# Theory of cluster Mott insulators, and, a spinon Fermi surface spin liquid in YbMgGaO4

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# Part 1. Theory of cluster Mott insulators

# Outline of Part 1

- A 2D cluster magnet: $LiZn_2Mo_3O_8$
- The theory of cluster Mott insulator in 2D
- Summary

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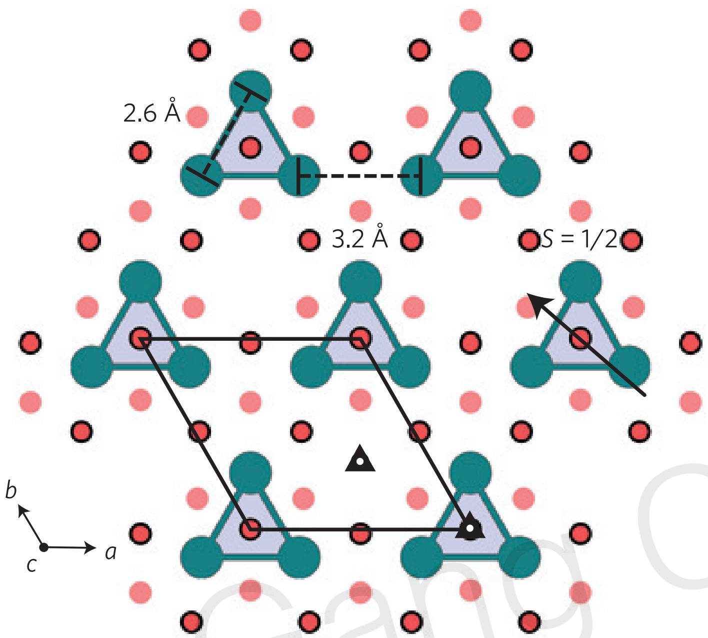
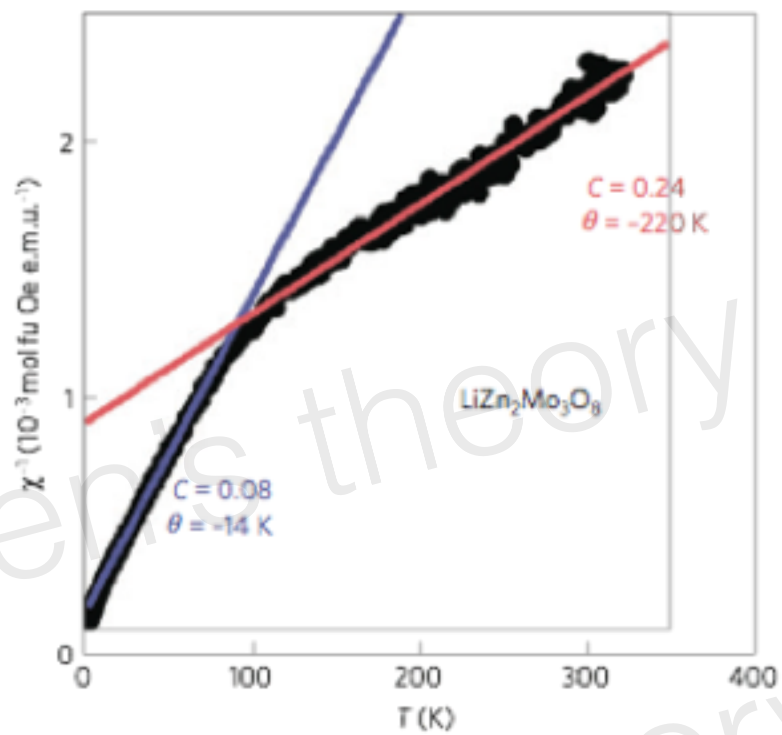
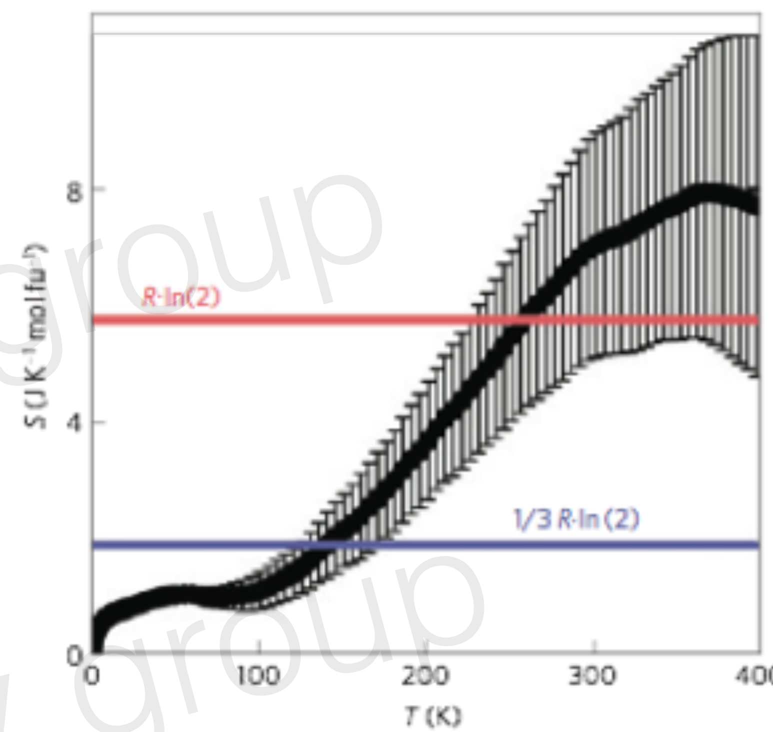
# One surprising experiment on $LiZn_2Mo_3O_8$

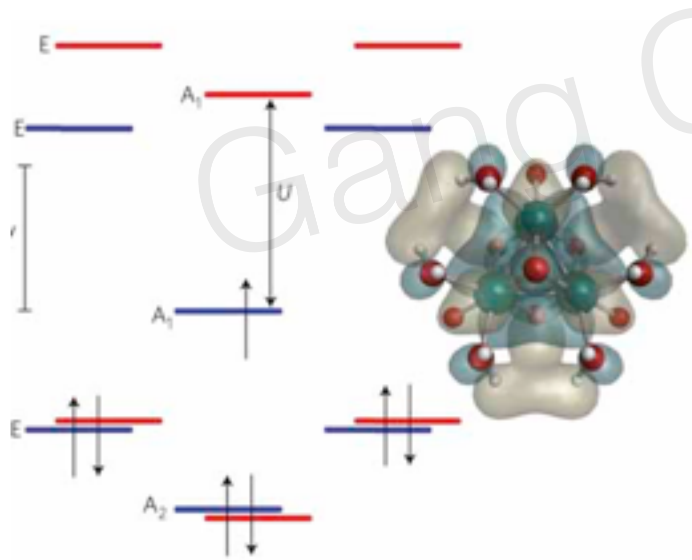
T. McQueen




Nature Material 2012





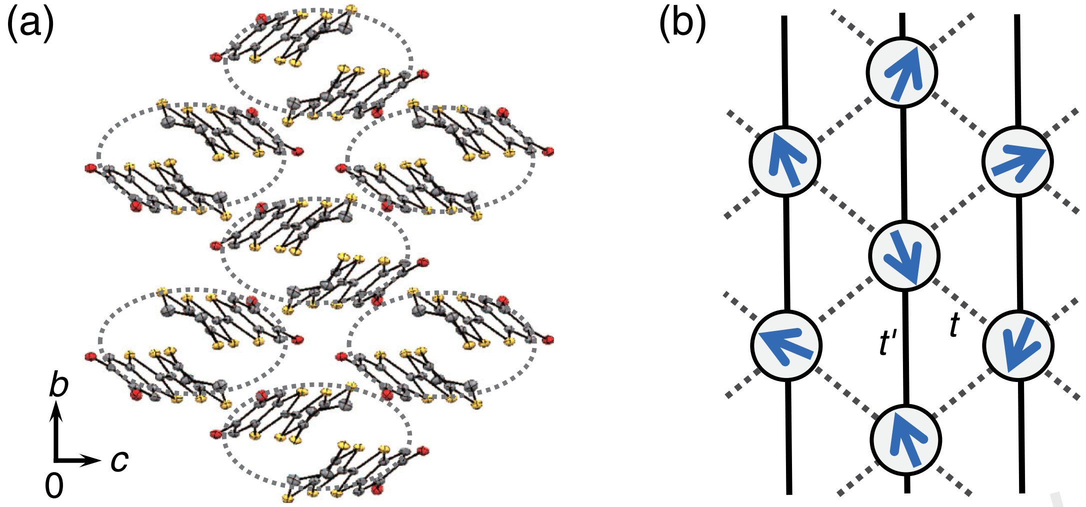

- Why unusual? neither model works.

    1. Triangular lattice Heisenberg model
    2. Triangular lattice Hubbard model at 1/2 filling

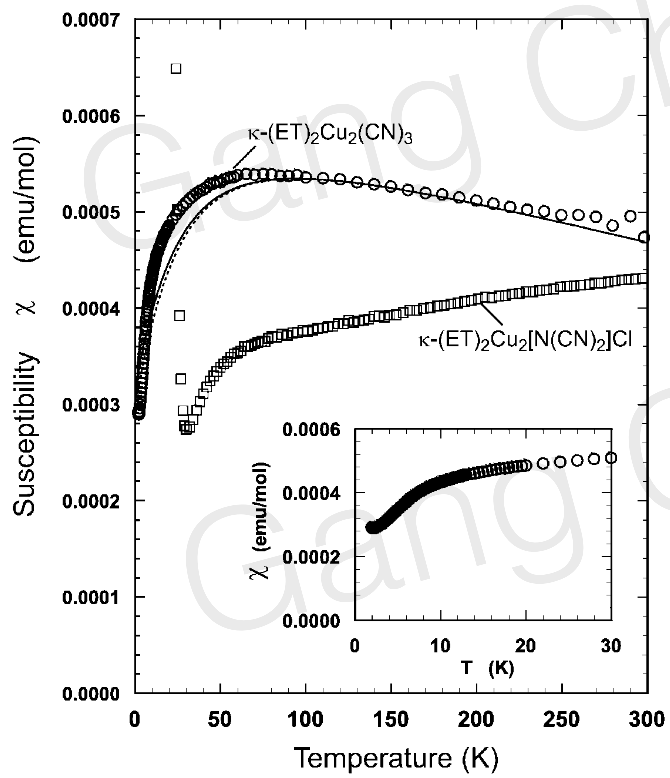
- Further low-temperature experiments: NMR, muSR, neutron scattering, proposed as a spin liquid candidate.

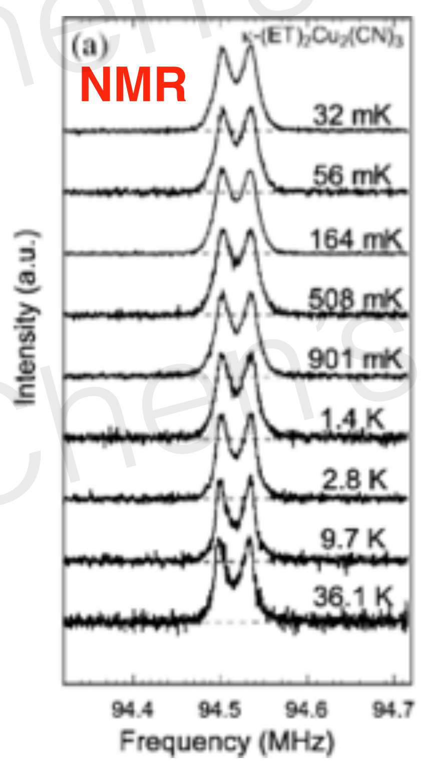
# Organic spin liquids?

Kanoda




kappa-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$,
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$,
kappa–H$_3$(Cat-EDT-TTF)$_2$ **a new one!**



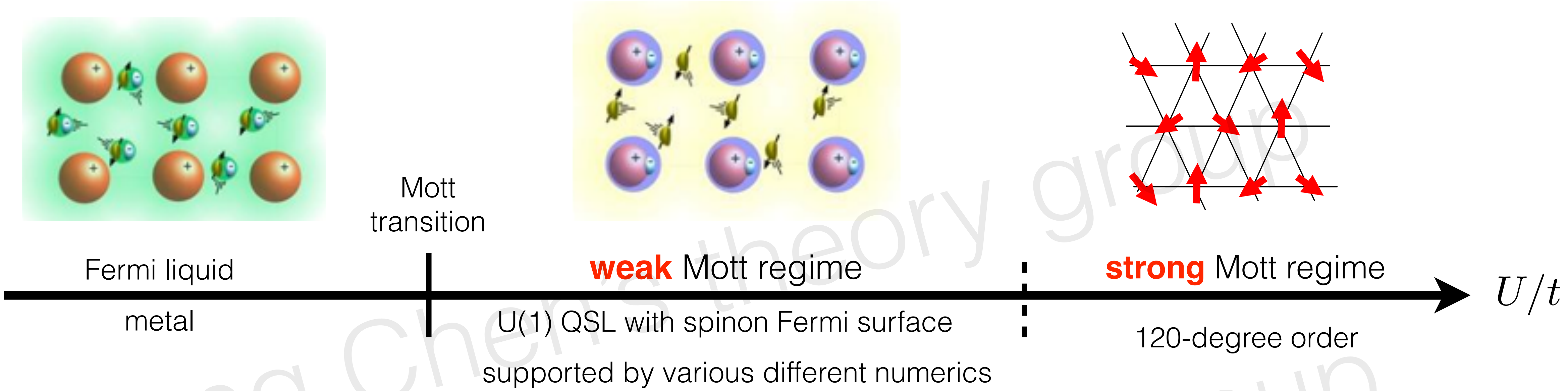

Other experiments: transport,
heat capacity, optical absorption, etc,
Unfortunately, **no neutron scattering** so far.

* No magnetic order down to 32mK
* Constant spin susceptibility at zero temperature

$$H = -t \sum_{\langle ij \rangle, \sigma} c^\dagger_{i\sigma} c_{j\sigma} + h.c. + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

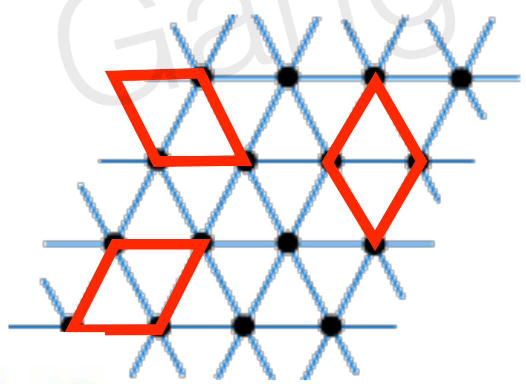

Sung-Sik Lee   T Senthil   P Lee

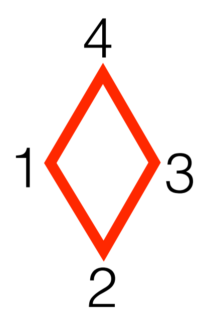
**Senthil's cartoon**




- **Physical mechanism** for weak Mott insulator spin liquids: perturbation in t/U

$$H_{\rm pert} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + K \sum_{1234} (P_{1234} + P^{-1}_{1234}) + \cdots$$






4-site ring exchange

$$(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_4)(\mathbf{S}_2 \cdot \mathbf{S}_3) - (\mathbf{S}_1 \cdot \mathbf{S}_3)(\mathbf{S}_2 \cdot \mathbf{S}_4)$$


Motrunich

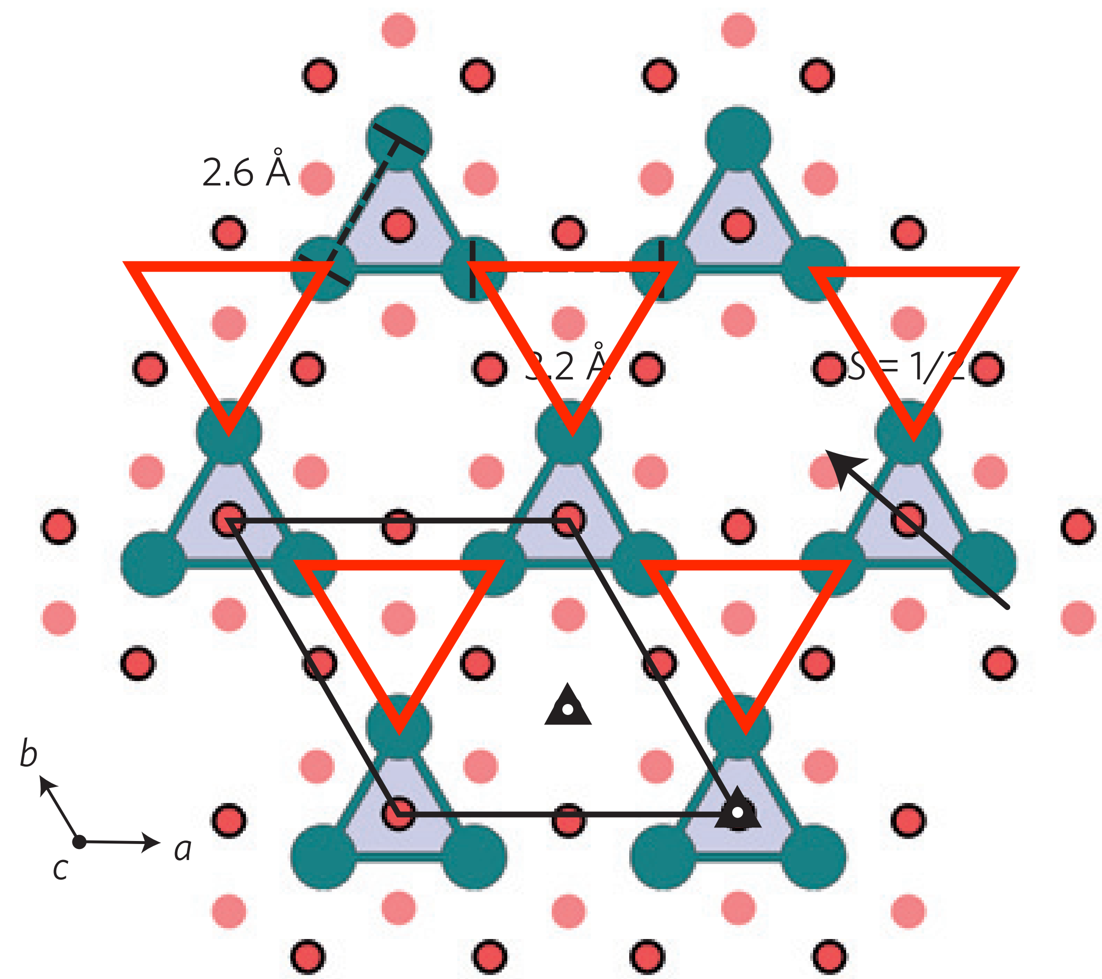

**Remark** **(on the mechanism NOT the properties)**:

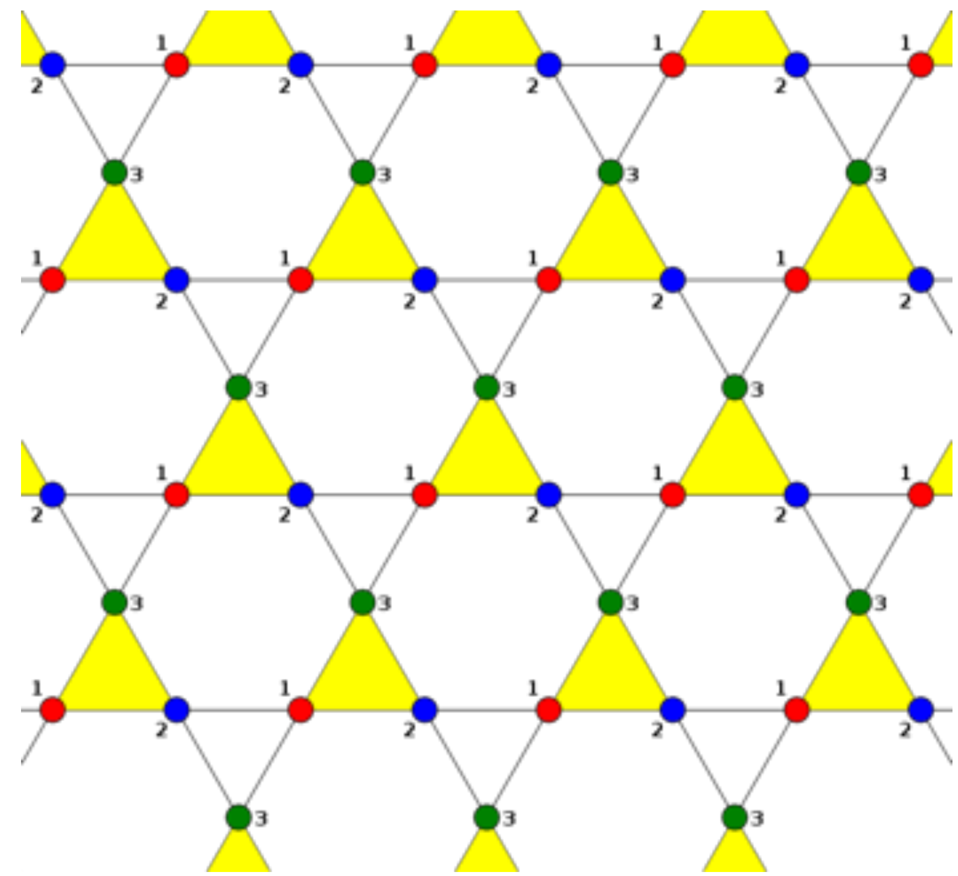
1. There is no sharp distinction between the charge fluctuations in the weak and strong Mott regimes.
2. Strong charge fluctuation in the weak Mott regime is a quantitative description.
3. Interesting physics occurs in the spin sector, but charge sector is completely trivial !

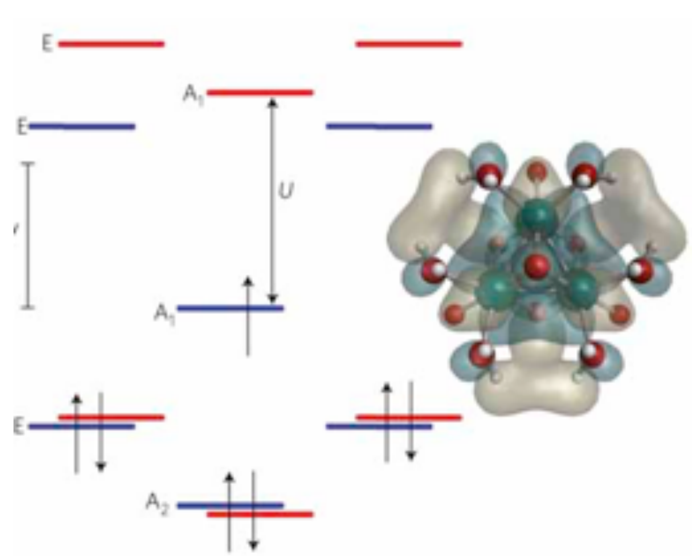
**Question / observation** **(this goes beyond just spin liquid):**

1. What if the change fluctuation is very strong, and in the most extreme case, the charge sector forms a **quantum charge liquid Mott insulator**?
2. What if the charge fluctuation leads to **some structure in the charge** sector? Spin sector is surely to be influenced in a non-trivial way. This would lead to a **striking experimental** consequence. If it is observed, it gives us confidence on the theoretical framework that we are developing.

# Cluster structure of $LiZn_2Mo_3O_8$



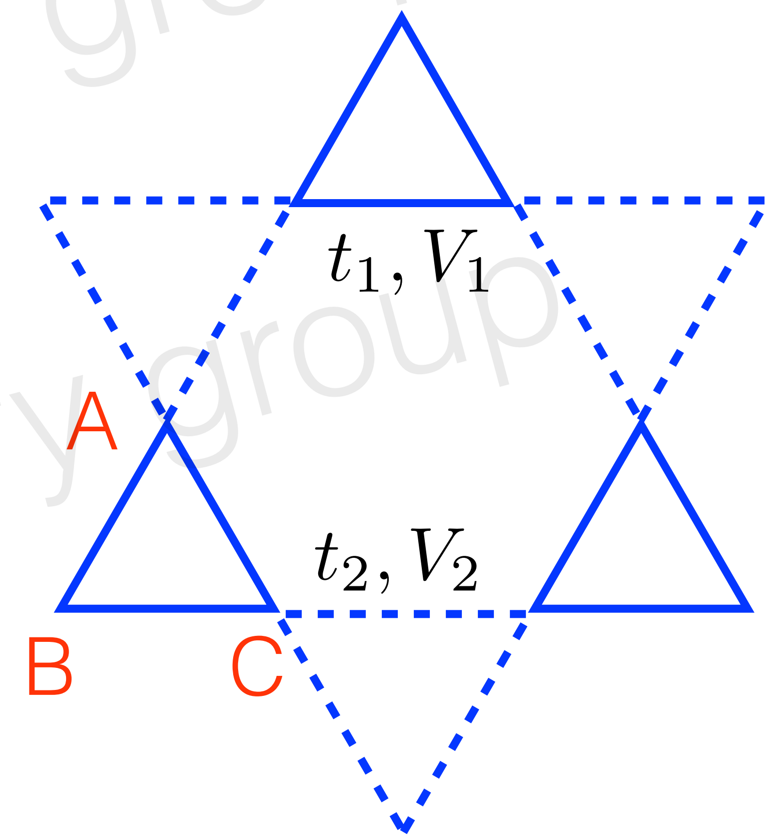


# Model

Claim: a single-band extended Hubbard model on an anisotropic Kagome lattice with **1/6 electron filling.**

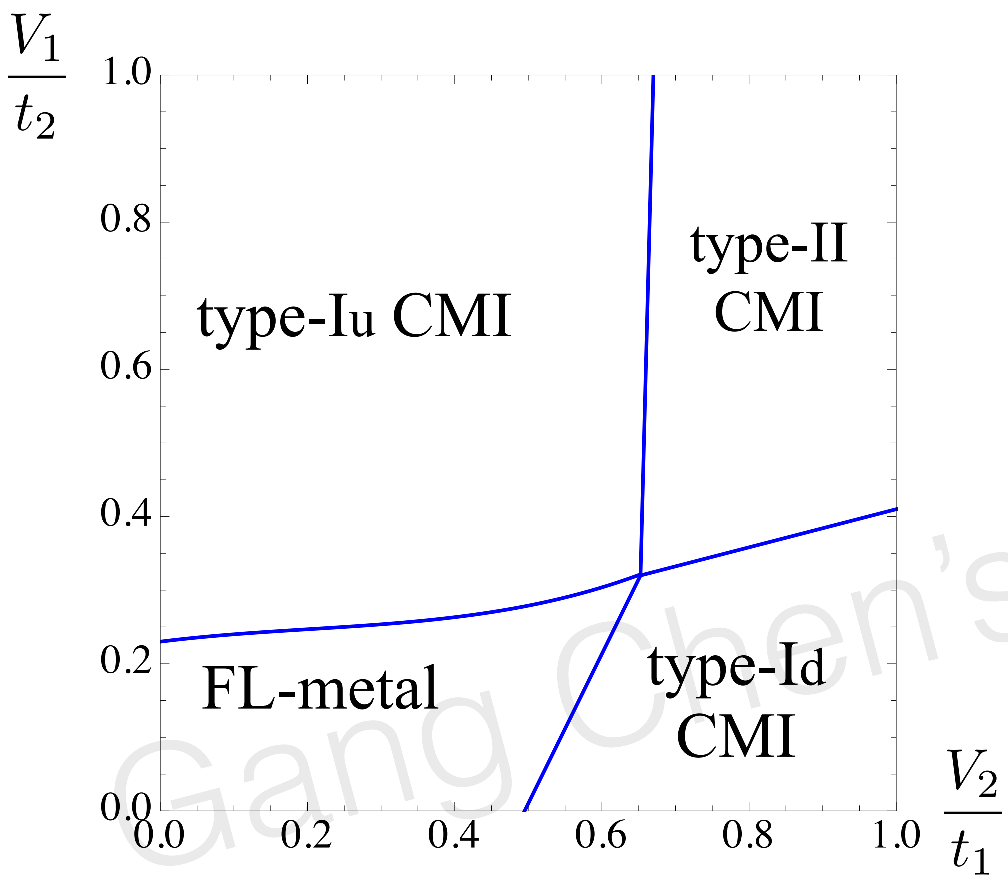
- Minimal model allowed by symmetry [require quantum chemistry understanding]

$$H = \sum_{\langle ij \rangle \in \mathrm{u}} [-t_1 (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + V_1 n_i n_j] + \sum_{\langle ij \rangle \in \mathrm{d}} [-t_2 (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + V_2 n_i n_j] + \sum_i \frac{U}{2} (n_i - \frac{1}{2})^2,$$




* Large U alone **cannot** localize the electron.
* $V_1$ and $V_2$ are needed: because it is 4d orbital, and also to localize the electron in the clusters.

# Generic phase diagram




spin sector is spin liquid

Here **t1/t2** = 4, no qualitative difference for different **t1/t2**

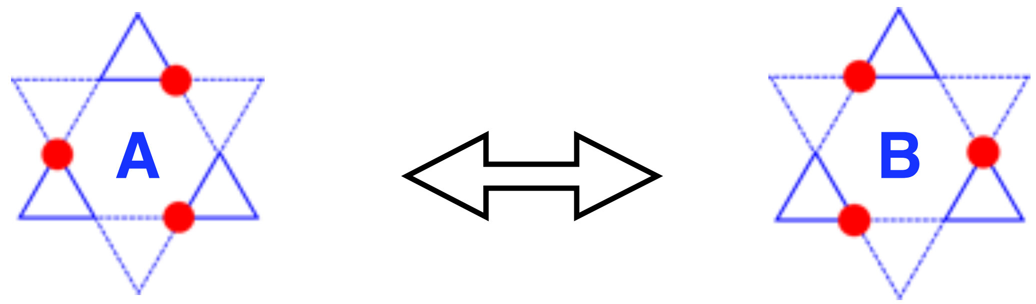
V2 is small, V1 is large

snapshots of electron occupation in type-I CMI

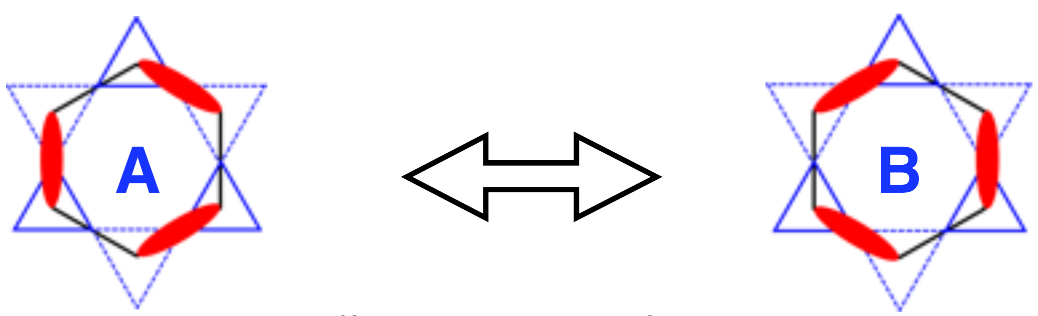
- A “simple” understanding:

* Electrons are localized in **one** type of triangles in type-I CMI;
* Electrons are localized in **both** types of triangles in type-II CMI.

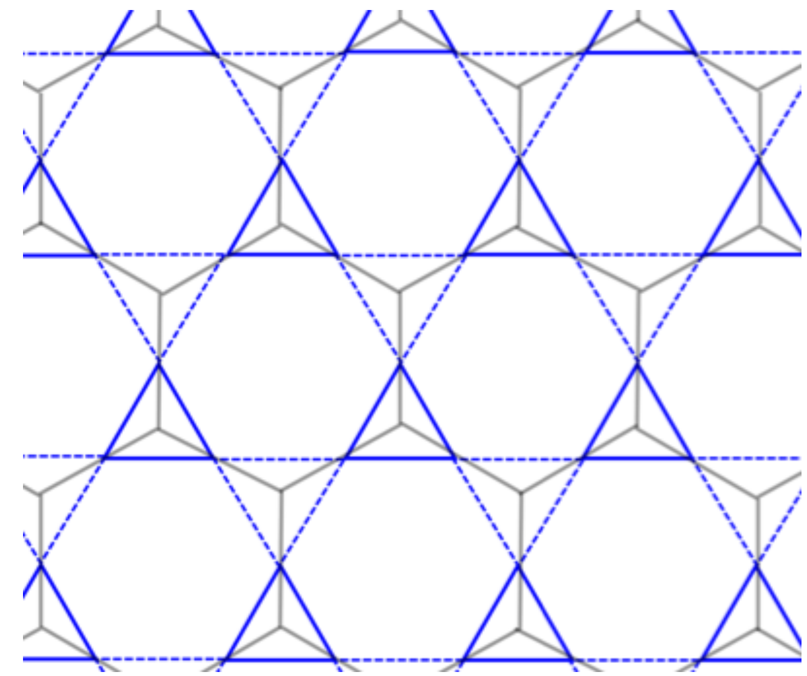
# Sub-Mott-gap process: correlated electron motion




3rd order process in type-II CMI

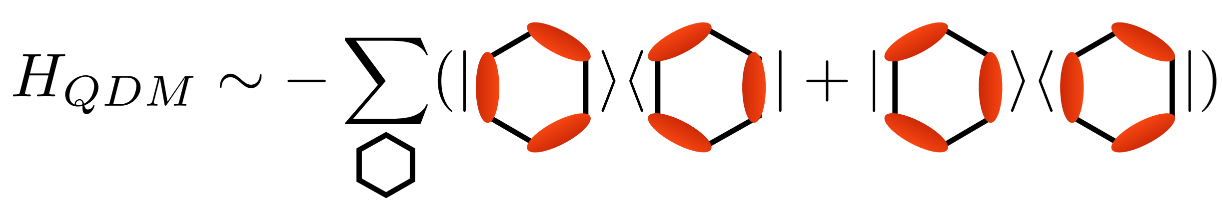



dimer resonating



Dual honeycomb lattice and Kagome lattice

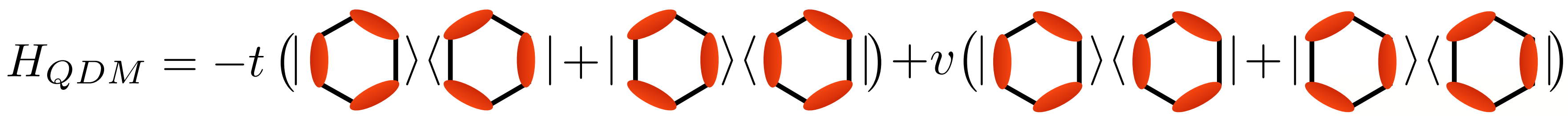
This collective tunnelling process **preserves the center of mass** of 3 electrons !

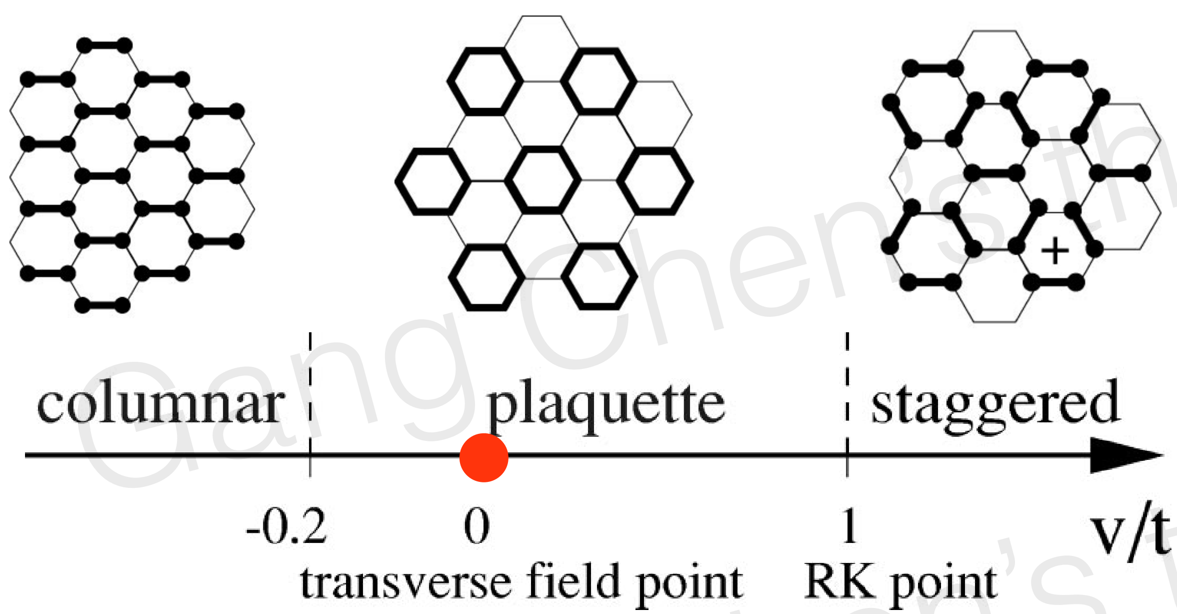


# Type-II CMI: plaquette charge order via QDM

R. Moessner S. Sondhi P. Chandra

- A model study in 2001



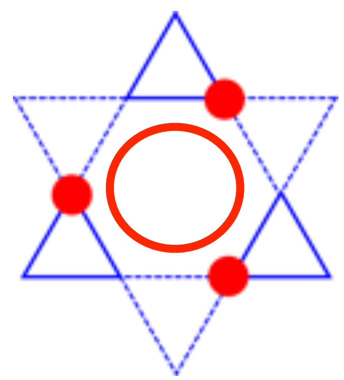

- Remarks:

* The plaquette charge order is a **local charge "RVB"**.
  (This is not Anderson's spin singlet RVB).
* One may simply view each resonating hexagon as a benzene molecule.
* It is a collective behaviour of 3 electrons.
* It is a **quantum** effect.

- **plaquette charge order**

- High energy d.o.f. (charge) usually influences low energy d.o.f. (spin). More practically, low d.o.f serves as a probe of the physical properties of the high energy d.o.f..

- Spin state reconstruction by the charge

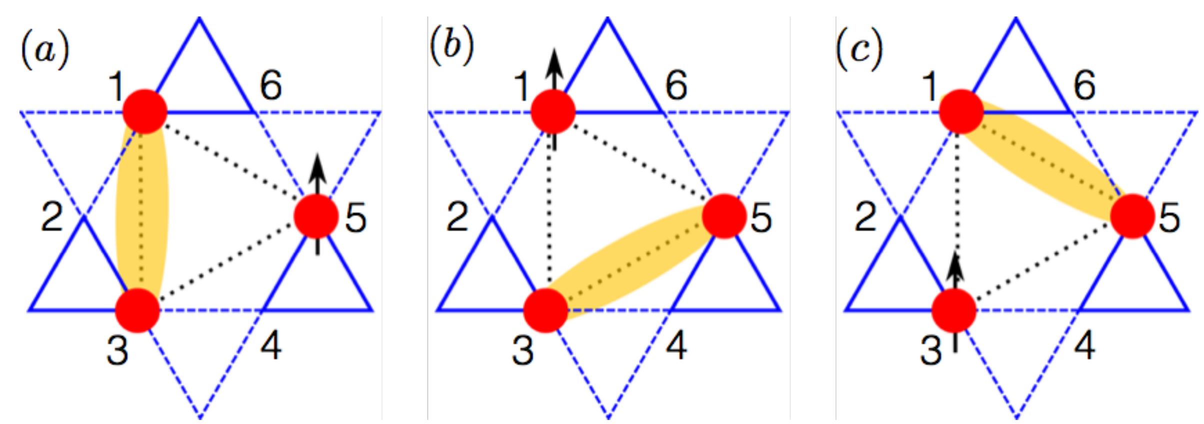


$$\frac{1}{2}\otimes\frac{1}{2}\otimes\frac{1}{2}=\frac{1}{2}\oplus\frac{1}{2}\oplus\cancel{\frac{3}{2}}$$

The total spin $S_{tot}=1/2$;
Pseudospin $\mathcal{T}=1/2$, nonmagnetic

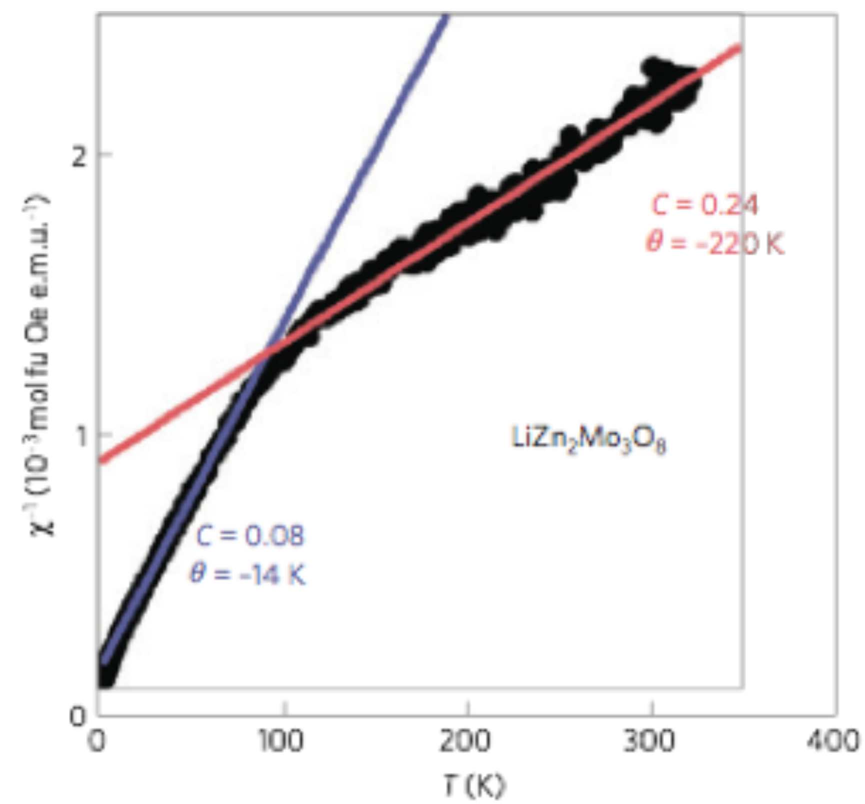
K. Kugel

D. Khomskii

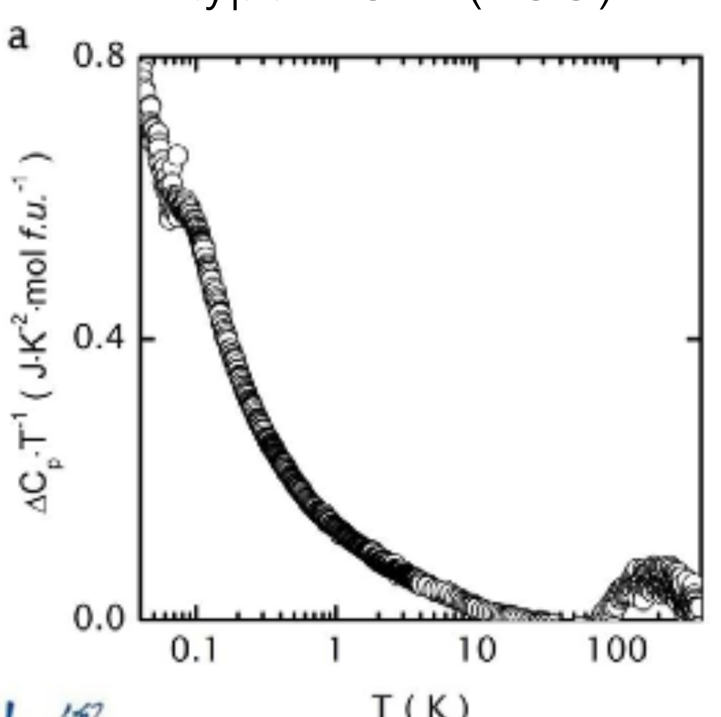



An effective Kugel-Khomskii model on the **emergent triangular lattice**

# Explanation for fractional spin susceptibility at finite temperatures

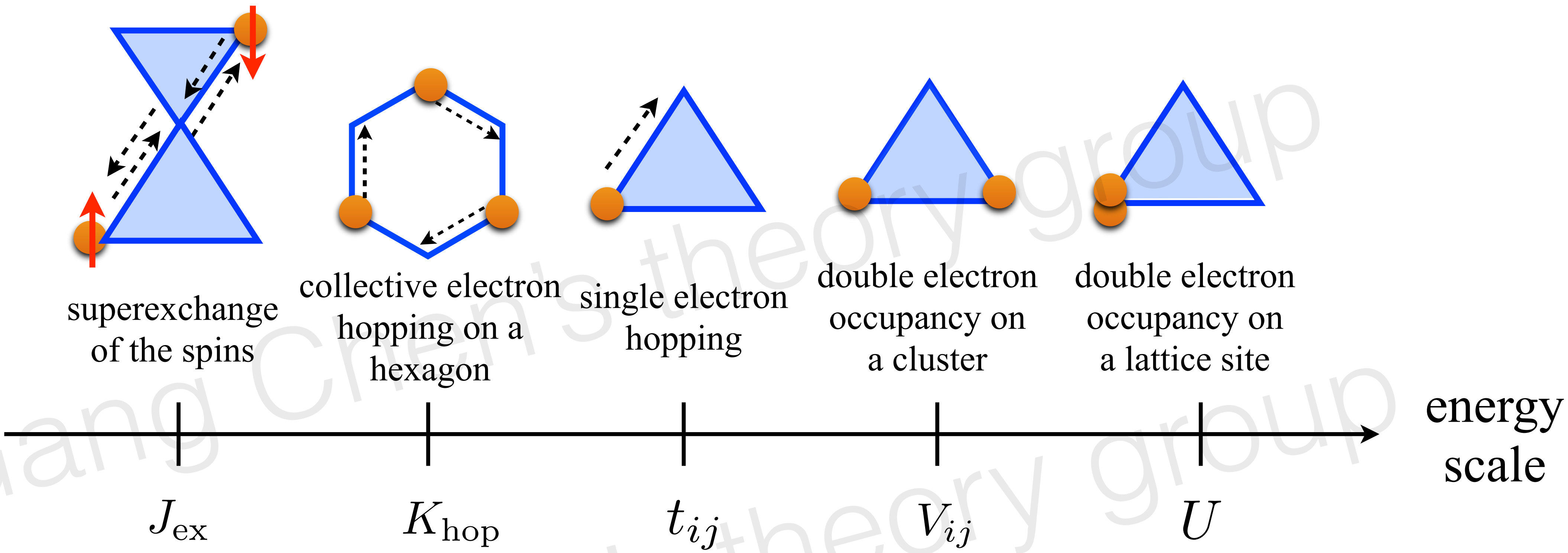



type-II CMI (PCO)




1. Expect 1st order finite temperature transition, peak at ~100K, (was interpreted as Li freezing.) smeared out 1st transition?
2. High resolution X-ray, RIXS
3. Nuclear quadrupolar resonance: electric field gradient (suggested to me by Baskaran)

# Summary: the cascade of energy scales



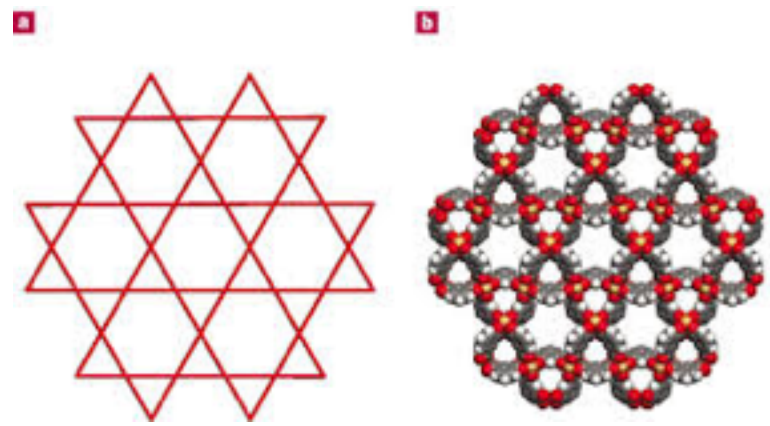




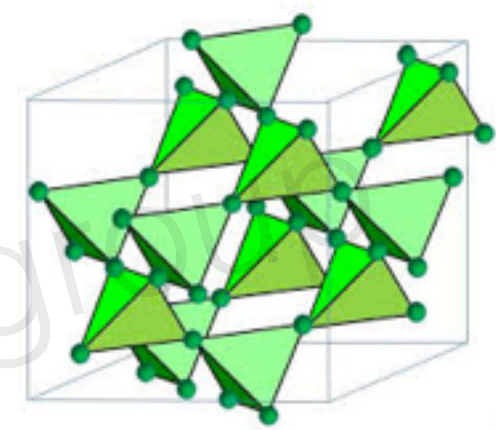

Below the Mott gap: usual superexchange process,
new collective charge fluctuation (hallmark).

# **Cluster Mott Insulator**: a new class of Mott insulators

Electrons (or bosonic particles) are localized on some cluster units instead of the lattice sites. These cluster units build the lattice.




triangle clusters in kagome
(**J. Atwood, nature mat 2002)**



tetrahedral cluster in pyrochlore



A large class of cluster magnets (Mott insulators)

$Mg_2Mo_3O_8$, $Mn_2Mo_3O_8$, $Fe_2Mo_3O_8$, $Co_2Mo_3O_8$, $Ni_2Mo_3O_8$, $Zn_2Mo_3O_8$, $Cd_2Mo_3O_8$

$LiScMo_3O_8$, $LiYMo_3O_8$, $LiInMo_3O_8$, $LiSmMo_3O_8$, $LiGdMo_3O_8$, $LiTbMo_3O_8$, $LiDyMo_3O_8$, $LiHoMo_3O_8$, $LiErMo_3O_8$, $LiYbMo_3O_8$

$NbO_2$, $Mg_3Nb_6O_{11}$, $Ba_{1.14}Mo_8O_{16}$, $NaMo_4O_6$, $GaTa_4Se_8$, $GaNb_4S_8$, $GaNb_4Se_8$ , many organic materials……

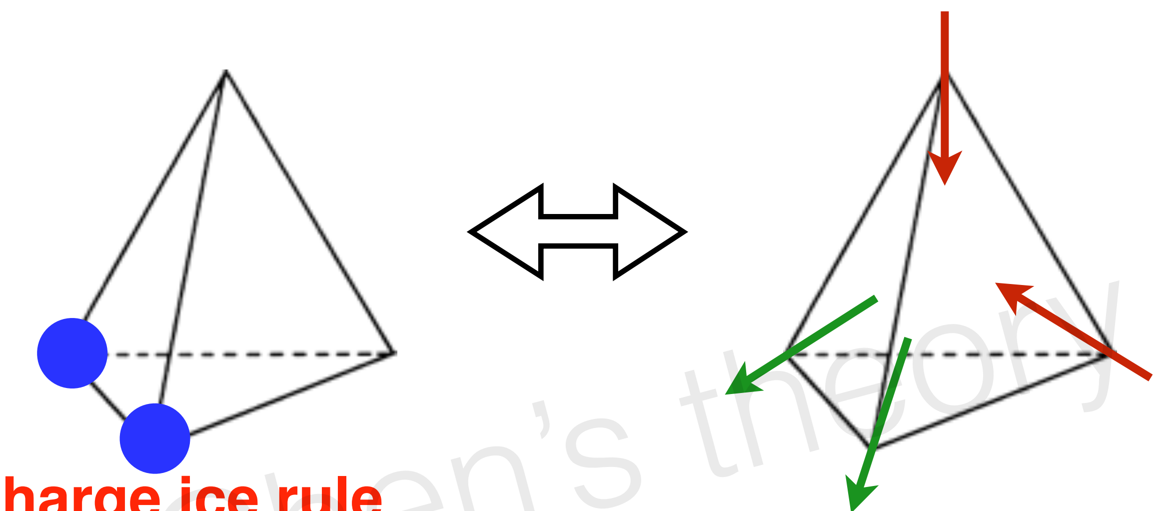
**Cluster magnets can even be systematically fabricated in organic chemistry !**

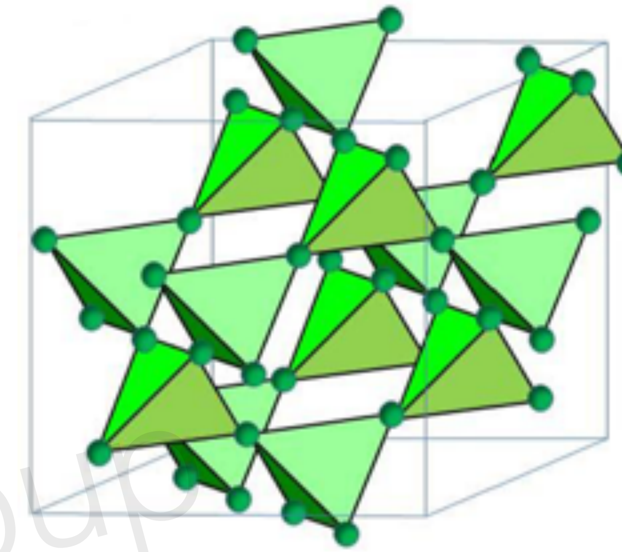
# Further extensions

- Extension to three dimensional materials
- Extension to bosonic systems.

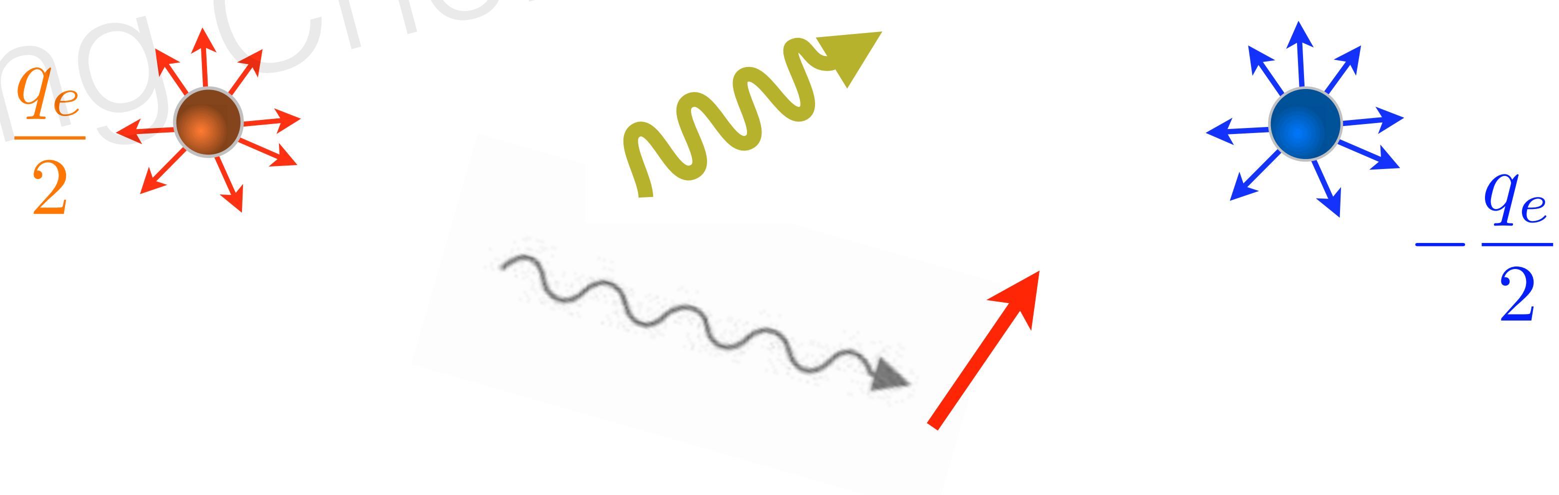
# 3D CMI as U(1) **quantum charge liquid**

$$H = -t \sum_{\langle ij\rangle,\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + h.c.) - \mu \sum_i n_i + V \sum_{\langle ij\rangle} n_i n_j + \frac{U}{2} \sum_i (n_i - \frac{1}{2})^2$$



- Low-energy physics of the charge is described by an **emergent (compact) quantum electrodynamics** in 3+1D. Charge excitation carries **1/2 the electron charge** !

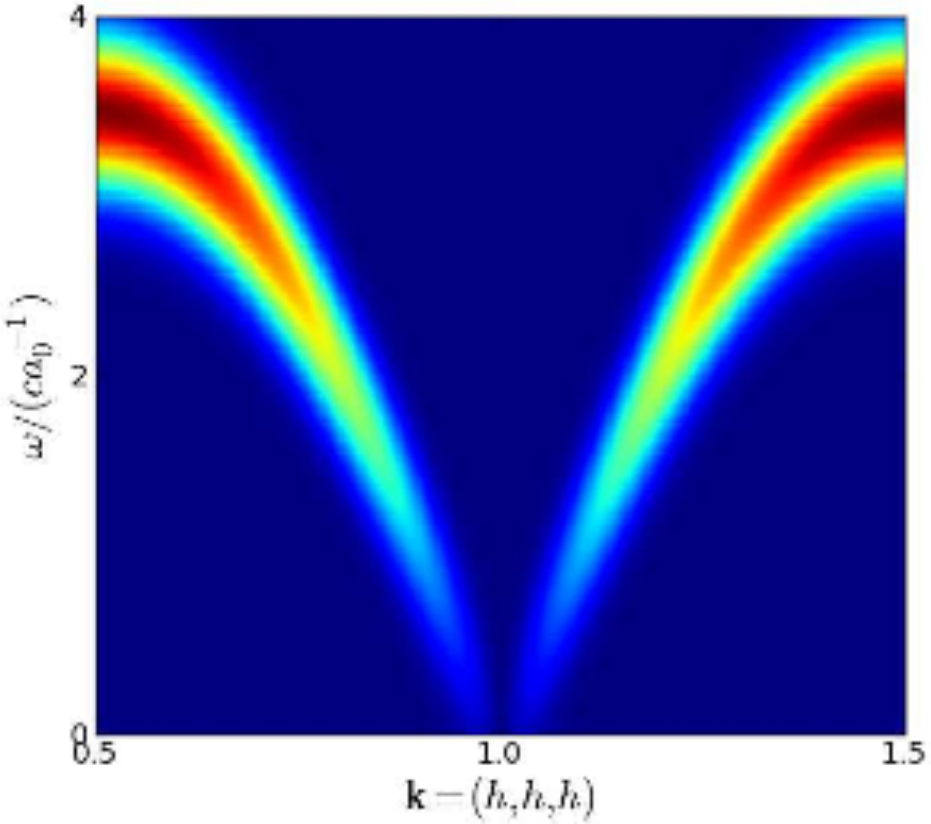



two internal U(1) gauge fields here

- (Inelastic) X-ray scattering measures U(1) gauge field correlation in the charge sector

$$\mathrm{Im}[E^{\alpha}_{-\mathbf{k},-\omega}E^{\beta}_{\mathbf{k},\omega}] \propto [\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{\mathbf{k}^2}]\,\omega\,\delta(\omega - v|\mathbf{k}|),$$

$$\mathbf{E}_{\mathbf{r}+\frac{1}{2}\mathbf{e}_\mu} \equiv L^z_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}\frac{\mathbf{e}_\mu}{|\mathbf{e}_\mu|} = (n_{\mathbf{r}+\frac{1}{2}\mathbf{e}_\mu} - \tfrac{1}{2})\frac{\mathbf{e}_\mu}{|\mathbf{e}_\mu|}$$




$$I(\omega) \sim \omega$$

emergent light in quantum charge ice !


Byron den Hertog / Science

$$\langle E^{\alpha}_{-\mathbf{k}}E^{\beta}_{\mathbf{k}}\rangle \propto \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{\mathbf{k}^2}$$

Pinch points in equal-time charge structure factor at T > ring hopping. “classical charge ice”

Hermele etc 2004
N Shannon etc 2012,
L Savary etc 2012

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# Framework: a new parton construction

- The slave rotor construction is used to describe the **conventional Mott insulator,** e.g. triangular lattice Hubbard model at 1/2 filling

$$c_{i\sigma} = e^{-i\theta_i} f_{i\sigma}$$

one U(1) gauge field

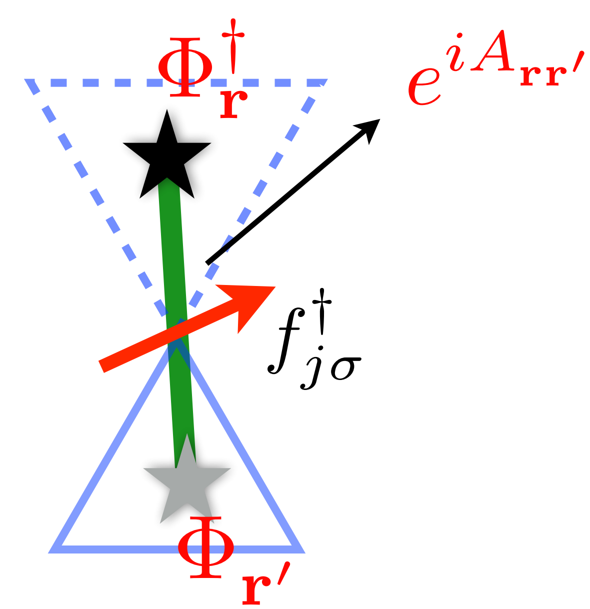
$e^{-i\theta_i}$: charge-$q_e$ spinless boson

$f_{i\sigma}$: charge-neutral spin-1/2 fermion

- A new parton gauge construction is required for cluster Mott insulators to **capture additional U(1) gauge structure** in the charge sector

$$c^\dagger_{j\sigma} \sim f^\dagger_{j\sigma} \Phi^\dagger_{\mathbf{r}} \Phi_{\mathbf{r}'} e^{iA_{\mathbf{r}\mathbf{r}'}}$$

two U(1) gauge fields: $U(1)_c \times U(1)_{sp}$



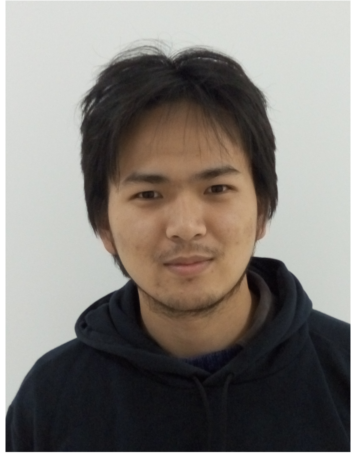
# 2. A rare-earth triangular lattice quantum spin liquid: $YbMgGaO_4$

Experimental collaborators

Yuesheng Li (Renmin)
Qingming Zhang (Renmin)

Yao Shen (Fudan)
Jun Zhao (Fudan)

Theoretical collaborators



Yaodong Li (Fudan)

more recently,
Yuan-Ming Lu (OSU)

Other contributors:

Wei Tong (High Magnetic field Lab, Hefei)
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Zhaorong Yang (Institute of Solid-State Physics, Hefei)
Xiaoqun Wang (Renmin, Shanghai Jiaotong)

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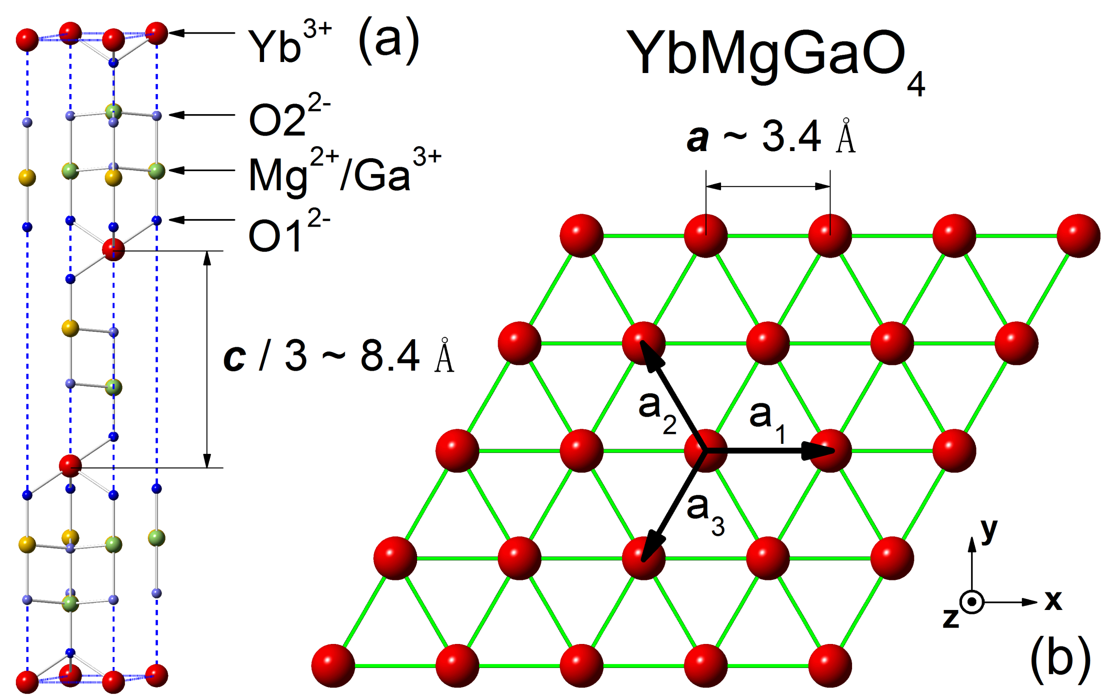
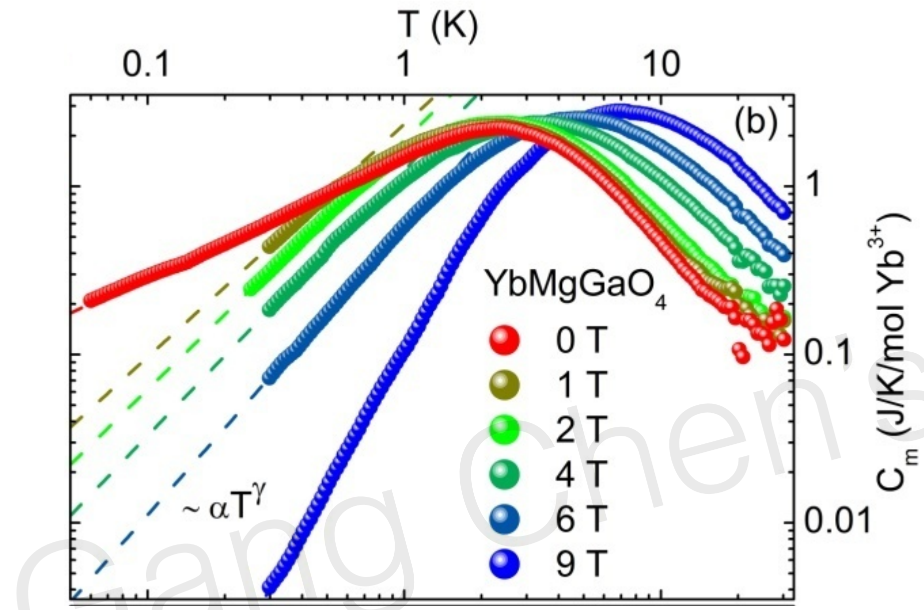
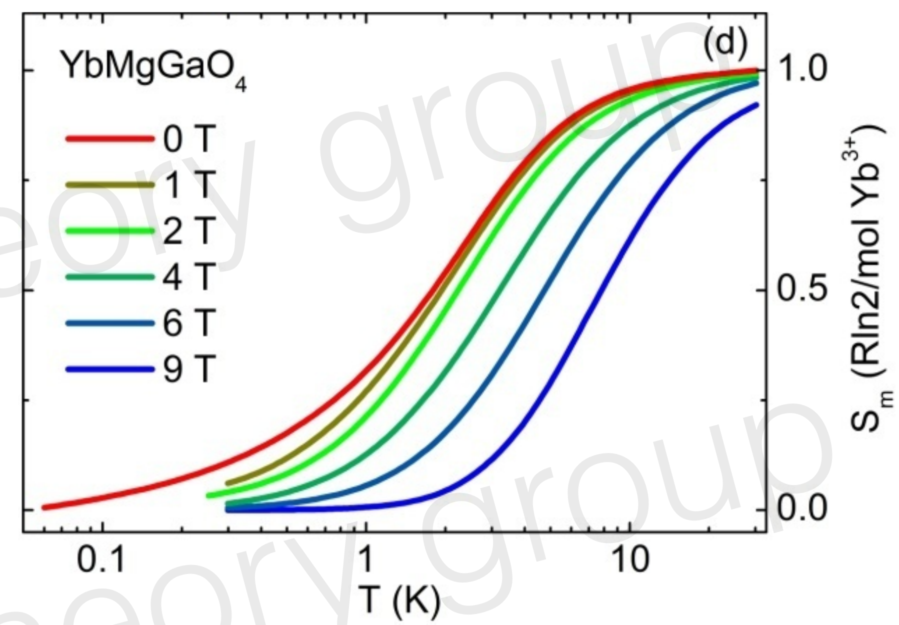
# A rare-earth triangular lattice quantum spin liquid: **$YbMgGaO_4$**




- Hastings-Oshikawa-Lieb-Shultz-Mattis theorem.
- Recent extension to spin-orbit coupled insulators (Watanabe, Po, Vishwanath, Zaletel, PNAS 2015).
- This is the **first** strong spin-orbit coupled QSL with odd number of electrons and effective spin-1/2.
- It is the **first** clear observation of $T^{2/3}$ heat capacity. I think it is spinon Fermi surface U(1) QSL.
- Inelastic neutron scattering is consistent with spinon Fermi surface results.
- We understand the microscopic Hamiltonian and the physical mechanism.

# YbMgGaO$_4$




- observation of $T^{2/3}$ heat capacity
- Entropy: effective spin-1/2 local moments

Our proposal for ground state: spinon Fermi surface U(1) QSL.



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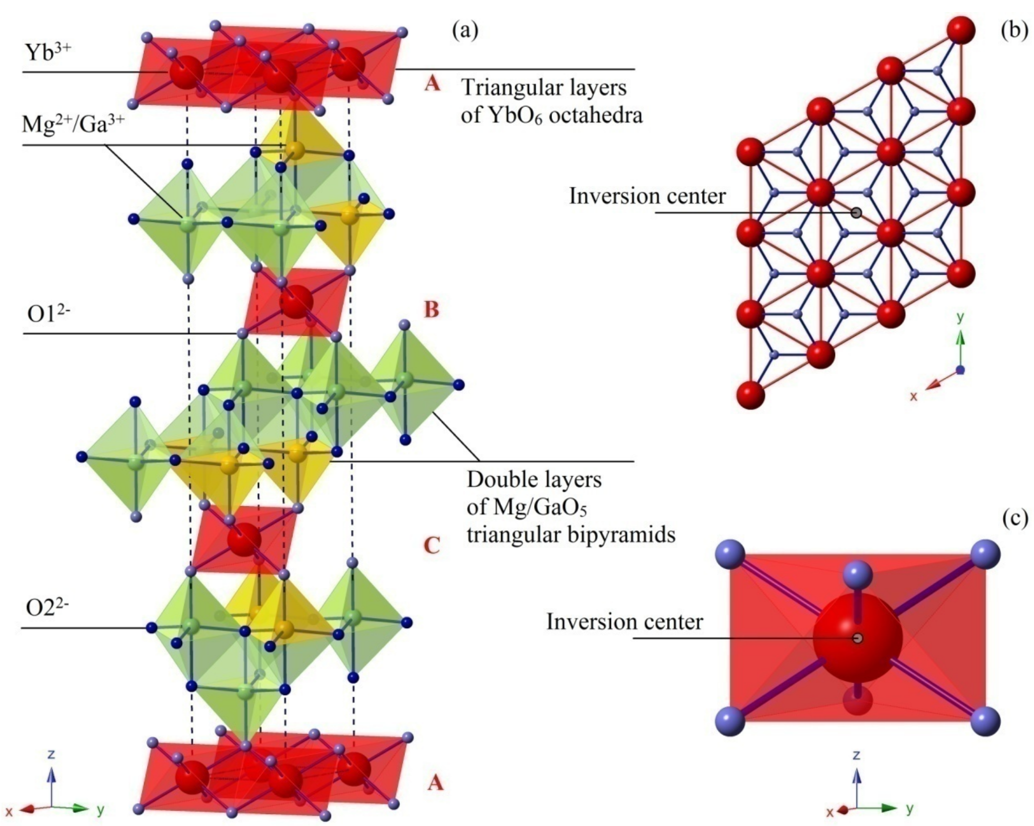
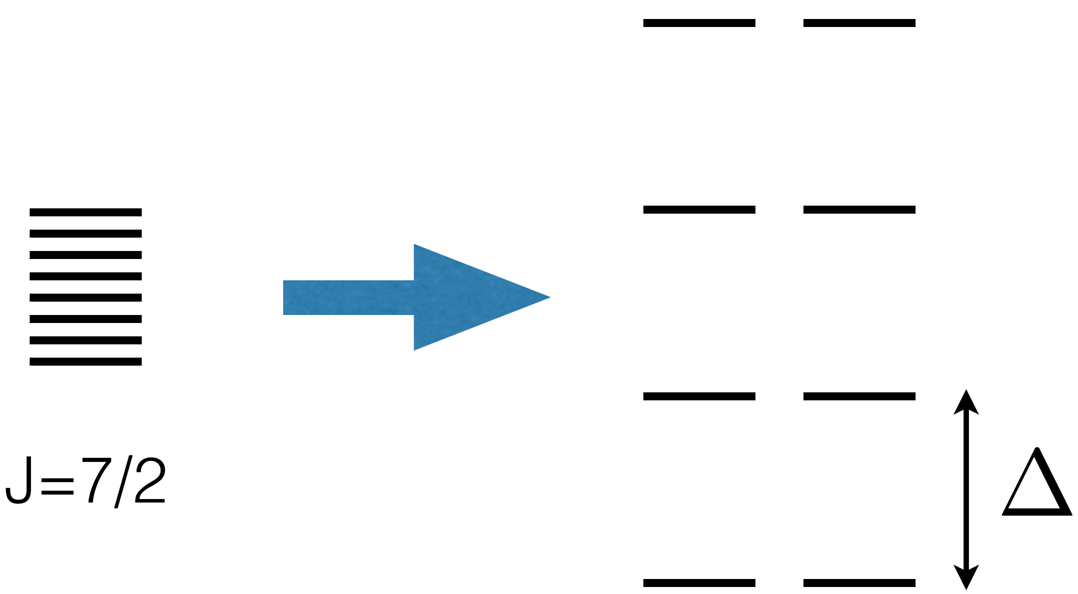
# Microscopics

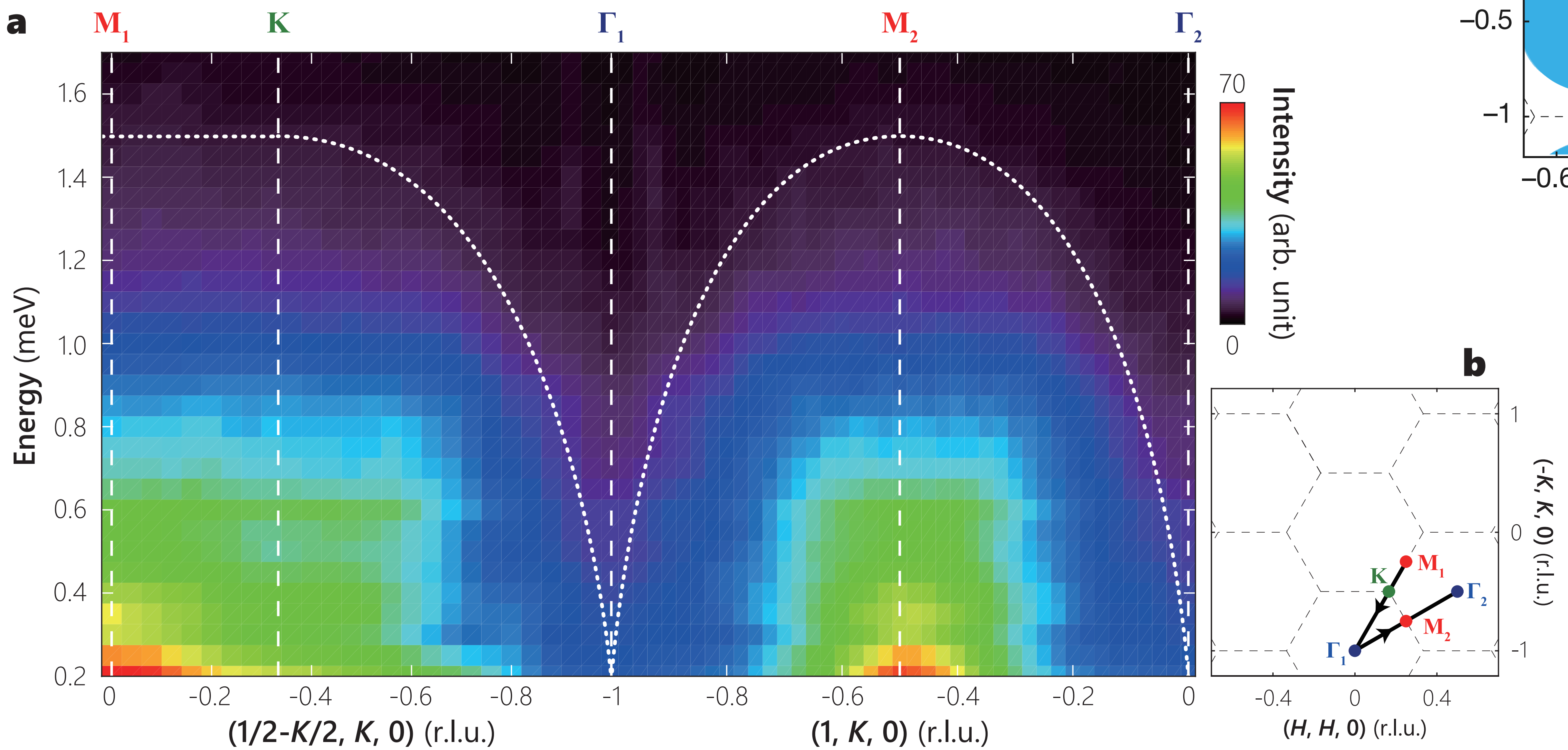


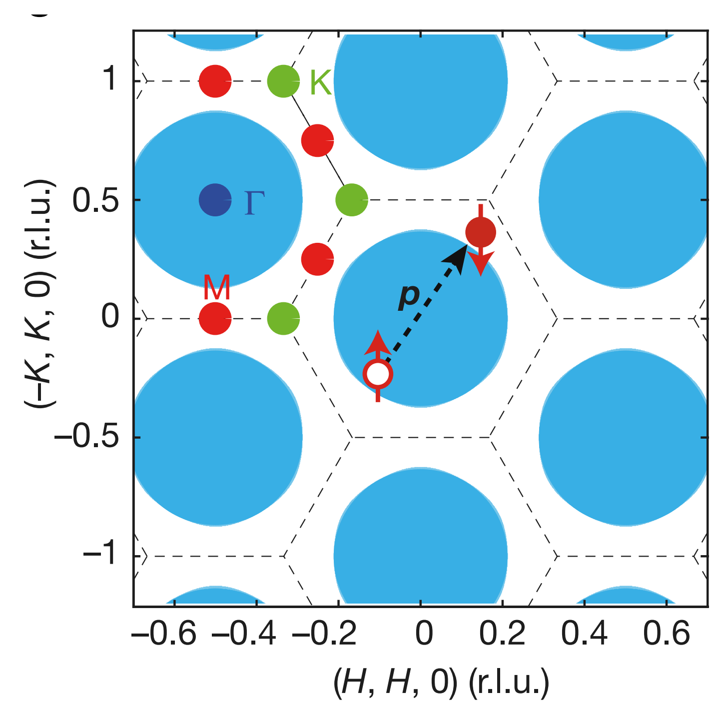

Yb$^{3+}$ ion: 4f$^{13}$ has J=7/2 due to SOC.




At $T \ll \Delta$, the only active DOF is the ground state doublet that gives rise to an effective spin-1/2.

# Advantage for neutron scattering



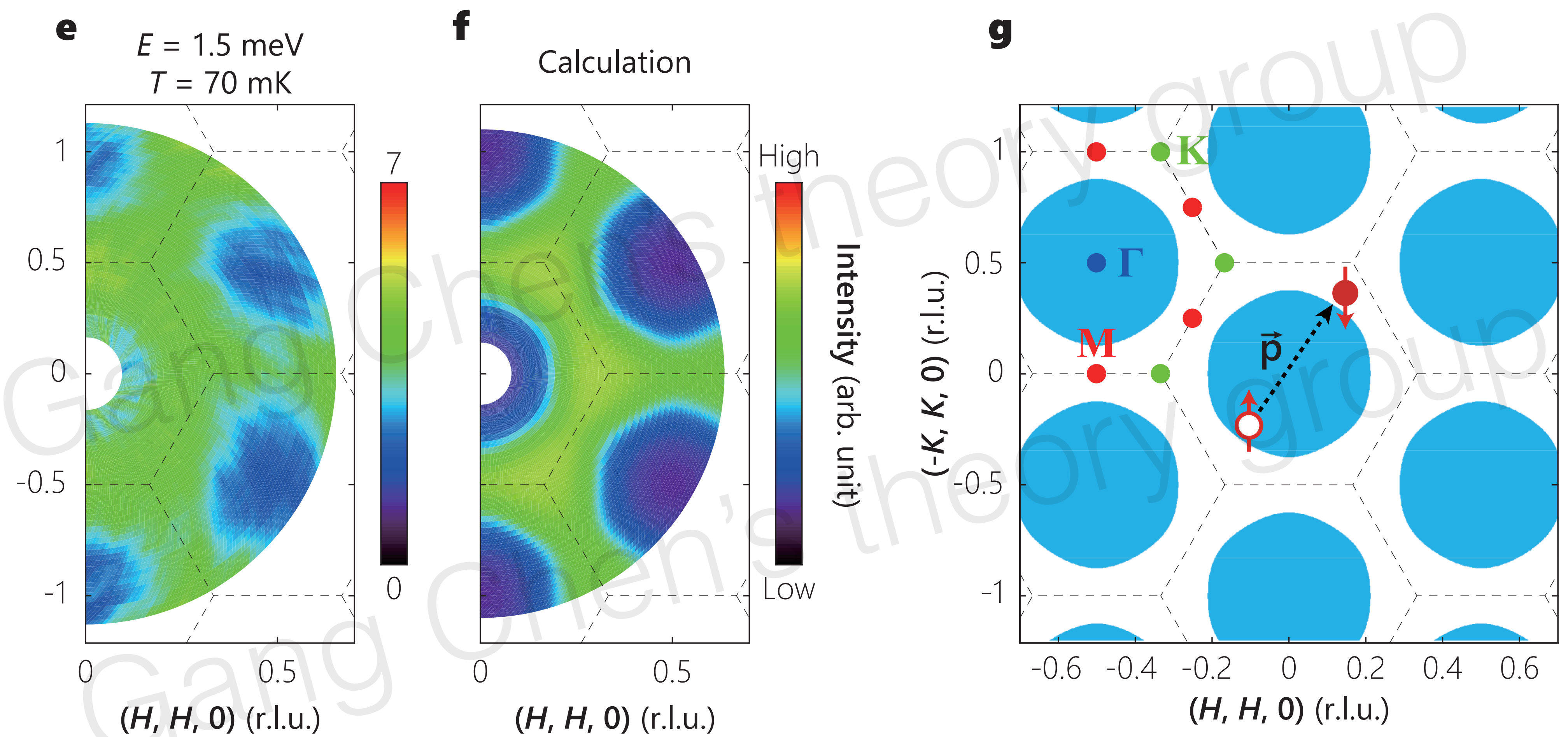




Continuum excitation

Near T=0, but not-very-low energy excitation

Yao Shen, …Gang Chen*, Jun Zhao* Nature

# Spinon continuum




Yao Shen, …Gang Chen*, Jun Zhao* Nature

# Two Major Questions

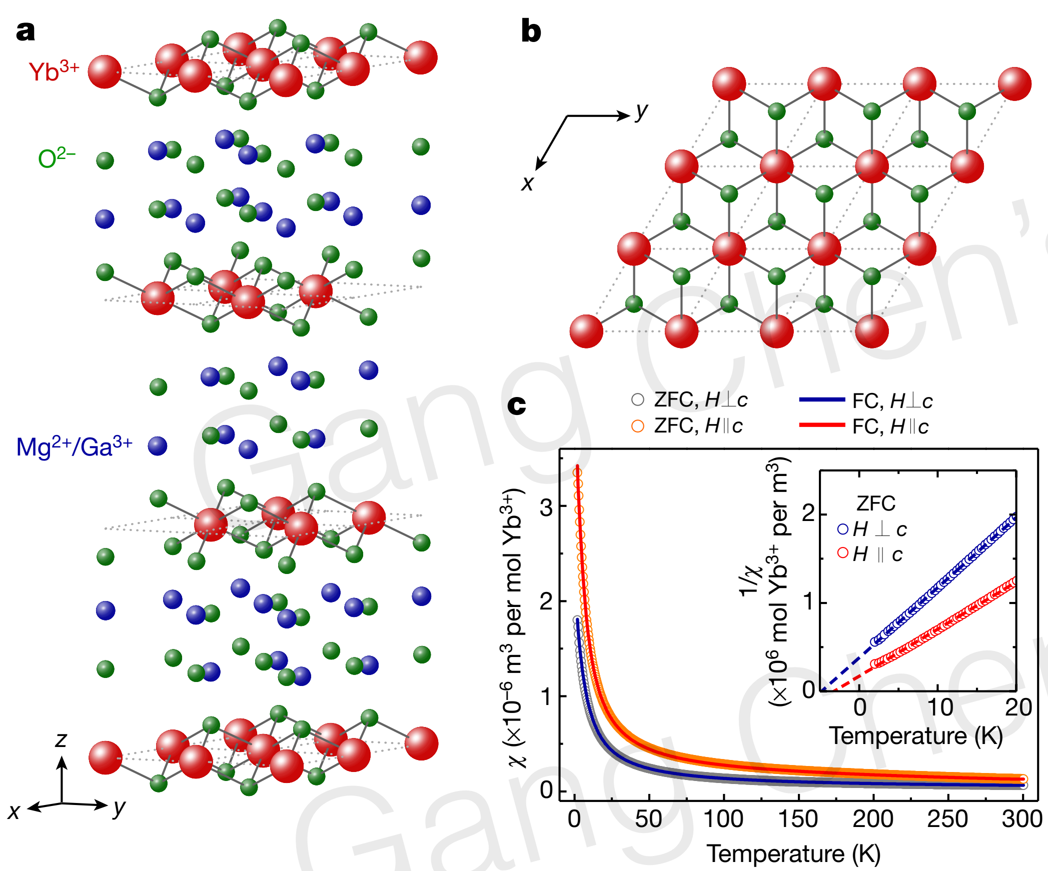
1. Whether the continuum represents the fractionalized spinon excitation? Probably most important !

   (Our new work will appear soon. )

2. What is the physical origin of the QSL physics ?

# Spin-orbit coupling

4f electron is very localized, and dipolar interactions weak.




$$
\begin{aligned}
\mathcal{H} = \sum_{\langle ij \rangle} \Big[ & J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) \\
& + J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-) \\
& - \frac{i J_{z\pm}}{2} (\gamma_{ij}^* S_i^+ S_j^z - \gamma_{ij} S_i^- S_j^z + \langle i \leftrightarrow j \rangle) \Big], \quad (1)
\end{aligned}
$$

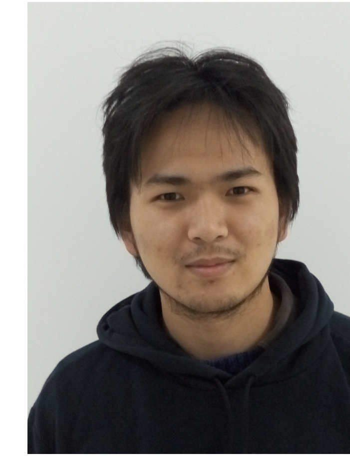
where $S_i^{\pm} = S_i^x \pm i S_i^y$, and the phase factor $\gamma_{ij} = 1, e^{i2\pi/3}, e^{-i2\pi/3}$ for the bond $ij$ along the $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$ direction (see Fig. 1), respectively. This generic Hamil-

The spin-1/2 XXZ model supports conventional order.

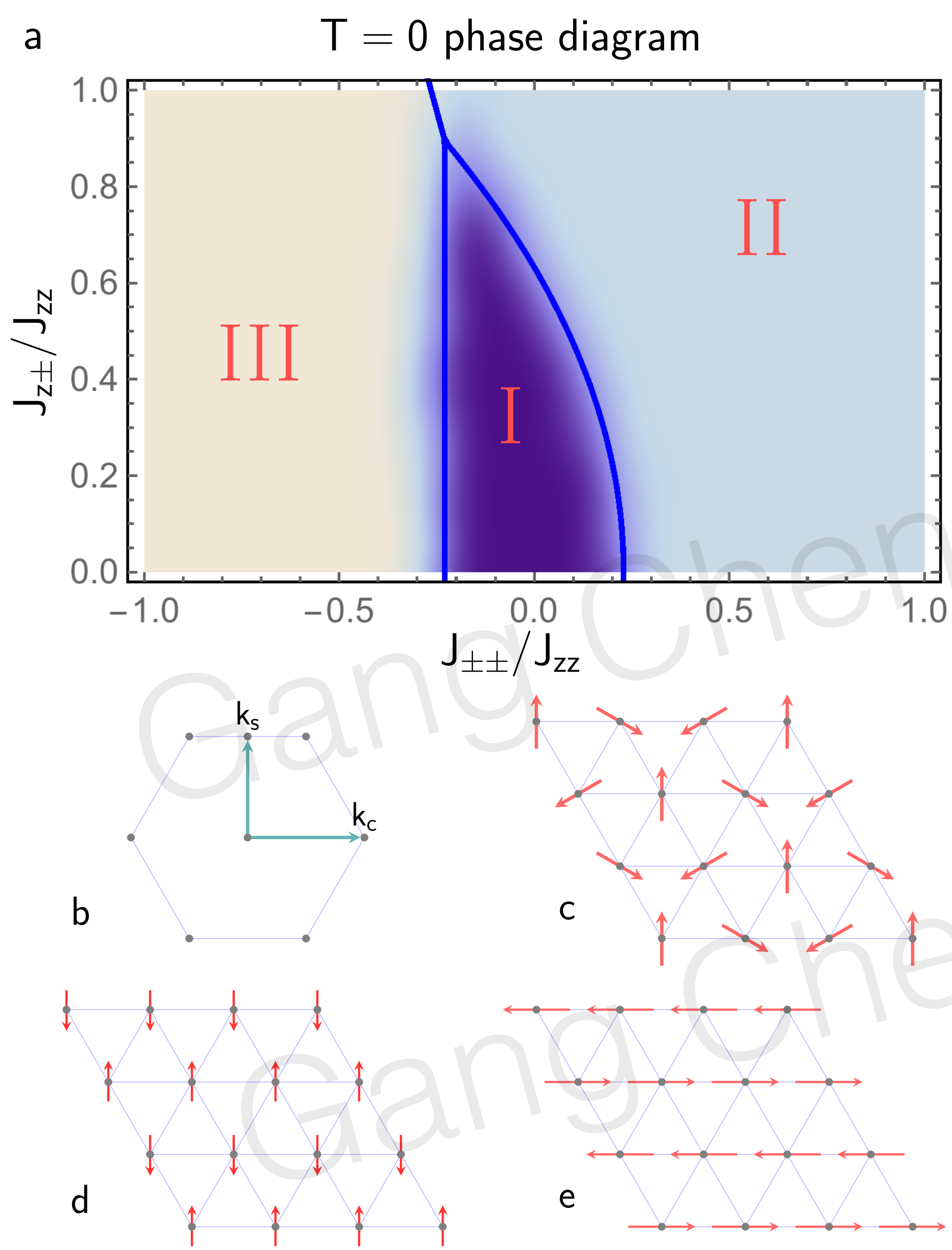
(Yamamoto, etc, PRL 2014)

Ga/Mg disorder may do something too. But not very clear at this stage.

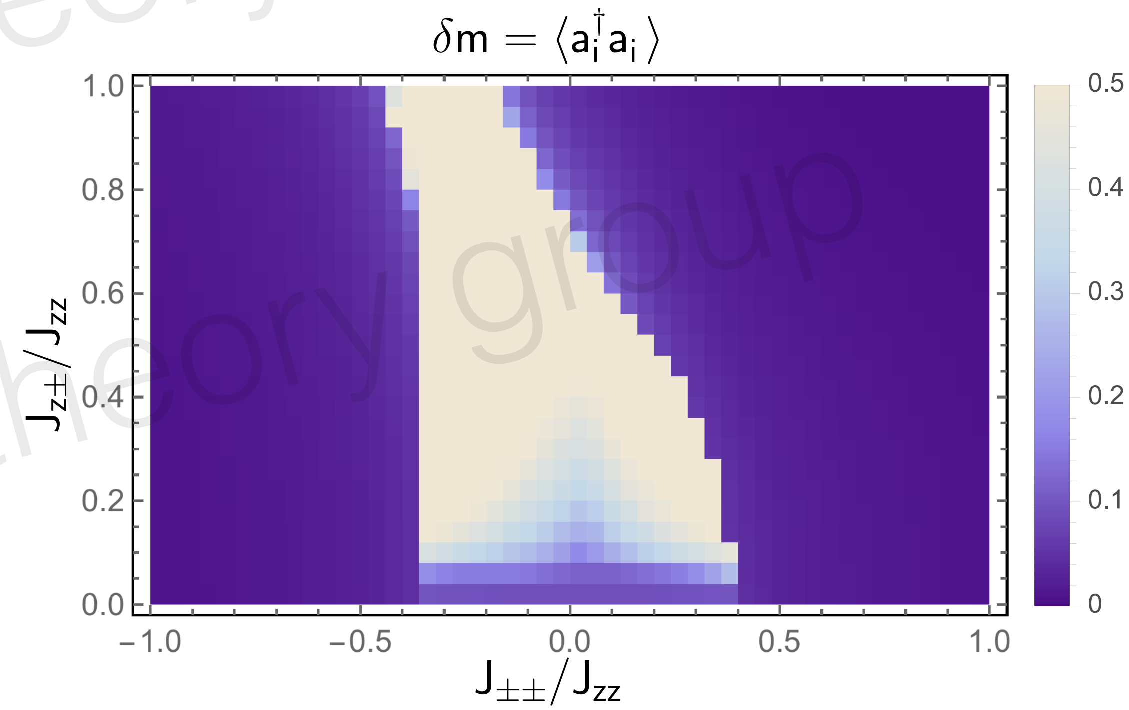
# Conservative treatment



Yao-Dong Li




mean-field phases



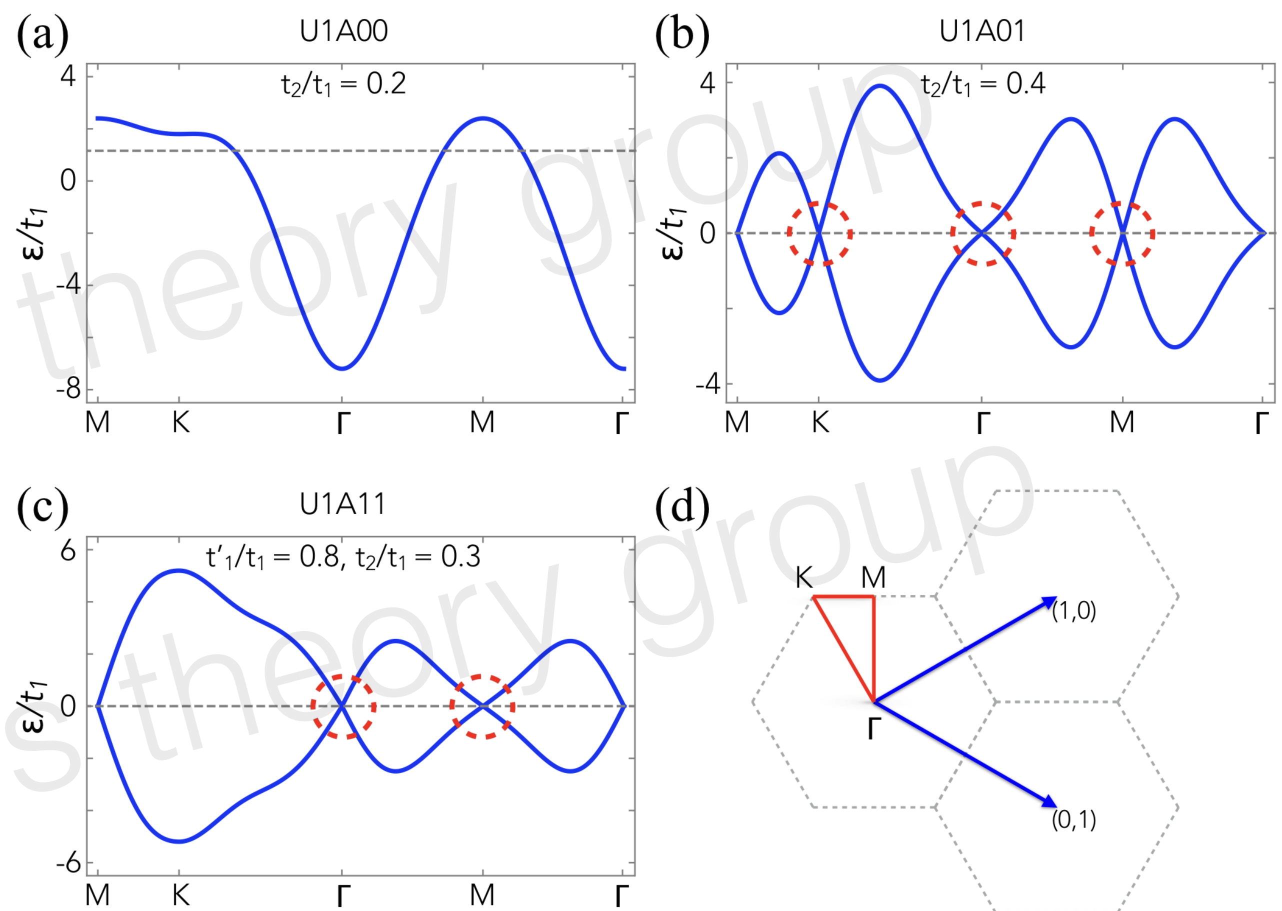

with quantum fluctuation

# Parton construction and PSG classification

$$\boldsymbol{S}_i = \sum_{\alpha\beta} \frac{1}{2} f^{\dagger}_{i\alpha} \boldsymbol{\sigma}_{\alpha\beta} f_{i\beta}$$

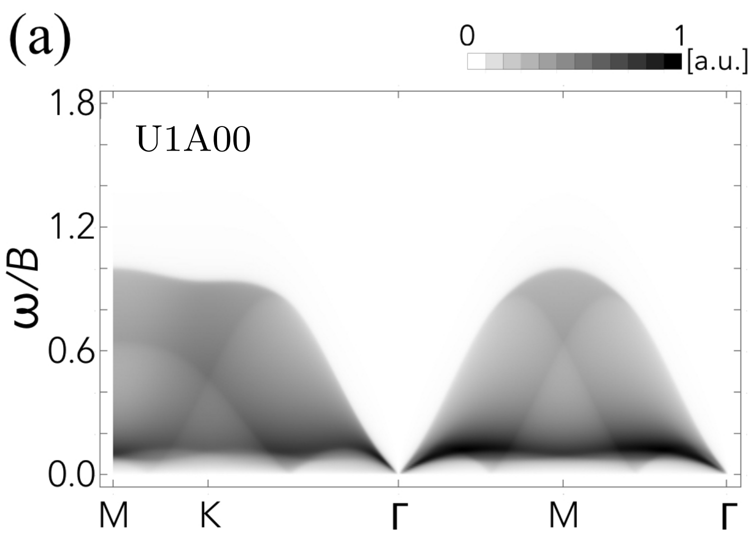
$$H_{\mathrm{MF}} = -\sum_{(\boldsymbol{r}\boldsymbol{r}')} \sum_{\alpha\beta} \left[ t_{\boldsymbol{r}\boldsymbol{r}',\alpha\beta} f^{\dagger}_{\boldsymbol{r}\alpha} f_{\boldsymbol{r}'\beta} + h.c. \right],$$

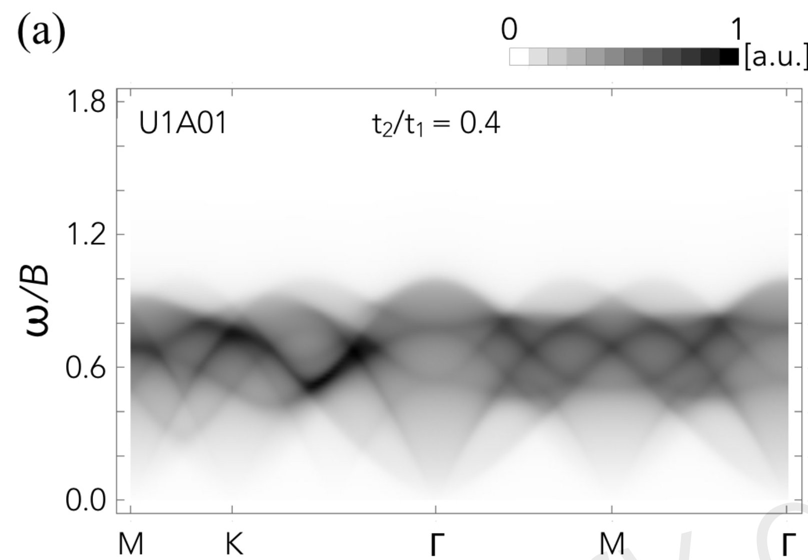
| U(1) QSL | $W_{\boldsymbol{r}}^{T_1}$ | $W_{\boldsymbol{r}}^{T_2}$ | $W_{\boldsymbol{r}}^{C_2}$ | $W_{\boldsymbol{r}}^{C_6}$ |
|---|---|---|---|---|
| U1A00 | $I_{2\times2}$ | $I_{2\times2}$ | $I_{2\times2}$ | $I_{2\times2}$ |
| U1A10 | $I_{2\times2}$ | $I_{2\times2}$ | $i\sigma^y$ | $I_{2\times2}$ |
| U1A01 | $I_{2\times2}$ | $I_{2\times2}$ | $I_{2\times2}$ | $i\sigma^y$ |
| U1A11 | $I_{2\times2}$ | $I_{2\times2}$ | $i\sigma^y$ | $i\sigma^y$ |

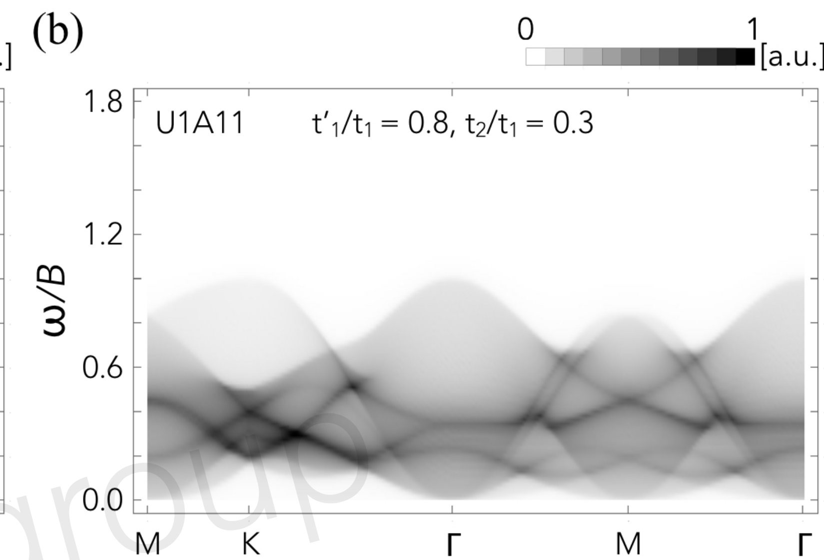


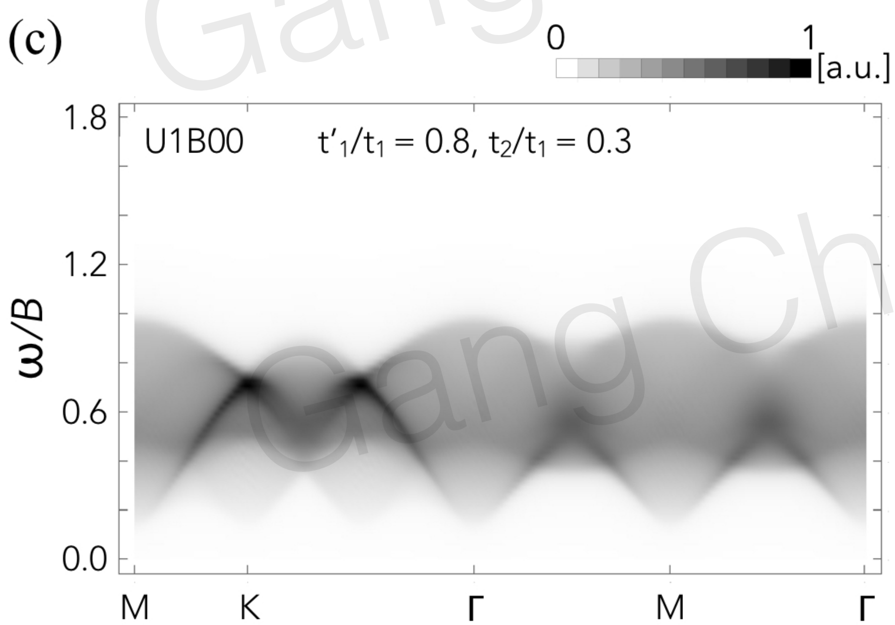



The U1A00 state is the spinon Fermi surface state that we proposed in Shen, et al, Nature.


(a)
0
1
[a.u.]
U1A00
1.8
1.2
0.6
0.0
ω/B
M
K
Γ
M
Γ


(a)
0
1
[a.u.]
U1A01
$t_2/t_1 = 0.4$
1.8
1.2
0.6
0.0
ω/B
M
K
Γ
M
Γ


(b)
0
1
[a.u.]
U1A11
$t'_1/t_1 = 0.8,\ t_2/t_1 = 0.3$
1.8
1.2
0.6
0.0
ω/B
M
K
Γ
M
Γ


(c)
0
1
[a.u.]
U1B00
$t'_1/t_1 = 0.8,\ t_2/t_1 = 0.3$
1.8
1.2
0.6
0.0
ω/B
M
K
Γ
M
Γ

# U1B state

# Summary

1. I present a theory for cluster Mott insulator that is motivated by $LiZn_2Mo_3O_8$.

2. I review the experiments and present some recent theory progress on YbMgGaO4.