Title: The theory of cluster Mott insulators: charge fluctuations and spin liquids

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Abstract: I will present recent theoretical work on cluster Mott insulators (CMI) in which interesting physics such as emergent charge lattices, charge fractionalization and quantum spin liquids are proposed. For the anisotropic Kagome system like LiZn2Mo3O8, we find two distinct CMIs, type-I and type-II, can arise from the repulsive interactions. In type-I CMI, the electrons are localized in one half of the triangle clusters of the Kagome system while the electrons in the type-II CMI are localized in every triangle cluster. Both CMIs are U(1) quantum spin liquids (QSL) in the weak Mott regime with a spinon Fermi surface and gapped charge excitations. In type-II CMI, however, the charge fluctuations lead to a long-range plaquette charge order that breaks the lattice symmetry, gives rise to an emergent charge lattice and reconstructs the mean-field spinon band structure of the underlying U(1) QSL. Such a reconstruction gives a consistent prediction of the "fractional spin susceptibility" that is observed in LiZn2Mo3O8. For the pyrochlore system, the CMI can further support a charge fractionalization with an emergent gauge photon in the charge sector in addition to the spin fractionalization in the spin sector.
The theory of cluster Mott insulators: charge fluctuations and quantum spin liquids

GANG CHEN


Collaborators: Hae-Young Kee, Yong-Baek Kim
Outline

- Motivation and introduction
- Cluster Mott insulator in 2D: theory and experiments
- Cluster Mott insulator in 3D: theory and experiments
- Summary
“Conventional”
Mott insulator and Mott transition

Triangular lattice Hubbard model at half filling

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

- **weak** Mott regime
  - Fermi liquid metal
  - U(1) quantum spin liquid with spinon Fermi surface
- **strong** Mott regime
  - 120-degree magnetic order
  - such a disordered regime is supported by various and different numerical studies

Many properties of spinon metal are “similar” to electron metal but with **subtle and important** differences!

Low EFT of QSL: spinon Fermi surface coupled with a fluctuating U(1) gauge field. It is a strong coupled theory, not resolved!
Organic triangular spin liquid?

3 organic candidates: k-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, κ-H$_3$(Cat-EDT-TTF)$_2$

a new one!

T. Isono et al, PRL 112, 177201 (2014)

Constant Pauli-like spin susceptibility at T->0 limit

magnetic torque measurement

also many other measurements
**Remark (on the mechanism NOT the properties of QSL):**
1. There is no sharp distinction between the charge fluctuations in the weak and strong Mott regimes, despite they are distinct spin phases.
2. Strong charge fluctuation in the weak Mott regime is a quantitative description.

**Question / observation:**
1. What if the charge fluctuation is very strong, and in the most extreme case, the charge sector forms a **quantum charge liquid**? Spin sector is even more likely to be in a QSL.
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 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.

My Goal of This Talk

1. Introduce the notion of cluster Mott insulator (they are interesting and they exist in nature, actually quite a lot, not studied)
2. Develop a new theoretical framework to understand the novel charge fluctuation and spin fluctuation
3. Apply to illustrative examples and explain the puzzling experiments.
One striking experiment on LiZn$_2$Mo$_3$O$_8$

Why striking and difficult?

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

Neither model works.

Nature Materials 2012
FQHE (Tsui, Stormer, and Gossard)
First exotic phenomenon known to us

Wen: all the electrons in the Laughlin state dance collectively under a rule that every electron moves 3 steps around one another.

What do electrons do in LiZn$_2$Mo$_3$O$_8$? Collective behaviours? Actually there are similarities.
LiZn$_2$Mo$_3$O$_8$ structure
Model

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

Minimal model allowed by symmetry

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

Large U alone cannot localize the electron. V1 and V2 are needed: because it is 4d orbital, and also to localize the electron in the clusters.
Generic phase diagram

\[ \frac{V_1}{t_2} \]

- type-I_d CMI
- type-II CMI
- FL-metal
- type-I_d CMI

\[ \frac{V_2}{t_1} \]

spin sector is spin liquid

no qualitative difference for different \( t_1/t_2 \)

snapshots of electron occupation in type-I CMI

\( V_2 \) is small, \( V_1 \) is large

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.
type-II CMI: **correlated** electron motion

3rd order process in type-II CMI

dimer resonating

\[ H_{QDM} \sim - \sum_{\text{dimer}} \left( |\downarrow\rangle \langle \downarrow| + |\uparrow\rangle \langle \uparrow| \right) \]

Charge sector is described by a compact U(1) gauge theory on the dual honeycomb lattice.
A new parton gauge construction to obtain proximate phases

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

charge-q_e 
spinless boson

charge-neutral 
spin-1/2 fermion

Original slave rotor construction works for the **conventional Mott insulator**

A new parton gauge construction is required for **cluster Mott insulators**.

**Gauge structure**

- \( U(1)_{sp} \)
  - one U(1) gauge field

- \( U(1)_c \times U(1)_{sp} \)
  - two U(1) gauge fields
A digression — two classes of parton-gauge theories

1. A formal way of introducing spinon + gauge

\[ S_i = \frac{1}{2} f_{i\alpha}^\dagger \sigma_{\alpha\beta} f_{i\beta} \quad \text{or} \quad S_i = \frac{1}{2} b_{i\alpha}^\dagger \sigma_{\alpha\beta} b_{i\beta} \]

and slave rotor representation
often blamed as "unjustified", often hard
to develop physical intuition

2. The microscopic model (at low/intermediate energies) already looks like a gauge theory
(e.g. Toric code . . .)

also in various contrived models
(Kitaev, Senthil, Motrunich etc)

\[ H = \sum_{\langle ij \rangle} \frac{E_{ij}}{2e} - t \sum_{\langle ij \rangle} a_{i\sigma}^\dagger e^{iA_{ij}a_{j\sigma}} - U \sum_i a_{i\uparrow}^\dagger a_{i\uparrow}^\dagger a_{i\downarrow} a_{i\downarrow} \]

Tchernyshyov's Kagome theory

Balents' quantum spin ice
A formalism

\[
H = -t_1 \sum_{\mu \neq \nu} \sum_{r \in u} l_{r, \nu}^{+} l_{r, \mu}^{-} f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \phi_{r, \mu}^{+} \phi_{r, \nu}\]

\[
- t_2 \sum_{\mu \neq \nu} \sum_{r \in d} l_{r, -\mu}^{+} l_{r, -\nu}^{-} f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \phi_{r, -\mu}^{+} \phi_{r, -\nu}\]

\[
+ \frac{V_1}{2} \sum_{r \in u} Q_r^2 + \frac{V_2}{2} \sum_{r \in d} Q_r^2, \quad (12)
\]

**Self-consistent mean field theory:** charge, spinon, gauge sectors

\[
H_{ch}^{u} = -\tilde{J}_1 \sum_{r \in u} \sum_{\mu \neq \nu} \phi_{r, \mu}^{+} \phi_{r, \nu} + \frac{V_1}{2} \sum_{r \in u} Q_r^2.
\]

\[
H_{ch}^{d} = -\tilde{J}_2 \sum_{r \in d} \sum_{\mu \neq \nu} \phi_{r, \mu}^{+} \phi_{r, \nu} + \frac{V_2}{2} \sum_{r \in d} Q_r^2.
\]

\[
H_{sp} = - \sum_{\mu \neq \nu} [t_1 \sum_{r \in u} f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} + t_2 \sum_{r \in d} f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma}].
\]

\[
H_A = - \sum_{\mu \neq \nu} [\tilde{K}_1 \sum_{r \in u} l_{r, \mu}^{+} l_{r, \mu}^{-} + \tilde{K}_2 \sum_{r \in d} l_{r, -\mu}^{+} l_{r, -\mu}^{-}].
\]

\[
\tilde{J}_1 = t_2 \langle l_{r, -\mu}^{+} \phi_{r, -\nu} \rangle \sum_{\sigma} \langle f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle, \quad r \in d,
\]

\[
\tilde{J}_2 = t_1 \langle l_{r, \mu}^{+} \phi_{r, \nu} \rangle \sum_{\sigma} \langle f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle, \quad r \in u,
\]

\[
\tilde{t}_1 = t_1 \langle l_{r, \mu}^{+} \phi_{r, \nu} \rangle \langle \tilde{f}_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle, \quad r \in u,
\]

\[
\tilde{t}_2 = t_2 \langle l_{r, -\mu}^{+} \phi_{r, -\nu} \rangle \langle \tilde{f}_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle, \quad r \in d,
\]

\[
\tilde{K}_1 = t_1 \sum_{\sigma} \langle f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle \langle \phi_{r, \mu}^{+} \phi_{r, \nu} \rangle, \quad r \in u,
\]

\[
\tilde{K}_2 = t_2 \sum_{\sigma} \langle f_{r \mu \sigma}^{\dagger} f_{r \nu \sigma} \rangle \langle \phi_{r, -\mu}^{+} \phi_{r, -\nu} \rangle, \quad r \in d.
\]
Generic phase diagram from **gauge** theory of charge sector

<table>
<thead>
<tr>
<th>Phases</th>
<th>$\langle \Phi_r \rangle$, $r \in u$</th>
<th>$\langle \Phi_r \rangle$, $r \in d$</th>
<th>$U(1)_c$</th>
<th>$U(1)_{sp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>FL-metal</td>
<td>$\neq 0$</td>
<td>$\neq 0$</td>
<td>Higgsed</td>
<td>Higgsed</td>
</tr>
<tr>
<td>type-I$_u$ CMI</td>
<td>$\neq 0$</td>
<td>$= 0$</td>
<td>Higgsed</td>
<td>Deconf</td>
</tr>
<tr>
<td>type-I$_d$ CMI</td>
<td>$= 0$</td>
<td>$\neq 0$</td>
<td>Higgsed</td>
<td>Deconf</td>
</tr>
<tr>
<td>type-II CMI</td>
<td>$= 0$</td>
<td>$= 0$</td>
<td>Confining?</td>
<td>Deconf</td>
</tr>
</tbody>
</table>

Sung-Sik Lee PRB 2008
type-II CMI: plaquette charge order via QDM

Moessner, Sondhi, Chandra 2001, also in several other numerical works (e.g. R. Melko)

$$H_{QDM} = -t\hat{T} + v\hat{V}$$
$$= -t (|\nabla\rangle\langle\nabla| + H.c.) + v (|\nabla\rangle\langle\nabla| + |\Delta\rangle\langle\Delta|).$$

Plaquette charge order:
- a local charge "RVB",
- a local collective behaviour!
- It is a quantum effect.
Lieb-Schultz-Mattis-Oshikawa-Hastings' theorem: apply to type-II CMI

- Tripled unit cell, host 3 electrons

BZ of type-I & type-II CMIs

Spinon band reconstruction

- Type-I CMI: no PCO
- Folding the lowest spinon band onto the BZ of type-II CMI

Increasing PCO in type-II CMI

Implication to susceptibility from bandwidth and filling
Another view: **spin state reconstruction**

3 spins act as one effective spin-1/2 and one pseudospin-1/2

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

spin $s=1/2$, pseudospin $T=1/2$, nonmagnetic

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

$$H_{KK} = \frac{J'}{9} \sum_{\mathbf{R}} \sum_{\mu=x,y,z} \left[ \mathbf{s}(\mathbf{R}) \cdot \mathbf{s}(\mathbf{R} + \mathbf{a}_\mu) \right]$$

$$\times \left[ 1 + 4\pi^\mu(\mathbf{R}) \right] \left[ 1 - 2\pi^\mu(\mathbf{R} + \mathbf{a}_\mu) \right]$$

$$\Theta_{CW} = -\frac{z_is(s+1)}{3} \left( \frac{J'}{9} \right) C = \frac{g^2 \mu_B^2 s(s+1)}{3k_B} \frac{N_\Delta}{3}$$

due to the reduced probability of spin interaction
Summary about LiZn$_2$Mo$_3$O$_8$

The emergence of PCO is the driving force of the 1/3 susceptibility anomaly. "Short-range quantum entanglement"
The spin GS of the system is probably a U(1) QSL with spinon Fermi surfaces."Long-range quantum entanglement"

![Diagram](image)

type-II CMI (PCO)

The crossover Kagome charge ice (KCI) regime is probably not sharply defined in LiZn$_2$Mo$_3$O$_8$ as it requires $V_2 >> T$ > ring hopping.

KCI: same Curie const as high-T one, slightly different Curie temperature
**Prediction A :** charge sector

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 Prof. Baskaran)

Disorders pin the charge density wave, broaden the phase transition.

W. L. McMillan PRB 1975
Further **prediction B**: low-T QSL

1. thermodynamics

   U(1) QSL with spinon Fermi surfaces

   \[ C_V \sim T^{2/3}, \quad \chi \sim \text{const} \]

   at very low temperature (<1K).

2. spectroscopic

   NMR: large density of low-energy spin excitations because of the reduced bandwidth

   \[ 1/(T_1 T) \propto D(E_F)^2 \]

   Neutron scattering: it would be nice to compare the prediction from the spinon band structure in future work. Single crystal data and better resolution are preferred.
What is type-I CMI?

Fig. 5. Inverse magnetic susceptibility as a function of temperature for Li₂InMo₃O₈. Data were taken under an applied field of 5000 Oe. Curie-Weiss fit is represented by the solid line.

no susceptibility anomaly!
Li₂InMo₃O₈ as a type-I CMI?
quantum spin liquid?
type-I CMI is a triangular lattice spin liquid

P Gall, etc. J Solid State Chem. 2013

M₂Mo₃O₈ (M=Mg, Mn, Fe, Co, Ni, Zn, Cd).
LiRMo₃O₈ (R=Sc, Y, In, Sm, Gd, Tb, Dy, Ho, Er, Yb) and many others.

Many materials mean many opportunities to discover new physics there.
ICE AGE 1: classical spin ice

ICE AGE 2: quantum spin ice

M. Gingras, R. Melko, M. Hermele, L. Balents,
M. Fisher, L. Savary, S. Lee, Y. Wan,
O. Tchernyshyov, G. Chen, Y.-P. Huang,........
C. Broholm, K. Ross, B. Gaulin........

So far, not confirmed experimentally!
partly because of very small energy scale.
Solution: d electrons, or others?

lots of materials
A little more about the motivation

1. Can we use other degrees of freedom to reveal quantum spin ice physics?

   electron = spin + charge + orbital (for condensed matter physicists only!)

     quantum spin ice (most famous !)
     quantum charge ice (the rest of the talk)
     quantum orbital ice (Gang Chen, unpublished)

2. Any other physical observables that do not have strong temperature constraint but still manifest the intrinsic properties of quantum spin ice?
   Not trivial ! (Gang Chen, working in progress !)
3D cluster Mott insulator

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

1/4 (or 1/8) electron filling

charge ice rule

Charge sector is a **Coulombic charge liquid (Quantum charge ice)**.
Charge fractionalization of the Coulombic charge liquid

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

\[ \frac{q_e}{2} \]

\[ c_{j\sigma}^\dagger \sim f_{j\sigma}^\dagger \Phi_{r}^\dagger \Phi_{r'} e^{i A_{rr'}} \]

fermionic spinon
Phase diagram and gauge description

FL metal $\rightarrow$ Cluster Mott Insulator $\rightarrow$ Coulombic charge liquid $\rightarrow$ QSL

Mott transition

<table>
<thead>
<tr>
<th>Phases</th>
<th>charge $\frac{q_e}{2}$ boson</th>
<th>$U(1)_c$</th>
<th>$U(1)_{sp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>FL-metal</td>
<td>Condensed</td>
<td>Higgsed</td>
<td>Higgsed</td>
</tr>
<tr>
<td>3D CMI</td>
<td>uncondensed</td>
<td>Deconf</td>
<td>Deconf</td>
</tr>
</tbody>
</table>

Here charge boson carries both $U(1)_c$ and $U(1)_{sp}$ gauge charge.
- (Inelastic) X-ray scattering measures U(1) gauge field correlation in the charge sector

$$\text{Im}[E_{-k,-\omega}^\alpha E_{k,\omega}^\beta] \propto [\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2}] \omega \delta(\omega - \nu |\mathbf{k}|),$$

$$E_{r+\frac{1}{2}e_\mu} \equiv L_{r,e_\mu} \frac{e_\mu}{|e_\mu|} = (n_{r+\frac{1}{2}e_\mu} - \frac{1}{2}) \frac{e_\mu}{|e_\mu|}$$

$$\langle E_{-k}^\alpha E_k^\beta \rangle \propto \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2}$$

emergent light in quantum charge ice!

N Shannon etc 2012,
L Savary etc 2012,
Gingras etc, 2007-now

Pinch points in equal-time charge structure factor at $T >$ ring hopping. “classical charge ice”
Mott transition: low-energy field theory

\[ \mathcal{L} = \mathcal{L}_\Phi + \mathcal{L}_f + \mathcal{L}_A + \mathcal{L}_a + \mathcal{L}_{bf} \quad (6) \]

\[ \mathcal{L}_\Phi = \left| \left[ \nabla - i(\mathbf{A}_\mu - \frac{a_\mu}{2}) \right] \Phi_1 \right|^2 + \left| \left[ \nabla - i(\mathbf{A}_\mu + \frac{a_\mu}{2}) \right] \Phi_{11} \right|^2 + m^2 \left( |\Phi_1|^2 + |\Phi_{11}|^2 \right) + u |\Phi_1|^4 + |\Phi_{11}|^4 + v |\Phi_1|^2 |\Phi_{11}|^2 \]

\[ \mathcal{L}_f = \bar{\psi}_\sigma (\nabla - \mu_f) \psi_\sigma + \frac{1}{2m_f} |(\nabla - i\mathbf{a}) \psi_\sigma|^2 \]

\[ \mathcal{L}_A = \frac{1}{4g_A^2} (\partial_\mu \mathbf{A}_\nu - \partial_\nu \mathbf{A}_\mu)^2, \quad \mathcal{L}_a = \frac{1}{4g_a^2} (\partial_\mu a_\nu - \partial_\nu a_\mu)^2 \]

\[ \mathcal{L}_{bf} = \lambda |\bar{\psi}_\sigma|^2 (|\Phi_1|^2 + |\Phi_{11}|^2). \]

\( \Phi_1, \Phi_{11} \) are charge bosons
\( \psi_\sigma \) are fermionic spinons
\( \mathbf{A}_\mu \) is \( U(1)_c \) gauge field
\( a_\mu \) is \( U(1)_{sp} \) gauge field

\[ z_c = 1, \quad z_s = 3 \]

different dynamical scalings for spin and charge

**FIG. 2.** The finite temperature crossover in the vicinity of the weakly first-order Mott transition.
Crossover in heat capacity and electric conductivity

1. Heat capacity crossover signals the \( z_s=3 \) dynamical exponent

Spinon-\( U(1)_{sp} \) gauge sector controls/dominates the thermodynamics

\[
C \approx \begin{cases} 
T \ln \ln 1 / T & T > |V - V_c|^{3/2} \\
\gamma_1 T \ln 1 / T & T < (V - V_c)^{3/2} \\
\gamma_2 T & T < (V_c - V)^{3/2}
\end{cases}
\]

- critical regime
- \( U(1) \) QSL
- FL metal

2. Electric resistivity signals the \( z_c=1 \) dynamical exponent

Ioffe-Larkin composition rule:

\[
\rho_c = \rho_f + (\rho_1^{-1} + \rho_1^{-1})^{-1}
\]

note: the resistivity gap in the Mott regime is single boson gap.

Mott transition itself is insulating. Crossover to metal in the metallic side.
Pyrochlore Mott insulators with fractional electron filling

usually associated with mixed valences

FIG. 2. Temperature dependence of the normalized electrical resistance $R_n = [R(T)/R(297 \text{ K})]$ of GaNb$_2$Se$_8$ (a) and GaTa$_2$Se$_8$ (b) at different pressures up to 28.5 GPa. The insets show the drop of $R_n$ at high pressures and low temperatures.

Superconductivity is actually interesting!

Both $U(1)_c$ and $U(1)_{sp}$ gauge fields can be higgsed down to $Z_2$ gauge fields.

The resulting CMI is $Z_2$ QCL + $Z_2$ QSL

Gang Chen, working in progress!

Metal-insulator transition:
but superconductivity intervenes!

M.M.Abd-Elmeguid etc, PRL 2004
**Question / observation:**

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**Summary**

1. I provide two specific examples about the physics of cluster Mott insulators.

2. There is a very interesting interplay between the charge and spin degrees of freedom in both 2D and 3D cluster Mott insulators.

3. Cluster Mott insulators are new physical systems that may host various emergent and exotic physics.