Magnetic properties of a spin-orbit Mott insulator and effects of spin vacancies

GDR MICO, Roscoff, 7/1/2013

Fabien TROUSSELET(1,2), G.KHALIULLIN(2), P.HORSCH(2)
(1): Institut Neel, Grenoble
(2): Max-Planck Institut für Festkörperforschung, Stuttgart

Outline

- A pseudospin model for Mott insulators with strong spin-orbit coupling (alkali iridates)
- Phase diagram in a magnetic field; susceptibility
- Effects of a spin vacancy in ordered & liquid phases
- About the actual spin order in alkali iridates

Spin-orbit coupling in iridates A_2IrO_3 [A=Na,Li,..]

$$H_0 = \lambda \vec{l} \cdot \vec{s}$$

- Ir^{4+} ion: configuration $5d^5$
- Crystal field (O^{2-} octahedron): → 1 hole in t_{2g} subspace
- Spin-orbit coupling λ → low-energy doublet

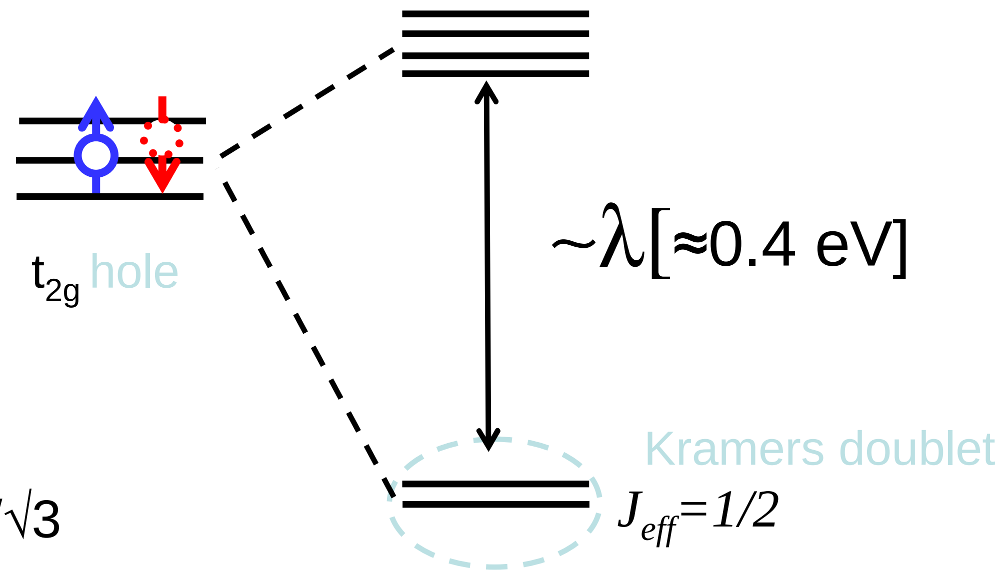
~ pseudospin-1/2 σ

- Eigenstates of σ^z:

$|\sigma^z=1/2\rangle=(|\uparrow,xy\rangle + |\downarrow,xz\rangle+i|\downarrow,yz\rangle)/\sqrt{3}$

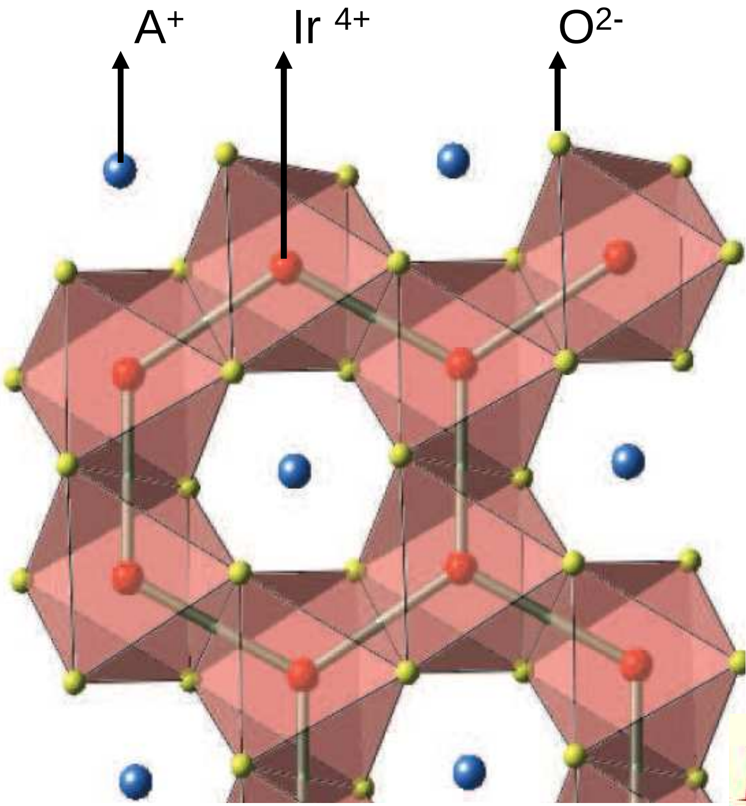
$|\sigma^z=-1/2\rangle=(|\downarrow,xy\rangle + |\uparrow,xz\rangle+i|\uparrow,yz\rangle)/\sqrt{3}$

→ $s_z=-1/2$(, $l_z=0$)



[from G.Jackeli]

Effective model for layered iridates A_2IrO_3



[from : Y.Singh et al, PRB'10]

Ir^{4+} : honeycomb lattice

on-site Coulomb interactions U, J_H

nearest neighbor hoppings
$|t_{Ir\text{-}O\text{-}Ir}|, |t_{Ir\text{-}Ir}| <(<) U$

Effective Hamiltonian for exchange processes :

$$H_0 = \sum_{<\vec{r},\vec{r}'>//\gamma} \left(-2\alpha\sigma^{\gamma}_{\vec{r}}\sigma^{\gamma}_{\vec{r}'} + (1-\alpha)\vec{\sigma}_{\vec{r}}\cdot\vec{\sigma}_{\vec{r}'} \right)$$

$\alpha = f(U, J_H, t_{Ir\text{-}O\text{-}Ir}, t_{Ir\text{-}Ir})$

„Kitaev“ couplings

Kitaev model on the honeycomb lattice

- S=1/2 spins with locally Ising-like interactions
  (! Non-SU(2) invariant):

$$H = -J \ \Sigma_{<i,j>//\gamma} \sigma_i^{\gamma}\sigma_j^{\gamma}$$

„Local easy axis" $\gamma \in \{x,y,z\}$ on γ-labeled bonds

- Exact solution using Majorana fermions

$\sigma_i^{\gamma}=i\, b_i\, c_{i\gamma}$

$b_i=b_i^{+}$, $b_i^2=1$

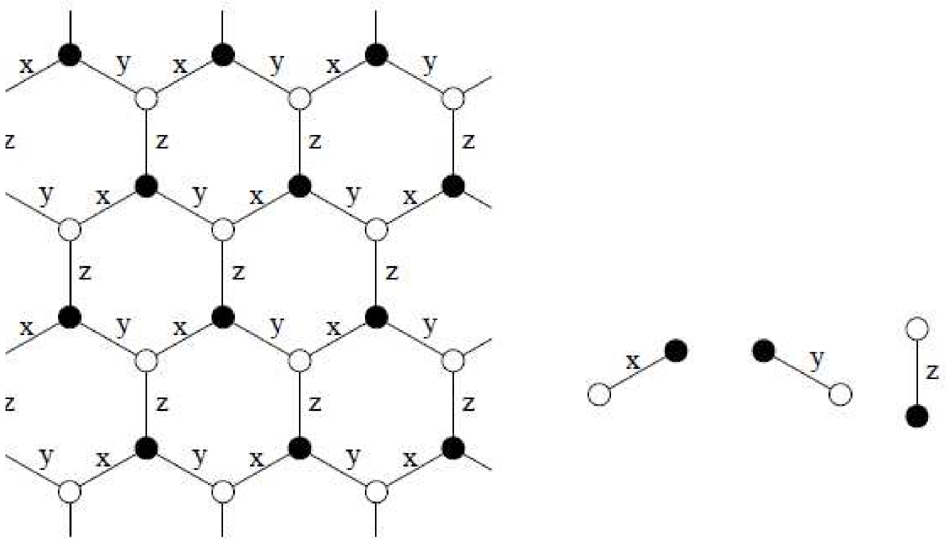
$c_{i\gamma}=c_{i\gamma}^{+}$, $c_{i\gamma}^{2}=1$

$\rightarrow c_{i\gamma}\, c_{j\gamma}$ conserved $\forall$ $<i\ j>$ // γ

- Spin liquid with Dirac-like excitation spectrum

$< \sigma_i^{\gamma}\sigma_j^{\gamma} > \approx 0.53$ for $<i\ j>$ // γ

$= 0$ otherwise

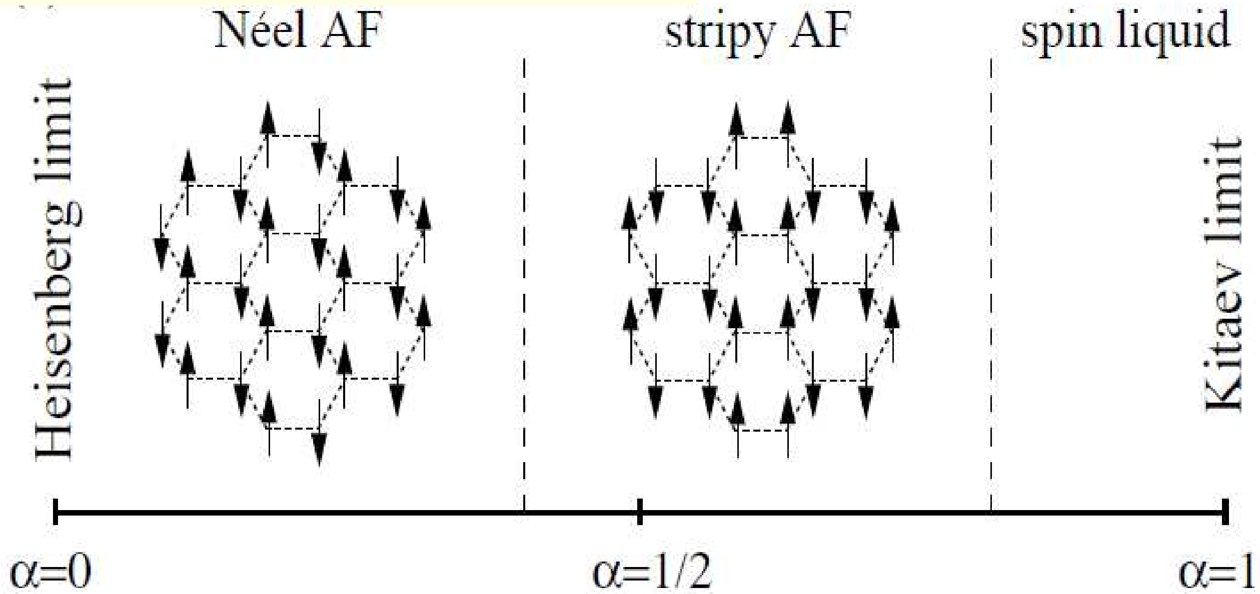


[A.Y.Kitaev, Ann.Phy'06]

Kitaev-Heisenberg model : phase diagram

Interpolation between (AF) Heisenberg and (FM) Kitaev models

$$H_0 = \sum_{<\vec{r},\vec{r}'>//\gamma} \left(-2\alpha\sigma^{\gamma}_{\vec{r}}\sigma^{\gamma}_{\vec{r}'} + (1-\alpha)\vec{\sigma}_{\vec{r}}\cdot\vec{\sigma}_{\vec{r}'} \right)$$



[J.Chaloupka et al., PRL'10]

3 T=0 phases:

- spin liquid [α>0.8]
- Neel phase [α<0.4]
- stripe phase [0.4<α<0.8]:

6 ordered patterns;
„entangled spin direction & ordering wave vector „

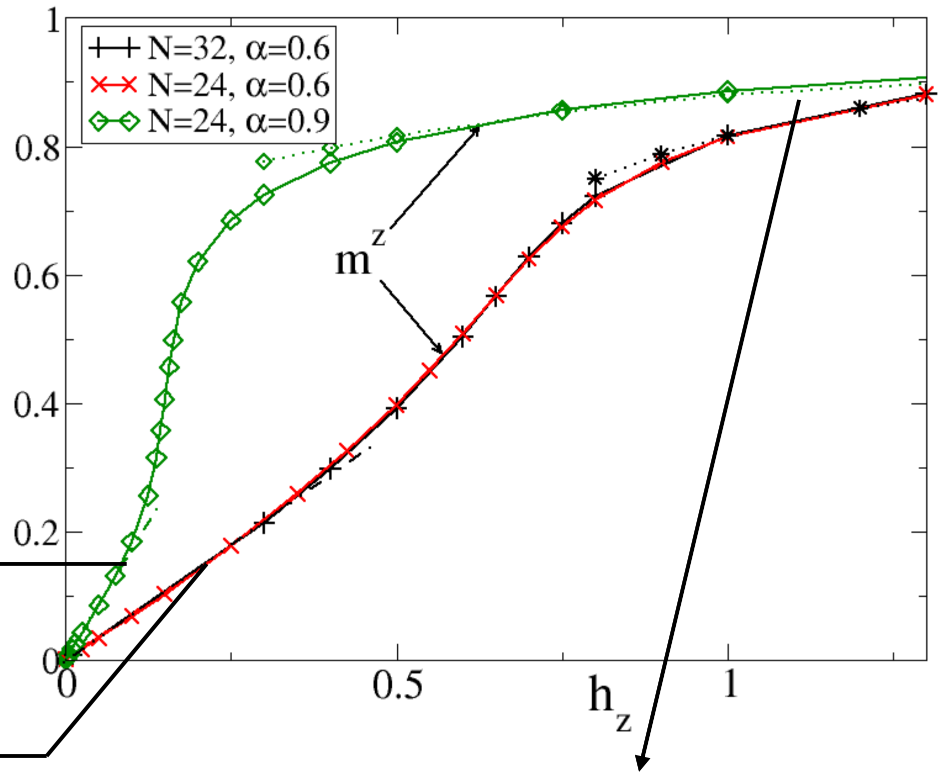
In iridates A_2IrO_3 : AF-order, not Neel, possibly stripe

Effect of a magnetic field

$H = H_0 - \Sigma_r h_\gamma \sigma_r^\gamma$

- Magnetization $m^z = \langle\sigma_{\mathbf{r}}^z\rangle$ in $\mathbf{h}=(0,0,h_z)$:

low- vs high-field regimes



$m^z \sim \chi^{\text{liquid}}\, h_z$

$m^z \sim \chi^{\text{stripe}}\, h_z$

spin canting mechanism

$(\langle\sigma_{\mathbf{r}}\rangle, \mathbf{h}) \sim \pi/2 - \chi.|h|$

$m^z \sim 1 - (\alpha/[h_z+4\alpha-2])^2$

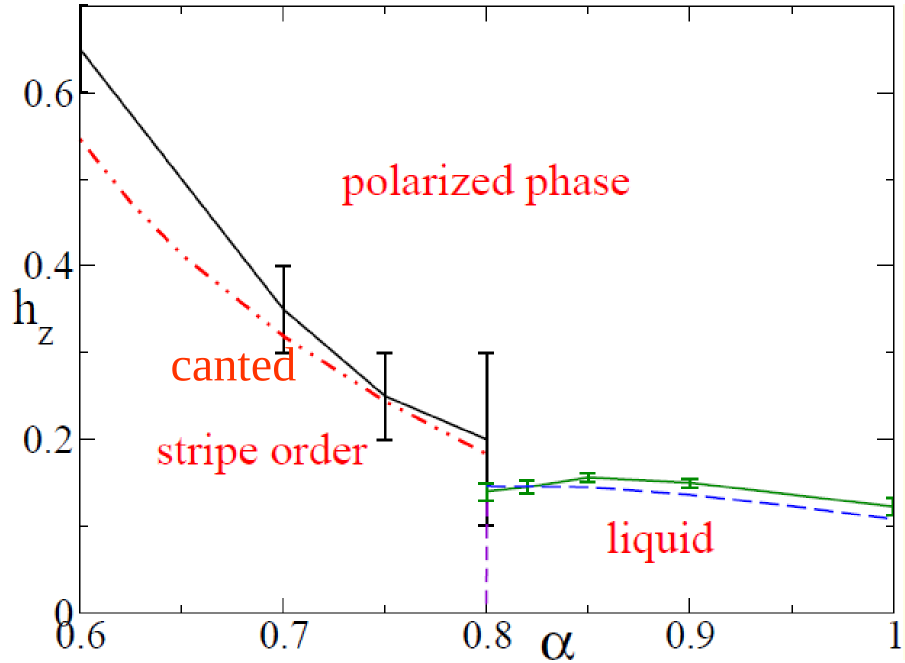
full polarization – qu.fluctuations

Phase diagram of K-H model in field (**h** // [001])

Determination of transition fields h_c:
Numerical & analytical estimates
[classical + 2nd order perturbation]

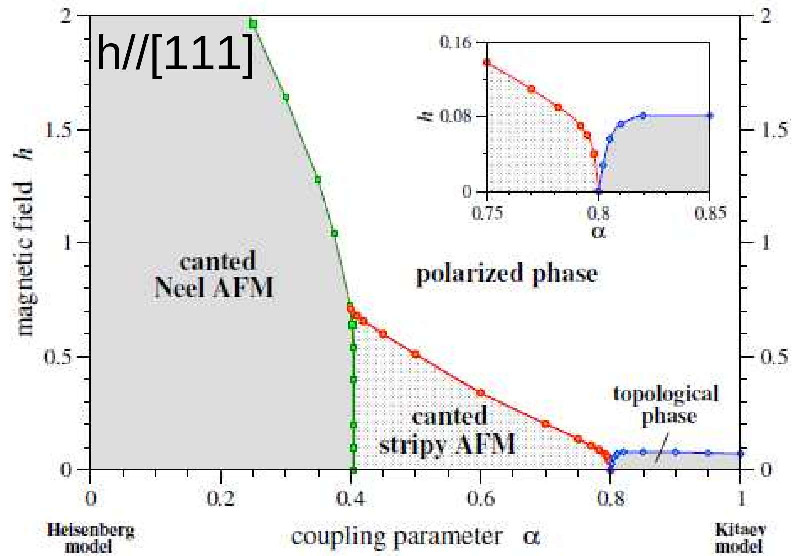
Frustration increases with α

[F.T. et al., PRB 84, 094405 (2011)]



NB: [111]-field → Q.C.P. at (α~0.8, h=0)
Connected to topological phase for α>0.8,
0<h<<1

[H.C. Jiang et al., PRB **83**, 245104 (2011)]



Susceptibility in the stripe phase

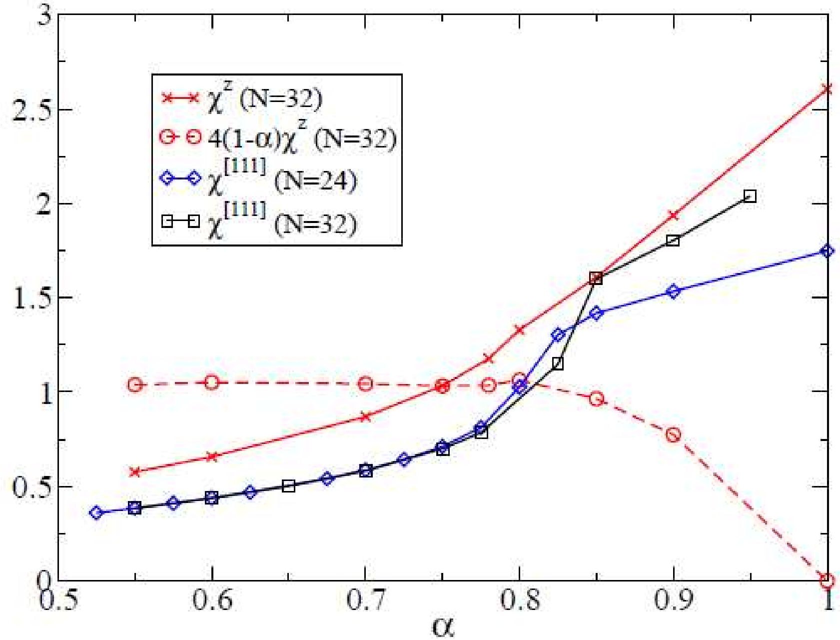
- T=0, h=0 susceptibility:
- Classical result (canted stripe phase) :

$$\chi \sim c/(1-\alpha)$$

h // [001] : c=1/4
h // [111] : c=1/6

Angular dependance of $\chi \leftrightarrow$
$\langle \sigma_r \rangle$ // x,y or z in stripe GSs

- Agreement with Exact Diagonalization results



[F.T. et al, PRB 84, 054409(2011)]

Allows comparison with χ measured in Na_2IrO_3 & Li_2IrO_3 ($T \rightarrow 0$):

consistent with AF stripe order
α(Na)≤ 0.6 , α(Li)≥ 0.7

Vacancy-induced magnetization: stripe phase

- Zero field: 6 degenerate patterns

[x-, y- or z- stripes; $\boldsymbol{\sigma}_r \rightarrow \boldsymbol{\sigma}_r$ symmetry]

- Small field h (//[001]):

  1 pattern selected

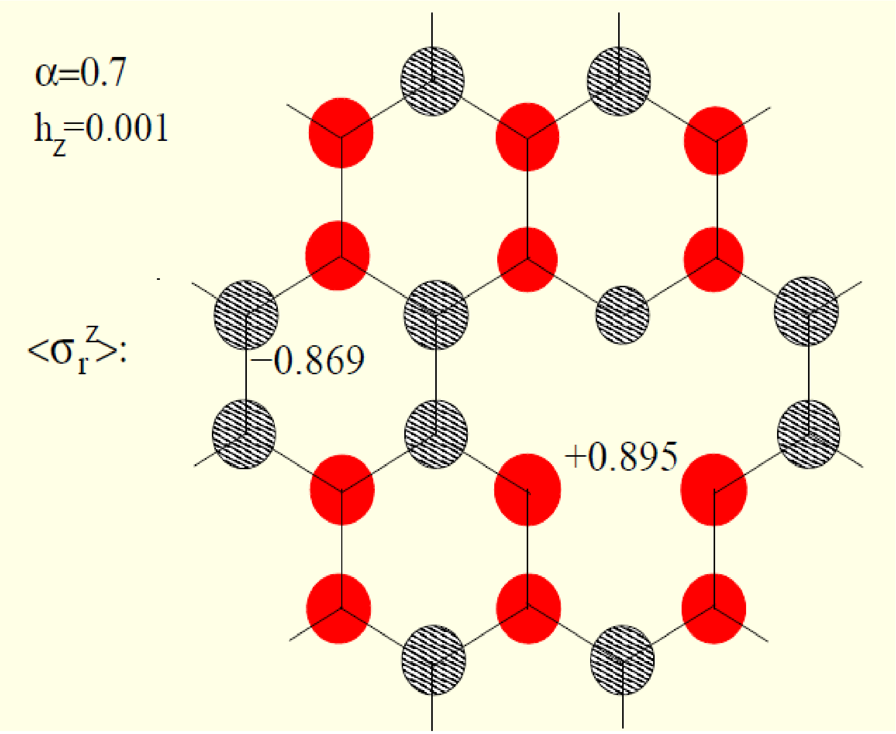
[$<\boldsymbol{\sigma}_r>$ //z, m^z_{total}=+1]

⇨ spin canting locally blocked

- No such mechanism for h//[111]

⇨ could account for $\chi^{[111]} > \chi^{[001]}$ at low T

(opposite to model predictions without vacancies)



Vacancy-induced magnetization: liquid phase

- $\alpha=1$, $|h_z|<<1$:

anomalous vacancy-induced

magnetization $m_z \sim h_z \ln(h_z)$

mostly on the z-neighbor

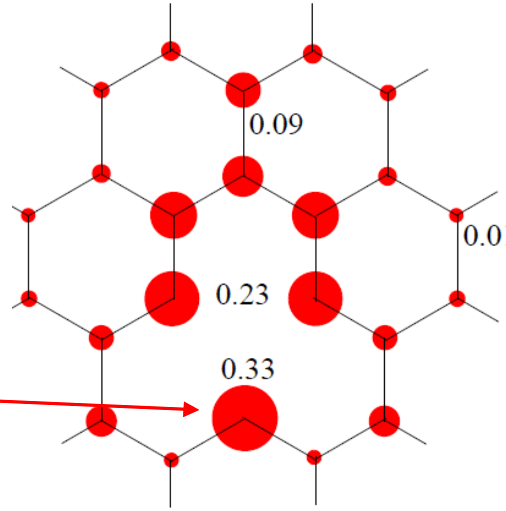
[A.Willans et al. PRL'10]

Effect of finite perturbations :

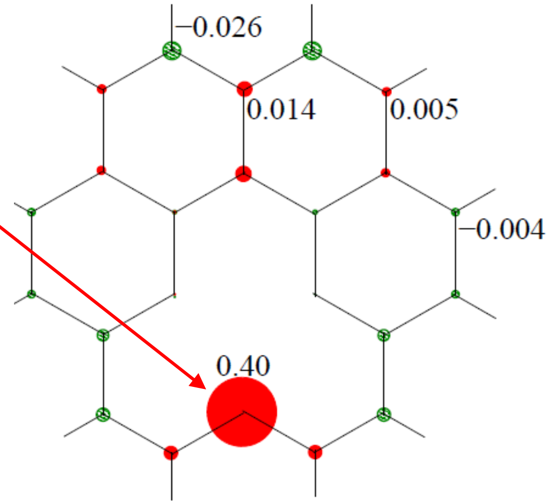
magnetization induced ($\delta\sigma_j^z$)

on further neighbors

[$\leftrightarrow$ $<\sigma_i^z\sigma_j^z>$ pattern without vacancy]



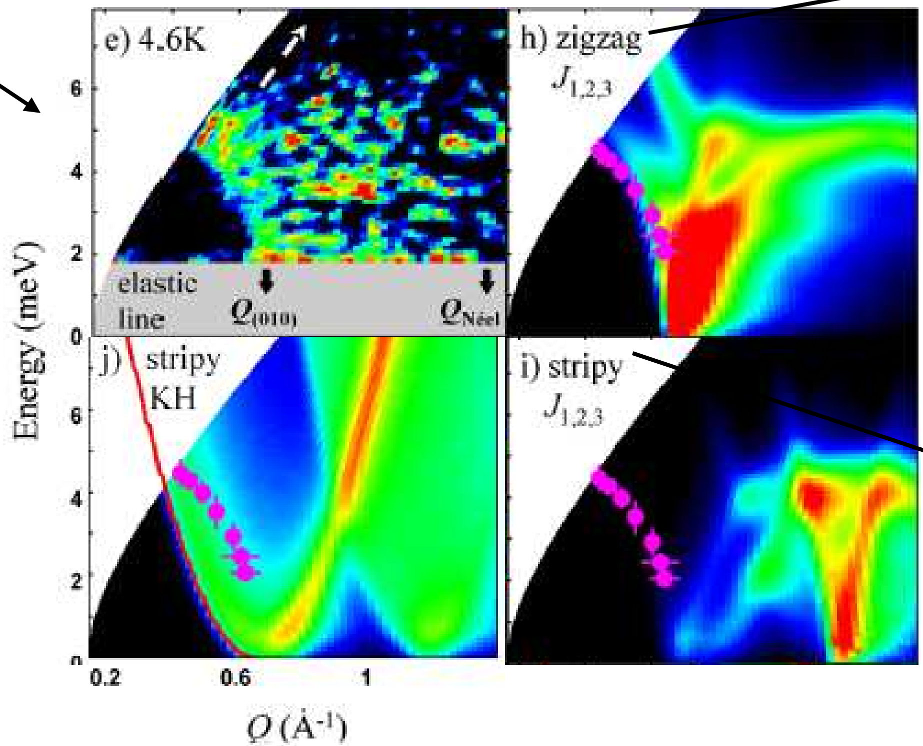
$\delta\sigma_j^z$: $h_z = 0.05$, $\alpha = 1$

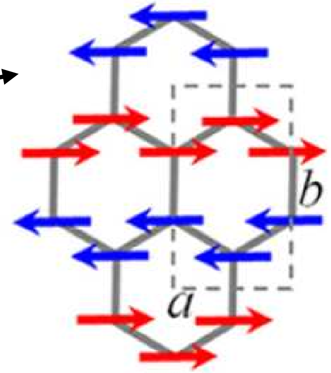


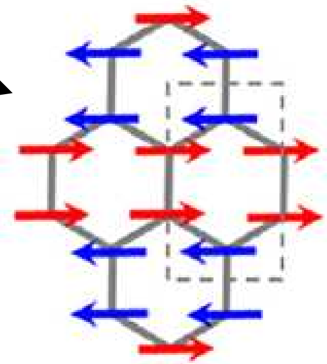
$\delta\sigma_j^z$: $h_z = \varepsilon$, $\alpha = 0.9$

Zigzag-type AF order evidenced in Na2IrO3

Recent experiments on Na2IrO3
(inelastic neutron scattering,
RIXS)







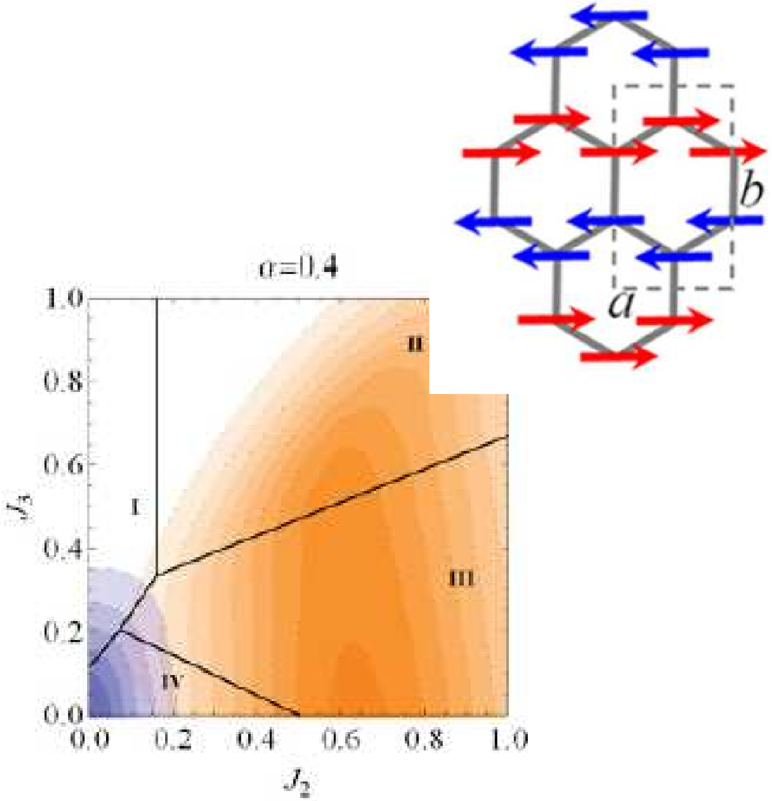
[S. Choi et al., PRL **108**, 127204 (2012)]

Incompatibe with low-T stripe order;

Compatible with zigzag(AF) order, beyond KH model

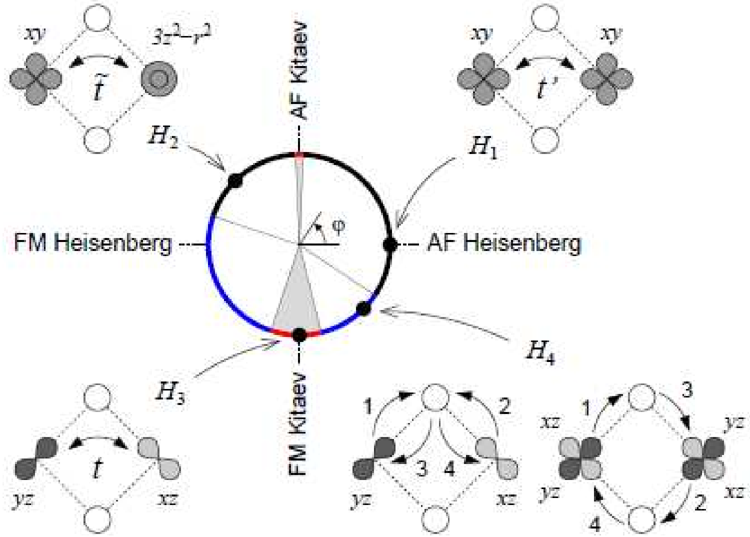
Possible scenari for the zigzag order

- Further neighbor couplings



[I.Kimchi & Y.Z.You, PRB **84**, 180407 (2011)]

- KH model.. with reversed signs for both types of couplings



Clue: e_g orbitals involved in exchange processes

[J.Chaloupka et al., 1209.5100]

Conclusions

- Kitaev-Heisenberg model for layered iridates A_2IrO_3
  → spin liquid & exotic AntiFerro (stripe) phases

- Pecularities of spin canting in the stripe phase
  (→anisotropy in spin susceptibility)

- Spin density around a vacancy
  → characterization of order via NMR (or spin-polarized AFM) ?

- Features also expected in the zigzag-type AF phase lastly evidenced in Na_2IrO_3

THANK YOU!

Interplay between vacancies in the stripe phase

- 2 neighbor vacancies:

Broken rotational symmetry
→ stripe patterns inequivalent

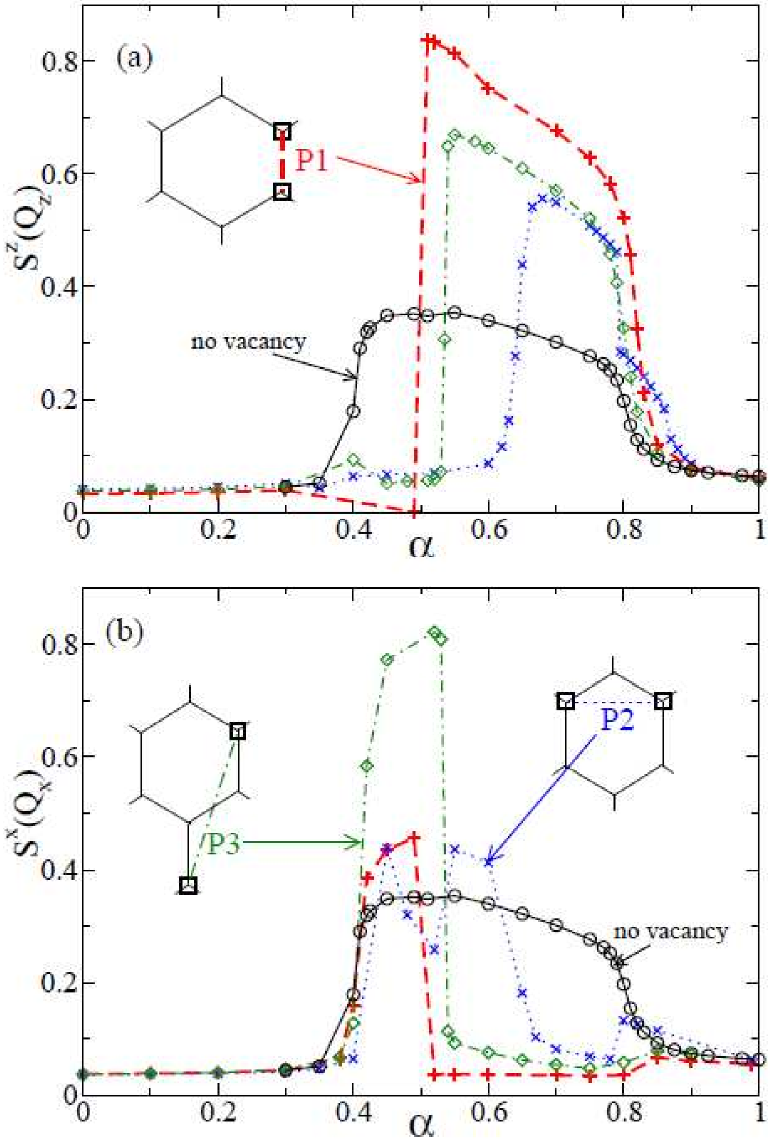
- 2 vacancies on a z-bond („P1“):

Energy difference between patterns
$\delta E = E_z - E_{x,y} \approx 2\text{-}4\alpha$
[E_γ ↔ pattern with spins // γ]

⇒ z-oriented stripes iff α>1/2
Corresponding structure factor

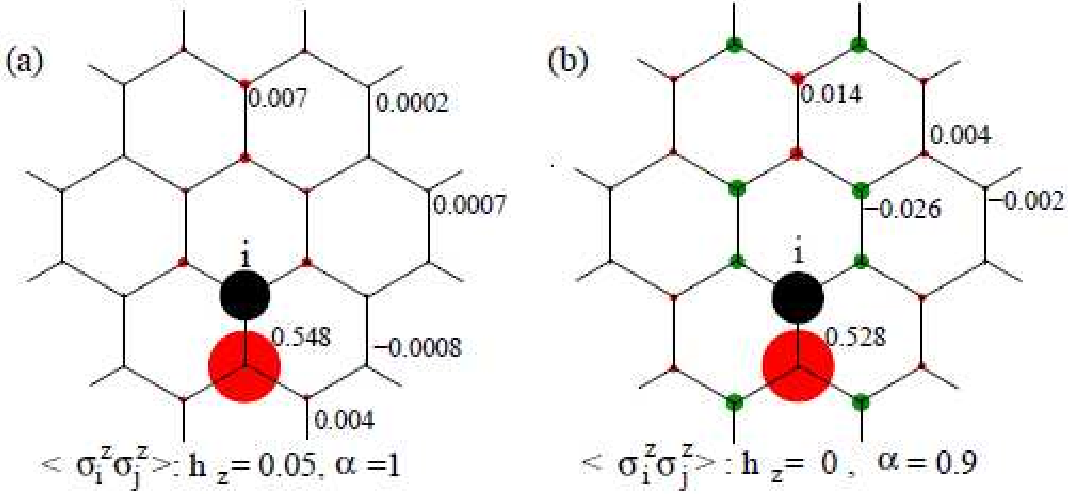
_Generalizable to further neighbor vacancies

_No magnetic field needed



Magnetization/Correlation patterns in the liquid phase

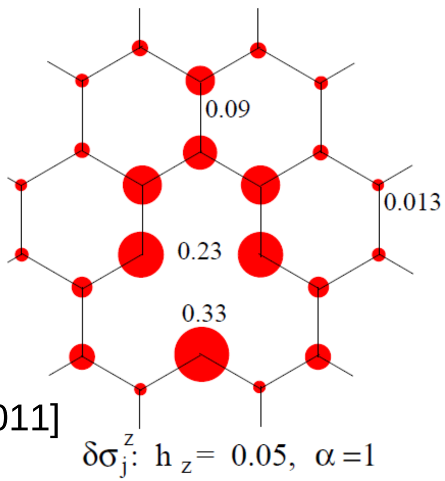
- Spin correlations between reference site i and site j:

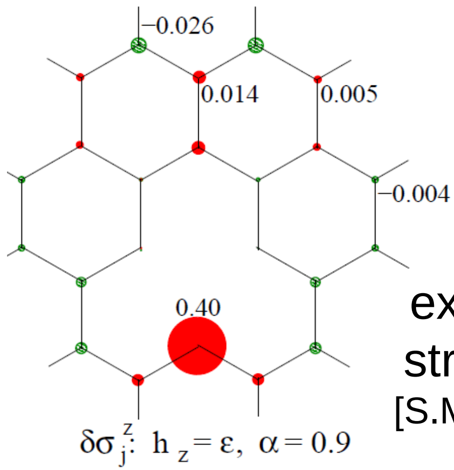


~ similar pattern as $<\sigma_j^z>$ with vacancy at i

Consistent with power-law decay + oscillations

[K.S. Tikhonov et al., PRL 2011]





Consistent with exponential decay + stripe-like oscillations

[S.Mandal et al., PRB 2011]