Wilson Ratio enhancement in a quantum spin liquid candidate: $Na_4Ir_3O_8$ (hyperkagome)

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Acknowledge Dr. Perry and Prof. Takagi
for sharing their experimental results

KITP conference 2012

Quantum spin liquid candidates

Triangle $\kappa-(ET)_2Cu_2(CN)_3, EtMe_3Sb[Pd(dmit)_2]_2,$

$LiZn_2Mo_3O_8$

$He-3$ on graphite layer

Kagome $Cu_3Zn(OH)_6Cl_2$(kapellasite),

$BaCu_3V_2O_8(OH)_2$(vesignieite),

$ZnCu_3(OH)_6Cl_2$(herbertsmithite)

FCC Ba_2YMoO_6

Pyrochlore *some* $R_2TM_2O_7$ R=rare earth, TM=transition metal — Quantum spin ice

Hyperkagome $Na_4Ir_3O_8$

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Hyperkagome $\boxed{Na_4Ir_3O_8}$

4d, 5d and f electrons: Spin-orbit coupling is expected to be important, and may lead to some new physics.

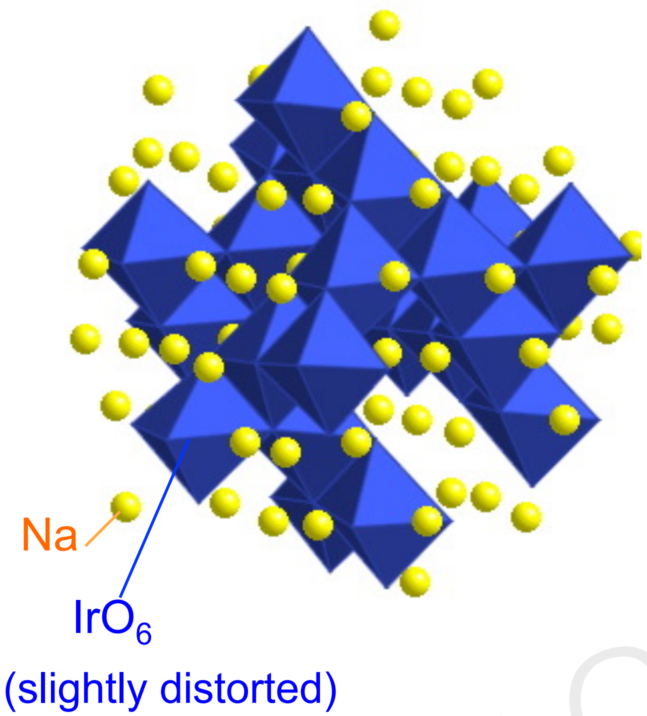
Outline

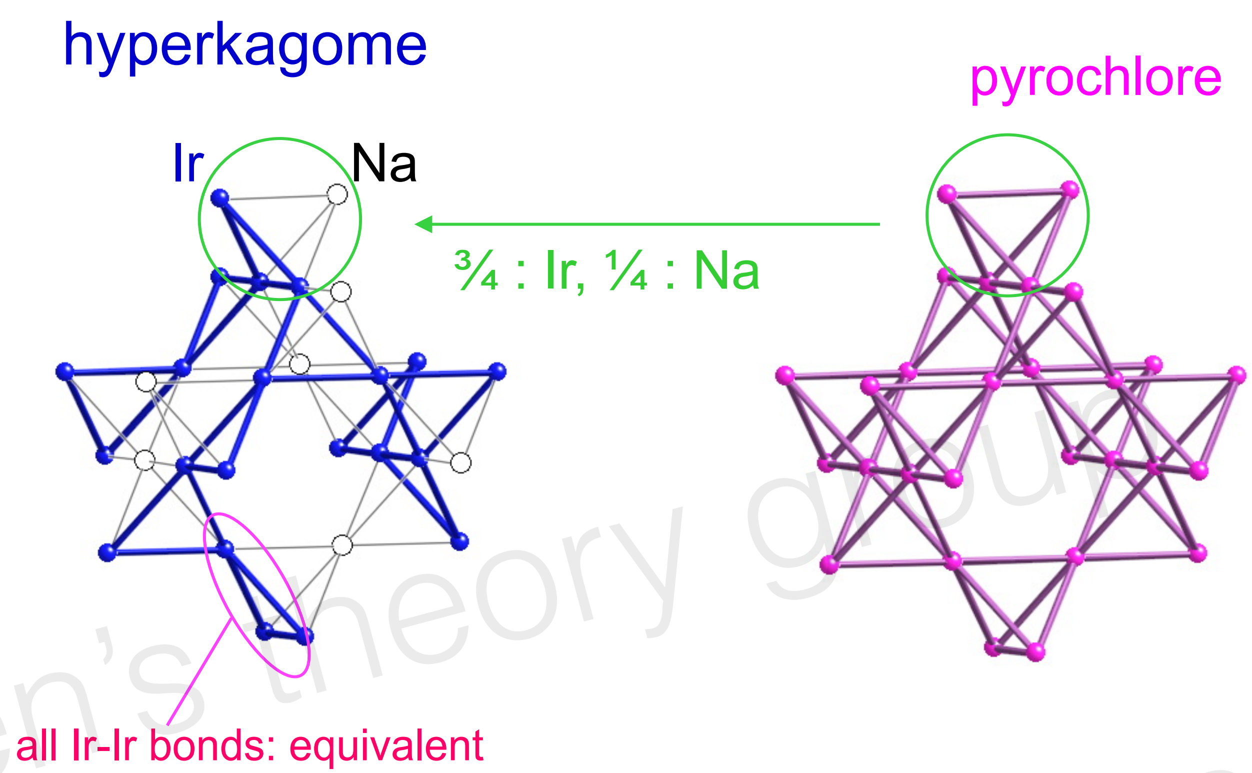
Part 1. Review experiments and current theories

Part 2. Present a possible explanation for the experiments

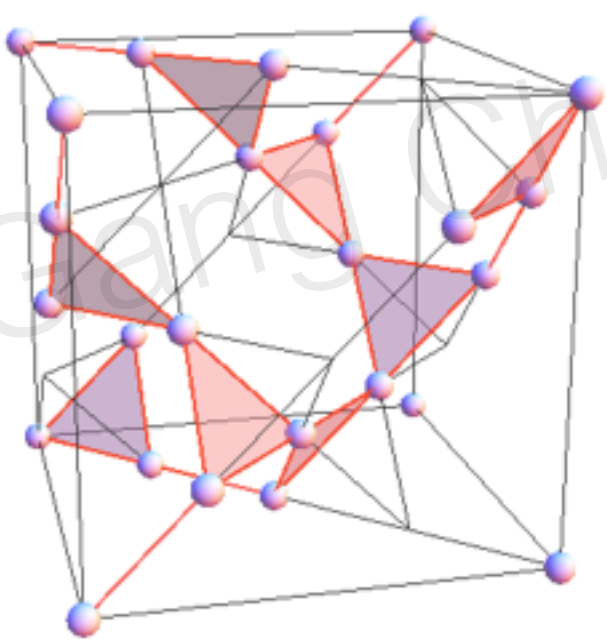
$Na_4Ir_3O_8$: a hyperkagome Ir sublattice

$Na_4Ir_3O_8$



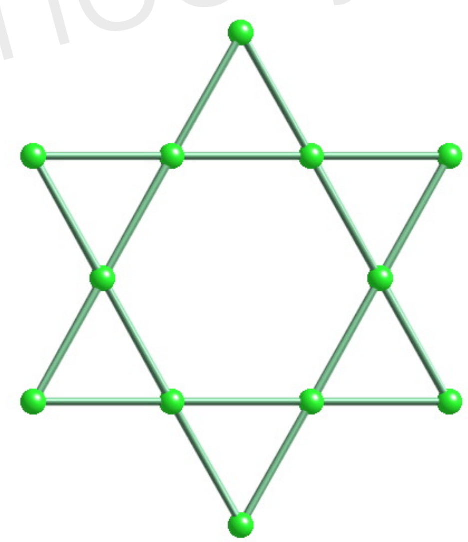


hyperkagome



12 sites per unit cell

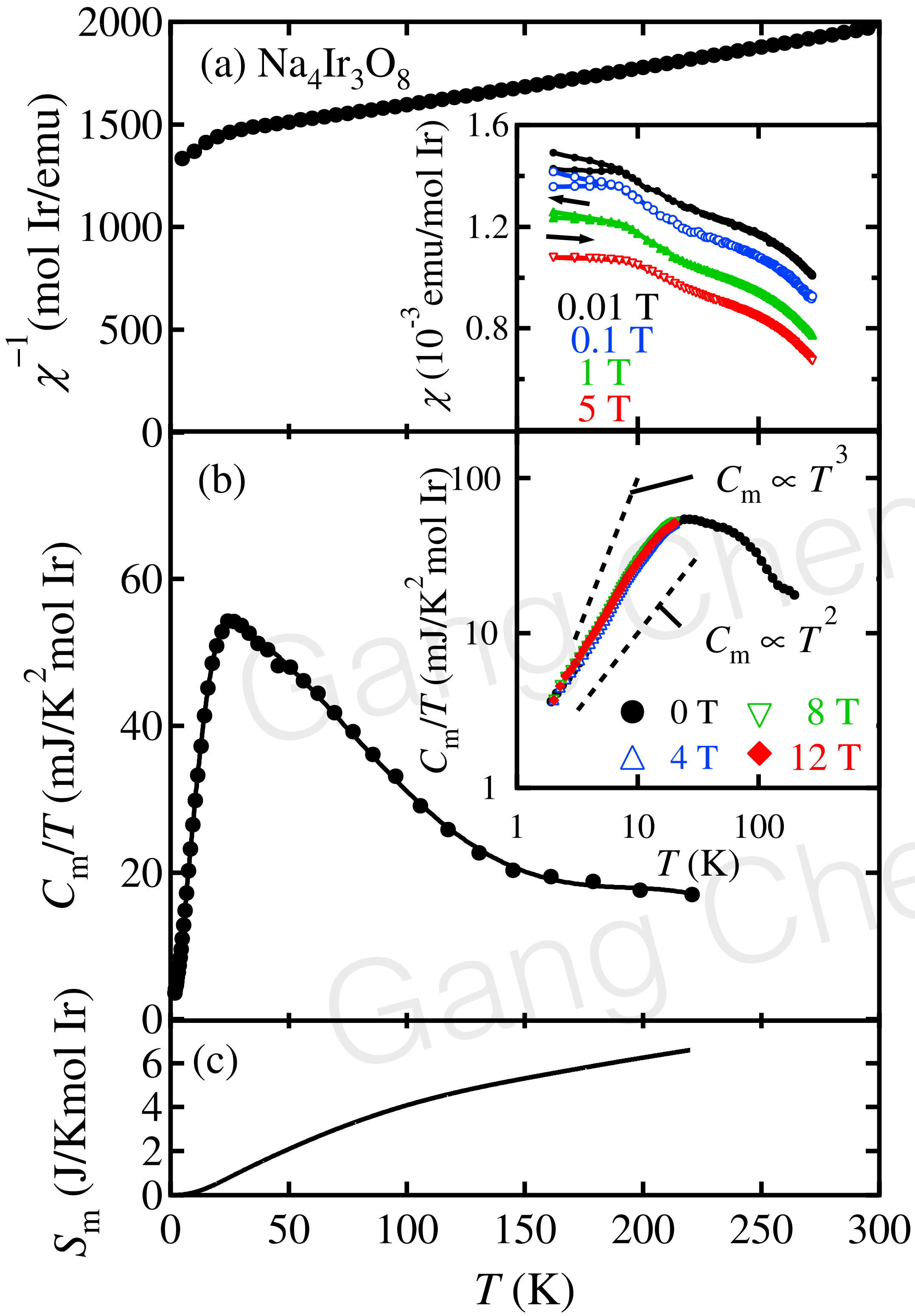
kagome

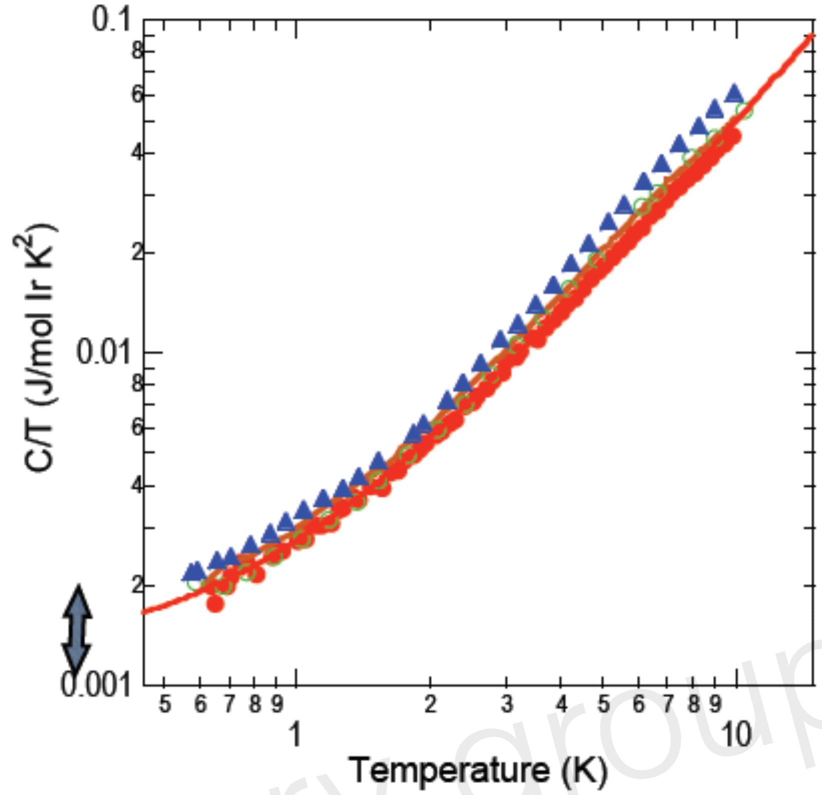


3 sites per unit cell

Y. Okamoto, et al, H. Takagi
Phys. Rev. Lett. **99**, 137207 (2007)

Polycrystal sample





From
Prof. Takagi's talk

Curie-Weiss fit:

$$\mu_{eff} = 1.96\mu_B$$ close to spin-1/2

$$\Theta_{CW} = -650\text{K}$$

No indication of ordering down to 2K from Cv and χ

NMR measurement confirms the absence of magnetic ordering. (See Prof. Takagi's talk on Monday)

$$\chi|_{T\to 0} = constant$$

$$C_v/T|_{T\to 0} = constant$$

Very large Wilson Ratio, 35!

Y. Okamoto, et al, H. Takagi
Phys. Rev. Lett. **99**, 137207 (2007)

Wilson Ratios of some QSL candidates

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

Material	Lattice	S	Θ_{CW} (K)	R*	Status or explanation
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$	Triangular†	½	−375‡	1.8	Possible QSL
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$	Triangular†	½	−(375–325)‡	? ~1.0-3.0	Possible QSL
Cu$_3$V$_2$O$_7$(OH)$_2$•2H$_2$O (volborthite)	Kagomé†	½	−115	6	Magnetic
ZnCu$_3$(OH)$_6$Cl$_2$ (herbertsmithite)	Kagomé	½	−241	?	Possible QSL
BaCu$_3$V$_2$O$_8$(OH)$_2$ (vesignieite)	Kagomé†	½	−77	4	Possible QSL
Na$_4$Ir$_3$O$_8$	Hyperkagomé	½	−650	70 30-40	Possible QSL
Cs$_2$CuCl$_4$	Triangular†	½	−4	0	Dimensional reduction
FeSc$_2$S$_4$	Diamond	2	−45	230	Quantum criticality

BEDT-TTF, bis(ethylenedithio)-tetrathiafulvalene; dmit, 1,3-dithiole-2-thione-4,5-ditholate; Et, ethyl; Me, methyl. *R is the Wilson ratio, which is defined in equation (1) in the main text. For EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ and ZnCu$_3$(OH)$_6$Cl$_2$, experimental data for the intrinsic low-temperature specific heat are not available, hence R is not determined. †Some degree of spatial anisotropy is present, implying that $J' \neq J$ in Fig. 1a. ‡A theoretical Curie–Weiss temperature (Θ_{CW}) calculated from the high-temperature expansion for an S = ½ triangular lattice; $\Theta_{CW} = 3J/2k_B$, using the J fitted to experiment.

Wilson Ratio quantifies spin fluctuations that enhance the susceptibility.

Fermi gas $W = 1$

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

Fe-Superconductor ($Fe_{1.04}Te_{0.67}Se_{0.33}$) $W = 5.7$

Basic physics in $Na_4Ir_3O_8$

- Strong spin-orbit coupling (Z=77)
- Multi-orbital bands, 3 t2g orbitals
- Close to metal-insulator transition (true for almost all iridates under current investigation)

With SOC, spin-rotational symmetry is broken,
large Wilson Ratio is certainly possible,
e.g. ordered AFMagnet with gapped spin-wave excitations

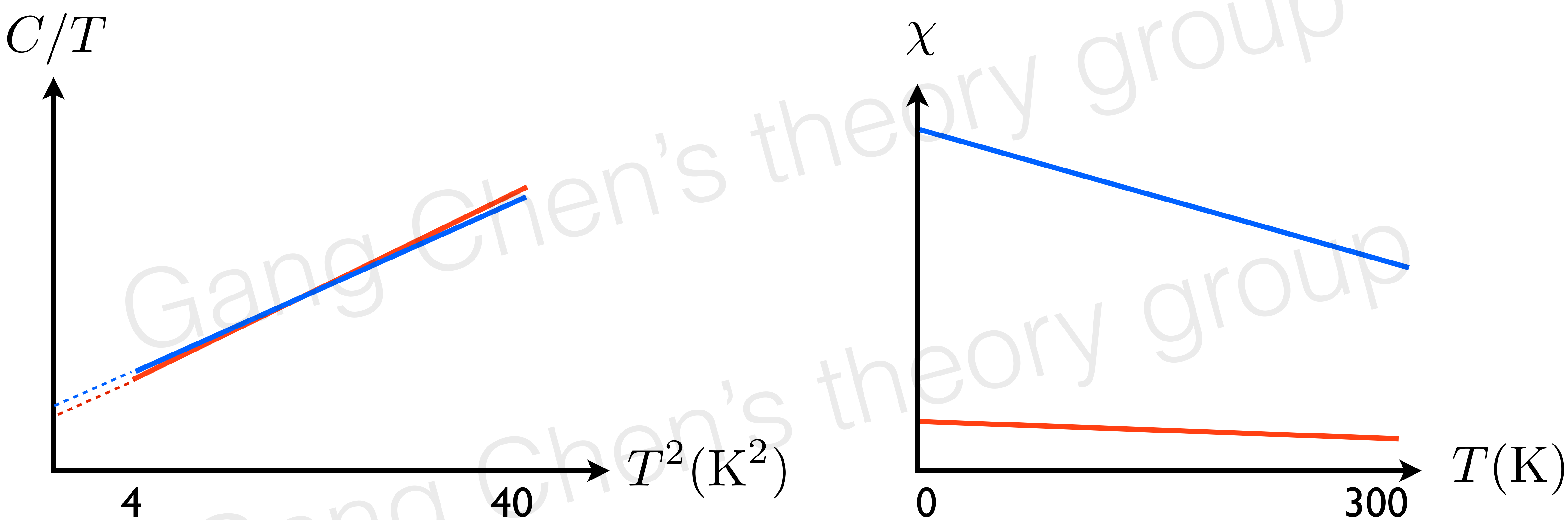
$$W = \infty$$

L. Balents, Nature **464**, 199 (2010)
G. Chen, et al, Phys. Rev. Lett. **102**, 096406 (2009)
D. Vollhardt, Rev. Mod. Phys. **56**, 99 (1984)
J. Yang, et al, JPSJ, **79**, 074704, (2010)

New data: schematic plots

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

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

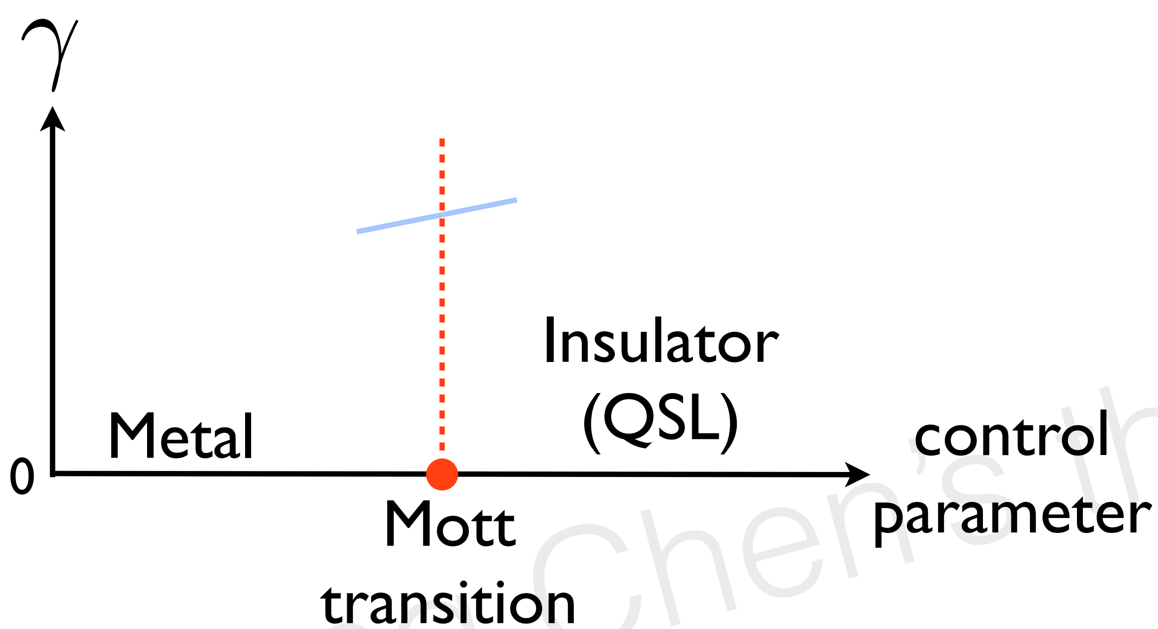


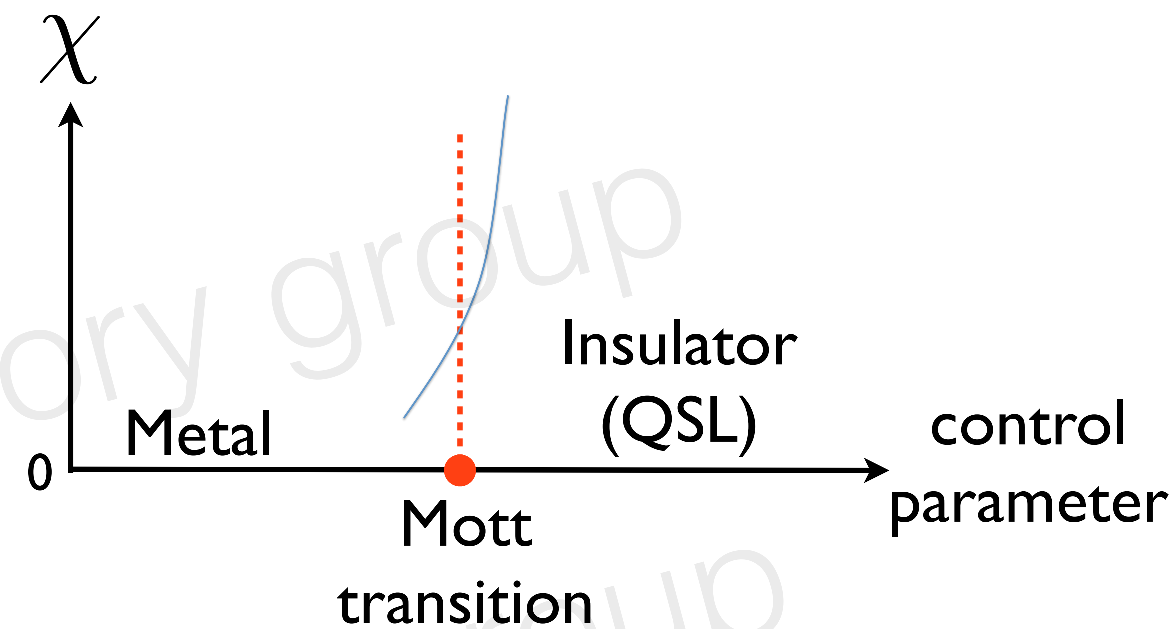
Small change (~18%) in linear-T heat capacity

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

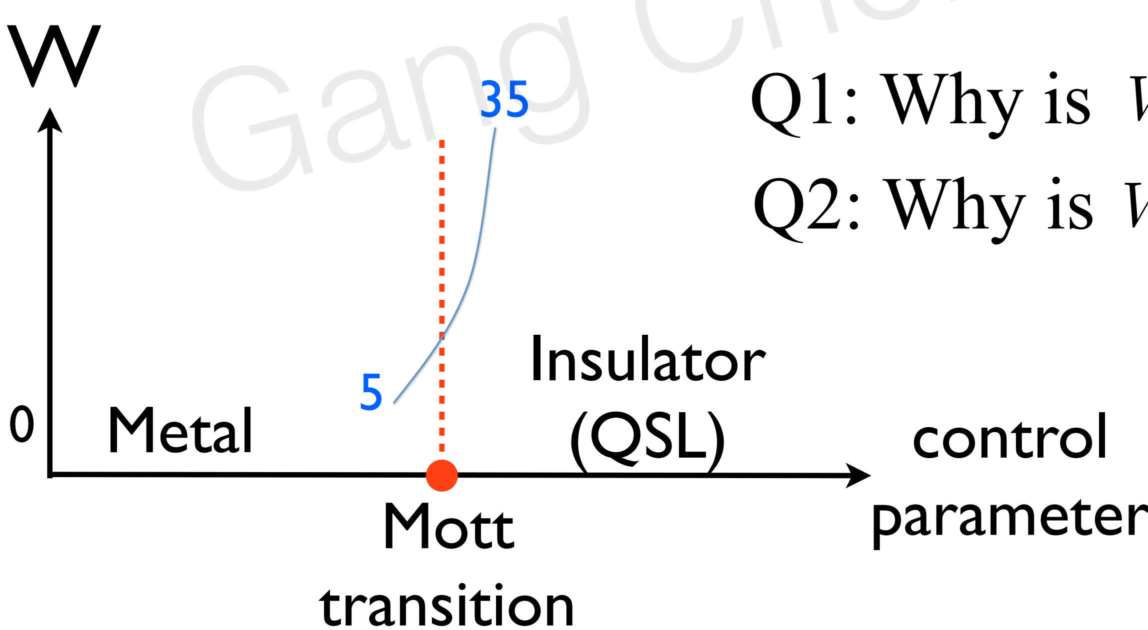
Summary of the experiments: schematic plots

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

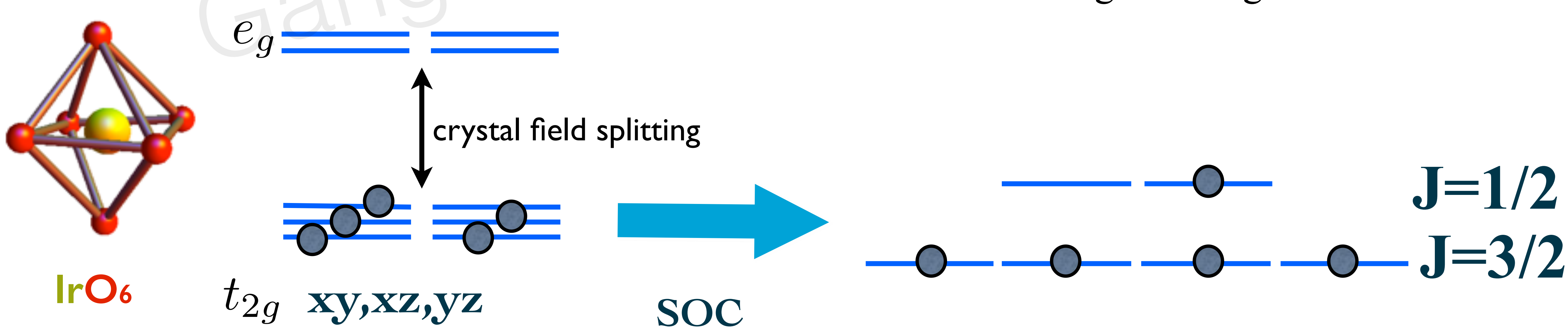
Current theoretical work on $Na_4Ir_3O_8$

U(1) QSL	M. Lawler, et al	Phys. Rev. Lett. **101**, 197202 (2008)
Z_2 QSL	M. Lawler, et al	Phys. Rev. Lett. **100**, 227201 (2008)
	Y. Zhou, et al	Phys. Rev. Lett. **101**, 197201 (2008)
VBS	E. J. Bergholtz, et al	Phys. Rev. Lett. **105**, 237202 (2010)

Other works focus on various other things

John M. Hopkinson, et al	Phys. Rev. Lett. **99**, 037201, (2007)
G. Chen, et al	Phys. Rev. B **78**, 094403, (2008)
D. Podolsky, et al	Phys. Rev. Lett. **102**, 186401, (2009)
T. Micklitz, et al	Phys. Rev. B **81**, 174417, (2010)
M. R. Norman, et al	Phys. Rev. B **81**, 024428, (2010)
D. Podolsky, et al	Phys. Rev. B **83**, 054401, (2011)

Formation of local moment in the strong Mott regime



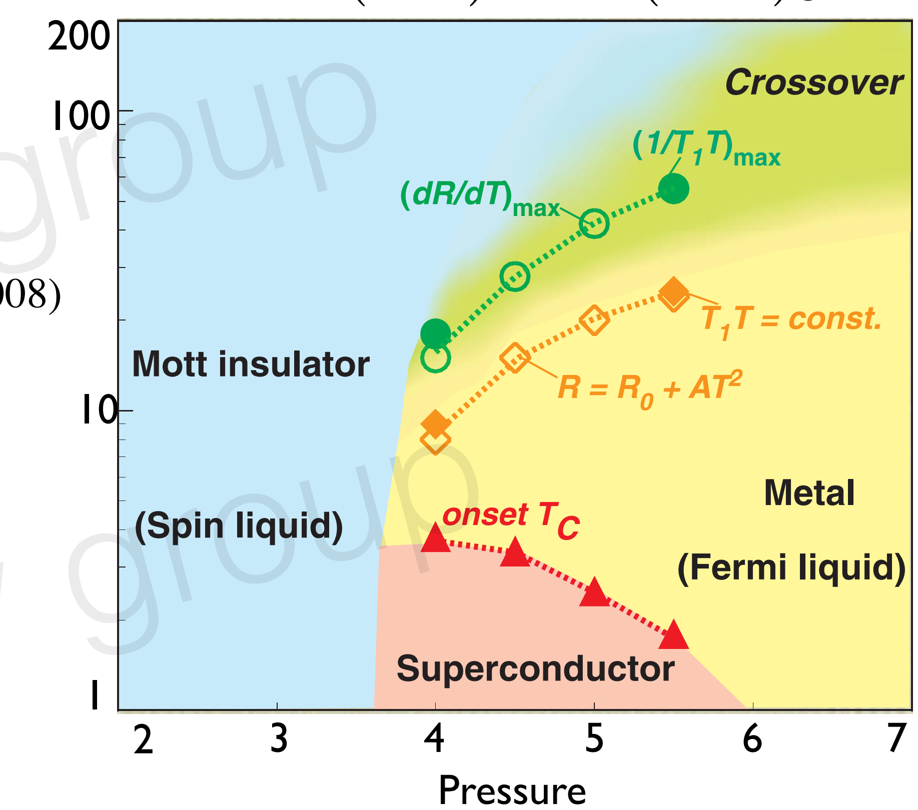
Theoretical proposals

U(1) QSL? M. Lawler, et al 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.

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



K. Kanoda's group 2003-

Z2 QSL (less likely) Y. Zhou, et al 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 suppressed Cv from metal to QSL, and also superconductivity in metallic side just like kappa-ET organics

VBS (less likely) E. J. Bergholtz, et al Phys. Rev. Lett. **105**, 237202 (2010)

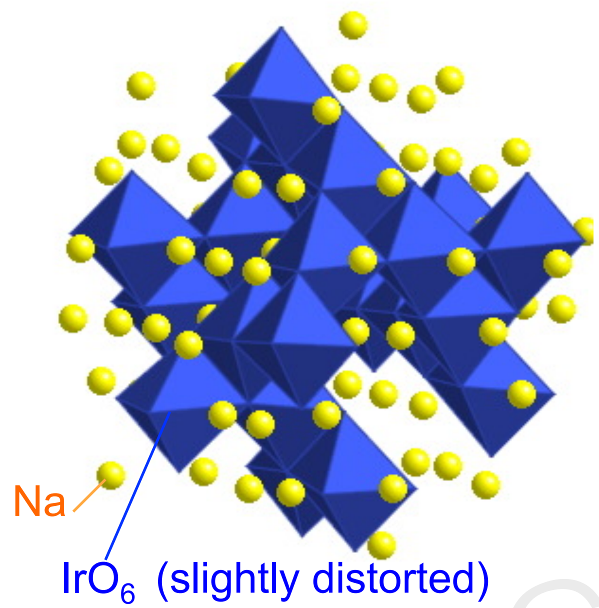
Similar series expansion like Huse+Singh's work on kagome

Complicated ground state: 72 sites in one cell

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

Extended Hubbard Model

$Na_4Ir_3O_8$



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

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

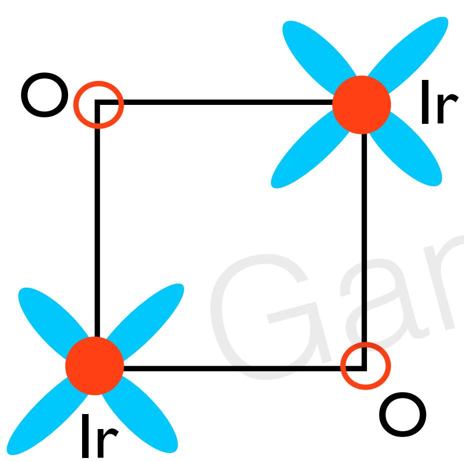
$\mathcal{H}_{soc}$ - Atomic spin-orbit coupling

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

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

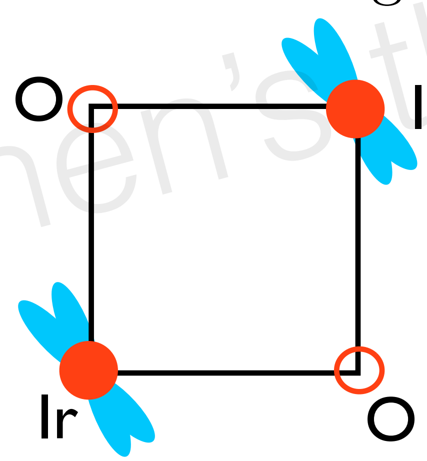
Tight-binding model

$\sigma-$bonding



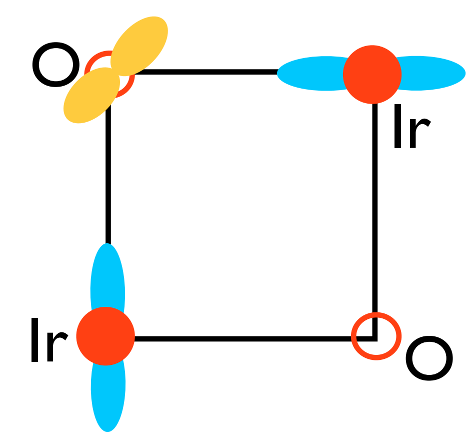
t_σ

$\pi-$bonding



t_π

indirect hop through oxygen



t_2

$$t_\sigma = 1, t_\pi = 0.2, t_2 = 0.5$$

No band insulator

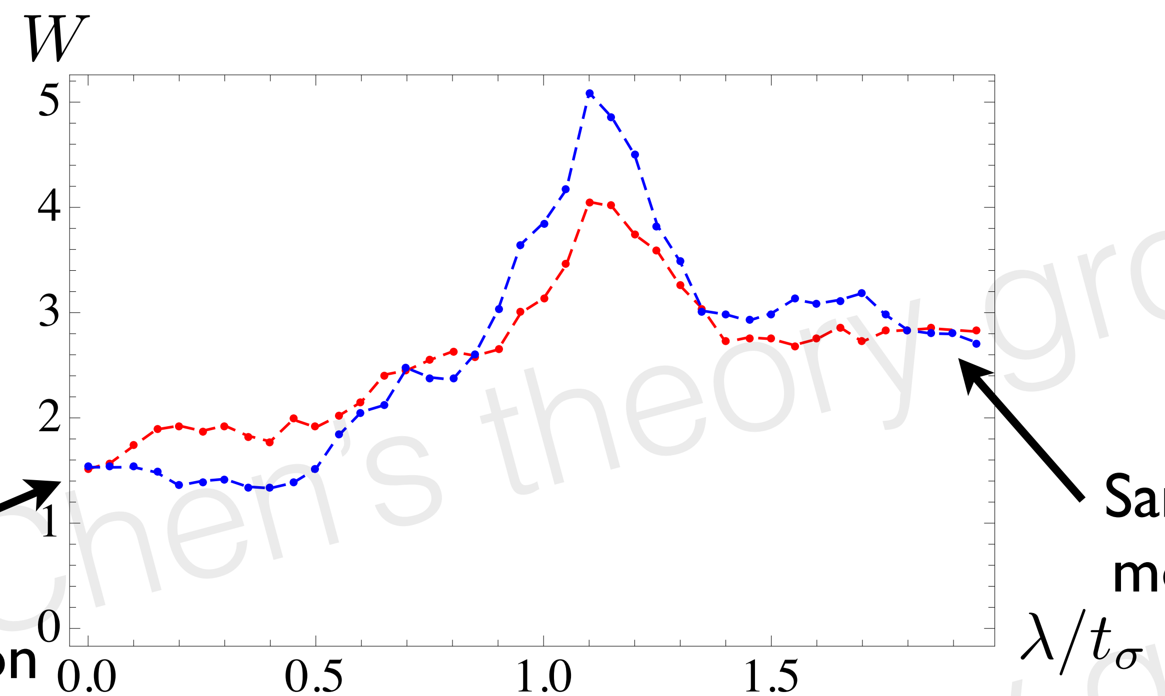
T. Micklitz, et al Phys. Rev. B **81**, 174417, (2010)
M. R. Norman, et al Phys. Rev. B **81**, 024428, (2010)

Wilson ratio for the non-interacting case

$$\mathcal{H}_0 = \mathcal{H}_{hop} + \mathcal{H}_{soc} + \mathcal{H}_{ion}$$

$$t_\sigma = 1, t_\pi = 0.2, t_2 = 0.5$$

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



two anisotropic parameters

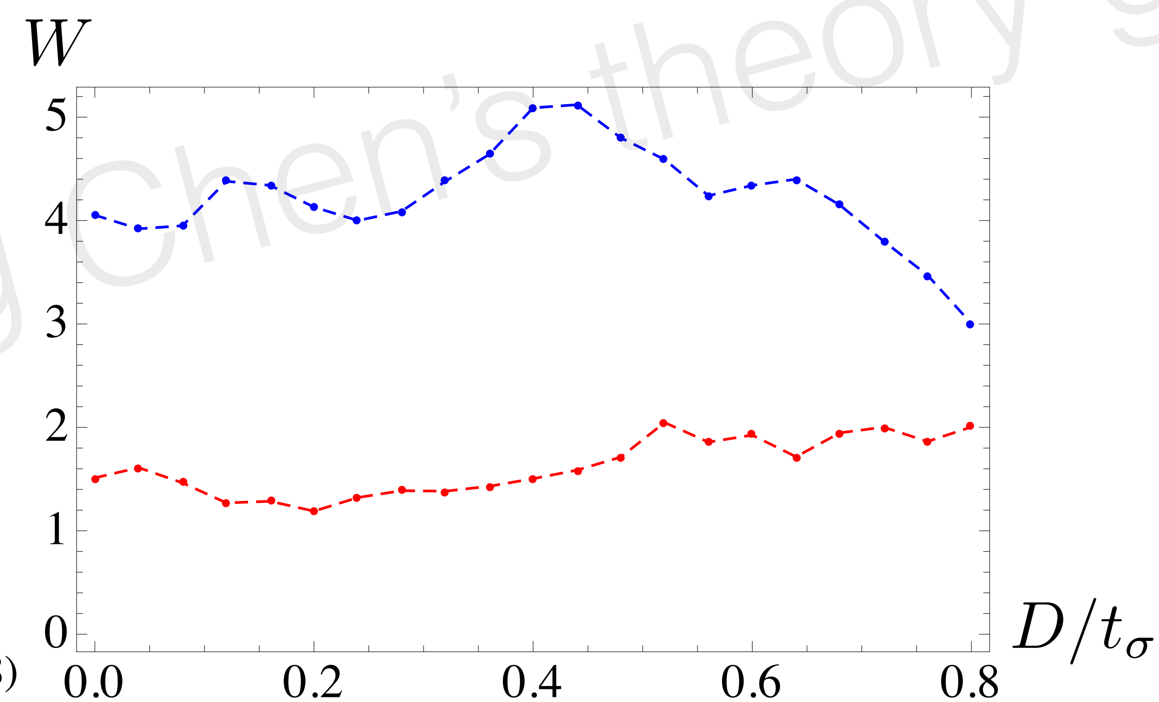
$D = 0$

$D = 0.4$

$W \neq 1$

is because of the hybridization of different orbitals

Same reason why Heisenberg model is relevant for Sr_2IrO_4



$\lambda = 0$

$\lambda = 1.1$

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)

Multi-orbital interactions

$$H_{int} = U\sum_{i,m}\hat{n}_{i,m,\uparrow}\hat{n}_{i,m,\downarrow} + \frac{U'}{2}\sum_{i,m\neq m'}\hat{n}_{i,m}\hat{n}_{i,m'}$$
$$+\frac{J}{2}\sum_{i,m\neq m'} d^{\dagger}_{im\sigma}d^{\dagger}_{im'\sigma}d_{im\sigma'}d_{im'\sigma} + \frac{J'}{2}\sum_{i,m\neq m'} d^{\dagger}_{im\uparrow}d^{\dagger}_{im\downarrow}d_{im'\downarrow}d_{im'\uparrow}$$

i is a position index.

m is an orbital index.

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

$J = J'$

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

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

$$\mathcal{H}_{ex-int} = -J\sum_{i,m\neq m'}\hat{n}_{i,m}\hat{n}_{i,m'} + \frac{J}{2}\sum_{i,m\neq m'} d^{\dagger}_{im\sigma}d^{\dagger}_{im'\sigma}d_{im\sigma'}d_{im'\sigma}$$
$$+\frac{J}{2}\sum_{i,m\neq m'} d^{\dagger}_{im\uparrow}d^{\dagger}_{im\downarrow}d_{im'\downarrow}d_{im'\uparrow}$$

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 mean field: slave-rotor theory

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

Original electron Hamiltonian

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

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

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

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

Slave-rotor approach to obtain fermionic spinons

(see Prof. Senthil's talk)

$$d_{im\alpha} = e^{-i\theta_i} f_{im\alpha}$$

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

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

Slave-rotor mean field Hamiltonian

R is unit cell index

i is sublattice index

$$H_f = Q_f \sum_{Rim,R'i'm'} (t^{ii'}_{mm'} f^\dagger_{im\sigma}(R) f_{im'\sigma}(R') + h.c.)$$
$$+ \frac{\lambda}{2} \sum_{Ri} \mathbf{L}_{mn} \cdot \boldsymbol{\sigma}_{\alpha\beta} f^\dagger_{im\alpha}(R) f_{in\beta}(R) + D \sum_{Ri\alpha} (L^\mu_i)^2_{mn} f^\dagger_{im\alpha}(R) f_{in\alpha}(R)$$

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

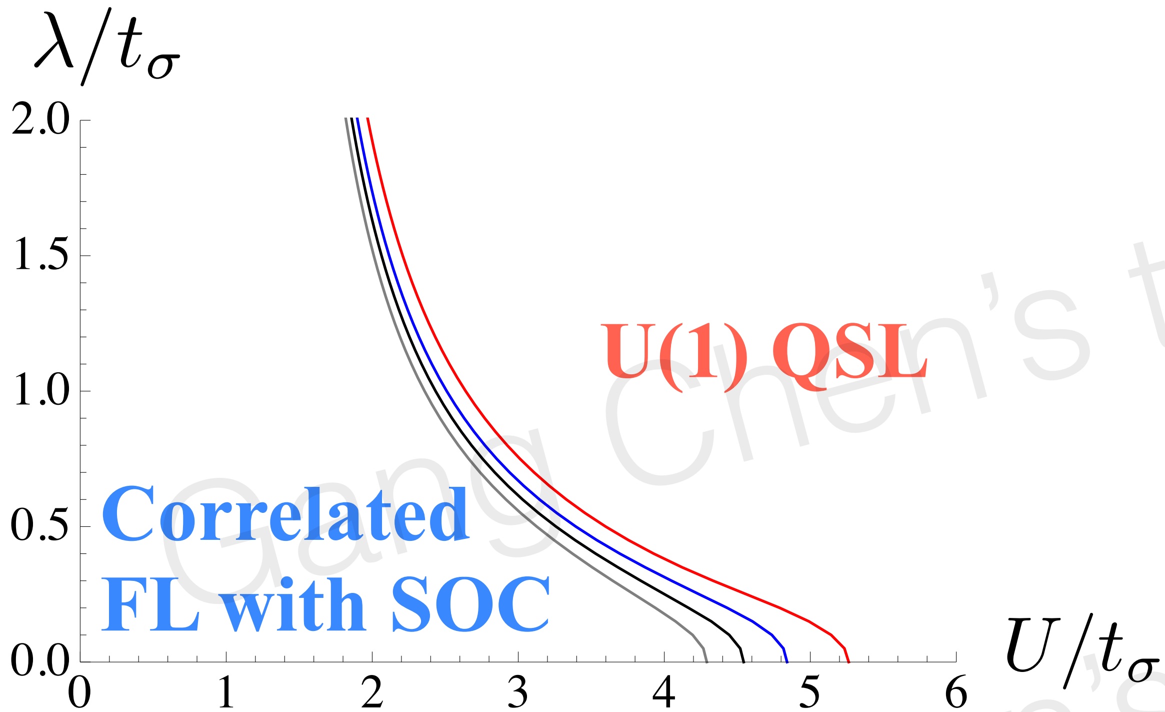
$$Q_f \equiv \langle e^{i\theta_i(R) - i\theta_{i'}(R')} \rangle_\theta \qquad Q_r \equiv \sum_{mm'\sigma} t_{mm'} \langle f^\dagger_{im\sigma} f_{i'm'\sigma}(R) \rangle_f$$

S. Florens and A. Georges,
Phys. Rev. B. **70**, 035114 (2004)

Slave-rotor phase diagram

$\langle e^{-i\theta_i} \rangle \neq 0,\ Z \neq 0,$ 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”.



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 (also see Prof. Balents' talk):

1. SOC enhances correlation effects. $\longrightarrow$ Strong correlation physics may be seen in 4d/5d electron system

2. Correlation effects enhance SOC. $\longrightarrow$ SOC may be also important even in 3d electron system in certain cases: $FeSc_2S_4$, ZnV_2O_4, etc

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

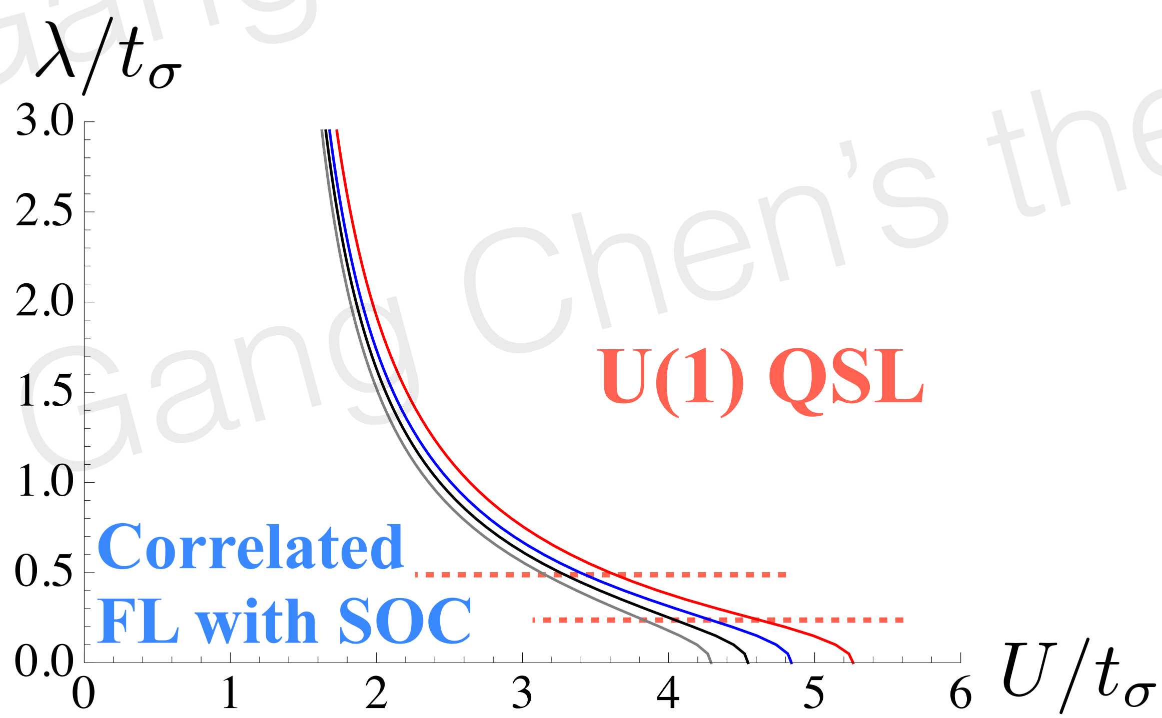
Onsite exchange interaction

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

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

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

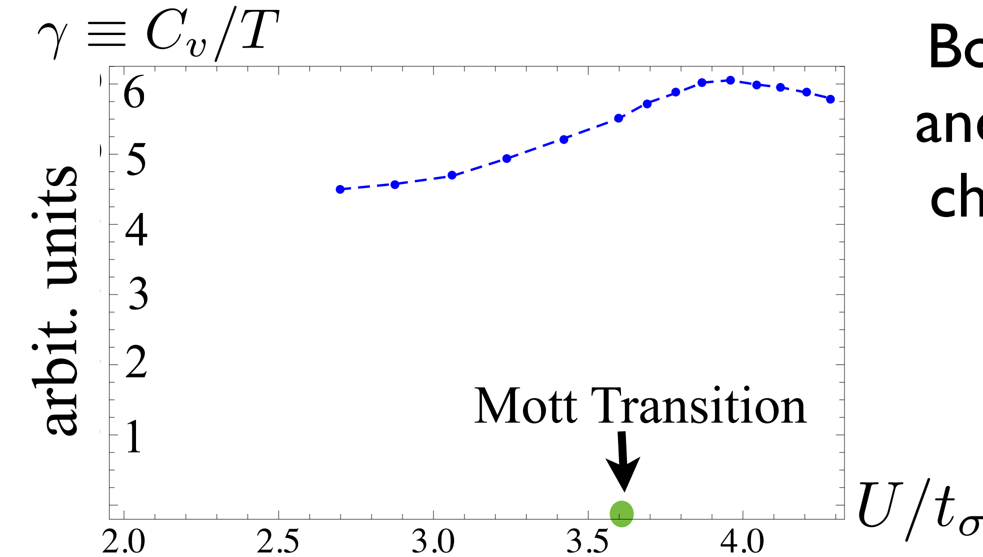
$$H_f \to H_f + H_{ex-int}$$



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

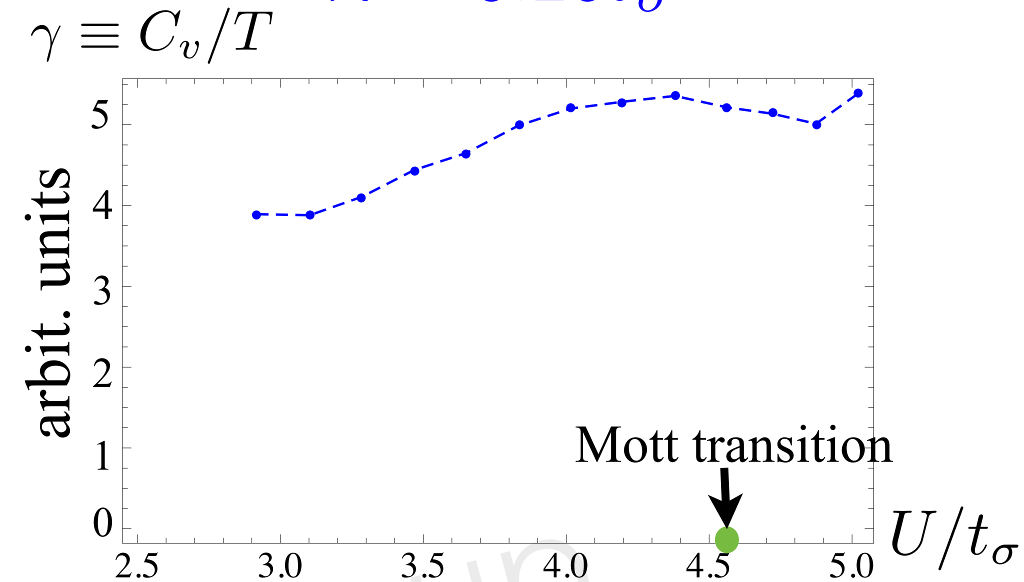
Study Wilson ratio
along the dashed line

$\lambda = 0.5t_\sigma$

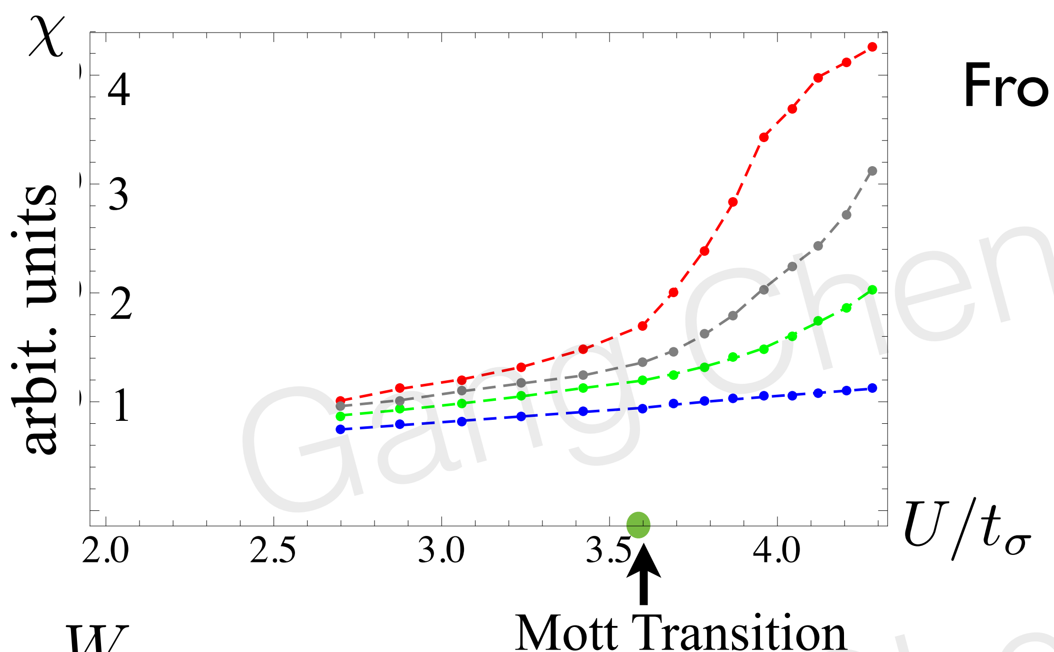


Both "effective mass" and fermi surfaces are changed due to SOC

$\lambda = 0.25t_\sigma$



$$J \lesssim 0.1U_c$$



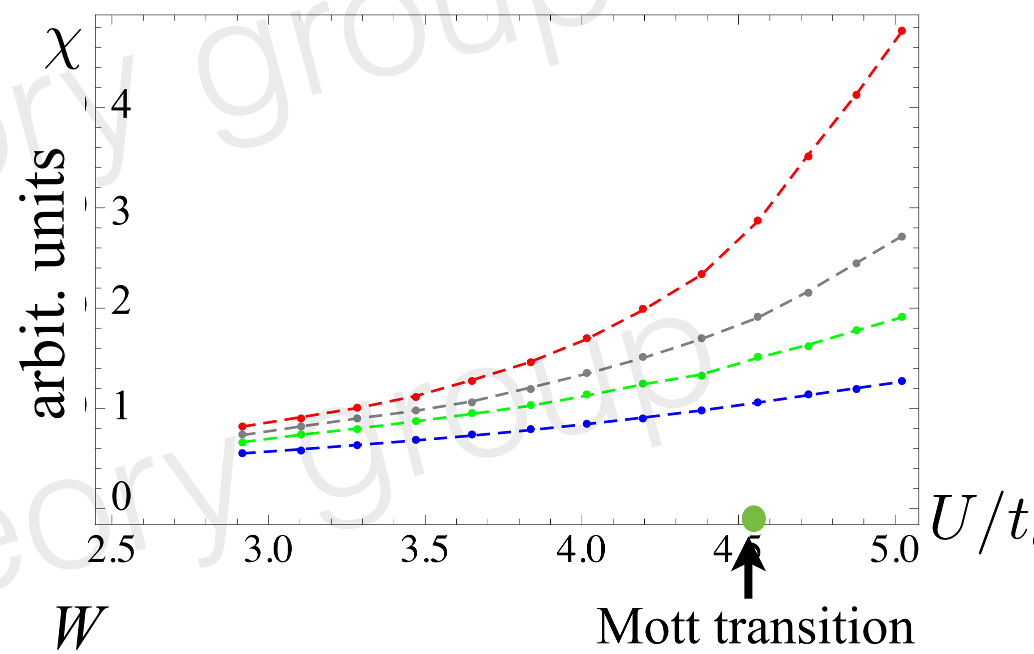
From bottom to top,

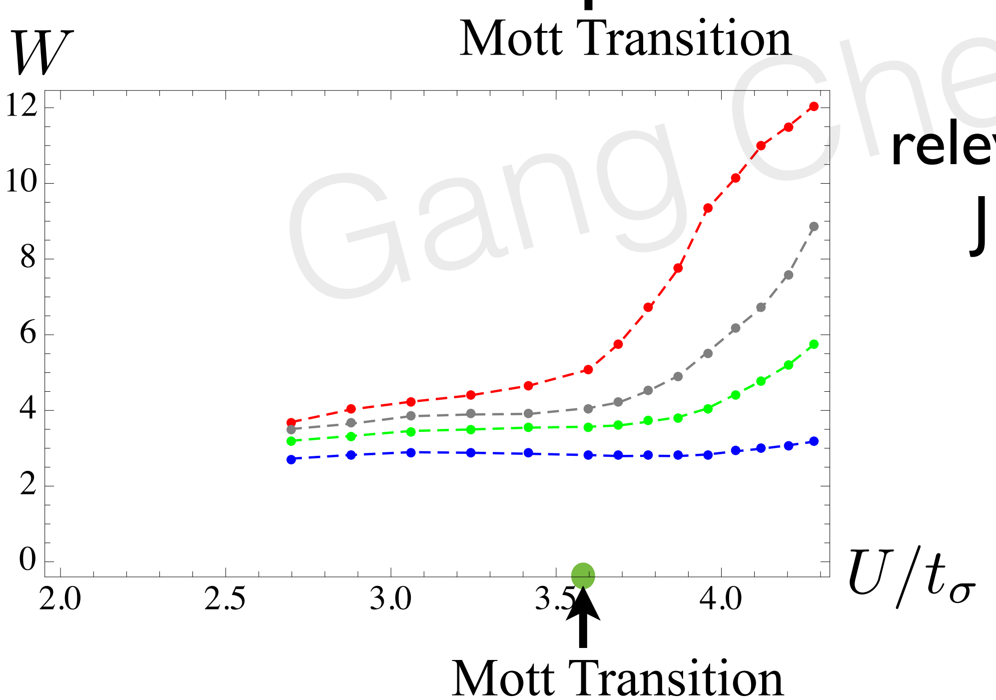
$J = 0$

$J = 0.2t_\sigma$

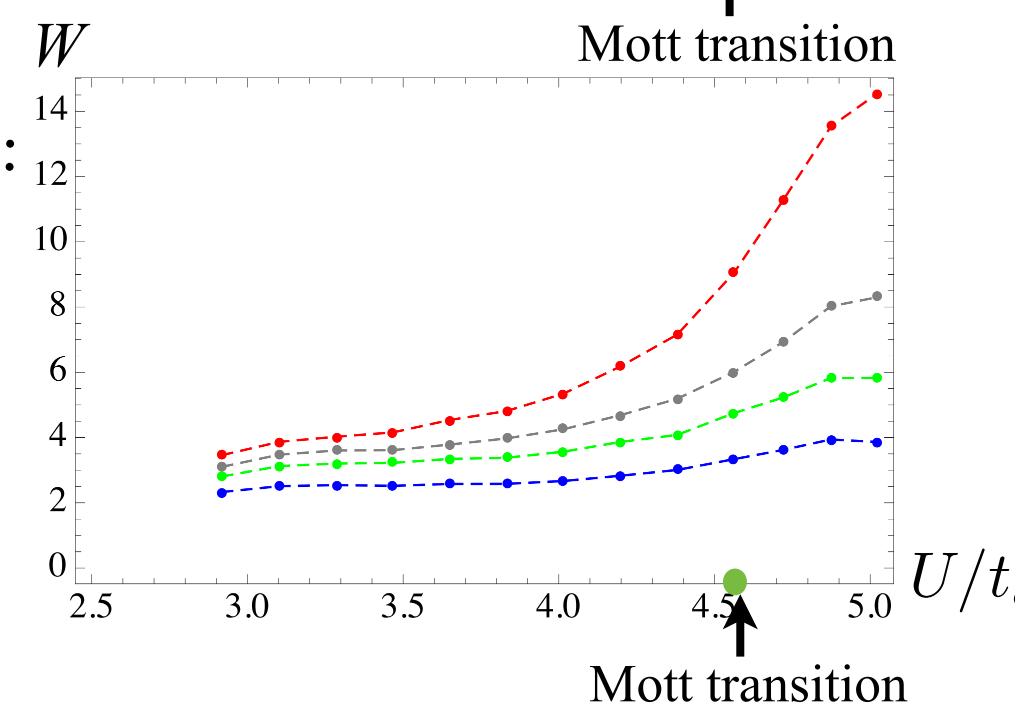
$J = 0.3t_\sigma$

$J = 0.4t_\sigma$





relevant energy scales: J and bandwidth



Summary

$Na_4Ir_3O_8$ 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.
(other possible explanation, gauge fluctuations?)

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 $R_2Ir_2O_7$, Os-compounds, etc