# HΦを利用した研究例の紹介

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1. Proximity to quantum spin liquids
2. $Na_2IrO_3$: Mott insulator in vicinity of QSL
3. Spin excitations by shifted Krylov subspace methods
4. Extension of Kitaev's spin liquid

CdMSi CBSM² Computational Science Alliance The University of Tokyo

# Proximity to Quantum Spin Liquid

# Quantum Liquid

Typical examples of quantum liquids:
Liquid helium 3, metals

Landau's Fermi liquid Theory

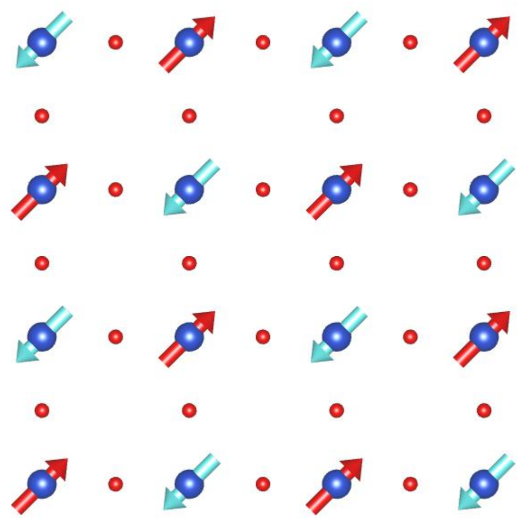
L. D. Landau, Zh. Eksperim. I Teor. Fiz 30, 1058 (1956).

Spontaneous symmetry breaking (SSB) in Fermi liquid:
Superfluidity, superconductivity, magnetic order

Spins in Mott insulators

R. Peierls, N. F. Mott (1937)

Example:
$La_2CuO_4$



$$J\left[\hat{S}_i^x\hat{S}_j^x + \hat{S}_i^y\hat{S}_j^y + \hat{S}_i^z\hat{S}_j^z\right]$$

Search for quantum spin liquid
P. W. Anderson, Mater. Res. Bull. 8, 153 (1973)

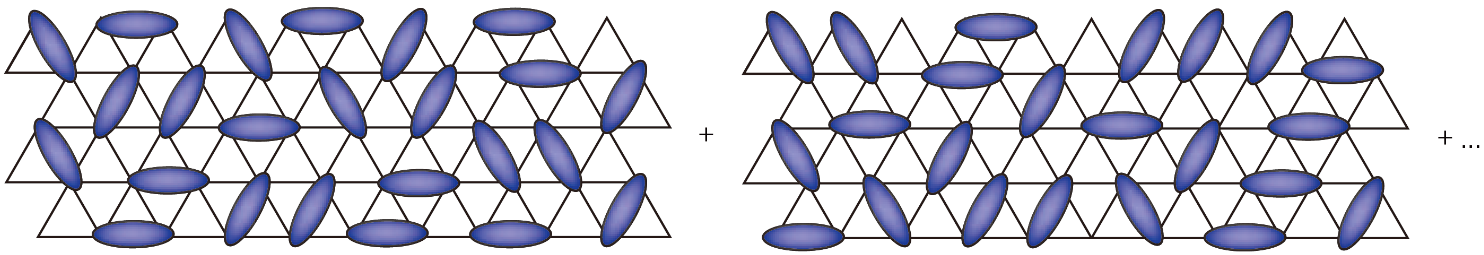
No universal basis for SSB in spins

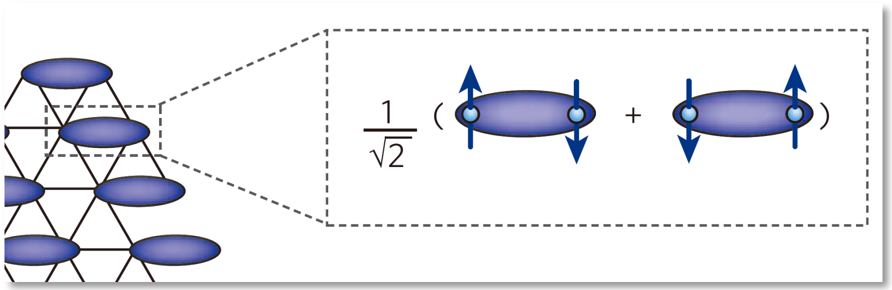
# Quantum Spin Liquid in Frustrated Magnets

RVB

P. W. Anderson, Mater. Res. Bull. 8, 153 (1973).
P. Fazekas & P. W. Anderson, Philos. Mag. 30, 423 (1974).

- A v.w.f. for S=1/2 Heisenberg model




Review: L. Balents, Nature 464, 199 (2010).

No spontaneous symmetry breakings at $T=0$

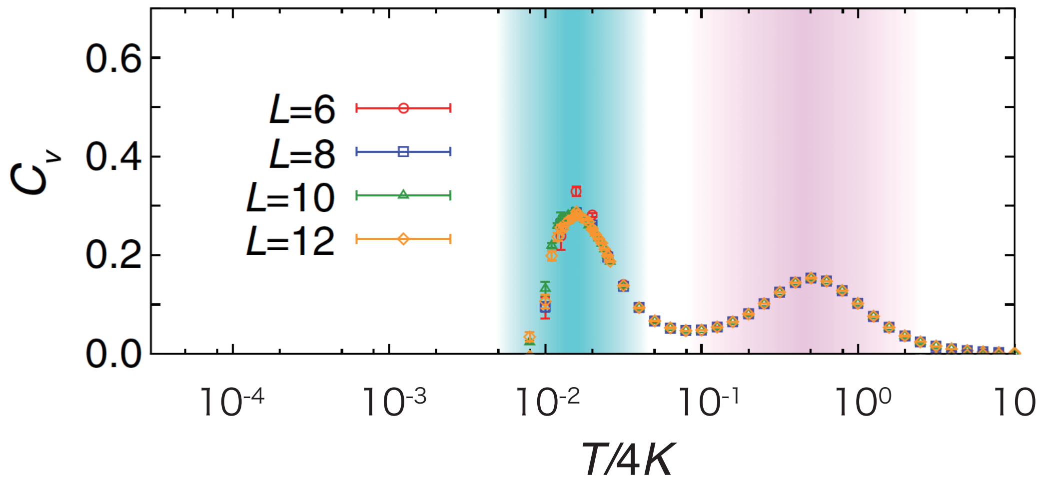
Fractionalization→

- Entropy remaining at low temperatures ($\sim O(10^{-1}\text{-}10^{-2}J)$)
- Continuum in spin excitations

# Thermal Excitations

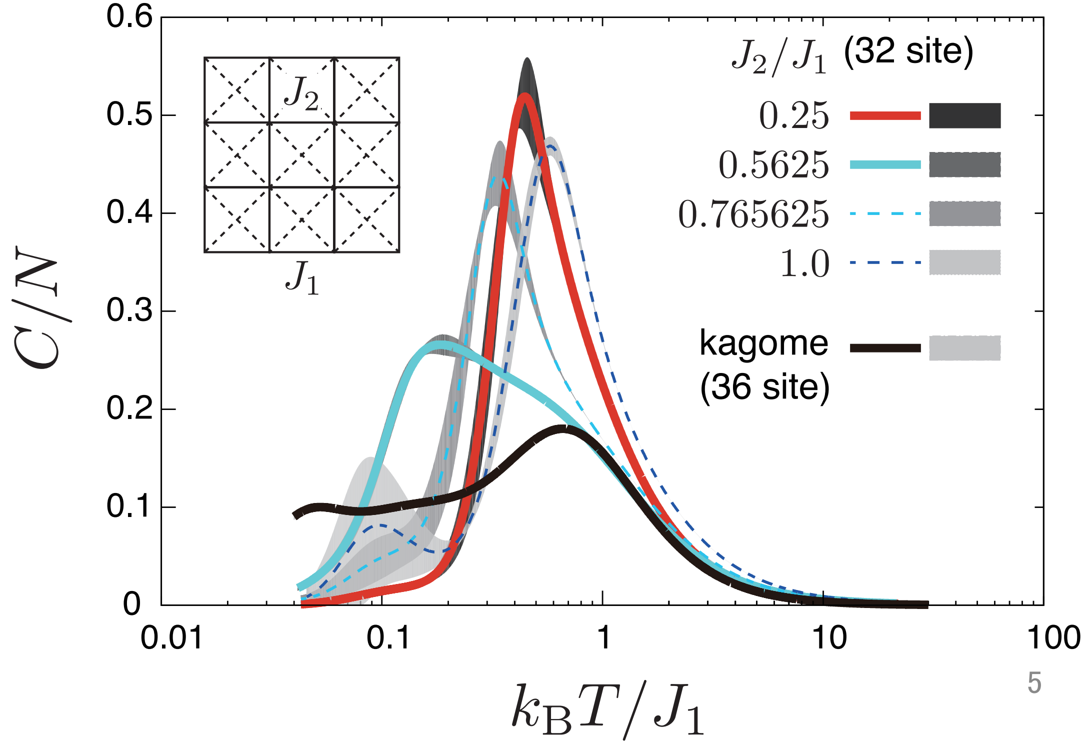
- Heat capacity of Kitaev's spin liquid

J. Nasu, M. Udagawa, & Y. Motome, Phys. Rev. B 92, 115122 (2015)




- Typical candidates: $J_1$-$J_2$ and kagome

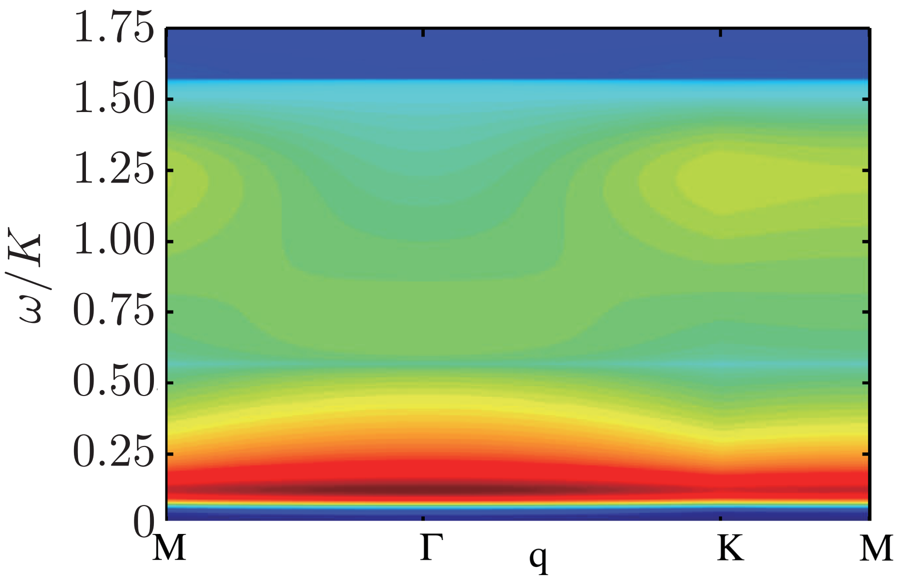
Y. Yamaji & T. Misawa, unpublished



# Spin Excitations

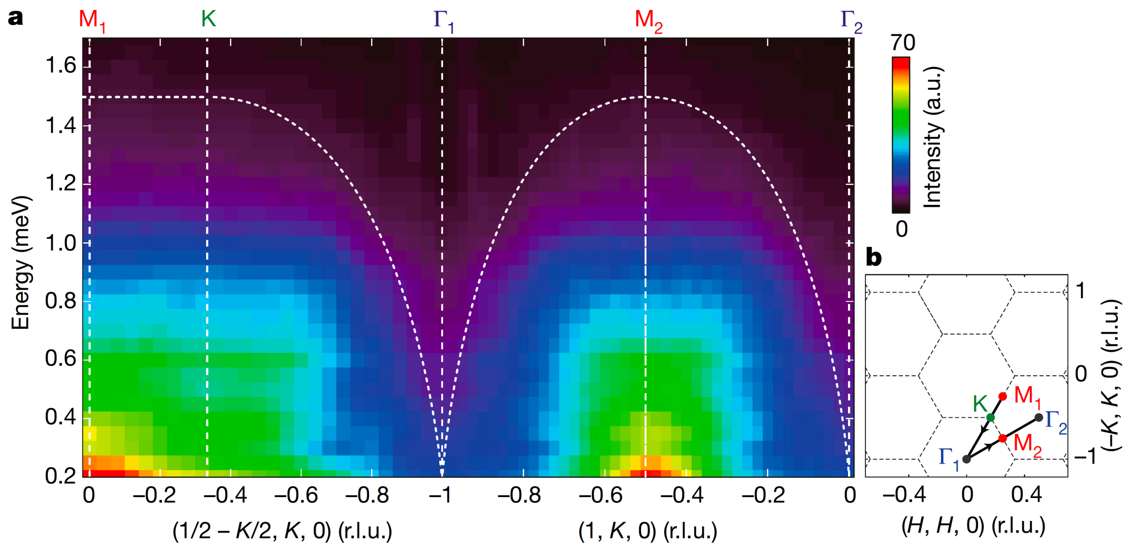
- S(Q,$\omega$) at $T$=0 of Kitaev's spin liquid

J. Knolle, D. L. Kovrizhin, J. T. Chalker, & R. Moessner, Phys. Rev. Lett. 112, 207203 (2014).




- Recent candidate (but not likely): $YbMgGaO_4$

$YbMgGaO_4$: Yb triangular lattice
Y. Shen, *et al.*, Nature 540, 559 (2016).



# Kitaev Model

## Kitaev Model

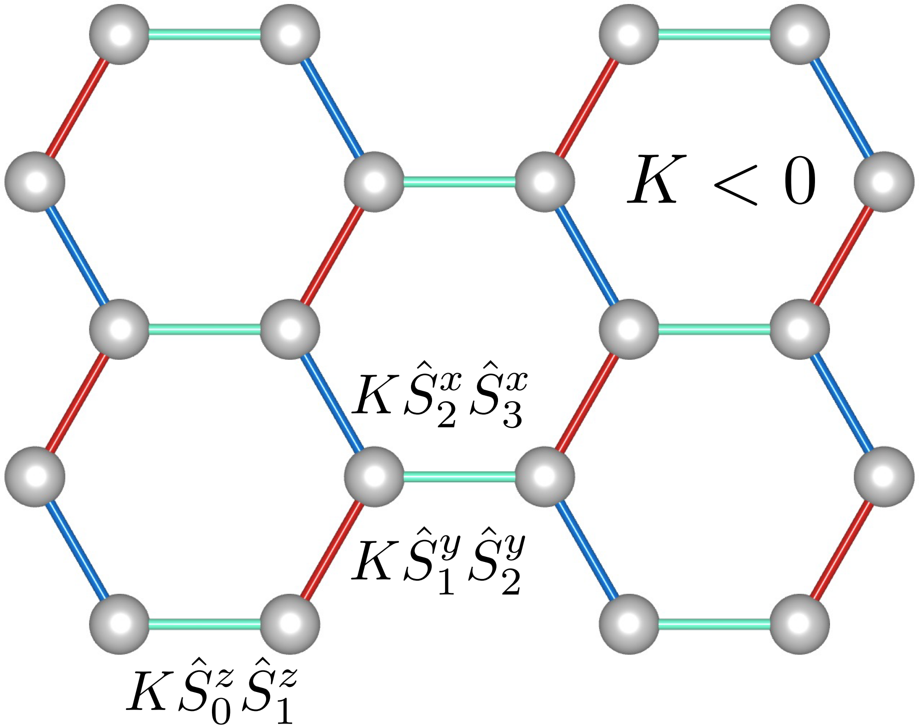
Exactly solvable

Kitaev, Annals Phys. 321, 2 (2006)

Gapless spin liquid:

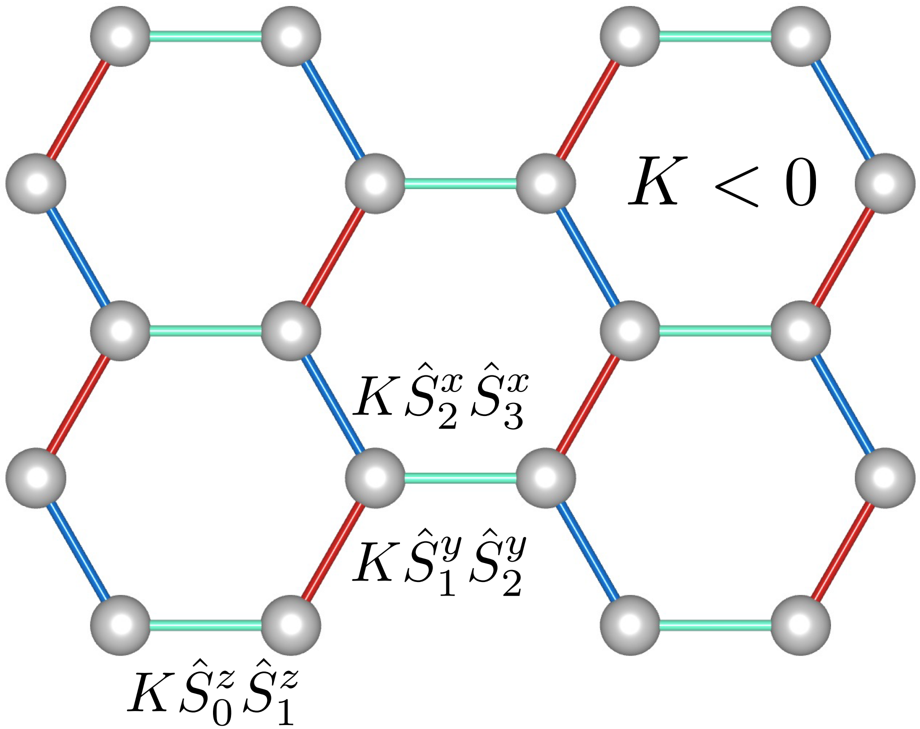
-No long-range order in 2D

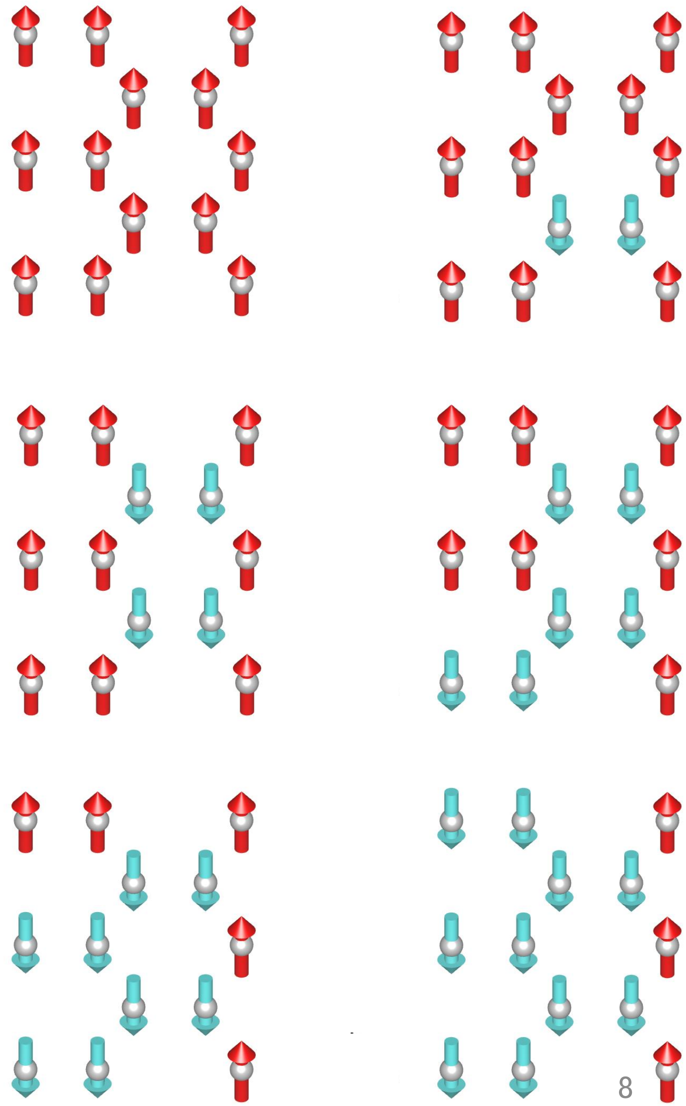
-Fermionic excitation (Majorana)



# An Example of Frustration in Magnets

Honeycomb lattice of S=1/2



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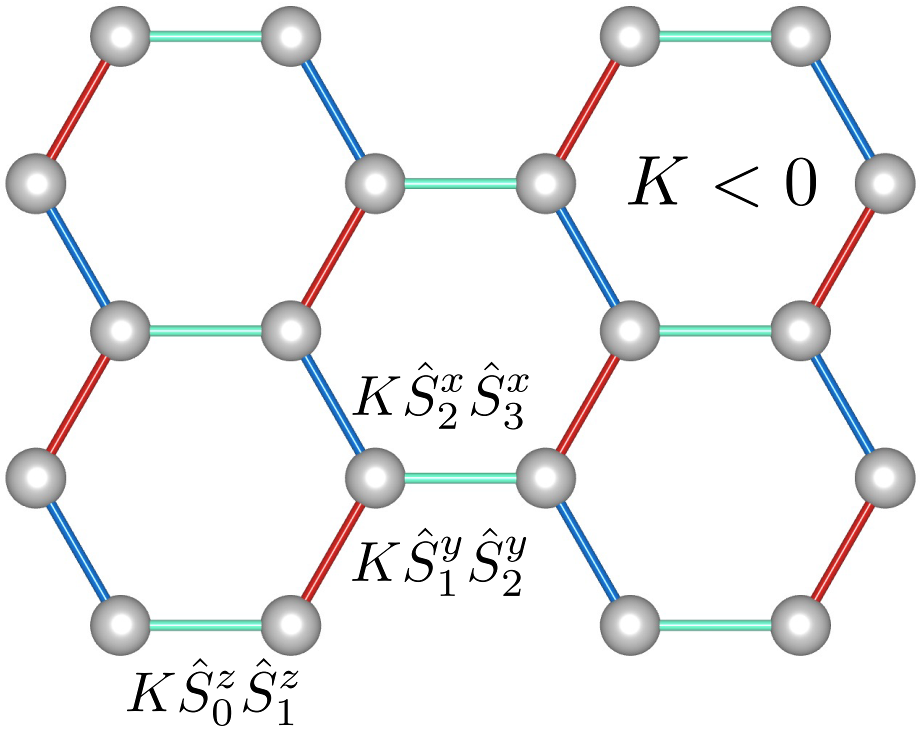
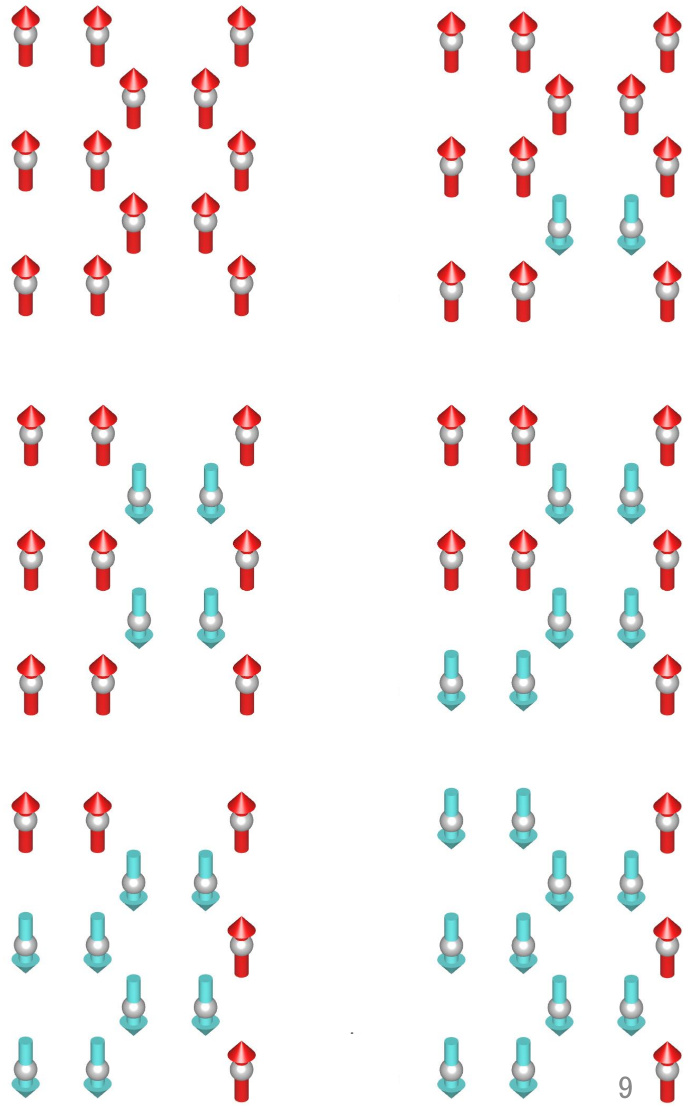
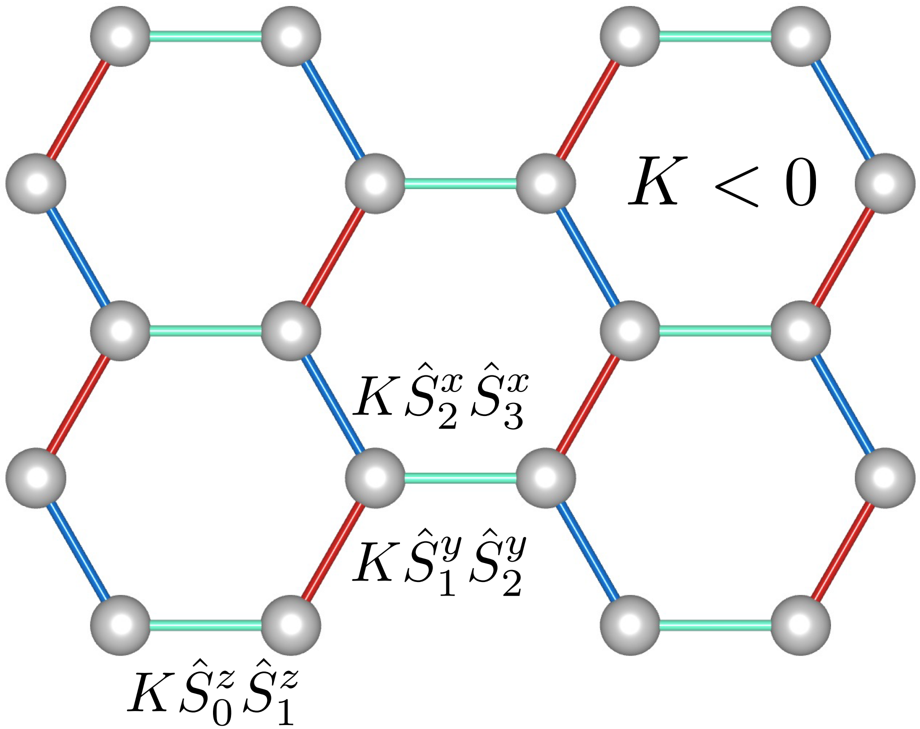


At least in UHF level,
$3 \times 2^{N/2}$ degenerated states !

# An Example of Frustration in Magnets

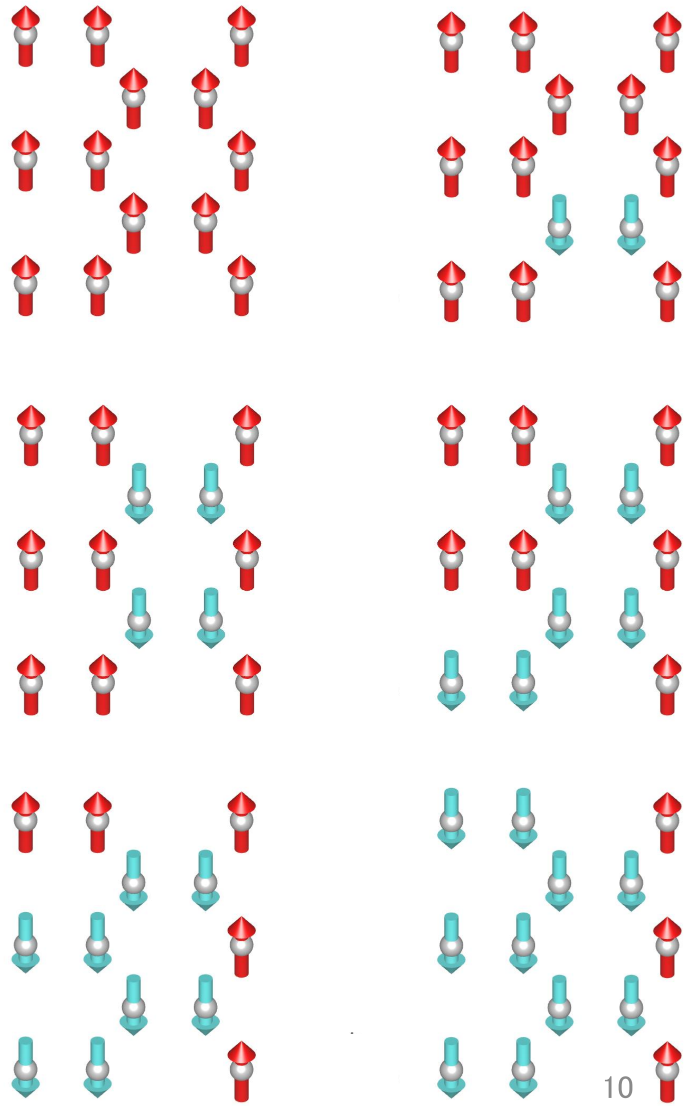
Honeycomb lattice of S=1/2




At least in UHF level,
3x2$^{N/2}$ degenerated states !

No long range order @ $T$=0
Quantum spin liquids

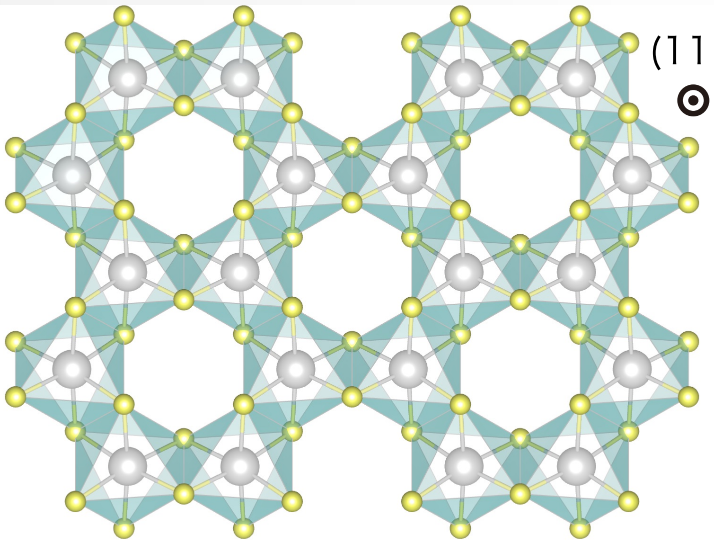
Kitaev, Annals Phys. 321, 2 (2006)



# Anisotropic Exchange Couplings in Honeycomb Iridates

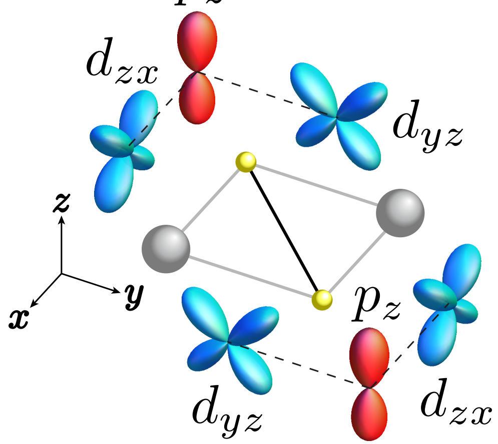
$A_2IrO_3$ $J_{eff}$=1/2 doublet

J. Chaloupka, G. Jackeli, and G.Khaliullin,
Phys. Rev. Lett. **105**, 027204 (2010)




Spin-orbit couplings →
Spin and lattice are locked each other

Ideal octahedron

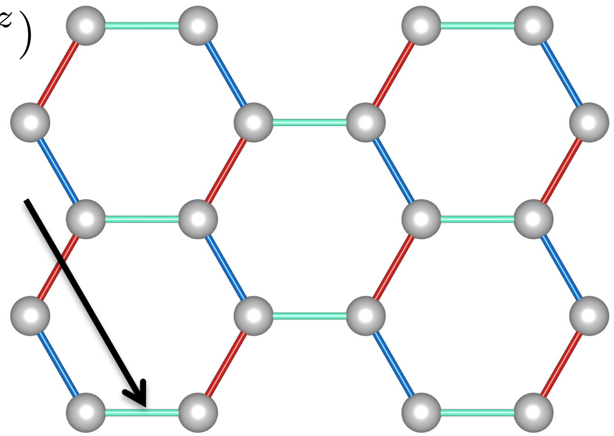



(001) mirror symmetry

$$(\hat{S}^x, \hat{S}^y, \hat{S}^z) \to (-\hat{S}^x, -\hat{S}^y, \hat{S}^z)$$

$$\begin{bmatrix} J & I & 0 \\ I & J & 0 \\ 0 & 0 & K \end{bmatrix}$$

C$_3$ around (111) axis



$$J(\hat{S}_i^x \hat{S}_j^x + \hat{S}_i^y \hat{S}_j^y) + K \hat{S}_i^z \hat{S}_j^z + I(\hat{S}_i^x \hat{S}_j^y + \hat{S}_i^y \hat{S}_j^x)$$

Rau-Kee 2014

*d-p-d* hopping + Hund →
Kitaev $K$<0

# *Ab Initio* Spin Hamiltonian

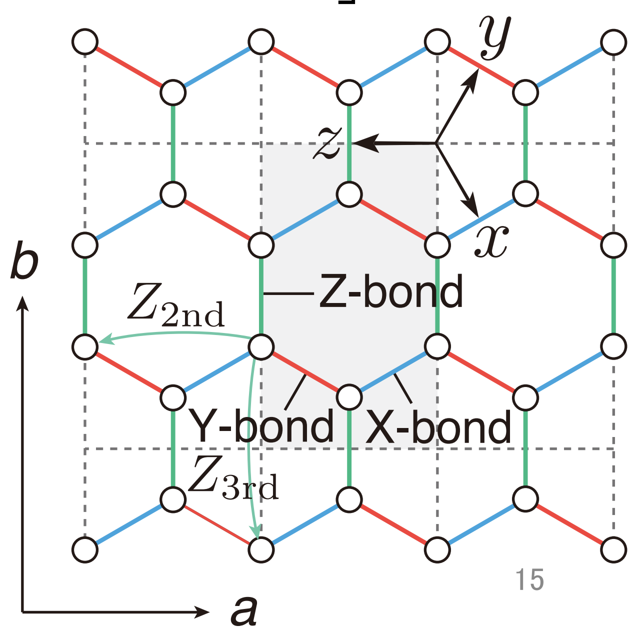
Y. Yamaji, Y. Nomura, M. Kurita, R. Arita, & M. Imada, Phys. Rev. Lett. 113, 107201 (2014).

$$\hat{H} = \sum_{\Gamma = X,Y,Z,Z_{2\mathrm{nd}},3} \sum_{\langle \ell, m \rangle \in \Gamma} \vec{\hat{S}}_\ell^T \mathcal{J}_\Gamma \vec{\hat{S}}_m \qquad \vec{\hat{S}}_\ell^T = (\hat{S}_\ell^x, \hat{S}_\ell^y, \hat{S}_\ell^z)$$

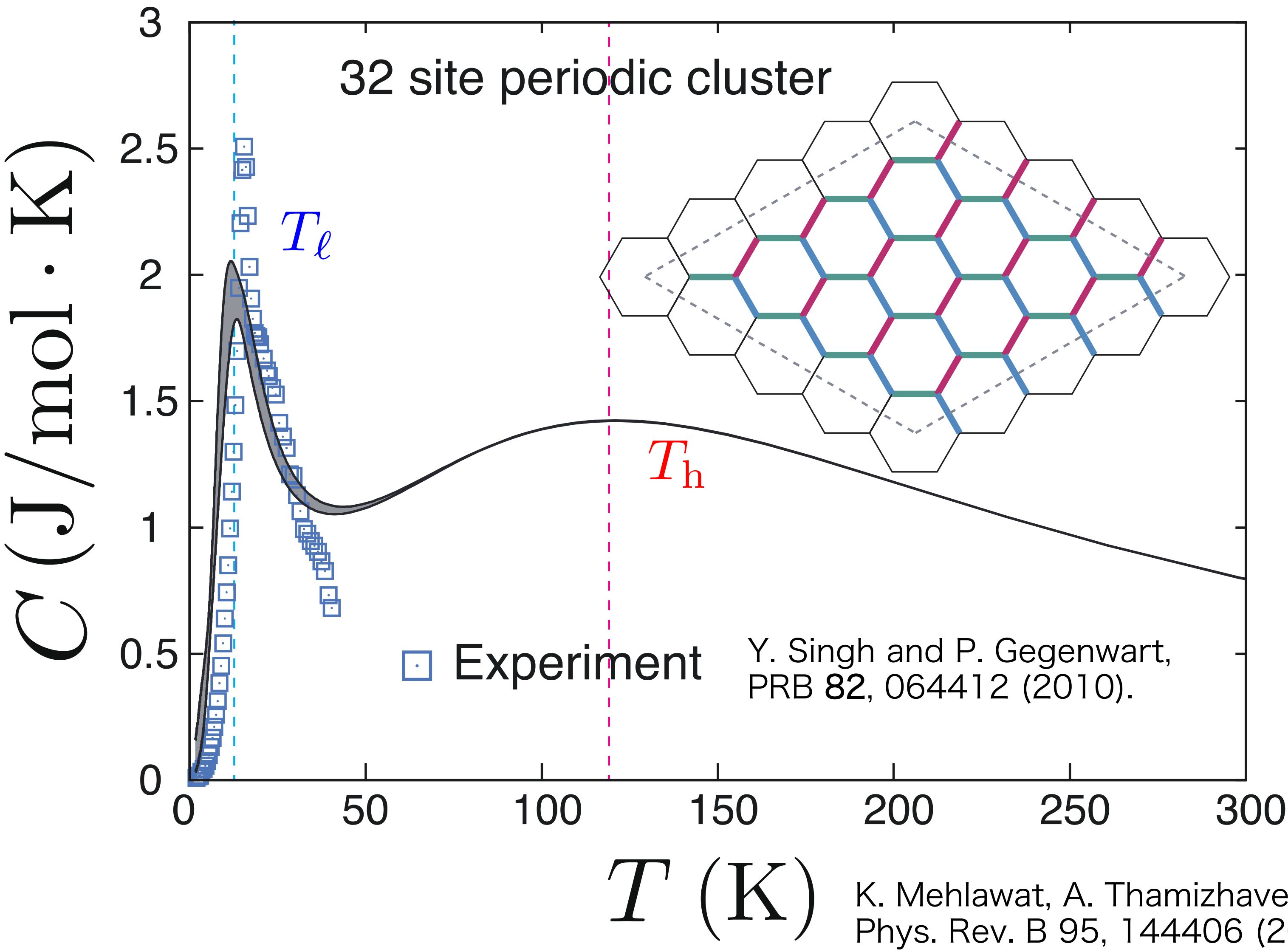
$$\mathcal{J}_X = \begin{bmatrix} \mathbf{-23.9} & -3.1 & -8.4 \\ -3.1 & 3.2 & 1.8 \\ -8.4 & 1.8 & 2.0 \end{bmatrix} (\mathrm{meV}) \qquad \mathcal{J}_{Z_{2\mathrm{nd}}} = \begin{bmatrix} -0.8 & 1.0 & -1.4 \\ 1.0 & -0.8 & -1.4 \\ -1.4 & -1.4 & -1.2 \end{bmatrix} (\mathrm{meV})$$

$$\mathcal{J}_Y = \begin{bmatrix} 3.2 & -3.1 & 1.8 \\ -3.1 & \mathbf{-23.9} & -8.4 \\ 1.8 & -8.4 & 2.0 \end{bmatrix} (\mathrm{meV}) \qquad \mathcal{J}_3 = \begin{bmatrix} 1.7 & 0.0 & 0.0 \\ 0.0 & 1.7 & 0.0 \\ 0.0 & 0.0 & 1.7 \end{bmatrix} (\mathrm{meV})$$

$$\mathcal{J}_Z = \begin{bmatrix} 4.4 & -0.4 & 1.1 \\ -0.4 & 4.4 & 1.1 \\ 1.1 & 1.1 & \mathbf{-30.7} \end{bmatrix} (\mathrm{meV})$$



# Specific Heat of $Na_2IrO_3$ by TPQ




K. Mehlawat, A. Thamizhavel & Y. Singh, Phys. Rev. B 95, 144406 (2017).

$T_h \sim 120$ K

# Spin Excitations

# Simulating Spectroscopy Measurements

**Linear response of ground state $|\psi\rangle$**

Green's function

$$G_{\hat{O}}(z) = \langle\psi|\hat{O}^{\dagger}(z\mathbf{1} - \hat{H})^{-1}\hat{O}|\psi\rangle$$

$$z \to \omega \quad (z \in \mathbb{C},\ \omega \in \mathbb{R})$$

**Excitation spectrum** $-\frac{1}{\pi}\mathrm{Im}G_{\hat{O}}(\omega + i\delta)$

Example of perturbation & response:

Magnetization of spins under magnetic fields

$$\hat{H}_{\mathrm{ex}} = e^{i\omega t}B_z\left(\frac{1}{N}\sum_{j=0}^{N-1}\hat{S}_j^z\right) \qquad \hat{O} = \frac{1}{N}\sum_{j=0}^{N-1}\hat{S}_j^z$$

# Shifted Krylov Subspace Method for Excitation Spectra

Green's function $G_{\hat{O}}(z) = \langle\psi|\hat{O}^{\dagger}(z\mathbf{1} - \hat{H})^{-1}\hat{O}|\psi\rangle$

$$z \to \omega \quad (z \in \mathbb{C},\ \omega \in \mathbb{R})$$

Liner equations

$$(z\mathbf{1} - H)\vec{x} = \vec{b} \qquad \vec{b} \doteq \hat{O}|\psi\rangle$$

$$\Rightarrow G_{\hat{O}}(z) = \vec{b}^{\dagger}\vec{x} \qquad \vec{x} \doteq (z\mathbf{1} - \hat{H})^{-1}\hat{O}|\psi\rangle$$

←Solvable by Shifted Krylov subspace method

A. Frommer (1995, 2003)

T. Sogabe, T. Hoshi, S. L. Zhang, and T. Fujiwara, *A numerical method for calculating the Green's function arising from electronic structure theory*, In Frontiers of Computational Science. pp.189-195, 2007.

→Stable and controlled convergence

# Shifted Krylov Subspace Method for Excitation Spectra

-Shift invariance of Krylov subspace

-Collinear residuals

A. Frommer, Computing 70, 87 (2003).

$$\vec{r}_n \propto \vec{r}_n^{\sigma}$$

$$(z\mathbf{1} - H)\vec{x} = \vec{b} \qquad ((z+\sigma)\mathbf{1} - H)\vec{x} = \vec{b}$$

$$\vec{r}_n = \vec{b} - (z\mathbf{1} - H)\vec{x}_n \qquad \vec{r}_n^{\sigma} = \vec{b} - ((z+\sigma)\mathbf{1} - H)\vec{x}_n^{\sigma}$$

Seed switch

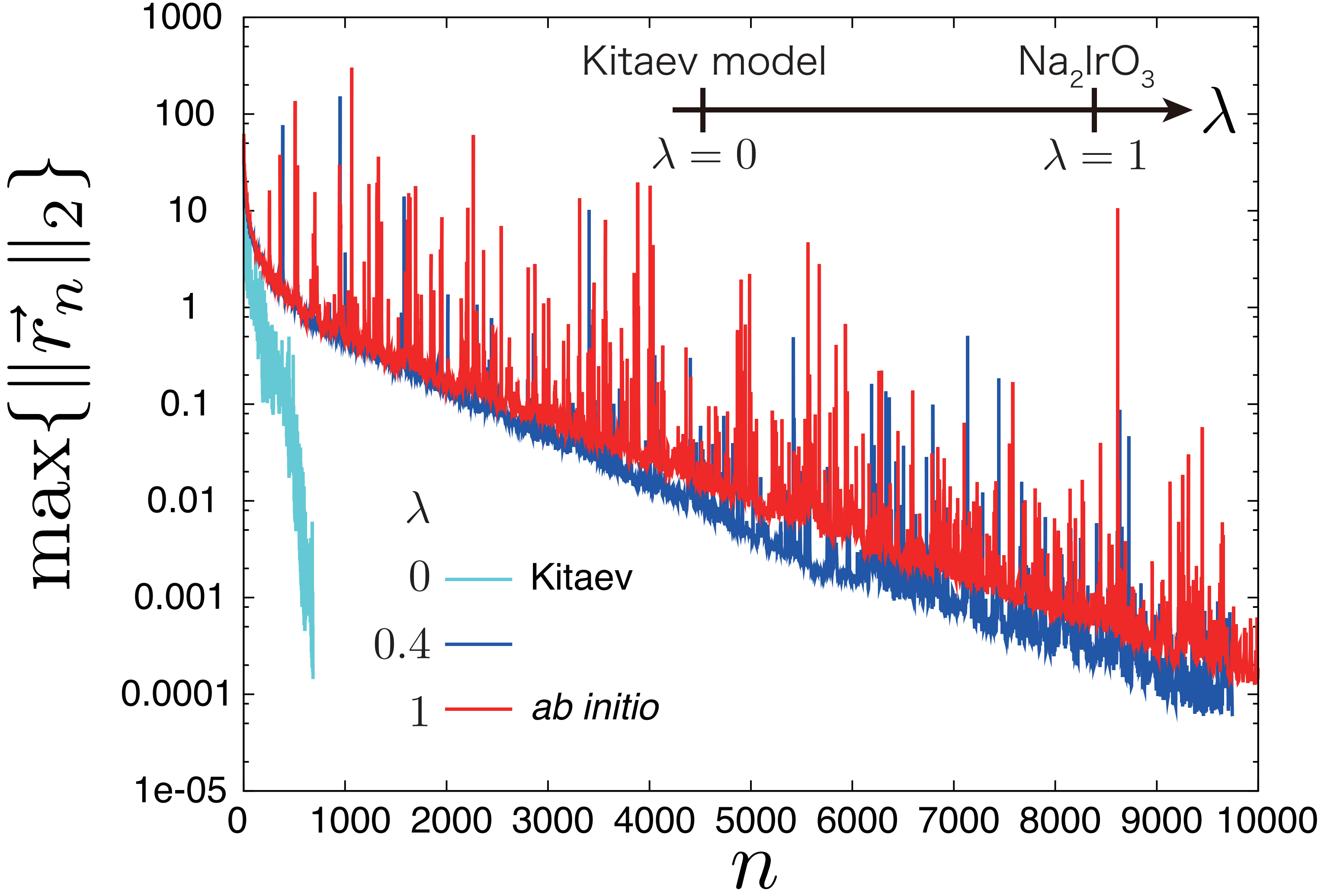
S. Yamamoto, T. Sogabe, T. Hoshi, S.-L. Zhang, & T. Fujiwara,
J. Phys. Soc. Jpn. 77, 114713 (2008).

Library K$\omega$ (released) by Dr. Kawamura (ISSP)

# 2-Norm of Residual Vector

Strong parameter dependence
in convergence of S(Q,$\omega$) with sBiCG

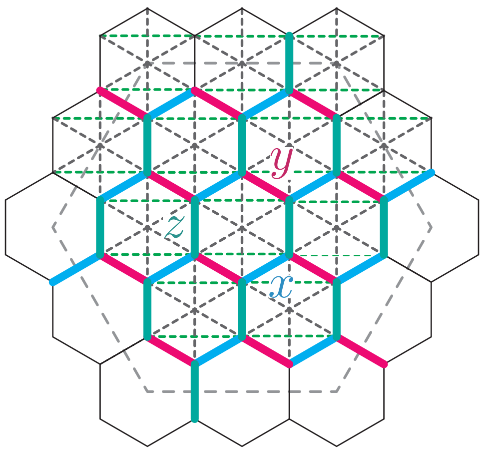
$$\hat{O} = \frac{1}{\sqrt{N}} \sum_{j=0}^{N-1} \hat{S}_j^z$$

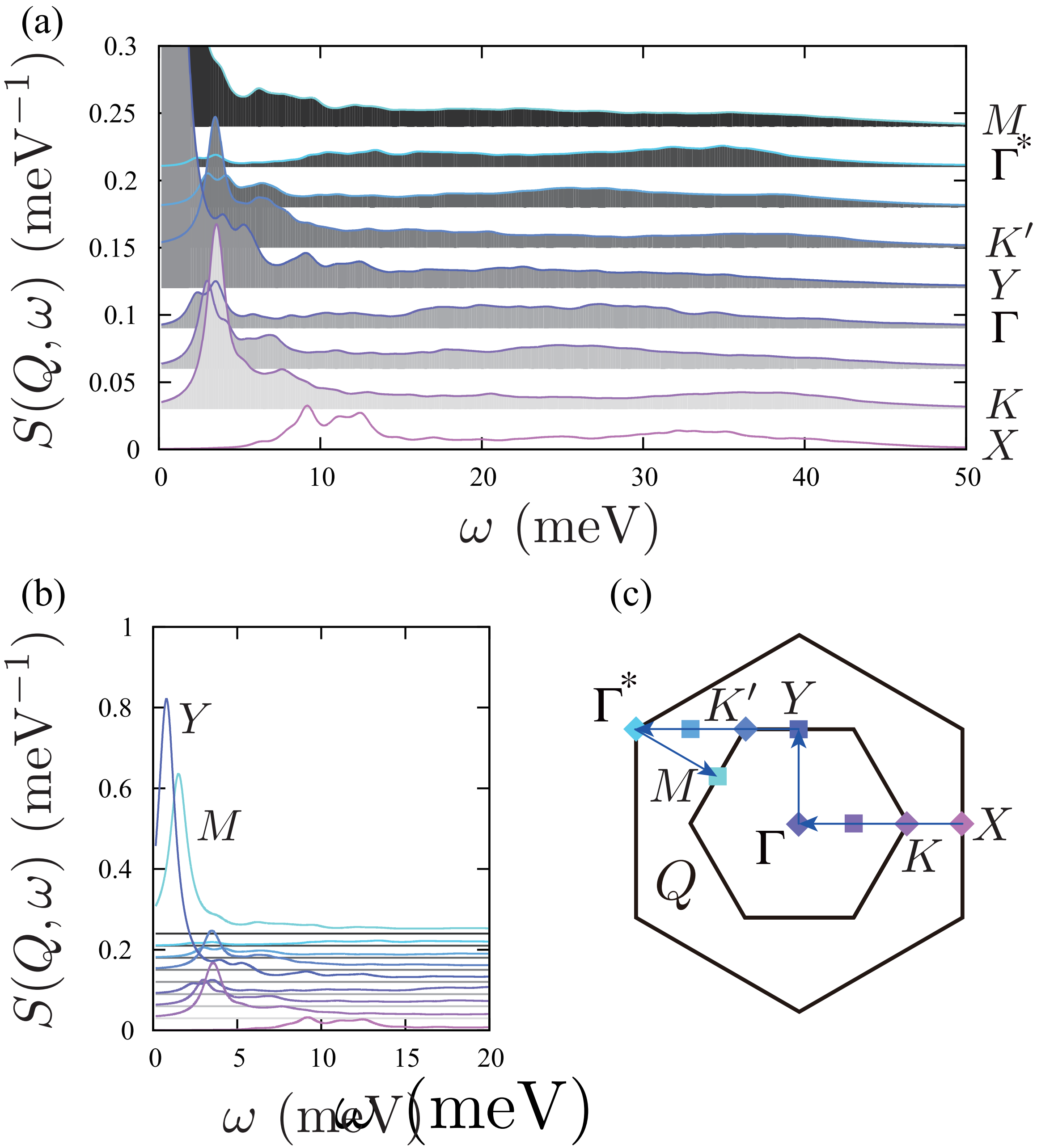


# Dynamical Spin Structure Factors

$\lambda = 1.0$

zigzag



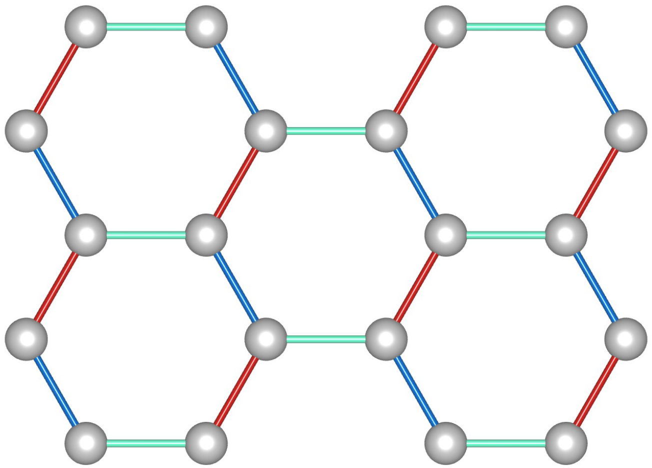
# Application of HΦ: Exploration of A “New” Spin Liquid

# Exploration of A "New" Spin Liquid

## $K$-$\Gamma$ model

cf.) Classical macroscopic degeneracy
I. Rousochatzakis & N. B. Parkins,
arXiv:1610.08463.

A. Catuneau, Y. Yamaji, G. Wachtel, H.-Y. Kee, & Y.-B. Kim,
arXiv:1701.07837.



$$\mathcal{J}_X = \begin{bmatrix} -(1-a)\cos\varphi & 0 & 0 \\ 0 & 0 & \sin\varphi \\ 0 & \sin\varphi & 0 \end{bmatrix}$$

$$\mathcal{J}_Y = \begin{bmatrix} 0 & 0 & \sin\varphi \\ 0 & -(1-a)\cos\varphi & 0 \\ \sin\varphi & 0 & 0 \end{bmatrix}$$

$$\mathcal{J}_Z = \begin{bmatrix} 0 & \sin\varphi & 0 \\ \sin\varphi & 0 & 0 \\ 0 & 0 & -(1+2a)\cos\varphi \end{bmatrix}$$

$$a = 0.1$$

$\alpha$-$RuCl_3$ H.-S. Kim & H.-Y. Kee,
Phys. Rev. B 93, 155143 (2016).

$\alpha$-$RuCl_3$ H.-S. Kim & H.-Y. Kee,
Phys. Rev. B 93, 155143 (2016).

One-body: *ab initio* TB

Coulomb: parameters

$$U = 3\ \mathrm{eV},\ \ J_{\mathrm{H}}/U = 0.15\ \mathrm{eV}$$

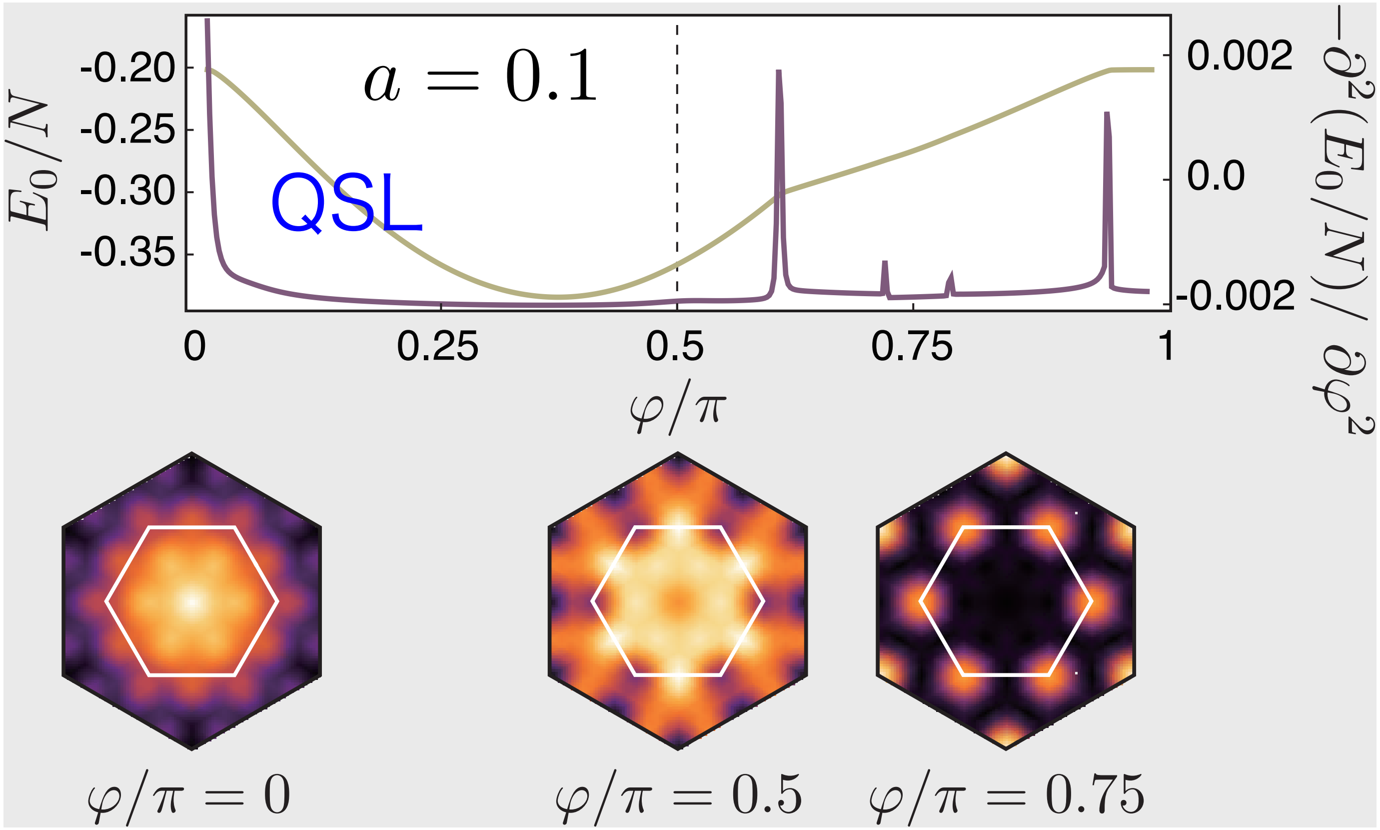
$$\mathcal{J}_X = \begin{bmatrix} -7.64 & -0.87 & -0.87 \\ -0.87 & -1.09 & 4.38 \\ -0.87 & 4.38 & -1.09 \end{bmatrix} \ (\mathrm{meV})$$

$$\mathcal{J}_Z = \begin{bmatrix} -0.74 & +3.71 & -1.04 \\ +3.71 & -0.74 & -1.04 \\ -1.04 & -1.04 & -9.34 \end{bmatrix} \ (\mathrm{meV})$$

# Extension of Kitaev's Spin Liquid

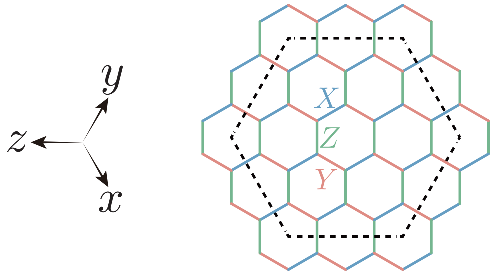
**Kitaev-Γ model**

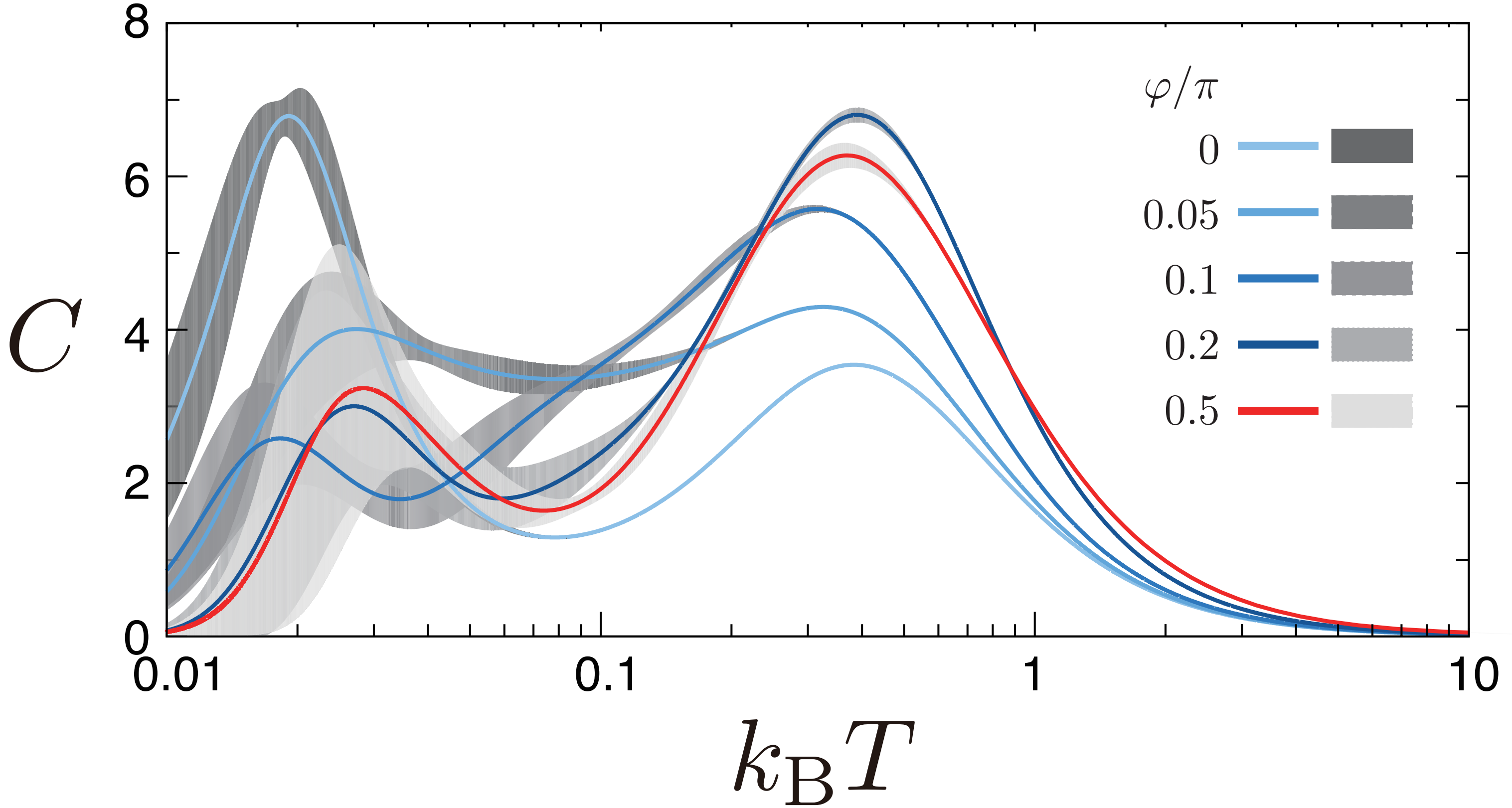
A. Catuneau, Y. Yamaji, G. Wachtel, H.-Y. Kee, & Y.-B. Kim, arXiv:1701.07837.



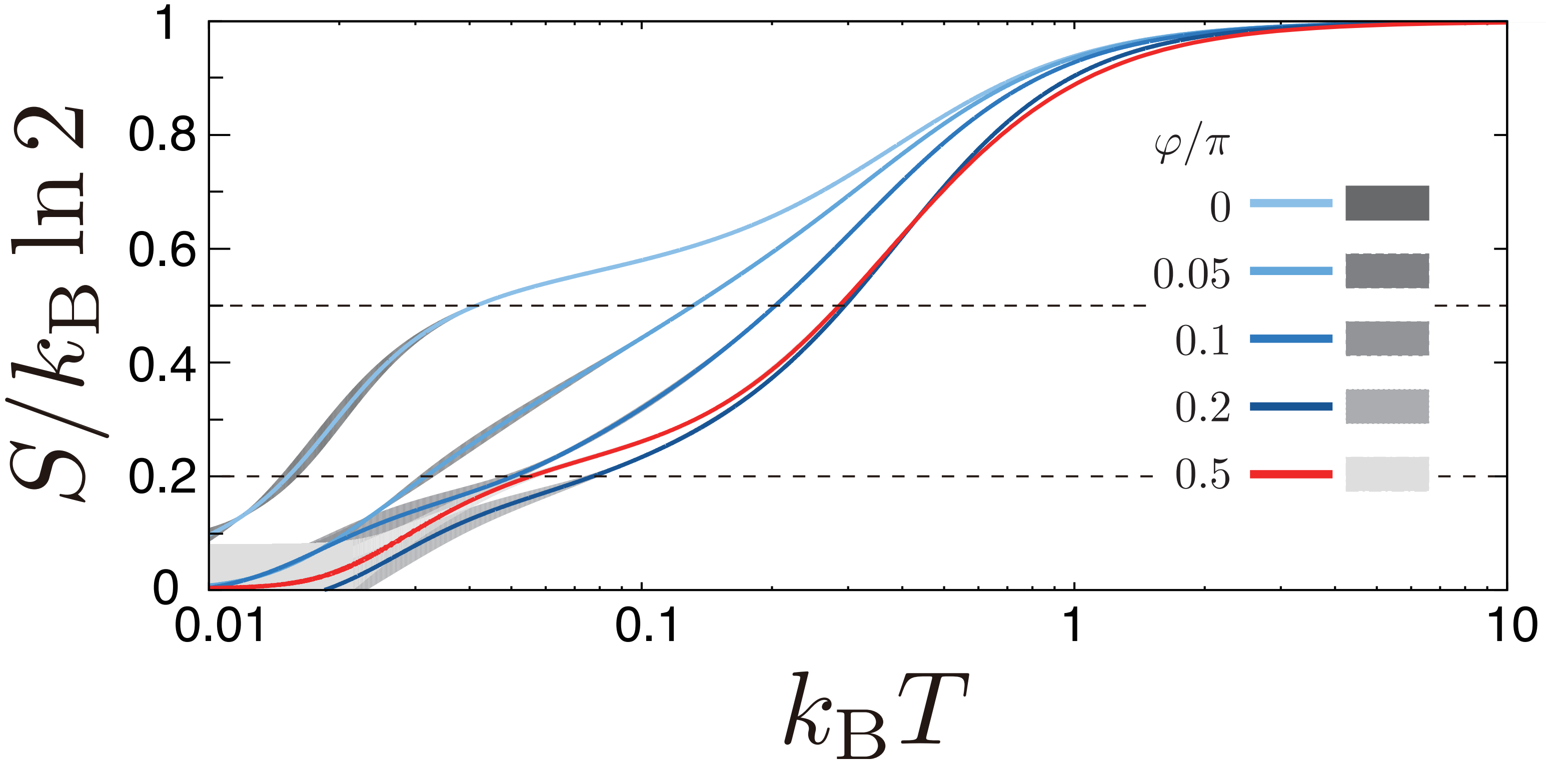

QSL adiabatically connected to Kitaev limit

# Heat Capacity

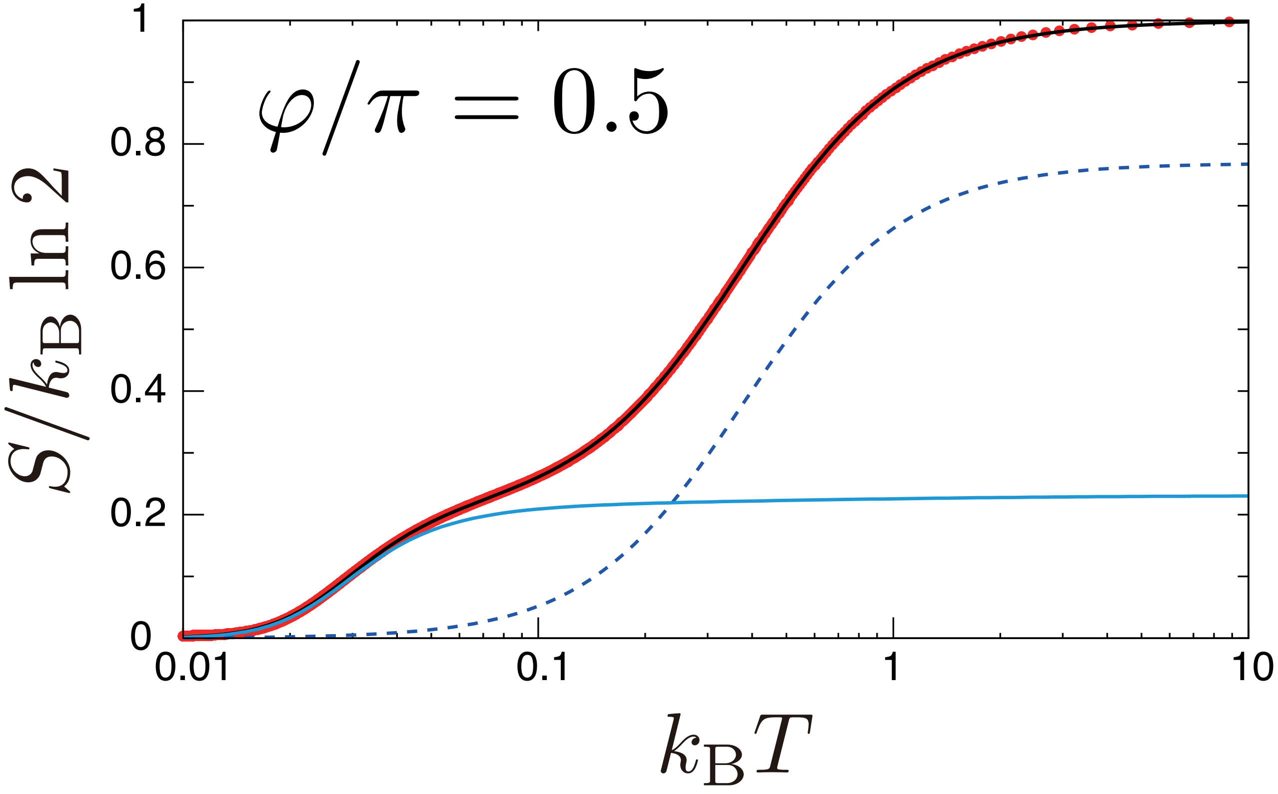


# Plateau of Entropy



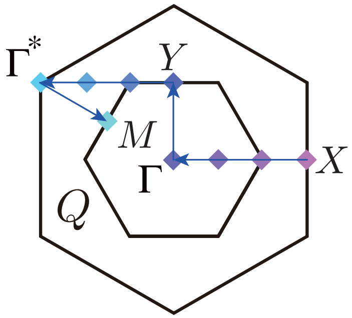
# Decomposition of Entropy

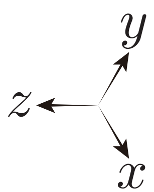


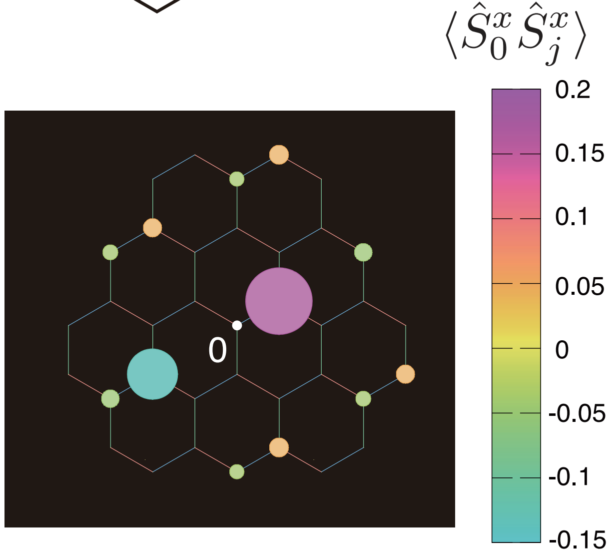

A new plateau appears

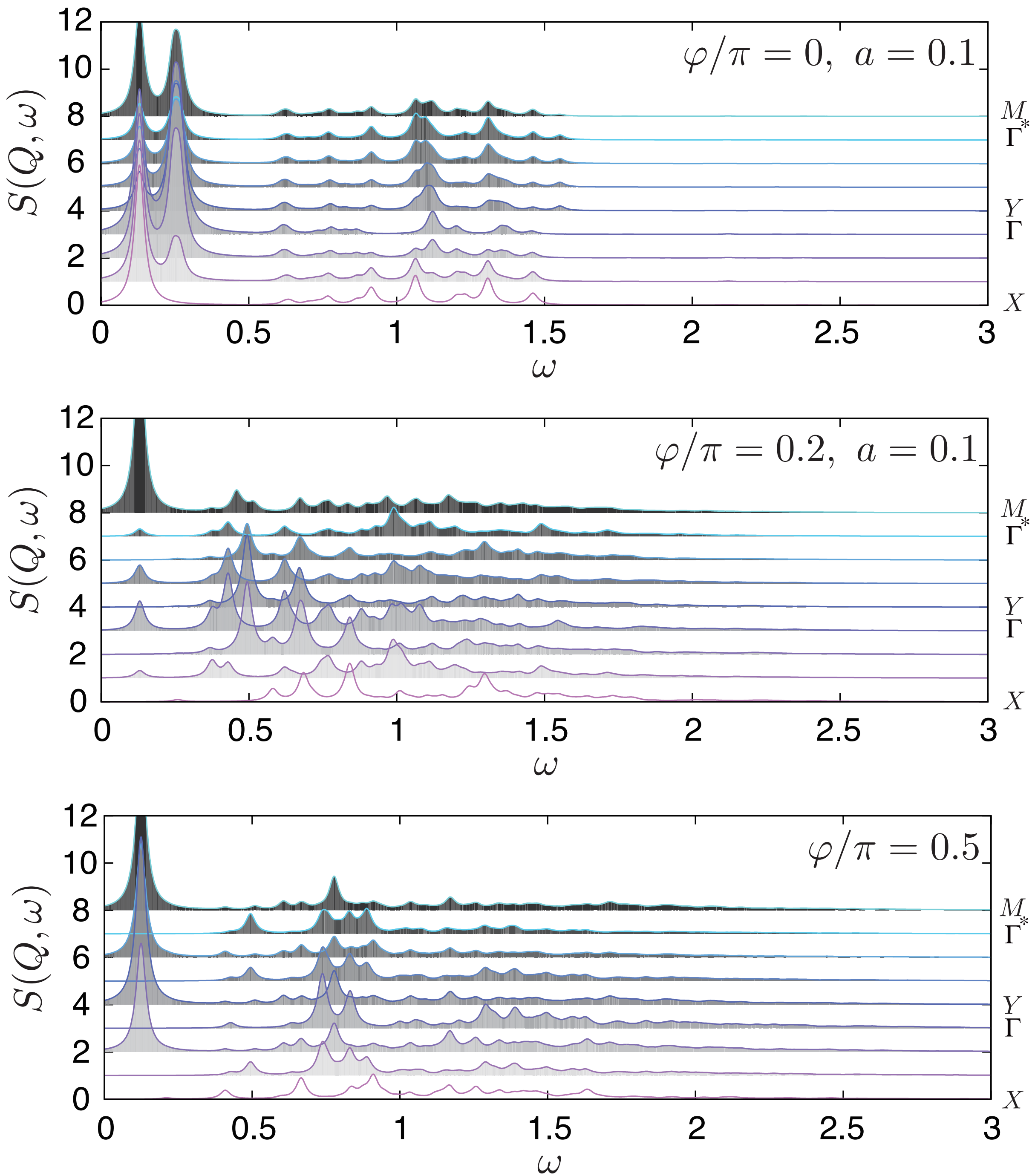
# $S(Q,\omega)$ by $K\omega$

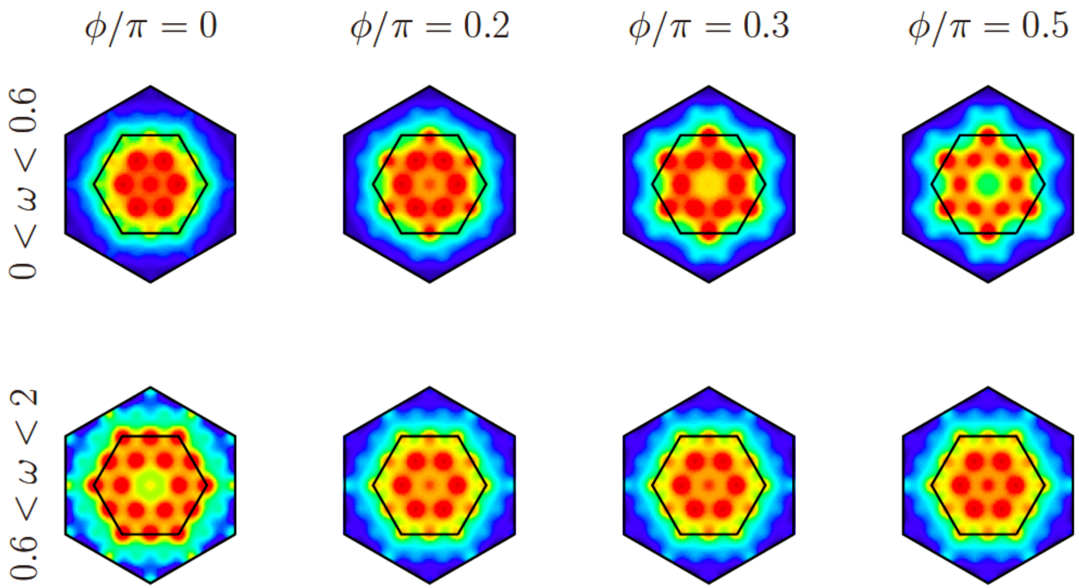
-Local excitations
⇔Short range correlations
-Continuum



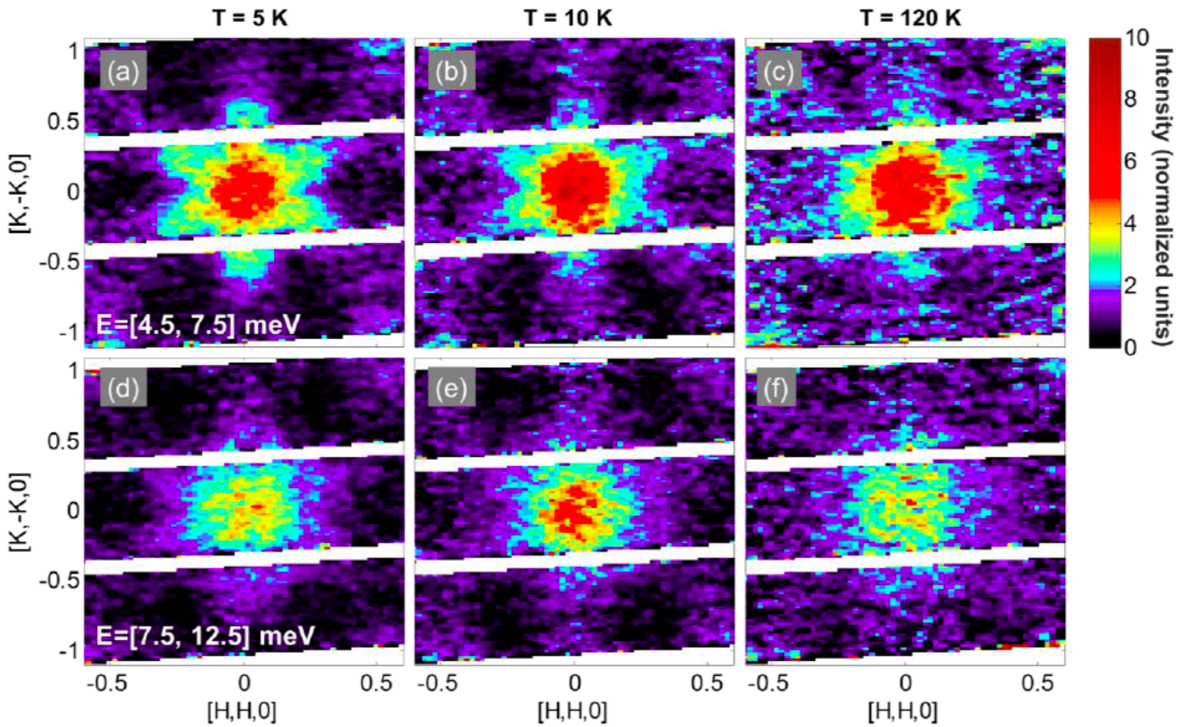





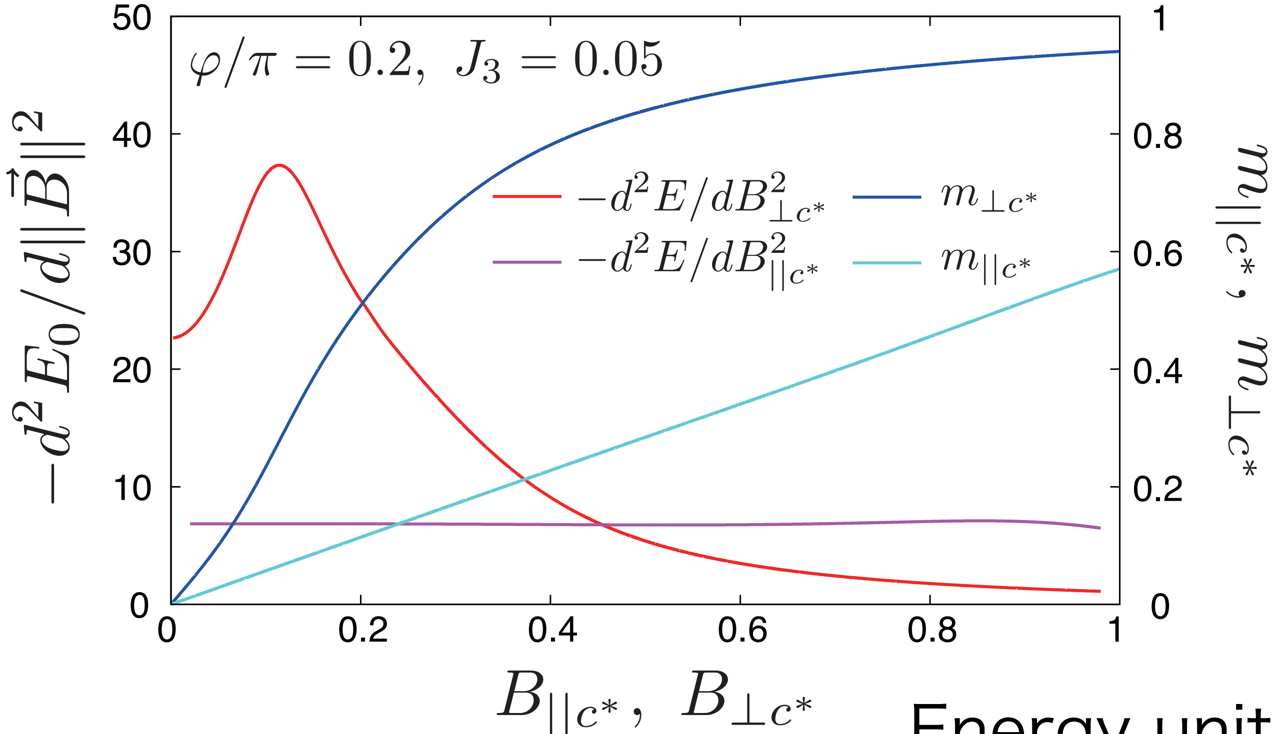

INS for $\alpha$-$RuCl_3$
A. Banerjee *et al.*,
arXiv:1609.00103.



# Relevance of $K$-$\Gamma$-$J_3$ to $\alpha$-$RuCl_3$

-Easy plane anisotropy
-Transition from zigzag to forced FM at ～10T

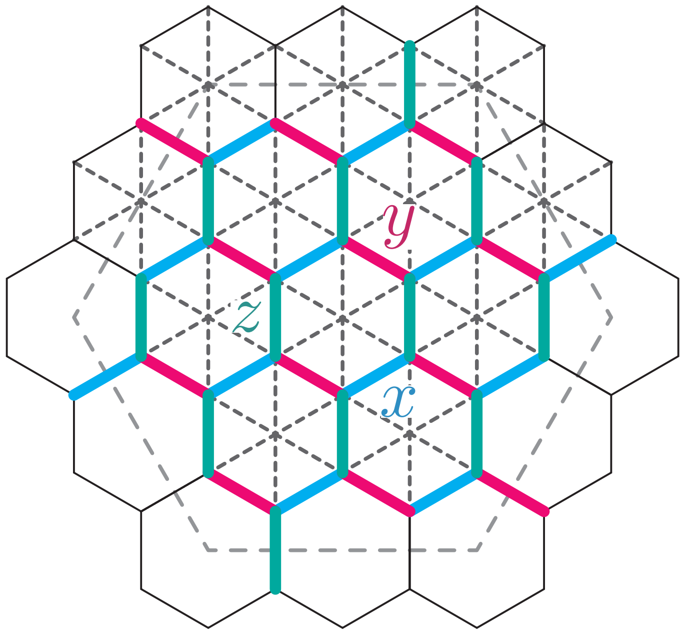
J. A. Sears, *et al.*, Phys. Rev. B **91**, 144420 (2015).
M. Majumder, *et al.*, Phys. Rev. B **91**, 180401 (2015).
R. D. Johnson, *et al*, Phys. Rev. B **92**, 235119 (2015).




Energy unit ～ 10 meV

# How to Simulate $K$-$\Gamma$-$J_3$ Model by HΦ

$$\hat{H} = \sum_{\Gamma=X,Y,Z,3} \sum_{\langle \ell,m \rangle \in \Gamma} \hat{\vec{S}}_\ell^T \mathcal{J}_\Gamma \hat{\vec{S}}_m \qquad \hat{\vec{S}}_\ell^T = (\hat{S}_\ell^x, \hat{S}_\ell^y, \hat{S}_\ell^z)$$




$$\mathcal{J}_X = \begin{bmatrix} -\cos\phi & 0 & 0 \\ 0 & 0 & \sin\phi \\ 0 & \sin\phi & 0 \end{bmatrix}$$

$$\mathcal{J}_Y = \begin{bmatrix} 0 & 0 & \sin\phi \\ 0 & -\cos\phi & 0 \\ \sin\phi & 0 & 0 \end{bmatrix}$$

$$\mathcal{J}_Z = \begin{bmatrix} 0 & \sin\phi & 0 \\ \sin\phi & 0 & 0 \\ 0 & 0 & -\cos\phi \end{bmatrix}$$

Nearest neighbor

$$\mathcal{J}_3 = \begin{bmatrix} J_3 & 0 & 0 \\ 0 & J_3 & 0 \\ 0 & 0 & J_3 \end{bmatrix}$$

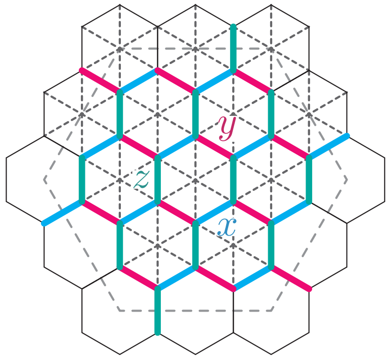
3rd neighbor

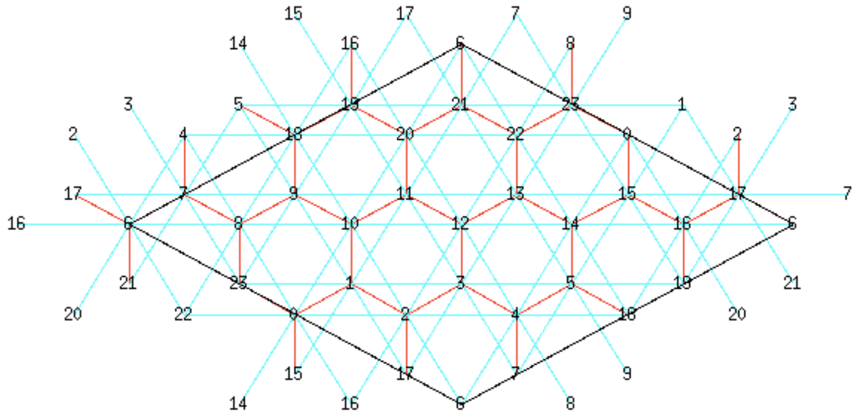
$$J_3[\hat{S}_\ell^x \hat{S}_m^x + \hat{S}_\ell^y \hat{S}_m^y + \hat{S}_\ell^z \hat{S}_m^z]$$

# How to Simulate $K$-$\Gamma$-$J_3$ Model

$$\phi/\pi = 0.2$$

```
model = "SpinGC"
method = "TPQ"
lattice = "Honeycomb"
a0w = 2
a0l = 2
a1w = 4
a1l = -2
J0x = -0.80901699437
J0yz = 0.58778525229
J0zy = 0.58778525229
J1zx = 0.58778525229
J1y = -0.80901699437
J1xz = 0.58778525229
J2xy = 0.58778525229
J2yx = 0.58778525229
J2z = -0.80901699437
h = 0.07071067811
Gamma = -0.07071067811
2S=1
```



$$\mathcal{J}_X = \begin{bmatrix} -\cos\phi & 0 & 0 \\ 0 & 0 & \sin\phi \\ 0 & \sin\phi & 0 \end{bmatrix}$$

$$\mathcal{J}_Y = \begin{bmatrix} 0 & 0 & \sin\phi \\ 0 & -\cos\phi & 0 \\ \sin\phi & 0 & 0 \end{bmatrix}$$

$$\mathcal{J}_Z = \begin{bmatrix} 0 & \sin\phi & 0 \\ \sin\phi & 0 & 0 \\ 0 & 0 & -\cos\phi \end{bmatrix}$$

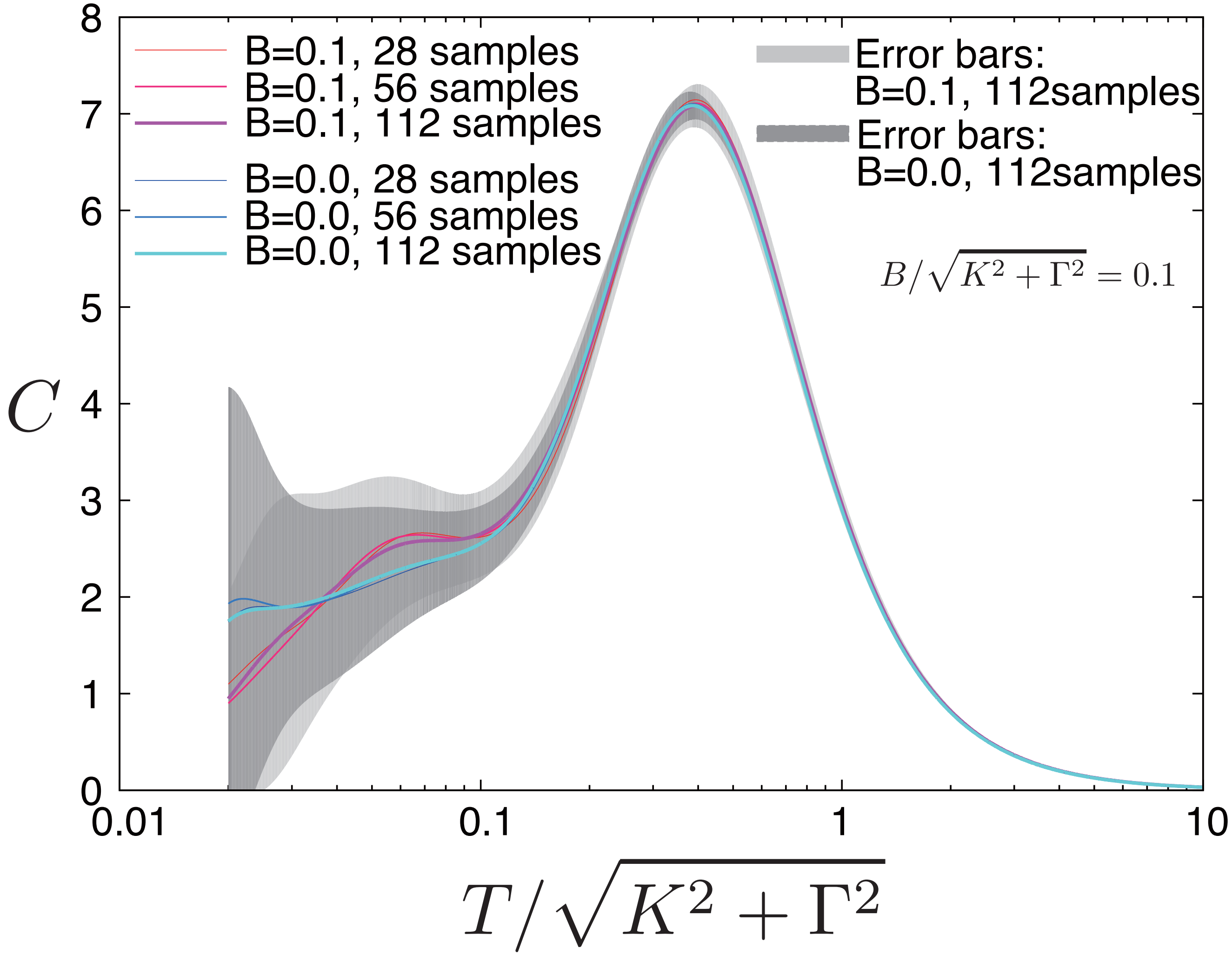
$$\vec{B} \propto (1, 0, -1)$$

$$\mathcal{J}_3 = \begin{bmatrix} J_3 & 0 & 0 \\ 0 & J_3 & 0 \\ 0 & 0 & J_3 \end{bmatrix}$$

Add `Exchange` and `Ising` by Expert mode

# Heat Capacity of $K$-$\Gamma$-$J_3$ Model

$$\phi/\pi = 0.2, \;\; J_3/\sqrt{K^2+\Gamma^2} = 0.05$$



# Typicality Approach

## Finite-temperature pure state

$$|\phi_\beta\rangle = e^{-\beta \hat{H}/2}|\phi_0\rangle$$

$$\langle \hat{O} \rangle_\beta^{\mathrm{ens}} = \frac{\mathbb{E}[\langle \phi_\beta | \hat{O} | \phi_\beta \rangle]}{\mathbb{E}[\langle \phi_\beta | \phi_\beta \rangle]}$$

M. Imada and M. Takahashi, J. Phys. Soc. Jpn. 55, 3354 (1986).
P. de Vries and H. De Raedt, Phys. Rev. B 47, 7929 (1993).
A. Hams and H. De Raedt, Phys. Rev. E 62, 4365 (2000).

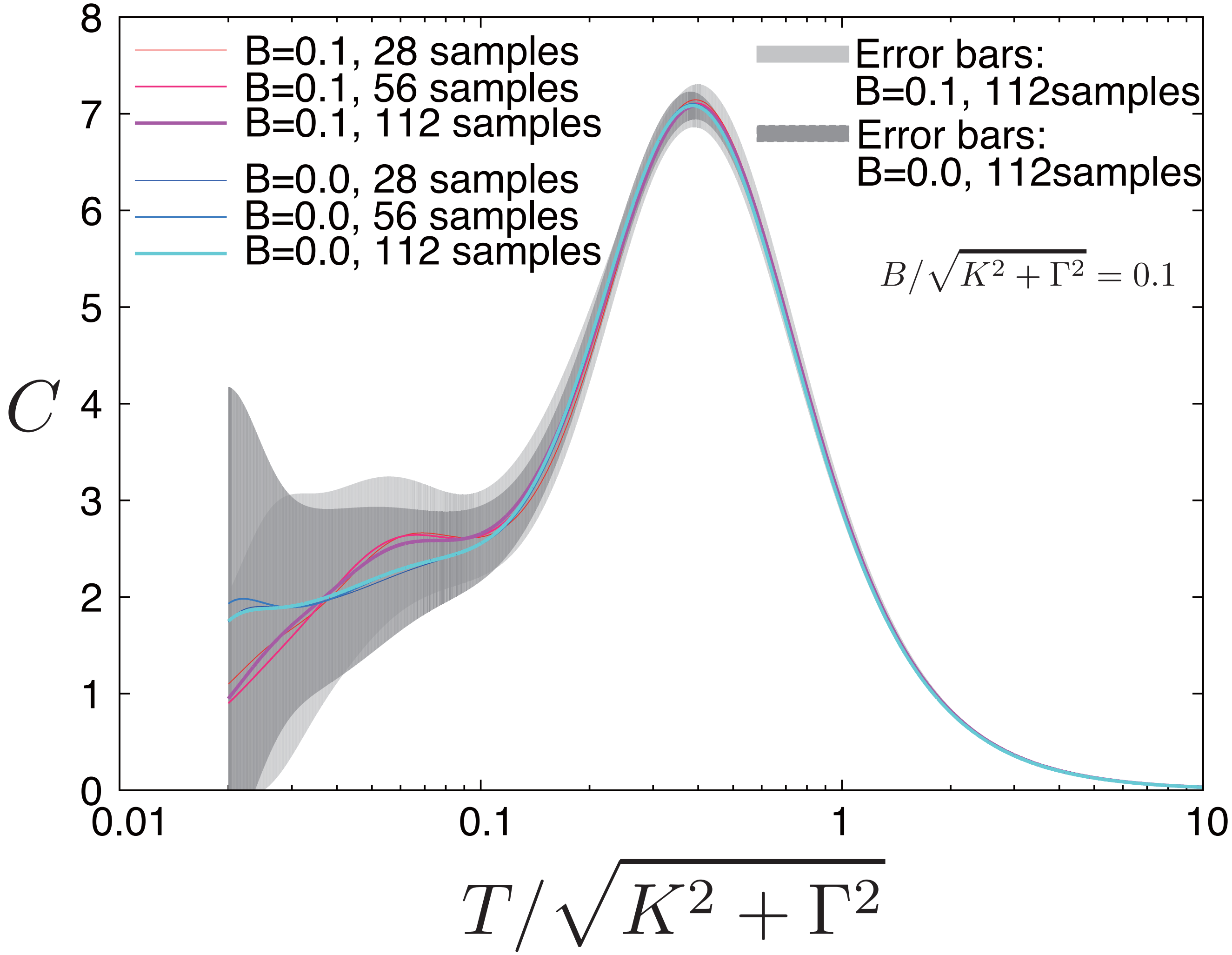
S. Sugiura and A. Shimizu, Phys. Rev. Lett. 111, 010401 (2013).

$$\sigma_O^2 = \mathbb{E}\left[\left(\frac{\langle \phi_\beta | \, \hat{O} \, | \phi_\beta \rangle}{\langle \phi_\beta | \phi_\beta \rangle} - \langle \hat{O} \rangle_\beta^{\mathrm{ens}}\right)^2\right]$$

$$\sigma_O^2 \leq \frac{\langle (\Delta O)^2 \rangle_{2\beta}^{\mathrm{ens}} + (\langle O \rangle_{2\beta}^{\mathrm{ens}} - \langle O \rangle_\beta^{\mathrm{ens}})^2}{\exp[2\beta\{F(2\beta) - F(\beta)\}]}$$

# Heat Capacity of $K$-$\Gamma$-$J_3$ Model

$$\phi/\pi = 0.2,\ \ J_3/\sqrt{K^2+\Gamma^2} = 0.05$$



# まとめ

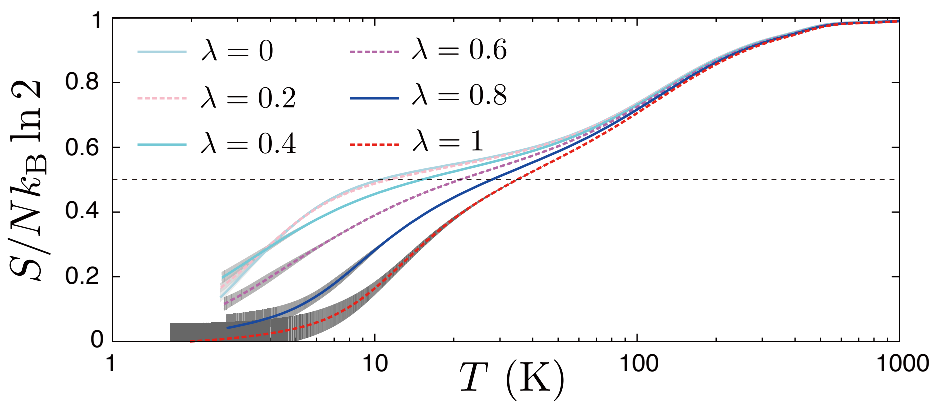
## HΦによる量子スピン液体近傍の熱励起とスピン励起

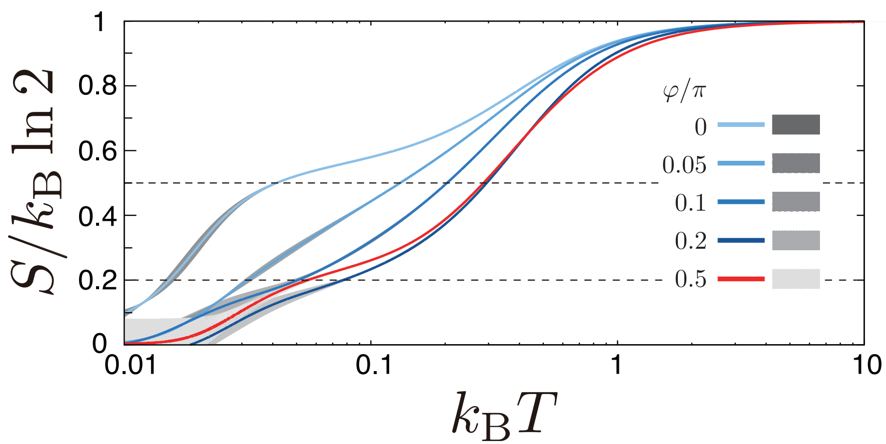
自発的対称性の破れのアリバイにかわる証拠探し

-低温まで残るエントロピー

-励起の局在性と連続スペクトル

- $Na_2IrO_3$
  低温まで残る比熱
  局在的連続スペクトルからスピン波的励起へ
- キタエフ-Γ模型における拡がった量子スピン液体相
  エントロピー・プラトーのクロスオーバー
  頑健な局在的連続スペクトル



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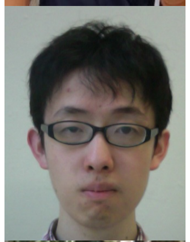

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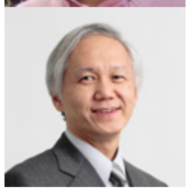

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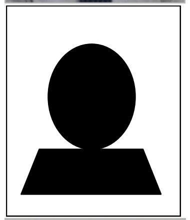

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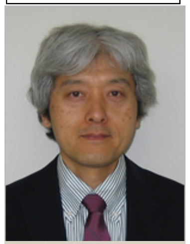

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