Emergent quantum criticality from spin-orbital entanglement in d^8 Mott insulators

Gang Chen
Fudan University, Shanghai, China
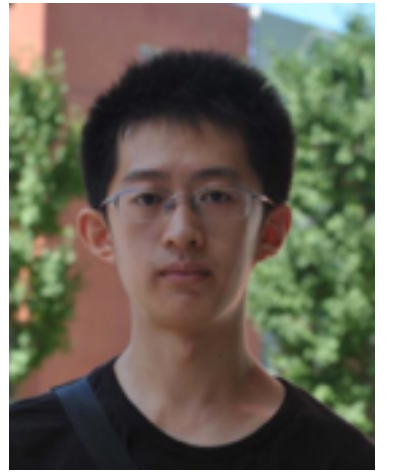
International conference on Highly Frustrated Magnetism (HFM-2020)

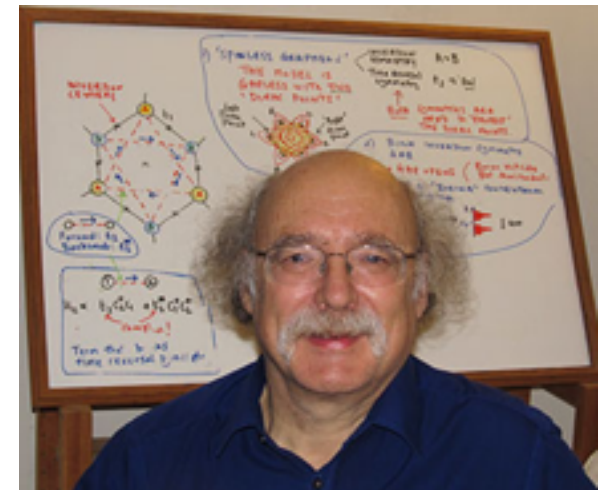## **May 2020**, Shanghai

Pudong, Shanghai

Outline

Fei-Ye Li
（**Fudan**）

1. Introduction to spin-1 quantum magnets

2. A S=1 diamond lattice antiferromagnet

3. First theoretical insight: the role of single-ion anisotropy

4. Updated theoretical insight: spin-orbital entanglement

Gang Chen, PRB 96, 020412 (2017)
Fei-Ye Li, **Gang Chen**, arXiv 1808.06154 (2018)

Spin-one Haldane chain

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

Duncan Haldane

S=1 chain

$$= \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

$$= |+\rangle\langle\uparrow\uparrow| + |0\rangle\frac{\langle\uparrow\downarrow| + \langle\downarrow\uparrow|}{\sqrt{2}} + |-\rangle\langle\downarrow\downarrow|$$

AKLT state

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

Symmetry Protected Topological Phase

Xiao-Gang Wen

Symmetry	$d = 0$	$d = 1$	$d = 2$	$d = 3$
$U(1) \rtimes Z_2^T$	Z	Z_2	Z_2	Z_2^2
Z_2^T	Z_1	Z_2	Z_1	Z_2
$U(1)$	Z	Z_1	Z	Z_1
$SO(3)$	Z_1	Z_2	Z	Z_1
$SO(3) \times Z_2^T$	Z_1	Z_2^2	Z_2	Z_2^3
Z_n	Z_n	Z_1	Z_n	Z_1
$Z_2^T \times D_2 = D_{2h}$	Z_2^2	Z_2^4	Z_2^6	Z_2^9

Table for **boson SPTs**
classified with group cohomology from symmetry and dimension.

It turns out, the well-known topological insulator is a fermion SPT that is protected by time reversal symmetry. Boson SPT must be stabilized by interaction.

Important question: understand the mechanism of SPT, which physical system can realize SPTs.

Senthil's suggestion

PhysRevB, 2015

Topological Paramagnetism in Frustrated Spin-One Mott Insulators

Chong Wang, Adam Nahum, and T. Senthil

Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

(Dated: January 7, 2015)

Time reversal protected three dimensional (3D) topological paramagnets are magnetic analogs of the celebrated 3D topological insulators. Such paramagnets have a bulk gap, no exotic bulk excitations, but non-trivial surface states protected by symmetry. We propose that frustrated spin-1 quantum magnets are a natural setting for realising such states in 3D. We describe a physical picture of the ground state wavefunction for such a spin-1 topological paramagnet in terms of loops of fluctuating Haldane chains with non-trivial linking phases. We illustrate some aspects of such loop gases with simple exactly solvable models. We also show how 3D topological paramagnets can be very naturally accessed within a slave particle description of a spin-1 magnet. Specifically we construct slave particle mean field states which are naturally driven into the topological paramagnet upon including fluctuations. We propose bulk projected wave functions for the topological paramagnet based on this slave particle description. An alternate slave particle construction leads to a stable U(1) quantum spin liquid from which a topological paramagnet may be accessed by condensing the emergent magnetic monopole excitation of the spin liquid.

T. Senthil

The frustrated diamond lattice model appears to describe well [56] the physics of the spinel oxide materials $MnAl_2O_4$ and $CoAl_2O_4$ [58] which belong to a general family of materials of the form AB_2O_4. The A site forms

There is no sharp question in 1D any more. So what is the 3D analogue of Haldane spin-1 phase?

APS March Meeting 2017

Monday–Friday, March 13–17, 2017; New Orleans, Louisiana

Session B48: Frustrated Magnetism: Spinels, Pyrochlores, and Frustrated 3D Magnets I

11:15 AM–2:15 PM, Monday, March 13, 2017
Room: 395

Sponsoring Units: GMAG DMP
Chair: Martin Mourigal, Georgia Tech

Abstract: B48.00006 : S = 1 on a Diamond Lattice in NiRh2O4

12:15 PM–12:27 PM

Preview Abstract

MathJax **On** | *Off*   ← Abstract →

T. McQueen

Authors:
  Juan Chamorro
    (Johns Hopkins University)

  Tyrel McQueen
    (Johns Hopkins University)

An S = 1 system has the potential of rich physics, and has been the subject of intense theoretical work. Extensive work has been done on one-dimensional and two-dimensional S = 1 systems, yet three dimensional systems remain elusive. Experimental realizations of three-dimensional S = 1, however, are limited, and no system to date has been found to genuinely harbor this. Recent theoretical work suggests that S = 1 on a diamond lattice would enable a novel topological paramagnet state, generated by fluctuating Haldane chains within the structure, with topologically protected end states. Here we present data on NiRh2O4, a tetragonal spinel that has a structural phase transition from cubic to tetragonal at T = 380 K. High resolution XRD shows it to have a tetragonally distorted spinel structure, with Ni2+ (d8, S = 1) on the tetrahedral, diamond sublattice site. Magnetic susceptibility and specific heat measurements show that it does not order magnetically down to T = 0.1 K. Nearest neighbor interactions remain the same despite the cubic to tetragonal phase transition. Comparison to theoretical models indicate that this system might fulfill the requirements necessary to have both highly entangled and topological behaviors.
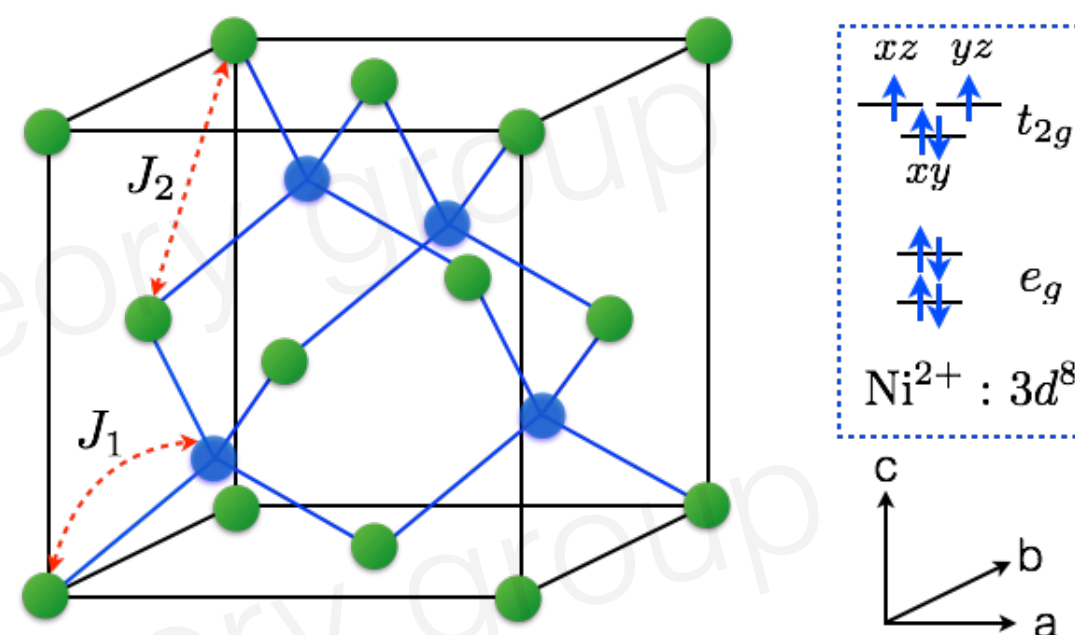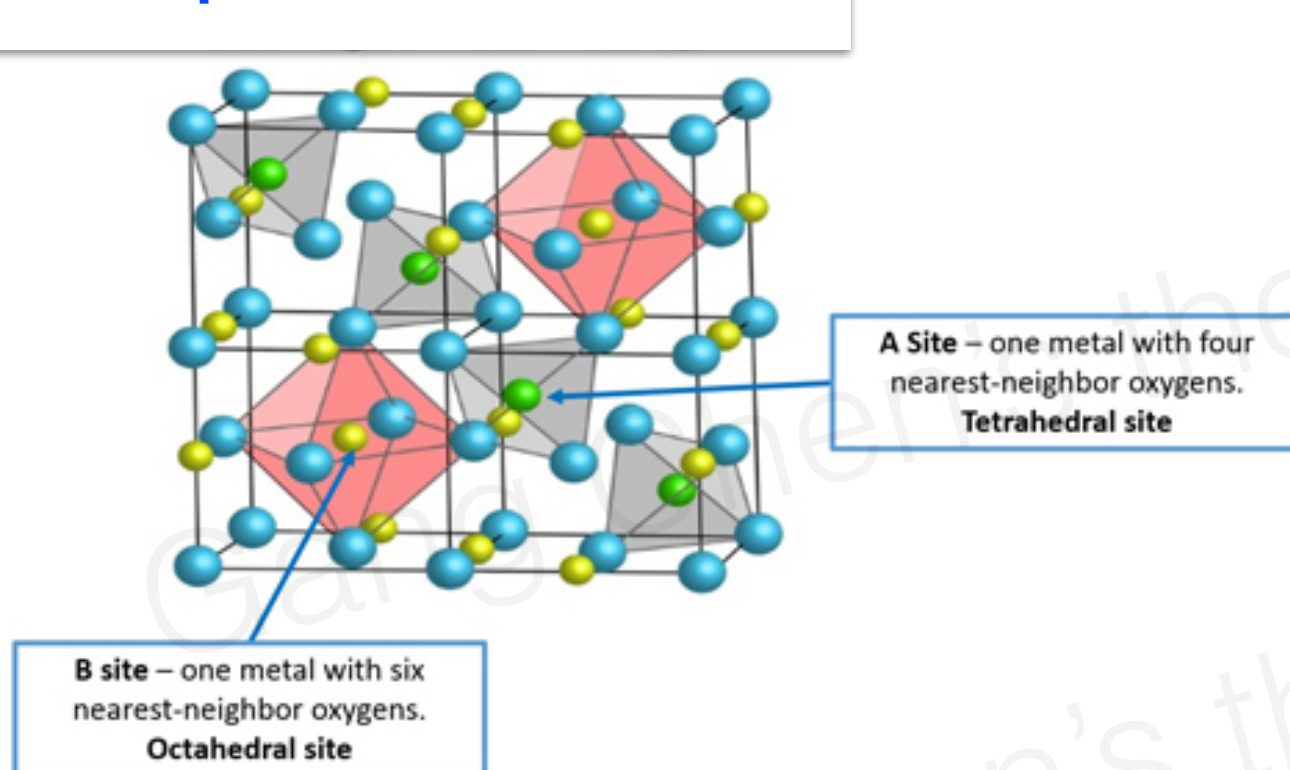
Experimental facts from the abstract
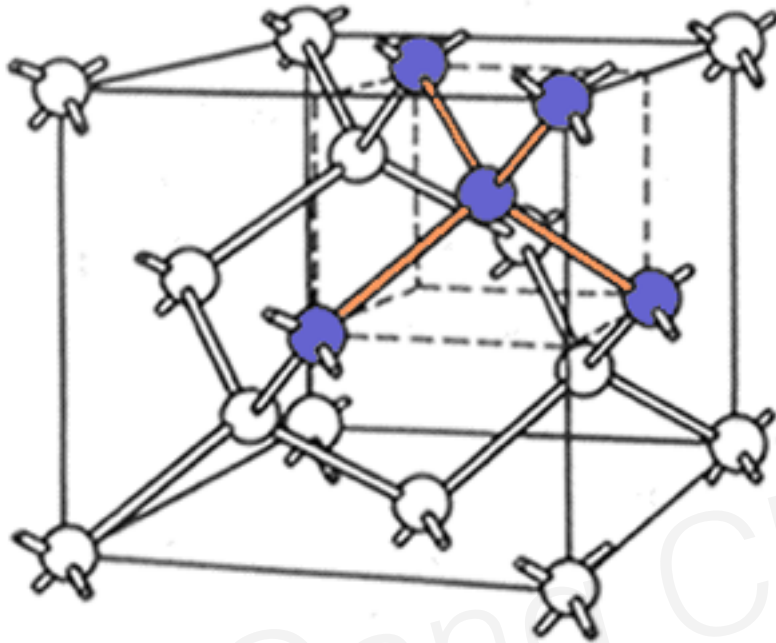
1. A S=1 magnet with diamond structure and Ni ion

2. The system remains disordered down to 0.1K

3. There is a cubic to tetragonal structure transition at 380K.

First theoretical insight:

the role of single-ion anisotropy

Minimal spin model

Spinel AB$_2$O$_4$

A Site – one metal with four
nearest-neighbor oxygens.
Tetrahedral site

B site – one metal with six
nearest-neighbor oxygens.
Octahedral site

J_2

J_1

xz yz

t_{2g}

xy

e_g

Ni^{2+} : $3d^8$

c

b

a

$$H = J_1 \sum_{\langle \boldsymbol{rr'} \rangle} \boldsymbol{S_r} \cdot \boldsymbol{S_{r'}} + J_2 \sum_{\langle\langle \boldsymbol{rr'} \rangle\rangle} \boldsymbol{S_r} \cdot \boldsymbol{S_{r'}} + D_z \sum_{\boldsymbol{r}} (S_{\boldsymbol{r}}^z)^2,$$

Immediate experimental
consequence

$$\Theta_{\mathrm{CW}}^z = -\frac{D_z}{3} - \frac{S(S+1)}{3}(z_1 J_1 + z_2 J_2),$$

$$\Theta_{\mathrm{CW}}^\perp = +\frac{D_z}{6} - \frac{S(S+1)}{3}(z_1 J_1 + z_2 J_2),$$

FUDAN UNIVERSITY
1905

Order-by-disorder and spiral spin-liquid in frustrated diamond-lattice antiferromagnets

DORON BERGMAN[1]*, JASON ALICEA[1], EMANUEL GULL[2], SIMON TREBST[3] AND LEON BALENTS[1]

[1] Department of Physics, University of California, Santa Barbara, California 93106-9530, USA
[2] Theoretische Physik, Eidgenössische Technische Hochschule Zürich, CH-8093 Zürich, Switzerland
[3] Microsoft Research, Station Q, University of California, Santa Barbara, California 93106, USA
*e-mail: doronber@physics.ucsb.edu
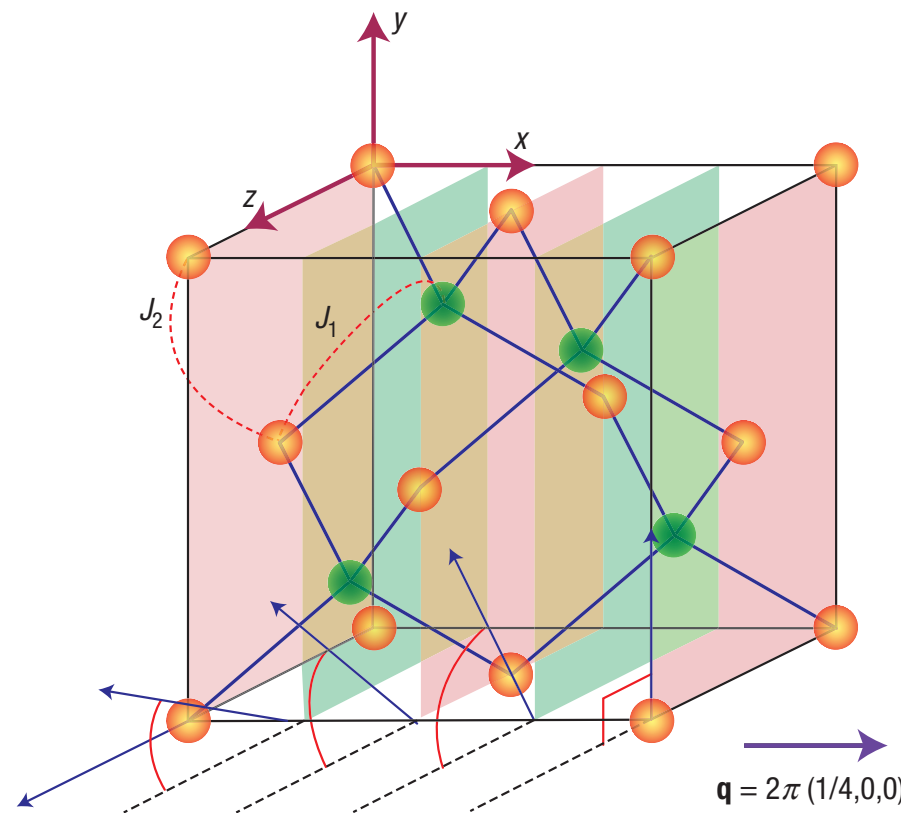
Sungbin Lee, Balents,
PRB 78, 144417 (2008)
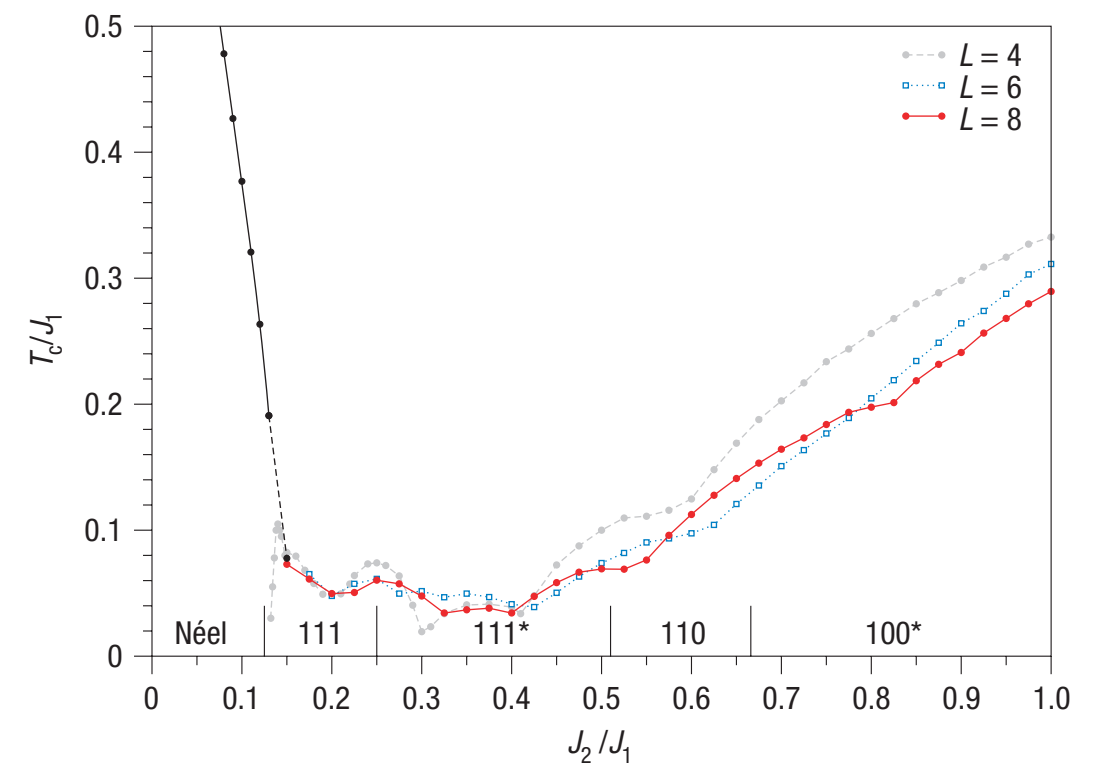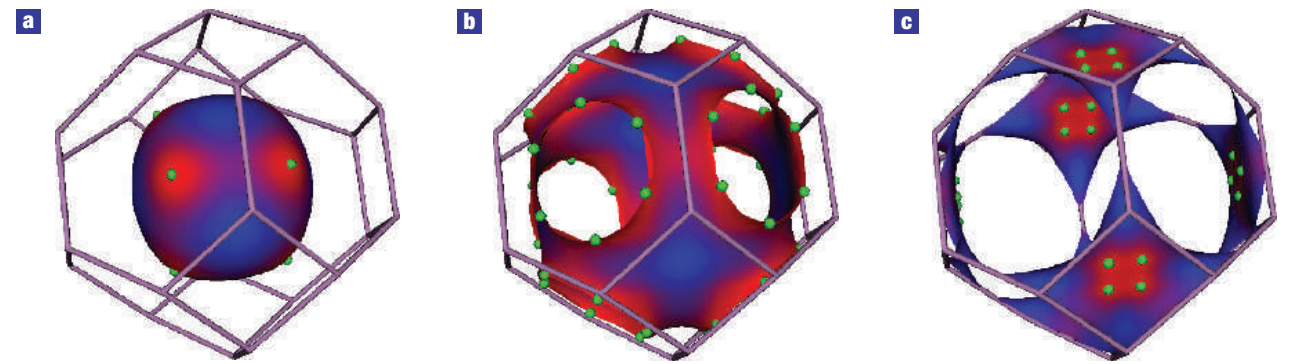
Figure 3 Phase diagram. Numerical results for the ordering temperature, T_c, versus the coupling strength, J_2/J_1, for systems with up to $N = 8 \times L^3 = 4,096$

The (quantum) phase diagram
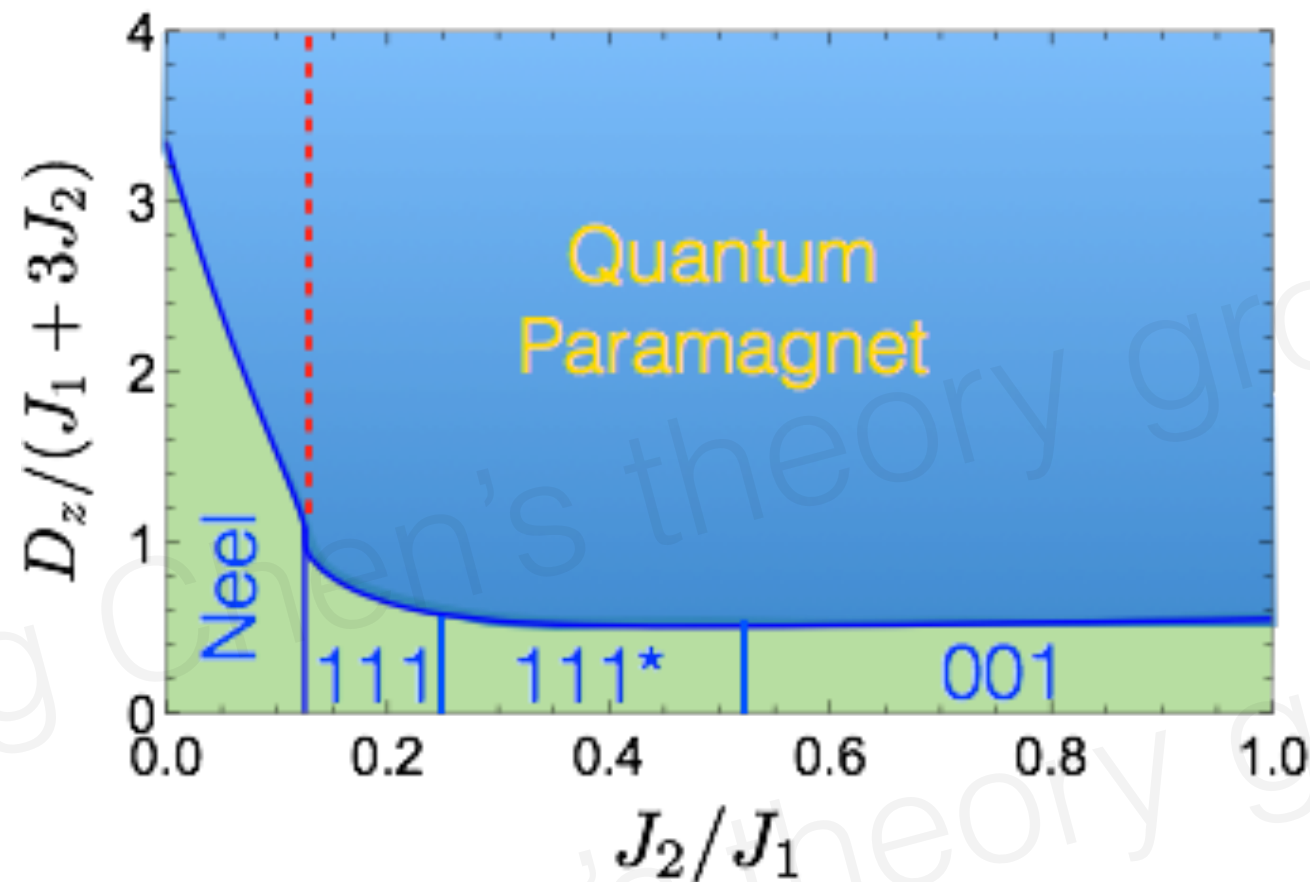
Deep in quantum paramagnet, the ground state is a trivial product state.
The state is trivial, but excitation and phase transition out of it can be non-trivial.

$$|\Psi\rangle = \prod_{\boldsymbol{r}} |S^z_{\boldsymbol{r}} = 0\rangle$$

Rotor treatment

$$S_r^z = n_r, \quad S_r^\pm = \sqrt{2}e^{\pm i\phi_r}, \text{ for } \phi_r \text{ and } n_r \text{ with } [\phi_r, n_{r'}] = i\delta_{rr'}.$$

With the rotor variables, the J_1-J_2-D_z spin model takes the form

$$H = \sum_{\langle rr' \rangle} J_1 [2\cos(\phi_r - \phi_{r'}) + n_r n_{r'}]$$

$$+ \sum_{\langle\langle rr' \rangle\rangle} J_2 [2\cos(\phi_r - \phi_{r'}) + n_r n_{r'}]$$

$$+ \sum_r D_z n_r^2. \tag{6}$$

paramagnet but take finite values in the ordered states. We here carry out a coherent state path integral and integrate out the number operator n_r. The resulting partition function is

$$Z = \int \mathcal{D}\Phi_r \mathcal{D}\lambda_r \exp\left[-\mathcal{S} - i\sum_r \lambda_r (|\Phi_r|^2 - 1) \right], \tag{7}$$

where the effective action for the rotor variable is

$$\mathcal{S} = \int d\tau \sum_{k \in \text{BZ}} (2D_z \mathbb{1}_{2\times2} + \mathcal{J}_k)_{ij}^{-1} \partial_\tau \Phi_{i,k}^\dagger \partial_\tau \Phi_{j,k}$$

$$+ \sum_{\langle rr' \rangle} J_1 \Phi_r^\dagger \Phi_{r'} + \sum_{\langle\langle rr' \rangle\rangle} J_2 \Phi_r^\dagger \Phi_{r'}, \tag{8}$$

Unconventional magnetic excitation

$$\sum_{i=1,2} \sum_{\boldsymbol{k} \in \text{BZ}} \frac{2D_z + \xi_{i,\boldsymbol{k}}}{\omega_{i,\boldsymbol{k}}} \coth\left(\frac{\beta\omega_{i,\boldsymbol{k}}}{2}\right) = 2, \qquad (9)$$

where $\omega_{1,\boldsymbol{k}}$ and $\omega_{2,\boldsymbol{k}}$ are the two modes of the magnetic excitations in the paramagnetic phase and are given by

$$\omega_{i,\boldsymbol{k}} = [(4D_z + 2\xi_{i,\boldsymbol{k}})(\Delta(T) + \xi_{i,\boldsymbol{k}})]^{\frac{1}{2}}, \qquad (10)$$

$J_2 = 0.05J_1$, $D_z = 3J_1$

$J_2 = 0.18J_1$, $D_z = 1.5J_1$

$J_2 = 0.4J_1$, $D_z = 1.5J_1$

$J_2 = 0.8J_1$, $D_z = 2J_1$

Unconventional magnetic excitation

J2=0.18J1   J2=J1/3
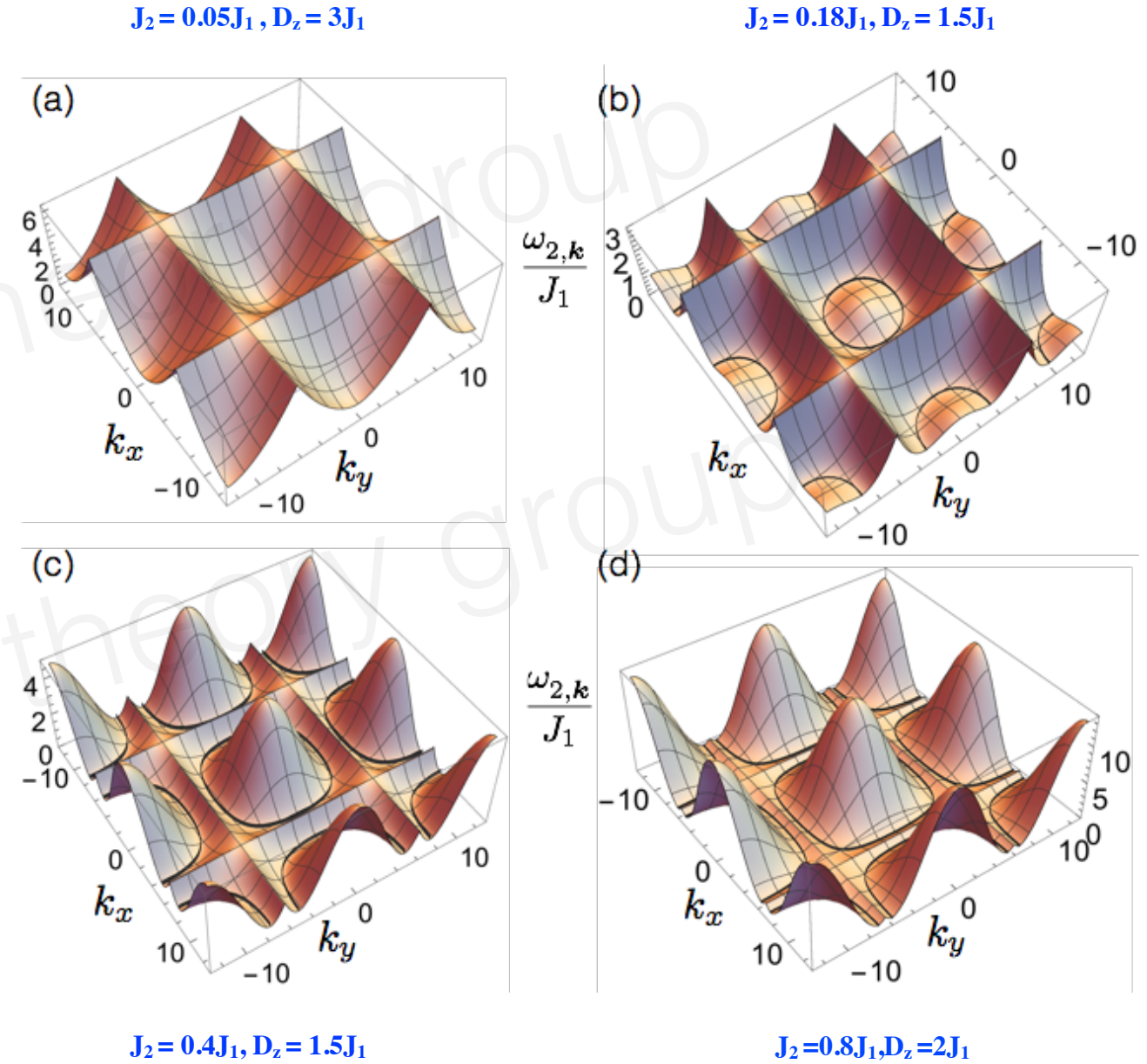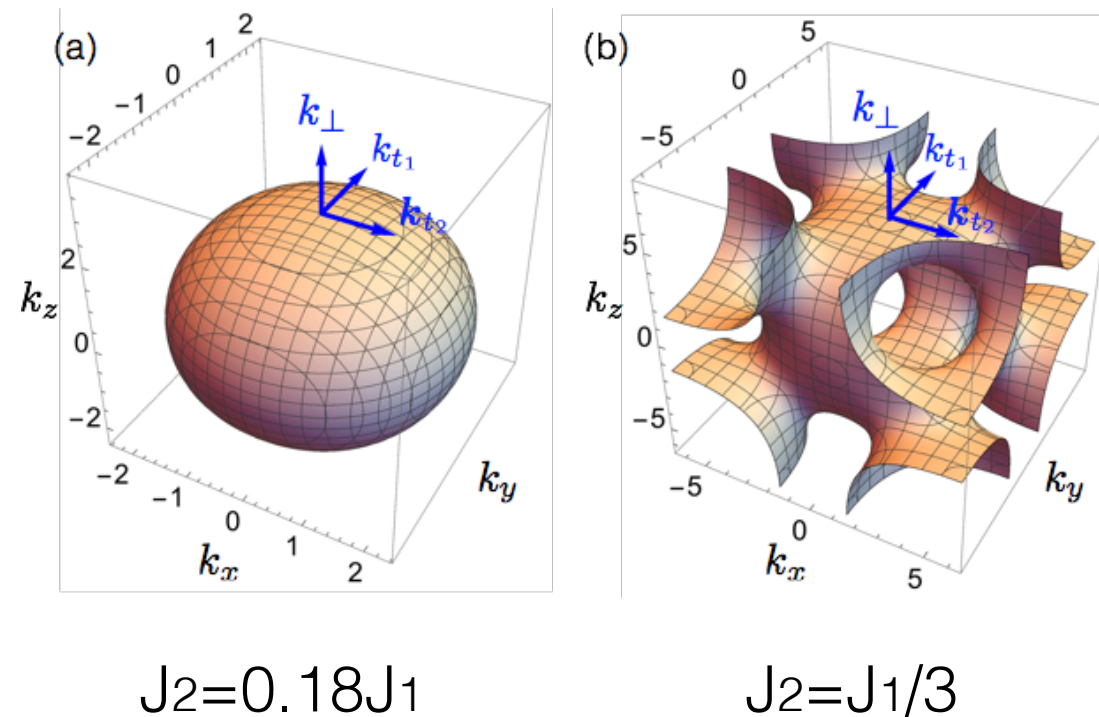
These are bosonic excitations. What is relevant for bosons is the lowest energy mode. Usually, the lowest energy modes occur at certain discrete momenta. But here, the lowest energy modes occur at a surface in the reciprocal space.

Frustrated Quantum Criticality: collapse of boson surface

(a)

(b)

(a)

J2=0.18J1

J2=J1/3
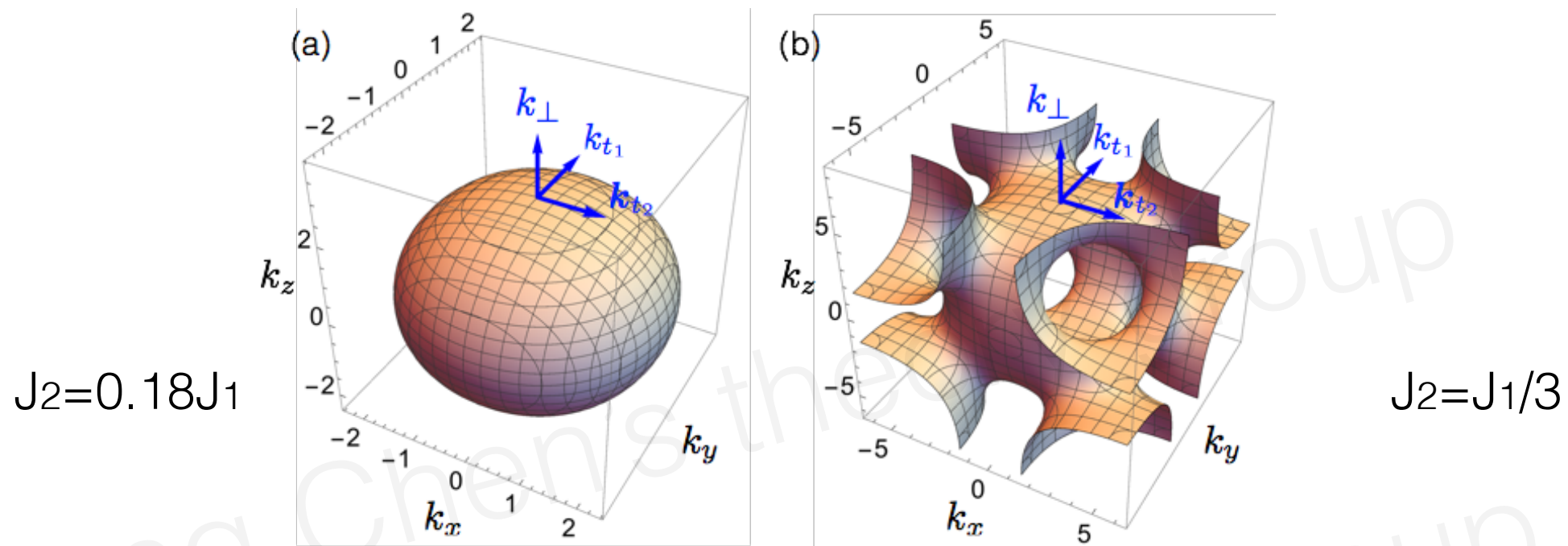
$$\cos\frac{k_x}{2}\cos\frac{k_y}{2} + \cos\frac{k_x}{2}\cos\frac{k_z}{2} + \cos\frac{k_y}{2}\cos\frac{k_z}{2} = \frac{J_1^2}{16J_2^2} - 1$$

These degenerate surfaces are NOT Fermi surface !

But at low temperature, the fluctuation of the system is governed by the surface, i.e. low-energy fluctuations are near the 2D surface.
We obtain a linear-T heat capacity **Cv ~ T,** which is like a Fermi surface.

Degeneracy breaking in the ordered side
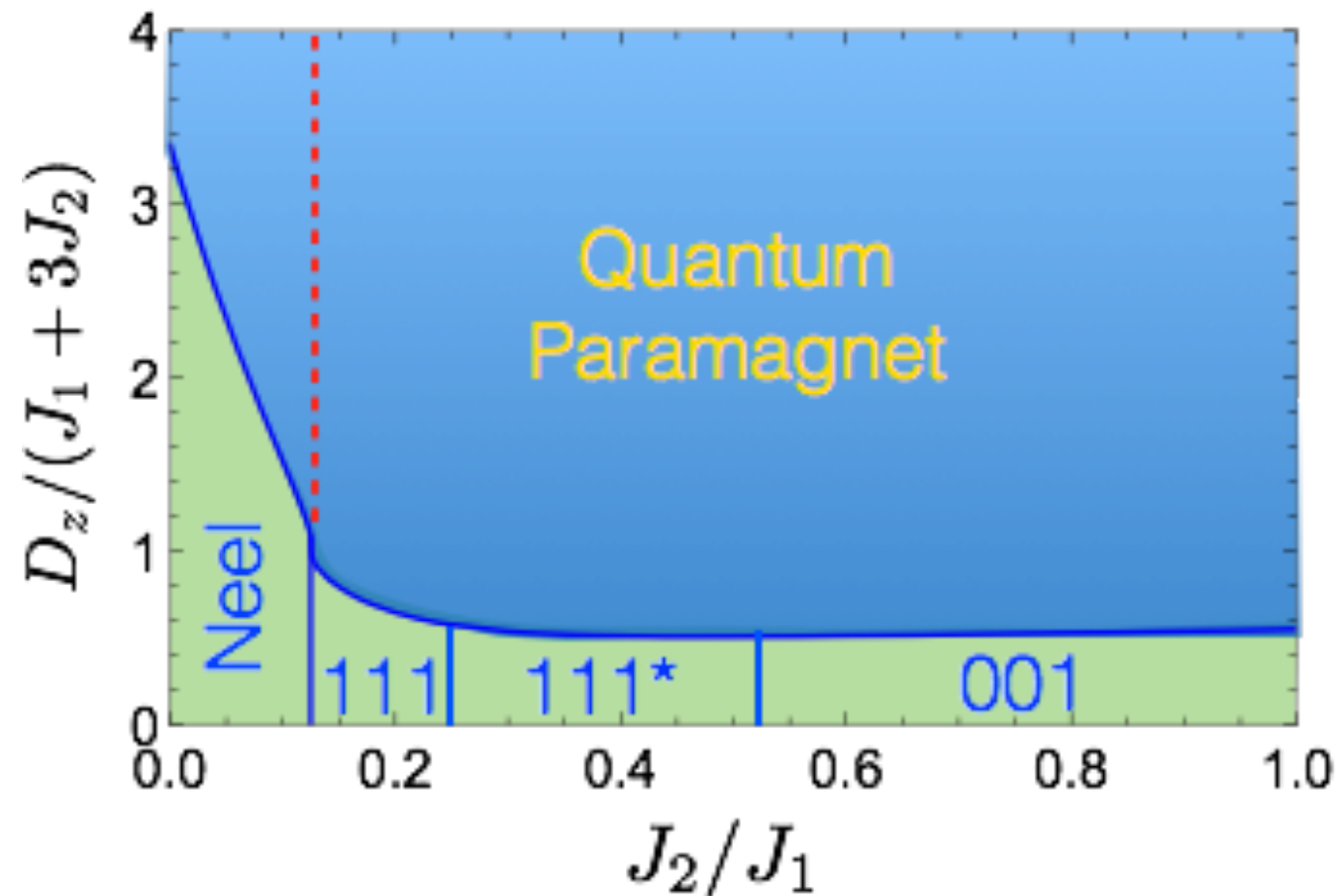
Here, because infinite number of boson modes are condensed, the system does not know which order to select.

So quantum fluctuation will pick up the order that gives the lowest quantum zero-point energy.
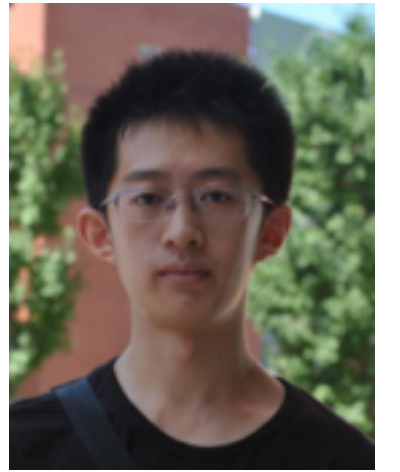
Fei-Ye Li
（**Fudan**）

Updated theoretical insight:
spin-orbital entanglement

Fei-Ye Li, **Gang Chen**, arXiv 1808.06154 (2018)

Magnetic entropy
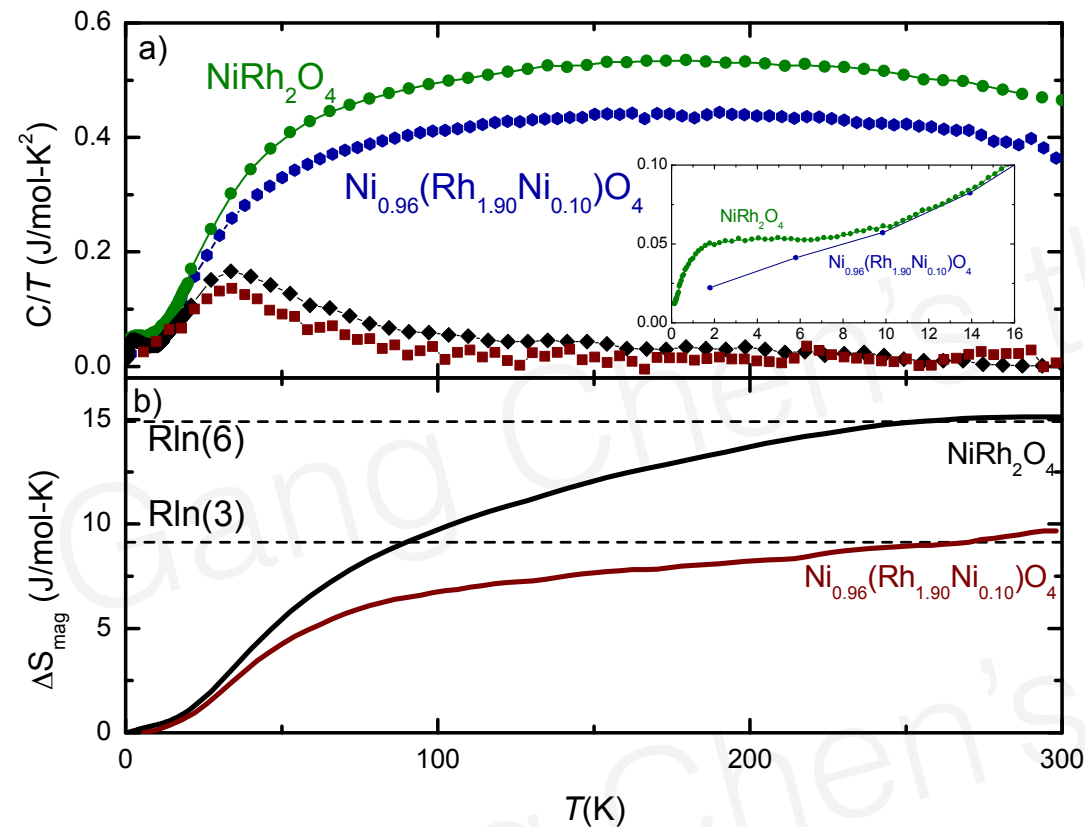
Mcqueen, etc,
PR material 2018

$Ni^{2+} : 3d^8$
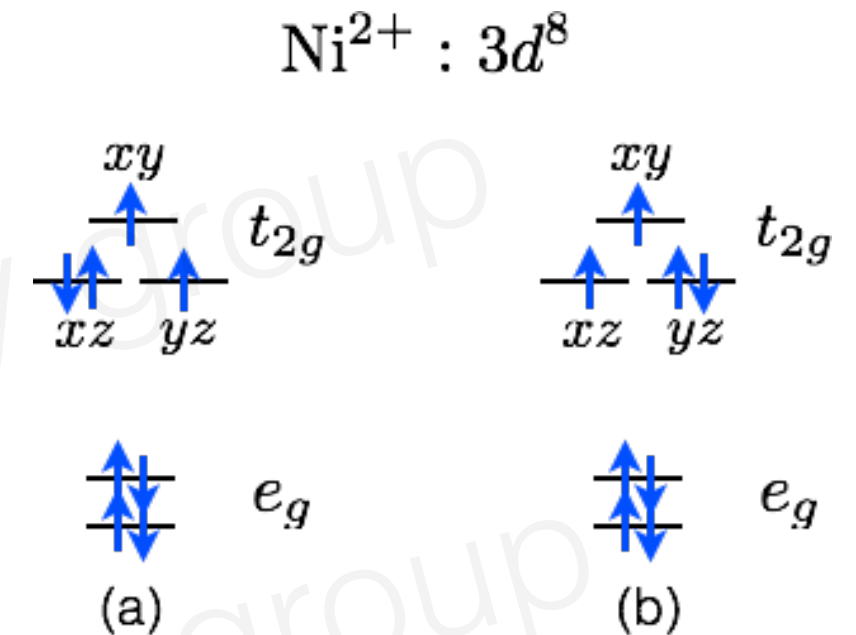
FIG. 5. (Color online.) The energy level diagram when the xz and yz orbitals are lower than the xy orbital. (a) and (b) are two equivalent electron filling schemes and can be represented by a pseudospin-1/2 orbital degree of freedom. See the text for the detailed discussion.

Gang Chen, PRB 2017

SOC is active here
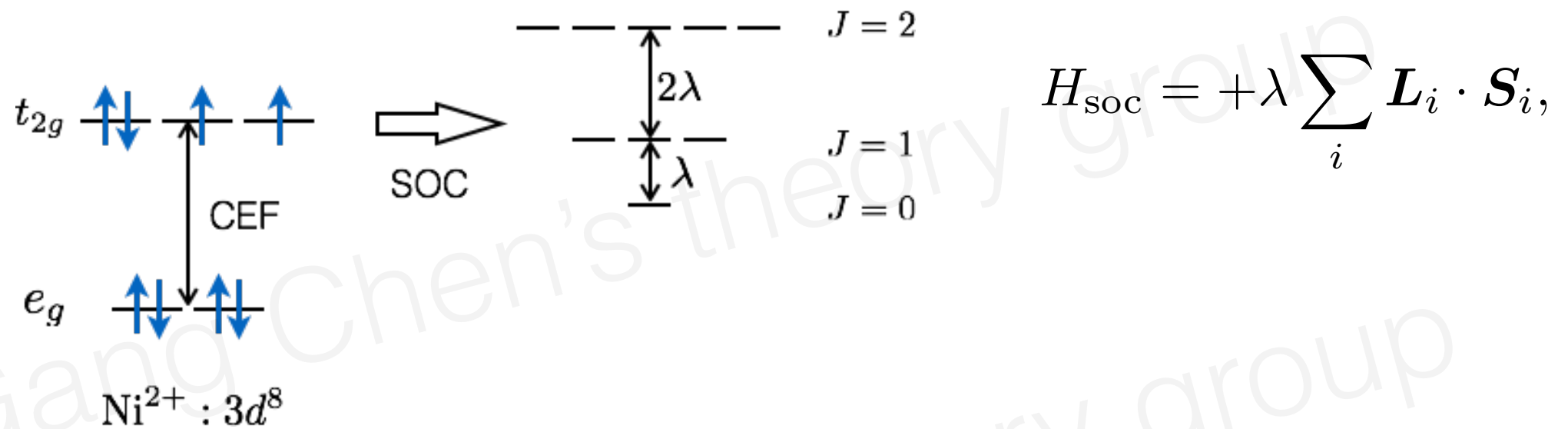
FIG. 1. (Color online.) The electron configuration of the Ni^{2+} ion in the tetrahedral crystal field environment. When the atomic spin-orbit coupling (SOC) is introduced, the electron states in the upper t_{2g} levels are further split into the spin-orbital entangled J states. In the figure, "CEF" refers to the crystal electric field splitting.

A quite useful correspondence !

TABLE I. The correspondence between different electron configurations in the tetrahedral and octahedral environments.

Ir, Os, Mo, …
4d/5d materials

William, **GC**, YB Kim, Balents,
Annual Review of Condensed Matter Physics

Minimal model and phase diagram
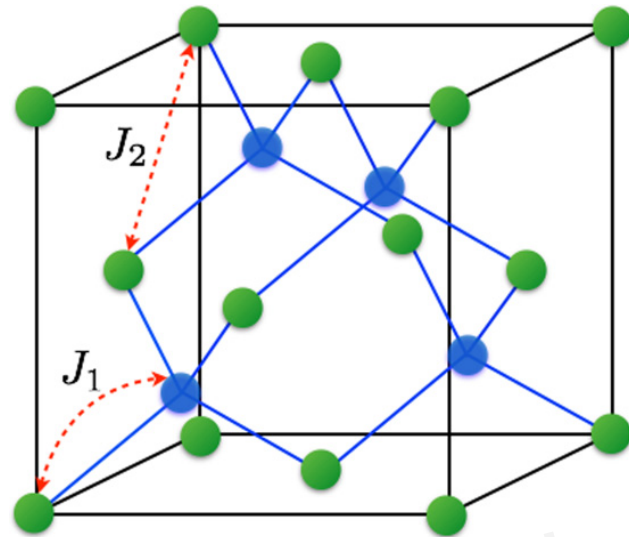
$$H_{\mathrm{ex}} = \sum_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

$$= \sum_{\langle ij \rangle} J_1 \, \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \sum_{\langle\langle ij \rangle\rangle} J_2 \, \boldsymbol{S}_i \cdot \boldsymbol{S}_j,$$
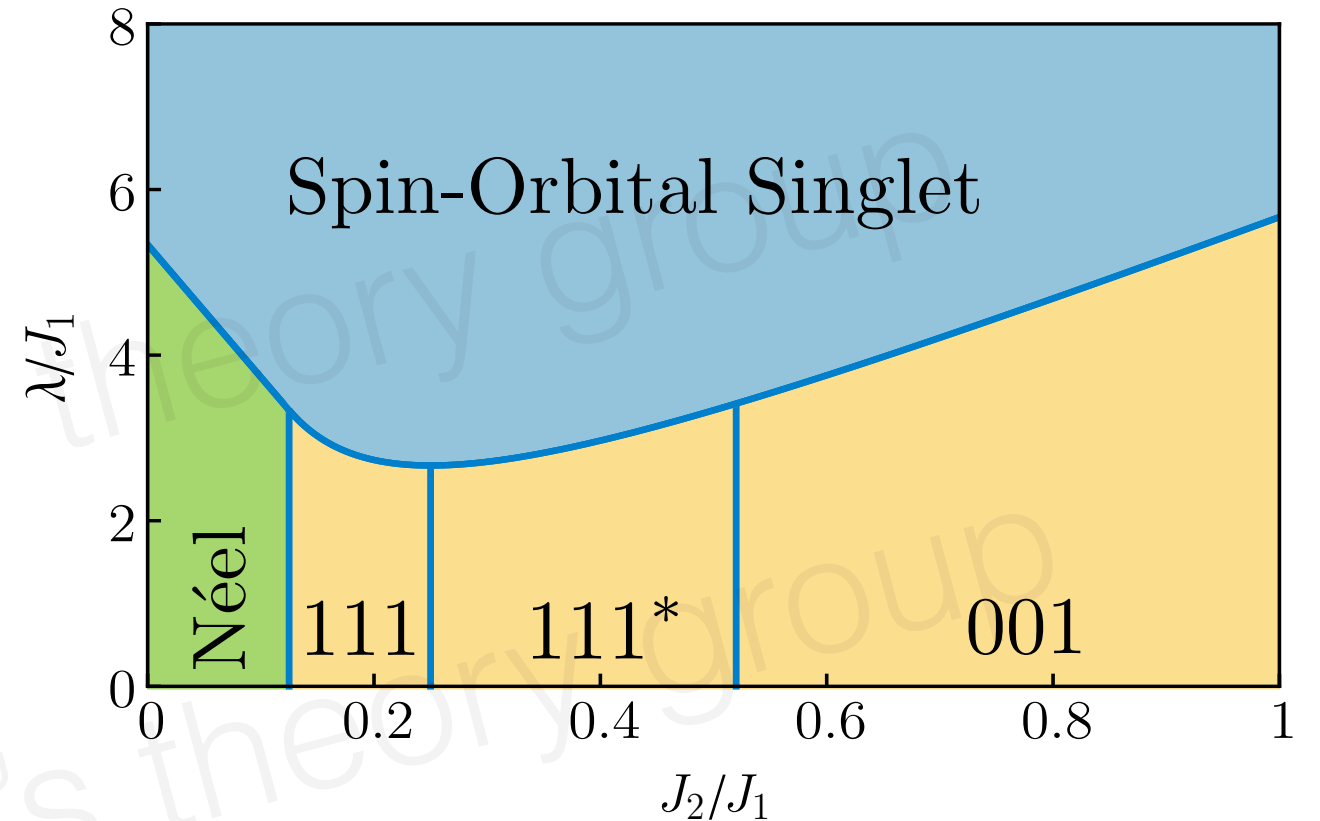
$$H = H_{\mathrm{soc}} + H_{\mathrm{ex}}.$$

Unique response to the external magnetic field
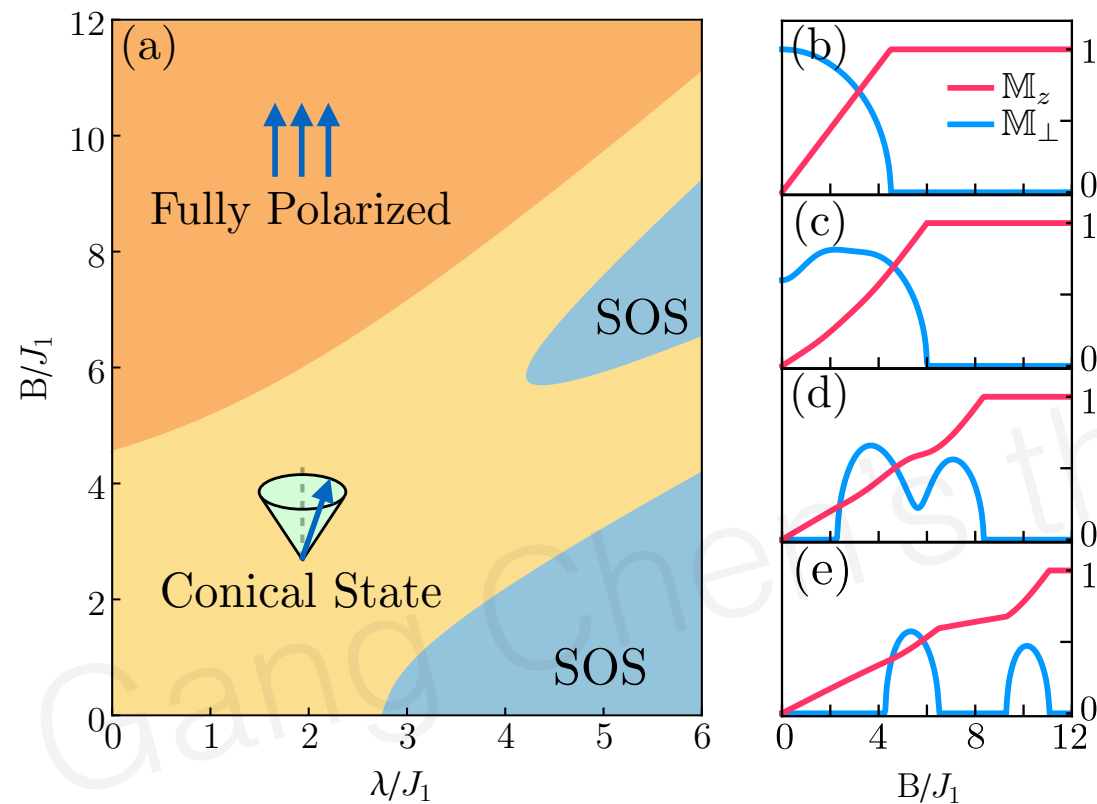
FIG. 3. (Color online.) The left panel is the phase diagram under external magnetic fields. We have fixed $J_2/J_1 = 1/4$ in the plot. "SOS" refers to spin-orbital singlet. There is a region that supports re-entrant transitions between the conical state and the SOS state as the field is varied. The right panel depicts the magnetization curves for $\lambda/J_1 = 0, 2, 4, 6$ from top to bottom.

$$H_{\text{Zeeman}} = -\sum_i B(L_i^z + 2S_i^z),$$

Fundamentally different from dimerized magnets !

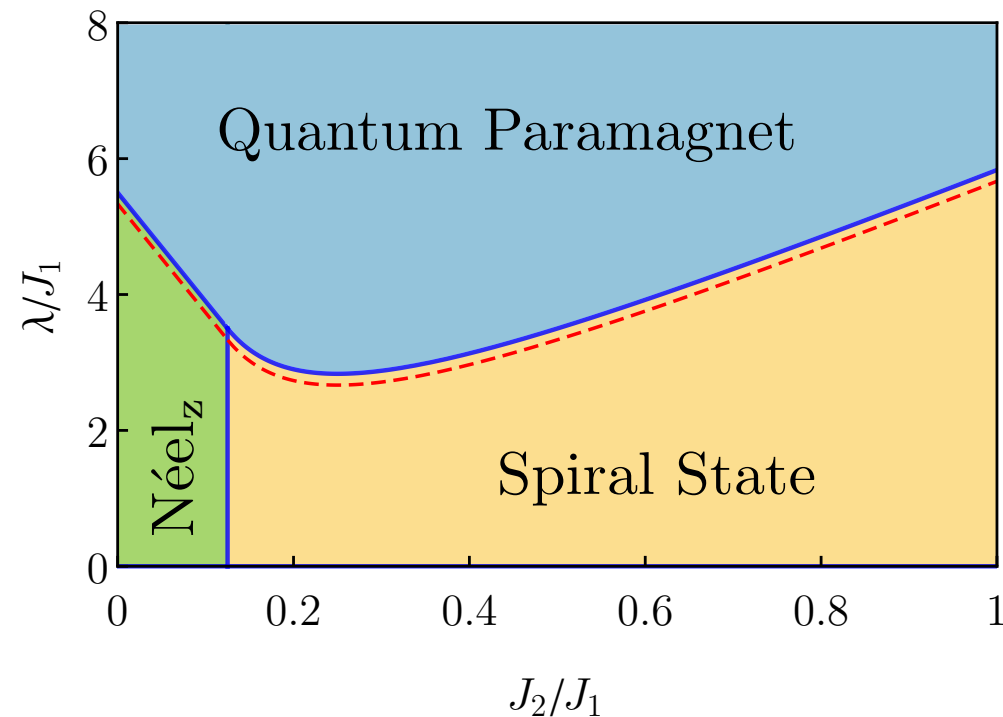# Comparison with other systems
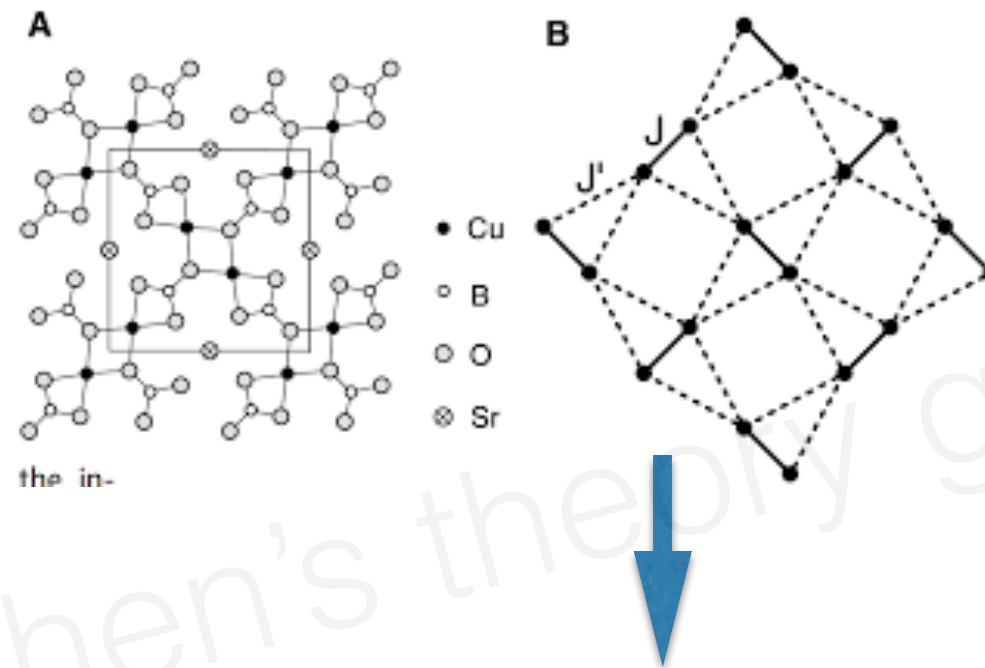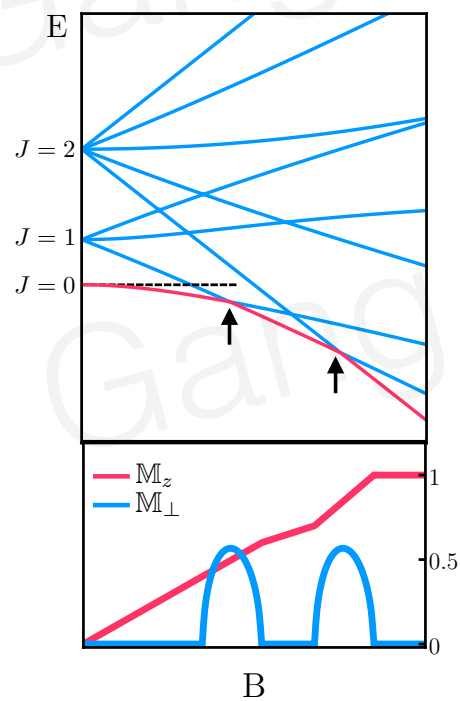
(a) d^8 Diamond AFM   (b) FeSc$_2$S$_4$ (c) S=1/2 dimer   (d) S=1 dimer
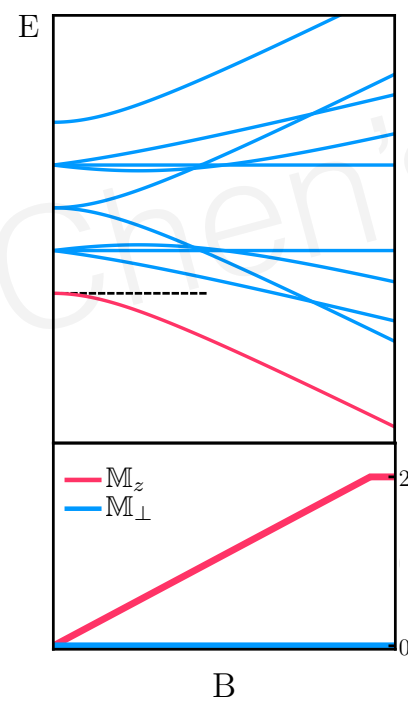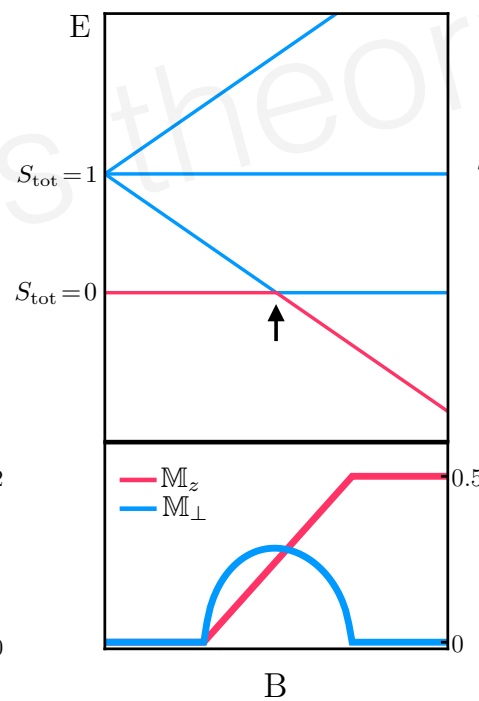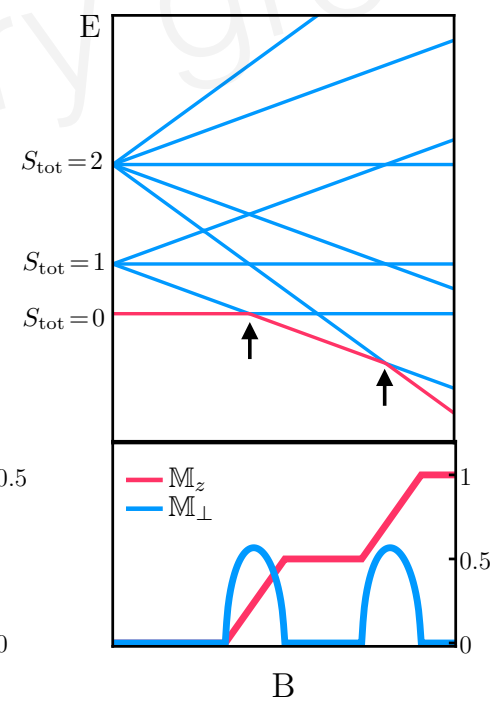
Strain: tetragonal distortion

FIG. 4. (Color online.) The phase diagram with uniaxial strain $U/J_1 = 0.5$. The Néel$_z$ phase indicates that the Néel order is along the z axis. For comparison, the red (dashed) line gives the original boundary between the quantum paramagnet and the magnetic ordered phases when $U/J_1 = 0$.

$$H_{\mathrm{Uni}} \quad = \quad -U \sum_i (L_i^z)^2.$$

Doping induced superconductivity
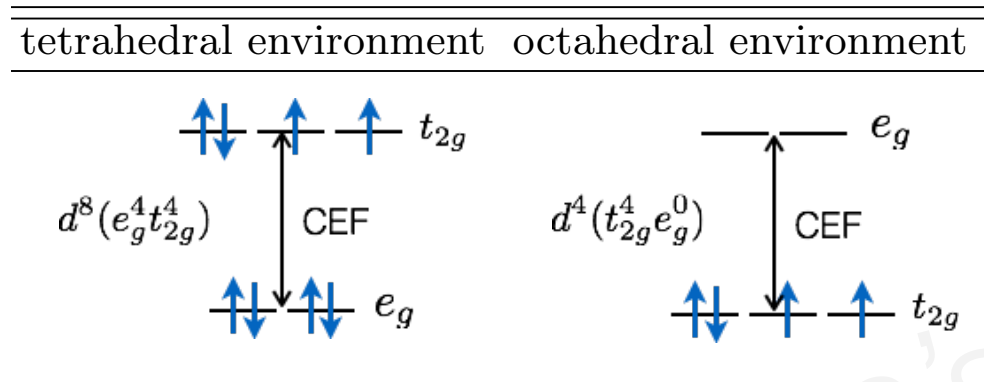
more examples. This correspondence immediately suggests some of the physics on one side may be extended to the other side. For instance, doping-induced ferromagnetism and unconventional superconductivity were proposed for the doped d^4 systems such as $CaRu_2O_4$ [15, 32]. Without much creativity, one may think such phenomena could be relevant for the doped d^8 materials with the tetrahedral crystal field environments.

Doping-Induced Ferromagnetism and Possible Triplet Pairing in d^4 Mott Insulators

Jiří Chaloupka[1] and Giniyat Khaliullin[2]

[1]*Central European Institute of Technology, Masaryk University, Kotlářská 2, 61137 Brno, Czech Republic*
[2]*Max Planck Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
(Dated: February 23, 2018)

We study the effects of electron doping in Mott insulators containing d^4 ions such as Ru^{4+}, Os^{4+}, Rh^{5+}, and Ir^{5+} with $J = 0$ singlet ground state. Depending on the strength of the spin-orbit coupling, the undoped systems are either nonmagnetic or host an unusual, excitonic magnetism arising from a condensation of the excited $J = 1$ triplet states of t_{2g}^4. We find that the interaction between J-excitons and doped carriers strongly supports ferromagnetism, converting both the nonmagnetic and antiferromagnetic phases of the parent insulator into a ferromagnetic metal, and further to a nonmagnetic metal. Close to the ferromagnetic phase, the low-energy spin response is dominated by intense paramagnon excitations that may act as mediators of a triplet pairing.
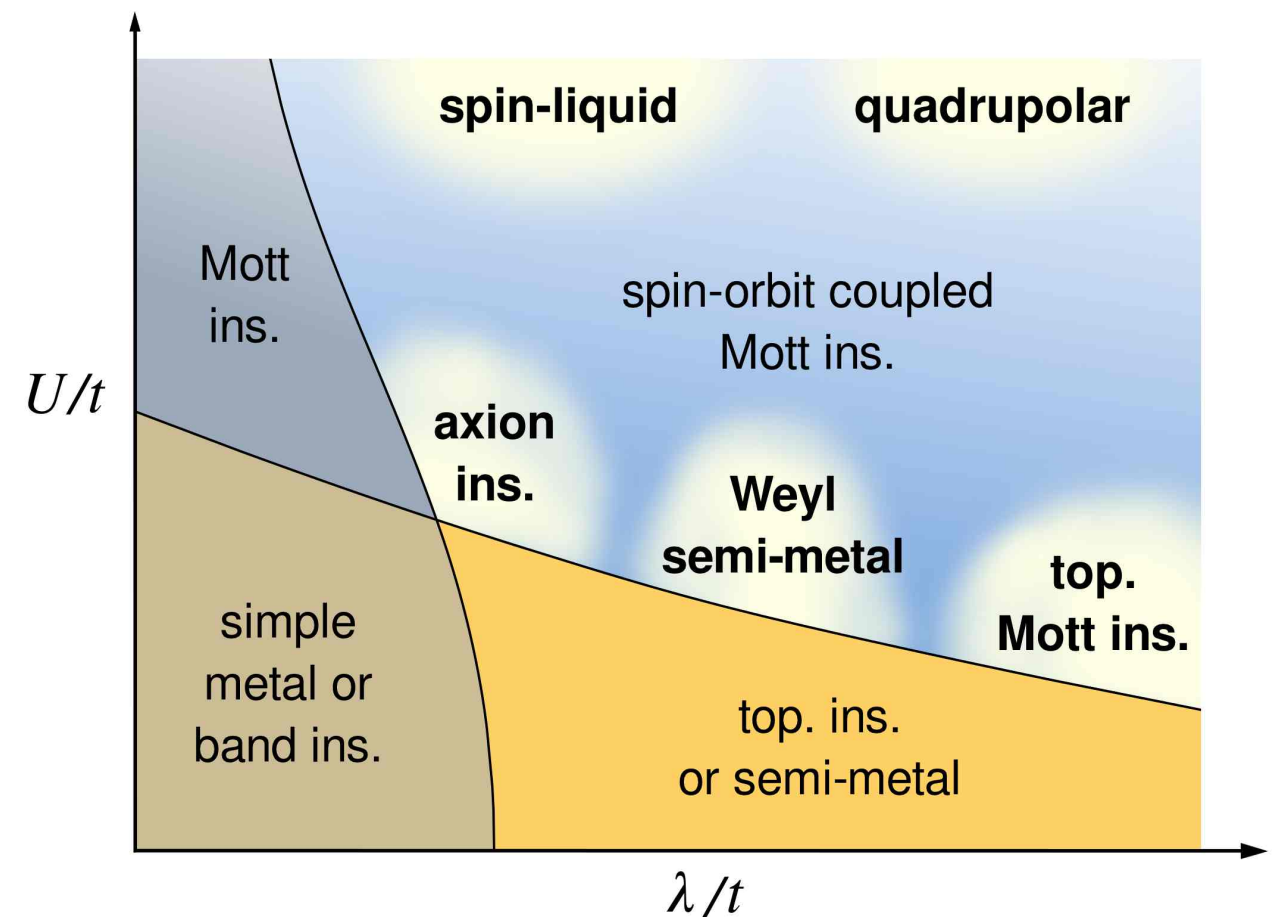
Spin-orbit entanglement meets correlation

William, **GC**, Kim, Balents, Annual Review of Condensed Matter Physics, 2014

Summary

1. We point out that $NiRh_2O_4$ spin-1 diamond lattice antiferromagnet is NOT the topological quantum paramagnet.

2. Through a minimal model, we find that the ground state can be a trivial quantum paramagnet. But due to the frustrated interaction, the excitations with respect to this trivial state develop an extensively degenerate minima in the reciprocal space.

3. Moreover, as the system approaches the phase transition to a magnetic order, these extensively degenerate low-energy bosonic modes condense at the same time, leading to an unusual critical behavior.