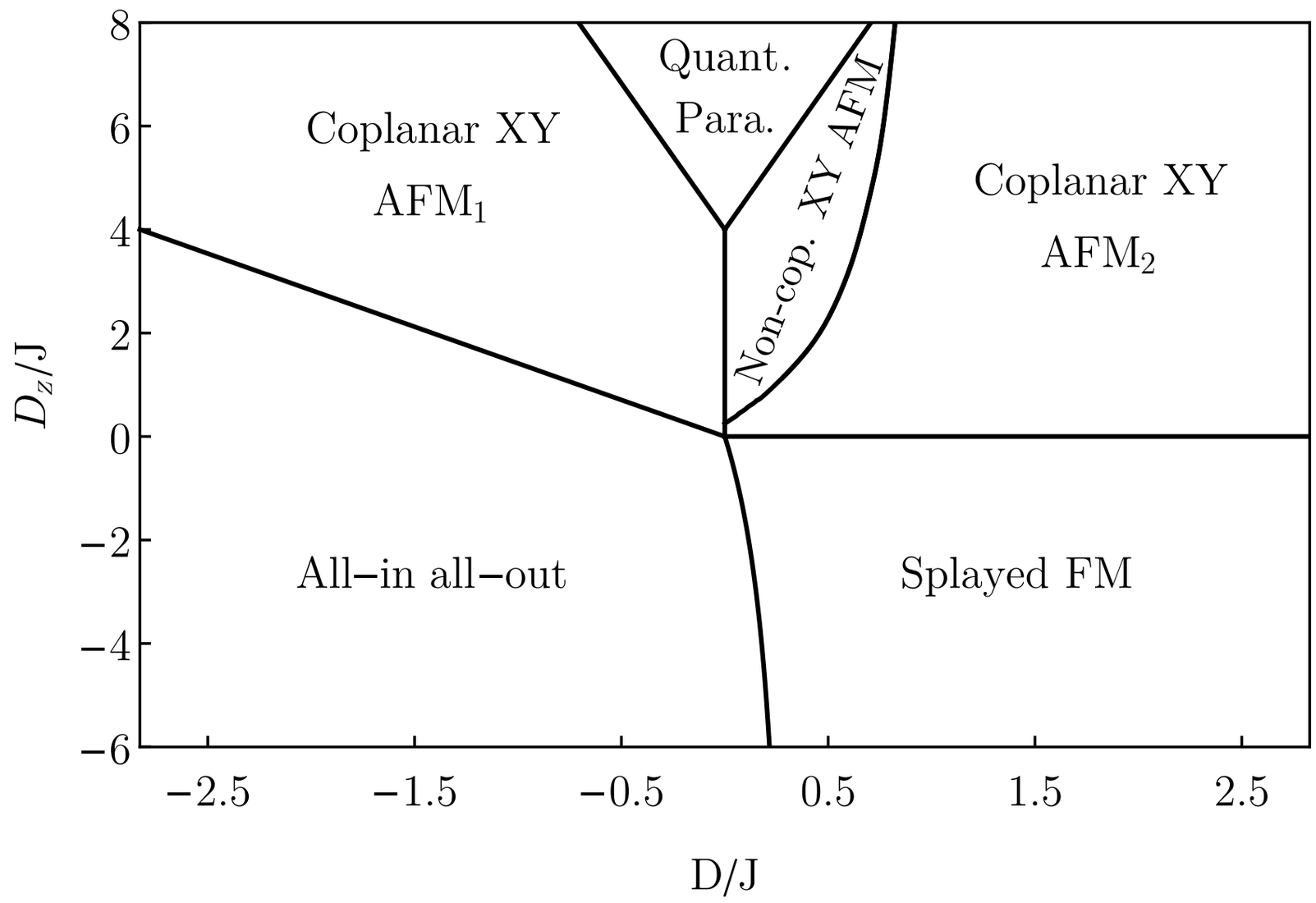
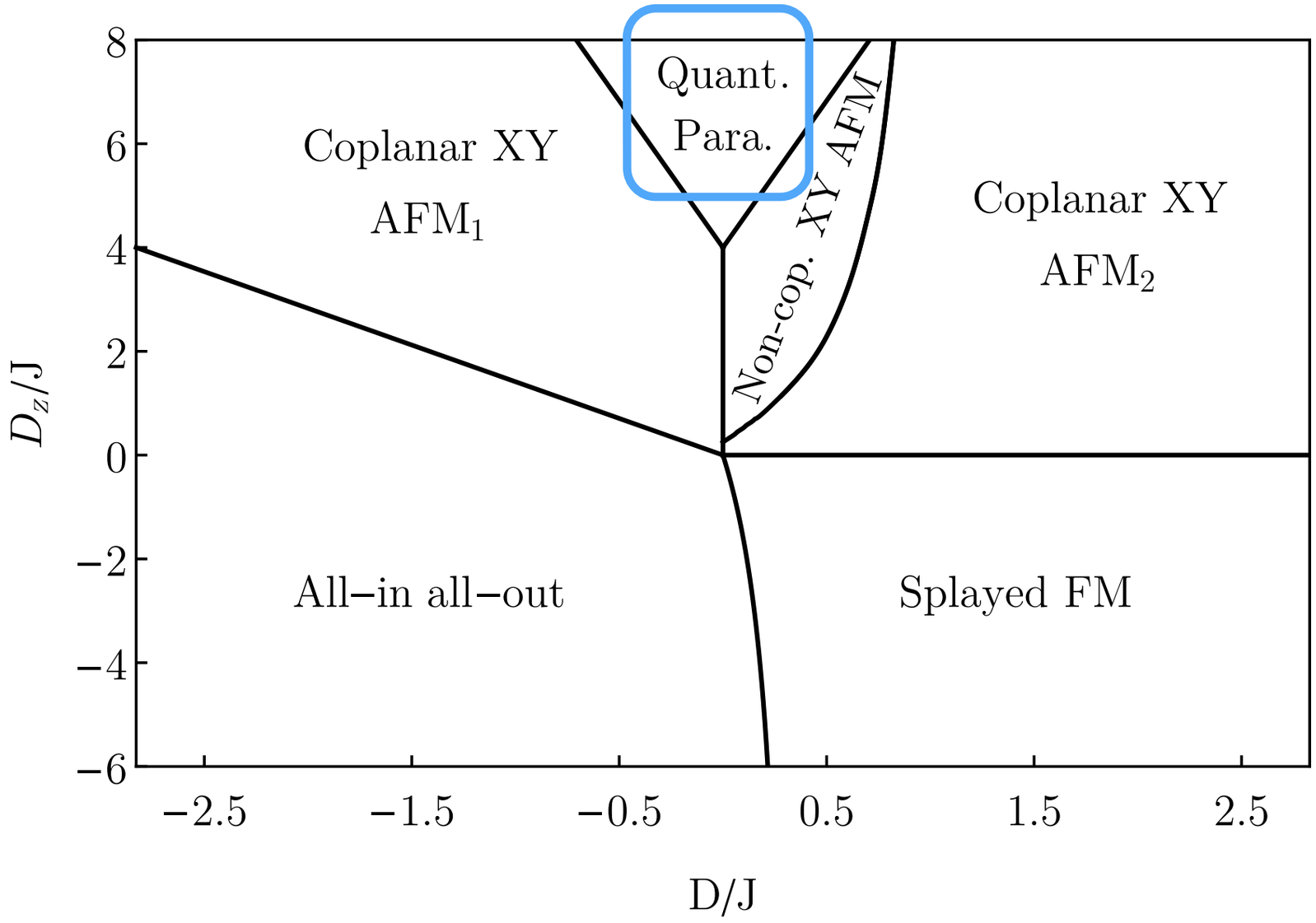
Phase diagram (overview)



Quant. Para. = quantum paramagnetic phase
Others are magnetic ordered phases

Quantum paramagnetic phase



Quant. Para. = quantum paramagnetic phase
Others are magnetic ordered phases

Quantum paramagnetic phase

$D_z \to +\infty$ (easy plane limit): $|\Psi\rangle = \prod_i |S_i^z \equiv \boldsymbol{S}_i \cdot \hat{z}_i = 0\rangle$

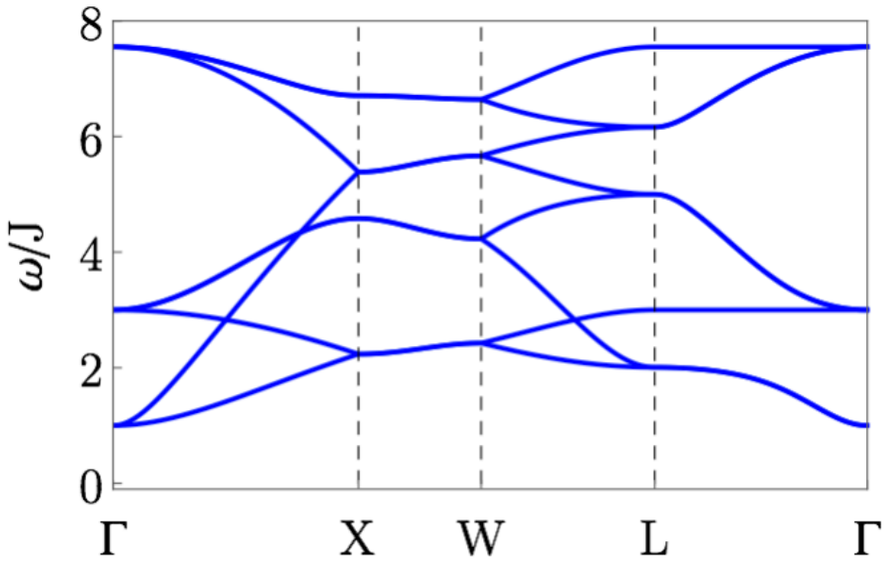
Flavor wave theory

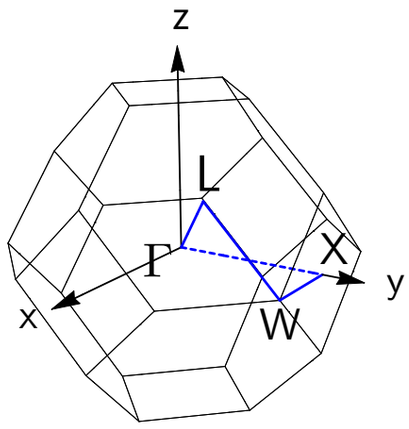
start from the ground state in easy plane limit, one can introduce two flavors of bosons to represent the spin Hamiltonian

$$a_{1(i)}^\dagger |S_i^z = 0\rangle = |S_i^z = 1\rangle$$

$$a_{\bar{1}}^\dagger |S_i^z = 0\rangle = |S_i^z = -1\rangle$$

$$H_{\rm fw} = \sum_{\boldsymbol{k}} \Psi_{\boldsymbol{k}}^\dagger M(\boldsymbol{k}) \Psi_{\boldsymbol{k}}$$





flavor wave excitations

8 branches = 4 sublattices x 2 flavors

A. Joshi, et al. PRB 60, 6584 (1999)

Quantum paramagnetic phase

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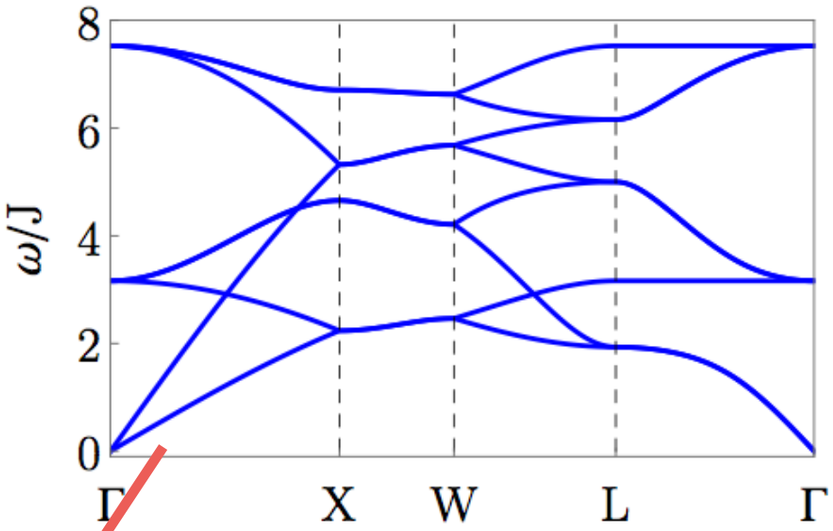
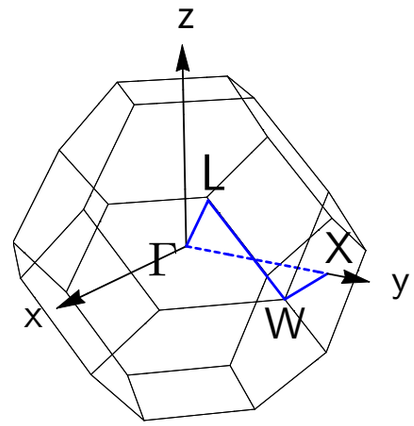




flavor wave excitations

8 branches = 4 sublattices x 2 flavors

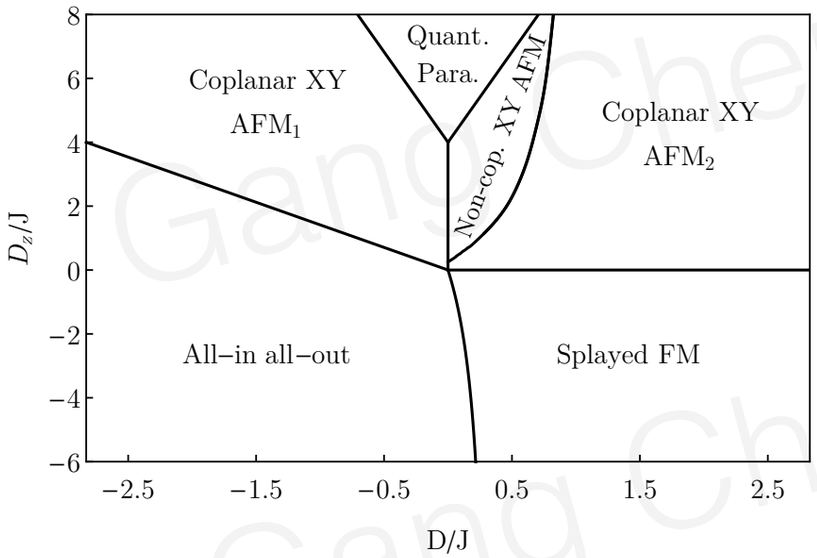
instability of the quantum paramagnet ⇨ magnetic order

A. Joshi, et al. PRB 60, 6584 (1999)

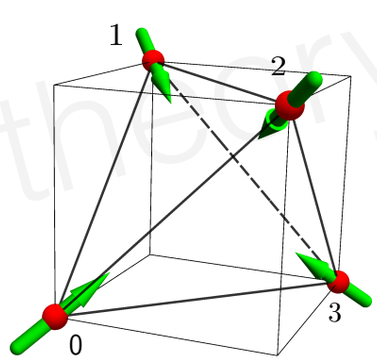
Competing magnetic orders

Mean-field theory

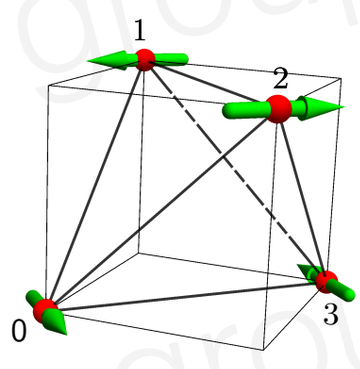
$$\langle H \rangle = \sum_{\langle ij \rangle} J\, \boldsymbol{m}_i \cdot \boldsymbol{m}_j + \boldsymbol{D}_{ij} \cdot (\boldsymbol{m}_i \times \boldsymbol{m}_j) + \sum_i D_z (\boldsymbol{m}_i \cdot \hat{z}_i)^2$$



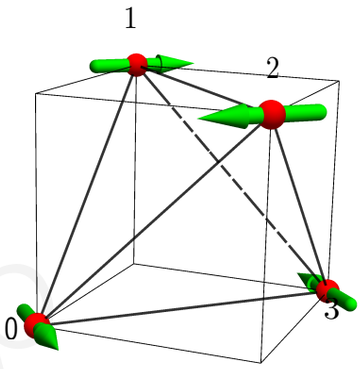
All ordered states have Q=0



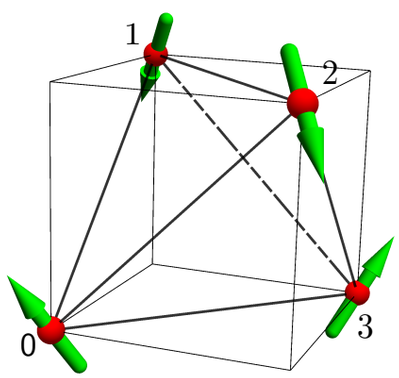
All-in all-out



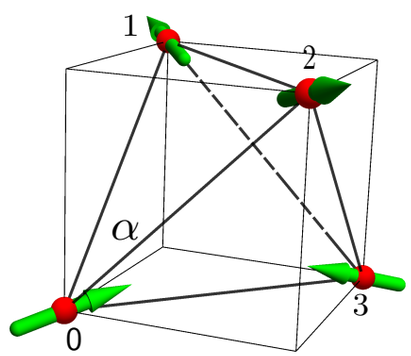
Coplanar
XY AFM1



Coplanar
XY AFM2



Non-coplanar XY AFM



Splayed FM

Gang Chen's theory group

Magnetic ordered phases

The ordered phases can be understood from degeneracy lifting:

J term requires $\sum_{i \in I_{\mathrm{u/d}}} \mathbf{S}_i = 0$ in one tetrahedron (huge degeneracy)

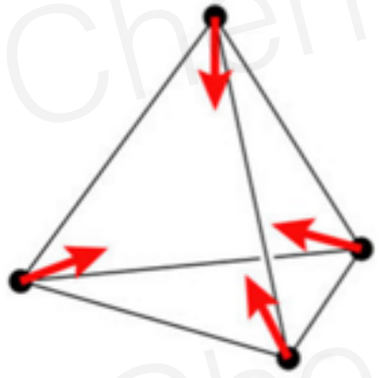
$\Downarrow$

$-|D| \sum_i (\mathbf{S}_i \cdot \hat{z}_i)^2$ $\qquad$ $|D| \sum_i (\mathbf{S}_i \cdot \hat{z}_i)^2$

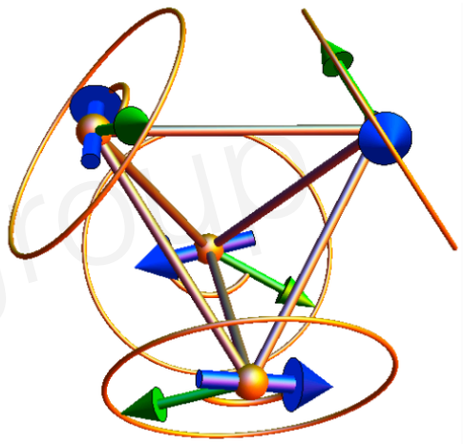
"Z-type" order:
local Z axis

all-in all-out

two-in two-out



"XY-type" order:
local XY plane



$\Downarrow$

Further selected by the sign of DM interaction
or
Quantum order by disorder

C. L. Henley **J. Appl. Phys.** 1987
L. Savary, K. A. Ross, B. D. Gaulin, J. P. C. Ruff and L. Balents **PRL** 2012

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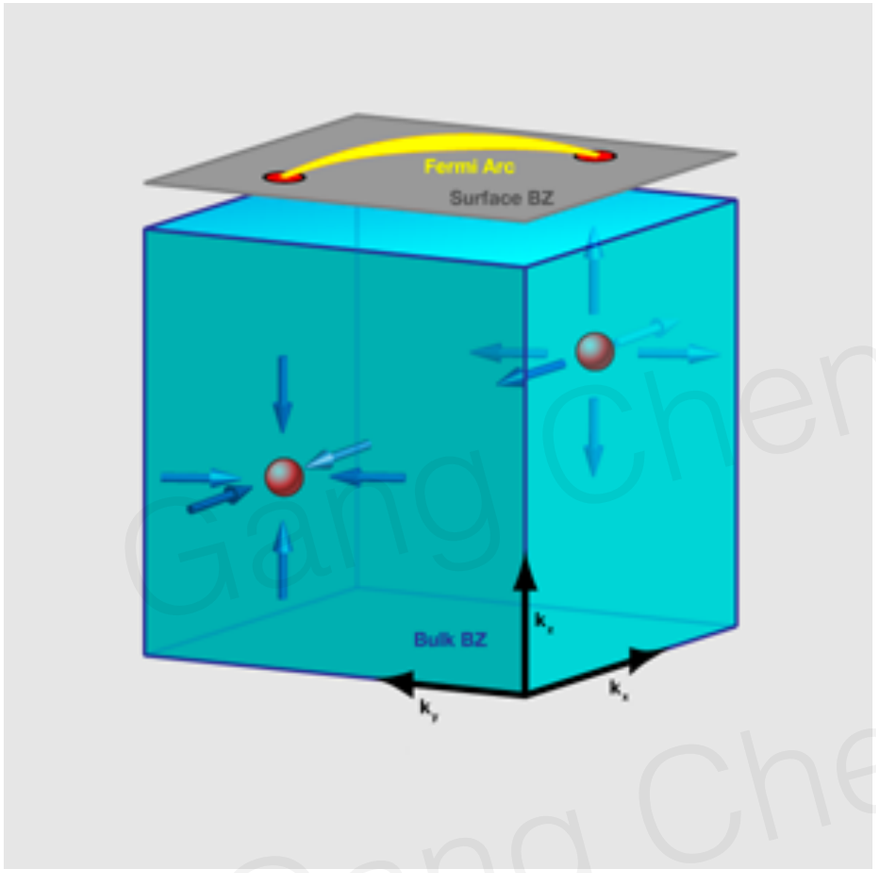
Weyl semimetal and extension



$$H_D = E_0\mathbb{1} + \mathbf{v}_0 \cdot \mathbf{q}\mathbb{1} + \sum_{i=1}^{3} \mathbf{v}_i \cdot \mathbf{q}\sigma_i . \qquad (1)$$

Energy is measured from the chemical potential, $\mathbf{q} = \mathbf{k} - \mathbf{k}_0$

Extension: Type-II Weyl semimetal, Dirac semimetal, nodal line semimetal, hourglass fermion, new fermions

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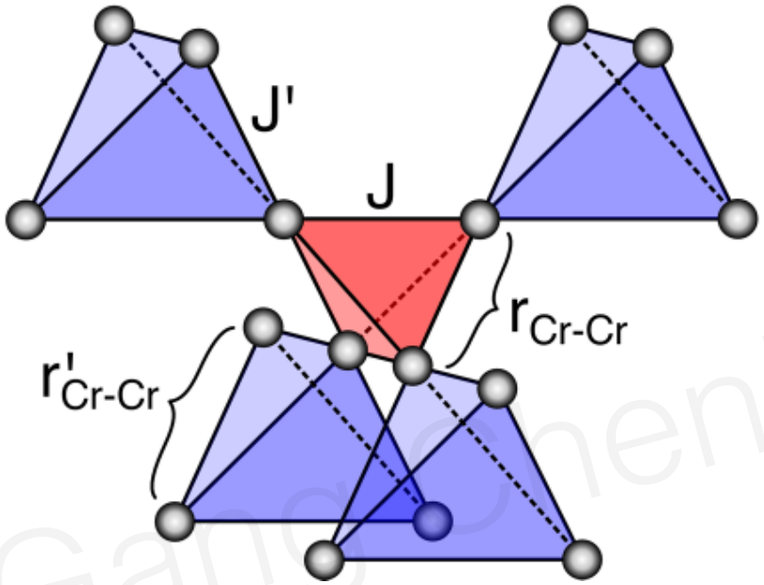
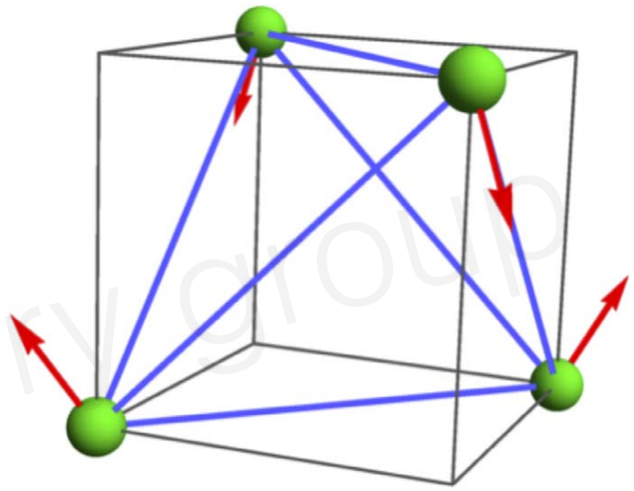
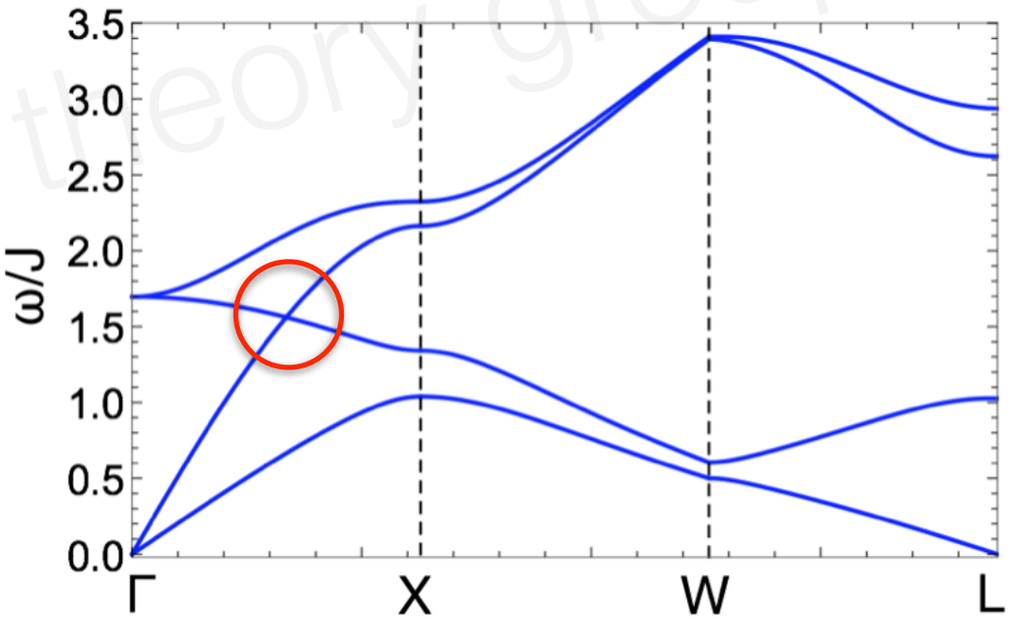
Topological magnons: Weyl magnon







F-Y Li, YD Li, Kim, Balents, Yu, **GC**,
Nature communications 2016

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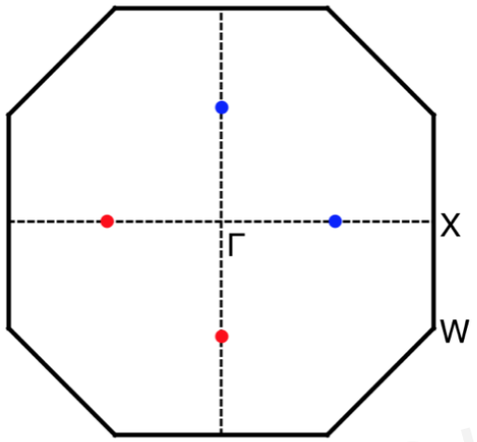
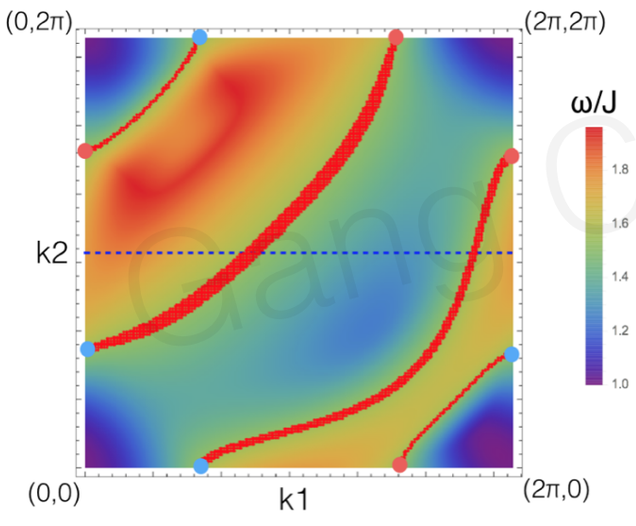
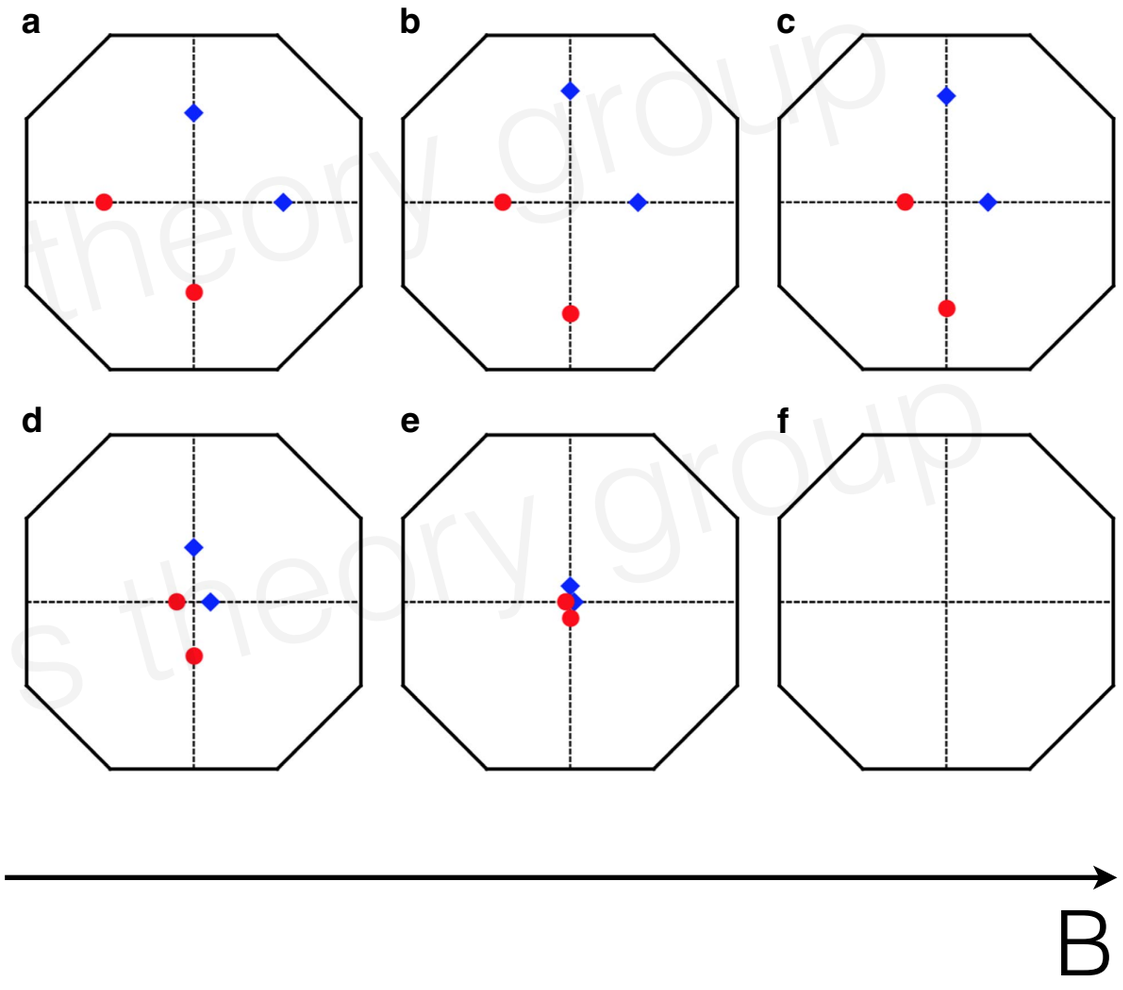
Unique properties of topological Weyl magnon



Magnon Weyl nodes



Magnon surface arcs



tune Weyl nodes with fields

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Focus session: topological magnons

Monday–Friday, March 4–8, 2019; Boston, Massachusetts

Session Index

Session V44: Topological Magnons

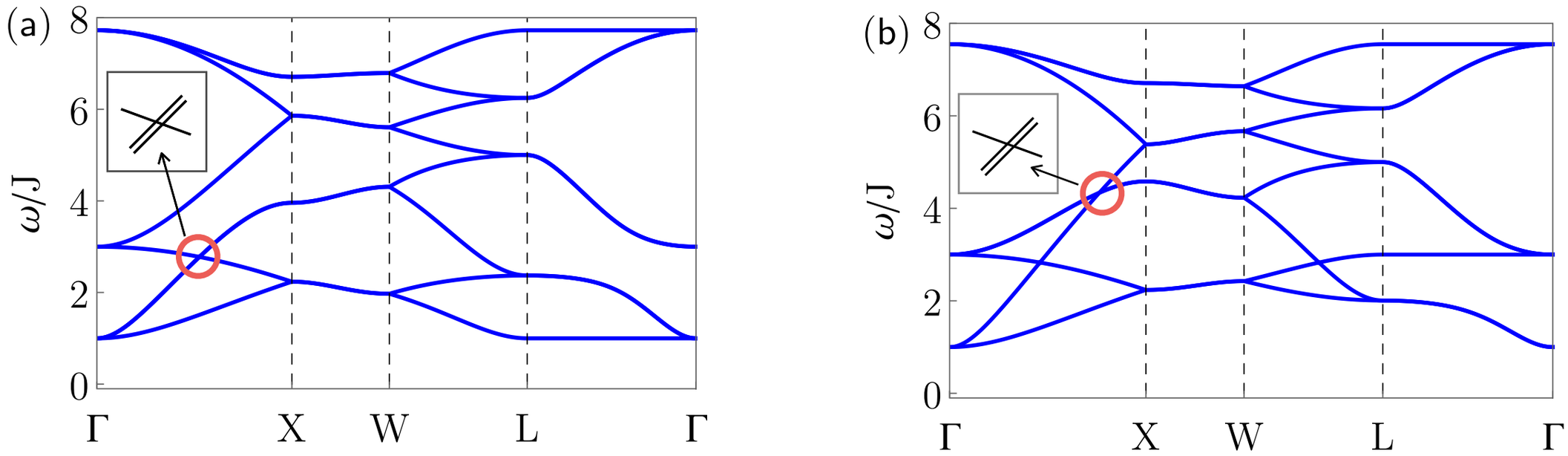
Invited

Room: *BCEC 210C*

Show Abstracts

Thursday, March 7, 2019 2:30PM - 3:06PM	V44.00001: Topological magnon bands in ultra-thin film pyrochlore iridates and iron jarosites Invited Speaker: Gregory Fiete
Thursday, March 7, 2019 3:06PM - 3:42PM	V44.00002: Topological spin excitations in a three-dimensional antiferromagnet Invited Speaker: Yuan Li
Thursday, March 7, 2019 3:42PM - 4:18PM	V44.00003: Topology of magnons: classification and application to honeycomb Kitaev magnets Invited Speaker: Yuan-Ming Lu
Thursday, March 7, 2019 4:18PM - 4:54PM	V44.00004: Discovery of coexisting Dirac and triply degenerate magnons in a three-dimensional antiferromagnet Invited Speaker: Jinsheng Wen
Thursday, March 7, 2019 4:54PM - 5:30PM	V44.00005: The surprising usefulness of magnons at intermediate and high energies: from frustration to topology Invited Speaker: Roderich Moessner

Topological magnons for spin-1 pyrochlores



Triple degeneracy

Topological magnons for spin-1 pyrochlores

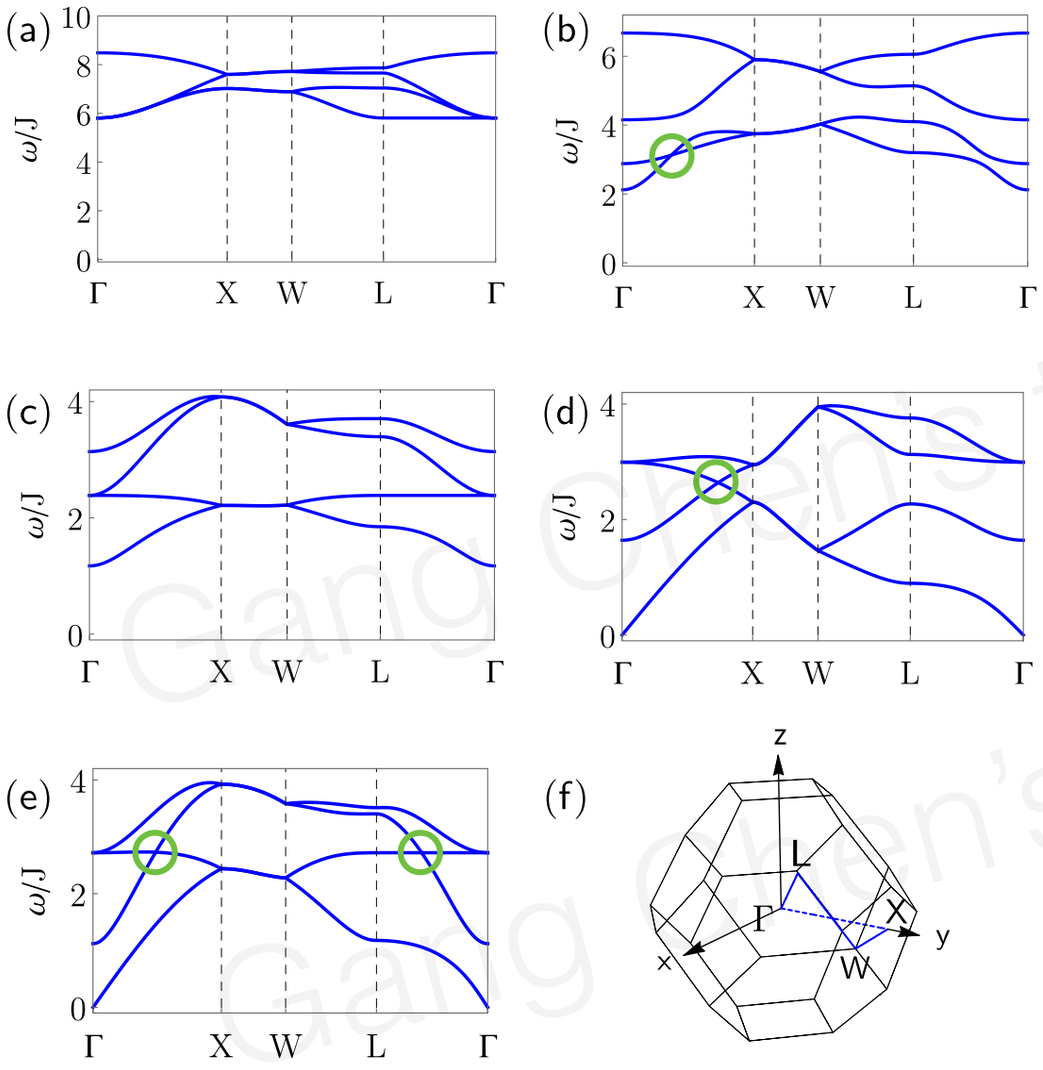


FIG. 7. Spin wave excitations of the ordered phases. The pa-

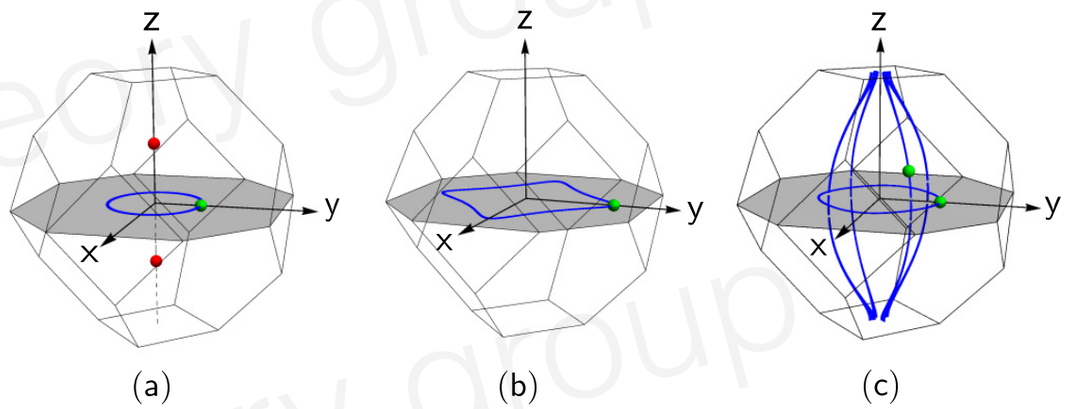


FIG. 8. The nodal lines and Weyl nodes of the spin wave excitation. (a) For the same parameters as in Fig. 7(b), there is a nodal

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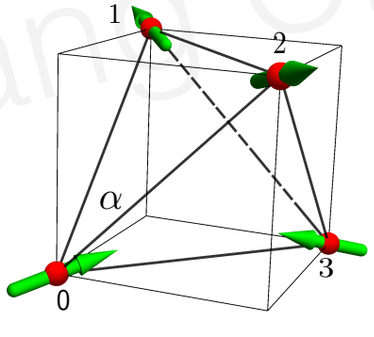
Materials' relevance

materials	magnetic ions	Θ_{CW}	magnetic transitions	magnetic structure
$NaCaNi_2F_7$	$Ni^{2+}(3d^8)$	-129K	glassy transition at 3.6K	spin glass
$Y_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-1250K	AFM transition at 76K	canted AFM $\boldsymbol{Q=0}$
$Tl_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-956K	structure transition at 120K	gapped paramagnet
$Eu_2Ru_2O_7$	$Ru^{4+}(4d^4)$	-	Ru order at 118K	Ru order
$Pr_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Pr^{3+}(4f^2)$	-224K	Ru AFM order at 162K	Ru AFM order
$Nd_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Nd^{3+}(4f^3)$	-168K	Ru AFM order at 143K	Ru AFM order
$Gd_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Gd^{3+}(4f^7)$	-10K	Ru AFM order at 114K	Ru AFM order $\boldsymbol{Q=0}$
$Tb_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Tb^{3+}(4f^8)$	-16K	Ru AFM order at 110K	Ru AFM order $\boldsymbol{Q=0}$
$Dy_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Dy^{3+}(4f^9)$	-10K	Ru AFM order at 100K	Ru AFM order
$Ho_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Ho^{3+}(4f^{10})$	-4K	Ru AFM order at 95K	Ru FM order $\boldsymbol{Q=0}$
$Er_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Er^{3+}(4f^{11})$	-16K	Ru AFM order at 92K	Ru AFM order $\boldsymbol{Q=0}$
$Yb_2Ru_2O_7$	$Ru^{4+}(4d^4)$, $Yb^{3+}(4f^{13})$	-	Ru AFM order at 83K	Ru AFM order
$Y_2Mo_2O_7$	$Mo^{4+}(4d^2)$	-200K	Mo spin glass at 22K	Mo spin glass
$Lu_2Mo_2O_7$	$Mo^{4+}(4d^2)$	-160K	Mo spin glass at 16K	Mo spin glass
$Tb_2Mo_2O_7$	$Mo^{4+}(4d^2)$, $Tb^{3+}(4f^8)$	20K	spin glass at 25K	spin glass



Splayed FM

on the systems with more known results. $Ho_2Ru_2O_7$ was studied using neutron scattering measurements in a nice paper [109] by C.R. Wiebe *et al.* The authors revealed the Ru moment order at ∼95 K and the Ho moment order at ∼1.4 K. The high temperature Ru magnetic order is consistent with the splayed FM with a splayed angle $\alpha \approx 41°$. Under the internal exchange

C. R. Wiebe, J. S. Gardner, S.-J. Kim, G. M. Luke, A. S. Wills, B. D. Gaulin, J. E. Greedan, I. Swainson, Y. Qiu, and C. Y. Jones, Phys. Rev. Lett. **93**, 076403 (2004).

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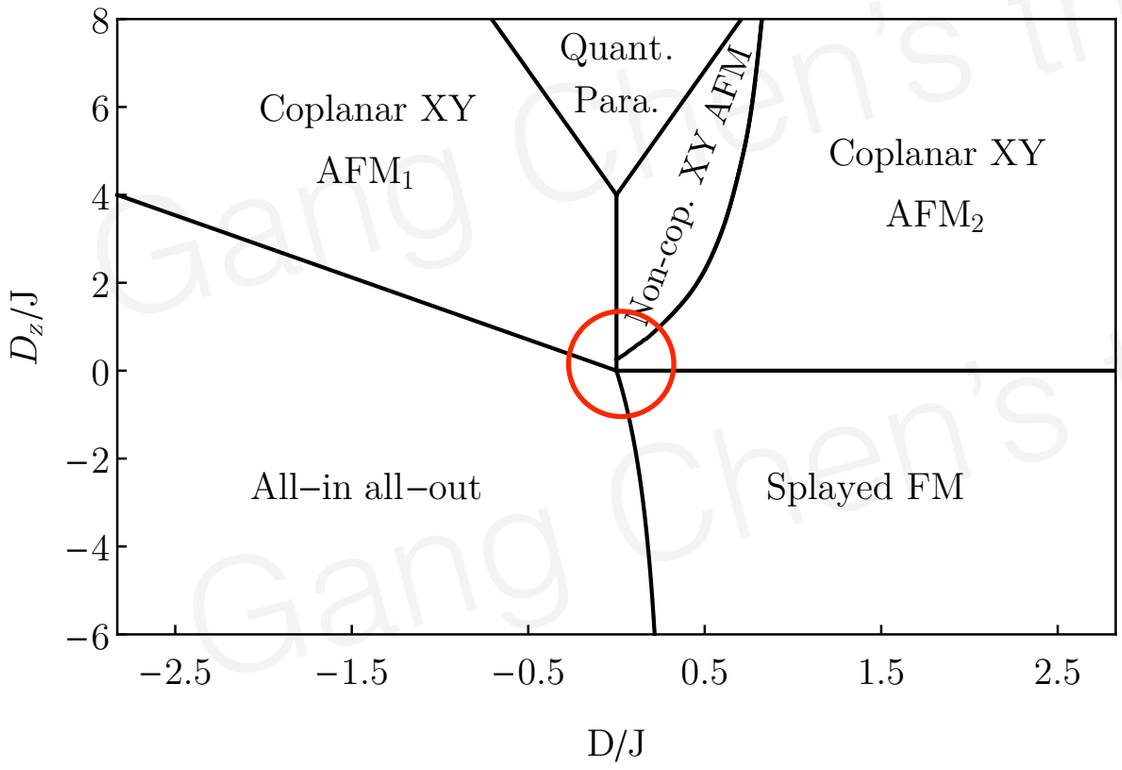
Discussion I

The Heisenberg point requires more “quantum” treatment



Heisenberg point $D = 0, D_z = 0$

⇩

classical ground states are extensively degenerate

⇩

strong quantum fluctuations and fundamental distinctions between spin-1/2 and spin-1

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Discussion II

Relation to spin-3/2 pyrochlores:

- The same model actually applies to the spin-3/2 pyrochlore materials (e.g. Mn-based pyrochlores)

- Local spin anisotropy acts on it quite differently, $D_z(\boldsymbol{S}_i \cdot \hat{z}_i)^2$ The quantum paramagnetic phase is absent since no Sz=0 state.

- The magnetic orders, if they occur, would be similar to the spin-1 pyrochlore system. The magnetic excitations would have similar properties, too.

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Fei-Ye Li
Fudan

Summary

1. We propose a minimal spin model for spin-1 pyrochlores

2. The competing phases and topological excitations are discussed.

3. Various materials' realization and relevance are clarified.

Fei-Ye Li, GC, PRB 98, 045109 (2018)