Search for quantum spin liquids in real materials

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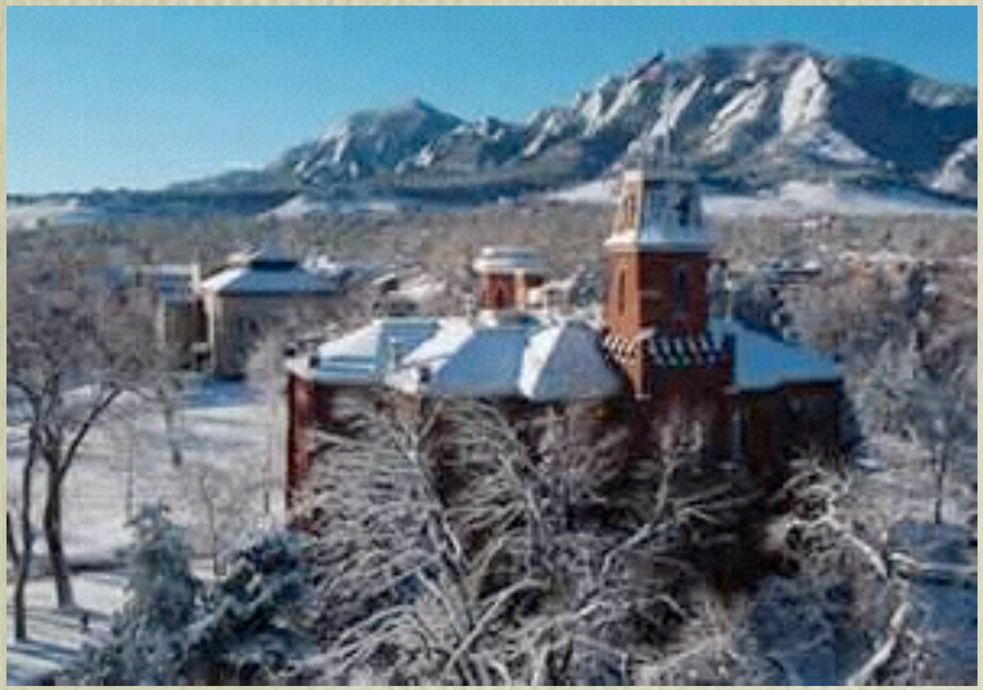


Outline

- Introduction to quantum spin liquids
- Materials' survey: failed and promising candidates for quantum spin liquids
- Summary and outlook

i will discuss one failed and one promising candidate for QSLs

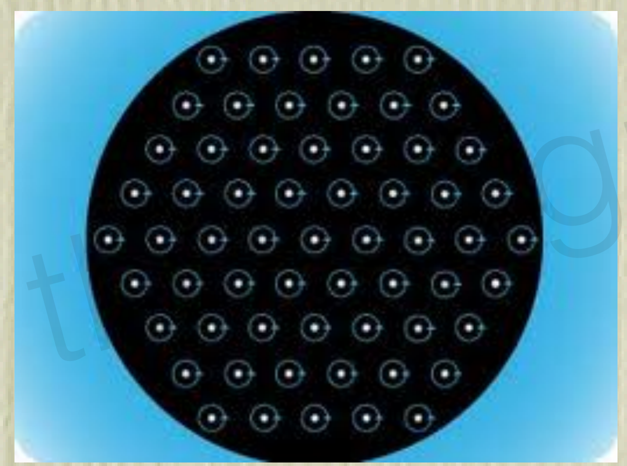
Physical systems usually order at low enough temperatures

e important goal of condensed matter is to understand different
ases of matter.

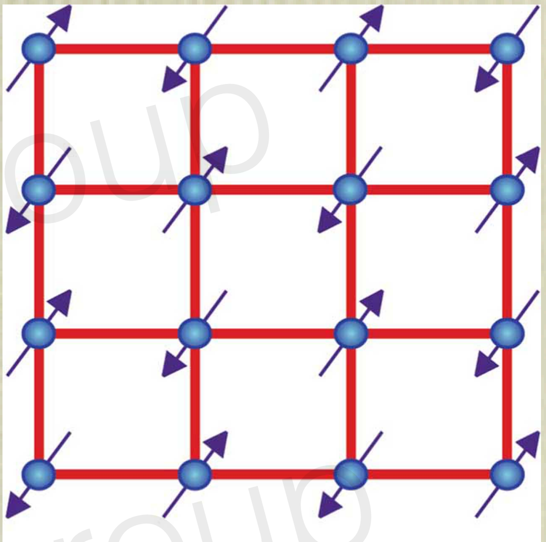
know the matter usualy orders in some way at low engout
nperature. e.g. in cyrstal, atoms devlops cyrstal order,
He-4, the sytsem devlops SF order,
spin ssytem ,spin develop magnetic order.

as pointed out by landau, all thses ordered phase can be
dsraodod from systemmtry breaking. Fore xmaple, He-4 SF breaks
internal U(1) symmetry ,and spin system breaks trnasitonl spin
on and time reveal .

toms
develops crystal order



He-4 liquid develops
superfluid order

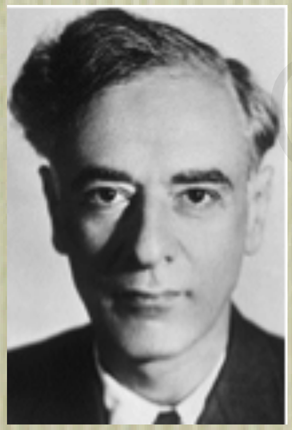


Spins develop
magnetic order

Break
translation symmetry
rotational symmetry
etc

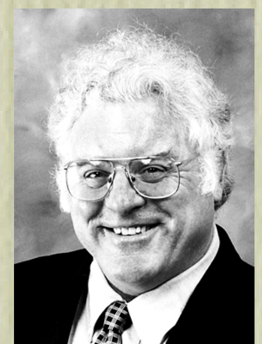
Break
an internal
U(1) symmetry

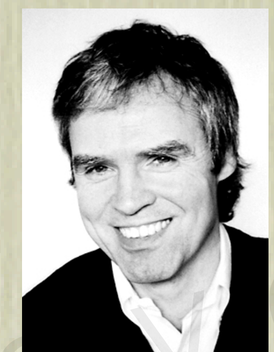
Break
spin rotation
translation symmetry
time reversal symmetry
etc

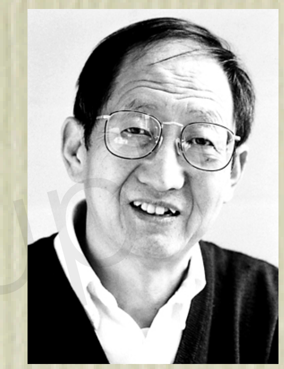


Landau

Modern exception: FQHE


Laughlin


Stormer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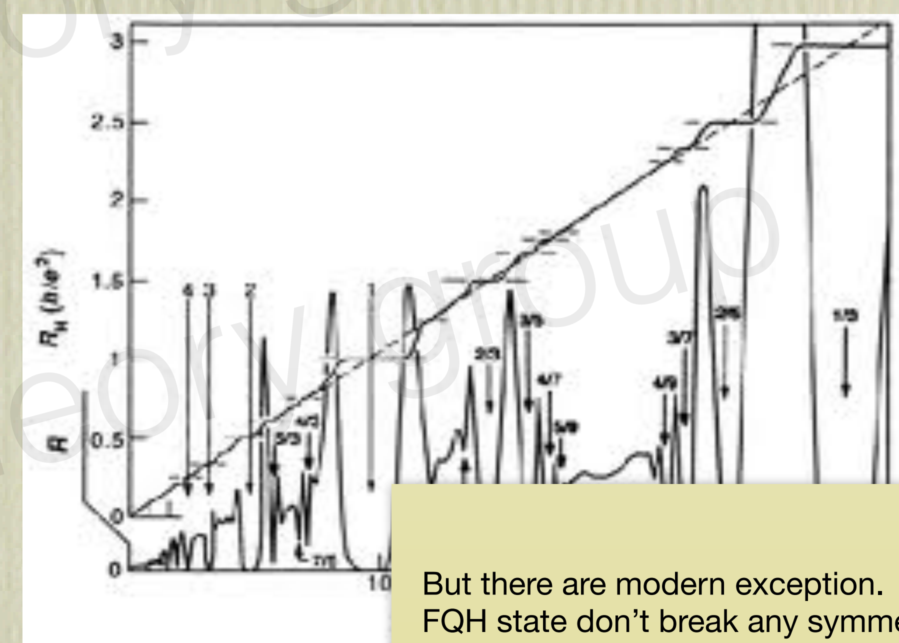


Gang Chen's theory group

* FQHE states don't break any symmetry and canno
characterized by symmetries.
* It has fractional excitations: excitations that carry
charge and statistics.
* Emergent (Chern-Simons) gauge fields.
* FQHE is a completely new phase of matter and ca
described within the Landau symmetry breaking par

But there are modern exception.
FQH state don't break any symmetry and cannot be cha
symmetry

FQH is very well understood in theory.

The key property of FQH is that, it
carries fractional excitation, that has fractional charge a
have emergent gauge fields.

Now we know FQH is a completely new phase matter.

In fact, it is an example of a new class of matter, This ne
matter cannot be described by landau symmetry breakin
Understanding this new class matter has attracted a lot
and experimental interest
in recent years.

Spin exceptions: quantum spin liquids

Motivation: Look for spin states/phases that
* do not break any symmetry (more precisely, symmetry is not essential to define the phase),
* do not have long-range spin order.
* have emergent gauge field and fractional excitation.

This novel phase is called “quantum spin liquid”.

QSL and a classical liquid (e.g. water):
e order but no long range order

snapshot of molecules
in liquid water

QSL is an example of this new class of matter that cannot be characterized by landau symmetry breaking theory.

QSLs don’t have spin long-range order,
have emergent gauge field and fractional excitation.

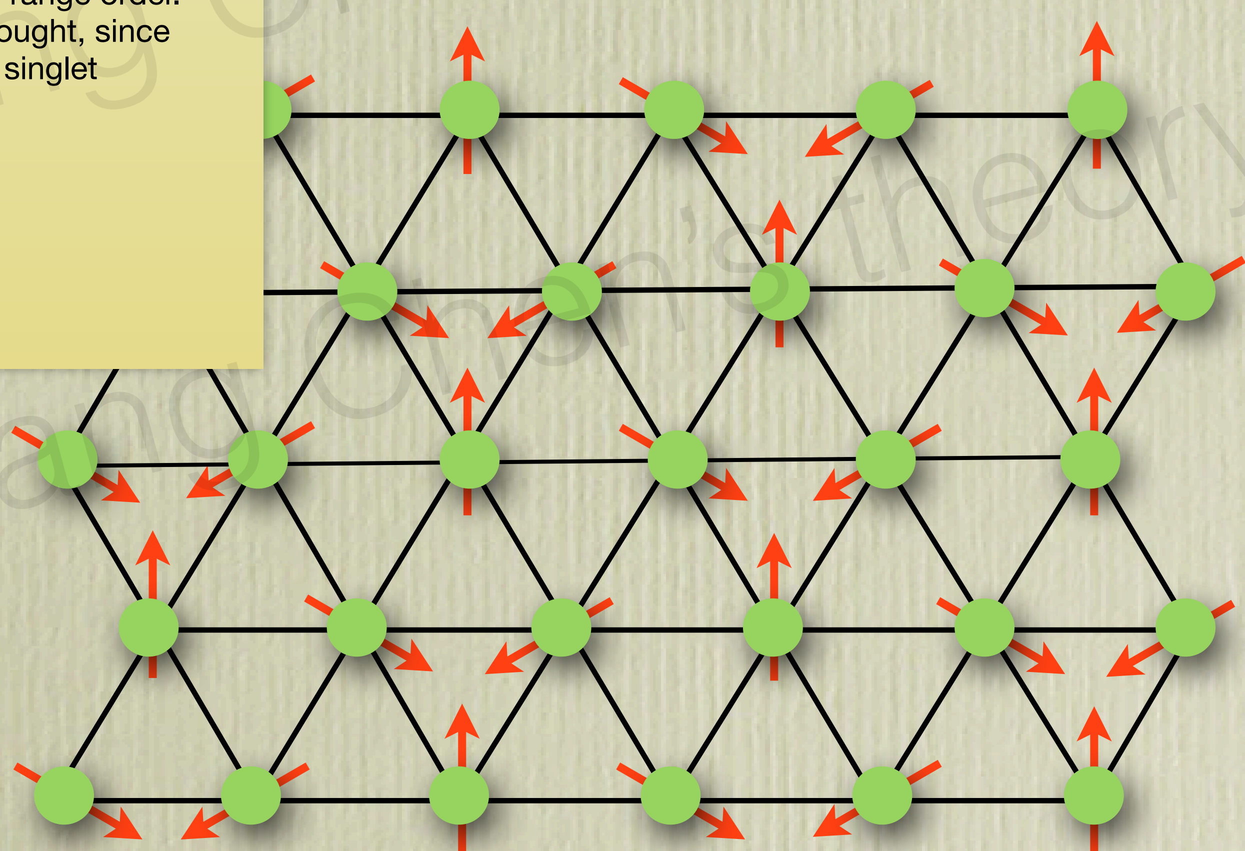
The name “liquid” comes from simple analogy with water liquid. In water liquid, there is density short range order, but there is no density short range order. Similarly, in spin liquid, there is spin short range order, but not spin long range order. This is the only analogy they have.

1973 Anderson's resonant valence bonds

Nearest-neighbor antiferromagnetic Heisenberg model on triangular lattice

$$H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \qquad J > 0, S = \frac{1}{2}$$

let me start with some history of QSL. 1973, anderson was thinking about the gs of NN AFM heisenberg model on triangular lattice with spin 1/2. now we know it has 120 degree long range order. but that time, he did not know. he thought, since spin is quantum, they tend to form a singlet

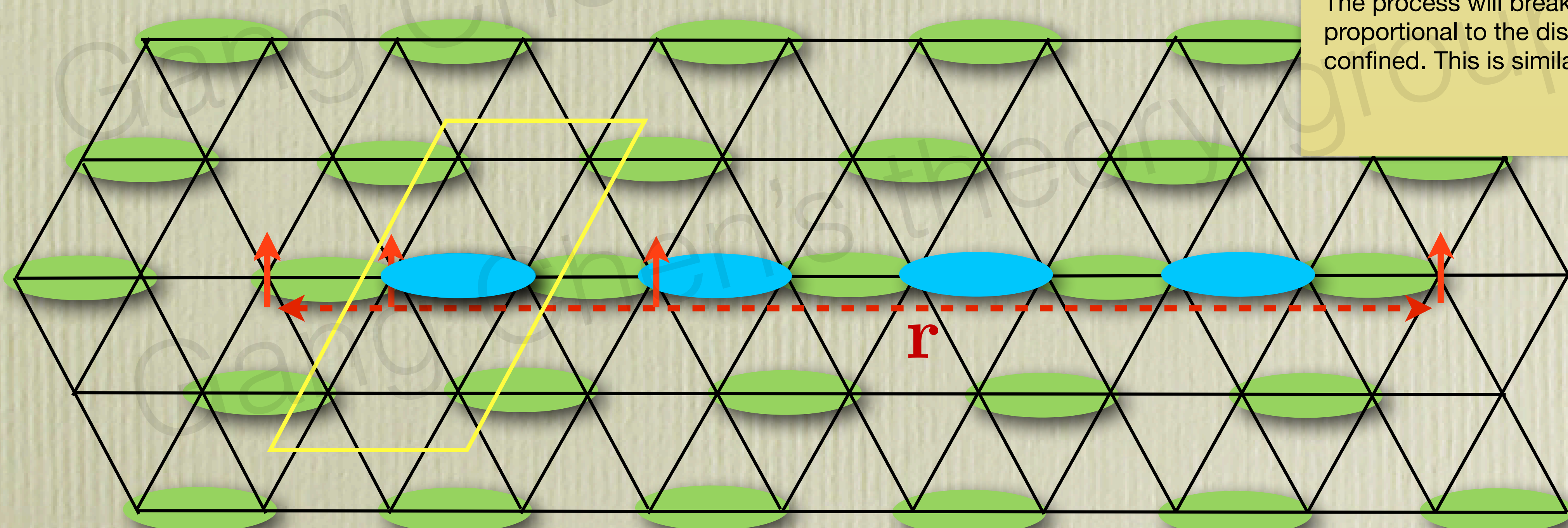


Long-range order
120-degree state

1973 Anderson's resonant valence bonds

dimer/singlet/ valence bond $= \frac{1}{\sqrt{2}}$ (

Valence bond solid: columnar dimer state (break tra



$E \propto |\mathbf{r}|$ "with a finite string tension", spinons (carrying spin-1/2) are linearly confined.

he thought, since spin is quantum, they tend to form a s
often called dimer or valence bond in literature. Let's fir
in which, the dimers develop a crystal order. If we break
create a triplit excitation.
The spin are carried by
two spinons (each spinon carry spin -1/2), and then sep

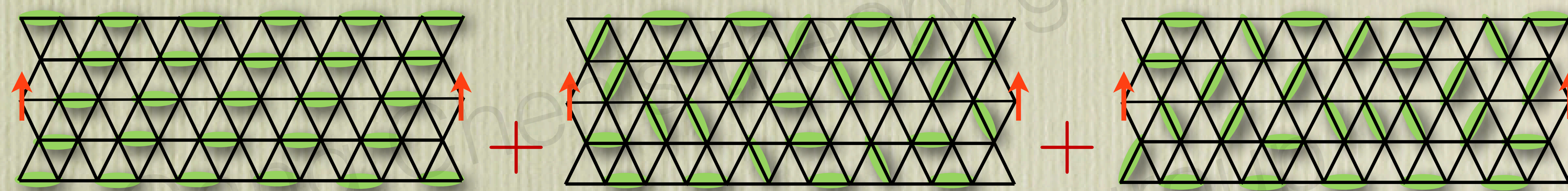
This process creates domain wall, it will cost a finite ene
spinons further, it will disrupt a lot of dimer in between.
be proportional to the separation between two spinons.
spinons are linearly confined. This is reminsticet of the c
confinement in qcd. One can imagine that the two spin
a string, and the string has a finite tension.

(one way to see this, is to compute the expectation valu
with respect to the excitated state.)

The process will break a lot of dimer in between, the en
proportional to the distance between the spinons. As a
confined. This is similar to the quark matter confinemen

1973 Anderson's resonant valence bonds

With a RVB state (Moessner, Sondhi, PRL2001),



+ all other dimer covering

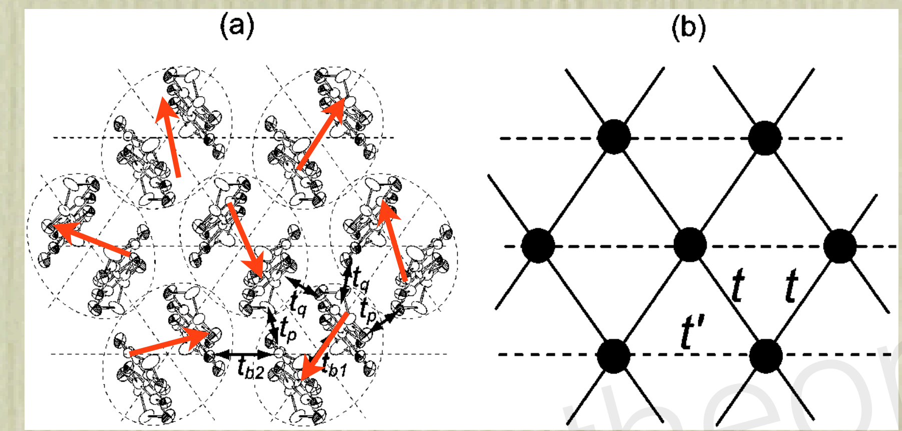
dimers are strongly fluctuating, the "string tension" vanishes and the spinons are *deconfined*.

1987, Anderson further proposed that RVB/spin liquid state might be relevant to high-Tc superconductor. Cuprates don't have such a QSL regime. The Mott insulator has AFM Neel order.

but if we have state which is a linear superpositions of all possible dimer

Kanoda

2003 Kanoda's organics



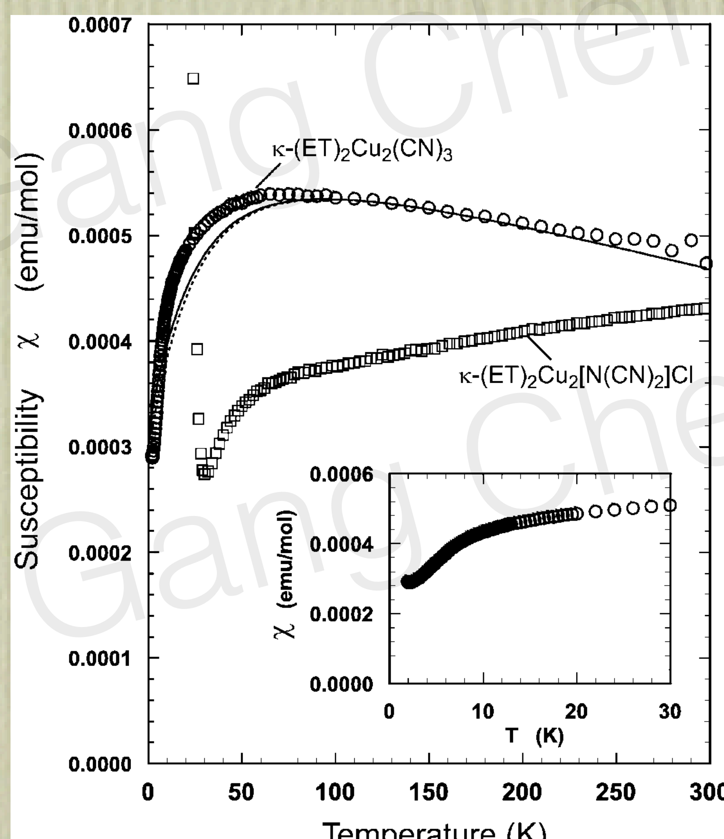
κ-$(ET)_2Cu_2(CN)_3$

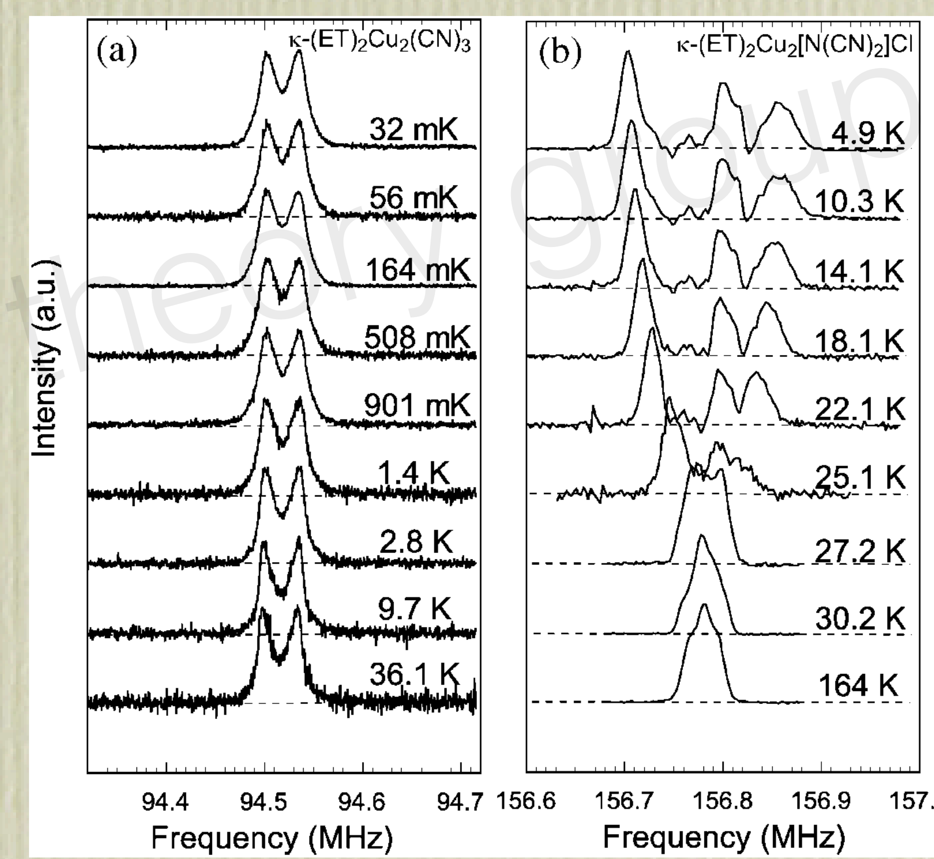
* molecular ET2 dimer
* triangular lattice, spi
* close to metal-insulat

For a long time, there is on

Experimental breakthrough
kanoda discovered an org
molecular dimer carries sp
the system is close to met
side.

Both spin susceptibility an
spin suscep is constant at

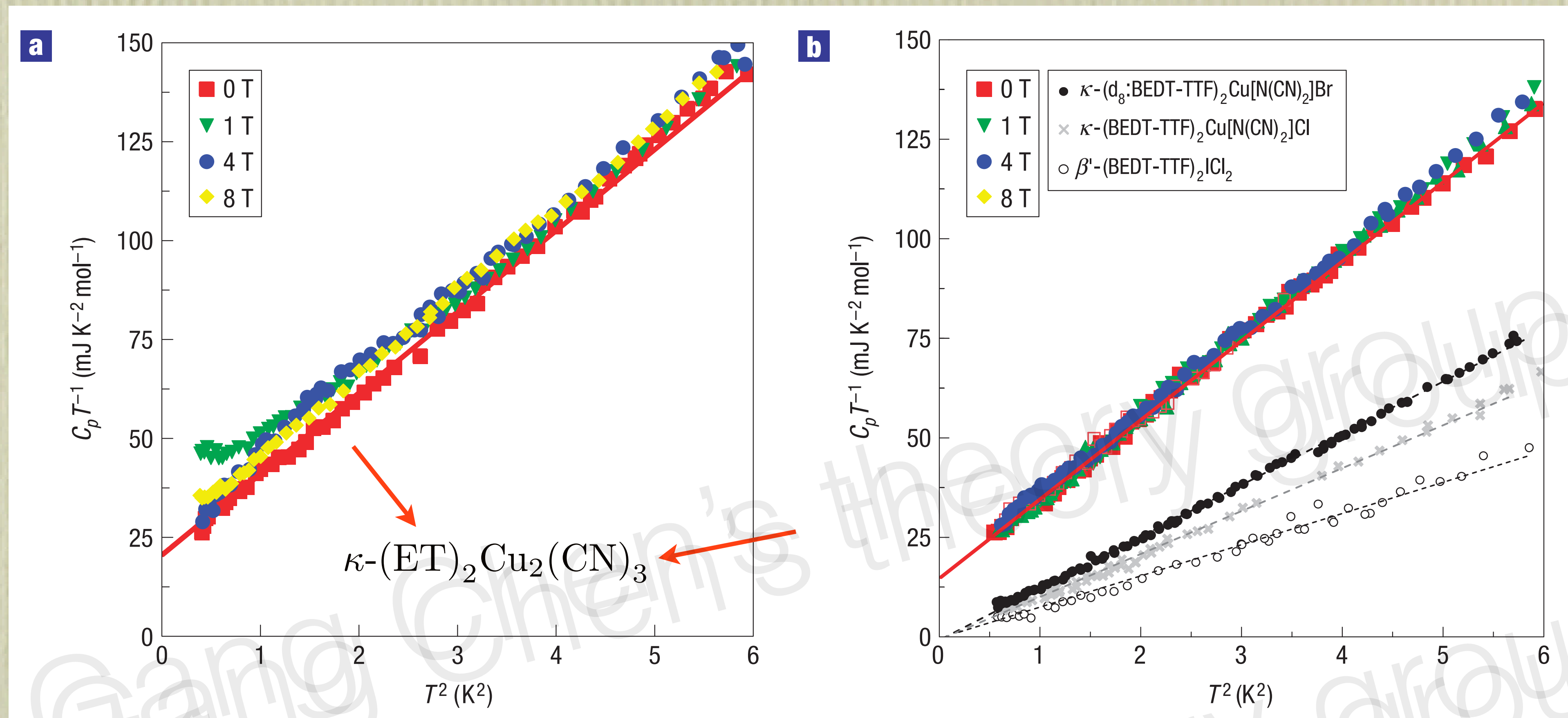




* No magnetic order down to 32mK
* constant spin susceptibility at zero temperature

Shimizu, etc, PRL, 91,107001

* Heat capacity



Yamashita, etc, Nature Physics, 4, 459, (2008)

$$\boxed{\begin{array}{ll} * & C_v \propto T \\ * & \chi \propto const \end{array}}$$

QSL with a spinon Fermi surface?

the heat capacity measurement obtain a linear-T dependence at low temperature limit.

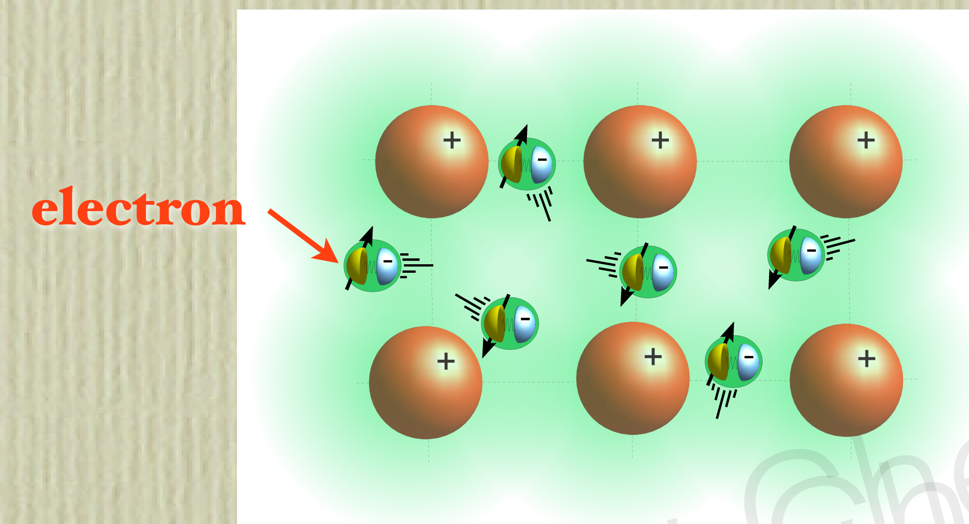
Constant spin suscep and linear-T heat capacity suggest the large density of low energy state. It is postulated to be spinon fermi surface.

QSL with spinon Fermi surface

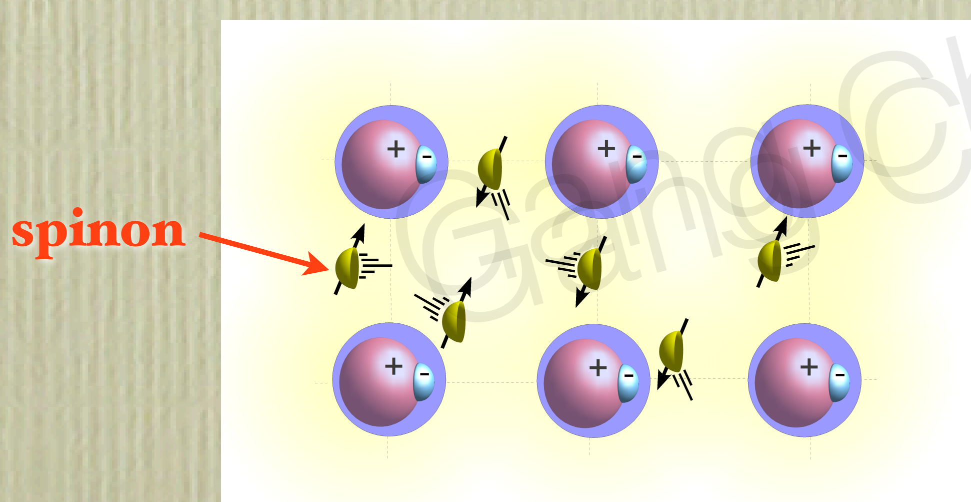
To describe the state, we split the electron operator int
the electron in this way enlarges the physical Hilbert sp
space is to project out the unphysical states. The othe
spinon and charge back into an electron. I am going to

In the metallic phase, charge boson is condensed, gau
in the mott insulator, boson are not condensed, the ga
charge are decofined. The charge is localized and the s

The low energy theory of this QSL is described by spin



Metal
"electrons swimming in the background of positively charge ions. "



Mott insulator: QSL (spinon metal)
"electron charge is pinned to the ion site while the spin still swims freely."

split the electron into cha

$$c_{\mathbf{r}\sigma} = b_{\mathbf{r}} f_{\mathbf{r}\sigma}$$

but glue them back to electron with gauge fields.

phases	charge boson	gauge field	spin-charge separation
metal	$\langle b_{\mathbf{r}} \rangle \neq 0$	higgsed	$c_{\mathbf{r}\sigma} = \langle b_{\mathbf{r}} \rangle f_{\mathbf{r}\sigma}$
Mott-QSL	$\langle b_{\mathbf{r}} \rangle = 0$	strongly fluctuating	Yes

Low-energy theory of Mott QSL
fermionic spinons coupled to U(1) gauge field.

theory: Motrunich, Lee, Lee, Senthil, etc

Many QSLs with many different low energy theories

QSLs	low-energy theory
U(1) QSL	QED (w/ monopole)
Dirac QSL	Dirac spinon couple to QED
nodal QSL	nodal spinon couple to Z_2 gauge
Majorana QSL	Majorana spinon couple to Z_2 gauge
Fermi surface QSL	spinon FS couple to U(1) or Z_2 gauge
……	……

Gang Chen's theory group

What's remarkable is
the low-energy theory has little to do with the microscopic theory, which is just some bosonic spin exchange model.

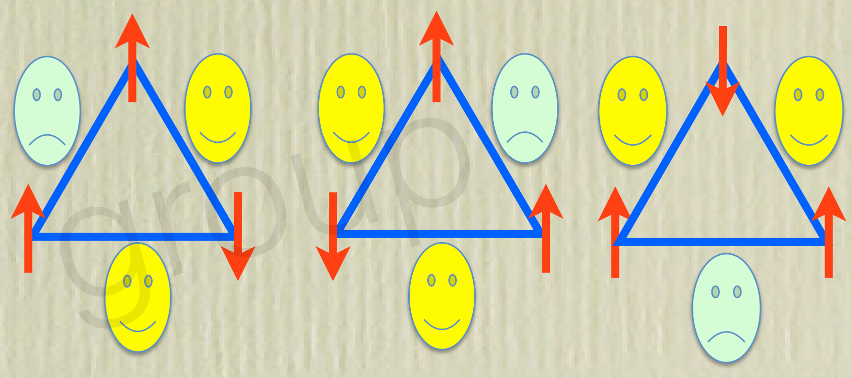
Wen, QFT of many-body systems

Now we know there are many different QSLs with quite different low energy theory. What's remarkable is that, t low energy theory has little to do with the microscopic theory, which is just some bosonic spin exchange mod

even though all come, the emergent low-E theory is completely different.

Where to search for QSL

- system with frustration (competing interaction)



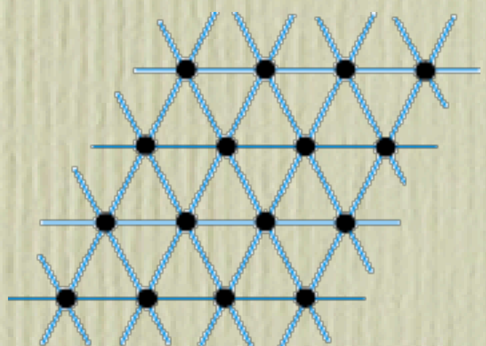
- quantum spins, e.g. S=1/2
- proximate to metal-insulator transition: large charge fluctuation
- others: strong spin-orbit coupling, quenched disorder, etc

Since qsl is stabilized by quantum fluctuation, we need to search among the systems with strong quantum fluctuation. THe following are some guidance.
The first one is frustrated system. frustration means competing interaction cannot be optimized simultaneously. A classical example is AFM ising model on triangular lattice. No matter how you arrange the spin, there is always one unhappy bond.

the second one is system with small spin moment, the most quantum one is spin 1/2.

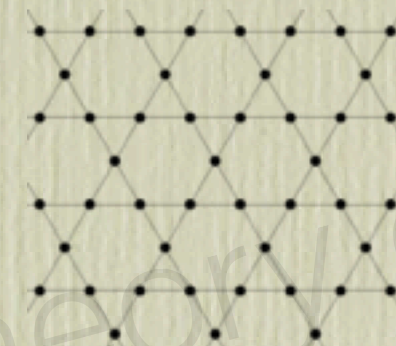
the third is system near metal-insua xtion, where the charge fluct are

Many candidate materials now!



triangular lattice

~~Cs_2CuCl_4~~
~~$NiGa_2S_4$~~
He-3 layers on graphite
κ-$(ET)_2Cu_2(CN)_3$
$EtMe_3Sb[Pd(dmit)_2]_2$
$Ba_3CuSb_2O_9$
$Ba_3NiSb_2O_9$
$LiZn_2Mo_3O_8$

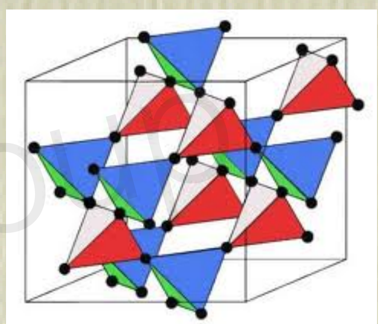


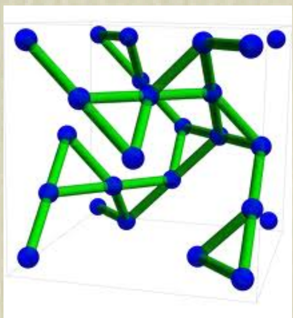
kagome lattice

$ZnCu_3(OH)_6Cl_2$
$Cu_3Zn(OH)_6Cl_2$
~~$Cu_3V_2O_7(OH)_2$~~
$BaCu_3V_2O_3(OH)_2$

Proximate to Mott transitions

κ-$(ET)_2Cu_2(CN)_3$
$EtMe_3Sb[Pd(dmit)_2]_2$
~~Na_2IrO_3~~
$Na_4Ir_3O_8$
IrO_2





Strong spin-orbit coupling

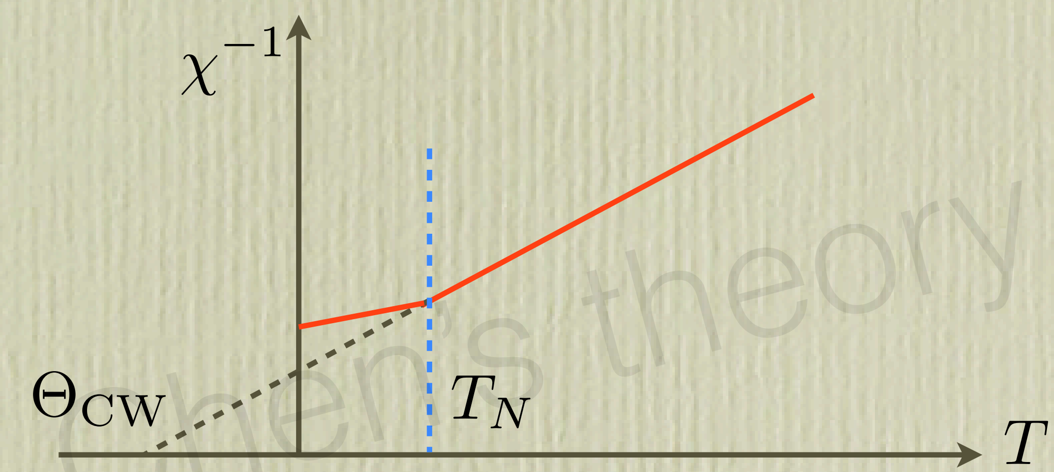
~~Na_2IrO_3~~
$Na_4Ir_3O_8$
IrO_2
Ba_2YMoO_6
$Yb_2Ti_2O_7$
$Pr_2Zr_2O_7$
~~$FeSc_2S_4$~~

since 2003, experimentalists have discovered many QSL candidates.

Some of them have already been ruled out to be QSL.

* Some common phenomenology

- No ordering down to lowest measurable temperature, i.e. a large frustration parameter $f \equiv \frac{|\Theta_{\mathrm{CW}}|}{T_N}$



- Constant $T = 0$ spin susceptibility.
- Power-law heat capacity. $C_v \sim T^{\alpha}$

* Challenge

Many materials have very similar phenomenology.
Are they all quantum spin liquid?
How to connect the experiments to the theory?
We need mutual feedback between theory and experiment.

There are some common phenomenology of these QSL can
high temp, the spin suscp obey curie-weiss law. There is no
down to lowest temp.
It is often useful to introduce an empirical experimental para
frustration parameter, which is given by the ratio between C
ordering temp. In real QSL, f is infinity. In exp, we cannot re
temperature.
For instance, if CW is 100K, and lowest temp in exp is 1K, th
pram is greater than 100.

the spin sucept is often const at low temp. the heat capacity
in temp at low T.

Although many materials have similar pheno, it does not me
QSL.
this is like biology.
you know,
whales and dolphin look like fish, but they are not fish.
they are more interesting and advanced creattures.

The challenge is to tell which material is qsl and the type of
tell which material is not qsl and if it is not, what is it. The lat
careful examination of the experiments and material.

failed examples

candidate materials	spin	lattice	f	explanation
Cs_2CuCl_4	1/2	triangle	f~8	dimensional reduction
$Cu_3V_2O_7(OH)_2$	1/2	kagome	f>30	magnetic order
Na_2IrO_3	1/2	honeycomb	f~10	magnetic order
$NiGa_2S_4$	1	triangle	f~10	spin nematics
$FeSc_2S_4$	2	diamond	f>900	spin-orbital singlet
....				

Balents, Nature 2010

If any one is interested in any of the materials, we can talk after the talk.

promising ones

Candidate QSL	spin	lattice	susceptibility	Cv	f	possible QSL
kappa-$(ET)_2Cu_2(CN)_3$	1/2	triangle	constant	$C_v \sim T$	$f>1000$	spinon FS
$EtMe_3Sb[Pd(dmit)_2]_2$	1/2	triangle	constant	$C_v \sim T$	$f>1000$	spinon FS
$ZnCu_3(OH)_6Cl_2$	1/2	kagome	------	------	$f>1000$	Dirac QSL
$Cu_3Zn(OH)_6Cl_2$	1/2	kagome	constant	$C_v \sim T$	$f>475$	Majorana QSL
$Na_4Ir_3O_8$	1/2	hyperkagome	constant	$C_v \sim T$	$f>300$	U(1) QSL with FS
$Pr_2Zr_2O_7$	1/2	pyrochlore	constant	------	$f>70$	U(1) (quantum spin ice)
…….						

Experimental work by C. Broholm's group, H. Takagi's group, Kanoda's group, Y. Lee's group, Lhuillier's group.

In the following part of the talk, I will discuss two materials, one is probably not QSL but still very intersting. The other is probably a QSL.

The first work is in collaboration w/

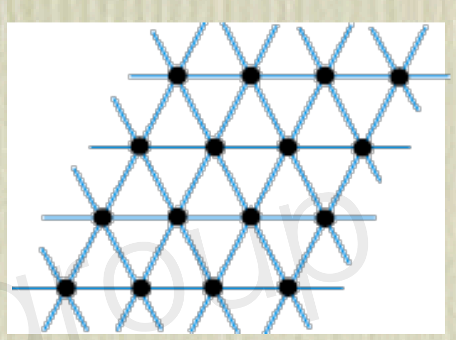
The second work is in collaboration w/

Plan

- A probably failed but very interesting QSL candidate: 6H-B-$Ba_3NiSb_2O_9$

Collaborators: * Michael Hermele (Univ of Colorado Boulder)
* Leo Radzihovsky (Univ of Colorado Boulder)

Refs: GC, Hermele, Radzihovsky, PRL 109, 016402, 2012

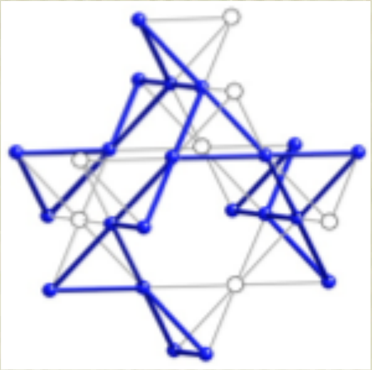


triangle

- A very promising QSL candidate: $Na_4Ir_3O_8$

Collaborators: * Yong-Baek Kim (Univ of Toronto)

Refs: GC, Kim, unpublished



hyperkagome

Gang Chen's theory group

last year, Dr. Balicas from high magnetic lab discovered three different structures of this material under high pressure. It is a spin-1 system. These are the exp data.

3NiSb2O9-a spin-1 AFM

Balicas

J. Cheng, etc, PRL, 107,197204 (2011)

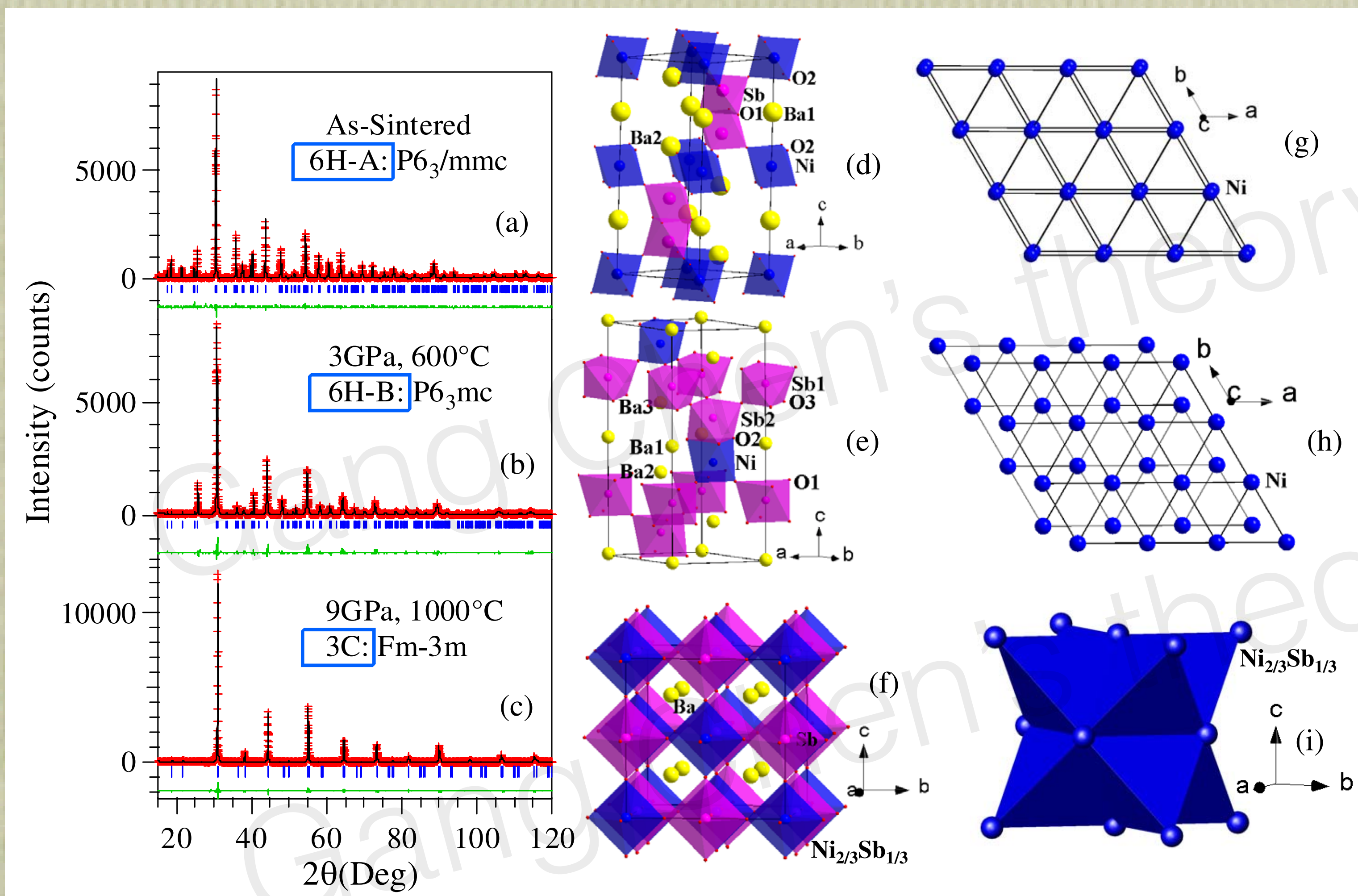
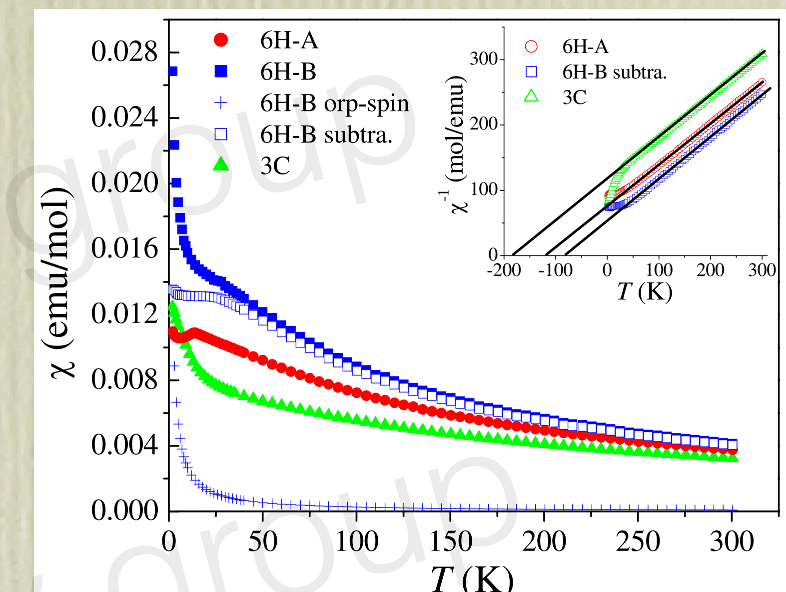
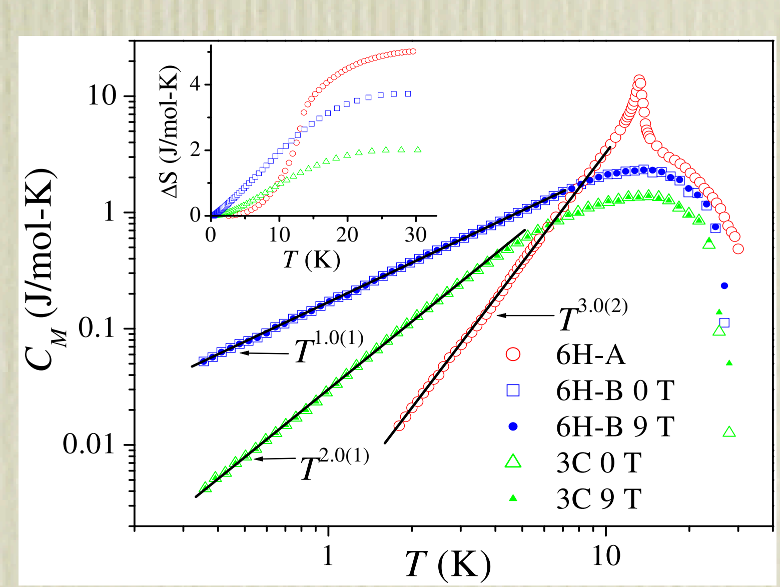


FIG. 1: Powder XRD patterns (crosses) at 295 K for the $Ba_3NiSb_2O_9$ polytypes: (a) 6H-A, (b) 6H-B, and (c) 3C. Solid curves are the best fits obtained from Rietveld refinements using FullProf. Vertical marks indicate the position of the Bragg peaks, with the curves at the bottom showing the difference between the observed and the calculated intensities. Schematic crystal structures for the $Ba_3NiSb_2O_9$ polytypes: (d) 6H-A, (e) 6H-B, and (f) 3C. Magnetic lattices composed of Ni^{2+} ions for the $Ba_3NiSb_2O_9$ polytypes: (g) 6H-A, (h) 6H-B, and (i) 3C.

three high-pressure structures

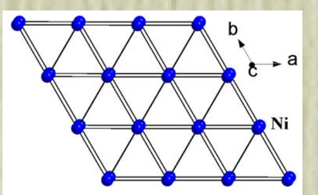


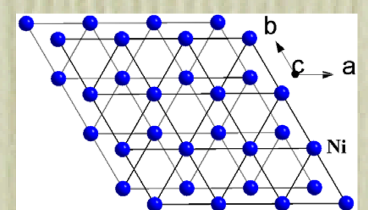
spin susceptibility

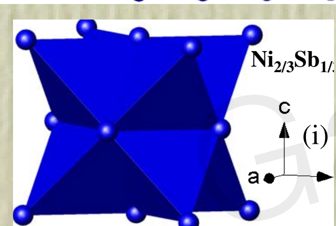


heat capacity

Summary of experiments on Ba3NiSb2O9







spin-1 system	structure	T=0 susceptibility	heat capacity	explanation
6H-A	A-A stacking	const	$C_v \propto T^3$	magnetic order
6H-B	A-B stacking	const	$C_v \propto T$	QSL?
3C	FCC with 1/3 dilution	const	$C_v \propto T^2$	QSL?

Other's work

Serbyn, Senthil, P. Lee, PRB 84,180403, 2011

* Z2 QSL with spinon Fermi surface
* Rely on ***large and positive*** biquadratic exchange $K(\mathbf{S}_i \cdot \mathbf{S}_j)^2$

Xu, Wang, Qi, Balents, Fisher, PRL 108, 087204, 2012

* Z2 QSL with quadratic dispersive spinons
* Spinon MFT only has pairing
* Energetically favorable with ***negative*** ring exchange

1

$\big[(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3$

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

Let me summary his experimental results in this tabl
all the traingular layers are identitical. 6H-B has A-E
shifted like graphite. 3C is a FCC lattice with 1/3 site

All the spin suscpt is constant at zero temp limit. The
exponent. 6H-A has AF LRO, T^3 heat capacity is d
hard to understand. Here I will focus on the 6H-B ma

There are already two other works on 6H-B material.

the first is by Mit group. senthil and patrick are prop
's gutts feeling when they see const and linear T.
But this state needs a large and postive biquaer , an
negative.

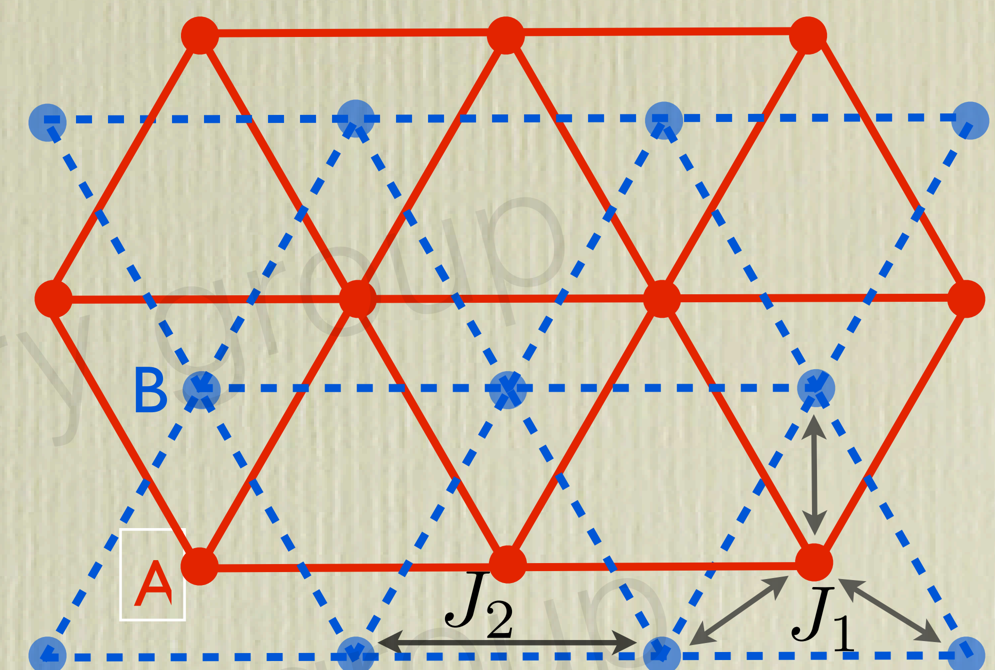
the other is from SB, by CenKe Xu, L Balent, M Fish
spinon dispersion.
their state requires the a negative ring exchange. Bu

Minimal model

* Exchange interaction

$$\mathcal{H}_{\text{ex}} = J_1 \sum_{\langle ij \rangle \in \text{AB}} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle ij \rangle \in \text{AA or BB}} \mathbf{S}_i \cdot \mathbf{S}_j,$$

Exchange is already frustrated, the magnetic order would be very weak if there is any.



Exchange between 2 neighboring triangular layers

* Single-ion anisotropy allowed by symmetry and S=1

$$\mathcal{H}_{ani} = D \sum_i (S_i^z)^2$$

* Single-ion anisotropy would suppress the weak magnetic order and favors a trivial quantum paramagnetic state

$$\prod_i |S_i^z = 0\rangle$$

* Expect: $J_1, J_2 \gg D$ weakly ordered state

$J_1, J_2 \ll D$ quantum paramagneti

Here, we explain the seemingly QSL phenomenology by a conventional mecha
linear-T heat capacity is quite challenging as it requires a constant DOS at low
to have a constant low energy DOS without introducing spinons. Here I provide
such an explanation without introducing spinons.

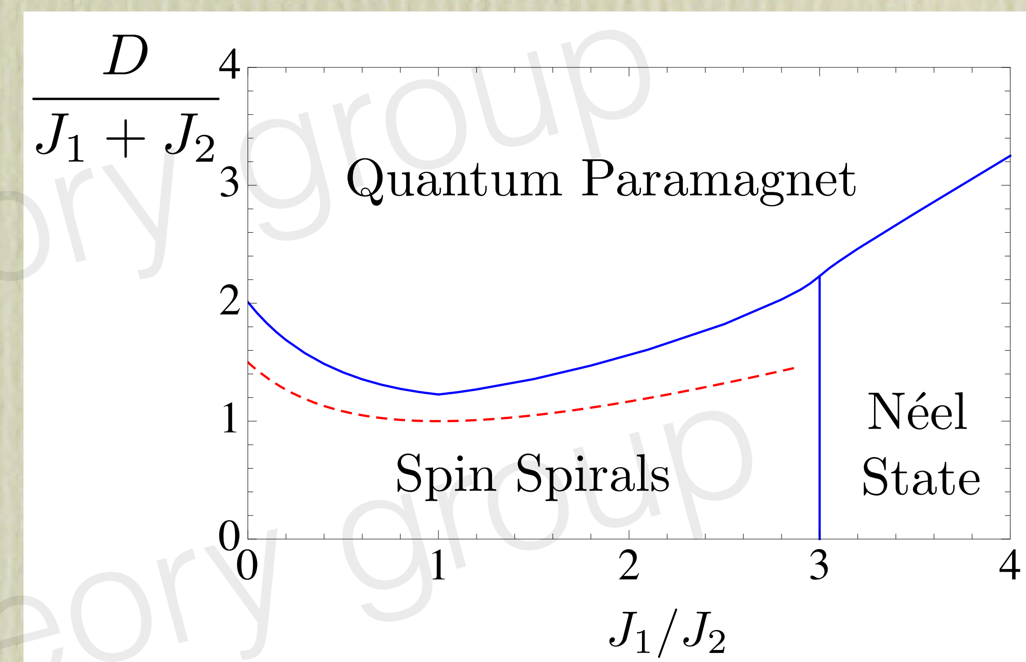
In our theory, we consider a minimal model. The first interaction is the spin exc
both intra and inter layer exchange. This exchange is frustrated, if it is orders, t
is going to be small.

Then we add single-ion anisotropy allowed by symmetry. If anisotropy is large,
paramagnetic state is favored with the Sz=0 at every state.

So we expect, when exchange dominant, we obtain weakly ordered state, whe
anisotropy is dominant, we obtain a quantum paramagnetic state.

Map spin to rotor

spin	rotor
S_i^z	n_i(integer valued)
S_i^+	$\sqrt{2}e^{i\phi_i}$
$[S_i^+, S_i^-] = 2\delta_{ij}S_i^z$	$[\phi_i, n_j] = i\delta_{ij}$
XY spin order, $\langle S^+ \rangle \neq 0$	condensed boson, $\langle e^{i\phi} \rangle \neq 0$
quantum paramagnet, $S^z = 0$	uncondensed boson, $\langle e^{i\phi} \rangle = 0$



Phase diagram
At dashed curve, quadratic dispersion is obtained, so is constant DOS.

Equivalent rotor Hamiltonian

$$H_{\rm rotor} = \frac{1}{2}\sum_{ij} J_{ij}\left[2\cos(\phi_i - \phi_j) + n_i n_j\right] + \sum_i D n_i^2$$

This is essentially an extended boson-Hubbard model and can be solved by standard methods.

To solve our minimal model, we simplify the problem and ma
rotor variable .
Sz map to rotor, S+ map to this phase variables. Spin orderin
corresponds the boson condensation of rotor. The quantum p
corresponds to a uncondensed rotor.

This is the equivalent rotor Hamiltonain. This is essentially a e
Bose-Hubbard model and can be solved by standard metho

Both the phase diagram and low energy spin excitation can b

Prediction

* Spin susceptibility

field in the plane, $\chi_0^{\perp} = \dfrac{2\mu_0(g\mu_B)^2}{D + 12(J_1+J_2)}$

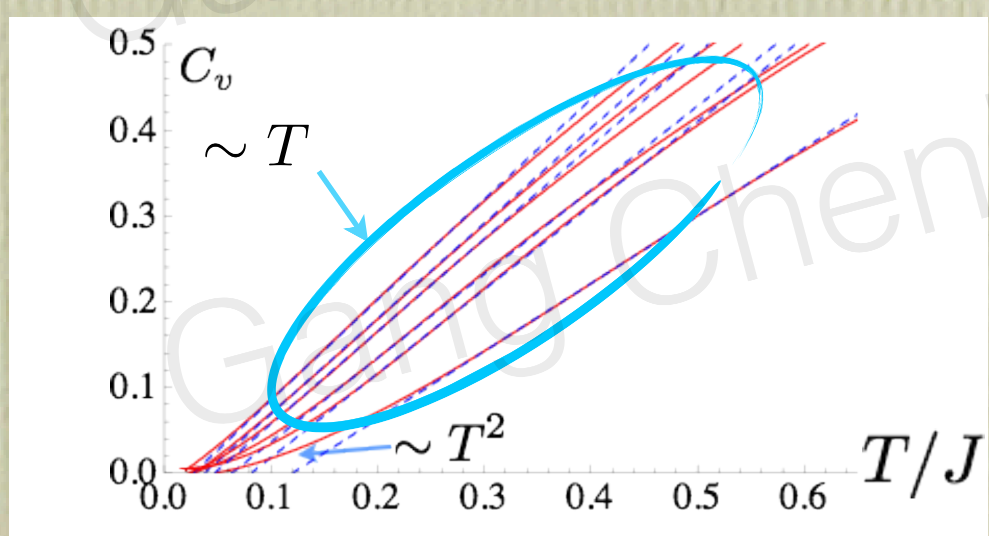
field normal to the plane, $\chi^z = 0$

Powder averag

$\chi_{av} = \dfrac{2}{3}\chi_0^{\perp}$

Experimental check: susceptibility on single-crystal sample.

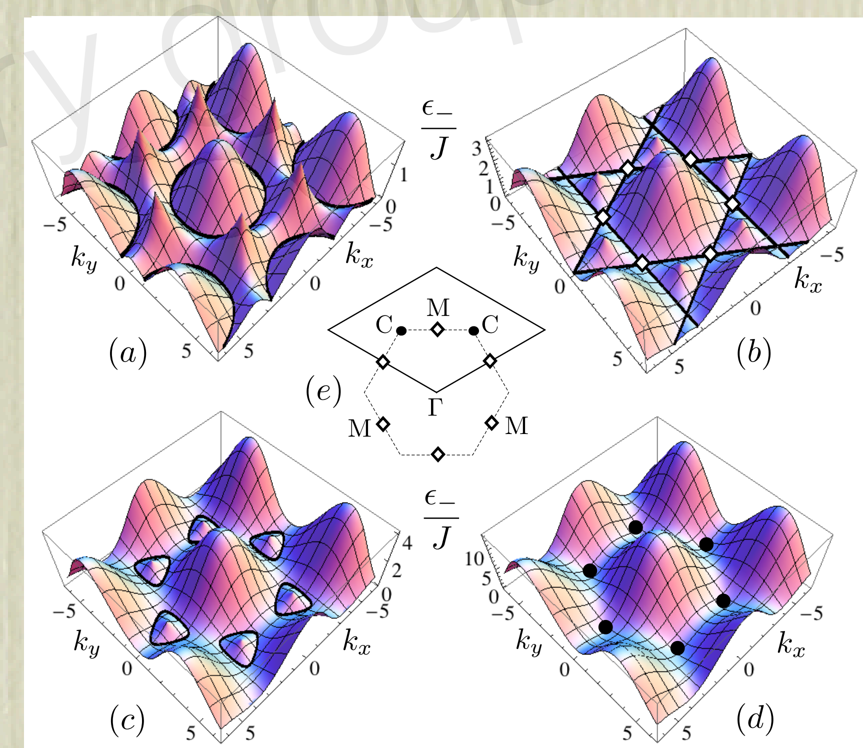
* Linear-T heat capacity: due to an emergent quadratic spin excitation near the criticality



$J_1 = J_2, 0.7J_2, 1.5J_2, 1.8J_2, 0.3J_2, 0$ with $D \approx D_c$ and $D = 1.24J, 1.311J, 1.36J, 1.472J, 1.576J, 2.01J$, respectively.

$(J = J_1 + J_2)$

Experimental check: dynamical spin structure factor from inelastic neutron scattering.



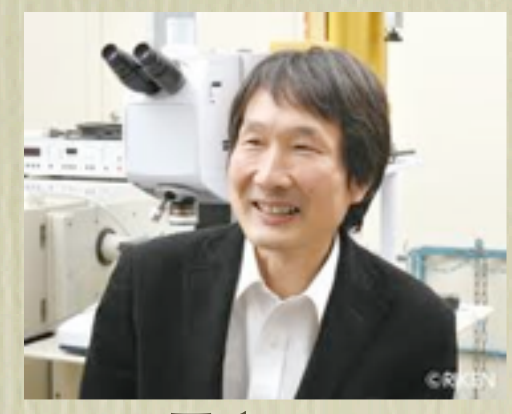
Evolution of low energy spin excitation at the critical point as J2/J1 increases.

Since the model still has U(1) symmetry, i
along z direction si zero. However, if the
powder sample, so spins sus is constant
single crystal smapela and measure susc

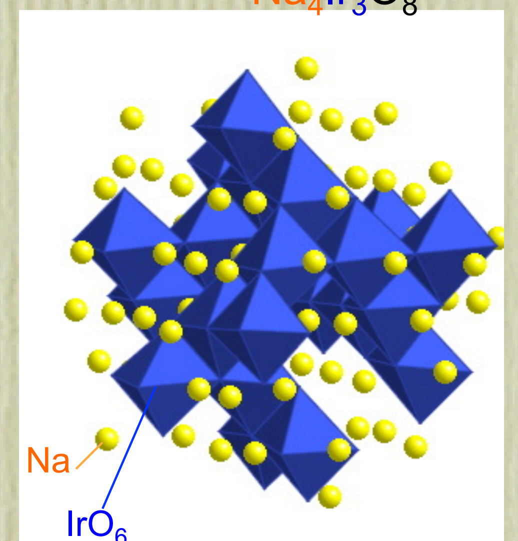
From the low-energy spin excitation, we
capacity at the quantum criticality. There
intermediate temperature. That's due to t
very low temperature, the dispersion bec
behavior. The crossover temperature is c

This is a plot of low energy spin excitatio
layer coupling. And these contours have
neutron scattering.

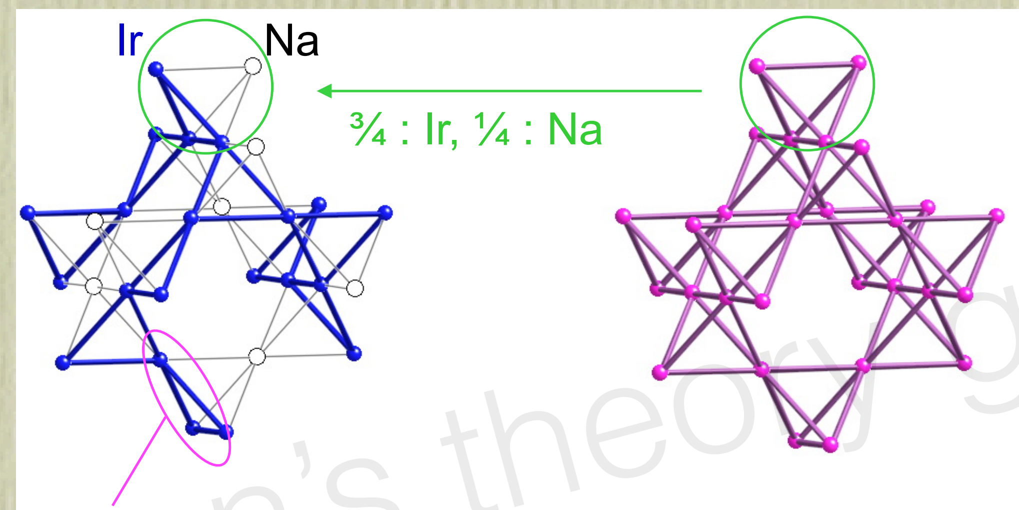
Na4Ir3O8-a promising QSL candidate



Takagi

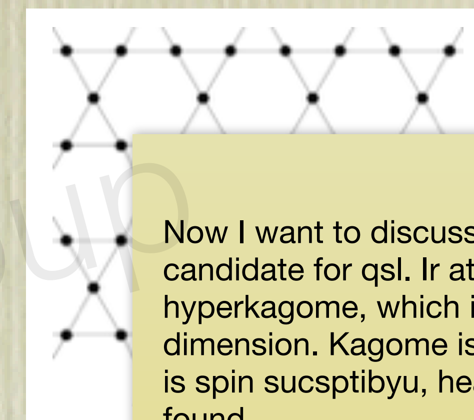


very little disorder

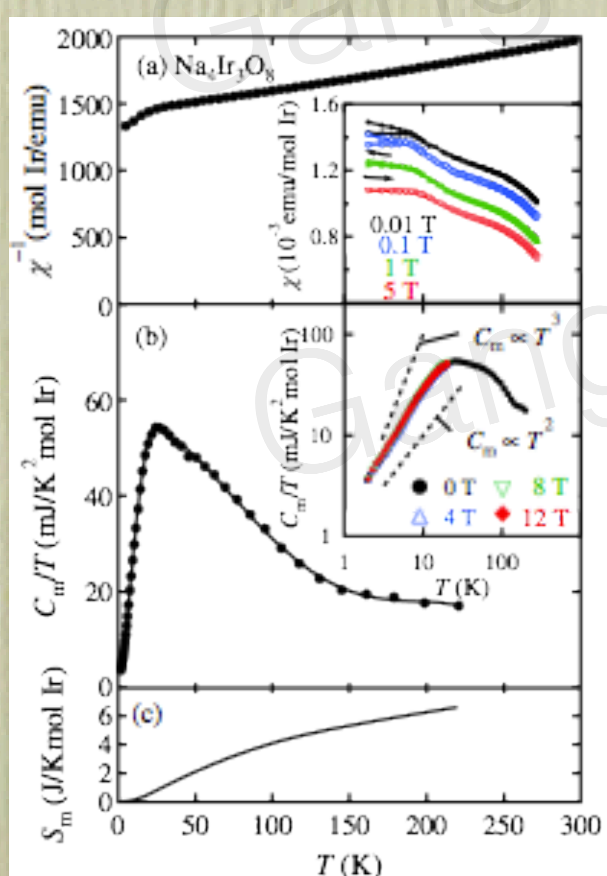


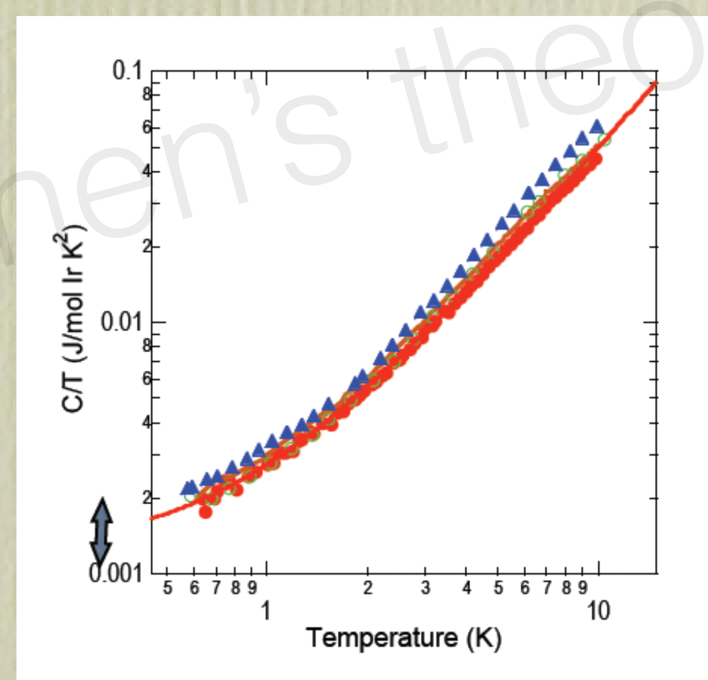
hyperkagome

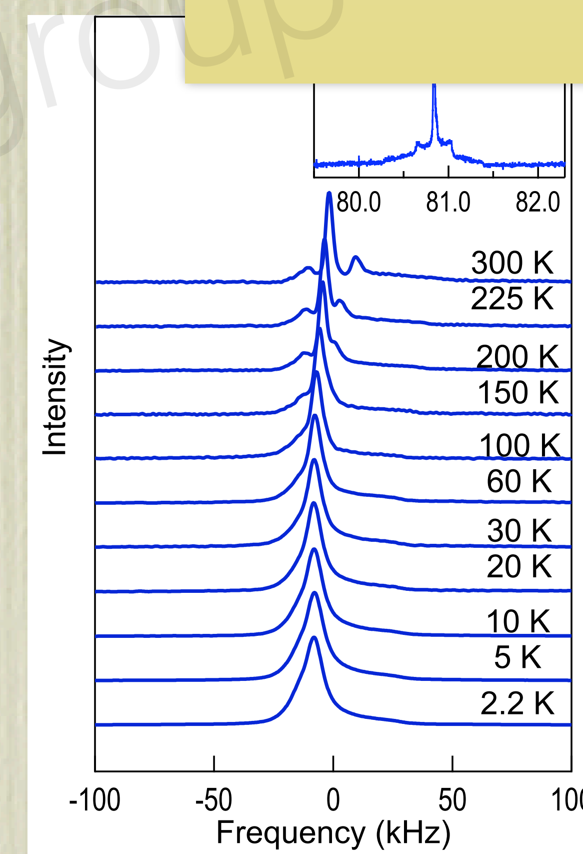
corner-sharing triangles in 3D



Now I want to discuss this material Na4
candidate for qsl. Ir atom carries the mo
hyperkagome, which is a corner sharing
dimension. Kagome is a corner sharing
is spin sucsptibyu, heat capacity, and N
found.







Okamoto, etc, Phys. Rev. Lett. **99**, 137207 (2007)

Na-NMR

Experiments on polycrystal sample

* spin-1/2 moment on hyperkagome lattice
* $\Theta_{\rm CW} = -650K$, no ordering down to 2K, $f > 325$
* very *large* T=0 spin susceptibility
* linear-T heat capacity

Wilson Ratio $\gamma \equiv \frac{C_v}{T}|_{T\to 0}$ $\quad W \equiv \frac{\pi^2}{3}\frac{\chi/\mu_B^2}{\gamma/k_B^2}$ $\quad W$ =35 in polycrys

Wilson Ratio quantifies spin fluctuations that enhance the sus

Table 1 | Some experimental materials studied in the search for QSLs Wilson Ratio

Material	Lattice	S	Θ_{CW} (K)	R*
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$	Triangular†	½	−375‡	1.8
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$	Triangular†	½	−(375–325)‡	? ~1.0-3.0
Cu$_3$V$_2$O$_7$(OH)$_2$•2H$_2$O (volborthite)	Kagomé†	½	−115	6
ZnCu$_3$(OH)$_6$Cl$_2$ (herbertsmithite)	Kagomé	½	−241	?
BaCu$_3$V$_2$O$_8$(OH)$_2$ (vesignieite)	Kagomé†	½	−77	4
Na$_4$Ir$_3$O$_8$	Hyperkagomé	½	−650	~~70~~ 30-40
Cs$_2$CuCl$_4$	Triangular†	½	−4	0

Free electron gas $\quad W = 1$

He-3 (almost localized fermi liquid) $\quad W = 4$

Fe-Superconductor (Fe1.04Te0.67Se0.33) $\quad W = 5.7$

Basic physics in Na4...

- Strong spin-orbit coupling (Z=77)
- Multi-orbital bands, 3 t$_{2g}$ orbitals
- Close to metal-insulator transition
  (true for almost all iridates under current investigation)

Any reasonable modeling should capture these three physics!

L. Balents, Nature **464**, 199 (2010)

GC, et al, Phys. Rev. Lett. **102**, 096406 (2009)

D. Vollhardt, Rev. Mod. Phys. **56**, 99 (1984)

J. Yang, et al, JPSJ, **79**, 074704, (2010)

the spin moment is spn 1/2, CW temp i
parameter is avery large. spins scuspt i
temperature.

Using the heat capacity and spin scusp
Which is quite large W=35. To give you
ratio, let's look at wilson ratio of some c
other qsl candiate is of order of unity.

Let's also look at some other systems.
He-3, which is interpret as
an almost localized fL has wilson ratio 4
Fe-SC, which is believed to be a mutli o

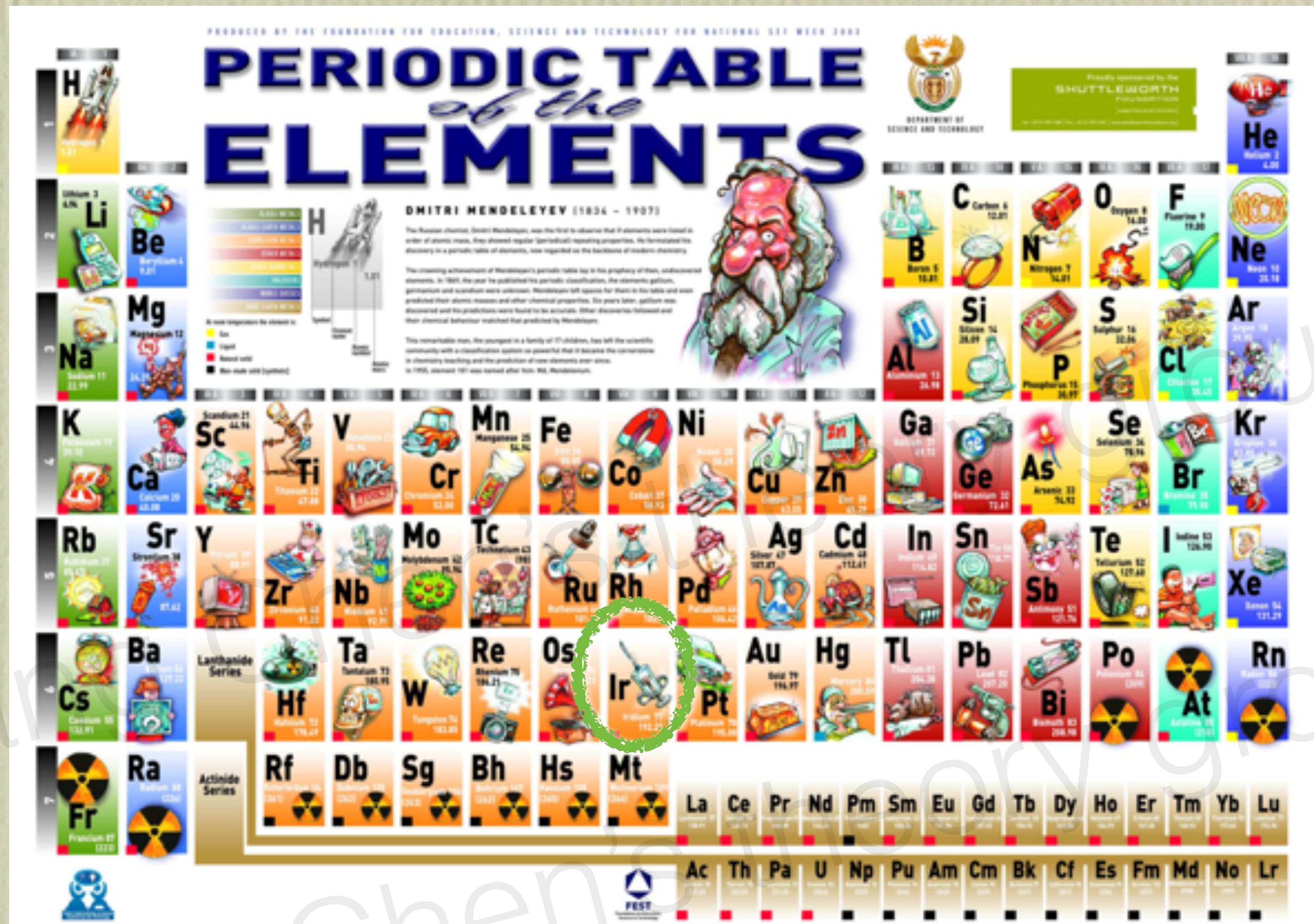
What distinguish NIO from these other
are the following three basic physics.

it has strong soc
it is mutli-obital band, all 3 t2g bands a
it is close to metal-insualtor xtion.

Other qsl candidates are described eith
heisenberg model with spin-rotational s

He-3 is close to Mott insulator xiton, bu
believed to be a multi-band system, bu

Iridium is very heavy!

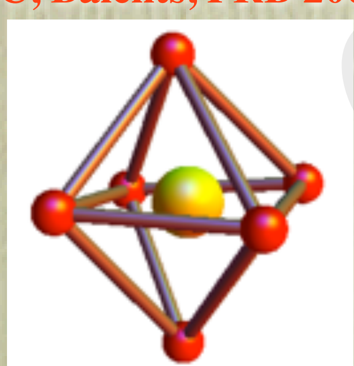


Ir is very heavy, so there is a large soc
the local moment in the mott regime w
first pointed out in this work.

d electron orbital split into upper eg a
low t2g. The lower t2g is further spit b
soc into upper spin j=1/2 and lower j=
Four electrons completely fill j=3/2, an
one electron fill j=1/2 and give a spin-
local moment.

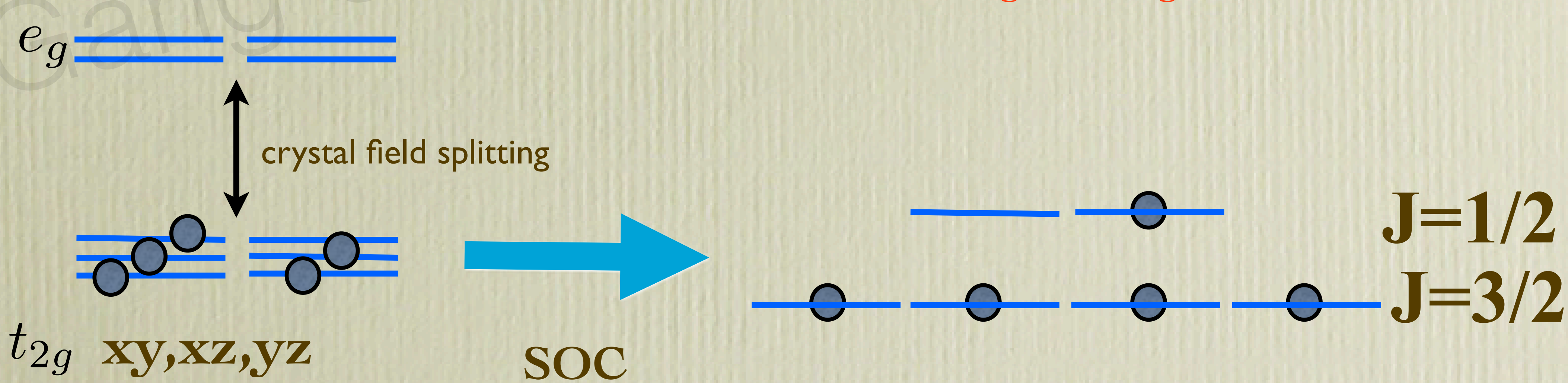
GC, Balents, PRB 2008

Formation of local moment in the strong Mott regime



IrO6

Ir^{4+}, d^5

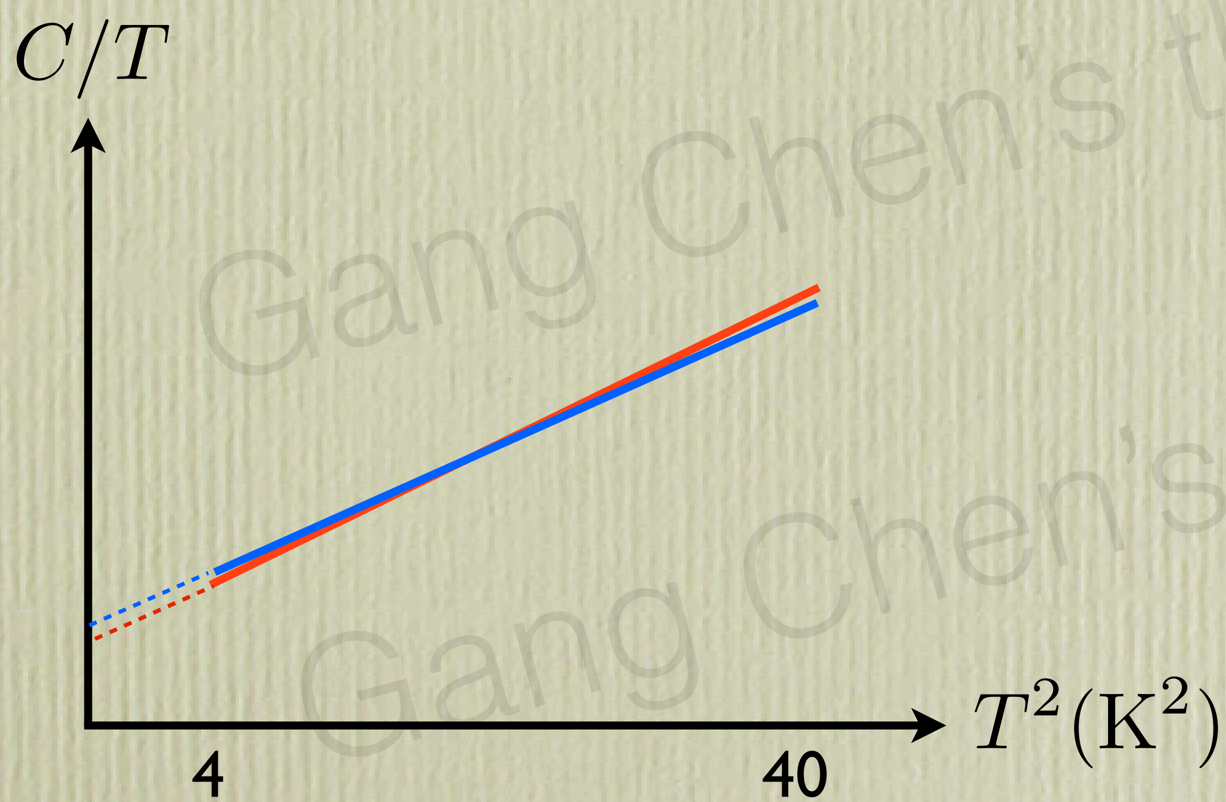


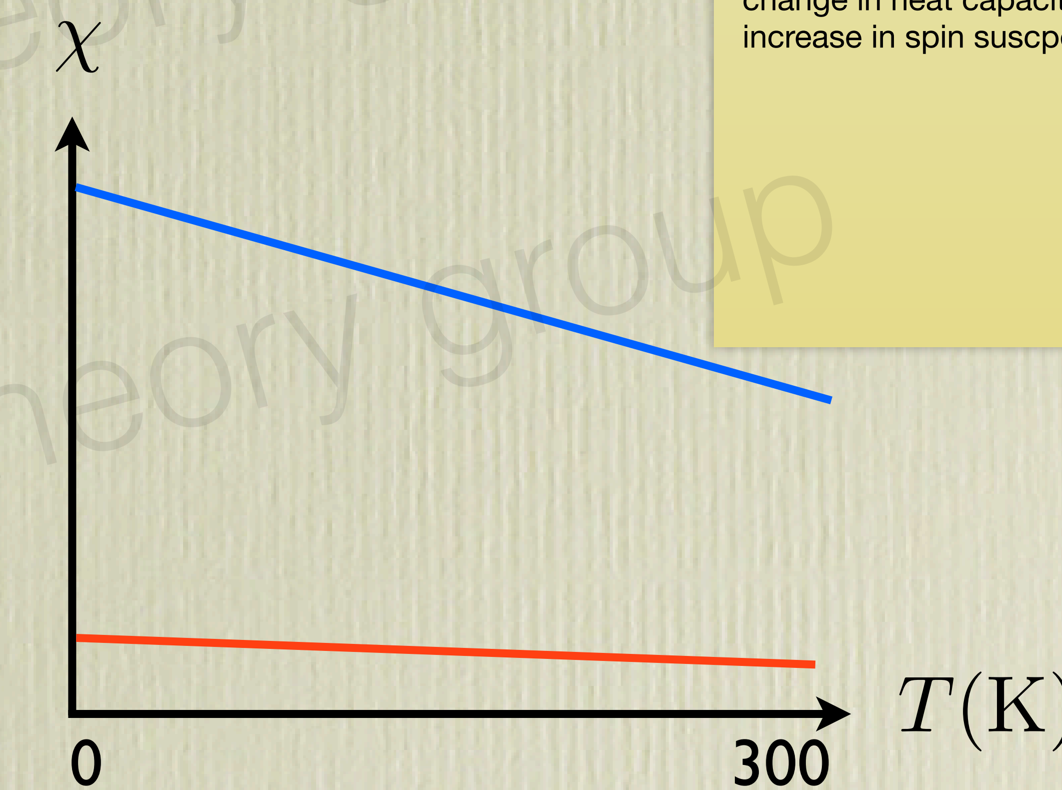
Unpublished new expts

— Single-crystal metallic sample (**R. Perry**, et al, unpublished, Prof. Takagi's group)

— Polycrystal insulating sample (Okamoto, et al, PRL 2007)

Now we have single crystal sample. D
on the ppprepation, thie signle cystralt
is metallic, some time is insuator. Anc
experimental list found, as the sytem
from metal to insualtor, thersei s very
change in heat capacity, but there is a
increase in spin suscpetiblity.





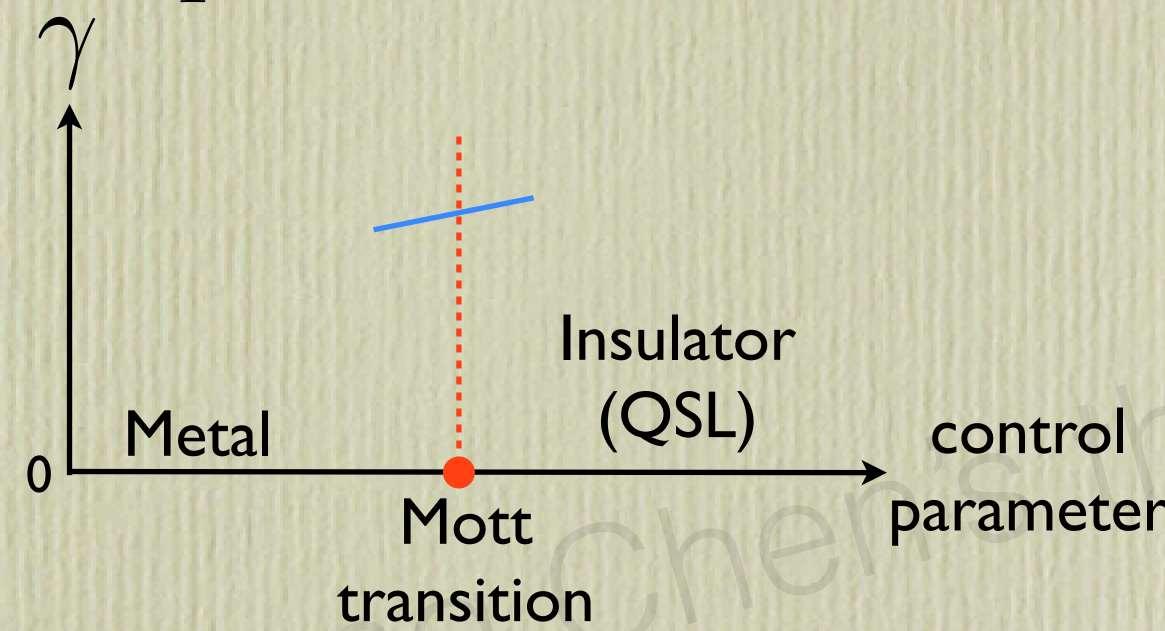
Small change (~18%) in
linear-T heat capacity

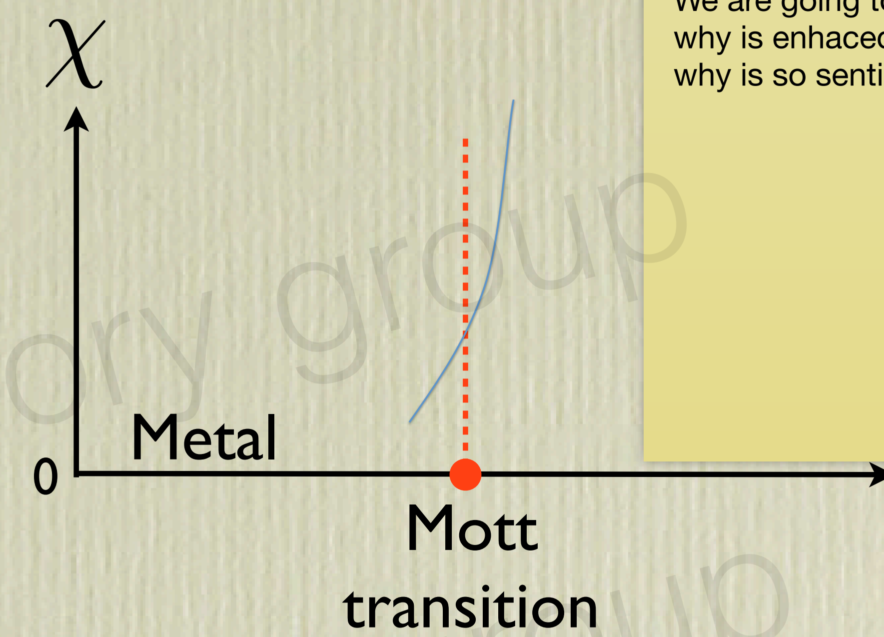
Large enhancement of magnetic susceptibility.
Susceptibility increases with resistivity
(several other single-crystal samples)

Summary of the new expts: schematic plots

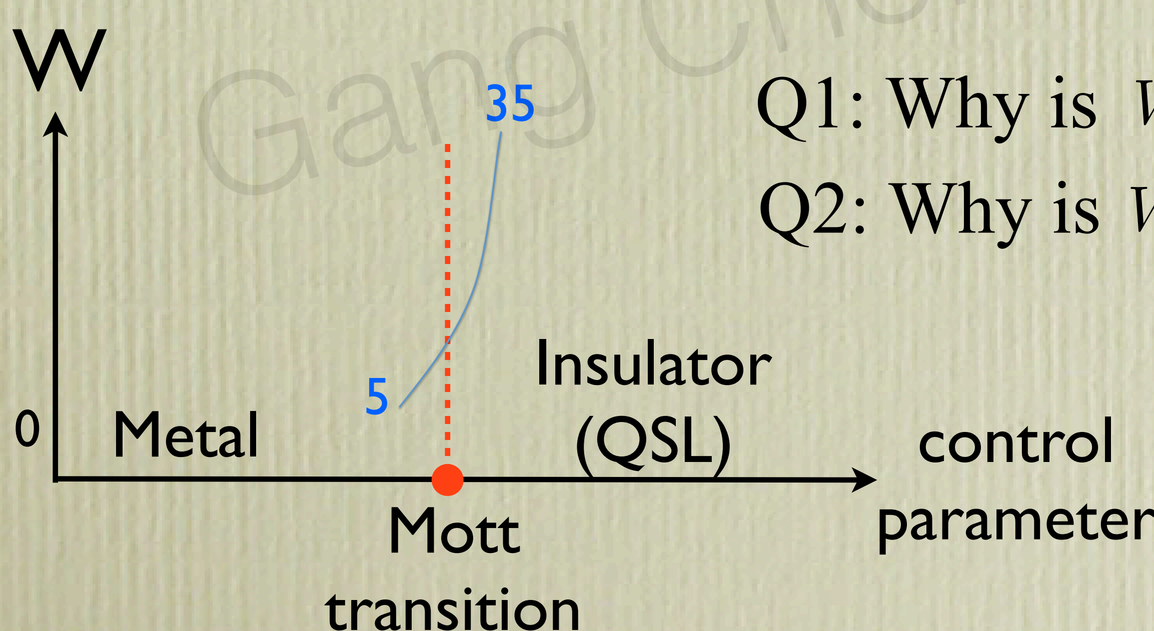
$\gamma \equiv \frac{C_v}{T}|_{T\to 0}$

control parameter: carrier concentration, chemical pressure, etc.





Wilson Ratio $W \equiv \frac{\pi^2}{3}\frac{\chi/\mu_B^2}{\gamma/k_B^2}$



Q1: Why is W(or χ) enhanced in the insulating phase?

Q2: Why is W(or χ) so sensitive to Mott transition?

Let me sumarize the new expts with the
plots. I imagnore there is a control pran
insuatlr xtion. this prameter can be che
somethign else which we don't know.

As the system cross the metal insuatlro
has little cahange,
but suscept is strong enhanaced, so is
from 5 to 35.

We are going to address the following t
why is enhaced in the niuslating phase
why is so sentistive to mott xtion.

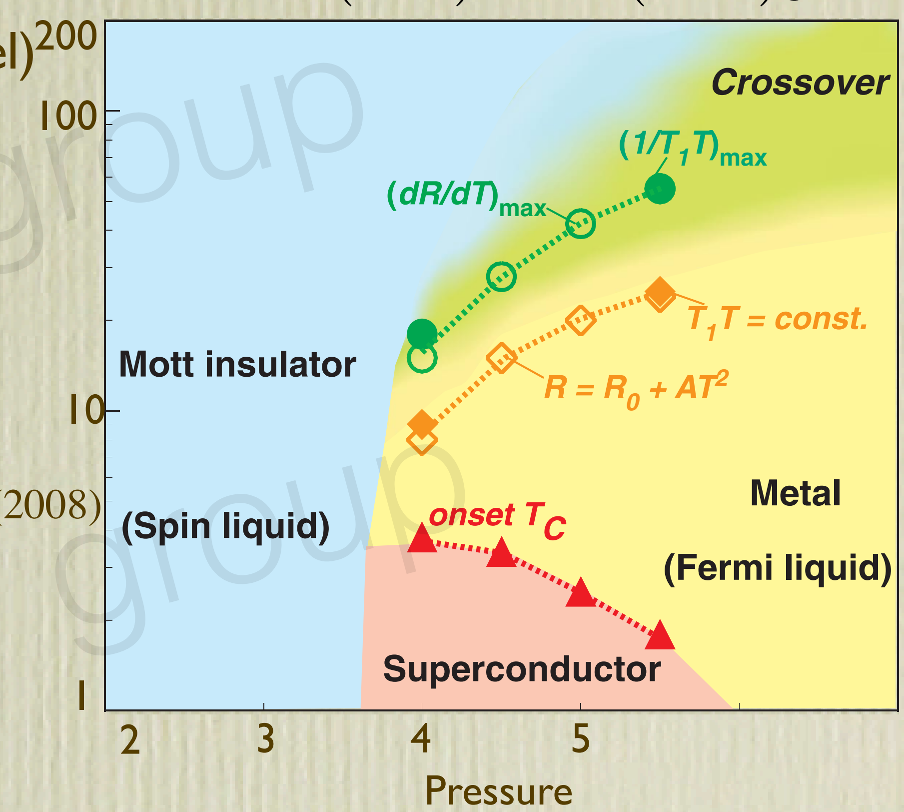
Theoretical proposals

U(1) QSL? M. Lawler, Kim, Balents, etc Phys. Rev. Lett. **101**, 197202 (2008)

Spinon fermi surface: (nearly) linear-T Cv, constant χ (Heisenberg model)

If other interactions are included to break spin-rotational symmetry, large W might be obtained for this state.

$\kappa-(ET)_2Cu_2(CN)_3$



Kanoda's group 2003-

Z2 QSL (less likely) Y. Zhou, P. Lee, etc Phys. Rev. Lett. **101**, 197201 (2008)

Suppress Cv by spinon pairing to enhance W (interesting)

Explain the susceptibility remaining constant by large SOC $\lambda \gg \Delta$

Expect superconductivity in conducting side just like kappa-ET organics
if no SC, expect suppressed Cv from metal to QSL.

VBS (less likely) R. Moessner, etc. Phys. Rev. Lett. **105**, 237202 (2010)

Similar series expansion like Huse+Singh's work on kagome

Complicated ground state: 72 sites in one cell

a bit hard to explain power-law Cv and constant χ over a large temperatur

all the three proposals are based on Heisenberg model.
Heisenberg model is not appropriate for this material. I
first proposal (given by my collaborator ,YBKim and L B
a U(1) QSL with SF. This state should have Wilson ratio

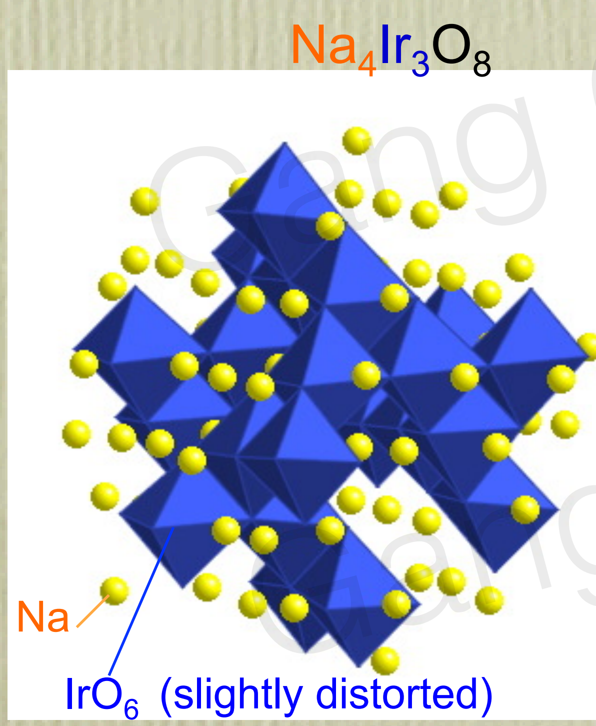
The second proposal is given Patrick Lee and his collab
QSL, with spinon pairing.
The proximate conducting state should have electron p
spinon binds charge boson., which means superductivi
observe that. However, this proximate SC phase was o
organics.

Extended Hubbard model

Basic physics in $Na_4Ir_3O_8$

- Strong spin-orbit coupling (Z=77)
- Multi-orbital bands, 3 t_{2g} orbitals
- Close to metal-insulator transition
  (true for almost all iridates under current investigation)

Here, we want to write down a honest model to capture the three basis physics of NaIO. We consider an extended Hubbard model. in the hamiltonian.
kinetic term, describing hopping,
soc coupling
single ion anisotropy due to the distortion of IrO6 octhadron.



$$\mathcal{H} = \mathcal{H}_{hop} + \mathcal{H}_{soc} + \mathcal{H}_{ion} + \mathcal{H}_{int}$$

$\mathcal{H}_{hop}$ - Tight-binding model

$\mathcal{H}_{soc}$ - Atomic spin-orbit coupling

$\mathcal{H}_{ion}$ - single-ion (crystal field) term due to IrO_6 distortion
(drive transition from TBI to metal in 227 iridates)

$\mathcal{H}_{int}$ - Multiorbital interactions

Wilson ratios in non-interacting

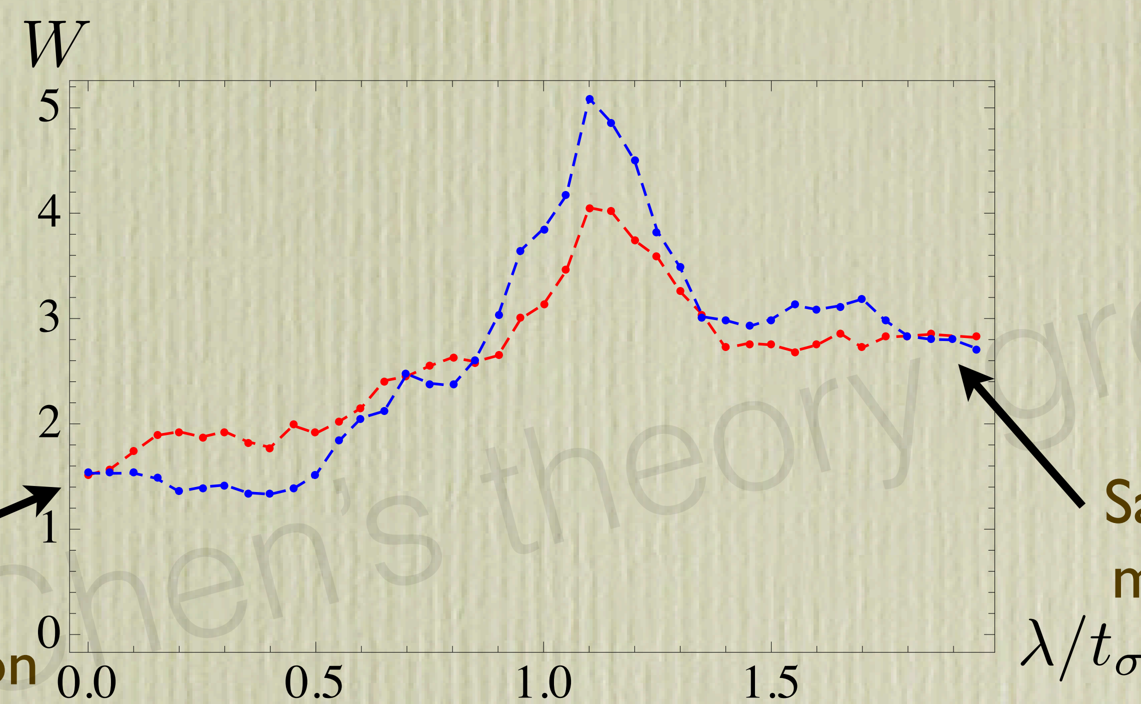
Let's first look the non interacting limit

WIslon raito against spinon orbit, t simg
hopping parameter

spin susc not only has contribution frm
from non-FS contribuiton.

two an

$$\mathbf{M}_i \equiv \mu_B(\mathbf{L}_i + 2\mathbf{S}_i)$$



$D = 0.4$

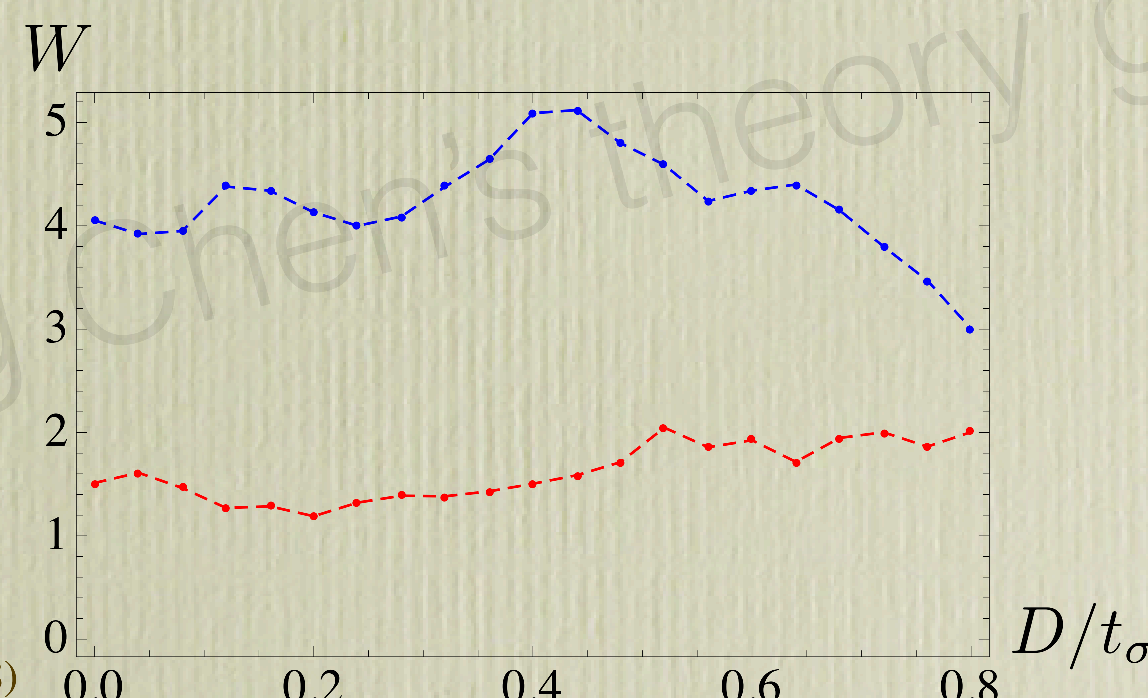
Same reason why Heisenberg model is relevant for Sr_2IrO_4

$W \neq 1$

is because of the hybridization of different orbitals

λ -SOC

t_σ -hopping parameter



$\lambda = 0$

$\lambda = 1.1$

G. Chen, et al Phys. Rev. B **78**, 094403, (2008)
G. Jackeli, et al Phys. Rev. Lett. **102**, 017205, (2009)
F. Wang, et al Phys. Rev. Lett. **106**, 136402, (2011)

Wilson Ratio is 5 at most.

Multi-orbital interaction

$$H_{int} = U\sum_{i,m}\hat{n}_{i,m,\uparrow}\hat{n}_{i,m,\downarrow} + \frac{U'}{2}\sum_{i,m\neq m'}\hat{n}_{i,m}\hat{n}_{i,m'}$$

$$+\frac{J}{2}\sum_{i,m\neq m'} d^\dagger_{im\sigma}d^\dagger_{im'\sigma}d_{im\sigma'}d_{im'\sigma} + \frac{J'}{2}\sum_{i,m\neq m'} d^\dagger_{im\uparrow}d^\dagger_{im\downarrow}d_{im'\downarrow}d_{im'\uparrow}$$

i is a position index.

m is an orbital index.

In atomic limit, $U = U' + J + J'$

$J = J'$

Rewrite interaction, $\mathcal{H}_{int} = \mathcal{H}_{c-int} + \mathcal{H}_{ex-int}$

$$\mathcal{H}_{c-int} = \frac{U}{2}\sum_i(\hat{n}_i - 5)^2$$

$$\mathcal{H}_{ex-int} = -J\sum_{i,m\neq m'}\hat{n}_{i,m}\hat{n}_{i,m'} + \frac{J}{2}\sum_{i,m\neq m'} d^\dagger_{imc}$$

$$+\frac{J}{2}\sum_{i,m\neq m'} d^\dagger_{im\uparrow}d^\dagger_{im\downarrow}d_{im'\downarrow}d_{im'\uparrow}$$

There are four terms in the mutliorbiatal interactior
m is the orbital inddex, i is the lattice site. sigme is

intraorbital coulomb
interobital comblomg
hunds
pair hopping

in atomic limit, we have this realtion. I am going to

I rewrite the inteaction into two parts, the charge i
exchange.The two terms have different physics. th
charge interaction. U si the energy scale for exciss
orccupation while the second part is the spin/orbit

Sicne these two parts descaribe iddferent physica
diferetnely.

U is the energy scale for excessive electron/charge occupation.

J is the energy scale for electron distribution among different spin and orbital states.

$\mathcal{H}_{ex-int}$ is like an onsite exchange interaction in the *Kugel-Khomskii* picture.

W. Ko, P.A. Lee, Phys. Rev. B. **83**, 134515 (2011)

Strong-coupling MFT: slave-Rotor

First I want to treat the charge interact
is the largest enrgy scale and is respo

To desribe the metal insulato transtion
appraoch, this similar as the slave bsc
organics dicscueed i nthe itnrudction.
cary spin. Rotor condenson correspo
torotr corresond to the QSL.

The hubbard model can be solved by
calcualation.

Slave-rotor approach to obtain fermionic spinons

$$d_{im\alpha} = e^{-i\theta_i} f_{im\alpha} \qquad \text{vs} \qquad \boxed{d_{im\alpha} = b_i f_{im}}$$

$$L_i(R) = \sum_{m\sigma} f^\dagger_{im\alpha}(R) f_{im\alpha}(R) - 5$$

$$[\theta_i, L_i] = i$$

$\langle e^{-i\theta_i} \rangle \neq 0,\ Z \neq 0,$ spin and charge are confined, we have a "correlated FL".

$\langle e^{-i\theta_i} \rangle = 0,\ Z = 0,$ we have a "U(1) QSL".

Original electron Hamiltonian (with the Hubbard-U interaction only)

$$H_{hop} = \sum_{Rim,R'i'm'} t^{ii'}_{mm'} d^\dagger_{im\sigma}(R) d_{im'\sigma}(R') + h.c.$$

$$H_{c-int} = \frac{U}{2} \sum_{Ri} \Big(\sum_{m,\alpha} d^\dagger_{im\alpha}(R) d_{im\alpha}(R) - 5 \Big)^2$$

$$H_{ion} = D \sum_{Ri\alpha} (L^\mu_i)^2_{mn} d^\dagger_{im\alpha}(R) d_{in\alpha}(R)$$

$$H_{soc} = \frac{\lambda}{2} \sum_{Ri} \mathbf{L}_{mn} \cdot \boldsymbol{\sigma}_{\alpha\beta} d^\dagger_{im\alpha}(R) d_{in\beta}(R)$$

Slave-rotor mean field Hamiltonian

$$H_f = Q_f \sum_{Rim,R'i'm'} (t^{ii'}_{mm'} f^\dagger_{im\sigma}(R) f_{im'\sigma}(R') + h.c.)$$

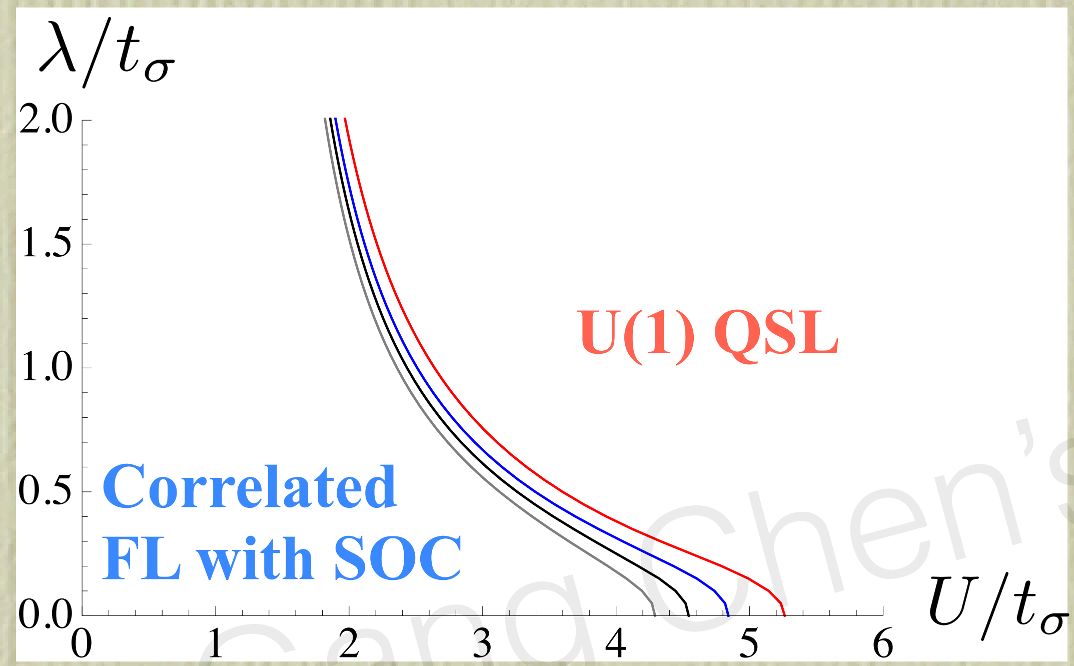
$$+ \frac{\lambda}{2} \sum_{Ri} \mathbf{L}_{mn} \cdot \boldsymbol{\sigma}_{\alpha\beta} f^\dagger_{im\alpha}(R) f_{in\beta}(R) \quad + D \sum_{Ri\alpha} (L^\mu_i)^2_{mn} f^\dagger_{im\alpha}(R) f_{in\alpha}(R)$$

$$H_L = \frac{U}{2} \sum_{Ri} L^2_i(R) + \sum_{Ri} (h L_i(R) + 5h) + Q_r \sum_{Ri,R'i'} e^{i\theta_i(R) - i\theta_{i'}(R')} + h.c.$$

$$Q_f \equiv \langle e^{i\theta_i(R) - i\theta_{i'}(R')} \rangle_\theta \qquad Q_r \equiv \sum_{mm'\sigma} t_{mm'} \langle f^\dagger_{im\sigma} f_{i'm'\sigma}(R) \rangle_f$$

S. Florens and A. Georges,
Phys. Rev. B. **70**, 035114 (2004)
D. Pesin, L. Balents, Nature Physics **6**, 376 (2010)

Phase diagram



From left to right, the single-ion anisotropies are

$D = 0.8t_\sigma$ $D = 0.4t_\sigma$ $D = 0.2t_\sigma$ $D = 0$

Three energy scales: SOC, correlation, bandwidth

Two observations:

1. SOC enhances correlation effects.

   Strong correlation physics may be seen in 4d/5d electron system.

2. Correlation effects enhance SOC.

   SOC may be also important even in 3d electron system in certain cases.

This is the mft phase diagram.

illustrate point 1,
4d,5d have strong soc, weak correaltion, but soc suppress bw, and enhance correlation.

pt 2, soc is weak in 3d, correaltion suppress bw and then enehance soc

D. Pesin, L. Balents, Nature Physics **6**, 376 (2010)

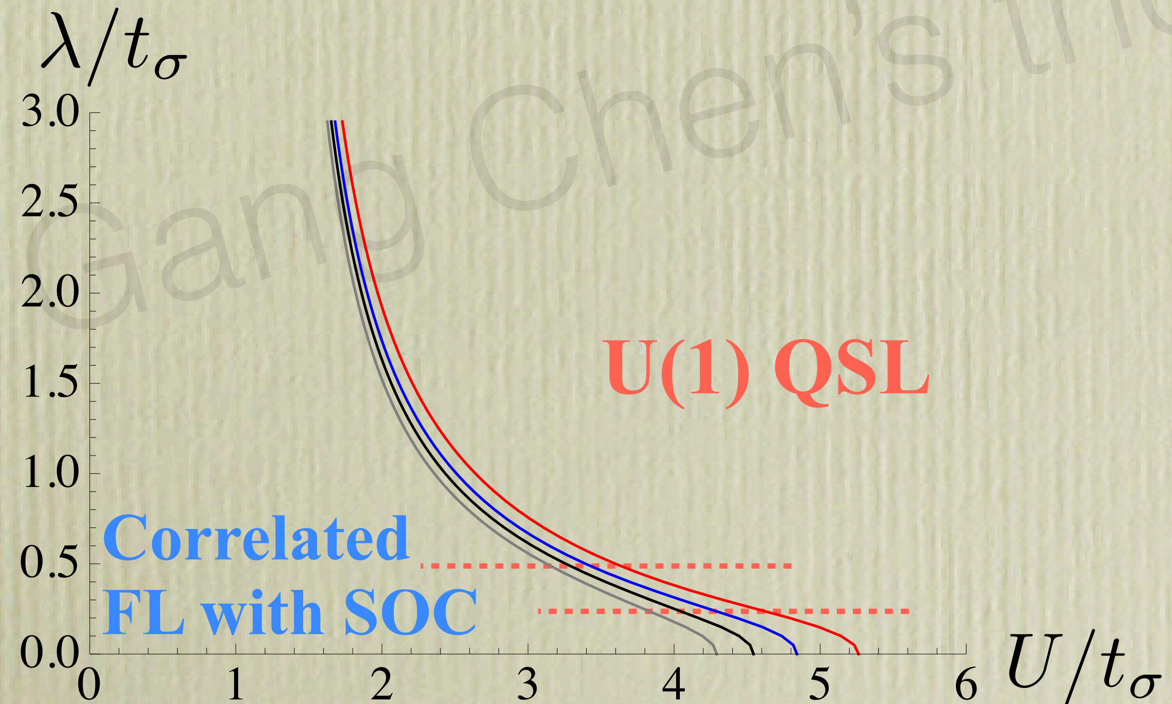
Onsite exchange

We put the onsite exchange interaction in the spinon mean field hamiltonian.

W. Ko, P.A. Lee, Phys. Rev. B. **83**, 134515 (2011)

$$H_{ex-int} = \sum_i \Big[-J \sum_{m\neq m'} f^\dagger_{im\sigma} f_{im\sigma} f^\dagger_{im'\sigma'} f_{im'\sigma'} + \frac{J}{2} \sum_{m\neq m'} f^\dagger_{im\sigma} f^\dagger_{im'\sigma'} f_{im\sigma'} f_{im'\sigma} + \frac{J}{2} \sum_{m\neq m'} f^\dagger_{im\uparrow} f^\dagger_{im\downarrow} f_{im'\downarrow} f_{im'\uparrow} \Big]$$

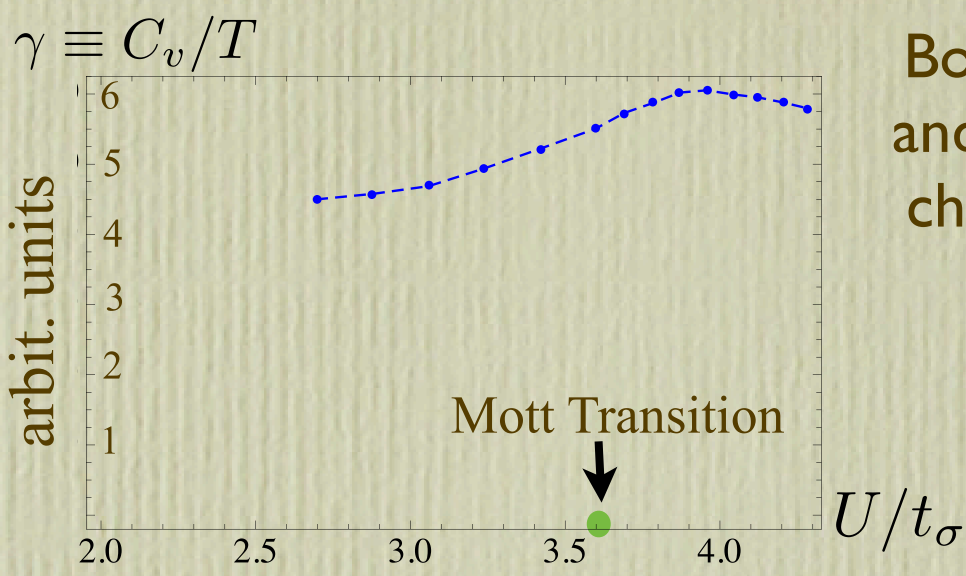
$$H_f \to H_f + H_{ex-int}$$



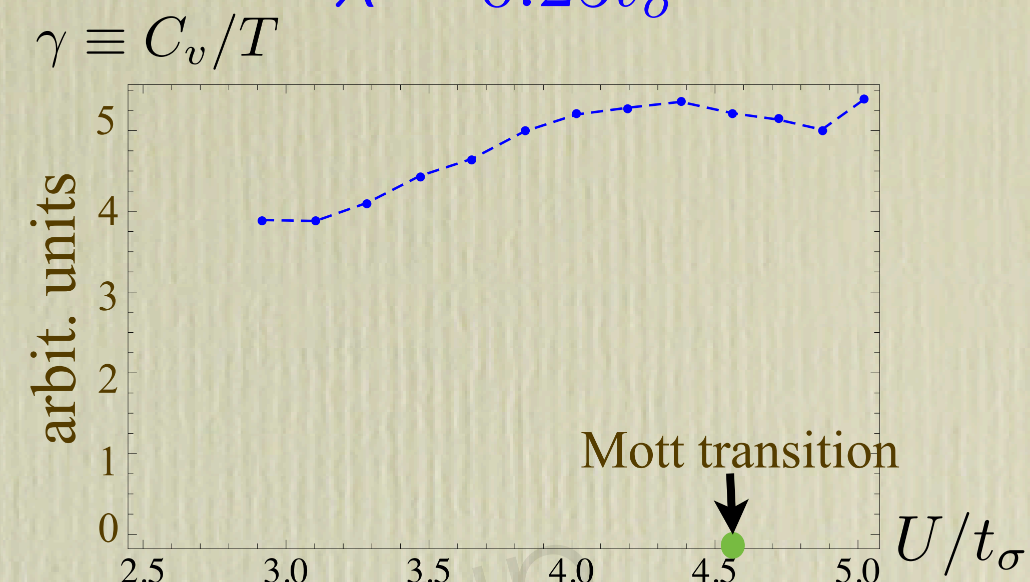
$$\mathbf{M}_i \equiv \mu_B (\mathbf{L}_i + 2\mathbf{S}_i)$$

Study Wilson ratio along the dashed line

$\lambda = 0.5t_\sigma$



$\lambda = 0.25t_\sigma$



Both "effective mass" and fermi surfaces are changed due to SOC

$$J \lesssim 0.1U_c$$

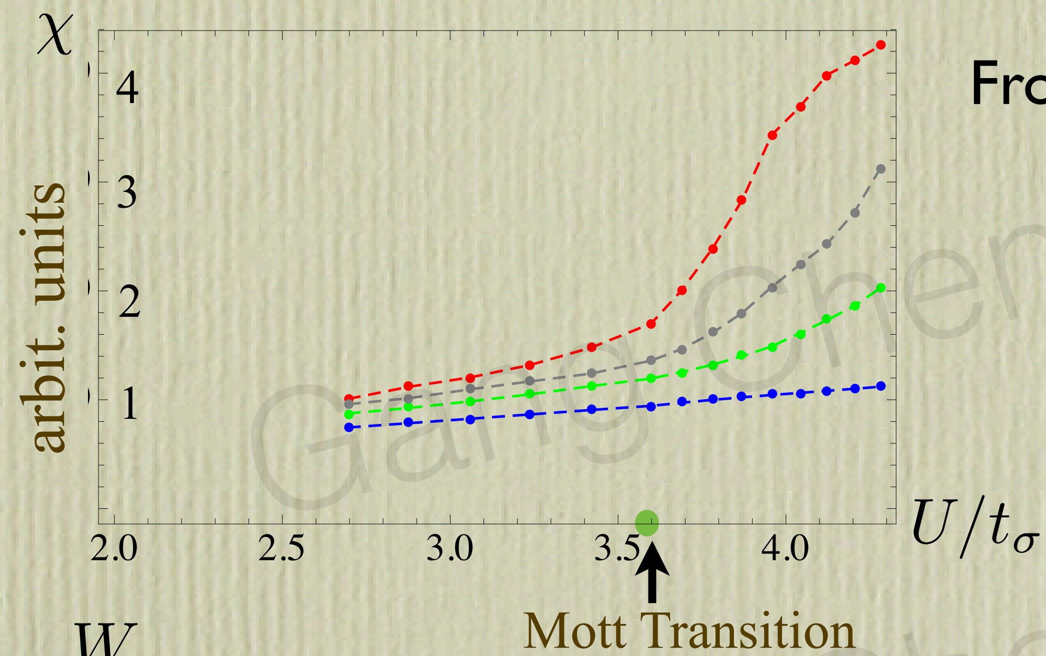
From top to bottom,

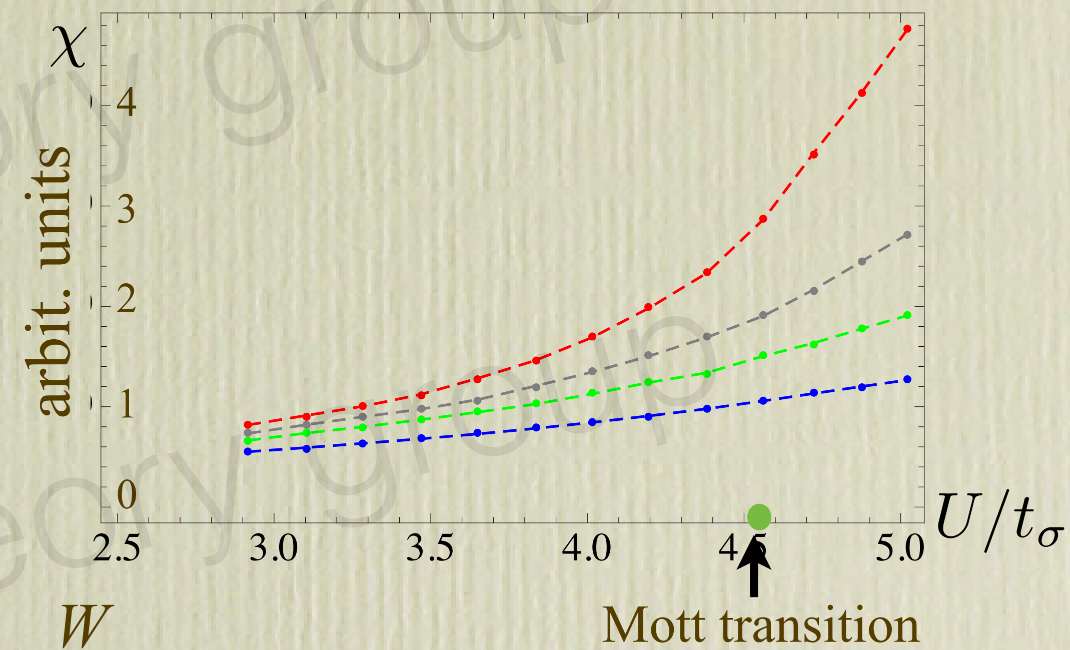
$J = 0.4t_\sigma$

$J = 0.3t_\sigma$

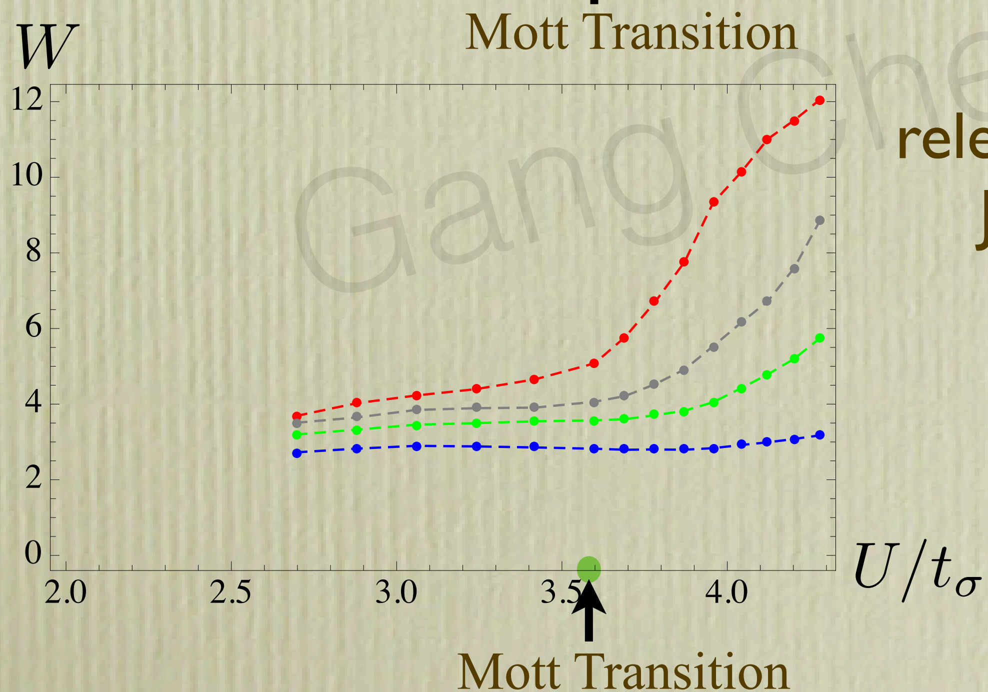
$J = 0.2t_\sigma$

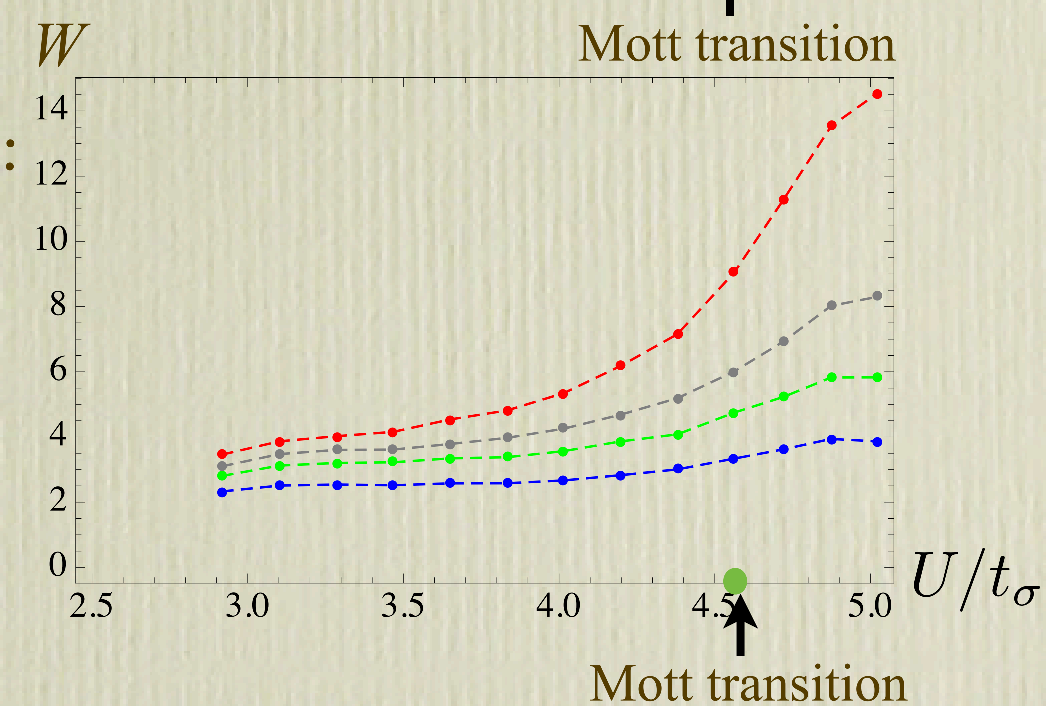
$J = 0.1t_\sigma$





relevant energy scales: J and bandwidth





Summary for $Na_4Ir_3O_8$

* $Na_4Ir_3O_8$ is likely to be a U(1) quantum spin liquid with spinon fermi surfaces.

* The large Wilson ratio might arise from the combined effect of spin-orbit coupling, correlation and onsite spin-orbital exchange.

For experiments,

* Other experiments: resonant inelastic x-ray scattering (planned), thermal conductivity (seems like a metal), quantum oscillations (too soft gauge field? O. Motrunich, PRB 2005)

* Can similar physics be observed in related materials?
e.g. nonmagnetic $R_2Ir_2O_7$ (pyrochlore lattice), etc

Summary and outlook

- I introduce the basis theoretical concepts and experiments related to QSLs.
- I review QSL candidate materials and explain the physics in both failed and promising examples.
- Searching for QSL in real materials provides a lot of opportunities for theoretical innovation, material synthesis, and experimental efforts.