

Title: Kitaev spin liquids in spin-orbit coupled correlated materials

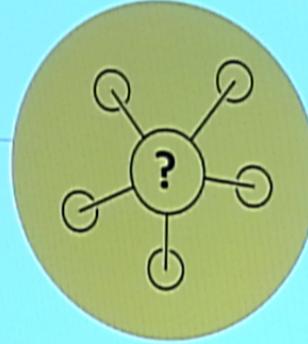
Speakers: Hae-Young Kee

Series: Condensed Matter

Date: May 14, 2019 - 3:30 PM

URL: <http://pirsa.org/19050020>

Abstract: Recently, a new family of correlated honeycomb materials with strong spin-orbit coupling have been promising candidates to realize the Kitaev spin liquid. In particular, a half-integer quantized thermal Hall conductivity was reported in alpha-RuCl₃ under the magnetic field. Using a generic nearest neighbour spin model with bond-dependent interactions, I will present numerical evidence of an extended regime of quantum spin liquids. I will also discuss how to achieve a chiral spin liquid near ferromagnetic Kitaev interaction in the presence of the magnetic field leading to the quantized thermal Hall conductivity.



Quantum Emergence

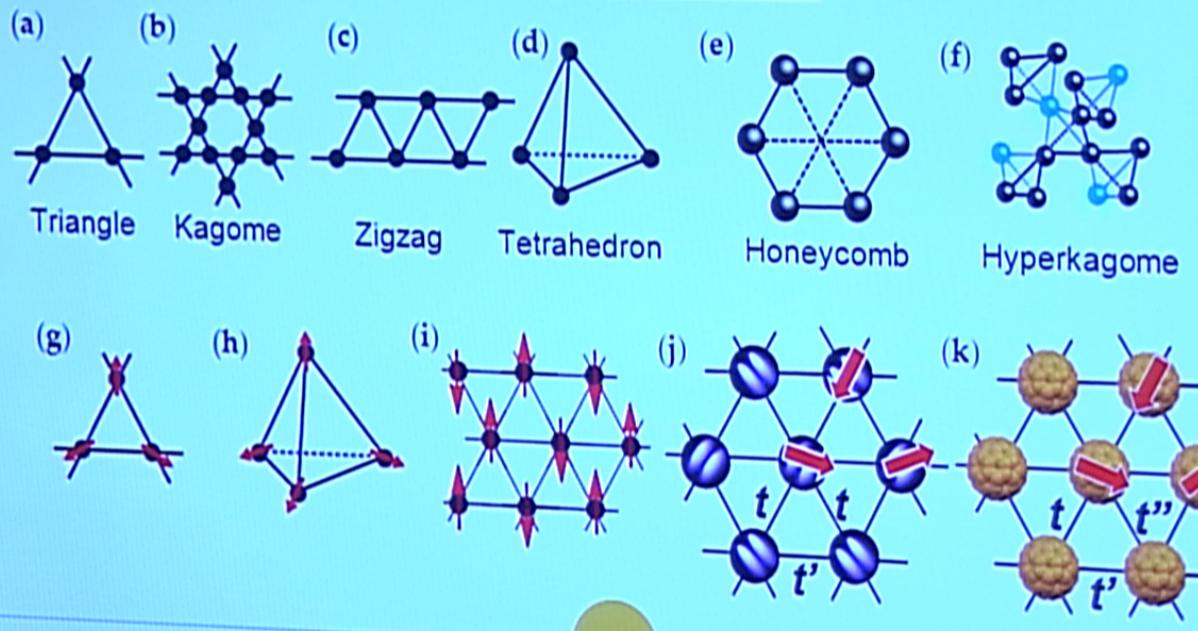
A new emergent quantum particle,
different from the boson and fermion

Spin liquids

7

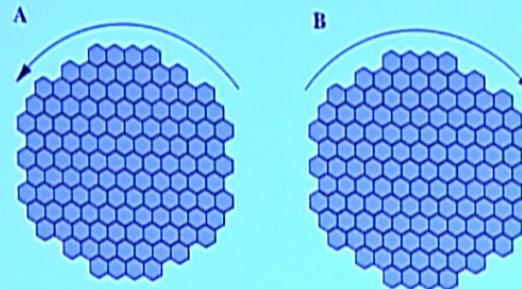
↳

**Frustrated magnetic (no magnetism) insulators:
many shapes and own characters**





smoking-gun signature



Chiral edge mode : $1/2$ quantized thermal Hall conductivity

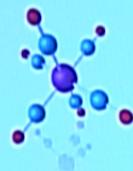
Chiral edge modes can carry energy, leading to potentially measurable thermal transport. (The temperature T is assumed to be much smaller than the energy gap in the bulk, so that the effect of bulk excitations is negligible.) For quantum Hall systems, this phenomenon was discussed in [56,57]. The energy current along the edge in the left (counter-clockwise) direction is given by the following formula:

$$I = \frac{\pi}{12} c_- T^2, \quad (57)$$

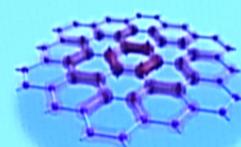
A. Kitaev, Annals of Physics 321, 2 (2006):
Anyons in exactly solved model and beyond

Topological quantum computation

Quantum
Information



Anyon?



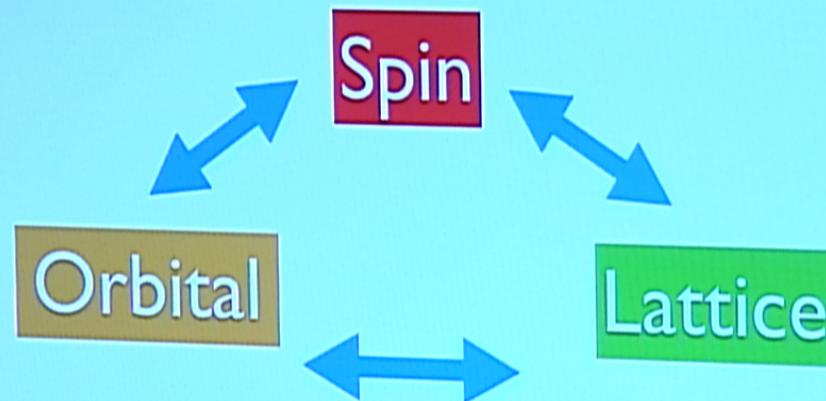
Quantum Materials



Where to find Anyons?

**How to realize KSL in
Solid-State Materials ?**

Solid State Systems

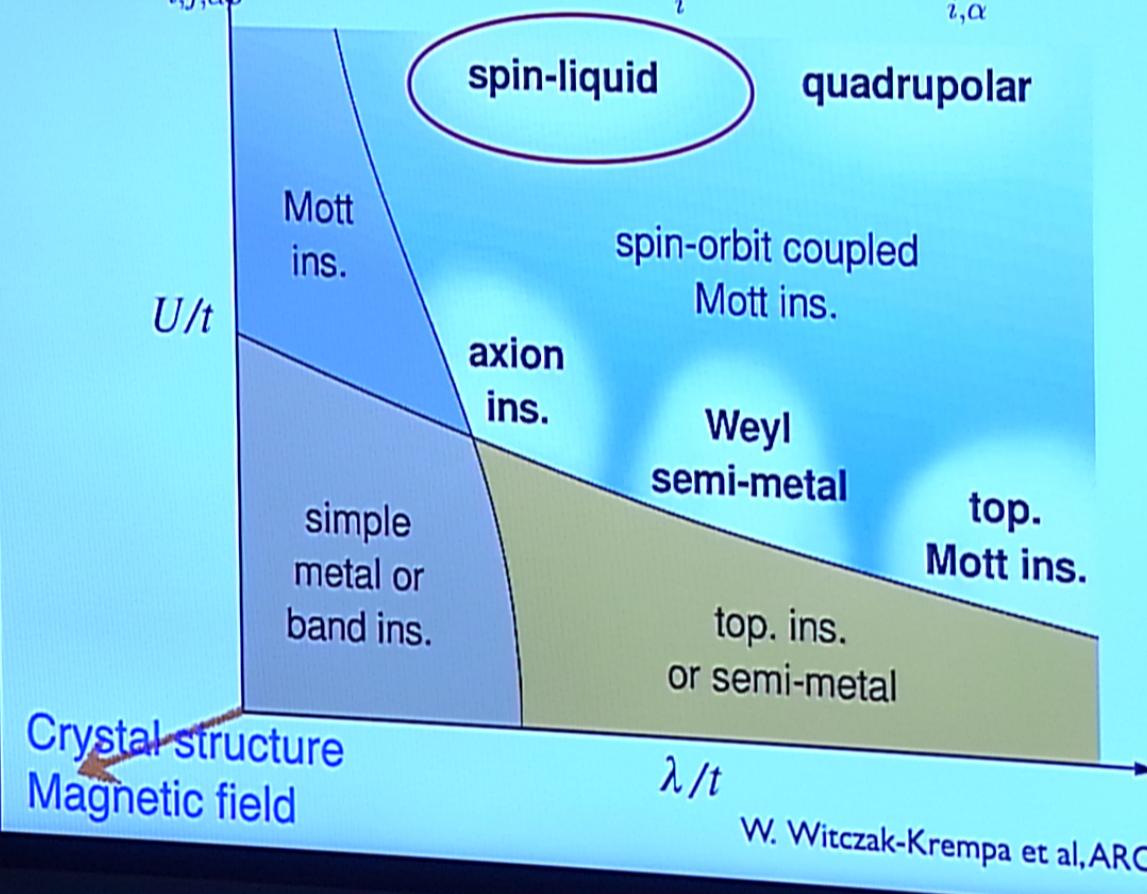


spin-orbit coupling in solids;
strong electron-electron interaction

Charge Insulator

↳ Correlation & spin-orbit coupling

$$H = \sum_{i,j;\alpha\beta} t_{ij,\alpha\beta} c_{i\alpha}^\dagger c_{j\beta} + \text{h.c.} + \lambda \sum_i \mathbf{L}_i \cdot \mathbf{S}_i + U \sum_{i,\alpha} n_{i\alpha} (n_{i\alpha} - 1) ,$$

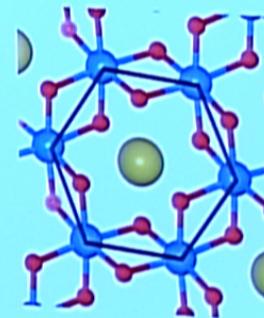


1	H														2	He
3	Li	4	Be													
11		12														
19	20															
K	Ca															
37	39															
Rb	St															
55	56															
Cs	Ba	*														
87	89	*														
Fr	Ra	*														
21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	
Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr	
39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	
Y	Zr	Nb	Mo	T	Ru	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe		
71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	
Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn	
103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118	
Lr	Rf	Db	Sg	Bh	Hs	Uun	Uuu	Uub	Uut	Uuq	Uup	Uuh	Uus	Uuo		
57	58	59	60	61	62	63	64	65	66	67	68	69	70			
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Trm	Yb			
89	90	91	92	93	94	95	96	97	98	99	100	101	102			
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No			

SOC + Large U/t + honeycomb structure

* candidates for Kitaev spin liquid

Iridium (5d): Na_2IrO_3 , Li_2IrO_3
Ruthenium (4d): RuCl_3



↳
Insulator with strong spin-orbit coupling: spin model

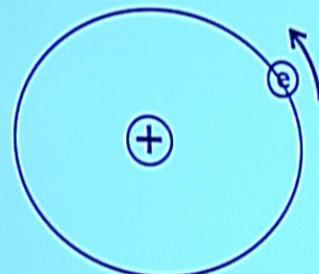
Kitaev spin liquid with strong spin-orbit coupling



- A. Spin-orbit coupling in a lattice
- B. Generic spin model in materials
- C. Field revealed Kitaev spin liquid

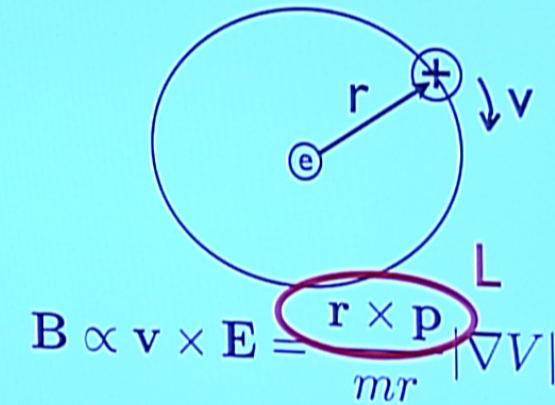
A. Spin-orbit coupling: relativistic effects

Atomic SOC



$$\mathbf{E} \propto -\nabla V$$

In the rest frame of e



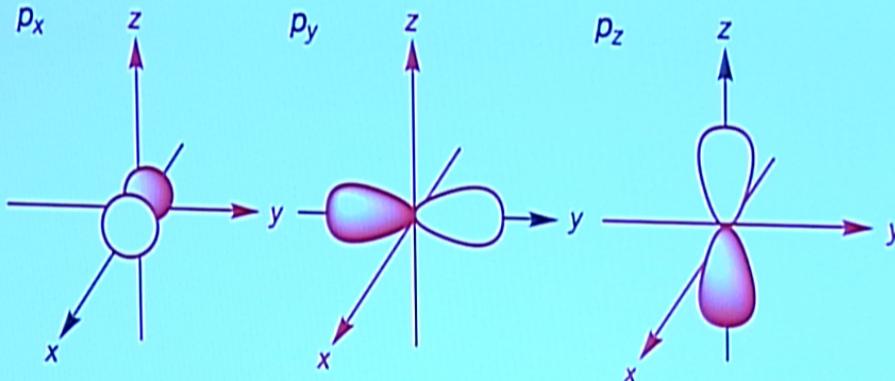
$$\rightarrow \mathbf{B} \cdot \mathbf{m}_s \propto \mathbf{L} \cdot \mathbf{S}$$

$$H_{soc} = \lambda \mathbf{L}_i \cdot \mathbf{S}_i : \text{atomic site } i$$

$$\lambda \propto Z^4 : \text{heavier atom, bigger SOC}$$

spin-orbit coupling and lattice structure

Consider p-orbitals



the three degenerate p orbitals are aligned along perpendicular axes

$$H_{soc} = \lambda \mathbf{L} \cdot \mathbf{S} = \frac{\lambda}{2} (J^2 - L^2 - S^2) \quad \mathbf{L} = 1 \quad \mathbf{S} = 1/2$$

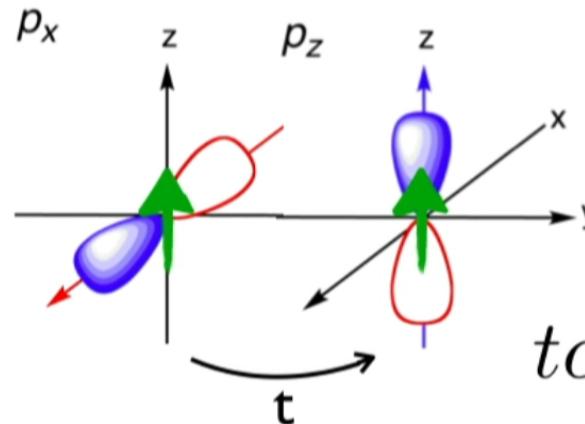
$J = 3/2$

$J = 1/2$

$$\begin{aligned} |\psi_{j_z=\frac{1}{2}}\rangle &= \frac{1}{\sqrt{3}} (|p_x \downarrow\rangle - i|p_y \downarrow\rangle + |p_z \uparrow\rangle) \\ |\psi_{j_z=-\frac{1}{2}}\rangle &= \frac{1}{\sqrt{3}} (|p_x \uparrow\rangle + i|p_y \uparrow\rangle - |p_z \downarrow\rangle) \end{aligned}$$

atomic site i ; no motion

Let's consider two sites: effects of SOC on hopping



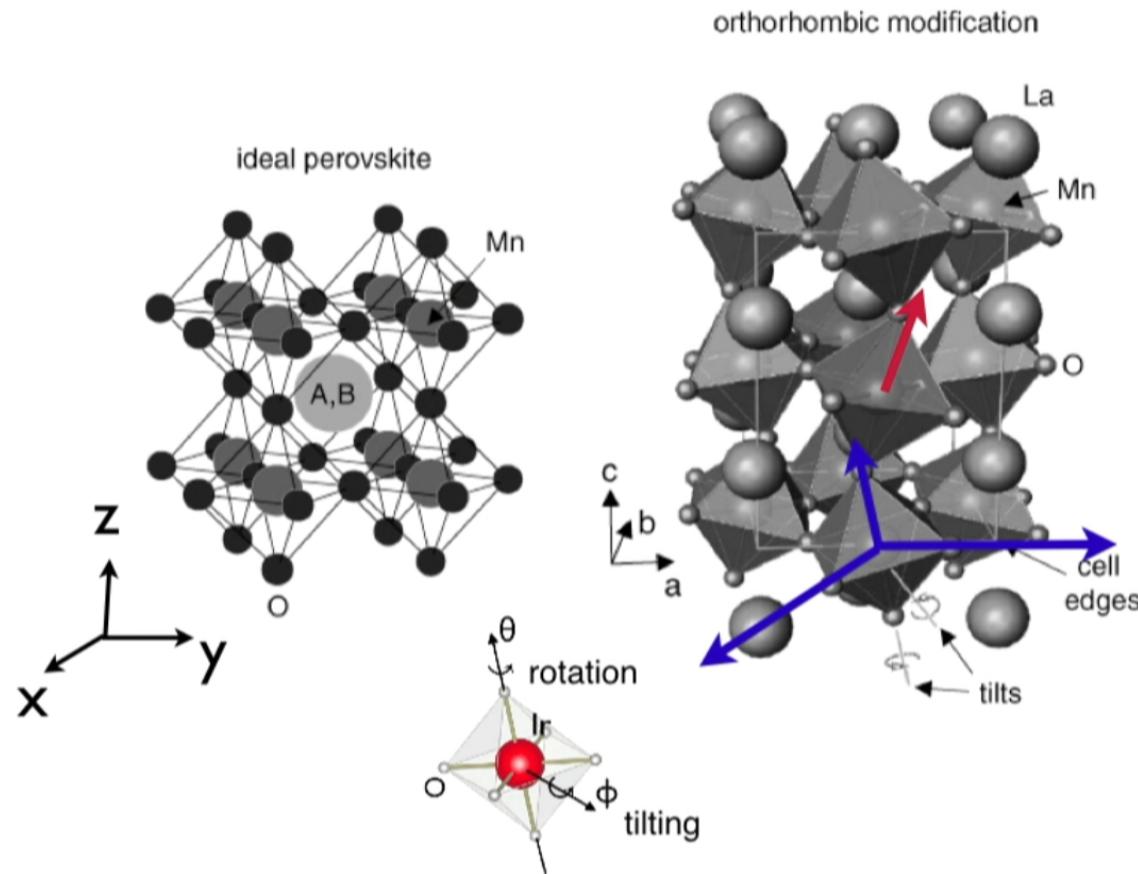
$$tc_{j,p_z,\uparrow}^{\dagger}c_{i,p_x,\uparrow} + h.c.$$

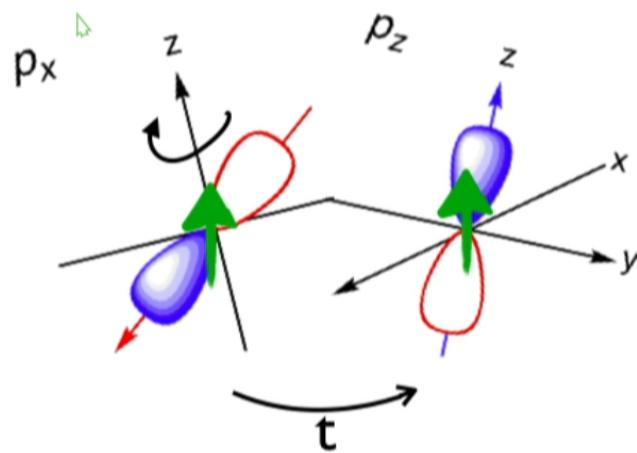
$$\begin{aligned} t_{i,j;p_z,p_x} &= \int d^3\mathbf{r} \phi_{p_z}^*(\mathbf{r} - \mathbf{R}_i) \delta\mathcal{H} \phi_{p_x}(\mathbf{r} - \mathbf{R}_j) \\ &= 0 \end{aligned}$$

because of symmetry: odd & even in mirror reflection z

when mirror symmetries broken $t \neq 0$

local axis is determined by other atoms inside the solids





$$tc_{j,p_z,\uparrow}^\dagger c_{i,p_x,\uparrow} + h.c.$$

$$\rightarrow tc_j^\dagger \boxed{j_z=\frac{1}{2}} c_i, \boxed{j_z=-\frac{1}{2}} + h.c.$$

; pseudospin (strong SOC) flips over hopping

$$|\psi_{j_z=\frac{1}{2}}\rangle = \frac{1}{\sqrt{3}} (|p_x \downarrow\rangle - i|p_y \downarrow\rangle + \boxed{|p_z \uparrow\rangle})$$

$$|\psi_{j_z=-1/2}\rangle = \frac{1}{\sqrt{3}} (\boxed{|p_x, \uparrow\rangle} + i|p_y, \uparrow\rangle - |p_z, \downarrow\rangle)$$

atomic SOC + crystal structure: Rashba-like SOC

$i \cos(k_y) \sigma_y$

with inversion symmetry

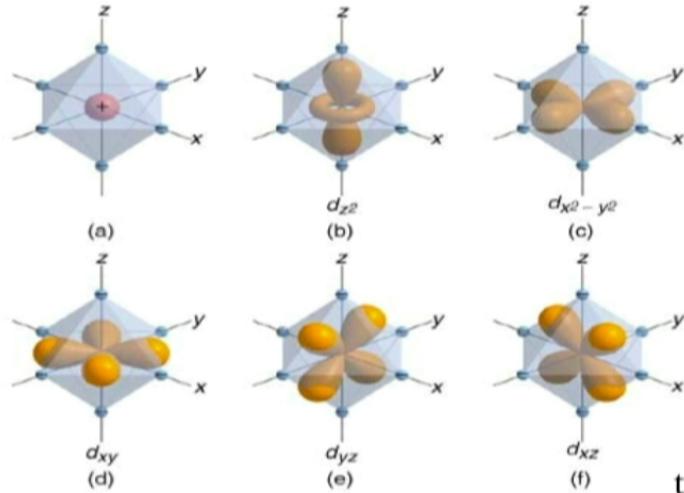
* **inversion is broken :** $\sin(k_y) \sigma_x$

$$\sin(k_y) \sigma_x - \sin(k_x) \sigma_y \sim (\mathbf{k} \times \vec{\sigma}) \cdot \hat{z}$$



Single ion: Crystal field splitting

Single d-electron in a crystal field (CF)



$$[-(\hbar^2/2m)\Delta + U(\mathbf{r}) + V_c(\mathbf{r})]\psi(\mathbf{r}) = \epsilon\psi(\mathbf{r})$$

$$V_c(\mathbf{r}) = \sum_{i=1}^6 Ze^2/|\mathbf{R}_i - \mathbf{r}| \quad \psi(\mathbf{r})_{n,l,m} = R_{n,l}(r)Y_{l,m}(\theta, \phi)$$

$$\begin{vmatrix} \epsilon_n + Dq - \epsilon & 0 & 0 & 0 & 5Dq \\ 0 & \epsilon_n - 4Dq - \epsilon & 0 & 0 & 0 \\ 0 & 0 & \epsilon_n + 6Dq - \epsilon & 0 & 0 \\ 0 & 0 & 0 & \epsilon_n - 4Dq - \epsilon & 0 \\ 5Dq & 0 & 0 & 0 & \epsilon_n + Dq - \epsilon \end{vmatrix} = 0$$

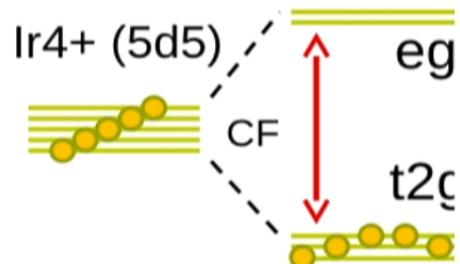
$$Dq = \langle \psi_{n,l=2,\pm 2} | V_c(\mathbf{r}) - (6Ze^2/a) | \psi_{n,l=2,\pm 2} \rangle$$

$$-4Dq = \langle \psi_{n,l=2,\pm 1} | V_c(\mathbf{r}) - (6Ze^2/a) | \psi_{n,l=2,\pm 1} \rangle$$

$$5Dq = \langle \psi_{n,l=2,\pm 2} | V_c(\mathbf{r}) - (6Ze^2/a) | \psi_{n,l=2,\mp 2} \rangle$$

two-fold e_g states: $\epsilon^{(e_g)} = \epsilon_n + 6Dq$, $\{d_{z^2}, d_{x^2-y^2}\}$

three-fold t_{2g} states: $\epsilon^{(t_{2g})} = \epsilon_n - 4Dq$, $\{d_{xy}, d_{yz}, d_{xz}\}$





Crystal Field Splitting & Spin-orbit coupling (SOC)

$$H_{SOC} = \lambda \mathbf{l} \cdot \mathbf{s} = \lambda \sum_{a,\sigma} \sum_{b,\sigma'} \langle a\sigma | l_x s_x + l_y s_y + l_z s_z | b\sigma' \rangle d_{a\sigma}^\dagger d_{b\sigma'}$$

$$l_x = \begin{bmatrix} d_{yz} & d_{zx} & d_{xy} & d_{z^2} & d_{x^2-y^2} \\ 0 & 0 & 0 & -\sqrt{3}i & -i \\ 0 & 0 & i & 0 & 0 \\ 0 & -i & 0 & 0 & 0 \\ \sqrt{3} & 0 & 0 & 0 & 0 \\ i & 0 & 0 & 0 & 0 \end{bmatrix}$$

$$l_y = \begin{bmatrix} 0 & 0 & -i & 0 & 0 \\ 0 & 0 & 0 & \sqrt{3}i & -i \\ i & 0 & 0 & 0 & 0 \\ 0 & -\sqrt{3}i & 0 & 0 & 0 \\ 0 & i & 0 & 0 & 0 \end{bmatrix}$$

$$l_z = \begin{bmatrix} 0 & i & 0 & 0 & 0 \\ -i & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2i \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -2i & 0 & 0 \end{bmatrix}$$

$p_x \quad p_y \quad p_z$

$$l_x = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}$$

$$l_y = \begin{bmatrix} 0 & 0 & i \\ 0 & 0 & 0 \\ -i & 0 & 0 \end{bmatrix}$$

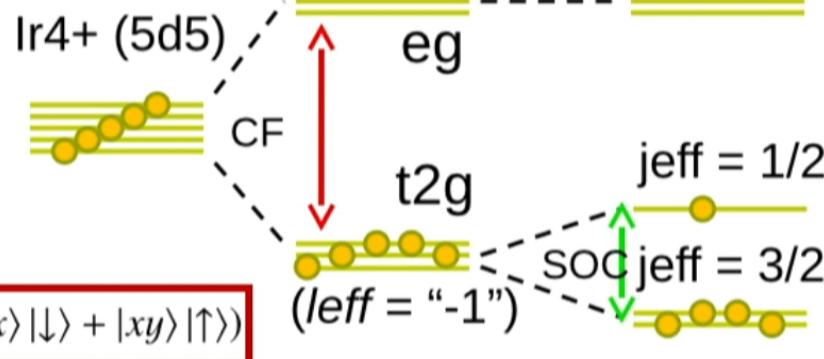
$$l_z = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

T-P equivalence

$$\mathbf{l}(t_{2g}) = -\mathbf{l}(p)$$

$l = 2$

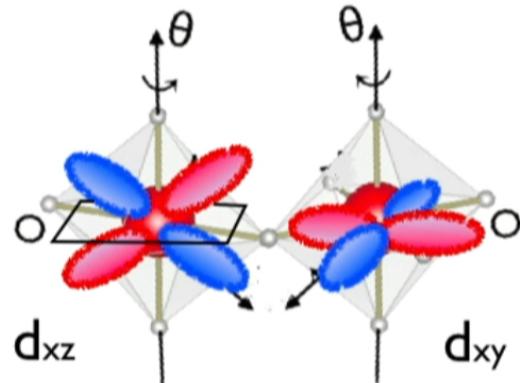
$$l_{eff} = -1$$



$J_{eff} = 1/2$ basis

$$|+\frac{1}{2}\rangle = \sqrt{\frac{1}{3}}(|yz\rangle |\downarrow\rangle + i|zx\rangle |\downarrow\rangle + |xy\rangle |\uparrow\rangle)$$

↳ SOC in Jeff=1/2 basis → pseudo-spin flip terms



$$|1/2\rangle \propto |yz\downarrow\rangle + i|zx\downarrow\rangle + |xy\uparrow\rangle$$

$$|-1/2\rangle \propto |yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle$$

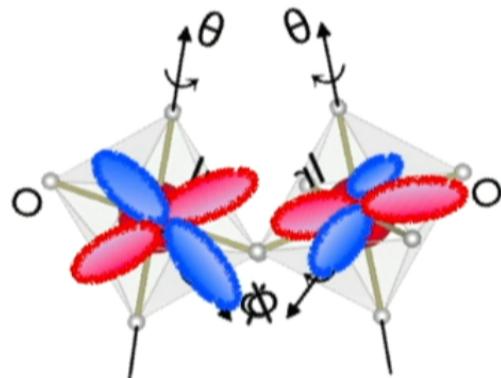
bulk
 Sr_2IrO_4

only rotation

$$t = 0$$

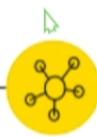
$$t d_{i,xz,\uparrow}^\dagger d_{j,xy,\uparrow}$$

$$\rightarrow t c_{j,j_z=\frac{1}{2}}^\dagger c_{i,j_z=-\frac{1}{2}} + h.c.$$



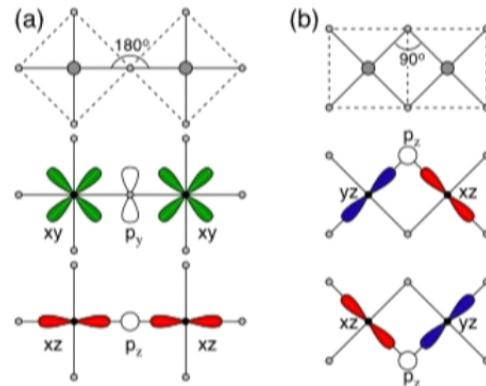
Orthorhombic lattice SrIrO_3 ,
dxz and dxy hopping between nearest
neighbour is allowed:

$$t \neq 0$$

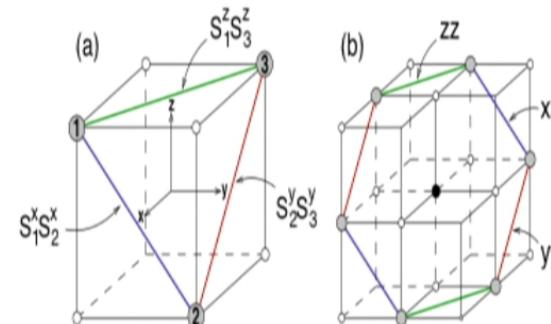


Compass model (large U limit)

Jackeli and Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)



edge-shared octahedra



strong SOC in t_{2g} states:

Kitaev-Heisenberg model

$$H_{ij}^\gamma = -K S_i^\gamma S_j^\gamma + J \mathbf{S}_i \cdot \mathbf{S}_j \quad \text{where} \quad K = \frac{8J_H t_0^2}{3U^2}$$

Material candidates: honeycomb Iridates (5d)

Na_2IrO_3 , Li_2IrO_3



B. Generic Spin Model (large U limit)

J. Rau, E. Lee, HYK, Phys. Rev. Lett. 112, 077204 (2014)

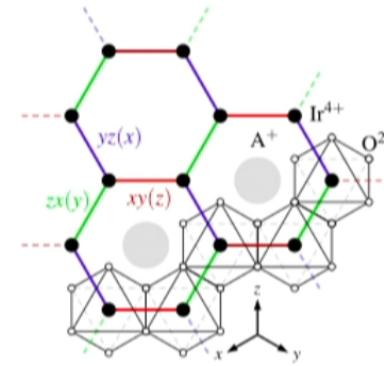
nearest neighbour:
ideal honeycomb

$$H = \sum_{\gamma \in x,y,z} H^\gamma,$$

bond-dep. interaction

$$H^z = \sum_{\langle ij \rangle \in z-bond} [K_z S_i^z S_j^z + \Gamma_z (S_i^x S_j^y + S_i^y S_j^x)] + J \mathbf{S}_i \cdot \mathbf{S}_j$$

$$H^x = H^z(x \rightarrow y \rightarrow z \rightarrow x)$$





Ordered phases nearby KSL

For $+\Gamma$

ED 24-site cluster

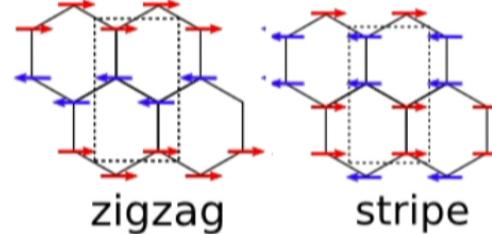
+J

$$(J, K, \Gamma) = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$$

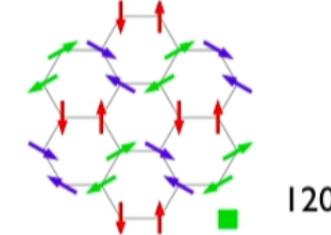
+K

-J

-K



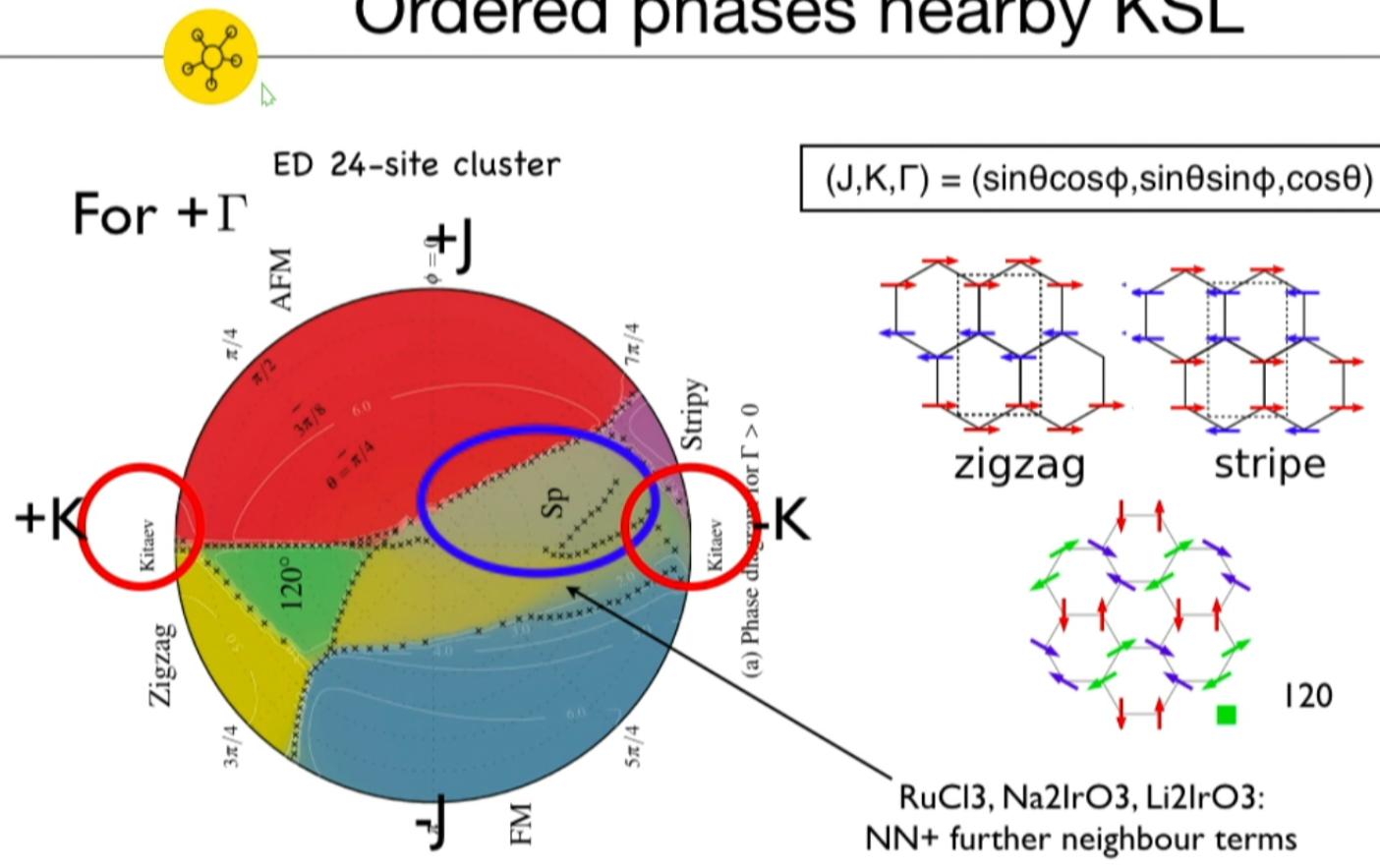
zigzag stripe



I20

J. Rau, E. Lee, HYK, Phys. Rev. Lett. 112, 077204 (2014)

Ordered phases nearby KSL



J. Rau, E. Lee, HYK, Phys. Rev. Lett. 112, 077204 (2014)

Kitaev materials: extends to 4d

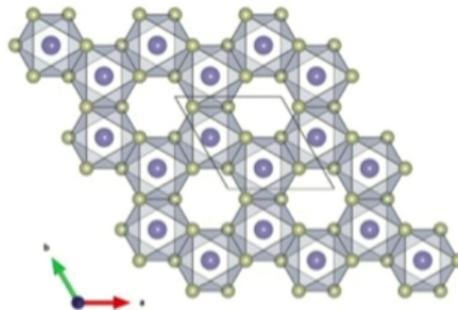
$\alpha - RuCl_3$



PHYSICAL REVIEW B 90, 041112(R) (2014)

α -RuCl₃: A spin-orbit assisted Mott insulator on a honeycomb lattice

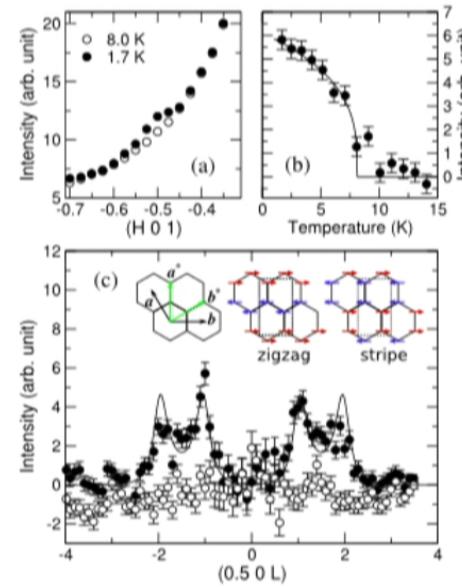
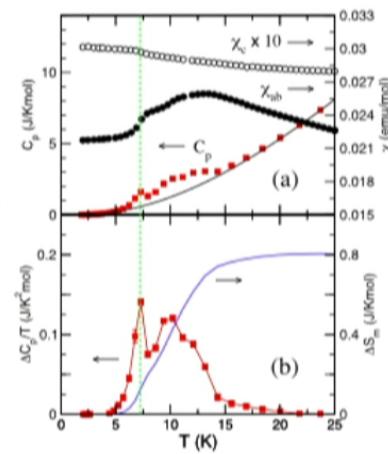
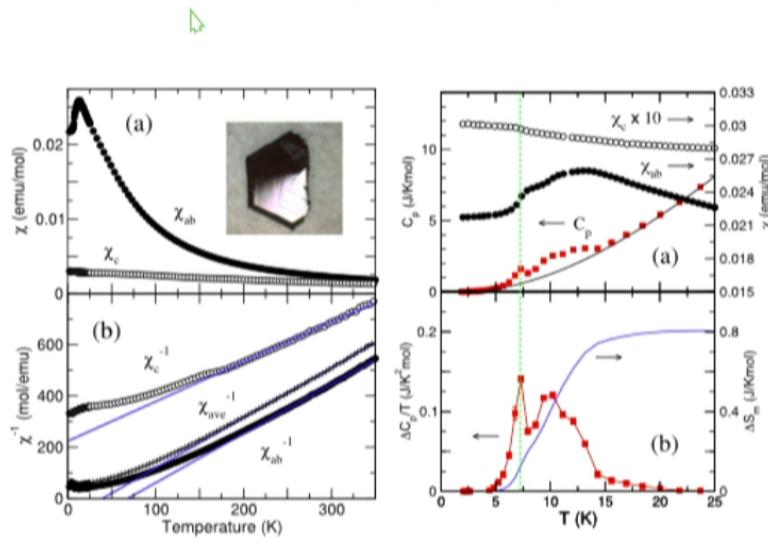
K. W. Plumb,¹ J. P. Clancy,¹ L. J. Sandilands,¹ V. Vijay Shankar,¹ Y. F. Hu,² K. S. Burch,^{1,3} Hae-Young Kee,^{1,4} and Young-June Kim^{1,*}



Ru³⁺ : 4d⁵

process in α -RuCl₃. Then a microscopic spin model relevant for α -RuCl₃ should be composed of both the nearest-neighbor Heisenberg and bond-dependent exchange terms denoted by Kitaev K and Γ [44–46].

Zigzag magnetic ordering below T_c



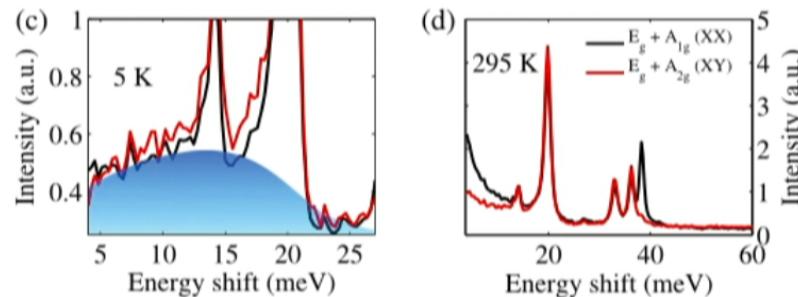
J. Sears et al, PRB 91, 144420 (2015)

R. D. Johnson et al, Phys. Rev. B 92, 235119 (2015).

H. B. Cao et al, Phys. Rev. B 93, 134423 (2016);

Proximate to Kitaev spin liquid

Raman spectrum



L. J. Sandilands et al,
Phys. Rev. Lett. 114, 147201 (2015).

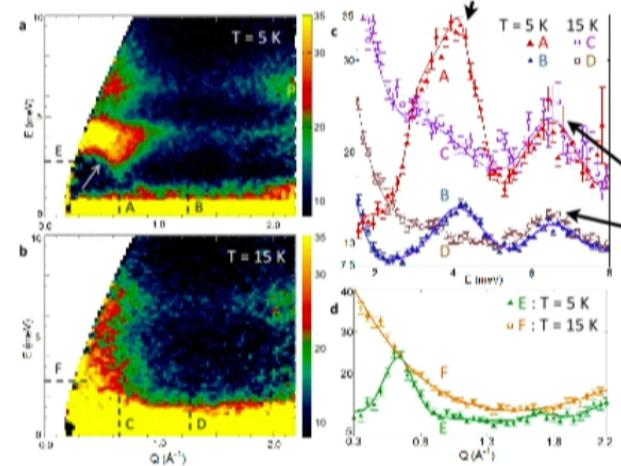
Magnetic excitations: inelastic neutron scattering

A. Banerjee... S. Nagler, Nat. Mat. 15, 733 (2016)

A. Banerjee... S. Nagler, Science (2017)

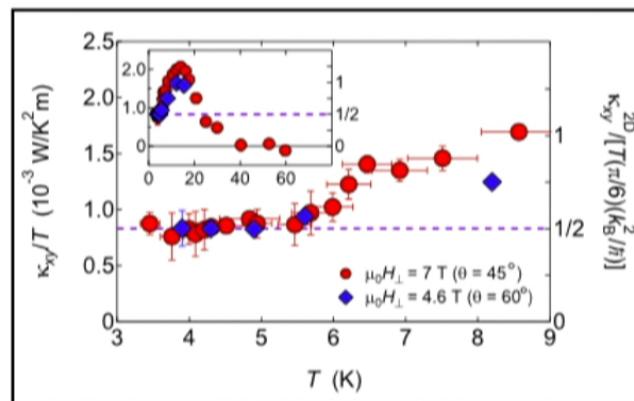
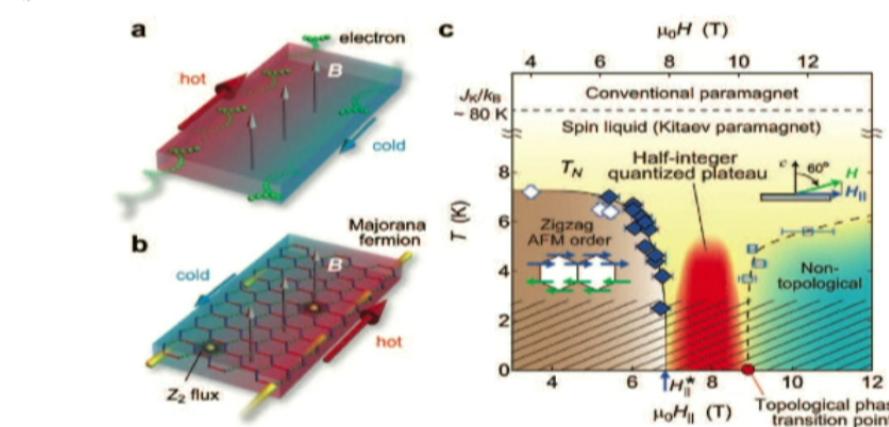
S.-H. Do, ..., S. Ji, Nat. Phys. 13, 1079 (2017)

• • •



Magnetic field - destroy the ordering?
Governed by Kitaev spin interaction?

Thermal Transport: alpha-RuCl₃



$$\frac{\kappa_{xy}^{2D}}{T} = c \frac{\pi}{6} \frac{k_B^2}{\hbar}$$

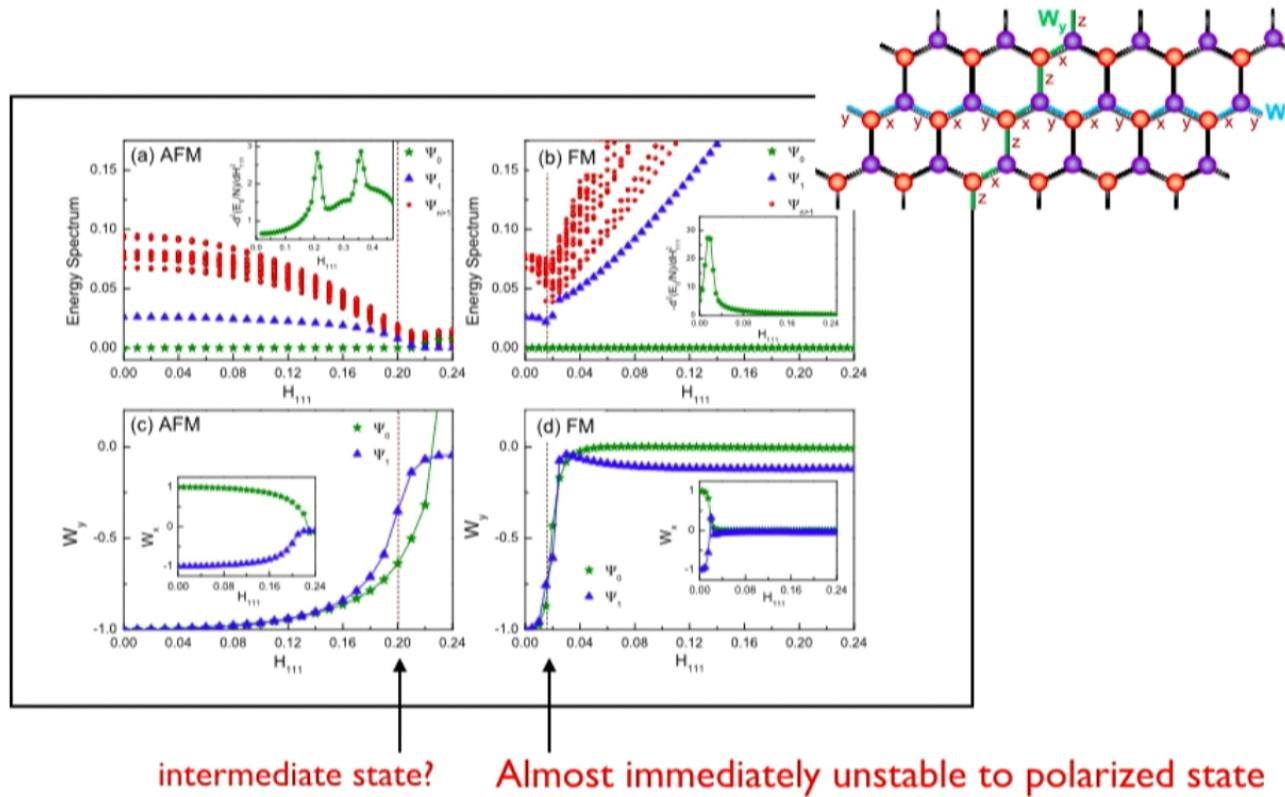
compare:

$$\sigma_{xy}^{QH} = \nu \frac{e^2}{h}$$

Kasahara,..Y. Matsuda, Nature (2018)

Real materials : not a pure Kitaev model (magnetic order at low T)

► Pure Kitaev model cannot explain intermediate-field state



Z. Zhu, I. Kimchi, D. Sheng, L. Fu, PRB 97, 241110 (2018)

List of proposed parameter sets

L.Janssen,E.Andrade, M.Vojta, PRB (2017)

Set	Material	J_1 [meV]	K_1 [meV]	Γ_1 [meV]	J_2 [meV]	K_2 [meV]	J_3 [meV]	Method	Ref.	Year
1	✓ α -RuCl ₃	-4.6	+7.0	—	—	—	—	fit to neutron scattering	34, 35	2016
1'	Na ₂ IrO ₃	-4.0	+10.5	—	—	—	—	fit to susceptibility & neutron scattering	29	2013
1+ Γ	✓ α -RuCl ₃	-12	+17	+12	—	—	—	DFT + t/U expansion	41	2015
2	Na ₂ IrO ₃	0	-17	0	0	—	+6.8	DFT + exact diagonalization	31	2016
2+ Γ	Na ₂ IrO ₃	+3	-17	+1	-3	+6	+1	DFT + t/U expansion, direction of moments	39, 42	2016
(2+ Γ)'	Na ₂ IrO ₃	+3	-17.5	-1	+5	—	+5	MRCI, fit to θ_{CW}	44	2014
(2+ Γ)''	α -RuCl ₃	+1.2	-5.6	-1	+0.3	—	+0.3	MRCI, fit to magnetization	13	2016
2/3	α -RuCl ₃	-1.7	-6.6	+6.6	0	—	+2.7	DFT + exact diagonalization	31	2016
3	α -RuCl ₃	—	-6.8	+9.5	—	—	—	fit to neutron scattering	32	2017
3'	α -RuCl ₃	—	-5.5	+7.6	—	—	—	DFT + t/U expansion	33	2016
3'''	α -RuCl ₃	-1	-8	+4	—	—	—	DFT + t/U expansion	✓ 37	2016
3+ J_3	α -RuCl ₃	-0.5	-5.0	+2.5	—	—	+0.5	fit to neutron scattering	38	2017

FM Kitaev and AFM Γ : starting point

 Dominant interactions: $-K + \Gamma$
other small interactions : magnetic ordering



magnetic field: kills the magnetic ordering
& reveal “the phase” set by
 $FM\ K + AFM\ \Gamma$

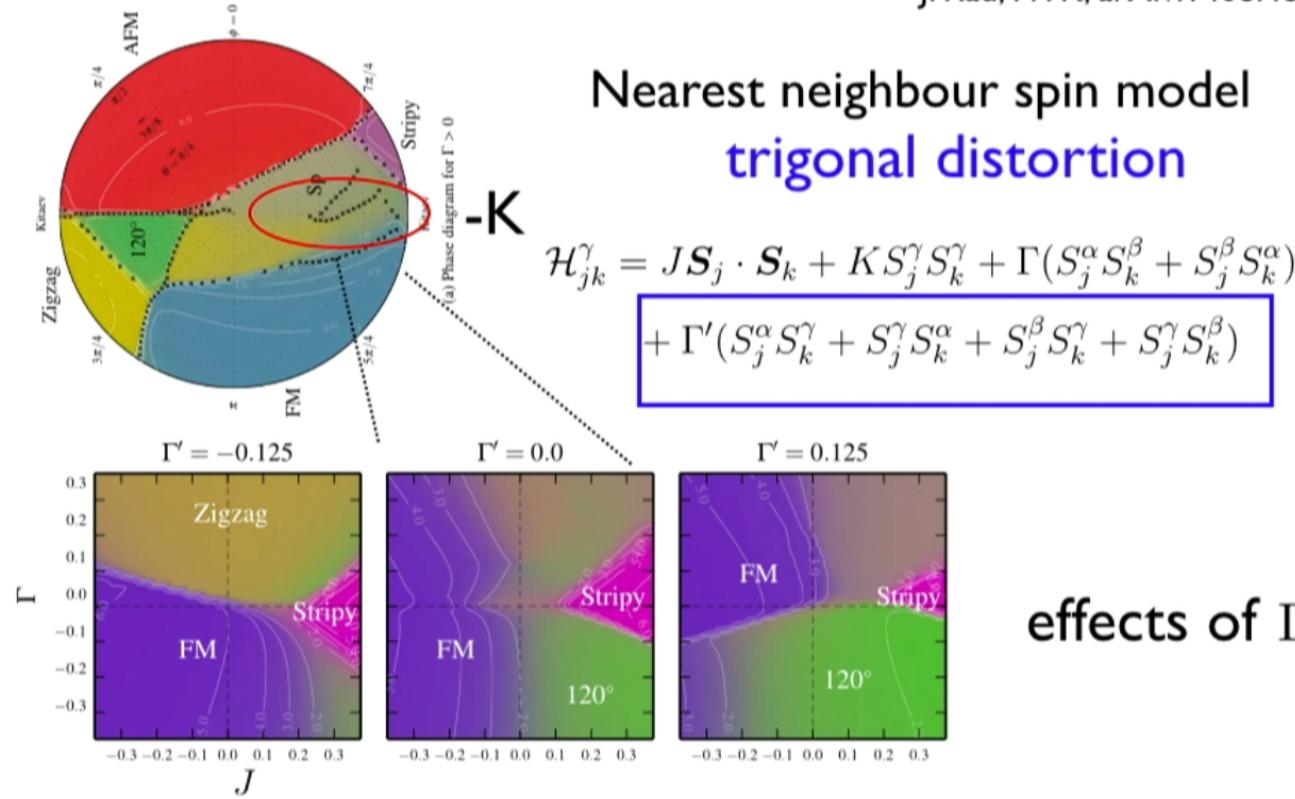
Does FM Kitaev + AFM Γ model support
spin liquid?

J. Gordon, A. Catuneanu, E. Sorensen, HYK, arXiv:1901.09943
(accepted for publication in Nature Comm.)

ZZ ordering in Kitaev model

J. Rau, HYK, arXiv:1408.4811

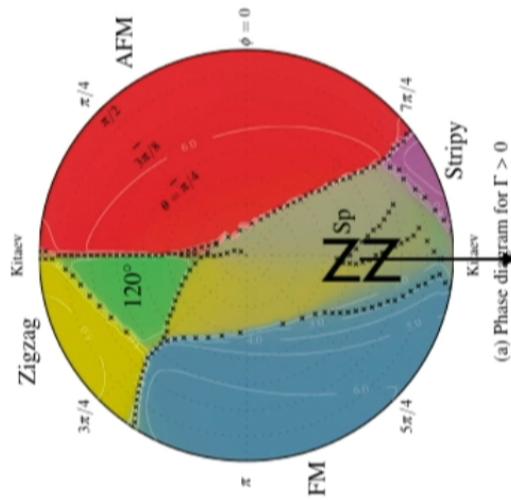
Nearest neighbour spin model
trigonal distortion



C. Field-revealed Kitaev spin liquid near ferromagnetic Kitaev (-K) region

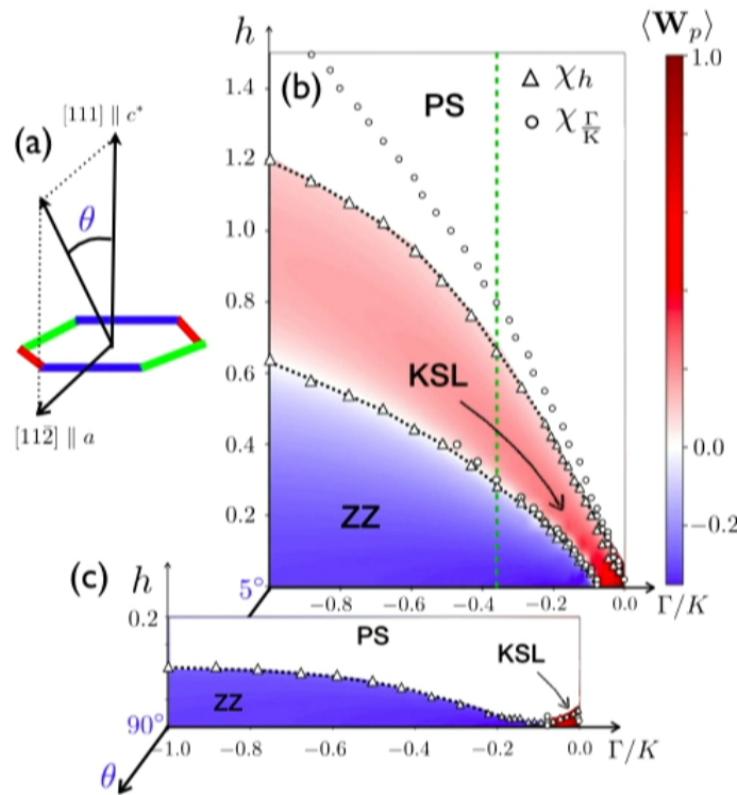


24 site ED

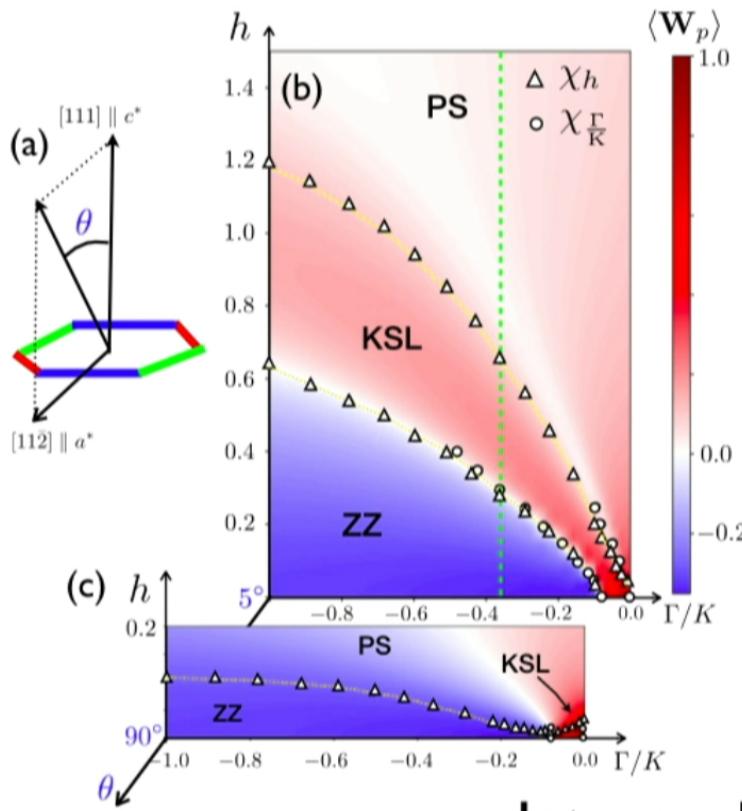


Trigonal distortion induces the ZZ ordering

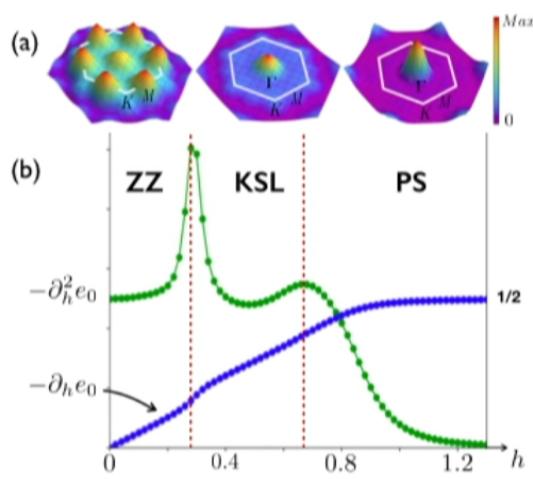
Plaquette op. $\langle W_p \rangle = \langle \sigma_x \sigma_y \sigma_z \sigma_x \sigma_y \sigma_z \rangle$



24 site ED

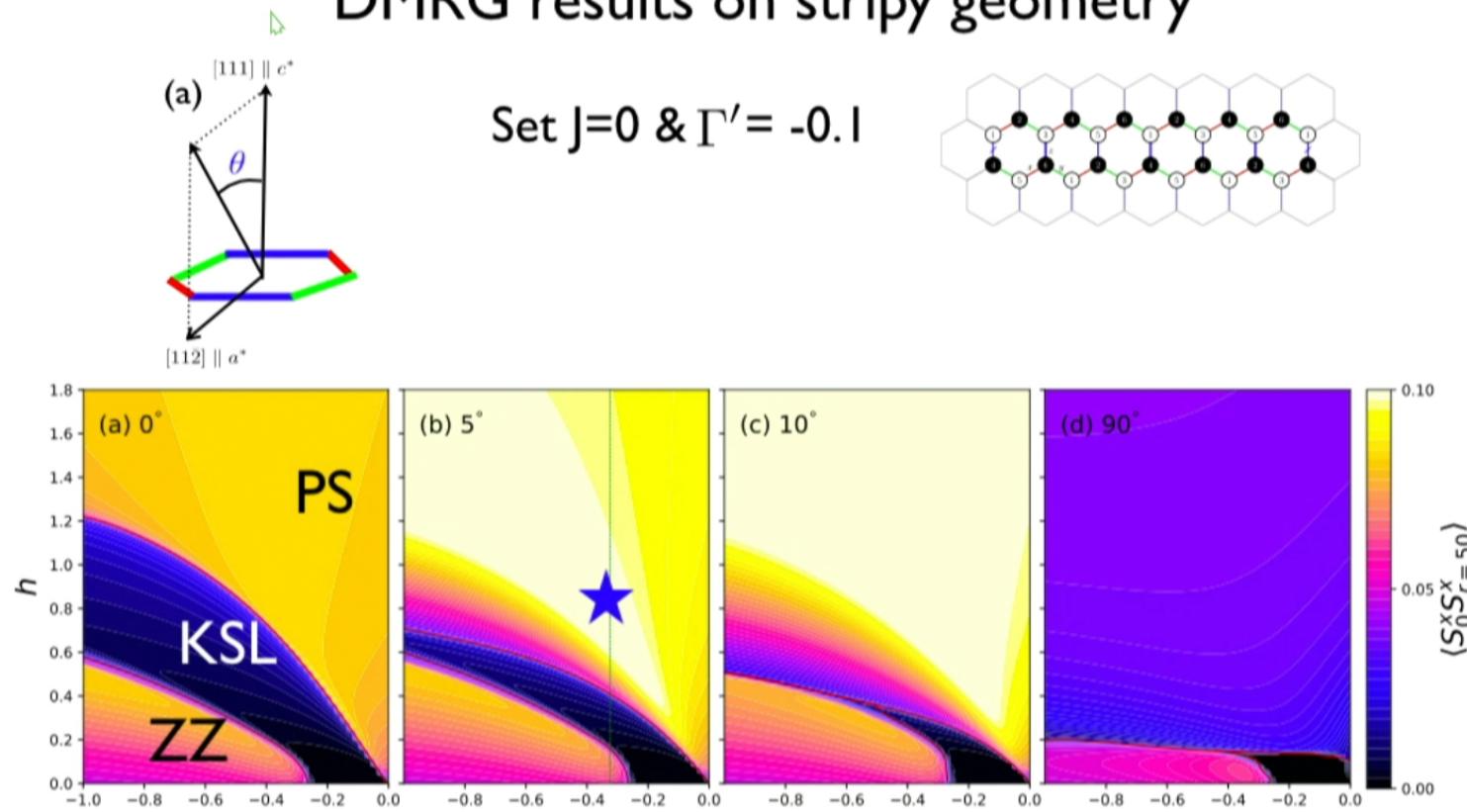


Set $J=0$ & $\Gamma = -0.03$



Intermediate field KSL is found
when Gamma is finite

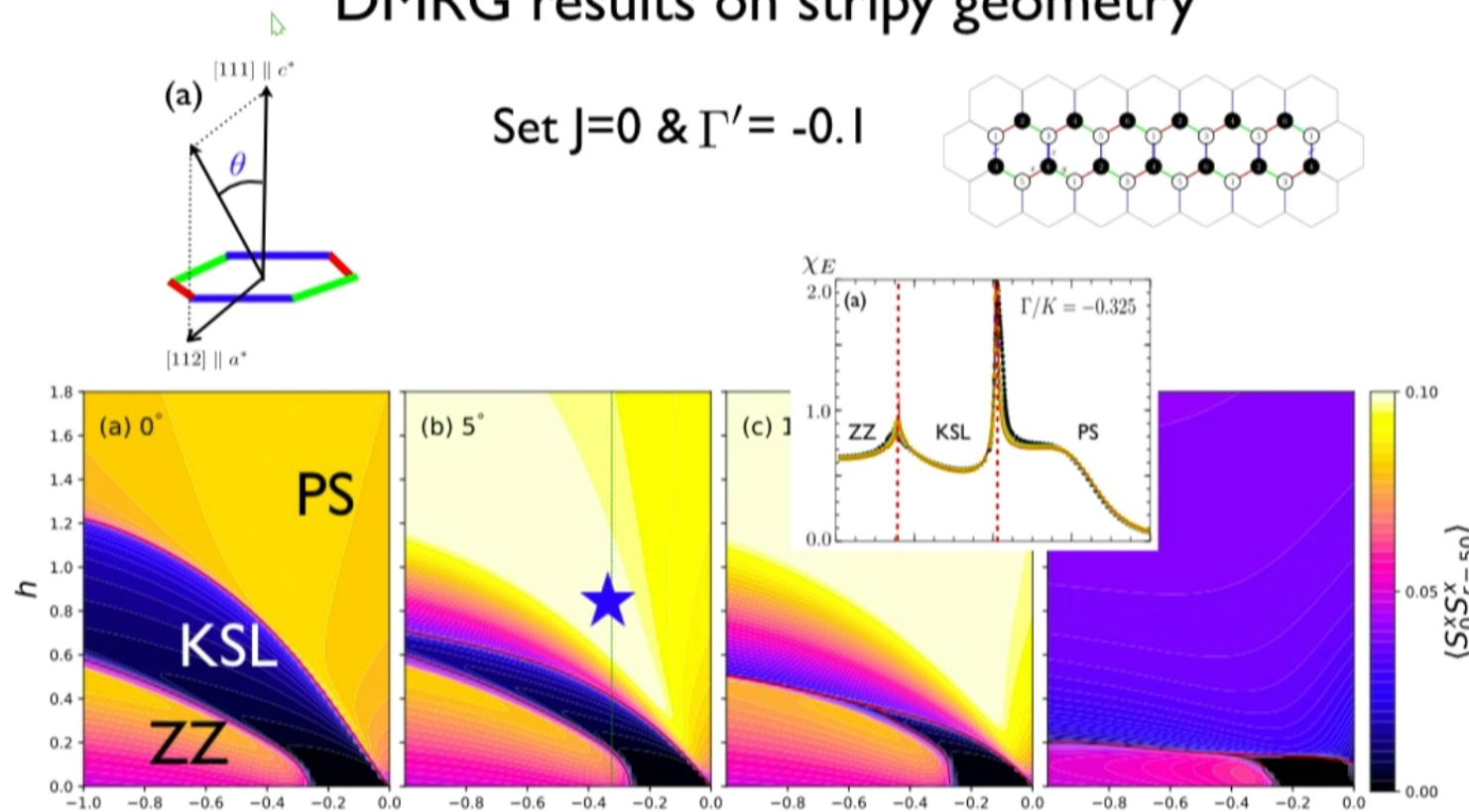
DMRG results on stripy geometry



Strong field-angle dependence

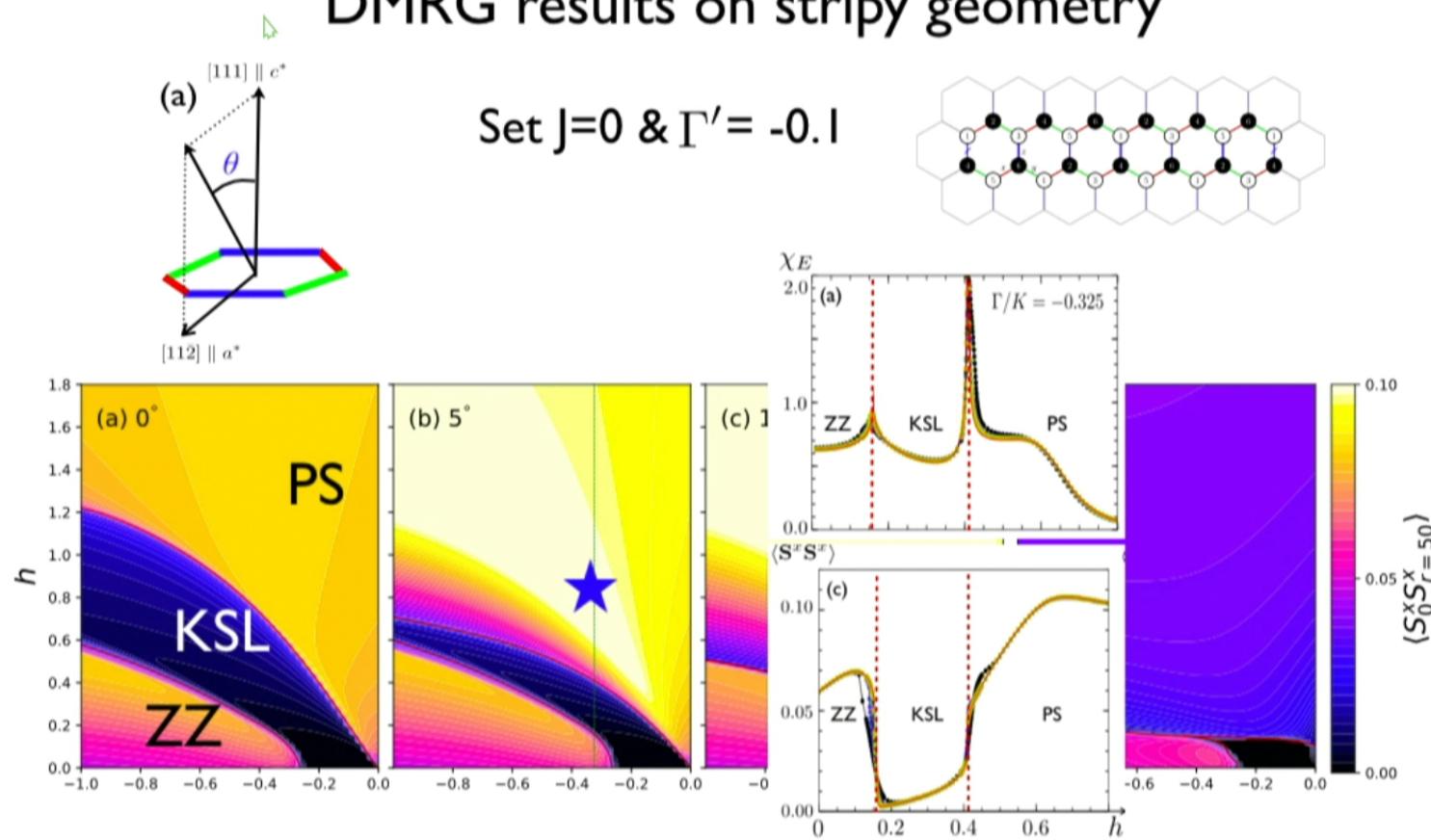
★ strong spin fluctuations in PS in high field regime

DMRG results on stripy geometry



DMRG results on stripy geometry

Set $J=0$ & $\Gamma' = -0.1$



Strong field-angle dependence

★ strong spin fluctuations in PS in high field regime

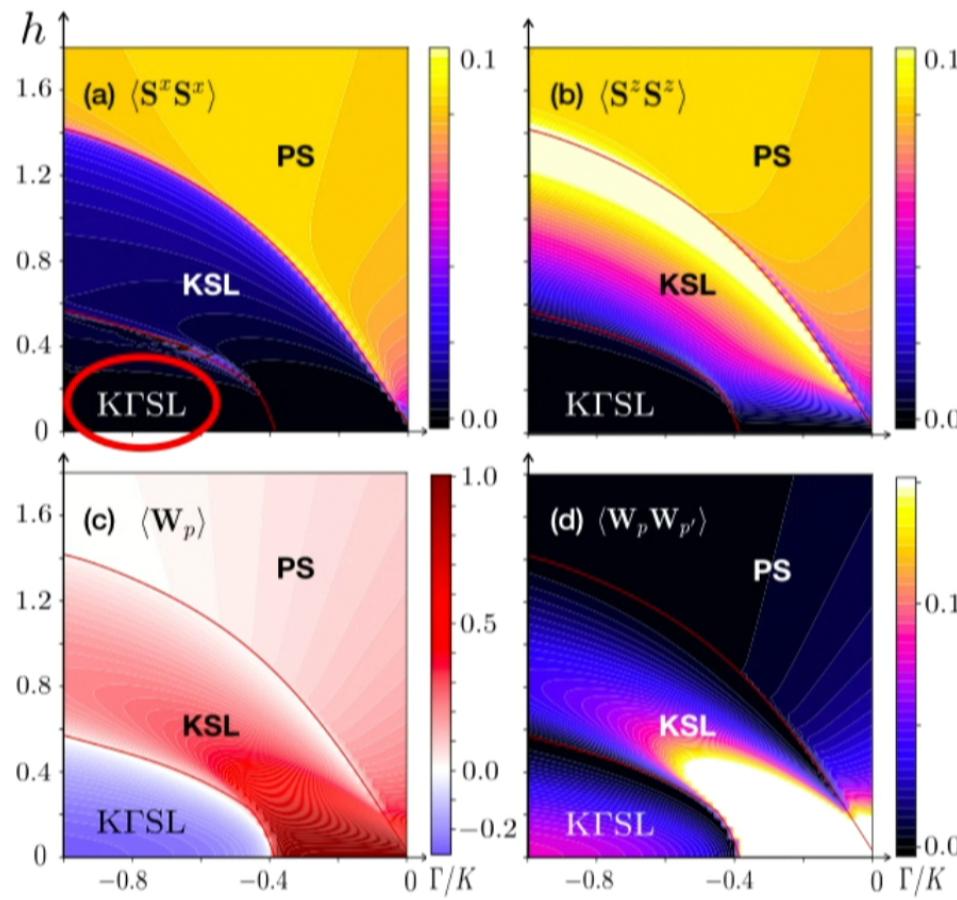
Another spin liquid nearby Kitaev spin liquid
near ferromagnetic Kitaev (-K) region?



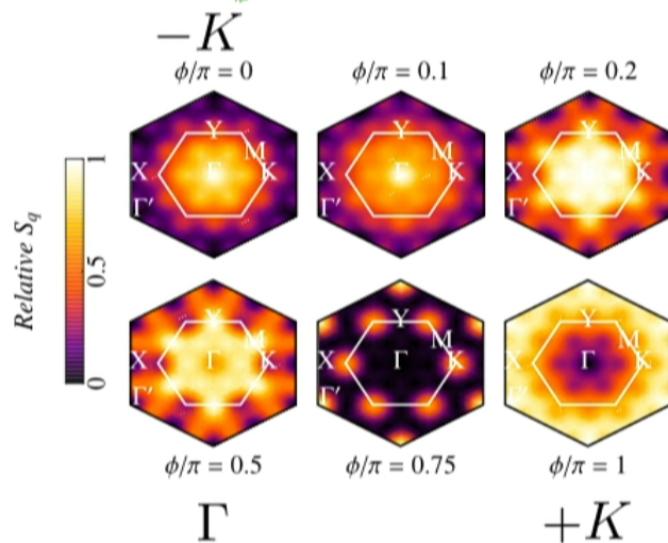
When $\Gamma' = 0$
: no ZZ ordering
: What is the phase set by -K + Gamma?

DMRG results on 2 leg stripy geometry

Set $J=0$ & $\theta=0$ & $\Gamma'=0$ without ZZ ordering



Static structure factor



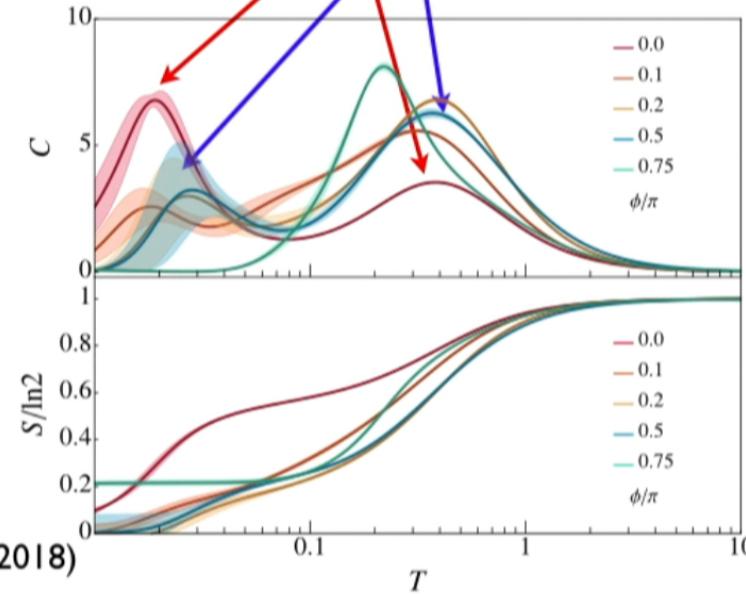
$-K + \Gamma$: wide
 regime of Spin liquids

vision gap: bigger

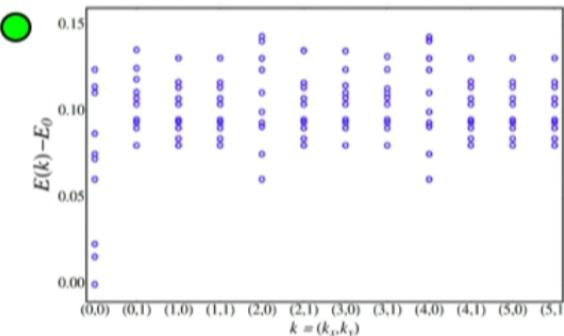
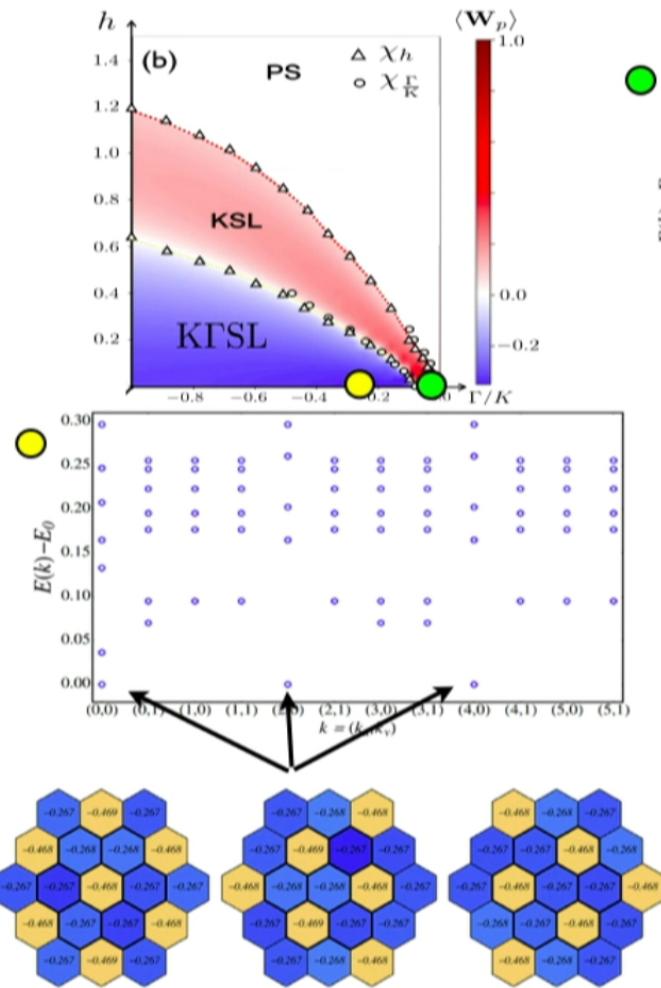
A. Catuneanu... HYK, npj Quantum Materials (2018)

Temperature dependence

double peaks



3-fold degenerate: vortex crystal spin liquid (24 site ED)



small anisotropy
lifts the degeneracy:
Does not appear in
leg-geometry either:
strong finite size effects:
Further studies are required

 Table 4
Numerical results for $J_x = J_y = J_z = 1$

$E_{\text{vortex}} \approx 0.1536, \quad \Delta E \left(\begin{array}{c} \text{hexagonal lattice} \\ \text{with vortices} \end{array} \right) \approx -0.04, \quad \Delta E \left(\begin{array}{c} \text{hexagonal lattice} \\ \text{with vortices} \end{array} \right) \approx -0.07.$			
	Phase	Vortex density	Energy per \diamond and per vortex
1		$\frac{1}{1}$	0.067 0.067
2		$\frac{1}{2}$	0.052 0.104
3		$\frac{1}{3}$	0.041 0.124
4		$\frac{2}{3}$	0.054 0.081
5		$\frac{1}{3}$	0.026 0.078
6		$\frac{2}{3}$	0.060 0.090
7		$\frac{1}{4}$	0.034 0.136
8		$\frac{2}{4}$	0.042 0.085
9		$\frac{3}{4}$	0.059 0.078
10		$\frac{1}{4}$	0.042 0.167
11		$\frac{3}{4}$	0.074 0.099
12		$\frac{1}{4}$	0.025 0.101
13		$\frac{2}{4}$	0.046 0.092
14		$\frac{3}{4}$	0.072 0.096

Vortices are shown in gray; for periodic phases the unit cell is indicated by a parallelogram.



RuCl₃ near Spin Liquid (possibly two spin liquids)

Majorana fermions & Z2 are coupled via Γ interaction,
but the phase is adiabatically connected to pure Kitaev under the field

Outlook

Kitaev materials: SOC + honeycomb Mott insulator

hyperhoneycomb spiral - near SU(2) $K = \Gamma : L37.00012$

Session L37: Iridates - honeycomb lattice & other geometries

higher-spin Kitaev materials: arXiv:1903.00011

P37.00006 by P. Stavropoulos

Session P37: Honeycomb lattice and other low-D models

doping: topological superconductor

strain induced phase transitions