キタエフスピン液体における
マヨラナ励起

求 幸年 (東大院工)

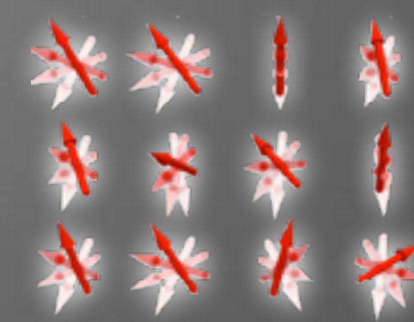
THE UNIVERSITY OF TOKYO

Contents

- **What is the quantum spin liquid?**
  difficulty in search for quantum spin liquids
- **breakthrough:** Kitaev model
  rare example of exact quantum spin liquids
  spin fractionalization into Majorana fermions
  realization in spin-orbit coupled Mott insulators
- **fingerprints of Majorana excitations at finite *T***
  comparison between theory and experiment on
  - specific heat and entropy
  - magnetic Raman scattering
  - thermal conductivity
  - inelastic neutron scattering
- **Summary and perspectives**

Fate of magnets

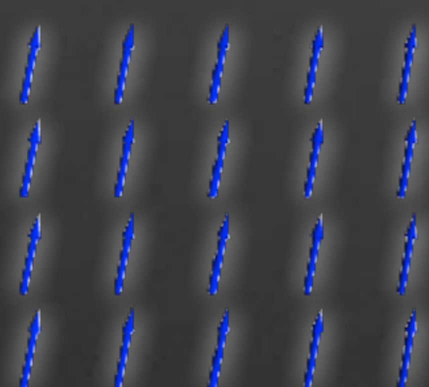
paramagnet



long-range order?

YES → **magnetic order**

- symmetry breaking
- phase transition
- collective excitations: magnon (boson)



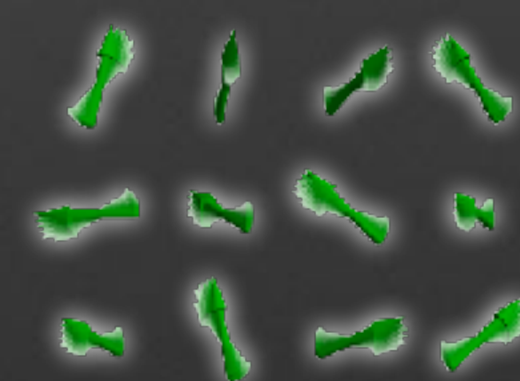
NO → *spin freezing?*

YES → **spin glass**

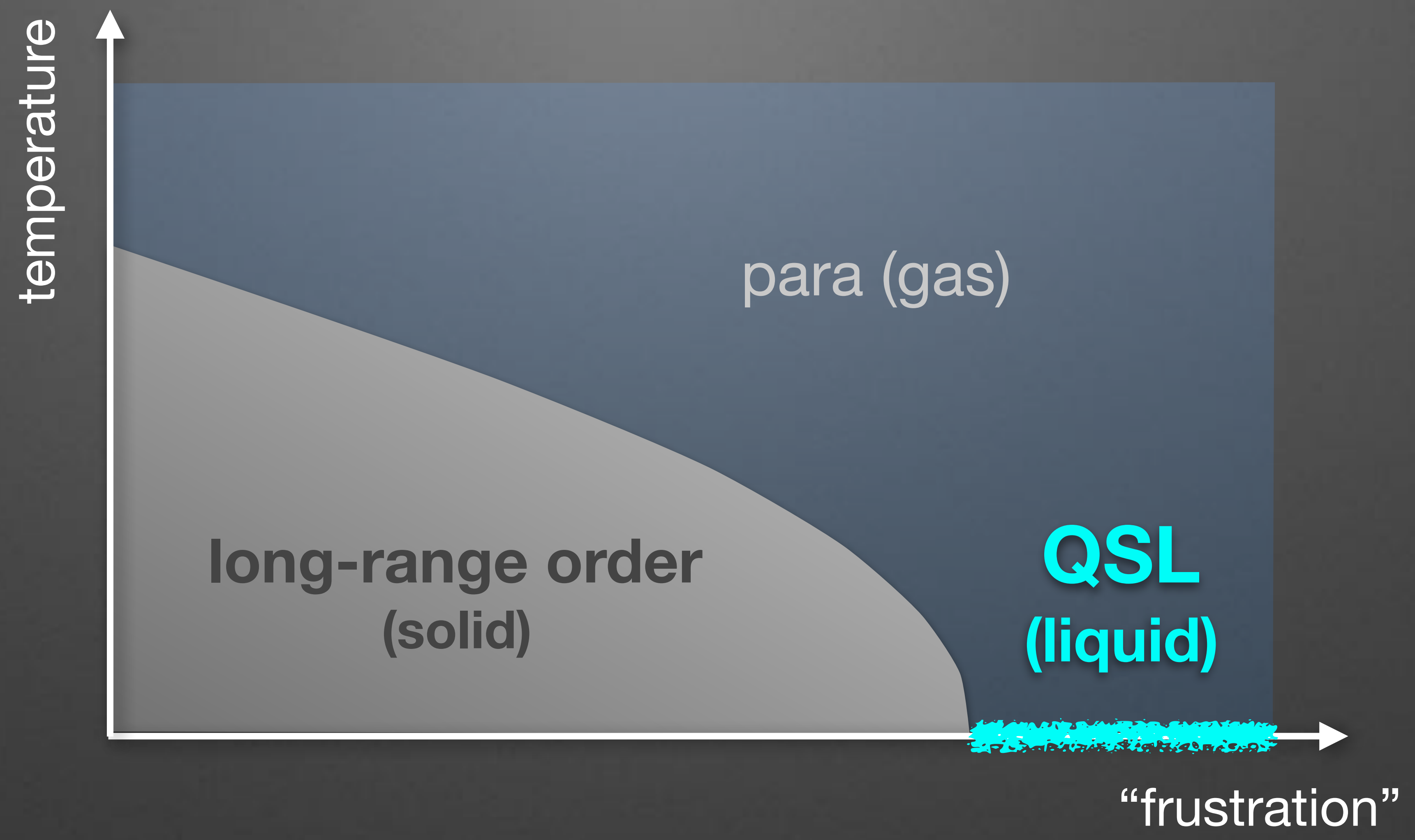
- randomness
- replica symmetry breaking

NO → **quantum spin liquid (QSL)**

- no symmetry breaking down to zero temperature
- no conventional order parameter



Quantum spin liquid



hard to identify experimentally
- no symmetry breaking down to zero temperature
- no conventional order parameter

➡ *alibi* (proof of absence) is impossible to prove

Proof positive of QSL?

- **topological order/degeneracy**

  usually well-defined only at zero temperature

  not local but global (long-range quantum entanglement)

  ➡ not easy to detect experimentally…

- **fractionalized excitations**

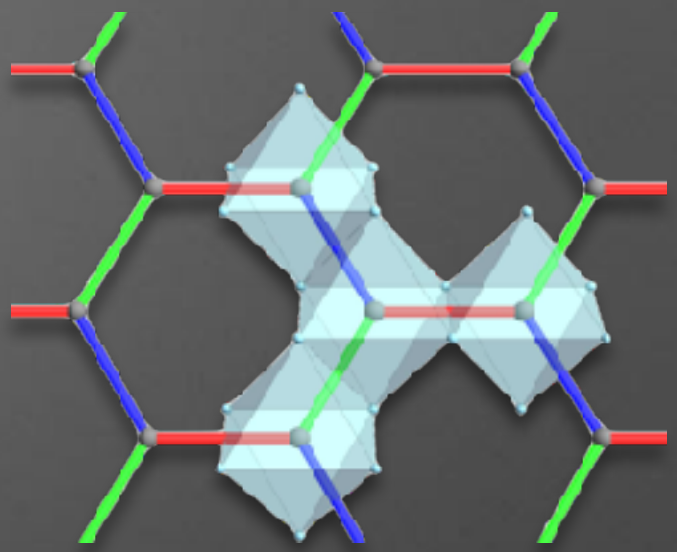
  fractionalized quasiparticles have their own energy scales

  ➡ clear fingerprints on spin dynamics and thermodynamics?

☐ How does the fractionalization show up in QSLs?

☐ How can we observe the signatures in real compounds?

Breakthrough



Kitaev model (A. Kitaev, 2006)

- **exact solution for the ground state**
  - exact quantum spin liquids
  - analytical expression for spin fractionalization

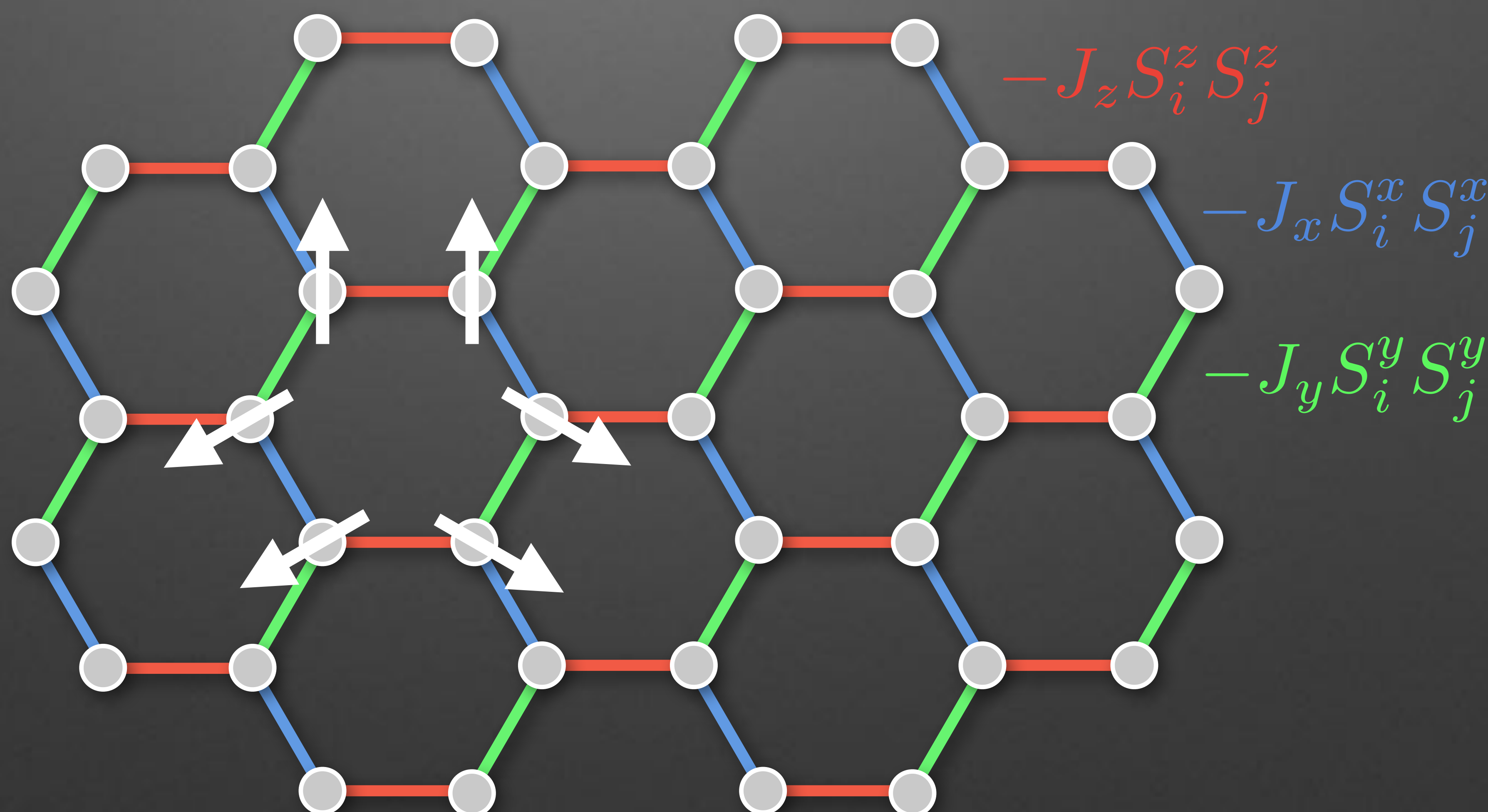
- **experimental realization in spin-orbit Mott insulators**
  (G. Jackeli and G. Khaliullin, 2009)
  - *4d* and *5d* electrons systems

explosive development in both theory and experiment
during the past decade

Kitaev model

A. Kitaev, Ann. Phys. **321**, 2 (2006)

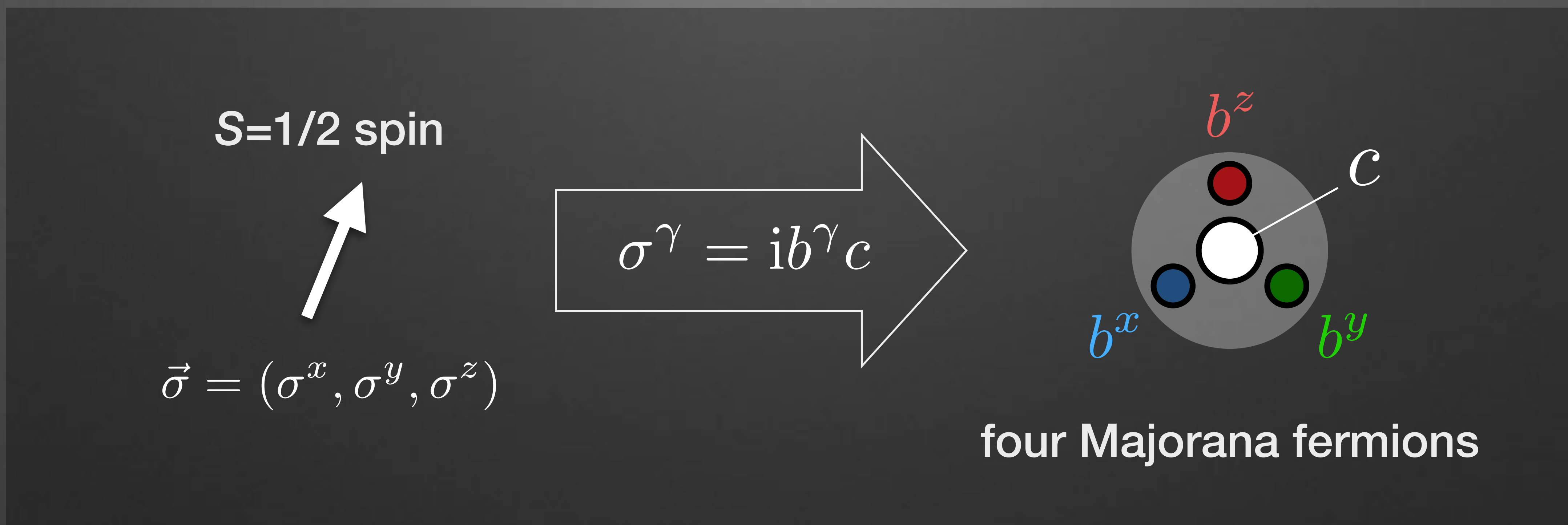
- honeycomb S=1/2 model with bond-dependent interactions



severe frustration: macroscopic degeneracy in the classical case

Majorana representation

A. Kitaev, Ann. Phys. **321**, 2 (2006)



NB1. The Majorana representation extends the Hilbert space; a projection to the original physical subspace is needed.

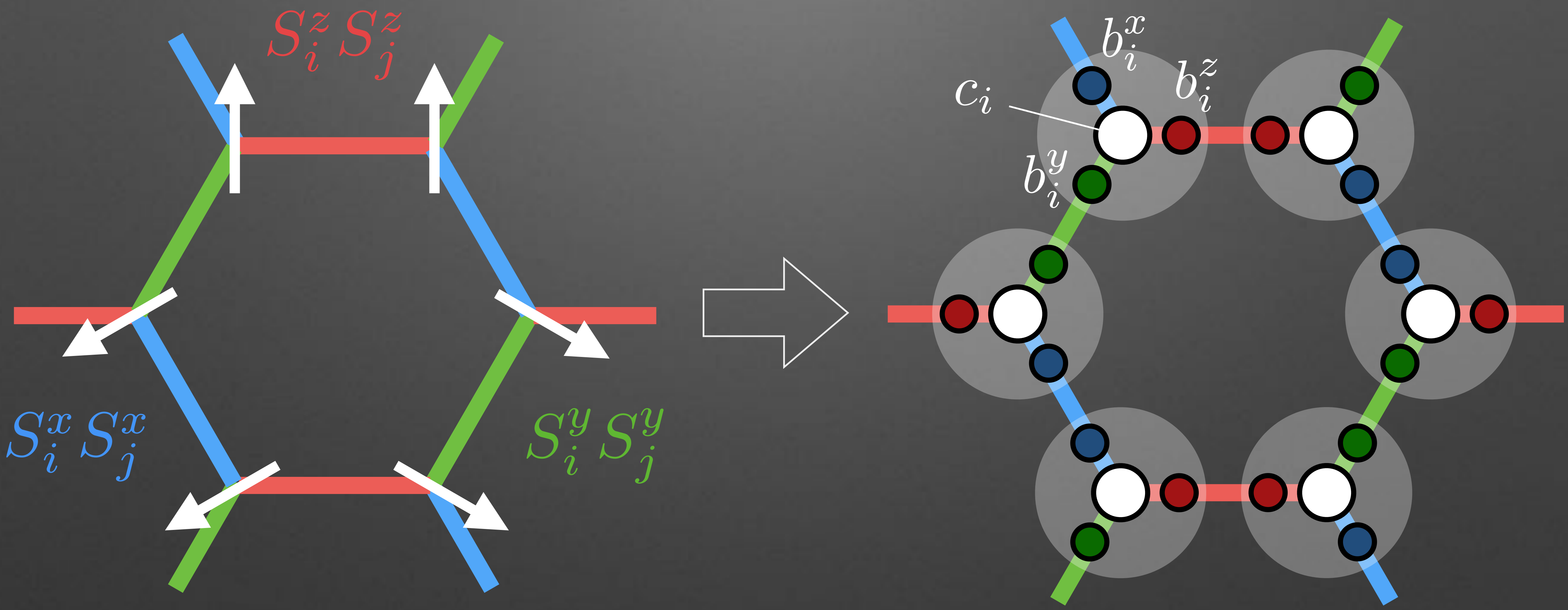
NB2. Majorana representation of spins is not unique.

- by three Majorana (Tsvelik, 1992; Shastry and Sen, 1997; Biswas et al., 2011)
- by two Majorana (Chen and Hu, 2007; Feng, Chang, and Xiang, 2007; Chen and Nussinov, 2008)

✳ We adopt the “two Majorana” representation in our numerical simulations.

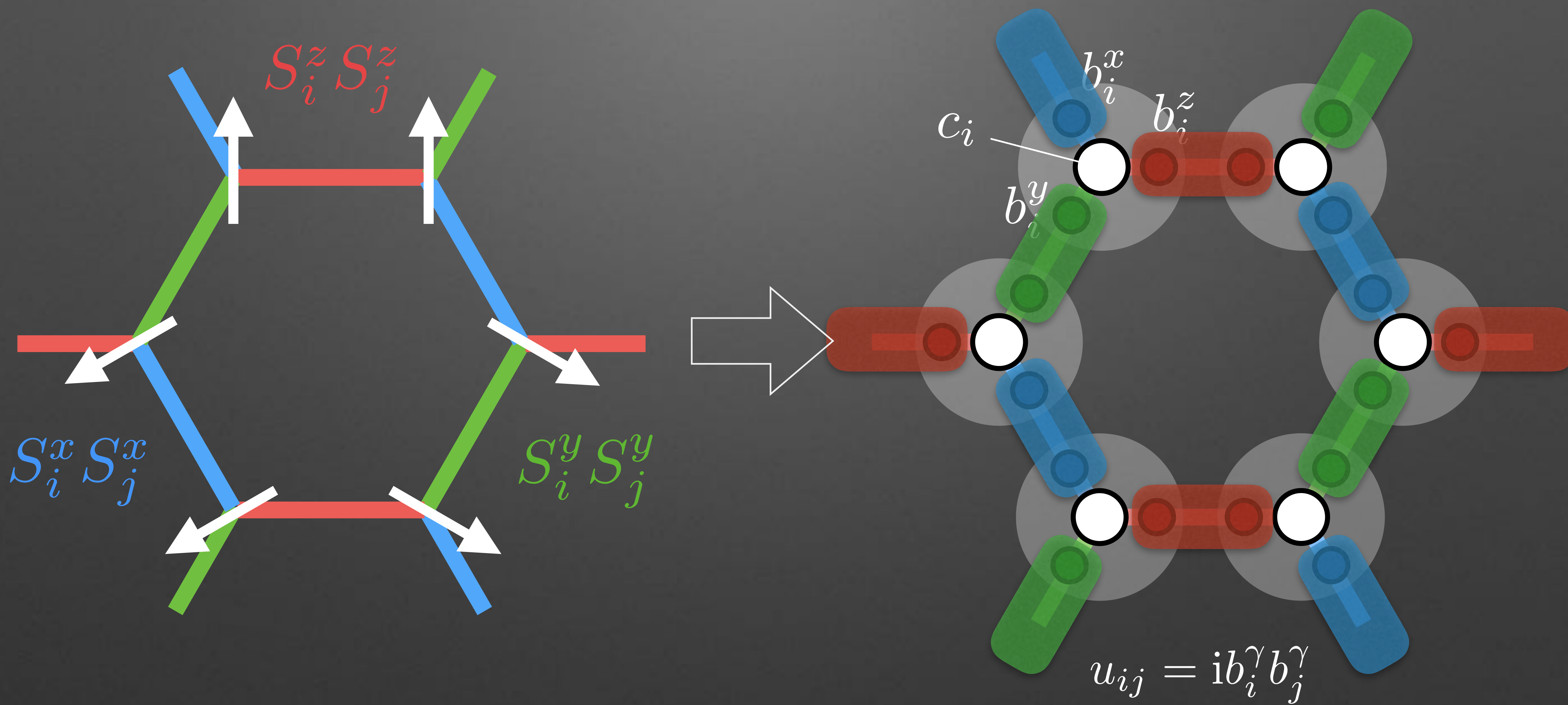
Majorana representation

A. Kitaev, Ann. Phys. **321**, 2 (2006)



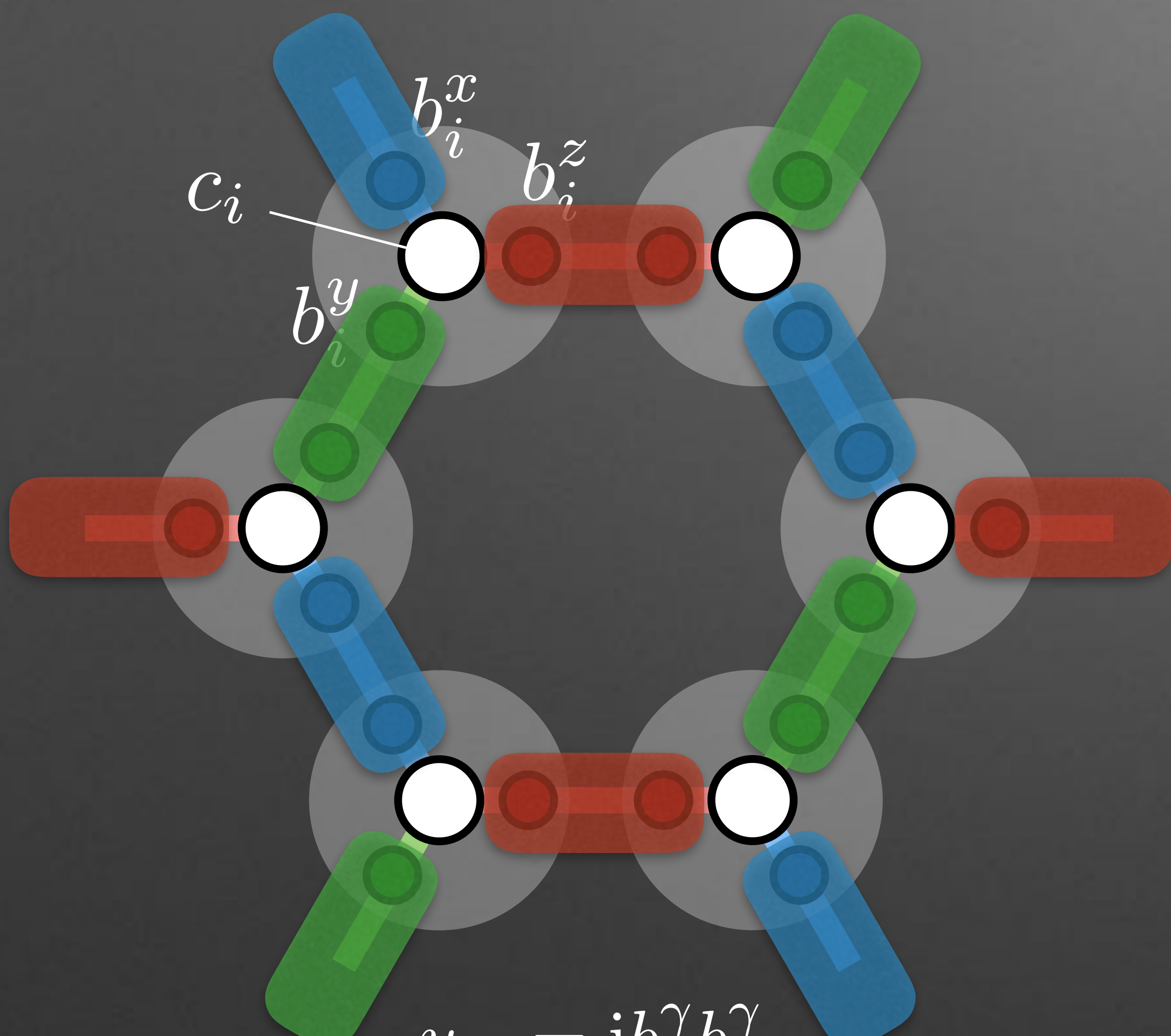
Majorana representation

A. Kitaev, Ann. Phys. **321**, 2 (2006)

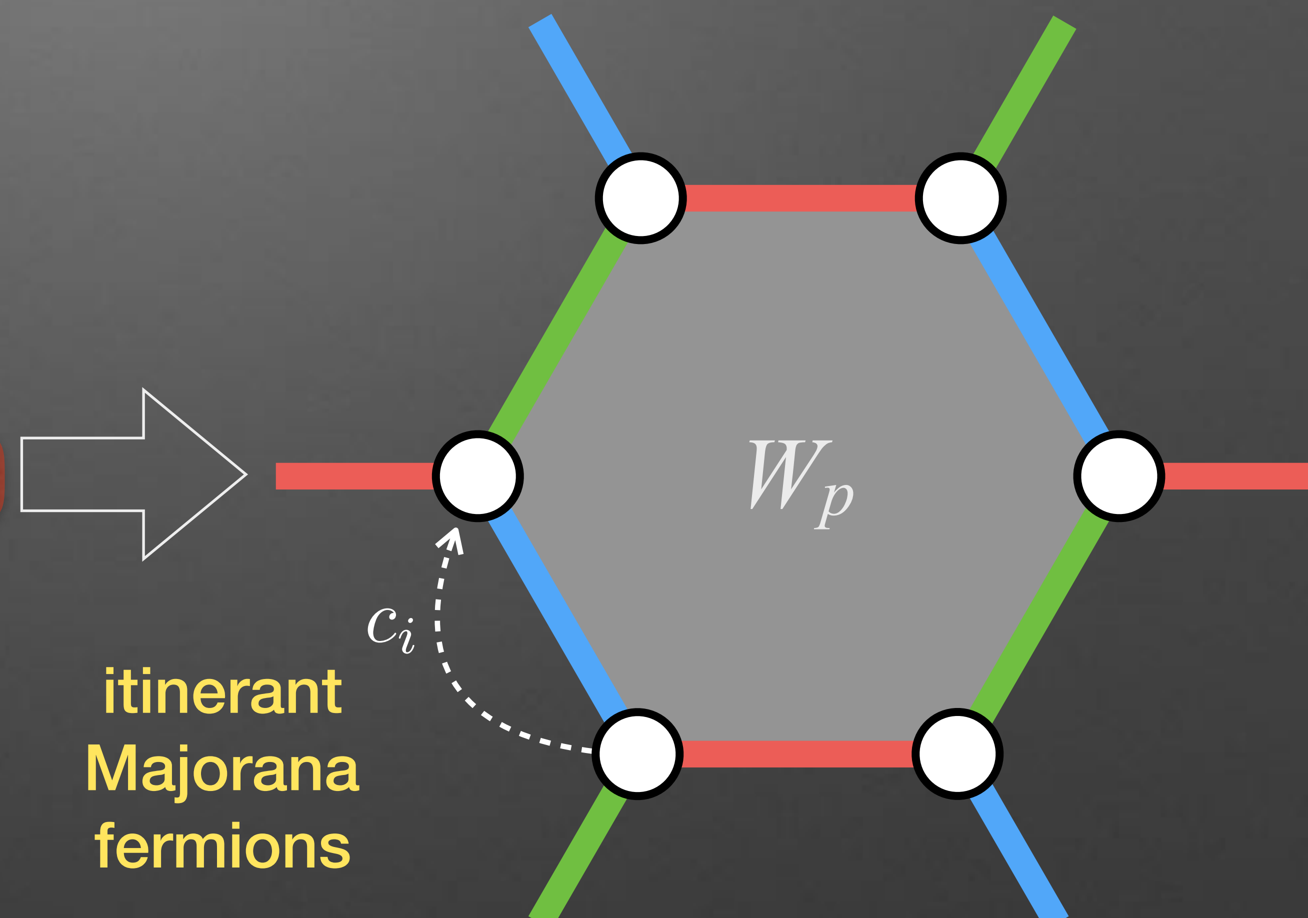


Majorana representation

A. Kitaev, Ann. Phys. **321**, 2 (2006)



$$u_{ij} = \mathrm{i} b_i^\gamma b_j^\gamma$$



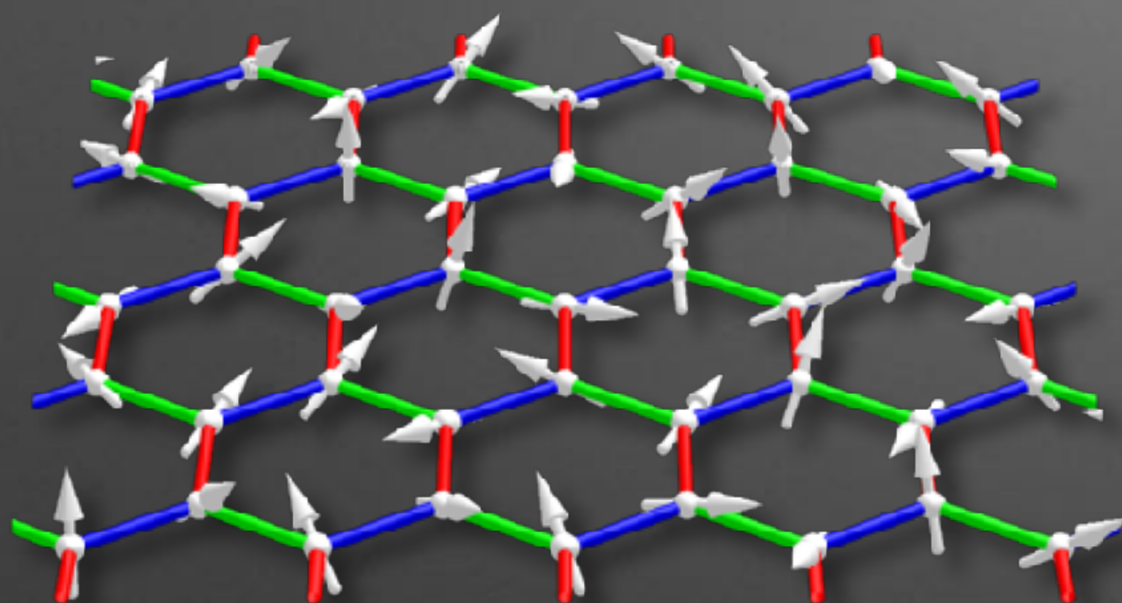
localized Z_2 fluxes $\quad W_p = \prod_{\langle i,j \rangle \in p} u_{ij}$

conserved quantity: $W_p = \pm 1$

$$[\mathcal{H}, W_p] = 0, \;\; W_p^2 = 1, \;\; [W_{p_1}, W_{p_2}] = 0$$

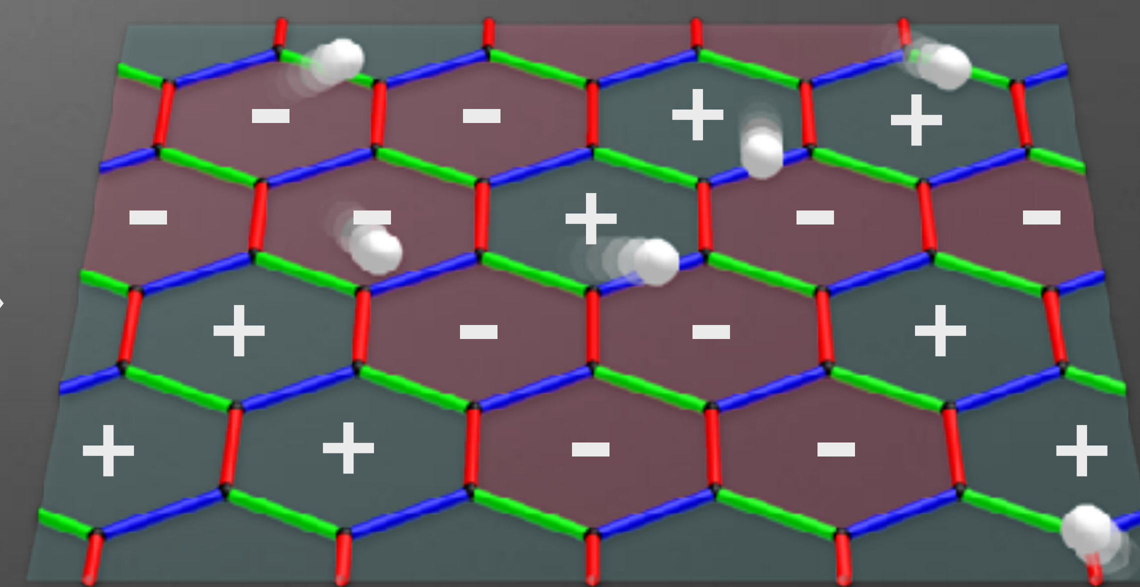
Majorana representation

A. Kitaev, Ann. Phys. **321**, 2 (2006)



S=1/2 model

quantum many-body problem



Majorana fermions moving on localized Z_2 fluxes

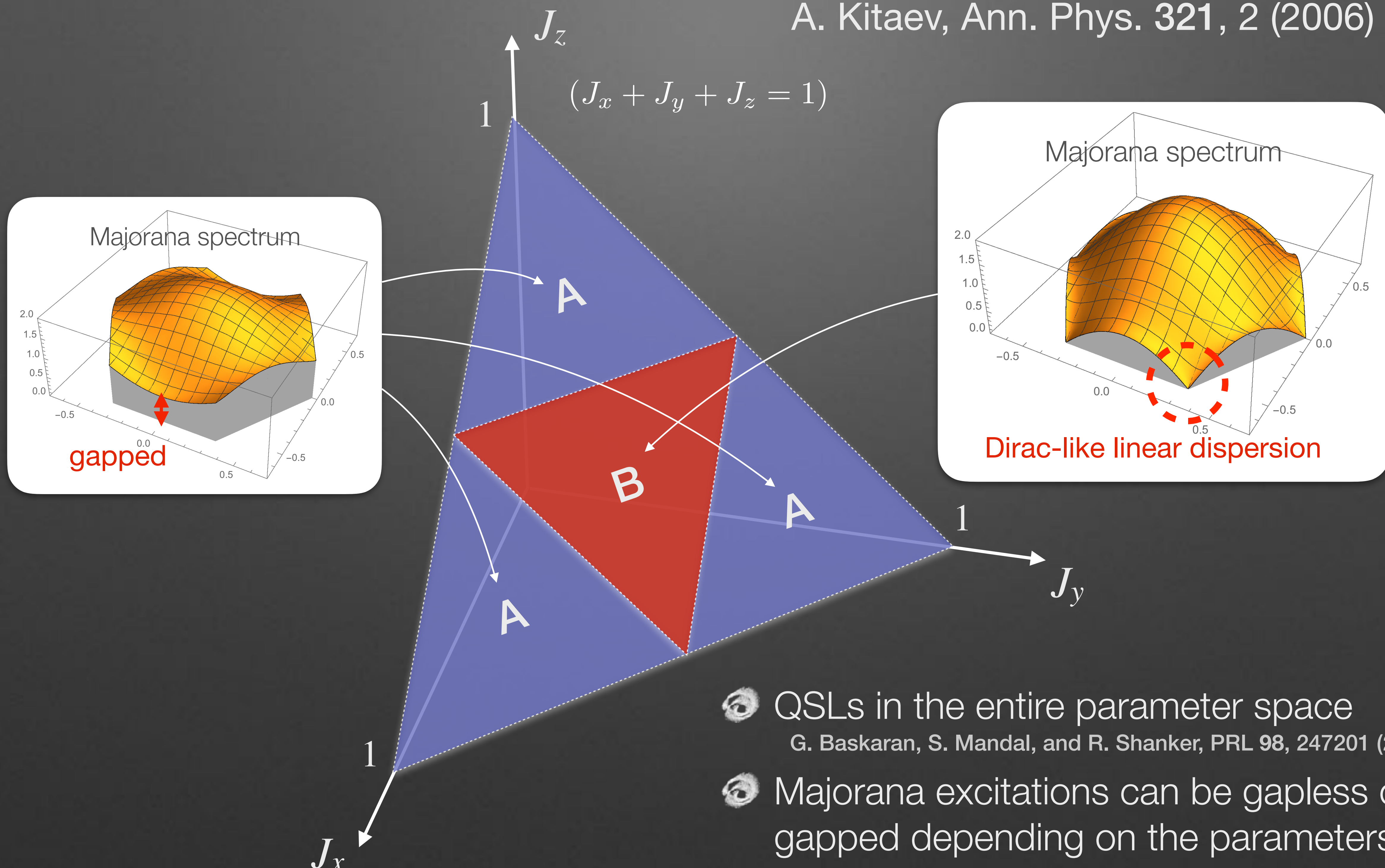
one-body problem

➡ exact ground state is available as <u>the flux free state: all W_p=+1</u>

NB. The exact solution is limited to the cases to which the Lieb theorem is applicable.

Ground state

A. Kitaev, Ann. Phys. **321**, 2 (2006)

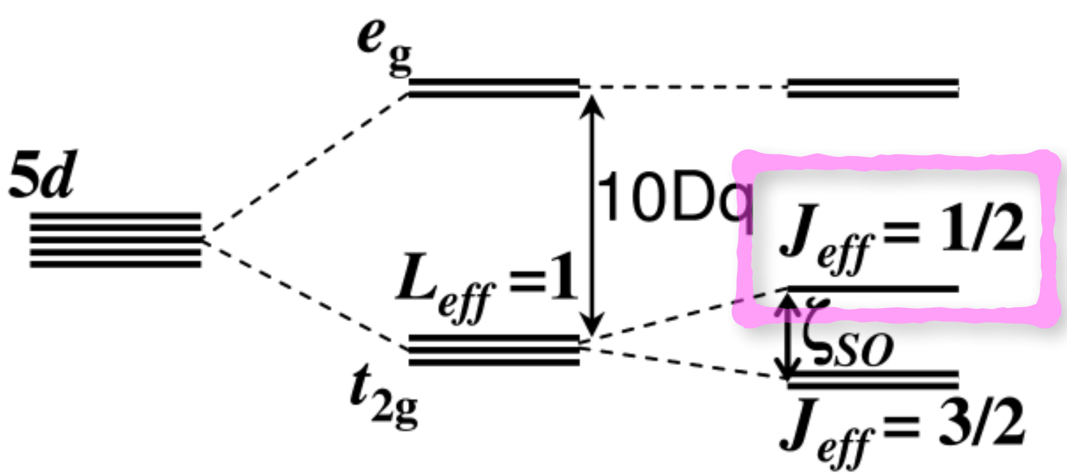


- QSLs in the entire parameter space
  G. Baskaran, S. Mandal, and R. Shanker, PRL **98**, 247201 (2007)
- Majorana excitations can be gapless or gapped depending on the parameters, while flux excitations are always gapped.

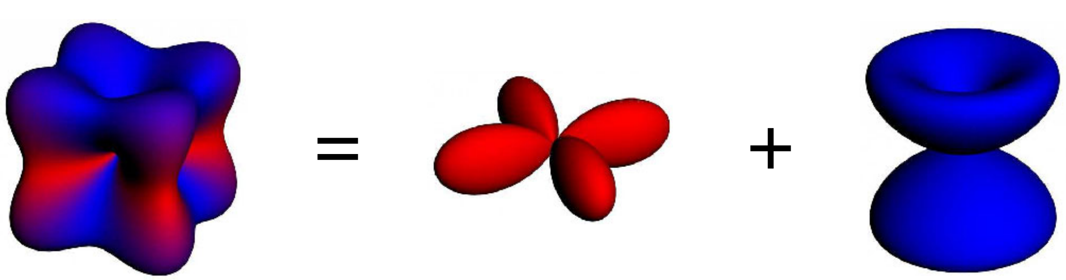
Experimental relevance

- two requisites for realizing the Kitaev-type anisotropic interactions
  G. Jackeli and G. Khaliullin, Phys. Rev. Lett. **102**, 017205 (2009)

spin-orbit Mott insulator with J_{eff}=1/2 Kramers doublet (e.g., Ir^{4+}, Ru^{3+})

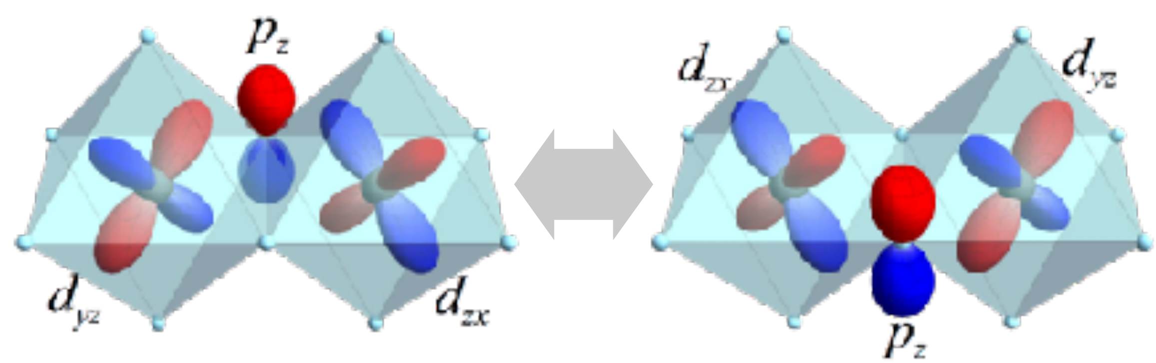


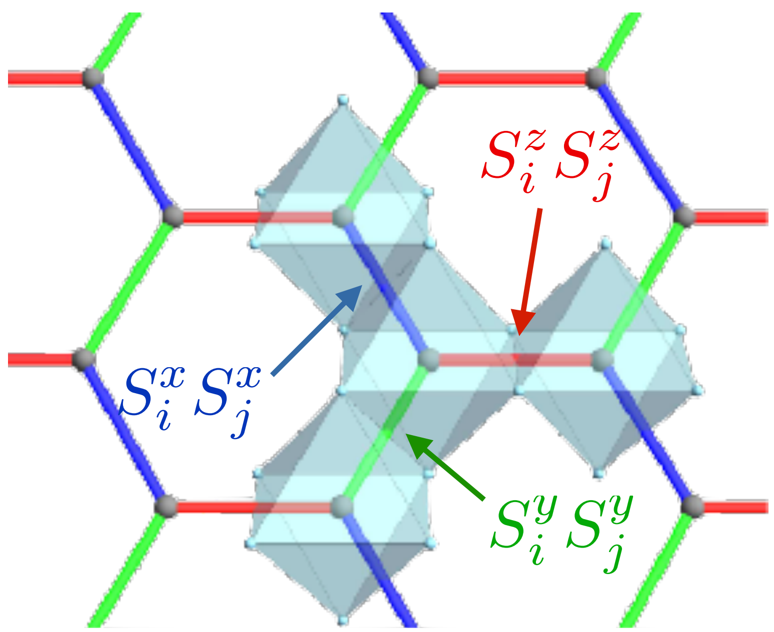
B. J. Kim *et al.*, PRL **101**, 076402 (2008)



G. Jackeli and G. Khaliullin, PRL **102**, 017205 (2009)

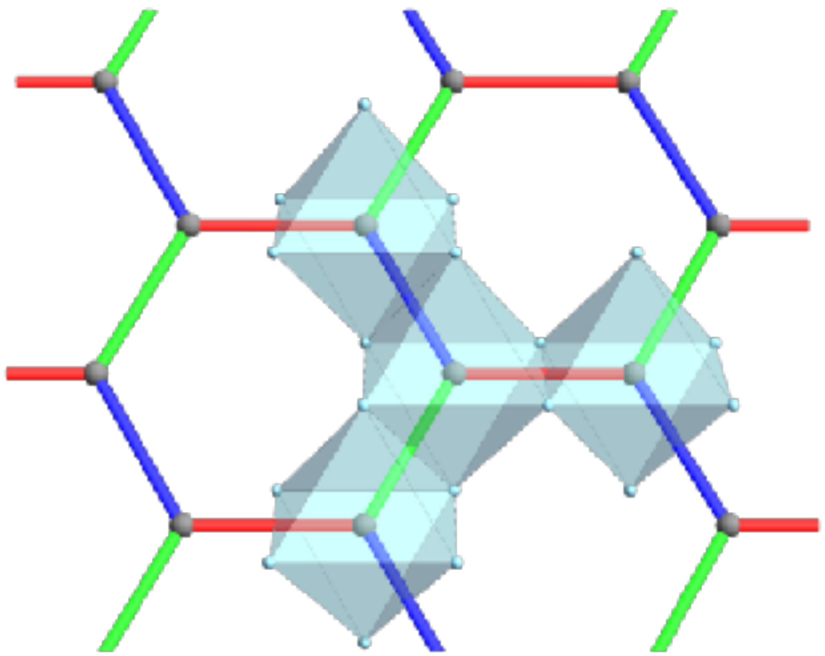
interference between *d-p-d* perturbations
(e.g., edge-sharing octahedra)





Candidate materials

2D honeycomb



Na_2IrO_3, Li_2IrO_3,...

M. J. O'Malley *et al.*, 2008
Y. Singh and P. Gegenwart, 2010
Y. Singh *et al.*, 2012, ...

α-$RuCl_3$

K. Plumb *et al.*, 2014
Y. Kubota *et al.*, 2015, ...

Li_2RhO_3

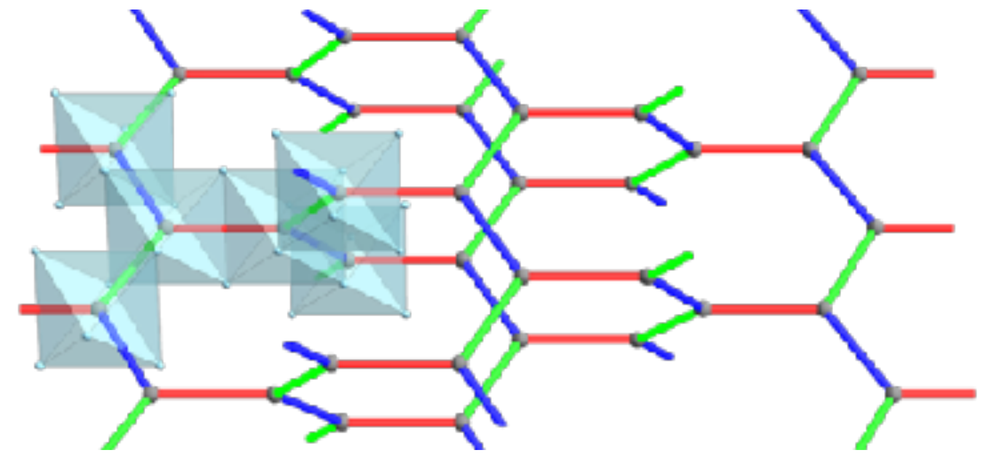
V. Todorova and M. Jansen, 2011
Y. Luo *et al.*, 2013, …

$H_3LiIr_2O_6$

K. Kitagawa *et al.*, 2018

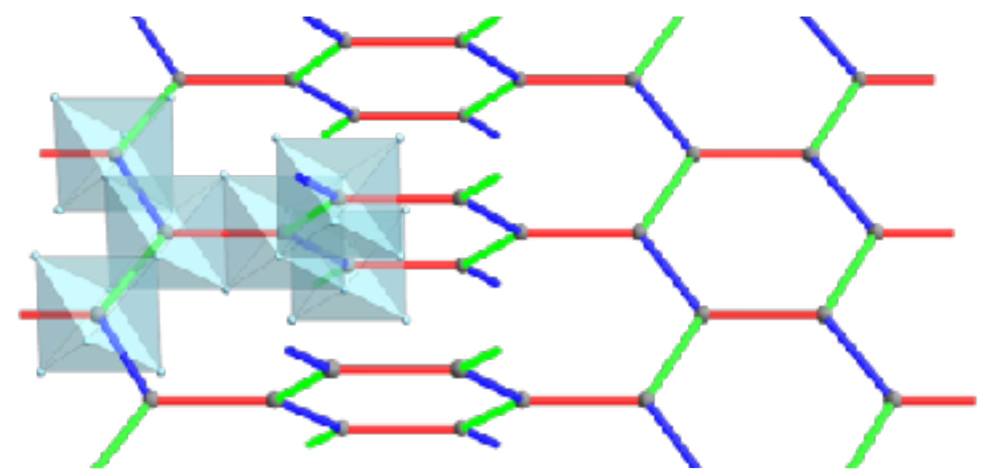
3D extensions

β-Li_2IrO_3 (hyper-honeycomb)



T. Takayama *et al.*, 2015

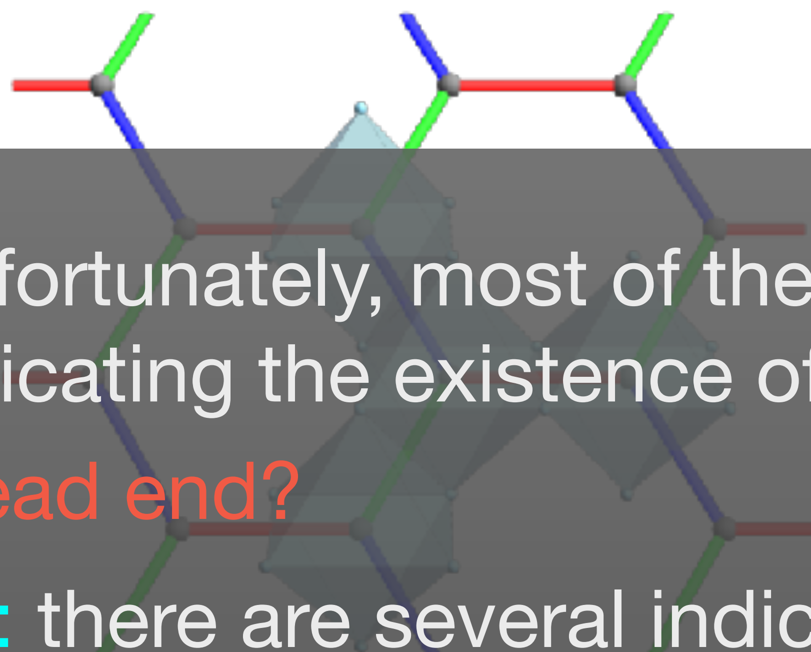
γ-Li_2IrO_3 (stripy-honeycomb)



K. A. Modic *et al.*, 2014

Candidate materials

2D honeycomb



Na_2IrO_3, Li_2IrO_3,... M. J. O'Malley *et al.*, 2008; Y. Singh and P. Gegenwart, 2010

K. Plumb *et al.*, 2014

Li_2RhO_3 V. Todorova and M. Jansen, 2011; Y. Luo *et al.*, 2013, …

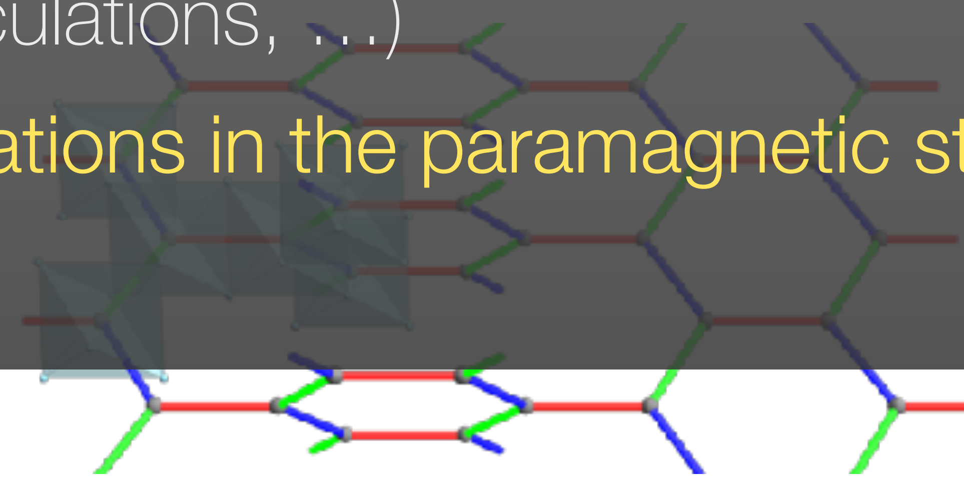
$H_3LiIr_2O_6$ K. Kitagawa *et al.*, 2018

3D extensions

β-Li_2IrO_3 (hyper-honeycomb)

T. Takayama *et al.*, 2015

γ-Li_2IrO_3 (stripy-honeycomb)



K. A. Modic *et al.*, 2014

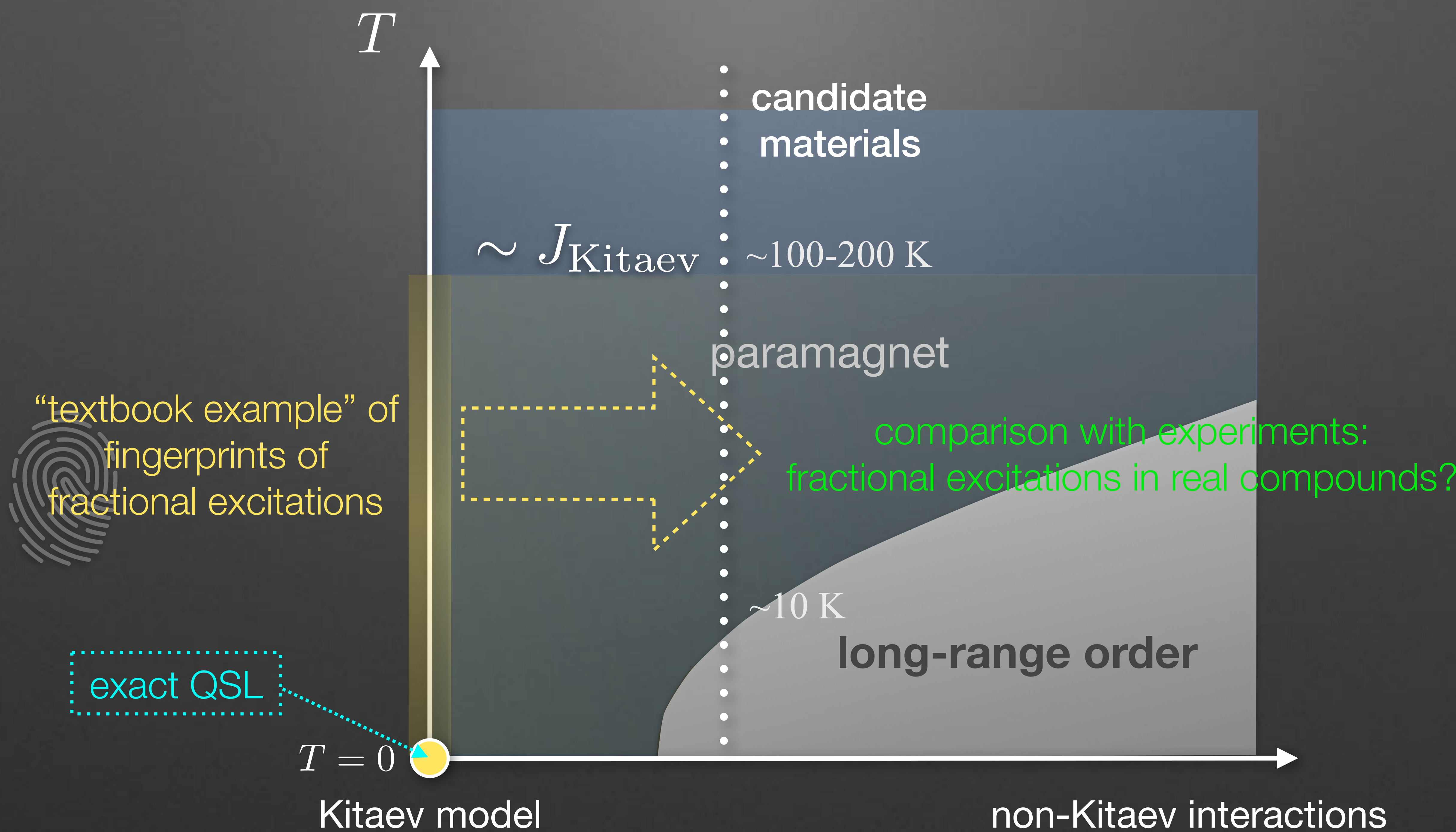
- Unfortunately, most of the candidates show AF order at low *T*, indicating the existence of non-Kitaev interactions

➡ dead end?

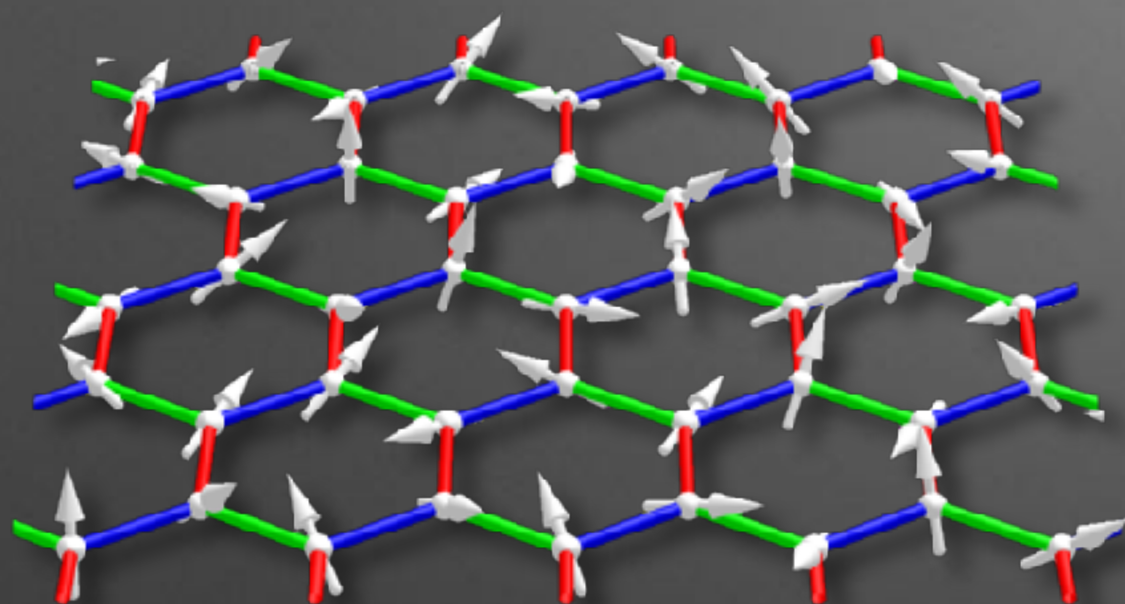
- No: there are several indications for the dominant Kitaev-type interactions (X-ray, first-principles calculations, …)

➡ signatures of the fractionalized excitations in the paramagnetic state above the critical temperature

Anticipated phase diagram



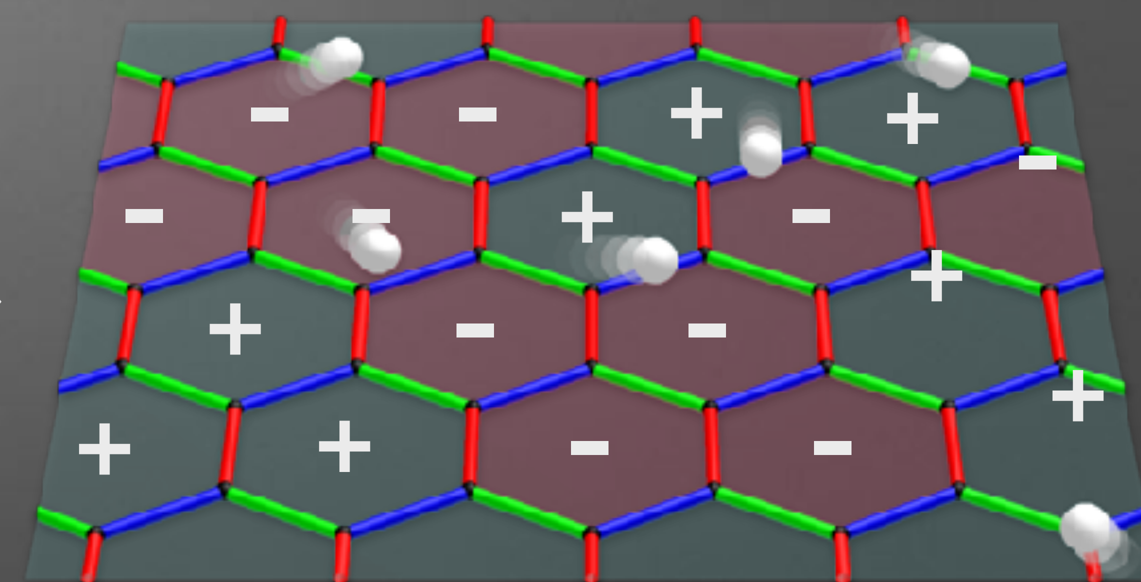
How to compute $T>0$



S=1/2 model

quantum many-body problem

- Conventional numerical techniques suffer from the negative sign problem due to severe frustration.



itinerant Majorana fermions + localized Z_2 fluxes

one-body problem

Our solution:
avoid the negative sign problem by using a Majorana representation

NB. We adopt the “two Majorana” representation via the Jordan-Wigner transformation.

Methods

- quantum Monte Carlo (QMC)
  - MC sampling on the gauge fields + exact diagonalization
    J. Nasu, M. Udagawa, and YM, PRL **113**, 197205 (2014)
  - MC sampling on the gauge fields + Green function based kernel polynomial method
    P. A. Mishchenko, Y. Kato, and YM, PRB **96**, 125124 (2017)
- QMC + continuous-time QMC
  J. Yoshitake, J. Nasu, and YM, PRL **117**, 157203 (2016)
  J. Yoshitake, J. Nasu, and YM, PRB **96**, 064433 (2017)

✓ **free from biased approximations**: numerically exact within the statistical errors

NB. We employed the maximum entropy method for analytical continuation of the dynamical quantities.

✓ **free from finite-size effects**: applicable to large enough clusters up to ~10^3 sites

We have computed …

- ✓ specific heat and entropy ➡ *thermal fractionalization*
- ✓ magnetic susceptibility
- ✓ equal-time spin-spin correlation
- ✓ NMR relaxation rate $1/T_1$
- ✓ dynamical spin structure factor $S(\mathbf{q},\omega)$

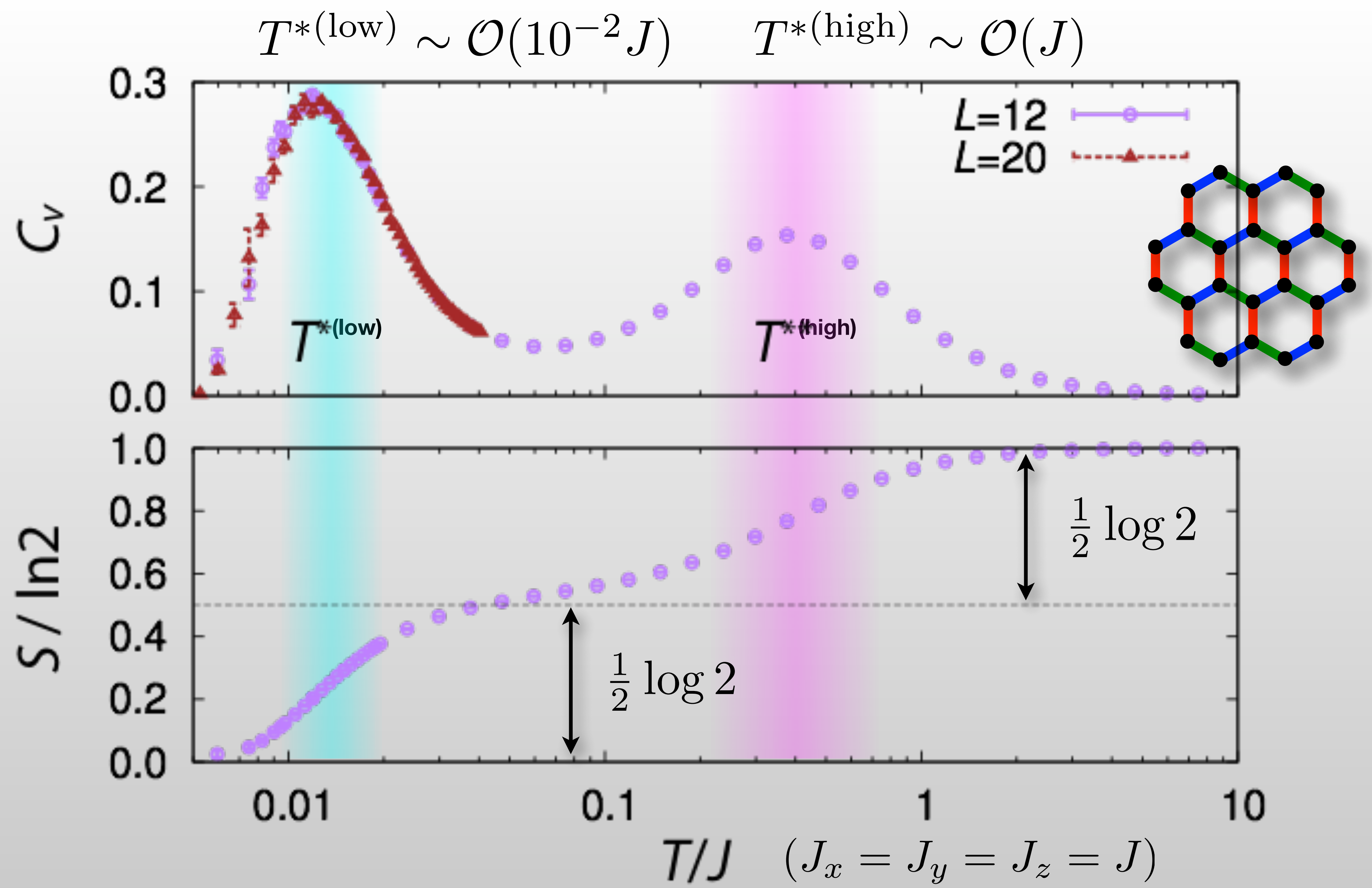
} *dichotomy between static and dynamical spin correlations*

- ✓ magnetic Raman scattering intensity
- ✓ thermal conductivity κ^{xx} and κ^{xy}

} *itinerant fermionic excitations*

All the quantities exhibit peculiar *T* and ω dependences
➡ smoking guns for fractional spin excitations !

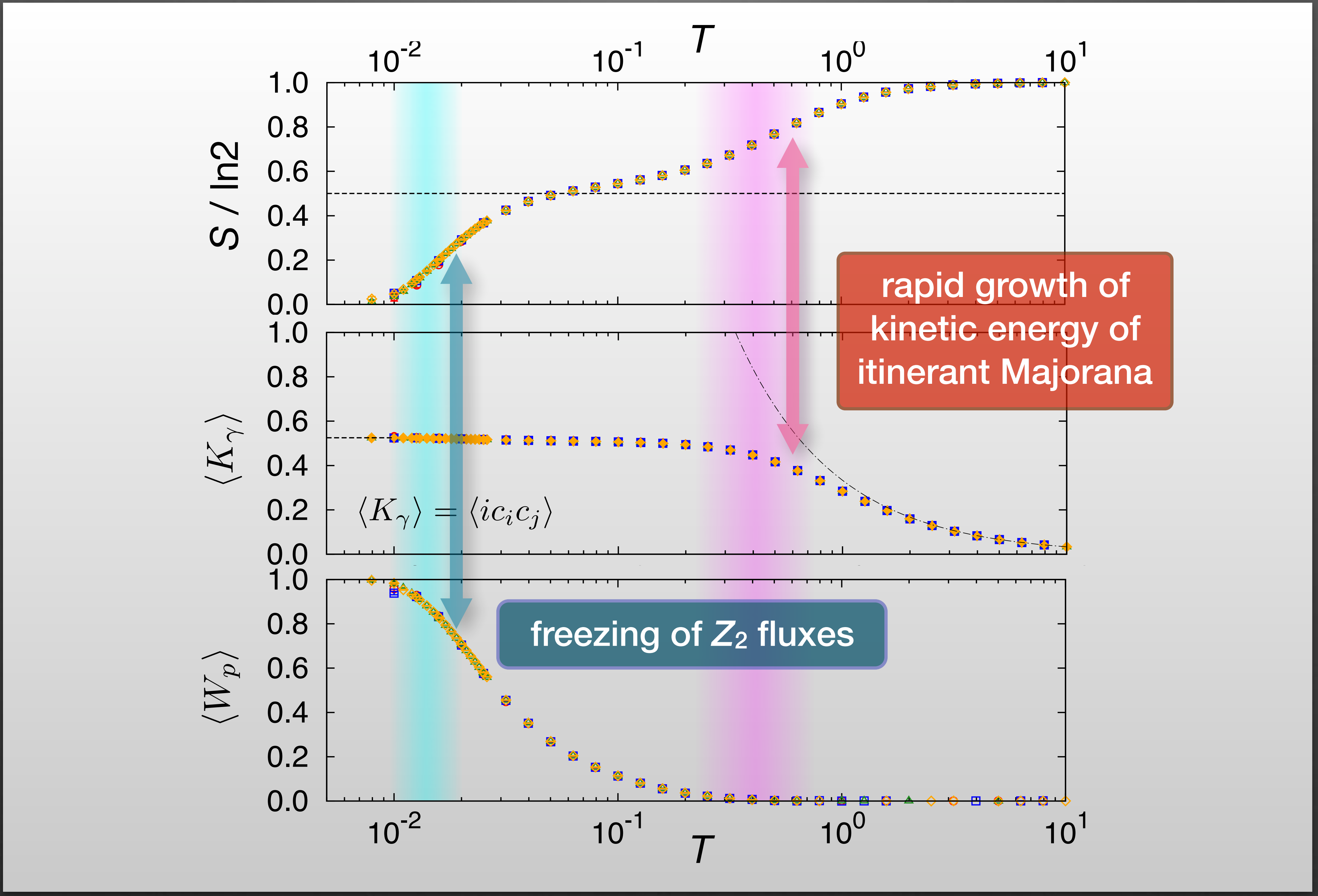
Specific heat and entropy



two crossovers: two-step release of the spin entropy

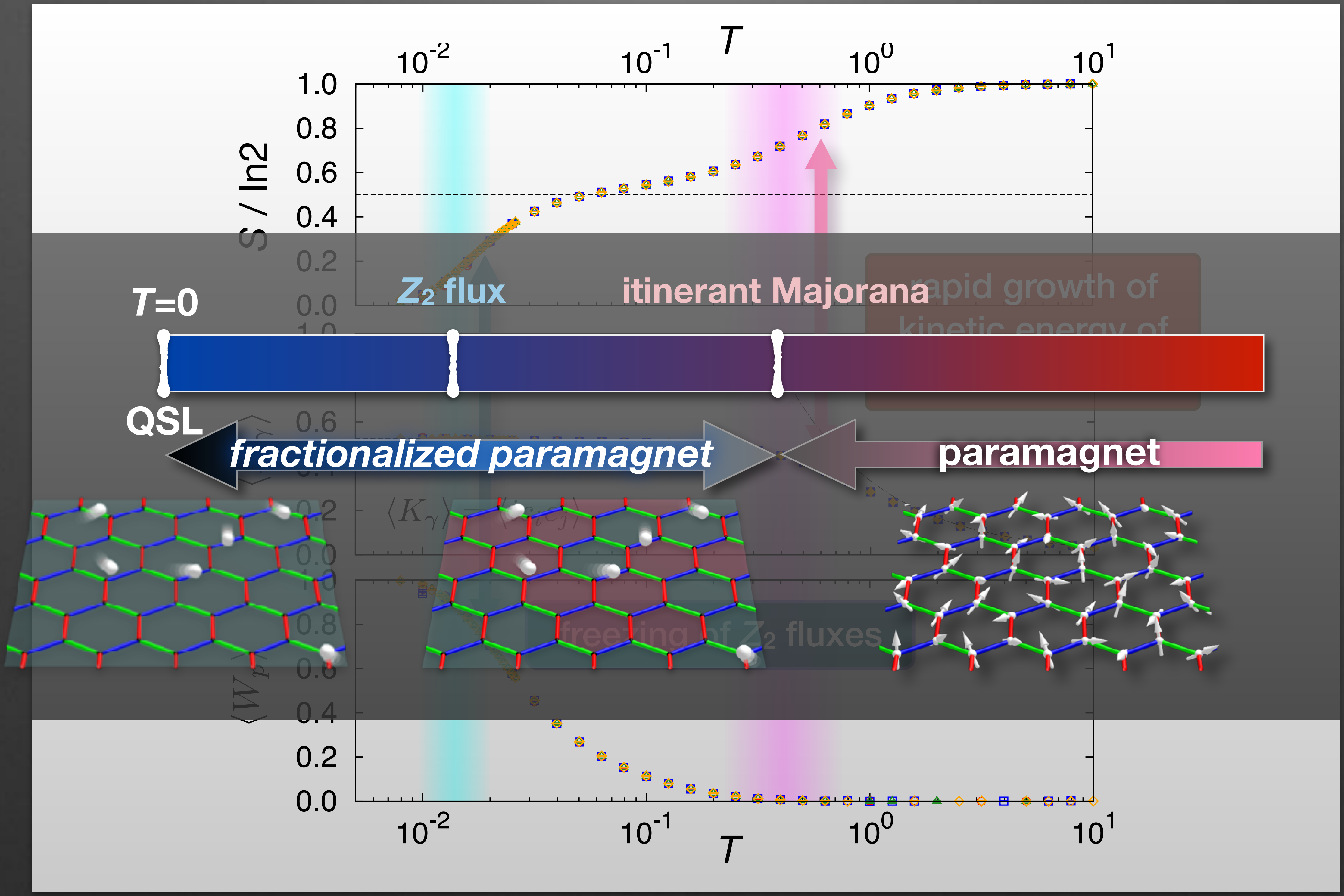
J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. B **92**, 115122 (2015)

Thermal fractionalization



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

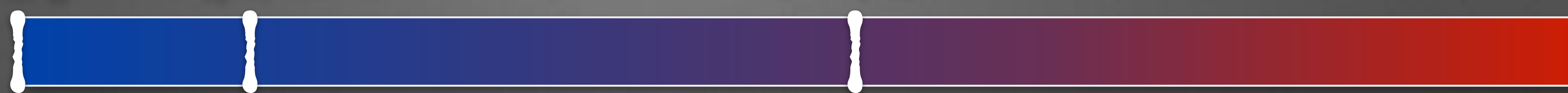
Thermal fractionalization

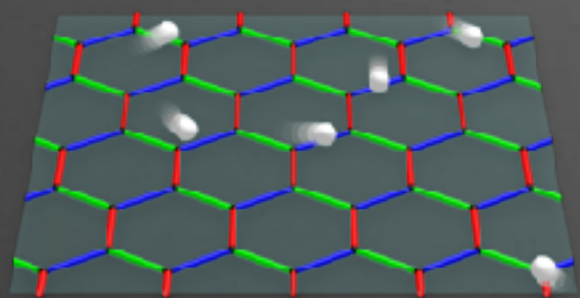


J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. B 92, 115122 (2015)

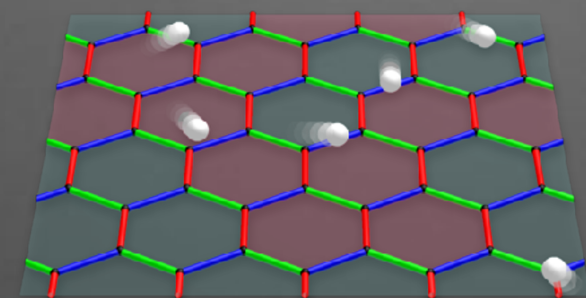
Thermal fractionalization

$T = 0$ $T \lesssim J/100$ $T \sim 0.4J$

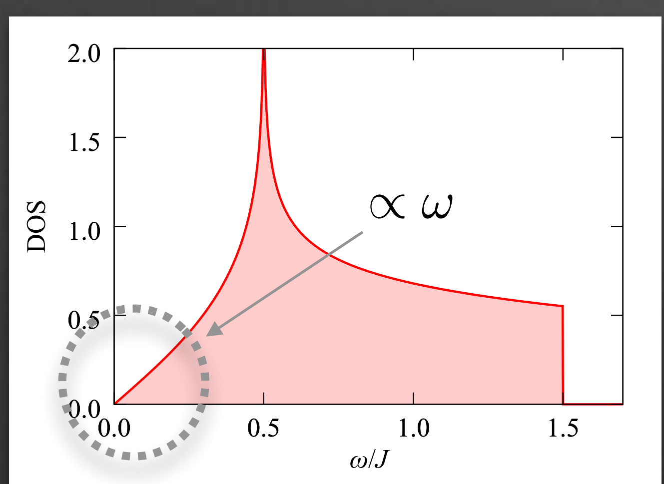




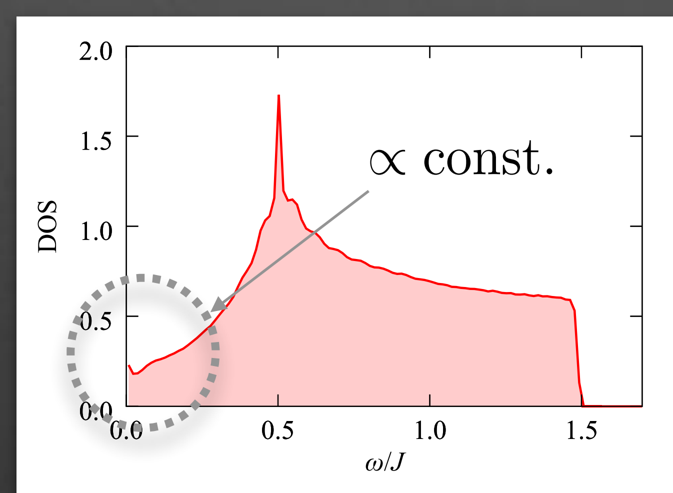
Majorana w/ no flux



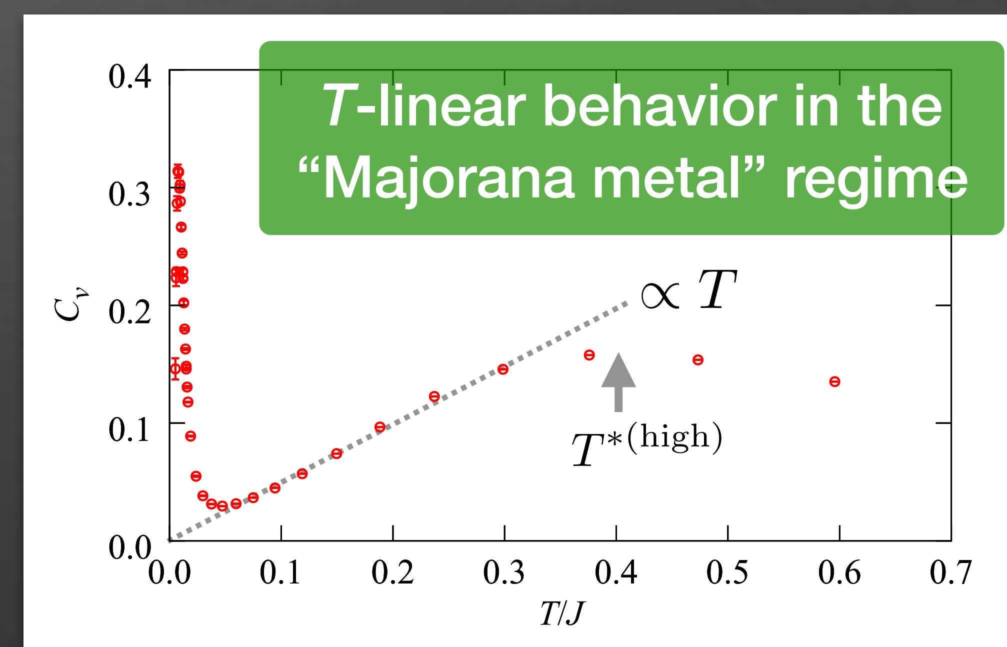
Majorana w/ disordered fluxes



Dirac-type semimetal



"Majorana metal"



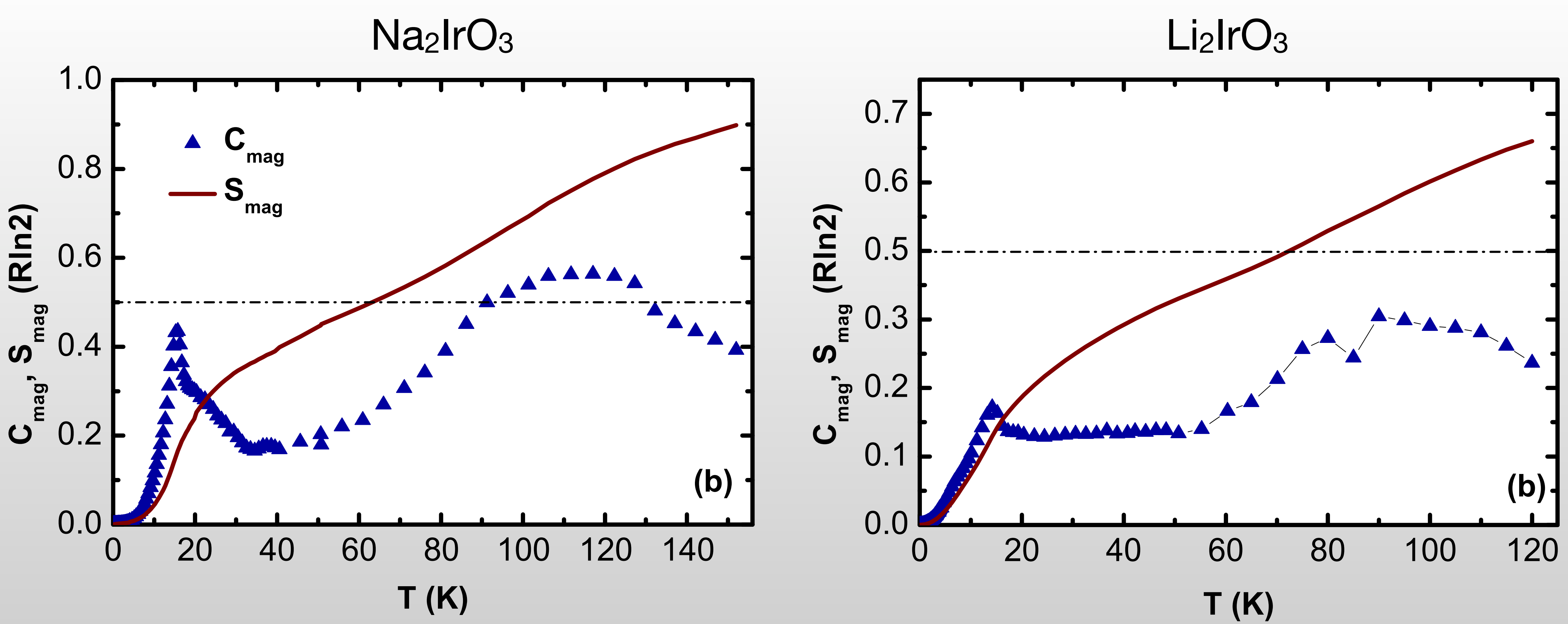
fermionic itinerant nature will be observable

NB. Anisotropy in J_x, J_y, J_z widens the T range of the "Majorana metal".

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

Specific heat and entropy: exp.

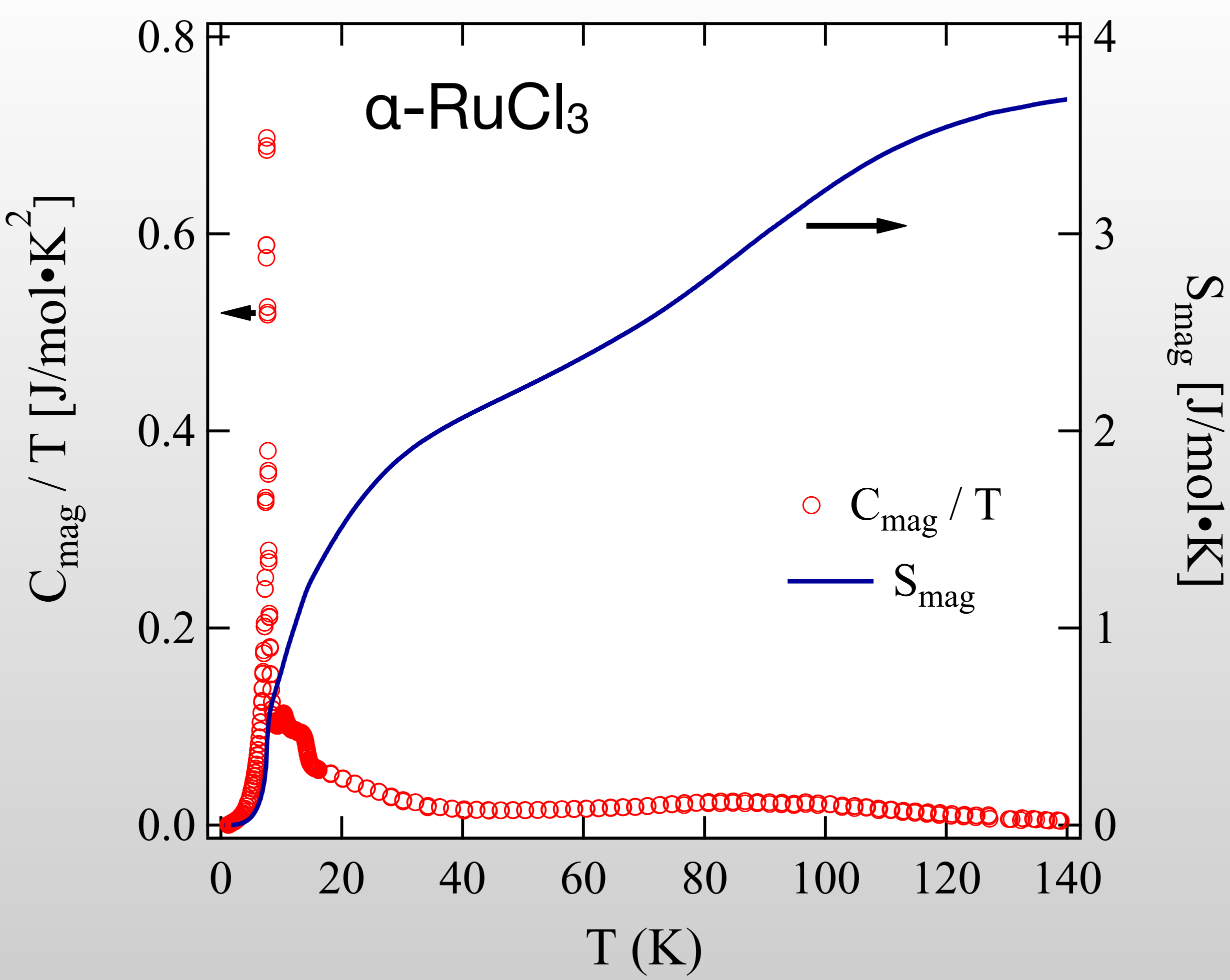
- broad hump at high *T* in the specific heat, and step-like shoulder in the entropy around (1/2)log2



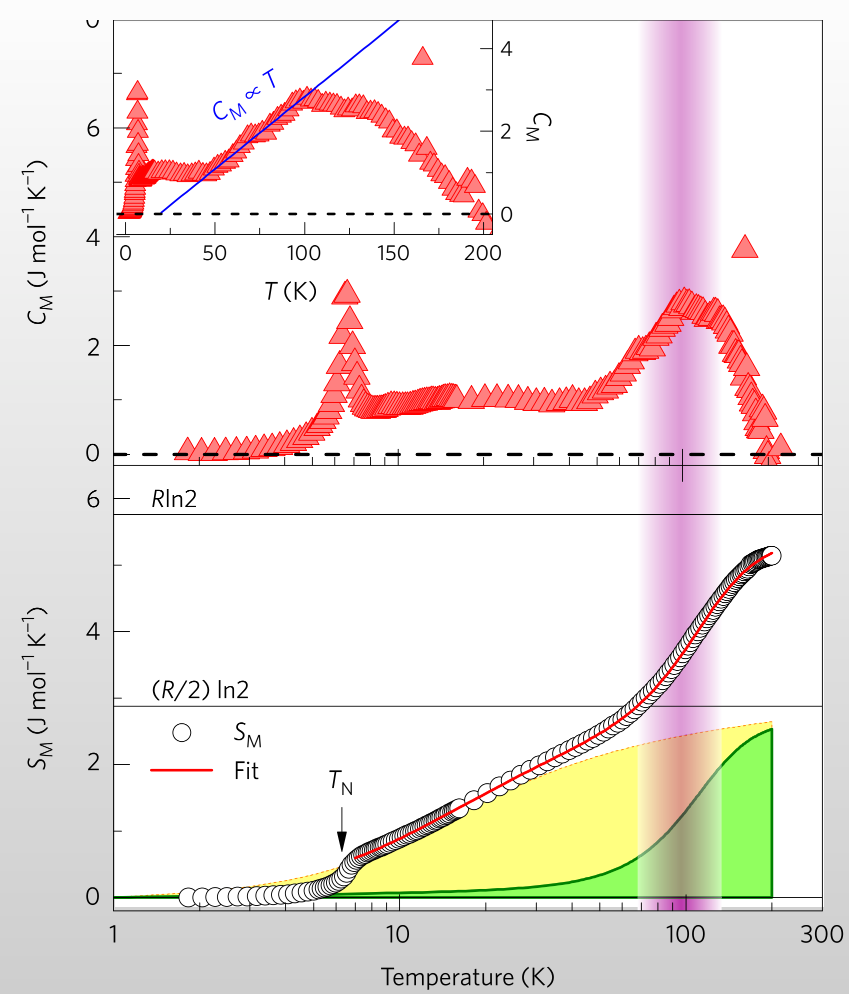
K. Mehlawat, A. Thamizhavel, and Y. Singh, PRB **95**, 144406 (2017)

Specific heat and entropy: exp.

- broad hump at high *T* in the specific heat, step-like shoulder in the entropy around (1/2)log2, and *T*-linear in mid *T*?



Y. Kubota *et al.*, Phys. Rev. B **91**, 094422 (2015)

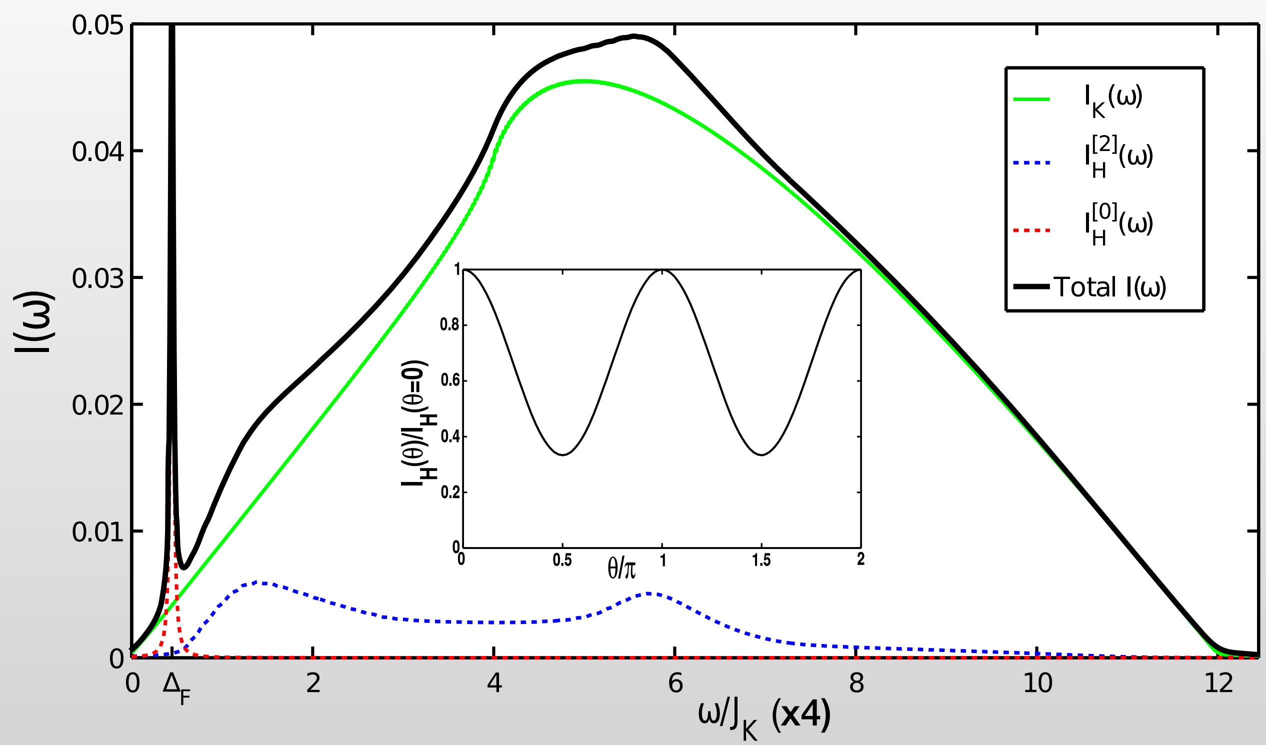


S.-H. Do *et al.*, Nat. Phys. **13**, 1709 (2017)

Raman scattering at low *T*

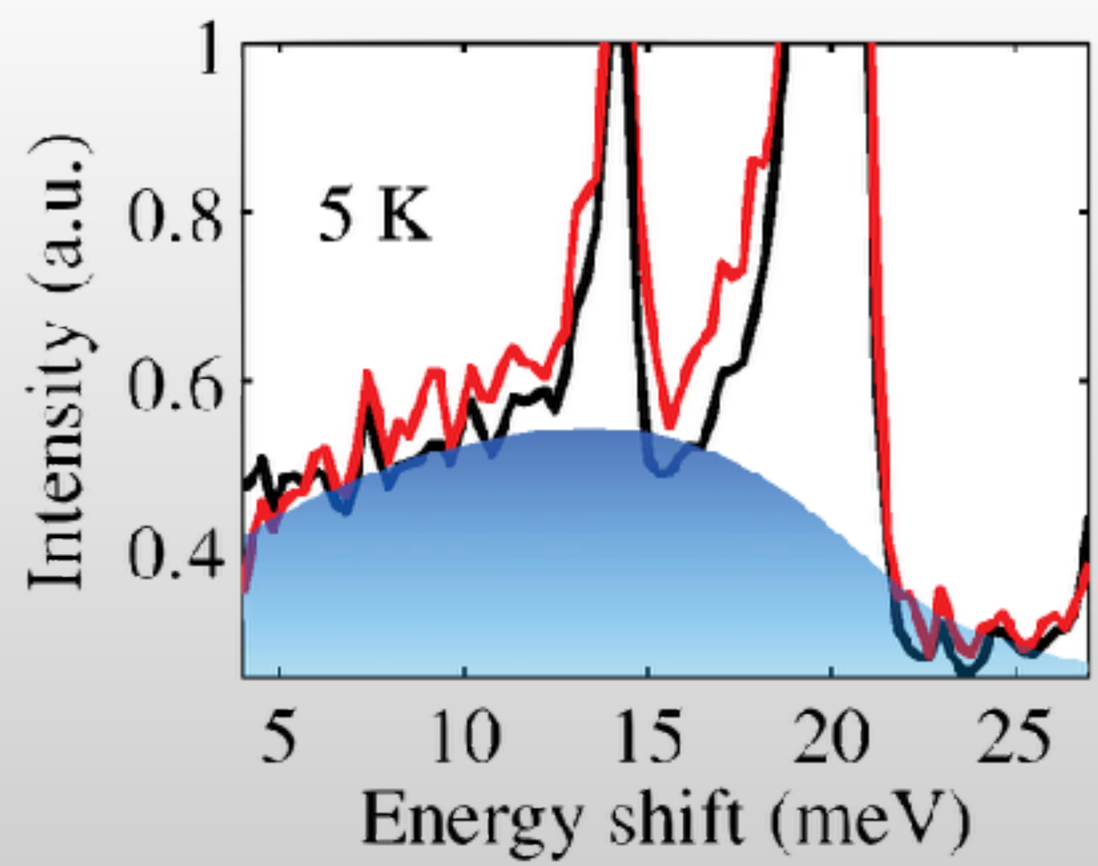
- anomalous incoherent component up to ~3J_{Kitaev}: signature of fractionalized excitations in the Kitaev QSL?

theory for the Kitaev model at *T*=0



J. Knolle *et al.*, PRL **113**, 187201 (2014)

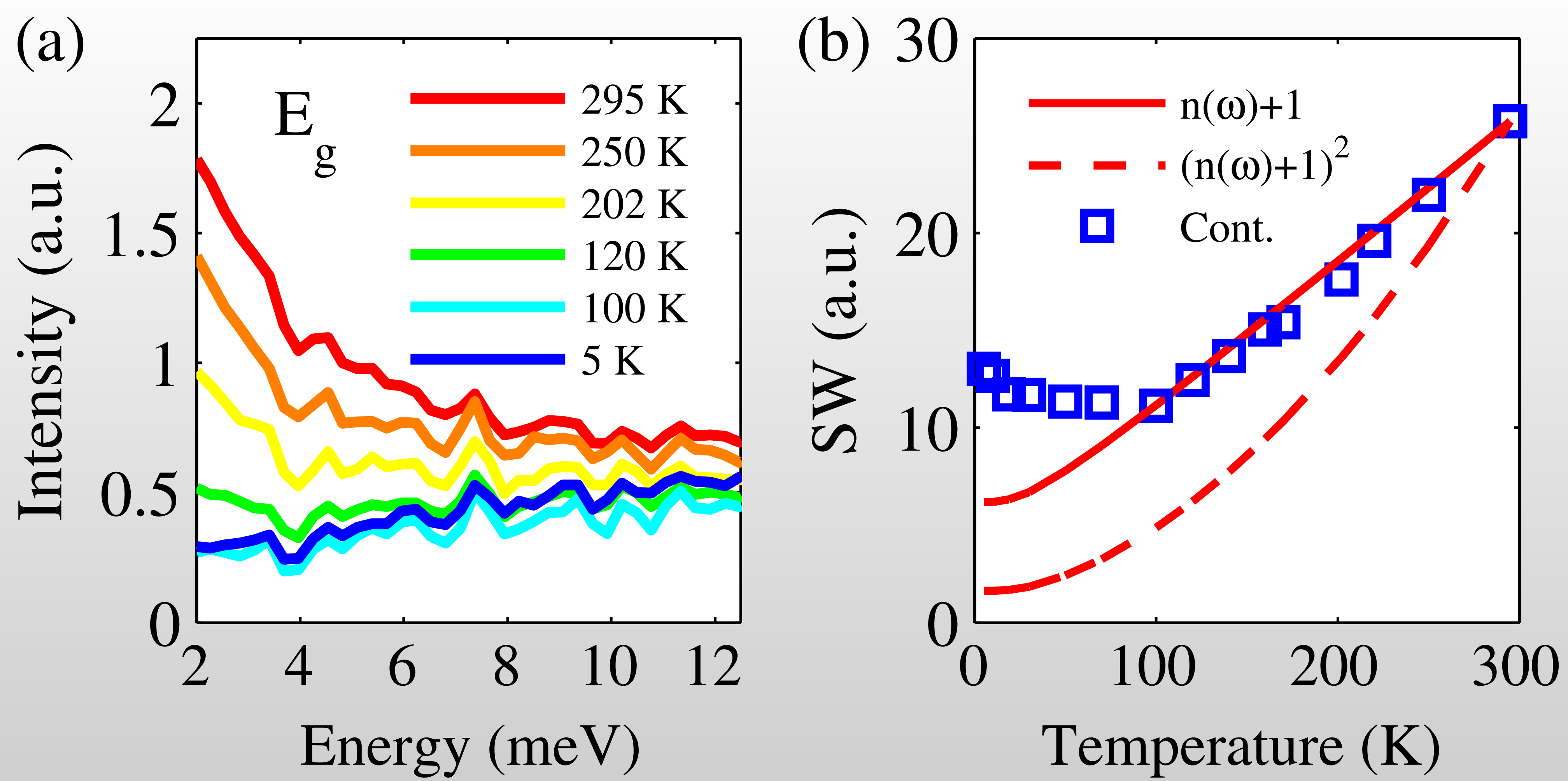
experiment for α-$RuCl_3$



L. J. Sandilands *et al.*, PRL **114**, 147201 (2015)

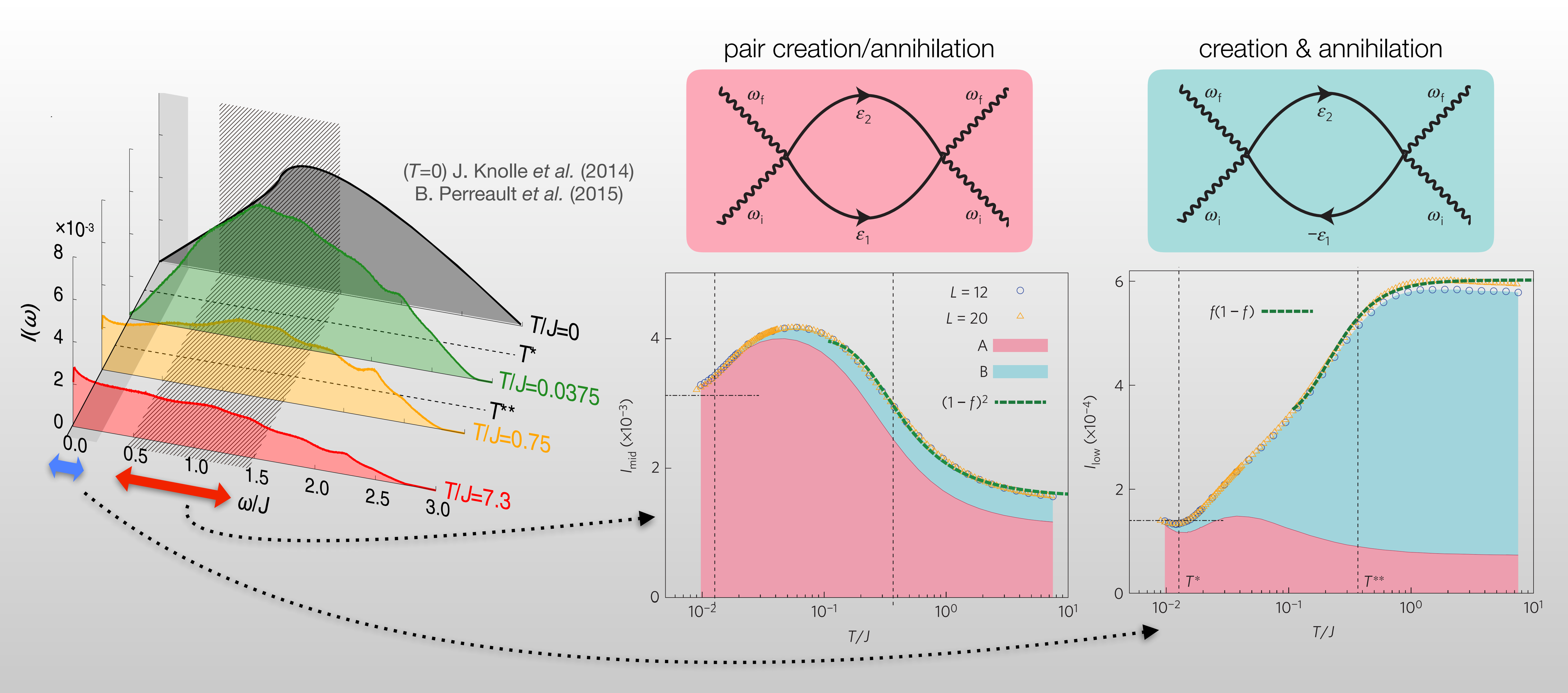
Raman scattering: experiment

- anomalous incoherent component, whose *T* dependence is not explained by bosonic contributions



L. J. Sandilands *et al.*, PRL **114**, 147201 (2015)

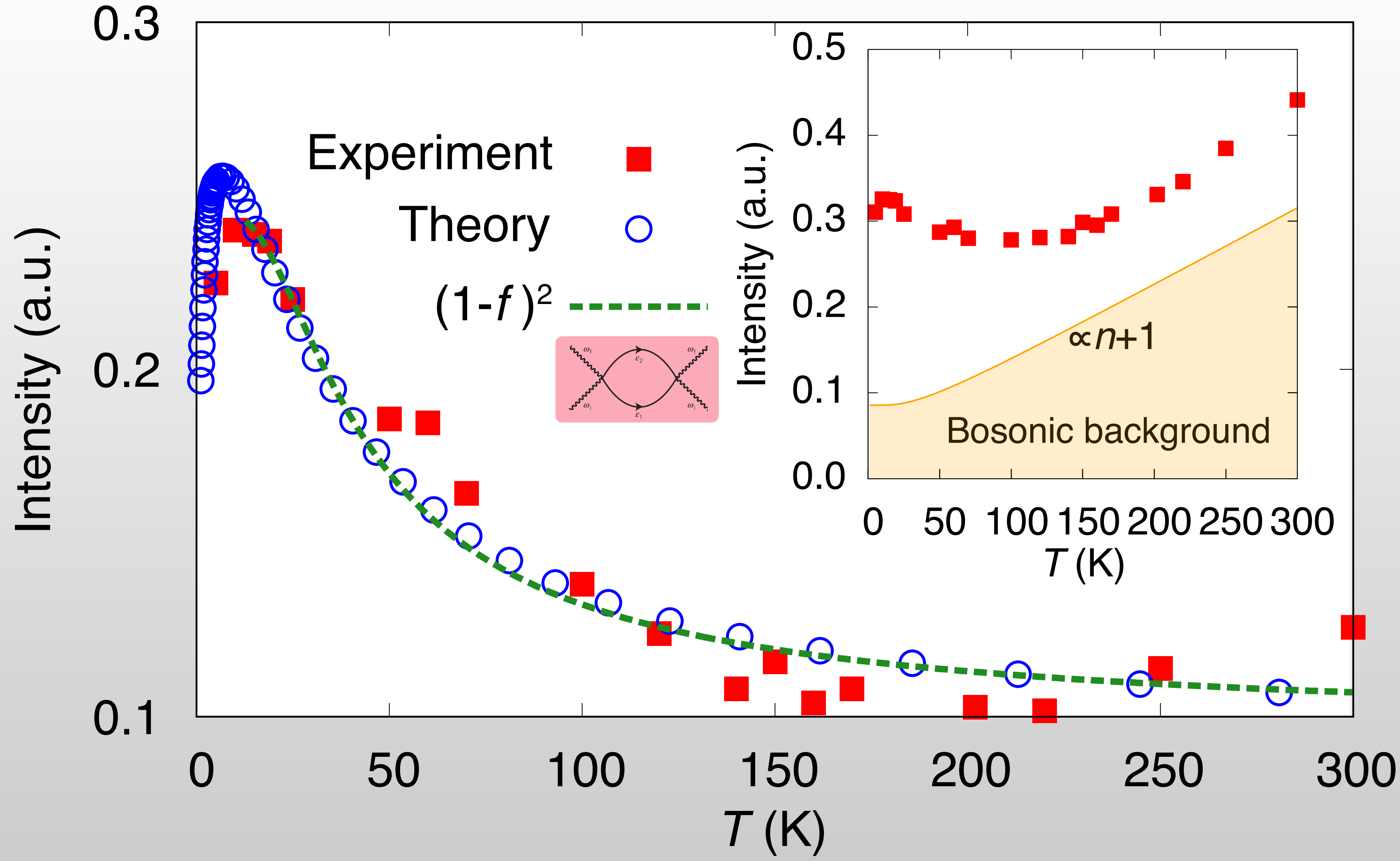
Raman scattering: theory



mid energy: dominated by pair creation/annihilation ~$(1-f)^2$
low energy: dominated by creation & annihilation ~$f(1-f)$
➡ **Both reflect fermionic excitations of Majonaras**

J. Nasu, J. Knolle, D. L. Kovrizhin, Y. Motome, and R. Moessner, Nat. Phys. 12, 912 (2016)

Raman scattering: comparison

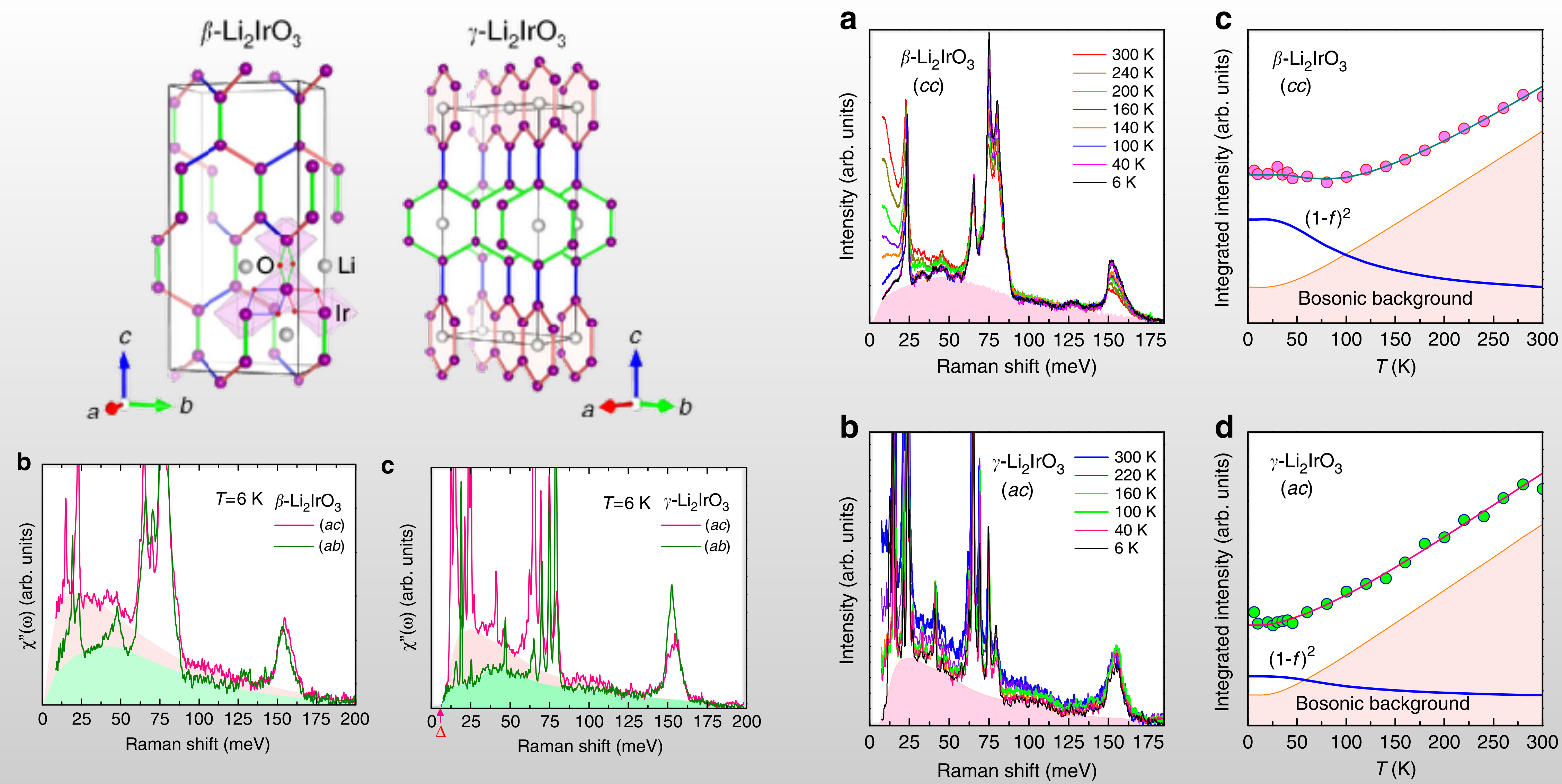


emergent fermionic excitations from spin fractionalization
in a wide temperature range from ~10K to room temperature

J. Nasu, J. Knolle, D. L. Kovrizhin, Y. Motome, and R. Moessner, Nature Physics **12**, 912 (2016)

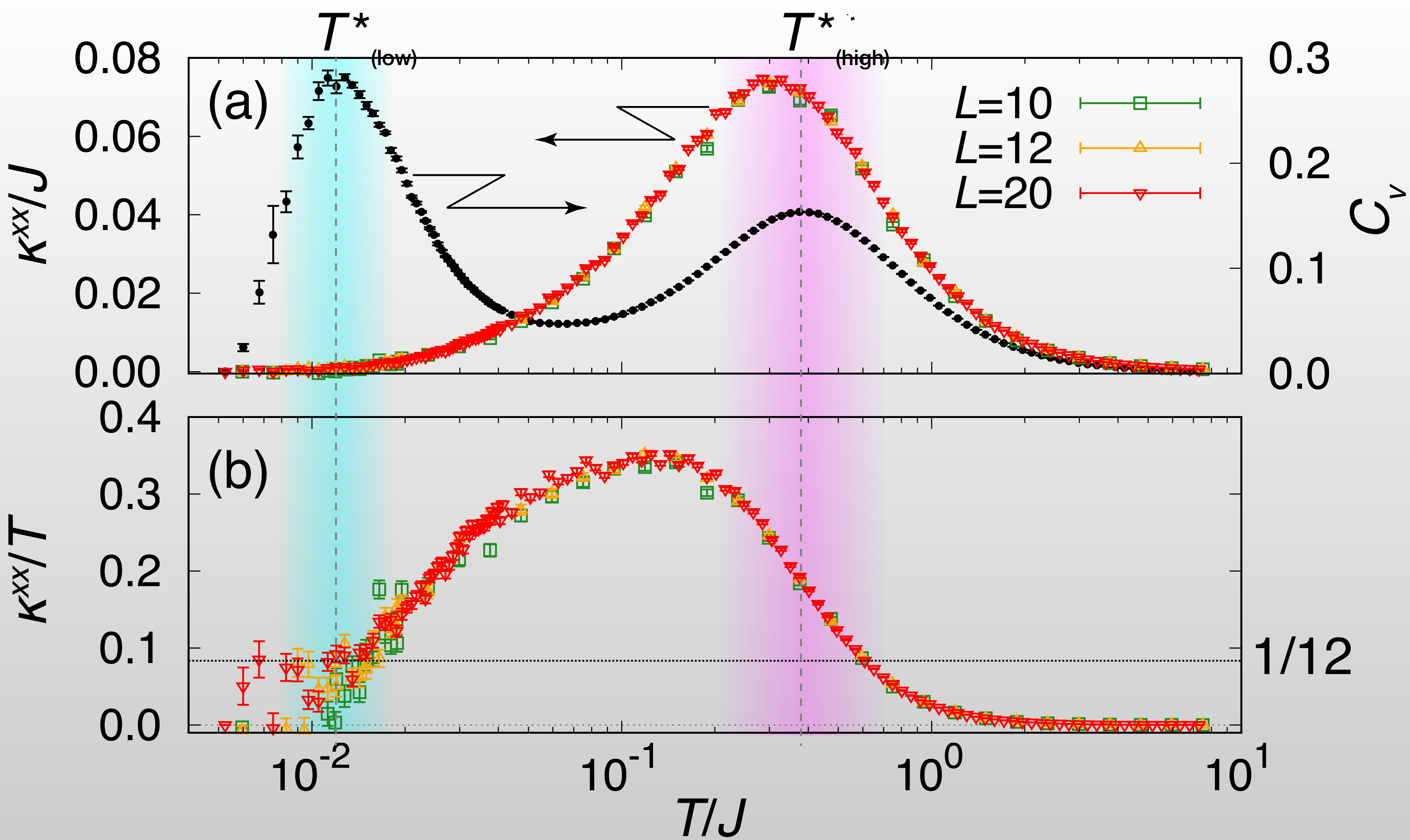
Raman scattering

- *similar (1-f)² behavior was observed also in 3D iridium oxides*



A. Glamazda *et al.*, Nat. Commun. 7, 12286 (2016)

Thermal conductivity: theory

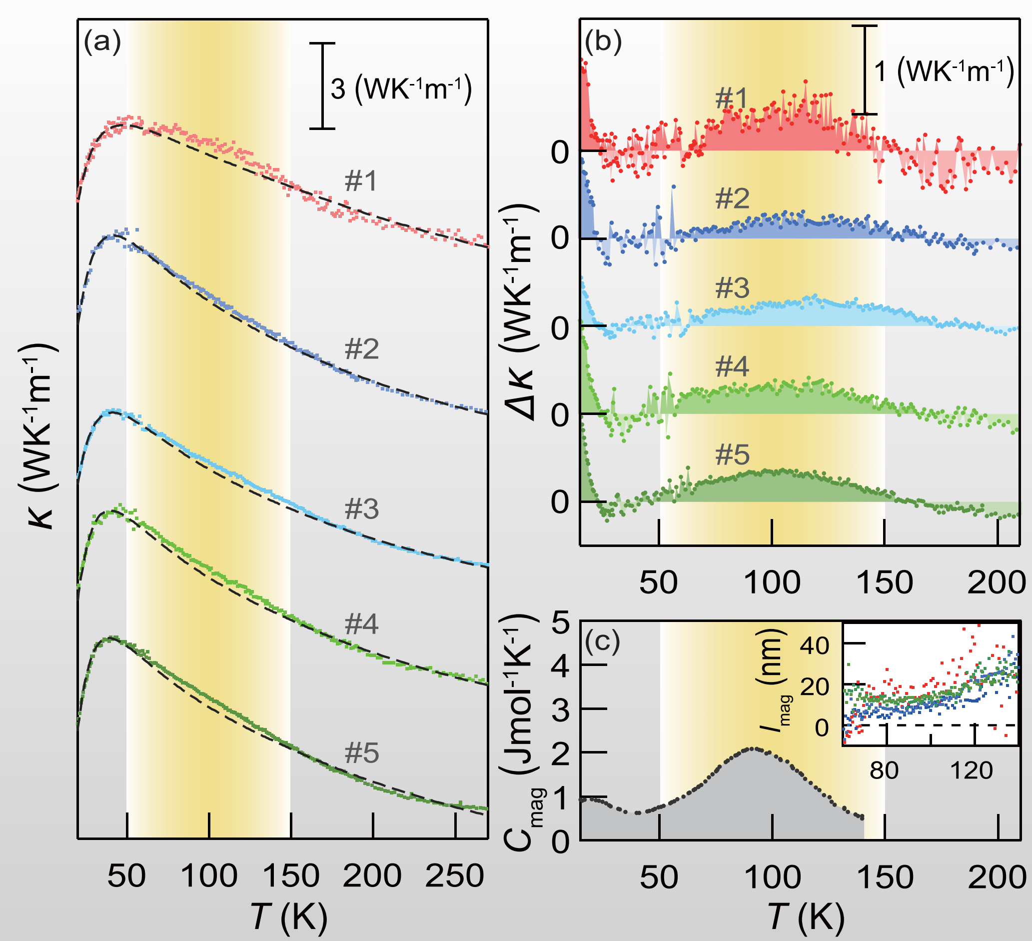


heat conduction below $T^{*(\mathrm{high})}{\sim}J$ by itinerant Majorana fermions

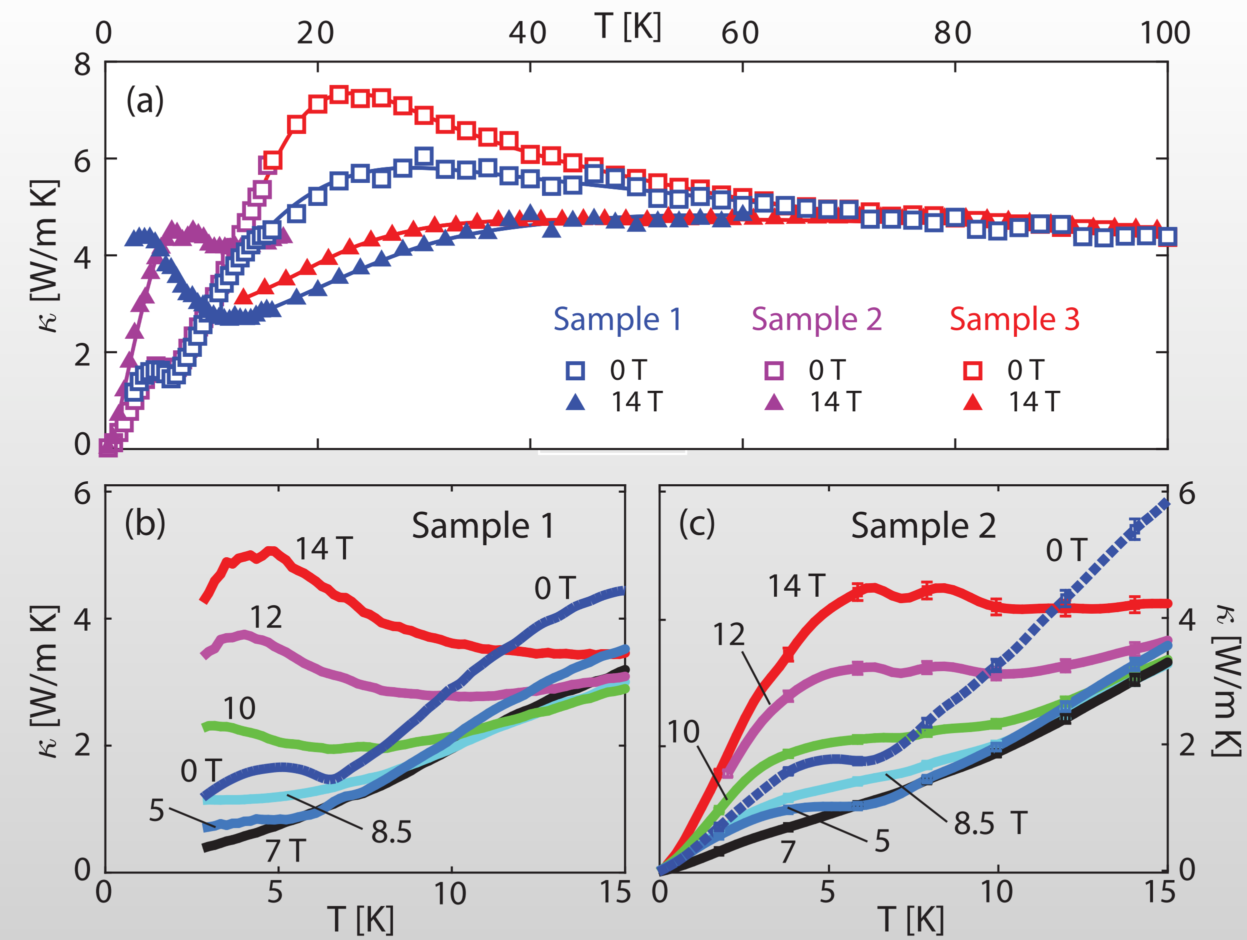
J. Nasu, J. Yoshitake, and Y. Motome, Phys. Rev. Lett. 119, 127204 (2017)

Thermal conductivity: exp.

- seemingly consistent, but scattered data … *difficult to measure at high T, possibly due to contributions from phonons*



D. Hirobe *et al.*, PRB **95**, 241112(R) (2017)

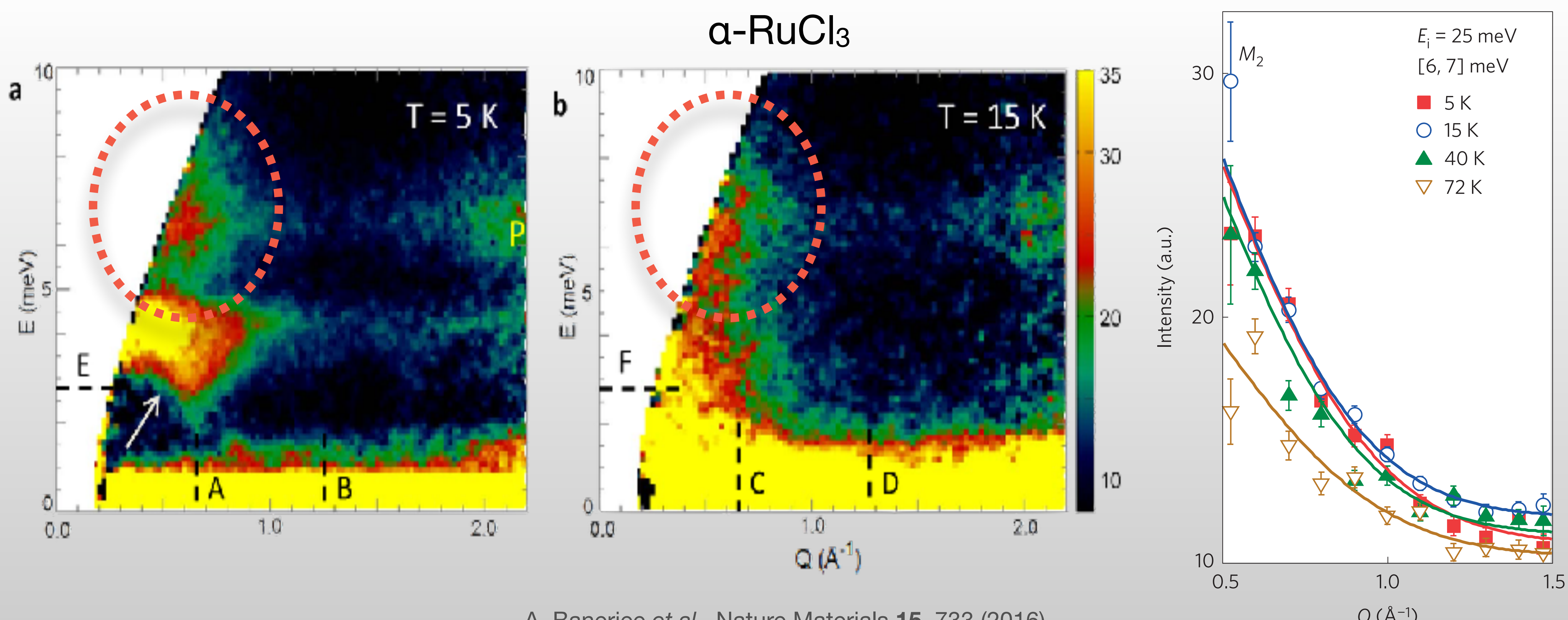


I. A. Leahy *et al.*, PRL **118**, 187203 (2017)

cf. R. Hentrich *et al.*, preprint (arXiv:1703.08623)

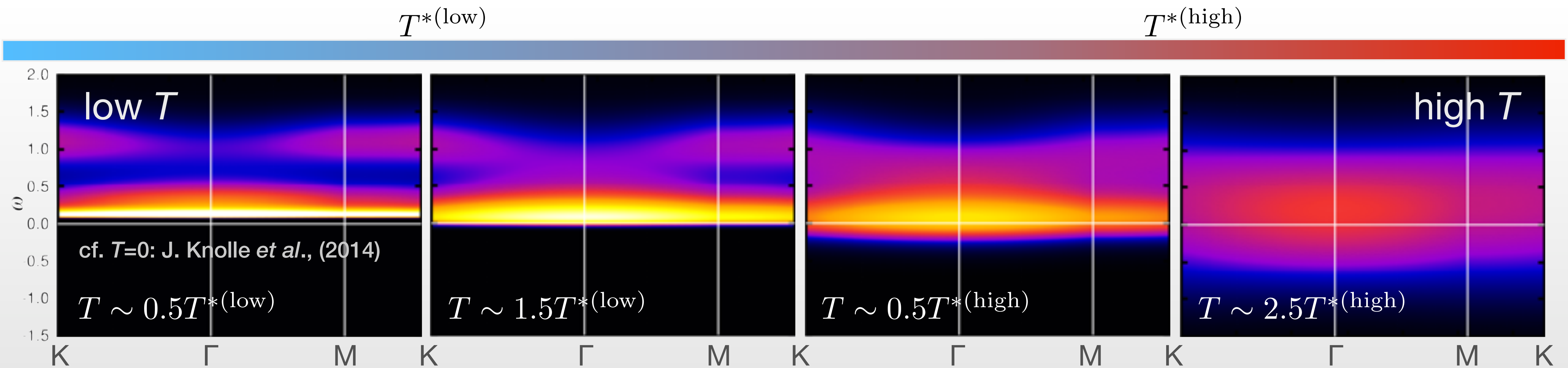
Inelastic neutron scattering

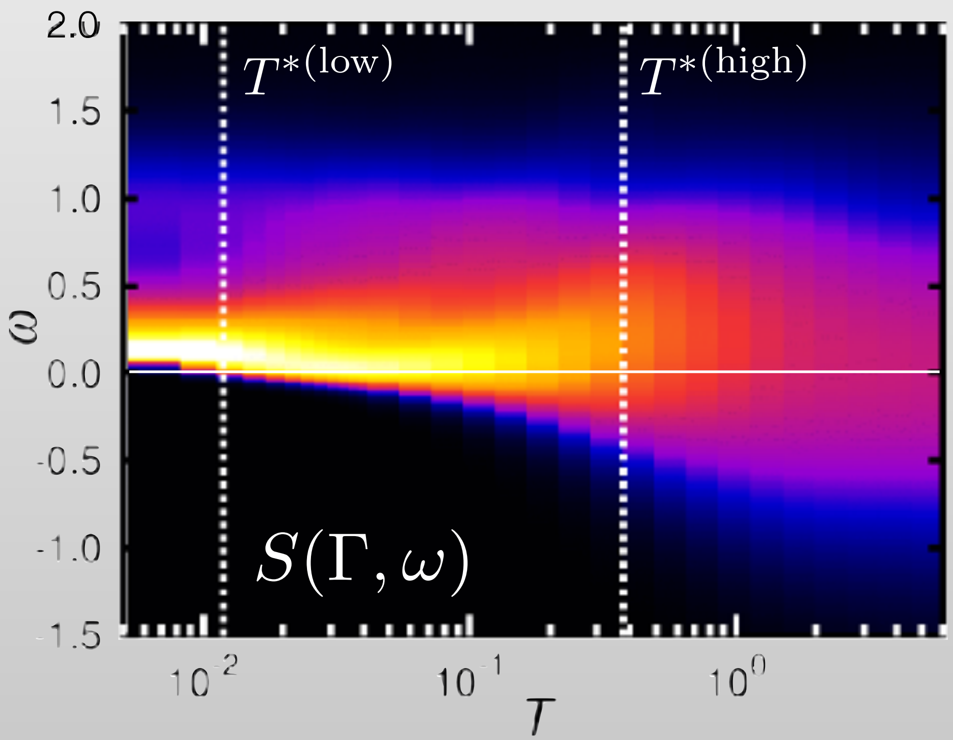
- continuum up to ~8 meV for both below and above T_N, persistent up to ~80 K (powder sample)



A. Banerjee *et al.*, Nature Materials **15**, 733 (2016)

$S(\mathbf{q},\omega)$: theory





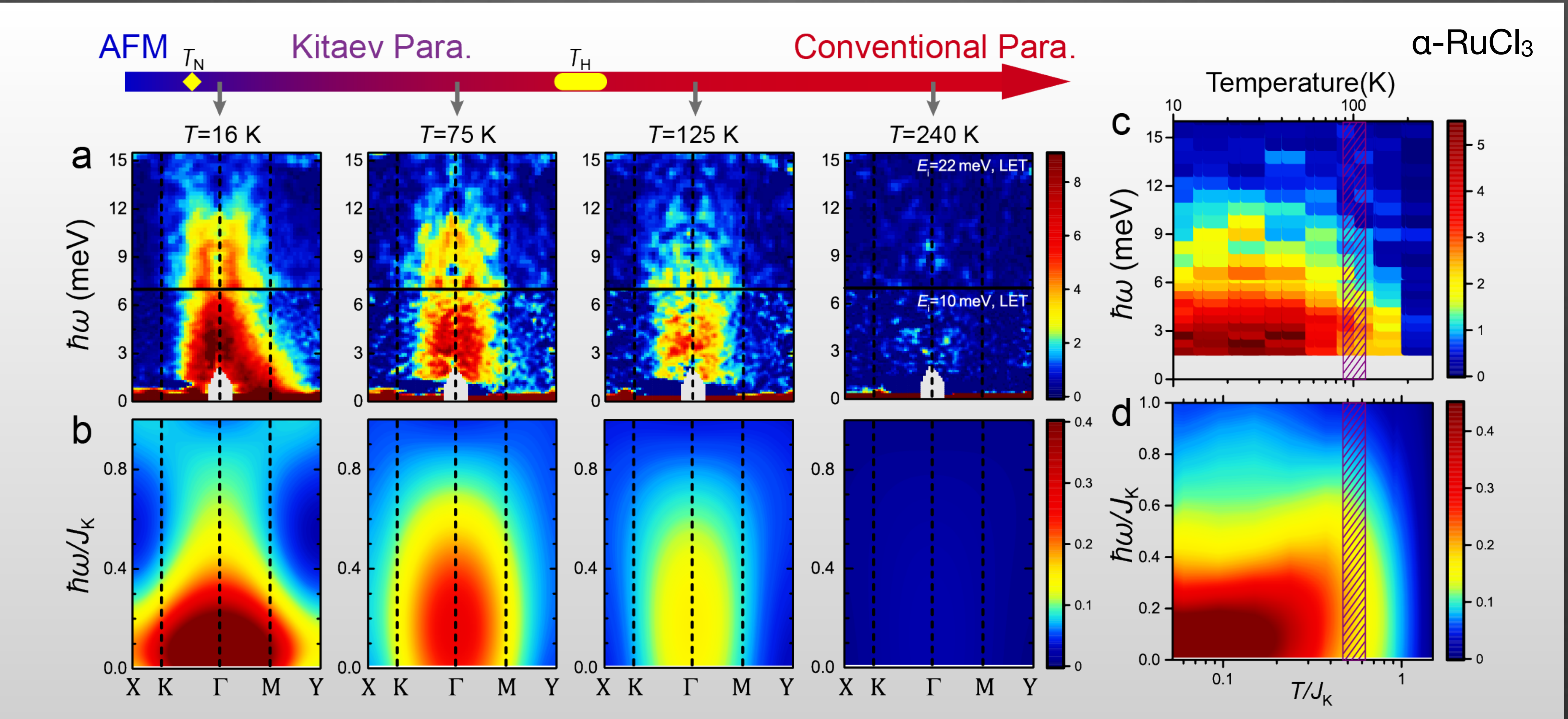
dichotomy of spin excitation:

- growth of continuum up to $\omega \sim J$ below $T \sim T^{*(\mathrm{high})}$
- growth of quasi-elastic response as approaching $T^{*(\mathrm{low})}$

J. Yoshitake, J. Nasu, and Y. Motome, Phys. Rev. Lett. **117**, 157203 (2016)
J. Yoshitake, J. Nasu, Y. Kato, and Y. Motome, Phys. Rev. B **96**, 024438 (2017)
J. Yoshitake, J. Nasu, and Y. Motome, Phys. Rev. B **96**, 064433 (2017)
cf. T. Suzuki *et al.*, Phys. Rev. B **92**, 184411 (2015), Y. Yamaji *et al.*, Phys. Rev. B **93**, 174425 (2016)

$S(\mathbf{q},\omega)$: comparison

- continuum up to ~12meV, persistent up to >100K (single crystal): *fairly good agreement with our theory in (T,ω,**q**) dependences*



S.-H. Do, S.-Y. Park, J. Yoshitake, J. Nasu, Y. Motome *et al.*, Nature Physics **13**, 1709 (2017)

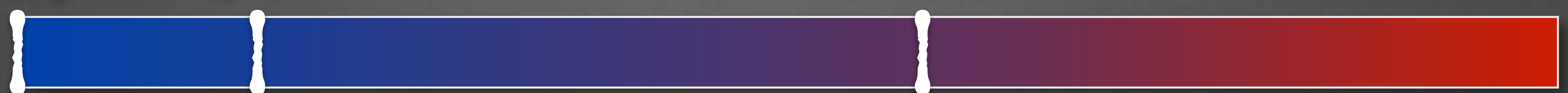
cf. A. Banerjee *et al.*, Nature Materials **15**, 733 (2016); Science **356**, 1055 (2017); npj Quantum Materials **3**, 8 (2018)

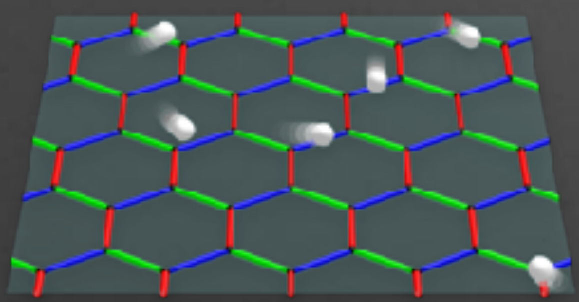
Summary: Majoranization

freezing of Z_2 fluxes

"Fermi degeneracy"
of itinerant Majorana

$T = 0$ $T \lesssim J/100$ $T \sim 0.4J$

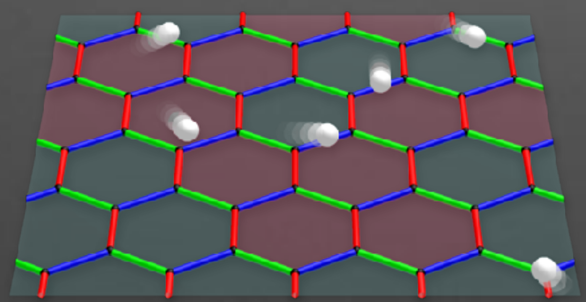




Kitaev QSL

Majoranas w/ no flux

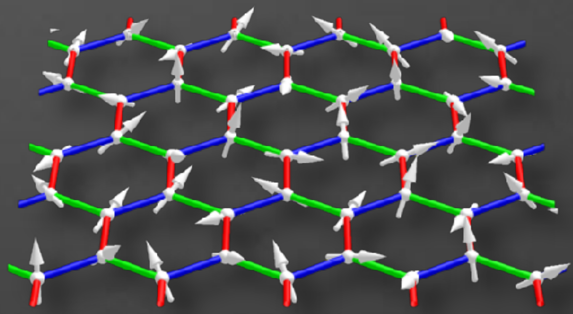
Dirac-type semimetal



fractionalized paramagnet

Majoranas w/ disordered fluxes

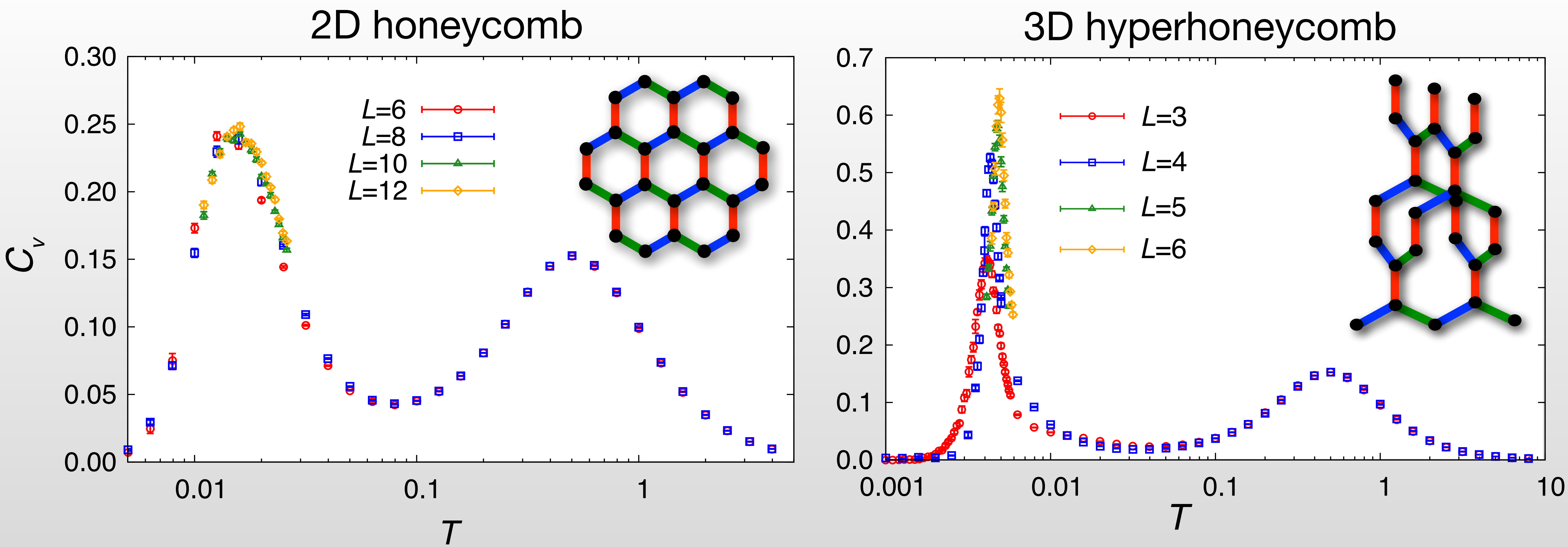
"Majorana metal"



conventional paramagnet

many signatures of spin fractionalization in physical observables ➡ useful for identifying the Kitaev QSLs

Supplemental: dimension matters



broad peak almost independent of the system sizes

➡ just a crossover

sharp peak growing and becoming narrower with the system size

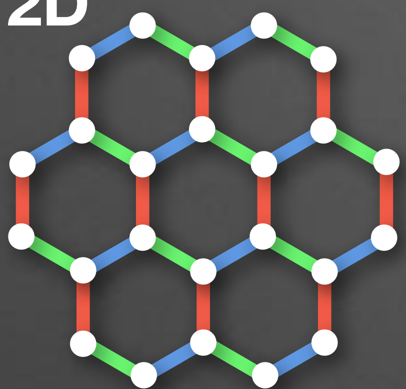
➡ phase transition !!

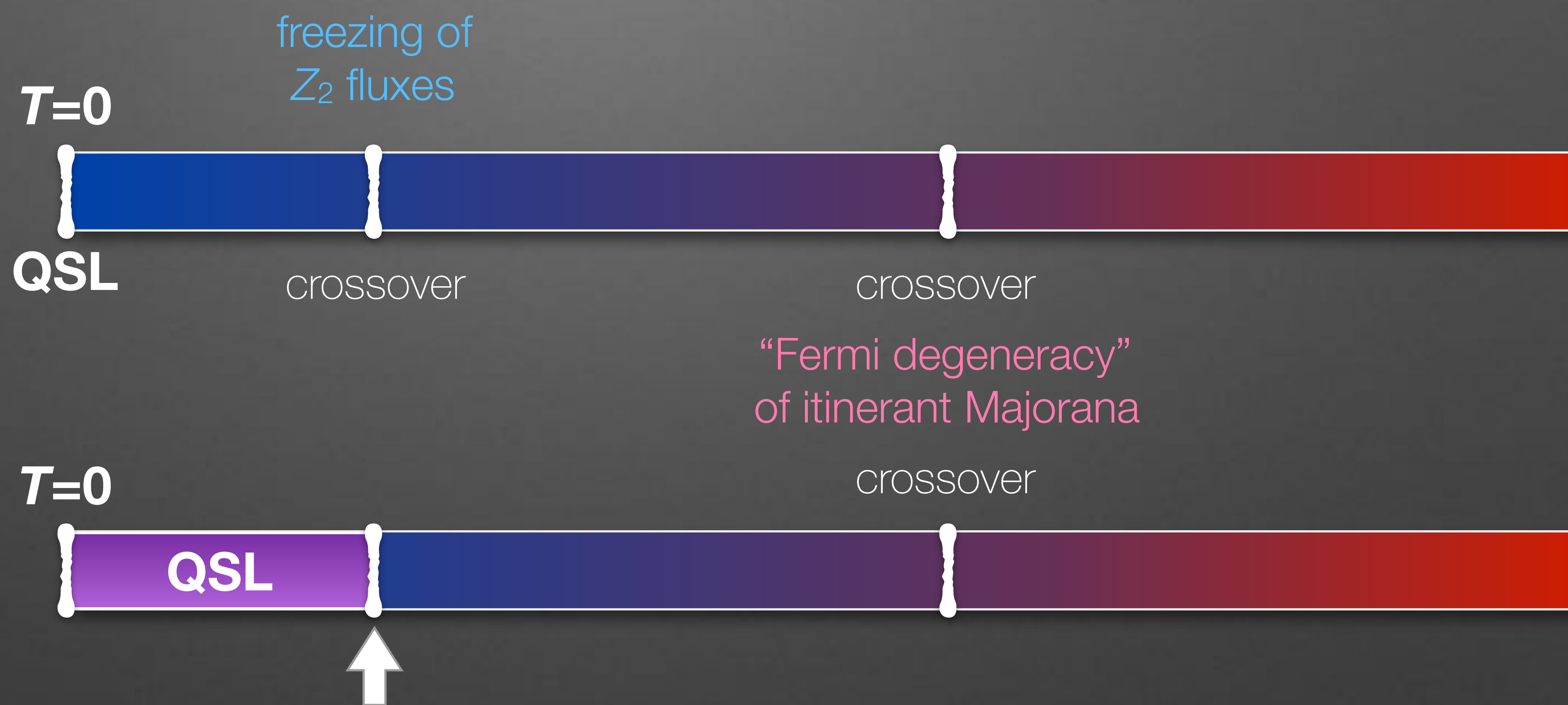
J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. Lett. 113, 197205 (2014)
J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. B 92, 115122 (2015)

Supplemental: “liquid-gas” transition

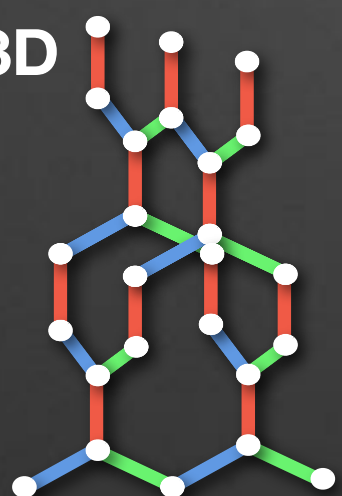
J. Nasu, M. Udagawa, and Y. Motome, Phys. Rev. Lett. **113**, 197205 (2014)

2D





3D



phase transition

- caused by proliferation of “loops” made of flipped Z_2 fluxes (hard local constraint for W_p specific to 3D)
- characterized by the Wilson loop (global order parameter)

NB. A similar transition was also observed on the hyperoctagon lattice.

P. A. Mishchenko, Y. Kato, and YM, PRB **96**, 125124 (2017)

Perspectives

☐ Further comparison between theory and experiment

- detailed analysis for Kitaev and non-Kitaev signatures for further critical comparison

☐ search for other candidate materials

- intercalation, exfoliation, …

K. Kitagawa *et al.*, Nature **554, 341** (2018)
M. Ziatdinov *et al.*, Nat. Commun. **7**, 13774 (2016)
B. Zhou *et al.*, J. Phys. Chem. Solids, in press

- d^7 high-spin systems

H. Liu and G. Khaliullin: Phys. Rev. B **97**, 014407 (2018)
R. Sano, Y. Kato, and Y. Motome: Phys. Rev. B **97**, 014408 (2018)

- f-electron systems

F.-Y. Li *et al.*, Phys. Rev. B **95**, 085132 (2017)
J. G. Rau and M. J. P. Gingras: preprint (arXiv:1802.03024)

☐ effect of a magnetic field

R. Yadav *et al.,* Sci Rep. **6**, 37925 (2016); I. A. Leahy *et al.,* Phys. Rev. Lett. **118**, 187203 (2017);
U. B. Wolter *et al.*: Phys. Rev. B **96**, 041405(R) (2017); S.-H. Baek *et al.,* Phys. Rev. Lett. **119**, 037201 (2017);
J. Zheng *et al.*: Phys. Rev. Lett. **119**, 227208 (2017); A. Little et al., Phys. Rev. Lett. **119**, 227201 (2017);
S. Winter *et al.*, Phys. Rev. Lett. 120, 077203 (2018); R. Hentrich *et al.,* preprint (arXiv:1703.08623);
Y. Kasahara *et al.,* preprint (arXiv:1709.10286); Z. Zhu *et al.*, preprint (arXiv:1710.07595);
P. A. McClarty *et al.*, preprint (arXiv:1802.04283), …

Perspectives

- ☐ experimental detection of the fingerprint of Z_2 fluxes
  - so far, all the signatures are for itinerant Majorana fermions
  - important toward quantum computations: how to generate and control the fractionalized excitations
- ☐ further exotic transitions in 2D and 3D extensions
  - transition to a chiral spin liquid
  - other symmetry breaking?
  - frustration in the Z_2 fluxes?
- ☐ … and more !

J. Nasu and YM, Phys. Rev. Lett. **115**, 087203 (2015)
Y. Kato *et al.*, Phys. Rev. B **96**, 174409 (2017)

Lattice	Majorana metal	TRS breaking
(10,3)a	Fermi surface	Fermi surface
(10,3)b	Nodal line	Weyl nodes
(10,3)c	Nodal line	Fermi surface
(9,3)a*	Weyl nodes	Weyl nodes
(8,3)a	Fermi surface	Fermi surface
(8,3)b	Weyl nodes	Weyl nodes
(8,3)c*	Nodal line	Weyl nodes
(8,3)n	Gapped	Weyl nodes
(6,3)	Dirac cones	Gapped

K. O'Brien, M. Hermanns, and S. Trebst, Phys. Rev. B **93**, 085101 (2016)

Collaborators

My group (Tokyo):

Yasuyuki Kato

Troels Bojesen

Petr Mishchenko

Junki Yoshitake

Ryoya Sano

Former members of my group:

Joji Nasu (TITECH)

Masafumi Udagawa (Gakushuin)

Others:

Yoshitomo Kamiya (RIKEN)

Johannes Knolle (Cambridge)

Dmitry L. Kovrizhin (Cambridge)

Roderich Moessner (MPIPKS)

Experimental colleagues:

Sungdae Ji, Kwang-Yong Choi,

J.-H. Park (POSTECH, Seoul)

Yasuhiro Shimizu, Masayuki Itoh (Nagoya)

Yuji Matsuda, Kasahara Yuichi (Kyoto)

Takasada Shibauchi (Tokyo)

Minoru Yamashita (ISSP)

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